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Field and Laboratory Methods used by the
Geological Survey of Canada in Geochemical Surveys
No. 10. RADON DETERMINATION APPARATUS
FOR GEOCHEMICAL PROSPECTING FOR URANIUM

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DEPARTMENT OF ENERGY, MINES AND RESOURCES

CONTENTS

Abstract	Page v
Introduction	1
Determination of radon in natural waters	2
Description of apparatus	2
Calibration and instrument performance data	5
On-site radon determinations of soil emanations	11
References	15
Appendix I. Tables 1-8	17
Appendix II. Procedure for measuring radon 222 in water samples	27

Illustrations

Figure 1. Details of the water gas cell used for the determination of radon in water	3
Figure 2. Radon extraction and cell filling line	4
Figure 3. Schematic of alpha counter	5
Figure 4. Photograph of radon detector equipped for on- site alpha activity measurements of soil gas emanations	6
Figure 5. Plot of alpha activity from a pure radon-air sample in the water gas cell as a function of time	7
Figure 6. Loss of radon from water in 1-litre plastic bottles	9
Figure 7. Typical plateau of the radon alpha activity obtained with the water gas cell and electronics described in this report	11
Figure 8. Cross-sectional view of soil gas cell and light cap in place above photomultiplier	12
Figure 9. Plot of alpha activity as a function of time obtained with the soil gas cell	13

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ABSTRACT

A radon detection instrument was developed comprising a portable nuclear scaler-timer, a photomultiplier, alpha-sensitive cells made with silver-activated zinc sulphide, and a simple vacuum system. The instrument is suitable for the absolute determination of radon in 30 to 40 water samples per day with a practical lower detection limit of 0.25 picocuries. Evacuated 130 ml cells are filled with air bubbled through 130 ml of sample and the alpha activity determined with the counter.

Relative radon and/or thoron concentrations in soil emanations were measured on site with the same instrument using a slightly modified cell. These tests take about 3 to 5 minutes and are carried out by inserting a fritted tube into a hole punched in the ground, pumping air from the hole into the cell with a rubber bulb, and measuring the alpha activity of the air with the counter.

INTRODUCTION

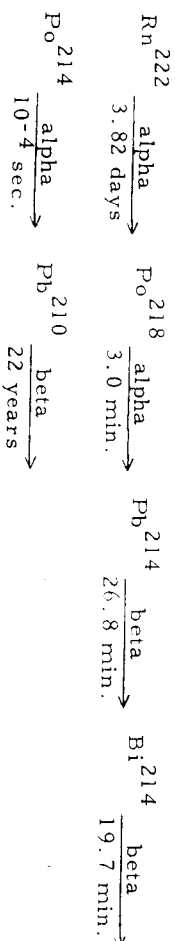
The radon determinations of natural waters and soil gases made in the Bancroft, Ontario and Ottawa-Hull areas during the 1967 field season (Dyck and Smith, 1968; Dyck, 1968) using a make-shift instrument, were encouraging enough to warrant constructing proper equipment for the systematic sampling programs encountered in geochemical exploration techniques. Although the ionization chamber offers much greater sensitivity, the relative simplicity and ruggedness of the zinc sulphide cell make it the preferred unit for routine applications.

There are many accounts of instruments for the measurement of radon (Sedlet, 1966). The instrument described here most closely resembles those described by Lucas, 1957; Higgins et al., 1961; and Rushing et al., 1964.

The basic principles involved in the detection of radon are: when alpha particles emitted by radon strike silver activated zinc sulphide, the sulphide de-excites by the emission of photons. These photons can be picked up by a photomultiplier, amplified, and recorded. Because radon is a gas at ordinary conditions it is easily separated from the other members of the radioactive series and brought into contact with activated zinc sulphide.

The radon isotopes of mass 222, 220, and 219, commonly known as radon, thoron, and actinon respectively, are radioactive decay products of the naturally radioactive series uranium 238, thorium 232, and uranium 235. They are gaseous at ordinary conditions and are therefore more mobile in natural environments than the other elements in the series. All three decay with the emission of an alpha particle to solid radioactive products. These products in turn emit several more alpha and beta particles, accompanied by gamma-ray de-excitation.

The members of the uranium 238 series which affect the efficiency of detection of radon in the instrument described below are:



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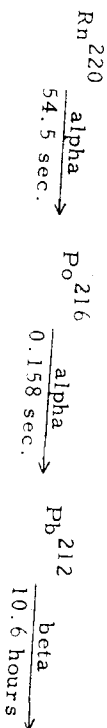
Ms. received 18 December, 1968

Critical reader: A.G. Darnley

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Ottawa 4, Canada.

Radon 219, because of its very short half life (3.9 sec.) and the low isotopic abundance of its parent uranium 235, need not be considered in this method.

The members of the thorium 232 series which could add to the alpha activity of radon 222 are:



The alpha-emitting daughter products of radon complicate matters somewhat. However, judicious choice of procedures of sampling and analysis, based on physicochemical differences of the various decay products, permits quantitative determinations of radon in natural waters to be made relatively easily. Radon in soil (determined by the method of this report) number of variables thus considered. The effect of these variables on quantitative interpretations.

This report describes apparatus and procedures which permit 30 to 40 quantitative determinations of radon in water per man day, or roughly the same number of relative radon concentration determinations in soil emanations.

THE DETERMINATION OF RADON IN NATURAL WATERS

A detailed procedure for the determination of the radon concentration in waters is included in the appendix. Essentially the method involves the removal of radon from an aliquot of the sample by passing air through it, filling an evacuated zinc sulphide cell with this air-radon mixture, and counting the alpha particle activity in the cell with a photomultiplier-scaler assembly.

