Figure 1.2 The ^{232}Th Decay Series (4n Series) (adapted from Sedlet, 1966)

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**LA THÈSE A ÉTÉ
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SPONTANEOUS DEPOSITION OF RADON DECAY PRODUCTS FROM AIR

by

JEFFREY WELLINGTON CARD

A thesis submitted to the Faculty Of
Graduate Studies in partial fulfilment
of the requirements for the degree of
Master of Science

Carleton University
Ottawa, Ontario
May, 1978

c 1978 Jeffrev Wellington Card

The undersigned hereby recommend to the Faculty Of
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ABSTRACT

Solid materials, including metals, plastics, and foils, exposed in tanks of ^{222}Rn -bearing air, collect significant amounts of radon decay-product activity. The amount of activity collected increases with the surface area of the solid collector, the volume of the tank in which it is exposed, and the duration of the exposure. A maximum amount of activity is approached at the end of a three-hour exposure. There is no significant difference in the ability of the different materials studied to collect radon decay products.

Most of the activity collected is in the form of ^{218}Po and ^{214}Pb at the time of collection. The ratio of the amount of activity collected as ^{214}Pb , to the amount collected as ^{218}Po , is greatest in larger tanks, but does not depend on the surface area of the solid collector or on the duration of the exposure.

Solid collectors exposed overnight in holes in the soil cover overlying the South March, Ontario uranium-copper occurrence collected radon decay products. Measurement of the activity on the collectors delineated an anomaly which had previously been outlined by conventional exploration techniques.

Solid collectors exposed for thirty days in the air in glass jars partially filled with soil samples from the occurrence also collected radon decay products in quantities which resulted in the delineation of the anomaly. The results of this work compared favourably with the results of analyses of duplicate samples for uranium using the conventional fluorimetric technique.

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CHAPTER 1. INTRODUCTION

1.1 The Relationship Between Radon And Uranium

The use of the inert gas, ^{222}Rn , to detect buried uranium ore deposits has become an accepted exploration technique. Three natural isotopes of radon occur. Each is a member of a separate decay series. The three isotopes, their half-lives, and the decay series in which each occurs are as follows:

<u>Isotope</u>	<u>Half-life</u>	<u>Decay Series</u>
^{222}Rn	3.823 days	^{238}U
^{220}Rn	55 seconds	^{232}Th
^{219}Rn	4 seconds	^{235}U

The decay schemes for the ^{238}U , ^{232}Th , and ^{235}U series, showing the relationship of each radon isotope to its precursors and decay products, are given in Figures 1.1, 1.2, and 1.3, respectively.

Most soils and rocks contain uranium in a concentration of a few parts per million. An ore deposit must, however, contain at least 500 parts per million uranium in order to be commercially viable. Since natural radon is produced only by decay of uranium or thorium through several transformations, the presence of anomalously large concentrations of radon in soil air, or in lake or stream waters, may indicate that there is a buried deposit of one of these elements.

Measurements of radon in the soil over buried uranium deposits or occurrences often result in the delineation of a radon anomaly. The usual explanation for such anomalies is that radon, created by decay from primary uranium, has emanated upwards through the

Figure 1.1 The ^{238}U Decay Series $\cdot (4n + 2)$ Series (adapted from Sedlet, 1966)

Figure 1.2 The ^{232}Th Decay Series (4n Series) (adapted from Sedlet, 1966)

Figure 1.3 The ^{235}U Decay Series ($4n + 3$ series) (adapted from Sedlet, 1966)

soil, as shown schematically in Figure 1.4a. The anomalies associated with some deposits may, however, be the result of emanation of radon created by decay from primary uranium in a buried deposit, secondary dispersion halo (Figure 1.4b). Another possibility is that the secondary dispersion halo is sufficiently large that it extends to the upper surface of the soil (Figure 1.4c). In this case, the measured radon may have migrated a rather short distance from its source. A radon measurement may reflect the combined effect of radon created in more than one of the sources.

Radon dissolved in natural waters may also indicate uranium mineralization. After being leached from buried deposits by groundwater, uranium series nuclides may be transported to lakes or streams. If the target has been glaciated, mechanical dispersion of mineralized source material may have broadened the area from which leaching occurs.

There is no *prima facie* reason to expect that radon, whether created in a deposit, or in a secondary dispersion halo, has been derived from nuclei which remained immobile during the several nuclear transformations between radon and primary uranium. In the ^{238}U decay series, the nucleus exists as six successive species before decaying to ^{222}Rn . Each of these species may respond differently to the geochemical environment in which it resides. Accordingly, it is more appropriate to interpret a ^{222}Rn anomaly as reflecting anomalous amounts of its immediate precursor, ^{226}Ra , at the source. ^{226}Ra may have been separated from its primary source, ^{238}U , or vice versa, and the application and interpretation of radon techniques must include consideration of these possibilities.

(a) Migration of radon from ore deposit

(b) Migration of radon from secondary dispersion halo

(c) Occurrence of radium (and radon) in secondary dispersion halo.

Figure 1.4 Possible Types Of ^{222}Rn Dispersion Patterns.
(Arrows depict pattern of movement of detectable radon. Dashed lines in (b) and (c) represent secondary dispersion haloes.)

Smith et al (1976) have emphasized that radon concentrations may be significantly influenced by the geochemical mobility of the precursor elements in the decay series, and have commented on the lack of knowledge on this topic.

In any environment, the relationship between the relative abundances of successive members of a decay series can be discussed in terms of the concept of radioactive equilibrium. Radioactive equilibrium between a nuclide and its decay product occurs when the activity of the decay product is equal to the activity that would be expected if the nuclide and the decay product had been kept together in a closed system for a very long time. For most practical purposes, a time of several half-lives of the decay product is a sufficiently long time to establish a very close approximation to equilibrium. If the half-life of the parent is several orders of magnitude longer than the half-life of the decay product, the equilibrium relationship is described as one of secular equilibrium. When secular equilibrium occurs,

$$\lambda_p N_p = \lambda_d N_d, \quad 1.1$$

where λ_p is the decay constant of the parent,

λ_d is the decay constant of the decay product,

N_p is the number of nuclei of the parent, and

N_d is the number of nuclei of the decay product.

A decay series involving three or more successive nuclear species is conveniently stated to be in radioactive equilibrium if the decay rates of all of the species are identical.

Radioactive decay is a random process, such that it is impossible

to predict when a given nucleus will decay. If there are many nuclei of a species, the observed decay rate at any time tends to be directly proportional to the number of nuclei. The relationship can be written as

$$\frac{dN}{dt} = -\lambda N, \quad 1.2$$

where $\frac{dN}{dt}$ is the decay rate,

λ is the decay constant (the constant of proportionality),

and N is the number of nuclei.

This equation can be solved to yield

$$\frac{N}{N_0} = e^{-\lambda T} \quad 1.3$$

where N_0 is the number of nuclei when time, T , = 0.

For a series of n nuclides in secular equilibrium,

$$\lambda_1 N_1 = \lambda_2 N_2 = \lambda_3 N_3, \quad 1.4$$

where λ_1 , λ_2 , and λ_n are the decay constants for the first, second, and n th members of the decay series, respectively,

and N_1 , N_2 , and N_n are the number of nuclei of the first, second, and n th members of the decay series, respectively.

The relative number of nuclei of each species can be expressed as

$$\frac{N_1}{N_2} = \frac{\lambda_2}{\lambda_1}; \dots \frac{N_1}{N_n} = \frac{\lambda_n}{\lambda_1}, \quad 1.5$$

or as

$$\frac{N_1}{N_2} = \frac{T_{1/2_1}}{T_{1/2_2}}; \dots \frac{N_1}{N_n} = \frac{T_{1/2_1}}{T_{1/2_n}}, \quad 1.6$$

where $T_{1/2_1}$, $T_{1/2_2}$, and $T_{1/2_n}$ are the half-lives for the first, second, and nth members of the decay series, respectively.

A convenient unit to describe the decay rate of a nuclear species is the Curie. One Curie is the activity of any species that is decaying at the rate of 3.7×10^{10} disintegrations per second. The picoCurie (10^{-12} Curies) is generally used to describe the activity of naturally-occurring nuclides. The units, picoCurie per liter, or picoCurie per gram, are often used to describe the concentration of radon in air or in water.

Of the three radon isotopes, ^{222}Rn is the only one that is of significant value in uranium exploration: ^{219}Rn is of little importance, because of the relatively low abundance of its uranium precursor, ^{235}U . The relative activity of ^{219}Rn to that of ^{222}Rn in a closed system in which both isotopes are in secular equilibrium with their respective uranium precursors can be computed as

$$\frac{N_{235\text{U}} \times \lambda_{235\text{U}}}{N_{238\text{U}} \times \lambda_{238\text{U}}},$$

where $N_{235\text{U}}$ and $N_{238\text{U}}$ are the number of atoms of ^{235}U and

^{238}U , respectively, and

$\lambda_{235\text{U}}$ and $\lambda_{238\text{U}}$ are the decay constants for ^{235}U and

^{238}U , respectively.

The terrestrial isotope ratio $N_{235\text{U}}/N_{238\text{U}}$ is 0.007. Accordingly, the relative activity is $0.007 \times \left[\lambda_{235\text{U}} / \lambda_{238\text{U}} \right]$, or 0.044.

^{220}Rn is also of little value in uranium exploration. This nuclide

is a member of the ^{232}Th decay series, and hence its presence does not indicate uranium mineralization. ^{232}Th is about four times as abundant as uranium in unmineralized rocks. Using this factor, the relative activity of ^{220}Rn to ^{222}Rn is 1.28, in a closed system in which both nuclides are in secular equilibrium with their respective uranium or thorium precursors. On this basis, techniques designed to measure ^{222}Rn may suffer from significant interference by ^{220}Rn if precautions are not taken.

1.2 Radon Migration

For radon from buried uranium deposits or from buried secondary dispersion haloes to be detectable near the surface of a soil, it must first escape from the host mineral grain in which it is included. After escape, it must migrate through the soil. The processes of escape and migration have been reviewed by Tanner (1964) and Card (1975).

The radioactive decay of a radium atom occluded in a mineral grain in a soil results in the emission of an alpha particle. The resulting radon atom recoils in the opposite direction with a momentum of the same magnitude as that of the alpha particle. These recoil atoms may be ejected from the grain if they are located sufficiently close to the grain boundary and if the direction of recoil carries them towards the boundary. The recoil atoms may have sufficient residual energy after crossing the grain boundary to allow them to penetrate a nearby grain, in which they may become occluded. For some of the atoms, sufficient energy may be lost in crossing pore-spaces to prevent penetration. These atoms are free to migrate through the pore-space network.

Of the atoms which have not escaped or which have become occluded in a nearby grain, a few may escape by diffusion through the grain to a grain boundary or fracture. The diffusion rate for gases in minerals is so small that only those atoms ending their recoil paths very close to a grain boundary have a chance to escape before decaying. The ratio of the number of radon atoms escaping to the number produced is called the emanation coefficient. Typical values of the emanation coefficient in fine-grained porous material are about 0.1. Measurements, by Barretto (1975) of radon escape from a number of powdered rock samples suggested that emanation coefficients were highest in granites and lowest in sandstones.

The pore spaces in which the escaping fraction comes to rest after recoil may be dry or may be partly or entirely filled with water. If the pore-spaces are water-saturated, radon migration depends both on diffusion and on patterns of movement of the water. If the pore spaces are dry, migration is controlled by processes of diffusion in air; the latter can be modified by meteorological factors.

The diffusion coefficient for radon in air has been given as $0.120 \text{ cm}^2/\text{sec}$ (Hirst and Harrison, 1939). A slightly lower value, $0.054 \text{ cm}^2/\text{sec}$, was observed for dry building sand (Bulashevich and Khairitdinov, 1959). Schroeder et al (1965) found diffusion coefficients which ranged between 0.02 and $0.10 \text{ cm}^2/\text{sec}$ in various types of soil. Blake and Page (1948) investigated the effect of soil porosity on gaseous diffusion rates, and showed that these rates were higher in more porous soils.

The diffusion coefficient for radon in water, 0.95×10^{-5} cm²/sec (Ramstedt, 1919), is about four orders of magnitude lower than the values for the dry media. On this basis, radon in stagnant ground-water has relatively little chance of diffusing to the soil surface.

Where the lower part of a soil is water-saturated and the upper part is dry, radon may exchange between the water-filled pore spaces and the air-filled pore spaces. Rutherford (1913) has noted that radon tends to become partitioned between the air and the water in a closed system according to the relation:

$$a = \frac{E_w \times V_a}{E_a \times V_w},$$

where a is the coefficient of absorption,

E_w and E_a are the total amounts of radon mixed with water and with air, respectively, and

V_w and V_a are the total volumes of water and air, respectively.

The coefficient of absorption, a , is strongly temperature-dependent. At 20.3 °C, $a = 0.250$, whereas at 0 °C, $a = 0.507$.

Meteorological factors may cause significant variations in the radon content of a soil. Measurements by Kraner et al (1964), and other workers, have shown how barometric pressure variations, rain, and wind movements can modify the concentration of radon in soils. Barometric pressure fluctuations tend to mix soil air with atmospheric air, and this results in a reduction of the radon content of the soil air. Radon in soil air may become dissolved in downward-percolating rain-water, or may be removed from the

soil into the atmosphere by physical displacement by the water. Cases are known where the radon content of a soil has increased after a rainfall, and this is believed to be due to a water-saturated upper layer of soil which acts as a barrier to radon escape. Winds result in short-term oscillations of the barometric pressure, a process known as "turbulent pumping", and radon is removed from soils by this means. Changes in soil temperatures give rise to convection currents, which transport radon out of the soil.

The radon concentration of atmospheric air is generally several orders of magnitude lower than that of soil air. A typical soil in non-mineralized terrains might be expected to contain a few hundred picoCuries of radon per liter of soil air. Hess (1943) found the time-average of many measurements of the radon content of New York air at twelve meters above ground to be 0.097 picoCuries per liter.

