

# **THORON COMPONENT OF ATMOSPHERIC RADON**



**By**

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Dated: August 2007

**This Work is Dedicated to  
My Parents**

# Table of Contents

<b>Table of Contents</b>	<b>v</b>
<b>List of Tables</b>	<b>vii</b>
<b>List of Figures</b>	<b>viii</b>
<b>Acknowledgements</b>	<b>ix</b>
<b>Abstract</b>	<b>x</b>
<b>Introduction</b>	<b>1</b>
<b>1 Review of Literature</b>	<b>2</b>
1.1 Physical properties of radon . . . . .	2
1.2 Source of radon . . . . .	3
1.2.1 Radon entry into the atmosphere . . . . .	4
1.2.2 Radon in mines . . . . .	6
1.2.3 Radon in outdoor air . . . . .	6
1.2.4 Radon in indoor air . . . . .	7
1.2.5 Radon in water . . . . .	11
1.3 Health hazards due to radon . . . . .	12
1.3.1 Discovery . . . . .	12
1.3.2 Effects of radon on the body . . . . .	14
1.4 Main objective of the study . . . . .	15
<b>2 Radon Measurement Methods</b>	<b>16</b>
2.1 Passive Measuring Methods . . . . .	16
2.1.1 Solid State Nuclear Track Detector . . . . .	17
2.1.2 Activated charcoal (AC) . . . . .	18
2.1.3 Charcoal liquid scintillation (LS) . . . . .	19
2.1.4 Electret ion chamber (EC) . . . . .	20
2.2 Active Measuring Methods . . . . .	20
2.2.1 Pump / collapsible bag (PB) . . . . .	20
2.2.2 Three-day integrating evacuated scintillation cells (SC) . . . . .	21
2.2.3 Grab radon sampling . . . . .	21

<b>3</b>	<b>Materials and Methods</b>	<b>23</b>
3.1	Materials . . . . .	23
3.1.1	The sampling head . . . . .	24
3.1.2	Filtering air . . . . .	25
3.2	Methods . . . . .	25
3.2.1	The counting process . . . . .	25
3.2.2	Mathematical model . . . . .	26
3.2.3	Defined Solid Angle Absolute Beta Counting Method . . . . .	31
<b>4</b>	<b>Data Analysis and Discussion</b>	<b>36</b>
4.1	Results . . . . .	36
4.2	Analysis . . . . .	39
4.3	Comparison With Other Areas of the World . . . . .	40
4.4	Diurnal variation of EEC of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ . . . . .	41
<b>5</b>	<b>Conclusion</b>	<b>46</b>
	<b>Bibliography</b>	<b>50</b>

# List of Tables

1.1	Physical properties of Radon . . . . .	3
1.2	Radon concentrations in dwellings (UNSCEAR, 2000) . . . . .	10
1.3	Thoron concentrations in outdoor and indoor air (UNSCEAR, 2000)	11
1.4	Average worldwide exposure to natural radiations (UNSCEAR, 2000)	13
4.1	Measurement Procedure in DSAABC Method . . . . .	37
4.1	Continued . . . . .	38
4.2	Computed activity concentration of Radon Progeny and Equilibrium Activity Concentrations . . . . .	43
4.3	Computed activity concentration of Thoron Progeny and Equilibrium Activity Concentrations . . . . .	44
4.4	Comparison of Indoor Radon Concentrations . . . . .	45

# List of Figures

1.1	Decay series of Radon isotopes . . . . .	4
1.2	Radon entry into the house (EPA, 2005) . . . . .	8
3.1	The experimental setup . . . . .	23
3.2	Air Sampling Head Blueprint . . . . .	24
3.3	Parts of the Sampling Head . . . . .	24
3.4	The Air Sampling Head assembled . . . . .	25
4.1	Concentration of daughters determined from gross $\beta$ count . . . . .	40
4.2	Comparison of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ Equilibrium Equivalent Concentrations: Indoor . . . . .	41
4.3	Comparison of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ Equilibrium Equivalent Concentrations: Outdoor . . . . .	42

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# Abstract

Air was sampled by the use of suction pump through a glass filter (> 90% filtration efficiency) and aerosols were deposited on the glass-fiber filter. The aerosol deposited on the filter paper was placed below a GM tube (with GM nuclear counter) within a time interval of less than one minute after the end of sampling. The gross  $\beta$  counts were registered in successive time intervals for more than six to ten hours (i.e. in intervals of 1 min for the first one hour; 2 min for the second hour; 4 min for the third hour etc. )

The activity concentration of progenies of  $^{222}\text{Rn}$  and EEC (equilibrium equivalent concentration) of  $^{222}\text{Rn}$  was done by fitting the observed gross  $\beta$  count to a mathematical model derived.

The indoor and outdoor variation of  $^{222}\text{Rn}$  was observed inside and outside of the nuclear physics laboratory (around four meters above the ground). The average EEC of  $^{222}\text{Rn}$  was found to be  $23.9 \text{ Bq/m}^3$  and  $3.4 \text{ Bq/m}^3$  for indoor and outdoor respectively.

Indoor radon concentration is 4 to 6 times higher than the outdoor concentration. Our result agrees with the results available in other studies.

# Introduction

Inhalation of the short-lived decay products of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  daughter products contribute the largest fraction of radiation exposure from natural sources. Studies of  $^{222}\text{Rn}$  and its progeny in various types of atmosphere are reported since the early fifties (Tzivoglou et al., 1953; Harley, 1953). In the decades that follow alpha and gamma detection techniques have been developed resulting in a steadily growing international database (Budnitz, 1974).

However, information on  $^{220}\text{Rn}$  and its daughters are scarce and or partly unavailable in literature (Steinhäusler, 1996). The activity concentrations of  $^{212}\text{Pb}$ , were measured at the nuclear science laboratory of the Physics Department (AAU) and the outdoor atmosphere behind the lab. The locale is more than 300 m far from the nearby street and there is no known mining site in the region and no known deposition of Uranium. There is no coal fired thermal power plant in the country.

Information on  $^{220}\text{Rn}$  and its daughters have their application in the description of air flow (Bigu, 1992); as tracer gases (Willet, 1985) and in the studies of atmospheric electrical phenomena (Hoppel et al., 1986). In recent years it has become clear that more knowledge about  $^{220}\text{Rn}$  and its daughters is required for reliable assessment of dose from radon progeny in air. It has been reported that the concentration of  $^{220}\text{Rn}$  can be as high as  $200 \text{ Bq m}^{-3}$  under certain circumstances and the outdoor air can also be a source of high  $^{220}\text{Rn}$  in indoor air (Porstendörfer, 1994).

In this work the atmospheric concentration of  $^{220}\text{Rn}$  was determined in the indoor and outdoor environment of air using Defined Solid Angle Absolute Beta Counting (DSAABC) method. The findings are compared with literature values.

# Chapter 1

## Review of Literature

Rutherford and R.B.Owens in 1899 noticed that some of the radioactivity of thorium compounds could be blown away. They were studying the emanations from thorium and wrote that the radiation from thorium varied in a sudden unexpected changes. 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. Rutherford and Owens had discovered thoron( $^{220}\text{Rn}$ ), half life 51 sec.

In 1899 Pierre and Marie curie, who were studying about emanations from radium, concluded that it stayed radioactive for several days. They detected  $^{222}\text{Rn}$ , half life 3.82 days, though they had doubts as to whether the activity was really in the form of a gas. In 1901 Rutherford and Ernst Dorn (in 1900) confirmed that the radiations emitted by radium were radioactive. That emitted gas, named as radon since 1923, identified as a radioactive noble gas[Nagaratnam].

The other isotope of radon, called actinon, was discovered in 1904 by Friedrich Giessel and Andre-Lousi Debierne. Besides the above three naturally existing isotopes, around 17 other isotopes of radon are known.

### 1.1 Physical properties of radon

Radon is a noble gas. It is mono-atomic, radioactive, soluble in water and seven times denser than air. Its most abundant isotope, and also the most dangerous, is  $^{222}\text{Rn}$ , generated by the decay of  $^{238}\text{U}$  contained in geological material.

Radon is the only radioactive noble gas which exists every where in different proportions. This chemically inert gas is one of the heaviest gases at room temperature. Radon<sup>222</sup>Rn and thoron<sup>220</sup>Th are belonging to the Uranic and Thoric groups respectively.

At standard temperature and pressure radon is colorless gas. But, when it is cooled below its freezing point, it will have a brilliant phosphorescence which turns to yellowish as the temperature is lowered and orange-red at the temperature air liquifies.

Physical properties of radon	
Name , Symbol , Number	radon , Rn , 86
Chemical series	noble gases
Group , Period , Block	18 , 6 , p
Appearance	colorless
Atomic mass	( 222 ) g / mol
Electron configuration	[Xe] 4f <sup>14</sup> 5d <sup>10</sup> 6s <sup>2</sup> 6p <sup>6</sup>
Electrons per shell	2,8,18,32,18,8
Phase	gas
Melting point	202 °K ( -71 °C )
Boiling point	211.3 °K ( -61.7 °C )
Heat of fusion	3.247 KJ / mol
Heat of vaporization	18.10 KJ / mol
Heat capacity	( 25 °C ) 20.786 J / ( mol . °k)

Table 1.1: Physical properties of Radon

## 1.2 Source of radon

Radon is an ubiquitous element found worldwide. It is produced by the radioactive decay of radium which is a member of the naturally occurring Uranium decay series. The most abundant isotope <sup>222</sup>Rn is a descendant of <sup>238</sup>U while <sup>220</sup>Rn (thoron) and <sup>219</sup>Rn (actinon) are the descendants of <sup>232</sup>U and <sup>235</sup>U respectively.

Uranium and thorium are primordial (meaning they were present when the earth was formed) radioactive elements. They are present throughout the earth's crust. Some types of rock are rich in these elements. Uranium (<sup>238</sup>U) eventually decays to radium (<sup>226</sup>Ra, half-life 1600 years), and subsequently to radon (<sup>222</sup>Rn, half-life 3.82 days). As an inert gas, radon diffuses through the soil to the atmosphere,

where it can accumulate in buildings, particularly in the lower levels. Radon is relatively innocuous; however, it decays to lead ( $^{210}\text{Pb}$ ) in a matter of minutes by emitting several alpha particles from elements in the decay sequence. The elements of this decay sequence are called radon daughters (or progeny).

There are at least seventeen known isotopes of radon. The decay series of the three abundant isotopes is shown below.

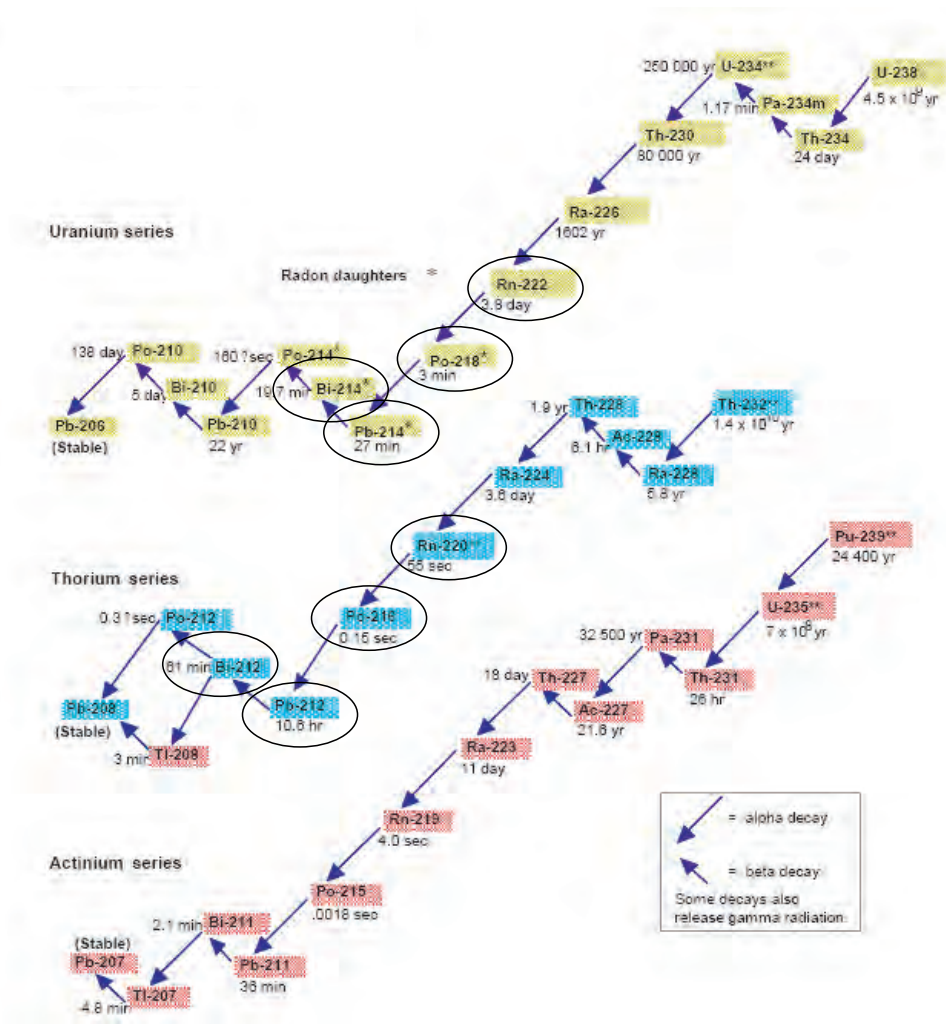


Figure 1.1: Decay series of Radon isotopes

**1.2.1 Radon entry into the atmosphere**

$^{222}\text{Rn}$  and  $^{220}\text{Rn}$  are the gaseous radioactive products of the decay of the radium isotopes  $^{226}\text{Ra}$  and  $^{224}\text{Ra}$  which are present in all terrestrial materials. Some of the

atoms of these radon isotopes are released from the solid matrix of the material by recoil when the radium decays.

