

**DETERMINATION OF THE ACTIVITY CONCENTRATIONS OF RADON-222
AND RADON-220 IN WATER AND SOIL SAMPLES FROM NEWMONT-
AKYEM GOLD MINE USING GAMMA SPECTROMETRY**

BY

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BSc CHEMISTRY, (2000)



**THIS THESIS IS SUBMITTED TO THE
UNIVERSITY OF GHANA, LEGON**

**IN PARTIAL FULFILLMENT OF THE REQUIREMENT FOR THE AWARD OF
MPHIL NUCLEAR SCIENCE AND TECHNOLOGY DEGREE**

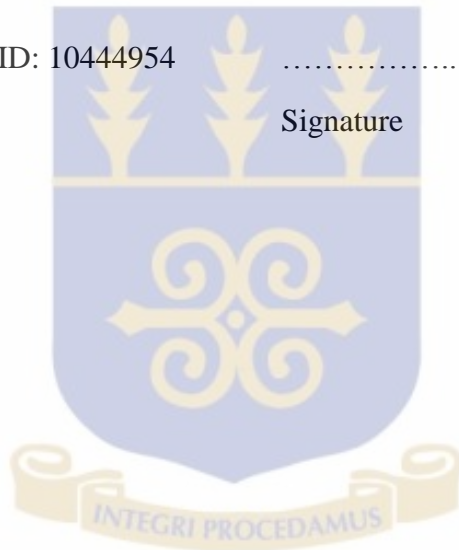
JULY, 2015

DECLARATION

I, **Wiseman Chisale Bekelesi** , hereby declare that this Thesis towards the Master of Philosophy Degree and material contained therein is my genuine work, and have ensured that wherever information was retrieved from previous publications-journals, seminars, textbooks, references have been outlined accordingly.

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DEDICATION

This work is dedicated to my entire family more particularly to my wife Patricia Lewis Bekelesi and my Father, Mr. Bekelesi Chisale Wiseman; Mother, Mrs. Alepha Langisani Bekelesi , my Uncle, Mr. Alfredo Micheta and my two brothers Tobias and Everson Bekelesi.



ABSTRACT

In this study a total of 28 water and soil samples were analysed by High Purity Germanium (HPGe) detector for ^{222}Rn (Radon), ^{220}Rn (Thoron) and other natural radionuclides (^{226}Ra , ^{232}Th plus ^{40}K) activity concentrations. The data generated was then used to assess radiological hazards posed to the public. The radon (Rn-222) concentration in the surface water varied from 0.015 to 1.701 Bq/L with a mean value of 0.913 ± 0.605 Bq/L against the global average of 11 Bq/L and thoron (Rn-220) was not detected in the surface water. For the soil samples, the concentration of radon ranged from 4.194 to 21.114 kBq/m³ with mean value of 10.829 ± 4.130 kBq/m³ while thoron ranged from 0.544 to 13.222 kBq/m³ with the mean value of 5.062 ± 3.051 kBq/m³. In order to assess hazards due to radon concentration in the water to the people, the annual effective doses (AED) were computed. For ingestion of radon in water the AED varied from 0.019 to 4.346 $\mu\text{Sv/y}$ with a mean value of 2.333 ± 1.546 $\mu\text{Sv/y}$ while for inhalation of radon from water varied from 0.019 to 2.146 $\mu\text{Sv/y}$ with mean value of 1.152 ± 0.763 $\mu\text{Sv/y}$ respectively. The total AED due to ingestion and inhalation of radon in water ranged from 0.057 to 6.492 $\mu\text{Sv/y}$ with a mean value of 3.486 ± 2.309 $\mu\text{Sv/y}$. Most people in the study area use soil as building material, thus the assessment of Radium equivalent activity (Ra_{eq}) and Internal hazard index (H_{in}), which are related to radon gas were carried out. The results varied from 19.708 to 69.880 Bq/Kg with mean value of 37.527 ± 15.508 Bq/Kg for Ra_{eq} which is lower than the global limit of 370 Bq/Kg. The H_{in} ranged from 0.065 to 0.248 Bq/Kg with a mean value of 0.132 ± 0.053 Bq/Kg, also lower than the agreed global value of one (1). The study also assessed AED

due to external and internal gamma and the values ranged from 0.060 to 0.175 mSv/y with a mean value of 0.11 ± 0.025 mSv/y against the global value of 1 mSv/y.

Since Annual Effective Dose due to inhalation of indoor radon from soil gas was not conducted it is difficult to conclude whether people at Akyem are safe from radon. However the results for water and soil gas radon have shown that the people may safely use the water for drinking and soil for building dwellings with very minimal radiological hazards.



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LIST OF SYMBOLS AND ACRONYMS

| | |
|-------------------------|--|
| AAS | Atomic Absorption Spectrometry |
| AED | Annual Effective Dose |
| AGM | Akyem Gold Mine |
| Ass | Associate |
| BEIR | Biological Effects of Ionizing Radiation |
| Bq | Becquerel |
| Bq/g | Becquerel per gram |
| Bq/Kg | Becquerel per kilogram |
| Bq/L | Becquerel per liter |
| Bq/m³ | Becquerel per cubic meter |
| °C | Degree Celsius |
| Concn | Concentration |
| DCF | Dose Conversion Factor |
| DG | Director General |
| Dr | Doctor |
| D_R | Dose rate |
| e.g. | For example |
| EIA | Environmental Impact Assessment |
| FWHM | Full Width at Half Maximum |
| GAEC | Ghana Atomic Energy Commission |
| GC-MS | Gas Chromatography-Mass Spectrometry |
| Ge (Li) | Germanium drifted in Lithium |
| GPS | Geographical Positioning System |

| | |
|--------------------------|---|
| Gy | Gray |
| Hin | Internal Hazard Index |
| HPGe | High Purity Germanium detector |
| Inh | Inhalation |
| Ing | Ingestion |
| J/Kg | Joule per kilogram |
| K-40 | Potassium-40 |
| KBq/m³ | Kilobecquerel per cubic meter |
| IAEA | International Atomic Energy Agency |
| ICP-MS | Inductively Coupled Plasma-Mass Spectrometry |
| ICRP | International Commission on Radiological Protection |
| ICRU | International Commission on Radiation Units and Measurement |
| ICMM | International Council on Mining and Metals |
| MBS | Malawi Bureau of Standards |
| MCA | Multi Channel Analyzer |
| MDA | Minimum Detectable Activity |
| M.Phil | Master of Philosophy |
| Mt | Mountain |
| NAA | Neutron Activation Analysis |
| NORM(s) | Naturally Occurring Radioactive Material(s) |
| Prof. | Professor |
| Ra-226 | Radium-226 |
| Raeq | Radium Equivalent Activity |
| RPI | Radiation Protection Institute |
| TENORM | Technologically Enhanced Naturally Occurring Radioactive Material |

| | |
|--|--|
| NAA | Neutron Activation Analysis |
| NaI (TI) | Sodium Iodide Thallium <i>activated detector</i> |
| NTN | National Toxics Network |
| NRC | National Research Council |
| LET | Linear Energy Transfer |
| RBE | Relative Biological Effectiveness |
| Rn-220 | Radon-220 |
| Rn-222 | Radon-222 |
| Th-232 | Thorium-232 |
| U-238 | Uranium-238 |
| UNSCEAR | United Nations Scientific Committee on the Effects of Atomic Radiation |
| WHO | World Health Organization |
| WL | Working Level |
| WLM | Working Level Month |
| γ | Gamma |
| Y | Year |
| Sv | Sievert |
| μSv | Micro Sievert |
| μScm^{-1} | MicroSiemens per centimeter |
| SRS | Safety Report Series |
| SSDL | Secondary Standard Dosimetry Laboratory |
| T1/2 | Half-life |
| TRS | Technical Report Series |
| pH | Power of Hydrogen |
| TDS | Total Dissolved Solids |

ACKNOWLEDGEMENT

Firstly I would like thank YHWH-Almighty Miracle Working God, the creator of entire seen and unseen Universe for being with me throughout the M.Phil Degree Programme.

I wish to express special gratitude to my supervisors Prof. E.O. Darko and Prof. Aba B. Andam for their guidance throughout the project work. Special thanks also go to Rev Dr. Akoto-Bamford, Prof. J.J Fletcher and Dr.D.K. Adotey for their untiresome effort throughout the exercise. Without their input the work would not thrive well. The programme would not have been possible without financial support of International Atomic Energy Agency (IAEA). I am also very grateful to management of Malawi Bureau of Standards (MBS) for their support throughout the two year's period of the M.Phil programme, special thanks also go to Director General (DG), MBS, Mr. Davlin Chokazinga for his encouragement towards advanced studies. Members of staff at the Radiation Protection Institute (RPI) of the Ghana Atomic Energy Commission especially Mr. Oscar Adukpo, David Kpegloh , Charles Kansaana , Ali Ibrahim and Ms. Rita Kpordzro; and Mr. Nicholas Opata of Ghana Nuclear Research Reactor ought to be thanked for their assistance throughout the research work. The Dean, Prof. Yaw-Serfor Armah also played a very crucial role in choosing right supervisors to preside over the research work. Special gratitude also go to my friends: John Mudiwa of Botswana ,Burnnet Msika and Ass. Prof. Samson Sajidu from Malawi.

Finally I wish to pass my heartiest appreciation to my family members likes of Alepha, Leonard, W² (Wiseman/Wisdom), Tobias and Everson Bekelesi; utmost Patricia Lewis Bekelesi and Nellipher Lewis Mchenga whose moral support and prayers made me reach the ultimate goal.

CHAPTER ONE

INTRODUCTION

In this chapter background information pertaining to Naturally Occurring Radioactive Materials (NORMs) in particular Rn-222 and Rn-220 will be explained. The purpose for conducting the research work as regards to the safety of the people and also its relevance, scope and limitations or delimitations if any, will be discussed.

1.1 Background Information

Humans are exposed to ionizing radiation (IR) from natural sources which is on a large scale in the earth's environment and remains in several geological formations in soils, rocks, plants, water and air. The public subjected to IR include natural radiation sources such as cosmic and terrestrial radiation which also involve inhalation or ingestion of natural radioactive materials [Damla, 2009; Kant et al., 2006].

Natural radioactive concentration depends mainly on geological and geographical positions, conditions and is found at various surfaces, locations in soils, water from several geological areas respectively [UNSCEAR, 2000; Duenas et al, 1997].

No human on planet Earth can refrain from exposure to ionizing radiation from natural sources [Tabar et al, 2013]. In this thesis IR will also be referred to simply as radiation.

Studies have shown that radon progeny contributes most of radiation exposure to the people [Garba et al, 2013]. Radon is an odorless and colorless gas that emanates from the earth and along with its decay products, gives out a series of high- and low-Linear Energy Transfer (LET) radiation [BEIR VII, 2006].

There are several isotopes of radon but ^{222}Rn (Radon) and ^{220}Rn (Thoron) are of interest because of their availability in the environment and due to their negative health impacts on the humans. In this document following popular usage Rn-222 will be referred to as “radon” and Rn-220 will be called “thoron”.

Each nuclide has its own contribution to radiation exposure, for instance radon’s half-life of 3.8 days is adequate enough to diffuse into the indoor environment and gives rise to indoor concentration and because of the relatively longer half-life, it can be inhaled into the respiratory tract and irradiate the cells. On the contrary the half-life of thoron is only 56 seconds which implies that its presence is limited to close proximity [Yamada, 2006].

Uranium (^{238}U) and Thorium (^{232}Th) are the ultimate progenitors of ^{222}Rn , ^{220}Rn respectively; and the immediate mother radionuclides of radon, thoron are radium-226, radium-224, respectively [WHO, 2001]. Despite that Rn-220 comes from the disintegration of Ra-224, it is often characterized as a decay product of Ra-228, which is a longer-lived parent ($t_{1/2} = 5.75\text{y}$) commonly analyzed in the environmental samples such as soil and water.

Ramachandran (2010) noted that both radon and thoron are omnipresent in both working and public environments, although their activity concentrations vary considerably.

The hazardous exposure to radiation due to radon have been earmarked globally mostly because of its association with the cancer of the lung. Both radon and thoron progenies disintegrate by giving out alpha (α) particles which are proceeded by emission of gamma (γ) radiation. While γ -rays are comparatively less harmful to the bronchi than α and β particles, they are largely deposited in other body tissues and organs. The half-life of radon (3.8 days) is long enough for it to spread while thoron half-life (56 seconds) is of

little significance compared to the latter [Banari et al, 2012]. However, recent studies in some countries have shown that in certain circumstances the doses from thoron and its progeny are notable and comparable to those from radon [Sciocchetti, 1992].

Exposure due to radon is the most variable and the International Atomic Energy Agency (IAEA) has been working hand in hand with the World Health Organization (WHO) in finding means of raising the awareness of radon as a public health and radiation protection issue and to support the Member States with technical guidance for establishment and implementation of radon action plans. Generally, the system of radiological protection ought to deal much with exposure due to natural sources of ionizing radiation more particularly radon [IAEA, 2014].

Scientific Committee under United Nations on the Effects of Atomic Radiation, postulated that the exposure to natural sources of radiation constitutes more than 60% of the population radiation dose, whereas 50% is as a result of inhalation and ingestion of natural radioactive gas radon and its decay products. It is assessed that exposure to radon through inhalation in closed rooms is the cause of about 10% of all deaths from the cancer of the lung [UNSCEAR, 2000; Forkapic et al, 2006].

However studies have shown that data on the thoron concentration are still very little since its determination is difficult [Yu et al, 2000].

While exposure to natural sources is unpreventable, there are conditions where exposures to natural sources of radiation may need attention. A good scenario is the accumulation of high concentrations of radon and thoron in air. Another case is the mining and/or processing of mineral or material where the activity concentrations of radionuclides of natural origin in the material itself, or in any material arising from the process, are

significantly elevated known as Naturally Occurring Radioactive Material (NORM) [IAEA, 2005].

The radionuclides embedded in bedrock are weathered off; chemically or physically and by means of transportation they end up deposited in rivers, lakes or seas. Other human practices such as mining and mineral processing increases the concentration both in end products or wastes to produce Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) [Langat, 2009].

Both NORM/TENORM radioactive elements found in the environment, include ^{238}U , ^{232}Th and ^{40}K and a series of their progeny such as ^{228}Ra , ^{224}Ra , ^{226}Ra as well as radon and thoron gases. These radioisotopes can pollute the environment and bring constraints to public well-being [Peroni et al, 2012].

Although the intensity of these radiation sources is much smaller than produced by human activities such as mining and mineral processing, no human can avoid these natural sources. Consequently, the effects of the natural radiation background happens to be a crucial health issue, particularly radon and thoron levels. The information on the radioactivity levels in the soil and water is necessary for the estimation of possible radiological hazards to human health [Chowdhury et al, 1998].

Upadhyay et al (2007) reported that measurement of radon, thoron and their progenies is critical because the radiation dose to the people as a result of inhalation of radon alone and its progeny cover over half of the dose from all sources of radiation, both naturally occurring and man-made. Further, a linkage between cancer of the lung and inhalation of radon and its progeny has been studied and recent epidemiological evidence suggests that

inhalation of radon and its decay products in domestic environment initiates the cancer of the lung.

Shoeib and Thabayneh (2014) noted that inhalation of radon and its progeny also causes skin cancer and kidney diseases besides lung cancer and that radiological impact commenced by radionuclides is as a result of radiation exposure of the body by the gamma rays and irradiation of lung tissues from inhalation of radon and its progeny. Thus in order to combat radiological hazard to the people, it is necessary to know the dose limits of community exposure to radon.

However it has not been easy to produce a standard for radon, in comparison to other radioisotopes in drinking water, mainly because of lack of guidelines concerning dosimetry baseline data on radon diffusing in water. In addition, radon poses a unique regulatory constraint as a consequence of it being migrating into the air which initiates a risk from the inhalation of its progeny. Thus, it is safeguarded as a radioisotope in the water, while the main part of the related danger occurs due to its contribution to the radon concentration in the air [NRC, 1999].

While mining has been seen as one of the principal sources of exposure to NORM/TENORM, the mining companies are not provided with guidelines for these radioactive materials in most countries including Ghana due to insufficient safeguards for their regulation by the Radiation Protection Board. The health concern of NORM/TENORM is focused primarily with the production and release of radon, thoron gases produced through the radioactive decay of ^{226}Ra , ^{224}Ra respectively. The inhalation of radon has been accompanied with high risk of the cancer of the lung [BEIR IV, 1988].

Ghana is conducting numerous mining activities which means that possibility of producing NORM which is the main source of radon and thoron gases is very high.

For radiation protection purposes it is preferably important to monitor the presence and concentration of radon decay products, while for the identification of the sources and origin of radon the measurement of its concentrations in air or water, and sometimes its exhalation from soil and building materials, are more significant [Tykva and Sabol, 1995].

While ingestion of radon in water poses a possible risk, however in normal conditions water collected on the surface seldom has high levels of radon diffused in it, comparable to underground water [NRC, 1999]. In contrast mining activities may elevate radon levels in the surface water [Mohammed, 2011].

Radon in water typically contributes only a small proportion to the indoor air concentration while most of radon in indoor air is generated from radon in the soil diffused into dwellings. Usually the danger to the people from the radon indoors is influenced by the radon from soil gas [NRC, 1999].

Studies have shown that radon activity concentrations in the soil few meters from the surface are very crucial in evaluating the doses for the indoor radon gas. While the indoor radon come from soil and water in comparison more radon come from the soil than from the water [Elmoniem, 2015].

This research was carried out with the aim of assessing radon and thoron levels in surface water and soil at Akyem-Newmont Gold mine and surrounding communities in the Eastern Region of Ghana following the mining and mineral processing activities conducted there. In addition the study looked at NORMs of concern which could play a

significant role in gamma ray radiological hazard assessment on humans. This was accomplished using gamma ray spectrometry system equipped with High Purity Germanium (HPGe) detector.

1.2 Research Problem

Darko et al (2005, 2010) reported that in Ghana there are more than 200 mining companies embarking on small, medium to large scale operation and that there is limited data on the levels of environmental radioactivity concentrations and public exposure due to mining and mineral processing activities.

Likewise Newmont- Akyem is not exceptional, not much work has been done on radon and thoron. Consequently, there exist inadequate information on potential health hazards to workers and the public in the surrounding areas from NORM/TENORM (particularly radon and thoron).

The workers at Newmont-Akyem and the communities surrounding the mine are likely threatened by a number of environmental problems such as ingestion of radon from surface water and inhalation of radon from soil which is used as building material for dwellings. Most of the previous studies conducted on water quality and soil focused on the chemical, physical and microbiological analysis but not much research has been done on NORMs/TENORMs particularly radon and thoron.

Akyem final Environmental Impact Assessment (EIA) report on existing environment (2008) showed that because of the seasonal nature of stream flow in the Akyem area, surface water availability during the dry season is scarce. During the wet season, water in streams and rivers in the Study Area is used for bathing, recreation, drinking, irrigation of

crops and aquatic life support. During the dry season, fetch points are dug by hand in the channels in remote areas to obtain drinking water and for other uses when there is no flow in the streams [Akyem EIS, 2008].

Therefore it was necessary to estimate natural radioactivity levels particularly radon and thoron in both water and soil. In addition it was necessary to assess radon and thoron levels in the soil since it is used as building material for dwellings.

According to Mohammed (2011) and IAEA (2006) findings, radon in surface water is not a major problem as most of it diffuses into the atmosphere, but its concentration may be influenced by various factors such as the geology of the area, bottom sediments and mining or mineral processing activities going on in the area.

With the mining and mineral processing activities conducted at Newmont-Akyem, it is likely that the levels of radon and thoron are elevated. It was therefore necessary to determine concentrations of radon and thoron in water and soil samples and compare the results with national and international standards as well as work done elsewhere.

1.3 Objectives

The main objective of this study was to assess the health hazards from NORMs in particular radon due to operational activities of Newmont-Akyem Gold Mine

Specific objectives:

1. To estimate radon (^{222}Rn), thoron (^{220}Rn) levels in water and soil samples based on activity concentrations of Ra-226, Ra-228 respectively.
2. To evaluate the radiological hazards due to NORMs through assessment of the:
 - i Annual Effective Dose (AED) due to Ingestion, Inhalation of radon in water
 - ii AED due to Terrestrial radiation

iii Radium Equivalent Activity and Internal Hazard Index

1.4 Justification

Water is very essential to both workers and the public around Newmont-Akyem mine, thus, its quality ought to be monitored. In a similar manner, it is vital to estimate radon and thoron levels as their progenitors ^{238}U and ^{232}Th are spread throughout the earth crust and soil is the main source of indoor radon. However, in Ghana, not much data on NORMs particularly radon and thoron in soil and water has been done for establishing database on radionuclides and radioactivity levels in the environment. The database is needed to enhance knowledge and understanding on the behavior of the radionuclides and their migration in the environment that can be applicable for radiological assessment.

Risk assessment and management of radon due to NORMs entering or present in the environment as a result of industrial activities associated with mining has not been carried out extensively almost in all the mines in Ghana.

Faanu (2011) reported that there is widespread lack of awareness and knowledge of the radiological hazards and exposure levels by law makers and operators in Ghana. As a consequence determination of radioactivity for the presence and concentration of radon in soils and water which could be used in building houses by communities in and around the Newmont-Akyem mine is important for developing safeguards.

1.5 Significance of the Project

In many African states including Ghana, mining industries have not been subjected to radiological regulatory control. Baseline data on NORMs including radon on the public exposure is limited [Darko et al, 2005]. As a result, the public does not have enough knowledge on the radiological hazards and exposure levels of NORMs including radon.

Therefore there is need to conduct more research work on NORMs to cover all gold mines in Ghana. This is very vital because currently Ghana is in the process of developing guidelines on standards pertaining to the regulation of NORMs in the mining sector [Faanu, 2011].

1.6 Scope and Limitations

This project work embarked on the following:

Firstly, before collecting water and soil samples, absorbed doses were measured by a survey meter at a height of 1m above the sampling ground level in order to determine absorbed dose. In addition the physico-chemical properties of water were tested using portable water kit. Then the water and soil samples were collected and transported to the laboratory for thorough sample preparation.

Secondly, the water and soil samples were measured by using Gamma Spectrometry System equipped with a High Purity Germanium (HPGe) detector. Concentrations of Rn-222, Rn-220 and other NORMs Ra-226, Th-232 and K-40 were calculated accordingly.

Thirdly, radiological hazard assessment was done based on:-

- Radon ingestion in water and inhalation of radon originating from water:
Where annual effective doses are computed based on UNSCEAR (2000) and ICRP 2007 expressions
- Terrestrial radiation: since people are exposed to gamma radiation, where annual absorbed doses are measured in the field and also calculated
- Hazard indices: due to use of soil as building material where Radium Equivalent Activity (Raeq) and Internal Hazard Index (Hin) are calculated.

Finally, one of the limitation that was faced in this research was inability for the direct measurement of indoor radon to determine AED for the public. Although activity concentrations for the two radioactive gases were measured it was difficult to assess hazards based on soil gas radon and thoron concentrations alone as literature shows that results found are limited [Varley and Flowers ,1998]. Therefore there is need to conduct another research whereby direct measurement of indoor radon is done in order to determine real AED due to radon emanating from the soil and water.

1.7 Structure of the Thesis

The Thesis consists of five main chapters:

- ❖ The first chapter outlines background information pertaining to this project work by looking at the main objective of the research, its significance , potential and the safety of the people
- ❖ The second chapter portrays more on literature centered on the research work, more particularly what has been accomplished by Ghanaian and International researchers. It has also examined the theoretical aspects taken in the previous studies and how the same techniques can be utilized in the present work
- ❖ Chapter three covers materials and methodology utilized where appropriate outline of the procedure applied in performing calculations has been explained
- ❖ The fourth chapter covers the results found and eventually thorough discussion of the findings thereby giving meaning to the results and their effects
- ❖ Finally, the fifth chapter talks about conclusion of the work, summarizing the work and giving out the positive and negative effects of the findings and

eventually also contains recommendations as regards to the results of the project work and also potential areas for future research

CHAPTER TWO

2.0. LITERATURE REVIEW

This section will discuss literature related to natural sources of ionizing radiation in particular radon and thoron ranging from background information, origin of radium and radon, thoron, their toxicity, secular equilibrium, and what has been done in Ghana on radon and thoron. In addition a review of a number of different measurement strategies available, ranging from analysis of water and soil samples, through dose-rate measurement, to gamma-ray spectrometry will be carried out.