Meteorological factors act not only to release radon from soils, but also to bring radon up from relatively great depths to shallower levels, where it may be more readily detected.

A convenient way to appraise the over-all effect of meteorological variables is to make a series of radon measurements at a single soil site under a variety of weather conditions. Several sites were monitored daily for many days by Bhatnagar (1973). For six typical sites the mean radon activities, with their standard deviations, were as follows:

Site	Number Of Observations	Radon Activity	
		Mean (cpm)	Standard Deviation (cpm)
A	46	223	50
B	52	313	97
C	52	349	65
D	42	458	122
E	52	305	75
F	34	210	70

Dyck (personal communication, 1976) has suggested that soil-gas radon concentrations, measured at different times, usually do not vary by more than a factor of two. Morse (1976) noted that, although radon concentrations in soil-gas varied from time to time, areas of anomalously high radon concentrations could still be recognized.

1.3 Use Of Radon In Uranium Exploration

The application of radon to uranium exploration includes:

- (i) the direct measurement of radon in water samples from lakes, streams, springs, and wells,
- (ii) the use of radon to obtain a measure of radium in soil samples and in sediment samples from lakes and streams, and
- (iii) the measurement of radon in soil air.

Exploration for uranium is a two-stage process. In relatively unexplored terrains, the initial reconnaissance stage involves work designed to find potential targets for more detailed follow-up

work.

At the reconnaissance stage, radon measurements in water samples and radium measurements in sediments may be made as part of a battery of exploration techniques. Typical exploration techniques include geological mapping, airborne gamma-ray spectrometry, and geochemical sampling for uranium analysis. The Canadian Uranium Reconnaissance Program involves both gamma-ray spectrometry and geochemistry (Cameron and Hornbrook, 1976). Of the anomalies discovered by the program, only 80% were common to both techniques (Cameron, 1976).

Because the gamma-ray spectrometer detects gamma rays originating at depths of only thirty centimeters or less (Breiner et al, 1976, and other workers), important relatively-shallowly-buried targets could be missed. Bjorklund (1976) noted that uranium anomalies in Finland, discovered using regional geochemical techniques, could not be investigated by gamma-ray techniques because of apparent leaching of uranium out of the upper parts of the till cover.

Radon and radium measurements may conveniently be made with relatively simple instruments in field laboratories; the rapid availability of radon and radium geochemical results helps to direct the exploration program. Techniques for measurement of uranium in natural waters and sediments include fluorimetry (Smith and Lynch, 1969), fission track analysis (Fleischer and Lqvett, 1968; Fleischer and Delany, 1976; and Wenrich-Verbeek et al, 1976), and measurements of laser-induced fluorescence (Robbins, 1978). Uranium may be measured in sediments by delayed

neutron activation (Bjorklund et al, 1976). Some of these uranium analysis techniques involve very sophisticated instruments and, as a result, samples must be sent out of the field for analysis. The technique involving laser-induced fluorescence has, however, been adapted for use in field laboratories.

Morse (1969) measured radium in lake sediments in the Bancroft area on a reconnaissance scale, and was able to outline areas previously known to be mineralized. Subsequently, Morse (1971) evaluated and compared the application of techniques involving ^{222}Rn , ^{226}Ra , and uranium.

In populated areas, where there is an abundance of wells, ground water can be sampled. Dyck et al (1975) have used radon and uranium in well waters as reconnaissance tools in the Maritime Provinces, and Dyck et al (1976) have performed similar work in the Cypress Hills region of Saskatchewan. Radon measurements on well-waters were conducted by Snihs (1973), to evaluate the health hazards associated with drinking water from wells in different terrains.

Detailed follow-up uranium exploration work may involve the measurement of radon in soil air or measurement of radium in soils. Studies by Dyck (1968), Dyck (1969), and Dyck et al (1975) have shown that buried uranium deposits can be outlined by radon measurements.

1.4 Techniques Of Measurement Of Radon

A table of properties of radon is presented in Appendix 1. The radioactive properties of radon and its short-lived decay

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products can be used to measure radon concentrations. Measurements may be made by counting the alpha particles emitted by radon, or by counting the beta particles or gamma rays emitted by its decay products.

Alpha particles, beta particles, and gamma rays of both high energy and high yield are of value in detecting and identifying nuclides. Energies and yields of the most useful alpha and beta particles and gamma rays for radon detection are shown in Table 1.1.

Most of the methods commonly used for detection of radon in uranium exploration involve the use of alpha-sensitive detectors.

Sédlet (1966) and Lucas (1957) have described the use of alpha-sensitive, zinc-sulfide-coated chambers to measure radon in gases. Dyck (1969), Caneer and Saum (1974), and Morse (1976) have reported the results of the application of the technique to the measurement of radon in soil gas. Soil samples are obtained by applying a vacuum through a hollow probe driven into the soil. This technique has been used as part of the present research, and is described in more detail in Section 3.4.1. Bristow and Jonasson (1972) have discussed the technique as part of a general review of vapour sensing applications to mineral exploration. A related technique involves the measurement of the beta particles given off by the radon decay products, rather than the alpha particles from radon.

Beck and Gingrich (1976) and Gingrich and Fisher (1976)

Table 1.1 Important Alpha, Beta, and Gamma Energies and Yields
In The ^{222}Rn Decay Series

Nuclide	Decay Mode	Alpha Decay		Beta Decay		Gamma Decay	
		Energy (MeV)	Yield (%)	Energy (MeV)	Yield (%)	Energy (MeV)	Yield (%)
^{222}Rn		5.49	100				
^{218}Po		6.00	100				
^{214}Pb				0.69	47		
				0.74	44		
				1.03	6		
^{214}Bi				3.26	19		
						1.764	17
^{214}Po		7.69	100				

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have described the Track Etch method of measuring radon in soils. An inverted cup, containing an alpha-sensitive track detector, is exposed in a hole in the soil for a few weeks. The track detector is then etched, and the tracks are counted using a microscope.

Semiconductor detectors have been described by Gaucher (1976). An inverted cup, containing an alpha-sensitive semiconductor is buried in the soil. The semiconductor is connected by cable to a counting unit; the total number of accumulated counts at any time can be read by connecting a reader unit to the counting unit.

The 1.764-MeV gamma ray from the radon decay product, ^{214}Bi , with a 17% yield, is used to detect uranium series nuclides by gamma-ray spectrometry. Gamma-ray spectrometers are carried in aircraft, in motor vehicles, and on foot. The spectrometer counts the number of pulses which occur within a preset energy "window" centered on 1.764 MeV. Use of this technique gives a measure of the ^{214}Bi content of the terrain, and the uranium concentration can be estimated by assuming secular equilibrium in the decay series between ^{238}U and ^{214}Bi . The 1.764-MeV gamma ray has a relatively high energy and a relatively high yield. Similar high-energy, high-yield gamma rays in the ^{232}Th and ^{40}K decay series are used to obtain a measure of thorium and potassium, in conjunction with the uranium measurements from the ^{214}Bi gamma rays.

Radon dissolved in natural waters may be readily detected

by bubbling air through the water sample and into an alpha-sensitive scintillation chamber.

A technique for the measurement of radium in soils has been described by Sutton and Soonawala (1975).

All of the alpha-particle-based techniques described above involve the counting of alpha particles, without identification of the specific alpha-emitting nuclides. The individual nuclides can be identified ~~either~~ by differences in their half-lives or by differences in the energies of their respective alpha particles. Half-life discrimination can be used with the alpha-scintillation method of radon detection to identify ^{220}Rn . The half-life of the latter nuclide is 55 seconds, and, if the count-rate decreases rapidly after introduction of soil gas into the scintillation chamber, the latter nuclide can be identified and, if desired, measured quantitatively.

Alpha-particle spectrometry, the discrimination of nuclides by the energies of their alpha particles, has not been extensively applied to uranium exploration, but a number of workers have used the technique for studies involving ^{238}U series nuclides. ^{226}Ra has been measured in ores and mill products by Zimmerman and Armstrong (1975), using alpha-particle spectrometry, following leaching and co-precipitation of the radium with barium, as barium sulfate. Killeen and Carmichael (1976) have developed a technique of alpha-particle spectrometry for determining the extent of disequilibrium in radioactive ores. Gorenstein et al (1974) have described measurements of ^{222}Rn and ^{210}Po by alpha-particle spectrometry conducted from the Apollo 15 and Apollo 16 spacecraft orbiting the moon. Lambert et al (1972) applied alpha-

particle spectrometry to studies involving radon outgassing from sand, in a closed system held under vacuum, to approximate lunar conditions. Abe et al (1966) used alpha-particle spectrometry to measure polonium thinly-plated onto silver discs.

In the two chapters which follow, a method of detection of radon involving the spontaneous deposition of radon decay products onto solid collectors will be considered. This work, a study of the spontaneous deposition of radon decay products, and the use of this process to outline a known uranium deposit, is the topic of the thesis.

CHAPTER 2. LABORATORY STUDY OF SPONTANEOUS DECAY PRODUCT DEPOSITION FROM AIR

2.1 The Physical Basis For Spontaneous Deposition

Radon, an inert gas, does not readily enter into chemical reactions and, because of this, radon atoms in a volume of air remain uncombined with other atoms. The processes of thermal diffusion and convection, however, result in movement of the atoms within the volume. In due course, each ^{222}Rn atom decays by emission of an alpha particle; this process results in the production of ^{218}Po . Because ^{218}Po is not an inert gas, it is very likely to enter into processes involving the gain or loss of valence electrons.

A newly-created ^{218}Po entity probably exists initially either as an atom or as an ion, or in combination with an oxygen molecule, forming an electrically-neutral polonium oxide molecule. In any event, in the absence of an applied electric field, the atom, ion, or molecule is free to migrate in air until it comes into contact with a solid surface, to which it may become attached. These processes (migration, followed by attachment) are the basis for spontaneous deposition.

Because ^{218}Po has a half-life of only 3.05 minutes, it is possible for some atoms of ^{218}Po to decay to ^{214}Pb before becoming attached to solid surfaces. In this latter event, it is the ^{214}Pb species which may be spontaneously deposited. Successive decays from ^{214}Pb to ^{214}Bi , to ^{214}Po , to ^{210}Pb , etc., may occur prior to deposition of a particular nucleus.

If the air contains dust or aerosol particles, then a decay

product atom or oxide molecule, which comes in contact with such a particle, may become attached to it. Results in the literature cited by Serdyukova and Savenko (1967) indicated that radon decay products are deposited on aerosols soon after creation and that about 90% of the unattached decay products in the air are of the ^{218}Po species. The fraction of unattached to total ^{218}Po particles in a variety of environments ranged between 0.005 and 0.12 (George, 1972). It appears from this that most of the decay products become attached very soon after they are created. The aerosol or dust particles carrying attached decay products may become deposited on solid surfaces. Accordingly, both attached and unattached decay products may be spontaneously deposited.

A wire or plate placed in a volume of radon-bearing air as a solid surface for deposition of decay products may conveniently be called a collector. When radon decay products are deposited on a collector, the air from which the decay products were collected becomes depleted with respect to the concentration that would be expected by assuming radioactive equilibrium with the supporting radon. When depletion occurs, any second or subsequent decay product may be more highly depleted than any preceding nuclide in the decay series. Measurements by Blanchard (1969) of ^{218}Po , ^{214}Pb , ^{214}Bi , ^{210}Pb , and ^{210}Po in the air of both stagnant and ventilated uranium mine working places showed a depletion in the activity of each nuclide in the series with respect to the activity of any precursor. Diffusion processes are modified by convection, which may act to replenish decay products in volumes which are depleted

due to spontaneous deposition.

Rutherford (1913) described the results of early experimental work involving the spontaneous deposition of radon decay products from air onto solid surfaces. The activity collected on a set of identical solid collecting materials exposed for a constant time in a tank of radon-bearing air was constant, regardless of the position of the collecting material in the tank. The emplacement of a collecting material behind a baffle in a corner of the tank resulted in the collection of the same amount of activity as a collecting material exposed elsewhere in the tank. Croft and Perry (1962) suspended discs of filter paper, hardboard, and aluminum at several heights in a sealed room contaminated with radon, and found that the amount of activity collected was independent of both height and collecting material.

The potential use of radon decay products in detailed geochemical exploration for uranium has not, to the writer's knowledge, been previously investigated. The present research involved a laboratory study of spontaneous deposition of radon decay products from air, followed by an investigation of the feasibility of using spontaneous deposition as a uranium exploration tool. The investigation involved the use of spontaneous deposition to delineate a known buried uranium deposit.

The nuclides involved in the study were the successive radon decay products, ^{218}Po , ^{214}Pb , ^{214}Bi , and ^{214}Po . As illustrated in Figure 1.1, the first three of these decay products have half-lives measured in minutes, and ^{214}Po has a half-life of a few microseconds. The

two polonium isotopes, ^{218}Po and ^{214}Po , are alpha-particle emitters, whereas ^{214}Pb and ^{214}Bi emit beta particles. The activity of the nuclides was measured using an alpha-particle detector, which allowed direct measurement of the alpha-emitters, and indirect measurement of the beta-emitters, following their decay to ^{214}Po . It was anticipated that ^{222}Rn would not become attached to any collector.

The present chapter describes a laboratory study undertaken to investigate some aspects of spontaneous deposition of radon decay products from air. The work involved the exposure of collectors in air tanks containing known concentrations of radon. (For brevity the tanks are hereinafter referred to as "radon tanks".) The effects of type of collector material, surface area of collector, duration of exposure, and size of radon tank were investigated. Both the total activity on collectors, and the form of the activity at the time of collection (whether as ^{218}Po , ^{214}Pb , or ^{214}Bi) were considered. It was hoped that this work would delineate optimum and acceptable parameters for an applied study involving the use of collectors to delineate uranium mineralization in the field.

2.2 Experimental Method

The evaluation of spontaneous deposition consisted of a number of trials that involved the exposure of a collector of known surface area in a radon tank and the subsequent counting of the collected decay-product activity in an alpha-particle counter.

