internal report **279**

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^{222}Rn and ^{220}Rn ACTIVITY FLUX FROM THE GROUND IN THE VICINITY OF RANGER URANIUM MINE

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SUMMARY

This report is prepared for submission to the Environmental Research Institute of the Supervising Scientist (ERISS) for part fulfillment of the requirement under a QUT-ERISS consultancy agreement.

The report contains up to date details of the work and measurements carried out to study the radon activity flux as a collaborative effort by QUT and ERISS staff. The main body of the text deals with the radon activity flux survey; chapter five describes an initial set up of a radon emanation measurement column and its application to study thoron emanation from a monazite sample; appendix one describes the field sites, appendix two is a site by site description of all measurements, appendix three includes the results of an intercalibration exercise of the equipment.

Extensive research into the production and transport of ^{222}Rn in the environment as been performed, however it is still not possible to determine what set of conditions will lead to elevated levels of ^{222}Rn. Very limited data is available for ^{220}Rn. The majority of studies have been performed in temperate climates with little data obtained in tropical regions. There are large variations in flux between regions. The relative importance of the various parameters on flux also varies between regions. Therefore it is difficult to apply results from previous studies to an unknown region.

A flow through accumulator technique was used to measure ^{222}Rn and ^{220}Rn flux in field and laboratory experiments. An activity flux survey was performed over an area of approximately 10 kilometres square, in the Jabiru region of the Northern Territory. The aim was to determine the association between radon flux and various soil characteristics and meteorological parameters in this tropical region. The average ^{222}Rn and ^{220}Rn flux in the region were

determined to be 64 ± 25 $mBq.m^{-2}.s^{-1}$ and 2.15 ± 0.21 $Bq.m^{-2}.s^{-1}$ respectively. The strongest correlation was found between radon activity flux and radium activity concentration in the soil. Weak non-linear relationships were observed between ^{222}Rn and ^{220}Rn flux and the ratio of the >2mm fraction to the <2mm fraction of a core soil sample (0-20cm deep) and with soil moisture. Strong correlation was found between gamma dose rate at 1m above ground and ^{220}Rn flux but not ^{222}Rn flux and no relationship was observed between ^{222}Rn and ^{220}Rn. Of the meteorological parameters observed weak correlation was observed between ^{222}Rn flux and air temperature. No significant effect was observed due to wind speed, barometric pressure, soil temperature or air soil temperature difference, and no significant correlation was found between ^{222}Rn flux and any of the meteorological parameters.

A study of the diurnal variations in flux was also performed. Significant diurnal variations were observed in both ^{222}Rn and ^{220}Rn flux. ^{222}Rn flux was lower through the middle of the day while ^{220}Rn flux was elevated at this time. No correlation was found between ^{222}Rn and ^{220}Rn flux, suggesting that different factors are dominating variations in these parameters.

A laboratory study was undertaken, into the effect of sample thickness and moisture on ^{220}Rn flux from a monazite sample. Flux increased rapidly with thickness to approximately 5cm after which no increase was observed. Soil Moisture had a large effect on flux, causing an increase in flux for water content less than approximately 6% by weight. This was followed by a dramatic decrease with little flux observed for water content over 10%.

A large part of this report constitutes the thesis submitted by Rebecca Todd for partial fulfillment of the requirements of the degree of Master of Applied Science (Medical Physics) at the Queensland University of Technology.

ACKNOWLEDGMENT

Financial and equipment support provided by ERISS is acknowledged. The equipment calibration excercise was conducted through an AINSE grant # 96/154. Bruce Ryan and Therese Fox at ERISS, Jaya Dharmasiri and Nick Giannakis at QUT provide significant technical and experimental support.

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CHAPTER 1. GENERAL INTRODUCTION

1.1 RADON IN THE ENVIRONMENT

Radon is a radioactive noble gas. There are three naturally occurring isotopes; ^{219}Rn, ^{220}Rn, and ^{222}Rn which are part of the actinium (^{235}U), thorium (^{232}Th), and uranium (^{238}U) primordial series. ^{219}Rn (actinon) is least significant due to it's short half-life and the limited abundance of ^{235}U which constitutes only 0.7% of natural uranium. The uranium and thorium decay series are illustrated in Figure1.1. ^{238}U and ^{232}Th are equally abundant in nature however, ^{222}Rn (radon) is generally considered to be a greater health risk than ^{220}Rn (thoron) due to its longer half life. In areas of elevated ^{220}Rn the dose due to the inhalation of its daughter ^{212}Pb may be significant. Some confusion may arise over the use of radon as it may refer to the element or the specific isotope ^{222}Rn. For the purpose of this paper the term radon will only be used when discussing the element while ^{222}Rn and ^{220}Rn will be used when referring to the specific isotopes.