2.1 Ionizing radiation and its source

Ionizing radiation is energy in the form of particles or waves that is sufficient to remove electrons from the shells of atoms. The nuclei of unstable atoms are one of the sources of radiation. As these radioisotopes seek to become more stable, their nuclei give out particles and high-energy waves in a process termed as radioactive decay. The major types of radiation emitted during radioactive decay are alpha and beta particles, including gamma rays. Radiation can come from natural sources or man-made radionuclides. Some radioisotopes, such as radium, uranium, and thorium, have been there since the creation of planet earth. The radioactive gas radon is one type of NORM produced as these radioactive materials decay and add over 50% of radiation exposure to public (Figure 2-1). Regardless of how they are generated, all radioisotopes emit radiation.

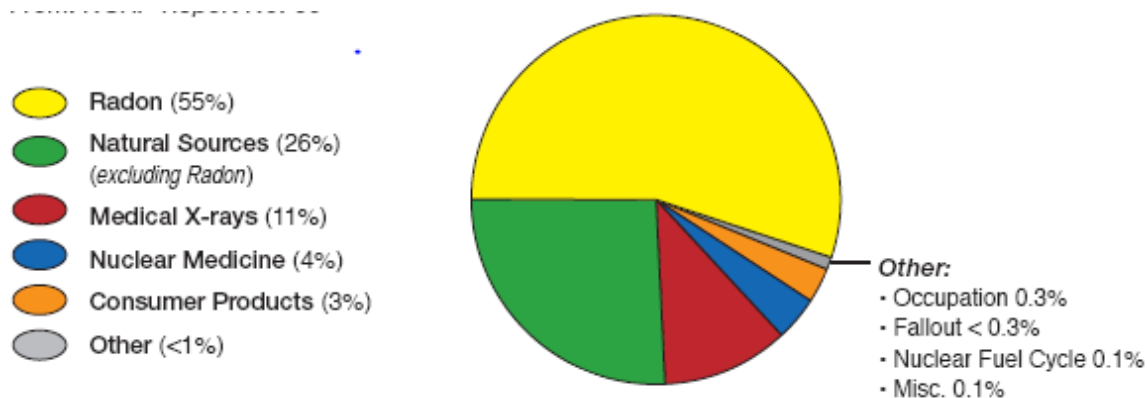


Figure 2- 1: Sources of radiation exposure [UNSCEAR, 2000; USEPA, 2007]

2.2 Properties of Radon

Radon is a natural inert radioactive gas, without any taste and smell, it has density of approximately 8 times bigger than that of air. It dissolves in water and easily mixes with other gases and water vapor, thus producing remarkable concentrations. The earth's crust contains trace amounts of ^{238}U and ^{232}Th which decompose to ^{222}Rn (radon) and ^{220}Rn (thoron) respectively. The first four offspring of radon— ^{218}Po , ^{214}Pb , ^{214}Bi , and ^{214}Po —are also radioactive and are altogether called radon decay products or simply progeny. They are all solid metals and have half-lives spanning from a fraction of a second to several minutes (Figure 2-2). ^{218}Po and ^{214}Po are alpha emitters, while ^{214}Pb and ^{214}Bi are beta/gamma emitters [Assad & Salih, 2008; NRC, 1999].

The well-known three naturally existing radon isotopes ^{222}Rn , ^{220}Rn and ^{219}Rn are produced on the alpha decay of their radium parents ^{226}Ra , ^{224}Ra and ^{223}Ra , respectively (Figs 2-2,2-3 & 2-4. ^{222}Rn , ^{220}Rn and ^{219}Rn are distinctively known as: radon, thoron, actinon respectively; they are all isotopes of radon [Ishimori et al, 2013]. Equations 1-3 illustrate mode of formation of the three isotopes of radon.

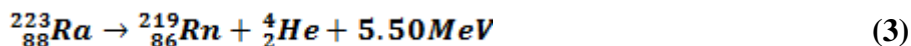
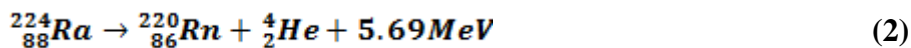
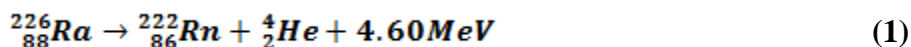


Table 2- 1: Physicochemical properties for ${}^{222}\text{Rn}$, ${}^{220}\text{Rn}$ and ${}^{219}\text{Rn}$

| Parameter | notation | ${}^{222}\text{Rn}$ | ${}^{220}\text{Rn}$ | ${}^{219}\text{Rn}$ |
|--------------------------------|--------------------|---------------------------------------|---------------------------------|--------------------------------|
| Half-life | $t_{1/2}$ | 3.8232(8)d | 55.8(3)s | 3.98(3)s |
| Decay constant | λ | $2.0984 \times 10^{-6}/\text{s}$ | $1.242 \times 10^{-2}/\text{s}$ | $1.74 \times 10^{-1}/\text{s}$ |
| Diffusion coefficient in air | D_{air} | $1 \times 10^{-5}\text{m}^2/\text{s}$ | | |
| Diffusion coefficient in water | D_{water} | $1 \times 10^{-9}\text{m}^2/\text{s}$ | | |

[Ishimori et al, 2013]

2.3 Origin of Radon in the environment

Radon, an inert gas under usual environmental conditions, is one of the NORMs and the decay product of radium. Radium is an alkaline earth metal with the atomic number 88, a Group IIA element in the Periodic Table. It was firstly discovered by Marie and Pierre Curie in 1898. At first, radium was not categorized as possible hazard for workers or ordinary people and was earlier on used for diagnostic and therapeutic

medical purposes until in mid-19th century that it was referred to as pollutant of the environment [IAEA, 2010]. The decay of the long lived radioisotopes ^{232}Th , ^{238}U and ^{235}U lead to four naturally occurring nuclides of radium. The ^{232}Th decay chain comes up with ^{228}Ra and ^{224}Ra while ^{238}U chain gives rise to ^{226}Ra , and ^{223}Ra being a member of the ^{235}U series [Lawrie et al, 2000].

Radium-226 and its decay products, members of the uranium chain, are responsible for a major proportion of the internal dose received by humans from NORM. ^{226}Ra with a half-life of 1600 years is an alpha emitter and decays, to radon-222, which has a half-life of 3.82 days (Fig 2-2). The decay of ^{222}Rn is preceded by the consecutive disintegration of a number of short-lived alpha and beta-particle emitting decay products. Radium itself adds little to the gamma-ray activity of the environment, but it does so indirectly through its gamma-ray-emitting decay products [BEIR VII, 2006].

All isotopes of radium disintegrate into stable ^{206}Pb , ^{208}Pb and ^{207}Pb respectively among all three series. ^{226}Ra , being an alpha emitter and the most common isotope with the longer half-life, is accompanied by gamma radiation in its decay process. Radium-224, Ra-228 have half-lives of 3.66days, 5.76 years respectively. Two radium isotopes ^{226}Ra and ^{228}Ra are very significant from radiologically protection viewpoint due to their relatively long half-life presence in nature and high dose conversion factors [IAEA, 2010].

^{226}Ra is found in all rocks and soils in different amounts. In nature, ^{226}Ra is generally in rough equilibrium with ^{238}U , so the concentrations for ^{238}U can be assumed to be a good guide to the expected range for ^{226}Ra [BEIR VII, 2006].

Radon-222 is daughter of ^{226}Ra in ^{238}U decay series. It is a radioactive noble gas that generally lacks activity toward other chemical agents. ^{222}Rn is the most important isotope because of its relatively long half-life compared to ^{219}Rn (4 seconds) and ^{220}Rn (56 seconds) in the ^{235}U and ^{232}Th which are immediate daughters of ^{223}Ra and ^{224}Ra respectively [Forkapic et al, 2006].

According to Ramachandran (2010), thoron was discovered by R.B.Owens and Ernest Rutherford in 1899. Important step in ^{220}Rn research took place in the atmospheric sciences where it was found out that atmospheric ions close to the earth's surface initiated generation of thunderstorms.

The chemical element radon in the periodic table is the most dense with atomic mass of 86 among the inert gases such as neon, argon, krypton and xenon.

Radon is the chief source (approximately 55%) of internal radiation to human life (Figure 2-1) [ICRP, 1993; USEPA, 2007]. Worldwide average year-long effective dose from natural sources of radiation is estimated to be 2.4 mSv, of which about 1.0 mSv comes from radioactive exposure [UNSCEAR, 2000].

Because of their different contribution to the total annual effective dose-radon and thoron and their decay products are responsible for 1.2 mSv and 0.07 mSv (world-wide average) respectively. Radon and its progeny are of primary concern. From the radiological point of view radon progeny are more important than radon itself. This is because the effective dose from the inhalation of radon contributes only to 5% of the total radon-related dose, i.e., 95% of this dose is delivered by the short-lived radon decay products ^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po and long-lived decay products ^{210}Pb , ^{210}Po [Tykva and Sabol, 1995]

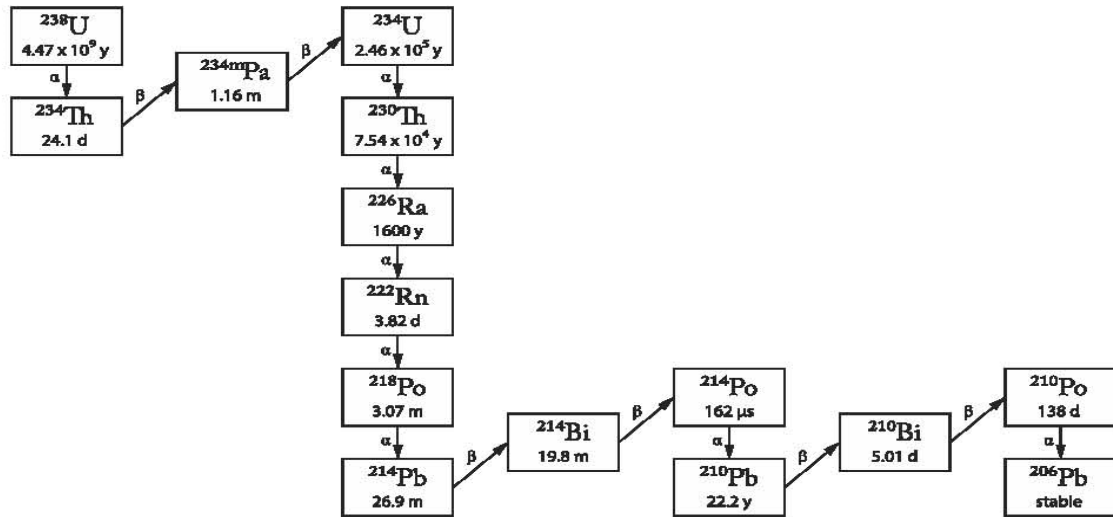


Figure 2- 2: Uranium decay series [Ishimori et al, 2013].

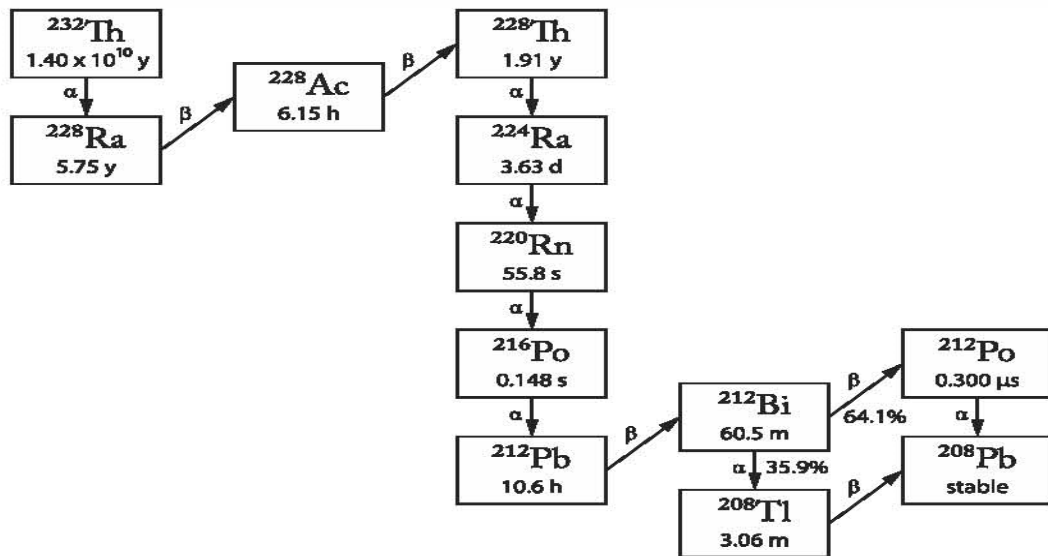


Figure 2- 3: Thorium decay series [Ishimori et al, 2013].

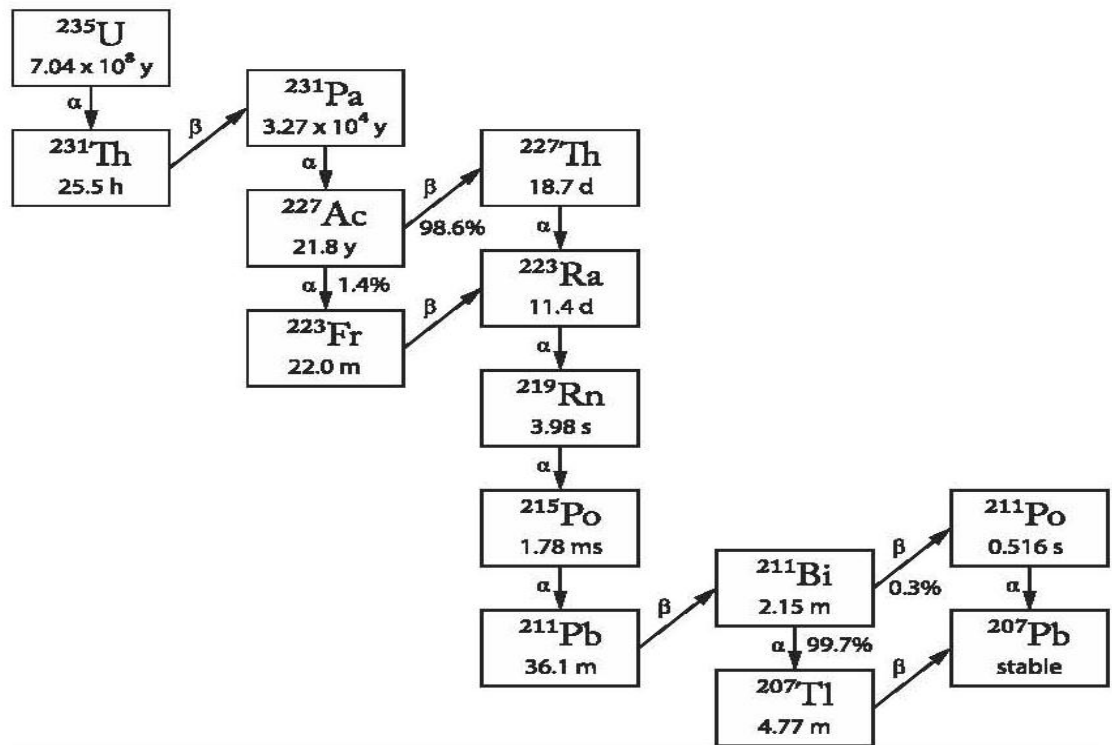


Figure 2- 4 : Actinium decay series [Ishimori et al, 2013].

2.4 Radon release to atmosphere

Radon atoms attached to soil particles are not easily released to the atmosphere because of low diffusion coefficient. However if they are located in an interstice gap between the grains they may diffuse readily to the surface. In summary radon can be given off to the atmosphere by emanation, transport and exhalation processes (Figure 2-5) [Ishimori et al, 2013].

Definitions

Emanation refers to radon atoms generated from the decay of radium in which radon escape from the grains.

Transport means diffusion and advective flow which causes the migration of the

emanated radon atoms through the soil profile to the ground surface.

Exhalation represents radon atoms that have been diffused to the ground surface and then given off into the atmosphere.

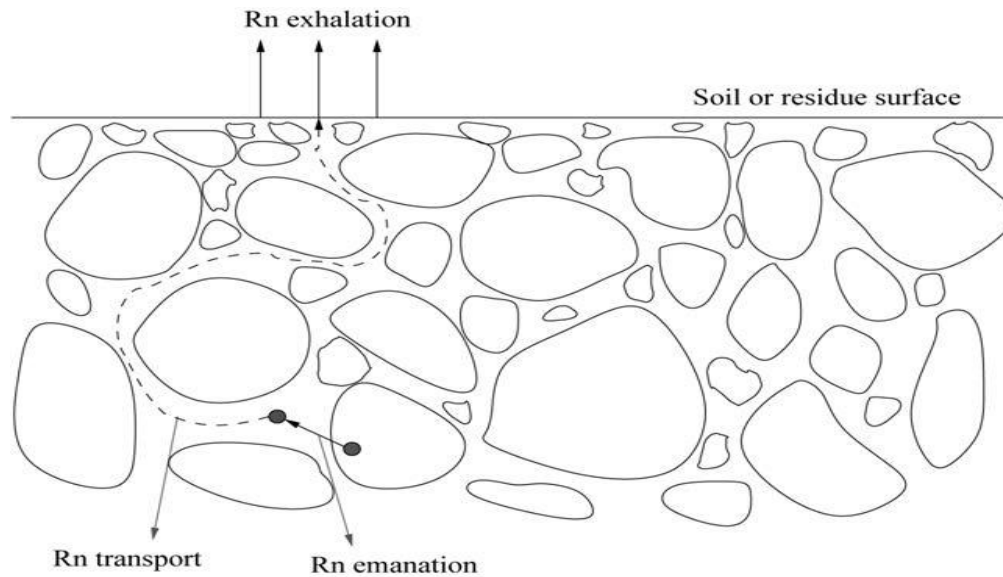


Figure 2- 5 : Processes leading to radon release to the atmosphere [Ishimori et al, 2013].

The dissemination of radium in soil grains are explained as the principal factor that affects emanation of radon. Past models of radon emission had postulated that radium is spread equally throughout the grain and that these grains are also spherical. However this is contrary to the common circumstance where uranium and radium movement lead to higher fraction of radium embedded on the surface of soil grains [Cameron, 2005].

2.5 Toxicity of Radium, Radon and Thoron to human beings

The radiation that liaises with the living tissue may cause transformation in the body of the person who has been subjected to, referred to as somatic effects and there is also a probability of causing variation in the person's offspring called the hereditary effects. Some somatic effects are non-stochastic or deterministic i.e. they do not take place unless a certain threshold dose has been surpassed. While other somatic and hereditary effects are stochastic i.e. the chance of the effect occurring increases with the amount of radiation acquired and there is no known threshold dose below which the effect does not occur [Michael Paul, 1989].

Radium gives out a variety of radiation, such as alpha particles and gamma rays. Alpha particles become harmful when they interact with the body. On the contrary subjection to gamma radiation is risky either inside or outside the body. Long-term exposure to radium adds the danger of causing several cancers such as cancer of the bone, liver and the breast, and lymphoma cancers [NTN, 2013]. External exposure to radium's gamma radiation enhances the risk of cancer of various types in the body organs. But the major impact of radium is its daughter radon which among others also causes the cancer of the lung [<http://www.epa.gov/radiation/radionuclides/radium.html> (2014-03-21)].

Radon and its progeny are steadily generated by decomposing of radium-226, a member of the naturally occurring uranium-238 series. Airborne concentrations of radon's short-lived decay products (Po-218, Pb-214, Bi-214, and Po-214) and long lived progeny Po-210 and Pb-210 are of concern due to their prospects for accumulating in the lung, which follows with irradiation of lung tissue by alpha emissions from Po-218 and Po-214. While thoron and its short-lived progeny are produced naturally and repeatedly from

decay of Th-232, in contrast with radon, it is noted that little thoron reaches the breathing zone because of its short half-life (56 seconds) compared with longer 3.8 days for radon [Strom & Reif, 1996].

Internal hazards of thoron are due to breathing in of dust comprising thorium and its progeny [Ramachandran, 2010].

2.6 Radon and cancer of the lung

Radon is only one of causes of the cancer of the lung (Table 2-2). The epidemic of the cancer of the lung worldwide largely agrees with trends in cigarette-smoking, the principal cause of lung cancer [BEIR VI, 1999].

Table 2- 2: Risk factors that may induce the cancer of the lung

| Rank | Cancer risk factor |
|------|---|
| 1 | Indulge in active cigarette-smoking |
| 2 | Get involved in passive cigarette-smoking |
| 3 | Radon itself |

[BEIR VI, 1999]

2.7 Exposure pathways

Worldwide exposure to radon and its progeny is usually considered as the only origin of radiation hazard at workplaces. However for those working underground the mine, gamma radiation and internal contamination could be significant source of hazard due to elevated concentration of radium and barium. Furthermore the release of water and deposits containing radium may also lead to the pollution of the environment in the vicinity of the mines [IAEA, 2005].

However, the home may be categorized as the main place where the public can readily be exposed to radon. The chief source of the radon in the home is the soil gas that penetrates from beneath and around the structure. Radon dissolved in water and the building materials that contain some proportion of radium may also release radon. Under some situations, water can contribute a remarkable percentage of airborne radon but water is inferior source of population exposure to radon progeny [BEIR VI, 1999].

The determination of radon in the environment is of great importance because of its alpha emitting nature. Radon monitoring in the air can be done indoor, outdoor and in underground mining environment. The sources of indoor radon are: building materials, water, basement air, soil and outdoor air among others [Richard, 1991]. Certain percentage of the radon given off into the outdoors is quickly diluted and its effects are ignored. However in confined spaces such as homes and office buildings radon can be elevated to harmful levels. Radon concentration in soil air changes during the year owing to variability in the permeability and water content of the soil as well as ventilation of the above horizons [Mehra & Bala, 2014].

2.8 Research on Radon in Ghana

Extraction of minerals from the ground is an unpleasantly rough task. In order to clarify how bad it is, it is estimated that production of only a single gold ring, requires generation of 20 tonnes of mine wastes. The waste could go into the environment (air, water, land, animals and people). Mining affects the quality of the air, and gold mining is categorized as one of the most unsafe industries worldwide following all the hazardous chemicals and gases it releases into the environment (<http://www.scribid.com/issue-analysis-effects-of-mining-on-the-environment>).

In most countries including Ghana the health issues from NORMs is affiliated mainly with the generation and release of radon gas [Faanu et al, 2011; UNSCEAR, 2000].

In Ghana there have been studies conducted on the indoor and outdoor radon in the environment by Andam and Amoo (2007) at Tarkwa and Prestea Goldfields ; Akwasi & Asumadu-Sakyi et al , (2012) around faulted areas in Accra,. Quashie et al, (2011) at Kassena Nankana Area of the Upper East Region of Ghana , and others.

However not much research has been conducted in Ghana on radon associated with NORMs in the mining industry. It has been outlined that Ghana has over 200 registered mining companies ranging from small, medium and large scale. Thus it is clear that a lot of research need to be done in Ghana concerning NORMs more particularly radon and thoron.

2.9 Regulations and guidance for Radon and Thoron in Ghana

Ghana lacks guidelines on regulations and recommendations for protection against radon and thoron progeny in the mine workplaces and areas surrounding the mines.

