

**RADIOLOGICAL ASSESSMENT AND HEAVY METALS
ANALYSIS AT KADUNA REFINING AND
PETROCHEMICAL COMPANY LIMITED, NIGERIA**

BY

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AHMADU BELLO UNIVERSITY,
ZARIA, NIGERIA**

OCTOBER, 2018

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BY

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MSc/Radiation Biophysics/P15SCP8023**

**A THESIS SUBMITTED TO THE SCHOOL OF POSTGRADUATE STUDIES,
AHMADU BELLO UNIVERSITY, ZARIA**

**IN PARTIAL FULFILLMENT FOR THE REQUIREMENTS FOR THE AWARD
OF MASTER DEGREE IN RADIATION BIOPHYSICS**

DEPARTMENT OF PHYSICS

FACULTY OF PHYSICAL SCIENCES

AHMADU BELLO UNIVERSITY,

ZARIA NIGERIA

OCTOBER, 2018

DECLARATION

I hereby declare that the work in this Thesis entitled Radiological and Heavy Metals Assessment of Kaduna Refining and Petrochemical Company Limited, Nigeria has been carried out by me in the Department of Physics, Ahmadu Bello University. The information derived from literature has been duly acknowledged in the text and a list of references provided. No part of this thesis was previously presented for another degree or diploma at this or any other Institution.

Mohammad Abubakar

Name of student

Signature

Date

CERTIFICATION

This Thesis entitled “RADIOLOGICAL AND HEAVY METALS ASSESSMENT OF KADUNA REFINING AND PETROCHEMICAL COMPANY LIMITED, NIGERIA” by MOHAMMAD ABUBAKAR (M.Sc. P15SCPY8023) meets the regulations governing the award of the degree of master of science (Radiation Biophysics) of the Ahmadu Bello University, and is approved for its’ contribution to knowledge and literary presentation.

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DEDICATION

This work is dedicated to my parents, Late Alhaji Abubakar and Hajiya Aisha Musa Yola.

ACKNOWLEDGEMENT

I will like to express my sincere gratitude to my supervisors, Dr. N.N. Garba and Dr. M. A. Onoja for their patience, understanding and guidance. I have greatly benefited from their long experience in teaching as well as in my research.

My indebtedness also goes to the Head of Department and the entire staff members of Physics department for always being there to assist me throughout my program.

My special appreciation goes to my lovely, caring and understanding brothers, Alhaji Adamu Garba, Abubakar Garba, Musa Garba, Late Sufyan Abubakar and Mohammad Ahmad for their support throughout my studies. I specifically acknowledge the spiritual and moral support of Alhaji Adamu Samaila and Ado Mundu, thank you so much, am also indebted to all my sisters most especially Hajiya Hauwa Abubakar and Hajiya Rabi Abubakar and the entire family for their constant love, concern and support towards my career.

I want to thank all my colleagues especially Mr. Suleiman Bello and Muhammad Bello Gusau whose advice and contribution saw me through the work. And to my friends who contributed in one way or the other to make this study a success, i say thank you.

My profound gratitude goes to the management of Center for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Kaduna Refining and Petrochemical Company Nigeria and National Institute for Radiation Protection and Research Ibadan for granting me opportunity to use their facilities during the field and experimental work.

Finally I wish to acknowledge Adama Minal Muhammad for her unwavering moral support, prayers and great words of encouragement that kept me moving at moments I felt alone. Thank you so much.

ABSTRACT

This study assessed the radiological and some heavy metal (Pb, Zn, Fe, Mn, Ni, Cr, Co and Cd) composition in soil and plant sample at five different locations inside Kaduna Refining and Petrochemical Company using gamma spectrometry (HPGe detector) and Atomic Absorption Spectroscopy (AAS). Activity concentrations of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs in soil and plant samples were determined using high purity germanium (HPGe) detector. All studied radionuclides were present in all the samples except for ^{137}Cs which was not detected in all the samples. The mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K were observed to be 61.98, 65.37 and 207.79Bq/kg in soil and 18.73, 12.73 and 278.79Bq/kg in plant samples. The mean radiation hazard levels in both soil and plant of study area were (154.35Bq/kg and 58.40Bq/Kg) for radium equivalent activity Ra_{eq} , (90.75 nGy/h and 28.18nGy/h) for absorb dose rate, (86.27 $\mu\text{Sv/y}$ and 34.56 $\mu\text{Sv/y}$) for annual effective dose equivalent, 0.42 for external hazard index (H_{ex}) and 0.57 for internal hazard index (H_{in}) respectively. The study shows that all the results obtained for the radiation hazard levels in all the samples were less than the world recommended average except for the AEDE and absorbed dose rate (D) in soil samples which were found to be higher than the world recommended value of 70 $\mu\text{Sv/yr}$ and 59 nGy/hr.

The study area was observed to be concentrated with Pb, Zn, Fe, Mn, Cr, Cd while Co and Ni however, were Below Detection Limit (BDL) in all the samples. The mean concentrations (ppm) of Pb, Zn, Fe, Mn, Cr, Cd for the soil samples were obtained to be 0.42, 0.02, 1.01, 0.21, 0.015 and 0.004 respectively. Similarly the mean concentrations (ppm) of Pb, Zn, Fe, Mn, Cr, Cd for the plant samples were obtained to be 0.18, 0.02, 0.22, 0.15, 0.01 and 0.003 respectively. The obtained results were found to be low when compared with the World Health Organization (WHO) recommended values on maximum limits of the studied heavy metals in soil samples.

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CHAPTER ONE

INTRODUCTION

1.1 Background of the Study

Natural environment is continuously bombarded with ionizing radiation from both natural and man-made sources (Ademola, 2008; Chad-Umoren, 2012). The most common radionuclides in soil and groundwater are the radioactive isotopes of the three natural decay series (^{238}U , ^{235}U , ^{232}Th) and ^{40}K . Natural sources of radiation constitute almost 80% of the collective radiation exposure of the World's population (UNSCEAR, 2000). Human beings are exposed naturally due to sources outside their bodies, mainly cosmic ray and gamma ray emitters in soil, water, food and air, etc (Agbalagba et al, 2012). Some artificial radionuclides are also present in the environment as a result of testing of nuclear weapons, accidents such as the Chernobyl accident and the Japan nuclear power plant disaster, and the routine discharge of radionuclides from nuclear installations. Once they enter into the environment, these radionuclides whether natural or man-made are taken up by plants and animals, as a result, they find their way into the human body through the food chain. The natural radioactivity of soil and sediment/sludge depends on their formation and transportation processes that were involved since soil formation; chemical and biochemical interactions influence the distribution patterns of Uranium and Thorium and their progenies (ECNR, 1995).

Furthermore, certain industrial activities such as crude oil exploration result in enhanced ionizing radiation in the environment. Ionizing radiations such as alpha (α), beta (β) and gamma (γ) radiations are often found in the petroleum matrix due to both contaminations by radionuclides in the earth's crust and the materials used in the drilling process (Chad-Umoren,

2012; Laogun et al., 2006). Gamma rays are highly penetrating and are products of the radioactive materials containing radon. These substances may be ingested or inhaled thereby exposing both the hydrocarbon industry personnel and members of the host communities to increase in the risk of lung cancer, eye cataracts and mental imbalance (Laogunet *al.*, 2006).

A strong correlation has been found between oil and gas activities and elevated environmental ionizing radiation (Avwiriet *al.*, 2007a; Avwiriet *al.*, 2007b; Chad-Umoren, 2012; Chad-Umoren and Briggs- Kamara, 2010; Ononugboet *al.*, 2011) which are attributed to the industries' input raw materials and effluents discharge such as gas flaring and other output products. Radiation monitoring is of primary importance for environmental protection purposes (El-Bahi, 2004).

The presence of heavy metals in the environment is of great ecological significance due to their toxicity at certain concentrations, translocation through food chains and non-biodegradability which is responsible for their accumulation in the biosphere (Aekolaet *al.*, 2008). Heavy metals like iron, tin, copper, manganese and vanadium occur naturally in the environment and could serve as plant nutrients depending on their concentrations. Mercury, lead, cadmium, silver, chromium and many others that are indirectly distributed as a result of human activities could be very toxic even at low concentrations. These metals are non-biodegradable and can undergo global ecological circles (Aekola et al., 2008).

1.2 Statement of the Problem

Apart from geophysical and geological factors, some human activities can also enhance the natural radiation background levels (Rose *et al.*, 1995). These activities include burning of fossils,

mining and milling operations (Saleh *et al.*, 2007). These operation brings large amount of otherwise buried materials containing Naturally Occurring Radioactive Materials (NORM). By ingesting and inhaling the radionuclides in the NORMS or by just staying close to large volume of NORM, people are inadvertently exposed to enhanced level of radiation which can result in health hazard and risk (Saleh *et al.*, 2007; Turhan and Gunduz, 2007).

In the hydrocarbon industry, oil spillage, gas flaring and drilling activities are believed to raise the natural background radiation of the environment (Sigalo and Briggs- Kamara, 2004).

Also, it has been reported that naturally occurring radioactive materials (NORMS) associated with oil and gas production contain radioactive uranium, thorium and their progenies Ra-226 and Ra-228 (Abison, 2001; Avwiriet *al.*, 2007a; Chad-Umoren, 2012).

Kaduna Refining and Petrochemical Company (KRPC) was established in 1974 as third refining company in Nigeria and is situated relatively far from the residential area (Ahmad Hadiza A. 2014). But due to the continuous increased in population, it has now been surrounded by settlements which includes Banks, Farming site, Houses. Based on the report by Sigalo and Briggs- Kamara (2004), KRPC may likely be vulnerable to oil spill pollution, oil related effluent discharges, gas flare and other anthropogenic activities, which may enhance the natural radioactivity level of the area, thereby exposing both hydrocarbon industry personnel and members of the public.

1.3 Aim and Objectives

This study is aimed at assessing the radiological and heavy metals composition within Kaduna Refining Petrochemical Company Nigeria.

Objectives of the Present Study

- i. To measure the gamma radiation dose level in the study area.
- ii. To determine the activity concentration of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs and their radiological implications
- iii. To determine some heavy elements contained in the plant and soil samples.
- iv. To estimate the pollution load index (PLI).
- v. To calculate the Contamination Factors (CF) for the assessment of level of pollution involved

1.4 Justification of the Study

Estimation of radiological hazard Indices through the determination of environmental radioactivity concentration will help in environmental monitoring and protection, strategic planning as well as protection of human health. The data generated in this study will provide an up to date knowledge of radioactivity and heavy metal concentrations in the study area. This data can further be utilized in future planning by the government against environmental pollution and potential threats to human health due to radon and other forms of radionuclides.

CHAPTER TWO

LITERATURE REVIEW

2.1 Concept of Radiation and Radioactivity

Radiation is a form of energy, such as light or heat. Radiation is characterized according to the frequencies and wavelengths. Infrared waves, microwaves, and radio waves occur at the lowest frequencies. Ionizing radiation occurs at the highest frequencies. The process of transforming a neutral atom or molecule into an electrically charged component is known as ionization. Ions are either positively charged or negatively charged, depending upon the number of protons and electrons present in the atom.

Ionizing radiation is produced as the result of the disintegration of an unstable isotope. These isotopes can decay in several ways. The ratio of neutrons to protons in a stable, light atom is approximately unity. This ratio can increase to about 1.5 in heavier elements. Unstable nuclei exist because of an imbalance in this ratio. The interaction of this ionizing radiation with specific biological material will result in the production of ion pairs. An ion pair is essentially an ionized atom together with its ejected electron. Therefore, ionizing radiation will affect the fundamental structures of biological materials through the production of these ion pairs (Fattibene *et al.*, 1999). Several different species of radionuclides will produce ionizing radiation. Heavy elements, which include radionuclides such as ^{238}U and ^{232}Th , are found in all common types of rock and soil. Furthermore, these radionuclides disintegrate into other nuclides that are also radioactive, thus forming a decay chain. Therefore, not only is the release of ionizing radiation cumulative over the existence of the lead radionuclide in a decay chain, but additive throughout all the energy releases from the progeny in the decay chain as well. Products formed in a nuclear reactor as the result of

nuclear fission are radioactive. Other radioactive isotopes can be produced by particle bombardment processes, such as neutron activation (Akers *et al.*, 1994).

2.2 Types of Ionizing Radiation

Ionizing radiation can be classified into two types namely; particle radiation and electromagnetic radiation.

2.2.1 Particle radiation

Particle radiation includes: alpha particles, Beta particles, Electrons, Muons, Neutrons, Mesons, Fission fragment and heavy ions. They cause direct ionization of materials they interact with except neutrons that caused indirect ionization because of its neutrality. The magnitude of ionization of this type of radiation depends largely on the magnitude of charge on the particle as well as the linear energy transfer (Knoll, 2010).

2.2.2 Electromagnetic radiation

Electromagnetic radiation constitutes a spectrum of radiation that ranges from very long to very short wavelength. The radiation of interest (capable of causing enormous ionization) are X and gamma rays. Gamma rays are energetic ($>10\text{keV}$) electromagnetic radiations produced by radioactive decay or other nuclear or subatomic processes. Although of the same nature, x-rays and gamma rays are termed x and γ due to their different origin: the first are due to energetic electron processes, the latter are produced by transitions within the atomic nuclei. Two sources of gamma rays are commonly distinguished; gamma rays following beta or alpha decay and gamma-rays following nuclear reactions. The first source is not a primary decay processes but usually accompanies alpha and beta decay. Typically, this type of radiation arises when the daughter

product resulting from alpha or beta decay is formed in an excited state. This excited state returns very rapidly ($< 10^{-9}$ s) to the ground state through the emission of a gamma photon (Dendy, 1999).

2.3 Sources of Radionuclide

Exposure to ionizing radiation originates from two major sources: naturally occurring and manmade sources

2.3.1 Natural sources

Apart from stable chemical elements, very low concentrations of radioactive elements occur naturally in the environment. Natural radionuclides are divided into three categories according to their origin and formation: primordial radionuclides, secondary radionuclides, and cosmogenic radionuclides (Magill, 2003).

2.3.1.1 Primordial radionuclides

Primordial radionuclides are radionuclides that originated with other (stable) nuclei in the course of cosmic nucleogenesis by thermonuclear reactions in the core of a star, which then exploded as Supernova and enriched the nucleus cloud from which the sun and the solar system originated. They became part of the Earth at the time when the solar system was formed about 4 to 5 billion years ago. To the present day, only radionuclides with a very long half-life (i.e., more than about 10^8 years) were preserved. The most widespread primordial radionuclide is ^{40}K with a half-life of 1.277×10^{10} years, disintegrates by β decay to ^{40}Ar (89%) and by electron capture to ^{40}Ca (11%); both isotopes are stable.

Another natural primary radionuclide is ^{232}Th , which has a half-life of 1.4×10^{10} years and gradually disintegrates by a decay into a number of radionuclides of the so-called thorium decay

chain (i.e., secondary radionuclides). However, the most important natural radionuclides of primary origin in the Earth's crust are ^{238}U , with a half-life of 4.468×10^9 years, and ^{235}U with a half-life of 7.038×10^8 years. Both of these isotopes of uranium are gradually transformed by α decay into a number of radionuclides of both uranium decay chains.

2.3.1.2 *Secondary radionuclides*

The decay of primary radionuclides continuously gives rise to a number of secondary radionuclides. Natural radionuclides ^{232}Th , ^{238}U , and ^{235}U decays by alpha (α) and beta (β) decay into nuclei, which are also radioactive, much like their other decay products (i.e., radioactive decay chains). In nature there are three radioactive decay chains: ^{232}Th , ^{238}U , and ^{235}U . To a certain extent these three natural decay chains are similar. Radon (the most stable isotope is ^{222}Rn) appears in the second half of the series; its decay products have a short half-life and disintegrate simultaneously by α and β decay.

