



ANALYSIS OF RADON PRODUCT ACTIVITIES FROM FILTERED AIR SAMPLES

By

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Dedication

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Acronyms and abbreviations

Å $\equiv$ Angstrom

ACs $\equiv$ Activity concentrations

Bq/l $\equiv$ Becquerel per liter

Bq/m³ $\equiv$ Becquerel per meter cube

CEC $\equiv$ Commission of European Committee

CR $\equiv$ Count Rates

DNA $\equiv$ Deoxyribonucleic Acid

E - PERM[®] $\equiv$ (Electret-Passive Environmental Radiation Monitors)

EPA $\equiv$ Environmental Protection Agency

EEDC $\equiv$ Equilibrium Equivalent Decay Concentration

EECs $\equiv$ Equilibrium Equivalent Concentrations

GM $\equiv$ Geiger-Müller

gy $\equiv$ Gray

IARC $\equiv$ International Agency for Research on Cancer

ICRP $\equiv$ International Commission For Radiological Protection

IAEA $\equiv$ International Atomic Energy Agency

MeV $\equiv$ Mega Electron Volt

NCRP $\equiv$ National Council on Radiation Protection

NORM $\equiv$ Naturally Occurring Radioactive Material

PET $\equiv$ Positron Emission Tomography

SSNTD $\equiv$ Solid State Nuclear Track Detector

SSJM $\equiv$ Steady State Jacobi Model

UNSCEAR $\equiv$ United Nation Scientific Committee on the Effects of Atomic Radiations

US EPA $\equiv$ United State Environmental Protection Agency

WHO $\equiv$ World Health Organization

WNA $\equiv$ World Nuclear Association

Abstract

Radon is a colorless, odorless, tasteless and radioactive gas. It forms naturally from the decay of radioactive elements such as uranium. It is a natural radioactive gas that occurs ubiquitously throughout the world. In this work, analysis of radon product activities from filtered air samples taken place based on grab sampling of known volume of air on a glass fiber filter with high retention power followed by gross beta counting of the filtered air sample counted over large number of successive time intervals using an end window Geiger - Müller (GM) counter were reviewed. This method results determination of concentrations of radon and thoron daughter products at a time. The gross beta count is determined using a mathematical model based on Bateman differential equations. Theoretically the radionuclides ^{218}Po , ^{214}Pb , ^{214}Bi , which are progenies of radon (^{222}Rn) and ^{212}Pb , ^{212}Bi , ^{208}Tl , which are progenies of thoron (^{220}Rn) have significant contributions to the total beta counting rate. To determine their individual contributions to the total count rate, the Weighted Least Square Method (WLSM) or the Steady State Jaccobi's Model (SSJM) can be applied. The review of this paper shown that using the Weighted Least Square Method (WLSM) results high random errors than that of the Steady State Jaccobi's Model (SSJM). In both methods accurate results can be obtained for ^{214}Pb , ^{214}Bi , ^{212}Pb , EEDC₂₂₂ and EEDC₂₂₀. However, the results for ^{218}Po and ^{212}Bi are much more inaccurate and the concentration of ^{208}Tl is difficult to determine. The Steady State Jaccobi's Model (SSJM) takes in to consideration air ventilation rate as a base line to determine radon progenies in a confined system. It gives better detection efficiency and lower the random error of radon daughter concentration.

Chapter 1

Introduction

Radon, it is a colorless, odorless, tasteless and radioactive noble gas. It is one of naturally occurring radioactive material (NORM) [1]. It is the heaviest member of the rare gas group ~ 100 times heavier than hydrogen and ~ 7.5 times heavier than air [1]. Although radon has about 39 known isotopes, the three main naturally occurring radon isotopes are radon (^{222}Rn), a member of the uranium decay series, thoron (^{220}Rn), a member of the thorium decay series, and actinon (^{219}Rn), a member of the actinium decay series. The first and very important to know is radon ^{222}Rn , which is a natural radioactive gas that occurs ubiquitously throughout the world. This isotope has an atomic number of 86 and a half life of 3.82 days. The second type of radon isotope is thoron, denoted by ^{220}Rn . Like ^{222}Rn , it has an atomic number 86, mass number 220 and half life of 55.3 seconds [1]. The third type of radon isotope is , ^{219}Rn which is derived from the most stable element of actinium (^{227}Ac) named "actinon" and is an alpha emitter with a half - life of 3.96 seconds [1]. The main characteristic of radon isotopes, like ^{222}Rn , ^{220}Rn and ^{219}Rn among other natural radioactive elements is the fact that their behavior is not affected by chemical processes [2, ?]. Concentration levels of radon depends strongly on geological and geophysical conditions. Once radon isotopes are formed as a result of the natural radioactive series in the earth's crust they can move freely through soil pores and rock fractures then escape into the atmosphere. Radon gases are exhaled from the Earth's surface into the free atmosphere, rapidly dispersed and diluted by natural convection and turbulence. When a dwelling is present, they may migrate into this structure and accumulate indoors in sufficient quantities to pose a health hazard. One can be continuously exposed to radon unknowingly. Air ventilation rate, in contrast, has negative impact on the concentration of radon and their progenies both in side and out of a dwelling. Radon is decay with the emission of alpha particles and produce daughter nuclei of polonium,

lead and bismuth [3]. These daughter nuclei emit alpha or beta particles [4, 3]. Among these daughter nuclei, accurate results can be obtained for ^{214}Pb , ^{214}Bi , ^{212}Pb , EEDC - 222 and EEDC - 220. However, the results for ^{218}Po and ^{212}Bi are much more inaccurate and ^{208}Tl is impossible to determine [3]. Hence, measurement of the concentration of radon and thoron decay products in air is an important aspect using different measuring techniques like grab sampling method to take a series remedial action to a health point of view. Concentration of short lived radon daughter products are measured in the air by using beta counting method. It is based on direct beta counting of filtered aerosol sample over successive time intervals by end-window Geiger - Muller counter.

1.1 Radioactivity

Radioactivity, discovered by Henry Becquerel in 1896, is the nature of an unstable atomic nucleus (radio nuclide) loses energy by emitting alpha, beta and gamma radiations to become more stable. Radioactive decay is the spontaneous emission of particles or electromagnetic radiation from an unstable atom due to a transmutation in its nucleus [5]. A material containing unstable nucleus is considered as radioactive material. Radioactive decay is the breakdown of unstable atom, occurs in unstable atomic nuclei that is, one that don't have enough binding energy to hold the nucleus (or nucleon) together due to unbalanced number of protons and neutrons ($Z > 82$) [6]. There are two types of decay chains, single step and many steps decay chain. For single step, the decay is a one step process and a stable atom is formed soon. The decay of ^{60}Co into ^{60}Ni is a best example. On the other hand, nucleus of the decay product, which is called its daughter is unstable and this results in other decay till the daughter nuclei is stable. This multiple step is called a decay series. The thorium ($4n$) series, the uranium ($4n+2$) series, the actinium ($4n+3$) series are the three naturally occurring unstable radioactive elements [6]. The fourth set, the neptunium ($4n+1$) series, produced artificially by a nuclear reaction and do not occur naturally. The decay of ^{238}U to ^{208}Pb is an example of decay series [6, 7].

1.2 The discovery of radon isotopes

1.2.1 The discovery of thoron (^{220}Rn)

The first radon isotope to be discovered was thoron (^{220}Rn). Thoron is the breakdown result of thorium (^{232}Th) series. Rutherford and R.B. Owens in 1899 noticed that some of the radioactivity of thorium compounds could be blown away [7]. They were studying the emanations from thorium and noticed that the radiation from thorium was not constant but varied in a sudden unexpected manner. Rutherford made the key observations such that, the emanation acts like an ordinary gas and that the intensity of radiation has fallen to one half of its value after an interval of about one minute [7]. Finally thoron (^{220}Rn), half life of 55.6 seconds was discovered in 1899 by R.B.Owens at Mc Gill University in collaboration with Ernest Rutherford. Information on ^{220}Rn and its daughters have their application in the description of air flow [8]; as tracer gases and in the studies of atmospheric electrical phenomena [9]. It has been reported that the concentration of (^{220}Rn) can be as high as 200 Bq / m^3 under certain circumstances and the outdoor air can also be a source of high ^{220}Rn in indoor air [9].

1.2.2 The discovery of radon (^{222}Rn)

Uranium decays to radium through a long series of steps with a half - life of 4.5 billion years. Radon (^{222}Rn) is a naturally occurring radioactive gas that originates from the decay of radium (^{226}Ra) in the primordial uranium (^{238}U) decay series. Pierre and Marie Curie, studying emanations from radium in 1899, noticed that it stayed radioactive for several days. The Curie's had detected ^{222}Rn , half life 3.82 days, but they still had doubts as whether the activity was really in the form of a gas or not and it was the second radon isotope to be discovered. Later work by Rutherford (1901) and also by Frederick Ernest Dorn, a German scientist in (1900) confirmed that the radiations emitted by radium were a radioactive gas. It was named 'niton' (Latin word, which means 'shining'). In 1908 by Ramsay and Gray, but soon after came to be called radium emanation, being named as a radioactive noble gas formed by the decay of radium, it was only since 1923 that it came to be called radon [10].

1.2.3 The discovery of actinon (^{219}Rn)

Actinon ^{219}Rn short lived another isotope of radon, whose half life is extremely short, only 3.92 seconds, chemically inert, that is a member of actinium series, was the third radon isotope to be discovered in 1904 by Friedrich Giesel and Andre - Louis Debierne. Now a day, about 39 isotopes of radon are known ranging from Rn - 193 to Rn - 231, from which the most abundant naturally existing isotopes of radon are ^{222}Rn , ^{220}Rn and ^{119}Rn [10].

1.3 Early radon measurements

In 1901, Elster and Geitel, were the first to make measurements of radon decay products by collecting them on a charged wire [11]. It was basically intended for the study of environmental phenomena like atmospheric electricity, atmospheric transport and exhalation of gases from soil. Due to the fact that, radon and its short lived daughters are carcinogenic agents and results series health hazard, serious investigations were followed not for atmospheric studies but to a great extent due to the health hazards caused by radon [11]. It is estimated that radon causes between 3 % to 14 % of all lung cancers in a country, depending on the average radon level and the smoking prevalence in a country [12]. Countries of the world have different levels of radon concentration as shown in the figure (Fig.1.1) below.

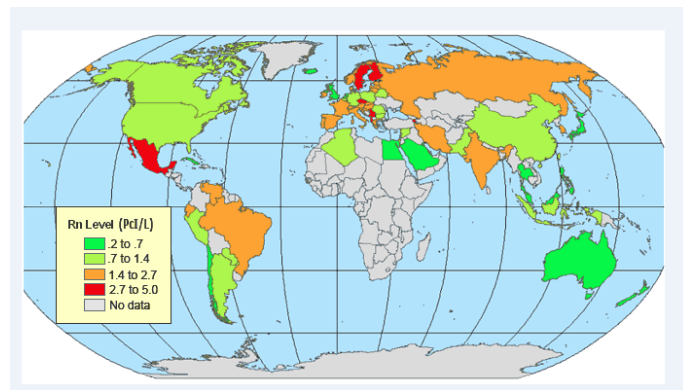


Figure 1.1: Arithmetic mean radon level by country (based on data up to 2007)

Some of them have high level of radon concentration like Asia and some of them have low concentration of radon. The radon concentration of Ethiopia is not known according to this data.

1.4 Chemical, physical and radiological information of radon

1.4.1 Chemical identities

Radon is a naturally occurring radionuclide. The largest source of radon in the environment is due to the ambient levels formed from uranium, thorium, actinium and their decay products in the soil [12]. Radon is a decay product of radium [12]. The chemical identity of radon isotopes and identification numbers for some of the radon isotopes like (^{218}Rn , ^{219}Rn , ^{220}Rn , ^{222}Rn , ^{226}Rn , ^{229}Rn , and ^{230}Rn) are representative examples.

1.4.2 Radiological properties

Although, radon has about 39 isotopes, the most stable and abundant type is ^{222}Rn with a half-life of 3.823 days, which decays into ^{218}Po [12]. Only three of 39 radon isotopes, which are ^{222}Rn , ^{211}Rn and ^{210}Rn , have half-lives greater than an hour. ^{222}Rn , ^{219}Rn and ^{220}Rn are an intermediate steps in the decay chain for ^{238}U , ^{235}U , ^{232}Th , respectively. The short lived radon daughters for each decay chain have minimum and maximum half lives as shown in table (1.1) below. .

decay series	minimum half life	maximum half life
actinium	1.78 m sec for ^{215}Po	36.1 min for ^{211}Pb
thorium	0.3 μ sec for ^{212}Po	10.6 h for ^{212}Po
uranium	164 μ sec for ^{214}Po	26.8 min for ^{214}Pb

Table 1.1: Minimum and maximum half lives of actinium, thorium and uranium decay series

The time taken for half of the amount of the substance to be expelled from the body is known as biological half-life. For long lived radon isotopes, the biological half-life can be shorter than the radiological half-life. This means that the nuclide will be expelled from our body before the majority of the nuclide decay. Example, Lead - 210. For that of short lived radon daughters, the biological half-life is longer than the radiological half-life which decay before they can be removed from the lung tissue. Example, ^{214}Po has half life of 164.3 μ sec.

1.4.3 Physical properties

Radon (Rn) is a radioactive, colorless, odorless and tasteless gas that occurs naturally as the decay product of the elements radium, which is the decedent of uranium, thorium and actinium . It is dens and the heaviest known gas. Radon has density of 9.73 kg / m^3 about 8 times the density of Earth's atmosphere at sea level (1.217 kg / m^3). At standard temperature and pressure radon is a colorless gas but when it is cooled below its freezing point, 202K ($-71 \text{ }^\circ\text{C}$), it has a brilliant phosphorescence which turns yellow as the temperature is lowered and orange-red at the temperature air liquefies, 211.2K ($-61.8 \text{ }^\circ\text{C}$).

1.4.4 Chemical properties

The main characteristic of radon among other natural radioactive elements is the fact that its behavior is not affected by chemical processes [12]. One compound that has been made is radon fluoride (RnF). In any event, such compounds are laboratory curiosities and have no commercial uses. Some of the physical, chemical and radiological properties of radon are given in the Table (1.2), below .

Name, Symbol, Atomic number (Z)	Radon, Rn , 86
Group, Period, Block	18, 6, P
Element category	noble gas
Atomic mass (<i>g/mol</i>)	Radon (222), Thoron (220), Actinon (219)
Color or appearance	Colorless
Physical state	Gas at $0 \text{ }^\circ\text{C}$ and 760 mm Hg
Melting point	$-71 \text{ }^\circ\text{C}$
Boiling point	$-61.8 \text{ }^\circ\text{C}$
Density at $20 \text{ }^\circ\text{C}$	$9.73 \times 10^{-3} \text{ g / cm}^3$
Half life of $^{222} \text{ Rn}$	3.8235 days
Half life of $^{220} \text{ Rn}$	55.6 sec
Half life of $^{219} \text{ Rn}$	3.96 sec
odor	odor less

Table 1.2: Physical, chemical and radiological properties of radon

1.5 Where does radon come from?

Radon is a naturally occurring gas and comes from the breakdown of naturally occurring radioactive elements (such as uranium and thorium) in soils and rocks [7, 12]. Basically,

radon atoms located within solid grains are unlikely to become available for release to the atmosphere due to their very low diffusion coefficients in solids. However, if they are located in the interstitial space between grains, they may diffuse to the surface. Therefore, there are about three mechanisms to be mentioned for radon entry in to our homes.

