

POOR ORIGINAL

MEASUREMENT OF ^{222}Rn FLUX WITH CHARCOAL CANISTERS

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ABSTRACT

A new method for determining the rate of ^{222}Rn (radon) emanation from a surface is described. A modified U. S. Army M11 gas mask canister containing activated charcoal is sealed to the surface for several hours to several days, depending on the anticipated flux. The amount of radon adsorbed in the charcoal canister is determined by counting gamma emission from radon daughters in equilibrium with the radon. Details of the canister calibration and the parameters that affect the measurements of radon flux using activated charcoal canisters are also described.

A. INTRODUCTION

Several methods have been used to measure the radon flux from surfaces. The most common method is the direct accumulation of radon in a closed container resting on the surface. An aliquot of the air from the accumulator is transferred either to an ionization chamber or to an alpha scintillation flask for analysis. Accumulators have been used to measure radon flux from soil surfaces extensively in the United States by Kraner *et al.*⁽¹⁾ and Wilkening *et al.*⁽²⁾ An alternate method consists of entraining the radon emanating from a small area of the ground in an airstream moving in a closed system through a charcoal trap or cold trap. At the end of the sampling period, the sealed trap is returned to the laboratory where the radon is transferred into an evacuated scintillation flask for analysis. This method is described by Wilkening and Hand⁽³⁾ and by Pearson.⁽⁴⁾ Recently Megumi and Mamuro⁽⁵⁾ developed a method whereby the radon emanating from the soil is adsorbed on a layer of granular, activated charcoal spread directly on the ground. The charcoal is bagged and returned to the laboratory where the analysis involves measuring the gamma activity from the 0.61 MeV gamma rays of ^{214}Bi (RaC). In another method described by Wilkening *et al.*,⁽²⁾ radon flux is calculated from measurements of airborne radon at varying heights in a vertical column of air. A balance is assumed between the radon input at the base of the column of air, and the decay of atoms within the column.

This paper describes a new procedure based on the one described by Megumi and Mamuro. This procedure which has been described briefly by this author⁽⁶⁾ is an improvement over the earlier methods. A modified U. S. Army M11 gas mask canister containing activated charcoal is placed directly in contact with the emanating source. After an exposure period from several hours to several days, the canister is removed from the source and placed in a gamma spectrometer. Radon adsorbed on the charcoal is determined by counting the radon daughters that have grown to equilibrium with the radon. As in the method proposed by Megumi and Mamuro, the canisters provide a time-integrated measurement of radon flux as opposed to the "instantaneous" measurement of the accumulator methods. The major advantage of the charcoal canisters over the traditional accumulator methods, as well as the Megumi and Mamuro method, is that numerous measurements can be made inexpensively due to the low cost and the ease of deployment and recovery of the canisters. Moreover, the canisters are reusable and are manufactured to rigid military specifications. In anticipation that the U. S. Army M11 canister may not be available at some time in the future, the characteristics of a commercial activated charcoal canister also were investigated. A canister that is

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manufactured by the Mine Safety Appliance Co. (MSA) was chosen arbitrarily from among several that are available.

B. DESCRIPTION OF THE METHOD

The U. S. Army M11 gas mask canister, developed by the Army Chemical Corps during World War II, has been described by West⁽⁷⁾ and Thomas.⁽⁸⁾ For use in flux measurements, the canisters are modified by removing the section containing a pleated filter and fitting a plastic or metal lid on the other threaded end. The remaining cylindrical metal canister is 6 cm deep by 10.5 cm in diameter, weighs 225 g and contains 148 g of activated charcoal. To measure radon emanation from concrete or similar smooth surfaces, the canister is sealed to the surface by applying a bead of pliable weather stripping around the outer edge that contacts the surface. To measure radon emanation from soil, the canister is twisted into the soil to a depth of 1 cm, and additional soil is packed around the edge to a depth of 2 cm. At the end of the exposure period the canister is removed from the surface, wiped clean, and the open end of the canister is sealed with a tight-fitting plastic or metal lid. The canister is then returned to the laboratory for analysis.