Description of apparatus

Pertinent details and materials of construction of the zinc sulphide cell used for the determination of radon in water are shown in Figure 1. The zinc sulphide coating is prepared by mixing silver activated zinc sulphide powder, such as the DuPont luminescent chemical type No. 1101, with Acrylic Cement (dichloroethylene with a bit of acrylic plastic dissolved in it) until a freely flowing paste is obtained. The coating is applied to the clean wall of the cell by pouring an aliquot of the mixture into the partly assembled cell, rotating the cell all the while until the solvent is evaporated. This takes about two to three minutes. To ensure that an infinite thickness (a layer which is equal to the range of the alpha particles) is obtained, this is recommended that two to three times the theoretical thickness (a layer used. The amount of light received by a photomultiplier from alpha decay in the cell depends on the amount of energy the alpha particle spends in the

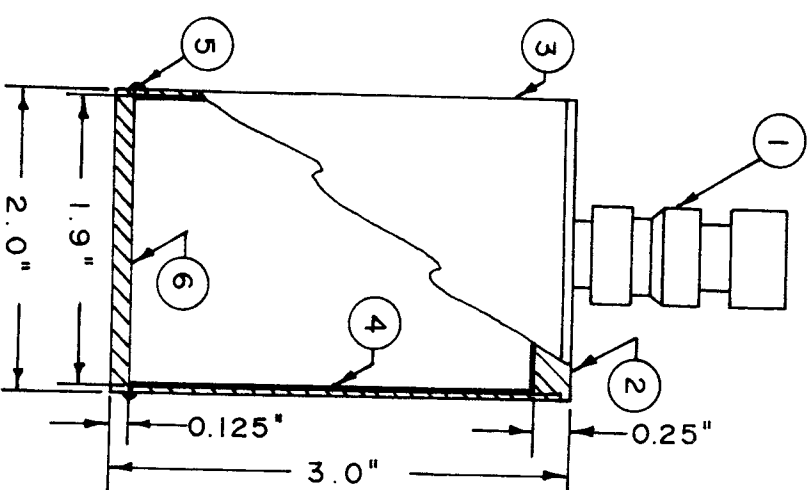


Figure 1.

Details of the water gas cell used for the determination of radon in water. 1-quick-connector, Swagelok 400-QC-1 Desc.; 2-endplate, brass; 3-copper tube; 4-silver activated zinc sulphide layer bonded to wall of tube with acrylic cement; 5-epoxy glue; 6-window, clear pyrex glass.

sulphide layer, the angle and distance between points of photon generation and detection, and on the light transmission efficiency of the window and filling gas. Such considerations plus the fact that the range of alpha particles from radon in air is about 30 ~~cm~~ ^{inches}, indicate that a hemispherical or cone-shaped cell design with dimensions similar to those shown in Figure 1 would be ideal. The cylindrical shape was chosen because it is easier to manufacture.

A schematic of the radon extraction system is shown in Figure 2. The parts are interconnected with metal and rubber tubing. The amount of rubber tubing should be kept to a minimum. Radon was found to have a considerable affinity for rubber. It may therefore take several hours to clean a rubber tube which has been exposed to a very active sample and crosscontamination can easily result if necessary precautions are not taken. The fritted disc at the bottom of the sample holder disperses air into fine bubbles resulting in more efficient scrubbing of the water sample. A simple vacuum

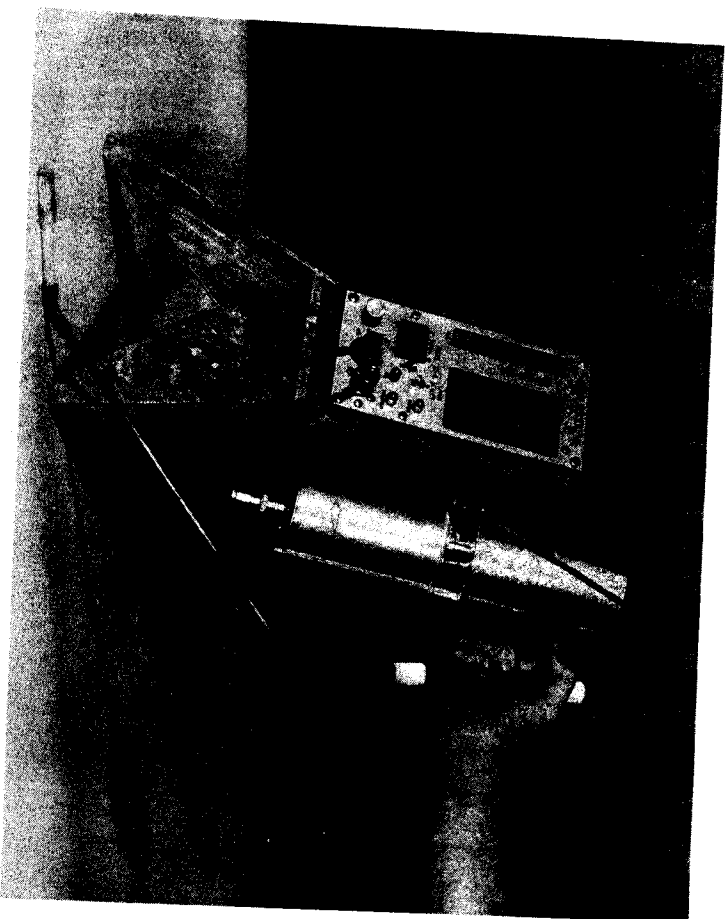


Figure 4. Photograph of radon detector equipped for on-site alpha activity measurements of soil gas emanations.

tables, in column 3. The ratio of these two values is expressed as the cell constant in the last two columns. It is evident from the values in this table that the counting error alone essentially accounts for the variation in cell constant. Experimental errors, and errors due to changes in detection efficiency of the cells are minimal at counting rates shown in the table. This observation remains essentially true even at much lower relative counting errors is evident from values tabled in Table 2. To speed up the calibration process of 63 cells and at the same time obtain better counting statistics, the cells were divided into batches of 8, each batch containing at least one of the cells calibrated with the radium standard. Each batch was then filled with radon from a strong radium solution, left until equilibrium was attained, and then counted. The batches at the bottom of Table 2 were repeated because the cells in them had given the largest deviation in the cpm from the mean of the first batch test. The reader may wonder about the reasons for the large number of cells. These were twofold. Firstly, they were split up into 3 batches of 21 cells each, each batch forming a complete radon detection unit together with a scaler-timer. The second reason becomes evident when studying Figure 5 on which is plotted the relationship of alpha activity to time obtained with a cell filled with radon 222 at time 0 (point A) and evacuated

