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Proceedings of the Technical Exchange Meeting on Passive Radon Monitoring

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September 21–22, 1987
Grand Junction, Colorado
U.S. Department of Energy

Sponsored by U.S. Department of Energy
Grand Junction Projects Office
Technical Measurements Center

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Introduction

The U.S. Department of Energy, Grand Junction Projects Office, was pleased to host a Technical Exchange Meeting on Passive Radon Monitoring, held at the Rodeway Inn, Grand Junction, Colorado, September 21-22, 1987. The purpose of the meeting was to bring together a number of scientists active in the development and use of passive radon monitoring instrumentation, primarily activated charcoal detectors and alpha track detectors.

To encourage the participants to discuss their current research efforts, the submission of formal reports for publication was not mandatory. However, many of those present expressed a desire to receive copies of the viewgraphs and other materials presented. Most have supplied extended abstracts or complete reports. These materials are reproduced here as a Technical Measurements Center Report for the benefit of those attending the meeting and for others interested in passive radon monitoring. Some minor editing of the reports was necessary to facilitate reproduction.

Dowell E. Martz, Editor
Technical Measurements Center
UNC Geotech
February 1988

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SECTION A

Studies of Our Diffusion Barrier Charcoal Adsorption Detectors

Bernard L. Cohen, University of Pittsburgh

STUDIES OF OUR DIFFUSION BARRIER CHARCOAL ADSORPTION DETECTORS

Bernard L. Cohen

The Radon Project, Inc.
Physics Department, University of Pittsburgh

Our method of determining the integration time constants for the charcoal canister detectors used by the University of Pittsburgh Radon Project is shown schematically in Figure 1. Eight sets of diffusion barrier charcoal adsorption detectors (DBCA) were exposed in a low radon atmosphere for one week, with one set moved into the radon exposure chamber each day and removed at the end of a 24 hour period. This provided eight sets of detectors with each set having received a 24 hour exposure to radon at 50 pCi/l and exposure to less than 0.5 pCi/l for the remainder of the week. At the end of the week all of the sets were counted using procedures normally applied to canisters receiving the regular one week exposure.

The excess counts above normal background for each set of detectors, plotted on both linear and logarithmic scales as a function of the number of days since the 24 hour exposure occurred, are shown in Figure 2. The integration time constant for the DBCA detectors, as determined from these plots, is 3.5 days.

Application of the 3.5 day time constant to typical test house data is shown in Figure 3, where the effect of the University of Pittsburgh diffusion barrier is compared theoretically with EPA radon measurements in a residential structure. The radon concentrations in the test house, plotted at intervals of 17 hours, is indicated in the lighter curve. The effect of the 3.5 day integration time of the University of Pittsburgh DBCA detectors smoothes out these time variations, as shown by the curve labeled DBCA.

Our methods for evaluating the time averaging effect of the 3.5 day integration time of the DBCA detector is explained in Figure 4, with the actual data used in Method 3 shown in Figure 5. All of the three methods indicated in Figure 4 suggest that a one week measurement using the University of Pittsburgh DBCA detectors will result in a radon concentration value that compares with the true one month average for that residence within a standard deviation of about 15%.

The disadvantage of the University of Pittsburgh diffusion barrier detector is that the available count rate is reduced by a factor of two. As indicated in Figure 6, this is more than compensated by counting four times longer than the non-diffusion barrier detectors are counted. While this requires additional counting equipment for the same throughput, no additional labor is required. We estimate the additional equipment cost, including the cost of adding the diffusion barrier to the charcoal canister, averages about $1 per measurement.

Figure 7 shows our determination of the humidity correction factor used for the University of Pittsburgh canisters. The average humidity correction factor is 7.7% per gram of weight gain. The distribution of weight gain in detectors employed in the regular measurement service, as a function of the month when the measurement was performed, is shown in Figure 8. The error bars indicate one standard deviation. By using a humidity correction factor that depends on the month the detectors are used rather than on actual measurements of the weight gain, we obtain results that should be correct to a 3.3% standard deviation without weighing the canister.

The variation in the radon collection efficiency of the University of Pittsburgh DBCA canisters with age is indicated in Figure 9, where old canisters (lower identification numbers) are compared in side by side measurements with newer canisters (high identification numbers). The differences in the response of the newest canisters, compared with the oldest canisters, is about 5.5%. This problem is largely removed by the normal attrition rate of the canisters.

The most important source of error in radon measurements using DBCA detectors is likely to be mistakes by the technician performing the measurement. Figure 10 illustrates the University of Pittsburgh Radon Project quality control system, which eliminates most of the large errors due to technician mistakes.

Figure 11 shows some of our data on Winter/Summer ratios for the same structures in various Zip Code areas. These data seem to indicate that, at least for the northeastern area of the United States, indoor radon levels were slightly higher in Winter 1985-86 than in Winter 1986-87, and slightly higher in Summer 1987 than in Summer 1986. We would be very interested in the data from other laboratories that might shed further light on this question.

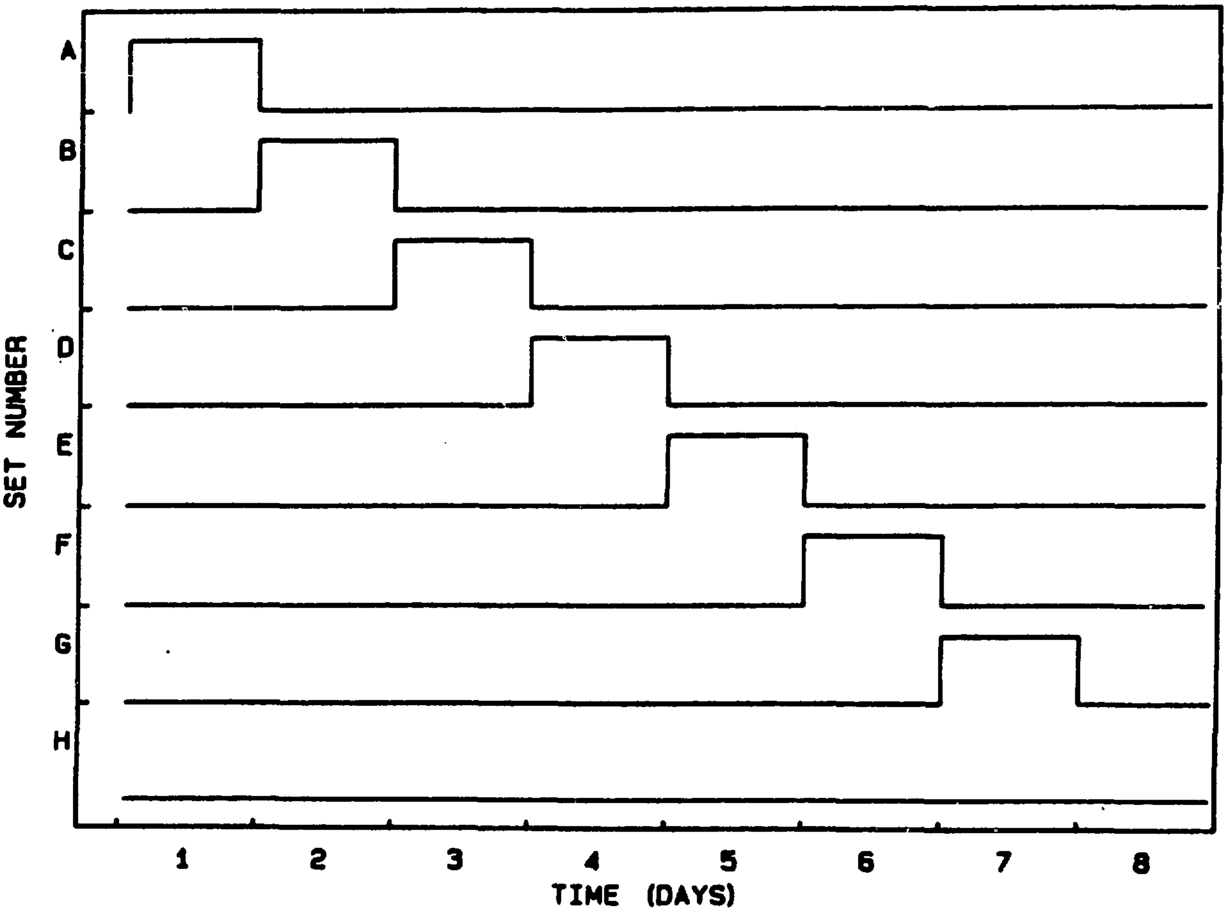

Figure 1. EXPOSURE HISTORY FOR EIGHT SETS OF CANISTERS USED FOR DETERMINING THE INTEGRATION TIME CONSTANT

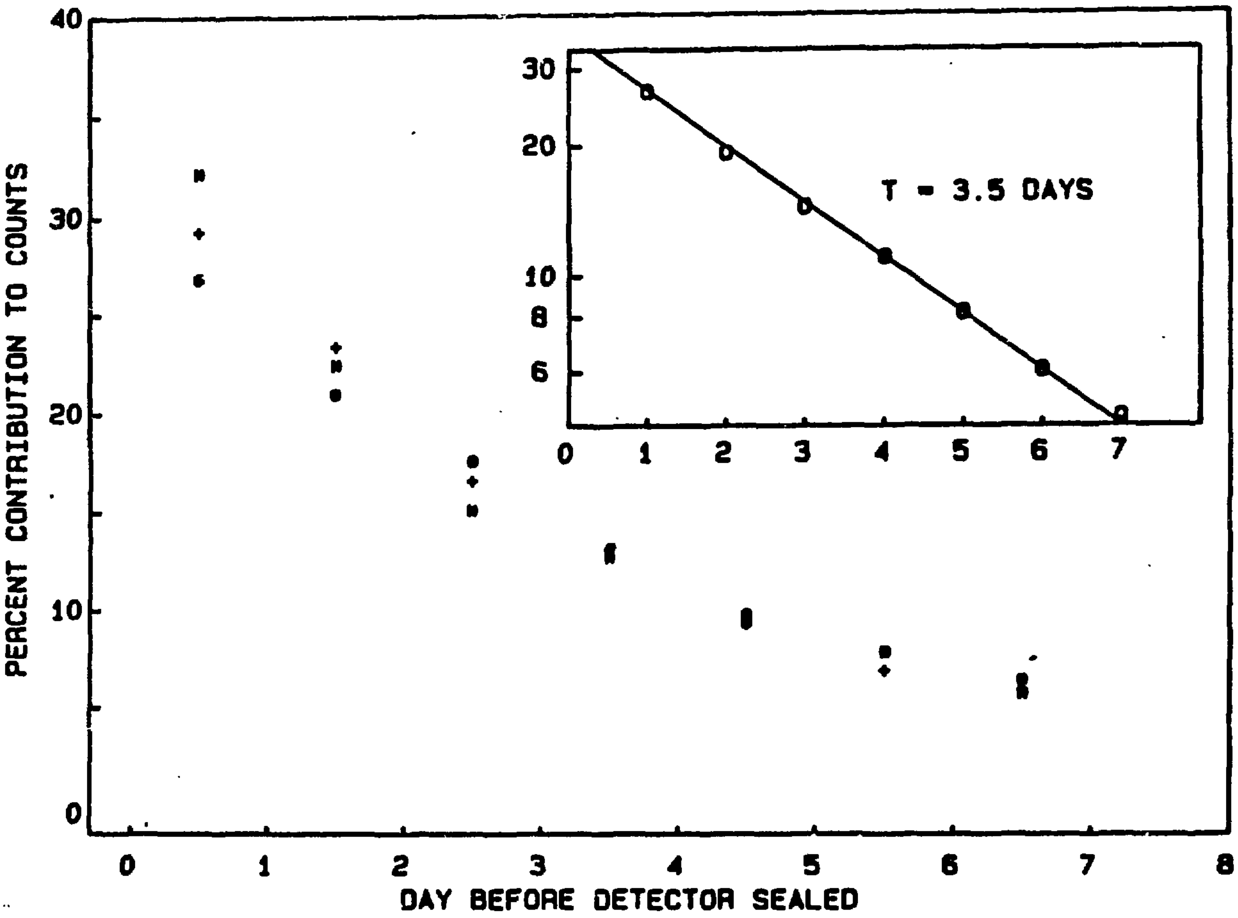

Figure 2. Plot of Percent Contribution as a Function of Time since Exposure Occurred

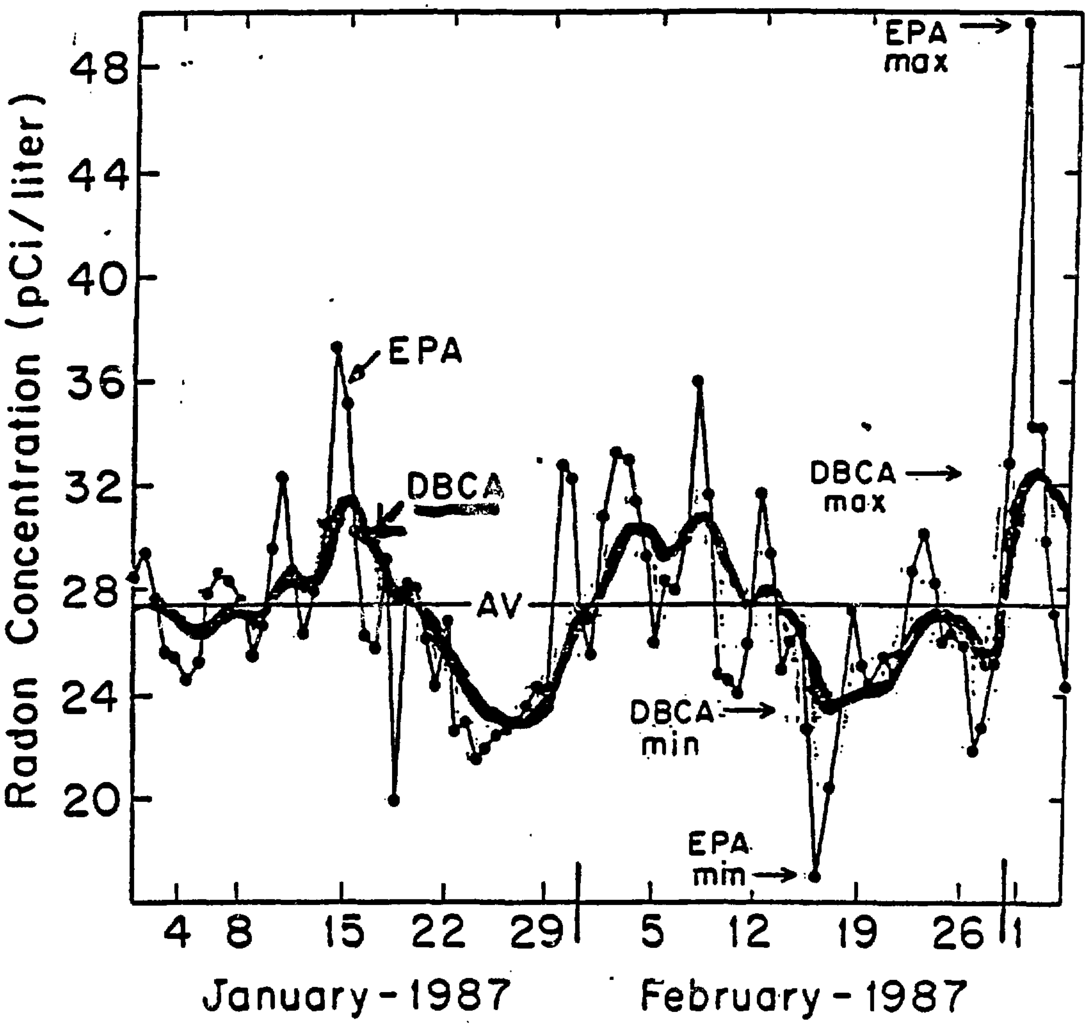

Figure 3. Effect of the 3.5 day integration time in smoothing the time variation of radon concentrations.