Of primary concern are the possible health effects due to the dose to the lungs upon inhalation of radon's daughter products. Progeny concentrations in the atmosphere are determined primarily by atmospheric radon concentrations. In turn the concentration of atmospheric radon depends on the radon activity flux and dispersion of radon in the atmosphere. Radon's progeny are all heavy metals and are chemically reactive therefore their migration from soil to the atmosphere is insignificant.

Radon exposure is the single most significant source of natural radiation. The worldwide average annual effective dose due to the inhalation of radon progeny is estimated to be 1.3 mSv. The total average annual dose from all

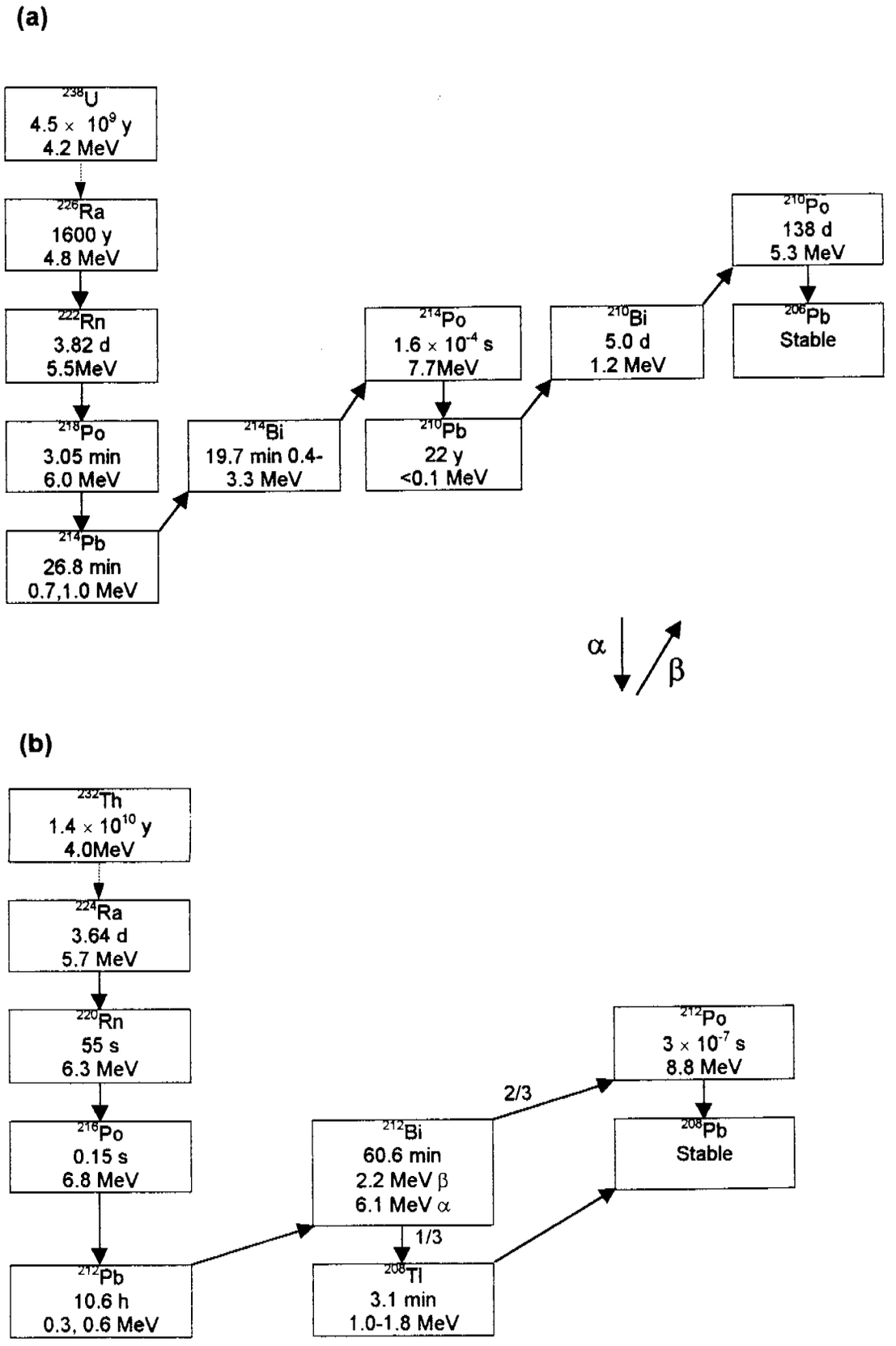


Figure 1.1 (a) Uranium and (b) Thorium Primordial Decay Series. For each isotope the half life and energy of the principal decay mode are shown.