2.2.1 Apparatus

2.2.1.1 Radon Tanks

Collectors were exposed in tanks of volumes 29 liters, 400 milliliters and 70 milliliters. A solution of 0.5 microCuries of radium was used as a source of radon. Radon was transferred from the radium solution to the tanks by use of a Varistaltic Pump. The apparatus is shown schematically in Figure 2.1. To ensure that the maximum amount of radon would be removed from the solution, and distributed uniformly throughout the internal tank volumes, the system was pumped for sixty minutes. The pumping was always performed the day before collectors were inserted.

The large tank was equipped with small ports for insertion and removal of collectors. The ports were plugged at all times, other than the few seconds required to insert or remove a collector, using silicone-covered rubber stoppers. To expose a collector, a wire hook was inserted into a rubber stopper, and the collector was suspended from the hook. The stopper was then inserted into a port. Collectors were suspended in the main volumes of each of the smaller tanks by long wire hooks inserted into the rubber stoppers at the tops of the chimneys.

2.2.1.2 Scintillation-Counting Apparatus

At the end of an exposure interval, the collector was removed from the radon tank and the alpha-particle activity due to collected decay products was counted. The counting configuration, shown in Figure 2.2, consisted of a silver-activated, zinc sulfide-coated scintillation chamber optically coupled to a photomultiplier.

Figure 2.1 Schematic Diagram Of Radon Tank Apparatus Showing Tanks, Pump, Bubbler, And Collectors In Place

Figure 2.2 Schematic Diagram Of Scintillation Counting Configuration, With Collector In Place

tube which, in turn, was connected to a Ludlum Model 2002 scaler.

A standard ^{233}U source (provided by Dr. A Baerg of the National Research Council) was used to determine the optimum voltage output for the scaler, and to determine the detection efficiency of the instrument configuration. The source consisted of a thin layer of ^{233}U bonded to a stainless steel disc about 3.0 centimeters in diameter. The active material was concentrated in a circle of 0.3 centimeters diameter concentric with the disc. The disc was thick with respect to the range of an alpha particle. The source had a total rated activity of 26.8 disintegrations per second. Approximately half of the alpha particles would travel into the disc, and not be detected, and this was taken into account during the calibration procedure. The optimum high voltage output for the scaler was determined by inserting the source into the scintillation chamber and observing the activity over a range of voltage settings. Over part of the range, the observed counts were constant, and these counts resulted in a "plateau" on a plot of counts per minute against voltage setting. The instrument was operated with voltage set at the mid-point of the plateau. Operation at the mid-point minimizes the effect of minor fluctuations in voltage output, resulting from temperature variations and other instrumental factors. With the source in the scintillation chamber, it was observed that the detection efficiency, counts per minute per disintegration per minute, at the mid-point of the plateau, was about 0.8 when the air pressure in the scintillation chamber was reduced to about one-quarter of an atmosphere. The detection efficiency under a pressure of one atmosphere was somewhat lower.

The energies of the alpha particles emitted by ^{233}U are relatively low (4.78 MeV and 4.82 MeV), whereas the energies of the alpha particles emitted by ^{218}Po and ^{214}Po are higher (6.00 and 7.69 MeV, respectively). To investigate the effect of atmospheric pressure on the detection efficiency for alpha particles from these decay products, two identical collectors were exposed for identical periods of time. The activity on one was counted at about one-quarter of an atmosphere, and the activity on the other was counted at one atmosphere. The results indicated that detection efficiencies for the decay products were not pressure-dependent, and this made it possible to conduct the measurements at atmospheric pressure. The ^{233}U standard was mounted at different positions in the scintillation chamber, and oriented towards different sectors of the walls, to show that detection efficiency would not vary markedly for activity originating at these different positions. Finally, pairs of identical collectors exposed for identical intervals were counted, successively, in centrally-mounted and eccentrically-mounted positions. Detection efficiencies did not vary with position.

2.2.2 Selection Of Collecting Materials

Suites of both metals and plastic materials were used as collectors. Most of the trials were performed using collectors that were thicker than the range of an alpha particle (about 30 microns), but a number of trials involved foils, materials thinner than the range of an alpha particle.

Silver, copper, aluminum, brass, platinum, and gold were the metals used. Silver was an obvious choice, because polonium readily deposits on silver in aqueous solution (Bagnall, 1966; DeBoeck, 1968;

Flynn, 1968; and Martin and Blanchard, 1969). Copper was chosen because of its chemical similarity to silver. Platinum and gold were chosen to represent the inert metals.

Containers in which radon-bearing air might be enclosed could conveniently be made of a plastic, such as plexiglass. Since some portion of the decay products in any volume is likely to become spontaneously deposited onto the walls of such a container, plexiglass was included in the suite of materials used as collectors. To obtain the maximum number of decay products on a collector, it would be best to use a radon container of a material with a relatively small collection efficiency for decay products, and a collector with a relatively large collection efficiency. In addition to plexiglass, the plastics, lexan and cellulose nitrate, were used as collectors.

The activity of spontaneously-deposited nuclei might be conveniently measured using a collector overlain on, and in immediate contact with, a detector. If the collector were in the form of a foil with a small thickness relative to the range of an alpha particle, the collection and measurement operations could be performed simultaneously, avoiding the necessity of transporting the collector to an alpha-particle detector. As examples, a foil collector could be overlain on a zinc sulfide screen (optically-coupled to a photomultiplier tube), a semiconductor detector, or an alpha-sensitive plastic track detector. Using this method, the best collecting material would be the one with the highest collection efficiency. Accordingly, several foil materials were included in the study: silver, brass, copper, and Kimfol.

Most of the collectors were in the form of wires or laths about 5 centimeters in length.

2.2.3 Exposure, Counting, And Error Evaluation Procedures

Most of the exposures of collectors were carried out over a ten-minute period, or overnight; and a few exposures were for one-hour or three-hour periods. Exposures of three hours or longer may be referred to as "steady-state" exposures because the activity on collectors does not increase markedly for exposures longer than three hours. The time that each collector was inserted into and removed from a radon tank was recorded. Each collector was placed in the scintillation chamber as soon as possible after removal from the radon tank. This took approximately two minutes. To avoid the effect of temporary excitation of the zinc sulfide by natural or artificial light, insertion of the collector into the scintillation chamber was done in the dark. The adequacy of this precaution was confirmed by inserting unexposed collectors into the scintillation chamber and noting that the instrument response was at a normal background level. The activity from each collector was counted for a series of consecutive one-minute intervals; counting was started as soon as the collector was inserted into the scintillation chamber and continued for thirty-five minutes after removal of the collector from the tank. To record the value of each count and to reset the counter at the end of each one-minute counting interval required about five seconds. The time of starting each count was recorded, to the nearest full minute.

Instrument background was determined before the insertion of

a collector into the scintillation chamber. Four consecutive one-minute background counts were taken, and the results were averaged. Instrument backgrounds were very low in comparison to the readings obtained from the exposed collectors. Signal-to-background ratios ranged from 20:1 to 1000:1, depending on exposure time, collector surface area, and tank volume. Occasionally, during insertion or removal of a collector, physical contact between the collector and the walls of the scintillation chamber would cause contamination, which increased instrument background by a few counts per minute. Such contamination was minor, in comparison with the activity of the contaminating collector.

There has been no evidence that decay products have escaped from the collectors during their residence in the scintillation chamber. When collectors were carefully inserted and removed, backgrounds before and after the counting operation were identical. Any activity leaving a collector would probably have become redeposited not only on the collector itself but also on the walls of the scintillation chamber. The activity on the walls of the scintillation chamber would be detectable as an increase in background. This was not observed.

The primary interest, in the present study, was in collected decay products of ^{222}Rn , and it was anticipated that ^{222}Rn itself would not be deposited on any collector. To confirm that ^{222}Rn was not deposited, a few collectors were exposed in a tank and then left overnight in the scintillation chamber. The only counts detected the following day were those equivalent to a normal background.

Accordingly, it appeared that all of the activity on collectors was in the form of radon decay products.

About half of the alpha particles from a collector are directed into the collector. During the scintillation-counting procedure, these alpha particles are not counted if the thickness of the collector is greater than the range of an alpha particle. For collectors in the form of foils with a thickness less than the range of an alpha-particle, some inward-directed alpha particles may emerge from the opposite side of the foil with sufficient residual energy to cause a count to register. To avoid the extra counts, each foil collector was prepared for exposure and counting by wrapping a sheet of the foil around a piece of lexan; in this way, only the foil material was exposed to the air in the radon tank, and the counting of inward-directed alpha particles was avoided.

For a typical exposure, the counting history was as follows:

<u>Time</u>	<u>Event</u>
4:15:00	Remove collector from tank
4:16:50	Start first one-minute counting interval
4:17:55	Start second one-minute counting interval
4:19:00	Start third one-minute counting interval
	⋮
4:43:10	
4:44:15	
4:45:20	Start of counting intervals for 28th to
4:46:25	32nd minutes after termination of exposure
4:47:30	

An arithmetic treatment was applied to the results for each exposure. The arithmetic mean of the five one-minute net counts for the 28th, 29th, 30th, 31st, and 32nd minutes was computed. The result was divided by the detection efficiency of the cell used (about 0.8, in terms of counts per minute per disintegration per minute), and multiplied by 2 to obtain the total activity on the collector (assuming that 50% of all alpha particles are directed into the collector, and not counted). The total activity was then divided by a factor to correct for loss of radon in the tanks by radioactive decay between the time of the last circulation of the pump and the time of termination of the exposure of the collector.

During the appraisal of a block of data obtained in the early stages of the study, it became clear that the observed counts were systematically high when compared with data for similar collectors inserted at a later date. These earlier results were normalized to the later data using information obtained from three matching pairs of collectors. The ratios were 0.534 for sterling silver exposed for ten minutes, 0.391 for silver exposed overnight, and 0.462 for lexan exposed overnight. The similarity of these three ratios suggests a systematic error. It has not been possible to explain this.

A number of replicate exposures were performed, to obtain an estimate of the precision of the measurements. Results of exposures of similar materials are shown in Table 2.1, including mean values and standard deviations. When these standard deviations are expressed as a percentage of their associated means, it can be seen that

Table 2.1 Summary Of Replicate Analyses Of Collectors
Exposed In The 29-Liter Tank

Material	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Mean	Standard Deviation	Standard Deviation As A Percentage Of Mean
A. Steady-State Exposures					
Silver	1.3	2160			
Silver	1.3	2211			
Silver	1.3	2143			
Silver	1.3	1828			
Silver	1.3	1958	1987	331	17
Silver	1.3	1748			
Silver	1.3	1389			
Silver	1.3	2460			
Lexan	5.0	2747			
Lexan	5.0	2842			
Lexan	5.0	3140	2417	704	29
Lexan	5.0	1878			
Lexan	5.0	1476			
Plexiglass	10.2	2618			
Plexiglass	10.2	3126			
Plexiglass	10.2	2973			
Plexiglass	10.2	3581	3119	649	21
Plexiglass	10.2	2312			
Plexiglass	10.2	4105			
B. Ten-Minute Exposures					
Silver	1.5	223			
Silver	1.3	277			
Silver	1.3	272	280	51	18
Silver	1.3	347			
Silver	9.3	362			
Silver	9.3	348	372	14	4
Silver	9.3	393			
Lexan	5.0	490			
Lexan	5.0	463	478	12	2
Lexan	5.0	480			

one standard deviation can amount to an error as high as 29%. Using the theory provided by Friedlander et al (1964), it can be shown that the error due to counting statistics alone is likely to be less than five percent for all exposures in the 29-liter tank. Accordingly, a significant portion of the total error appears to be due to factors other than counting statistics. Some of the factors which may have contributed to the uncertainty are: variable radon leakage from the tanks; fluctuations in the detection efficiency of the scintillation-counting apparatus; and differences in the collection efficiency associated with the position of the collectors in the tank during the exposure intervals.

2.3 Discussion Of Results

A complete tabulation of the results for all exposures is given in Appendix 2. The tabulation includes the results of over one hundred exposures, each involving at least 35 minutes of counting. The following findings are significant:

- (i) activity was detected on all collectors in amounts that were very large with respect to instrument background;
- (ii) the amount of activity was greatest on larger collectors;
- (iii) greater amounts of activity were recorded for longer exposure times;
- (iv) more activity was collected from the air in the large tank than from the air in the smaller tanks; and
- (v) the amount of activity was independent of the nature of the material which formed the collector.

Appendix 2 reveals that most of the exposures resulted in the

collection of hundreds or thousands of counts per minute of activity. The amount of activity was very great in comparison with instrument background, which was usually about two or three counts per minute.

The relationship between the activity on a collector and exposure time is clearly shown in Table 2.2. The table, which provides data for a metallic collector and two plastic collectors, indicates that maximum activity is reached after an exposure of three hours duration. The activity on collectors exposed for three hours is essentially identical to the activity on collectors exposed overnight. After three hours, the rate of decay is presumably equal to the rate of collection, so that there is a state of equilibrium between the rate at which decay products are being collected and the rate at which they are decaying.

A summary of the total activity measured on collectors exposed for three hours or longer in the 29-liter tank is given in Figure 2.3. The positive correlation between activity and surface area indicates that the activity is greatest on collectors with higher surface areas. The collectors having small surface areas have, however, collected significant amounts of activity with respect to the amount of activity on the collectors of large surface area; materials of surface area between 4.1 square centimeters and 10 square centimeters have collected about 40% more activity than metals of surface area less than 2 square centimeters.