The canisters are analyzed with a gamma spectrometer. We use an 8 cm by 8 cm NaI scintillation detector coupled to a 400-channel multichannel pulse height analyzer with a teletype printer. The canister is placed on the detector, with the exposed side down, and is counted any time later than five hours after the end of the exposure period. This allows the radon daughters to reach equilibrium with the adsorbed radon. The integration limits of the multichannel analyzer are set to include the gamma activity from ²¹⁴Pb (0.242 MeV, 0.294 MeV, and 0.352 MeV) and from ²¹⁴Bi (0.609 MeV).

After analysis, the canister is prepared for reuse by purging the radon with 1000 liters of room air, heated to a minimum of 60°C. Any time later than five hours after the regeneration of the canister, an integral background count of the canister is recorded. In addition to gamma-counting the canisters, a count of a reference sample (e.g., ²²⁶Ra), corrected for counter background, is recorded to check the counter response.

Radon flux, J , is calculated by dividing the net gamma activity due to radon adsorbed on the canister (corrected for decay), by the calibration factor of the counter for the canisters, the area of the canister, and the exposure time. The following equation applies:

$$J = \frac{(\text{Net Counts})(\lambda_1^2)}{(E)(A)(1 - e^{-\lambda_2 t})(e^{-\lambda_2 t'} - e^{-\lambda_2 t''})} \quad (1)$$

where

J = radon flux, pCi m⁻² min⁻¹

E = calibration factor of the counter for the canisters, cpm/pCi

A = area of the canister, m²

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λ_1, λ_2 = radon decay constants, $1.26 \times 10^{-4} \text{ min}^{-1}$ and $7.55 \times 10^{-3} \text{ h}^{-1}$, respectively
 t = exposure period, hours
 t' = time between end of sampling and start of counting, hours
 t'' = time between end of sampling and end of counting, hours

C. CALIBRATION

Calibration was obtained by a direct comparison with the traditional accumulator method. Canisters and accumulators were exposed simultaneously to several test surfaces. The exposed canisters were gamma-counted and the radon flux as determined by the accumulators was used to calculate the calibration factor for the canisters.

A test slab of concrete containing an artificially high level of ^{226}Ra (520 pCi per gram) served as a convenient emanation source for the comparison measurements between the canisters and the accumulators in the laboratory. Canisters and accumulators were exposed simultaneously side-by-side on this concrete slab. Positions of canisters and accumulators were alternated to average spatial and temporal variations in radon flux. Both the canisters and the accumulators were sealed to the concrete with a pliable weather stripping material. Exposure periods for the accumulators were limited to two hours or less to avoid the suppression of radon emanation. At the end of each exposure period, the air inside the accumulators was sampled with an alpha scintillation flask which was then analysed for radon content. Exposure periods for the canisters ranged from one hour to one week.

Additional comparisons of the flux-can method and the canister method were subsequently made at several locations outside the laboratory.

Figure 1 shows calibration data obtained for several surfaces. The radon flux from these surfaces ranged from $5.2 \text{ pCi m}^{-2} \text{ min}^{-1}$ for a concrete floor to $1.2 \times 10^4 \text{ pCi m}^{-2} \text{ min}^{-1}$ for a uranium mill tailings pile. Figure 1 indicates that the calibration factor for the canisters, $0.194 \pm 0.005 \text{ cpm per pCi of radon}$, is independent of exposure time from one hour to one week. From this figure, it may be inferred that the adsorption efficiency of the canister is also independent of time for these exposures up to one week and the canisters can be used to obtain time-integrated measurements of flux. Assuming constant flux during the exposure time and taking account of its half-life, radon will increase to approximately two-thirds of its maximum accumulation in the canister in six days. Thus there is little to be gained by longer exposures.