However international recommendations for protection against radon have been outlined by the International Commission for Radiological Protection (ICRP) in several publications. In addition, the ICRP considers that the dose conversion convention should be used to translate radon exposure into effective dose, which should then be added to the other effective doses (external and internal doses) for comprehensive assessment. The International Atomic Energy Agency (IAEA) embraced the ICRP recommendations, and also made recommendations for thoron [Strom & Reif, 1996].

2.10 Sample analysis

Both U-238 and Th-232 radionuclides contain not less than 12 radioisotopes of which some emit alpha, beta particles followed by gamma emission. This implies that measurement of each of the members of the decay series is not realistic in terms of time and cost constraints [Lumbardo and Mucha, 2008].

The decay chains of U-238 and Th-232 are subsequently affiliated with gamma transitions. The determination of these radioactivity sources can be conducted through a wide set of methods, such as gamma-ray spectroscopy, alpha spectrometry, neutron activation analysis, just to mention a few [IAEA, 2006].

2.10.1 Determination of Radium and Radon

Since not all progeny of the natural decay series give out gamma, the activity depends on theory of secular equilibrium and the spotting of gamma emitters down the series. For example, Ra-228 does not release a gamma ray for quantification by gamma

spectrometry but its closest daughter, Ac-228 emits a gamma ray that can be quantified. Nonetheless the assumption of secular equilibrium is credible as it is established relatively quickly between ^{228}Ra and ^{228}Ac since the half-life of ^{228}Ac is relatively short (6.1 hours) [Lumbardo and Mucha, 2008].

The availability of ^{226}Ra depends on the amount of ^{238}U . Concurrently these two radioelements are often found to be in disequilibrium due to their different chemical properties thus they have to be handled separately. Radium-226 is determined from the γ -ray emissions of the ^{222}Rn progeny (^{214}Pb and ^{214}Bi) after the sample container is sealed and stored for 4 weeks to ensure secular equilibrium between ^{226}Ra and ^{222}Rn . Likewise, ^{224}Ra can be presumed to be in secular equilibrium with ^{220}Rn for air-tight samples and its concentration can be obtained from the measurement of the ^{220}Rn γ -emitter decay products (^{212}Pb , ^{212}Bi and ^{212}Tl). However, the midway radioelements in the ^{232}Th series of ^{224}Ra have relatively short half-life, hence a γ -spectrometry of the ^{232}Th content bearing in mind the peaks of the intermediate γ -emitters, ^{228}Ra and ^{228}Ac is a good sign of the rate of ^{220}Rn generation within the reference soil (<http://www.tdx.cat/rn.and.tn.fundamentals.pdf>).

2.11 Measurement Techniques

2.11.1 Gamma (γ) ray measurement

Gamma-ray spectrometry is one of the powerful non-destructive analytical tools for the measurement of the gamma emitters both qualitatively and quantitatively. Its outstanding benefit is that it allows the concurrent measurement of many radioisotopes in a bulk sample, without unnecessary time consuming radiochemical sample separation and preparation [Saidou , 2008; IAEA, 1989].

In terms of solid materials comprising soils and sediments the sample can often be put into the sample holder or container after only very minimal sample preparation (e.g. drying, ashing, sieving etc). In order to ensure sample uniformity it is advisable to use the larger sample portions [IAEA, 2010].

The purpose of gamma spectrometric monitoring of environmental samples according to IAEA (2004) is to quantify the activity concentration of gamma ray emitting radioisotopes and the accompanied error analysis of the results. The technique is mostly applied to non-destructive analysis of environmental samples but it can also be conducted in destructive analysis [IAEA, 2004]

Gamma spectrometry is one of the techniques often used in the determination of several environmental radioisotopes. Simultaneous analysis of the absolute activity of different gamma peaks in a wide energy range can be obtained and in order to compute the absolute activity, the detector full-energy peak efficiency has to be determined.

There are two main types of detectors frequently used for gamma ray measurement and these are: Sodium Iodide Thallium activated (NaI(Tl)) and High Purity Germanium (HPGe) detectors. The capture of γ -ray in an absorber such as NaI(Tl) or HPGe detector

occur by any of three processes: pair production, photoelectric and Compton effect. In energy determination of γ -rays it is essential to capture the total energy and to minimize the loss of energy by escape of the scattered γ -rays from Compton interaction. The resolution is the determining component in the ability of the gamma spectrometry system to distinguish between photons of γ -rays of the same energy.

The following figure illustrates the resolution of the NaI(Tl) and HPGe detectors

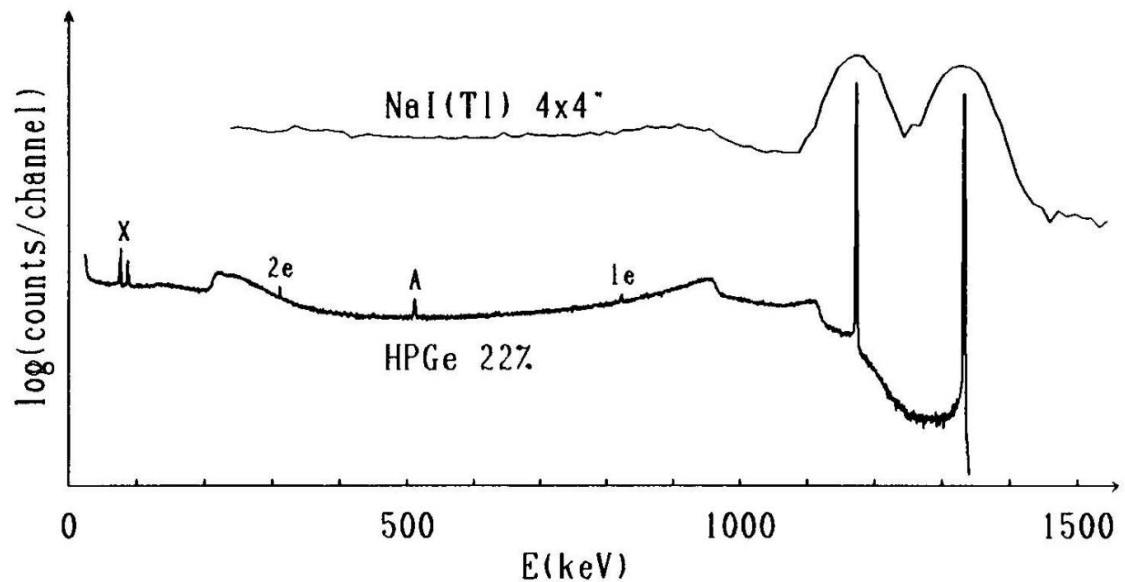


Figure 2- 6 : Comparison between HPGe and NaI (Tl) detectors [Choppin, 2002]

Gamma-ray spectra are measured with modular equipment consisting of a detector, an amplifier, a pulse-height analyzer, memory, and a permanent data storage apparatus. Germanium drifted in Lithium, Ge (Li), or most recommended High Purity Germanium detectors (p-type or n-type) are used for the determination of complex gamma (γ)-ray spectra due to their excellent energy resolutions. These Ge detectors, however, are very durable and need to be cooled by liquid nitrogen which is also costly. Thallium

activated Sodium-Iodide crystals, can be functional at ambient temperatures. On the other hand, their energy resolutions limit their use [HASL-300, 1997].

2.11.2 Germanium detector operational characteristics

Room-temperature operation of germanium detectors of any type is impossible in contrast to sodium iodide detectors because of the large thermally-induced leakage current that would result due to the small band gap (0.7 eV). Thus it is necessary to supply liquid nitrogen to germanium detectors to lessen the leakage current to the point that the associated noise does not spoil their excellent energy resolution. Under normal circumstances, the temperature is decreased to 77 K by means of an insulated dewar flask where a supply of liquid nitrogen is stored in thermal contact with the detector [Knoll, 2000].

However for Germanium Lithium [Ge(Li)] detectors, the low temperature must be maintained unceasingly to control unnecessary redistribution of the drifted lithium that will rapidly take place at ambient temperature. Lithium drifting is eliminated in HPGe detectors, and they can be allowed to warm to room temperature between uses. Fabrication techniques have been designed to the point that modern detectors will withstand indefinite cycling of the temperature. This operational benefit is the major reason that HPGe detectors have supplanted Ge(Li) configurations. Figure 2-7 below shows HPGe detector [Knoll,2000].

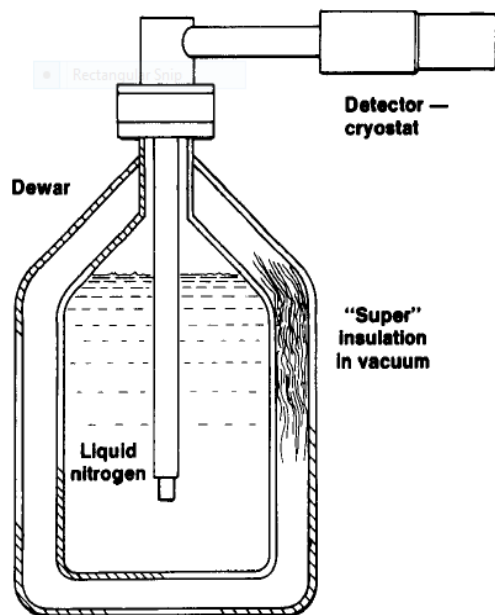


Figure 2- 7 : Germanium detector cooled by liquid nitrogen [Knoll, 2000]

2.11.3 Calibration of Germanium Detector Systems

The calibration of the Ge system is critical to achieving credible results [ANSI, 1999].

Proper calibration safeguards that gamma ray spectra are correctly clarified by means of activity concentration and the energy. Before the samples are measured the analyst has to do the following tasks [Gilmore, 1995]:

- Energy calibration which is for the identification of radionuclides
- Peak width calibration used as a function of energy
- Efficiency calibration which links decay rate against count rate applicable for quantification purposes

It is essential that calibration of gamma spectrometers is conducted properly before the sample can be measured. Standards are often obtained from various manufacturers (or prepared in the lab).

2.11.3.1 Energy Calibration

Energy calibration is achieved by determining energies of corresponding peaks using a standard, which may contain Co-57, Sn-113, Hg-203, Cd-109, Ce-144, Cs-137, Co-60, and Y-88. Usually the sources comprises well known peak energies of particular radioisotopes of interest which include the whole energy region showing a wide spectral range from 50 to 2000 keV over which the spectrometer is to be used. The energy calibration can either be performed manually or automatically [Gilmore, 1995; Luca et al, 2012]. The statistics of subsequent measurements demand that adequate counts be obtained in each energy peak so that sufficient uncertainty levels are attained [ANSI, 1999].

The energy calibration consists in the experimental determination of a function under normal conditions a first or second degree polynomial, depicting the energy dependence of the channel number in the spectrum [Luca et al, 2012]:

$$E_{\gamma} = A + B \cdot Ch + Ch^2 \quad (4)$$

Where E_{γ} is the gamma-ray energy,

Ch is the spectral channel number for the center of the peak corresponding to E_{γ} (usually the channel with the maximum number of counts),

A, B and C are constants to be determined for calibration.

2.11.3.2 Peak width calibration-Energy Resolution

One of the vital terms in gamma ray spectrometry is the energy resolution which is associated with the detector response. The resolution is referred to as the ability of the detector to differentiate between two radiations whose energies lie close to each other.

It is usually represented by means of the full width at the half of the maximum pulse height distribution [Fuad, 2008]:

The resolution calibration is also called Full-Width at Half-Maximum (FWHM) calibration which sets up a function to portray the peak width versus the spectral energy.

It is an important parameter featuring the system performance in separating several photon emissions in a narrow energy range [Luca et al, 2012].

Energy resolution ordinarily defined as the quotient of FWHM and the location of the peak centroid (H_0) is given by [Fuad 2008 ; Knoll,2000]: .

$$R = \frac{FWHM}{H_0} \quad (5)$$

Where:

R, H_0 corresponds to energy resolution, peak energy respectively

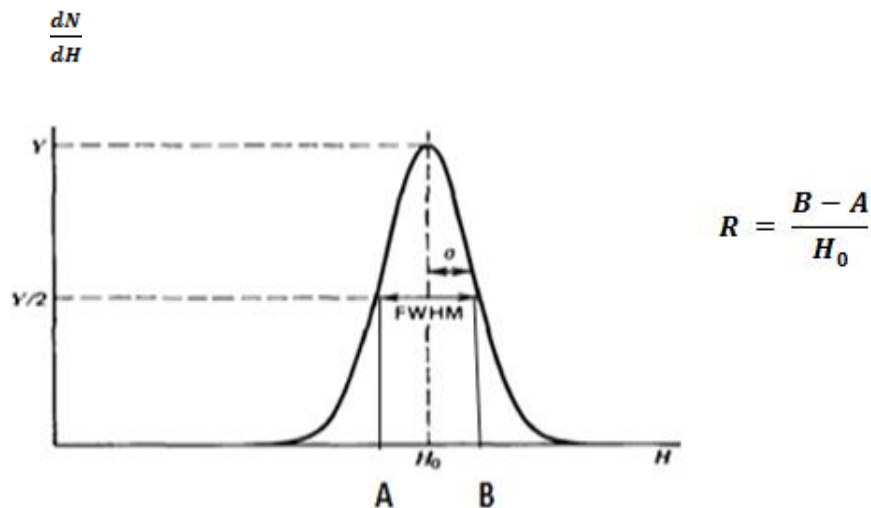


Figure 2- 8: Definition of detector resolution [Knoll, 2000]

The following Figure 2-9 outlines the difference between good and bad resolution.

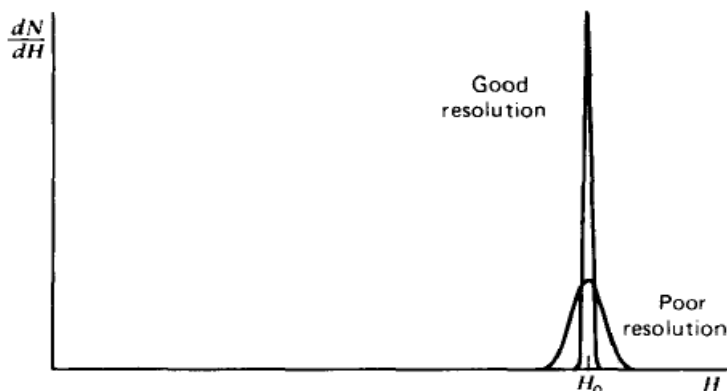


Figure 2- 9: Good resolution and relatively poor resolution [Knoll, 2000].

2.11.3.3 Efficiency calibration

It is frequently used practice, when determining the activity of gamma emitters with HPGe detectors, to compare measurements of an unknown source with those of a calibrated source of equivalent geometry and density. The total efficiency is termed as the ratio of the number of pulses counted in the spectrum to the number of photons emitted by the source [Daraban et al, 2013].

The efficiency of gamma spectrometry is not absolute, so the objective behind efficiency calibration of the system is the determination of a factor corresponding to each gamma line that normalizes their activity concentration to its absolute value [Bakr et al, 2011].

The efficiency calibration involves the calculation of the efficiency of the semi-conductor detector system as a function of energy. This comprises effects from the intrinsic detector crystal, the detector-source geometry, the materials surrounding the detector and the absorption in the source matrix. The efficiency calibration is needed for each source-detector combination such as disc source, Marinelli container etc [IAEA, 2004].

The efficiency calibration using standard sources of known initial activity is a procedure intermittently performed for the HPGe detector, as in Vukanac et al (2008) where the connection between the energy of the gamma radiation and the number of the channel is established. Upon the identification of the energy by standard sources, the efficiency value is computed taking into account the chance of disintegration for each energy. This data is required for the calibration of the detector efficiency, according to the following formula [IAEA, 2004; Daraban et al, 2013]:

$$\varepsilon = \frac{N}{A\gamma t_s MK} \quad (6)$$

Where:

ε is the efficiency at photopeak energy,

N is the number of counts

A is absolute/specific activity of the standard

γ is the emission probability of the gamma line corresponding to the peak energy,

t_s is the sample live counting time

M is the mass [kg] of the measured standard,

K is the correction factor

$$K = \frac{1}{e^{-\lambda T_d}}$$

Where

λ is the decay constant of individual radionuclides and

T_d is the time between sampling and time of counting

2.11.4 Minimum Detectable Activity (MDA)

Another important calibration parameter is the Minimum Detectable Activity (MDA) for the radionuclides of interest (based on background measurement) [Luca et al, 2012].

Credible data is defined as being spectral data in which the peaks of interest are well shaped, positioned and have good "signal to noise." This is an important consideration; just having more data doesn't make it better. One tactic of the quality of a spectrum is the minimum detectable activity (MDA) of the detector system. The resolution, background and efficiency of the detector are affiliated to the MDA. This connection may be simply equated as: [Gilmore and Hemmingway, 1995]:

$$\text{MDA (E)} = \frac{\sqrt{B_c E_R}}{\varepsilon(E)} \quad (7)$$

Where:

B_c is the background counts

E_R is the energy resolution of the detector and

$\varepsilon(E)$ is the efficiency of the detector

The MDA changes with energy because the quantities on which it relies on vary with energy. The formula above (equation 7) is also notable in guiding the analyst towards the right choice of the detector.

Equation 7 above shows that MDA is inversely proportional to the efficiency and directly proportional to the square root of the product of background counts and the energy

resolution. Therefore it is expected that the biggest detector will give a desirable MDA for samples containing minimal activity concentration [www.ortec-online.com].

The calibrations are also finished by: the time stability check for a reference peak position, net counting rate and FWHM, and the determination of a standard source of the same type (radionuclide, matrix and volume), using the calibration data and checking the values obtained with the true values. The two figures obtained should agree within the limit of their expanded uncertainty [Luca et al, 2012].

2.11.5 Calculation of uncertainty

Usually the quantity of interest called the measurand is not the one that can be absolutely analyzed and its value is derived from values of several other quantities. The activity concentration (or specific activity) of radioisotopes (analytes) in environmental samples is a function of various quantities like gamma ray emission probability, detector efficiency, counting rate, corrections factors etc and each of these quantities have an associated uncertainty [Bambynek, 1987].

The combined standard uncertainty of the measurand, y according to IAEA-TD-1401 can be derived by means of the error propagation law of Gaus. In this manner the combined standard uncertainty of y , $u_c(y)$ is computed according to terms of component uncertainties $u(x_i)$ as follows: [IAEA, 2004]

$$u_c(y(x_1, \dots, x_n)) = \sqrt{\sum_{i=1}^n \left(\frac{\partial y}{\partial x_i} \right)^2 (u(x_i))^2} \quad (8)$$

Where $y(x_1, x_2, \dots, x_n)$ is a function of several quantities x_1, x_2, \dots, x_n

Equation 8 above is true if and only if the quantities x_i are independent, and if $u(x_i) \ll x_i$. However in most cases this equation has been applicable even when the above mentioned restrictions are not encountered.

If the variables x_1, x_2, \dots, x_n are not independent, the relationship is more rigorous and the following expression is found with assumption that the derivative of function y can be calculated:

$$u_c(y(x_1, \dots, x_n)) = \sqrt{\sum_{i=1}^n \left(\frac{\partial y}{\partial x_i}\right)^2 (u(x_i))^2 + 2 \sum_{i=1}^n \sum_{j>i}^n \left(\frac{\partial y}{\partial x_i}\right) \left(\frac{\partial y}{\partial x_j}\right) \text{cov}(x_i, x_j)} \quad (9)$$

Where $\text{cov}(x_i, x_j)$ is the covariance between x_i and x_j .

The expanded uncertainty u is found by finding the product of the combined standard uncertainty by a suitable coverage factor k_a . The unidentified value of the measurand (the activity concentration) is thought to lie within $y \pm u$ with a confidence level of approximately a.

From equation 6, specific activity concentration (A) of measurand in the sample is given by:

$$A = \frac{N}{\varepsilon \gamma t_s M K} \quad (10)$$

Then relative combined standard uncertainty $\frac{u_c(A)}{A}$ of the equation above will be:

$$\frac{u_c(A)}{A} = \sqrt{\frac{u(M)}{M}^2 + \frac{u(N)}{N}^2 + \frac{u(\gamma)}{\gamma}^2 + \frac{u(\varepsilon)}{\varepsilon}^2 + \frac{u(K)}{K}^2} \quad (11)$$

2.11.6 Dosimetric quantities and Unit of Measurement

(a) Becquerel, Gray, Sievert, Dose and Working Level Month

Radiation is measured in units called Becquerel (Bq) and one Bq is termed as the activity of a quantity of radioactive material in which one nucleus decays per second (s^{-1}). For the sake of radiation assessment purposes, UNSCEAR (1988) came up with the following quantities and unit of measurement:

- Absorbed dose whose unit is joule per kilogram called the Gray (Gy), is a more sound assessment of the extent of radiation hazard to the body. It is defined as the energy absorbed per unit mass of the body.
- Equivalent dose which has unit of Sievert (Sv) , consists of the absorbed dose and the extent to which it can cause biological damage (termed quality factor)
- Effective dose equivalent (EDE) is also in joule per kilogram, but unlike the absorbed dose it is given the name Sievert (Sv) just like with equivalent dose. EDE gives the weighted aggregate of the doses of various tissues in the body

(b) Amount of Radon in the soil

According to (UNSCEAR, 2000; Nazaroff et al, 1988), the amount of radon, CRn, in soil gas, in the absence of radon transport can be calculated from:

$$C_{Rn,Tn} = C_{Ra-226,228} \cdot f \cdot \rho_s \cdot \varepsilon^{-1} (1 - \varepsilon)(m[K_T - 1] + 1)^{-1} \quad (12)$$

Where:

$C_{Rn-222,220}$: is the radon, thoron concentration in soil (Bq/m³)

$C_{Ra-226,228}$: activity concn.in dry mass of ²²⁶Ra, ²²⁸Ra in soil (Bq/kg),

f: soil emanation factor: radon (0.2) and thoron (0.1),

ρ_s : is the density of soil, (kgm⁻³),

ε : is the porosity, (0.25),

m : is the porosity fraction that is water filled and is zero if the soil is dry

k_T : is the radon partition coefficient between water and air phases and if the soil samples are dried before measurement, then m is zero, thus the last term of the equation 12 above is omitted.

(c) Radioactive decay equilibrium

When a radioisotope disintegrates, it does not disappear but modifies itself into a new type of higher binding energy. It can mathematically further be shown that if any radionuclide one (X_1) decays with decay constant (λ_1), forming a daughter nucleus two (X_2), which then decays with decay constant (λ_2), schematically this can be presented as:

$$X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} \quad (13)$$

Then net rate of formation of the daughter atoms X_2 is the difference between rate of generation of X_2 and her rate of disintegration given by:

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \quad (14)$$

Where N_1 and N_2 are the numbers of atoms X_1 and X_2 present at time t . The equation 6 above shows that the number of offspring or daughter nuclei increases following the decay of the parent and reduces as a result of its own decay [Loveland and Morrissey, 2006].