natural sources is 2.4 mSv (UNSCEAR, 1993). Due to the large populations of China and India the results for these countries have a large influence on the world wide annual effective dose (UNSCEAR, 1993). In Australia the dose from radon progeny is one of the most important natural sources, along with terrestrial gamma radiation and cosmic radiation.

NCRP's report no.78 (1984) lists the major sources of global atmospheric radon. At least 80% of atmospheric radon originates from exhalation from soil and rock formations. The other major source is radon dissolved in groundwater, however the contribution to human exposure is generally small due to the low dose following ingestion as opposed to inhalation. Other minor sources include: emanation from oceans, phosphate residues, uranium mill tailings, coal residues, and natural gas. While, on a global scale, soil is clearly the most significant source of atmospheric radon, the relative significance of each source is site dependent.

1.2 PROPERTIES OF RADON AND IT'S PROGENY

Details of the properties of radon contained in the Handbook of Chemical Physics (1983) are summarised below. At room temperature radon is a colourless gas. As it is a noble gas it is essentially inert. It occupies the last place in the zero group of the periodic table and is the heaviest known gas. It has a boiling point of -61.8 °C, melting point of 71°C and a density in gaseous form of 9.73 g/l. It is readily soluble in water (230 $cm^3.kg^{-1}$ at 20°), and some organic compounds (eg toluene). Radon is readily absorbed onto charcoal, a property that is often used in radon concentration measurements. The principal radiation energies and intensities of the three isotopes are contained in Table 1.1. Radon progeny are all heavy metals, including a series of Bi, Po and Pb isotopes and finishing with a stable isotope of lead.

Upon the decay of atmospheric radon the progeny may, attach to aerosols or surfaces, or remain unattached.

Table 1.1 Properties of the Primordial Series Radon Isotopes. (NCRP, 1988)

Isotope	Half Life	Principal Radiation Energies and Intensities			
		Alpha		Gamma	
		MeV	%	MeV	%
^{219}Rn	3.96 s	6.819	81	0.271	10
		6.553	12		
^{220}Rn	55.60 s	6.288	100		
^{222}Rn	3.824 d	5.490	100		

1.3 RADON EMANATION AND TRANSPORT FROM SOIL GAS TO AIR

The movement of radon from the soil to the atmosphere consists of two stages, emanation and transport from the soil gas to the atmosphere. Soil can be thought of as a porous network of soil grains or particles. The isotopes preceding radon in the primordial series' are solids and migration from the soil grains to the pore space is generally negligible. Radon emanates from the soil grains into the pore space primarily by recoil upon the decay of its parent radium. This process is illustrated in Figure 1.2. The typical recoil range in minerals is of the order of 0.03 μm for ^{220}Rn atoms and 0.02-0.07 μm for ^{222}Rn (Tanner, 1980). Diffusion also occurs however the diffusion length in solids is in the range 10^{-7} to 10^{-26} μm (Nazaroff, 1992) making this mechanism of migration from soil particles negligible. The fraction of radon, which emanates into the pore space is termed the emanating fraction or emanation coefficient. Experimental results indicate emanating fractions in soil are in the range 0.05-0.7, which is considerably higher than expected from theoretical models (Nazaroff, 1992). This

discrepancy has been explained in two ways. Firstly that the radium is distributed primarily on the surface of the grain (Morowska, 1993; Nazaroff, 1992). Secondly chemical and predominantly radiation damage due to previous decays facilitate the escape of radon (Nazaroff, 1992).