EVALUATION OF EFFECTIVENESS OF TIME AVERAGING

GOALS:

1. Estimate deviation of a single one week measurement from the true one month average radon concentration.

2. Express this deviation as a standard deviation, σ, where σ represents the following:
   68% probability a single measurement is within one σ of the true one month average, and 95% probability it is within two σ of the one month average.

METHODS:

1. Continuous monitoring, calculate from known response function using test house results for several different months.

   Result: $\sigma = 14\%$

2. For each of 5 houses with high radon levels, measurement each week for a month.

   Result: $\sigma = 15\%$

3. Choose high radon houses from measurement service, ask to do another (free) measurement 1 month later in same room, with same ventilation conditions. From 101 houses, all parts of US, various rooms.

   Result: $\sigma = 18\%$

   -- Some may have tried different rooms, different ventilation.

   -- Actual measurements > 1 month apart.

   ---> 18% is probably a high estimate.

CONCLUSION: $\sigma = 15\%$

Figure 4. Summary of studies used to evaluate the effect of the diffusion barrier and 3.5 day integration time on values obtained with the DBCA detectors.

Figure 5. MEASUREMENTS IN SAME HOUSE 4 - 6 WEEKS APART

(Zip - December Msmt - January Msmt)

82637 - 17.3 - 18.0
22091 - 7.1 - 8.8
17103 - 9.0 - 11.1
17603 - 9.1 - 8.3
72601 - 7.8 - 23
37027 - 4.5 - 3.4
17070 - 4.5 - 7.6
54701 - 7.6 - 10.9
07977 - 5.4 - 4.5
37064 - 6.9 - 8.8
80020 - 5.7 - 6.6
14435 - 5.2 - 10.1
17402 - 7.4 - 8.2
45414 - 5.0 - 5.0
18015 - 6.6 - 6.5
07853 - 8.9 - 9.7
17551 - 6.0 - 7.1
49509 - 7.4 - 8.7
80004 - 16.1 - 12.3
17512 - 8.0 - 7.9
21738 - 4.5 - 4.6
28021 - 16.0 - 14.9
21541 - 6.4 - 3.8
20817 - 9.7 - 10.4
08648 - 17.0 - 16.9
45429 - 6.4 - 5.3
08527 - 8.4 - 7.7
07924 - 12.6 - 12.5
07920 - 6.3 - 8.5
45305 - 4.8 - 6.2
18017 - 12.5 - 13.6
18018 - 12.5 - 8.9
21754 - 7.8 - 5.9
21754 - 6.5 - 6.8
37126 - 11.8 - 9.3
18106 - 16.4 - 16.7
49418 - 15.4 - 14.0
28021 - 8.8 - 6.7
37064 - 9.1 - 6.6
21401 - 6.1 - 6.7
28739 - 4.8 - 4.9
07801 - 16.0 - 7.7
07930 - 3.6 - 6.0
17501 - 12.5 - 9.2
80439 - 7.3 - 5.9

83814 - 9.0 - 9.7
37064 - 7.0 - 10.2
07928 - 12.6 - 11.6
72601 - 9.4 - 7.8
22186 - 4.5 - 3.2
20879 - 9.8 - 7.5
45415 - 7.6 - 6.8
37027 - 6.7 - 7.4
15217 - 5.5 - 6.0
80301 - 9.5 - 9.0
37064 - 6.8 - 5.2
20854 - 8.8 - 13.8
80517 - 7.8 - 5.4
80439 - 5.7 - 4.5
07920 - 5.6 - 7.2
18103 - 11.4 - 9.5
17339 - 10.0 - 14.1
07928 - 6.6 - 6.3
80104 - 4.8 - 4.0
19504 - 5.2 - 2.8
80013 - 12.8 - 13.7
82520 - 16.2 - 16.1
80121 - 5.2 - 5.8
17603 - 14.2 - 15.9
49001 - 5.0 - 6.5
17368 - 10.9 - 11.8
08540 - 6.0 - 3.7
13207 - 13.4 - 13.5
99021 - 5.1 - 5.0
20894 - 5.0 - 8.8
21131 - 10.8 - 9.1
18103 - 17.3 - 19.0
08833 - 12.6 - 20.6
22102 - 4.7 - 6.9
58201 - 12.3 - 14.9
18105 - 9.0 - 8.1
02159 - 8.9 - 9.0
13661 - 5.5 - 4.9
57042 - 5.5 - 6.6
07961 - 6.7 - 7.2
83467 - 5.8 - 5.4
37027 - 5.2 - 3.7
99223 - 6.7 - 10.4
60544 - 13.5 - 16.0
17601 - 6.8 - 6.8

37027 - 5.2 - 8.3
48105 - 16.7 - 18.0
07930 - 10.0 - 11.3
06441 - 6.1 - 7.4
80524 - 8.6 - 10.9
22312 - 9.6 - 9.4
61810 - 5.6 - 5.6
18064 - 14.2 - 13.8
18063 - 5.0 - 5.6
61550 - 6.0 - 6.5
80222 - 13.8 - 13.3

Average % deviation from average = 9.9%

Figure 6. EFFECT OF CUTTING SIGNAL IN HALF - AND COUNTING LONGER

S = signal
B = background
E = % of error = $\sqrt{(S + B)}/S$
C = counts = S + B

For B >> S then

E = $\sqrt{B}/S$ (approximately) Then if the signal S is cut in half one can count four times longer to double S and quadruple B, thus keeping the same E.

Examples:

C = 140, B = 100, S = 40 $\pm\sqrt{140}$
E = $\sqrt{140}/40$ = 30%

C = 120, B = 100, S = 20 $\pm\sqrt{120}$
E = $\sqrt{120}/20$ = 55%

C = 480, B = 400, S = 80 $\pm\sqrt{480}$
E = $\sqrt{480}/80$ = 27%

For B << S

E = $1/\sqrt{S}$ (approximately) Then if the signal is cut in half one can count twice as long to obtain the same total S counts and keep the same error.

Examples:

C = 500, B = 100, S = 400 $\pm\sqrt{500}$
E = $\sqrt{500}/400$ = 5.6%

C = 900, B = 100, S = 800 $\pm\sqrt{900}$
E = $\sqrt{900}/800$ = 3.8%

C = 1000, B = 200, S = 800 $\pm\sqrt{1000}$
E = $\sqrt{1000}/800$ = 4.0%

CONCLUSIONS:

- -- Intensity loss by a factor of 2 can be more than compensated by counting 4x longer
- -- This requires 4x as many counting systems, but no more labor.
  Our 22 systems cost $80,000, have been used for >80,000 measurements, no deterioration
- -- Cost/measurement << $1(+ $.15 to install diffusion barrier)

TRANSITION BETWEEN VERY LOW AND VERY HIGH HUMIDITY IN U. OF PITTSBURGH RADON CHAMBER

Ends of 1 wk exposures	Weight Gains (g)	% Change in Calib. Factor	% Change in CF Dif. in Wt Gain
1/18 --> 1/25	0.71 --> 2.88	17.3	8.0
2/22 --> 3/1	0.80 --> 4.15	21.2	6.3
3/1 --> 3/9	4.15 --> 0.30	28.2	7.3
3/9 --> 3/18	0.30 --> 3.63	19.6	5.9
3/18 --> 3/26	3.63 --> 0.45	28.7	9.0
3/26 --> 4/3	0.45 --> 3.75	41.0	12.4
4/3 --> 4/11	3.75 --> 0.45	28.9	8.8
4/18 --> 4/30	0.68 --> 4.10	16.0	4 7
6/12 - 8/7 to 7/3 - 8/28	3.2 --> 0.40	18.3	6.5
		-------	-------
		Av. = 24%	7.7%/g

Figure 7. Effect of humidity on DBCA values.

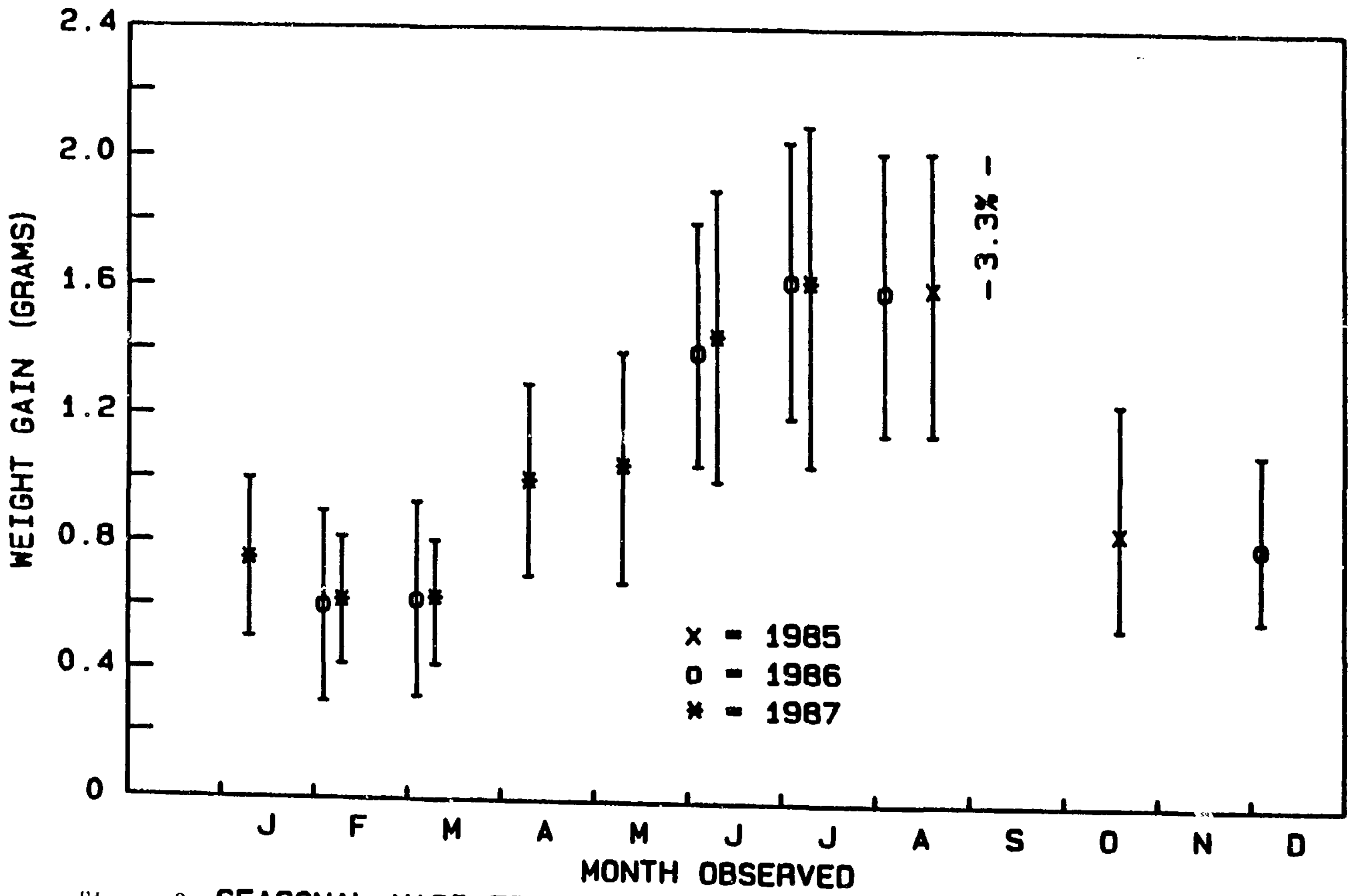

Figure 8. SEASONAL VARIATION OF WEIGHT GAIN DUE TO HUMIDITY

SENSITIVITY OF CANISTERS VS. AGE (TIMES RECYCLED)

Data from side-by-side exposure in U of P chamber

	Aver.	$\frac{\text{New}}{\text{Old}} - 1$
Feb 13 - 20 (>10 recycles for old)		
Old - Can No. 975, 1122, 1305, 5720	23.3	8.2%
New - Can No. 23729, 23732, 23736, 23737	25.3	
Feb 23 - Mar 2 (> 10 recycles for old)		
Old - Can No. 3040, 3890, 5354	26.4	6.7%
New - Can No. 23733, 23734, 23738	28.2	
March 20 - April 6 (> recycles for old)		
Old - Can No. 2357, 2613, 3629	29.1	6.7%
New - Can No. 26188, 26189, 26193	30.8	
April 6 - April 13 (old had only 2 - 3 recycles)		
Old - Can No. 1293, 3520, 3797, 4296	40.7	0.2%
New - Can No. 26184, 26185, 26190, 26192	40.8	

Av = 5.5%

Data from EPA-RMP Round 3

Can No.	0-5K	5-8K	8-12K	12-14K	14-16K	16-17K	17-18K
Ratio	0.97	0.97	0.98	1.00	1.04	0.99	1.06

Δ = 5.0%

Data from EPA-RMP Round 4

Can No.	0-5K	5-10K	10-15K	15-20K	20-25K	25-30K
Ratio	0.98	1.01	1.03	1.02	1.06	1.04
Samples	7	10	14	20	20	22

Δ = 5.6%

Average = 5.5%

Figure 9. Sensitivity of DBCA detectors versus age.