To establish that the canister adsorbs the radon quantitatively, several canisters were used to sample air containing a known amount of radon by means of an air sampling pump. The total air volume sampled in each case was less than 100 liters to prevent radon from being desorbed from the canister.⁽⁸⁾ Radon concentration in the air was determined by the two-filter method described by Thomas and LeClare.⁽⁹⁾ The integrated gamma activity of these canisters was $0.195 \pm 0.007 \text{ cpm per pCi of radon}$, confirming that radon is completely adsorbed by the canister.

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D. PROPERTIES OF CHARCOAL CANISTERS

To measure the reproducibility of the canister method, as it would be used to measure radon flux in the field, multiple exposures were made on several test surfaces. For each surface the canisters were deployed simultaneously in a relatively small area (1-2 m²) to minimize variation. The results of these tests are presented in Table 1. Coefficients of variation, corrected for counting errors, ranged from 0.06 for an outdoor location in the high phosphate region of Florida, to 0.15 for a measurement on soil in New Jersey.

The sensitivity of the canister method for measuring environmental levels of radon flux was calculated assuming a counter background of 58 cpm and a canister background of 24 cpm, which are typical values for our system. The lower limit of detection (LLD), which is defined as the smallest amount of sample activity that will yield a net count for which there is confidence at a predetermined level that activity is present, ⁽¹⁰⁾ was calculated for a counter calibration of 0.194 cpm per pCi of radon. Values are given as functions of counting time and confidence level in Table 2. An LLD of 69 pCi, corresponding to a 10-minute count and 95% confidence level, is equivalent to the radon accumulated on a canister in a four-day exposure at a flux of 2 pCi m⁻² min⁻¹.

As a substitute for the M11 canister, a commercial charcoal canister was investigated. Mine Safety Appliance Co. (MSA) manufactures an activated charcoal cartridge, Type CMA #459315, that is used with a gas mask against organic vapors. The cartridge is 2.5 cm high by 7.2 cm in diameter and contains 36 g of activated charcoal, with a mesh size similar to that of the M11 canister. Several of the MSA cartridges were exposed simultaneously with the M11 charcoal canisters for exposure periods up to one week. The results indicate that the MSA canister is equally suitable for measuring radon flux.

E. PROPERTIES AFFECTING CANISTER PERFORMANCE

Since analysis of the charcoal canisters is delayed at least five hours after the end of exposure and often may be delayed for much longer periods because of transportation or convenience, radon may be lost from the charcoal. Tests were performed to measure the rate of loss under different conditions and to ascertain proper procedures to prevent loss. Canisters were exposed to the concrete test slab and after being stored in the laboratory under various conditions were gamma-counted periodically to determine the effective half-life of the radon adsorbed on the canisters.

The results for canisters stored at normal ambient temperature (25°C) and pressure (760 mm Hg) are shown in Table 3. If there were no loss of radon from the canister other than by decay, the effective half-life observed would be 3.825 d (91.8 h). In all tests, the effective half-life is less than 91.8 h, indicating some loss. Canisters sealed with a tight-fitting plastic lid (2 mm thick) or with a metal lid gave an effective half-life that was not significantly different from that of radon.

The temperature was then varied over a range from 0°C to 60°C. The results are included in Table 3. Again, canisters fitted with plastic or metal lids lost the least amount of adsorbed radon.

Since on occasion the canisters were to be used in the field and returned to the laboratory for analysis by air freight, the effect of reduced pressure that might exist in an aircraft cargo hold was also investigated. Canisters were subjected to a combination of pressures and temperatures inside a large chamber that could be evacuated to 220 mm Hg, corresponding to an elevation of 31,000 feet. For this series of experiments, the exposure period in the chamber was limited to three hours to simulate an air flight of 2000 miles. Table 3 also lists these results. The maximum radon loss from canisters with lids that would occur during a three-hour aircraft flight would be less than 1%.