◇ **Emanation** - it is a process of radon atoms formed from the decay of radium, escape from the grains (mainly because of recoil) into the interstitial space between the grains.

◇ **Transport** - it is a process in which diffusion and advection flow cause the movement of the emanated radon atoms through the residue or soil profile to the ground surface.

◇ **Exhalation** - it is a process of radon atoms that have been transported to the ground surface and then exhaled or escape from the soil, where it can enter homes, schools and the workplace through cracks and other holes in the foundation.

Since all the above three radon entry mechanisms are difficult totally to control, any home can have a radon problem. There are trace amounts of uranium in the soil all over the globe. Some areas have higher radon concentrations and higher chance of having radon problem than other areas. But it can be a problem in homes, schools and buildings anywhere in work place where the air ventilation rate is very low [12]. The three mechanisms mentioned above for radon entry in to our homes is indicated in the figure (fig 1.2)

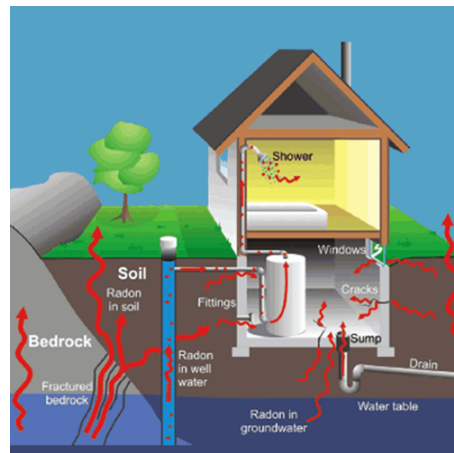


Figure 1.2: Radon entry mechanisms in to homes (EPA, 2005)

1.6 Radon and mining

The extraction of valuable minerals or other geological materials from an ore, vein, lode, seam reef or placer deposit is known as Mining. It is also an ancient, multi-disciplinary industry recognized as being arduous and liable to injury and disease [13]. Mining activities results a negative environmental impact. Different studies on mining activities have consistently shown an increased risk of lung cancer with cumulative exposure to the widely known radon isotopes (^{222}Rn and ^{220}Rn) [13]. The concentration of radon isotopes, radon (^{222}Rn and ^{220}Rn) and their progeny form 54.8 % of the effective dose of natural radiation received by the U.S population [14]. Most of the time we may exposed to radioactive radon isotopes and their short lived progeny through the inhalation of the radioactive radon daughter particles produced by mining and milling [15]. The short lived radon progenies, like, Po - 218, Bi - 214 and Po - 214 are major carcinogenic particles to human beings [16]. The decay products are all solids and are readily deposited on the bronchial airways and inside the alveoli of the lungs during breathing [17]. The radiation dose received from radon (^{222}Rn) is very low than the dose received from the daughters because of its long half-life, 3.83 days [18].

1.7 Radon in water

Ground water has much of radon than surface water because. Water has the ability to trap easily radon particles, hence underground water has much of radon particles [19]. Studying the properties of radon in ground water has an importance on both the health effect it can causes on human beings and the geological and hydrogeological contributions on the environment [20]. The radiological health risks posed by water borne radon and its progenies has significant effect on the human beings [21]. The radium in groundwater decayed from uranium (^{238}U) in rocks for example reaches houses with groundwater when pumped for household activities [22]. The exposure to high radon radiation has a carcinogenic effect to human organs including dissolved radon in ground waters [23]. The Commission of European Committee (CEC), suggests a concentration reference level of 100 Bq / l for radon in drinking water [24].

1.8 Radon entry in to the atmosphere

The main isotopes of radon, ^{222}Rn and ^{220}Rn are released from the solid matrix of the uranium material by recoil when the radium decays. For a radon atom to escape from the mineral grain into the pore space, the decay must occur within the recoil distance of the grain surface. The range of recoil distance for ^{222}Rn is 20-70 nanometers (nm) in common minerals, 100 nanometers (nm) in water, and 63 micrometers (μm) in air [25]. In addition to recoil distance, the location of radium atom in the mineral grain and the direction its recoil (whether it is to the surface or to the interior of the grain), determines the concentration of radon in the atmosphere. The process of radon atoms escape through rock or soil pore space from a radium-bearing grain is called the emanation coefficient, the emanation factor or the emanation power. Grain size and shape are two important factors that control the emanation of radon in soil. Microscopic fractures and fissures, called nano-pores, and pits or openings caused by previous radioactive decays provide additional pathways for radon release [25]. The main mechanism for the entry of radon into the atmosphere is by diffusion [25].

1.9 Radon in outdoor air

Meteorological and geological factors and the presence of local radon emitting sources determines the concentration of radon in out door environment [25]. Factors such as the strength of the source, the emanation rates, temperature, humidity and moisture, winds and the distance from the source, all have an impact on the radon concentration. There is evidence that moisture is the principle factor affecting the emanation rate and the highest radon levels are encountered during dry periods. Solar heating during the daytime tends to induce some turbulence, so that radon is more readily transported upwards [25].

1.10 Radon in indoor air

Cracks in solid foundations, construction joints, cracks in walls, gaps in suspended floors, cavities inside walls, and the water supply are the known entry mechanisms of radon into buildings [25]. The concentration of radon in water has direct relation to indoor radon

concentration. Radon concentrations in the same location may differ by a factor of two over a period of an hour [25]. The radon concentration level depends on the to the underlying rocks. The air flow and ventilation rates influence the radon concentration levels [25]. Hence, countries of the world may have different level of indoor radon concentrations, as shown in table (1.3) below .

<i>S.No.</i>	Country	Averege indoor radon level (Bq / m^3)
1	India(Bareilly)	87
2	Czech Republic	140
3	Spain	86
4	Finland	123
5	Australia	97
6	Belgium	48
7	France	68
8	Ireland	89
9	Italy	70
10	Luxembourg	115
11	Romania	112
12	Sweden	108
13	Turkey(Giresun)	130
14	USA	46

Table 1.3: Comparison of indoor radon levels with different countries, (Deepak Verma, 2013)

Factors such as design, construction, and ventilation of the house affect the pathways and sources that can draw radon indoors [26]. The chances of increased level of indoor radon and thoron are higher in areas of higher uranium laden soil. But always this may not be the case. Some houses in areas with lots of uranium in the soil have low levels of indoor ^{222}Rn and ^{220}Rn and other houses on uranium poor soils have high levels of indoor ^{222}Rn and ^{220}Rn [26]. Permeability of the soil can affect radon concentration. The presence of successful mitigation strategies, like identifying and sealing a limited number radon entry pathways or effectively ventilating the house can reduce radon concentration by about 30 %. Hybrid ventilation systems normally change radon concentration by less than 50 % [26]. Elevation has also its own impact on the concentration of radon. The concentration of radon gas is decreases with the elevation above the ground level. Ground floors have higher concentration of radon than higher floors [27]

1.11 Health hazards due to radon

Inhalation is the main means for the entry of radon into human body [28]. Most of the radon gas inhaled are also exhaled because of their longer half life. Some of them also can be cleared by the lung's natural defense system, swallowed or coughed out once they entered in to our body [29]. But, the daughters of radon (^{222}Rn) like, ^{218}Po , ^{214}Pb , ^{214}Bi , and ^{214}Po and that of daughters of radon ^{220}Rn like, (^{216}Po , ^{212}Pb , ^{212}Bi , and ^{212}Po) attach to aerosols and can be inhaled in to our lungs. Most of the above short lived radon progenies are carcinogenic and can cause series health problems, except for $\text{Pb} - 210$, because it has longer radiological and biological half-life [29].

The inhaled radionuclides impose radiation dose on our body and it can be also as high as 59% of the total radiation dose received from the natural background radiation [29]. There are two types of biological effects due to radiation exposure. The stochastic effect and the deterministic effect [29].

- **Stochastic effect:** In stochastic effect, the effect occurs by chance and without a threshold level of dose, whose probability is proportional to the dose and whose severity is independent of the dose. The chance of the effect occurring is statistical in nature. Examples are cancer and genetic effects [29].

- **Deterministic effects:** In deterministic effects, both the incidence and the severity increase above a threshold level with increasing dose. It has a threshold level of dose magnitude of 0.1 gy, below which the effect doesn't occurs [29]. In this case, groups of cells or tissues are extensively damaged due to exposure to very high radiation doses and the effects may appear within short period of time, even in an hours or in a few weeks [30]. The exposures to radiation other than cancer and genetic effects are deterministic. The heavily ionizing and short range particles of radon daughter products, like polonium ^{214}Po with energy of ($E = 7.7\text{ MeV}$) and the other polonium particle, ^{218}Po with energy ($E = 6.0\text{ MeV}$) can cause deterministic effects in a body of an organism [31].

The study also estimated that the risk for smokers versus non smokers is 25 times higher

[32]. Radon related health risk is the second for smokers and the first for non smokers carcinogen, according to the classification used by the World Health Organization [31, 32]. World Health Organization recommended to set a national reference level of 100 Bq / m³ as a minimum and 200 Bq / m³ as a maximum one for radon concentration [33].

1.12 Radon and its radiations

1.12.1 Ionizing radiations

Ionizing radiation is radiation with enough energy so that during an interaction with an atom in the form of particles or waves, it can remove tightly bound electrons from the orbit of an atom, causing the atom to become charged or ionized [34]. Examples of ionizing radiation are alpha particles, beta particles, positrons, gamma rays, and X-rays [34]. The breakdown of uranium, thorium and actinium results radiation of alpha and beta particles, including gamma rays. Radiation can be either from natural sources or man-made radio nuclide [35].

Radon have been existing on the earth's atmosphere being one the type of the naturally occurring radioactive material (NORM) and adds over 50% of radiation exposure to public as shown in the figure (1.3), below.

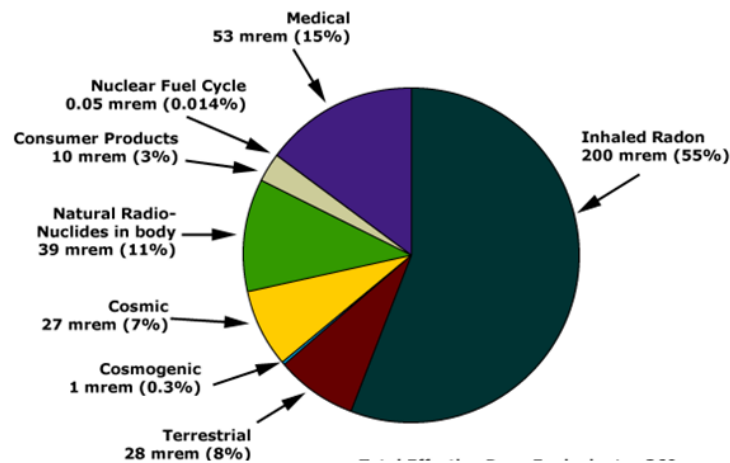


Figure 1.3: Radiation exposure levels in percent

1.12.2 Non ionizing radiations

Non-ionizing radiation is any type of electromagnetic radiation that does not have enough energy per quantum (photon energy) to ionize and to remove completely an electron from an atom. Examples of non-ionizing radiation includes visible light, infrared, ultraviolet ray microwaves, radio waves and radio frequency energy from cell phones. Most types of non-ionizing radiation have not been found to cause cancer [35]. Instead of producing charged ions when passing through matter, non-ionizing radiation has sufficient energy only to exit an electron to a higher energy state [35].

1.13 Radioactive decay chain

Radon, a radioactive gas, it decays and produces radon progenies, also known as radon daughters or decay products. Radon decays by emitting alpha and beta particles and goes from the gaseous state into being metallic polonium, bismuth and lead daughter particles [28, 35]. The intermediate radioactive decay product in the Uranium (^{238}U), actinium (^{235}U) and the Thorium (^{232}Th) are ^{222}Rn , ^{219}Rn and ^{220}Rn respectively [28, 35].

The aggregate mean life of the daughter products of radon ^{222}Rn which are (^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po) is 71.57 min, the corresponding aggregate mean life of thoron ^{220}Rn with bi-products of (^{216}Po , ^{212}Pb , ^{212}Bi , ^{212}Po) is 16.81 h and the aggregate mean life of ^{219}Rn with bi- products of (^{215}Po , ^{211}Pb , ^{211}Bi , ^{207}Tl) is 1.03 h. The ^{238}U , ^{232}Th and ^{235}U series with the half lives, decay mode, energy of emission and yield of the nuclear decay are given below in fig (1.4), fig (1.5) and fig (1.6) respectively.

Decay chain of uranium series

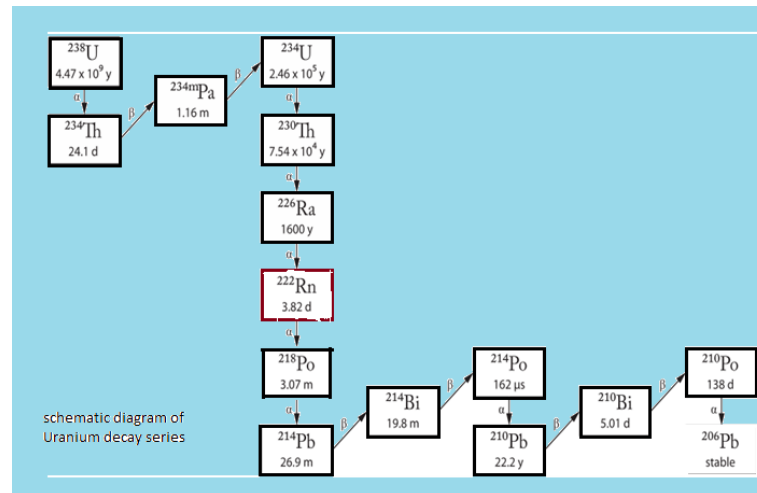


Figure 1.4: Uranium decay series (EPA, 2005)

Decay chain of Thorium series .

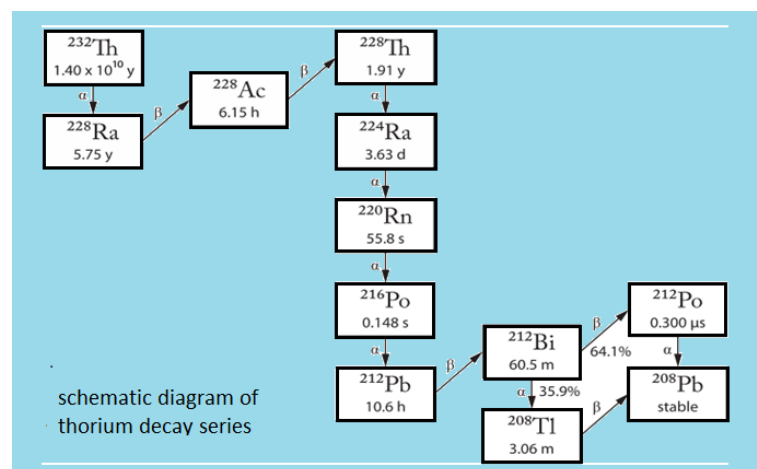


Figure 1.5: Thorium decay series (EPA, 2005)

Decay chain of actinium series .