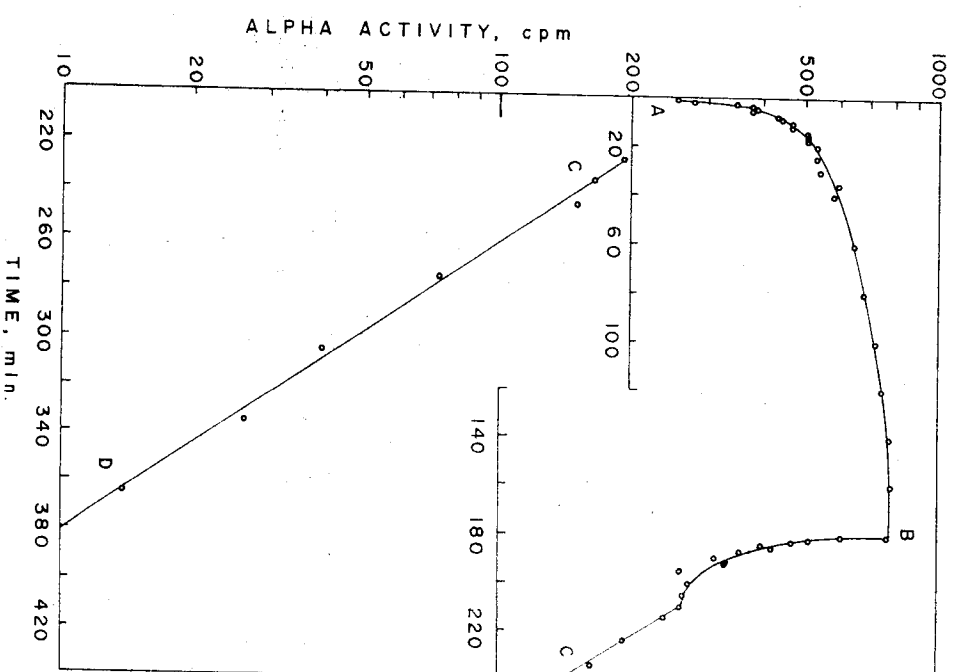


Figure 5. Plot of alpha activity from a pure radon-air sample in the water gas cell as a function of time; A-instant of filling of cell; B-removal of radon from cell; CD-decay of radon daughters deposited on wall of cell during time AB.

after 3 hours (point B). The constantly increasing alpha activity from A to B is the result of the growth of radon daughter products which are also radioactive. After about 3 hours, equilibrium between growth and decay of the daughter products and radon is reached and the whole series from radon to polonium 214 decays with the characteristic half life of radon 222. Lead 210, because of its very long half life of 22 years does not contribute measurably

to the total alpha activity. When radon is removed from the cell, the activity decreases - rapidly at first due to the decay of the 3-minute polonium 218 (B to C) then more slowly with a composite half life of about 35 minutes due to the 27-minute lead 214 and the 20-minute bismuth 214 (C to D). Because the decay products of radon are solid they cannot unlike radon, be removed by evacuation. However they decay with time. Hence, the cells need a 'cooling-off' period after they have been exposed to a highly active sample. It is therefore important to count as soon as possible after filling and remove the radon from the cell immediately after counting. Because of the sharply rising counting rate right after filling and the possibility of thoron interference, it is best to wait 5 to 10 minutes before counting.

Lucas (1957) found that cells with windows which were not coated with an electrically conducting layer on the inside, gave poor counting statistics. To find out if this was so in our case, two tests were carried out using 3 cells with coated and 3 with uncoated windows. The tests consisted in filling all six cells simultaneously with radon and counting the activity in each cell a number of times. The results of these tests are shown in Table 3. Apart from a 6.6 ± 2.1 per cent lower average efficiency of the cells with coated windows, there is no marked difference in the reproducibility of the counting rates of the two types of cells, i.e. the deviation of individual measurements from the average of each cell is less than the standard deviation of the measurement in the case of coated and uncoated windows. Even the maximum deviation of the average counting rate of individual cells from that of all cells of a kind falls within the limits of error: 5.4 per cent for cells with uncoated windows and 4.8 per cent for coated windows. The drop in counting efficiency with coated windows can be explained by assuming that the coating reduced the light transmission efficiency of the window.

The degassing efficiency of the bubbler and the loss of radon from samples stored in bottles have an important bearing on the reproducibility of the method. The results of 2 degassing efficiency tests are shown in Table 4. The procedure employed was that described in Appendix II, except that the standard solution was degassed 4 times in succession at 20 minute intervals. Between 94 and 95 per cent of the radon removable by the outlined procedure is removed from the sample in the first 4 minute degassing period. Tests using 2 minute degassing periods give only slightly lower radon removal efficiency. Because the standard radon source and the unknown samples are treated alike in this method, a reasonably constant degassing efficiency of 95 per cent is satisfactory.

Radon loss from water stored in plastic bottles, taking natural decay into account, was found to be appreciable. Even greater losses were observed when bottles were only partly filled. The results of two radon loss tests from one litre plastic bottles are shown in Figure 6. Loss from filled bottles is exponential with time (solid points), and can amount to nearly 50 per cent in 3 to 4 days. Much greater losses occur from partly filled bottles. About 90 per cent of the radon content can escape from a half filled bottle in

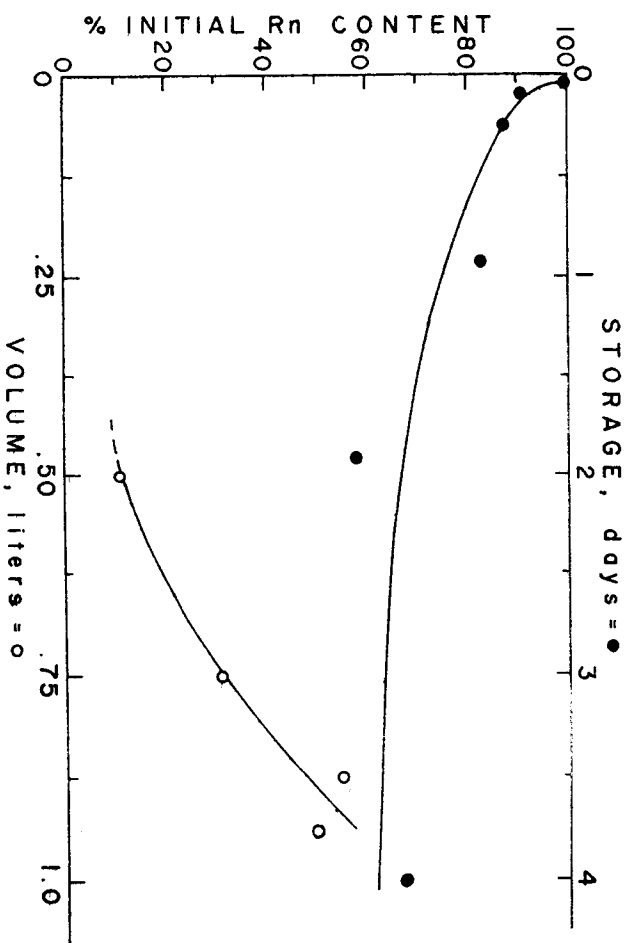


Figure 6. Loss of radon from water in 1-litre plastic bottles as a function of storage time of filled bottles (solid points), and as a function of bottle fullness (circles).