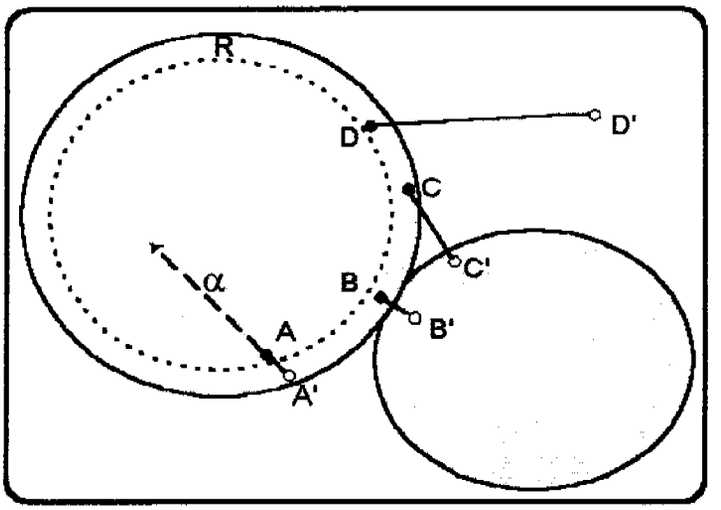


Figure 1.2 Emanation of Radon From Soil Grains
The above diagram is a schematic only and is not drawn to scale. Two soil grains surrounded by air are shown. They are depicted as circles to simplify the diagram. Radium atoms decay producing an alpha particle and a radon atom. The recoil range, R, of the radon atoms is shown. When inside this recoil range radon particles may emanate from the soil grain. The recoil range is air is considerably larger than in the soil grain.

Upon emanation the radon may : lodge in the pore space (D), be embedded in the adjacent grain leaving a track of damage (B or C), or remain in the original grain (A). Moisture has a large effect on the emanation coefficient as the small recoil range of radon in water increases the probability that the radon will lodge in the pore space.

Once in the pore space the isotopes migrate by forced advection and diffusion. Forced advection is a pressure driven flow of the soil gas, and is described by Fick's law. Diffusion, described by Darcy's law, is the flow of a component of the soil gas due to a concentration gradient. It is thought that on a long term basis diffusion is the dominant transport mechanism (Schery 1984). However this may not be true in the immediate vicinity of buildings

due to sustained pressure differences induced by the buildings geometry and operation; and the blocking of molecular diffuison by the buildings structure (Nazeroff, 1992).

There are a number of similar equations quoted which describe the total transport process. One such equation, quoted by Schery (1984) is:

$$\frac{\partial C}{\partial t} = \frac{1}{\varepsilon}\frac{\partial}{\partial z}\left(D\frac{\partial C}{\partial z}\right) - \frac{1}{\varepsilon}\frac{\partial}{\partial z}(\upsilon C) - \lambda C + \varphi$$

Where: C= interstitial radon concentration in soil (atoms.m^{-3})

J= flux density (atoms.$m^{-2}.s^{-1}$)

υ= volume interstitial fluid flowing per unit area per unit time ($m.s^{-1}$)

D= effective diffusion coefficient ($m^2.s^{-1}$)

ε= porosity (dimensionless)

λ= radon decay constant (s^{-1})

φ= source term (atoms.$m^{-3}.s^{-1}$)

The parameter, activity flux, is used to describe the rate which radon is emitted from the ground. Activity flux is the activity per unit area per unit time. The units that are generally used are mBq.$m^{-2}.s^{-1}$. Although concentrations of ^{238}U and ^{232}Th in the ground are comparable, ^{220}Rn activity flux is generally much greater than ^{222}Rn flux. The difference in flux is due to the difference in the half lives of the two isotopes. This is illustrated below.

$$\text{Activity Flux} = \text{Activity /Area /Time}$$

$$= \lambda N \text{ /Area /Time}$$

$$\lambda_{Rn\text{-}222} = 1.26 \times 10^{-4}\ s^{-1}$$

$$\lambda_{Rn\text{-}220} = 1.25 \times 10^{-2}\ s^{-1}$$

1.4 PROJECT JUSTIFICATION AND AIM

Extensive research into the production and transport of ^{222}Rn in the environment has been performed, covering the effects of radium distribution in soil grains, and the effect of meteorological parameters on flux, through to transport of radon and it's progeny in the atmosphere. However many of the studies present conflicting results and it is not yet possible to predict accurately what conditions will cause elevated ^{222}Rn concentrations. Very limited data is available for ^{220}Rn. For both isotopes the majority of studies have been performed in temperate climates. Details on radon flux from Australian soils and in particular tropical regions are sparse.