The decay must occur within the recoil distance of the grain surface so that a radon atom to escape from the mineral grain into the pore space. The range of recoil distance for  $^{222}\text{Rn}$  is 20-70 nm in common minerals, 100 nm in water, and 63  $\mu\text{m}$  in air (UNSCEAR, 2000). Radon atom entering the pore space are then transported by diffusion and advection through this space until they in turn decay or are released into the atmosphere(exhalation). Radon generation and transport in porous materials involve solid, liquid, and gas phase in the process of emanation, diffusion, advection, absorption in the liquid phase, and adsorption in the solid phase.

The additional pathways for radon release are microscopic fractures and fissures, called nanopores, and pits or openings caused by previous radioactive decays provide . Particularly in sand-sized and larger grains, nanopores can increase the specific surface area of the grain, enhancing emanation by one or two orders of magnitude.

The moisture soil plays an important role in the emanation of radon and its diffusion in soil for several reasons. Soil moisture, in the form of a thin film of water surrounding soil grains, directly affects radon emanation by capturing the radon that recoils from the solid matrix. These captures increase the likelihood that radon atom will remain in the pore space instead of crossing the pores and imbedding themselves in adjacent soil grains.

Molecular diffusion is the main mechanism for the entry of radon into the atmosphere. Although diffusive entry of radon into the outdoor atmosphere usually dominates, there is also some advection caused by wind and changes in barometric pressure. Measurements of exhalation rates of radon from soil show a variability of radon concentrations in near surface pore spaces. Concentrations of  $^{222}\text{Rn}$  in soil gas vary over many orders of magnitude from place to place and show significant time variations at any given site. Data have shown that there were prominent increases in radon concentrations in outdoor air and in ground water just before the larger earthquake at Kobe, Japan in 1995 (UNSCEAR, 2000).

Under normal conditions, thoron concentrations in soil gas would be roughly comparable to or perhaps somewhat less than the  $^{222}\text{Rn}$  concentrations because of

the generally similar production rates in rocks and soils and their behavior in the ground. On the other hand, high thoron entry rates from the ground are rarely encountered. Whereas fractures in the ground and/or bedrock allow  $^{222}\text{Rn}$  to be pulled to the surface from substantial depths (and volumes), the time frame may be such that most of the thoron present at these depths decays before reaching the surface.

### **1.2.2 Radon in mines**

There are many sources of radon. Radium's availability as part of ores is the major source of radon in mines. Production and release into the mine atmosphere depends on the grades, its emanation power, porosity and permeability of host rock to radon and moisture content of the ore. Water coursing through the ore body remains under high pressure which dissolves significant amounts (up to 23%) of radon available in the ore spaces through which it passes; this acts as an efficient medium of transport for radon carrying it from far-off places and discharging it into the mine cavity. The moment radon-laden water enters the mine openings and flows through the mine galleries, dissolved radon is released due to depressurization and constant agitation. (UNSCEAR, 2000)

Radon produced by the decay of radium located in the grains of the ore matrix moves out by recoil into the pore spaces and remains locked up there. Mining operations such as drilling and blasting bring fresh radon into the mine air.

On entering the mine air by various processes remains there till radon gas is carried along in the ventilation current and discharged above ground. Because of its comparatively long half life (3.82 days) compared with the residence time of air in a mine (approx. 40 mins.), radon concentration increases in proportion to its rate of release and travel time along different passages of the mine. (UNSCEAR, 2000)

### **1.2.3 Radon in outdoor air**

Concentrations of radon in the outdoor environment are affected not only by the magnitude of the exhalation rates in the general area but also by atmospheric

mixing phenomena. Solar heating during the daytime tends to induce some turbulence, so that radon is more readily transported upwards and away from the ground. At night and in the early morning hours, atmospheric (temperature) inversion conditions are often found, which tend to trap the radon closer to the ground. This means outdoor radon concentrations can vary diurnally by a factor of as much as ten. There are also seasonal variations related to the effects of precipitation or to change in the prevailing winds. These effects must be taken into account when interpreting the available measurements (UNSCEAR, 2000)

Although data are scarce for thoron, considerable variability from place to place would be expected because of thoron's short half-life. Even more important is the fact that thoron's short half-life results in a very steep vertical gradient in its atmospheric concentration at any location. A few measurements show that concentrations a few centimeters above the ground surface and concentrations at a height of 1 m vary by a factor of about 10. This gradient would be expected to vary considerably with atmospheric conditions. Thus, pronounced time variations would be expected at any height above the ground at any location. (UNSCEAR, 2000)

#### **1.2.4 Radon in indoor air**

In the early years of the 20th century, the newly discovered thorium and radium emanation (i.e. thoron and radon), the active deposit (radon daughters) and radium itself were investigated in laboratories in more detail. Indoor air is a dominant exposure for humans, where more than half of the body's intake during a lifetime is air inhaled in the home.

Exposure to the short-lived daughters of radon is not confined to the underground miners. Uranium is widely distributed constituent of the earth's crust, typically in 2-4 parts per million and in consequence is found in most materials commonly used by the building industry. Radon, being a noble gas, diffuses from the subsoil below the building into room air, where it and its daughters are available for inhalation by the room occupants (Cliff, 1978).

All building materials that originate from minerals may contain amounts of radionuclides such as uranium and thorium, which are created from their radioactive

decay chains. Of these, the most significant is radium (Ra-226). Presence of Ra-226 in building materials affects persons living in dwellings either by inhalation of radon daughters, that decay from radium and are released from the building materials to the indoor air, or by hard gamma radiation released from the building material as a consequence of the radioactive decay of the natural radionuclides (7-2). In addition to the building materials, the natural gas used domestically (7-3) and the underground-derived water supply (7-4) are potential sources of indoor radon and its daughters.

Outdoors, where it is diluted to low concentrations in the air,  $^{222}\text{Rn}$  poses significantly less risk than indoors. In the indoor environment, however,  $^{222}\text{Rn}$  can accumulate to significant levels. The magnitude of  $^{222}\text{Rn}$  concentration indoors depends primarily on the type of construction materials of the building and the amount of uranium deposition in the underlying soil. The soil composition under and around a house affects  $^{222}\text{Rn}$  levels and the ease with which  $^{222}\text{Rn}$  migrate into a house. Normal pressure differences between the house and the soil can create a slight under pressure in the house that can draw  $^{222}\text{Rn}$  gas from the soil into the building.  $^{222}\text{Rn}$  moves more rapidly through permeable soils, such as coarse sand and gravel, than through impermeable soils, such as clays. Fractures in any soil or rock allow  $^{222}\text{Rn}$  to move more quickly. In a very small number of houses  $^{222}\text{Rn}$  levels are generally highest in basements and ground floor rooms that are in contact with the soil. Factors such as design, construction, and ventilation of the house affect the pathways and sources that can draw  $^{222}\text{Rn}$  indoors (Tassos and Haralabos, 2003).

Radon Gets in Through:

1. Cracks in solid floors
2. Construction joints
3. Cracks in Walls
4. Gaps in Suspended floors
5. Gaps around service pipes
6. Cavities inside Walls
7. The Water Supply

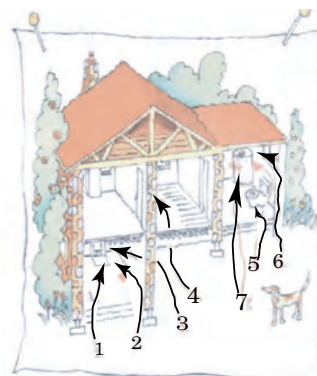


Figure 1.2: Radon entry into the house (EPA, 2005)

The chances of increased level of indoor radon is higher in areas of higher uranium laden soil. But some houses in areas with lots of uranium in the soil have low levels of indoor  $^{222}\text{Rn}$ , and other houses on uranium-poor soils have high levels of indoor  $^{222}\text{Rn}$ . This shows the amount of  $^{222}\text{Rn}$  in a house is affected by other factors in addition to the presence of uranium in the underlying soil. One example is the permeability of the soil (Tassos and Haralabos, 2003). Considering all of the factors mentioned above, the factors that determine the entry rate are many, varied, and very site-specific.

Successful mitigation strategies, such as identifying and sealing a limited number of entry pathways or effectively ventilating the soil immediately adjacent to the foundation, tend to work because radon entry into structures can be fairly readily prevented, or at least substantially reduced. Other techniques aim at reducing the building/ground pressure differential that drives the advection; the radon convections are typically reduced by 80-90 percent. Improvements in ventilation systems normally change radon concentration by less than 50 percent (UNSCEAR, 2000).

Because of thoron's short half-life, it was once thought that the only mechanism for significant thoron entry would be infiltration of outdoor air diffusion from building materials. But recent investigations have shown that entry through the foundation can also be possible. Given the comparable concentrations of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  usually found in outdoor air, soil gas, and building material pore spaces, it is not unexpected that indoor air concentrations of the two gases (ground floor level only) are often roughly comparable (UNSCEAR, 2000).

Concentration of radon and its progeny are usually higher in indoor air than in outdoor air; exceptions are in tropical areas, the concentrations in well-ventilated buildings are essentially the same in outdoor air. There is also diurnal variations in radon levels. Maxima occurs in the winter months around December-January and minima in summer around June-July. Values are maximum in nights, and around twice the values at noon.

Radon, being inert gas, can diffuse far away from its origin. However, in indoor air, where only the concentration of  $^{222}\text{Rn}$  is critical, given the short half-lives of the other isotopes  $^{220}\text{Rn}$  is sometimes present in wall materials, but its contribution can often be removed by an extra layer of paint.

Table 1.2: Radon concentrations in dwellings (UNSCEAR, 2000)

Region	Country	Population In 1996 (10 <sup>6</sup> )	Radon concentration (Bqm <sup>-3</sup> )			
			Arith. mean	Geom. mean	Max. Value	Geom. Standard Deviation
<b>Africa</b>	Algeria	28.78	30		140	
	Egypt	63.27	9		24	
	Ghana	17.83			340	
<b>North America</b>	Canada	29.68	34	14	1720	3.6
	United States	269.4	46	25		3.1
<b>South America</b>	Argentina	35.22	37	26	211	2.2
	Chile	14.42	25		86	
	Paraguay	4.96	28		51	
<b>East Asia</b>	China	1232	24	20	380	2.2
	Honk Kong	6.19	41		140	
	India	944.6	57	42	210	2.2
	Indonesia	200.45	12		120	
	Japan	125.4	16	13	310	1.8
<b>West Asia</b>	Armenia	3.64	104		216	1.3
	Iran(Islamic Rep.of)	69.98	82		3070	
	Kuwait	1.69	14	6	120	
	Syria	14.57	44		520	
<b>North Europe</b>	Denmark	5.24	53	29	600	2.2
	Estonia	1.47	120	92	1390	
	Finland	5.13	120	84	20000	2.1
<b>West Europe</b>	Austria	8.11		15	190	
	Belgium	10.16	48	38	12000	2
	France	58.33	62	41	4690	2.7
	Germany	81.92	50	40	>10000	1.9
<b>Eastern Europe</b>	Bulgaria	8.47		22	250	
	Czech Republic	10.25	140		20000	
	Hungary	10.05	107	82	1990	2.7
<b>South Europe</b>	Albania	3.4	120	105	270	2
	Croatia	4.5	35	32	92	
	Spain	39.67	86	42	15400	3.7
<b>Oceania</b>	Australia	18.06	11	8	420	2.1
	New Zealand	3.6	20	18	90	
Median Pop.-weighted average			46	37	480	2.2
			39	30	1200	2.3

Table 1.3: Thoron concentrations in outdoor and indoor air (UNSCEAR, 2000)

Region / Country	Equilibrium equivalent concentration (Bq m <sup>-3</sup> )		<sup>220</sup> Rn/ <sup>222</sup> Rn EEC ratio	
	Outdoors	Indoors	Outdoors	Indoors
<b>North America</b>				
United States	0.09 (0.03-0.3)	0.5 (0.03-4.7)		0.04
<b>East Asia</b>				
China	0.4	0.8	0.05	0.07
Hong Kong	0.3 (0.1-0.5)	0.8 (0.4-1.2)	0.04	0.06
Japan	0.09 (0.03-0.12)	0.6 (0.4-0.9)		0.1
Malaysia	0.5 (0.3-1.8)	1.1 (0.4-2.5)	0.08	0.08
<b>North Europe</b>				
Norway		0.7 (0.07-1.1)		0.04
Sweden		0.3 (0.1-0.6)		0.01
<b>West Europe</b>				
France		0.8 (0.6-13.3)		0.03
<b>United Kingdom</b>		0.3 (0.07-1.1)		0.02
<b>Central Europe</b>				
Germany		0.5 (0.1-1.0)		
Moldova	0.2	1.0 (0.1-6.4)	0.04	0.05
Romania	0.3 (0.1-0.6)	1.1 (0.1-6.4)	0.05	0.04
<b>East Europe</b>				
Russia		1.1-7.1		0.09 (0.02-0.24)
<b>South Europe</b>				
Italy		12 (0.5-7.6)		0.11 (0.01-0.38)
Slovenia	0.12 (0.05-0.37)		0.013	
Range	0.09-0.5	0.2-12	0.01-0.08	0.01-0.5

### 1.2.5 Radon in water

Radon is also found in the water, in homes, in particular, homes that have their own well rather than municipal water. When the water is agitated, as when showering or washing dishes, radon escapes into the air. However, radon from domestic water generally contributes only a small proportion (less than one percent ) of the total radon in indoor air.