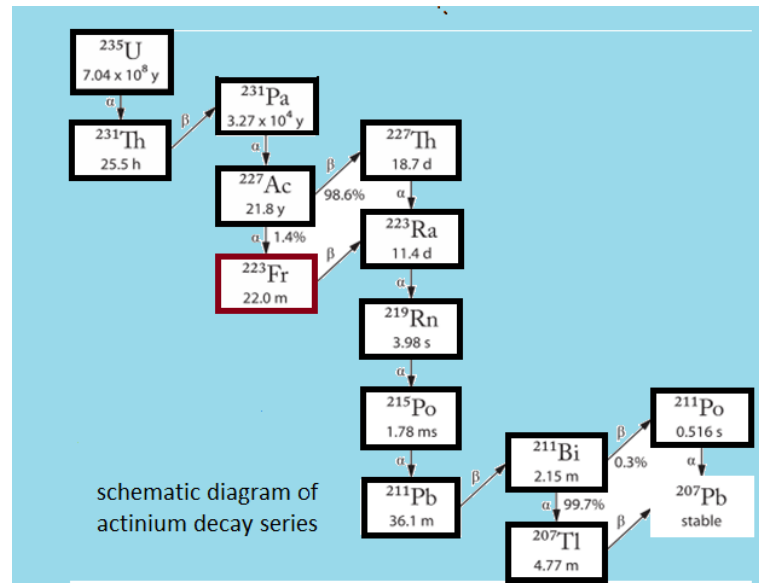


Figure 1.6: Actinium decay series (EPA, 2005)

1.14 Rationale of the study

High concentration of radon, thoron and their progeny are carcinogenic. From the health point of view, one must be aware of his / her environs radon, thoron and their progenies concentration and take the necessary steps to reduce their levels by knowing the devices and the methods used to measure their concentrations. Measurement of radon concentrations using filtered air samples is simple active method from the point of view of instrument to determine daughter concentration in air. However, it requires long time observation and analysis of data is cumbersome.

1.15 Objectives of the study

The work of this project focuses on review of measurement of activity concentrations of radon daughter products from filtered air using beta counter detector, an end window Geiger - Müller (GM) counter with the Least Square Weighted Method (LSWM) and the Steady State Jaccobi's Model (SSJM).

1.15.1 General objectives

- ◆ To review differential equations used to calculate concentrations of radon daughter products from filtered air.

- ◆ To compare determination of concentrations of radon daughter products from filtered air using the Least Square Weighted Method (LSWM) and the Steady State Jaccobi's Model (SSJM).

1.15.2 Specific objectives

- ◆ To review techniques of measurement of radon daughter products from filtered air using beta counter detector, an end window Geiger - Müller (GM).

- ◆ To compare radon daughter products determination efficiencies using the Least Square Weighted Method (LSWM) and the Steady State Jaccobi's Model (SSJM).

Chapter 2

Review of radon measurement related literatures

2.1 Dosimetry of radon

Radiation dosimetry is the measurement, calculation and assessment of ionizing radiation dose absorbed by an object or by the human body [36]. Measuring the concentration of radon and thoron concentration enables us to know the level of exposures to their by - products to cause health effects [36].

There are three naturally existing radon (Rn) isotopes in the environment which are ^{222}Rn , a member of the uranium decay series, ^{220}Rn , a member of the thorium decay series, and ^{219}Rn , a member of the actinium decay series. The two radon isotopes, which are ^{220}Rn and ^{219}Rn , often referred to as thoron and actinon respectively [36]. Actinon, due to its short half life, (3.98 sec) and its very low radioactivity concentrations, its radiological impact can be taken negligible compared with that of ^{222}Rn in the environment. The impact of thoron, (^{220}Rn), is also minimal because of its half life 55.6 seconds. Relatively the impact of thoron (^{220}Rn) compared with actinon (^{219}Rn) is significant [36].

Radon and thoron are identified by their radiation or their daughter products. Rn - 222 is produced by the radioactive decay of Ra - 226, which is part of the decay chain of U - 238. Thus any material containing radium or uranium, for example, soil and mineral deposits, is a potential source of radon [36]. Since Ra - 226 has a half-life of over 1600 years, its concentration is practically invariant with time and its decay product (Rn - 222), is constant. This makes radon is present everywhere in the environment [36].

2.2 Radon concentration measuring methods

A number of studies have been conducted to know the concentration of radon isotopes in different places since their discovery. The effects of its radiation on matter and the properties of the emitters are the two important things to be studied during radon measurement techniques [37]. Although many studies on radon (^{222}Rn) were conducted all over the world, there have only been a few studies on thoron (^{220}Rn) [37]. In some areas of the world studies have shown high concentrations of thoron (^{220}Rn), although its concentration exponentially decreases with a distance from its source because of its short half - life (55.6 sec). The concentrations of radon (^{222}Rn) and thoron (^{220}Rn) are independent of each other and no clear correlations between them. The concentration of thoron could have been predicted from widely available radon information [38]. Atmospheric radon (^{222}Rn) and thoron (^{220}Rn), originates exclusively from the earth's surface [39].

Radon, thoron and their progeny measurement techniques are generally classified as active and passive methods. In active measurement method, the radon and thoron concentrations are measured at a given measuring point in time and analysis is done on the gas samples by pumping of a known volume of air. In the passive radon and thoron concentrations measuring methods, the measurement is usually made over a long period of time and the result is reported being a mean over the measured time intervals. The sampling techniques are obtained by the natural diffusion of gases [40]. In this chapter both methods are discussed below.

2.3 Passive radon concentration measuring methods

The passive radon concentration measuring devices do not need power to function. They work by simply absorbing radon in the air. Electrets ion chamber (EC), Activated charcoal (AC), Solid State Nuclear Track Detector (SSNTD) and Charcoal liquid scintillation (LS) methods are representative examples of passive radon concentration measuring methods.

2.3.1 Electret ion chamber (EIC)

Electret Ion Chambers are made up of both an electret and an ionization chamber. The ionization chamber is constructed of electrically - conductive plastic, with a volume and shape that has been very accurately characterized and it displays strong anti-static properties (that prevents an electrical charge from accumulating on the chamber's exterior and interior surfaces) [41].

The second component is the electret, (electrically positively charged Teflon disc) mounted inside an electrically conducting chamber, which is housed in an electrically - conductive plastic holder. When the electret is loaded into an ionization chamber, an Electret Ion Chamber (known as an E - PERM[®]) is created. As radon decay inside the ionization chamber alpha and beta particles are released fly through the ionization chamber, strike other molecules in the air. This effect is called ionization and results air molecule to be charged either a positively or negatively. The electret serves both as a source of the electric field and as a sensor.

The ions formed inside the chamber are collected by the electret, as shown in fig (2.1).



Figure 2.1: Electret ion chamber (EIC)

These negatively charged ions are attracted to the positively charged electret surface, and a reduction in voltage occurs. The radon concentration of a given environment is a direct function of the reduction in voltage over a known time period. EICs can be deployed for exposure periods of two days to 12 months, depending upon the thickness of the electrets and the volume of the ion chamber.

2.3.2 Activated charcoal (AC)

For this method, an airtight container with activated charcoal is opened in the area to be sampled and radon in the air adsorbs onto the charcoal granules. At the end of the sampling period, the container is sealed and may be sent to a laboratory for analysis. This method needs low cost, easy operation, measures small volume. It is not suitably used for outdoor and humidity place. Timely analysis is needed after sampling (less than 7 days) [41].

2.3.3 Solid State Nuclear Track Detector (SSNTD)

Solid State Nuclear Track Detector (SSNTD) also known as etched track detector or a dielectric track detector, consists of a small piece of plastic or film enclosed in a container. When radon and thoron diffuses into the container and strike the detector they produce a narrow trail of damage tracks about $50\text{\AA}$ in diameter along its path. This is called ‘latent track’ because it cannot be seen with our naked eyes. At the end of the measurement period, the detectors are returned to the laboratory and in a caustic solution (NaOH) that accentuates and enlarge the damaged tracks so they can be counted using an automated counting system [41]. The number of tracks per unit of analyzed detector area produced per unit of time minus the background radiation is proportional to the radon concentration [41], as shown in fig (2.2) below.

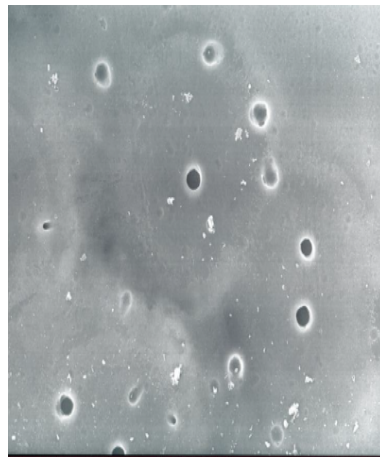


Figure 2.2: A picture of damaged part after etching

2.3.4 Charcoal liquid scintillation (CLS)

The charcoal liquid scintillation devices consist of a polyethylene vial with an active charcoal bag, filter paper and a desiccant. The radon gas and some of the progeny will attach to the charcoal surface. At the end of the exposure (typically 2 to 7 days), the device is resealed securely and returned to the laboratory for analysis.

2.4 Active measuring methods

In this category of measurement methods, some of the active measuring methods, like: Grab radon sampling method, Pump / collapsible bag method, Double - filter method are the prominent one to be discussed particularly in this work.

2.4.1 Pump / collapsible bag (PB)

It is one of the older and simpler methods of making an integrated measurement of the concentration of radon over a period of time. A sample bag impervious to radon is filled over a 24 hour period. This is usually accomplished by a pump programmed to pump small amounts of air at predetermined intervals during the sampling period. After sampling, analysis procedures takes place.

2.4.2 Double - filter method

The double - filter method is the instantaneous measurement of radon which belongs to active sampling method. It can measure radon and radon daughters at same time. Using this method, the measurement time is short, highly sensitive and gives accurate values. The detection limit was $3.3 \text{ Bq} / \text{m}^3$.

2.4.3 Grab radon / scintillation cell (GS) method

This device has sealed flask or cell that has zinc sulfide phosphor coating on its interior surfaces and a sample of air is pumped into. The window end of the cell is placed on a photomultiplier tube to count the scintillations (light pulses) produced when alpha particles from radon decay strike the zinc sulfide coating on the inside of the cell. The number of pulses are proportional to the radon concentration in the cell.

2.4.4 Grab radon / activated charcoal (GC) method

Air is pumped through activated charcoal. The pump with a charcoal cartridge is not flow dependent but must remain operational at the sampling location until the charcoal collects enough radon to be in equilibrium with the radon at the sampling collection. Depending on the cartridge design and airflow, sampling takes from 15 minutes to 1 hour. A sampling duration of 1 hour has been found to be optimal for most systems. After sampling, the cartridge is placed in a sealed container and taken to a laboratory.

2.4.5 Grab radon pump / collapsible bag (GB) method

This method uses a sample bag made of material impervious to radon. At the sample site, a skilled technician using a portable pump fills the bag with air, and then transports it to the laboratory for analysis.

2.4.6 Grab radon sampling using Geiger - Muller counter

This is another method of grab sampling which is based on direct beta counting of filtered aerosol sample over successive time intervals by end-window Geiger - Müller counter. The method is used for simultaneous determination of radon and thoron decay products. The concentrations of radon daughter products can be analyzed based on the Weighted Least Square Method. The work of this paper focuses on this method.

Chapter 3

Experimental method

3.1 Data gathering methods

To measurement concentrations of radon daughter products we use a differential mathematical formulas. The mathematical formulas for the activity concentration of radon progenies and gross beta counting of a filtered aerosol sample over successive time intervals by an end - window Geiger - Müller (GM) counter can be analyzed. The determination techniques of their activity concentrations is based on the following procedures:

1. Sampling known volume aerosol particles and capture on a glass - fiber filter with high retention efficiency, greater than 99 % like Sartorius, SM 1340, for a fixed flow rate.
2. Placing the filtered air sample under an end-window (GM) counting system, with in a couple of minutes after the end of sampling.
3. Record the gross count in successive intervals of time for sufficiently large number of intervals until the end of the measuring time interval.
4. Analysis of the count rate versus time and fitting of experimental points with the mathematical model.
5. Determination of EECs from the best - fit of experimental points and the mathematical model.

3.2 Geiger - Müller (GM) counting system

Materials used in the experimental method of sampling air are listed as follow.

- ◇ Beta detector

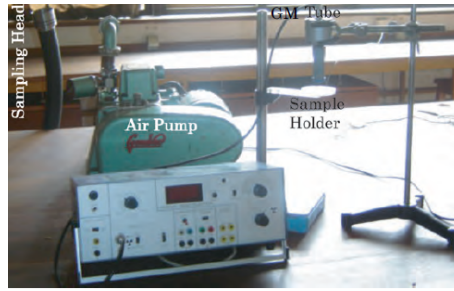


Figure 3.1: Basic materials used to sample aerosols, adapted from Tsega Birhan's paper

- ◇ Geiger - Müller counter (GM)
- ◇ Sampling head
- ◇ End window Gm tube
- ◇ Stop watch
- ◇ Shielded (leads)
- ◇ Filter holder (shelve)
- ◇ Power Supply unit
- ◇ Air suction pump
- ◇ Glass fiber filter

3.2.1 Sampling the air

Measurements of radon enriched aerosols can be taken in a room of a sampling site. Because air molecules are able to diffuse easily, the doors and windows of the sampling site must be closed before taking air samples. The air sampling technique can be using a special instrument, called, a suction pump fitted with a sampling head designed for this purpose as shown in (fig 3.1) below.

Closing doors and windows of a sampling site can result to minimize the air ventilation rate. The magnitude of air flow rate through a suction pump to suck air can be fixed at the beginning of sampling. A What man®-glass fiber filter paper, which has more than 99% retention power of aerosols can be used to trap aerosols [41]. Finally the filter paper has to be carefully removed from the sampling head, suspended at a certain height above the floor and place under the (GM) tube to take counting of beta particles.