COMPUTER (CMPT) CHECKS

1. Can No. copied correctly on sheet & into CMPT
   -- verification code (e.g., 128735 RGX) checked

2. Can No. on sheet vs measuring system (MS) No. OK
   -- ascending numerical order checked

3. Can in MS specified on sheet
   -- as cans removed, verif. code on can vs MS no. typed into CMPT, checked vs sheet

4. Transcription errors and "date sealed" (for decay correction)
   -- Can No., pCi/L, & "date sealed" on sheet typed into CMPT by typist, checked vs info from measurement

5. Instrument problems, knob bumped, very large statistical fluctuation
   -- pCi/L from different γ-ray peaks checked for consistency, if >2.5 SD, remeasured

6. Data for corrections cannot cause large error
   -- Temp, time between sealing and counting, & days exposed in reasonable range

SUPERVISOR CHECKS

a. Can No. written by client = Can No. used in MS

b. CMPT warnings on #5 & #6 above

c. Technician errors (#1, #2, #3 above) recorded on MS print-out

d. Anything abnormal flagged by technician

e. Date, time backgrounds sensitivities in CMPT
   <u>not necessary to check</u>

-- Result not sent to correct client

   -- mailing label by client, part of same sheet

Figure 10. Summary of computer and operator checks on potential data handling errors.

WINTER/SUMMER RATIOS VS GEOGRAPHICAL AREA

Living Areas of Homes - i.e., not in basements

Zip Codes	W85-86 S86	W86-87 S86	W86-87 S87
010-070	1.89	1.26	0.96
070-090	1.63	1.29	1.29
090-150	1.79	1.63	1.20
150-197	1.74	1.37	1.61
197-247	1.18	1.13	0.82
247-300	(1.15)	(1.74)	2.07
300-400	(1.58)	1.68	.149
400-500	1.62	1.19	0.67
500-600	(2.60)	1.98	(0.82)
600-630	----	1.10	----
630-700	----	(1.09)	----
700-800	----	(1.08)	----
800-840	1.47	2.02	(1.42)
840-900	(1.76)	----	----
900-990	(1.65)	1.46	(1.46)
AVER -	1.61	1.46	1.26
INC.() -	(1.67)	(1.43)	(1.26)

At least 100 measurements <u>in each season</u> ( ) --> 40-100 measurements in one or both.

Figure 11. Some historical data used to evaluate quality control.

SECTION B

A Comparison of Laboratory and Field Measurements of Radon

L. D. Michaels and A. S. Viner, Research Triangle Institute
T. Brennan, Camroden Associates

A COMPARISON OF LABORATORY AND FIELD MEASUREMENTS OF RADON

L. D. Michaels and A. S. Viner
Research Triangle Institute

T. Brennan
Camroden Associates

Charcoal canisters have found wide application as simple, inexpensive, passive devices for the monitoring of radon in homes. However, these devices are not ideal to this application since desorption of the gas from the charcoal occurs during sampling. Consequently, in homes where large variations in radon concentration occur over a short period of time, charcoal canisters may be inappropirate sampling devices.

Ten homes in Clinton, New Jersey were selected for the demonstration of radon reduction techniques in the spring of 1986. Premitigation monitoring was performed in the basement of each house using a Pylon AB-5 continuous radon monitor fitted with a PRD passive radon detector. Figure 1 shows the results of this monitoring in two different homes monitored simultaneously. The upper curve shows a home that was vacant and closed up during the first 3 days of monitoring and then lived in under normal conditions, while the lower curve shows radon concentration in a home that was occupied under normal living conditions for the full 5-1/2 days of monitoring. Diurnal variations over one or two orders magnitude were commonly encountered when measurements were made in these ten homes.

A chamber experiment was designed to simulate diurnal variations under controlled conditions in order to test the ability of charcoal canisters to accurately predict the average radon concentration in the chamber for exposures ranging from 24 to 96 hours. Charcoal canisters used in both chamber and field experiments were the standard open-faced US EPA type canisters. Figure 2 is a schematic diagram of the 2 m^3 chamber used in this experiment. Radon concentration, temperature, and relative humidity were held constant in the chamber. Canisters were moved from the chamber to the adjoining room in 12-hour cycles. The radon concentration in the room was also monitored using a Pylon AB-5 continuous monitor. Figure 3 shows the radon concentration versus time as experienced by the canisters under these conditions. A ratio defined as the charcoal canister monitoring results in pCi/l divided by the average continuous monitor results for the canister sample duration was calculated. The results are shown in Figure 4. The shaded portion of the plot corresponds to a value of ±25 percent of the continuous monitor average. This is the range of variability required in the US EPA radon monitoring proficiency program [EPA87]. The dashed curve shows the same ratio calculated for canisters continuously exposed to radon in the chamber. The results of the experiment which simulated the indoor radon variations encountered in Clinton indicate that the canisters significantly underestimate radon concentrations for exposure times greater than approximately 48 hours, while canisters exposed to a constant concentration of radon in the chamber performed within the guidelines for all but the longest sampling period.

Field measurements made in the Clinton homes using charcoal canisters collocated with continuous monitors were analyzed in the same manner. The results of 25 paired data points are shown in Figure 5. The data show a wide scatter with no significant time-dependent correlation. Figure 6 shows the mean value of the field data for each exposure period together with the results of chamber experiments previously discussed. The accuracy of the field data is within the recommended bounds for all but 24-, 72-, and 120-hour exposures.

While charcoal canisters can be extremely precise predictors of average radon concentrations in homes, under conditions of varying radon concentration, their ability to accurately predict average indoor levels of radon is questionable. Based on theory, when desorption occurs, significant underpredictions are possible [Co84, Na87]. Since the patterns of radon concentrations in a given home are not known *a priori*, the design and use of charcoal canisters deserves close analysis.

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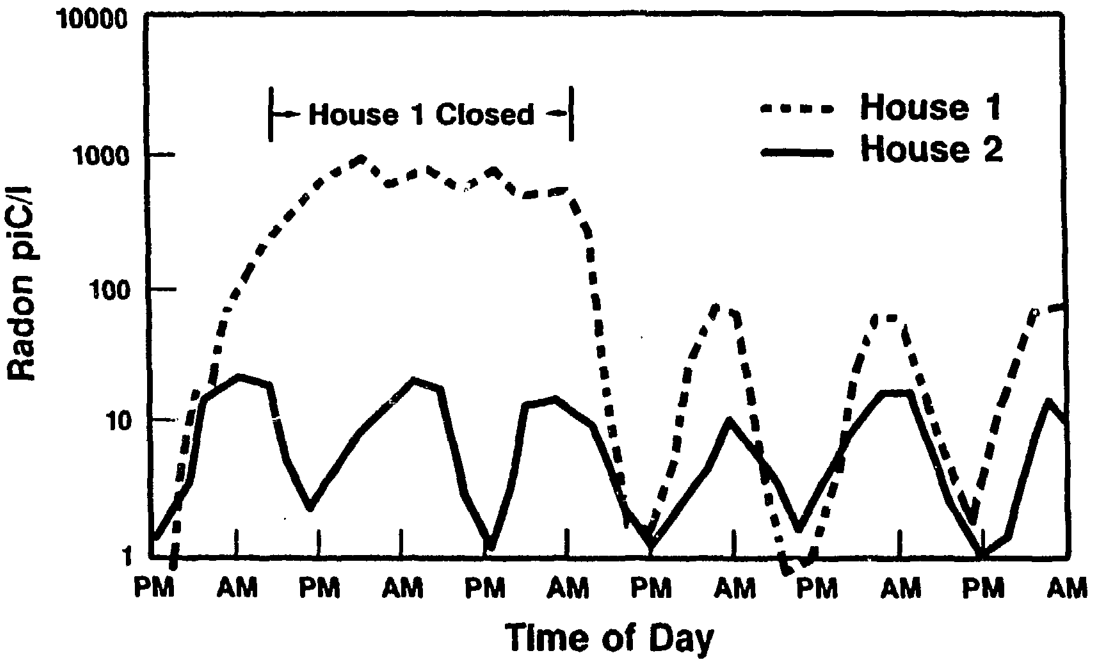

Figure 1. Simultaneous continuous radon monitoring results in two homes in Clinton, New Jersey.

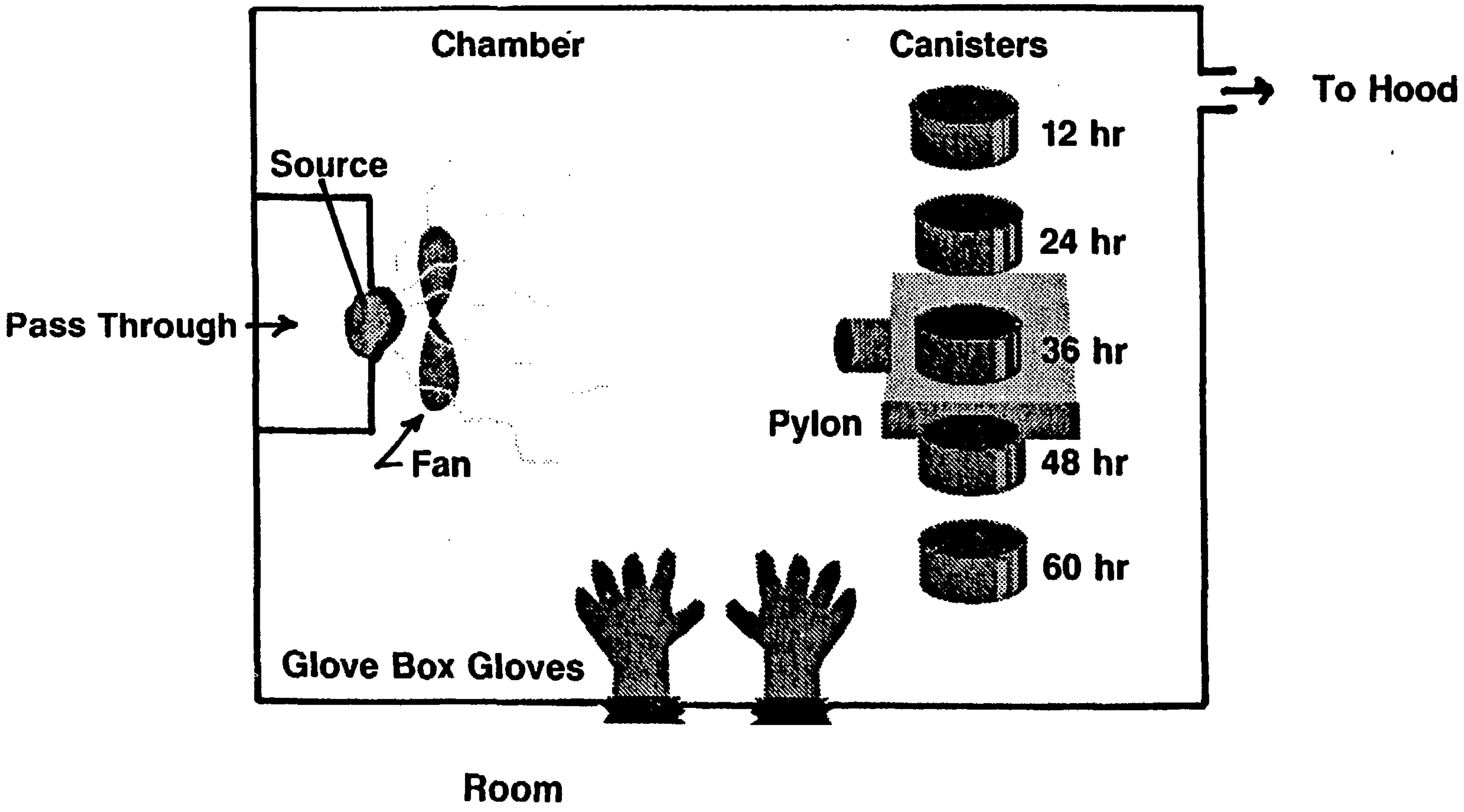

Figure 2. Experiment #1. Constant radon concentration. Schematic diagram of exposure chamber and constant radon concentration experiments.

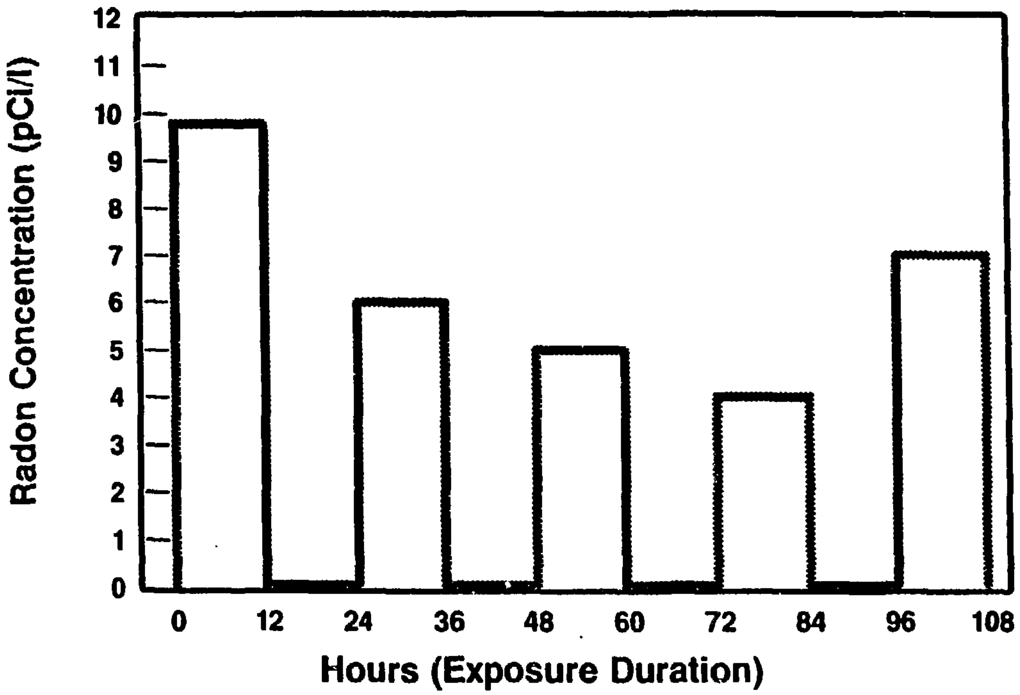

Figure 3. Radon concentration experiment #2. Varying concentration.