Radon is a radioactive noble gas, one of the heaviest gases. All three natural decay chains result in stable isotopes of lead.

2.3.1.3 *Cosmogenic radionuclides*

Cosmogenic radionuclides are natural radionuclides that currently originate by nuclear reactions when high-energy cosmic radiation passes through the Earth's atmosphere. Examples include radiocarbon (^{14}C) and tritium (^3H).

2.3.1.4 *Artificially produced radionuclides*

For the demands of present Science and Technology, industry, and health services, these few radionuclides of natural origin are far from sufficient. Therefore we must produce radionuclides

artificially. Artificial radionuclides can be produced by nuclear reactors, by particle accelerators, or by radionuclide generators.

2.4 Interactions of Radiation with Matter

The most important interactions of radiation with matter are interactions of alpha (α) or beta(β) particles and gamma (γ) photons with electrons of an atom. α and β particles are classed as ionizing particles. This is because they carry an electric charge that causes the atoms to separate into ions. Each separation creates an ion pair. γ rays are said to be indirectly ionizing radiation (Knoll 2010).

2.4.1 Interaction of gamma rays with matter

When a beam of gamma ray photon is incident on any material it removed individually in a single event. The event may be an actual absorption process, in which case, photon disappears or the photon may be scattered out of the beam. When a gamma rays passes through matter, probability for absorption is proportional to thickness of the layer, the density of the material, and absorption, cross section of the material. The total absorption shows an exponential decrease of intensity with distance from the incident surface.

$$I(x) = I_0 e^{-\mu x} \dots \dots \dots (2.1)$$

where, x is the distance from the incident surface,

$\mu = n\sigma$ is the absorption coefficient, measured in cm^{-1}

n is the number of atoms per cm^3 of the material (atomic density),

σ -is the absorption cross section in cm^2

Three processes are mainly responsible for absorption of γ - rays. These are as follows

1. Photoelectric Effect 2. Compton Effects 3. Pair Production

Which of these processes contributes the most is mainly dependent on the atomic number (Z) of the material and the energy (E) of the photon. The predominant mode of interaction of gamma rays with matter depends on the energy of incident photons and the atomic number of the material with which they are interacting. At low energies and with high Z materials the photoelectric effect is main interaction process. At intermediate energies and in low Z materials, the Compton scattering is dominating. At very high energies pair production is the most dominant interaction process.

2.4.1.2 *Photoelectric effect*

The photoelectric effect, in which the photon disappears, is an interaction between a photon and a tightly bound electron whose binding energy is equal to or less than the energy of the photon. The primary ionizing particle resulting from this interaction is the photoelectron, whose energy is given by

$$E_{pe} = h\nu - \Phi \dots \dots \dots (2.2)$$

The photoelectron dissipates its energy in the absorbing medium mainly by excitation and ionization. The binding energy Φ is transferred to the absorber by means of the fluorescent radiation that follows the initial interaction. These low-energy photons are absorbed by outer electrons or in other photoelectric interactions not far from their points of origin. The photoelectric effect is favored by low-energy photons and high-atomic-numbered absorbers (Cember, 1996).

2.4.1.3 *Compton effect*

Characteristic of the Compton Effect is that only part of its total amount of energy is transferred from the entering photon to an electron. The freed electron, which is called Compton electron

(recoil electron), reaches a certain velocity that is dependent on the energy transferred to the electron. The rest of the energy continues as a photon of lower energy in another direction, and is therefore called a scattered photon. Because of the lower energy the scattered photon has a longer wavelength than the original (Knoll 2010).

The Compton process occurs only then when the photon energy passes the limiting value of the photoelectric process. Since the impulse and the energy are divided among the Compton electron and the scattered photon, the law of preservation of impulse is complied with, and the process occurs with the electrons from the outer shells as well. For this reason, the atomic number (Z) of the material is less influential. The freed Compton electrons can, depending on the energy content, ionize other atoms along their routes. The scattered photon continues its way and continues to enter into Compton processes up until the energy is reduced to such an extent that a photoelectric process takes place. Only then the photon has disappeared.

Because the electron binding energy is very small compared to the gamma ray energy, the kinetic energy of electron is nearly equals to the energy lost by the gamma

$$E_e = E_\gamma - E' \dots \dots \dots (2.3)$$

where, E_e – energy of scattered electrons

E_γ - Energy of incident of gamma ray

E' - energy of scattered of gamma ray

2.4.1.4 *Pair production and Annihilation*

With photon energies larger than 1.022 MeV pair production may occur as an alternative to the Compton process. When such a high energetic photon comes close to a nucleus, transformation of

energy into mass can occur because of the electric field of the nucleus. With this the photon is converted into an electron and a positron with the same mass, but the reverse charge. If the photon energy is, for example, 2 MeV, $2 \times 0.511 = 1.022$ MeV goes to the electron-positron pair and the remainder (0.978 MeV) is divided as kinetic energy among the electron and the positron. In this process, in which the original photon disappears completely, the surplus of impulse is transferred to the nucleus. Summarizing, it can be posed that a photon, in comparison with a β -particle, loses a large part of its energy in a long route of interaction, and eventually disappears completely. The penetrating ability of photons in matter is therefore a lot bigger than that of the β -particles. On its way through the matter a photon produces 'hot' electrons (Photo, Compton, and Pair forming electrons) which can cause ionizations. That is why photon radiation is called indirectly ionizing.

2.5 Biological Effect of Ionization Radiation

The fact that ionizing radiation produces biological damage has been known for many years. The first case of human injury was reported in the literature just a few months following Roentgen's original paper in 1895 announcing the discovery of x-rays. As early as 1902, the first case of x-ray induced cancer was reported (ICRP, 1991). The long - term biological significance of smaller repeated doses of radiation, however was not appreciated until relatively recently and most of our knowledge of the biological effects of radiation has been accumulated since World War II (ICRP, 1991).

The types of injury attributable to ionizing radiation are subdivided, for purposes of risk assessment and radiological protection, into two broad categories: stochastic effects and non-stochastic effects. Stochastic effects are viewed as probabilistic phenomena, varying in frequency but not severity as a function of the dose, without any threshold; non-stochastic effects are viewed as deterministic phenomena, varying in both frequency and severity as a function of the dose, with

clinical thresholds. Included among stochastic effects are heritable effects (mutations and chromosome aberrations) and carcinogenic effects. Both types of effects are envisioned as unicellular phenomena which can result from non-lethal injury of individual cells, without the necessity of damage to other cells.

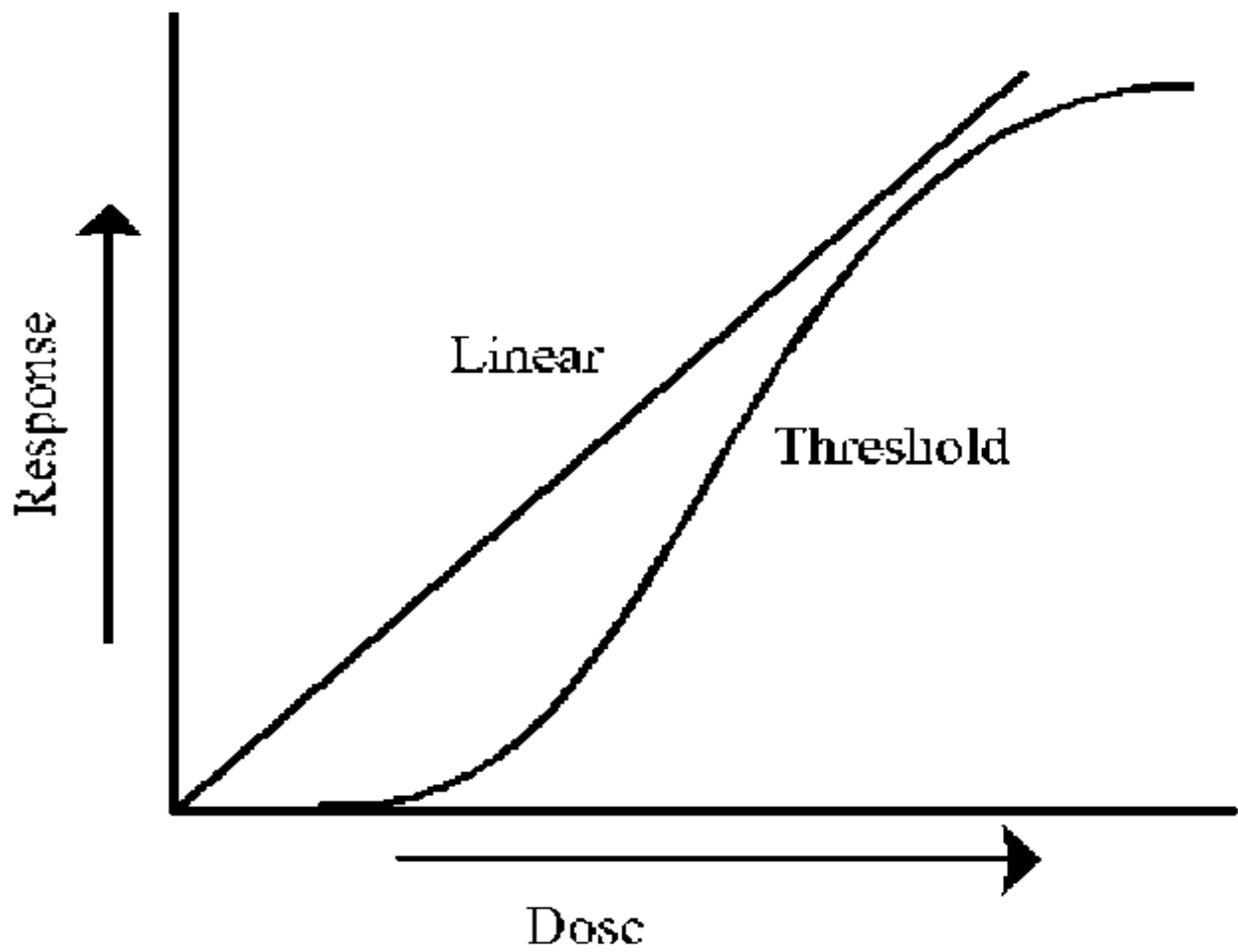


Fig. 2.0: Dose response Relationship (ICRP, 1991).

For the induction of mutations and chromosome aberrations in the low to intermediate dose range, the dose-response curve with high Linear Energy Transfer (LET) radiation generally conforms to a linear non-threshold relationship and varies relatively little with the dose rate. While for cataract of the lens, impairment of fertility and atrophy of blood forming organs in the high dose range, the dose-response curve is represented by the threshold relationship as shown in Figure 2.0. In contrast, the curve with low-LET radiation generally conforms to a linear-quadratic relationship, rising less steeply than the curve with high-LET radiation and increasing in slope with increasing dose and dose rate. The dose-response curve for carcinogenic effects varies widely from one type of neoplasm to another in the intermediate-to-high dose range, in part because of differences in the way large doses of radiation can affect the promotion and progression of different neoplasms. Information about dose-response relations for low-level irradiation is fragmentary but consistent, in general, with the hypothesis that the neoplastic transformation may result from mutation, chromosome aberration or genetic recombination in a single susceptible cell.

If the amount of radiation involved is large enough, acute doses may result in effects which can manifest themselves within a period of hours or days. Here the latent period of time elapsed between the radiation insult and the onset of effects is relatively short and grows progressively shorter as the dose level is raised. When the radiation is delivered to the whole body, the signs and symptoms which comprise these short-term effects are known as the Acute Radiation syndrome (ICRP, 1991). The long-term effects of radiation are those that manifest years after the original exposure. The latent period then is much longer than that associated with the acute radiation syndrome. From the standpoint of public health significance, the possibility of long-term effects on the large number of people receiving low and chronic exposures cause greater

concern than the short-term radiation effects from acute exposures which involve only a few individuals.

2.6 Radioactivity

Radioactivity is defined in accordance to the radioactive decay law (Turner, 1995). According to this law: The rate of disintegration of a radioactive atom N_t is proportional to the original number of atom N_0 , or alternatively in terms of activity; the activity of a radioactive nuclei A_t is directly proportional to the original activity A_0 of the nuclei. It is expressed as:

$$\frac{dN_t}{dt} \propto N_0 \text{ or } A_t \propto A_0 \dots \dots \dots (2.4)$$

where: N_t = Number of radioactive atom remaining at time t.

N_0 = Original number of nuclei.

A_t = The activity of the radioactive atom at a time t.

A_0 = original activity of the nuclei.

In radioactivity, the nucleus of an unstable atom is spontaneously transformed into a more stable one accomplished by different mechanisms, some of which include: alpha (α) particle emission, beta negative (β^-) (or negatron) and positive (β^+) emission and orbital electron capture (Cember, 1996). Each of these mechanisms may or may not be accompanied by gamma radiation as the spontaneous nuclear transformation of an unstable nucleus into more stable ones takes place. As presented by (Cember, 1996), the two factors that determine the exact mode of radioactive transformation are:

- (i) The mass-energy relationship among the parent nucleus, daughter nucleus and the emitted particle.
- (ii) The particular type of nuclear instability that is whether the neutron to proton ratio is either too high or too low for the type of nuclide consideration.

The substances whose nuclei undergo these transformation are to be radioactive and the

Transformation process is known as radioactive decay. The

Elements of radiation mentioned above as alpha particle, beta particle, and gamma rays carry energy along, which they deposit on matter they impinge. Alpha apart from depositing their energies, they also ionize the different media they interact with either directly or indirectly. The amount of energy absorbed (or deposited) per unit mass on any matter they impinge is called the absorbed dose measured in gray (Gy) or Joule per kilogram.

2.6.1 Serial radioactive decay and decay equilibrium

A serial radioactive decay is produced when one or more radionuclides are produced in a chain (Turner, 1995), suppose that a parent radionuclide A, having half-life T_a and decay constant λ_a , decays to a daughter radionuclide B of half-life T_b and constant λ_b , which in turn decays to a radionuclide C as illustrated below:



The rate of change, $\frac{dN_b}{dt}$ in the number of daughter atoms, N_b , per unit time equals the rate $\lambda_a N_a$ at which they are produced minus their decay rate. by applying radioactive decay equation, we then have the equation showing the number of daughter atoms as follows:

$$N_b = \frac{\lambda_a N_{0,a}}{\lambda_b - \lambda_a} (e^{-\lambda_a t} - e^{-\lambda_b t}) \dots\dots\dots (2.6)$$

In a particular case where the parent, A is long-lived and its daughter, B decays at a much faster rate such that $T_a \gg T_b$, a point is soon reached at which the instantaneous amount of A, that decays is equal to that of B. Under these conditions B is said to be in secular equilibrium with its parent A. (Cember, 1996).

As time t, increases beyond seven daughter half-lives (for practical purposes) when $\lambda_a \ll \lambda_b$, when $T_a \gg T_b$.

$$\lambda_b N_b = \lambda_a N_a \quad \text{or} \quad \frac{\lambda_a}{\lambda_b} = \frac{N_b}{N_a} \dots\dots\dots (2.7)$$

From the above equations, it is evident that at secular equilibrium, the activity of the daughter equals that of the parent, and that the ratio of the decay constant of the parent and daughter are in the inverse ratio of the equilibrium concentrations of the parent and daughter (Cember, 1996).

From the above it is possible to deduce from calculation the activity of particular long-lived parent like ^{232}Th by using the activity of its short-lived daughter ^{208}Tl when in secular equilibrium with it.