To investigate the effect of surface area in more detail, two metallic materials and two plastic materials were selected for further study. All of the collectors were in the form of plates. A broader range of surface areas was investigated than was used

Table 2.2 Relationship Between Activity On A Collector
And Duration Of Exposure In 29-Liter Tank

<u>Material</u>	<u>Duration Of Exposure</u>	<u>Number Of Determinations</u>	<u>Mean Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)</u>
Silver	10 minutes	3	299
Silver	3 hours	3	1655
Silver	overnight	4	2118
Silver	3 days	1	2143
Lexan	10 minutes	3	478
Lexan	3 hours	2	2509
Lexan	overnight	3	2355
Plexiglass	10 minutes	1	812
Plexiglass	3 hours	2	3179
Plexiglass	overnight	6	3269

Figure 23 Total Activity On Collectors Of Different Materials And Surface Areas Exposed In The 29-Liter Tank For Three Hours Or Longer

to obtain the results given in Figure 2.3. The results of the detailed surface area study, given in Figure 2.4, confirm that surface area is an important parameter in determining the total activity on a collector. Greater amounts of activity were found on collectors with larger surface areas. It is also interesting to note that there were no significant differences in the amounts of activity on the different materials of similar surface area.

The results of the ten-minute exposures, presented in Figure 2.5, also show a positive relationship between activity and surface area. The results are more-or-less identical to the results for the steady-state exposures, although the amount of collected activity was much smaller. A few metallic collectors of surface area between 7 square centimeters and 10 square centimeters were included in the study, for comparison with metals of lower surface area. For these metals, the relationship between activity and surface area is similar to that observed for the longer exposures.

The results of the exposures in the 400-milliliter and 70-milliliter tanks are presented in Table 2.3. It is useful to compare these results with those from similar materials of similar surface area and shape exposed in the 29-liter tank. Collectors exposed in the 29-liter tank have collected more activity than those exposed in the 400-milliliter tank; collectors exposed in the 400-milliliter tank have similarly collected more activity than those exposed in the 70-milliliter tank. These observations clearly indicate that the amount of activity collected depends on tank volume.

An analysis of the shape of the decay curve after termination

Figure 2.4. Relationship Between Total Activity On Collectors And Surface Area, For Four Suites Of Collecting Materials, Exposed For Three Hours Or Longer In The 29-Liter Tank

Figure 25 Total Activity On Collectors Of Different Materials And Surface Areas Exposed In The 29-Liter Tank For Ten Minutes

Table 2.3 Comparison Of Total Activity On Collectors
Exposed In 70-Milliliter, 400-Milliliter,
And 29-Liter Tanks

	Total Activity On Collectors At 30 Minutes After Termination Of Exposures		
	<u>29- Liter Tank</u>	<u>400- ml Tank</u>	<u>70- ml Tank</u>
Ten-Minute Exposures			
Silver	280	-	45
Plexiglass	812	122	8
Lexan	478	135	76
One-Hour Exposures			
Silver	1131	127	-
Plexiglass	-	258	82
Lexan	-	224	42
Steady-State Exposures			
Silver	1987	156	42
Plexiglass	2922	352	113
Lexan	2745	320	131

Note: The number of digits presented has arithmetic
significance only.

of each exposure was performed, so as to obtain an indication of the relative numbers of particles collected as ^{218}Po , ^{214}Pb , and ^{214}Bi . If an equilibrium assemblage of the three nuclides were collected, then for each ^{218}Po atom collected, the number of ^{214}Pb atoms collected would be simply $\left(\lambda_{^{218}\text{Po}}/\lambda_{^{214}\text{Pb}}\right)$, and the number of ^{214}Bi atoms collected would be $\left(\lambda_{^{218}\text{Po}}/\lambda_{^{214}\text{Bi}}\right)$. If the nuclides are not collected in equilibrium proportions,

the number of ^{214}Pb atoms collected may be expressed as $F_1 \times \left(\lambda_{^{218}\text{Po}}/\lambda_{^{214}\text{Pb}}\right)$ atoms per ^{218}Po atom. Similarly, the number of ^{214}Bi atoms collected may be expressed as $F_2 \times \left(\lambda_{^{218}\text{Po}}/\lambda_{^{214}\text{Bi}}\right)$ atoms per ^{218}Po atom. The evaluation of F_1 and F_2 yields a measure of the relative number of atoms of each species collected.

A mathematical method of deriving F_1 and F_2 is given in Appendix 3. The method requires the measurement of the total activity on a collector for three different finite time intervals after termination of an exposure. For the present study the counting intervals were of five minutes duration; the first interval represented the total number of counts from three minutes after termination of the exposure to eight minutes after termination of the exposure; the second and third intervals represented the counts from the thirteenth to eighteenth minutes, and from the twenty-eighth to thirty-third minutes, respectively. The ratios of the values yielded in these finite counting intervals allowed the determination of F_1 and F_2 . It has been assumed that each species of nuclide was provided to the collector at a constant rate as a function of time. Although this assumption is an oversimplification,

it is believed that important differences in the relative numbers of particles of each species may be recognized by application of the mathematical analysis.

Values of F_1 and F_2 for the ten-minute and steady-state exposures in the 29-liter tank are given in Tables 2.4 and 2.5; the values for the detailed surface area study are given separately in Table 2.6, and the values for the smaller tanks are given in Table 2.7.

The mean values and standard deviations of F_1 and F_2 , based on replicate determinations, are as follows:

Material	Number Of Determinations	F ₁		F ₂	
		Mean	Standard Deviation	Mean	Standard Deviation
A. Steady-state Exposures					
Silver	8	.20	.16	-.03	.06
Lexan	5	.15	.07	.03	.04
Plexiglass	6	.15	.08	.03	.07
B. Ten-minute Exposures					
Silver (small surface area)	4	.18	.05	.03	.05
Silver (large surface area)	3	.20	.03	.00	.02
Lexan	3	.23	.02	.02	.01

The negative values of F_1 and F_2 for some exposures can be explained by the uncertainties associated with the measurements; the negative values have no physical significance.

For all exposures in the 29-liter tank (Tables 2.4, 2.5, and 2.6), F_1 is greater than F_2 . Neither F_1 nor F_2 appear to depend on the nature of the collecting material. F_1 and F_2 appear to

Table 2.4 Summary Of Collection Ratios, F_1 And F_2 , For
Collectors Exposed For Ten Minutes In The
29-Liter Tank

Material	Surface Area (cm ²)	Number Of Determinations	F_1	F_2
Silver	1.3	3	.17	.03
Copper	1.4	1	.11	.07
Brass	1.4	1	.16	.04
Silver	1.5	1	.19	.04
Sterling silver	1.5	1	.13	.01
Platinum	1.6	1	.13	.04
Gold-filled silver	1.9	1	.15	.03
Sterling silver	3.0	1	.13	.02
Lexan	5.0	3	.23	.03
Aluminum foil	5.7	1	.15	.03
Lexan	7.1	1	.20	.03
Aluminized mylar	7.3	1	.30	.04
Aluminum	7.4	2	.12	.04
Kimfol	7.6	1	.19	.05
Mylar	7.7	1	.22	.00
Cellulose nitrate	8.2	1	.26	.04
Silver	9.3	3	.20	.00
Aluminum foil	9.5	1	.20	.02
Copper	9.6	2	.21	.02
Lexan	9.7	1	.36	.06
Plexiglass	10.5	1	.23	.03
Brass foil	10.5	1	.14	.00
Brass foil	12.0	1	.31	.13
Silver foil	13.1	1	.29	.02

Note: The number of digits presented has arithmetic significance only.

Table 2.5 Summary Of Collection Ratios, F_1 And F_2 For
Collectors Exposed For Three Hours Or Longer
In The 29-Liter Tank

Material	Surface Area (cm ²)	Number Of Determinations	F_1	F_2
Gold	0.3	1	.04	-.03
Sterling silver	1.1	1	.08	-.04
Silver	1.3	8	.20	-.03
Copper	1.4	2	.24	.05
Brass	1.4	1	.03	.01
Platinum	1.6	1	.15	.00
Aluminum foil	2.4	1	.05	-.06
Lexan	5.0	4	.17	.03
Cellulose nitrate	5.6	1	.13	.01
Brass foil	6.4	3	.12	.03
Mylar	6.4	1	.16	-.02
Aluminum	6.8	1	.11	.00
Silver foil	7.5	1	.39	.17
Kimfol	7.6	3	.05	-.04
Aluminum foil	9.5	1	.08	-.01
Plexiglass	10.2	6	.18	.04
Kimfol	10.8	1	.13	.08
Silver foil	11.3	1	.09	.00
Plexiglass	12.0	1	.07	-.02
Silver foil	13.0	1	.20	.04

(See also Table 2.6)

Note: The number of digits presented has arithmetic significance only.

Table 2.6 Summary Of Collection Ratios, F_1 And F_2 , For
Collectors Of Different Surface Areas
(Steady-State Exposures)

Material	Surface Area (cm^2)	F_1	F_2
Copper	1.0	.11	.04
	2.0	.06	-.03
	4.1	.04	-.02
	8.1	.03	-.04
	16.1	.04	.00
	34.3	.10	.00
Silver	1.0	.06	.04
	2.0	.12	.00
	8.1	.10	.00
Lexan	1.0	.25	.04
	2.2	.01	-.06
	4.5	.07	.01
	9.8	.05	.01
	19.2	.08	.01
Plexiglass	3.7	.13	.09
	5.6	.10	.03
	11.3	.03	.00
	19.0	.06	-.01

Note: The number of digits presented has arithmetic significance only.

Table 2.7 Comparison Of Collection Ratios, F_1 And F_2 ,
For Collectors Exposed In 70-Milliliter,
400-Milliliter, And 29-Liter Tanks

	29- Liter Tank		400- ml Tank		70- ml Tank	
	F_1	F_2	F_1	F_2	F_1	F_2
Ten-Minute Exposures						
Silver	.17	.03	-	-	.06	-.12
Plexiglass	.23	.03	.09	.06	-.04	.05
Lexan	.36	.06	.14	.00	.06	.03
One-Hour Exposures						
Silver	.14	.01	.06	-.03	-	-
Plexiglass	-	-	.06	.02	-.01	-.01
Lexan	-	-	.10	.02	-.02	-.03
Steady-State Exposures						
Silver	.17	.03	.06	.00	-.23	-.13
Plexiglass	.18	.04	.01	-.04	.08	.07
Lexan	.15	.03	-.02	-.01	.03	-.10

Note: The number of digits presented has arithmetic significance only.

be independent of the duration of the exposure; this can be seen by comparing Tables 2.4 and 2.5. From the data in Table 2.6, it appears that F_1 and F_2 are independent of surface area. From Table 2.7, it appears that lower values of F_1 result from the exposure of collectors in smaller tanks. If activity is collected, either on the walls of a tank or on a collector, soon after the transformation from ^{222}Rn to ^{218}Po , then the values of F_1 and F_2 should be very small. Theoretically, in the limiting case, where all ^{218}Po atoms are collected immediately after creation, F_1 and F_2 must be equal to zero. It appears that the residence time for decay products in the smaller tanks is shorter than the residence time in the larger tank. This is consistent with the view that decay products are collected on the walls of the smaller tanks relatively soon after the transformation from ^{222}Rn .

The data gathered in the laboratory study suggest that all of the materials tested are similar in their ability to collect decay products. The factors of importance in determining the amount of activity collected are collector surface area, tank volume, and duration of exposure.

The writer prefers to regard the results described in this chapter as only semi-quantitative. Refinements to the experimental method would, no doubt, improve the reliability and precision of the results. It is believed, however, that the present work has successfully achieved its objective of identifying the important factors that may be of value in applying spontaneous deposition to uranium exploration.

CHAPTER 3. SPONTANEOUS DECAY PRODUCT DEPOSITION STUDIES APPLIED TO AN AREA OF KNOWN URANIUM MINERALIZATION

3.0 Purpose Of The Studies

The results of the laboratory study indicated that significant radon decay product activity could be collected on solid collectors by spontaneous deposition. It was postulated that this knowledge could be applied to delineate a known uranium deposit. If so, the knowledge might be useful in helping to discover hidden uranium ore deposits.

The application of the collection principle was tested both in situ and in the laboratory, using soil samples. The test results were compared with the results of two conventional uranium exploration techniques; emanometry and fluorimetry. This work is described in the present chapter.

3.1 The South March Uranium-Copper Occurrence

The South March uranium-copper occurrence was discovered fortuitously in October, 1972 by airborne gamma-ray spectrometry (Grasty et al, 1973). Subsequent examinations of the March Formation resulted in the discovery of two additional mineralized zones. Later work at the main South March occurrence involved detailed radiometric work and soil geochemistry (Charbonneau et al, 1975). The showing responds to reconnaissance-scale geochemical surveys involving analysis of radon in stream waters and uranium in stream sediments (Jonasson and Dyck, 1974). Ford (1975) has provided maps at the scale of 1 centimeter = 91 meters, showing the results of detailed radiometric and geochemical work, and these maps are included in the Geological Survey of Canada Open File Report #264.

The anomaly at the main South March occurrence is associated with two intraformational conglomerates of the March Formation, a unit composed of interbedded sandstone, conglomerate and dolomite. Stratigraphically, the unit overlies the Nepean Formation, a sandstone unit resting unconformably on Precambrian granitoid rocks. The overlying unit is the Oxford Formation, of dolomite and limestone.

The mineralization takes the form of uraniferous hydrocarbons, with associated chalcopyrite and pyrite. Ford (1975) has marshalled evidence in favour of an epigenetic origin involving the circulation of low-temperature aqueous fluids.

On the basis of the favourable response of many exploration techniques to the South March occurrence, it was decided that the property would be suitable as a test site to evaluate the collector techniques.