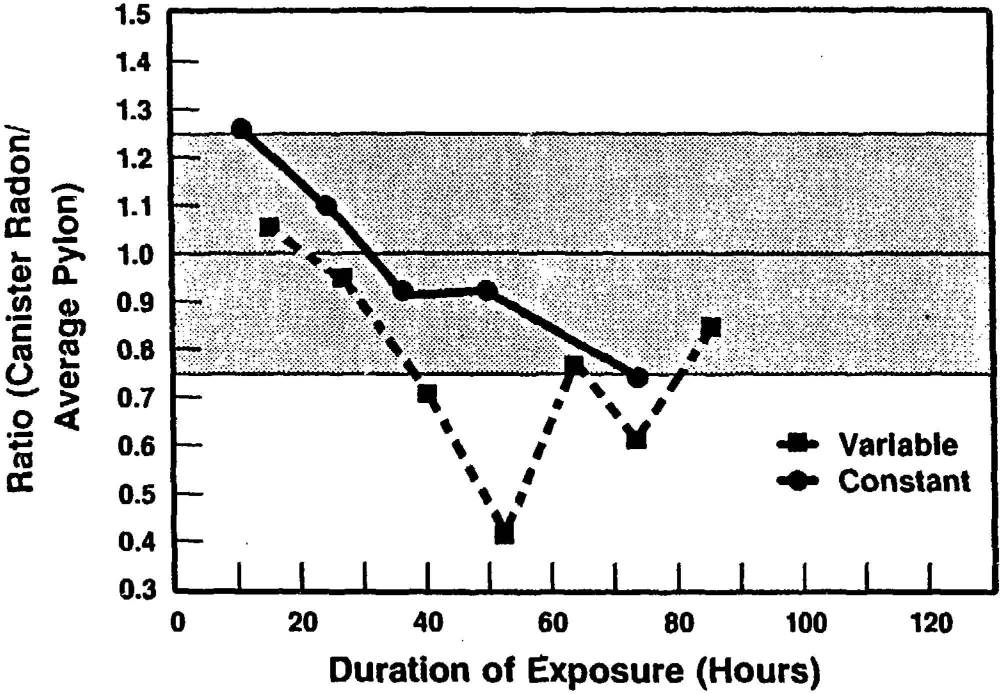

Figure 4. Chamber Experiment Results Plotting the Ratio of Canister to Continuous Radon Concentration Measurement.

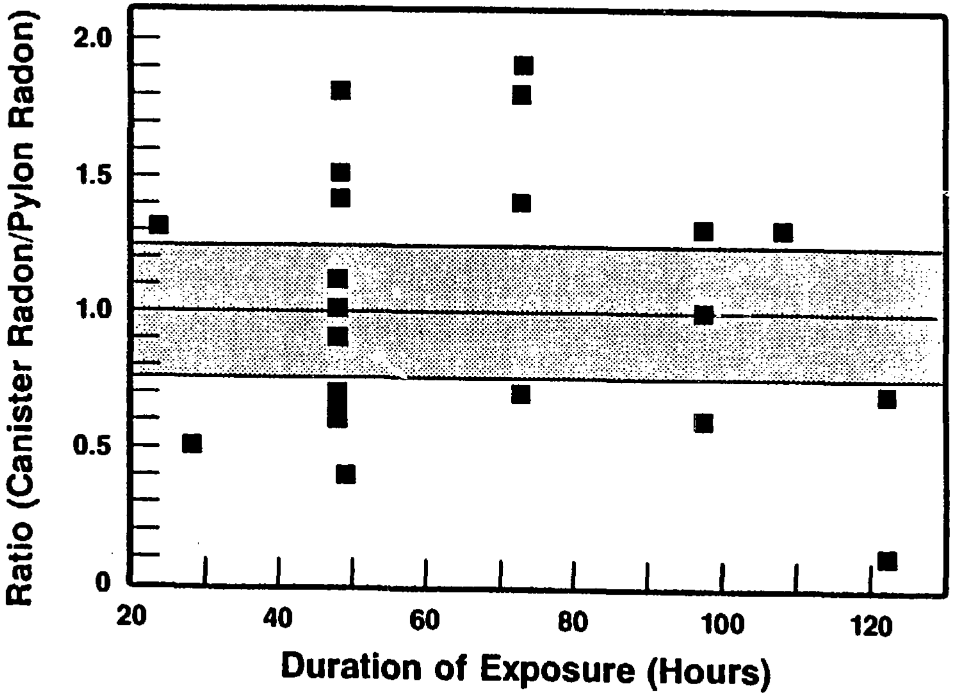

Figure 5. Ratio of canister to Pylon radon concentration measurement.

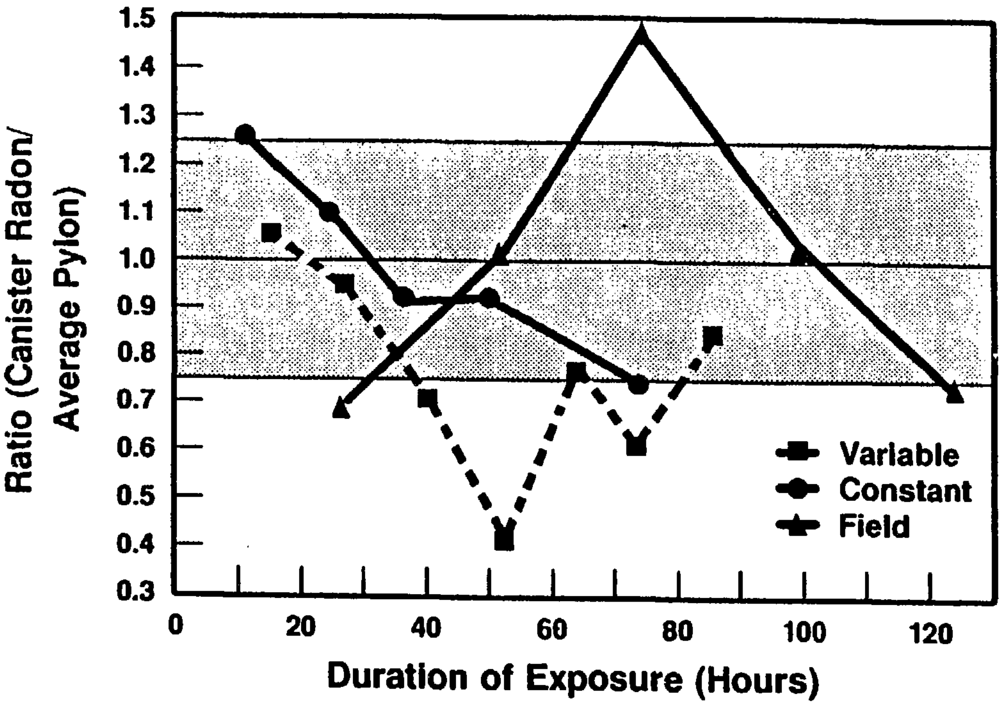

Figure 6. Comparison of Field and Chamber Results Plotting the Ratio of Canister to Continuous Radon Concentration Measurement.

SECTION C

The Response of Charcoal Canister Detectors to Time-Variant ^{222}Rn Concentrations

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Technical Measurements Center,
Grand Junction, CO. Sept. 21-22, 1987.

LBL-*****

The Response of Charcoal Canister Detectors to Time-Variant ^{222}Rn Concentrations

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INTRODUCTION

Utilizing charcoal adsorption to collect Rn in a canister has proven to be an easy and inexpensive way to get quick diagnostic measurements of ^{222}Rn levels. They are well suited to γ-ray counting and typically need to be exposed for only a few days, unlike α-track devices with recommended exposure times of at least a month. For these reasons, charcoal canisters seem to be preferred over other passive samplers for use in measurements requiring rapid reporting of results.

But because Rn desorbs from charcoal as well as it adsorbs onto it, some have expressed concern that the canisters are more affected by Rn levels at the end of the exposure than by the concentrations at the start. In one case, charcoal canister readings have differed by about a factor of four when they should have read the same mean concentration (Ge84). The manner in which these charcoal canisters respond to time-variant Rn levels is especially significant in light of the evidence that typical Rn concentrations undergo diurnal as well as seasonal variations which can vary by as much as an order of magnitude.

In response to this problem, Cohen and Nason (Co86) have described a canister which utilizes Prichard and Marien's concept of a "diffusion barrier" (Pr85). Rather than taking the whole lid off for a measurement and exposing all of the charcoal face to the environment, a small hole in the lid is opened which effectively limits the desorption, or back-diffusion, of Rn from the canister into the outside environment, while still allowing enough Rn adsorbtion onto the charcoal to be easily counted. Its designers claim that the new sampler has an integrating time constant of over three days; however, a comparison of the response of these "diffusion-limited" canisters to rapid variations in Rn levels with that of their "open face" counterparts requires further study.

MATHEMATICAL MODEL---OPEN FACE CANISTER

As described by Cohen (Co83), analysis of an open face canister having a charcoal bed depth of l cm (see Fig. 1) consists of solving the one-dimensional diffusion equation.

$$\frac{\partial y}{\partial t} = D\frac{\partial^2 y}{\partial x^2} - \lambda y, \quad y = f(x,t). \tag{1}$$

where $0 \leq x \leq l$ is the height above the bottom of the canister, $y(x,t)$ the concentration of ^{222}Rn in the charcoal in pCi/cm^2, t the time in seconds, D is the diffusion constant, and λ is the radioactive decay constant for ^{222}Rn.

Assuming that the lid is taken off at time zero, and that the canister previously had no ^{222}Rn in it beforehand, the initial condition is then

$$\text{at } t = 0, \ \ y = 0 \text{ for } 0 \leq x \leq l. \tag{2}$$

Then for the case when the bottom of the canister is impermeable, and the top is exposed to the Rn in the environment, the boundary conditions are

$$\text{at } x = 0, \ \ \frac{\partial y}{\partial x} = 0, \tag{3}$$

and

$$\text{at } x = l, \ \ y = k\rho C(t), \tag{4}$$

where k is the adsorption coefficient of charcoal, ρ the measured density of charcoal including porosity, and $C(t)$ is the concentration of Rn in the air as a function of time.

It would be impossible to solve Eq. (1) for any general function $C(t)$, but one should expect that any actual $C(t)$ can be reasonably approximated by a series of linear functions. Thus, if T is the total time of the exposure, then $[0,T]$ can partitioned into $[t_0 = 0,\ t_1,\ldots,t_i,t_{i+1},\ldots,t_{n-1},t_n = T]$ such that

$$C(t) \approx C_i + m_i t, \ \ \text{for} \ \ t_i \leq t \leq t_{i+1}. \tag{5}$$

Then, for $t_i \leq t < t_{i+1}$, Eq. (1) needs to be solved with the generalized initial condition

$$\text{at} \quad t = t_i, \ \ y = g_i(x), \tag{6}$$

where $g_0(x) = 0$, and also with the simplified boundary conditions,

$$\text{at} \quad x = 0, \ \frac{\partial y}{\partial x} = 0, \tag{7a}$$

$$\text{at } x = l, \ y = C_i + m_i t. \tag{7b}$$

First, a few substitutions:

$$t' = D\frac{t-t_i}{l^2}, \ x' = \frac{x}{l}, \ \lambda' = \frac{l^2\lambda}{D}, \tag{8a-c}$$

and

$$m'_i = \frac{l^2 m_i}{D}, \ \text{for} \ \ i = 0,1,\ldots,n. \tag{8d}$$

One can now express Eq. (1), initial condition (6), and boundary conditions (7a,b) in terms of the new variables t', x', λ', and m'_i as the differential equation,

$$\frac{\partial y}{\partial t'} = \frac{\partial^2 y}{\partial x'^2} - \lambda' y, \tag{9}$$

with initial condition

$$\text{at} \quad t' = 0, \ y = g_i(x'), \tag{10}$$

and with boundary conditions

$$\text{at} \quad x' = 0, \ \frac{\partial y}{\partial x'} = 0, \tag{11a}$$

$$\text{at} \quad x' = 1, \ y = C_i + m'_i t'. \tag{11b}$$

Since this is a *linear* differential equation, consider Eq. (9) with $y_1(1,t') = C_i$ as the second boundary condition instead of (11b). Separation of variables then yields a solution (Cr75):

$$y_1(x',t') = C_i + \sum_{j=1}^{\infty} \left[A1_j e^{-(\gamma_j^2+\lambda')t'} + A2_j\right] \cos \gamma_j x', \tag{12}$$

where

$$\gamma_j = (j - \frac{1}{2})\pi, \tag{13a}$$

$$A1_j = \frac{2(-1)^j \gamma_j C_i}{\gamma_j^2 + \lambda'} + 2\int_0^1 g_i(x') \cos \gamma_j x' dx', \tag{13b}$$

$$A2_j = \frac{2(-1)^j \lambda' C_i}{\gamma_j(\gamma_j^2 + \lambda')}. \tag{13c}$$

Now, to solve Eq. (9) with the boundary condition $y_2(1,t') = m'_i t'$, consider the simplified differential equation,

$$\frac{\partial y'_2}{\partial t'} = \frac{\partial^2 y'_2}{\partial x'^2}, \tag{14}$$

with initial condition

$$\text{at} \quad t' = 0, \ \ y'_2 = 0, \tag{15}$$

and with boundary conditions

$$\text{at} \quad x' = 0, \ \ \frac{\partial y'_2}{\partial x'} = 0, \tag{16a}$$

$$\text{at} \quad x' = 1, \ \ y'_2 = \frac{m'_i}{\lambda'}\left(e^{\lambda' t'} - 1\right) \tag{16b}$$