Turner (1995) argued that when the half-life, T_a of a parent is greater than that of the daughter, T_b but not greatly so (i.e, $T_a > T_b$ and not $T_a \gg T_b$), the activity of the daughter $\lambda_b N_b$ builds up steadily initially as the time, t increases, $e^{-\lambda_b t}$ eventually becomes negligible with respect to $e^{-\lambda_a t}$, since $\lambda_b > \lambda_a$. Knowing that the product of the number (N) of atoms in a substance and its decay constant is its activity, he then obtained

$$A_b = \frac{\lambda_b A_a}{\lambda_b - \lambda_a} \dots\dots\dots (2.8)$$

where A_a and A_b are respective activities of A and B measured Bq/kg.

Any initial rise (as shown in equation above) in the daughter activity, a corresponding fall in the activity of the parent activity is obtained at the same time. The form of equilibrium occurs after a maximum is attained in the activity of the daughter which clearly depicts another type of equilibrium which is the transient equilibrium (Turner, 1995). Further also, a situation can arise where no equilibrium occurs, principally because the activity of the daughter builds up and falls, i.e. $T_a < T_b$ indicating that the parent have a shorter half-life, it eventually decays away and only the daughter is left. (Turner, 1995).

2.6.2 Activity

The quality that characterizes the speed of radioactive decay is called activity (or emitter activity). The activity A is defined as the number of decays per unit time interval, or the loss in the number of undecayed nuclei per unit time interval:

$$A_t = -\frac{dN_t}{dt} = \lambda N_t \dots\dots\dots (2.9)$$

The unit of activity is the Becquerel (Bq), one of disintegration per second. The older unit of activity was the curie (1Ci = 3.7×10^{10} Bq).

2.6.3 Specific activity

The concentration of radioactivity or the relationship between the mass of radioactive material and the activity is called the specific activity. Specific activity is the number of Becquerel (or curies) per unit mass or volume (Cember, 1996).

$$SA = \frac{\lambda N}{Nm/N_a} = \frac{\lambda N_a}{M} \dots\dots\dots (2.10)$$

2.6.4 half life and mean life time

A clearer quantity used to characterize the radionuclide instead of the decay constant λ is half-life. The half-life $T_{1/2}$ is the time it takes for the number of atoms to decrease to half the initial value (i.e. $N/N_0 = e^{-\lambda t}$). Thus the half-life is related to the decay constant through the equation (Cember, 1996).

$$T_{1/2} = \frac{0.693}{\lambda} \dots \dots \dots (2.11)$$

Half – lives vary greatly among different types of atoms, from less than a second to billions of years. For example, it will take about 4.5 billion years for half of the atoms in a mass of ^{238}U to spontaneously disintegrate, but only 24,000 years for half of the atoms in a mass of ^{239}Pu to spontaneously disintegrate. In contrast ^{131}I , commonly used in medicine, has a half – life of only 8 days.

The average life is defined simply as the sum of the lifetime of the individual atoms divided by the total number of atoms originally present (Cember, 1996).

$$\tau = \frac{1}{\lambda} \dots \dots \dots (2.12)$$

If the expression for the transformation constant in terms of the half-life and mean life is found to be:

$$\tau = 1.44T_{1/2} \dots \dots \dots (2.13)$$

2.7 Radiation Protection

2.7.1 Objective of radiation protection

Man benefits greatly from the use of X-rays, radioisotopes, and fissionable materials in medicine, industry, research, and power generation. However, the realization of these gains entails the routine exposure of persons to radiation in the procurement and normal use of source as well as exposures from accidents that might occur. Since any radiation exposure presumably involves some risk to the individuals involved, the level of exposures allowed should be worth the result that is achieved. In principle, therefore, the activities that involve radiation protection are to balance the risks and benefits from activities involving radiation. If the standards are too lax, the risks may be unacceptably large; if the standards are too stringent, the activities may be prohibitively expensive or impractical, to the overall detriment to society. The balancing of risks and benefits in radiation protection cannot be carried out in an exact manner. The risks from radiation are not precisely known, particularly at the low level of allowed exposures, and the benefits are usually not easily measurable and often involve matters that are personal value judgments. Because of the existence of legal radiation-protection standards, in use everywhere their acceptance rests with society as a whole rather than with particular individuals or groups. Even if the risks from low-level radiation were established quantitatively on a firm scientific basis, the setting of limits would still represent a social judgment in deciding how great risks to allow. The setting of highway speed limits is an example of such a societal decision - one for which extensive quantitative data are available at the levels of risks actually permitted and accepted (turner, 1995)

2.7.2 Dosimetric quantities

2.8.2.1 Equivalent Dose

The equivalent dose, $H_{T,R}$ in a tissue or organ T due to radiation R, is defined as the product of the average absorbed dose $D_{T,R}$ in T from R and a dimensionless radiation weighing factor for each radiation (ICRP, 1991).

$$H_{T,R} = W_R D_{T,R} \dots \dots \dots (2.14)$$

The values of W_R specified by the NCRP are in Table 2.1. (The values recommended by the ICRP are the same, except that $W_R = 5$ for protons in the next to last entry). When the radiation consists of components with different W_R , then the equivalent dose in T is given by summing all contributions: $H_{T,R} = W_R D_{T,R}$ with $D_{T,R}$ expressed in Gy ($\text{Gy} = 1 \text{ J kg}^{-1}$), $H_{T,R}$ and H_T are in Sv ($1 \text{ Sv} = 1 \text{ J kg}^{-1}$). The equivalent dose replaces the dose equivalent for a tissue or organ. The two are conceptually different. Whereas dose equivalent in an organ is defined as a point function in terms of the absorbed dose weighted by a quality factor everywhere, equivalent dose in the organ is given simply by the average absorbed dose weighted by the factor W_R . For radiation types and energies not included in the Table 2.1, the ICRP and NCRP give a prescription for calculating an approximate value of W_R as an average quality factor, Q. For this purpose, the quality factor Q is defined in terms of the Linear Energy Transfer (LET).

Table 2.1 : Radiation Weighting Factors W_R

Radiation	W_R
X and γ rays, electrons, positrons and muons	1
Neutrons, Energy <10kev	5
Neutrons, Energy 10kev-2Mev	20
Neutrons, Energy >2Mev-20Mev	10
Neutrons, Energy >20Mev	5
Protons other than recoil proton & energy >2Mev	2
Alpha particles, Fission Fragments and non-relativistic heavy nuclei	20

(ICRP, 1991)

2.7.2.2 Effective Dose

Since different tissues of the body respond differently to radiation, the probability for stochastic effects resulting from a given equivalent dose will generally depend upon the particular tissue or organ irradiated. To take such differences into account, the ICRP and NCRP have assigned dimensionless tissue weighting factors W_T which add to unity when summed over all tissues T. The equivalent H_T in a given tissue weight by W_T gives a quantity that intended to correlate with the overall detriment to an individual, independently of T. The detriment includes different mortality and morbidity risks for cancers, severe genetic effects, and the associated length of lost. Table 2.5 implies, for example, that an equivalent dose of 1 mSv to the lung entails the same overall detriment for stochastic effects as an equivalent dose to the thyroid of $(0.12/0.05) \times (1 \text{ mSv}) = 2.4 \text{ mSv}$.

Table 2.2: Tissue Weighting Factors W_T ,

Tissue or organ	W_T
Gonads	0.20
Bone marrow	0.12
Stomach	0.12
Lung	0.12
Bladder	0.05
Breast	0.05
Liver	0.05
Esophagus	0.05
Thyroid	0.05
Skin	0.01
Bone surface	0.01
Remainder *	0.05

Note: The data refer to a reference population of equal numbers of both sex and a wide range of ages (ICRP, 1991).

In the definition of effective dose, they apply to workers, to the whole population, and to either sex. The W_T value is based on rounded values of the organ's contribution to the total detriment.

The risk for all stochastic effects for an irradiated individual is represented by the effective dose, E, defined as the sum of the weighted equivalent doses over all tissues: Like $H_{T,E}$ is expressed by Sv. The risk for all stochastic effects is dependent only on the value of the effective dose, whether or not the body is irradiated uniformly. In the case of uniform, whole body irradiation, HT is the same throughout the body. Then since the tissue weighing factors sum to unity, the value of the equivalent dose everywhere. The effective dose replaces the earlier effective dose equivalent. The latter quantity was defined the same way as E with H_T being the organ or tissue dose equivalent. The values of W_T are simplified and rounded for a reference population of equal numbers of males and females over a wide range of ages. As stated in ICRP 1991 report No. 116 (p.22), they “should not be used to obtain specific estimates of potential health effects for a given individual”

2.7.2.3 Committed equivalent dose

When a radionuclide is taken into the body, it can become distributed in various tissues and organs and irradiate them for some time. For the single intake of a radionuclide at time t_0 , the committed equivalent dose over a subsequent time in an organ or tissue T is defined as

$$H_T(\tau) = \int_{t_0}^{t_0+\tau} H_T(t) dt \dots\dots\dots (2.15)$$

where H_T is the equivalent dose rate in T at time t. Unless otherwise indicated, an integration time $\tau = 50$ y after intake is implied for occupational use and 70 y for members of the public.

2.7.2.4 *Committed effective dose*

By extension, the committed effective dose $E(\tau)$ following the intake of a radionuclide is the weighted sum of the committed equivalent doses in the various tissues. The effective half – life of a radionuclide in a tissue is determined by its radiological half – life and its metabolic turnover rate. For radionuclides with effective half – lives of no more than a few months, the committed quantities are practically realized within one year after intake. If a radionuclide is retained in the body for a long time, then the annual equivalent and effective doses it delivers will be considerably less than the committed quantities. The committed effective dose replaces the earlier committed effective dose equivalent.

2.8 Characteristics, and Transport of Heavy Metal Contamination in the Soil.

2.8.1 Characteristics of heavy metal contamination in soil

- (1) **Wide distribution:** With the development of economy and society, heavy metal contamination has become increasingly common in the world. It is almost a serious threat to every country. In the world's top ten environmental events, two events have related to heavy metal contamination (Yang and Sun, 2009).
- (2) **Strong latency** Heavy metal contamination is colorless and odorless, so it is difficult to be noticed. It does not explicitly damage the environment in a short period. Nevertheless, when it exceeds the environmental tolerance, or when environmental conditions have changed, heavy metals in the soil may be activated and cause serious ecological damage. So heavy metal contamination is usually chemical Time Bombs (CTBs) (Wood, 1974)
- (3) **Irreversibility and remediation hardness:** If the air and water are polluted, the pollution problem can be reversed certainly by dilution and self-purification after switching off the sources of pollution. However, it is difficult to use dilution or self-purification techniques

to eliminate heavy metal contamination and to get soils improved. Some soils contaminated by heavy metals are likely to take one or two hundred years to be remediated (Wood, 1974). Therefore, heavy metal contamination needs relatively high cost of remediation and the remediation cycle is relative long.

2.8.2 Heavy metal transport

Generally, heavy metals enter into the body system through air, food and water and bioaccumulate over a period of time (Lin, 1998). Bioaccumulation means an increase in the concentration of a chemical in a biological organism over time, compared to the chemical's concentration in the environment (Lin, 1998). Emission of heavy metals to the environment occur via a wide range of processes and pathways, including to the air (e.g. during combustion, extraction and processing), to surface waters (via runoff and releases from storage and transport) and to the soil (and hence into ground waters and crops) (Lin, 1998). Certain contaminants move through the air and deposited as dust or by precipitation. Heavy metals enter the aquatic environment through atmospheric deposition. They usually remain either in soluble or suspension form and finally tend to settle down to the bottom or are taken up by organisms (Wuana and Okieimen, 2011). Sediments are hosts of toxic metals and can therefore allow for the detection of heavy metals that may be either absent or in low concentration in the water column ((Lin, 1998). The accumulated heavy metals in the sediments can remain present for many years. Also chemicals may be carried by winds and deposited on the surface of soils (Wuana and Okieimen, 2011).

2.9 Instrumentation

2.9.1 Measurement of natural radioactivity in environmental samples

There are many different types of instruments available for measuring ionizing radiation in samples. Some of the instruments include: gas filled detectors (ionization chamber counters, proportional counters, and Geiger-Muller counters); scintillation counters; and solid state detectors (semiconductor detectors). The basic requirement of the instruments is that, the radiation interacts with the detector in such a manner that the magnitude of the instrument's response is proportional to the radiation effect or the radiation property being measured (Cember, 1996 and IAEA, 1989). For the detector to respond, the radiation must have undergone one of the following interactions: Photoelectric effect; Compton scattering; Pair production.

The result of the interaction in a detector is the appearance of a given amount of electric charge within the detector's active volume (Cember, 1996). Ionizing radiation (gamma rays) interacts with atoms in the sensitive volume of the detector to produce electrons by ionization. The collection of the electrons leads to an output pulse (signal). The energy required to produce ionization event in semiconductor detectors is 3.5 eV in contrast to the gas filled detectors which requires a mean high energy of 30-35 eV (Cember, 1996). A semiconductor is a substance that has electrical conducting properties midway between a conductor and an insulator. The most commonly used semiconductor materials are silicon and germanium. These elements belong to group IV of the periodic table implying each element has four (4) valence electron and will form crystal that consist of a lattice of atoms joined together by covalent bonds. Through the absorption of energy, the covalent bonds could be disrupted. Energy of 1.12 eV is required to knock out one of the valence electrons in silicon resulting in free electron and "hole" in the position formerly occupied by the valence electron. The free electron and hole can move about in

the crystal lattice. Electrons adjacent the hole can jump into the hole and leave behind another hole. If the semiconductor is connected in a closed electrical circuit, current will flow through the semiconductor (Cember, 1996). This implies that, the operation of the semiconductor radiation detector depends on having excess electrons or excess holes. A semiconductor with excess electrons is called n-type semiconductor, whilst the one with excess holes is called p-type semiconductor. These are achieved by adding an impurity to the crystal, either with excess number of electrons or an excess number of holes. If an atom of an element in group V such as phosphorus, arsenic, antimony is added to a pure silicon or germanium, four covalent bonds will be formed leaving behind an excess electron which is free to move about in the crystal and to participate in the flow of electric current. Under this condition, the crystal is of the n-type. On the other hand, p-type semiconductor is produced by adding an impurity from a group III such as boron, aluminum, gallium or indium which have three valence electrons. As a result, only three covalent bonds are formed in the crystal lattice. The deficiency of one electron results in a hole leading to the formation of p-type semiconductor detector (Cember, 1996). If a p region in silicon or germanium is adjacent to an n region, an n-p junction is created. If a forward bias is applied to the junction by connecting the p region to the positive terminal and the n region to the negative terminal, current will flow across the junction. However, if the polarity of the applied voltage is reversed, by connecting the n region to the positive terminal and the p region to the negative terminal, a condition known as reversed bias is achieved. Under this condition, no current will flow across the junction. The region around the junction is swept free by the potential differences created by the holes and electrons in the p and n regions. This region is known as the depletion layer and it is the sensitive volume of the solid-state detector.

Thus, when ionizing radiation passes through the depletion layer, electron-hole pairs are produced as a result of ionizing collisions between the ionizing radiations and the crystal. The electric field then sweeps the holes and electrons apart, giving rise to a pulse in the load resistor as the electrons flow through the external circuit. Semiconductor detectors are especially useful for nuclear spectroscopy because of their inherently high energy resolution.

Nuclear spectroscopy is based on the analysis of radioactive isotopes by measuring the energy distribution of the source. The spectrometer separates the output pulses from the detector according to size. The output of the spectrometer provides detailed information that is useful in identifying unknown radioisotopes. Nuclear Spectrometers are available in two types, either in Single Channel Spectrometer (SCA) or Multi-Channel Analyzer (MCA). The main use of the SCA is to discriminate between a desired radiation and other radiations that may be considered noise using a pulse height selector. On the other hand, MCA has an Analogue-to-Digital Converter (ADC) to sort out all the output pulses according to their energies. The MCA also has a computer-type memory for storing the information from the ADC. Most MCA are built with a number of channels varying by a factor of 2 over a range of 128 to 4096 each with a storage capacity of 10^5 to 10^6 counts per channel (Cember, 1996).