Equation 6 can be rearranged while collecting similar terms and becomes:

$$dN_2 + \lambda_2 N_2 dt = \lambda_1 N_2 dt \quad (15)$$

We know that

$$N_1 = N_1^0 e^{-\lambda_1 t} \quad (16)$$

We then have:

$$dN_2 + \lambda_2 N_2 dt = \lambda_1 N_1^0 e^{-\lambda_1 t} dt \quad (17)$$

Eqn.8 is a first order differential and solving by method of integrating factors we get:

$$N_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t} \quad (18)$$

Remembering that activity (A_2) is the product of decay constant (λ_2) and the number of atoms (N_2) then activity A_2 will be given as:

$$A_2 = \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + A_2^0 e^{-\lambda_2 t} \quad (19)$$

At secular equilibrium where

$\lambda_1 \ll \lambda_2$ we have :

$$\lambda_2 N_2 = \lambda_1 N_1^0 (1 - e^{-\lambda_2 t}) \quad (20)$$

Where as t becomes large nuclei X_2 decay at the same rate at which they are formed:

$$\lambda_2 N_2 = \lambda_1 N_1^0.$$

This result can also be immediately obtained from equation 14 above, taking:

$$\frac{dN_2}{dt} = 0$$

For practical reasons, equilibrium may be thought of being achieved after seven daughter half-lives (more than 99% of daughter nuclides grow-up) as shown in fig 2-10

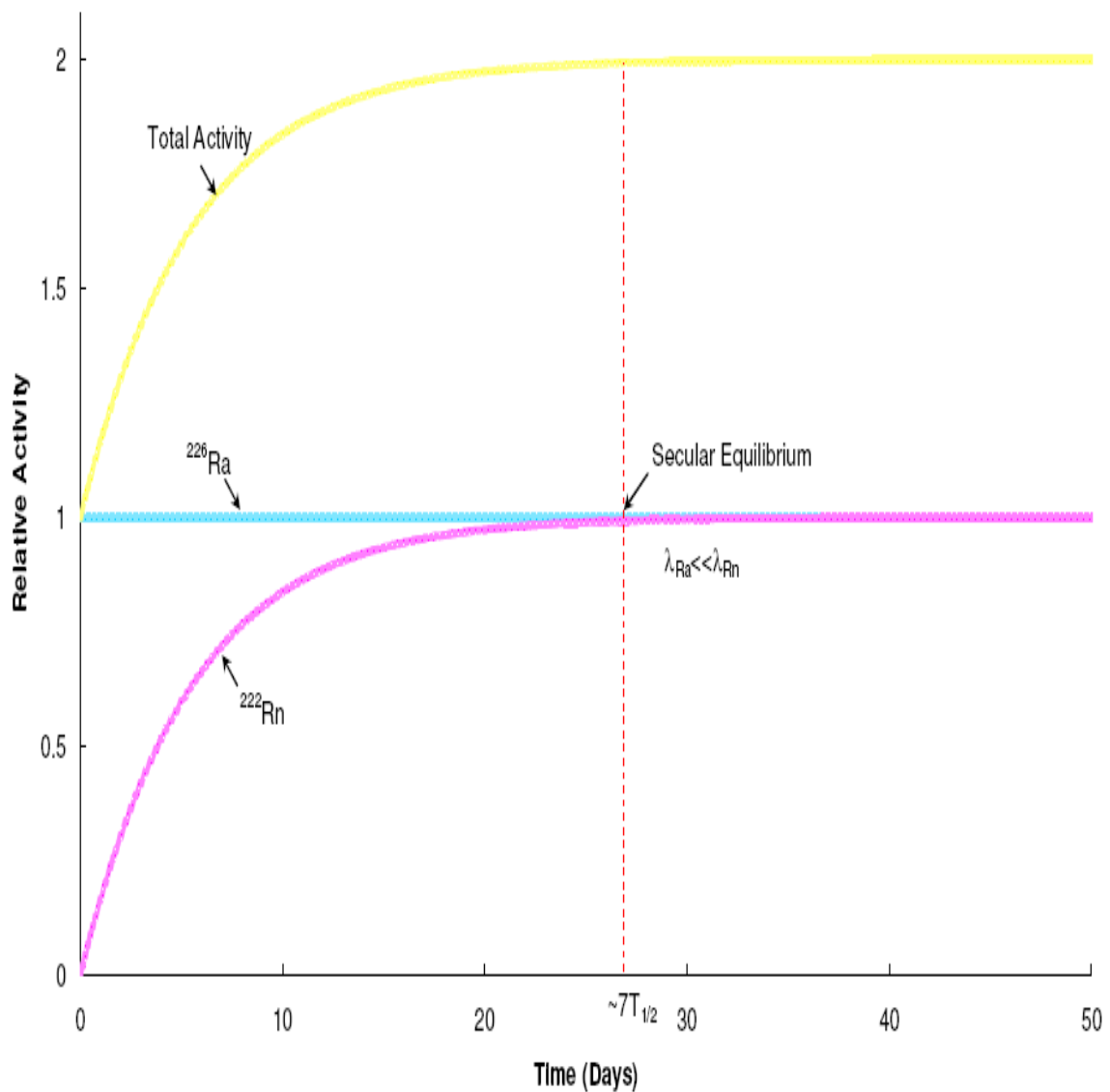


Figure 2- 10: Growth of a short lived daughter (^{222}Rn) from a much longer lived parent (^{226}Ra) until reaching Secular Equilibrium [Todsadol 2012; Xhixha, 2012]

(d) Amount of Radon in water

The activity concentrations of ^{222}Rn and ^{220}Rn in the water samples can be calculated using equations 21 and 22.

Since the daughters Rn-222, Rn-220 activities A_D, A_P will be in secular equilibrium with their parents Ra-226, Ra-228 respectively then according to equation 20 above,

knowing that activity of daughter, $A_D = \lambda_2 N_2$ and that of parent,

$A_P = \lambda_1 N_1^0 (1 - e^{-\lambda_2 t})$ and that $A_D = A_P$ at secular equilibrium then :

$$A_{\text{Rn-222}} = A_{\text{Ra-226}} [1 - e^{-(\lambda_{\text{Rn-222}})T_d}] \quad (21)$$

$$A_{\text{Rn-220}} = A_{\text{Ra-228}} [1 - e^{-(\lambda_{\text{Rn-220}})T_d}] \quad (22)$$

Where $A_{\text{Ra-226}}, A_{\text{Ra-228}}$ are activity concentration of parents $^{226}\text{Ra}, ^{228}\text{Ra}$ respectively

$\lambda_{\text{Rn-222}}, \lambda_{\text{Rn-220}}$ = decay constant of daughters radon, thoron respectively

T_d is the time between sampling the water and time of counting

CHAPTER THREE

METHODOLOGY

This section outlines the background information about Ghana, the study area, Newmont -Akyem Gold mine, the geology, sampling and sample preparation techniques employed and analysis methods used. A detailed description of the calculation of activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K and radiological hazard assessment are also given.

3.1 Background information about Ghana

3.1.1 Geographical Location, climate, demography and the Economy of Ghana

Ghana, located in West Africa, covers about 240,000 square kilometres (as big as United Kingdom). Ghana is one of the five African countries along the northern coastline of the Gulf of Guinea, and has Cote d'Ivoire to the west, Burkina Faso to the north and Togo to the east. The country comprises mostly low savannah regions with a hilly central belt of forest. It lies instantly north of the equator. Ghana's climate can be categorized as tropical and its seasons can be split into two different types: the cold or the wet season and the hot or the dry season. The population of Ghana in 2014 Census was estimated at about 26,700,000, an increase from 2013's estimate of 26,000,000. Geographically, Ghana is the country which is very close to the center of our planet Earth.

(<http://worldpopulationreview.com/countries/ghana-population>)(2014/12/15)

Ghana economy depends largely on agriculture and mining. Despite economic turmoil, Ghana is recognized as one of the most developed countries in Sub-Saharan Africa.

The domestic economy of Ghana relies on subsistence farming, which constitutes for about 37% of the gross domestic product ("GDP"). Over 60% of Ghanaians grow food

crops (plantain, cassava, maize, yams, rice, groundnuts, etc) and are for local consumption.

The remarkable cash crop in Ghana is cocoa and other cash crops include palm oil, rubber, coffee and coconuts. Cattle farming is done in northern Ghana. The most important source of foreign earning is gold mining, followed by cocoa and timber products. Manganese, bauxite and diamonds are also mined. Gold represents Ghana's major export commodity which puts Ghana as the world's tenth and Africa's second largest producer of gold.

According to an edition in Cable Network News (CNN) Marketplace Africa (2014) on the continent's key industries and corporations, Ghana was ranked number seven while Botswana took position one followed by Republic of South Africa while in terms of the safest nations in Africa, Ghana was ranked second. Tourism is also growing rapidly in Ghana (<http://edition.cnn.com/business/africa-prosperity-rankings-2014>) (2014/12/18).

3.1.2 Gold Mining in Ghana

Ghana has been in gold trading since the middle ages and now it is a well-focused business and recently gold mining overtook cocoa as Ghana's leading export activity. It has been reported that on average over 300 metric tonnes of gold are produced every year giving over 13 million ounces (oz) [Goldfields, 2007]. Current mining operations are situated in the south-western part of Ghana. Most of the mines in Ghana are open cut/cast mines unlike the underground ones [ICMM, 2007]. Faanu (2011) reported that in Ghana there were a total of 212 mining companies as of 2008. Literature has shown that mining of gold and other metals elevate NORMs whose dangers have been tackled in the background information of this thesis work.

3.1.3 Descriptions of the study area-Akyem Gold Mine

The Newmont Ghana's -Akyem Gold Mine (AGM) (Fig 3-1) is situated roughly three kilometers west of the district capital New Abirem, 133 km west of Koforidua, the regional capital and 180 km northwest of Accra, the national capital [Akyem Annex F, 2008]. Akyem in the Eastern region of Ghana is found amongst the following communities: Afosu, New Abirem, Old Abirem, Mamanso, Yayaaso, Adausena, Adjenua, Hweakwae, Ntronang and Yaw Tano.

The Eastern region of Ghana covers a land area of 19,323 kilometres and constitutes 8.1% of the total land area of Ghana. It is the sixth largest region in terms of land area and lies between latitudes 6° and 7° North between longitudes $1^{\circ} 30'$ West and $0^{\circ} 30'$ East. The region shares common boundaries with the Great Accra, Central, Ashanti, Brong Ahafo and Volta Regions. The region contains minerals such as gold, diamond, bauxite-tantalite, limestone, kaolin and clay. Gold and diamond are however the only minerals that are being mined commercially.

The Akyem Project in Ghana is managed by Newmont Mining Corporation (NMC) through its subsidiary Newmont Ghana Gold Limited and operating company Golden Ridge Resources Limited (Ltd). NMC is one of the world's leading producer of gold and copper. NMC was founded in 1921 and publicly started trading since 1925. The Company has approximately 30,000 employees and contractors worldwide and Ghana alone has about 2000 employees and contractors working at Akyem Gold mine.

[www.newmont.com/Africa/Akyem-Ghana] (2014/10/07).

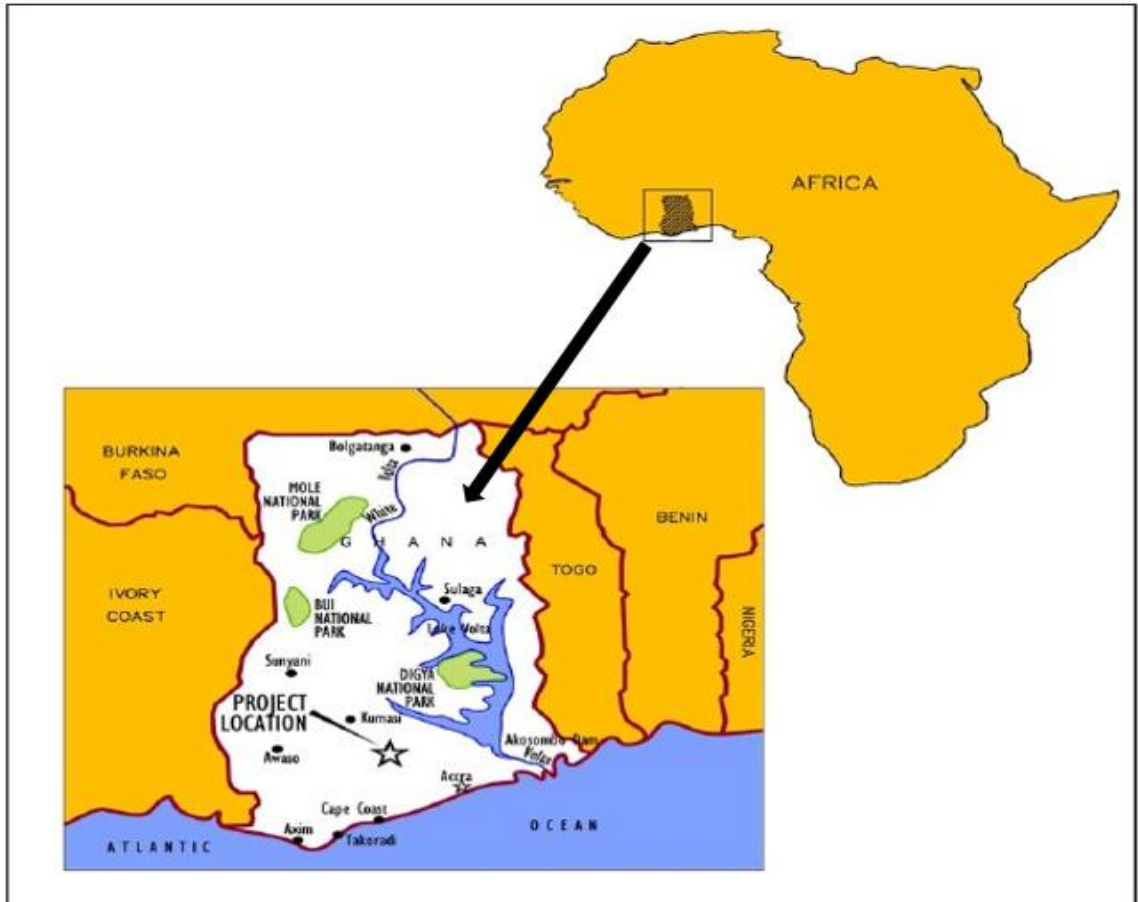


Figure 3- 1: Location map Akyem Project [AGM-Annex F, 2008]

3.1.4 Climate of the study area

According to Akyem EIS (2008) on the existing environment, the Study Area is situated within the wet semi-equatorial climatic zone of Ghana which is characterized by an annual maxima rainfall pattern; wet seasons normally occur from March to July and September to mid-November. The climate type is determined by movement of the Inter-Tropical Convergence Zone, which oscillates yearly about the equator attracting air masses from the north and south. Southern air masses, originating from the anticyclone of St. Helena in the South Atlantic, come up with relatively cool, moist weather. Northern

air masses, locally called "Harmattan," originate from the sub-tropical Azores anticyclone and its extension over the Sahara desert. These air masses come with hot, dry weather from December through February.

Mean monthly temperatures in the Study Area, are relatively constant, changing from about 25 to 27 degrees Celsius (°C) [Akyem EIA, 2008].

3.1.5 The Geology

According to Coffey Mining Pty Ltd, (2011), the geology of Ghana comprises primarily of rocks of the Birimian and to a lesser extent of units of the Tarkwaian.

The Birimian consists of volcanic belts, which can be tracked for hundreds of kilometers along strike but are usually only 20 to 60 km wide, detached by wider basins of mainly marine clastic sediments. These structures are very important in the development of gold deposits.

The Tarkwaian rocks consist of a characteristic sequence of metasediments existing within a large band along the interior of the Ashanti Belt. They host important gold deposits in the Tarkwa district. Equivalent rock types appear in other belts of the region but in relatively restricted areas.

The Akyem Mining Area is situated at the northern end and along the southeast margin of the Ashanti Belt. The Ashanti Belt has produced approximately 40 million ounces (oz) of gold since mining started.

The east-northeast striking Akyem deposit is embedded in Birimian rocks on the southern limb of a main regional fold structure called the Noyem Syncline. The basement of the

minerals corresponds to a near-planar thrust zone dipping approximately 60 degrees to the south-southeast.

The geologic section at the Akyem deposit is outlined graphically as shown in Figure 3-2 and depicts rock units, the shear or thrust fault zone, ore zone, and proposed Akyem pit outline. Main geologic units in the hanging wall of the thrust fault include greywacke, a quartz-epiclastic unit, graphitic shear breccia and graphitic rubble. Mafic metavolcanics consist of the foot wall of the shear zone (Figure 3-2).

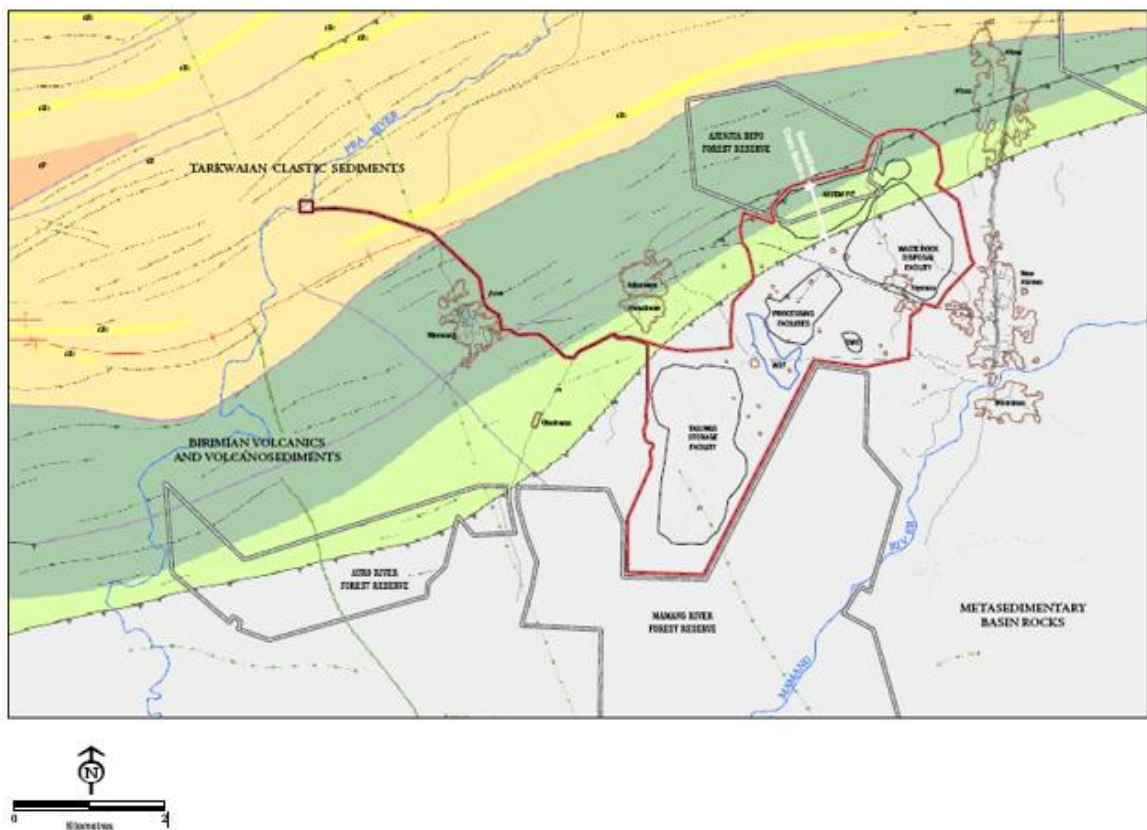











Figure 3- 2: Geology of Akyem Project [AGM-EIS, 2008]

Legend

| Feature | Description | Feature | Description |
|---|--------------------------------------|---|--|
|  | Tarkwa Phyllite |  | Banket Series: quartz pebble conglomerate, auriferous |
|  | Tarkwa Phyllite |  | Proposed Mining Area |
|  | Basalt, dolerite: Amphibolite; |  | Forest Reserve |
|  | Volcaniclastics and metasediments |  | Villages |
|  | Metasedimentary rocks | | |

3.1.6 Soil types

Soil in the suggested Mining Area was generated from two principal geologic units and their associated alluvial deposits. Soil formed on Lower Birimian rocks have the largest distribution, while the granitic unit is far less prevalent and happen only in the southern-most areas within the Proposed Mining Area. Soil series present on similar topographic positions in the two units express similar drainage and development with comparable series found in sequence from uplands to lowlands in each geologic unit.

3.1.7 Surface water

The Study Area is situated in the Pra River basin which drains its water south to the Gulf of Guinea (Atlantic Ocean). It is located from the Longitude 6°16'40''N to 6°22'30''N and from Latitude 0°59'10''W to 1°6'40''W. The Pra River is the second biggest river in Ghana with a total drainage area of approximately 23,170 square kilometres. All of the proposed mine facilities (with the exception of the Pra River pumping station and a portion of the water pipeline) are situated in the Mamang River drainage, which meets the Pra River close to the community of Kotokuom southwest of the Study Area. Locations to the north and west of the proposed mine component drain directly to the Pra River.

Selected surface water monitoring stations identified for the Study Area are listed in Annex 1 and are shown in Figure 3-3.

The Mamang River watershed covers an area of approximately 475 square kilometres. Mean annual discharge calculated for the Mamang River at its mouth is approximately 2.9 m³/sec. Streams supplying the Study Area that are tributaries to the Pra and Mamang Rivers (including those originating from the Ajenjua Bepo Forest Reserve) are ephemeral, flowing only during the wet seasons. The main tributaries in the area include the Afosu, Adentosu, Akwasi, and Adenkyerensu.

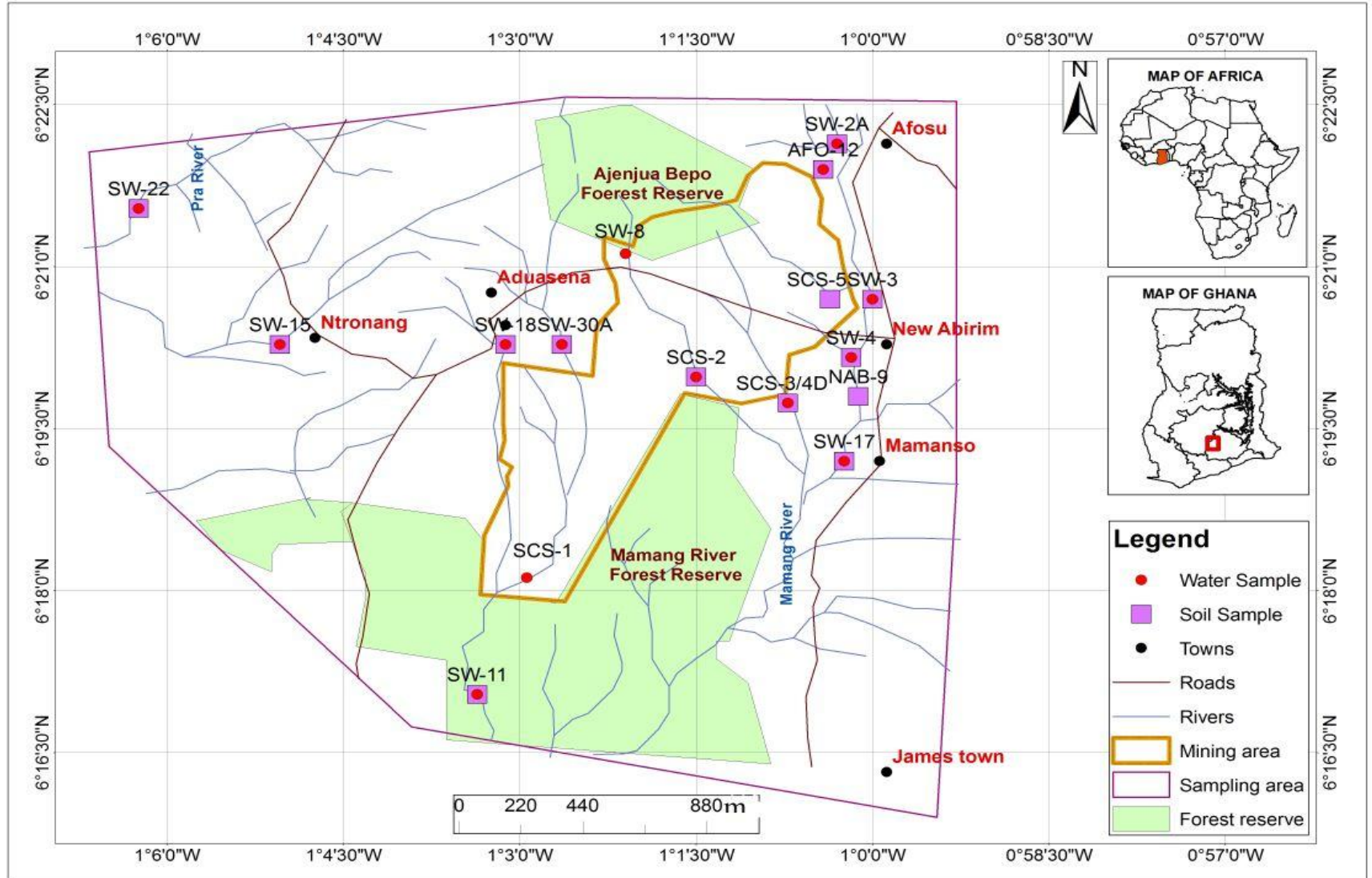


Figure 3- 3: Layout of Akyem Gold Mine showing water and soil sampling points

3.1.8 Population and Demography

According to the National Census (2000) there was a recording of population from 8 settlements within the Study Area that ranged from 570 people in Yayaaso to over 3,500 inhabitants in Afosu. In addition to settlements in the Study Area, there are multiple hamlets and individual farmsteads inhabited by farmers from other regions of the country. The population of the Study Area is projected to have grown from 15,433 people in 2000 to 19,418 persons (excluding hamlets and farmsteads) in 2008 (Table 3-1).