To test the effect of humidity on the canister's capacity for radon, pairs of canisters were exposed to 100% relative humidity for periods ranging from several hours to three days and then weighed to determine the amount of water adsorbed. The canisters were then exposed to the concrete slab along with dry canisters. The results are presented in Table 4. Only the pair of canisters exposed for 67 hours to 100% RH at 25°C show a significant difference in response (at the 95% confidence level) from the response of dry canisters. Based on these observations, it can be predicted that canisters exposed to wet soil for a period of several days will not adsorb enough water to reduce the capacity of the canisters for radon by more than 10-15%.

F. APPLICATIONS OF THE CHARCOAL CANISTERS

Applications of the canisters include investigations of spatial and temporal variations in radon flux. Figure 2 shows the spatial variation of flux from a tailings pile at an uranium mill. Flux varies by a factor of two over a distance of several hundred feet. Figure 3 shows variations of flux over one year at the same tailings pile. For the most part, variations from week to week were quite small. However, on two occasions, dramatic changes occurred. A thousand-fold reduction in flux persisted for several weeks following the formation of an ice cover on the pile and a ten-fold reduction in flux occurred immediately after a 3 cm rainfall.

The canisters can also be used indoors to locate and measure sources of radon. Adapters were built for mounting canisters in corners and across wall-wall or wall-floor joints. Radon input from these locations is often several orders of magnitude greater than that from an adjoining surface. In addition, this phenomenon is often observed for canisters mounted over cracks in a concrete foundation.

G. CONCLUSION

At least two types of activated charcoal canisters, the U. S. Army M11 and the MSA type GMA #459315, have been shown to be convenient and reliable for measuring radon flux. Presumably, other commercial charcoal canisters

would serve equally well. Because of their simplicity and low cost, the canisters may be deployed in great numbers to map the radon flux from large areas. In addition, deployment of the canisters requires much less manpower than is required to obtain measurements with accumulators. However, to attain adequate sensitivity, the canisters must be exposed for several hours to several days. The longer exposure time required with the canister yields an average (time-integrated) value of flux, rather than the series of "instantaneous" values which are possible with the accumulator.

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Table 1. Reproducibility of canister measurements

Type of Surface	No. of Measurements	Exposure Time (hours)	Radon Flux ($\text{nCi m}^{-2} \text{ min}^{-1}$)	Coefficient of Variation
Concrete Floor	10	73	5.2	0.14
NJ Soil, Loc. I	12	24-48	67	0.14
NJ Soil, Loc. II	18	6.0-28	134	0.15
Florida Soil	8	21	600	0.06
Radioactive Concrete	21	2-55	2380	0.09

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Table 2. Sensitivity (LLD) of canister method

Counting Time (min)	LLD, pCi ^{222}Rn	
	90% Confidence Level	95% Confidence Level
10	54	69
100	17	22
400	8.5	11
1000	5.4	6.9

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Table 2. Effective half-life of radon adsorbed on charcoal canisters
(^{222}Rn decay: $t_{1/2} = 91.8$ h)

Storage Conditions	Effective Half-Life (h)	Loss (%/h)
(I) <u>Room T + P</u>		
No Cover	68.1	0.26
Plastic Bag, 0.004"	82.0	0.09
Plastic, or metal lid	89.4	0.02
(II) <u>Effect of Temperature (P = 760 mm Hg)</u>		
No Cover @ 0°C	75.8	0.16
No Cover @ 60°C	37.4	1.1
Plastic Bag @ 0°C	89.4	0.02
Plastic Bag @ 60°C	63.9	0.33
Plastic, or metal lid @ 0°C	91.8	0
Plastic, or metal lid @ 60°C	88.0	0.03
(III) <u>Effect of Pressure</u>		
Plastic Bag @ 27°C, 760 mm Hg	81.1	0.10
Plastic Bag @ 27°C, 430 mm Hg	67.6	0.27
Plastic Bag @ 27°C, 220 mm Hg	67.0	0.28
Plastic Bag @ 4°C, 430 mm Hg	76.6	0.15
Plastic, or metal lid @ 27°C, 220 mm Hg	89.4	0.02

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Table 4. Effect of adsorbed water on canister capacity for radon