Municipal water systems hold and treat water, which helps to release radon, so that levels are very low by the time the water reaches homes. But those who have private wells, particularly in areas of high radium soil content, may be exposed to high levels of radon (EPA, 2005).

## 1.3 Health hazards due to radon

### 1.3.1 Discovery

Though the hypothesis of correlation between lung cancer and radon inhalation was supported by more precise radon measurements carried out in the 1920, leading Ludewig and Lorenzer in 1924 to make a definitive statement of the link, the hypothesis was not generally accepted. An expert pathological study group in Germany in its 1926 report ascribed the cancer to inhalation toxic ore dusts (Nagaratnam, 1994).

Association between cancer and radioactivity had been demonstrated as early as 1902, starting from the first cases of skin cancer induced by x-rays or radium emanation. Since detailed measurements showed a high concentration of radon in the air of mines at Schneeberg and Jachymov, a relation between radon and lung cancer came to be suspected. Probably the first person to suspect this casual line was H.E.Muller, a miner from Saxony.

It is interesting to recall that as early 1907, Lord Rutherford in a lecture in Canada, speculated that the inhalation of radon and progeny may play some part in the physiological processes, but it was no more than a wild speculation. In 1936 Rajewsky initiated a comprehensive study programme involving radon measurements in Schneeberg mines, measurements of the alpha activity in the tissue samples and histopathological analysis of lung tissues from miners who had died of lung cancer.

At that time, the average radon concentration in most mines at Schneeberg was in the range of 70 – 120 KBqm<sup>-3</sup>. In one time, however, the mean level was 500 KBqm<sup>-3</sup>, and most workers here died from lung cancer; it was called 'death mine'. On the basis of these studies Rajewsky was able to establish conclusively in 1940 the casual link. However, till 1945, no quantitative dose-response relation could be worked out, nor was the possible role of inhalation of short-lived radon progeny realized (Nagaratnam, 1994).

Table 1.4: Average worldwide exposure to natural radiations (UNSCEAR, 2000)

Source of exposure	Average effective dose (mSv) Average	Typical range
<b>Cosmic radiation</b>		
Directly ionizing and photon component	0.28	
Neutron component	0.1	
Cosmogenic radionuclides	0.01	
<b>Total cosmic and cosmogenic</b>	<b>0.39</b>	<b>0.3-1.0</b>
<b>External terrestrial radiation</b>		
Outdoors	0.07	
Indoor	0.41	
<b>Total external terrestrial radiation</b>	<b>0.48</b>	<b>0.3-0.6</b>
<b>Inhalation exposure</b>		
Uranium and thorium series	0.006	
Radon ( $^{222}\text{Rn}$ )	1.15	
Thoron ( $^{220}\text{Rn}$ )	0.1	
<b>Total inhalation exposure</b>	<b>1.26</b>	<b>0.2-10</b>
<b>Ingestion exposure</b>		
$^{40}\text{K}$	0.17	
Uranium and thorium series	0.12	
<b>Total ingestion exposure</b>	<b>0.29</b>	<b>0.2-0.8</b>
<b>Total</b>	<b>2.4</b>	<b>1-10</b>

The radiological impact from the above nuclides is due to radiation exposure of the body by the gamma rays and irradiation of the lung tissues from inhalation of Radon and its progeny. From the natural risk point of view, it is necessary to know the dose limits of public exposures and to measure the natural environmental radiation level provided by ground, air, water, foods, building interiors, etc., for the estimation of the exposures to natural radiation sources.

Although, the progeny of radon are now a well recognized cause of lung cancer, radon itself has again become a topic of controversy and public health concern because in 1950s it has been found to be a ubiquitous indoor air pollutant to which all persons are exposed.

Arguably, the most significant event to draw the radon problem to the attention of the media and ultimately the public at large were the remarkable circumstances surrounding a person called Stanley Watras and the event is commonly referred as The Watras incident. Stanley Watras was living in Boyertown, near to Philadelphia, and was an employee of the Limerick Nuclear Power plant in Pennsylvania. The home of the unfortunate Watras was built on an excavated vein of uranium in the Reading Prong, a Precambrian rock body relatively high in uranium. During December 1984, Watras set off radiation alarms on his way into work. This happened every working day for two weeks and he was regularly decontaminated while the authorities attempted to find the radioactive source. Eventually, the source was found to be radon gas in his domestic dwelling, not a discharge at the plant itself. The levels of radon in his home appeared astonishing to those monitoring at the time, being in the region of 100 000 Bqm – 3. The risks associated with living in that house were estimated to be equivalent to smoking 135 packs of cigarettes per day (National Research Council, 1999).

### **1.3.2 Effects of radon on the body**

The health hazard of radon is principally due to its short-lived daughters' inhalation, especially  $^{218}\text{Po}$  and  $^{214}\text{Po}$ , which use their physical properties to spread or attach like aerosols do, trapped in the lung and depositing their alpha-particle energies in the tissue, producing higher ionization density than beta particles or gamma-rays.

Since the life time of  $^{222}\text{Rn}$  is long relative to breathing times most of the

inhaled  $^{222}\text{Rn}$  will be exhaled rather than decaying or becoming lodged in the lungs and later decaying. In contrast, the immediate, promptly decaying daughters of  $^{222}\text{Rn}$ : ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ , and  $^{214}\text{Po}$ ) attach to a surface, typically of aerosols, which can be inhaled. They then deposit on epithelial surfaces within the lung, and shortly decay.

The result is that the sensitive surfaces of the bronchi are irradiated by these decays, the most energetic and destructive of which are the heavily ionizing, short range particles from the polonium isotopes  $^{216}\text{Po}$  ( $E= 7.7 \text{ MeV}$ ) and  $^{218}\text{Po}$  ( $E= 6.0 \text{ MeV}$ ) (Akeel and Hasan, 2003). The alpha particles from the radon daughters can irradiate the respiratory tract. The radon daughters from  $^{226}\text{Ra}$  contribute the greatest doses to the lungs of people. Thorium ( $^{232}\text{Th}$ ) also decays to radon ( $^{220}\text{Rn}$ ) by a similar decay series. The very short half-life of thoron prevents it from diffusing very far before it decays to thoron progeny. Therefore the dose from thoron is often inconsequential in homes compared to radon ( $^{222}\text{Rn}$ ).

Most of the radon ingested with water is excreted through the urine over several days. There is some risk from drinking water with elevated radon, because radioactive decay can occur within the the body when tissues, such as the stomach lining, would be exposed. However, particles emitted by radon and its decay product in water prior to drinking quickly lose their energy and are taken up by other compounds in water, and do not themselves pose a health concern (EPA, 2005).

## 1.4 Main objective of the study

Nowadays it is known that high concentration of radon is carcinogenic. People must be aware of environs radon concentration and take the necessary steps for the reduction. Also measuring the radon level of a given locale as part of accumulation of environmental radiation data is necessary.

Future generations may use the data for possible nuclear activities. In this aspect, Ethiopia seem lagging behind even by African standards. Therefore this work can be regarded as one of the first serious attempts toward measurement and accumulation of base line data in environmental radiation. It can also serve as a pioneering work for further researches in the area.

# Chapter 2

## Radon Measurement Methods

Many techniques have been developed over the years for measuring radon and radon progeny in air. Radon and radon progeny emit alpha and beta particles and gamma rays. Therefore, numerous techniques have been developed for measuring these radionuclides based on detecting alpha particles, beta particles, or gamma rays, independently or in some combination.

Radon measurement techniques are generally classified as *passive* and *active* methods. In the passive methods of measuring radon the measurement is usually made over a long period of time and the result is reported as the average over the measured time interval. In active measurement method, the radon concentration is measured at given measuring point in time.

In this chapter both methods are described and the different methods used for each category is discussed. These methods are standard methods which are described in detail in "indoor radon and radon decay product measurement device protocols" published by U.S. environmental protection agency (EPA, 1992).

### 2.1 Passive Measuring Methods

In this category of measurement methods; Solid state nuclear track detector(SSNTD), Activated charcoal(AC), Charcoal liquid scintillation(LS), and Electret ion chamber(EC) methods are the prominent ones.

### **2.1.1 Solid State Nuclear Track Detector**

Solid State Nuclear Track Detector (SSNTD) consists of a small piece of plastic or film enclosed in a container. Radon diffuses into the container and particles emitted by the radon and its decay products strike the detector and produce submicroscopic damage tracks. At the end of the measurement period, the detectors are returned to the laboratory. Plastic detectors are placed in a caustic solution that accentuates the damaged tracks so they can be counted using an automated counting system. The number of tracks per unit area is correlated to the radon concentration in air. The number of tracks per unit of analyzed detector area produced per unit of time minus the background is proportional to the radon concentration.

SSNTD is mostly made up of detection material and detection chamber. The most popular member of SSNTDs family detection material is CR-39. It has good sensitivity, stability against various environmental factors, and high degree of optical clarity. The detector chamber is a cylindrical cup. Carbon is impregnated in the wall material, polypropylene, to enhance electrical conductivity and to avoid the problem of electrostatic charge. Radon enters the holder with a half-time for entry about 1 minute, which is short compared with the radon half-life of 3.82 days. This means that the radon concentration inside the detector chamber quickly approaches that of outside. It can be shown that the long-term average radon concentration inside the detector chamber is the same as that outside, despite any variations in the outside concentration. But the radon concentration may be overestimated because the short half-time for entry will allow some thoron to enter the detector (Ahn and Lee, 2005).

The optimal use of any track detector is largely dependent on standardization of various etching parameter. CR-39 samples are irradiated using an alpha source. Irradiated CR-39 samples can be etched in a solution. NaOH solution is most popular etchant and has been extensively studied. Varying concentrations of NaOH solutions can be used at different temperatures and periods. The etched tracks can be observed using an optical microscope. Calibration experiment can be carried out to evaluate the relationship between the track density recorded and the radon concentration. (Ahn and Lee, 2005)

### 2.1.2 Activated charcoal (AC)

In the last years, a lot of work has been done on the measurement of radon by adsorption on activated charcoal based on its well known property of adsorbing different gases and vapors, including radon. In most cases, the quantity of  $^{222}\text{Rn}$  adsorbed is later measured by counting the gamma ray emissions of both  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ . This is possible due to the short half lives of these progeny.

ACs are passive devices requiring no power to function. The passive nature of the activated charcoal allows continual adsorption and the desorption of radon. During the measurement period, the adsorbed radon undergoes radioactive decay. Therefore, the technique does not integrate uniformly radon concentrations during the exposure period. As with all devices that store radon, the average concentration calculated using the mid-exposure time is subject to error if the ambient radon concentration varies substantially during the measurement period.

A device used most commonly consists of a circular container filled with activated charcoal. One side of the container is fitted with a screen that keeps the charcoal in but allows air to diffuse into the charcoal.

In some cases, the charcoal container has a diffusion barrier over the opening. For longer exposures, this barrier improves the uniformity of response to variations of radon concentration with time. Desiccant is also incorporated in some containers to reduce interference from moisture adsorption during longer exposures. Another variation of the charcoal container has charcoal packaged inside a sealed bag, allowing the radon to diffuse through the bag. All ACs are sealed with a radon-proof cover to outer container after preparation.

The measurement is initiated by removing the cover to allow radon-laden air to diffuse into the charcoal bed where the radon is adsorbed onto the charcoal. At the end of measurement period, the device is resealed securely and returned to laboratory for analysis.

At the laboratory, the ACs are analyzed for radon decay products by placing the charcoal, still in its container, directly on a gamma detector. Corrections may be needed to account for the the reduced sensitivity of the charcoal due to adsorbed water. This correction may be done by weighing each detector when it is prepared and then reweighing it when it is returned to the laboratory for analysis. Any

weight increase is attributed to water adsorbed on the charcoal. The weight of water gained is correlated to a correction factor. This conversion factor is used to correct the analytical results.

This correction is not needed if the configuration of the AC is modified to reduce significantly the adsorption of water and if the user has demonstrated experimentally that, over a wide range of humidities, there is negligible change in the collection efficiency of the charcoal within the specified exposure period.

The advantage of this technique is that it is completely passive, of very low cost, higher efficiency and a lower detection limit compared with gamma-spectrometry methods. This measurement method can be fully automated and is completely independent of humidity till 7 days of exposure.