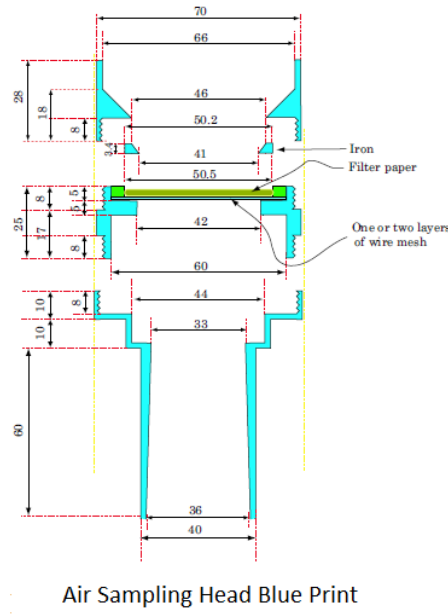


Figure 3.2: The sampling head in blue print & photograph, adapted from Tsega Birhan's paper

3.3 Counting the activities on the filtered samples

With in short period of time, not more than one or two minutes after sampling is ended, the filter paper has to be placed below the Geiger - Müller (GM) tube. Counting of the activity can be done using a GM tube counting system. The back ground counting rate has to be also measured.

3.4 Mathematical modeling of the counts

Defined Solid Angle Absolute Beta Counting (DSAABC) method has to be used to measure the activities of the filtered air samples. Some of the features are outlined below. Activity concentration of airborne radon and thoron decay products are determined by similar mathematical computations as follows [41].

1. Using differential equations which describe the buildup and the decay during sampling of the daughters.
2. Using differential equations which describe the decay of the daughters on the sample

after collection is ended.

Three radon and three thoron progenies in total of six and a long lived beta emitter from the unknown source were assumed to be determined. Let the numbers 1 up to 7 represent ^{218}Po , ^{214}Pb and ^{214}Bi for radon decay products and ^{212}Pb , ^{212}Bi and ^{208}Tl for thoron decay products and U for unknown beta emitters with long half life respectively.

3.4.1 Nomenclature

The radon decay products are represented from 1 up to 3, as follow

$$\begin{aligned} 1 &\Longleftrightarrow ^{218}\text{Po} \\ 2 &\Longleftrightarrow ^{214}\text{Pb} \\ 3 &\Longleftrightarrow ^{214}\text{Bi} \end{aligned}$$

The thoron decay products are represented from 4 up to 6, as follow

$$\begin{aligned} 4 &\Longleftrightarrow ^{212}\text{Pb} \\ 5 &\Longleftrightarrow ^{212}\text{Bi} \\ 6 &\Longleftrightarrow ^{208}\text{Tl} \end{aligned}$$

Unknown β emitters with very long and very short half lives are represented by number 7

$$7 \Longleftrightarrow U$$

3.4.2 Assumed constants in the mathematical equations

T = air sampling time.

η = air sampling flow rate.

t_b = time at the beginning of a counting interval.

t_e = time at the end of a counting interval.

C = total β counts over the interval (t_b, t_e).

L = number of counting intervals.

X = β counting efficiencies for the isotopes one by one ($i = 2, 3, \dots, 7$);

λ_i = decay constants ($i = 1, 2, \dots, 7$).

3.4.3 Variables

n_i = numbers of atoms in a unit volume of the sampled air, ($i = 1, 2, \dots, 7$);

a_i = activities of isotopes in a unit volume of the sampled air, ($i = 1, 2, \dots, 7$);

N_i = numbers of atoms in the filter, ($i = 1, 2, \dots, 7$);

N_{T_i} = numbers of atoms in the filter at the end of sampling, ($i = 1, 2, \dots, 7$);

t = time elapsed since the end of sampling ($t = 0$ at the end of same sampling);

CR = total beta counting rate.

During sampling period, the values of η and n_i are taken to be constant. In the mean time of sampling of aerosols, we get both building up and decaying daughter products. The differential equations which represents the buildup and decay of atoms on the sample are given below:

$$dN_1 = \eta n_1 dt - \lambda_1 N_1 dt \quad N_1(0) = (0) \quad (3.1a)$$

$$dN_2 = \eta n_2 dt - \lambda_2 N_2 dt + \lambda_1 N_1 dt \quad N_2(0) = (0) \quad (3.1b)$$

$$dN_3 = \eta n_3 dt - \lambda_3 N_3 dt + \lambda_2 N_2 dt \quad N_3(0) = (0) \quad (3.1c)$$

$$dN_4 = \eta n_4 dt - \lambda_4 N_4 dt \quad N_4(0) = (0) \quad (3.1d)$$

$$dN_5 = \eta n_5 dt - \lambda_5 N_5 dt + \lambda_4 N_4 dt \quad N_5(0) = (0) \quad (3.1e)$$

$$dN_6 = \eta n_6 dt - \lambda_6 N_6 dt + \lambda_5 N_5 dt \quad N_6(0) = (0) \quad (3.1f)$$

$$dN_7 = \eta n_7 dt - \lambda_7 N_7 dt \quad N_7(0) = (0) \quad (3.1g)$$

The solution for the buildup and decay equations shown in equations 3.1 above are given by Equations 3.2 as follow:

$$N_1 = \eta n_1 \left[\frac{1 - e^{-\lambda_1 t}}{\lambda_1} \right] \quad (3.2a)$$

$$N_2 = \eta n_1 \left[\frac{1 - e^{-\lambda_2 t}}{\lambda_2} - \frac{e^{-\lambda_2 t} - e^{-\lambda_1 t}}{\lambda_1 - \lambda_2} \right] + \eta n_2 \left[\frac{1 - e^{-\lambda_2 t}}{\lambda_2} \right] \quad (3.2b)$$

$$\begin{aligned} N_3 = & \eta n_1 \left[\frac{1 - e^{-\lambda_3 t}}{\lambda_3} - \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{\lambda_3 - \lambda_2} - \frac{\lambda_2 (e^{-\lambda_2 t} - e^{-\lambda_3 t})}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{\lambda_2 (e^{-\lambda_2 t} - e^{-\lambda_3 t})}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \right] \\ & + \eta n_2 \left[\frac{1 - e^{-\lambda_3 t}}{\lambda_3} - \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{\lambda_3 - \lambda_2} \right] + \eta n_3 \left[\frac{1 - e^{-\lambda_3 t}}{\lambda_3} \right] \end{aligned} \quad (3.2c)$$

$$N_4 = \eta n_4 \left[\frac{1 - e^{-\lambda_4 t}}{\lambda_4} \right] \quad (3.2d)$$

$$N_5 = \eta n_4 \left[\frac{1 - e^{-\lambda_5 t}}{\lambda_5} - \frac{e^{-\lambda_5 t} - e^{-\lambda_4 t}}{\lambda_4 - \lambda_5} \right] + \eta n_5 \left[\frac{1 - e^{-\lambda_5 t}}{\lambda_5} \right] \quad (3.2e)$$

$$\begin{aligned} N_6 = & \eta n_4 \left[\frac{1 - e^{-\lambda_6 t}}{\lambda_6} - \frac{e^{-\lambda_5 t} - e^{-\lambda_6 t}}{\lambda_6 - \lambda_5} - \frac{\lambda_5 (e^{-\lambda_5 t} - e^{-\lambda_6 t})}{(\lambda_6 - \lambda_5)(\lambda_5 - \lambda_6)} + \frac{\lambda_5 (e^{-\lambda_6 t} - e^{-\lambda_4 t})}{(\lambda_4 - \lambda_6)(\lambda_4 - \lambda_5)} \right] \\ & + \eta n_5 \left[\frac{1 - e^{-\lambda_6 t}}{\lambda_6} - \frac{e^{-\lambda_5 t} - e^{-\lambda_6 t}}{\lambda_6 - \lambda_5} \right] + \eta n_6 \left[\frac{1 - e^{-\lambda_6 t}}{\lambda_6} \right] \end{aligned} \quad (3.2f)$$

$$N_7 = \eta n_7 \left[\frac{1 - e^{-\lambda_7 t}}{\lambda_7} \right] \quad (3.2g)$$

N_{Ti} ($i = 1, 2, \dots, 7$), which is number of atom in the filter at the end of sampling can be expressed by replacing t by T , because end of sampling time is equal to the air sampling time (T). Hence, N_{Ti} ($i = 1, 2, \dots, 7$) can be expressed as follow

$$N_{T1} = \eta n_1 \left[\frac{1 - e^{-\lambda_1 T}}{\lambda_1} \right] \quad (3.3a)$$

$$N_{T2} = \eta n_1 \left[\frac{1 - e^{-\lambda_2 T}}{\lambda_2} - \frac{e^{-\lambda_2 T} - e^{-\lambda_1 T}}{\lambda_1 - \lambda_2} \right] + \eta n_2 \left[\frac{1 - e^{-\lambda_2 T}}{\lambda_2} \right] \quad (3.3b)$$

$$N_{T3} = \eta n_1 \left[\frac{1 - e^{-\lambda_3 T}}{\lambda_3} - \frac{e^{-\lambda_2 T} - e^{-\lambda_3 T}}{\lambda_3 - \lambda_2} - \frac{\lambda_2(e^{-\lambda_2 T} - e^{-\lambda_3 T})}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{\lambda_2(e^{-\lambda_2 T} - e^{-\lambda_3 T})}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \right] \\ + \eta n_2 \left[\frac{1 - e^{-\lambda_3 T}}{\lambda_3} - \frac{e^{-\lambda_2 T} - e^{-\lambda_3 T}}{\lambda_3 - \lambda_2} \right] + \eta n_3 \left[\frac{1 - e^{-\lambda_3 T}}{\lambda_3} \right] \quad (3.3c)$$

$$N_{T4} = \eta n_4 \left[\frac{1 - e^{-\lambda_4 T}}{\lambda_4} \right] \quad (3.3d)$$

$$N_{T5} = \eta n_4 \left[\frac{1 - e^{-\lambda_5 T}}{\lambda_5} - \frac{e^{-\lambda_5 T} - e^{-\lambda_4 T}}{\lambda_4 - \lambda_5} \right] + \eta n_5 \left[\frac{1 - e^{-\lambda_5 T}}{\lambda_5} \right] \quad (3.3e)$$

$$N_{T6} = \eta n_4 \left[\frac{1 - e^{-\lambda_6 T}}{\lambda_6} - \frac{e^{-\lambda_5 T} - e^{-\lambda_6 T}}{\lambda_6 - \lambda_5} - \frac{\lambda_5(e^{-\lambda_5 T} - e^{-\lambda_6 T})}{(\lambda_6 - \lambda_5)(\lambda_5 - \lambda_6)} + \frac{\lambda_5(e^{-\lambda_6 T} - e^{-\lambda_4 T})}{(\lambda_4 - \lambda_6)(\lambda_4 - \lambda_5)} \right] \\ + \eta n_5 \left[\frac{1 - e^{-\lambda_6 T}}{\lambda_6} - \frac{e^{-\lambda_5 T} - e^{-\lambda_6 T}}{\lambda_6 - \lambda_5} \right] + \eta n_6 \left[\frac{1 - e^{-\lambda_6 T}}{\lambda_6} \right] \quad (3.3f)$$

$$N_{T7} = \eta n_7 \left[\frac{1 - e^{-\lambda_7 T}}{\lambda_7} \right] \quad (3.3g)$$

n_i ($i = 1, 2, \dots, 7$) can be expressed from equations (3.3 a) to (3.3 g) as follows:

$$n_1 = \frac{\lambda_1 N_{T1}}{\eta [1 - e^{-\lambda_1 T}]} \quad (3.4a)$$

$$n_2 = \frac{\lambda_2 N_{T2}}{\eta [1 - e^{-\lambda_2 T}]} - n_1 \left[1 - \frac{\lambda_2(e^{-\lambda_2 T} - e^{-\lambda_1 T})}{[1 - e^{-\lambda_2 T}](\lambda_1 - \lambda_2)} \right] \quad (3.4b)$$

$$n_3 = \frac{\lambda_3 N_{T3}}{\eta [1 - e^{-\lambda_3 T}]} - n_2 \left[1 - \frac{\lambda_3(e^{-\lambda_2 T} - e^{-\lambda_3 T})}{[1 - e^{-\lambda_3 T}](\lambda_3 - \lambda_2)} \right] - n_1 \left[1 - \frac{\lambda_3(e^{-\lambda_2 T} - e^{-\lambda_3 T})}{[1 - e^{-\lambda_3 T}](\lambda_3 - \lambda_2)} \right] \\ - \frac{\lambda_2 \lambda_3 (e^{-\lambda_2 T} - e^{-\lambda_3 T})}{(\lambda_1 - \lambda_2) [1 - e^{-\lambda_3 T}] (\lambda_3 - \lambda_2)} \\ + \frac{\lambda_2 \lambda_3 (e^{-\lambda_3 T} - e^{-\lambda_1 T})}{\lambda_1 - \lambda_2 [1 - e^{-\lambda_3 T}] (\lambda_1 - \lambda_3)} \quad (3.4c)$$

$$n_4 = \frac{\lambda_4 N_{T4}}{\eta [1 - e^{-\lambda_4 T}]} \quad (3.4d)$$

$$n_5 = \frac{\lambda_5 N_{T5}}{\eta [1 - e^{-\lambda_5 T}]} - n_4 \left[1 - \frac{\lambda_5(e^{-\lambda_5 T} - e^{-\lambda_4 T})}{[1 - e^{-\lambda_5 T}](\lambda_4 - \lambda_5)} \right] \quad (3.4e)$$

$$\begin{aligned}
n_6 = & \frac{\lambda_6 N_{T5}}{\eta[1 - e^{-\lambda_5 T}]} - 0.36n_5 \left[1 - \frac{\lambda_6(e^{-\lambda_5 T} - e^{-\lambda_6 T})}{[1 - e^{-\lambda_6 T}](\lambda_6 - \lambda_5)} \right] - 0.36n_4 \left[1 - \frac{\lambda_6(e^{-\lambda_5 T} - e^{-\lambda_6 T})}{[1 - e^{-\lambda_6 T}](\lambda_6 - \lambda_5)} \right. \\
& - \frac{\lambda_2 \lambda_3 (e^{-\lambda_2 T} - e^{-\lambda_3 T})}{(\lambda_1 - \lambda_2)(1 - e^{-\lambda_3 T})(\lambda_3 - \lambda_2)} \\
& \left. + \frac{\lambda_5 \lambda_6 (e^{-\lambda_6 T} - e^{-\lambda_4 T})}{(\lambda_4 - \lambda_6)(1 - e^{-\lambda_6 T})(\lambda_4 - \lambda_5)} \right] \quad (3.4f)
\end{aligned}$$

$$n_7 = \frac{\lambda_7 N_{T7}}{\eta[1 - e^{-\lambda_7 T}]} \quad (3.4g)$$

3.4.4 Decay after sampling

After the end of sampling, the differential equations describing the decay of each radionuclides are solved with known boundary conditions: $N_i(t) = N_{Ti}$, at $t = 0$ and the decay parameters of the deposited radionuclides to obtain the number of radionuclides $N_i(t)$ on the filter at any latter time t . In the case of $N_6(t)$, due to the fact that only 36 % of ^{212}Bi ($i=5$) and therefore ^{212}Pb ($i=4$) decay via ^{208}Tl because of its branching decay, we introduce a factor of 0.36. Thus:

$$N_1(t) = N_{T1} e^{-\lambda_1 t} \quad (3.5a)$$

$$N_2(t) = N_{T1} \left[\frac{\lambda_1 (e^{-\lambda_2 t} - e^{-\lambda_1 t})}{\lambda_1 - \lambda_2} \right] + N_{T2} e^{-\lambda_2 t} \quad (3.5b)$$