The basis for nuclear spectroscopy is the location of spectral lines arising from the total absorption of charge particles or photons. For this reason, the resolution of the detector is important if spectral lines closed together are to be separated and observed. Energy resolution may be viewed as the extent to which a detector is able to distinguish between two closely lying energies (radioisotopes). The formal definition of energy resolution is given in terms of the full width at half maximum (FWHM) divided by the location of the peak centroid, E , as in equation (2.16).

$$\text{Resolution} = \frac{\text{Full width at half maximum}}{E} \dots \dots \dots (2.16)$$

Gamma radiation can also be measured using a scintillation detector consisting of Sodium Iodide crystal activated with thallium [NaI (Tl)] and optically coupled to a photomultiplier tube. The thallium activator present as an impurity in the crystal structure to the extent of 0.2 % converts the energy absorbed in the crystal to light. Sodium Iodide (NaI) (Tl) detectors have higher efficiency than high purity germanium (HPGe) detectors because of the high density of the crystal and high effective atomic number (Cember, 1996).

The energy resolution of scintillation detectors [NaI (Tl)] is normally between 7- 9% for gamma radiation of energy of about 1 MeV whilst for semiconductor detectors (HPGe), the energy resolution is of the order of 0.1% (Cember, 1996). The smaller the value of the energy resolution, the better the detector's ability to resolve between two isotopes whose energies lie close to each other. Semiconductor detectors have better resolution than scintillation detectors.

2.9.2 Theory of atomic absorption spectroscopy (AAS)

Atomic Absorption Spectrometry is an analytical technique which is used to determine the concentration of metals in solution using Atomic Absorption Spectrophotometer. The equipment consists of the following components:

- A lamp compartment which contains hollow cathode lamps of the analyte of interest.
- Atomizing chamber which vaporizes and atomizes the sample in the flame transforming it into unexcited ground state atoms to absorb light at specific wavelength. The source of energy for the production of free atoms is usually heat commonly from an air/acetylene or nitrous – oxide/acetylene flame. Usually, the sample is introduced as an aerosol into the flame and the

burner aligned in the optical path so that the light beam passes through the flame, where the light is absorbed.

- An optical system which directs light from the source through the atom population into the monochromator. The monochromator isolates the specific analytical wavelength of the light emitted by the hollow cathode lamp from the other non-analytical lines including those of the fill gas.

- A photomultiplier tube to measure the light output accurately.

- The display of the results of the analysis.

2.9.2.1 *Principle of AAS*

The principle of operation of Atomic Absorption Spectrometer is based on the fact that, ground state metals absorb light at specific wavelength. The metal ions in solution are converted to atomic state by means of a flame. Light of appropriate wavelength is supplied and the amount of light absorbed is measured against a standard curve. The technique requires that a liquid sample is aspirated, aerosolized and mixed with combustible gases such as acetylene and nitrous oxide. The mixture is ignited in a flame with temperature in a range of 2100-2800 °C. During the combustion, atoms of the element of interest in the sample are reduced to free, unexcited ground state atoms which absorbs light at the characteristic wavelength. The characteristic wavelengths are element specific and this is determined when light beam from a lamp which consists of the element of interest is passed through the flame. A photomultiplier detects the amount of reduction of the light intensity due to absorption by the analyte which is directly related to the amount of element in the sample and the results displayed.

2.10 Heavy Metal Assessment Index

In the statistical technique, indices were applied to assess the contamination load in soil. There are two types of indices i.e., single index and integrated index. Integrated indices are the geo-statistical techniques applied on aggregate elemental contaminants rather than single elements pollution, but each of integrated indices could be obtained from single pollution load index as suggested by Qingjie and Jun (2008).

2.10.1 Contamination factor

Contamination factor is the quotient obtained by dividing the concentration of metals related to the target area by reference area. The contamination factor can be calculated through the following formula as suggested by Harikumaret al. (2009).

$$CF = C_n/B_n \dots\dots\dots (2.17)$$

In the above equation, C_n is the concentration of metals in the target area, and B_n is the metals concentration of the reference area. The classifications according to contamination factor results are as follows (Hakanson, 1980):

Table 2.3: Classification of contamination factor.

Contamination factor	Classification
CF<1	Low
1≤CF<3	Moderate
3≤CF<6	Considerable
CF≥6	Very high

2.10.2 Pollution load index

Pollution load index (PLI) is simple statistical technique used to determine elemental contents in soil beyond the reference concentration and provide comprehensive information about the metals toxicity in respective samples (Tomlinson *et al*, 1996). Pollution load index can be determined through the following formula:

$$PLI = \sqrt[n]{CF_1 * CF_2 * CF_3 * \dots * CF_n} \dots\dots\dots (2.18)$$

where PLI represents the pollution load index, CF is the contamination factor, and n is the number of elements. The $PLI > 1$ indicates polluted, while $PLI < 1$ indicates no pollution

2.11 Review of Related Works

Nada *et al.*, (2016) have done studies on the measurement of natural radioactivity from different location inside Al-Dora refinery, Iraq by using high purity germanium detector. For solid samples, the results have shown that the specific activity, for ^{238}U ranged from $(32.62 \pm 5.71 \text{ Bq/kg})$ to $(9.39 \pm 3 \text{ Bq/kg})$, for ^{232}Th was ranged from $(26.56 \pm 5.15 \text{ Bq/kg})$ to $(6.28 \pm 2.51 \text{ Bq/kg})$, for ^{40}K was ranged from $(277.59 \pm 16.66 \text{ Bq/kg})$ to $(51.22 \pm 7.16 \text{ Bq/kg})$, for ^{137}Cs was ranged from $(3.05 \pm 1.75 \text{ Bq/kg})$ to (B.D.L). But regarding the radiation hazard indices in (both solid and liquid), for Radium Equivalent Activity (Raeq) was ranged between $(5.21 - 79.84 \text{ Bq/kg})$, for Absorbed Gamma Dose Rate ($D\gamma$) was ranged between $(37.53 - 2.49) \text{ nGy/h}$, for indoor (AEDE) ranged from $(0.012 - 0.184)$ and for outdoor (AEDE) ranged from $(0.003 - 0.046)$ in mSv/y , for internal hazard index (H_{in}) ranged from $(0.025 - 0.079)$ and for external hazard index (H_{ex}) ranged from $(0.014 - 0.216)$, respectively. The study shows that all the studied sites in Al-Dora refinery do not pose any significant source of radiation hazard and are safe for use.

Ehsanpouret al., (2014) have evaluate the radiological quality of ^{226}Ra , ^{232}Th and ^{40}K in some samples of water resources collected in Anarak-Khour a deserted area, Iran has been measured by direct gamma ray spectroscopy using high purity germanium detector in. The concentration ranged from ≤ 0.5 to 9701 mBq/L for ^{226}Ra ; ≤ 0.2 to 28215 mBq/L for ^{232}Th and $< \text{MDA}$ to 10332 mBq/L for ^{40}K . The radium equivalent activity was well below the defined limit of 370Bq/L. The calculated external hazard indices

Onoja (2015), have done studies on environmental pollution around Kaduna Refining and Petrochemical Company (KRPC) and reported that the activity concentration of 3 gamma emitting radionuclides determined (^{226}Ra , ^{232}Th and ^{40}K) where in the range of 16.0-101.8($46.6 \pm 2.8 \text{ BqKg}^{-1}$), 13.9-103.2 ($63.6 \pm 1.4 \text{ BqKg}^{-1}$) and 51.8-257.2 ($165.9 \pm 6.1 \text{ BqKg}^{-1}$) respectively. Radium equivalent (Ra_{eq}) activities were all below the threshold value of 370 BqKg^{-1} ; while the hazard indices were all below unity.

Bashir et al (2014) assess the radiological risk in flooded soil of Kudenda, Kaduna State Nigeria. The results of the activity concentration of ^{235}U , ^{232}Th , and ^{40}K was found to be 81 ± 1.3 \leftrightarrow 4514.4, 38 ± 1.3 \leftrightarrow 149.6 \pm 3.9 and 400.5 ± 3.9 \leftrightarrow 873.7 \pm 11.6 Bqkg^{-1} respectively. The AEDE was found to vary from 0.06 ± 0.003 to $0.17 \pm 0.005 \text{ mSvy}^{-1}$ with arithmetic mean value of $0.11 \pm 0.004 \text{ mSvy}^{-1}$ which is higher than the world wide effective dose of 0.07 mSvy^{-1} .

Okeyodeet al (2011) determined the composition of copper, manganese, iron, zinc, lead, nickel, magnesium and sodium in the soil samples from some selected dumpsites and 20m away from the dumpsites within Abeokuta metropolis in Ogun state, Nigeria, using atomic absorptions spectrophotometer. At dumpsites, the concentrations of iron, manganese, zinc, lead, nickel, magnesium, sodium and copper ranged from 86.89 – 277.33mg/l, 1.9731 – 5.6644mg/l, 0.3299 –

4.847mg/l, 0.4132 – 2.8137mg/l, 0.0896 – 0.1376mg/l, 1.9731 – 5.6644mg/l, 0.9203 – 29.00mg/l and 0.1645 – 5.4191mg/l respectively. The concentrations of metals in solid at the decomposed biodegradable wastes dumpsites and 20m away from dumpsites showed that there is an evidence of relative increase in concentrations of metals in solids at dumpsites compared to those 20m away from dumpsites.

Agbalaba *et al.* (2012) analyzed the natural radioactivity levels and estimated the hazard indices, radium equivalent activities, representative level index, external and internal hazard index, absorbed dose rate and the effective dose rate in soil and sediment of ten oil and gas field in Delta state using gamma-ray spectroscopy. Results shows that the activity levels of the radionuclides showed enhanced activity concentrations across the area under study. The mean activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K were found to be 41.0 ± 5.0 , 29.0 ± 4.0 and 412.5 ± 20.0 Bq/kg respectively. The values obtained for ^{226}Ra and ^{40}K were slightly higher than the world average.

Kant *et al.* (2010) have done the studies on measurement of radium activity and radon exhalation rate and the reported activity concentration of ^{238}U , ^{232}Th and ^{40}K in various fly ash samples which varied from 99 ± 2 to 203 ± 4 , 145 ± 2 to 288 ± 4 , and 355 ± 5 to 516 ± 6 Bqkg⁻¹ respectively. The radium equivalent activity varied from 317 to 614 Bqkg⁻¹ with radon activity varied between 214 and 590 Bqm⁻³. The absorbed dose varied from 143 to 277 nGyh⁻¹. The indoor annual effective dose varied from 0.7 to 1.36mSv and the outdoor annual effective dose varied from 0.17 to 0.34 mSv. The authors also reports that the activity concentration of U-238, Th-232 and K-40 are below the permissible levels.

A study by Avwiriet *al.*, (2012) was carried out to determine some radiological parameters and radiation health hazard for Ogulogu-Olo, in Ezeagu Local Government Area and AmaguUmuene, in Udi Local Government Area of Enugu State, south-east Nigeria. The three natural radionuclides (^{40}K , ^{226}Ra and ^{232}Th) were found in the two boreholes that were studied. However their concentrations were below the respective world average values (UNSCEAR, 2000).

In view of these, this research is undertaken to assess the radiological and heavy metals concentrations in soil and plant inside Kaduna Refining and Petrochemical Company Nigeria

CHAPTER THREE

MATERIALS AND METHOD

3.1 Materials

The following materials were used to carry out the research work

Hand Held Geographical positioning system (GPS),

Masking Tape,

Polyethylene Bags

Portable radiation survey meter

Pulverizer,

Marinelli beaker

Mortar and Pestle

Hand gloves

HPGE Detector Canberra (Model GC80-23)

Atomic Absorption Spectrometer (model AA6800 shimazu, Japan)

3.2 Methodology

3.2.1 Study area

The study area is KRPC which is located at Chikun Local Government Area of Kaduna State, Nigeria. KRPC lies between latitudes $10^{\circ} 22'$ to $10^{\circ} 27'$ North and longitudes $7^{\circ} 25'$ to $7^{\circ} 31'$ East. The facility lies between latitudes $10^{\circ} 24'$ to $10^{\circ} 26'$ North and longitudes $7^{\circ} 29'$ to $7^{\circ} 31'$ East.

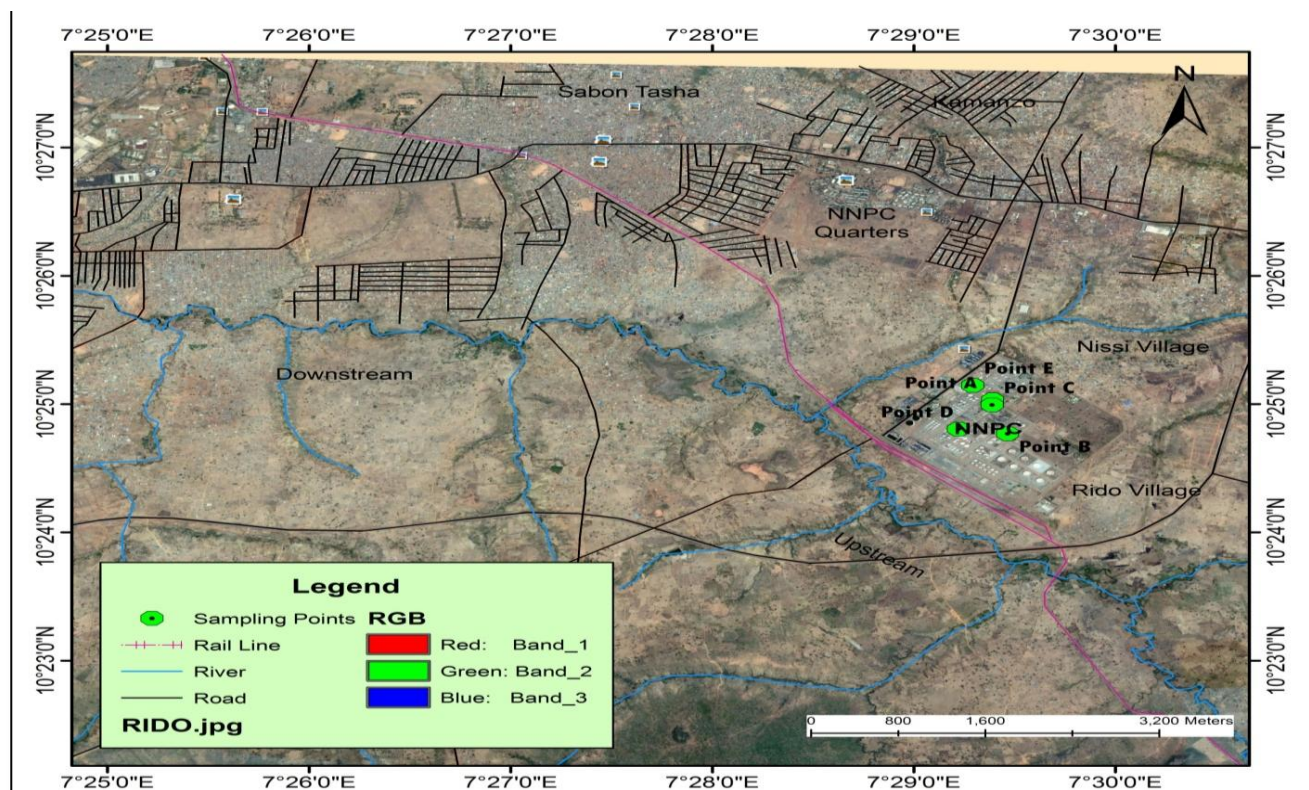


Plate 3.1: Study area showing sampling points

Source: GIS Lab Geography Department A.B.U. Zaria.