Using separation of variables, one arrives at the following solution:

$$y'_2(x',t') = \frac{m'_i}{\lambda'}\left[e^{\lambda' t'}\frac{\cosh\sqrt{\lambda'}x'}{\cosh\sqrt{\lambda'}} - 1\right] + \sum_{j=1}^{\infty} B_j e^{-\gamma_j^2 t'} \cos\gamma_j x' \tag{17}$$

where γ_j is the same as in Eq. (13a) and

$$B_j = \frac{2(-1)^{j-1} m'_i}{\gamma_j(\gamma_j^2 + \lambda')}. \tag{18}$$

Then, the following transformation,

$$y_2(x',t') = \lambda' \int_0^{t'} y'_2 e^{-\lambda' t'} dt' + y'_2 e^{-\lambda' t'}, \tag{19}$$

yields $y_2(x',t')$ as a solution to Eq. (9) with initial condition

$$\text{at} \quad t' = 0, \ \ y_2 = 0, \tag{20}$$

and with boundary conditions

$$\text{at } x' = 0, \ \frac{\partial y_2}{\partial x'} = 0, \tag{21a}$$

$$\text{at } x' = 1, \ y_2 = m'_i t'. \tag{21b}$$

Finally, from superposition, adding $y_1(x',t')$ and $y_2(x',t')$ yields the desired final solution,

$$y(x',t') = C_i - \frac{m'_i}{\lambda'} + \frac{\cosh\sqrt{\lambda'}\,x'}{\cosh\sqrt{\lambda'}}\left[m'_i t' + \frac{m'_i}{\lambda'}\right] + \sum_{j=1}^{\infty}\left[A1'_j e^{-(\gamma_j^2+\lambda')t'} + A2'_j\right]\cos\gamma_j x', \tag{22}$$

where

$$\gamma_j = (j - \frac{1}{2})\pi, \tag{23a}$$

$$A1'_j = \frac{2(-1)^j\gamma_j}{\gamma_j^2 + \lambda'}\left[C_i - \frac{\gamma_j m'_i}{\gamma_j^2 + \lambda'}\right] + 2\int_0^1 g_i(x')\cos\gamma_j x' dx', \tag{23b}$$

$$A2'_j = \frac{2(-1)^j\lambda'}{\gamma_j(\gamma_j^2 + \lambda')}\left[C_i - \frac{m'_i}{\gamma_j^2 + \lambda'}\right] \tag{23c}$$

and one can always substitute back to get the solution in terms of the original variables t, x, λ, and m_i.

Although the solution calls for infinite sums to be evaluated, one sees that for $j > 100$, the terms become very small indeed. Fourier orthogonality can also be used to evaluate the integral in Eq. (23b) and speed up the calculations. Using this orthogonality, errors which might have crept into the calculations for the time interval $[t_i, t_{i+1}]$ will not be magnified in the calculations for the time interval $[t_{i+1}, t_{i+2}]$.

Hence, knowing $y(x,t)$ for all t, the total amount of ^{222}Rn in the canister at any time T_0 can be found by integrating $y(x,T_0)$ from 0 to l, *i.e.*

$$\text{Rn in Canister at time } T_0 = \int_0^l y(x,T_0),dx. \tag{24}$$

MATHEMATICAL MODEL—DIFFUSION BARRIER CANISTER

The most rigorous way of modeling a diffusion barrier canister such as Cohen's would be to solve the diffusion equations along both the height and radial axes. But rather than devising a finite difference method or some other numerical approximations to solve such complex two-dimensional differential equations, the picture of the diffusion barrier was simplified somewhat in order to utilize the one-dimensional solution derived above (See Fig. 2). By assuming air in the immediate space above the charcoal bed is well mixed, an iterative process can be used to deduce the concentration of Rn in this well-mixed air for different times during the exposure.

Assuming a well-mixed layer of air above the charcoal, the main differential equation which describes the concentration of Rn in that layer of air is,

$$\frac{\partial C_i}{\partial t} = \frac{1}{V}\left[P_1(C_o - C_i) - \frac{\partial Q_{Rn}}{\partial t}\right] - \lambda C_i, \qquad (25)$$

where

A_{db} is the area of the diffusion barrier;

C_i is the Rn concentration in the layer of well-mixed air;

C_o is the Rn concentration in the outside air;

D_{air} is the diffusion coefficient of Rn in air, about 0.12 cm^2/s at room temperature (Co83);

L_{db} is the length of the diffusion barrier;

$P_1 = \dfrac{D_{air}A_{db}}{L_{db}}$;

Q_{Rn} is the amount of Rn in the charcoal bed;

V is the volume of well-mixed air; and

λ is the decay constant for Rn.

But Q_{Rn} is a function of C_i, while in turn, C_i is a function of Q_{Rn}. This makes it very hard to solve Eq. (25) exactly, but again if the total exposure time is partitioned into small enough time intervals, $C_i(t)$ will look nearly linear in each interval. Hence, knowing that $C_i(t) \approx C_{ij} + h_j t$, the differential equation can again be solved. This solution can then be implemented on a computer to predict the amount of Rn found in the canister after any specified Rn concentration profile.

EXPERIMENTS--DIFFUSION OUT

Before these models can be verified and put into use, various parameters such as diffusion coefficients, adsorption coefficients, and diffusion barrier dimensions have to be known more exactly. For this set of experiments, three different types of canisters were used. The "EPA" canister is essentially an open face type detector holding 70 g of charcoal having a bed depth of 1.8 cm and a cross-sectional diameter of 10 cm (Ro87). The "COH" canister is the diffusion barrier type designed by Cohen and Nason. It holds 25 g of a fine mesh charcoal having a bed depth of about 1.4 cm. The diffusion barrier consists of a hole with a 1.9 cm diameter cut into the lid and sits 1.0 cm above the charcoal bed. A nylon screen and desiccant taped beneath this hole also serves to impede the diffusion of Rn into the canister somewhat (Co86). Finally, non-commercial modifications were done on the EPA canisters to make them diffusion-limited. These modified canisters are henceforth called "LBL" and were made by cutting a hole in the lid and soldering a piece of copper tubing over the hole. It was hoped that by having the canister look almost exactly like Fig. 2b, parameters such as L_{db}, A_{db}, and V would be known exactly and one could test the model, using these parameters, against the experimental results.

The parameters for these various detectors, both open face and diffusion barrier, were determined by first loading the canister with ^{222}Rn and then watching how fast it desorbed from the canister in a space with low Rn concentrations over a period of a few days. In order to assure a uniform distribution of Rn within the charcoal at the start of each run, the canisters were taken apart and the charcoal spread out on thin beds in a room laden with Rn in order attain almost instantaneous equilibrium between the air and charcoal. At least three hours was given for the Rn daughters, particularly ^{214}Bi, to grow in. An initial count was taken with a NaI γ-ray detector; then the lid or hole was opened and the analyzer was set to cycle in order to take readings at various time intervals. These experiments were done with a 3 x 3 NaI detector in a low-background lead-shielded counting enclosure, through which air was circulated. Looking at the counts with energies above 506 keV and below 712 keV yielded the number of counts beneath the 609 keV peak, giving a number which should be proportional to the activity of the ^{214}Bi at that time.

The data obtained from use of these canisters are shown in Figs. 3 - 5. These data and the fits obtained from the appropriate model are discussed below.

EXPERIMENTS--TIME VARIANT CONCENTRATIONS

This set of experiments initially tried to accomplish two goals---to calibrate the canisters and to show how the canisters responded to Rn profiles that went from high to low concentrations and vice-versa. Also a fourth type of charcoal canister was used in these runs, labeled "FLT" for flow-through. This canister actually contained charcoal sandwiched between two essentially open faces, rather than having one side impermeable as with the EPA canister. Mathematically, we treated this situation as equivalent to two regular open face canisters glued back to back. Hence, the open face model can still be used for predicting the response of the FLT canister by using a bed depth parameter that is half the actual height of the canister.

Two rooms were used in these experiments, one with a high ^{222}Rn content, about 200 pCi/L, and the other with a lower ^{222}Rn content, about 30 pCi/L. The actual ^{222}Rn concentrations were measured over time using continuous radon monitors. It was hoped that the concentrations in both rooms could be maintained at nearly constant levels and thereby provide a calibration standard. But the room ventilation rates were to too high for this and the Rn concentrations turned out to be time-variant instead as shown in Fig. 6. Several canisters were switched from room to room during the middle of the exposure to expose them to high--low, and low--high variations.

Calibration of the various detectors was achieved by using the parameters determined in the diffusion out experiments. Thus, the actual mean Rn concentration value could then be compared to the concentrations as determined by the various charcoal canister devices, and these measurements could then, in turn, be compared with the model predictions.

RESULTS AND DISCUSSION

In order to determine the effective diffusion coefficients for radon in charcoal the diffusion out experiments were conducted for both the EPA and COH devices. In the latter case, the lid containing the diffusion barrier was removed from the can, providing an open-face canister from which the radon removal rate would be controlled solely by $D_{charcoal}$, as in the case of the open-face EPA devices. As shown in Fig. 3, the open face model tends to predict diffusion out at a higher rate than exponential immediately after the lid is opened. Although the experimental values for the actual EPA canister run do not agree exactly with the model's predictions for the first five hours, it should be noted that by looking carefully at Fig. 3 that the initial data points do not lie on a straight line as the later data appear to do. This seems to indicate that the prediction of diffusion out faster than exponential at the very start is in fact correct. One possible reason as to why the open face model doesn't entirely agree with the actual experimental values is that the metal lip around the face of the charcoal bed might act as somewhat of a diffusion barrier. Another possible reason is that the boundary condition, Eq. (4) might not be entirely true. There might well be some sort of time lag between the Rn concentration at the top layer of charcoal and the Rn concentration in the air.

The effective diffusion coefficient for each device was determined from the fit to the experimental data using the open-face model where $D_{charcoal}$ is the only free parameter. These values for the EPA and COH devices are shown in Table 1, along with the measured charcoal bed depths. For the FLT devices, diffusion-out experiments were not conducted, and an effective radon diffusion coefficient in charcoal was adopted from Co86.

The diffusion barrier model does, on the other hand, appear to agree quite well with the experimental results for the COH and LBL canisters. In the case of the COH devices, having previously determined the radon-in-charcoal diffusion coefficient, the model then has two free parameters, the area and length of the diffusion barrier. The size of the opening in the diffusion barrier can be measured, although the physical length of the diffusion barrier for the COH devices is somewhat ambiguous. We therefore chose to use the model to determine the effective length of the diffusion barrier, fixing the size of the opening. Thus, with the desiccant and screen attached, the COH canister appears to have an effective $L_{db} = 2.28$ cm when $A_{db} = 2.84$ cm^2. The model then yields an "integration time constant" of about three days as Cohen and Nason (Co86) have previously described.

By actually measuring the LBL canister's diffusion barrier, one arrives at a value of $L_{db} = 2.7$ cm and $A_{db} = 1.54$ cm^2. Using these values as parameters for the diffusion barrier model, the predicted reduction in radon in the charcoal bed is in quite close agreement with the actual experimental values as shown in Fig. 5. This provides additional confidence that the model is a reasonably accurate descriptor of the actual physical situation. All the parameters used in the various models are summarized in Table 1.

In the exposures to time-varying Rn concentrations in two rooms of the Indoor Air Quality Research House, several exposure variations were used, as summarized in the second column of Table 2. The average radon concentration for each exposure condition was determined from the continuous radon monitors, which provided radon concentration data every 30 min. These data were also used as input for either the open-face or diffusion barrier models.

Results of the chamber exposures are illustrated in Fig. 7 for the open-face canister, with the top half of each figure depicting the radon concentration profile to which the detector was exposed in each case. The bottom portion of the figure shows the model simulation of the radon activity in the canister. Similar model runs have been made for the other charcoal devices. The results of the experiments and the modeling are summarized in Table 2.

Two points should be emphasized in reviewing this table. First, the comparisons between the model and the measured charcoal device concentrations are quite good, for both the open face and the diffusion barrier devices. The largest discrepancies are in rows 6 and 7, one test using the EPA device and one using the FLT device. Second, these results also show that the diffusion-limited charcoal detectors, namely COH and LBL, do in fact integrate better than their open face counterparts, EPA and FLT.

The last column in Table 2 shows the comparison between the average of the actual radon concentrations to which the devices were exposed and the radon concentrations derived from the charcoal device measurements. The largest variations are seen in the case of the open face devices (EPA and FLT), where back-diffusion losses are important in going from high to low radon concentration atmospheres. These ratios are similar to those observed by George (Ge84) and Ronca-Battista and Gray (Ro87). The fact that the devices do not over-predict radon concentrations when exposed to the high radon environment last is an artifact of the experimental conditions, since radon was injected into the test chamber as a "spike", and the radon concentrations subsequently declined (primarily through ventilation) during the exposure period.

Another important use of models such as these is to be able to investigate the behavior of these devices parametrically. Two such runs are shown in Fig. 8, where the response characteristics of an open face and a diffusion barrier device can be compared. An important result of the simulation is to show that the predicted behavior of the charcoal devices is linear, and thus the responses to changes in radon concentrations can be superimposed. Note that in Figure 8a, the contribution of the first radon peak diminishes greatly as the second radon peak is recorded with the open face canister. In the case of the diffusion-limited device, the second peak does not have as great an influence on the total radon concentration adsorbed by the canister. Other parametric studies will be pursued to see how device parameters influence the response characteristics.

CONCLUSIONS

These results seem to verify the usefulness of mathematical models in predicting the response of the various types of charcoal detectors to time-variant ^{222}Rn concentrations. Such models may also play a role in the future design, optimization and/or use of charcoal canisters for various sampling strategies. Additional work needs to be done in determining the parameters for ^{222}Rn diffusion, adsorption, and canister configurations.

One primary result of this experimental and modeling work has been to demonstrate the limitations in using charcoal canisters to obtain integrated readings over short periods of time when the actual ^{222}Rn concentrations are changing. Even the diffusion-limited canisters showed errors of more than 20%, with an accompanying increase in uncertainties due to counting statistics. It is clear that when using charcoal adsorption techniques to deetermine indoor radon concentrations that inaccuracies can be great and that charcoal canisters should only be used as a screening devices, and not as an end measurement in itself. Diffusion-limited sampling is a benefit in this regard, but still should not be relied upon as an accurate measurement technique.