The map of radiometrically-determined uranium concentrations by Ford (1975) was used as a base map. A map showing the uranium contour lines and the traverse lines used for the present evaluation is given in Figure 3.1. Sample stations were located at 22.9-meter (75-foot) intervals along the portion of the baseline that crossed the better part of the anomaly, and at 45.7-meter (150-foot) intervals in the "background" area. Sample stations on the cross-lines were located at 45.7-meter (150-foot) intervals. The lines were surveyed by Brunton compass and surveyor's chain, and pickets were used to mark the stations at 45.7-meter (150-foot) intervals

A 19.2-centimeter (7.5-inch) diameter post-hole auger was used

- 5 contours - ppm U
 • - survey stations
 W-W' - profile lines for presentation of results
 CL No. 1 - cross-line number
 BL 0 + 50 ft - baseline, with station number
- 0 feet 600
 0 meters 200

Figure 3.1 Location of baseline and cross-lines at the South March uranium-copper occurrence, with contour lines showing distribution of radiometrically-determined uranium (adapted from Ford, 1975)

to dig a cylindrical hole to bedrock at each station. The average hole was about 30 centimeters in depth. All but two holes reached bedrock, and these holes were terminated at a depth of 55 centimeters. It was not known whether sample depth and soil-profile properties would affect the results of the laboratory analyses. Accordingly, soil samples were taken at different depths, at intervals of approximately 15 centimeters, and at shorter intervals whenever the soil profile changed radically. The soil samples were air-dried and stored for later use.

3.2 In Situ Collector Survey

The purpose of the in situ collector survey was to discover whether collectors exposed in a soil-air environment would collect radon decay products in a manner that would result in the delineation of the two main peaks of the uranium anomaly.

3.2.1 Method

The end of a 6-centimeter-long, 1.5-millimeter-diameter, piece of cleaned silver wire was bent into a hook and suspended from a piece of wire wedged crosswise in each post-hole. A piece of 30-centimeter-square, 0.95-centimeter-thick, plywood was placed over each hole, as a cover, and loose soil was packed around the edges, to form a reasonable seal. The following day, each wire was inserted into the scintillation chamber, and the collected radon decay-product activity was counted. Five consecutive one-minute counts were recorded, and the results, after subtracting instrument background (about two counts per minute), were averaged.

To obtain a measure of reproducibility, replicate measurements were made at three sites, following overnight exposures. At BL 0 + 425, the first trial resulted in 25 counts per minute, and the second trial resulted in 23 counts per minute. The pair of values from BL 0 + 1025 was 4 and 6, and the pair of values from BL 0 + 1250 was 6 and 6.

3.2.2 Discussion Of Results

The results of the in situ collector survey are tabulated in Appendix 4 and are presented in Figures 3.2 and 3.3 as profiles along the baseline and cross-lines respectively. At several sites the holes had filled with water, and it was impossible to insert a collector. These sites are identified by circles in Figures 3.2 and 3.3; the values associated with the circles do not represent actual readings, but are simply linear interpolations between the values at the preceding and succeeding stations. The points represented by circles are, accordingly, arbitrary, and do not necessarily represent the values that would have been obtained had it been possible to insert collectors.

The baseline profile, presented in Figure 3.2, shows two main peaks more-or-less coincident with the radiometrically-determined high areas mapped by Ford (1975). The cross-line profiles include several sites at which measurable activity was collected, but the results shown in these profiles are less dramatic than those portrayed in the baseline profile.

A few of the holes were imperfectly sealed, and the collectors exposed at these sites would probably have collected more decay product activity if better seals had been possible. Stations between

Figure 32 In Situ Collector Profile Along The Baseline

Figure 33 In Situ Collector Profiles Along The Cross-Lines

BL 0 + 2150 and BL 0 + 2900 were imperfectly sealed due to the extreme thinness of the soil cover, and station BL 0 + 500 was also imperfectly sealed, due to the presence of many large angular boulders in the soil.

At one site, BL 0 + 425, the activity on a collector was counted for two hours, using the usual one-minute counting intervals. A scrutiny of the decay curve for the two hours indicated that no decay products of ^{220}Rn had been collected, and that all of the activity was due to collected decay products of ^{222}Rn . On this basis, it has been assumed that ^{220}Rn decay products have not been collected during the survey.

The results of the in situ collector survey show that spontaneous deposition can be applied to outline an area of known uranium mineralization.

3.3 Sealed Collector Survey

After a review of the favourable results from the in situ collector survey, it was apparent that spontaneous deposition might also be used to measure radium in soil samples.

If a soil sample is placed in the bottom of a sealed container, some of the radon atoms created by decay of radium in the soil emanate into the upper air-filled portion of the container; after decay of the radon, some of the decay products are deposited on a collector suspended in the air-space. Assuming that the number of decay products deposited is proportional to the amount of radium in the soil sample, it is possible to obtain a measure of the radium concentration of the soil sample.

3.3.1 Method

Aliquots of the dried samples, collected as described in Section 1 of the present chapter, were weighed into 227-milliliter glass jars with screw tops. In most cases about 50 grams of soil were used, but, for a few low-density organic samples, smaller aliquots of sample were used. In all cases, the sample occupied between one-fifth and one-third of the volume of the jar. Two holes were drilled in the lid of each jar, and both ends of a short length of copper wire were inserted. Each end of the length of copper wire was bent into the shape of a hook. A cleaned copper collecting plate 3.8 centimeters in width by 4.5 centimeters in length, with two small holes drilled into one end, was attached to the hooks. The holes in the lid were sealed with quick-drying epoxy glue, and the lid was twisted firmly into place. A typical jar, with sample and collector in place is shown in Figure 3.4.

Samples were allowed to sit for four weeks to allow complete equilibrium between radium and radon to be approximated within the closed system. After this period, each collector was, in turn, removed from the jar, and inserted into the scintillation chamber for counting. Ten consecutive one-minute counts were taken, commencing two minutes after removal of the collector from the jar. The ten counts were averaged, and instrument background was subtracted, to obtain a numerical value representative of the activity on the collector.

It was suspected that decay products of ^{220}Rn might interfere with the results of the analysis. The apparent decay rate for

Figure 3.4 Collecting Plate Suspended Over Soil In Glass Jar For Collection Of Radon Decay Products

an assemblage of ^{220}Rn decay products is dominated by the 10.64-hour half-life of ^{212}Pb . If ^{220}Rn were present, its decay product activity should still be significant after 4.5 hours. The activity due to the ^{222}Rn decay products would, however, have decayed to negligible levels during this time. To test for this effect, some collectors were set aside after the original ten counts had been made, and were recounted after about 4.5 hours.

3.3.2 Discussion Of Results

The results of the sealed collector analyses are tabulated in Appendix 5, and are plotted in Figures 3.5 and 3.6; the horizontal scale used in the latter figures is the same as that used in Figures 3.2 and 3.3. The general pattern shown in Figures 3.5 and 3.6 is similar to that for the in situ collector survey (compare, particularly, Figure 3.2 and Figure 3.5); the two main anomalous zones are clearly represented.

The results of analyses of samples taken at different depths are plotted, and the approximate depth of each sample is indicated. Two or more data points have thus been generated at those stations at which samples from more than one depth were taken. Two profile lines have been drawn; an upper line connecting the highest values at each sample site, and a lower line connecting the lowest values at each site. An appraisal of the anomaly would generate similar conclusions, regardless of whether the upper or lower line were considered. On this basis, it appears that the anomaly can be reproduced regardless of the depth at which samples are taken from the holes.

Figure 3.5 Sealed Collector Profile Along The Baseline

Figure 36 Sealed Collector Profiles Along The Cross-Lines

Of the 75 samples analyzed by the sealed collector method, 34 were checked for ^{220}Rn . The highest number of counts from ^{220}Rn decay product activity, measured after 4.5 hours, was 3 counts per minute. If this amount is multiplied by a factor of 1.35, to obtain a rough estimate of the activity of the ^{220}Rn decay products on collectors at the time of the initial counting, the total value, 4, is obtained. The total counts at the anomalous zone are sufficiently high that the activity contributed by ^{220}Rn can be ignored.

3.4 Emanometry Readings

An emanometry survey was planned at the South March uranium-copper occurrence. Emanometry techniques are well-established, and it was hoped that the results from the emanometry survey could be used as a basis for evaluation of the in situ and sealed collector results.

3.4.1 Method

The emanometry technique was virtually identical to that described by Dyck (1969). A soil-gas sampling probe was inserted into a pilot hole produced by driving a ten-inch nail into the soil. The probe consisted of a hollow plexiglass tube. Air drawn from the soil by a manually-operated vacuum pump entered the tube, and was drawn through a drierite trap into a scintillation chamber. Samples were drawn from a depth of 20 centimeters, using five strokes of the vacuum pump. Counting was performed for ten consecutive one-minute counting intervals. Immediately after termination of the counting, the radon in the scintillation chamber and in

the air-lines was flushed out by applying ten or fifteen strokes on the vacuum pump, with the soil-gas probe exposed to atmospheric air. The counting configuration consisted of the identical components described in Chapter 2, with the exception that a component with two tubes, for air-flow into and out of the scintillation chamber, was incorporated into the cell housing cover.

The soil overlying the South March uranium-copper occurrence was extremely wet during September, October, and November, 1977, due to many days of intermittent rain. As a result it was difficult to conduct a comprehensive emanometry survey. It was found that, at the majority of stations, the necessary soil-air pumping procedure resulted in mud being drawn into the air intake line. Several attempts were made on different days. Eventually, the emanometry survey was abandoned, and it was decided to use the results of a fluorimetric analysis for uranium as a control, in lieu of emanometry. Emanometry results were, however, available for a few sites, and these allow a comparison of the technical factors involved in emanometry with those involved in the collector surveys.

3.4.2 Discussion Of Results

The ten consecutive one-minute counts recorded at station BL 0 + 1400 are typical, and are as follows: 341, 260, 253, 296, 299, 278, 289, 287, 334, and 324. The highest count was obtained during the first minute. A significant portion of this count can be attributed to interference by ^{220}Rn in the soil-air sample. In comparison, ^{220}Rn decay products were a minor source

of interference in the sealed collector survey, and did not appear to affect the in situ collector survey results.

After ^{220}Rn has completely decayed, successive counts from emanometry tend to increase, due to the decay of ^{218}Po which has been deposited as a result of the decay of ^{222}Rn in the chamber. Should ^{222}Rn be the only alpha-particle-emitting nuclide in the chamber, the successive counts would show little variation. Successive counts from collector exposures tended to decrease, due to the relatively short half-lives of the decay products, particularly that of ^{218}Po .

Instrument background counts were made before each emanometry reading. Each background count tended to be affected by the preceding readings. A typical set of background readings, obtained by a succession of one-minute counting intervals at station BL 0 + 1400 is as follows: 163, 139, 128, 119, 99, 88, 72, 69, 67, and 73. In contrast, the background at the start of the day, before the first reading, was about 3 counts per minute. The high background after the station reading was due to decay products deposited on the walls of the scintillation chamber. Successive counts from the first to seventh decreased steadily; after seven counts the counts remained nearly constant. The decay pattern was similar to those observed in the laboratory exposure of collectors in radon tanks for ten minutes. The relatively stable background after seven minutes was used for the measurements at the subsequent station. Instrument backgrounds for the collector surveys were not affected by such "memory effects". All of the decay products remained on the collectors and were removed with the collector after the counting was completed.

Results of the available emanometry readings are as follows:

<u>Station</u>	<u>Radon Activity</u> <u>(counts per minute)</u>
BL 0 + 50	63
BL 0 + 575	34
BL 0 + 950	156
BL 0 + 1025	143
BL 0 + 1250	101
BL 0 + 1400	302

To eliminate the interfering effects of ^{220}Rn , the data from each site is presented as the mean value of the number of counts above background from the sixth to tenth minutes of counting.

Results are available from only 6 of the 19 sites visited. The wetness of the ground prevented a complete survey and, as a result, the data is inadequate to allow interpretation of the emanometry response to the anomaly. The in situ collector survey was performed within a few days of the attempted emanometry survey, under nearly identical soil moisture conditions, and only 4 of the 19 stations at which emanometry was attempted failed to provide in situ collector results.

3.5 Fluorimetric Analysis

Aliquots of the 75 samples used for the sealed collector survey were analyzed for uranium by Bondar-Clegg and Company Limited, of Ottawa, using conventional fluorimetric techniques. The results were used as a basis of evaluation of the results obtained by application of the collector techniques.

3.5.1 Method

An aliquot of each sample was sieved to isolate the -80-mesh fraction. 0.1 grams of the fraction were placed in a test tube and 2 milliliters of 70% nitric acid were added. After a waiting period of one hour, the contents of the test tube were diluted to a volume of 10 milliliters. 0.1 milliliters of solution were taken from the test tube, placed in a small platinum cup, and evaporated to dryness. A sodium fluoride pellet was set in the cup, and the cup was rotated over a flame for a few minutes to fuse the residue with the pellet. Uranium fused with sodium fluoride is fluorescent, and the intensity of the fluorescence gives a measure of the uranium content. The intensity of the fluorescence was measured on a fluorimeter calibrated with a set of standards of known uranium content. The uncertainty associated with fluorimetric measurements, based on replicate analyses of internal standards by Bondar-Clegg and Company, is approximately as follows (stated in terms of relative standard deviation): 20% for samples bearing about 1 part per million U; 15% for samples bearing about 10 parts per million U; 10% for samples bearing about 50 parts per million U; and 5% for samples bearing about 100 parts per million U.

3.5.2 Discussion Of Results

The results of the fluorimetric analysis are given in Appendix 5, and are plotted in Figures 3.7 and 3.8.

The baseline profile, Figure 3.7, is similar to the profile obtained by the sealed collector analysis. The profiles along

Figure 37 Fluorimetry Profile Along The Baseline

Figure 38 Fluorimetry Profile Along The Cross-Lines

the cross-lines, however, include several sites with anomalously high values of uranium, which were not identified as anomalous by the sealed collector approach.

The cross-lines traversed a swamp, and the soil samples which gave the high fluorimetric values were greatly different in texture and composition from most of the baseline samples. The samples from the cross-lines which gave the high readings consisted of black organic muck in the form of partly and completely decomposed organic matter. Little or no mineral matter was present. Most of the baseline samples were composed largely of mineral matter. The tendency for uranium to become fixed in a reducing environment suggests that the high uranium values may be due to entrapment of uranium by organic matter.

The results of the sealed collector survey along the baseline appear to reproduce the anomaly as effectively as the results from the fluorimetric analyses.