3.2.2 Measurement of gamma radiation dose rate

The gamma radiation dose levels in the study area were measured with a portable radiation survey meter. All measurements were made at one meter above the ground surface, as is the

standard practice (ICRU, 1994). The arithmetic mean (AM) of the readings was taken as representative value for each location. This procedure is aimed at reducing minor fluctuations of the exposure level within the site.

3.2.3 Sample collection

Two categories of samples (soil and plant) were carried out in five different location inside KRPC (Linear Alkyl Benzene Plant (A), Refinery site (B), Waste Deposit site(C), Asphalt production site(D) and Ware house(E)). For purpose of comparison with outside environment free from the activities going on in the study area sampling was made at a location outside the study area to serve as control area.

3.2.3.1 *Soil sample*

1kg of top soil were collected from three spots at each of the above mentioned locations. All the soil samples from each spot were mixed thoroughly as a composite sample that is representative for each location. The samples were then packed in a well labeled polythene bag.

3.2.3.2 *Plant sample*

In the case of plant sample mango leaves (*Mangifera indica*) have been collected from two mango trees close to the location identified for soil sample collection. The sample location was marked using a geographical positioning system (GPS) and all the samples were then taken to Center for Energy Research and Training (CERT) for processing.

3.2.4 Sample preparation

3.2.4.1 *Sample preparation for gamma spectroscopy*

At the laboratory, the samples were air dried in trays for 7 days. The samples were then grounded into a fine powder using a ball mill grinder and sieve through a 2 mm pore size mesh

into a previously weighed one (1) litre Marinelli beaker. The Marinelli beakers with its content were weighed again to determine the weight of the sample. The beakers were covered and sealed with a paper tape to prevent the escape of the gaseous radionuclides in the sample. The samples were stored for 30 days to attain secular equilibrium between the long-lived parent radionuclide and their short-lived daughter radionuclides in the ^{238}U and ^{232}Th decay series and counted on a high purity germanium (HPGe) detector at National Institute for Radiation Protection and Research, University of Ibadan.

3.2.4.2 *Sample preparation and analysis for atomic absorption spectroscopy*

All samples were air-dried at ambient laboratory temperature. Soil and plant samples were grounded using mortar and pestle and sieved to pass through 2 mm sieve and stored for chemical analysis. With the aid of spatula and weighing bottle, 0.5g of each soil and plant sample was obtained using a high sensitive chemical balance. This was placed in a Teflon beaker and transferred to a fume-cupboard for digestion. The digestion was carried out using concentrated nitric (10.0 mL) and concentrated hydrochloric (5.0 mL) acids in the ratio of 2:1 and the oven was maintained at 100 °C. After one hour, the mixture was allowed to cool before leaching the residue with 5.0 cm³ of 20% HNO₃. Digested samples were filtered and made up to 100 mL with deionized water. Solution of samples were then taken and aspirated into Atomic Absorption Spectrometer (model AA6800 shimazu, Japan) for analysis.

3.2.5 Experimental

3.2.5.1 *Gamma Spectroscopy*

With radiochemical, analysis it is not possible to separate isotopes of the same element. Many nuclides present in environmental matrix, emit beta and alpha particles as well as gamma rays. Gamma ray spectrometry offers a convenient analytical method for the identification of isotopes

due to discrete energy of gamma rays. The method is not only qualitative but also quantitative. In the present study HPGe gamma ray spectrometer was used for the identification and measurement of the activities of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs in plants and soil samples.

3.2.5.2 Experimental Set-Up

The detection and measurement of radionuclides in the samples were carried out by gamma ray spectrometry system using a vertical co-axial cylindrical High Purity Germanium (HPGe) detector of 172 cm^3 active volume and with 80% relative efficiency. The p-type HPGe detector supplied by CANBERRA (Model-GC80-23) had a resolution of 2.3 keV at 1.33MeV of Cobalt-60 gamma-ray line. The detector was coupled to a 16 k-channel analyzer. The spectra of all samples were perfectly analyzed using Genie-2000 spectra analysis software (which matched various gamma energy peaks to a library of all possible radionuclides) to calculate the concentrations of ^{238}U , ^{232}Th and ^{40}K and ^{137}Cs . The detector was enclosed in a cylindrical shielding container made of Lead and Iron with 11.3 cm thickness, 51 cm height and 28 cm internal diameter and having a fixed bottom and moving cover to reduce the external gamma-ray background. All the samples were counted for 10800s. Prior to the measurement of the samples, the environmental gamma background at the laboratory site was determined with identical plastic container used in the sample measurement.

3.2.5.3 Calibration of the Detector

The energy calibration of the germanium detector system was done by using some standard radioactive point sources with known energies. These sources should be counted for a long enough period in order to produce well-defined photo peaks and then calibrated according to their energies. In the present work, the point source (^{60}Co) was used in the system calibration.

The absolute efficiency calibration of the detector was determined using a multi gamma ray standard sources of one litter capacity Marinelli beaker. This multi-gamma ray radionuclides (MGS6M315) consists of the four radioelements: EU-155, Sb-125, Mn-55, Zn-64 and K-40 which have the same shape and size, were used to compute the efficiencies over a wide energy range. To undertake detector efficiency calibration, the spectra of standard radioactive sources with known energies and activities were accumulated for long enough time by the detector to produce well-defined photo peaks such as 86,400 seconds (24 hours).

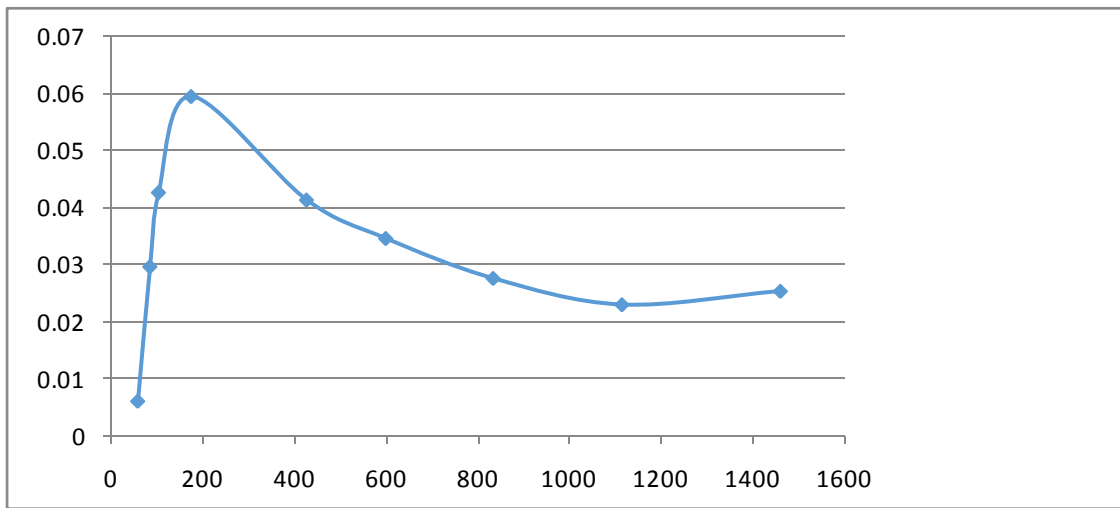


Fig 3.2: The Efficiency calibration curve of (HPGe) detector using multi-gamma ray standard sources.



Plate 3.1: Experimental Assembly

3.2.5.4 Determination of activity concentrations

The activity concentrations of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs in the soil and plants samples were calculated using the following analytical expression (Nafaa, 2006).

$$A = 1000 * \frac{\frac{N}{T}}{m\varepsilon P_{\gamma}} \dots \dots \dots (3.1)$$

where N is net counts of the photo peak, T is the live time of measurements (sec), m is the mass of the sample used (g), ε is the counting efficiency of the specific nuclide's energy and P_{γ} is the gamma transition probability through the specific energy.

3.4 Radiological Hazard Assessment

3.4.1 Absorbed dose rate in air (D)

In order to assess any radiological hazard, the exposure to radiation arising from radionuclide present in soil can be determined in terms of many parameters. A direct connection between

radioactivity concentrations of natural radionuclide and their exposure is known as the absorbed dose rate in the air at 1 meter above the ground surface. The mean activity concentrations of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K (Bqkg^{-1}) in the soil samples are used to calculate the absorbed dose rate given by the following formula.

$$D \text{ (nGy/h)} = 0.462A_U + 0.604A_{Th} + 0.0417A_k \dots \dots \dots (3.2)$$

Where D is the absorbed dose rate in nGyh^{-1} , A_U , A_{Th} and A_k are the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K , respectively. The dose coefficients in units of nGyh^{-1} per Bq.kg^{-1} were taken from the UNSCEAR (2000).

3.4.2 Annual effective dose equivalent (AEDE)

The absorbed dose rate in air at 1 meter above ground surface does not directly provide the radiological risk to which an individual is exposed (Jibiri et al; 2007). The absorbed dose can be considered in terms of the annual effective dose equivalent from outdoor terrestrial gamma radiation which is converted from the absorbed dose by taking into account two factors, namely the conversion coefficient from absorbed dose in air to effective dose and the outdoor occupancy factor. The annual effective dose equivalent can be estimated using the following formula (UNSCEAR, 2000 and Belivermis et al; 2010):

$$\text{AEDE } (\mu\text{Sv/y}) = D \text{ (nGy/hr)} \times 8760\text{h} \times 0.7\text{Sv/Gy} \times 10^{-3} \dots \dots \dots (3.3)$$

The values of those parameters used in the UNSCEAR report (2000) are 0.7 Sv.Gy^{-1} for the conversion coefficient from absorbed dose in air to effective dose received by adults and 0.2 for the outdoor occupancy factor (UNSCEAR, 2000).

3.4.3 Radium Equivalent Activity (Ra_{eq})

Due to a non-uniform distribution of natural radionuclide in the soil samples, the actual activity level of ^{238}U , ^{232}Th and ^{40}K in the sample was evaluated by means of a common radiological index named the radium equivalent activity (Ra_{eq}). It is the widely used index to assess the radiation hazards and was calculated using Equation 3.4 (Al-Hamarnech and Awadallah, 2009).

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K \dots\dots\dots(3.4)$$

where A_U , A_{Th} and A_K are the activity concentration of ^{238}U , ^{232}Th and ^{40}K in Bq.kg^{-1} , respectively. The permissible maximum value of the radium equivalent activity is 370 Bq.kg^{-1} (UNSCEAR, 2000) which corresponds to an effective dose of 1 mSv for the general public (Ajayi, 2009).

3.4.4 External and internal hazard indices (H_{ex} and H_{in})

To limit the radiation exposure attributable to natural radionuclide in the sample to the permissible dose equivalent limit of mSv.y^{-1} , the external hazard index based on a criterion have been introduced which is given by (Al-Hamarnech and Awadallah, 2009).

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \dots\dots\dots(3.5)$$

In order to keep the radiation hazard insignificant, the value of external hazard index must not exceed the limit of unity. The maximum value of H_{ex} equal to unity corresponds to the upper limit of radium equivalent activity 370 Bq.kg^{-1} (Al-Hamarnech and Awadallah, 2009).

In addition to external hazard index, radon and its short –lived products are also hazardous to the respiratory organs. So, the internal exposure to radon and its daughter products was quantified by the internal hazard index H_{in} which is given by;

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \dots\dots\dots(3.6)$$

CHAPTER FOUR

RESULTS AND DISCUSSION

4.1 INTRODUCTION

This chapter present the measured gamma radiation dose rate, evaluated activity concentration of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs and their radiological implication as well as the concentration of some heavy metals (Pb,Zn, Fe, Mn, Ni, Co, Cr and Cd)in soil and plant samples. Contamination factor and pollution load index were estimated to ascertain the level of pollution involved in the study area.

4.2 Gamma Radiation Dose Rate Measurement

The measured gamma radiation dose level ranges from 0.11 to 0.15 with a mean value of 0.12 $\mu\text{Sv/hr}$ in the target and 0.08 $\mu\text{Sv/hr}$ in the control area as presented in Table 4.1.The mean value obtained in the study area was found to be roughly twice that of the world average value of 0.056 $\mu\text{Sv/hr}$ (UNCEAR, 2000).

Fig 4.1 shows the variation of measured gamma radiation dose level in target and control areas. It is seen that the mean gamma radiation dose rate recorded in the target area were higher than that recorded in the control area. This could be due to anthropogenic activities such as gas flaring, oil spill pollution that is being carried out in the study area which are believed to enhance the background radiation level of the studyarea.

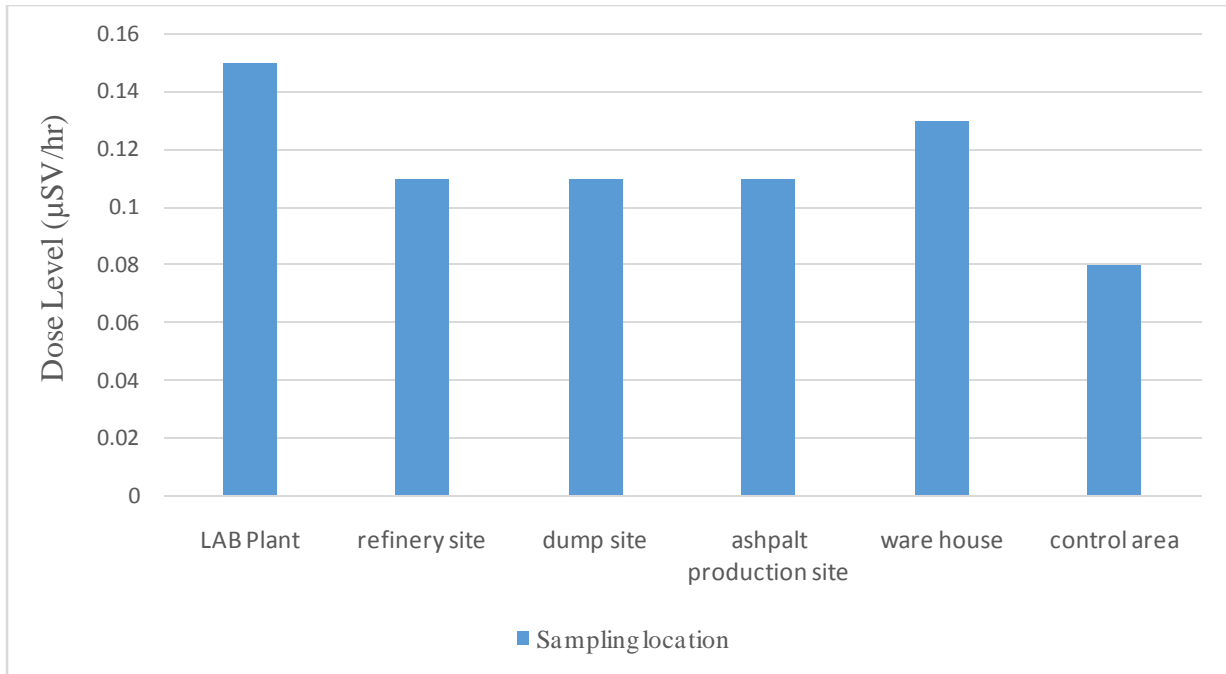


Figure 4.1. Gamma Radiation Dose level (µSv/hr) in the target and control areas.