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Table 1: Modeling Parameters

Open-face Devices:

Canister	Bed Depth (cm)	D (cm^2/s)
EPA	1.8	1.6×10^{-5}
COH	1.4	1.45×10^{-5} *
FLT	1.2	1.3×10^{-5} **

Diffusion Barrier Devices:

Parameter	COH	LBL
D_{air} (cm^2/s)	0.12	0.12
$D_{charcoal}$ (cm^2/s)	1.45×10^{-5}	1.60×10^{-5}
$K_{abs} \frac{([pCi/g]}{[pCi/cm^3])}$	3300	3300 ***
L_{db} (cm)	2.28	2.70
A_{db} (cm^2)	2.84	1.54
V (cm^3)	38.0	78.5
$L_{charcoal}$ (cm)	1.4	1.8
$M_{charcoal}$ (g)	25.0	70.0

* Measured using open-face COH devices.

** Adopted literature value (Co86).

*** Adopted value from Ro87.

Table 2: Response of Various Charcoal Canister Devices to Time-varying Radon Concentrations and a Comparison of Model and Measurement Results

Device	Exposure Condition*	Avg. [Rn]** (pCi/L) (1)	Charcoal Device		[(2) - (1)]/(1) (%)
			Measured (2)	Model Prediction	
Open-face Devices:					
EPA:	H1	196	155	167	-21
	H1-3	148	94	95	-36
	H1-4	132	76	77	-43
	L1-4	22	9.2	8.2	-58
	L1,2H3,4	64	67	70	+5
	H1,2L3,4	90	20	15	-78
FLT:	H1-4	132	89	68	-33
	H1,2L3,4	90	9.4	7.6	-90
	L1,2H3,4	64	66	67	+3
Diffusion Barrier Devices:					
COH:	H1	196	191	194	-3
	H1-3	148	136	135	-8
	L1,2H3	57	59	62	+4
	H1,2L3	118	91	97	-22
LBL:	H1',2L3,4	79	69	65	-12
	L1',2H3,4	65	75	69	+15

* See Figure 6 for corresponding Rn concentration profiles.

** Average during exposure period determined from continuous radon monitor data.

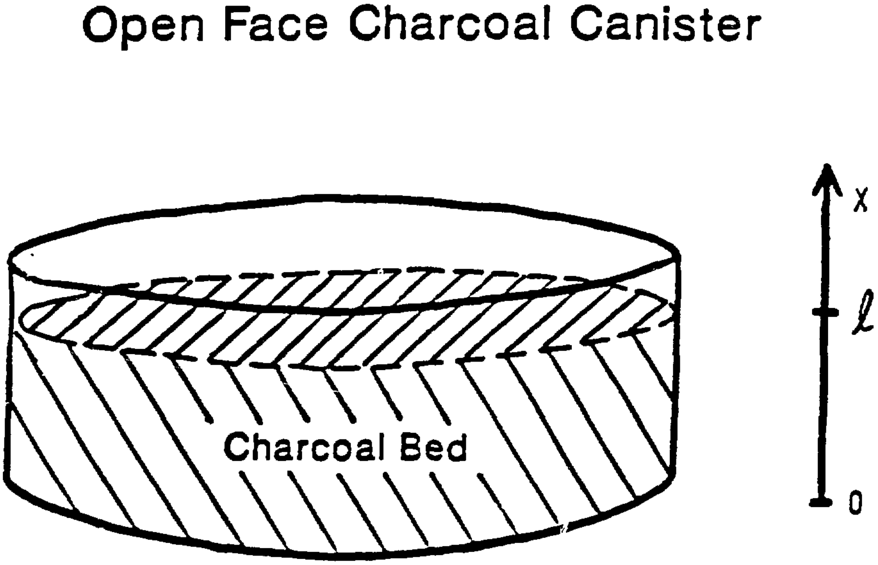


Figure 1.

One-dimensional open-face canister, as modeled.

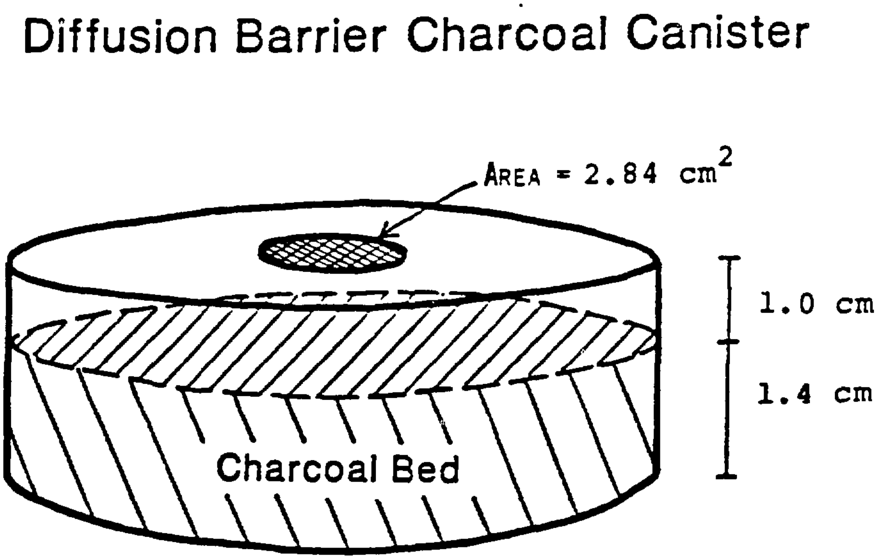


Figure 2A: Sketch of Cohen's diffusion-limited charcoal canister.

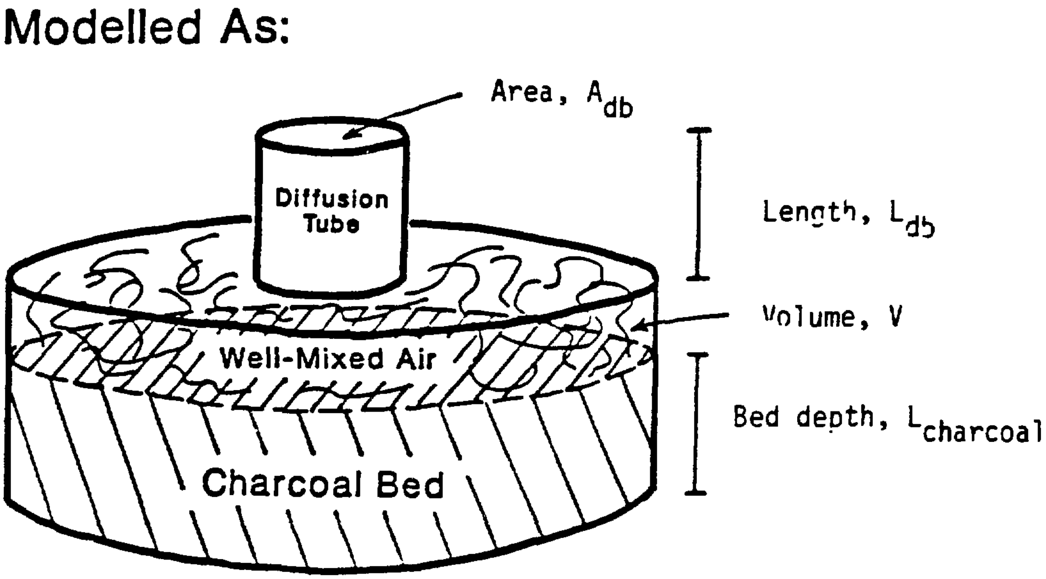


Figure 2B: Sketch of Cohen (COH) and LBL canisters, as modeled.

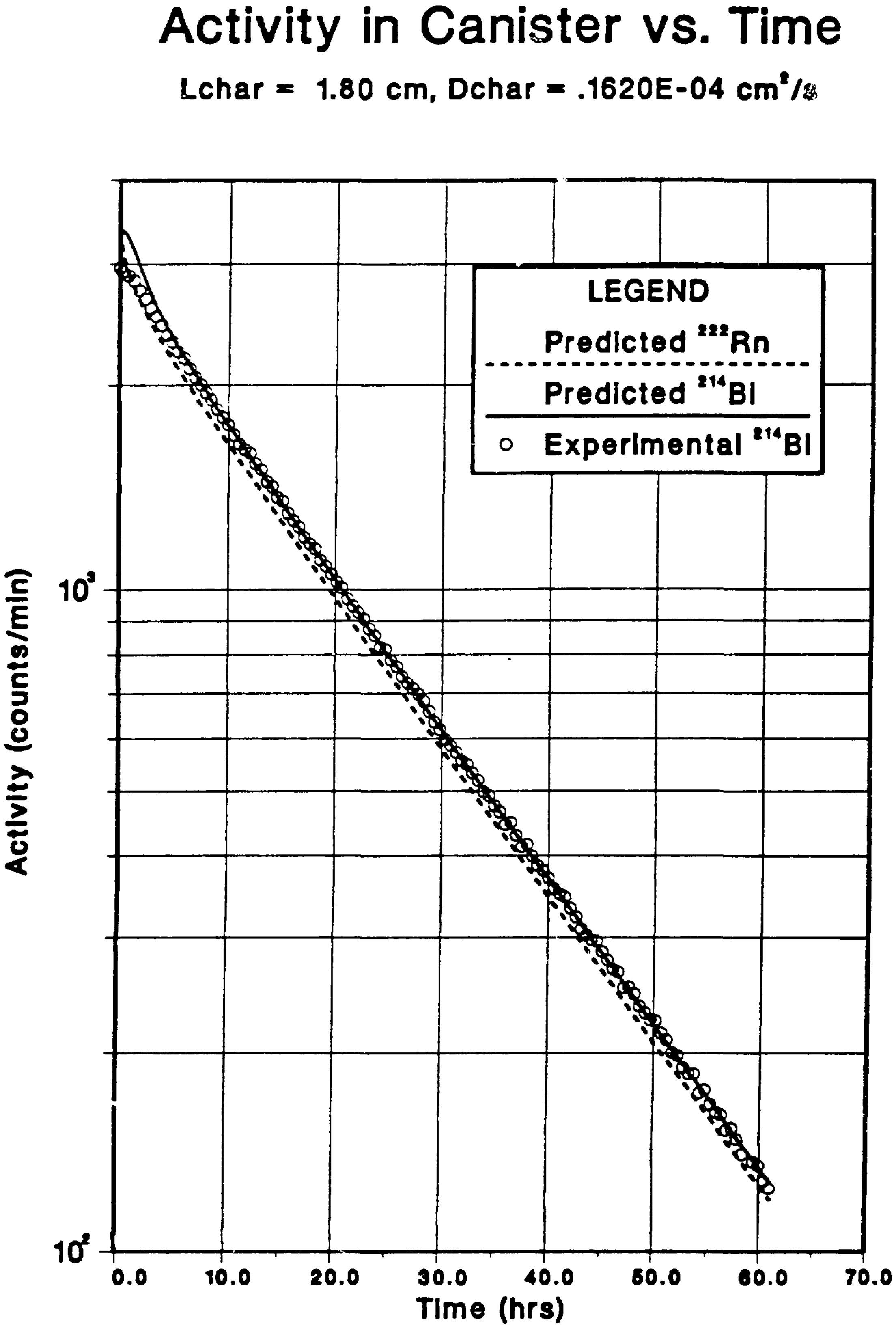


Figure 3: Diffusion-out data for open-face EPA canisters. Observed ^{214}Bi activity is plotted as open circles, while the predicted fit to the data, using the open-face model, is depicted by the solid line.

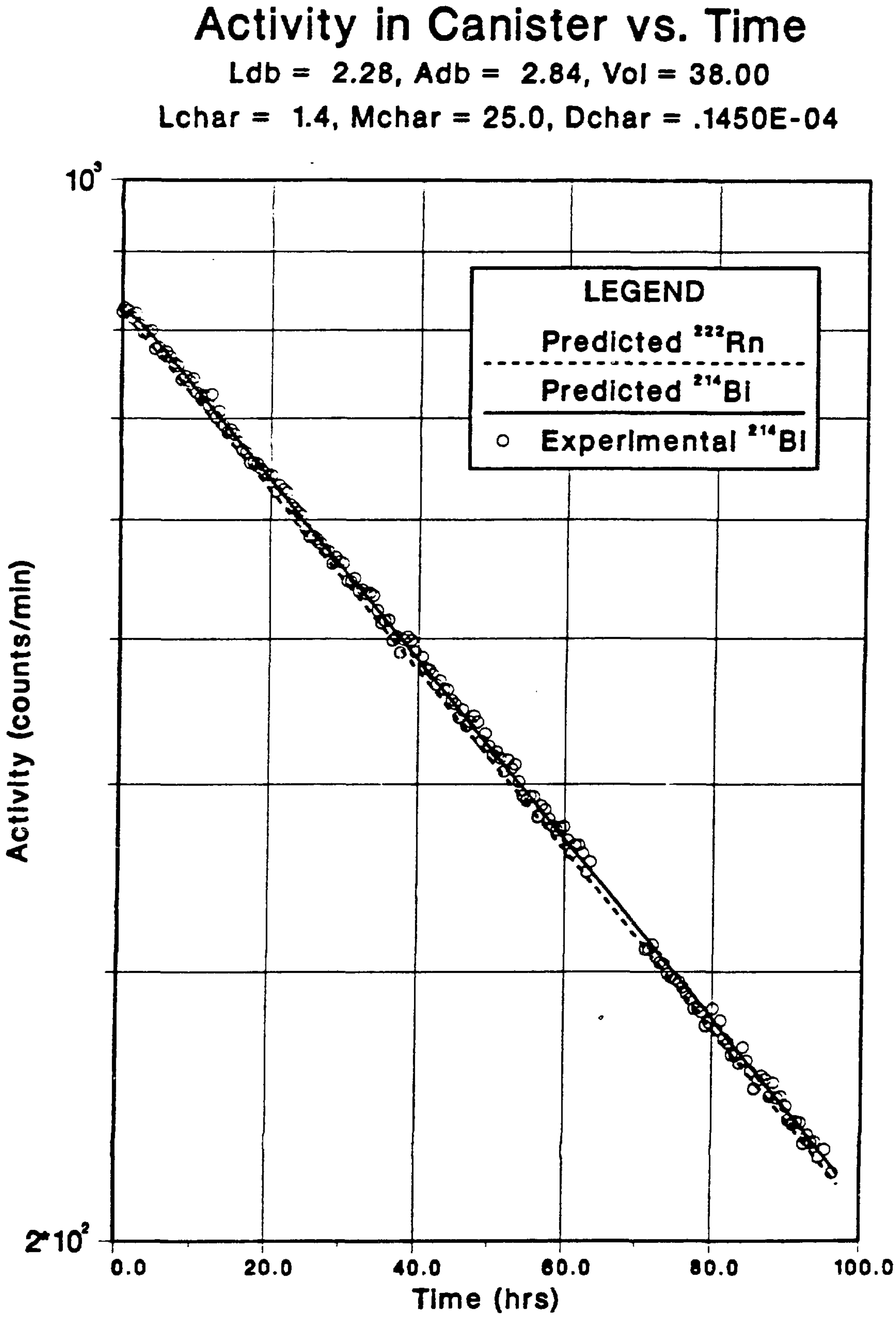


Figure 4: Diffusion-out data for diffusion-limited COH canisters. The predicted fit to the data is based on the diffusion barrier model.