3.6 Comparison Of Results Of Different Surveys

The baseline profiles of the results of the in situ collector, sealed collector, and fluorimetric surveys are presented together in Figure 3.9. It is interesting to note that the three profiles are essentially identical, suggesting that any one of the three techniques used could delineate the anomaly.

Of the three successful techniques, the in situ collector method is the only one which gives immediate results. A sealed collector survey could give reasonably prompt results in a field laboratory if a four-day, rather than a four-week, waiting period were used.

Figure 3.9 Comparison Of Results From In Situ And Sealed Collector Surveys With Results Of Fluorimetric Analysis

After four days the radon concentration of the air in the jars should be about one-half of that which would be expected after four weeks, and this radon concentration would probably be adequate for most purposes.

It is conceivable that in situ collectors would collect decay products from radon provided by radium decaying near the walls of the holes in the soil and from radium decaying at depths greater than could conveniently be penetrated by the post-hole auger. The sealed collectors have collected decay products from radon provided only by radium contained in the soil sample. Applying the models presented in Figure 1.4, the in situ collector technique may detect radon generated in any of the three cases, (a), (b), and (c); the sealed collector technique detects only radon created in case (c). In a dry, porous environment where radon might be expected to emanate upwards from a buried source, the in situ collector technique should be used. In other environments, radon may be capable of migrating only short distances; the long-lived radon precursors, ^{238}U , ^{234}U , ^{230}Th , and ^{226}Ra may, however, have been transported from a relatively deep source to the upper soil horizons. In the latter event, the response of radon-detecting instruments may be due largely to radon provided by nearby radium, and it may be convenient to apply the sealed collector technique.

Both the sealed collector and fluorimetric survey results readily outlined the uranium occurrence, regardless of the depths from which the samples were taken. Because of the nature of the in situ survey, depth was not controlled, but the reproduction of the two peaks of the anomaly is consistent with the results from the other two

surveys.

Of the two collector survey techniques, the results from the sealed collector analysis are particularly gratifying. The reproduction of the anomaly is excellent. The sealed collector technique was applied under conditions involving neither wet chemistry nor heating. No sample-sieving was required, and no reagents were needed. The technique was non-destructive to the samples, and collectors and jars were re-usable. A very minimum of technical skill was required to weigh the samples and to subsequently measure their activity. Background readings taken after analyzing each sample were essentially constant, indicating that no memory effects were associated with the measurements.

To the writer's knowledge, neither the in situ nor the sealed collector technique has ever been attempted before in connection with exploration geochemistry, and the quality of the results of the collector surveys compare favourably with those obtained by fluorimetry.

CHAPTER 4. CONCLUSIONS

Both laboratory and field experiments have shown that spontaneous deposition of radon decay products has potential application in uranium exploration and in monitoring mine-air and other atmospheres which contain significant amounts of radon.

The results of this thesis clearly show that solid substances exposed in a radon-bearing atmosphere collect substantial amounts of radon decay-product nuclides. The amount of decay-product activity collected depends on several factors. Greater amounts of activity are collected on larger collectors, and greater amounts of activity are collected in larger containers. The amount of activity collected increases with the duration of the exposure until approaching a maximum steady-state value after about three hours. There appears to be no significant difference in the ability of different materials to collect decay products.

Essentially all of the decay-product activity is in the form of ^{218}Po and ^{214}Pb at the time of collection. The ratio of the number of decay products collected as ^{214}Pb to the number collected as ^{218}Po is greatest in larger containers of radon-bearing air. The ratio does not depend significantly on the duration of the exposure or on the surface area of the collectors.

The application of the collection principle to delineating anomalies under field conditions was successful. Collectors exposed overnight in covered holes in the soil overlying the South March uranium-copper occurrence collected sufficient decay-product activity to allow delineation of a decay-product anomaly. In addition, collectors exposed in the air in glass jars partially filled with soil samples

from the occurrence collected sufficient decay-product activity, after a four-week exposure for generation of radon, to allow the delineation of a radium anomaly.

The results of the application of the collector techniques compared favourably with the results of an analysis of a duplicate suite of soil samples for uranium, using the conventional fluorimetric technique. The anomalies defined by the collector techniques were also consistent in form with the results of other surveys by earlier workers using accepted uranium exploration techniques.

A number of areas for further research can be suggested. Laboratory work might include:

- (i) an evaluation of the effect of collector shape on the amount of decay product activity collected; and
- (ii) a study of the effect of placing a charge on the collector to increase the amount of activity collected.

Useful field studies could involve:

- (i) an evaluation of the collector methods in other areas in which uranium mineralization occurs;
- (ii) a study of the effect of using charged collectors in conjunction with either the in situ or sealed collector techniques;
- (iii) a determination of the optimum sealed collector jar volume and shape, and the optimum soil sample size;
- (iv) an evaluation of possible improvements to the sealed collector technique by applying sample preparation techniques (sieving, leaching, crushing, etc.); and
- (v) an evaluation of the collectibility of ^{220}Rn decay

products, and further development of methods of discriminating against these nuclides.

In addition, the utility of the collector techniques might be enhanced by instrument engineering studies involving the design of an efficient method of simultaneously exposing the collectors and counting the collected activity.

Studies of the migration of radon and its precursors in various soil environments would provide a basis for deciding whether the in situ collector technique, the sealed collector technique, or both techniques should be applied during the evaluation of the uranium potential of particular terrains.

It is hoped that the results of the present work will be of value in guiding the direction of further research.

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APPENDIX 1. PHYSICAL AND CHEMICAL PROPERTIES OF RADON

Atomic mass	222.0175
Weight of one atom	368.52×10^{-24} gm
Atomic radius	2.4 Å
Atomic volume	58 Å^3
Melting point	-71 °C
Boiling point	-61.8 °C
Density of gas	9.73 gm/liter
Specific gravity of liquid	4.4 (at -62 °C)
Specific gravity of solid	4
Ionization potential	10.746 eV
Viscosity	2.13×10^{-4} poise (at 0 °C)
Solubility in water	510 cc/1000 gm
Colour	Colourless at ordinary temperatures
Compounds reported	Radon fluoride Radon clathrate
Other properties	-readily absorbed on charcoal or silica gel -brilliantly phosphorescent at freezing point

Data compiled from Green (1972), Moore (1972),
and Weast (1973).

APPENDIX 2. TABULATION OF TOTAL ACTIVITIES ON COLLECTORS,
AND COLLECTION RATIOS, FOR COLLECTORS EXPOSED
IN THE LABORATORY STUDY OF SPONTANEOUS DECAY
PRODUCT DEPOSITION

Part 1. Ten-Minute Exposures In The 29-Liter Tank

Material	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
			F ₁	F ₂
(a) <u>Metal Collectors</u>				
Silver	1.5	223	.19	.04
Silver	1.3	277	.12	-.01
Silver	1.3	272	.23	.10
Silver	1.3	347	.18	.01
Silver	9.3	362	.22	.02
Silver	9.3	348	.22	.01
Silver	9.3	393	.17	-.01
Sterling silver	1.5	255	.13	-.01
Sterling silver	2.9	295	.13	-.02
Gold-filled silver	1.9	238	.15	.03
Copper	1.4	153	.11	.07
Copper	9.6	446	.27	.08
Copper	9.6	435	.14	-.05
Brass	1.4	570	.16	.04
Platinum	1.6	168	.13	.04
Aluminum	7.4	384	.13	-.04
Aluminum	7.4	352	.12	.03

Note: The number of digits presented in this appendix
has arithmetic significance only.

Material	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
			F ₁	F ₂

(b) Plastic Collectors

Plexiglass	10.5	812	.23	.03
Lexan	5.0	490	.20	.01
Lexan	5.0	480	.23	.03
Lexan	5.0	463	.25	.04
Lexan	7.1	334	.20	.03
Lexan	9.7	478	.36	.06
Cellulose nitrate	8.3	737	.26	.04
Mylar	7.7	488	.22	.00

(c) Foil Collectors

Silver foil	13.1	1857	.29	.02
Brass foil	10.5	1482	.14	.00
Brass foil	12.0	1255	.31	.13
Aluminum foil	5.7	341	.15	.03
Aluminum foil	9.5	523	.20	.02
Kimfol	7.6	744	.19	.05

(d) Miscellaneous Collectors

Aluminized Mylar	7.3	731	.30	.04
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Material	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
			F ₁	F ₂

(b) Plastic Collectors

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Lexan	5.0	490	.20	.01
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Lexan	5.0	463	.25	.04
Lexan	7.1	334	.20	.03
Lexan	9.7	478	.36	.06
Cellulose nitrate	8.3	737	.26	.04
Mylar	7.7	488	.22	.00

(c) Foil Collectors

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Brass foil	10.5	1482	.14	.00
Brass foil	12.0	1255	.31	.13
Aluminum foil	5.7	341	.15	.03
Aluminum foil	9.5	523	.20	.02
Kimfol	7.6	744	.19	.05

(d) Miscellaneous Collectors

Aluminized Mylar	7.3	731	.30	.04
---------------------	-----	-----	-----	-----

Part 2. One-Hour Exposures In The 29-Liter Tank

Material	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
			F ₁	F ₂
Silver	1.3	1131	.14	.01

Part 3. Steady-State Exposures In The 29-Liter Tank

Material	Exposure Time (hours)	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
				F ₁	F ₂
(a) Metal Collectors					
Silver	17.8	1.3	2160	.17	.01
Silver	22.8	1.3	2211	.28	.02
Silver	74.6	1.3	2143	.54	.17
Silver	3.0	1.3	1828	.23	.04
Silver	22.6	1.3	1958	.16	.03
Silver	3.0	1.3	1748	.10	.02
Silver	3.0	1.3	1389	.01	-.04
Silver	21.0	1.3	2460	.12	-.01
Sterling silver	20.7	1.1	2066	.08	-.04
Copper	20.7	1.4	2694	.01	-.02
Copper	19.8	1.4	2028	.48	.13
Brass	30.6	1.4	2793	.03	.01
Platinum	19.0	1.6	2555	.15	.00
Gold	19.6	0.3	1753	.04	-.03
Aluminum	18.7	6.8	3301	.11	.00
(b) Plastic Collectors					
Plexiglass	21.3	10.2	4105	.18	.01
Plexiglass	20.8	10.2	2618	.23	.17
Plexiglass	21.3	10.2	3126	.06	-.01
Plexiglass	22.3	10.2	2973	.08	-.02
Plexiglass	23.0	10.2	3581	.25	.06
Plexiglass	3.0	10.2	2312	.10	-.02
Plexiglass	25.5	11.3	3213	.10	.03
Plexiglass	3.0	12.0	4046	.07	-.02
Lexan	17.3	5.0	2747	.12	-.01
Lexan	18.6	5.0	2842	.21	.00
Lexan	3.0	5.0	3140	.25	.08
Lexan	3.0	5.0	1878	.10	.03
Lexan	19.6	5.0	1476	.09	.04

Material	Exposure Time (hours)	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
				F ₁	F ₂
(b) <u>Plastic Collectors</u> (continued)					
Cellulose nitrate	18.3	5.6	3119	.13	.01
Mylar	23.0	6.4	5672	.16	-.02
(c) <u>Foil Collectors</u>					
Silver foil	23.4	7.5	3467	.39	.17
Silver foil	49.6	11.3	3582	.09	.00
Silver foil	19.8	13.0	4535	.20	.04
Brass foil	23.9	6.4	2782	.08	.01
Brass foil	26.3	6.4	2577	.09	-.01
Brass foil	28.2	6.4	3388	.13	.04
Aluminum foil	50.4	2.4	1190	.05	-.06
Aluminum foil	20.7	9.5	3461	.08	-.01
Kimfol	23.2	7.6	1576	.05	-.04
Kimfol	65.4	7.6	2179	.04	-.06
Kimfol	18.6	7.6	1637	.05	-.02

Part 4. Steady-State Exposures In The 29-Liter Tank
(Surface-Area Study)

Material	Exposure Time (hours)	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
				F ₁	F ₂
Copper	69.6	1.0	997	.11	.04
Copper	70.5	2.0	2766	.06	.03
Copper	71.3	4.1	3590	.14	.02
Copper	72.2	8.1	2857	.03	.04
Copper	73.2	16.1	4742	.04	.00
Copper	73.6	34.3	12972	.10	.00
Silver	3.4	1.0	932	.06	.04
Silver	4.2	2.0	2377	.12	.00
Silver	5.1	8.1	2714	.10	.00
Lexan	66.1	1.0	428	.25	.04
Lexan	67.1	2.2	1030	.01	.06
Lexan	68.8	4.5	1336	.07	.01
Lexan	69.5	9.8	1964	.05	.01
Lexan	70.7	19.22	4598	.08	.01
Plexiglass	43.8	3.7	1122	.13	.09
Plexiglass	45.3	5.6	2410	.10	.03
Plexiglass	19.5	11.3	2077	.03	.00
Plexiglass	20.5	19.0	3088	.06	.01

Part 5. Ten-Minute Exposures In The 70-Milliliter And
400-Milliliter Tanks

Material	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
			F ₁	F ₂
<u>(a) Exposures In 400-Milliliter Tank</u>				
Plexiglass	10.2	122	.09	.06
Lexan	4.5	135	.14	.00
<u>(b) Exposures In 70-Milliliter Tank</u>				
Silver	1.3	47	.01	.05
Silver	1.3	42	.11	.08
Plexiglass	12.0	8	.04	.05
Lexan	7.0	76	.06	.03

Part 6. One-Hour Exposures In The 70-Milliliter And
400-Milliliter Tanks

Material	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
			F ₁	F ₂
(a) <u>Exposures In 400-Milliliter Tank</u>				
Silver	1.3	127	.06	-.03
Lexan	7.2	224	.10	.02
Plexiglass	12.0	258	.06	.02
(b) <u>Exposures In 70-Milliliter Tank</u>				
Lexan	7.2	42	-.02	-.03
Plexiglass	12.0	82	-.01	-.01

Part 7. Steady-State Exposures In The 70-Milliliter And
400-Milliliter Tanks

Material	Exposure Time (hours)	Surface Area (cm ²)	Total Activity On Collector At 30 Minutes After Termination Of Exposure (Disintegrations per minute)	Collection Ratios	
				F ₁	F ₂
(a) <u>Exposures In 400-Milliliter Tank</u>					
Silver	18.2	1.3	156	.06	.00
Plexiglass	22.6	12.0	352	.01	-.04
Lexan	18.3	7.2	320	-.02	-.01
(b) <u>Exposures In 70-Milliliter Tank</u>					
Silver	19.4	1.3	42	-.23	-.13
Plexiglass	24.6	10.2	113	.08	.07
Lexan	18.6	7.0	131	.03	-.10

APPENDIX 3. DERIVATION OF A MATHEMATICAL EXPRESSION FOR THE
COLLECTION RATIOS, F_1 AND F_2 , IN TERMS OF
OBSERVED COUNTING RATIOS

Rutherford (1913) has provided equations which may be applied to estimate the relative proportions of decay products that become attached to a collector as ^{214}Pb and ^{214}Bi with respect to the number of decay products collected as ^{218}Po . To apply these equations it is necessary to assume that the rate of deposition of each of the three species is constant with time.