The Health Assessment conducted noted that the main cause of morbidity of people in the Mining Area is malaria, contributing up to 82 percent of illnesses . Other diseases are diarrhea; as well as skin, eye, ear and throat ailments and this may explain lower populations for Afosu , Ahausena in 2008 than they were in 2000 respectively

Table 3- 1: An estimate of population in Akyem Gold Mining Project Study Area

| Community | Population (2000) | Population (2008) |
|-------------------------|--------------------------|--------------------------|
| New Abirem | 1967 | 3990 |
| Afosu | 3511 | 5325 |
| Mamanso | 2000 | 2663 |
| Yayaaso | 570 | 700 |
| Ahausena | 1462 | 1429 |
| Hweakwae | 1249 | 1249 |
| Ntronang | 2732 | 2756 |
| Old Abirem | 1411 | 1306 |
| Hamlets/farmsteads | 531 | No projection |
| Total Study Area | 15,433 | 19,418 |

3.1.9 Open-pit mining and ore processing at Akyem

The Akyem open-pit mine comprises two lobes engulfing a total area of 139 hectares.

The open-pit mining operation includes drilling and blasting techniques for the extraction of the ore which is loaded into haul trucks by means of shovels and backhoes. The waste rock is disposed in the open-pit while the important ore is taken to the processing plant, built over an area of 85 ha within the mine site (<http://www.mining-technology.com/projects/akyem-gold-mine>) (2014/10/09).

3.2 Sampling, sample preparation and measurement

The method of the measurement of the activity concentration of NORMs particularly radon and thoron in environmental samples by gamma spectrometry was done according to Fig 3-4 modified from Bakr et al (2011). The figure also depicts measurement of absorbed gamma dose by the survey meter and water physico-chemical properties such as: pH, Temperature, Total Dissolved Solids (TDS) and Conductivity using portable water testing kit.

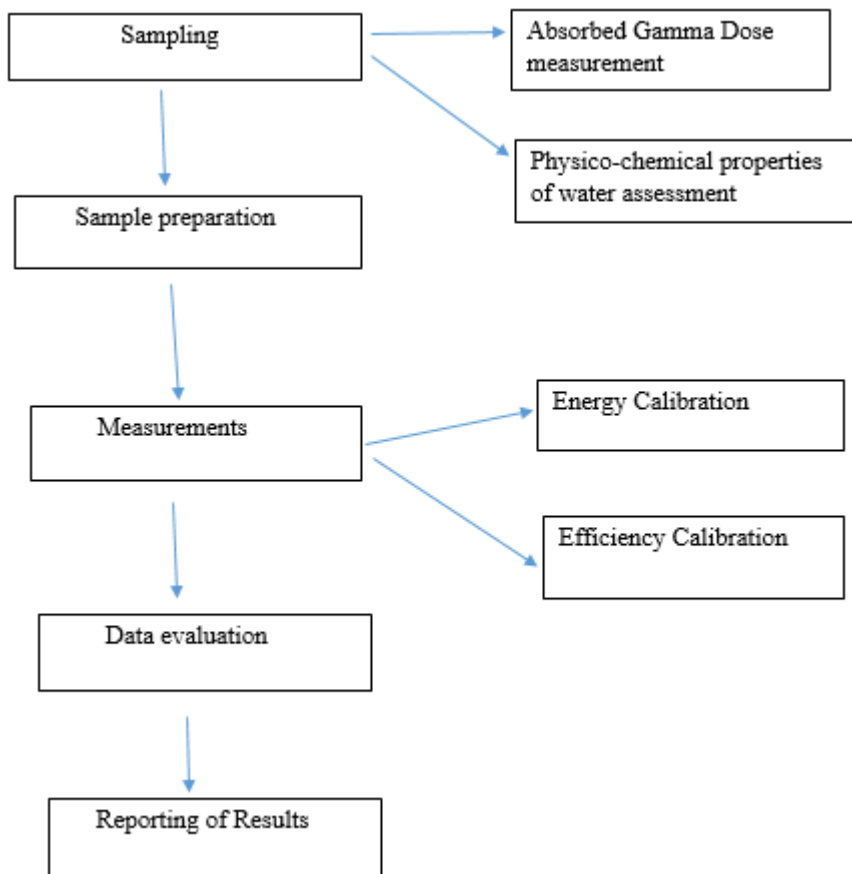


Figure 3- 4 : Analytical steps for determination of radioactivity contents in soil or water samples

3.2.1 Sampling

Activity determination of gamma-ray-emitting radionuclides in environmental samples is a difficult task, mainly, due to the low level of activity concentration. The main problem associated with low-level activity measurements is inadequate counting statistics. This is prevented by increasing the volume or mass of the sample collected [Vukanac et al, 2008].

Sampling was done based on significant exposure pathways such as direct gamma exposure, radon and thoron inhalation and ,ingestion of radon and thoron through contaminated water source.

A total of 28 soil and water samples were collected from designated sampling locations in Akyem Project study area (Fig. 3.3 and Annex 1).

3.2.2 Water and soil samples

The water samples were collected from identified locations that were chosen to characterize water quality in the region and to focus on locations where impacts could occur from the suggested Akyem Gold Mining Project (Annex 1 and Fig 3-3). In order to characterize the quality of the water, the following parameters were measured; pH, conductivity, temperature and total dissolved solids (TDS). The water samples were collected into two and half litres (2.5L) plastic bottles and acidified on site with 1M HNO₃ to avoid radionuclides from sticking to the sides of the container. For activity concentration determination, the water samples were taken to the laboratory and transferred into 1 litre Marinelli beakers. The water samples were not filtered before preparation and measurement. The beakers were thick enough to avoid the permeation of radon and thoron. The beakers were closed using screw caps and plastic tape wrapped over the caps and then stored for 4 weeks before measurement. This step was essential to make sure that radon and thoron gases do not escape from the sample matrix.

3.2.3 Soil samples

Soil samples were collected at the sampling points where the water samples were also taken (Fig.3-3 and Annex 1). The soil samples were carried to the laboratory where plant roots and other unwanted materials were identified and removed. The samples were then dried in an oven at 105 °C for 12-24 h, ground, and sieved through a 2-mm sieve for homogeneity of the soil sample. About 1000g of dry sample was weighed into a plastic container, which was capped and sealed by means of masking tape. The container was

sealed to ensure that none of the gaseous daughter products of uranium and thorium generated, particularly radon and thoron, could escape. The prepared samples were then stored for one month before counting to ensure that equilibrium had been established between radium and its short-lived daughters. Detailed gamma-ray spectrometry analysis was performed on the soil samples.

3.3 Measurement of samples by Gamma Spectrometry

The purpose of gamma spectrometric measurement for environmental samples is to determine the activity of gamma emitting radionuclides.

In gamma-ray spectrometry with germanium detectors, the pulse height scale is calibrated with respect to absolute gamma-ray energy in order for various peaks in the spectrum to be properly identified. In various normal applications, the gamma rays anticipated to appear in the spectrum are well known in advance and the corresponding peaks are readily identified by inspection. However in other applications, where unknown gamma-ray spectra may be encountered which do not provide clear calibration of the energy scale; a separate calibration gamma-ray source is ordinarily used to supply peaks of known energy in the spectrum [Knoll, 2000].

To precisely quantify activity concentrations, correct and reliable determination of the efficiency curve is very essential. Detection efficiency is a rigorous function of the energy, spectrometer characteristics, measurements geometry, volume and density of the sample, etc. In order to calibrate the spectrometer in a proper manner, calibration standards are prepared with matrices with identical chemical composition and density, similar concentration for the pertinent radioisotopes, and in the same geometry and counting configuration as the genuine samples [IAEA, 1989].

3.3.1 Calibration of gamma spectrometry system

Prior to the measurements, the detector and measuring assembly were calibrated for energy and efficiency to ensure both qualitative and quantitative analysis of the samples were conducted. The energy and efficiency calibrations were executed using mixed radionuclide calibration standard uniformly spread in solid water, certificate number 9031-OL-146/14 with approximate mass 1000g, volume 1000 mL and density 1.0 g cm^{-3} in a 1.0 L Marinelli beaker. The standard was procured from Czech Metrology Institute Inspectorate for ionizing radiation and it contained radionuclides with known energies, initial activity (Bq) and combined standard uncertainty (Annex 2).

The calibration standard source had physical dimensions, chemical composition, and density similar to the samples that were analyzed, so that the deviation in the measured activity was minimized. With regard to the geometry, the deviation was reduced to almost zero by ensuring that the bottle had the same dimensions and that all the heights were similar for the standard source and the samples [Harb et al, 2008].

(a) Energy calibration

The aim of energy calibration according to IAEA-Tecdoc-1401 is to derive a relationship between a peak position in the spectrum and the corresponding gamma-ray energy. Most of the time, it is done before measuring the sample [IAEA, 2004].

The energy calibration (channel number of the Multi-Channel Analyzer against the Gamma-ray energy) of the detector system was effected at a fixed Gain using standards with known radionuclides. The standards were sealed in a container and expected to give

out various γ -ray energies covering the range of interest, usually 50 keV to 2000 keV in order to test for system linearity [HASL-300, 1997].

Upon establishing energy calibration points over the whole desired energy range, a calibration curve connecting energy to channel number was plotted. Common techniques involve the least-square fitting of a polynomial of the form:

$$E_i = \sum_{n=0}^N a_n C_i^n \quad (23)$$

where E_i is the energy referring to the channel number C_i . A Polynomial of order $N = 4$ or 5 usually is sufficient for typical germanium spectrometers, depending on the severity of nonlinearity that is available [Knoll, 2000].

(b) Efficiency calibration

Efficiency calibration as a function of energy provides the efficiency value at any energy in a given range. The broader the range in energy, the bigger the number of radionuclides whose concentration can be measured [Harb et al, 2008].

The same standard that was used in energy calibration was also used in efficiency calibration.

3.3.2 Sample measurement

After the spectrometer system had been arranged, the energy and efficiency calibrations were carried out, then the samples were counted and spectrometry analysed.

Following the general principles of quantitative analytical chemistry, each sample was moved to the specimen container and put in the same way as was done in the course of system calibration. Each sample was counted for 36,000 seconds to obtain a γ -ray spectrum which would meet the minimum admissible counting uncertainty [HASL-300, 1997].

3.3.3 Data Evaluation

(a) Determination of activity concentration of NORMs in soil and water

The activity concentration of ^{226}Ra , ^{228}Ra , ^{232}Th and ^{40}K were calculated using the following equation [Darko et al, 2010]

$$\text{Then, } A_{\text{sp}} = \frac{N_{\text{sam}} \cdot \exp(-\lambda T_d)}{P_E \cdot T_c \cdot \epsilon \cdot M_{\text{sam}}} \quad (24)$$

Where:

N_{sam} - net counts of the radionuclide in the sample

P_E - gamma ray emission probability (gamma yield)

ϵ - total counting efficiency of the detector system

T_d - Delay time between sampling and counting

$\exp(-\lambda T_d)$ - Correction factor between sampling and counting

T_c - sample counting time

M_{sam} - mass of sample (kg) or volume (L)

(b) Soil and Water Radon

i Radon in the soil

The activity concentrations of ^{222}Rn and ^{220}Rn in the soil samples were calculated using equation 12 explained in the Literature Review of this thesis work given by:

$$C_{Rn,Tn} = C_{Ra-226,228} \cdot f \cdot \rho_s \cdot \varepsilon^{-1} (1 - \varepsilon)(m[K_T - 1] + 1)^{-1} \quad (12)$$

N/B: measured Akyem-soil density (ρ_s) of 1600 kg/m^3 was used.

ii Radon in the water

The activity concentrations of ^{222}Rn and ^{220}Rn in the water samples were calculated using equations 21 and 22 respectively, clarified in the Literature Review under dosimetric quantities of this research work given by:

$$A_{Rn-222} = A_{Ra-226} [1 - e^{-(\lambda_{Rn-222})T_d}] \quad (21)$$

$$A_{Rn-220} = A_{Ra-228} [1 - e^{-(\lambda_{Rn-220})T_d}] \quad (22)$$

(c) Assessment of Radiological hazards

This was achieved by calculating the Radium equivalent activity, Internal hazard index, Absorbed dose and Annual Effective Doses.

3.4 Radium equivalent activity, Internal hazard index and Absorbed dose

i Radium equivalent activity

The radium equivalent activity (Ra_{eq}) is a weighted addition of activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K in which the sum of their weighed proportion produces the same gamma-ray dose rates as given by the following formula [Nada , 2004]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (25)$$

ii Internal Hazard Index (H_{in})

Another factor signifying radiological hazard due to radon and its progeny being an internal radiation exposure hazard is the Internal Hazard Index as shown in equation (25) [Saher et al, 2013]:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810 \quad (26)$$

Where: C_{Ra} , C_{Th} and C_K represents activity concentration (in Bq/Kg) of ^{226}Ra , ^{232}Th and ^{40}K respectively. In order to keep the radiation negligible, the value of Ra_{eq} must be less than 370 while H_{in} must be less than unity (1).

iii Absorbed Dose

Two methods were used to assess the external doses that result from deposition of Radioisotopes in soil surfaces: direct determination and calculations according to radionuclide deposition densities with reference to UNSCEAR, (2000).

(1) Absorbed Dose Rate measurement by Survey meter

External gamma dose rate was measured and estimated at each location 1m above ground surface using a calibrated Rados Universal Survey Meter, model RDS-200 manufactured by Rados Technology of Finland. The survey meter had a sensitivity of $0.01\text{-}10 \mu\text{Sv h}^{-1}$ in the energy range up to 1.3 MeV. It was calibrated at the Secondary Standard Dosimetry Laboratory (SSDL) of the Radiation Protection Institute (RPI) of Ghana Atomic Energy

Commission (GAEC) with a calibration factor given. An aggregate of five measurements (in $\mu\text{Gy/h}$) were made at 1 meter above the ground at each location.

(2) Absorbed Dose Rate calculation

Calculation of absorbed dose rate at each sampling location was based on NORMs distribution densities according to [Mohanty et al, 2004; Saher et al, 2013]

$$D_R = [0.621C_{\text{Th}} + 0.462C_{\text{Ra}} + 0.0417C_{\text{K}}] \text{ nGyh}^{-1} \quad (27)$$

where: D_R :Dose rate

3.5 Annual Effective Doses

i Estimation of Annual Effective Dose due to terrestrial radiation

Effective dose is meant for use as a safeguard quantity. The main uses of effective doses are the proposed dose assessment for planning and optimization in radiological protection, and demonstration of compliance with dose limits for regulatory objectives [ICRP, 2007].

To evaluate the year-long effective dose rates, the conversion coefficient from absorbed dose in the air to effective dose (0.7Sv.Gy^{-1}) and outdoor occupancy factor (0.2Sv.Gy^{-1}) suggested by UNSCEAR, (2008) was applied.

According to (UNSCEAR, 2008) world average indoor and outdoor occupancy factors are 0.8 and 0.2 respectively. However in Ghana, the average of time spent indoor and outdoor (occupancy Factors) are 0.6 and 0.4 respectively [Asumadu-Sakyi et al, 2012].

Therefore, the effective dose rate in units of mSv/y was estimated using the formula according to UNSCEAR, (1998) but used occupancy factors for Ghana such that:

Indoor Effective Dose rate (mSv y⁻¹)

$$= \text{Dose rate (nGyh}^{-1} \times 8760\text{h)} \times 0.4 \times 0.7 \text{ SvGy}^{-1} \times 10^{-6} \quad (28)$$

Outdoor Effective Dose rate (mSv y⁻¹)

$$= \text{Dose rate (nGyh}^{-1} \times 8760\text{h)} \times 0.6 \times 0.7 \text{ SvGy}^{-1} \times 10^{-6} \quad (29)$$

ii Estimation of Annual Effective Dose due to ingestion of Rn-222 in water

The estimated annual effective dose by ingestion was calculated because of habitual consumption of water by dwellers. It was computed using the formula below [Asumadu-Sakyi et al, 2012]:

$$AED_{ing} = C_{Rn-222} \times AW_{ing} \times DCF_{ing} \quad (30)$$

Where:

AED_{ing} is the Annual Effective Dose due to ingestion (mSv/y)

C_{Rn} is the concentration of ²²²Rn in the ingested drinking water (Bq / m³)

AW_{ing} is the annual intake of drinking water (L/y) ~ 730 L/y and

DCF_{ing} is the ingested dose conversion factor for ²²²Rn of $3.5 \times 10^{-9} \text{ Sv h}^{-1} (\text{Bqm}^{-3})^{-1}$

was used as suggested by UNSCEAR (2008).

Annual effective dose due to intake of ²²²Rn from drinking water is calculated considering that an adult (Age > 18) consumes an average of 730 L water annually.

iii Estimation of AED due to ingestion of Ra-226, Th-232 and K-40 in water

In order to control radiation exposure to the people, evaluation of total annual effective dose due to ingestion of ^{226}Ra , ^{40}K and ^{232}Th in water samples was done based on the activity concentration and dose conversion factors of chosen radionuclides using the following formulation [Darko et al,2013]

$$H_{ing}(W) = \sum_{j=1}^3 DCF_{ing} (^{226}\text{Ra}, ^{40}\text{K}, ^{232}\text{Th}) * A_{sp} * I_w \quad (31)$$

where, DCF_{ing} -dose conversion factors by ingestion of selected radioisotopes in water samples by adult members of the public staying in the study areas were taken to be 2.8×10^{-7} Sv/Bq for ^{226}Ra , 6.2×10^{-9} Sv/Bq for ^{40}K and 2.3×10^{-7} Sv/Bq for ^{232}Th . A_{sp} is activity concentration of each picked radionuclide in water samples and I_w is daily water taken to be 2L/day.

iv Estimation of Annual Effective Dose due to inhalation of Rn-222 from water

According to Crawford and Cothorn (1987) concentration of radon in the indoor air originating from the water only can be calculated based on knowing the ratio of the concentration of Rn-222 in the indoor air ($C_{a_{Rn-222}}$) to the concentration of Rn-222 in the water ($C_{w_{Rn-222}}$).

Most literature have established this ratio either by theoretical computation or by direct measurement. UNSCEAR 1977, 1982 and NCRP, 1984 gave calculated values of $(C_{a_{Rn-222}}) / (C_{w_{Rn-222}}) = R$ ranging from 1×10^{-5} to 20×10^{-4} with the mean value of 1×10^{-4} .

According to UNSCEAR (2000) the AED due to inhalation of Rn-222 from water is calculated by:

$$AED_{inh_{Rn-222}} \text{ (mSv/y)} = Ca_{Rn-222} \times R \times DCF_{Rn-222} \times F_{Rn-222} \times P_{Rn-222} \quad (32)$$

Where :

Ca_{Rn-222} = Concentration indoor radon in $Bq\ m^{-3}$

DCF = Dose conversion factor ($9.0 \times 10^{-6} \text{ mSv}\ h^{-1} (Bq\ m^{-3})^{-1}$)

$R = (Ca_{Rn-222}) / (Cw_{Rn-222}) = 1 \times 10^{-4}$

F = Radon equilibrium factor (0.4)

P = Indoor occupancy time per person ($\frac{9\text{hrs}}{24\text{hrs}} \times 24\text{hrs} \times 365 \text{ days} = 3504\text{hy}^{-1}$)

v Estimation of AED due to inhalation of Rn-222 and Rn-220 in the indoor air

Varley and Flowers (1998) noted that it is limited to estimate AED due to indoor radon or thoron based on results of soil gas radon concentrations. Therefore this is left for future research work to conduct direct indoor radon and thoron measurements in houses and working places within and around Akyem Gold mine and use appropriate expressions to compute the AED due to radon and thoron .

CHAPTER FOUR

4.0. RESULTS AND DISCUSSIONS

There were altogether 28 water and soil samples that were collected from the study area of Akyem Gold Mine and the surrounding communities.

The HPGe detector was calibrated with respect to Energy and Efficiency in order to identify energy of the radionuclides and quantify the radionuclides respectively. This was achieved by using standard radionuclides in a one litre Marinelli beaker with solid water as the matrix. It was very essential to carry out calibration in order to ascertain the quality and the reliability of measurements.

Fig.4-1 and 4-2 depict Energy and Efficiency calibration whereby Energy is plotted against Channel number, Natural logarithm of Efficiency is plotted against Natural logarithm of Energy respectively.