### **2.1.3 Charcoal liquid scintillation (LS)**

The frequently used type of LS device is a capped liquid scintillation vial which contains charcoal. In some cases, the vial contains a diffusion barrier over the charcoal which improves the uniformity of response of the device to variations of radon concentration with time, particularly for longer exposures. Some LS devices include a few grams of desiccant which reduces interference from moisture adsorption by the charcoal. All LS devices are sealed with a radon proof closure after preparation.

A measurement with LS device is initiated by removing the radon proof closure to allow radon-laden air to diffuse into the charcoal where the radon is adsorbed. At the end of the exposure (typically two to seven days), the device is resealed securely and returned to the laboratory for analysis.

At the laboratory, the devices are prepared for analysis by radon adsorption techniques. This technique transfers reproducibly a major fraction of the radon adsorbed on the charcoal into a vial of liquid scintillation fluid containing the dissolved radon are placed in a liquid scintillation counter and counted for a specified number of minutes (e.g. 10 minutes) or until the standard deviation of the count is acceptable (e.g., less than 10 percent)

### **2.1.4 Electret ion chamber (EC)**

There are short-term and long-term ECs. They require no power, and function as true integrating detectors, measuring the average concentration during the measurement period.

The EC contains a charged electret (an electrostatically charged disk of Teflon) which collects ions formed in the chamber by radiation emitted from radon and radon decay products. When the device is exposed, radon diffuses into the chamber through filtered opening. Ions which are generated continually by the decay of radon and radon decay products are drawn to the surface of the electret and reduce its surface voltage. The amount of voltage reduction is related directly to the average radon concentration and the duration of exposure period. ECs can be deployed for exposure periods of two days to 12 months, depending upon the thickness of the electret and the volume of the ion chamber chosen for use. These deployment periods are flexible, and valid measurements can be made with other deployment periods depending on the application.

The electret must be removed from the EC chamber and the electret voltage measured with a special surface voltmeter both before and after exposure. To determine the average radon concentration during the exposure period, the difference between the initial and final voltages is divided first by a calibration factor and then by the number of exposure days. A background concentration equivalent of ambient gamma radiation is subtracted to compute radon concentration. Electret voltage measurements can be made in a laboratory or in the field.

## **2.2 Active Measuring Methods**

Grab radon sampling method, Pump/collapsible bag method and Three-day integrating evacuated scintillation cells method are the prominent radon measurement methods in this category. Brief description of these methods is presented in this section.

### **2.2.1 Pump / collapsible bag (PB)**

One of the older and simpler methods of making an integrated measurement of the concentration of radon over a period of time is to collect a sample of the ambient

air in a radon-proof container over the desired sampling time period and measure the resulting radon concentration in the container. One practical method is to use a small pump with a very low and uniform flow rate to pump ambient air into an inflatable and collapsible radon proof bag.

### **2.2.2 Three-day integrating evacuated scintillation cells (SC)**

This method typically uses Lucas type scintillation cells that have been outfitted with a restricter valve attached to the main valve. Samples are collected by opening the valve on an evacuated cell. The restricter valve is set so that the cells fill from a 30-inch mercury (Hg) vacuum to about 80% of its capacity over a three day period. At the end of the measurement period, the valve is closed and returned to the analysis laboratory. Since the volume of the cell is known, the exact volume of filtered air collected over the three day measurement period can be calculated from the vacuum gauge reading at the end of the sampling period.

The sample is analyzed on an alpha scintillation counter. Prior to counting, the pressure in the cell is brought to one atmosphere by adding radon-free air so that the sample is analyzed under the same conditions that prevailed during calibration of the cell. To allow radon and progeny to grow into equilibrium and to allow any radon decay products that may have been collected to decay, the sample should be collected no sooner than four hours after the end of the measurement.

### **2.2.3 Grab radon sampling**

In this method three techniques which are frequently used. These are

**Grab radon / scintillation cell (GS) method** In this method sample is collected by pumping air through an activated charcoal. A sample of air is drawn into and sealed in a flask or cell that has zinc sulfide phosphor coating on its interior surfaces. One surface of the cell is fitted with a clear window that is put in contact with a photomultiplier tube to count scintillations resulting from alpha disintegrations from the air sample interacting with the zinc sulfide coating. The number of pulses is proportional to the radon concentration in

the cell. The cell is counted about four hours after filling to allow the short-lived radon decay products to reach equilibrium with the radon. Correction factors are applied to the counting results to compensate for decay during the time between collection and counting and for the decay during counting if the counting time is long ( $>$  one hour). In a variation of this method, used in some portable instruments, air is pumped continuously through a flow-through-type scintillation cell for just a few minutes. Alpha particles resulting from the decay of radon gas and decay products are counted as the gas is swept through.

**Grab radon/ activated charcoal (GC) method** Air pumped through activated charcoal to collect the sample is used in this method. A charcoal-filled cartridge is placed into a sampler and air is pumped through the carbon cartridge. 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. A sampling duration of one hour has been found to be optimal for most systems. The cartridge must be weighed prior to and after sampling in order to correct for the reduced sensitivity of the charcoal due to adsorbed water. The cartridges are analyzed by placing them on a sodium iodide gamma scintillation system or a germanium gamma detector.

**Grab radon pump / collapsible bag (GB) method** This method uses the same technology described in the pump / collapsible bag devices (PB) which will be discussed in the next subsection. The GB method discussed here differs only in that the bag is filled over a much shorter collection period than the PB.

**Grab radon and thoron progeny 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. This method can be used for simultaneous measurement of radon and thoron decay products. The experiment which will be discussed in this paper was done using this method. So the method will be explained in detail in the next chapter.

# Chapter 3

## Materials and Methods

### 3.1 Materials

The materials used in the experimental work were Sampling head, Glass fiber filter, End-window GM tube, Nuclear counter containing high voltage supply, and Air suction pump. The experimental setup is shown in figure 3.1

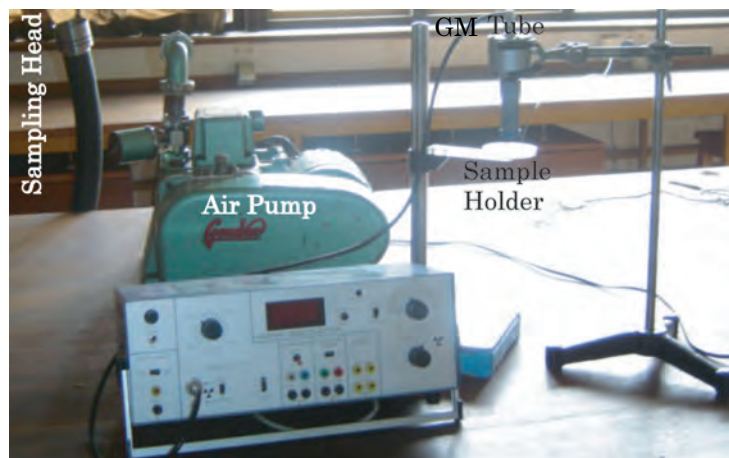


Figure 3.1: The experimental setup

In this kind of setup Papp (Papp, 1997) suggested, high sampling flow rate and long duration of sampling to lower the limit of detection and improve the accuracy. In this experiment 20 minutes of air sampling time was used and the duration of counting was from 6 to 10 hours.

### 3.1.1 The sampling head

The design, which is same as that used by Z. Papp et al. details of the sampling head are shown in figure 3.2. It was crafted by ANBO Engineering in Addis Ababa.

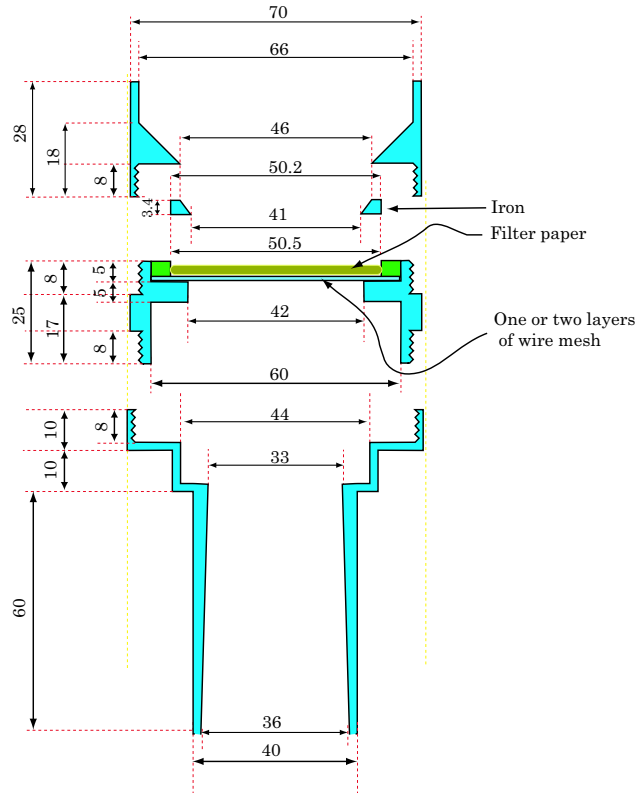


Figure 3.2: Air Sampling Head Blueprint



Figure 3.3: Parts of the Sampling Head



Figure 3.4: The Air Sampling Head assembled

### 3.1.2 Filtering air

Aerosol samples were collected on a glass fiber filter by drawing air through it. The filter used has a thickness of  $0.0095 \text{ g cm}^{-2}$  and dimensions of  $2.9 \text{ cm} \times 2.8 \text{ cm}$ . Aerosols were collected on a circular spot of  $1.8 \text{ cm}$  diameter. To pump air Genevao rotary piston vacuum pump was used. The air flow rate, during sampling, was  $112 \text{ litres/min}$ . A rubber tube connected the sampling head and the pump.

## 3.2 Methods

### 3.2.1 The counting process

In the counting process, we used a GM tube with a nuclear counter. The GM counter has a window thickness of  $0.002 \text{ g cm}^{-2}$  and a diameter of  $1.8 \text{ cm}$ . The operating voltage the GM counter was determined to be  $440 \text{ V}$ , except in the last three experiments(of the 38) which we used  $500 \text{ V}$ .

We sampled indoor (Nuclear physics lab. of A.A.U.) and outdoor air alternatively. Air was sampled on the filter for about 20 minutes(sometimes with some more seconds) from 6:02:00 to 6:22:00 (except minor variations)for 38 days. After a short time of the end of sampling (1 minute for most of the cases), the filter was placed below the GM tube. The distance between the tube and the filter was  $6 \text{ mm}$ . The total beta counts over a series of successive time intervals (for 1, 2, 4, 8, 12, 16 min, each for an hour) were recorded.

The background counting rate was also measured by using clean filter and was found to be  $33.75/\text{min}$ .

### 3.2.2 Mathematical model

Activity concentration of airborne radon decay products and thoron decay products were determined by similar mathematical computations used by Raabe and Wrenn (Raabe and E.Wrenn, 1969) and Papp and Daróczy (Papp and Daróczy, 1997)

The computations proceed as follows:

1. The differential equations which describe the collection and the decay during collection of the daughters are solved.
2. The differential equations which describe the decay of the daughters on the sample after collection is complete are solved.
3. A weighted least-squares regression analysis is performed to mathematically fit the observations of counts during various counting periods to the general decay equation for the sample. This provides the number of atoms of each daughter present on the sample at the end of collection period from which the number concentrations of the daughters in the atmosphere are calculated.

Let the numbers 1 to 7 represent  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  for radon decay products ;  $^{212}\text{Pb}$   $^{212}\text{Bi}$  and  $^{208}\text{Tl}$  for thoron decay products and U for unknown beta emitter with long half life respectively.

#### Constants:

$K$  = Air sampling flow rate;

$T$  = Air sampling time;

$t_b$  = beginning of a counting time interval;

$t_c$  = end of a counting time interval;

$C$  = total beta counts measured over the time interval ( $t_b$ ,  $t_c$ );

$M$  = number of the counting time intervals ;

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

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

#### 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 , ( $i = 1,2,\dots,7$ ) ;

$N_{Ti}$  = 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 sampling );  
 CR = total beta counting rate .