There is a large variety of factors which effect radon flux. As a result there a large variations in flux between regions. The relative importance of the various parameters is also site dependent, hence the conflicting reports from different studies. The relative importance of the various effects also depends on the size of the region and the time frame of interest. For example, barometric pressure may dominate variations in flux with time at one site, however on a regional scale it may not be significant in determining flux.

There are a number of points which are generally true. In general higher radium levels lead to higher flux, although some believe it is the emanating fraction not the radium content which is most important (Schery, 1989). Moisture content tends to increase flux to a maximum due to an increase in the emanation coefficient and then decrease with the decreasing diffusion length until no flux is observed (Schery 1984, Strong 1982, Ball 1991). Higher soil permeability and porosity increase emanation while soil density has no effect (Schery 1984 and 1989). The effect of other parameters such as; soil and air temperature, atmospheric pressure, wind, vegetation and humidity are not as clear.

Very few studies have been performed in tropical regions. It cannot be assumed that results from studies in temperate climates can be transferred to tropical climates. Australia's major uranium deposits are located in the regions of Jabiru (Northern Territory) and Roxby Downs (South Australia). It is therefore important to examine the transport of radon in these different climates.

Limited data is available for ^{220}Rn as it is generally not a domestic problem. However due to Australia's mineral sands industry there may be sites where ^{220}Rn is important.

The primary aim of this project was to examine the effects of various meteorological parameters and soil characteristics on ^{222}Rn and ^{220}Rn flux from the ground, in particular in tropical northern Australia. Analysis of the association between activity flux and: radium activity concentration, soil grain size, moisture content, atmospheric pressure, soil and air temperature and other relevant parameters was performed. This was achieved through an activity flux survey of the region surrounding Jabiru in the Northern Territory. A preliminary study into ^{220}Rn flux from a monazite sample was also performed.

1.5 PROJECT OUTLINE

This report consists of four components. Chapter 2 contains a description of the working principles behind and the specific design of the emanometer used to obtain flux measurements.

Chapter 3 contains a description of the ^{222}Rn and ^{220}Rn activity flux survey conducted in the Jabiru and Jabiru East Regions of the Northern Territory. It includes the experimental method, and the results of the survey.

An examination of the diurnal variations in ^{222}Rn and ^{220}Rn flux along with various meteorological parameters at a number sites is presented in Chapter 4.

Finally Chapter 5 details laboratory experiments performed to provide insight into ^{220}Rn exhalation.

CHAPTER 2. MEASUREMENT OF RADON FLUX

2.1 METHODS OF RADON MEASUREMENT

Due to it's unique properties there are many techniques which may be used to determine radon concentration. These techniques are all based on its radioactive properties. Ionisation chambers have low detection limits however they are not commonly used as they are costly and not practical for field work. Scintillation cells (typically zinc sulphide) may be used as closed or flow through types and provide either integrated or partially integrated results. They are commonly used as they are; rugged, versatile, sensitive, reliable, relatively low cost and simplistic (IAEA, 1992).

The adsorption of radon onto charcoal was first noticed by Rutherford in 1906 (Harley, 1992). Measurement of radon concentration with activated charcoal uses preweighed amounts of charcoal contained in canisters, which are exposed to the radon source for a few hours to days. After exposure they are sealed and later analysed by gamma spectroscopy. They are simple, flexible, rugged and low cost (IAEA,1992). There are practical problems such as desorption and decay which need to be considered when using this technique (Harley, 1992).

There are a number of less commonly used techniques. Nuclear track detectors may be used to measure radon by placing them in containers with a diffusion barrier to exclude solid decay products (Harley, 1992). They provide a passive, fully integrated result, however they are not suitable for flux measurements (IAEA, 1992). Solid state surface barrier detectors are suited to flux measurements however they are expensive and require high

levels of care and maintenance (IAEA, 1992). Thermo luminescent detectors and electrets have also been used.

There are three methods commonly used to determine ^{222}Rn and ^{220}Rn flux from the soil: accumulation, flow-through, and adsorption. The accumulation technique uses a container placed over the test surface. The radon concentration inside the container at one time or at several specific times is used to determine the rate of radon accumulation. The concentration is measured using one of the instantaneous detectors mentioned above. NCRP (1988) lists a number of requirements which must be satisfied to obtain accurate results : the accumulation time must be short compared to radon's half life, the concentration inside the container must be less than the soil gas to prevent back diffusion, the measuring device should not significantly affect the exhalation, and good seal must be sustained around the edge of the container.