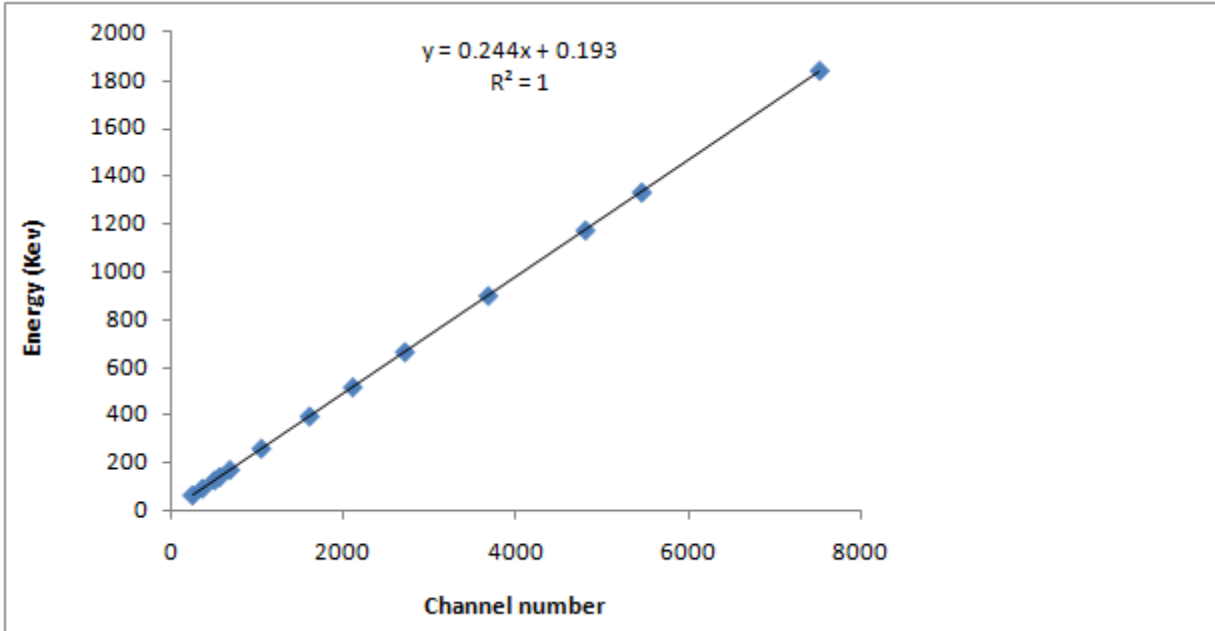


Figure 4- 1: Energy calibration curve as a function of channel number against Energy

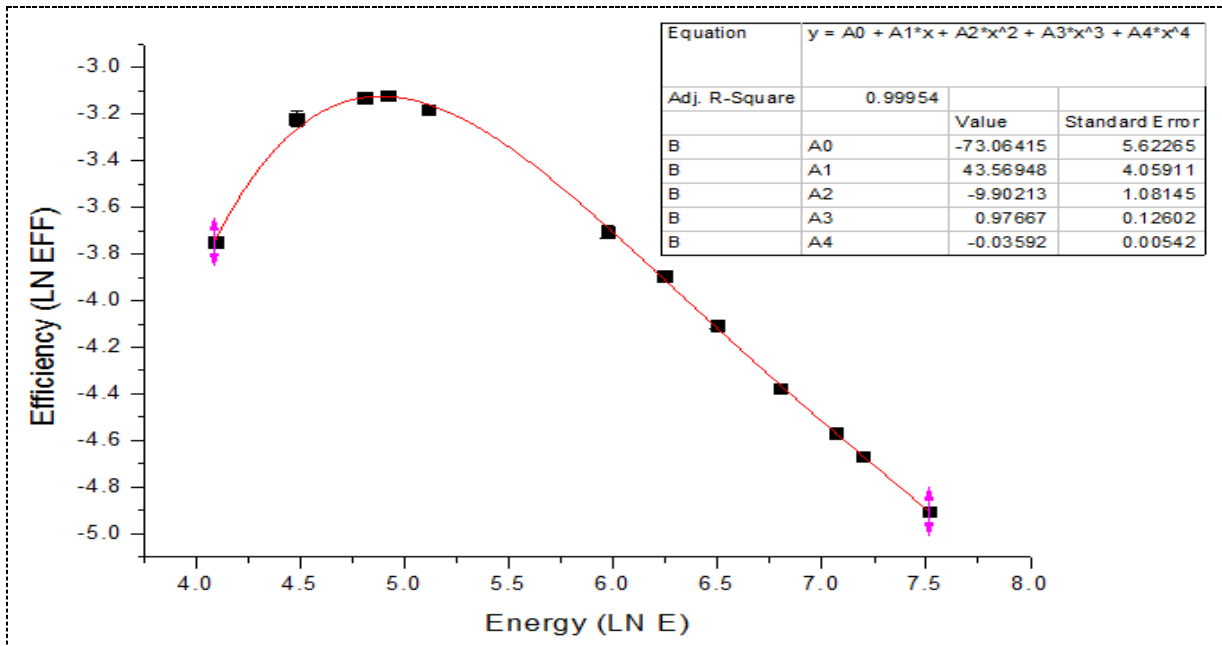


Figure 4- 2: Efficiency calibration curve as a function of natural log of energy against natural log of efficiency

4.1 Concentration of Radium, Radon and Annual Effective Dose (AED) due to radon water ingestion and inhalation

4.1.1 Ra-226 and Rn-222 activity concentrations in water

The concentrations of Ra-226 and Rn-222 are shown in Table 4-1 and the concentration of Ra-226 ranges from a low of 0.015 (SW-3) to a high of 1.708 Bq/L (SW-22) with a mean of 0.917 ± 0.608 Bq/L and radon has a range of 0.015 to 1.701 Bq/L with a mean value of 0.913 ± 0.605 Bq/L from samples taken from SW3, SW22 respectively (Fig 4-3). The high concentration of Ra-226, Rn-222 at sampling point SW-22 situated at Pra River west of Study Area near Teshie-Praso consists mainly of phyllites slates which according to Ramola et al (2006) usually contains higher radium-radon levels compared to sampling points consisting of mainly sedimentary, quartzite rocks.

Rn-220 was difficult to detect in surface water since the values found were below the Minimum Detectable Activity (MDA), thus thoron results were ignored in this thesis.

4.1.2 Comparison of Rn-222 in water with other studies

The results obtained in this project were compared with data from various studies in Ghana and other parts of the world (Table 4-2 and Figures 4-4). Most studies in Ghana have dealt much with borehole water and literature has shown that the surface water contains less radon dissolved in it (as most of it diffuses into the atmosphere) than the borehole water and this study found similar results with much lower mean value of 0.91 Bq/L against the threshold of 11.1 Bq/L according to USEPA, 1999.

4.1.3 AED associated with Radon and other natural radionuclides

4.1.3.1 AED due to ingestion and inhalation of radon gas in water

Table 4-1 also shows the Annual Effective Doses (AED) due to ingestion and inhalation of radon in water which ranges from 0.038 to 4.346 $\mu\text{Sv/y}$ with a mean value of $2.333 \pm 1.546 \mu\text{Sv/y}$ and from 0.019 to 2.146 $\mu\text{Sv/y}$ with mean value of $1.152 \pm 0.763 \mu\text{Sv/y}$ respectively adding to aggregate total annual dose spanning from 0.057 to 6.492 $\mu\text{Sv/y}$ with a mean value of $3.486 \pm 2.309 \mu\text{Sv/y}$. The same results are shown (Fig 4-6) from which it is clear to notice that the people at the catchment area of Akyem may ingest more radon dissolved in water than they inhale the indoor radon gas originating from the water even though the doses are generally insignificant. The increase in the amount of radon dissolved in water could be attributed to mining and mineral processing activities taking place at Akyem and the higher AED could be due to use of surface water for drinking in which each person consumes over 700L per year.

Comparison of AED radon ingestion and inhalation has shown that people at Akyem Newmont has an average dose of 3.486 $\mu\text{Sv/y}$ (Table 4-1 and Figure 4-5) compared to the AED limit of 100 $\mu\text{Sv/y}$ (WHO,2004).

The findings reveal that the people at Newmont-Akyem mine may freely use the surface water for drinking and other domestic purposes as the doses due to ingestion and inhalation are much lower than the global recommended values.

4.1.3.2 AED due to ingestion of natural radionuclides (^{226}Ra , ^{232}Th and ^{40}K)

The total AED due to ingestion of ^{226}Ra , ^{232}Th and ^{40}K in the surface water is shown in Table 4-3 and varies from 33.726 to 590.087 $\mu\text{Sv/y}$ with the average value of $175.490 \pm 15.343 \mu\text{Sv/y}$ lower than the recommended value of 1000 $\mu\text{Sv/y}$ according to WHO report of 2011, in comparison to the sum of ingestion and inhalation of radon in the water ranging from 0.057 to 6.492 $\mu\text{Sv/y}$ with mean value of $3.486 \pm 2.309 \mu\text{Sv/y}$ (Table 4-1). The results show that the AED from the radon ingestion in water is much lower than that from the ingestion of Ra-226, Th-232 and K-40 in water.

Table 4- 1: Concentration (Radium, Radon) and AED (Ingestion, Inhalation) of Radon -water samples

| Sample Location | Concentration, Bq/L | | AED, $\mu\text{Sv/y}$ | | |
|---------------------------------------|-------------------------------------|--------------------------------------|-------------------------------------|-------------------------------------|--|
| | Ra-226 | Rn-222 | Ingestion | Inhalation | $\Sigma_{\text{AED}}(\text{ing, inh})$ |
| AFO-12 | 0.876 ± 0.121 | 0.872 ± 0.120 | 2.228 ± 0.307 | 1.100 ± 0.151 | 3.328 ± 0.229 |
| SW-3 | 0.015 ± 0.001 | 0.015 ± 0.001 | 0.038 ± 0.003 | 0.019 ± 0.001 | 0.057 ± 0.002 |
| SW-8 | 0.639 ± 0.116 | 0.636 ± 0.115 | 1.625 ± 0.294 | 0.802 ± 0.145 | 2.427 ± 0.219 |
| SW-11 | 0.799 ± 0.093 | 0.796 ± 0.093 | 2.034 ± 0.238 | 1.004 ± 0.117 | 3.038 ± 0.177 |
| SW-15 | 1.568 ± 0.127 | 1.561 ± 0.126 | 3.988 ± 0.322 | 1.969 ± 0.159 | 5.957 ± 0.240 |
| SW-18 | 1.507 ± 0.125 | 1.500 ± 0.124 | 3.833 ± 0.317 | 1.892 ± 0.156 | 5.725 ± 0.237 |
| SW-22 | 1.708 ± 0.124 | 1.701 ± 0.123 | 4.346 ± 0.314 | 2.146 ± 0.155 | 6.492 ± 0.235 |
| SW-30A | 0.094 ± 0.013 | 0.094 ± 0.013 | 0.240 ± 0.033 | 0.119 ± 0.016 | 0.359 ± 0.025 |
| SCS-2 | 1.391 ± 0.124 | 1.385 ± 0.123 | 3.539 ± 0.314 | 1.747 ± 0.155 | 5.286 ± 0.235 |
| SCS-3/4D | 0.576 ± 0.107 | 0.573 ± 0.107 | 1.464 ± 0.273 | 0.723 ± 0.135 | 2.187 ± 0.204 |
| Minimum | 0.015 ± 0.001 | 0.015 ± 0.001 | 0.038 ± 0.003 | 0.019 ± 0.001 | 0.057 ± 0.002 |
| Maximum | 1.708 ± 0.124 | 1.701 ± 0.123 | 4.346 ± 0.314 | 2.146 ± 0.155 | 6.492 ± 0.235 |
| Average \pm StDev | 0.917 ± 0.608 | 0.9133 ± 0.605 | 2.333 ± 1.546 | 1.152 ± 0.763 | 3.486 ± 2.309 |

AED: Annual Effective Dose, **ing:** ingestion, **inh:** inhalation

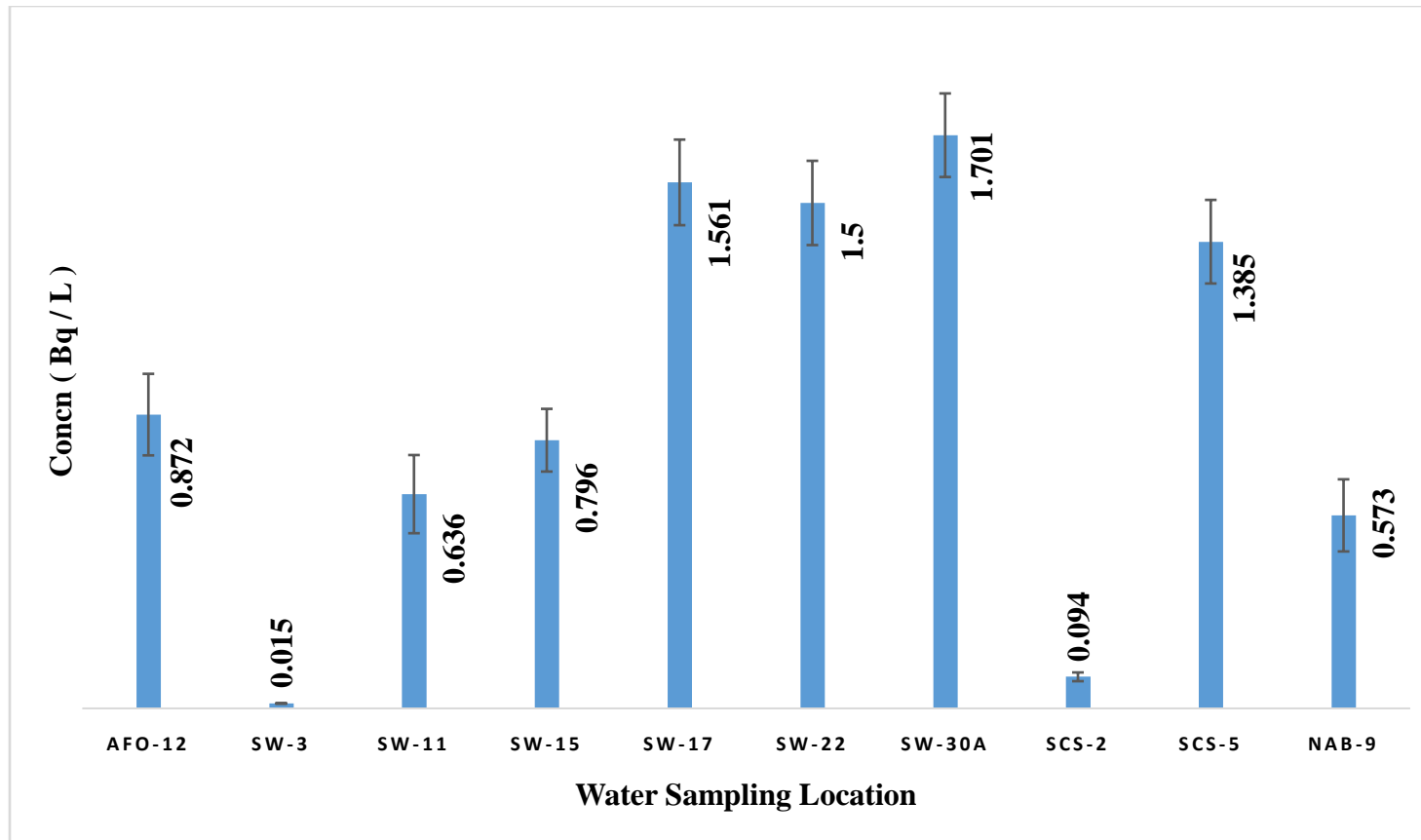


Figure 4- 3: Radon-222 concentration of selected water samples

Table 4- 2: Comparison with other studies of Radon concentration and Annual Effective Dose due to water ingestion

| Country | Type of Sample | Measurement Technique | Concn (Bq/L) | AED, μSvy^{-1} | Reference |
|---------|----------------|-----------------------|-----------------------|---------------------------|---------------------|
| | | | Rn-222 | | |
| Ghana | Surface water | HPGe | 0.91 | 3.49 | This study |
| Ghana | Borehole | HPGe | 8.10 | 59.20 | Darko et al,2010 |
| Ghana | Borehole | HPGe | 4.38×10^{-5} | 1.14 | Asumadu et al,2012 |
| India | Tap water | Radon Emanometry | 53.03 | 0.01 | Rajesh et al, 2014 |
| India | water | RAD 7 Radon Monitor | 4.7 | 13 | Rohit et al, 2015 |
| Iraq | Tap water | Alpha Track Detectors | 4.69 | 11.55 | Asaad & Salih, 2008 |
| Global | Drinking water | - | 11.1 | 100.00 | USEPA,1999;WHO,2004 |

Concn: Concentration**AED :**Annual Effective Dose**HPGe:** High Purity Germanium detector

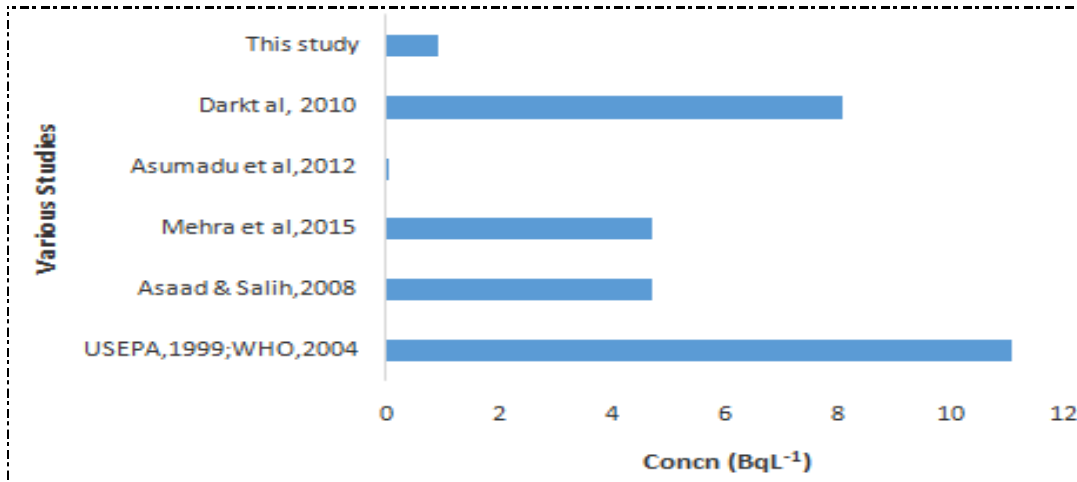


Figure 4- 4: Comparison of Rn-222 concentration in water with other studies

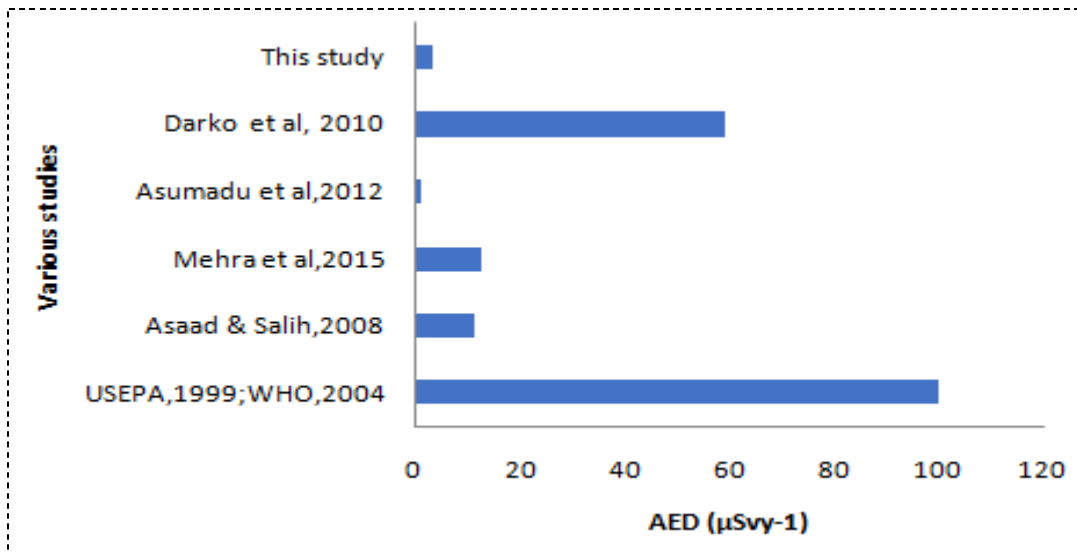


Figure 4- 5: Comparison of AED due to ingestion of radon in water

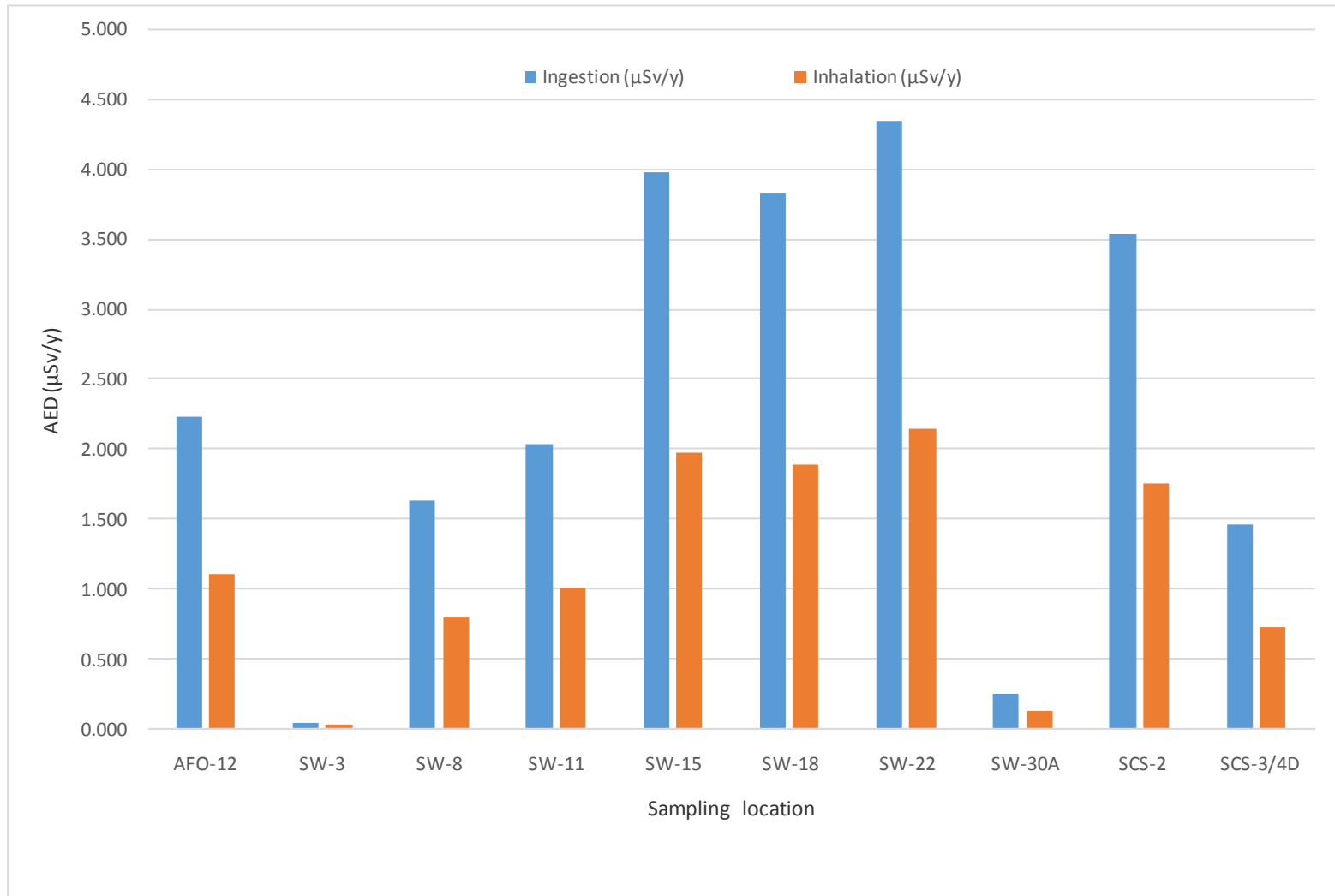


Figure 4- 6: Distribution of Annual Effective Dose due to ingestion and inhalation of Rn-222 in water

Table 4- 3: AED due to ingestion of other natural radionuclides in water apart from Rn-222

| Sample Location | Concentration, Bq/L | | | <i>AED</i> _{Ing} , μ Sv/y |
|------------------------------------|-------------------------------------|-------------------------------------|-------------------------------------|--|
| | Ra-226 | Th-232 | K-40 | |
| AFO-12 | 0.876 \pm 0.121 | 1.521 \pm 0.201 | 1.347 \pm 0.488 | 440.527 \pm 60.485 |
| SW-3 | 0.015 \pm 0.001 | 0.147 \pm 0.076 | 1.321 \pm 0.458 | 33.726 \pm 15.038 |
| SW-8 | 0.639 \pm 0.116 | - | 1.155 \pm 0.501 | 135.839 \pm 25.774 |
| SW-11 | 0.799 \pm 0.093 | 0.073 \pm 0.023 | - | 175.572 \pm 22.871 |
| SW-15 | 1.568 \pm 0.127 | 0.061 \pm 0.012 | 1.620 \pm 0.496 | 38.073 \pm 30.014 |
| SW-18 | 1.507 \pm 0.125 | 1.321 \pm 0.011 | 0.734 \pm 0.467 | 533.149 \pm 29.306 |
| SW-22 | 1.708 \pm 0.124 | 1.412 \pm 0.066 | 0.861 \pm 0.476 | 590.087 \pm 38.377 |
| SW-30A | 0.094 \pm 0.013 | 1.235 \pm 0.0.027 | 1.809 \pm 0.506 | 234.758 \pm 9.481 |
| SCS-2 | 1.391 \pm 0.124 | 0.055 \pm 0.015 | 1.174 \pm 0.495 | 298.868 \pm 29.900 |
| SCS-3/4D | 0.576 \pm 0.107 | 0.985 \pm 0.169 | 0.029 \pm 0.048 | 283.247 \pm 50.463 |
| Minimum | 0.015 \pm 0.001 | 0.055 \pm 0.015 | 0.029 \pm 0.048 | 33.726 \pm 9.481 |
| Maximum | 1.708 \pm 0.124 | 1.521 \pm 0.201 | 1.809 \pm 0.506 | 590.087 \pm 31.171 |
| Mean \pm StDev | 0.917 \pm 0.608 | 0.681 \pm 0.662 | 1.005 \pm 0.610 | 175.490 \pm 15.343 |

AED: Annual Effective Dose**Ing:** Ingestion

4.2 Radon and Thoron in the soil

4.2.1 Concentration of Soil gas Radon and Thoron

The results of Rn-222 and Rn-220 are shown in Table 4-4 and Fig 4-7 with concentrations varying from 4.194 to 21.114 kBq/m³ and a mean value of 10.829 ± 4.130 kBq/m³ for radon and 0.544 to 13.222 kBq/m³ with mean value of 5.062 ± 3.051 kBq/m³ for thoron.