Assuming that  $k$  and  $n_i$  are constant during the sampling period the differential equations describing the buildup and decay of atoms on the sample are :

$$dN_1 = kn_1 dt - \lambda_1 N_1 dt, \quad N_1(0) = 0 \quad (3.1a)$$

$$dN_2 = kn_2 dt - \lambda_2 N_2 dt + \lambda_1 N_1 dt, \quad N_2(0) = 0 \quad (3.1b)$$

$$dN_3 = kn_3 dt - \lambda_3 N_3 dt + \lambda_2 N_2 dt, \quad N_3(0) = 0 \quad (3.1c)$$

$$dN_4 = kn_4 dt - \lambda_4 N_4 dt, \quad N_4(0) = 0 \quad (3.1d)$$

$$dN_5 = kn_5 dt - \lambda_5 N_5 dt + \lambda_4 N_4 dt, \quad N_5(0) = 0 \quad (3.1e)$$

$$dN_6 = kn_6 dt - \lambda_6 N_6 dt + \lambda_5 N_5 dt, \quad N_6(0) = 0 \quad (3.1f)$$

$$dN_7 = kn_7 dt - \lambda_7 N_7 dt, \quad N_7(0) = 0 \quad (3.1g)$$

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

$$N_1 = kn_1 \left[ \frac{1 - e^{-\lambda_1 t}}{\lambda_1} \right] \quad (3.2a)$$

$$N_2 = kn_1 \left[ \frac{1 - e^{-\lambda_2 t}}{\lambda_2} - \frac{e^{-\lambda_2 t} - e^{-\lambda_1 t}}{\lambda_1 - \lambda_2} \right] + kn_2 \left[ \frac{1 - e^{-\lambda_2 t}}{\lambda_2} \right] \quad (3.2b)$$

$$N_3 = kn_1 \left[ \frac{1 - e^{-\lambda_3 t}}{\lambda_3} - \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{\lambda_3 - \lambda_2} - \frac{\lambda_2 (e^{-\lambda_2 t} - e^{-\lambda_3 t})}{(\lambda_3 - \lambda_2)(\lambda_1 - \lambda_2)} + \frac{\lambda_2 (e^{-\lambda_3 t} - e^{-\lambda_1 t})}{(\lambda_1 - \lambda_3)(\lambda_1 - \lambda_2)} \right] \\ + kn_2 \left[ \frac{1 - e^{-\lambda_3 t}}{\lambda_3} - \frac{(e^{-\lambda_2 t} - e^{-\lambda_3 t})}{\lambda_3 - \lambda_2} \right] + kn_3 \left[ \frac{1 - e^{-\lambda_3 t}}{\lambda_3} \right] \quad (3.2c)$$

$$N_4 = kn_4 \left[ \frac{1 - e^{-\lambda_4 t}}{\lambda_4} \right] \quad (3.2d)$$

$$N_5 = kn_4 \left[ \frac{1 - e^{-\lambda_5 t}}{\lambda_5} - \frac{e^{-\lambda_5 t} - e^{-\lambda_4 t}}{\lambda_4 - \lambda_5} \right] + kn_5 \left[ \frac{1 - e^{-\lambda_5 t}}{\lambda_5} \right] \quad (3.2e)$$

$$N_6 = kn_4 \left[ \frac{1 - e^{-\lambda_6 t}}{\lambda_6} - \frac{e^{-\lambda_5 t} - e^{-\lambda_6 t}}{\lambda_6 - \lambda_5} - \frac{\lambda_5 (e^{-\lambda_5 t} - e^{-\lambda_6 t})}{(\lambda_6 - \lambda_5)(\lambda_5 - \lambda_6)} + \frac{\lambda_5 (e^{-\lambda_6 t} - e^{-\lambda_4 t})}{(\lambda_4 - \lambda_6)(\lambda_4 - \lambda_5)} \right] \\ + kn_5 \left[ \frac{1 - e^{-\lambda_6 t}}{\lambda_6} - \frac{e^{-\lambda_5 t} - e^{-\lambda_6 t}}{\lambda_6 - \lambda_5} \right] + kn_6 \left[ \frac{1 - e^{-\lambda_6 t}}{\lambda_6} \right] \quad (3.2f)$$

$$N_7 = kn_6 \left[ \frac{1 - e^{-\lambda_7 t}}{\lambda_6} \right] \quad (3.2g)$$

$N_{T_i}$  ( $i = 1, 2, \dots, 7$ ), 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 ( $T$ ). So  $N_{T_i}$  ( $i = 1, 2, \dots, 7$ ) can be expressed as:

$$N_{T1} = kn_1 \left[ \frac{1 - e^{-\lambda_1 T}}{\lambda_1} \right] \quad (3.3a)$$

$$N_{T2} = kn_1 \left[ \frac{1 - e^{-\lambda_2 T}}{\lambda_2} - \frac{e^{-\lambda_2 T} - e^{-\lambda_1 T}}{\lambda_1 - \lambda_2} \right] + kn_2 \left[ \frac{1 - e^{-\lambda_2 T}}{\lambda_2} \right] \quad (3.3b)$$

$$N_{T3} = kn_1 \left[ \frac{1 - e^{-\lambda_3 T}}{\lambda_3} - \frac{e^{-\lambda_2 T} - e^{-\lambda_3 T}}{\lambda_3 - \lambda_2} - \frac{\lambda_2(e^{-\lambda_2 T} - e^{-\lambda_3 T})}{(\lambda_3 - \lambda_2)(\lambda_1 - \lambda_2)} + \frac{\lambda_2(e^{-\lambda_3 T} - e^{-\lambda_1 T})}{(\lambda_1 - \lambda_3)(\lambda_1 - \lambda_2)} \right] \\ + kn_2 \left[ \frac{1 - e^{-\lambda_3 T}}{\lambda_3} - \frac{(e^{-\lambda_2 T} - e^{-\lambda_3 T})}{\lambda_3 - \lambda_2} \right] + kn_3 \left[ \frac{1 - e^{-\lambda_3 T}}{\lambda_3} \right] \quad (3.3c)$$

$$N_{T4} = kn_4 \left[ \frac{1 - e^{-\lambda_4 T}}{\lambda_4} \right] \quad (3.3d)$$

$$N_{T5} = kn_4 \left[ \frac{1 - e^{-\lambda_5 T}}{\lambda_5} - \frac{e^{-\lambda_5 T} - e^{-\lambda_4 T}}{\lambda_4 - \lambda_5} \right] + kn_5 \left[ \frac{1 - e^{-\lambda_5 T}}{\lambda_5} \right] \quad (3.3e)$$

$$N_{T6} = kn_4 \left[ \frac{1 - e^{-\lambda_6 T}}{\lambda_6} - \frac{e^{-\lambda_5 T} - e^{-\lambda_6 T}}{\lambda_6 - \lambda_5} - \frac{\lambda_5(e^{-\lambda_5 T} - e^{-\lambda_6 T})}{(\lambda_6 - \lambda_5)(\lambda_5 - \lambda_6)} + \frac{\lambda_5(e^{-\lambda_6 T} - e^{-\lambda_4 T})}{(\lambda_4 - \lambda_6)(\lambda_4 - \lambda_5)} \right] \\ + kn_5 \left[ \frac{1 - e^{-\lambda_6 T}}{\lambda_6} - \frac{e^{-\lambda_5 T} - e^{-\lambda_6 T}}{\lambda_6 - \lambda_5} \right] + kn_6 \left[ \frac{1 - e^{-\lambda_6 T}}{\lambda_6} \right] \quad (3.3f)$$

$$N_{T7} = kn_6 \left[ \frac{1 - e^{-\lambda_7 T}}{\lambda_6} \right] \quad (3.3g)$$

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

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

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

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

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

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

$$n_6 = \frac{\lambda_6 N_{T6}}{k(1 - e^{-\lambda_6 T})} - 0.36n_5 \left[ 1 - \frac{\lambda_6(e^{-\lambda_5 T} - e^{-\lambda_6 T})}{(\lambda_6 - \lambda_5)(1 - e^{-\lambda_6 T})} \right] \\ - 0.36n_4 \left[ 1 - \frac{\lambda_6(e^{-\lambda_5 T} - e^{-\lambda_6 T})}{(\lambda_6 - \lambda_5)(1 - e^{-\lambda_6 T})} - \frac{\lambda_5 \lambda_6 (e^{-\lambda_5 T} - e^{-\lambda_6 T})}{(\lambda_6 - \lambda_5)(\lambda_4 - \lambda_5)(1 - e^{-\lambda_6 T})} \right. \\ \left. + \frac{\lambda_5 \lambda_6 (e^{-\lambda_6 T} - e^{-\lambda_4 T})}{(\lambda_4 - \lambda_6)(\lambda_4 - \lambda_5)(1 - e^{-\lambda_6 T})} \right] \quad (3.4f)$$

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

The solutions of the equation for the decay after the end of sampling are:

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

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

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

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

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

$$N_6 = 0.36 N_{T4} \left[ \frac{\lambda_4 \lambda_5 (e^{-\lambda_5 t} - e^{-\lambda_6 t})}{(\lambda_6 - \lambda_5)(\lambda_4 - \lambda_5)} - \frac{\lambda_4 \lambda_5 (e^{-\lambda_6 t} - e^{-\lambda_4 t})}{(\lambda_4 - \lambda_6)(\lambda_4 - \lambda_5)} \right] \\ + .36 N_{T5} \left[ \frac{\lambda_5 (e^{-\lambda_5 t} - e^{-\lambda_6 t})}{\lambda_6 - \lambda_5} \right] + N_{T6} e^{-\lambda_6 t} \quad (3.5f)$$

$$N_7 = N_{T7} e^{-\lambda_7 t} \quad (3.5g)$$

The count rate (CR) can be described by the equation

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

in the above equation  $^{218}\text{Po}$  is not considered because it is mostly alpha emitter. Replacing  $N_i$  by substituting (3.5a) to (3.5g) and integrating with respect to  $t$

between  $t_b$  and  $t_c$ , it follows:

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

where

$$X_1 = \frac{\epsilon_2 \lambda_2 (e^{-\lambda_1 b} - e^{-\lambda_1 c})}{\lambda_2 - \lambda_1} - \frac{\epsilon_2 \lambda_1 (e^{-\lambda_2 b} - e^{-\lambda_2 c})}{\lambda_2 - \lambda_1} + \frac{\epsilon_3 \lambda_2 \lambda_3 (e^{-\lambda_1 b} - e^{-\lambda_1 c})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} - \frac{\epsilon_3 \lambda_1 \lambda_3 (e^{-\lambda_2 b} - e^{-\lambda_2 c})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} + \frac{\epsilon_3 \lambda_1 \lambda_2 (e^{-\lambda_3 b} - e^{-\lambda_3 c})}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)} \quad (3.8a)$$

$$X_2 = \epsilon_2 (e^{-\lambda_2 b} - e^{-\lambda_2 c}) + \frac{\epsilon_3 \lambda_3 (e^{-\lambda_2 b} - e^{-\lambda_2 c})}{\lambda_3 - \lambda_2} - \frac{\epsilon_3 \lambda_2 (e^{-\lambda_3 b} - e^{-\lambda_3 c})}{(\lambda_3 - \lambda_2)} \quad (3.8b)$$

$$X_3 = \epsilon_3 (e^{-\lambda_3 b} - e^{-\lambda_3 c}) \quad (3.8c)$$

$$X_4 = \epsilon_4 (e^{-\lambda_4 b} - e^{-\lambda_4 c}) + \frac{\epsilon_5 \lambda_5 (e^{-\lambda_4 b} - e^{-\lambda_4 c})}{\lambda_5 - \lambda_4} - \frac{\epsilon_5 \lambda_4 (e^{-\lambda_5 b} - e^{-\lambda_5 c})}{(\lambda_5 - \lambda_4)} + \frac{0.36 \epsilon_6 \lambda_5 \lambda_6 (e^{-\lambda_4 b} - e^{-\lambda_4 c})}{(\lambda_5 - \lambda_4)(\lambda_6 - \lambda_4)} - \frac{0.36 \epsilon_6 \lambda_4 \lambda_6 (e^{-\lambda_5 b} - e^{-\lambda_5 c})}{(\lambda_5 - \lambda_4)(\lambda_6 - \lambda_5)} + \frac{0.36 \epsilon_6 \lambda_4 \lambda_5 (e^{-\lambda_6 b} - e^{-\lambda_6 c})}{(\lambda_6 - \lambda_5)(\lambda_6 - \lambda_4)} \quad (3.8d)$$

$$X_5 = \epsilon_5 (e^{-\lambda_5 b} - e^{-\lambda_5 c}) + \frac{0.36 \epsilon_6 \lambda_6 (e^{-\lambda_5 b} - e^{-\lambda_5 c})}{(\lambda_6 - \lambda_5)} - \frac{0.36 \epsilon_6 \lambda_5 (e^{-\lambda_6 b} - e^{-\lambda_6 c})}{(\lambda_6 - \lambda_5)} \quad (3.8e)$$

$$X_6 = \epsilon_6 (e^{-\lambda_6 b} - e^{-\lambda_6 c}) \quad (3.8f)$$

$$X_7 = \epsilon_7 (e^{-\lambda_7 b} - e^{-\lambda_7 c}) \quad (3.8g)$$

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

$$C_m = \sum_{i=1}^7 N_{Ti} X_{mi}, \quad (m = 1, 2, \dots, M) \quad (3.9)$$

$C_m$  are the measured total beta counts, and  $X_{mi}$  are the values of  $X_i$  for different time intervals, respectively.

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

From these concentrations the Equivalent Equilibrium Concentration (EEC) of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  can be obtained. EEC can be given in terms of the individual decay product concentrations of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  as:

$$\text{EEC}_{\text{Rn}} = 0.105 C_1 + .515 C_2 + 0.380 C_3 \quad (3.10)$$

where  $C_1$ ,  $C_2$ , and  $C_3$  represent concentrations of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  respectively.

$$\text{EEC}_{\text{Th}} = 0.913 C_1 + 0.087 C_2 \quad (3.11)$$

where  $C_1$ ,  $C_2$  represent  $^{212}\text{Pb}$  and  $^{212}\text{Bi}$  respectively.

The Defined solid angle absolute beta counting (DSAABC) method was used to determine the counting efficiencies  $\epsilon_i$ s in (3.8a) to (3.8g), one by one. This method will be explained briefly in the next section.

### 3.2.3 Defined Solid Angle Absolute Beta Counting Method

#### Introduction

Defined solid angle absolute  $\beta$ -counting (DSAABC) is one of the oldest methods for the determination of the activity of  $\beta$ -emitting radionuclides. It was developed nearly 60 years ago by authors like B.P.Burt (Burt, 1949) and others.