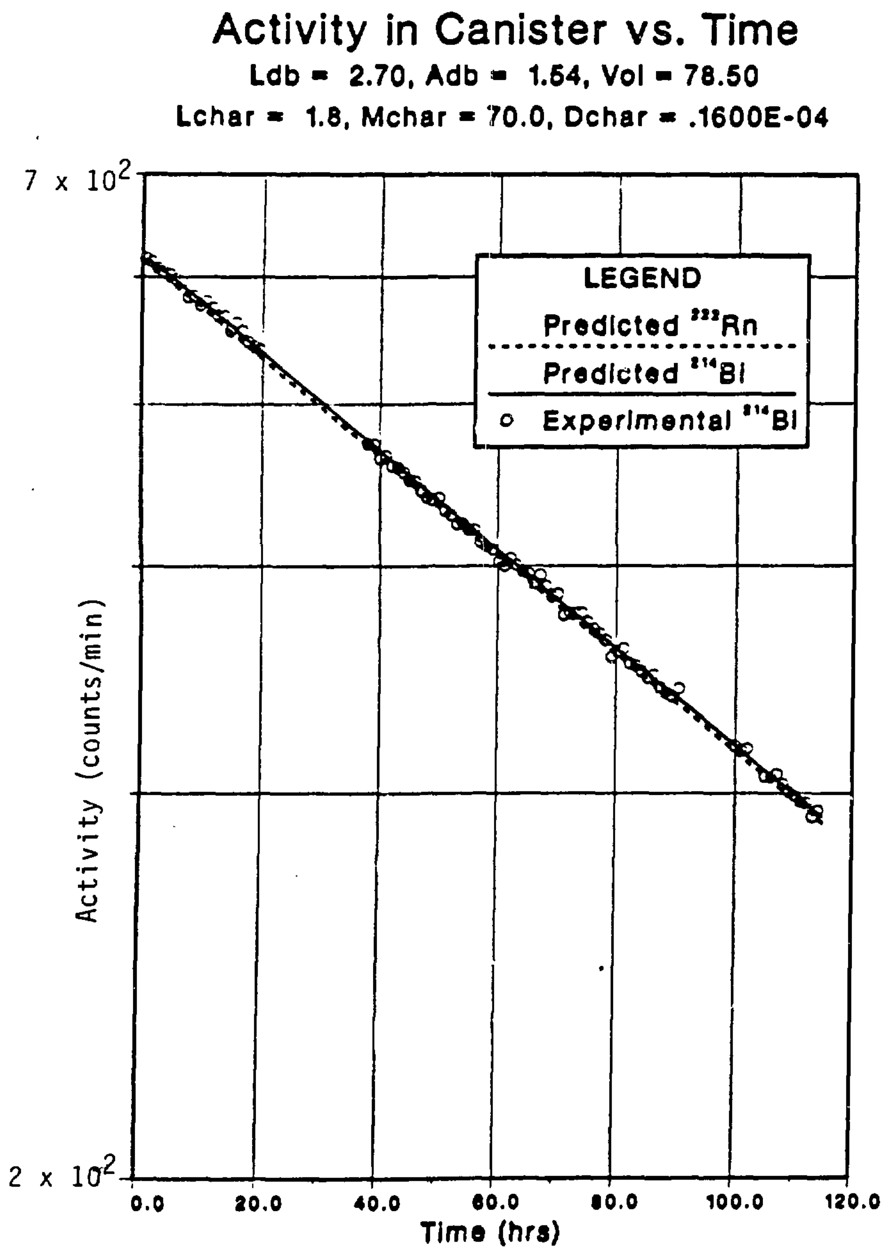
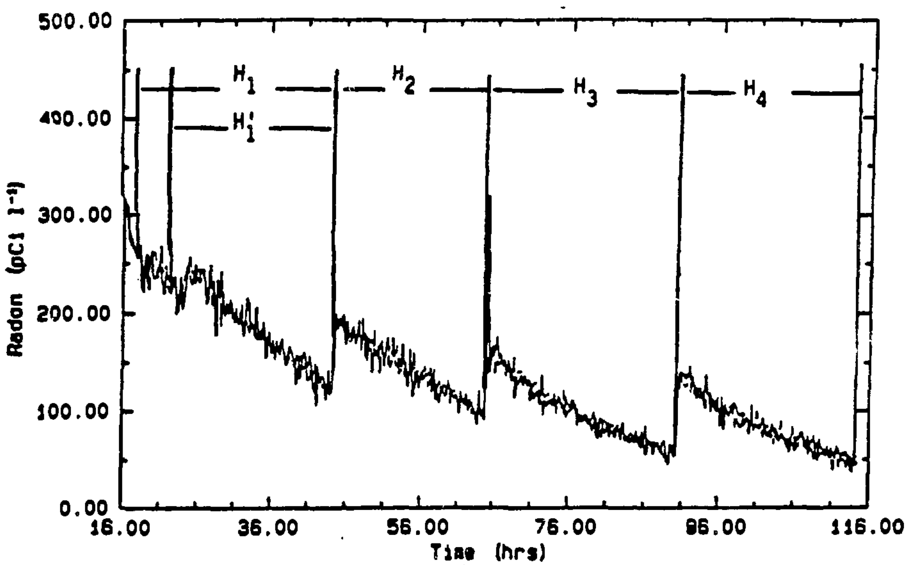
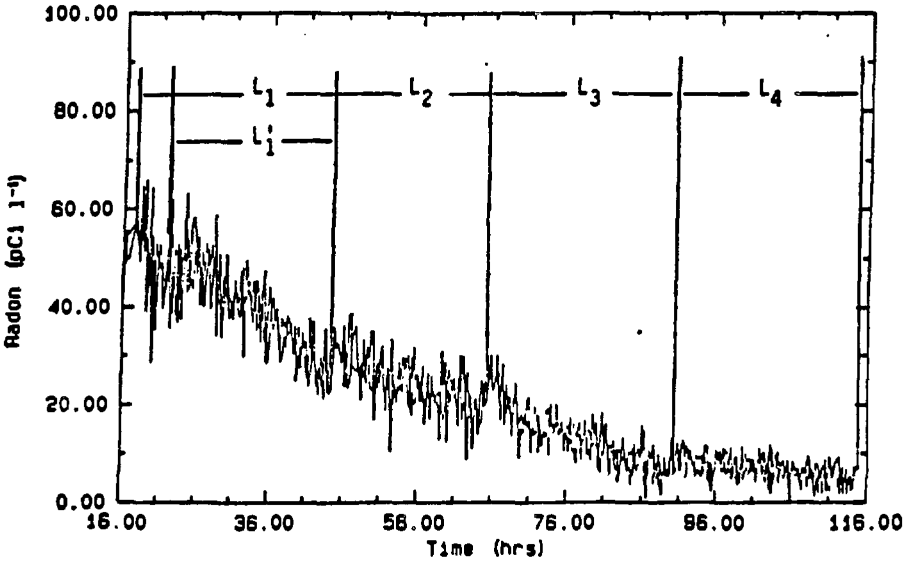


Figure 5. Diffusion-out data for diffusion-limited LBL device. The predicted fit of the data is based on the diffusion barrier model.



(a) High Conc. Room



(b) Low Conc. Room

Figure 6: Variations of radon concentrations in the two rooms used in the chamber exposures.

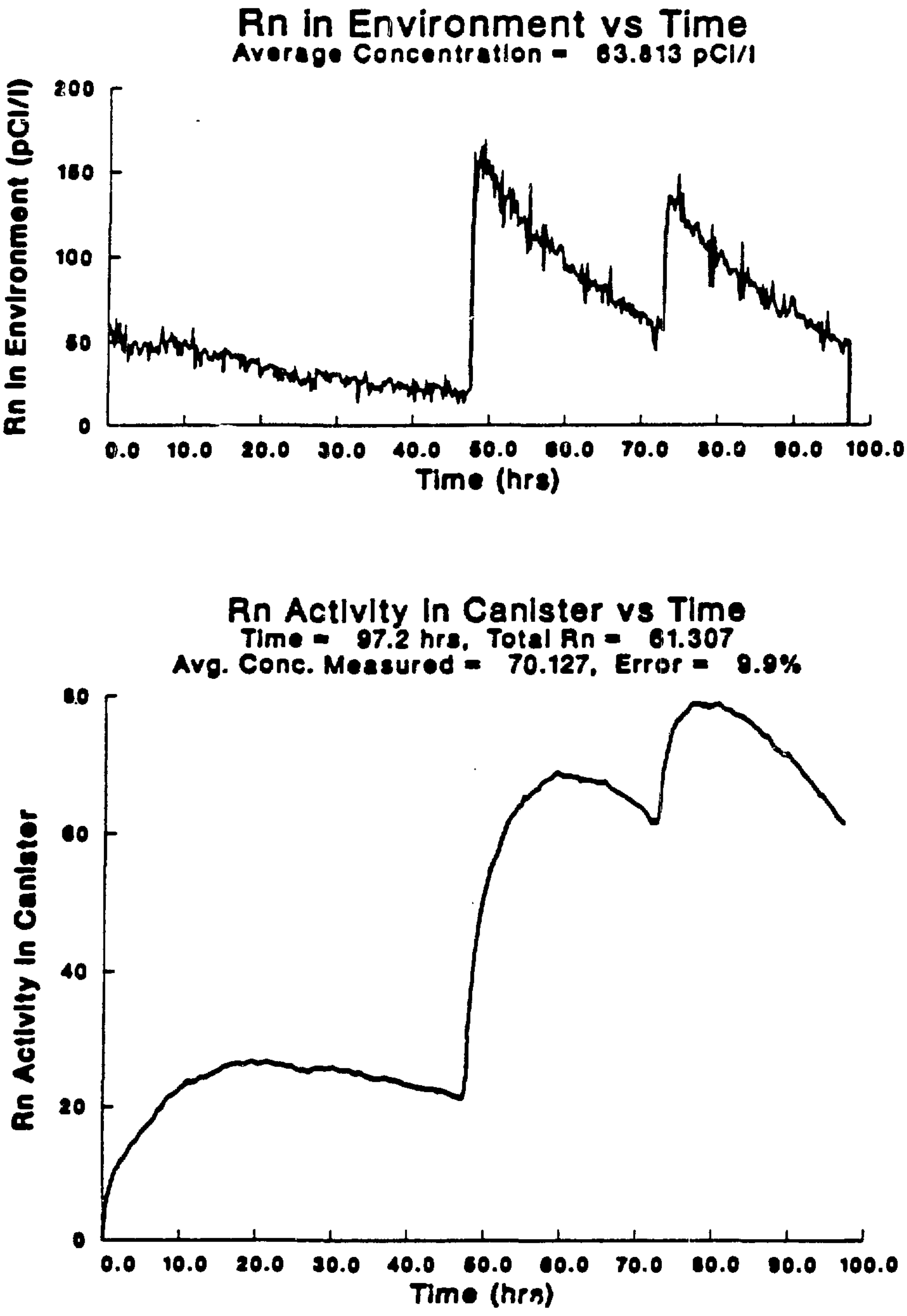


Figure 7A: Radon concentrations and model predictions of radon adsorbed in the open-face EPA canister under exposure conditions of low concentration followed by high radon concentration.

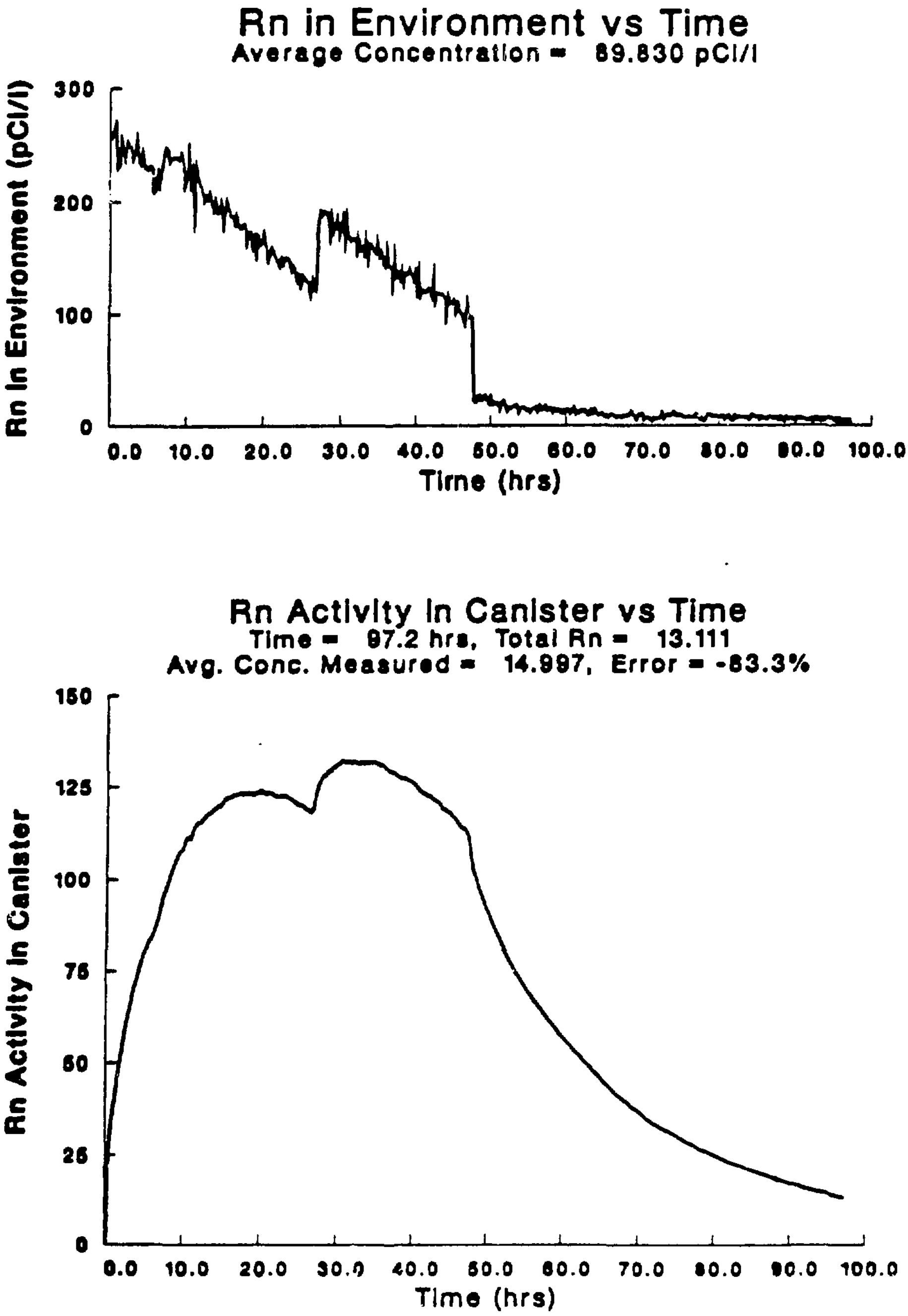


Figure 7B: Same as Figure 7A, except the exposure conditions are reversed.