$$\begin{aligned}
N_3(t) = & N_{T1} \left[\frac{\lambda_2 \lambda_1 (e^{-\lambda_2 t} - e^{-\lambda_3 t})}{(\lambda_3 - \lambda_2)(\lambda_1 - \lambda_2)} - \frac{\lambda_2 \lambda_1 (e^{-\lambda_3 t} - e^{-\lambda_1 t})}{(\lambda_1 - \lambda_3)(\lambda_1 - \lambda_2)} \right. \\
& \left. + N_{T2} \left[\frac{\lambda_2 (e^{-\lambda_2 t} - e^{-\lambda_3 t})}{\lambda_3 - \lambda_2} \right] + N_{T3} e^{-\lambda_3 t} \right] \quad (3.5c)
\end{aligned}$$

$$N_4(t) = N_{T4} e^{-\lambda_4 t} \quad (3.5d)$$

$$N_5(t) = N_{T4} \left[\frac{\lambda_4 (e^{-\lambda_5 t} - e^{-\lambda_4 t})}{\lambda_4 - \lambda_5} \right] + N_{T5} e^{-\lambda_5 t} \quad (3.5e)$$

$$N_6(t) = 0.36N_{T4} \left[\frac{\lambda_4\lambda_5(e^{-\lambda_5t} - e^{-\lambda_6t})}{(\lambda_6 - \lambda_5)(\lambda_4 - \lambda_5)} - \frac{\lambda_4\lambda_5(e^{-\lambda_6t} - e^{-\lambda_4t})}{(\lambda_4 - \lambda_6)(\lambda_4 - \lambda_5)} \right] + 0.36N_{T5} \left[\frac{\lambda_5(e^{-\lambda_5t} - e^{-\lambda_6t})}{\lambda_6 - \lambda_5} \right] + N_{T6}e^{-\lambda_6t} \quad (3.5f)$$

$$N_7(t) = N_{T7}e^{-\lambda_7t} \quad (3.5g)$$

3.4.5 The β count Rate (CR)

The gross β - count, at a given time interval is the sum of the contributions by each beta emitter in the series and by an unknown beta emitter (if any) deposited on the filter paper. Therefore the count rate at a given time t is given by [42]:

$$CR = \sum_{i=2}^7 \varepsilon_i \lambda_i N_i \quad (3.6)$$

where, ε_i is the absolute detection efficiency of the detection system for the β - particles emitted by radionuclide i and given by $\varepsilon = CR/A$. In the above equation polonium (^{218}Po) is not taken in to consideration because mostly it is alpha emitter. Replacing N_i by substituting equation (3.2) to (3.3) and integrating with respect to time 't' between t_b and t_e , it becomes:

$$C = \sum_{i=1}^7 N_{Ti} Y_i \quad (3.7)$$

Where,

$$Y_1 = \left[\frac{\varepsilon_2 \lambda_2 (e^{-\lambda_1 t_b} - e^{-\lambda_1 t_e})}{(\lambda_2 - \lambda_1)} - \frac{\varepsilon_2 \lambda_1 (e^{-\lambda_2 t_b} - e^{-\lambda_2 t_e})}{\lambda_2 - \lambda_1} + \frac{\varepsilon_3 \lambda_2 \lambda_3 (e^{-\lambda_1 t_b} - e^{-\lambda_1 t_e})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} - \frac{\varepsilon_3 \lambda_1 \lambda_3 (e^{-\lambda_2 t_b} - e^{-\lambda_2 t_e})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} + \frac{\varepsilon_3 \lambda_1 \lambda_2 (e^{-\lambda_3 t_b} - e^{-\lambda_3 t_e})}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)} \right] \quad (3.8a)$$

$$Y_2 = \varepsilon_2 (e^{-\lambda_2 t_b} - e^{-\lambda_2 t_e}) + \frac{\varepsilon_3 \lambda_3 (e^{-\lambda_2 t_b} - e^{-\lambda_2 t_e})}{\lambda_3 - \lambda_2} - \frac{\varepsilon_3 \lambda_2 (e^{-\lambda_3 t_b} - e^{-\lambda_3 t_e})}{\lambda_3 - \lambda_2} \quad (3.8b)$$

$$Y_3 = \varepsilon_3 (e^{-\lambda_3 t_b} - e^{-\lambda_3 t_e}) \quad (3.8c)$$

$$Y_4 = \varepsilon_4 (e^{-\lambda_4 t_b} - e^{-\lambda_4 t_e}) + \left[\frac{\varepsilon_5 \lambda_5 (e^{-\lambda_4 t_b} - e^{-\lambda_4 t_e})}{(\lambda_5 - \lambda_4)} - \frac{\varepsilon_5 \lambda_4 (e^{-\lambda_5 t_b} - e^{-\lambda_5 t_e})}{\lambda_5 - \lambda_4} + \frac{0.36\varepsilon_6 \lambda_5 \lambda_6 (e^{-\lambda_4 t_b} - e^{-\lambda_4 t_e})}{(\lambda_5 - \lambda_4)(\lambda_6 - \lambda_4)} - \frac{0.36\varepsilon_6 \lambda_4 \lambda_6 (e^{-\lambda_5 t_b} - e^{-\lambda_5 t_e})}{(\lambda_5 - \lambda_4)(\lambda_6 - \lambda_5)} + \frac{0.36\varepsilon_6 \lambda_4 \lambda_5 (e^{-\lambda_6 t_b} - e^{-\lambda_6 t_e})}{(\lambda_6 - \lambda_5)(\lambda_6 - \lambda_4)} \right] \quad (3.8d)$$

$$Y_5 = \varepsilon_5(e^{-\lambda_4 tb} - e^{-\lambda_4 te}) + \frac{0.36\varepsilon_6\lambda_6(e^{-\lambda_5 tb} - e^{-\lambda_5 te})}{\lambda_6 - \lambda_5} - \frac{0.36\varepsilon_6\lambda_5(e^{-\lambda_6 tb} - e^{-\lambda_6 te})}{\lambda_6 - \lambda_5} \quad (3.8e)$$

$$Y_6 = \varepsilon_6(e^{-\lambda_6 tb} - e^{-\lambda_6 te}) \quad (3.8f)$$

$$Y_7 = \varepsilon_7(e^{-\lambda_7 tb} - e^{-\lambda_7 te}) \quad (3.8g)$$

Assume that the total beta counts were measured over L successive time intervals. This series of measurements can be described by a series of linear equations as:

$$C_L = \sum_{i=1}^7 N_{Ti} Y_{Li}, [L = 1, 2, 3, \dots, N] \quad (3.9)$$

C_L are the measured total beta counts, and Y_{Li} are the values of Y_i for different time intervals, respectively.

- ◆ N_{Ti} could be estimated from equation (3.9) using the least square fitting method.
- ◆ n_i could be computed using equations from (3.4a) to (3.4g).

To make full use of the statistical nature of the decay process in the determination of concentration of daughters products, counting has to be made for a large number of time intervals [42].

3.5 Determination Equilibrium Equivalent Concentrations (EECs)

From these concentrations the Equivalent Equilibrium Concentration (EECs) of both ^{222}Rn and ^{220}Rn can be obtained. EEC can be given in terms of the individual decay product concentrations of ^{222}Rn and ^{220}Rn according to the formula [42, 43].

$$EEC_{Rn} = 0.105C_1 + 0.515C_2 + 0.380C_3$$

where, C_1 , C_2 , and C_3 represent concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi respectively and also

$$EEC_{Rn} = 0.913C_1 + 0.087C_2$$

where, C_1 and C_2 represents ^{212}Pb and ^{212}Bi respectively.

Chapter 4

Discussion

Measurements of radon daughter products taken place based on garb sampling of known volume of air using Section Pump [4, 44, 45], were the main concern of this work. The gross count obtained from the measurement and the duration of the measuring time intervals are put in a computer programme, that computes the concentration of radon progeny and the equilibrium equivalent concentration for the two radon isotopes [4, 44, 45].

The computer program shown a decline decay curve in the observed count rates, based on the mathematical model developed in equations (3.3a - 3.3b). The best fit curve specifies the concentration of radon daughter products as shown in the fig (4.1), below.

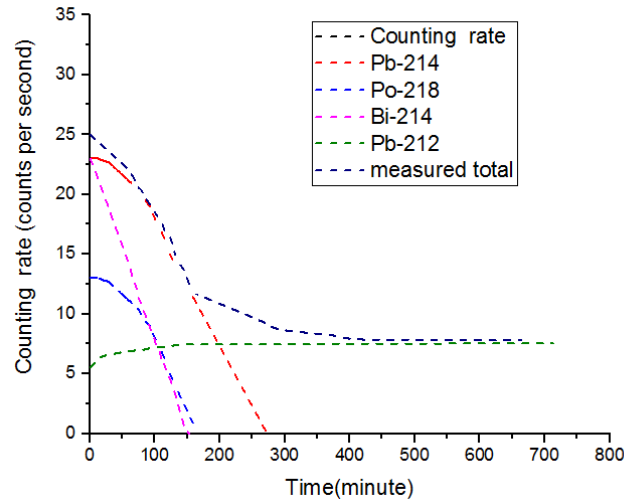


Figure 4.1: Concentration of radon daughter products determined from gross β counts

4.1 The decay graph

From the decay graph shown above, we get the following results.

- ◇ The beta count rate measured over the time interval as a function of time.
- ◇ Contribution of the individual decay products for all the counting intervals, as a function of time.
- ◇ Activities of isotopes in a unit volume of sampled air.
- ◇ Sum of the individual contributions for some counting intervals.
- ◇ The Equilibrium Equivalent Concentration (EEC) of ^{222}Rn and ^{220}Rn .

4.2 Contributors to the total beta count rates

The progenies ^{218}Po , ^{214}Pb , ^{214}Bi which are daughter products of radon (^{222}Rn) and ^{212}Pb , ^{212}Bi , ^{208}Tl , which are daughter products of thoron (^{220}Rn), are expected to be significant contributors to the total beta count rates. Polonium (^{218}Po), is the only radon progeny which indirectly causes contributions in the activities of ^{214}Pb and ^{214}Bi . Based on their half lives, the computation split out the total (measured) count rate between the radionuclides on the basis of the different shapes of their theoretical decay curves [4, 44, 45].

4.3 Artificial and natural long lived beta emitters

The assumption of the existence of some artificial short lived nuclide, like the bi - products of PET laboratories in a special environment such as ^{18}F , fission products from a nuclear accident and naturally occurring long lived nuclide that are present in the sampled air in low concentrations have mathematically correct but physically meaningless concentration values [4, 44, 45]. Thus, the contributions of such β emitters have negligible effect in the graph.

4.4 Accuracy and random errors in the graph

The duration of sampling, the delay between sampling and beta counting, the activity concentration levels and the duration of beta counting are the basic factors to determine the

accuracy (random errors) of the activity concentrations of the radionuclides. Their values are different for different radionuclides. From the graph we see that, for example, lead, ^{212}Pb , half life of 10.6 hr, can be determined more accurately if long duration of sampling and long duration of beta counting is taken [4, 44, 45]. On the other hand, a short duration of sampling and a short delay between sampling and beta counting results better detection for ^{218}Po . The decay curves of nuclide ^{218}Po and ^{214}Pb have similar shapes. The collective contribution of ^{212}Bi and ^{208}Tl to the total counting rate is relatively small throughout in the decay curve [46, 47, 48]. It implies, the AC ^{21}Bi and AC ^{208}Tl are determined only with high relative random errors. From the count rates, the three radionuclides concentrations (^{218}Po , ^{212}Bi and ^{208}Tl) results uncertainties in the measurements and accurate results are obtained for ^{214}Pb , ^{214}Bi , ^{212}Pb , EEDC Rn $_{222}$ and EEDC Rn $_{220}$. The EEDC Rn $_{222}$ and EEDC Rn $_{220}$ almost depends on the concentration of ^{214}Pb , ^{214}Bi , ^{212}Pb .

The concentrations of radon daughters can be obtained by taking their successive precursor ratios, according to steady - state Jacobi's model, (SSJM) [47, 48]. These ratios are modeled using the system of balance equations. In most cases, fore example, values of the ratios of AC ^{214}Pb / AC ^{218}Po and AC ^{212}Bi / AC ^{212}Pb are typically within the range of 0.2 - 0.8 [46, 47, 48, 49]. The model results to obtain better detections of concentrations of radon daughter and minimize random errors when the measurement is taken in the base line of air ventilation rate (0.55 h^{-1}), [47, 48, 49], as shown in fig (4.2) below.

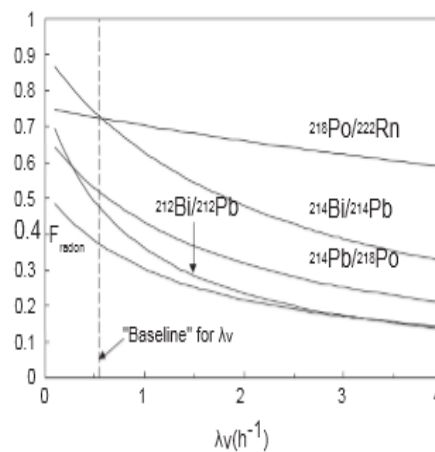


Figure 4.2: The dependence of some ratios of ACs of radon - thoron progeny on air ventilation rate constant (λv) according to the (SSJM), adapted from Z. Papp, etal. paper.

From the above graph we see that the air ventilation rate constant (λ_v) is a significant factor to determine the ratios of the activity concentrations of radon daughters, [46, 47, 48, 49]. Hence, the activity concentrations of the daughter products are outlined as follow.

◆ Let the following letters (X, Y, Z and W), are constants and their values are between 0 and 1 for constant air ventilation rate at its base line.

$$AC^{218}Po / AC^{222}Rn = X \implies AC^{218}Po = X [AC^{222}Rn]$$

$$AC^{214}Bi / AC^{214}Pb = Y \implies AC^{214}Bi = Y [AC^{214}Pb]$$

$$AC^{214}Pb / AC^{218}Po = Z \implies AC^{214}Pb = Z [AC^{218}Po]$$

$$AC^{212}Bi / AC^{212}Pb = W \implies AC^{212}Bi = W [AC^{212}Pb]$$

From the above relations we get three conditions.

1. When the air ventilation rate is below the base line, the coefficients are related as

$$Y > X > W > Z$$

2. When the air ventilation rate is equals to the base line, the coefficients are related as

$$Y = X \text{ and } Z > W$$

3. When the air ventilation rate is above the base line, the coefficients are related as

$$X > Y > Z > W$$

. By the same argument, i obtained computed conditional concentrations of radon daughter products by fixing air ventilation rate at its baseline 0.16 hr^{-1} , shown in fig (4.3), below.

The air ventilation rate is obtained by applying the following relations.