Table 4.1 ; mean gamma radiation dose rate in study and control area

Location	Geographical Location	Gamma Dose rate ($\mu\text{Sv/hr}$)
Lab Plant	N10°25'02.0''	0.15
	E007°29'23.4''	
Refinery Area	N10°24'40.1''	0.11
	E007°29'27.9''	
Dump Site	N10°24'46.2''	0.11
	E007°29'44.2''	
AshpaltProd.Site	N10°24'48.3''	0.11
	E007°29'13.3''	
Ware House	N10°25'08.3''	0.13
	E007°29'17.5''	
Control Area	N10°26'09.3''	0.08
	E007°29'37.2''	
Mean (this study)		0.12
Worldave.(UNCEAR, 2000)		0.056

4.3 Activity Concentrations of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs

Tables 4.2 and 4.3 shows the specific activity concentrations in soil and mango leaves (*mangiferaindica*) samples collected at five different locations inside Kaduna Refining and Petrochemical Company Nigeria and control. The mean and range of specific activity concentrations for ^{238}U , ^{232}Th and ^{40}K in the soil samples were found to be $61.98 \pm 9.30 \text{ Bq/kg}$ with a range of 43.11 ± 6.18 (refinery site) to $76.005 \pm 10.86 \text{ Bq/kg}$ (LAB plant) for ^{238}U , $65.37 \pm 7.79 \text{ Bq/kg}$ ranging from 44.50 ± 6.37 (refinery) to $93.88 \pm 8.55 \text{ Bq/kg}$ (ware house) for ^{232}Th and $207.79 \pm 6.91 \text{ Bq/kg}$ ranging from 73.65 ± 4.58 (ware house) to $376.00 \pm 9.82 \text{ Bq/kg}$ (LAB plant) for ^{40}K respectively. Similarly the mean and range of specific activity concentrations for ^{238}U , ^{232}Th and ^{40}K in the mango leaves samples was found to be $18.73 \pm 4.258 \text{ Bq/kg}$ with a range of 13.73 ± 4.27 (LAB plant) to $30.17 \pm 6.44 \text{ Bq/kg}$ (dump site) for ^{238}U , $12.73 \pm 3.66 \text{ Bq/kg}$ ranging from 10.58 ± 5.11 (refinery) to $14.09 \pm 3.00 \text{ Bq/kg}$ (ware house) for ^{232}Th and $278.79 \pm 11.90 \text{ Bq/kg}$ ranging from 218.79 ± 9.94 (refinery site) to $346.09 \pm 13.17 \text{ Bq/kg}$ (LAB plant) for ^{40}K respectively. ^{137}Cs was not detected in all the samples collected from both target and control areas.

Fig 4.2-4.4 present the activity concentration of ^{238}U , ^{232}Th and ^{40}K for both soil and plant samples in targets and control area. It is seen that ^{40}K is dominant radionuclide in both soil and plant samples of target and control area. Furthermore, the mean specific activities of the radionuclides were compared with the world wide mean values of 35, 30, and 400 Bq/kg for ^{238}U , ^{232}Th and ^{40}K respectively (UNSCEAR, 2000). The mean specific activities of ^{238}U and ^{232}Th in both the target and control areas were above the world average values. The mean activity concentration of ^{40}K for both target and the control area were below the world mean values. When compared with other national and international studies, the mean activity concentrations of

^{238}U , ^{232}Th and ^{40}K obtained from the present study were found to be higher than the results obtained by (Onoja, 2015) and (Nada *et al*, 2016) as shown in Table 4.4. This variations could be due to activities that is being carried out in the study area or differences in geological conditions (UNCEAR, 2000).

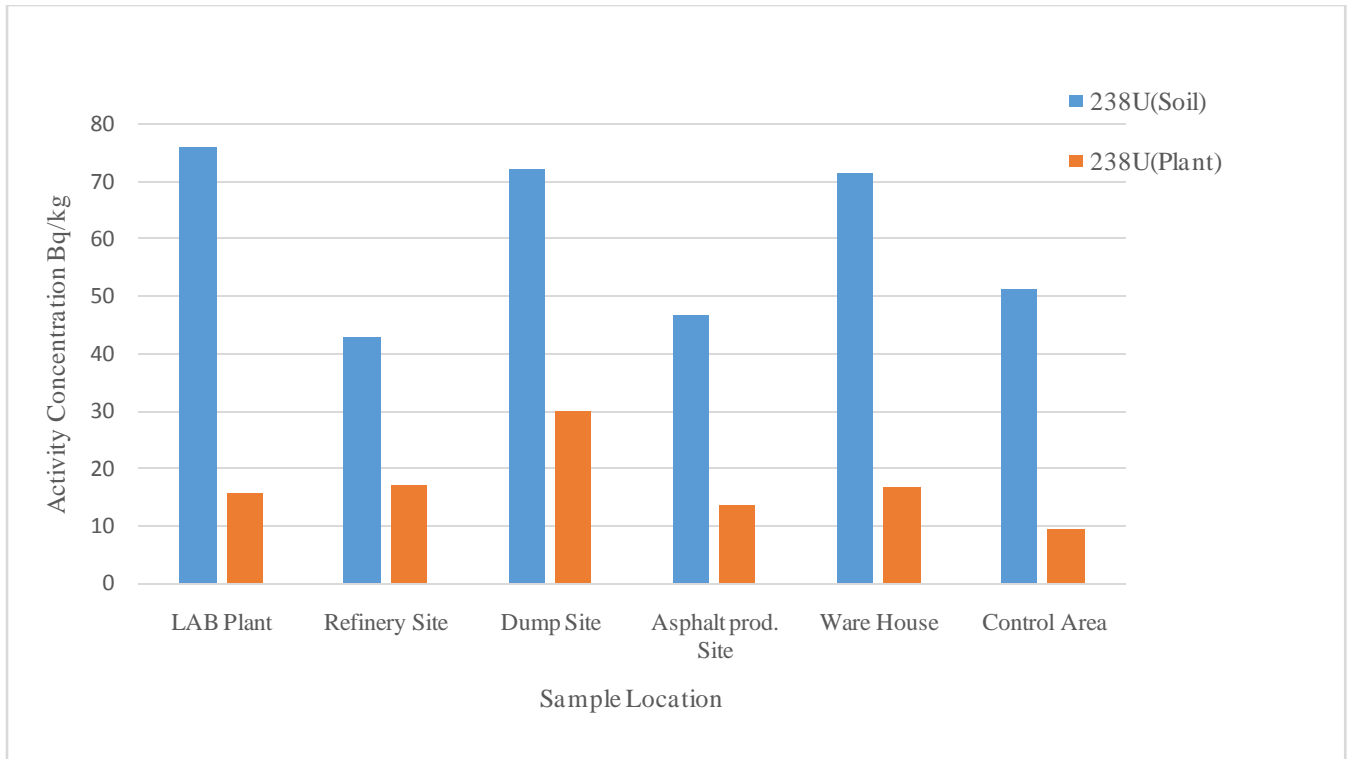


Figure 4.2.: activity concentration of ^{238}U in soil and plant samples

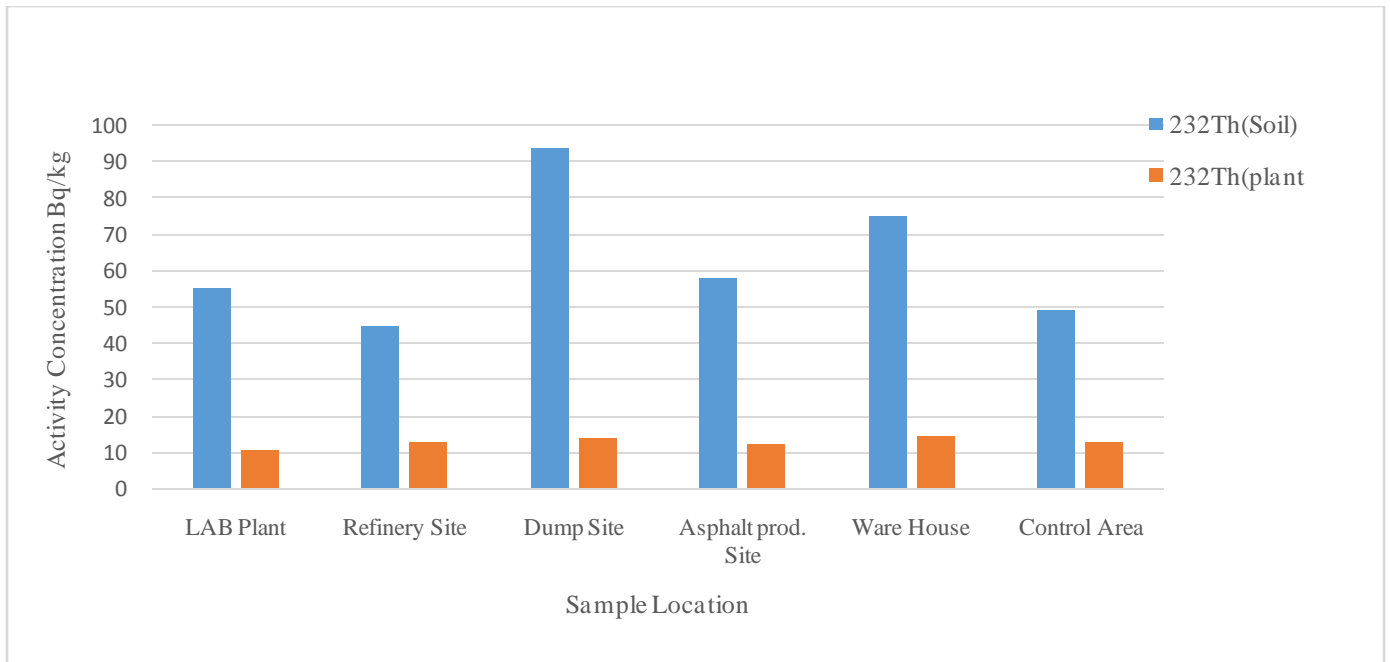


Figure 4.3: activity concentration of ^{232}Th in soil and plant samples

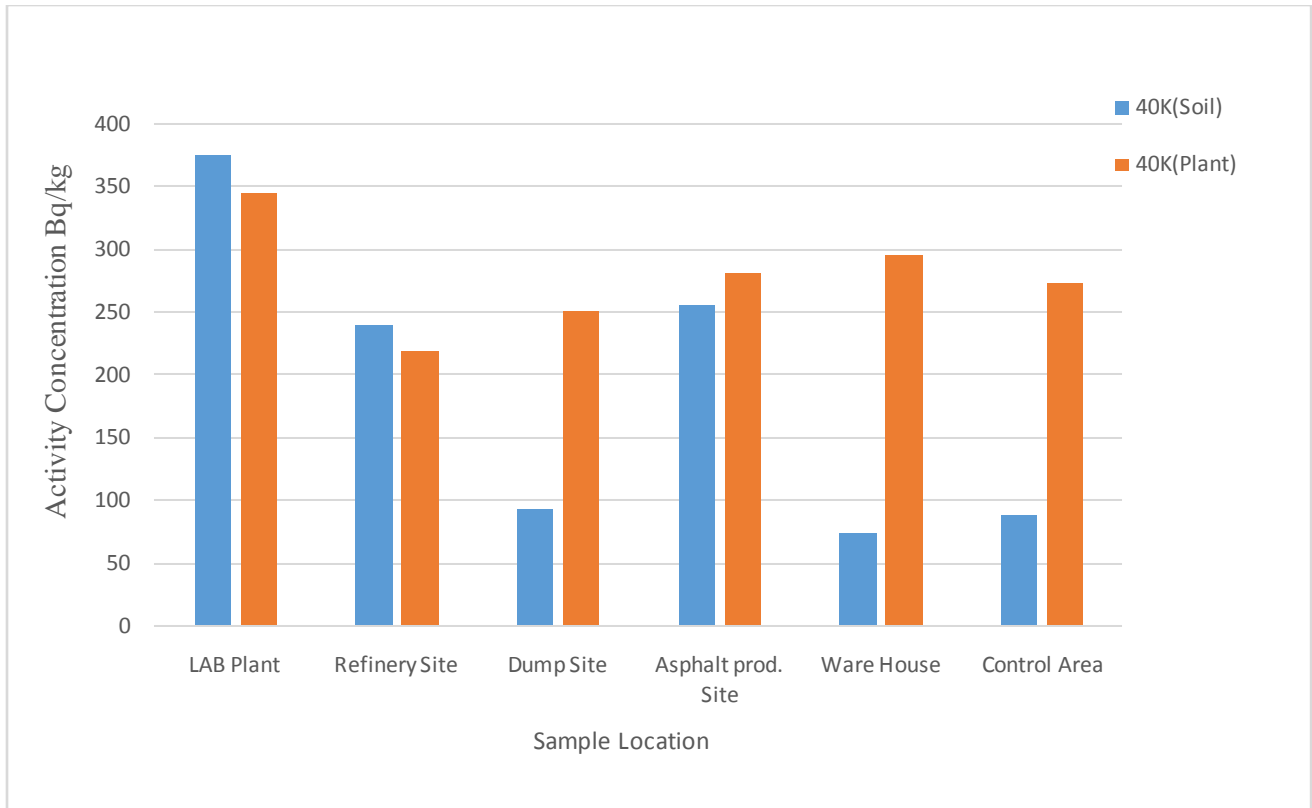


Figure 4.4: activity concentration of ^{40}K soil and plant samples

Table 4.2: Mean specific activity concentration in soil samples.

Sample code	²³⁸U(Bq/kg)	²³²Th(Bq/kg)	⁴⁰K(Bq/kg)	¹³⁷Cs (Bq/kg)
Lab plant	76.01 ± 10.86	55.23 ± 6.24	376.00 ± 9.82	ND
Refinery	43.11 ± 6.18	44.50 ± 6.37	240.11 ± 7.62	ND
Dump site	72.39 ± 10.52	93.88 ± 8.55	93.27 ± 4.71	ND
Asphalt site	46.73 ± 9.91	57.99 ± 7.63	255.91 ± 7.80	ND
Ware house	71.67 ± 9.03	75.26 ± 10.17	73.65 ± 4.58	ND
Minimum	43.11 ± 6.18	44.50 ± 6.37	73.65 ± 4.58	
Maximum	76.01 ± 10.86	93.88 ± 8.55	376.00 ± 9.82	
Mean	61.98± 9.30	65.37± 7.79	207.79± 6.91	ND
Control area	51.52 ± 7.52	48.97 ± 5.81	87.23 ± 3.78	ND

ND: Not Detected.

Table 4.3: Mean specific activity concentration in plant samples.

Sample code	$^{238}\text{U}(\text{Bq/kg})$	$^{232}\text{Th}(\text{Bq/kg})$	$^{40}\text{K}(\text{Bq/kg})$	$^{137}\text{Cs}(\text{Bq/kg})$
LAB Plant	15.87 ± 3.33	10.58 ± 5.11	346.09 ± 13.17	ND
Refinery	17.00 ± 3.30	12.95 ± 3.71	218.79 ± 9.94	ND
Dump site	30.17 ± 6.44	13.96 ± 2.58	251.73 ± 11.42	ND
Asphalt site	13.73 ± 4.27	12.04 ± 3.89	281.53 ± 12.28	ND
Ware house	16.90 ± 3.95	14.09 ± 3.00	295.81 ± 12.71	ND
Minimum	13.73 ± 4.27	10.58 ± 5.11	218.79 ± 9.94	
Maximum	30.17 ± 6.44	14.09 ± 3.00	346.09 ± 13.17	
Mean	18.73 ± 4.26	12.73 ± 3.66	278.79 ± 11.90	ND
Control area	9.45 ± 1.72	12.60 ± 4.51	274.04 ± 11.92	ND

ND: Not Detected

Table 4.4: Comparison with mean activity concentration values for ^{238}U , ^{232}Th and ^{40}K in soil samples with other related works.

Mean Activity Concentration (Bq/kg)			Reference
^{238}U	^{232}Th	^{40}K	
61.98	65.37	207.79	This study
-	63.6	165.9	Onoja, 2015
19.15	18.10	195.98	Nada <i>et al</i> , 2016
35	30	400	UNCEAR 2000.