Some research findings such as Tabar et al (2003) and others have shown that the soil gas radon concentration may differ widely due to weather pattern, conditions and soil varieties. The season of sampling may also affect radon soil concentration due to disturbance of site condition by fault movement.

4.2.2 Correlation between Radium and Radon

There is correlation between Ra-226 and Rn-222 in soil gas. Fig 4-8 shows that Rn-222 is a linear function of Ra-226 with an excellent linear coefficient of ($R^2 = 1$). The availability of Ra-226, Ra-228 and Rn-222, Rn-220 shows that there is source of Uranium-238 and Th-232 bearing minerals within the adjacent geologic units of Akyem. The best correlation of 1 for plot of Ra-226 against Rn-220 implies that most of the radon in the soil gas comes from Ra-226. While the correlation for radon, thoron in the soil was found to be 0.7 (Fig 4-9) which may mean that radon gas in the soil co-exist with thoron.

4.2.3 Comparison of soil gas radon and Thoron with other studies

Table 4-6 compares radon results in this study with various research findings carried out in Ghana and around the world. It is clear to note that the radon values in soil gas at Akyem area are within the range of those reported in Ghana and different parts of the world except a few. Moreover the values determined in this study are much below the acceptable levels according to USEPA (2005).

4.3 Soil Gas Rn-222 and Rn-220

Table 4- 4: Rn-222 and Rn-220 concentration calculated from soil samples

| Sample Location | Concentration, Bq/Kg | | Concentration, kBq/m ³ | |
|---------------------|-----------------------|-----------------------|-----------------------------------|-----------------------|
| | Ra-226 | Ra-228 | Rn-222 | Rn-220 |
| AFO-12 | 9.332 ± 0.560 | 7.602 ± 0.302 | 8.958 ± 0.538 | 3.649 ± 0.290 |
| SW-2A | 12.400 ± 0.514 | 1.133 ± 0.359 | 11.904 ± 0.658 | 0.544 ± 0.345 |
| SW-3 | 9.339 ± 0.402 | 7.554 ± 0.310 | 8.965 ± 0.515 | 3.626 ± 0.298 |
| SW-4 | 9.766 ± 0.424 | 8.628 ± 0.310 | 9.375 ± 0.543 | 4.141 ± 0.298 |
| SW-11 | 9.859 ± 0.623 | 8.499 ± 0.335 | 9.464 ± 0.798 | 4.080 ± 0.322 |
| SW-15 | 4.369 ± 0.226 | 4.509 ± 0.232 | 4.194 ± 0.289 | 2.164 ± 0.223 |
| SW-17 | 9.185 ± 0.400 | 9.354 ± 0.329 | 8.817 ± 0.511 | 4.490 ± 0.316 |
| SW-18 | 6.547 ± 0.304 | 6.682 ± 0.288 | 6.285 ± 0.388 | 3.207 ± 0.276 |
| SW-22 | 21.994 ± 1.307 | 27.545 ± 0.520 | 21.114 ± 1.672 | 13.222 ± 0.499 |
| SW-30A | 12.286 ± 0.519 | 10.183 ± 0.370 | 11.795 ± 0.664 | 4.888 ± 0.355 |
| SCS-2 | 10.605 ± 0.453 | 11.933 ± 0.363 | 10.181 ± 0.580 | 5.728 ± 0.348 |
| SCS-3/4D | 16.355 ± 0.665 | 17.562 ± 0.457 | 15.701 ± 0.851 | 8.430 ± 0.439 |
| SCS-5 | 14.572 ± 0.606 | 14.605 ± 0.428 | 13.989 ± 0.775 | 7.010 ± 0.411 |
| NAB-9 | 11.312 ± 0.704 | 11.841 ± 0.349 | 10.859 ± 0.902 | 5.684 ± 0.335 |
| Minimum | 4.369 ± 0.226 | 1.133 ± 0.359 | 4.194 ± 0.289 | 0.544 ± 0.290 |
| Maximum | 21.994 ± 1.307 | 27.545 ± 0.520 | 21.114 ± 1.672 | 13.222 ± 0.499 |
| Mean ± StDev | 11.280 ± 4.301 | 10.545 ± 6.357 | 10.829 ± 4.130 | 5.062 ± 3.051 |

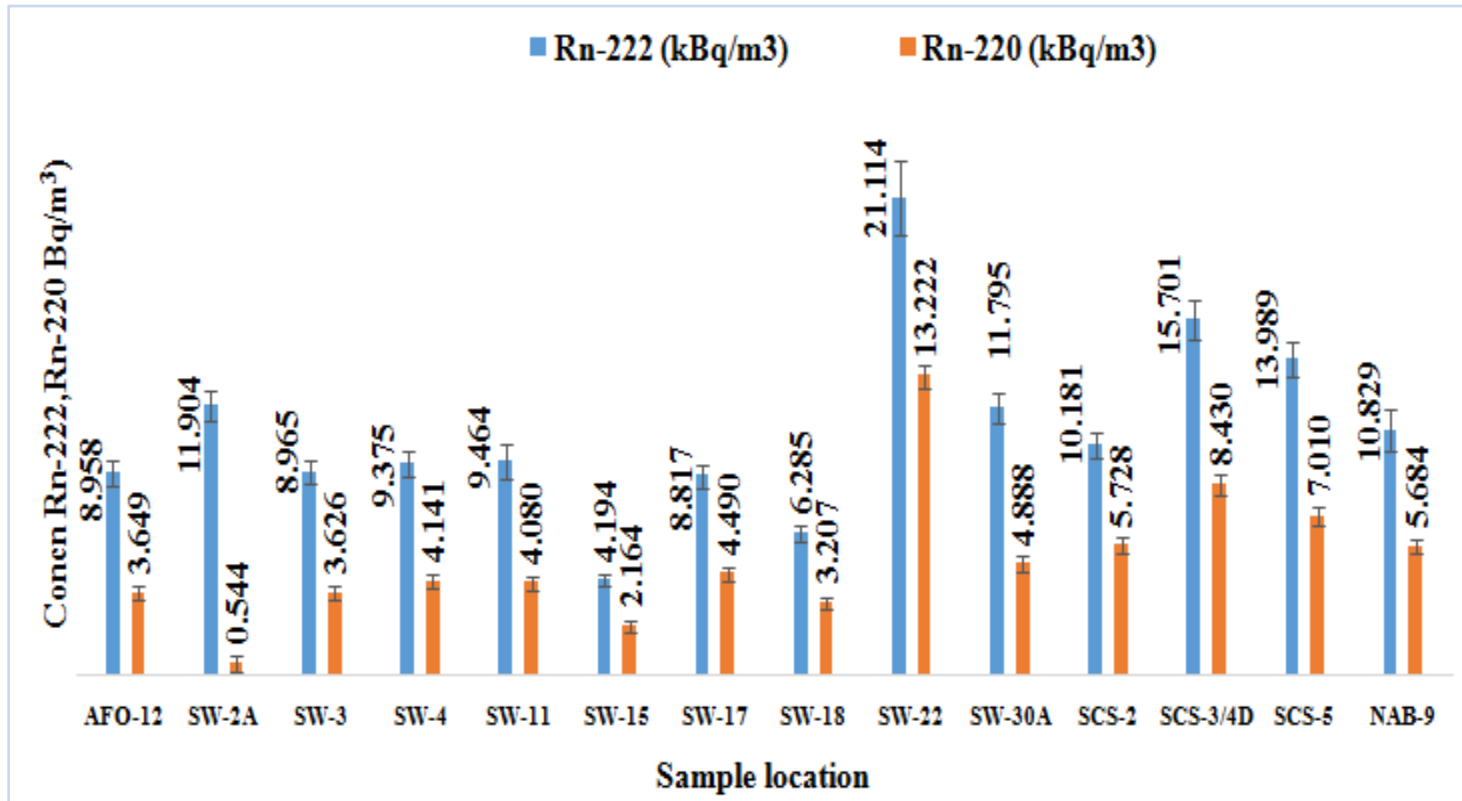


Figure 4- 7: Calculated Soil Gas Rn-222, Rn-220 in the soil samples

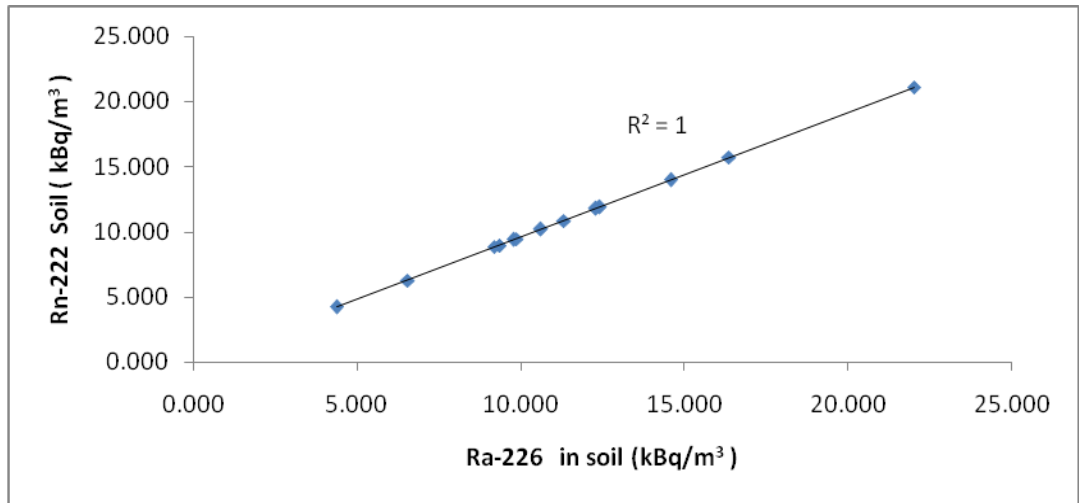


Figure 4- 8: Correlation between Ra -226 and Rn-222 in soil gas

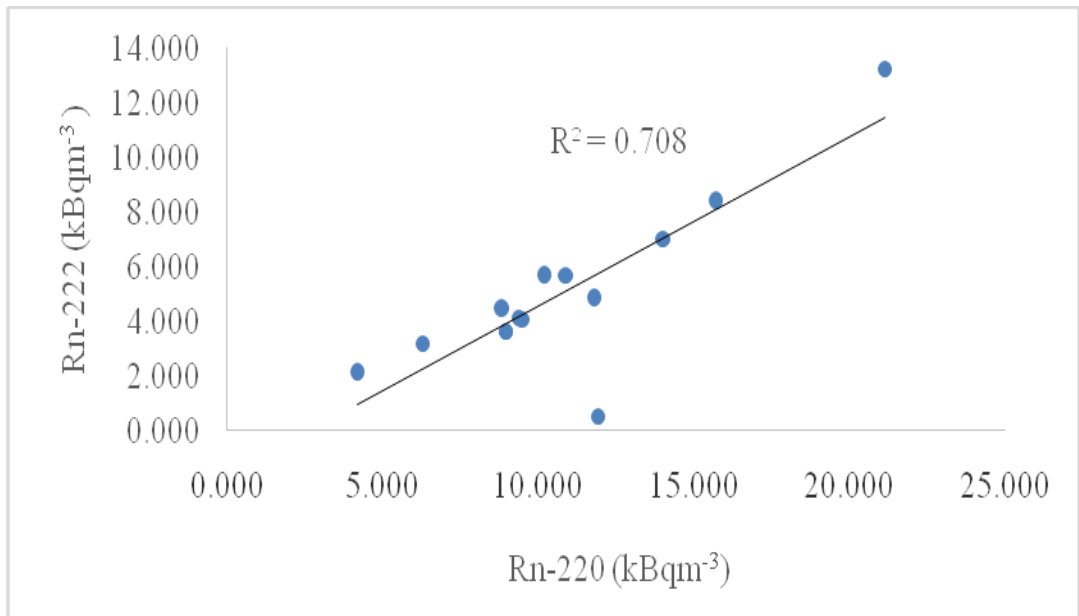


Figure 4- 9: Correlation between Rn -222 and Rn-220 in soil gas

Table 4- 5: Comparison of Soil Gas Radon with other studies

| Country | Sampling Location | Measurement Technique | Concentration, Bq/m ⁻³ | | Ratio | Reference |
|---------|-------------------|-----------------------|-----------------------------------|-------------|---------------|----------------------|
| | | | Rn-222 | Rn-220 | Rn-220/Rn-222 | |
| Ghana | G.Mine | HPGe | 4,194 –21,114 | 544 –13,222 | 0.444 | This study |
| Ghana | G.Mine | Alpha Guard | 56-268 | - | - | Andam & Amoo,1994 |
| Ghana | Mine | Alpha Guard | 43-878 | - | - | Andam & Amoo,1994 |
| Ghana | Mine | HPGe | 12,500-41,300 | - | - | Faanu,2011 |
| Ghana | Fault | - | 9,910 – 42,10 | - | - | Amponsah P, 2008 |
| Turkey | Geotherm. Area | SSNTD | 98 – 8594 | | | Tabar et al, 2013 |
| Italy | Volcanic Mt | RAD7 Radon Meter | 232-104,300 | 10-23,350 | 0.503 | Giammanco et al,2007 |
| India - | | RAD7 Radon Meter | 3,200-17,200 | - | - | Mehra et al, 2015 |
| India | *Dwellings | SSNTD | - | - | 0.530 | Ramachandran , 2010 |
| Russia | - | - | 1,700 – 24,000 | - | - | Iakovleva et al,2003 |
| Sudan | soil | SSNTD | 5,500 -15,100- | | - | Elmoniem, 2015 |

G.: Gold **Mt.:**Mountain **SSNTD:** Solid State Nuclear Track Detector *Indoor thoron and radon

4.4 Absorbed dose

The absorbed dose was measured 1 m above the ground at each sampling point using the Survey meter. The measured absorbed dose was then compared with calculated absorbed dose based on the concentration of Ra-226, Th-232 and K-40 (Table 4-6, Fig 4-10). Almost in all sampling points the measured absorbed dose rates were higher than those calculated. This could be attributed to background radiation and human errors. The mean absorbed dose rate measured, calculated are $57, 19 \text{ nGyh}^{-1}$ respectively against the global values of 54, 60 nGy/h [UNSCEAR, 2008, 2000] respectively. The mean absorbed dose rate measured was slightly lower than that of the recommended value and some sampling points even reported higher values of measured absorbed dose rate.

4.3.1 AED, Radium equivalent activity and Internal hazard index

Table 4-7 shows the average values of Radium equivalent activity (Ra_{eq}), Internal hazard index (H_{in}) and annual effective dose (mSv/y). The Ra_{eq} was calculated and ranged from 19.708 to 69.880 Bq/Kg with mean value of $37.527 \pm 15.508 \text{ Bq/Kg}$ than the global limit of 370 Bq/Kg. The value for H_{in} ranged from 0.065 to 0.248 Bq/Kg with a mean value of $0.132 \pm 0.053 \text{ Bq/Kg}$ which is lower than the agreed value of unity. While for the total annual effective dose due to external and internal gamma dose the range was 0.060 - 0.175 mSv/y with the mean value of $0.11 \pm 0.025 \text{ mSv/y}$ against the world value of 1 mSv/y.

4.3.2 Comparison of Ra_{eq} , H_{in} and AED with the other studies

Table 4-8 gives comparison of Ra_{eq} , H_{in} and AED with the other studies in Ghana and elsewhere. It is clear to note that values of Ra_{eq} , H_{in} and AED obtained were much lower than those recommended according to study carried out by UNSCEAR, (2008) which found the values of 370, 1, 1 Bq/Kg respectively. Table 4-8 also shows that the concentration of Ra-226, Th-232, K-40 in this study were 11, 12,113 Bq/Kg respectively against the global values of 37, 33, 400 Bq/Kg respectively.

4.5 The Absorbed Dose

Table 4- 6: NORMs concentration and measured versus calculated absorbed dose rate

| Sample Location | Concentration NORMs, Bq/Kg | | | Absorbed dose rate, nGy/h | |
|------------------------|----------------------------|-----------------------|-------------------------|---------------------------|------------------------|
| | Ra-226 | Th-232 | K-40 | Calculated | Measured |
| AFO-12 | 9.332 ± 0.560 | 7.995 ± 0.720 | 24.268 ± 0.966 | 10.321 ± 0.757 | 82.500 |
| SW-2A | 12.400 ± 0.514 | 12.182 ± 1.063 | 26.368±1.018 | 14.493 ± 0.967 | 43.333 |
| SW-3 | 9.339 ± 0.402 | 8.212 ± 0.760 | 71.956±1.811 | 12.518 ± 0.753 | 98.000 |
| SW-4 | 9.766 ± 0.424 | 9.434 ± 0.850 | 92.708±2.173 | 14.402 ± 0.837 | 47.143 |
| SW-11 | 9.859 ± 0.623 | 9.513 ± 0.882 | 111.542±2.502 | 15.303 ± 0.958 | 64.000 |
| SW-15 | 4.369 ± 0.226 | 4.870 ± 0.475 | 108.775±2.433 | 9.767 ± 0.515 | 40.000 |
| SW-17 | 9.185 ± 0.400 | 10.153 ± 0.908 | 86.716±2.070 | 14.372 ± 0.861 | 44.000 |
| SW-18 | 6.547 ± 0.304 | 7.583 ± 0.721 | 90.825±2.123 | 11.721 ± 0.698 | 47.143 |
| SW-22 | 21.994 ± 1.307 | 29.666 ± 2.405 | 70.969±1.857 | 32.081 ± 2.228 | 44.000 |
| SW-30A | 12.286 ± 0.519 | 11.180 ± 1.011 | 298.659±5.749 | 27.937 ± 1.893 | 42.857 |
| SCS-2 | 12.286 ± 0.519 | 12.403 ± 1.053 | 99.826±2.301 | 24.772 ± 1.110 | 62.500 |
| SCS-3/4D | 10.605 ± 0.453 | 19.214 ± 1.637 | 157.359±3.306 | 19.487 ± 1.080 | 68.333 |
| SCS-5 | 16.355 ± 0.665 | 15.962 ± 1.385 | 302.900±5.819 | 25.708 ± 1.484 | 74.000 |
| NAB-9 | 14.572 ± 0.606 | 12.821 ± 1.109 | 50.063±1.466 | 28.422 ± 1.468 | 34.000 |
| Minimum | 4.369 ± 0.226 | 4.870 ± 0.475 | 24.268±0.966 | 9.767 ± 0.515 | 34.000 |
| Maximum | 21.994 ± 1.307 | 29.666 ± 2.405 | 302.900±5.819 | 32.081 ± 2.228 | 98.000 |
| Average ± StDev | 11.280 ± 4.301 | 12.228 ± 6.182 | 113.781 ± 86.411 | 18.665 ± 7.59 | 56.558 ± 18.734 |

ADR: Absorbed Dose Rate

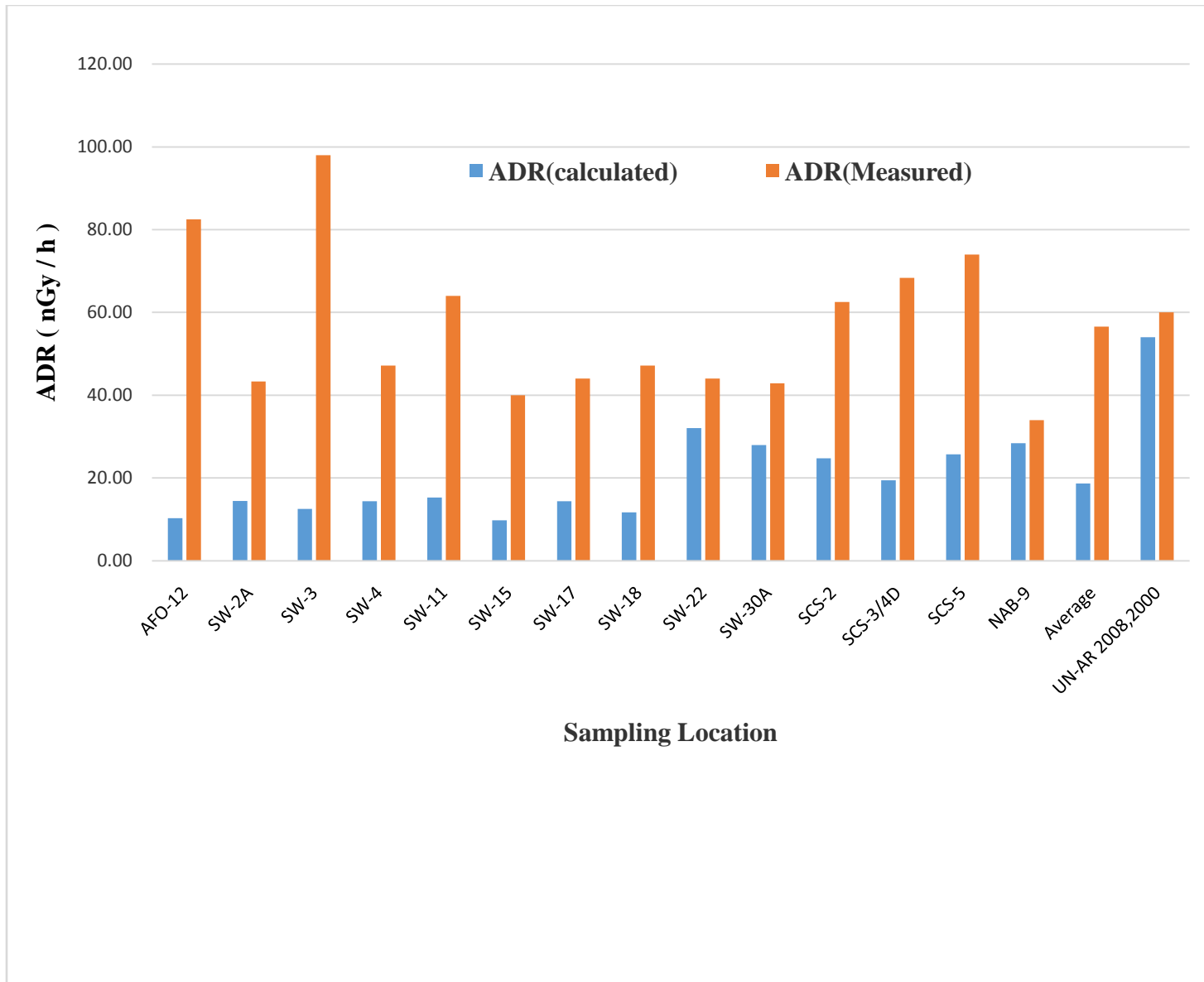


Figure 4- 10: Absorbed dose rate (ADR) measured 1 m above the ground versus that calculated