This method mostly concerns on the idea that the counting rate of a beta emitter relative to the activity rate depends upon the geometry, the percent of solid angle about the source subtended by the sensitive volume of the counter, and absorption of the radiations by the air and window of the counter and scattering from the source support and assembly.

So the counting efficiencies for the radon decay products and thoron decay products can be evaluated one by one using the defined solid angle absolute beta counting (DSAABC).

However, several problems may arise if someone wants to apply this method in the field of environmental radioanalysis. The most difficult one is that the counting rate is influenced by numerous effects which have to be precisely considered if accurate activity results are needed.

It is often said that it is not worth the trouble to deal with the problems of this method because

1. High accuracy cannot be reached
2.  $\beta$  emitters cannot be easily identified
3. relative counting using certified reference samples is simpler and more accurate
4. more suitable methods are available

The first of the above arguments is indisputable but high accuracy is not necessarily needed in all fields of environmental radioanalysis. As to the second argument, identification of the emitters are exactly known. It has to be emphasized that relative counting as against absolute one may be more accurate only if the reference sample contains the same radionuclide(s) and has exactly the same dimensions, elemental composition and inner distribution of activity as those of the sample to be measured. The complete fulfilment of the above requirements is not easy in most cases, so the relative method may be either simpler or more accurate than the absolute one, but seldom may be both at the same time. It is true that modern methods for the detection of  $\beta$  particles or  $\gamma$ -rays, such as liquid scintillation counting (LSC) or semiconductor  $\gamma$ -spectrometry, are better choices in most cases. But there are special cases when the DSAABC method using an end-window Geiger-Müller (GM) can compete with the above methods or may be even more suitable. Samples have to be liquefied and in most cases also some chemical separations are needed for LSC. Semiconductor  $\gamma$ -spectrometers have low efficiency and cannot be used for the determination of pure  $\beta$ -emitters. If, for example, low activities, have to be measured or the chemical treatment of the sample should be avoided for some reason then the DSAABC method is a cheap and technically simple one (Papp, 1997).

### **Counting efficiency determination**

Assume that the  $\beta$ -radiation of a thin sample containing one  $\beta$ -emitting radionuclide is measured by an end-window GM counter through a thin absorber in a fixed geometry. The nuclide has  $n$  different  $\beta$ -transitions with energies,  $E_{\beta i}$  and intensities,  $I_{\beta i}$  ( $i= 1,2,\dots,n$  ;  $\sum_{i=1}^n I_{\beta i} \leq 1$ ), emits conversion electrons with  $p$  different energies,

$E_{c_j}$  and intensities,  $I_{c_j}$  ( $j=1,2,\dots,p$ ) and  $\gamma$ -quanta and X-rays with  $q$  different energies,  $E_{\gamma_k}$  and intensities,  $I_{\gamma_k}$  ( $k= 1,2,\dots,q$ ). According to Papp (Papp, 1997), the counting rate, CR, is related to the activity  $A$ , by

$$CR = Af_m f_c f_g \left[ f_{se} \left( \sum_{i=1}^n I_{\beta_i} f_{W_i} f_{S_i} f_{A_i} f_{B_i} f_{H_i} + \sum_{j=1}^p I_{c_j} f_{C_{c_j}} f_{ac_j} \right) + \sum_{k=1}^q I_{\gamma_k} f_{c_{\gamma_k}} f_{s_{\gamma_k}} f_{a_{\gamma_k}} \right] \quad (3.12)$$

where

$f_m$  is for multiple counting

$f_c$  is for loss due to random coincidence

$f_g$  is for the solid angle geometry

$f_{se}$  is for the intrinsic sensitivity of the counter for electrons

$f_{W_i}$  are for the absorption of  $\beta$ -particles by the window and further absorbers

$f_{S_i}$  are for the effect of the sample in scattering and absorption of  $\beta$ -particles

$f_{A_i}$  are for air scattering

$f_{B_i}$  are for backscattering

$f_{H_i}$  are for the effect of the housing in scattering  $\beta$ -particles into the counter

$f_{C_{c_j}}$  are for the true coincidences of conversion electrons with  $\beta$ -particles

$f_{ac_j}$  are for the absorption of conversion electrons

$f_{c_{\gamma_k}}$  are for the true coincidences of  $\gamma$ -quanta and X-rays with  $\beta$ -particles and conversion electrons

$f_{s_{\gamma_k}}$  are for the intrinsic sensitivity of the detector for  $\gamma$ -quanta and X-rays

$f_{a_{\gamma_k}}$  are for the absorption of  $\gamma$ -quanta and X-rays

$A$  can be determined from CR if the correction factors in (3.10) are measured, computed, estimated or eliminated in some way. Errors are introduced in this way into the final activity result. So it is recommended to choose the detector, the counting geometry and the structural materials like that the values of the factors be as near unity and their errors be as low as possible. The counting efficiency  $\epsilon$  can be computed as  $\epsilon = CR/A$ .

In the discussion to determine the factors one by one Papp (Papp, 1997), tried to attain the highest accuracy obtainable with reasonable effort. The less important factors and the errors of the evaluations have not been examined intensively.

$f_{se}$  and  $f_m$  are assumed to be close to unity for good GM tubes if the high voltage is chosen at the beginning of the plateau.

$f_A$  and  $f_H$  are close to unity if small sample-detector distance and spacious housing covered from inside by low atomic number material are used.

$f_{Ccj}$  is equal to the probability of not detecting  $\beta$ -particle emitted simultaneously with the conversion electron. It can be estimated by the formula

$$f_{Ccj} = 1 - f_g(I_{\beta i} f_{Wi} f_{Si} f_{Bi}) \quad (3.13)$$

$f_{acj}$  can be estimated using experimental data on the transmission of monoenergetic electron beams.

$f_{s\gamma}$  can be measured by using  $^{54}\text{Mn}$  and  $^{57}\text{Co}$  reference point sources and by analyzing the  $\gamma$ -background in absorption curves measured by  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  point sources.

$f_{c\gamma k}$  can be estimated as

$$f_{c\gamma k} = 1 - f_g(I_{\beta i} f_{Wi} f_{Si} f_{Bi} + I_{cj} f_{Ccj} f_{acj}) \quad (3.14)$$

where  $i$  and  $j$  are the indexes of the corresponding  $\beta$  transition and conversion electron.

$f_{a\gamma}$  differs from unity only for X-rays. It can be estimated as

$$f_{a\gamma} = \exp(-\mu_\gamma W)$$

where  $W$  is the total thickness of the absorbers (detector window, air, added absorber) and  $\mu_\gamma$  is the mass absorption coefficient which can be evaluated from literatures.

$f_c$  becomes an important factor only in case of high values of CR (above 100-200  $\text{sec}^{-1}$ ). It can be computed for GM counters as  $f_c = 1 - CR\tau$ , where  $\tau$  is the resolving time of the counter

$f_g$  can be determined from the solid angle subtended by a circular aperture of radius  $R$  at a circular source of radius  $r$ , coaxial with the aperture, the distance

of which from the aperture is equal to  $Z$ , in case of  $r < R$  which is

$$G = \frac{1}{2} \left[ 1 - \frac{Z}{D} \right] - \frac{3r^2 R^2 Z}{16D^5} + \frac{5r^4 R^2 Z}{32D^9} \left[ Z^2 - \frac{3}{4} R^2 \right] - \frac{35r^6 R^2 Z}{256D^{13} \left[ Z^4 - \frac{5}{2} R^2 Z^2 + \frac{5}{8} R^4 \right]} \quad (3.15)$$

where  $D = (Z^2 + R^2)^{1/2}$  if  $R$  and  $Z$  are equal to the radius of the sensitive volume of the detector and the distance from the sample to the bottom surface of the sensitive volume, respectively. It can be assumed that the sensitive volume is cylindrical and its diameter is equal to that of the window.

$f_W$  can be evaluated by the expression

$$f_W = \exp(-\mu_a W)$$

where  $\mu_a$  is the mass-absorption coefficient and  $W$  is the summed thickness of the absorbers (window, absorber sheet and air)  $\mu_a$  in function of  $E_\beta$  can be determined experimentally from the results of absorption measurements.

$f_s$  is the most difficult factor that has complex character. It is reasonable to treat the different effects contributing in  $f_s$  separately. They are: self-absorption ( $f_{sa}$ ), self-scattering ( $f_{ss}$ ) and internal backscattering ( $f_{ib}$ ). Thus  $f_s$  is written as the product of three factors, which are for the above effects:

$$f_s = f_{sa} f_{ss} f_{ib}$$

$f_{ss}$  is close to unity if

1. near  $2\pi$ -type geometry is used;
2. sample is made from low  $Z$  material;
3. most of the activity is on or close to the upper surface of the sample

$f_{sa} = [1 - \exp(-\mu_s d)] / (\mu_s d)$ , where  $\mu_s$  is the self absorption coefficient and  $d$  is the thickness of the sample.

$f_{ib}$  is generally a phenomenon of second order but it becomes more significant in case of strongly decreasing exponential activity distribution.

using this method efficiencies 0.14313, 0.123516, 0.11697, 0.0774 and 0.1319 were obtained for  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ,  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}$ ,  $^{208}\text{Tl}$  respectively.

# Chapter 4

## Data Analysis and Discussion

### 4.1 Results

Indoor measurements were taken in the nuclear physics laboratory located on the first floor of physics building and Outdoor measurements were taken from the same elevation by pulling the sampling head out of the window 1 m away from the building and in the upright position. Air was sampled during the morning hour of the day (between 6:00 AM and 6:30 AM)

A total of 38 measurements were made and each was recorded for more than 8 hrs in successively increasing intervals of time. A typical measurement procedure is indicated in table 4.1. The first and the second columns in table 4.1 were recorded from the Geiger counter apparatus. The third term indicates the serial of the intervals of measurement and the fourth and the fifth columns indicate the beginning and end of diurnal time for the corresponding interval. The sixth column indicates time as measured from the end of sampling as origin. i.e. the time in the six column thus indicate the age of the filtered air sample as measured from the end of sampling. The seventh column shows the length of measurement of each time interval whereas the eighth column is the gross count in the interval.

The age of the filter, the length of the measuring time interval and the gross count obtained were fed in a computer programme, that computes the concentration of progeny and the equilibrium equivalent concentration for the two isotopes, from the observed decline in the observed count rates.



Table 4.1: Continued

7:23:00	52184	61	7:23:00	7:24:00	61	1	548	548
7:24:00	52732	62	7:24:00	7:26:00	62	2	1047	524
7:26:00	53779	63	7:26:00	7:28:00	64	2	1064	532
7:28:00	54843	64	7:28:00	7:30:00	66	2	1051	526
7:30:00	55894	65	7:30:00	7:32:00	68	2	1042	521
7:32:00	56936	66	7:32:00	7:34:00	70	2	1017	509
7:34:00	57953	67	7:34:00	7:36:00	72	2	969	485
7:36:00	58922	68	7:36:00	7:38:00	74	2	934	467
7:38:00	59856	69	7:38:00	7:40:00	76	2	911	456
7:40:00	60767	70	7:40:00	7:42:00	78	2	890	445
7:42:00	61657	71	7:42:00	7:44:00	80	2	859	430
7:44:00	62516	72	7:44:00	7:46:00	82	2	883	442
7:46:00	63399	73	7:46:00	7:48:00	84	2	817	409
7:48:00	64216	74	7:48:00	7:50:00	86	2	836	418
7:50:00	65052	75	7:50:00	7:52:00	88	2	783	392
7:52:00	65835	76	7:52:00	7:54:00	90	2	774	387
7:54:00	66609	77	7:54:00	7:56:00	92	2	729	365
7:56:00	67338	78	7:56:00	7:58:00	94	2	772	386
7:58:00	68110	79	7:58:00	8:00:00	96	2	770	385
8:00:00	68880	80	8:00:00	8:02:00	98	2	720	360
8:02:00	69600	81	8:02:00	8:04:00	100	2	736	368
8:04:00	70336	82	8:04:00	8:06:00	102	2	693	347
8:06:00	71029	83	8:06:00	8:08:00	104	2	688	344
8:08:00	71717	84	8:08:00	8:10:00	106	2	672	336
8:10:00	72389	85	8:10:00	8:12:00	108	2	626	313
8:12:00	73015	86	8:12:00	8:14:00	110	2	615	308
8:14:00	73630	87	8:14:00	8:16:00	112	2	616	308
8:16:00	74246	88	8:16:00	8:18:00	114	2	584	292
8:18:00	74830	89	8:18:00	8:20:00	116	2	625	313
8:20:00	75455	90	8:20:00	8:22:00	118	2	614	307
8:22:00	76069	91	8:22:00	8:24:00	120	2	549	275
8:24:00	76618	92	8:24:00	8:28:00	122	4	1114	279
8:28:00	77732	93	8:28:00	8:32:00	126	4	1087	272
8:32:00	78819	94	8:32:00	8:36:00	130	4	1078	270
8:36:00	79897	95	8:36:00	8:40:00	134	4	1062	266
8:40:00	80959	96	8:40:00	8:44:00	138	4	972	243
8:44:00	81931	97	8:44:00	8:48:00	142	4	977	244
8:48:00	82908	98	8:48:00	8:52:00	146	4	957	239
8:52:00	83865	99	8:52:00	8:56:00	150	4	1134	284
8:56:00	84999	100	8:56:00	9:00:00	154	4	698	175
9:00:00	85697	101	9:00:00	9:04:00	158	4	919	230
9:04:00	86616	102	9:04:00	9:08:00	162	4	842	211
9:08:00	87458	103	9:08:00	9:12:00	166	4	875	219
9:12:00	88333	104	9:12:00	9:16:00	170	4	837	209
9:16:00	89170	105	9:16:00	9:20:00	174	4	853	213
9:20:00	90023	106	9:20:00	9:24:00	178	4	814	204
9:24:00	90837	107	9:24:00	9:32:00	182	8	1594	199
9:32:00	92431	108	9:32:00	9:40:00	190	8	1548	194
9:40:00	93979	109	9:40:00	9:48:00	198	8	1560	195
9:48:00	95539	110	9:48:00	9:56:04	206	8.067	1470	182
9:56:04	97009	111	9:56:00	10:04:10	214	8.167	1522	186
10:04:10	98531	112	10:04:00	10:12:00	222	8	1434	179
10:12:00	99965	113	10:12:00	10:20:00	230	8	1502	188
10:20:00	101467	114	10:20:00	10:28:00	238	8	1422	178
10:28:00	102889	115	10:28:00	10:40:20	246	12.33	2178	177
10:40:20	105067	116	10:40:00	10:52:20	258	12.33	2053	166
10:52:20	107120	117	10:52:00	11:04:00	270	12	1985	165
11:04:00	109105	118	11:04:00	11:16:00	282	12	1939	162
11:16:00	111044	119	11:16:00	11:28:00	294	12	1937	161
11:28:00	112981	120	11:28:00	11:44:00	306	16	2524	158
11:44:00	115505	121	11:44:00	12:00:00	322	16	2546	159
12:00:00	118051	122	12:00:00	12:30:00	338	30	4799	160
12:30:00	122850	123	12:30:00	13:11:20	368	41.33	6352	154
13:11:20	129202	124	13:11:20	13:50:20	409.33	39	5931	152
13:50:20	135133	125	13:50:20	14:57:00	448.33	66.67	9508	143
14:57:00	144641	126	16:25:20	17:10:20	603.33	45	5784	129
16:25:20	0	127	18:16:01	19:15:00	714.02	58.98	7025	119
17:10:20	5784	128	21:39:10	22:09:02	917.17	29.87	3092	104
18:16:01	3950	129	22:09:02	22:43:00	947.03	33.97	3501	103
19:15:00	10975							