Exposure Time to 100% RH (h)	Absorbed Water (g)	Ratio: $\frac{\text{Radon Absorbed by Wet Canister}}{\text{Radon Absorbed by Dry Canister}}$
(I) $T = 25^{\circ}\text{C}$		
7	4.2 ± 1.0	1.06 ± 0.07
31	11.9 ± 0.8	0.96 ± 0.06
67	16.3 ± 2.3	0.86 ± 0.08
(II) $T = 20^{\circ}\text{C}$		
24	3.7 ± 0.1	0.98 ± 0.01
48	8.8 ± 1.5	0.90 ± 0.09
72	12.1 ± 0.3	0.93 ± 0.05
96	13.2 ± 0.3	0.90 ± 0.05

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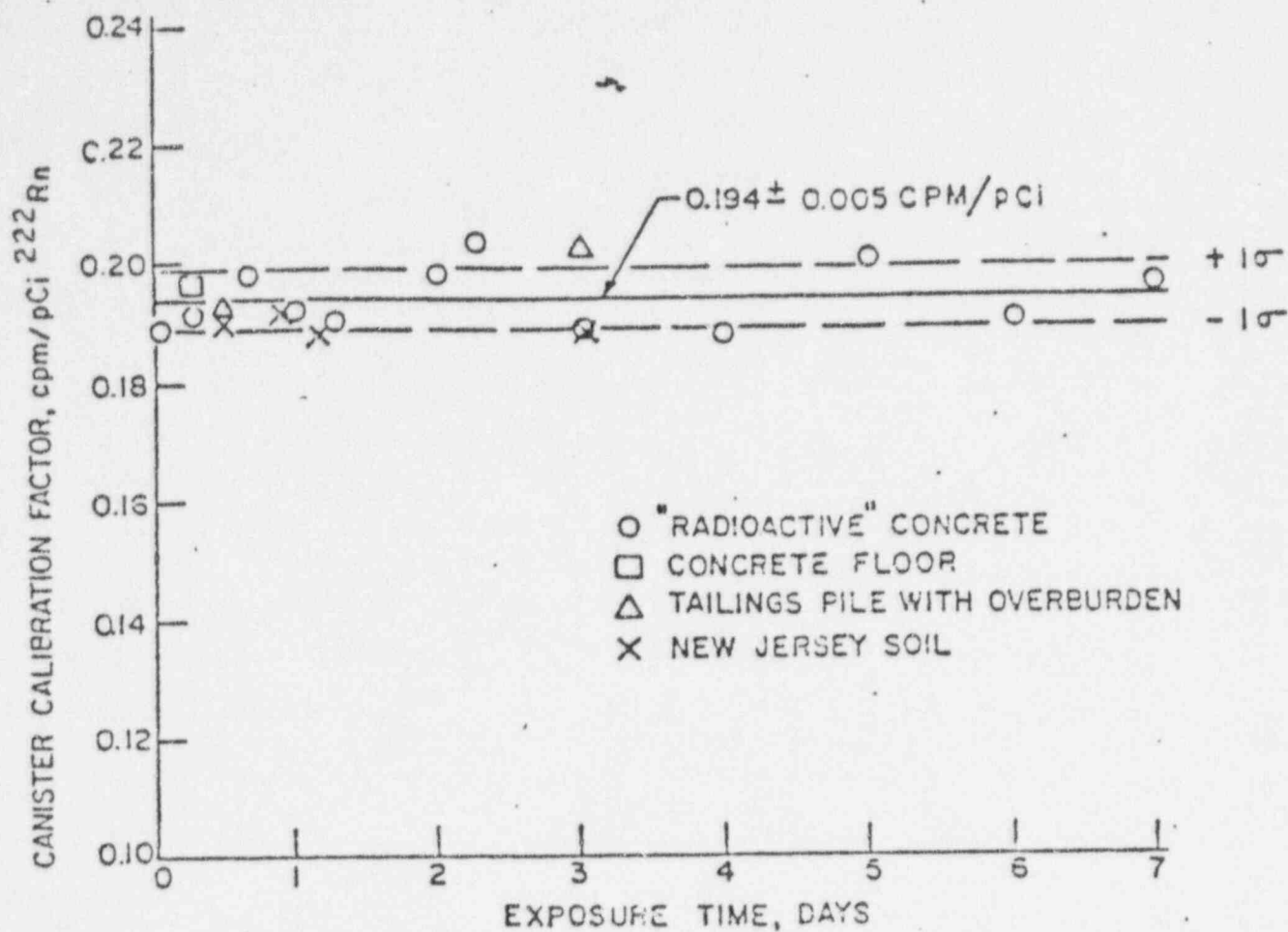