Table 4- 7: Annual effective dose, Radium Equivalent Index (Ra_{eq}) and Internal hazard index (H_{in}) of soil samples

| Sample Location | Concentration, Bq/Kg | | AED, mSv/y | | |
|---------------------|------------------------|----------------------|-----------------------|-----------------------|--------------------------|
| | Ra_{eq} | H_{in} | External | Internal | $\Sigma_{AED}(ext, int)$ |
| AFO-12 | 22.633 ± 1.664 | 0.086 ± 0.006 | 0.025 ± 0.0019 | 0.038 ± 0.0028 | 0.063 ± 0.003 |
| SW-2A | 31.851 ± 2.112 | 0.120 ± 0.007 | 0.036 ± 0.0024 | 0.053 ± 0.0036 | 0.089 ± 0.004 |
| SW-3 | 26.623 ± 1.628 | 0.097 ± 0.005 | 0.031 ± 0.0018 | 0.046 ± 0.0028 | 0.077 ± 0.003 |
| SW-4 | 30.395 ± 1.807 | 0.108 ± 0.006 | 0.035 ± 0.0021 | 0.053 ± 0.0031 | 0.088 ± 0.003 |
| SW-11 | 32.051 ± 2.077 | 0.113 ± 0.007 | 0.038 ± 0.0023 | 0.056 ± 0.0035 | 0.094 ± 0.004 |
| SW-15 | 19.708 ± 1.092 | 0.065 ± 0.004 | 0.024 ± 0.0013 | 0.036 ± 0.0019 | 0.06 ± 0.002 |
| SW-17 | 30.380 ± 1.857 | 0.107 ± 0.006 | 0.035 ± 0.0021 | 0.053 ± 0.0032 | 0.088 ± 0.003 |
| SW-18 | 24.384 ± 1.498 | 0.084 ± 0.005 | 0.029 ± 0.0017 | 0.043 ± 0.0026 | 0.072 ± 0.003 |
| SW-22 | 69.880 ± 4.889 | 0.248 ± 0.017 | 0.058 ± 0.0055 | 0.087 ± 0.0082 | 0.145 ± 0.008 |
| SW-30A | 51.271 ± 2.407 | 0.172 ± 0.008 | 0.069 ± 0.0046 | 0.103 ± 0.0070 | 0.172 ± 0.007 |
| SCS-2 | 36.028 ± 2.136 | 0.126 ± 0.007 | 0.061 ± 0.0027 | 0.091 ± 0.0041 | 0.152 ± 0.004 |
| SCS-3/4D | 55.948 ± 3.260 | 0.195 ± 0.011 | 0.048 ± 0.0026 | 0.072 ± 0.0040 | 0.12 ± 0.004 |
| SCS-5 | 60.720 ± 3.034 | 0.203 ± 0.010 | 0.063 ± 0.0036 | 0.095 ± 0.0055 | 0.158 ± 0.006 |
| NAB-9 | 33.501 ± 2.403 | 0.121 ± 0.008 | 0.070 ± 0.0013 | 0.105 ± 0.0054 | 0.175 ± 0.005 |
| Minimum | 19.708 ± 1.092 | 0.065 ± 0.004 | 0.024 ± 0.0013 | 0.036 ± 0.0019 | 0.06 ± 0.002 |
| Maximum | 69.880 ± 4.889 | 0.248 ± 0.017 | 0.070 ± 0.0013 | 0.105 ± 0.0054 | 0.175 ± 0.005 |
| Mean ± StDev | 37.527 ± 15.508 | 0.132 ± 0.053 | 0.044 ± 0.0170 | 0.066 ± 0.0248 | 0.11 ± 0.025 |

Table 4- 8: Comparison of Annual effective dose, Radium Equivalent Index and Internal hazard index with other studies in soil samples by HPGe detector

| Country | Concentration, Bq/Kg | | | | | AED, mSv/y | Reference |
|---------|----------------------|--------|---------|--------|-------|------------|------------------|
| | Ra-226 | Th-232 | K-40 | Raeq | Hin | | |
| Ghana | 11.28 | 12.228 | 113.781 | 37.527 | 0.132 | 0.11 | This study |
| Ghana | 13.61 | 24.22 | 162.08 | 61 | 0.2 | 0.17 | Faanu, 2011 |
| Kenya | 40.7 | 35.8 | 574.4 | 140.69 | 0.48 | 0.17 | Aguko et al,2013 |
| Global | 37 | 33 | 400 | 370 | 1 | 1 | UNSCEAR,2008 |

4.6 Physico-chemical properties of water in relation to Radon-222 concentration

Before water was taken from the sampling points it was analysed for physico-chemical properties in order to compare with radon concentration (Table 4-9).This is because the power of Hydrogen (pH), temperature, conductivity and total dissolved solids are said to have some effects on the radon concentration thereby contributing to the radiological characteristic of the water. Figures 4-11 through 4-14 show plot of each parameter against radon concentration, from which there have not been any correlation established. Fig 4-15 depicts physico-chemical parameters and radon concentration in which the data were normalized to unity and expressed as percentage (%) index. Comparison of the trends in the four parameters with the radon concentration as shown in Fig.4-15, however, the comparison does not reveal conclusive probably due to close proximity of the sampling points or the geographical locations. The study area also has similar geological structures and meteorological pattern. Parameter variations may also be due to statistical waverings since the sampling positions are within short distances of reach to each other [Darko et al, 2010].

Table 4- 9: Physico-chemical properties of water in relation to Radon-222 concentration

| Sample Location | Rn-222 | Physico-Chemical properties of water | | | |
|------------------------------------|-------------------------------------|--------------------------------------|------------------------------------|------------------------------------|------------------------------------|
| | Bq/L | pH | Temperature, °C | Conductivity, μScm^{-1} | TDS, mgL^{-1} |
| AFO-12 | 0.872 | 7.45 | 26.7 | 194.8 | 140.1 |
| SW-2A | <MDA | 7.53 | 25.2 | 247.6 | 160.9 |
| SW-3 | 0.015 | 7.6 | 27 | 192.7 | 125.6 |
| SW-4 | <MDA | 7.49 | 30.8 | 294.4 | 191.3 |
| SW-8 | 0.636 | 7.72 | 26.4 | 234.4 | 152.2 |
| SW-11 | 0.796 | 7.8 | 25.8 | 121.1 | 78.8 |
| SW-15 | 1.561 | 7.6 | 25.5 | 192.4 | 125.5 |
| SW-17 | <MDA | 7.12 | 26.4 | 213.3 | 138.6 |
| SW-18 | 1.5 | 7.72 | 25.9 | 250.4 | 162.8 |
| SW-22 | 1.701 | 7.65 | 27.8 | 280.3 | 186.5 |
| SW-30A | 0.094 | 7.45 | 26.5 | 345.7 | 224 |
| SCS-1 | <MDA | 7.76 | 29.5 | 117.3 | 77 |
| SCS-2 | 1.385 | 7.96 | 31.4 | 166.9 | 108.3 |
| SCS-3/4 | 0.573 | 7.89 | 30.6 | 311.1 | 107.8 |
| Minimum | 0.015 | 7.12 | 25.2 | 117.3 | 77 |
| Maximum | 1.701 | 7.96 | 31.4 | 345.7 | 224 |
| Mean \pm StDev | 0.913 \pm 0.605 | 7.62 \pm 0.21 | 27.54 \pm 2.13 | 225.9 \pm 67.9 | 141.4 \pm 42.2 |

TDS: Total Dissolved Solids

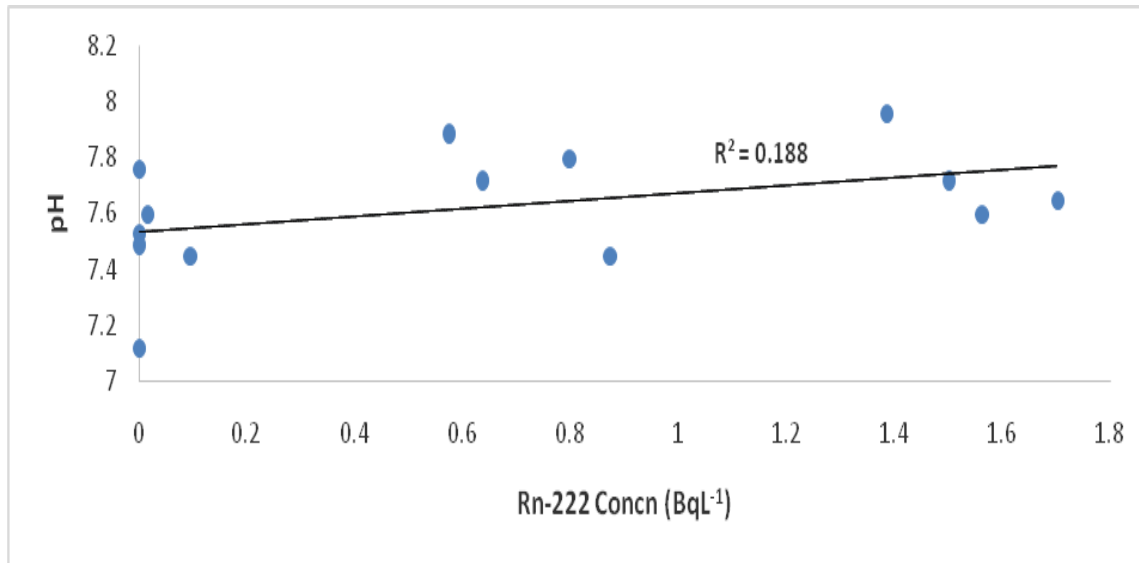


Figure 4- 11: Correlation between pH and Rn-222

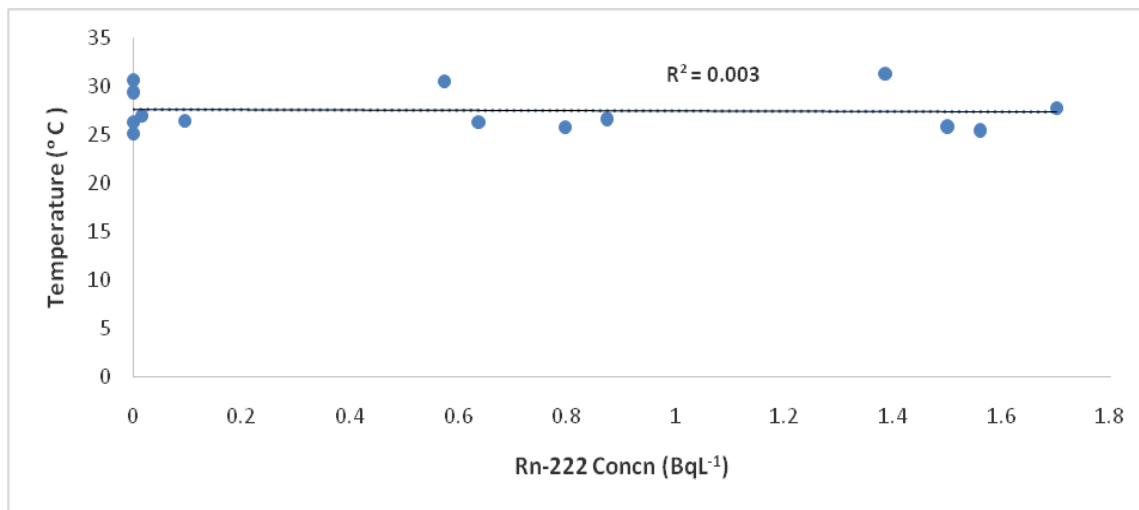


Figure 4- 12: Correlation between Temperature and Rn-222

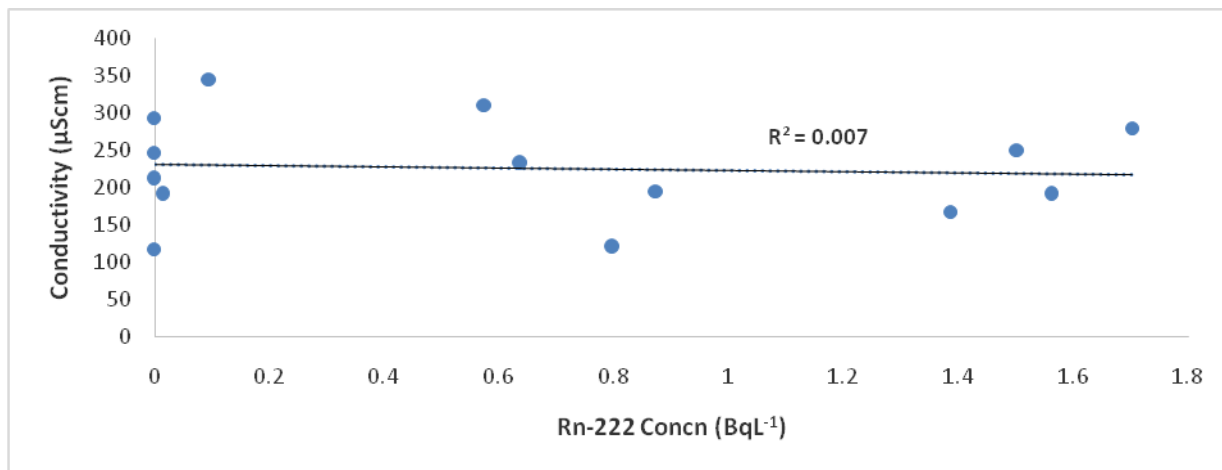


Figure 4- 13: Correlation between Conductivity and Rn-222

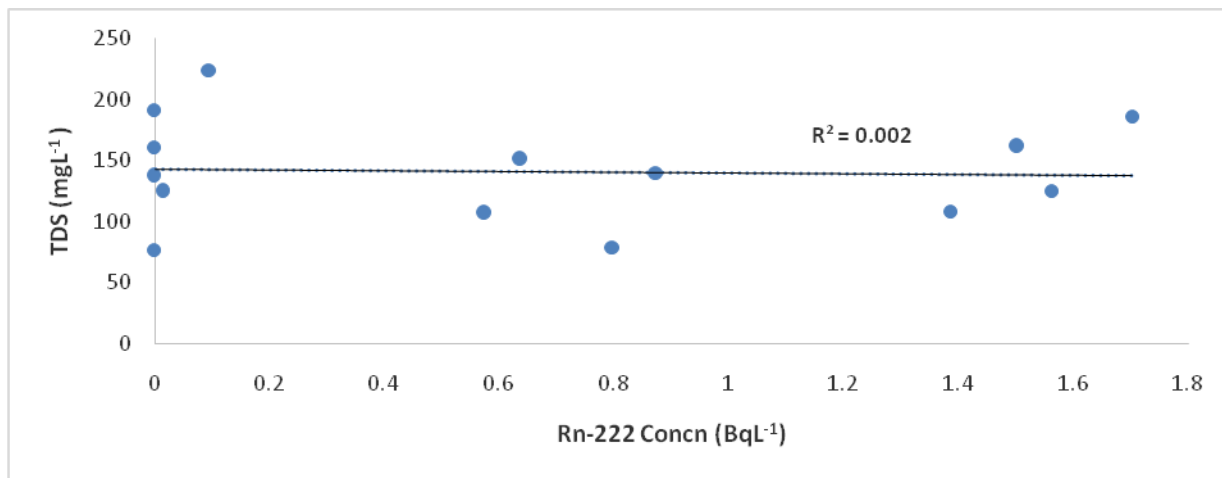


Figure 4- 14: Correlation between TDS and Rn-222

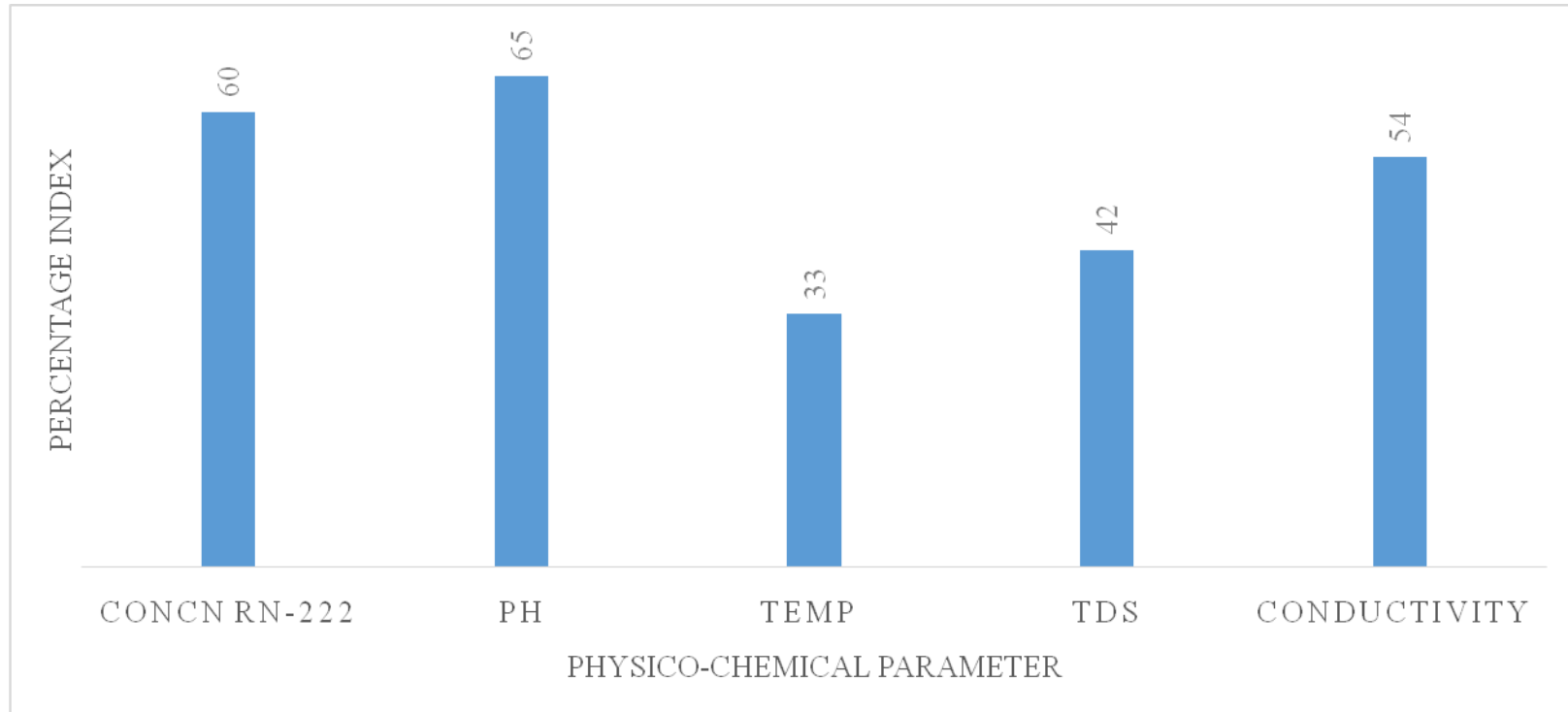


Figure 4- 15 : Percentage Index physico-chemical parameters versus that of Rn-222

CHAPTER 5

5.0. CONCLUSION AND RECOMMENDATIONS

5.1 CONCLUSION

The purpose of the project work was to determine levels of ^{222}Rn , ^{220}Rn plus ^{226}Ra , ^{232}Th and ^{40}K in water and soil samples at Newmont-Akyem Gold Mine and assess hazards posed by these natural radionuclides to the public. Three exposure pathways were outlined for the study area starting with direct external gamma ray exposure from natural radioactivity concentrations in soil, then internal exposure from drinking water containing natural radioactivity, finally inhalation of radon gas originating from water. The communities covered during this work include Afosu, New Abirem, Mamanso, Ntronang, James town, Aduasena to mention just a few.

Radon, Thoron and other natural radionuclides assessment in water and soil samples were measured by Gamma Spectrometry System equipped with High Purity Germanium detector. It has been observed that the Rn-222 concentration in the surface water samples is much lower than the international agreed value of 11.1Bq/L [USEPA, 1999]. The total annual effective dose for Rn-222 in water at the study area is 3.5 $\mu\text{Sv/y}$ which is very low compared to the world agreed value of 100 $\mu\text{Sv/y}$ according to WHO (2004). One of the explanation for this is that most radon in the surface water diffuses into the atmosphere.

People at Newmont-Akyem use the surface water for drinking, washing clothing, cooking and other domestic purposes, so with these findings it can be concluded that the communities surrounding the mine are not being exposed to significant levels of NORMs. This means they can freely use the water with very minimal radiological

hazards, but pollutants like metals, organochlorine pesticide residues, polycyclic aromatic hydrocarbons etc. need to be assessed for drinking water quality purposes.

Most people in the study area use soil and water for constructing dwellings, therefore it was necessary to assess if there are any radiological hazards associated with soil. This was achieved by determining Radium equivalent activity (Ra_{eq}) and Internal hazard index (H_{in}) which are related to radon gas. Both values found for Ra_{eq} , H_{in} were much less than the values of 370 Bq/Kg and One(1) respectively as recommended by UNSCEAR (2008).

The average absorbed dose rate and the AED due to Ra-226, Th-232 and K-40 are less than the world values. In terms of soil gas radon and thoron they lie within the acceptable levels.

Although soil gas radon and thoron concentrations were determined, it was difficult to find AED based on soil gas radon and thoron as literature indicates that it gives limited results. Thus, in this project it was difficult to conclude whether people at Akyem are safe from radon without AED based on radon indoor direct measurement. However the results for water and soil gas radon have shown that the people may safely use the water and soil with very minimal radiological hazards.

5.2 RECOMMENDATIONS

From this research it has been noted that there is a strong positive correlation between the soil gas radon and Ra-226. Literature has shown that about 2% of Rn-222 in the soil emanates into the atmosphere. Therefore it is recommendable to :

- Conduct direct indoor radon measurement in order to know the contribution ratio of the Rn-222 in the soil, water to that indoor air
- Use indoor radon concentration values to estimate the AED to the public due to inhalation of radon emanating from both the soil and water into the indoor air

In addition there is need:

- For an intensive research work to find out whether there exists the relationship between the radon concentration and the physico-chemical properties of water in the sampling area
- To conduct research at Akyem-Newmont communities on water quality to measure heavy metals by Atomic Absorption Spectrometry (AAS), Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) or Neutron Activation Analysis (NAA) Techniques, pesticides residues and other Persistent Organic Pollutants (POPs) by Gas Chromatography-Mass Spectrometry (GC-MS)

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ANNEXES**Annex 1: Soil and water sample identification and locations**

| Soil Sample | Water Sample | Location |
|--------------------|---------------------|---|
| AFO-12 | AFO-12 | Afosu drilled/ hand pump |
| SW-2A | SW-2A | Afosu stream below Ninki (SW-2A is upstream of former SW-2 station) |
| SW-3 | SW-3 | Afosu stream above Yaayaaso (moved downstream) |
| SW-4 | SW-4 | Afosu stream above Abirem-Yaayaaso road (moved upstream) |
| - | SW-8 | Sentifa stream above confluence with Adenkyerensu stream |
| SW-11 | SW-11 | Adenkyerensu stream below proposed Tailing Storage Facility (TSF) |
| SW-15 | SW-15 | Aprapon stream near Ntronang |
| SW-17 | SW-17 | Afosu stream upstream of Mamang confluence |
| SW-18 | SW-18 | Beayaa stream at Hweakwae village |
| SW-22 | SW-22 | Pra River west of Study Area near Teshie-Praso |
| SW-30A | SW-30A | Adenkyerensu stream above proposed TSF |
| - | SCS-1 | Sediment Control Structure 1 |
| SCS-2 | SCS-2 | Sediment Control Structure 2 |
| SCS-3/4D | SCS-3/4D | Sediment Control Structure 3 |
| SCS-5 | - | Sediment Control Structure 5 |
| NAB-9 | - | New Abirem hand dug well |

Annex 2: The standards used in energy and efficiency calibration of HPGe detector

| Radionuclide | Half-life (dy) | Activity(Bq) as on 20/03/2014 | Uncertainty (%) | Energy (keV) |
|---------------------|---------------------------|--|----------------------------|---------------------|
| Am-241 | 157800 | 4694 | 1.1 | 60 |
| Cd-109 | 462.6 | 14540 | 1.4 | 88 |
| Ce-139 | 137.5 | 1355 | 1.1 | 166 |
| Co-57 | 271.26 | 1156 | 1.1 | 122 |
| Co-60 | 1925.4 | 2697 | 1.1 | 1173,1333 |
| Cs-137 | 11019 | 2689 | 1.3 | 662 |
| Sn-113 | 115.1 | 4000 | 2.2 | 392 |
| Sr-85 | 64.78 | 4570 | 1.5 | 514 |
| Y-88 | 106.6 | 5323 | 1.2 | 898,1836 |