3 days (circles in Fig. 6). A summary of radon and uranium loss tests from 8-ounce plastic bottles relative to 8-ounce glass bottles is presented in Table 5, Part A. The last four samples in Part A show zero radon and uranium content and are therefore not included in the comments that follow. The average radon content of the samples collected in plastic bottles is 11 per cent lower than that of the samples collected in glass bottles from the Scientific Glass Co.; the average radon content of samples collected in Fisher glass bottles is only 1 per cent lower. The 1 per cent is well within the limits of experimental error, the 11 per cent not. The uranium analyses do not show the trend observed for radon.

Part B, Table 5, lists radon concentrations of a suite of samples collected at the same time and site. The large variation in radon concentration observed did not correlate with bottle fullness or position of bottle. The large peak at samples 3 to 6 and the smaller high at samples 11 to 13 can best be explained by assuming that the act of walking into the sampling site caused sufficient disturbance in the water to bring to the surface a highly concentrated radon solution from the bottom.

Electronic difficulties are not serious with transistorized equipment. However, the low counting rates encountered in this work make severe restrictions on the tolerable noise level of the photomultiplier. For example, one picocurie of radon in equilibrium with its daughters gives 4 cpm in the setup described here, and about 2 cpm if counted 10 minutes after filling the cell. As only about one eighth litre is used for analysis, a 1 pc/l sample will give approximately one quarter cpm. Unless the background is in the same region, the instrument will not detect radon concentration levels of about 1 pc/l. The actual background of the new cells was 0.1 cpm. After a summer's use the background of some of the cells had increased slightly, probably as a result of build-up of lead 210.

The detection efficiency of a photomultiplier decreases exponentially with the age of the tube. It is therefore necessary to correct for this change when absolute measurements are desired. One of the simplest ways to detect such changes, as well as shifts in gain of the main amplifier and changes in the high voltage supply, is to measure the plateau of the counting system. A typical plateau of the unit described here is shown in Figure 7. By operating near the midpoint of the flat portion of the plateau, considerable changes in the applied voltage or gain of the system can occur without changing the counting rate or efficiency of the system appreciably.

In relating radon concentrations of water samples to source strength it is important to know the detection limit as well as the reproducibility or errors of the method. As stated above the limit of detection is 1 or 2 pc/l. The use of larger cells and samples, sample enrichment, and longer counting times would lower the detection limit but decrease the output. The errors associated with the method can be divided into 4 types: instrumental, experimental, sampling, and statistical. Instrumental errors include detection efficiency changes of cells and photomultiplier, changes in the gain of amplifiers, high voltage drifts, and faulty operation of other components such as pressure gauges, timers, and valves. The main experimental errors are pressure, volume, and time measurements, and the degassing procedure explained earlier. Sampling errors will result mainly from the escape tendency of radon. To minimize these, glass bottles with well-sealing caps should be used and filled to the top except for one air bubble. This bubble is necessary to prevent the tightly sealed bottle from cracking during temperature changes. The largest uncertainty in the radon values results from the inhomogeneous distribution of radon in natural water bodies. Experience has shown (see Table 5, Part B) that the act of sampling can disturb the water and change the radon concentration at the site by as much as a factor of 5. In view of such large variations, the other errors including the errors associated with the randomness of radioactive decay, become negligible. With constant radon sources the statistical counting error is dominant in all but very active samples (several hundred pc or more).

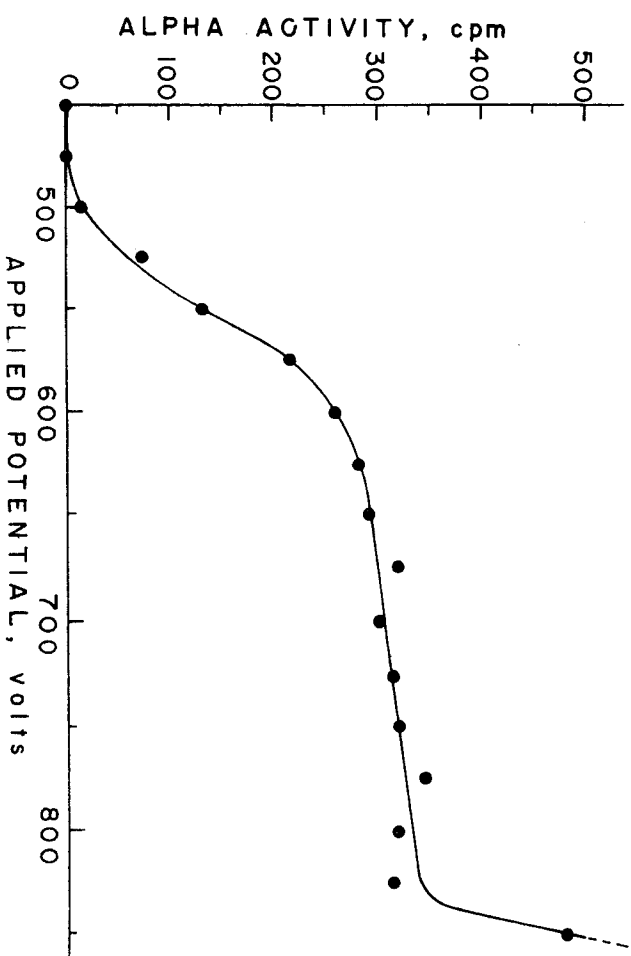


Figure 7. Typical plateau of the radon alpha activity obtained with the water gas cell and electronics described in this report.

ON-SITE RADON DETERMINATIONS OF SOIL EMANATIONS

Semiquantitative determinations of radon and/or thoron in soil can be carried out relatively quickly and easily with the instrument already described using a modified cell and procedure for the transfer of the emanation. The assembled components used successfully for the determination of on-site radon-thoron levels in soils is shown in Figure 4. Except for the cell and the cap on the photomultiplier, the components are the same as those used in the determination of radon in water. The whole assembly as shown weighs about 15 pounds. Some of this weight is due to a battery charger and other options in the scaler-timer not really necessary in a portable instrument. A cross-section of the modified cell and the cap is shown in Figure 8. The cell consists of a metal cylinder coated on the inside with silver-activated zinc sulphide. The tapered rubber rings, one fastened to the photo-tube and the other to the inside of the cap, provide a reasonably air tight chamber when cell and cap are put in place. Soil gas is sucked into the rubber bulb pump and forced into the cell through inlet No. 2 by displacement. The pump displaces about 50 ml with one puff. A porous plug at the intake end of the gas line prevents dust from entering the cell. As soon as a hole is made, the tube with the porous end is inserted into the hole to the desired depth and the air transferred to the cell and counted for 3 one minute periods. The desirability of counting several intervals becomes apparent from studying

Figure 8.
Cross-sectional view of soil gas cell and light cap in place above photomultiplier. 1-gas outlets; 2-gas inlet; 3-tapered rubber sleeves; 4-copper cylinder; 5-silver activated zinc sulphide cement; 6-removable cap of photomultiplier shield; 7-fixed photomultiplier shield; 8-photomultiplier, RCA 6342-A or equivalent.