Figure 1. Canister calibration factor versus exposure time.

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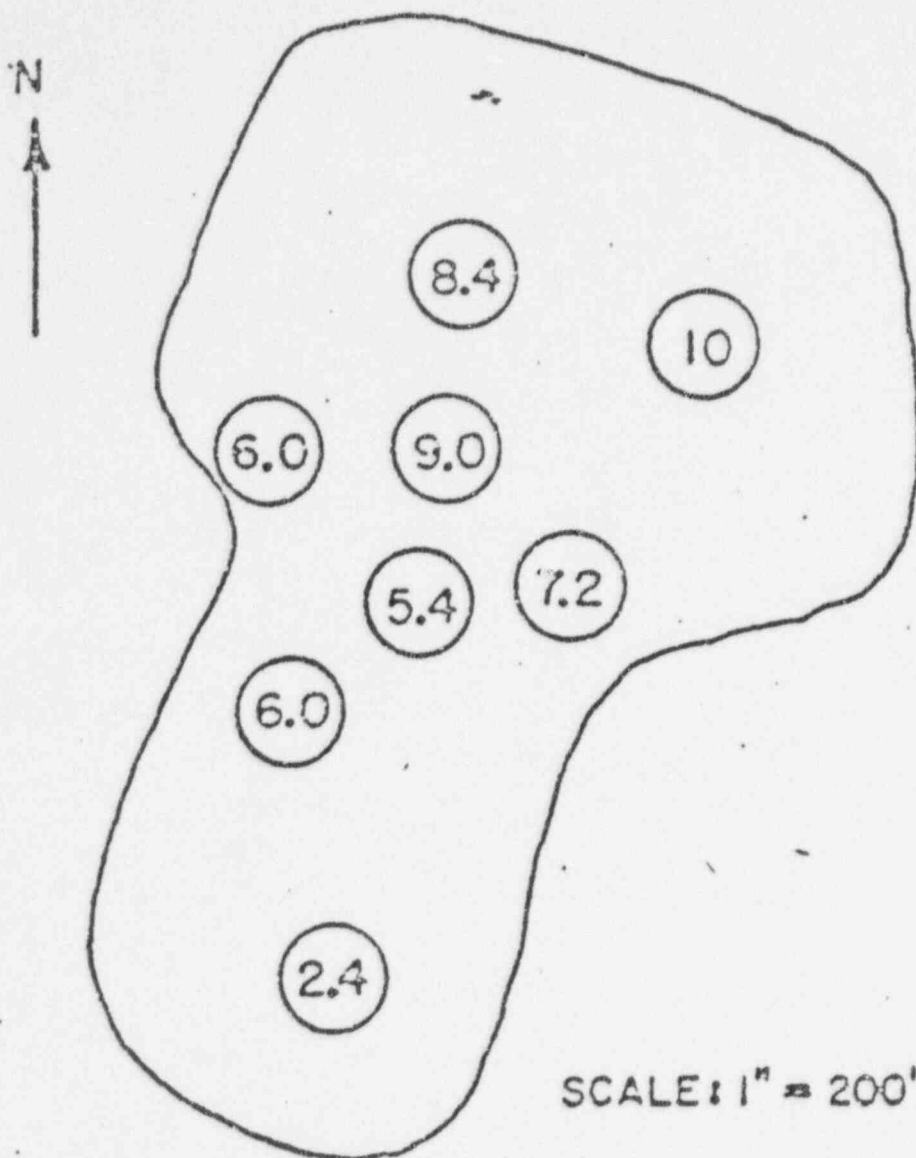


Figure 2. Spatial variation of radon flux from tailings pile (radon flux in $10^3 \text{ pCi m}^{-2} \text{ min}^{-1}$).