## 4.2 Analysis

In the computation the following assumptions were fed in the program

- The assumption of the existence of longlived  $\beta$ - emitter in the sampled air has resulted in mathematically correct but physically meaningless concentrations that fit to the observed gross count. Thus the concentration of longlived  $\beta$ - emitter was assumed to be zero in all the calculations.
- The daughter products of  $^{220}\text{Rn}$  namely  $^{212}\text{Bi}$  ( $t_{\frac{1}{2}} = 61 \text{ min}$ ) and  $^{208}\text{Tl}$  ( $t_{\frac{1}{2}} = 3 \text{ min}$ ) are linked together when the graph is plotted. This stems from the fact that the activities of these two isotopes are very small as compared to that of  $^{222}\text{Rn}$  daughters.
- When  $^{208}\text{Tl}$  and  $^{212}\text{Bi}$  are in a very small concentration, the best fit result is obtained by assuming their concentration to be zero. This is justified by the fact that the EEC of  $^{220}\text{Rn}$  is mainly contributed (>91%) by  $^{212}\text{Pb}$ . See equation (3.11)

From the plot using the program provided by Papp the following results were observed

- Activities of isotopes in a unit volume of sampled air
- Standard deviation which can be regarded as random error
- The beta count rate measured over the time interval specified for all the counting intervals as a function of time
- Contribution of the individual decay products for all the counting intervals, as a function of time.
- Sum of the individual contributions for some counting intervals
- The Equilibrium Equivalent Concentration (EEC) of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$

Based on the data of table 4.1 the results observed is shown in figure 4.1.

Using the same procedure the computation was done for the rest of the gross count measurements. One instance of the computed activities of Radon daughters

and the equilibrium equivalent concentration of the two radon isotopes is shown in table 4.2.

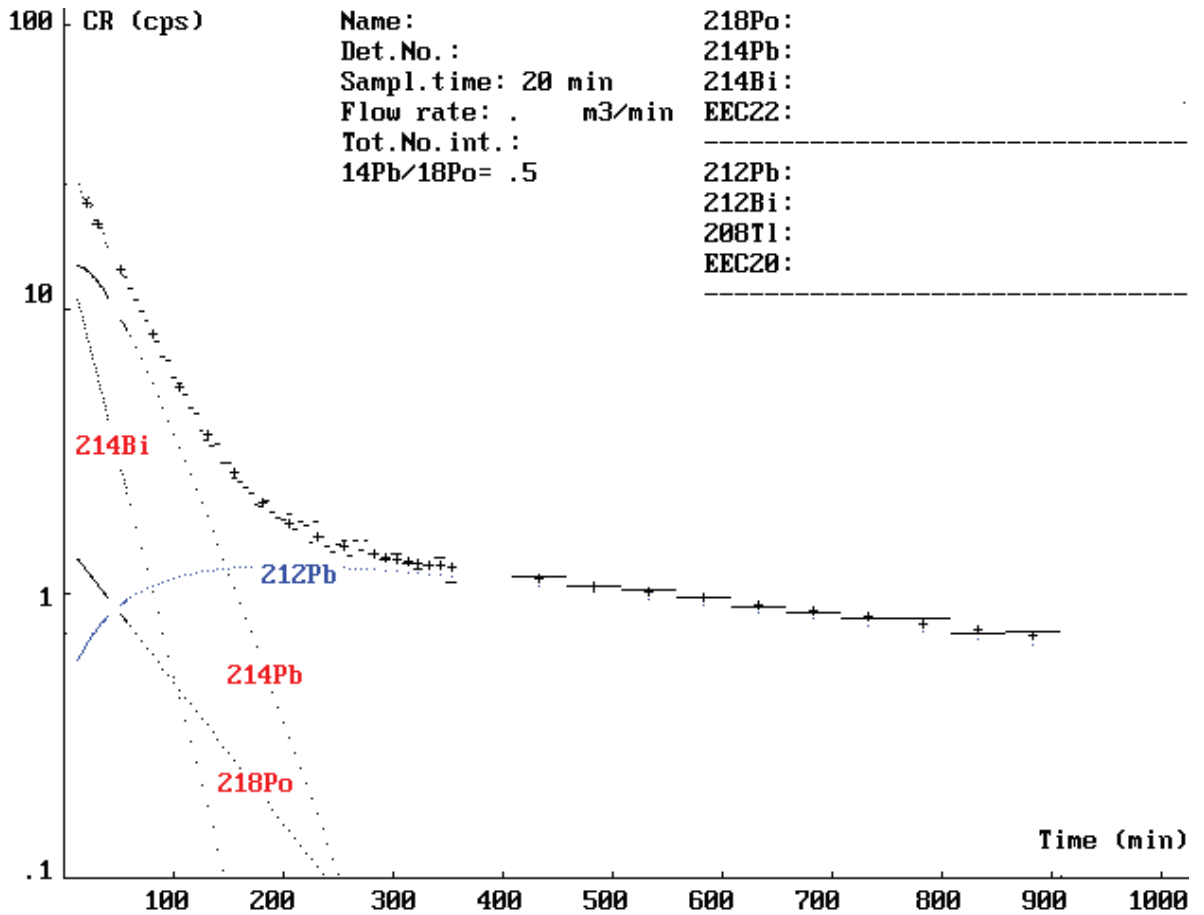


Figure 4.1: Concentration of daughters determined from gross  $\beta$  count

The naming of the data files and hence the experiment consists of the first three letters of the month (some times four) of the date of measurement and a suffix ID if the sampled air is indoor and a suffix OD if the measurement is made outdoor.

### 4.3 Comparison With Other Areas of the World

The indoor radon concentration determined in our lab is  $23.9 \text{ Bq/m}^3$ . It is about 38.5% less than the global average  $39 \text{ Bq/m}^3$  reported by WHO (WHO, 2005)

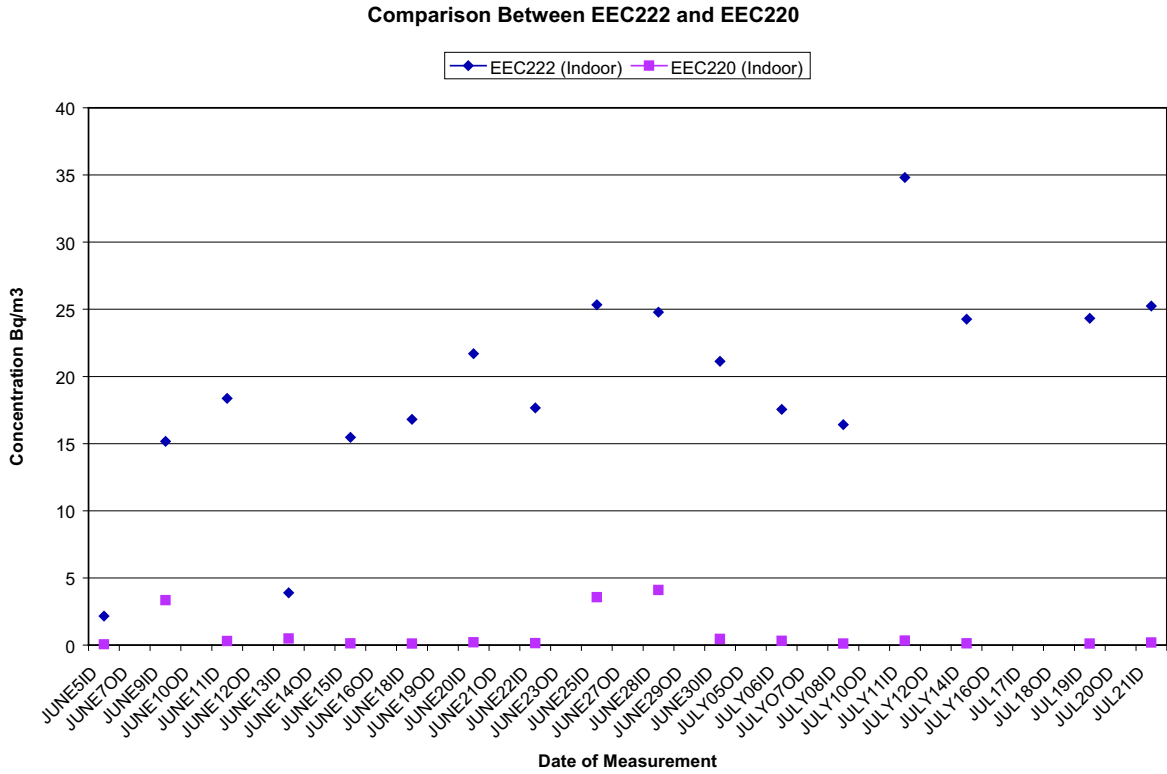


Figure 4.2: Comparison of <sup>222</sup>Rn and <sup>220</sup>Rn Equilibrium Equivalent Concentrations: Indoor

### 4.4 Diurnal variation of EEC of <sup>222</sup>Rn and <sup>220</sup>Rn

The diurnal variation of EEC <sup>222</sup>Rn and <sup>220</sup>Rn is due to atmospheric mixing phenomena. During the day time solar heating tends to induce some turbulence. This results easy transportation of radon upwards. At night and in the morning hours, the atmosphere mixup will be minimum which will trap radon to the ground. This phenomena results in that the EEC concentration will be maximum in the morning and minimum during day time.

From the datas collected in this experiment, the EEC variation of <sup>222</sup>Rn and <sup>220</sup>Rn was analyzed. The results obtained are shown in table ?? and the corresponding plot is shown in fig. ?? and fig. ??.