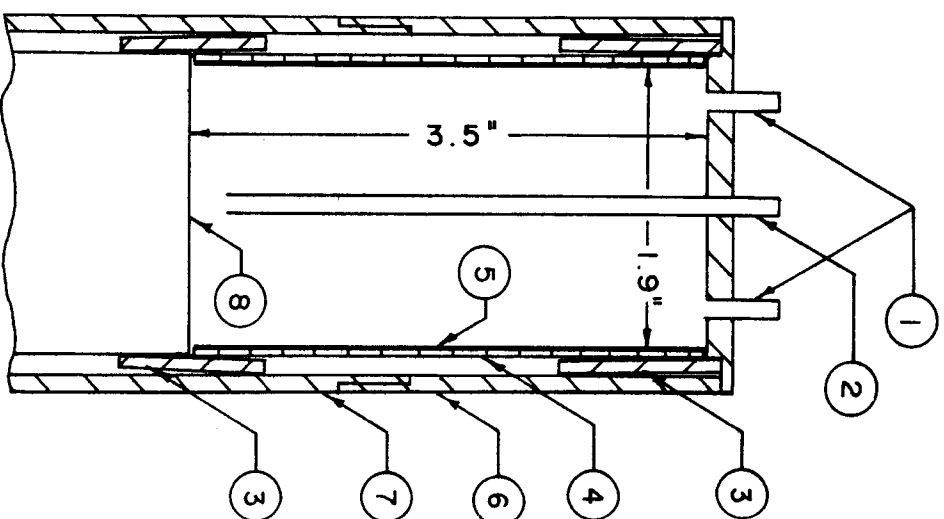


Figure 9 where the alpha activity of several synthetic gas mixtures in the soil counter are plotted as a function of time. The solid points were obtained from a filling of a pure thoron-air mixture. The one minute half life of this activity confirms the presence of nearly pure thoron. The straggling near the 8-minute mark suggests the presence of a trace of radon; the last alpha emitter in the thorium series is branching and decaying with an approximate half life of 17 hours due to the 16.6-hour half life of lead 212, and should therefore not be detectable at the level shown. The rising curves were obtained with pure radon-air mixtures; the upper one from the soil gas cell and the lower one from the water gas cell previously described. Because the soil gas cell is not vacuum tight and the exhaust is left open during routine measurements, there is a possibility of radon escape. However the two curves rise at about the same rate indicating that loss in the open ended cell

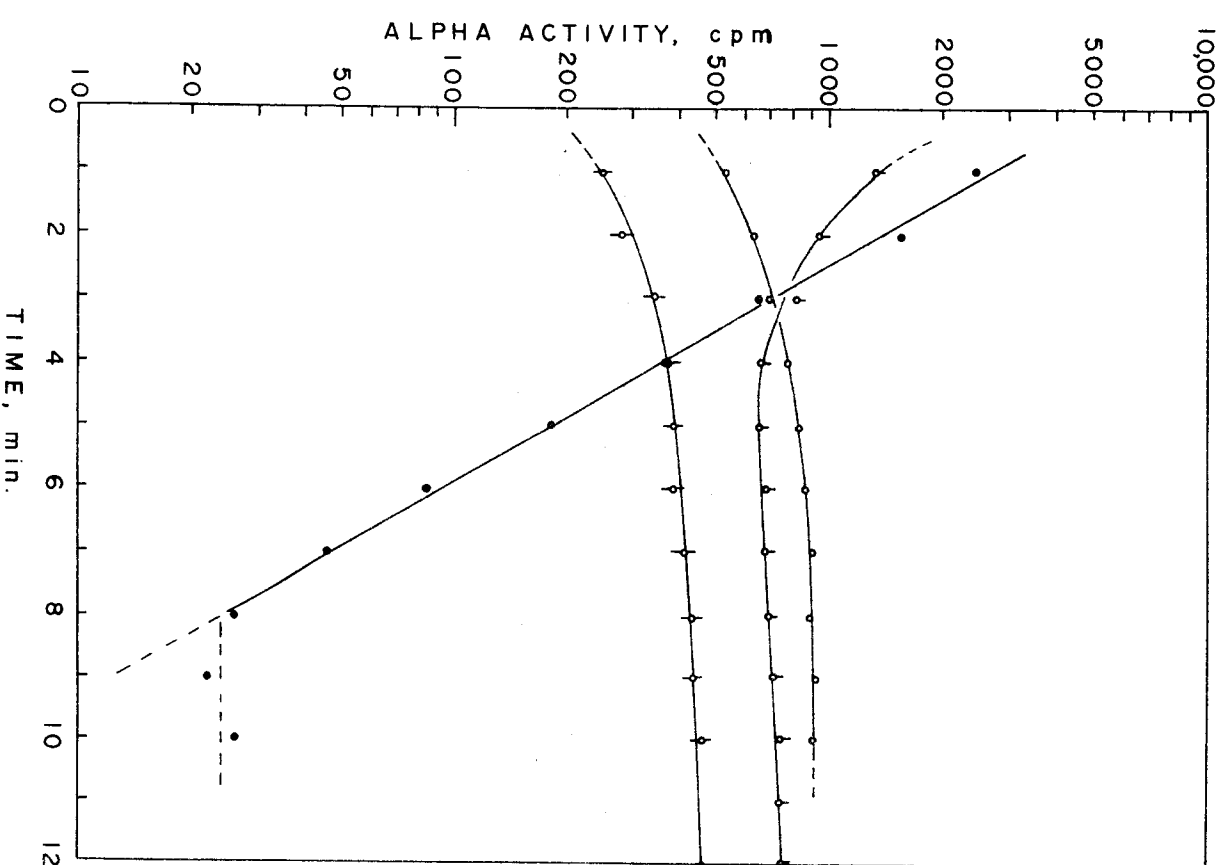


Figure 9. Plot of alpha activity as a function of time obtained with the soil gas cell. Solid points--pure thoron-air; circles--pure radon-air; circles with one tail--mixture of radon and thoron-air; circles with two tails--pure radon-air in the water gas cell.