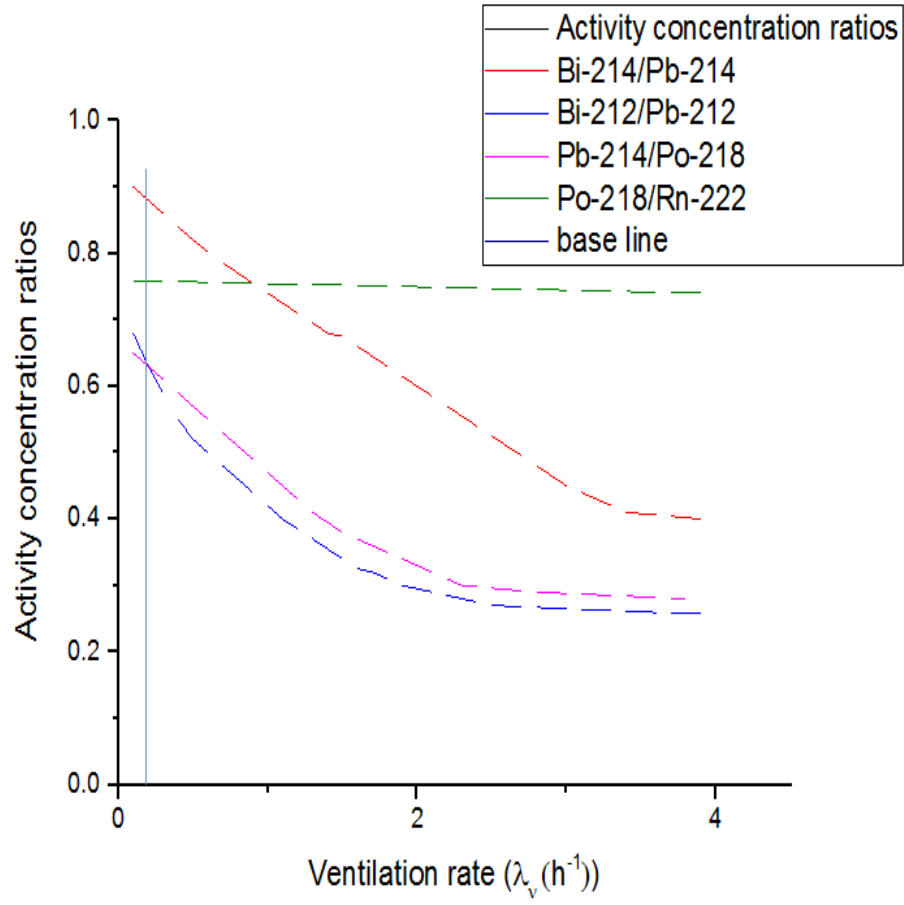


Figure 4.3: Computed concentration ratios of radon daughter products determined from gross β counts, based on Jacobi's model

Air exchange rate is the rate at which stale indoor air with fresh out door air in a room. The amount of fresh air (make up air) flow through the room (m^3/hr) can be obtained using a formula of:

$$n = \frac{Q}{V} \quad (4.1)$$

where

n = air changes per hour or air change rate

Q = fresh air (make up air) flow through the room (m^3/hr)

V = volume of the room (m^3)

The computed graph is similar to that of the graph obtained from Z. Papp.

Chapter 5

Conclusion and recommendations

5.1 Conclusion

Depending on the setted objectives, the main concern of this work is to compare concentrations of radon daughter products from filtered aerosols in terms of the Weighted Least Square Method (WLSM) and the Steady State Jaccobi's Model (SSJM). Measurements of radon and their daughters are important to determine whether or not a room needs to have radon reduction remedial for the well being of the occupants from the health point of view. During measurements of radon daughter products a room must to be well closed so as to obtain low air ventilation rate and adequate radon accumulation on the filter paper.

Measurements of radon based on garb sampling of known volume of air on a glass fiber filter with high retention power followed by gross beta counting of the filtered air sample count over large number of successive time intervals using an end window Geiger - Müller (GM) was reviewed. The measuring method lets us to determine the decay products of radon and thoron separately and simultaneously. The total or gross beta count is determined using the settled differential equations. Defined Solid Angle Absolute Beta Counting (DSAABC) is applied to evaluate the counting efficiencies for the decay products one by one [50]. Air flow rate, duration of sampling, delay between sampling and counting, background and dead time of counter, counting geometry, and filter parameters determine the accuracy (random error) of the activity concentration.

The analysis of the concentrations of each radon daughter products was conducted based on two different methods, the weighted Least Square Method (WLSM) and the Steady State Jaccobi's Model (SSJM). The two methods have no any correlations. In case of the Weighted Least Square Method (WLSM), the experimental count rate is fitted to the mathematical method to determine Equilibrium Equivalent Concentrations of Radon isotopes (EEC Rn). From figure 4.1,above although the radionuclides ^{218}Po , ^{214}Pb , ^{214}Bi , which are progenies of radon (^{222}Rn) and ^{212}Pb , ^{212}Bi , ^{208}Tl , which are progenies of thoron (^{220}Rn) was assumed to be significant contributions to the total beta count rate, only the three progenies, ^{214}Pb , ^{214}Bi , ^{212}Pb resulted accurate values. The EEDC of Rn $_{222}$ and EEDC of Rn $_{220}$ are almost dependant on the concentration of ^{214}Pb , ^{214}Bi , ^{212}Pb . However, the results for ^{218}Po and ^{212}Bi are much more inaccurate because of their short half lives. They need very low sampling time and very low time delay between sampling and beta counting. Unlike to this, the daughter product lead, (^{212}Pb), can be determined more accurately if long duration of sampling and long duration of beta counting is applied. The concentration of radon based on the weighted least square method was found to be 21.6 Bq/m³.

The other way mechanism which used to obtain the concentrations of radon daughter products was the Steady - State Jacobi's Model (SSJM), and was applied in this work for the same aerosols sample used in the Weighted Least Square Method above. This method is dependant on air exchange rate or air ventilation rate of a room to determine concentrations of radon daughter products. It is simply taking the ratios of radon daughter products of succussive precursor. These ratios are modeled using the system of balance equations. This results the values of the ratios of AC ^{214}Pb / AC ^{218}Po and AC ^{212}Bi / AC ^{212}Pb are typically within the range 0.2 - 0.8. This ratio value is in the range of 0 and 1 [47, 48, 49]. The most typical values of the ratios of radon daughter products at the base line of air ventilation rate constant are 0.52 for AC ^{214}Pb / AC ^{218}Po and 0.48 for AC ^{212}Bi / AC ^{212}Pb [47, 48, 49]. Using the Steady State Jaccobi's Model for the same sample of aerosols, the Equilibrium Equivalent Concentration (EEC) of radon was obtained to be 24.6 Bq/m³.

When we compare the two methods, the Steady State Jaccobi's Model decreases the uncertainty in the evaluation of the activity concentrations of radon daughter products [49].

In this cases better detection of radon is obtained using the Steady State Jaccobi's Model. The weighted least square method has some what magnified random errors and results low accuracy than the Steady State Jaccobi's Model.

Using both methods, for the two radionuclides ^{208}Tl (half life of 3.053 min) and that of ^{218}Po (half life of 3.10 min), due to their comparable half lives, it is impossible to get their concentrations at the same time and totaly impossible to get directly the specific concentration of ^{208}Tl from the measurement. We assumed the concentration of ^{208}Tl being 0.3 times that of its precursor because the decay branching ratio is 0.36. Its concentration is obtained being very low because of high random error encountered during the process of measurement. Although we have assumed the seventh naturally existing long lived and the very short lived artificial beta emitter radionuclide to be determined mathematically under chapter 3, the actual result doesn't shown their concentrations graphically. Thus the concentration of long lived and very short lived β emitters are seen being negligible physically in the graph due to their uncontrollable half lives.

5.2 Recommendations

Here after reviewing analysis of radon product activities from filtered air samples using garb radon sampling measurement, using an instrument the Geiger - Müller (GM) counter, the list square weighted method is sophisticated and cumbersome to determine radon daughter concentrations. The other reviewed radon daughter concentrations determination techniques is the Steady States Jaccobi's Model (SSJM). This method maximizes the detection efficiency of radon progenies as the air flow rate is lowered because it takes in to consideration the air flow rate as a base line.

Chapter 6

Appendix

Fitted graphs of radon daughter contributions using the Weighted Least Square Method (WLSM) and graphs of radon prosector daughter concentrations ratios determined by the Steady State Jaccobi's (SSJM) Model (SSJM) and the analyzed their random error values.