The flow through method is based on the same principals as the accumulation method however air in the container is continually replaced. The radon concentration in air drawn from the container is measured using a scintillation detector or other semi-integrating device (IAEA, 1992). The concentration of radon is proportional to the flux rate and the surface area (IAEA, 1992). This system reduces the build-up of radon and therefore has less effect on the concentration gradient than the accumulator technique. The flow rate must be high enough to prevent build up but low enough to ensure that the conditions under the container resemble natural conditions (IAEA, 1992).

Radon is readily adsorbed onto certain materials, for example activated charcoal. In the adsorption method a canister of this material is exposed for a known amount of time (typically 1-3 days). They are then sealed and

analysed using gamma spectroscopy (IAEA, 1992). This technique is less sensitive than the other techniques to due the smaller sampling area. However it can be left to obtain and integrated value over a period of time. The flux is determined using the radon activity collected, surface area exposed, time of exposure and a correction for radon decay (IAEA, 1992).

NCRP's publication "Measurement of Radon and Radon Daughter in Air" (1998) describes some less commonly used techniques. The vertical profile method is based on the principal that the total amount of radon in a vertical column of fixed area represents the radon that has exhaled from the same area of soil. It is a large scale, costly technique requiring aircraft or balloons for measurements. Soil concentration gradient may be used to determine flux provided pressure conditions are stable and rain and wind do not interfere. This technique requires independent measurements of the diffusion constant in soil and soil radon concentrations.

2.2 EQUIPMENT DETAILS

Flux measurements were performed using a flow through type radon/thoron emanometer and accompanying software manufactured by ANSTO. The emanometer consists of a drum, which is placed over the surface to be measured, two pumps, two scintillation detectors and relevant supporting electronics and a lap top computer. The drum is approximately conical in shape with a volume of 0.01846 m^3 and covers an area of 0.259 m^2. To obtain accurate measurements; conditions in the drum must closely match those outside and the air in the drum must be well mixed (Schery et al, 1989). To achieve this air in the drum is continually replaced via inlet and outlet hoses at the top of the drum with a ventilation rate of approximately 1 $L.min^{-1}$. These hoses are approximately 10m long to allow the drum to be placed at a distance from the detectors. Two small pumps control the ventilation rate. A

small 12V fan (dimensions: 82×82×25 mm) located in the centre of the drums interior ensured mixing of the air.

Air drawn from the outlet passes through two zinc sulphide alpha scintillation detectors which are separated by a length of pipe. As a result a sample of air reaches the second detector six minutes after passing through the first, allowing distinction of ^{222}Rn and ^{220}Rn counts. The first detector counts both ^{222}Rn and ^{220}Rn while the second detector counts predominantly ^{222}Rn. The delay time is more than 6half lives of ^{220}Rn. Figure 2.1 describes the flow of air through the emanometer.

To determine the flux from a particular surface the drum was placed over the area to be measured with the rim placed 1cm into the soil. Error in the placement of the drum is often the most important source of error in the measurements (Whittlestone). To minimise this error a specially designed cutting device was used to dig a circular hole 1cm deep, which the drum was placed in (Figure 2.2). The edge of the drum must be adequately sealed, and extra dirt was placed around the rim when required. The operation manual