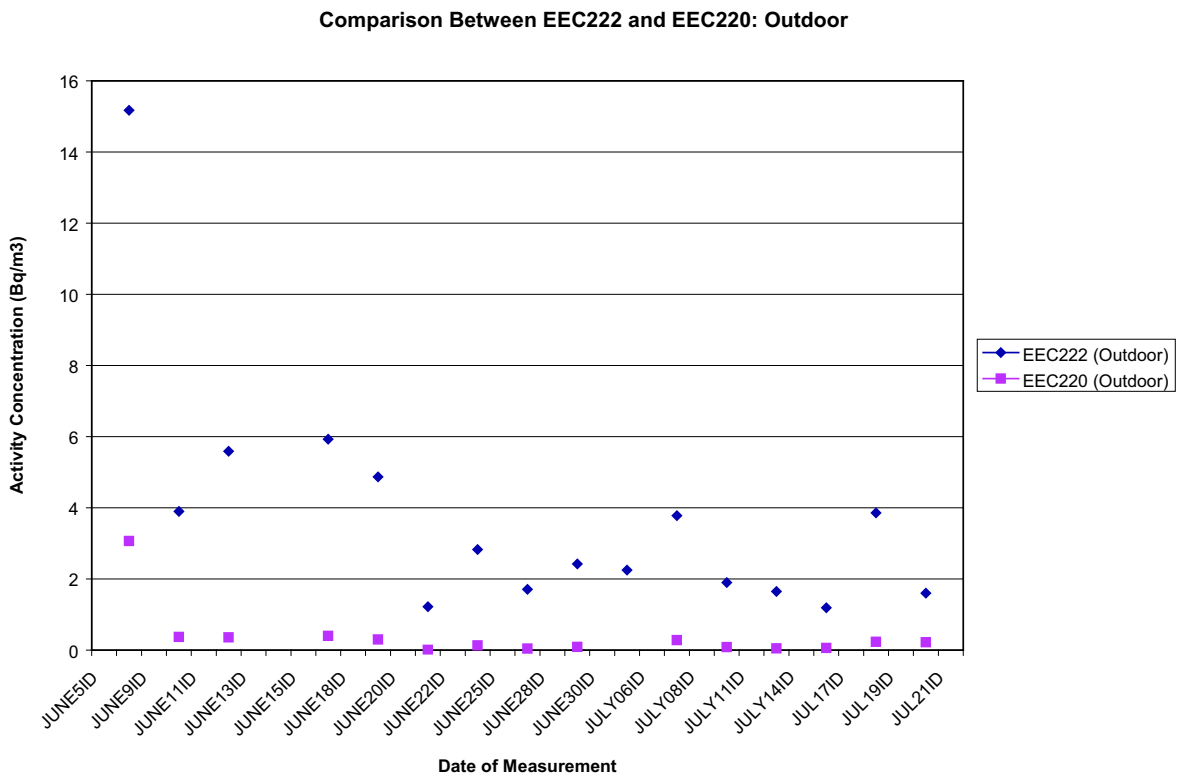


Figure 4.3: Comparison of <sup>222</sup>Rn and <sup>220</sup>Rn Equilibrium Equivalent Concentrations: Outdoor

Table 4.2: Computed activity concentration of Radon Progeny and Equilibrium Activity Concentrations

Label	Radon Daughters and EEC							
	INDOOR MEASUREMENTS				OUTDOOR MEASUREMENTS			
	Po-218	Pb-214	Bi-214	Rn-EEC	Po-218	Pb-214	Bi-214	Rn-EEC
JUNE5ID	3.09 ± 0.62	2.32 ± 0.46	1.69 ± 0.39	2.16 ± 0.29				
JUNE7OD					30.3 ± 3.46	15.15 ± 1.73	11.01 ± 4.69	15.17 ± 2.02
JUNE9ID	30.3 ± 3.46	15.15 ± 1.73	11.01 ± 4.69	15.17 ± 2.02				
JUNE10OD					8.41 ± 0.51	4.21 ± 0.26	2.24 ± 0.66	3.9 ± 0.29
JUNE11ID	37.7 ± 4.12	18.85 ± 2.06	12.36 ± 1.62	18.37 ± 1.3				
JUNE12OD					14.35 ± 0.79	7.17 ± 0.4	1.01 ± 1.03	5.59 ± 0.45
JUNE13ID	9.93 ± 0.5	4.97 ± 0.25	0.78 ± 0.64	3.9 ± 0.28				
JUNE14OD								
JUNE15ID	30.16 ± 2.24	15.08 ± 1.12	11.93 ± 1.1	15.47 ± 0.78				
JUNE16OD					11.37 ± 0.4	5.68 ± 0.2	4.76 ± 0.53	5.93 ± 0.23
JUNE18ID	29.52 ± 2	14.76 ± 1	16.08 ± 0.98	16.81 ± 0.67				
JUNE19OD					9.32 ± 0.5	4.66 ± 0.25	3.91 ± 0.66	4.87 ± 0.29
JUNE20ID	39.56 ± 3.67	19.78 ± 1.83	19.35 ± 1.83	21.69 ± 1.24				
JUNE21OD					2.04 ± 0.21	1.02 ± 0.1	1.25 ± 0.26	1.22 ± 0.11
JUNE22ID	35.13 ± 2.62	17.56 ± 1.31	12.99 ± 1.28	17.66 ± 0.88				
JUNE23OD					5.31 ± 0.38	2.66 ± 0.19	2.38 ± 0.49	2.83 ± 0.22
JUNE25ID	150.46 ± 15.28		25.18 ± 4.24	25.34 ± 2.27				
JUNE27OD					3.58 ± 0.31	1.79 ± 0.16	1.1 ± 0.4	1.71 ± 0.17
JUNE28ID	46.2 ± 2.45	27.72 ± 1.47	14.87 ± 3.88	24.79 ± 1.67				
JUNE29OD					4.56 ± 0.31	3.65 ± 0.25	0.15 ± 0.6	2.42 ± 0.26
JUNE30ID	37.16 ± 8.91	18.58 ± 4.45	20.17 ± 4.66	21.13 ± 3.05				
JULY05OD					5.06 ± 0.41	2.53 ± 0.2	1.11 ± 0.56	2.25 ± 0.24
JULY06ID	33.74 ± 5.6	16.87 ± 2.8	13.98 ± 2.77	17.55 ± 1.88				
JULY07OD					6.61 ± 0.91	3.31 ± 0.45	3.63 ± 1.23	3.78 ± 0.53
JULY08ID	28.54 ± 2.16	14.27 ± 1.08	15.99 ± 1.07	16.42 ± 0.73				
JULY10OD					3.14 ± 0.24	1.71 ± 0.12	1.73 ± 0.31	1.9 ± 0.14
JULY11ID	70.5 ± 6.23	35.25 ± 3.12	24.33 ± 3.18	34.82 ± 2.11				
JULY12OD					3.89 ± 0.28	1.94 ± 0.14	0.64 ± 0.35	1.65 ± 0.15
JULY14ID	47.47 ± 2.79	23.73 ± 1.4	18.58 ± 1.62	24.27 ± 0.99				
JULY16OD					2.33 ± 0.21	1.16 ± 0.11	0.9 ± 0.28	1.19 ± 0.12
JUL17ID								
JUL18OD					7.92 ± 0.34	3.96 ± 0.17	2.59 ± 0.45	3.86 ± 0.19
JUL19ID	46.16 ± 2.66	23.08 ± 1.33	19.98 ± 1.67	24.33 ± 0.97				
JUL20OD					3.2 ± 0.25	1.6 ± 1.3	1.16 ± 0.33	1.6 ± 0.14
JUL21ID	47.89 ± 5.05	23.95 ± 2.53	20.74 ± 3.43	25.24 ± 1.92				
Count	17	16	17	17	16	16	16	16
Minimum	3.09	2.32	0.78	2.16	2.04	1.02	0.15	1.19
Maximum	150.46	35.25	25.18	34.82	30.3	15.15	11.01	15.17
Average	42.559	18.245	15.295	19.125	7.5869	3.8875	2.4731	3.7419
Standard Dev	31.613	7.9478	6.7284	7.8465	6.9693	3.4555	2.6138	3.4086
95% Confidence	15.027	3.8944	3.1984	3.7299	3.4149	1.6932	1.2807	1.6702

Table 4.3: Computed activity concentration of Thoron Progeny and Equilibrium Activity Concentrations

Label	Thoron Daughters and EEC							
	INDOOR MEASUREMENTS				OUTDOOR MEASUREMENTS			
	Pb-212	Bi-212	Tl-208	Tn-EEC	Pb-212	Bi-212	Tl-208	Tn-EEC
JUNE5ID	0.183 ± 0.033	1.149 ± 0.483	0.413 ± 0.174	0.267 ± 0.052				
JUNE7OD					3.368 ± 0.187			3.065
JUNE9ID	3.68 ± 0.187			3.35 ± 0.17				
JUNE10OD					0.406 ± 0.032			0.369
JUNE11ID	3.311 ± 0.225	1.629 ± 2.526	0.585 ± 0.907	3.164 ± 0.301				
JUNE12OD					0.39 ± 0.045			0.355
JUNE13ID	0.543 ± 0.031			0.49 ± 0.03				
JUNE14OD								
JUNE15ID	3.135 ± 0.077	3.748 ± 1.189	1.346 ± 0.427	3.188 ± 0.125				
JUNE16OD					0.438 ± 0.023			0.399
JUNE18ID	3.745 ± 0.069	6.532 ± 1.06	2.346 ± 0.0381	3.987 ± 0.112				
JUNE19OD					0.326 ± 0.03			0.297
JUNE20ID	4.208 ± 0.122	7.129 ± 1.912	2.561 ± 0.687	4.462 ± 0.2				
JUNE21OD					0.008 ± 0.015			0.007
JUNE22ID	3.21 ± 0.189	5.07 ± 1.379	1.823 ± 0.95	3.84 ± 0.14				
JUNE23OD					0.147 ± 0.025			0.134
JUNE25ID	3.912 ± 0.168			3.56 ± 0.15				
JUNE27OD					0.046 ± 0.021			0.042
JUNE28ID	4.512 ± 0.147			4.11 ± 0.13				
JUNE29OD					0.095 ± 0.02			0.086
JUNE30ID	4.072 ± 0.27	8.352 ± 4.504	3 ± 1.618	4.444 ± 0.463				
JULY05OD								
JULY06ID	3.482 ± 0.192	6.38 ± 2.951	2.292 ± 1.06	3.734 ± 0.31				
JULY07OD					0.305 ± 0.037			0.278
JULY08ID	2.96 ± 0.071	6.049 ± 1.129	2.173 ± 0.406	3.229 ± 0.117				
JULY10OD					0.091 ± 0.016			0.083
JULY11ID	5.682 ± 0.199	6.711 ± 3.198	2.411 ± 1.149	5.772 ± 0.332				
JULY12OD					0.052 ± 0.019			0.047
JULY14ID	4.521 ± 0.069	4.382 ± 1.325	1.574 ± 0.476	4.509 ± 0.131				
JULY16OD					0.063 ± 0.01			0.057
JUL17ID								
JUL18OD					0.252 ± 0.02			0.229
JUL19ID	4.476 ± 0.05	3.761 ± 1.184	1.351 ± 0.425	4.413 ± 0.113				
JUL20OD					0.239 ± 0.013			0.217
JUL21ID	4.61 ± 0.085	4.04 ± 1.998	1.451 ± 0.718	4.56 ± 0.19				
Count	17	12	12	17	15			15
Minimum	0.183	1.15	0.41	0.27	0.01			0.01
Maximum	5.682	8.35	3.00	5.77	3.37			3.07
Average	3.5436	5.28	1.89	3.58	0.42			0.38
Standard Dev	1.38	0.83					0.76	
95% Confidence	0.65	0.42					0.38	

Table 4.4: Comparison of Indoor Radon Concentrations

<b>Region</b>	Country	Population In 1996 ( $10^6$ )	Mean Radon concentration ( $\text{Bqm}^{-3}$ )
<b>Africa</b>	Algeria	28.78	30
	Egypt	63.27	9
	Ghana	17.83	
<b>America</b>	Canada	29.68	34
	United States	269.4	46
	Argentina	35.22	37
	Chile	14.42	25
	Paraguay	4.96	28
<b>Asia</b>	China	1232	24
	Honk Kong	6.19	41
	India	944.6	57
	Indonesia	200.45	12
	Japan	125.4	16
	Iran	69.98	82
	Kuwait	1.69	14
Syria	14.57	44	
<b>Europe</b>	Denmark	5.24	53
	Estonia	1.47	120
	Finland	5.13	120
	Belgium	10.16	48
	France	58.33	62
	Germany	81.92	50 9
	Ireland	3.55	
	Czech Republic	10.25	140
	Hungary	10.05	107
	Albania	3.4	120
	Croatia	4.5	35
	Slovenia	1.92	87
	Spain	39.67	86

# Chapter 5

## Conclusion

In this work, the method of defined solid angle absolute beta counting, using an end-window Geiger-Müller counter, was tried and tested for the first time in the nuclear science laboratory of the physics department. In view of the portability and low cost aspect of the method it is promising for future wide area survey of radon.

Using the above method concentration of radon progeny and EEC of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  was determined. The diurnal variation of the EEC concentration of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  was studied and the results obtained are in agreement with earlier works in other parts of the world.

In the future, this method may be used to study various aspects of radon and thoron concentrations in air. Some potential areas of research include:

- Radon concentration in various parts of the country (emphasis on mining areas);
- Seasonal variation of radon and radon progeny concentration at locations of interest;
- Radon and progeny measurement in all parts of Ethiopia so as to establish "Radon map of Ethiopia";
- Earth quake prediction in places which are exposed to it as sudden change in radon concentration can be related to turbulence inside the earth's crust.

## **Improvements Proposed on the Method**

The method of data acquisition may be improved by automation. i.e. the count of the GM tube may be directly fed, with a suitably designed interface, to a computer in a progressively increasing interval of time rather than manual counting. This simplifies and slashes the time required to feed the gross count of hundreds of intervals manually and thereby reduce the labor required to analyze the data.

Improving the shielding of the detection system may also help to minimize the variation of the background radiation.

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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 material used for the thesis have been duly acknowledged.

Name: \_\_\_\_\_

Signature: \_\_\_\_\_

This Thesis has been submitted for examination with my approval as university advisor.

Name: \_\_\_\_\_

Signature: \_\_\_\_\_