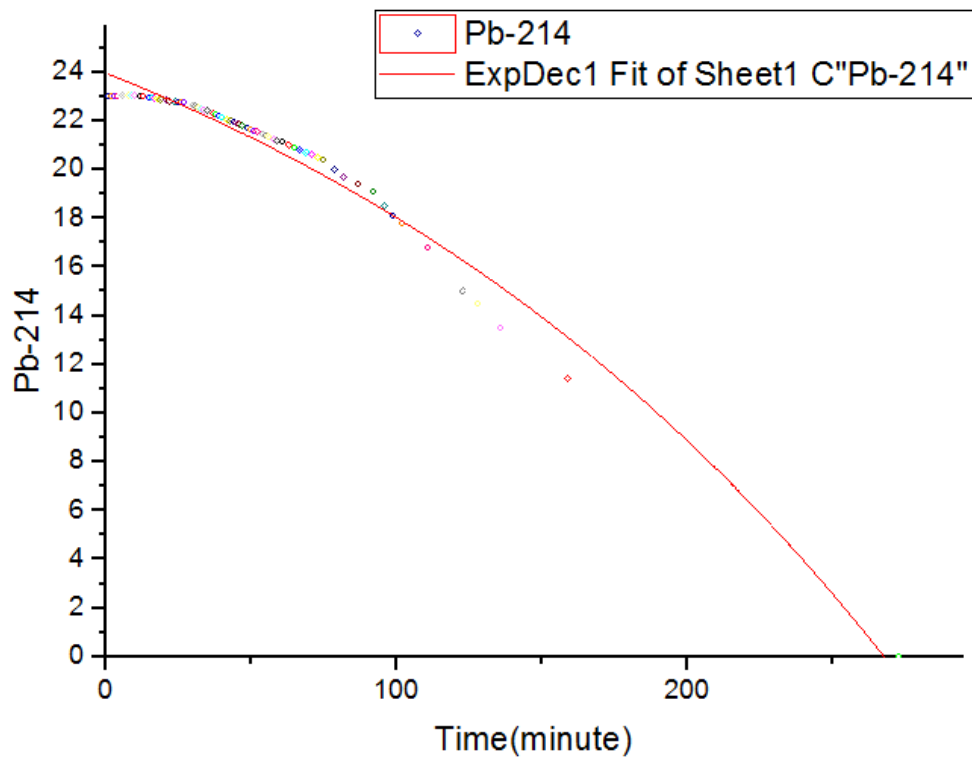


Figure 6.1: fitted graph of Pb - 214 with the Least Square Method

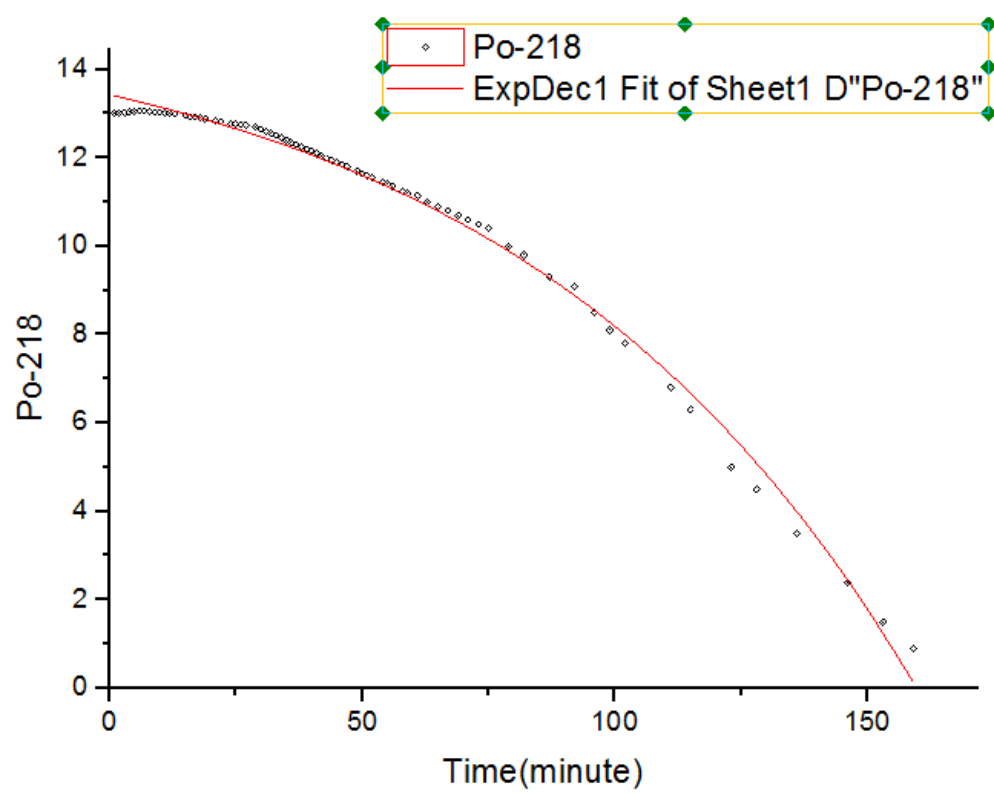


Figure 6.2: fitted graph of Po - 218 with the Least Square Method

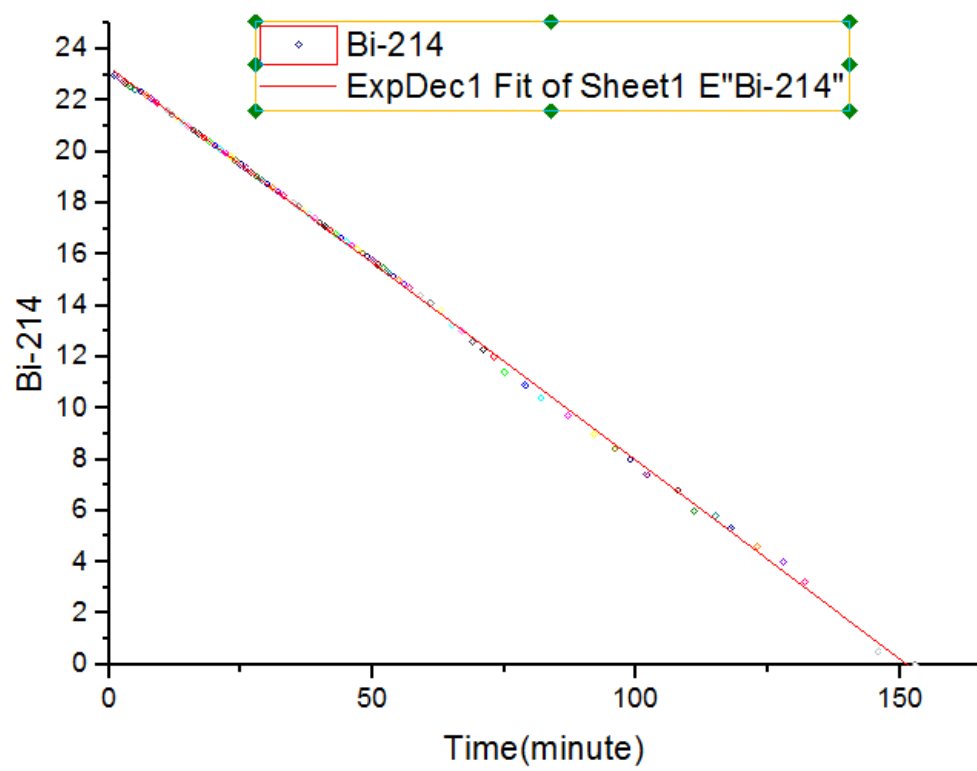


Figure 6.3: fitted graph of Bi - 214 with the Least Square Method

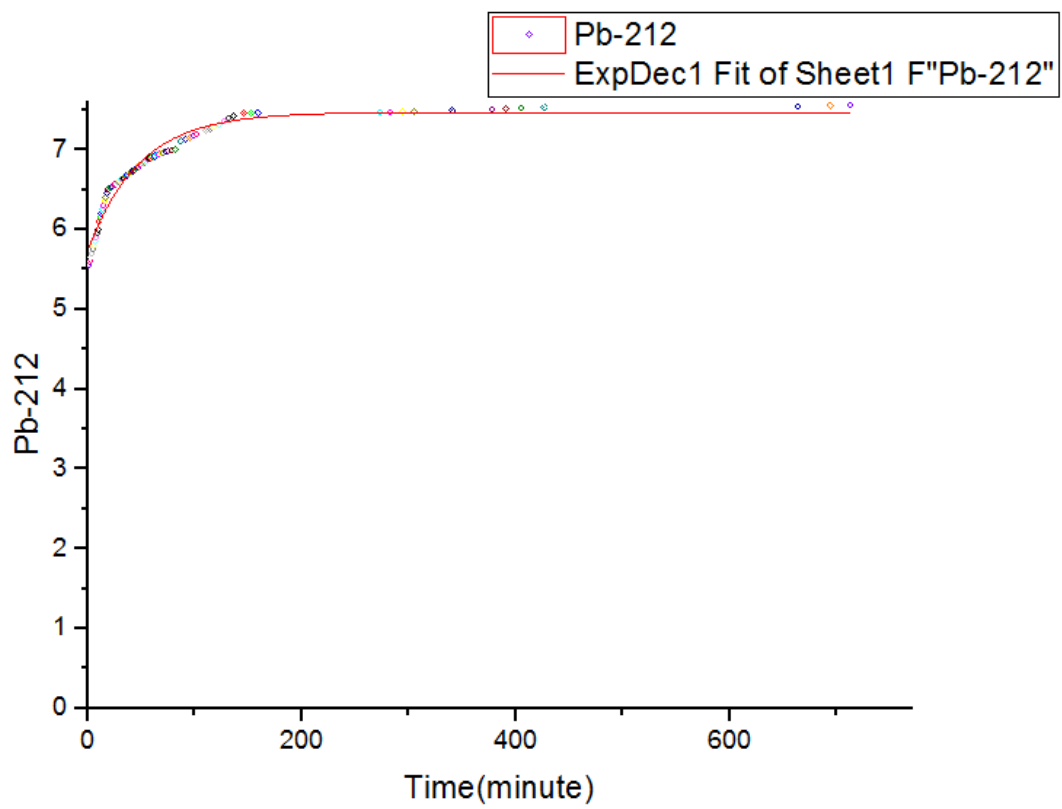


Figure 6.4: fitted graph of Pb - 212 with the Least Square Method

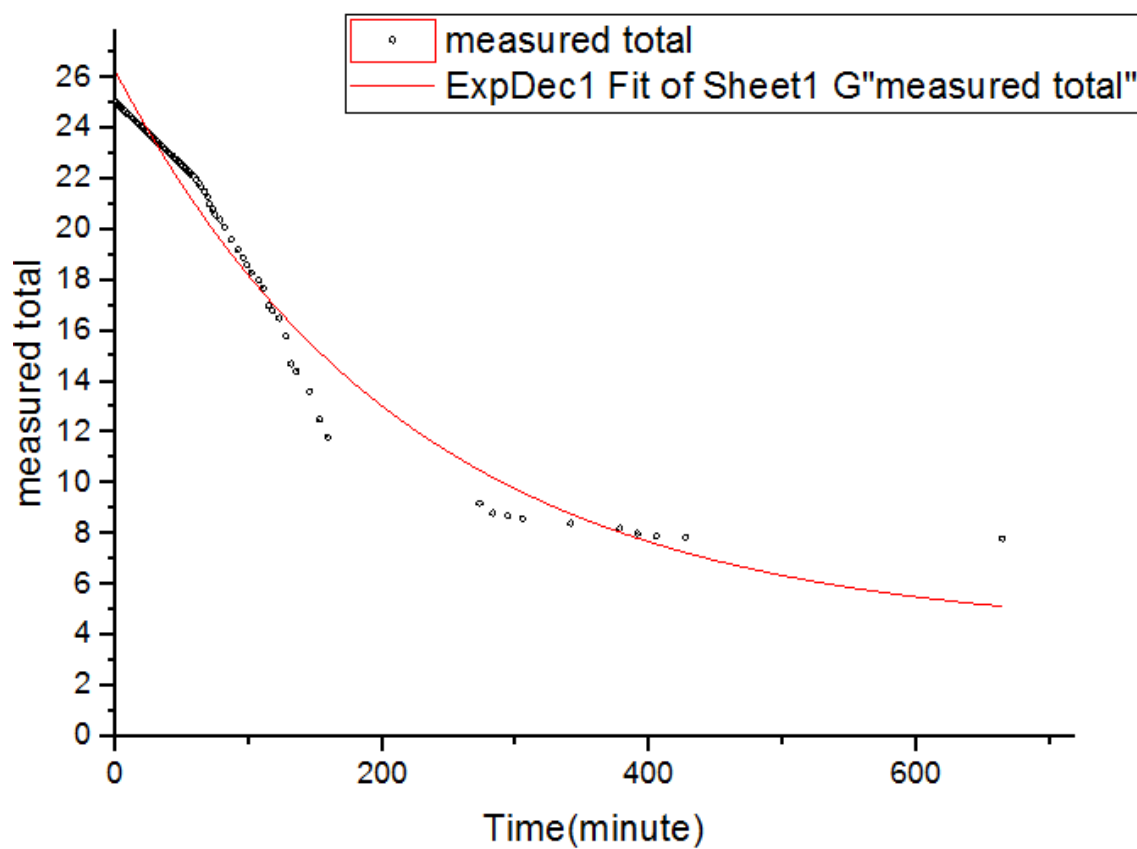


Figure 6.5: fitted graph of measured total concentration of radon with the Least Square Method