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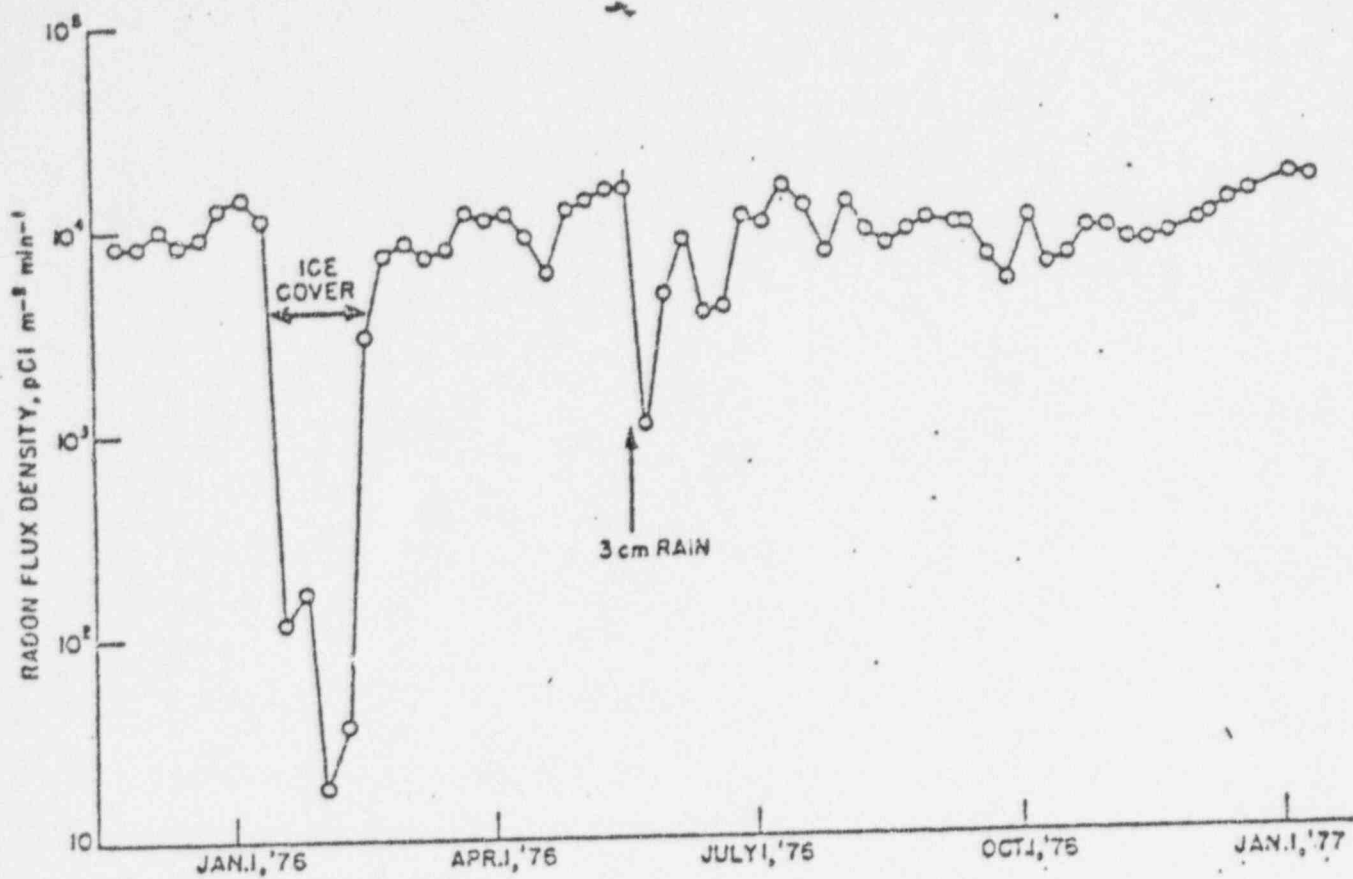


Figure 3. Time variation of radon flux from tailings pile.