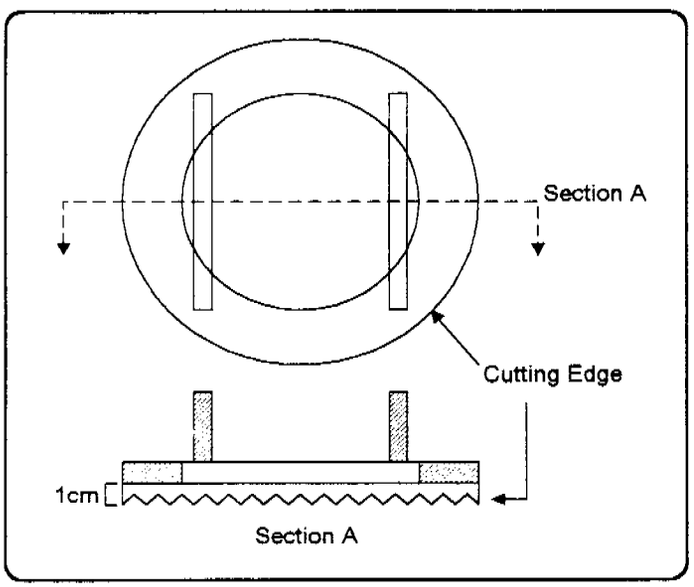
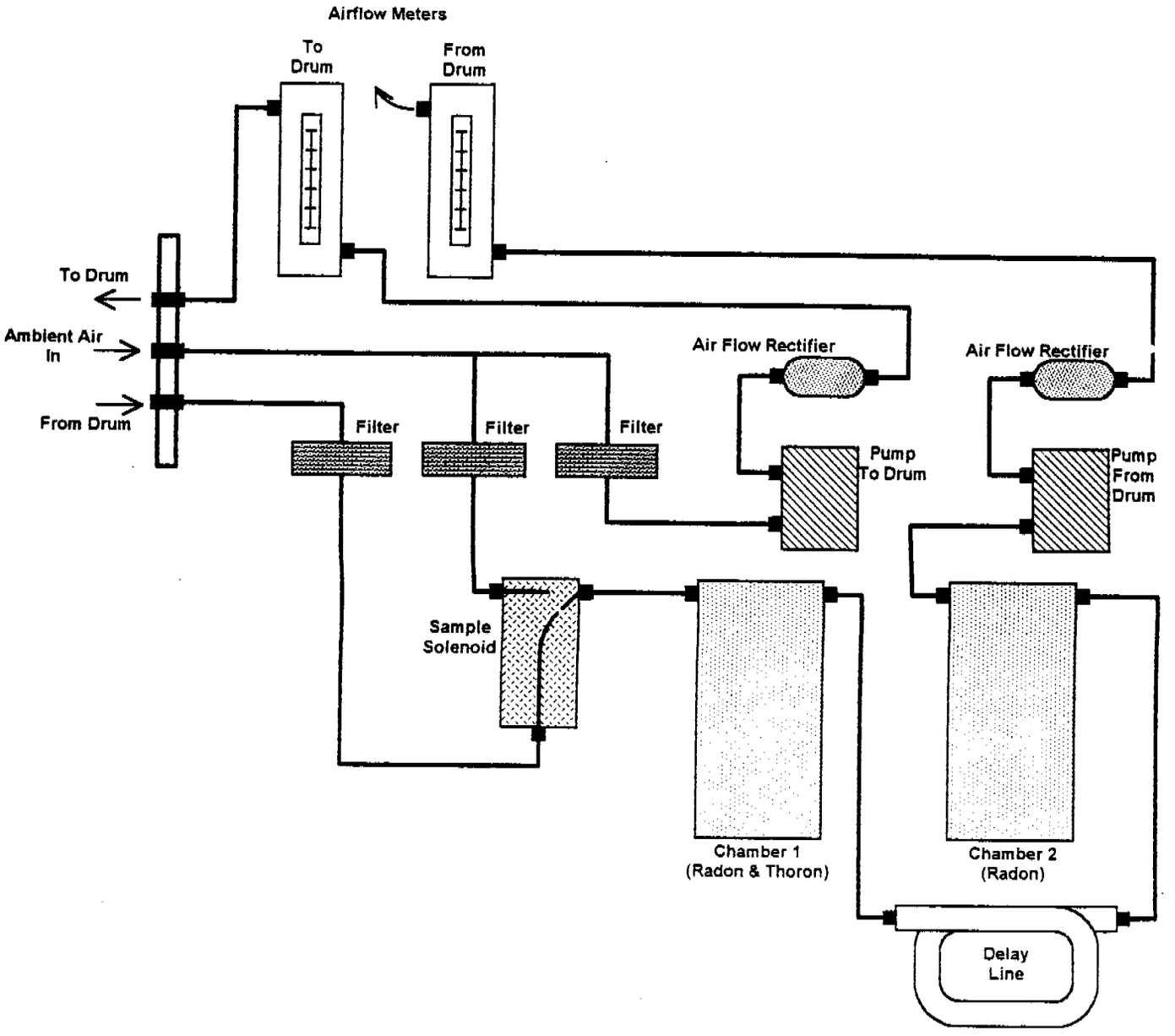


Figure 2.2 Schematic Diagram of the Cutting Device Used to Accurately Place the Emanometer

Figure 2.1 Schematic Diagram of Air Flow Through the Radon/Thoron Emanometer

Diagram Supplied by N. Giannakas,

Queensland University of Technology Centre for Medical and Health Physics



states that a tarpaulin may need to be placed over the drum in particularly windy conditions however this was never necessary.