is negligible during the first 10 minutes. The curve exhibiting a minimum at the four-minute mark was obtained with a radon-thoron-air mixture. In this case the activity decreases at first due to the short half life of thoron and then when the thoron has all decayed the activity assumes the slope portrayed by pure radon. Thus by counting a soil gas sample immediately after collecting for several intervals, a rough measure of the relative amounts of radon and thoron can be obtained. For example, I know from calibration tests that the soil gas cell has a detection efficiency of approximately 4 cpm/pc for radon in equilibrium with its decay products. By comparing curve AB in Figure 5 to the lower pure radon curve in Figure 9 (these curves were obtained from the same filling) it can be seen that a cpm of 76.0 at equilibrium is equivalent to a cpm of 250 at the one minute mark. Hence this particular filling contained about 200 pc of radon. Therefore, the 1 minute radon counting efficiency of the instrument is about 1.2 cpm/pc, and the 10 minute counting efficiency of the instrument is about 12 cpm/pc. If the thoron decays almost instantaneously, the efficiency of the instrument for thoron will be about 1/10 that of radon, or roughly 2 cpm/pc. From the 10 minute count of the radon-thoron mixture one can estimate a radon content of 320 pc. Converting this to one minute cpm and subtracting it from the total, one obtains a thoron content of about 500 pc.

To relate radon and thoron emanations quantitatively to uranium and thorium concentrations is difficult. Too many factors such as radioactive disequilibrium of the series in the soil as a result of weathering, changes in the emanation efficiency due to soil density and grain size, moisture content, distribution of radioactive particles, etc. could cause erroneous extrapolations. However, precise quantitative results are not of prime importance in prospecting. The important thing is that there be correspondence between radon in soil emanations and uranium in the area nearby. That this correspondence exists was demonstrated last summer in the field. The results of the field tests will be published separately. Two tests, one to determine the optimum number of puffs to fill the cell, and the other to measure the variation in radon content of soil emanations at a site, are recorded in Tables 7 and 8. As can be seen, the cpm decreases fairly consistently with increasing number of puffs. No doubt the flushing action of atmospheric air displacing the air pumped out of the hole is responsible for the decrease. With a constant thoron source, seven puffs are required to reach maximum cpm. On the basis of these tests it was decided that 4 puffs would be a good number to use. Fewer puffs, it was felt, would lead to larger concentration fluctuations, and hence give poorer reproducibility, even though slightly higher cpm are obtainable with fewer puffs. Tests for the homogeneity of radon emanations were carried out at 3 natural sites. Results obtained with the soil gas cell are shown in Table 8, Part A. The soil gas samples were taken from 1-foot-deep holes punched in the ground with a needle bar and spaced 6 to 8 inches apart. The average deviation from the mean cpm at a site is ± 30 per cent. However, individual measurements can differ by a factor of two. The change in cpm with time at these test sites clearly indicates the predominance of radon at sites 1 and 2, and of thoron at site 3.

The results in Part B show that nearly twice the efficiency is obtainable with evacuated cells. However, convenience and speed made the gas displacement technique the preferred one. As was the case with water analyses, prolonged exposure of the cell to high radon concentrations results in an increase in the background count of the cell. It is therefore desirable to have about a dozen of these cells for replacement as required. Also the affinity of radon for rubber makes it advisable to use as little rubber as is possible in the gas lines, and place the pump after the cell in the flow system.

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TABLES*

Table 1.. Calibration of cells using a 250 pc/l
N.B.S. radium 226 standard.

Cell No.	cpm**	pc/l	Cell constant, cpm/pc/l	
			Single	Average
8	38.7 ± 1.9	128.9	0.300	0.328
	14.0 ± 1.2	41.4	0.337	
	14.1 ± 1.2	41.4	0.340	
16	26.5 ± 1.3	76.0	0.349	0.347
	13.7 ± 1.2	41.4	0.316	
	15.2 ± 1.2	41.4	0.366	
	15.1 ± 1.2	41.4	0.359	
	17.7 ± 1.3	41.4	0.427	
24	23.4 ± 1.5	76.0	0.308	0.351
	16.5 ± 1.3	41.4	0.398	
	32.2 ± 1.8	104.8	0.307	
32	26.5 ± 1.6	76.0	0.348	0.329
	14.4 ± 1.2	41.4	0.347	
	13.0 ± 1.1	41.4	0.314	
	13.9 ± 1.2	41.4	0.335	
	37.5 ± 1.9	104.8	0.357	
40	14.1 ± 1.2	41.4	0.340	0.344
	33.1 ± 1.8	104.8	0.316	
	41.7 ± 2.0	128.9	0.323	
48	14.6 ± 1.2	41.4	0.352	0.331
	33.1 ± 1.8	41.4	0.323	
	43.8 ± 2.1	128.9	0.339	
56	13.2 ± 1.1	41.4	0.313	0.325
	13.4 ± 1.2	41.4	0.323	
	13.4 ± 1.2	41.4	0.323	
64	14.9 ± 1.2	41.4	0.359	0.335

* The radon determinations presented in these tables were carried out by J. C. Pelchat of the Radiochemistry Laboratory of the Geological Survey of Canada.

** These counting rates were obtained employing the procedure described in Appendix II.

Table 2. Relative calibration of cells.
Column (1) lists the cells in a batch of 8, and
column (2) the average counting rate of two ten-
minute counting intervals.