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Measurement of Radon
Questions and Answers

- Q: HOLUB: What polarity is required to collect the radon daughters on the walls of the Lucas chamber?
- A: LUCAS: Radon daughters are collected on the walls when the walls are negative with respect to the electrode; radon daughters are collected on the electrode when the electrode is negative with respect to the walls.
- Q: CLINE: How do you correct for radon decay during the sampling period?
- A: SILL: We take the mid-point of the collection time as the time of collection. In a 48-hour sampling period with a radon half-life of about 4 days and using the mid-point, you will have, on the average, as much too high at the beginning as you have too low at the end of the sampling period. The point I want to make is that the error from this correction will be less than 15% under the most pessimistic assumption, and is very likely to be 2 - 3%. Further, I would like you to remember that I'm talking about a detection limit of about 0.01 pCi/l for a one-hour counting period so that this order of uncertainty is totally negligible.
- Q: ROLE: What is the need for a 10% precision flow meter when you are collecting a 20 - 50 liter sample in 48 hours and from that sample analyzing a 6 liter aliquot?
- A: SILL: The only alternative for the precision-low digital flow meter would be a meter that we could not depend on to adequately measure the flow. In operation, we collect the sample at a particular rate for the 48-hour sampling period.
- Q: FLEISHER: In your continuous digital environmental radon measuring instrument, do you detect both radium A and radium B?
- A: WRENN: Yes, there are two alpha emitting daughters (RaA and RaC) for each radon decay and you are able to measure one alpha from each, which is the theoretical upper limit in the sensitive volume. In this instrument, we get between 80 - 90% of the upper limit of response. Although the signal is developed by the alpha emitting daughters, the response is proportional to the radon.
- * COMMENT: PRICHARD: In Houston, Texas we see effects below 25% relative humidity; this is seldom the case in Texas, but might occur in places like New Mexico. In any case, above a relative humidity of 25%, the response is flat.
- Q: PRICHARD: In looking at the printout from the readings taken in Grand Junction, Colorado, most of the peaks occurred between noon and midnight. Is this correct?
- A: WRENN: It is possible. The peak periods seem to depend more on what man does rather than what nature does. There are peaks shown that can be correlated with the air changes in a house; e.g., air conditioning, open windows, etc.

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Q: GROER:

You stated that when the ground gets saturated with water, the radon and radon flux goes down. According to your experiment, is this generally true or does it change with soil type?

A: WILKENING:

Your question has to do with water saturation. When we first made measurements following a small amount of rain, we thought the radon would be lower but it wasn't. But when you get up to 80-90% of moisture in the soil, the radon flux drops down. This is true whether the water is frozen or liquid.

Q: WORTH:

What is your technique for measuring radon migration in soil?

A: WILKENING:

The work we did considered two parameters: One is molecular diffusion in the pores of the soil and the other is pressure induced transport.

The radon emanating power of soil particles is important and influences the radon concentrations in the soil gas. For this determination, we do a radium-226 analysis of the soil to obtain quantity of radium per gram. Then we take a sample of the same soil and determine the radon emitting from the soil. The ratio is the number you want.

Q: KAUTZ:

We expose canisters in duplicate and triplicate at each location, within one meter of each other. Variation in radon flux for these canisters is less than 10%. However, over a distance of 50 to 100 feet, the spatial variations may range from a factor of 2 to 10, depending on the type and condition of the soil. Numerous flux measurements should be made simultaneously on the given land mass to cancel out temporal variations in radon flux due to changes in the meteorological conditions.

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