4.4 Assessment of Radiological Hazard Indices

The Hazard indices presented in chapter 3 have been calculated from the measured activity concentrations of the radionuclides studied in soil and plant samples. The radiological hazard parameters were computed for each sample in Table 4.5 and 4.6. The values of these radiological hazard indices show that radium equivalent activity ranged from 125.24 ± 15.87 to 184.96 ± 23.93 Bq/kg with a mean value of 154.35 ± 26.73 Bq/kg in soil samples and 52.36 ± 9.38 to 69.52 ± 11.01 Bq/kg with a mean value of 58.40 ± 10.41 Bq/kg in plant samples. The mean and standard deviation of absorbed dose rate were calculated to be 90.75 ± 14.55 nGy/h in soil and 28.18 ± 4.74 nGy/h in plant samples. Similarly the mean values of annual effective dose equivalent are 86.27 ± 14.60 μ Sv/y in soil and 34.56 ± 5.81 μ Sv/yr in plant samples. Furthermore the mean values of External and internal hazard indices were calculated to be 0.42 ± 0.07 and 0.57 ± 0.10 in soil samples. The present study shows that all the results obtained for the hazard indices in all the samples are less than the world recommended average except for AEDE and absorbed dose rate (D) in soil samples which are higher than the world average value (70μ Sv/yr and 59 nGy/hr) recommended by UNSCEAR (2000).

Table 4.5: The mean calculated hazard indices and radiological parameters in soil samples.

Location	Ra_{eq} (Bq/Kg)	D_Y (nGy/hr)	AEDE(μSv/yr)	H_{ext}	H_{in}
LAB Plant	183.94±20.54	85.09±9.30	104.36±11.41	0.50±0.06	0.70±0.08
Refinery	125.24±15.87	57.57±7.13	70.60±8.74	0.34±0.04	0.46±0.06
Dumpsite	128.27±51.87	57.85±22.76	70.95±27.92	0.35±0.14	0.49±0.16
Asphalt	149.35±21.42	68.27±9.64	83.72±11.82	0.40±0.06	0.53±0.09
W/House	185 ±23.93	82.92±10.68	101.69±13.10	0.50±0.07	0.69 ±0.09
Minimum	125.24 ±15.87	57.57 ±7.13	70.60±8.74	0.34±0.04	0.46±0.06
Maximum	184.96±23.93	85.09±9.30	104.36±11.41	0.50±0.07	0.70±0.08
Mean	154.35±26.73	90.75±14.55	86.27±14.60	0.42±0.07	0.57±0.10

Table 4.6: The mean calculated hazard indices and radiological parameters in plant samples.

Location	Ra_{eq} (Bq/Kg)	D_Y (nGy/hr)	AEDE(μSv/yr)
LAB plant	57.64 \pm 11.64	28.33 \pm 5.26	34.75 \pm 6.45
Refinery	52.36 \pm 9.38	25.02 \pm 4.25	30.68 \pm 5.21
Dumpsite	69.52 \pm 11.01	33.11 \pm 5.06	40.60 \pm 6.20
Asphalt	52.63 \pm 10.79	25.56 \pm 4.90	31.35 \pm 6.01
Ware house	59.83 \pm 9.22	28.89 \pm 4.22	35.44 \pm 5.17
MIN	52.36 \pm 9.38	25.02 \pm 4.25	30.68 \pm 5.21
MAX	69.52 \pm 11.01	33.11 \pm 5.06	40.60 \pm 6.20
MEAN	58.40 \pm 10.41	28.18 \pm 4.74	34.56 \pm 5.81

4.5 Heavy metals Assessment

Fig 4.5-4.9 presented the concentrations in (ppm) of the heavy metals analyzed (Pb, Zn, Fe, Ni, Mn, Cr, Co and Cd) in soil and plant samples collected at five different locations within the study area. Heavy metals were detected at varying concentrations in all the samples except for Ni and Co which were below detection limit (BDL) in all the samples. Tables 4.7 and 4.8 summarize the concentrations (ppm) of the 8 heavy metals in soil and plant samples collected in the study area. The mean and standard deviations of the concentrations of Pb, Zn, Fe, Mn, Ni, Cr, Co and Cd for soil samples in the study area were 0.42 ± 0.18 , 0.02 ± 0.02 , 1.01 ± 1.18 , 0.21 ± 0.27 , BDL, 0.02 ± 0.01 , BDL and 0.004 ± 0.00 respectively. The Highest concentration corresponds to Fe and the lowest corresponds to Cd. The increasing trend was: $Co < Ni < Cd < Cr < Zn < Mn < Pb < Fe$. Also the mean and standard deviations of the concentrations of Pb, Zn, Fe, Ni, Mn, Cr, Co and Cd in the study area for plant samples were 0.18 ± 0.18 , 0.02 ± 0.02 , 0.22 ± 0.11 , 0.15 ± 0.13 , BDL, 0.01 ± 0.01 , BDL and 0.003 ± 0.003 respectively. The Highest concentration corresponds to Fe and the lowest corresponds to Cd. The increasing trend was: $Co < Ni < Cd < Cr < Zn < Mn < Pb < Fe$.

Table 4.9 and 4.10 presented the average concentrations (ppm) of each of the studied heavy metals corresponding to each sample collection site in the target and control areas respectively. The average concentrations observed in the target area were higher than that in the control area for all the studied heavy metals. The average concentration in each sampling location was found to be at different variation and this could be attributed to various anthropogenic activities been carried out in the study area. The Average concentrations of soil samples in the target and control area were compared with the world health organization guideline on the maximum limits of toxic metals in soil (WHO, 1997) as provided in Table 4.11.

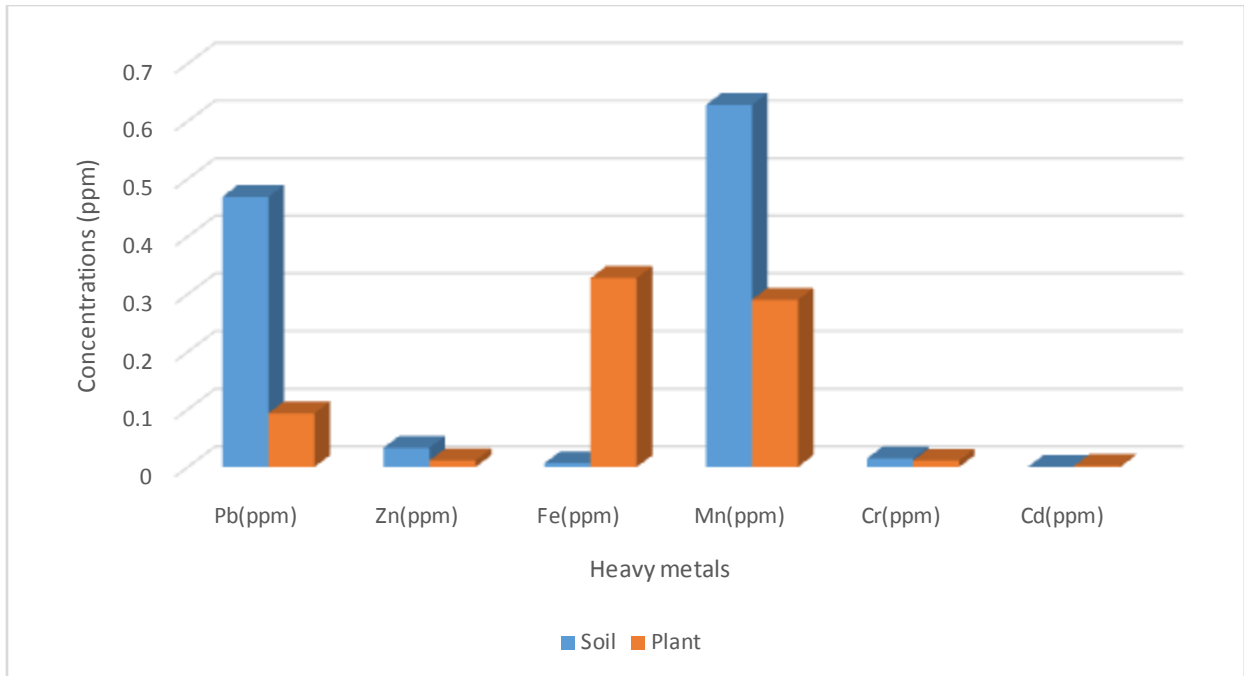


Figure 4.5: Heavy metals concentration in soil and plant sample in Lab plant.

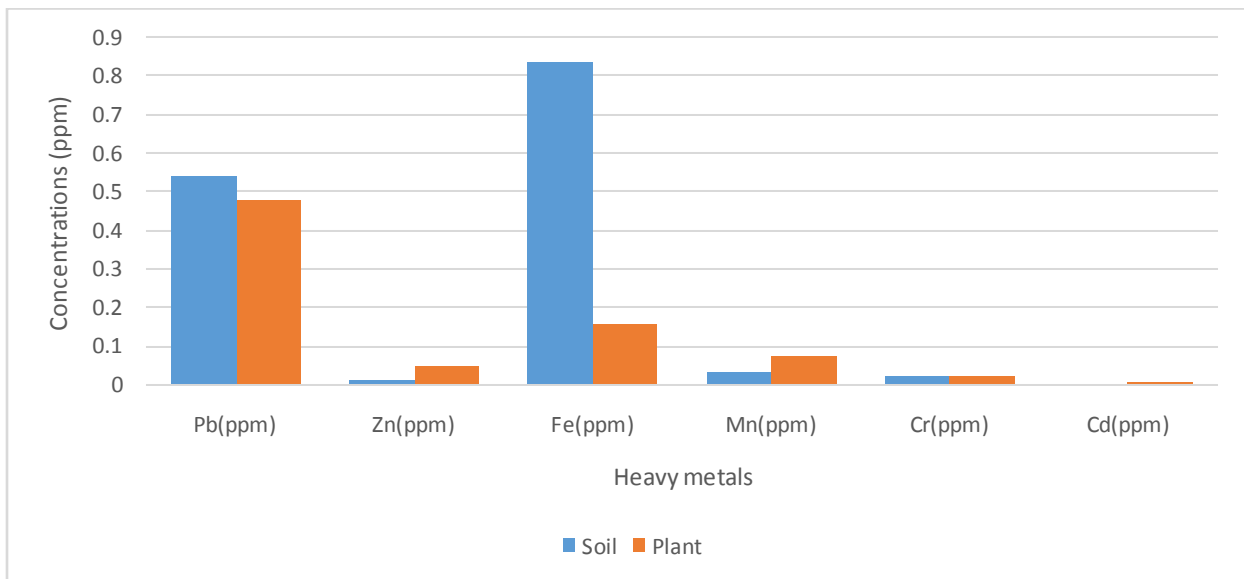


Figure 4.6: Heavy metals concentration in soil and plant sample in Refinery site.

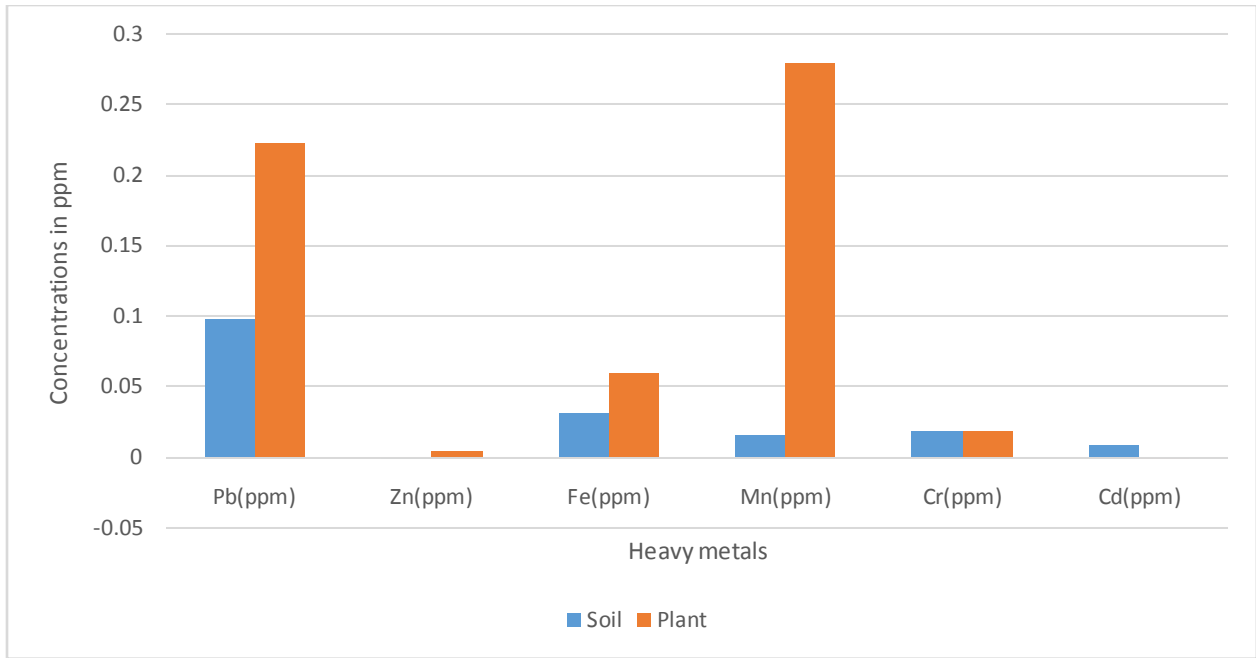


Figure 4.7: Heavy metals concentration in soil and plant sample in Waste deposit site.

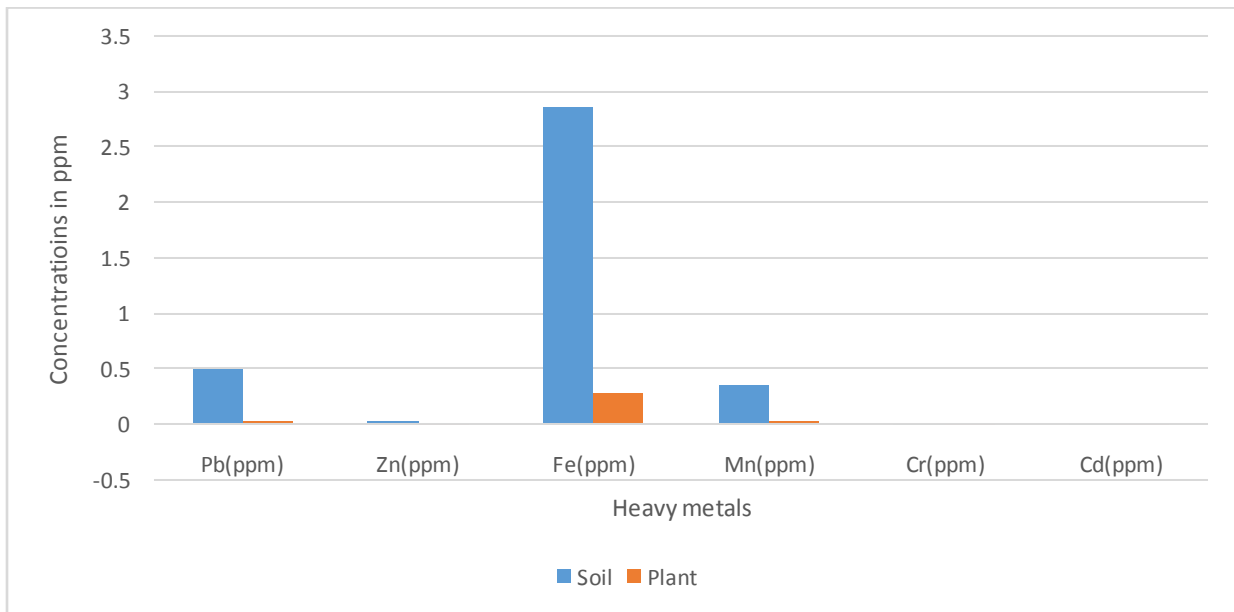


Figure 4.8: Heavy metals concentration in soil and plant sample in Asphalt production site.