(1)		(2)		(1)		(2)	
1	201.6 ± 3.2	9	140.3 ± 2.7	17	378.9 ± 4.4		
2	203.2 ± 3.2	10	145.6 ± 2.7	18	359.8 ± 4.2		
3	193.8 ± 3.1	11	141.5 ± 2.7	19	369.9 ± 4.3		
4	190.3 ± 3.1	12	146.5 ± 2.7	20	385.3 ± 4.4		
5	190.3 ± 3.1	13	146.8 ± 2.7	21	377.2 ± 4.4		
6	206.0 ± 3.2	14	142.9 ± 2.7	22	375.9 ± 4.4		
7	205.2 ± 3.2	15	137.8 ± 2.6	23	376.2 ± 4.4		
8	196.2 ± 3.1	16	143.7 ± 2.7	24	371.2 ± 4.3		
25	130.0 ± 2.5	33	112.5 ± 2.4	41	119.7 ± 2.5		
26	134.8 ± 2.5	34	111.5 ± 2.3	42	122.7 ± 2.5		
27	123.8 ± 2.5	35	111.3 ± 2.3	43	126.2 ± 2.5		
28	122.8 ± 2.4	36	116.3 ± 2.4	44	127.3 ± 2.5		
29	128.7 ± 2.5	37	116.0 ± 2.4	45	123.9 ± 2.5		
30	124.6 ± 2.5	38	114.0 ± 2.4	46	124.8 ± 2.5		
31	127.0 ± 2.5	39	114.3 ± 2.4	47	125.9 ± 2.5		
32	130.0 ± 2.5	40	109.7 ± 2.3	48	124.9 ± 2.5		
49	98.2 ± 2.2	57	228.0 ± 3.4				
50	98.0 ± 2.2	58	225.4 ± 3.4				
51	97.7 ± 2.2	59	232.2 ± 3.4				
52	93.7 ± 2.2	60	229.3 ± 3.4				
53	98.0 ± 2.2	61	233.5 ± 3.4				
54	94.3 ± 2.2	62	232.9 ± 3.4				
55	94.2 ± 2.2	63	232.9 ± 3.4				
56	94.8 ± 2.2						
5	108.4 ± 2.3	40	101.7 ± 2.3				
15	117.0 ± 2.3	41	96.1 ± 2.2				
18	112.2 ± 2.4	55	98.0 ± 2.2				
28	114.5 ± 2.4	62	99.8 ± 2.2				
6	115.4 ± 2.4	36	97.6 ± 2.2				
10	118.0 ± 2.4	44	101.0 ± 2.2				
20	117.7 ± 2.4	49	101.9 ± 2.3				
26	114.6 ± 2.4	61	98.4 ± 2.2				

Table 3. Reproducibility tests of tin oxide coated
and uncoated pyrex glass cell windows.

Test No. 1		Net decay corrected 10 min. counts taken at equilibrium				
Cell No.		1st count				
		1st count	2nd count	3rd count	4th count	Average
1	uncoated	474 ± 22	489 ± 22	491 ± 22	409 ± 23	491 ± 11
2		459 ± 21	458 ± 21	458 ± 21	461 ± 21	459 ± 10
3		497 ± 22	523 ± 23	513 ± 23	482 ± 22	504 ± 11
4		482 ± 22	485 ± 22	478 ± 22	490 ± 22	484 ± 11
5	Coated	444 ± 21	447 ± 21	435 ± 21	433 ± 21	440 ± 10
6		467 ± 22	462 ± 22	454 ± 21	461 ± 22	461 ± 10
Test No. 2						
1	uncoated	356 ± 19	358 ± 19	363 ± 19	362 ± 19	360 ± 10
2		338 ± 18	339 ± 18	338 ± 18	349 ± 19	341 ± 9
3		374 ± 19	377 ± 19	358 ± 19	378 ± 20	372 ± 10
4		353 ± 19	347 ± 18	340 ± 18	336 ± 18	344 ± 9
5		311 ± 18	319 ± 18	318 ± 18	324 ± 18	318 ± 9
6	coated	332 ± 18	341 ± 18	330 ± 18	323 ± 18	331 ± 9

Table 4. Radon degassing efficiency of bubbling assembly standard radium 226 solution.

Degassing	Test No.			
	cpm*		per cent	
	cpm*	per cent	cpm	per cent
1st	32.7	95.9	40.9	93.0
2nd	1.2	3.5	2.2	5.0
3rd	0.1	0.3	0.6	1.3
4th	0.1	0.3	0.3	0.7
totals	34.1	100.0	44.0	99.9

* The counting rates are those obtained in the 10- to 20-minute interval after filling of cells and are corrected for radon growth between degassing.

TABLE 5.

Part A. Radon and uranium loss tests from 8 ounce plastic bottles relative to 8 ounce glass bottles.

Sample Location	Collection date 1968	Radon, pc/l*			Uranium, ppb*	
		glass "Scientific"	glass "Fisher"	plastic	glass "Scientific"	glass "Fisher" plastic
Kingsmere Lake	27/02	2	3	15	2.2	1.7
	"	11	13		2.4	3.6
	"	69	51		2.9	3.1
	"	83	68		3.4	3.1
	"	136	168		1.2	1.2
Chelsey Brook	upstream	230		188	2.6	
	downstream	133		103	0.9	2.9
Fortune Lake	upstream	336		229	0.7	0.5
	downstream	20		19	1.0	0.2
Pinks Lake	18/03	47		34	0.6	0.4
	"	7		6	0.4	0.6
	"	326		257	0.2	0.2
	"	623	605	528	0.2	0.4
	"					0.2
middle surface	"	0	0	0	0.0	0.0
	"	0	0	1	0.0	0.0
	"	1	0	0	0.0	0.0
	"	1	0	1	0.0	0.0

Part B. Radon concentrations in pc/l in water samples collected in 8 ounce bottles from a "hot spot" in Fortune Lake, Galtineau National Park on May 27, 1968. Samples were collected at the surface of the lake 5 feet from shore where the depth of water was approximately 1 foot.

Bottle fullness	Scientific Glass		Plastic		Fisher Glass	
	up ⁺	down	up	down	up	down
full	591 (1) ⁺⁺	416 (2)	645 (7)	582 (8)	869 (13)	713 (14)
1/4" space	1108 (3)	2196 (4)	498 (9)	553 (10)	693 (15)	624 (16)
1" space	1199 (5)	1151 (6)	828 (11)	740 (12)	(17)	634 (18)

* Radon analyses of the water samples were carried out on the same day or the day after collection; all uranium analyses were carried out on May 15.

⁺ Up and down refers to the position of bottle neck between collection and analysis of the sample.

⁺⁺ Numbers in brackets refer to the order in which the samples were collected.