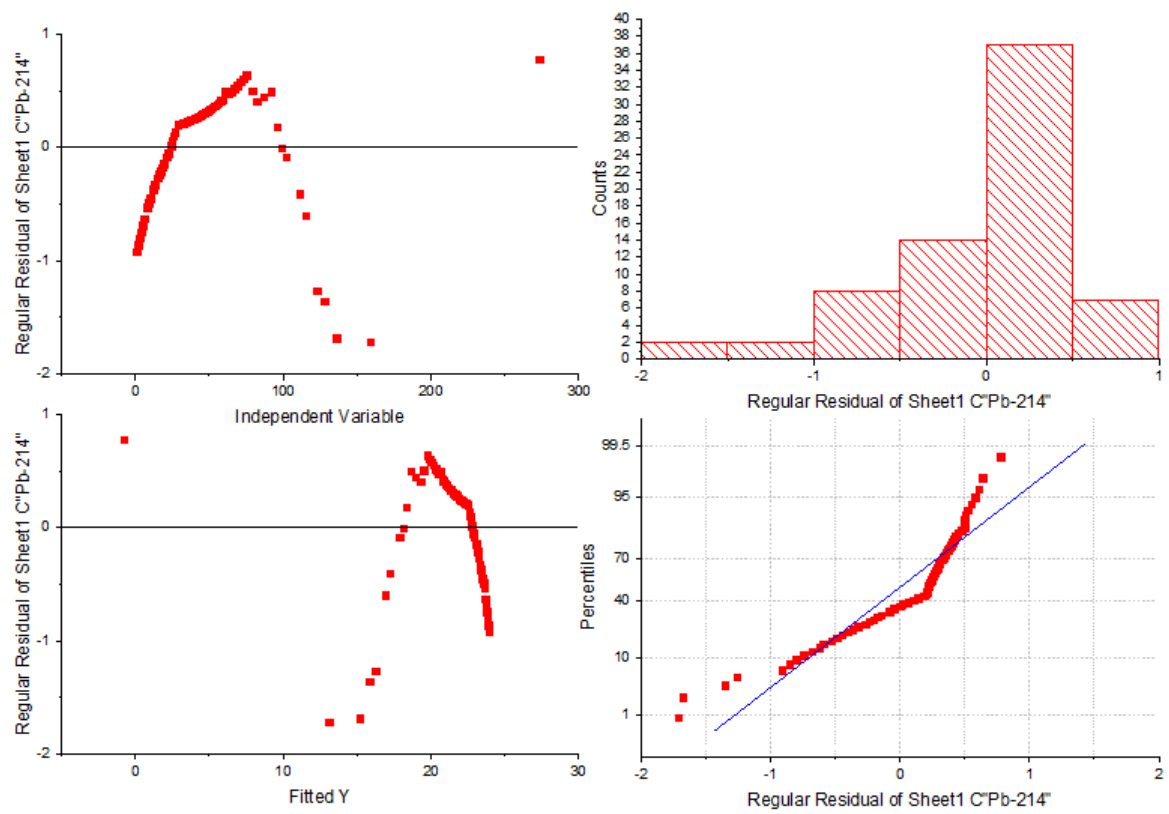


Figure 6.6: fitted graph of Pb - 214 with the Least Square Method and its error

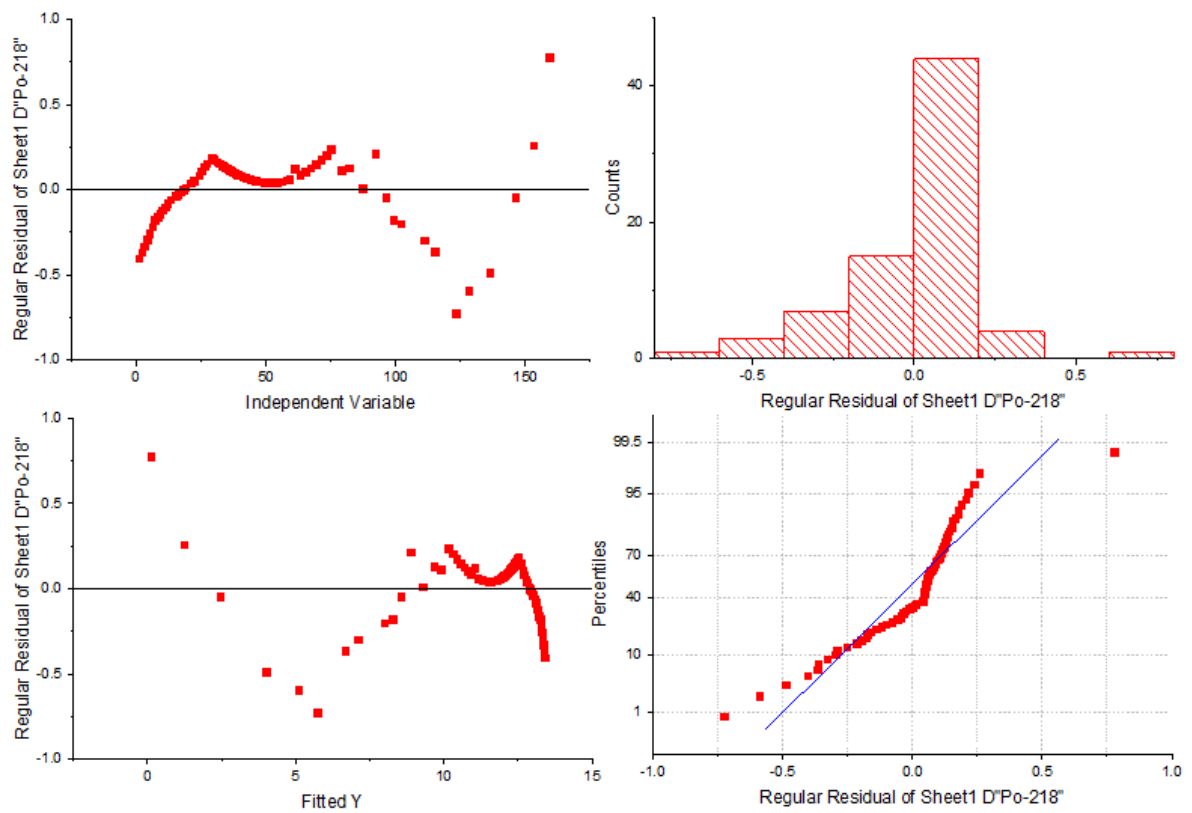


Figure 6.7: fitted graph of Po - 218 with the Least Square Method and its error

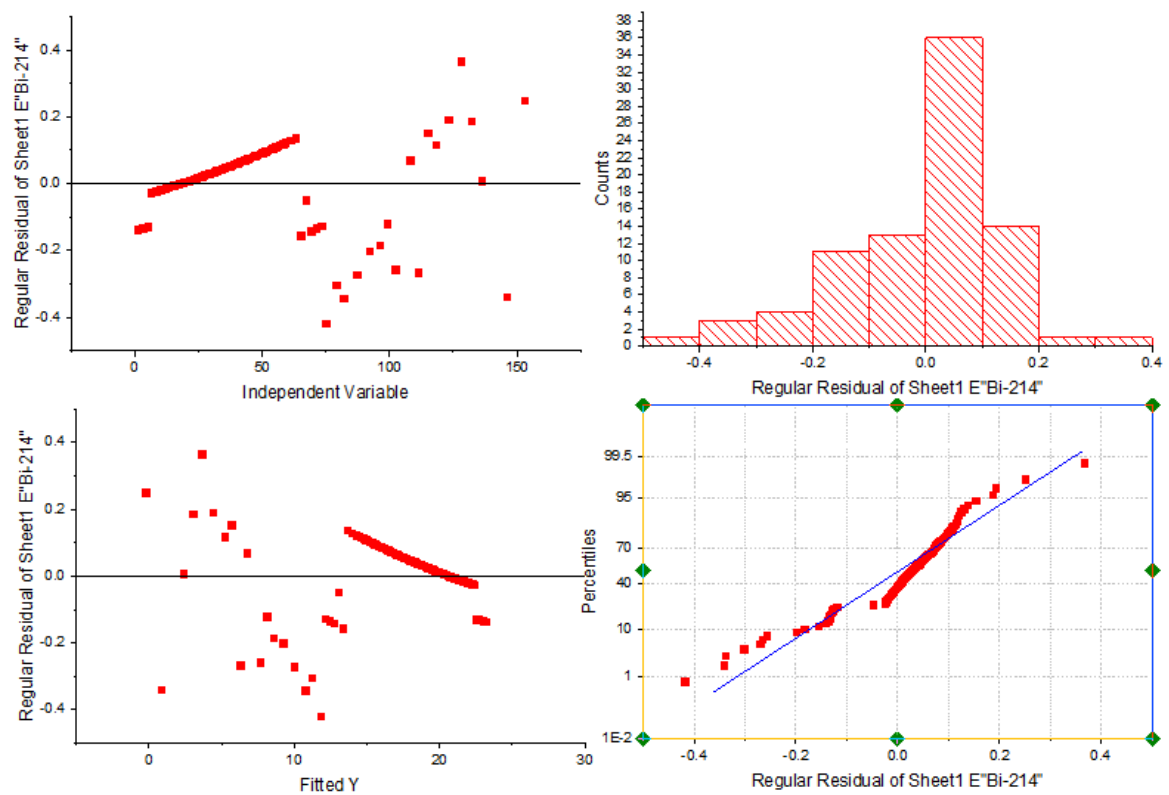


Figure 6.8: fitted graph of Bi - 214 with the Least Square Method and its error

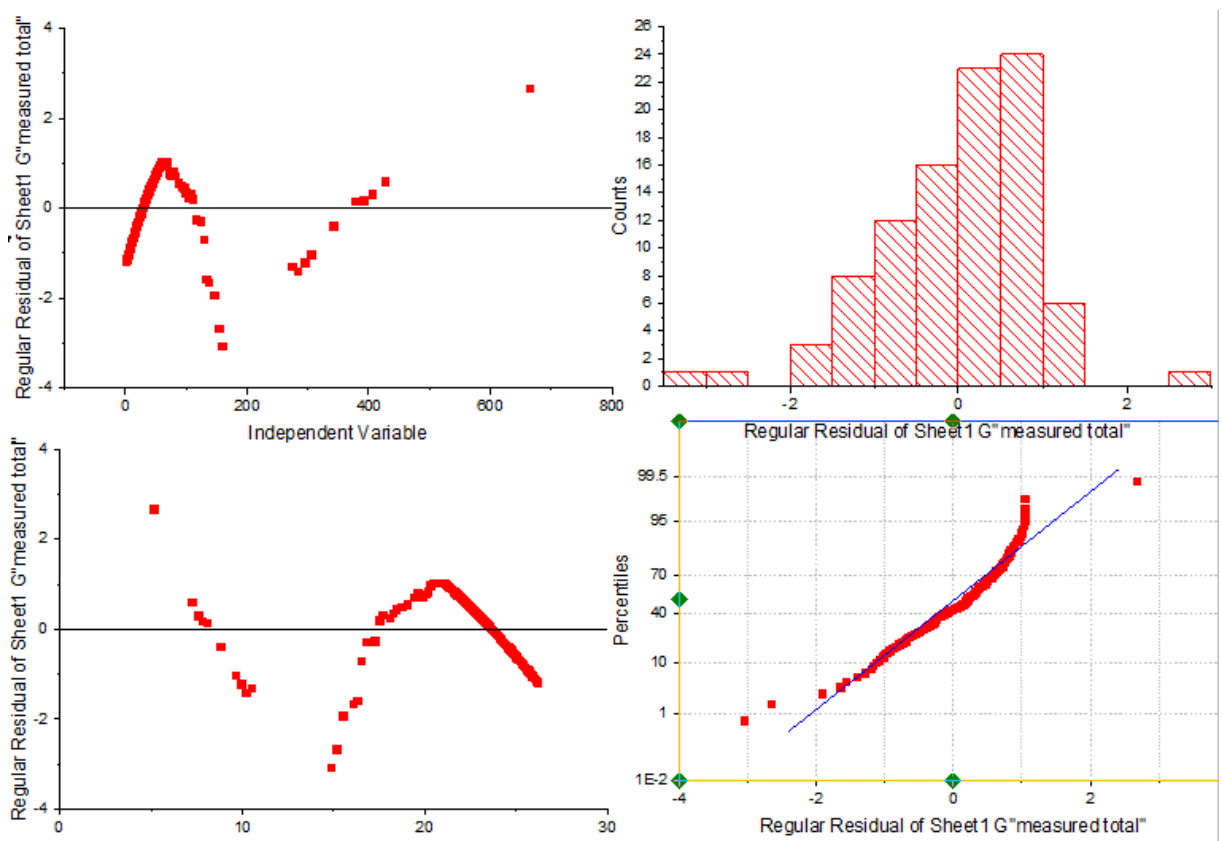


Figure 6.9: fitted graph of total concentration of radon with the Least Square Method and its error

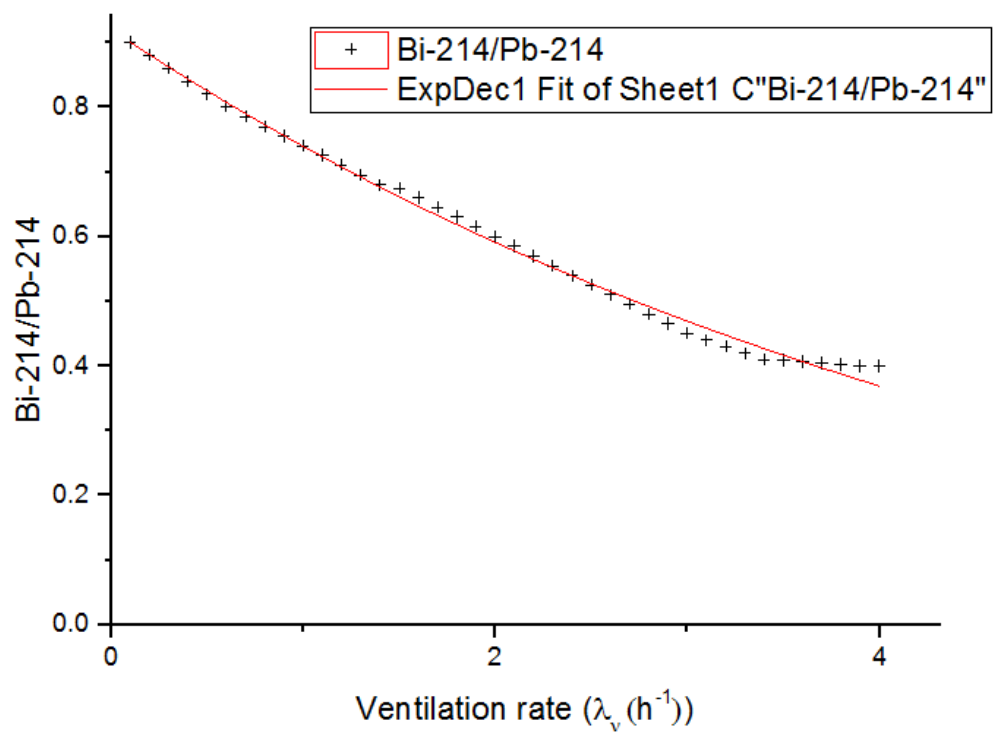


Figure 6.10: fitted graph of ratio concentrations of Bi - 214 / Pb - 214

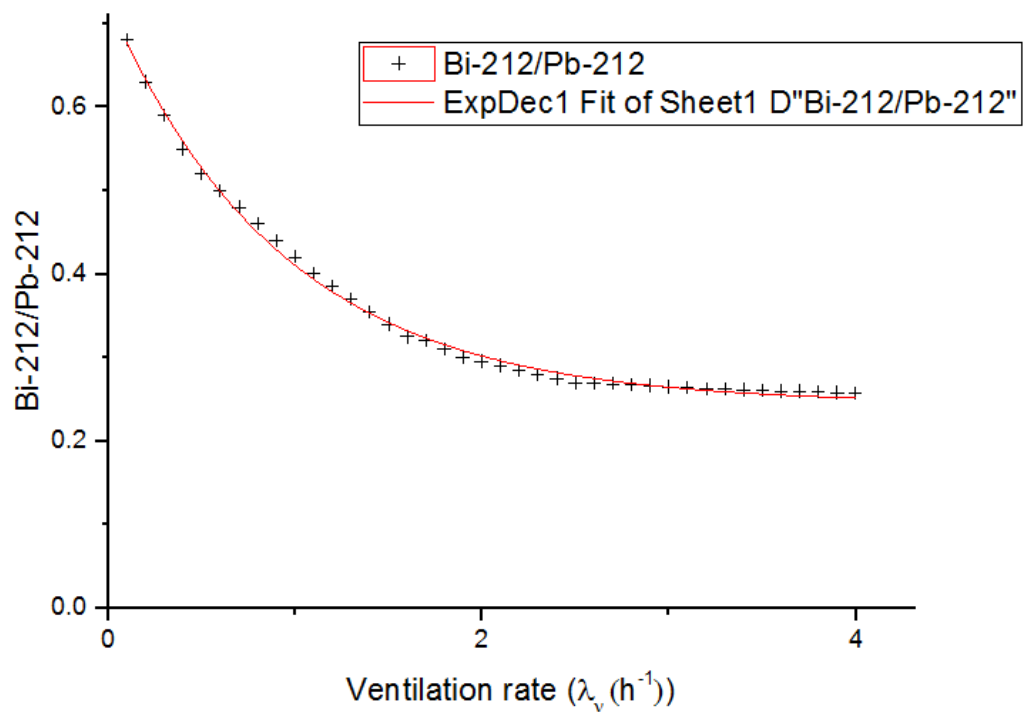


Figure 6.11: fitted graph of ratio concentrations of Bi - 212 / Pb - 212

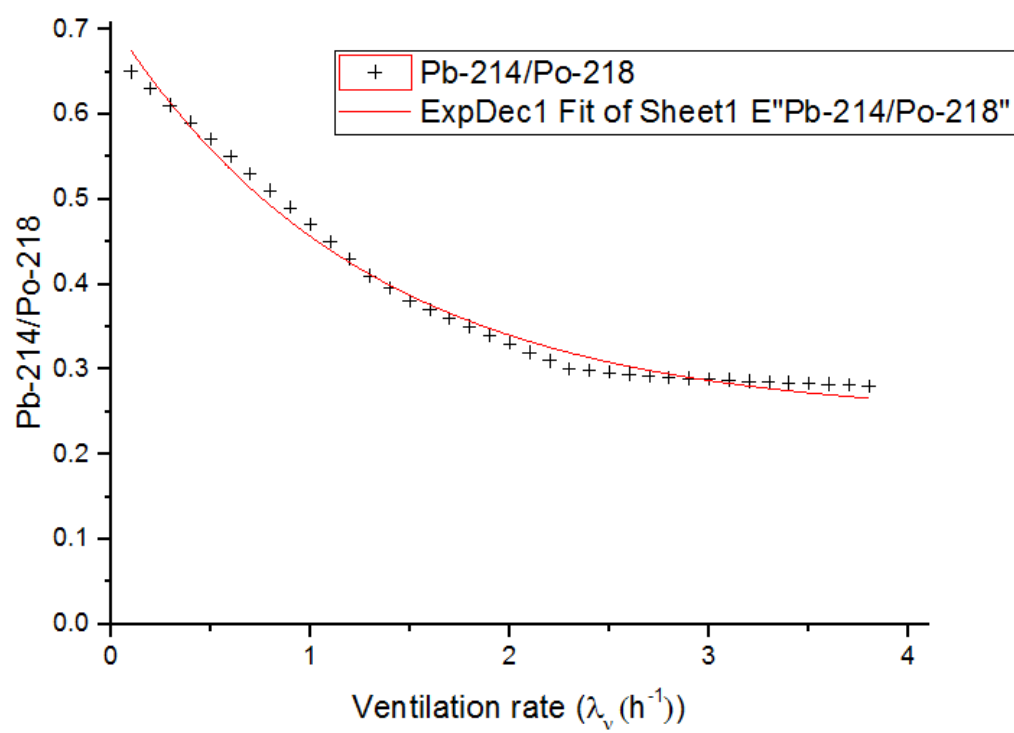


Figure 6.12: fitted graph of ratio concentrations of Pb - 214 / Po - 218

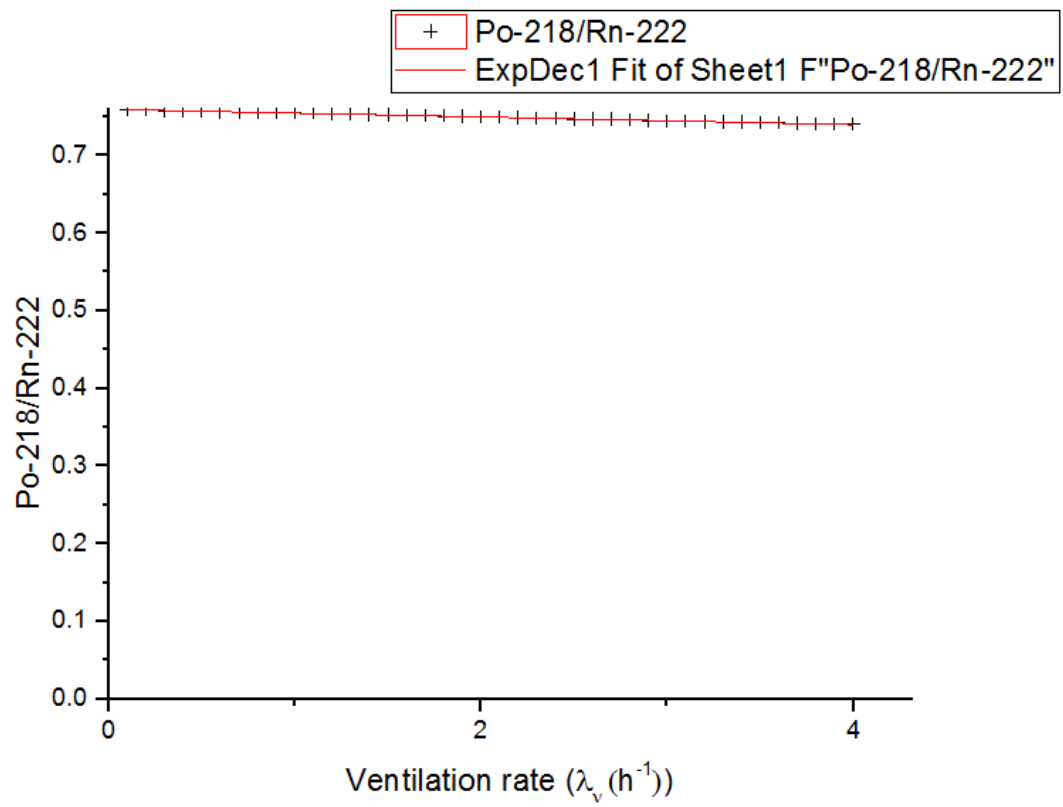


Figure 6.13: fitted graph of ratio concentrations of Po - 218 / Rn-212

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DECLARATION

I, the under signed declare that the thesis is my original work, has not been presented for a degree in any other university and that all sources of materials have been duly acknowledged.

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