Let us assume that n_0 atoms of ^{218}Po become attached every minute. The number of atoms of ^{214}Pb that become attached can be expressed as $\frac{\lambda_1}{\lambda_2} \times F_1 \times n_0$,

where λ_1 is the decay constant for ^{218}Po (.22726 min^{-1}),

λ_2 is the decay constant for ^{214}Pb (.02586 min^{-1}), and

F_1 is the fraction of the radioactive-equilibrium proportion of ^{214}Pb atoms collected with respect to n_0 atoms of ^{218}Po .

Similarly, the number of atoms of ^{214}Bi that become attached is $\frac{\lambda_1}{\lambda_3} \times F_2 \times n_0$,

where λ_3 is the decay constant for ^{214}Bi (.03519 min^{-1}), and

F_2 is the fraction of the radioactive-equilibrium proportion of ^{214}Bi atoms collected with respect to n_0 atoms of ^{218}Po .

Assuming that only ^{218}Po is collected, and that collection proceeds at the rate of n_0 atoms per minute, the number of atoms of ^{218}Po , ^{214}Pb , and ^{214}Bi on a collector, after an

exposure of duration T is given, using Rutherford's case 4,

as:

$$N(\text{Po}, \text{Po}) = \frac{n_0}{\lambda_1} \times (1 - e^{-\lambda_1 T}), \quad \text{A3.1}$$

$$N(\text{Pb}, \text{Po}) = \frac{n_0}{\lambda_2} \times (1 - (\lambda_1 e^{-\lambda_2 T} - \lambda_2 e^{-\lambda_1 T}) / (\lambda_1 - \lambda_2)), \quad \text{A3.2}$$

and

$$N(\text{Bi}, \text{Po}) = \frac{n_0}{\lambda_3} \times (1 - \lambda_3 (a e^{-\lambda_1 T} + b e^{-\lambda_2 T} + c e^{-\lambda_3 T})), \quad \text{A3.3}$$

where $N(\text{Po}, \text{Po})$ is the number of atoms of ^{218}Po at

time T which, when collected, were

in the form of ^{218}Po ,

$N(\text{Pb}, \text{Po})$ is the number of atoms of ^{214}Pb at

time T which, when collected, were

in the form of ^{218}Po ,

$N(\text{Bi}, \text{Po})$ is the number of atoms of ^{214}Bi at

time T which, when collected, were

in the form of ^{218}Po ,

$$a = \frac{\lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}$$

$$b = \frac{\lambda_1}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)}$$

$$c = \frac{\lambda_1 \lambda_2}{\lambda_3 (\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}$$

and

Similarly, assuming that $\frac{\lambda_1}{\lambda_2} \times F_1 \times n_0$ atoms of ^{214}Pb are collected each minute, the number of atoms of ^{214}Pb and ^{214}Bi from this source, after an exposure of duration T , is given by:

$$N(\text{Pb}, \text{Pb}) = F_1 \times \frac{\lambda_1}{\lambda_2} \times \frac{n_0}{\lambda_2} \times (1 - e^{-\lambda_2 T}), \quad \text{and} \quad \text{A3.4}$$

$$N(\text{Bi}, \text{Pb}) = F_1 \times \frac{\lambda_1}{\lambda_2} \times \frac{n_0}{\lambda_3} \times (1 - (\lambda_2 e^{-\lambda_3 T} - \lambda_3 e^{-\lambda_2 T}) / (\lambda_2 - \lambda_3)) \quad \text{A3.5}$$

where $N(\text{Pb}, \text{Pb})$ is the number of atoms of ^{214}Pb at

time T which, when collected, were

in the form of ^{214}Pb , and

$N(\text{Bi}, \text{Pb})$ is the number of atoms of ^{214}Bi at time T which, when collected, were in the form of ^{214}Pb .

Again, assuming that $\frac{\lambda_1}{\lambda_3} \times E_2 \times n_0$ atoms of ^{214}Bi are collected each minute, the number of atoms of ^{214}Bi from this source after an exposure of duration T , is given by:

$$N(\text{Bi}, \text{Bi}) = E_2 \times \frac{\lambda_1}{\lambda_3} \times \frac{n_0}{\lambda_3} \times (1 - e^{-\lambda_3 T}), \quad \text{A3.6}$$

where $N(\text{Bi}, \text{Bi})$ is the number of atoms of ^{214}Bi at time T which, when collected, were in the form of ^{214}Bi .

Collection terminates at time T , and we wish to find the activity on the collector at any subsequent time t . Of the three decay products considered, only ^{218}Po emits alpha particles. ^{214}Bi , however, decays to become ^{214}Po , an alpha-emitting nuclide with a half-life so short (1.64×10^{-4} seconds) that secular equilibrium with ^{214}Bi can be conveniently assumed.

For this reason, no significant error is introduced by assuming, for this mathematical argument, that ^{214}Bi is an alpha-emitter. Decay of ^{214}Po produces ^{210}Pb , a beta-emitter with a long half-life (19.4 years). Hence we are concerned with two, and only two, alpha emitters, ^{218}Po and ^{214}Po .

At time T , the total number of atoms of each of the three species is as follows:

$$\Sigma \text{Po} = N(\text{Po}, \text{Po}) \quad \text{A3.7}$$

$$\Sigma \text{Pb} = N(\text{Pb}, \text{Po}) + N(\text{Pb}, \text{Pb}) \quad \text{A3.8}$$

$$\Sigma \text{Bi} = N(\text{Bi}, \text{Po}) + N(\text{Bi}, \text{Pb}) + N(\text{Bi}, \text{Bi}) \quad \text{A3.9}$$

It is now convenient to make some substitutions.

$$\text{Let } X = \frac{(1 - e^{-\lambda_1 T})}{\lambda_1} \quad \text{A3.10}$$

$$Y = \frac{1}{\lambda_2} \times (1 - (\lambda_1 e^{-\lambda_2 T} - \lambda_2 e^{-\lambda_1 T}) / (\lambda_1 - \lambda_2)) \quad \text{A3.11}$$

$$Y_1 = \frac{\lambda_1}{\lambda_2} \times \frac{1}{\lambda_2} \times (1 - e^{-\lambda_2 T}) \quad \text{A3.12}$$

$$Z = \frac{1}{\lambda_3} \times (1 - \lambda_3 (a e^{-\lambda_1 T} + b e^{-\lambda_2 T} + c e^{-\lambda_3 T})) \quad \text{A3.13}$$

$$Z_1 = \frac{\lambda_1}{\lambda_2} \times \frac{1}{\lambda_3} \times (1 - (\lambda_2 e^{-\lambda_3 T} - \lambda_3 e^{-\lambda_2 T}) / (\lambda_2 - \lambda_3)) \quad \text{A3.14}$$

$$\text{and } Z_2 = \frac{\lambda_1}{\lambda_3} \times \frac{1}{\lambda_3} \times (1 - e^{-\lambda_3 T}) \quad \text{A3.15}$$

We may now rewrite A3.7, A3.8, and A3.9 as:

$$\Sigma \text{Po} = n_0 X \quad \text{A3.16}$$

$$\Sigma \text{Pb} = n_0 (Y + (F_1 \times Y_1)) \quad \text{A3.17}$$

$$\Sigma \text{Bi} = n_0 (Z + (F_1 \times Z_1) + (F_2 \times Z_2)) \quad \text{A3.18}$$

to yield expressions in terms of F_1 and F_2 .

From Rutherford's case 1, the number of atoms of ^{218}Po at time t is given as:

$$N(\text{Po}, \Sigma \text{Po}) = \Sigma \text{Po} \times e^{-\lambda_1 t} \quad \text{A3.19}$$

The number of atoms of ^{214}Bi derived from atoms which, at $t = 0$, were in the form of ^{218}Po is:

$$N(\text{Bi}, \Sigma \text{Po}) = \Sigma \text{Po} \times (a e^{-\lambda_1 t} + b e^{-\lambda_2 t} + c e^{-\lambda_3 t}) \quad \text{A3.20}$$

$$\text{where } a_1 = \frac{\lambda_1 \lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}$$

$$b_1 = \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \quad , \text{ and}$$

$$c_1 = \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}$$

The number of atoms of ^{214}Bi derived from atoms which, at $t = 0$, were in the form of ^{214}Pb is:

$$N(\text{Bi}, \sum \text{Pb}) = \sum \text{Pb} \times \frac{\lambda_2}{(\lambda_3 - \lambda_2)} \times (e^{-\lambda_2 t} - e^{-\lambda_3 t}). \quad \text{A3.21}$$

The number of atoms of ^{214}Bi remaining from atoms which, at $t = 0$, were in the form of ^{214}Bi is:

$$N(\text{Bi}, \sum \text{Bi}) = \sum \text{Bi} \times e^{-\lambda_3 t}. \quad \text{A3.22}$$

The activities of each group at time t are:

$$A(\text{Po}, \sum \text{Po}) = \sum \text{Po} \times \lambda_1 \times e^{-\lambda_1 t}, \quad \text{A3.23}$$

$$A(\text{Bi}, \sum \text{Po}) = \sum \text{Po} \times \lambda_3 \times (a e^{-\lambda_1 t} + b e^{-\lambda_2 t} + c e^{-\lambda_3 t}), \quad \text{A3.24}$$

$$A(\text{Bi}, \sum \text{Pb}) = \sum \text{Pb} \times \lambda_3 \times \frac{\lambda_2}{(\lambda_3 - \lambda_2)} \times (e^{-\lambda_2 t} - e^{-\lambda_3 t}), \text{ and} \quad \text{A3.25}$$

$$A(\text{Bi}, \sum \text{Bi}) = \sum \text{Bi} \times \lambda_3 \times e^{-\lambda_3 t}. \quad \text{A3.26}$$

The total activity, $\sum A$, at time t is obtained by summing equations A3.23 to A3.26. Accordingly:

$$\begin{aligned} \sum A = & \left[\sum \text{Po} \times \lambda_1 \times e^{-\lambda_1 t} \right] \\ & + \left[\sum \text{Po} \times \lambda_3 \times (a e^{-\lambda_1 t} + b e^{-\lambda_2 t} + c e^{-\lambda_3 t}) \right] \\ & + \left[\sum \text{Pb} \times \lambda_3 \times \frac{\lambda_2}{(\lambda_3 - \lambda_2)} \times (e^{-\lambda_2 t} - e^{-\lambda_3 t}) \right] \\ & + \left[\sum \text{Bi} \times \lambda_3 \times e^{-\lambda_3 t} \right]. \end{aligned} \quad \text{A3.27}$$

Rearranging the right side of A3.27 yields:

$$\begin{aligned} \sum A = & \left[e^{-\lambda_1 t} \times ((\sum \text{Po} \times \lambda_1) + (\sum \text{Po} \times \lambda_3 \times a)) \right] \\ & + \left[e^{-\lambda_2 t} \times ((\sum \text{Po} \times \lambda_3 \times b) + (\sum \text{Pb} \times \lambda_3 \times \frac{\lambda_2}{(\lambda_3 - \lambda_2)})) \right] \\ & + \left[e^{-\lambda_3 t} \times ((\sum \text{Po} \times \lambda_3 \times c) - \sum \text{Pb} \times \lambda_3 \times \frac{\lambda_2}{(\lambda_3 - \lambda_2)} + (\sum \text{Bi} \times \lambda_3)) \right]. \end{aligned} \quad \text{A3.28}$$

Equation A3.28 may be integrated between t_1 and t_2 , to obtain the total expected number of counts between $t = t_1$ and $t = t_2$, as follows:

$$\begin{aligned} \sum_{t_1}^{t_2} A = & \left\{ \frac{e^{-\lambda_1 t_2} - e^{-\lambda_1 t_1}}{-\lambda_1} \times \left[\left(\sum P_o \times \lambda_1 \right) + \left(\sum P_o \times \lambda_3 \times a_1 \right) \right] \right\} \\ & + \left\{ \frac{e^{-\lambda_2 t_2} - e^{-\lambda_2 t_1}}{-\lambda_2} \times \left[\left(\sum P_o \times \lambda_3 \times b_1 \right) \right. \right. \\ & \quad \left. \left. + \left(\sum P_b \times \lambda_3 \times \frac{\lambda_2}{(\lambda_3 - \lambda_2)} \right) \right] \right\} \\ & + \left\{ \frac{e^{-\lambda_3 t_2} - e^{-\lambda_3 t_1}}{-\lambda_3} \times \left[\left(\sum P_o \times \lambda_3 \times c_1 \right) \right. \right. \\ & \quad \left. \left. - \left(\sum P_b \times \lambda_3 \times \frac{\lambda_2}{(\lambda_3 - \lambda_2)} \right) \right. \right. \\ & \quad \left. \left. + \left(\sum B_i \times \lambda_3 \right) \right] \right\} \end{aligned} \quad A3.29$$