TABLE 6. DECAY OF RADON 222 AS A FUNCTION OF TIME
HALF-TIME 91.800 HOURS (3.825 DAYS)

FRACTION OF RADON REMAINING AFTER X HOURS					
HOURS	0.0	1.0	2.0	3.0	4.0
0.0	1.0000	.9924	.9850	.9776	.9702
5.0	.9629	.9557	.9485	.9413	.9343
10.0	.9273	.9202	.9133	.9065	.8996
15.0	.8929	.8862	.8795	.8729	.8663
20.0	.8598	.8533	.8469	.8405	.8342
25.0	.8279	.8217	.8155	.8094	.8033
30.0	.7973	.7913	.7853	.7794	.7735
35.0	.7677	.7619	.7562	.7505	.7449
40.0	.7393	.7337	.7282	.7227	.7173
45.0	.7119	.7065	.7012	.6959	.6907
50.0	.6855	.6803	.6752	.6701	.6651
55.0	.6601	.6551	.6502	.6453	.6405
60.0	.6356	.6309	.6261	.6214	.6167
65.0	.6121	.6075	.6029	.5984	.5939
70.0	.5894	.5850	.5806	.5762	.5719
75.0	.5676	.5633	.5591	.5549	.5507
80.0	.5465	.5424	.5384	.5343	.5303
85.0	.5263	.5223	.5184	.5145	.5106
90.0	.5068	.5030	.4992	.4954	.4917
95.0	.4880	.4843	.4807	.4771	.4735
100.0	.4699	.4664	.4629	.4594	.4559

Table 7. Determination of optimum number of puffs
(pump strokes) to move soil gas into soil
gas cell from one-foot-deep holes in the
soil. Holes at a site were made 6 to 8
inches apart. Each count was obtained with
gas from a new hole.

Site No. 1		Site No. 2	
No. of puffs	cpm	No. of puffs	cpm
3	203	5	56
6	191	10	76
9	171		
		5	39
3	207	10	23
6	199		
9	168	5	34
		10	31
3	180		
6	160	5	68
9	121	10	44
3	146	5	41
6	222		
9	140	10	44

Table 8. Tests for homogeneity of radon emanations from one-foot-deep holes in the soil. Holes at a site are 6 to 8 inches apart. Four puffs were used to fill cells.

Site No.	Hole No.	Net cpm			
		1st min.	2nd min.	3rd min.	Average
A. Soil gas cells					
1	1	93	120	130	114
	2	124	158	141	141
	3	136	136	136	136
	4	113	100	107	107
	5	134	128	127	129
	6	115	97	115	109
2	7	135	135	110	127
	1	208	194	223	208
	2	223	212	241	225
	3	279	302	301	294
	4	342	342	346	343
	5	396	449	450	432
3	6	298	345	357	333
	7	194	194	243	210
	1	70	38	13	40
	2	70	59	32	54
	3	95	48	14	52
	4	68	39	29	45
B. Evacuated water gas cells	5	128	83	44	85
	6	47	30	22	33
1	8	200	207	217	208
	9	210	245	244	233
	10	260	268	298	281
3	7	124	41	42	69
	8	145	108	46	100
	9	118	61	46	75

APPENDIX II
Procedure for measuring radon 222 in
Water Samples

Procedure for measuring radon 222 in water samples

Switch counter on and let it warm up for about 2 minutes before making measurements.

Evacuate cell and admit air to cell through drierite trap. (Make sure that drierite is not used up.)

Place cell on photomultiplier tube after checking if pm tube and cell window are clean. Put cap on pm tube and engage high voltage switch. Wait one minute for the de-excitation of light in the cell with counting switch on. Zero scaler and timer and count "background" for 4 to 5 minutes.

Evacuate cell, transfer 130 ml of the water sample from bottle to clean bubbler without turbulence and connect bubbler to vacuum line as soon as possible. Close valve to pump, open stopper above bubbler slowly, set interval timer to ten minutes, slowly open stopper below bubbler watching pressure gauge and let air bubble through the water into cell fast enough to fill cell in 4 minutes. If necessary use air pressure to fill cell completely. Nine minutes after the start of the filling procedure place cell on pm tube and connect the high voltage by replacing the light shield. Observe the light de-excitation. If cell is exposed to strong light it will take more than a minute to de-excite. It is best therefore, to place the cell on a flat opaque surface a few minutes before counting.

Exactly ten minutes after the start of the filling procedure zero the scaler and timer and take two five-minute counts.

Record date, sample number, time of day, cell number, background counts/minutes, sample counts/minutes.

Immediately after completion of counting of the sample, evacuate the cell and flush three times with air by filling and evacuating the cell. (Avoid using the same cell until all cells have been used once: the radon daughters decay with a half life of 35 minutes. Hence if not allowed to stand for some time, the background of the cell and therefore, the error in the next determination, could be high).

As soon as a sample count is completed, count another background, etc. As soon as a cell is clean, fill another cell with radon from another sample, etc. Time operations in such a way as to complete a background and sample count every ten to fifteen minutes depending on the counting rate of the sample; the higher the sample counting rate with respect to the background counting rate, the shorter the counting time required.

The whole system should be checked at least once a week with the standard radium solution. The test can be carried out as follows: degass for about ten minutes enough of the standard to fill a sample bottle. Fill

bottle with the degassed standard noting the time and date. One or two days later measure the radon that has accumulated in the standard in the same way as usual, and compare this to the amount produced by the radium during the storage time i. e. 250 times fraction of radon grown into solution since degassing. For example: the 250 picocurie standard was degassed 29 hours ago. Look up the decay of radon in 29 hours (=0.803, Table 6). The growth of radon is $\approx 1 - 0.803 = 0.197$. The amount of radon produced by the standard is therefore =

$$250 \times 0.197 = 49.25$$

Sample calculation:

Background	= 4 counts/5 min. = 0.8 cpm
Sample	= 12 counts/5 min. 25 counts/10 min. = 2.5 cpm
Net	= 2.5 - 0.8 = 1.7 cpm

If the sample was collected 21 hours ago, the decay corrected net rate is:

$$\begin{aligned} &= 1.7/0.853 = 1.99 \text{ cpm where } 0.853 \text{ is the fraction of radon} \\ &\text{remaining in the sample after 21 hours, and is taken from the radon 222} \\ &\text{decay table. The radon concentration in the sample at the time it was col-} \\ &\text{lected, assuming there is no radium in the sample, is:} \\ &= 1.99/0.333 = 5.98 \text{ pc/l} \end{aligned}$$

Where 0.333 is the experimentally determined cell constant expressed in cpm/pci (counts per minute per picocurie per litre). This constant is applicable only in the case where the sample was counted in the 10- to 20-minute interval from the start of the filling procedure. Counting at another interval will require corrections depending on the degree of equilibrium attained between radon and its decay products. The error due to the randomness of radioactive decay will be the largest single error in the determination in all but very active samples. In the example above the standard error of the background rate = $(4) 1/2/5 = \pm 0.4$ cpm. The error of the sample rate = $(25) 1/2/10 = \pm 0.5$ cpm the error of the net rate = $((0.5)^2 + (0.4)^2) 1/2 = \pm 0.64$ cpm or ± 38 per cent.