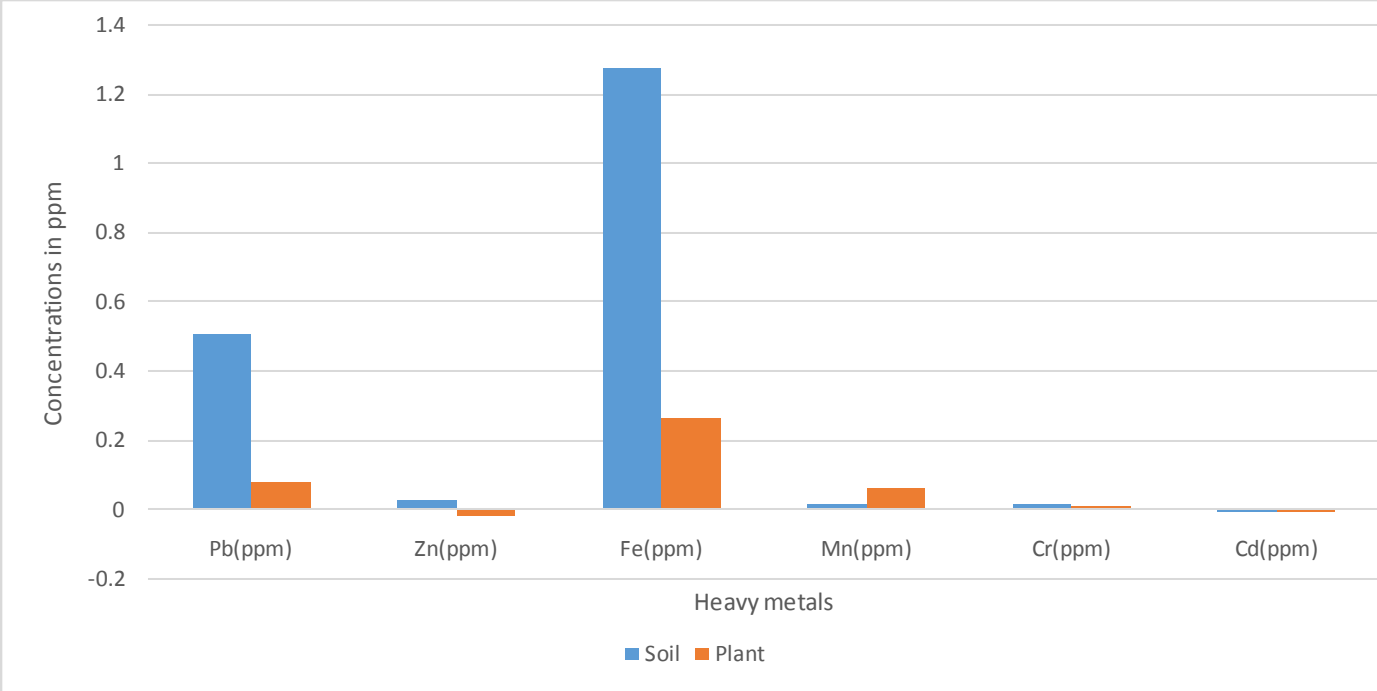


Figure 4.9: Heavy metals concentration in soil and plant sample in Ware house.

Table 4.7: Univariate descriptive Statistics of the concentration of the heavy metals (ppm) in soil samples

Toxic metal	Mean	Minimum	Maximum	Standard deviation
Pb	0.42	0.10	0.54	0.18
Zn	0.02	BDL	0.03	0.02
Fe	1.01	0.01	2.90	1.18
Mn	0.21	0.02	0.63	0.27
Ni	BDL	BDL	BDL	BDL
Cr	0.02	0.00	0.02	0.01
Co	BDL	BDL	BDL	BDL
Cd	0.01	0.001	0.008	0.003

Table 4.8: Univariate descriptive Statistics of the concentration of the toxic metals plant samples (ppm)

Toxic metal	Mean	Minimum	Maximum	Standard deviation
Pb	0.18	0.02	0.48	0.18
Zn	0.02	BDL	0.05	0.02
Fe	0.22	0.06	0.33	0.11
Mn	0.15	0.02	0.29	0.13
Ni	BDL	BDL	BDL	BDL
Cr	0.01	0.003	0.01	0.01
Co	BDL	BDL	BDL	BDL
Cd	0.003	0.0004	0.01	0.003

Table 4.9: Average Concentration of the heavy metals (ppm) in soil corresponding to each of the studied site in the target and control Area

Pb	Zn	Fe	Mn	Ni	Cr	Co	Cd			
Lab plant(soil)			0.47	0.03	0.007	0.63	BDL	0.02	BDL	0.01
Refinery soil			0.54	0.01	0.84	0.03	BDL	0.02	BDL	0.002
Dump site(soil)			0.10	BDL	0.03	0.02	BDL	0.02	BDL	0.01
Asphalt site(soil)			0.49	0.033	2.87	0.34	BDL	0.003	BDL	0.01
Ware house(soil)			0.51	0.03	1.28	0.02	BDL	0.02	BDL	0.001
Control area(soil)			0.03	0.01	0.07	0.02	BDL	0.02	BDL	0.01

Table 4.10: Average Concentration of the heavy metals (ppm) in plant corresponding to each of the studied site in the target and control Area

Pb	Zn	Fe	Mn	Ni	Cr	Co	Cd		
Lab plant		0.09	0.01	0.33	0.29	BDL	0.01	BDL	0.002
Refinery		0.48	0.05	0.16	0.07	BDL	0.02	BDL	0.004
Dump site		0.22	0.004	0.06	0.28	BDL	0.02	BDL	0.00
Asphalt site		0.02	BDL	0.27	0.02	BDL	0.02	BDL	0.01
Ware house		0.08	BDL	0.27	0.07	BDL	0.01	BDL	0.001
Control area		0.06	0.003	0.07	0.07	BDL	0.003	BDL	0.004

Table 4.11: Average Concentration of the elements in the Background and Sampled Area and The World Health organization Limit for Heavy metal concentrations (ppm)

Parameter	Pb	Zn	Fe	Mn	Ni	Cr	Co	Cd
Average Background Concentration	0.03	0.01	0.07	0.02	BDL	0.02	BDL	0.01
Average Concentration of Sample points	0.42	0.02	1.01	0.21	BDL	0.02	BDL	0.004
WHO recommended level	200	30	80	3.00	N/A	100	-	100

4.5.1 Contamination Factor and Pollution Load Index

Table 4.12 and 4.13 presented the summary statistics of the calculated contamination factors of the analyzed samples. The contamination factor was used to assess the level of contamination of each element in the studied soil and plant samples. Based on the categories discussed in chapter 2, the elemental concentrations could be categorized as follows for soil samples (i) Cr and Cd in the low contamination category (ii) Zn in the moderate contamination category (iii) Pb, Fe and Mn in the very high contamination factor category. While for plant samples (i) Cr, Zn and Cd were in the low contamination category (ii) Fe in the moderate contamination category (iii) Pb, and Mn in the very high contamination factor category.

Table 4.14 presented the statistics of pollution load indices in samples collected within the study area. The pollution load indices (PLI) was used to assess the overall pollution of the site resulting from studied heavy metals. The pollution load indices indicated that the study area is highly polluted with all observed heavy metals (mean = 75.96).

Table 4.12: Univariate descriptive Statistics for the Contamination factors of the Soil Samples Analyzed.

Toxic Metal	Mean	Minimum	Maximum	Standard deviation
Pb	15.06	3.49	19.29	6.54
Zn	1.86	1.00	2.99	1.39
Fe	13.67	0.09	39.03	15.99
Mn	9.31	0.67	15.33	12.23
Cr	0.72	0.16	1.00	0.33
Cd	0.43	0.06	0.99	0.44

Table 4.13: Univariate descriptive Statistics for the Contamination factors of the plant Samples Analyzed.

Toxic Metal	Mean	Minimum	Maximum	Standard deviation
Pb	6.35	0.58	17.05	6.55
Zn	0.76	0.35	4.02	2.04
Fe	2.94	0.81	4.46	1.46
Mn	6.53	0.94	13.01	5.78
Cr	0.71	0.53	0.86	0.14
Cd	0.33	0.05	0.82	0.30

4.14: The mean Pollution Load Index in each site and the average in the whole study area

Sample Site	Mean Pollution Load Index
Lab Plant	6.07
Refinery Site	28.70
Dump Site	0.02
Asphalt Site	332.36
Ware House	12.66
Mean Pli In The Study Area	75.96

4.5.2 Correlation between heavy metals concentrations

In an attempt to unravel the relationship between the heavy metal concentrations in the sampling location, the result obtained at each sampling location were subjected to correlation analysis using Microsoft excel 2013. The correlation coefficient were presented in Tables 4.15- 4.19. The results of this correlation indicated a significant Positive correlation between the pairs Pb/Zn, Pb/Mn, Pb/Cr, Zn/Mn, Zn/Cr, Mn/Cr and Cr/Cd in LAB plant, Pb/Fe, Zn/Cr, Mn/Cd in refinery area, Pb/Cr, Zn/Fe, Fe/Mn in dump site, Pb/Zn/Fe/Mn in asphalt production site and Pb/Zn, Pb/Fe, Zn/Fe, Fe/Cr, Mn/Cd, Cr/Cd in ware house. Similarly a Significant negative Correlation between the pairs Pb/Fe, Zn/Fe, Fe/Mn, Fe/Cr, Mn/Cd, in LAB plant, Pb/Zn, Zn/Fe, Mn/Cr, Cr/Cd in refinery area, Pb/Fe, Pb/Mn, Pb/Cd, Zn/Cd, Zn/Cr in the dump site, Pb/Cr, Zn/Cr, Mn/Cr, Cr/Cd in asphalt production site and Pb/Mn, Zn/Mn, Fe/Mn in ware house were observed.

Table 4.15: Pearson correlation matrix for concentration of pairs of elements (ppm) in lab plant.

	Pb	Zn	Fe	Mn	. Cr	Cd
Pb	1					
Zn	0.904	1				
Fe	-0.986	-0.963	1			
Mn	0.941	0.707	-0.870	1		
Cr	0.986	0.963	-1	0.872	1	
Cd	-0.395	0.034	0.235	-0.683	0.545	1

Table 4.16: Pearson correlation matrix for concentration of pairs of elements (ppm) in refinery site.

	Pb	Zn	Fe	Mn	Cr	Cd
Pb	1					
Zn	-0.926	1				
Fe	0.974	-0.816	1			
Mn	-0.140	-0.243	-0.362	1		
Cr	-0.140	0.516	0.073	-0.956	1	
Cd	0.093	-0.461	-0.136	0.973	-0.998	1

Table 4.17: Pearson correlation matrix for concentration of pairs of elements (ppm) in dump site.

	Pb	Zn	Fe	Mn	Cr	Cd
Pb	1					
Zn	-0.262	1				
Fe	-0.555	0.948	1			
Mn	-0.662	0.897	0.991	1		
Cr	0.835	-0.750	-0.921	-0.956	1	
Cd	-0.710	-0.847	-0.635	-0.524	0.283	1

Table 4.18: Pearson correlation matrix for concentration of pairs of elements (ppm) in asphalt production site.

	Pb	Zn	Fe	Mn	Cr	Cd
Pb	1					
Zn	0.991	1				
Fe	0.986	0.956	1			
Mn	0.997	0.980	0.995	1		
Cr	-0.610	-0.708	-0.468	-0.0551	1	
Cd	0.011	0.141	-0.157	-0.061	-0.799	1

Table 4.19: Pearson correlation matrix for concentration of pairs of elements (ppm) in ware house.

	Pb	Zn	Fe	Mn	Cr	Cd
Pb	1					
Zn	0.955	1				
Fe	0.984	0.887	1			
Mn	-0.767	-0.922	-0.640	1		
Cr	0.393	0.103	0.550	0.289	1	
Cd	-0.322	-0.587	-0.148	0.854	0.745	1

CHAPTER FIVE

SUMMARY, CONCLUSION AND RECOMMENDATIONS

5.1 Summary

The activity concentrations of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs were assessed in soil and plant samples collected from five different locations inside KRPC. The result obtained for ^{238}U and ^{232}Th were above the world average values, ^{137}Cs was not detected in all the samples. The radiological implications were all below the standard limits except for annual effective dose equivalent and the absorbed dose rate in soil samples. This implies KRPC personnel and other members of the public are within the safety limits of radiation exposure.

The studied heavy metals (Pb, Zn, Fe, Mn, Ni, Cr, Co and Cd) were detected at varying concentrations in both the soil and plant samples. Based on the values for the contamination factor and pollution load index, it can be seen that the soil and plant samples are highly contaminated with Pb, Fe, and Mn.

5.2 Conclusion

The measured gamma radiation level ranged from 0.11 to $0.15\mu\text{Svhr}^{-1}$ with a mean value of $0.12\mu\text{Svhr}^{-1}$. This is roughly twice the world average value of $0.056\mu\text{Svhr}^{-1}$ (UNCEAR, 2000).

Gamma spectrometry was used to assess the activity concentration of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs . All the studied radionuclides were detected at varying concentrations except for ^{137}Cs which was not detected in all the samples. The mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil samples were estimated to be 61.98 ± 9.30 , 65.37 ± 7.79 and 207.79 ± 6.91 Bq/kg for soil and 18.7 ± 4.3 , 12.7 ± 3.7 and 278.8 ± 11.9 Bq/kg for plant samples. The mean activity

concentrations for ^{238}U and ^{232}Th were higher than the world average values of 35 and 30 Bq/kg, but ^{40}K lies within the world average value of 400 Bq/kg. The overall result indicates that the natural occurring radionuclides are not uniformly distributed in soil and plant samples. Radiological hazard indices were estimated from the activity concentration results obtained for the observed radionuclides in soil and plant samples collected from the study area. The results show that all the hazard indices (H_{ex} and H_{in}) were less than unity. In conformity with the values of the hazard indices, the radium equivalent activity has a mean value of 154.35 ± 26.73 Bq/kg in soil and 58.40 ± 10.41 Bq/kg in plant samples which is lower than the threshold value of 370 Bq/kg, this implies that KRPC personnel and members of the public are subjected to an effective dose lower than 1 mSv/yr. AEDE and absorbed dose (D) for soil were higher than the standard value of $70 \mu\text{Sv}$ and 59nGy/hr .

The study used flame atomic absorption spectrometry to assess the concentrations of some heavy metals (Pb, Zn, Fe, Mn, Ni, Cr, Co and Cd) in soil and mango leaves (*mangifera indica*) collected from the study area. The mean concentrations of the studied heavy metals were found to be far below the WHO recommended values on the maximum of heavy metals in soil, although even at lower concentration these heavy metals can become a potential threat to human health due to their non-biodegradability in the biosphere. The obtained concentrations of the studied heavy metals in both soil and plant samples were used in computation of the hazard indices using pollution load index (PLI) and contamination factors. Based on the calculated values obtained for these indices it has been concluded that the soil and plant samples collected from the study area have been highly contaminated with the studied heavy metal especially Pb, Fe and Mn.

5.3 Recommendations

In view of the major findings revealed by this study, the following recommendations were made:

- i. This kind of study should be carried out in and around other industries to ensure compliance in order to sustain our fragile environment.
- ii. Appropriate measures should be put in place by government and petroleum companies to check the discharge of waste into the environment.
- iii. Measuring of heavy metals and other contaminants should be a continuous process.
- iv. Sensitization and awareness workshop should be carried out on effect of ionizing radiation.
- v. Government agencies responsible for the safety of the environment should enforce all existing legislations on the environment especially radioactivity.

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