To simplify A3.29

$$\begin{aligned} \text{let } E_1 &= \frac{e^{-\lambda_1 t_2} - e^{-\lambda_1 t_1}}{-\lambda_1}, \\ E_2 &= \frac{e^{-\lambda_2 t_2} - e^{-\lambda_2 t_1}}{-\lambda_2}, \text{ and} \\ E_3 &= \frac{e^{-\lambda_3 t_2} - e^{-\lambda_3 t_1}}{-\lambda_3}, \end{aligned}$$

and let $\lambda_4 = \lambda_3 \times a_1$,
 $\lambda_5 = \lambda_3 \times b_1$,
 $\lambda_6 = \frac{\lambda_3 \times \lambda_2}{\lambda_3 - \lambda_2}$, and
 $\lambda_7 = \lambda_3 \times c_1$, so that

$$\begin{aligned} \sum_{t_1}^{t_2} A = & \left\{ E_1 \times \left[\left(\sum P_o \times \lambda_1 \right) + \left(\sum P_o \times \lambda_4 \right) \right] \right\} \\ & + \left\{ E_2 \times \left[\left(\sum P_o \times \lambda_5 \right) + \left(\sum P_b \times \lambda_6 \right) \right] \right\} \\ & + \left\{ E_3 \times \left[\left(\sum P_o \times \lambda_7 \right) - \left(\sum P_b \times \lambda_6 \right) + \left(\sum B_i \times \lambda_3 \right) \right] \right\} \end{aligned} \quad A3.30$$

To obtain $\sum_{t_1}^{t_2} A$ as a function of F_1 and F_2 , substitute the expressions for $\sum P_0$, $\sum P_b$, and $\sum B_i$ from A3.16, A3.17, and A3.18, respectively, into A3.30, so that

$$\begin{aligned} \sum_{t_1}^{t_2} A = & \left\{ E_1 \times [(n_0 \times \lambda_1) + (n_0 \times \lambda_4)] \right\} \\ & + \left\{ E_2 \times [(n_0 \times \lambda_5) + ((n_0(Y + (F_1 \times Y_1))) \times \lambda_6)] \right\} \\ & + \left\{ E_3 \times [(n_0 \times \lambda_7) - ((n_0(Y + (F_1 \times Y_1))) \times \lambda_6) \right. \\ & \quad \left. + ((n_0(Z + (F_1 \times Z_1)) \right. \\ & \quad \left. + (F_2 \times Z_2)) \times \lambda_3)] \right\} \quad \text{A3.31} \end{aligned}$$

To simplify,

$$\begin{aligned} \text{let } G = & E_1 \times (X \times \lambda_1) + (X \times \lambda_4) + (E_2 \times (X \times \lambda_5)) \\ & + (E_3 \times (X \times \lambda_7)), \end{aligned}$$

and substitute into A3.31, so that;

$$\begin{aligned} \sum_{t_1}^{t_2} A = & n_0 \times \left\{ G + [E_2 \times ((Y \times \lambda_6) + (F_1 \times Y_1 \times \lambda_6))] \right. \\ & - [E_3 \times ((Y \times \lambda_6) + (F_1 \times Y_1 \times \lambda_6))] \\ & + [E_3 \times ((Z \times \lambda_3) + (F_1 \times Z_1 \times \lambda_3) \\ & \quad \left. + (F_2 \times Z_2 \times \lambda_3))] \right\} \quad \text{A3.32} \end{aligned}$$

A3.32 may be rearranged, to give:

$$\begin{aligned} \sum_{t_1}^{t_2} A = & n_0 \times \left\{ [G + (E_2 \times Y \times \lambda_6) - (E_3 \times Y \times \lambda_6) \right. \\ & \quad \left. + (E_3 \times Z \times \lambda_3)] \right. \\ & + (F_1 \times [(E_2 \times Y_1 \times \lambda_6) - (E_3 \times Y_1 \times \lambda_6) \\ & \quad \left. + (E_3 \times Z_1 \times \lambda_3)]) \right. \\ & \left. + (F_2 \times [E_3 \times Z_2 \times \lambda_3]) \right\} \quad \text{A3.33} \end{aligned}$$

A3.33 may be simplified by letting:

$$H = G + (E_2 \times Y \times \lambda_0) - (E_3 \times Y \times \lambda_0) + (E_3 \times Z \times \lambda_3)$$

$$L = (E_2 \times Y_1 \times \lambda_0) - (E_3 \times Y_1 \times \lambda_0) + (E_3 \times Z_1 \times \lambda_3); \text{ and}$$

$$M = (E_3 \times Z_2 \times \lambda_3),$$

yielding

$$\sum A = n_0 \times (H + (F_1 \times L) + (F_2 \times M)). \quad A3.34$$

The activity expected during three different counting intervals may be expressed by selecting three sets of starting and ending time-points: t_1' and t_2' ; t_1'' and t_2'' ; and t_1''' and t_2''' ; and evaluating A3.34 for each of the three intervals.

For the first counting interval, from t_1' to t_2' ,

define $H' = H$; $L' = L$; and $M' = M$.

For the second counting interval, from t_1'' to t_2'' ,

define $H'' = H$; $L'' = L$; and $M'' = M$.

For the third counting interval, from t_1''' to t_2''' ,

define $H''' = H$; $L''' = L$; and $M''' = M$.

Then, using the appropriate values for t_1 and t_2 in each case,

$$\text{let } \sum A' = n_0 \times [H' + (F_1 \times L') + (F_2 \times M')], \quad A3.35$$

$$\sum A'' = n_0 \times [H'' + (F_1 \times L'') + (F_2 \times M'')], \text{ and } A3.36$$

$$\sum A''' = n_0 \times [H''' + (F_1 \times L''') + (F_2 \times M''')]. \quad A3.37$$

In this way,

$\sum A'$ represents the total number of expected counts during the first finite counting interval, from t_1' to t_2' ,

$\sum A''$ represents the total number of expected counts during the second finite counting interval, from t_1'' to t_2'' , and

$\sum A'''$ represents the total number of expected counts during the third finite counting interval, from t_1''' to t_2''' .

Now,

let $R_1 = \frac{\sum A'''}{\sum A''}$, and $R_2 = \frac{\sum A'''}{\sum A''}$.

The values R_1 and R_2 represent the ratios of the expected number of counts over two different counting intervals to the expected number of counts at a third counting interval. Writing R_1 and R_2 in terms of the expressions for A'' , A''' , and A'''' yields:

$$R_1 = \frac{H''' + (F_1 \times L''') + (F_2 \times M''')}{H'' + (F_1 \times L'') + (F_2 \times M'')}, \text{ and} \quad A3.38$$

$$R_2 = \frac{H''' + (F_1 \times L''') + (F_2 \times M''')}{H'' + (F_1 \times L'') + (F_2 \times M'')}. \quad A3.39$$

A3.38 and A3.39 may be rewritten, respectively, as:

$$F_2 \times (R_1 M' - M''') = (F_1 \times (L''' - R_1 L'')) + (H''' - R_1 H''), \text{ and} \quad A3.40$$

$$F_2 \times (R_2 M' - M''') = (F_1 \times (L''' - R_2 L'')) + (H''' - R_2 H''). \quad A3.41$$

A3.40 and A3.41 may be simplified by letting:

$$N_1 = R_1 M' - M''',$$

$$N_2 = L''' - R_1 L'',$$

$$N_3 = H''' - R_1 H',$$

$$N_4 = R_2 M' - M'',$$

$$N_5 = L'' - R_2 L', \text{ and}$$

$$N_6 = H'' - R_2 H',$$

so that;

$$F_2 \times N_1 = (F_1 \times N_2) + N_3, \text{ and} \quad \text{A3.42}$$

$$F_2 \times N_4 = (F_1 \times N_5) + N_6. \quad \text{A3.43}$$

A3.42 and A3.43 represent two equations with two unknowns,

F_1 and F_2 . The equations may be solved for F_1 and F_2 to yield:

$$F_1 = \frac{(N_6 \times N_1) - (N_3 \times N_4)}{(N_2 \times N_4) - (N_5 \times N_1)}, \text{ and}$$

$$F_2 = \frac{(F_1 \times N_5) + N_6}{N_4}.$$

This gives F_1 and F_2 in terms of variables which can themselves be expressed in terms of $\lambda_1, \lambda_2, \lambda_3, T, t_1', t_2', t_1'', t_2'', t_1''', t_2'''$, R_1 and R_2 . R_1 and R_2 are known from observation, and the other variables are determined from the parameters of the decay series, exposure durations, and counting intervals. Hence, the values of the collection ratios, F_1 and F_2 , can be determined.

*APPENDIX 4. TABULATION OF RESULTS OF IN SITU COLLECTOR SURVEY

<u>Profile End-Point Designation</u>	<u>Station</u>	<u>Activity On Collector (counts per minute)</u>
W - W'	BL 0 + 50	1.4
	125	0.0
	200	0.0
	275	no reading
	350	no reading
	425	26.9
	500	0.0
	575	3.6
	650	0.0
	725	1.8
	800	no reading
	875	2.0
	950	1.2
	1025	7.0
	1100	5.1
	1175	1.7
	1250	7.6
	1325	no reading
	1400	1.2
	1475	3.2
	1550	1.2
	1625	no reading
	1700	0.0
	1775	0.4
	1850	no reading
	1925	0.8
	2000	0.2
	2075	0.0
	2150	no reading
	2300	0.0
	2450	0.0
	2600	0.0
	2750	0.0
	2900	0.0
X - X'	CL No. 1, 1 + 0 SE	1.8
	3 + 00 SE	1.0
	4 + 50 SE	no reading
	6 + 00 SE	no reading
Y - Y'	CL No. 2, 1 + 50 SE	1.4
	3 + 00 SE	0.6
	4 + 50 SE	no reading

Note: The number of digits presented in this appendix has arithmetic significance only.

Profile
End-Point
Designation

Station

Activity
On
Collector
(counts per minute)

2 - Z'

CL No. 3, 1 + 50 SE
3 + 00 SE
4 + 30 SE

no reading
0.2
0.0

APPENDIX 5.. TABULATION OF RESULTS OF SEALED COLLECTOR AND
FLUORIMETRIC ANALYSES

Profile End-Point Design- ation	Station	Sample Number	Depth (cm)	Sealed Collector Results -Activity On Collector (counts per minute)	Fluorimetric Results (parts per million U)
W - W'	BL 0 +				
		50 SM 1	15	8.5	3.0
		125 2	15	9.9	1.3
		200 3	15	5.9	6.6
		200 4	30	4.5	1.3
		200 5	46	0.1	0.7
		275 6	5	6.8	6.7
		275 7	20	2.6	0.9
		275 8	36	2.3	1.0
		275 9	51	3.0	1.0
		350 10	8	11.5	60.0
		350 11	23	4.2	2.4
		425 12	8	80.4	28.0
		425 13	23	53.2	24.0
		425 14	38	48.0	21.0
		500 15	15	43.6	23.0
		500 16	30	87.3	40.0
		575 17	15	13.1	2.1
		650 18	15	12.6	7.0
		725 19	15	4.4	1.6
		800 20	8	10.9	10.2
		800 21	23	6.5	14.8
		875 22	15	10.9	5.0
		950 23	15	17.2	5.5
		950 24	30	9.7	5.9
		1025 25	15	49.2	12.6
		1025 26	30	46.5	13.0
		1100 27	15	42.8	14.8
		1175 28	15	57.5	16.2
		1250 29	15	51.8	20.5
		1325 30	15	58.0	15.8
		1400 31	15	37.5	23.0
		1400 32	30	57.1	18.0
		1475 33	15	14.3	5.0
		1550 34	15	4.2	1.3
		1550 35	30	4.5	1.6
		1625 36	15	4.8	4.9
		1625 37	30	4.5	2.9
		1700 38	13	12.6	25.0
		1700 39	30	5.5	9.0

Note: The number of digits presented in this appendix
has arithmetic significance only.

Profile End-Point Designation	Station	Sample Number	Depth (cm)	Sealed Collector Results -Activity On Collector (counts per minute)	Fluorimetric Results (parts per million U)
W - W'	BL 0 +				
		1775	SM 40 15	23.4	5.8
		1775	41 30	14.4	2.9
		1850	42 8	3.4	2.4
		1850	43 23	8.5	1.9
		1850	44 38	11.7	1.3
		1925	45 15	1.1	0.3
		2000	46 15	1.2	0.1
		2075	47 15	1.1	0.0
		2150	48 15	4.6	2.2
		2300	49 8	1.9	1.5
		2450	50 8	4.1	1.0
		2600	51 13	1.6	0.4
		2750	52 15	3.8	1.6
		2900	53 15	1.9	1.3
X - X'	CL No. 1,				
	1 + 50 SE	54	13	30.4	21.0
	1 + 50 SE	55	28	10.7	2.1
	1 + 50 SE	56	43	5.5	1.3
	3 + 00 SE	57	15	1.8	47.0
	3 + 00 SE	58	30	1.9	65.0
	3 + 00 SE	59	46	7.7	111.0
	4 + 50 SE	60	15	6.4	9.6
	4 + 50 SE	61	30	4.9	7.6
	6 + 00 SE	62	15	2.5	0.6
	6 + 00 SE	63	33	2.7	1.0
	6 + 00 SE	64	48	4.0	1.1
Y - Y'	CL No. 2,				
	1 + 50 SE	65	10	6.8	2.1
	1 + 50 SE	66	25	3.1	1.6
	3 + 00 SE	67	10	2.4	21.0
	3 + 00 SE	68	28	3.6	1.0
	3 + 00 SE	69	46	3.7	0.7
	4 + 50 SE	70	10	6.8	3.6
Z - Z'	CL No. 3,				
	1 + 50 SE	71	8	10.2	23.0
	1 + 50 SE	72	23	6.9	10.5
	1 + 50 SE	73	38	3.3	1.7
	3 + 00 SE	74	15	3.4	4.5
	4 + 30 SE	75	15	3.8	1.5

END

1 6 [REDACTED] [REDACTED] 7 9

FIN