The equipment may be run in two modes: background mode, where ambient air is drawn from an inlet on the side of the electronics box, or drum mode where air is drawn from the drum. This allows background measurements to be performed. Sources of background are: ^{222}Rn or ^{220}Rn daughters remaining in the cells after a cycle, radon in the ambient air, contamination of the air lines and drum, and ^{210}Pb which has built up in the scintillation cells (Whittlestone). Background is only a problem when measuring sites with particularly high flux. To determine activity flux a number of background measurements are taken followed by an emanation cycle. The number of count periods per cycle, nc, and the length of these (the count time), cp, may be adjusted. During the last count period ambient air is sampled. This allows the sample of air in the first detector to reach the second detector before the cycle is complete. After each measurement the equipment is left to count in background mode until the background reaches a suitable level. This assists in reducing the concentration of daughter products remaining in the detector after a measurement.

The same basic procedure was used at each site and during laboratory work. At least two ambient cycles were performed before each measurement. Each emanation cycle consisted of 6 count periods of 6 minutes each. Following each cycle the detector was run in background mode until the counts reached a suitable level (typically 30 min).

From the count rates measured the ^{220}Rn and ^{222}Rn flux densities are determined using the accompanying software.

Whilst performing preliminary measurements it was found the high voltage supplied to the detectors increased with temperature. Before determining the effect of temperature on the emanometer's performance the effect of variations in high voltage was determined. Therefore the effect of voltage supply on the emanometer's performance was examined. A radium source of approximately 180Bq (radon emanation rate of 1.5 Bq/g/s) electrostatically deposited on a metal disk was placed centrally under the drum. Emanation cycles were run with the usual six steps but a count period of only 1min. This prevented excessive build up of background in the detectors. The high voltage was increased from approximately 700V to 850 V. The total counts measured in the first detector over the six count periods was examined as a function of the high voltage (Figure 2.3).

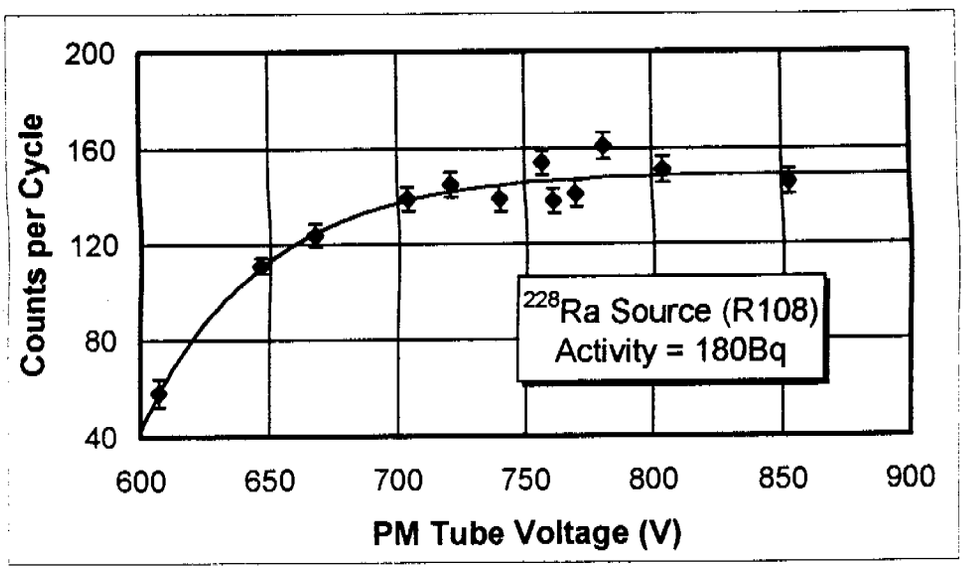


Figure 2.3 Detector Response to Variations in the High Voltage Supplied to the Photomultiplier Tube.

In the range 720V to 850V there is no significant variation in the total counts. It can therefore be inferred that in this voltage range there would be no significant variation in emanation results due high voltage variations. Throughout subsequent use the high voltage range was limited to 740-780V.

This was achieved by covering the equipment to reduce heating when in the field and by adjusting the gain.

Regular test of the detectors response to high voltage variations and flux from a standard source should be performed. These provide an indication of any system variations or faults (Conversation with Whittlestone).