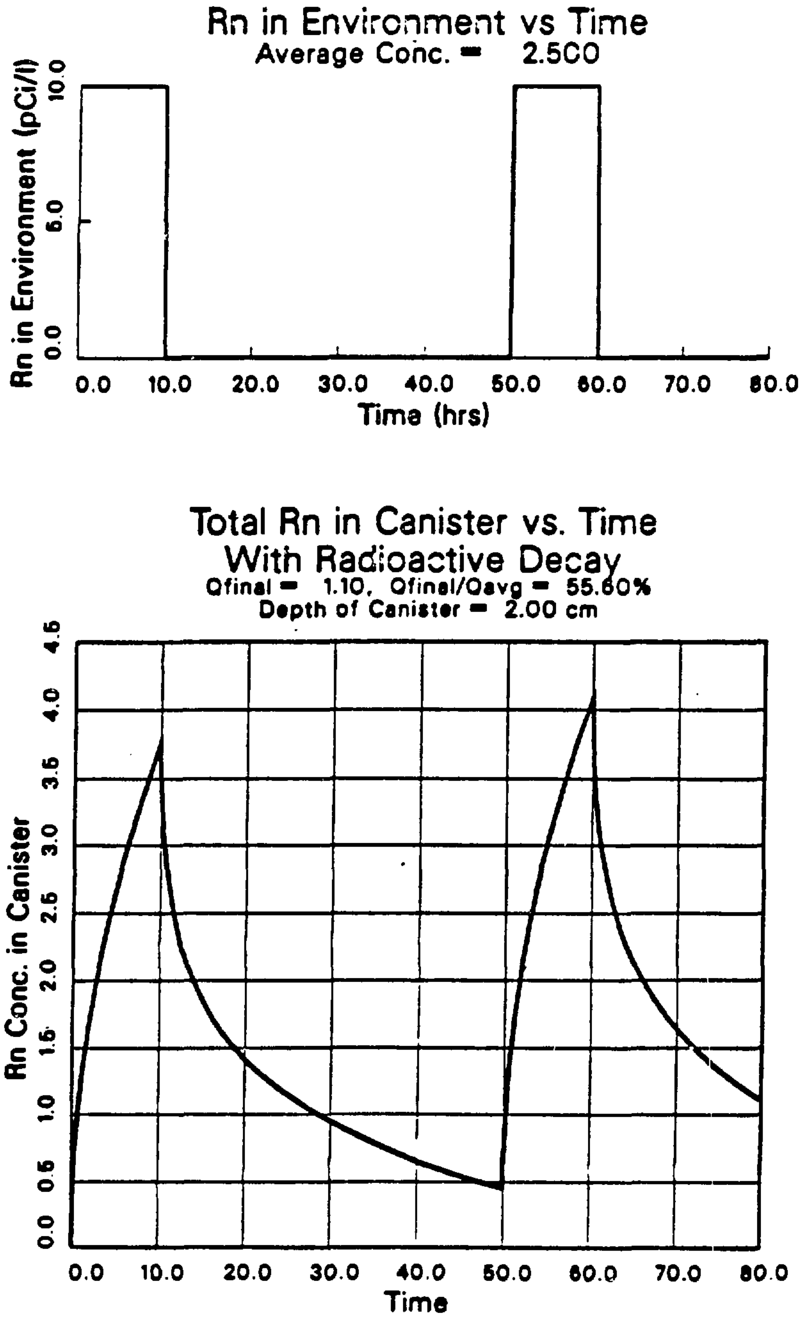


Figure 8A: Simulation of radon concentration profile and resulting radon adsorbtion in an open-face canister.

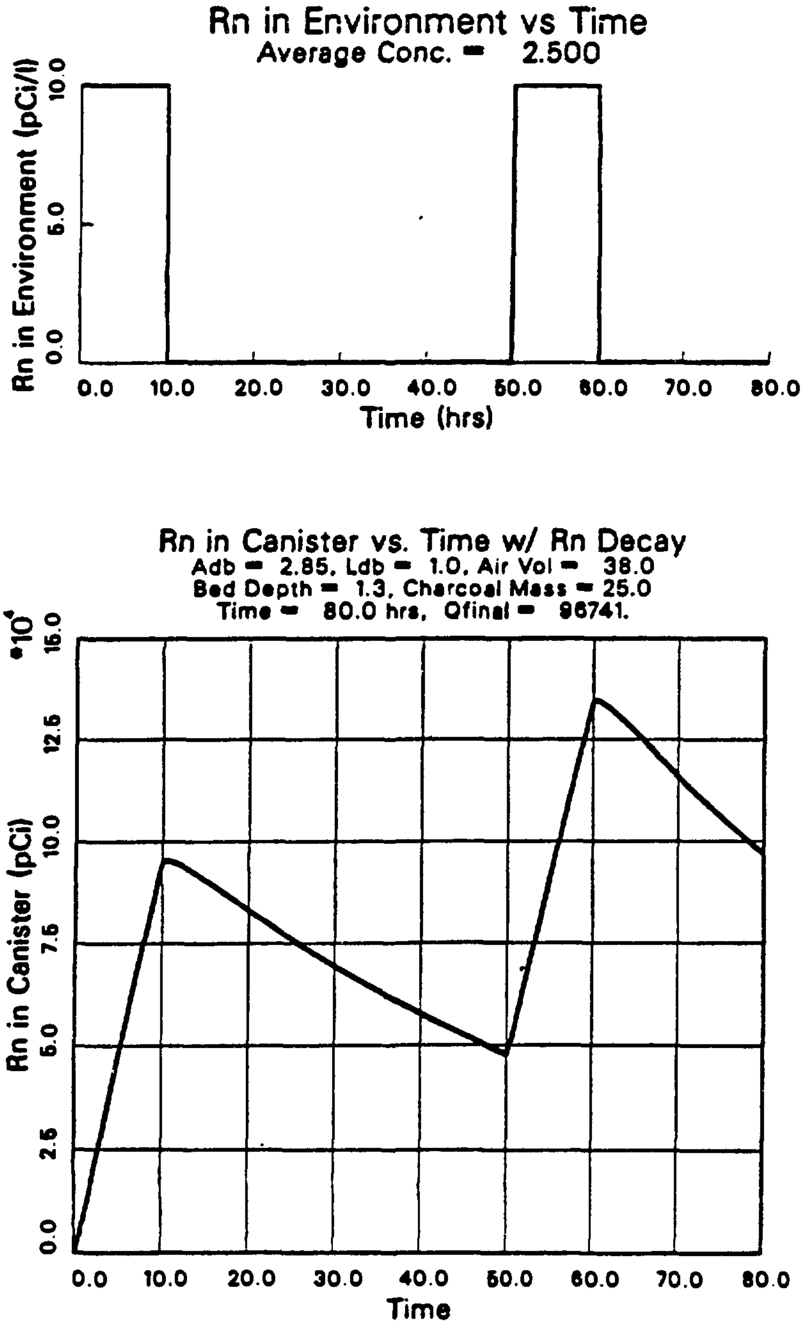


Figure 8B: Same as Figure 8A except using a diffusion-limited canister.

SECTION D

Influence of Temporal Variations of Radon Concentrations on Passive Radon Monitoring

Howard M. Prichard, University of Texas

INFLUENCE OF TEMPORAL VARIATIONS OF RADON CONCENTRATIONS ON PASSIVE RADON MONITORING

Howard M. Prichard

University of Texas

Radon sampling systems can be characterized by the effective half-life of the stored information. Continuous radon monitors and alpha track systems have very long effective half-lives, so that extending the sampling interval does not inherently imply the loss of information stored early in the sampling interval. Electret and TLD based radon monitoring systems may or may not have fading problems, depending on the situation. If the information consists of accumulated radon, then the 92 hour half life of radon imposes a limit on the optimum sampling duration. A constant flow pump filling a leak-free impermeable bag with ambient air has the best performance in this class of systems, having the effective half-life of radon. Passive sampling with activated carbon is generally more convenient, however, and hence has come into wide use.

At room temperatures activated carbon is not an infinite sink for radon, and its ability to retard the motion of radon decreases as temperature increases. It can be shown that, for dry air at a given temperature, radon loss from an activated carbon radon detector by back diffusion is well approximated by an exponential loss model. Consequently, the combined back diffusion and decay loss is also exponential, and the total system can be described as having an effective half-life that can be determined simply by taking repeated gamma measurements on a open detector taken from a high radon environment.

Measurements on a number of commercially available radon detectors have shown effective half-lives ranging from 14 to 61 hours. In general, the longer the effective half-life, the less will be the influence of temperature variations and moisture uptake that affect the performance of the detector. Effective half-life also has a pronounced effect on the response of the detector to various temporal radon profiles. The greater the ratio of sampling time to effective half-life, the greater will be the under estimation of radon arriving early in the sampling interval and over estimation of radon arriving later. The consequences of sampling for intervals much greater than the effective half-life are illustrated by computer simulations on real indoor radon data.

(The use of hourly radon data contributed by Dr. Naomi Harley, New York University Medical Center is gratefully acknowledged.)

SECTION E

The Influence of Changing Exposure Conditions on Measurements of Radon Concentrations with the Charcoal Adsorption Technique

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DRAFT

The Influence of Changing Exposure Conditions on Measurements of Radon Concentrations with the Charcoal Adsorption Technique

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ABSTRACT

The charcoal adsorption technique is now widely used to measure indoor radon concentrations. Deployment periods commonly used range from two to seven days. The passive nature of the activated charcoal allows continual adsorption and desorption of radon, and the adsorbed radon undergoes radioactive decay during the exposure period. Therefore, the technique does not uniformly integrate radon concentrations during the exposure period. The technique can be calibrated to yield very precise results for exposures during relatively constant radon concentrations. During a deployment period of several days in a home, however, radon concentrations can vary by a greater magnitude than under controlled laboratory conditions. Charcoal canisters were exposed in an environmental chamber to controlled radon concentrations, and temperature and humidity conditions were varied during the exposure period. The collection efficiency of the charcoal was investigated for a variety of exposure conditions and exposure periods. The conclusions of these investigations led to a change in exposure time from 96 hours to 48 hours in the United States Environmental Protection Agency charcoal canister measurement program, and an improved understanding of the limitations and capabilities of the charcoal adsorption technique.

I. Introduction

In response to growing concern about exposures to indoor radon concentrations, the Environmental Protection Agency (EPA) Office of Radiation Programs undertook a large-scale effort to provide assistance to States and Federal Agecnies to measure radon concentrations in thousands of homes nation-wide. The activated carbon adsorption technique developed by George (Ge84) was chosen for this program because the measurement method is simple, easy, and inexpensive. The radon adsorption processes, the influences of environmental factors, and the dynamics of the changing activity of the radon in the charcoal bed, however, are more complicated than may first appear. EPA conducted a series of experiments to investigate radon collection on the activated carbon adsorber used by the EPA Eastern Environmental Radiation Facility (EERF) to gain a more complete understanding of the measurement method.

This technique has been reviewed by Cohen (Co83; Co86), by George (Ge84), and by Prichard (Pr85). The EPA modelled its activated carbon canister after the model developed and described by George (Ge84). As these investigators noted, the technique does not produce integrated measurements of radon concentrations during the exposure period. Instead, the amount of radon in the carbon bed is changing in response to three separate processes. In addition, the rate of change of these three processes is influenced by environmental factors such as air temperature, humidity, and the presence of other indoor air pollutants. These three processes are discussed on a conceptual basis here.

The first process is radon adsorption. When the carbon bed is exposed to air, the gases in the air, including radon, diffuse into the carbon bed. The rate of this diffusion is proportional to the difference in concentration of the radon gas in the air and in the bed. As the gases diffuse into the carbon, radon becomes trapped in the carbon. One theory of adsorption of molecules onto carbon is described by Langmuir (Langmuir, in Hassler, 1974). Langmuir describes wandering molecules of vapor in the carbon colliding with an unoccupied surface space and adhering. At first when the surface is bare, the number of molecules adhering exceeds the number desorbing. As the surface becomes covered, other gas molecules find fewer unoccupied spaces, but there is also an increase in the number of molecules escaping from the surface. When the rate of desorption equals the rate of adsorption, equilibrium is reached. Section 4 of this report provides data illustrating the rate of radon adsorption, and how it is affected by the humidity of the air to which the carbon was exposed. Since water vapor competes with radon for adsorption in the carbon bed, increasing humidity decreases the collection efficiency of radon on the carbon.

The second process is radon desorption from the carbon bed. The radon atoms do not become permanently bound in the carbon bed; instead, a fraction of radon atoms are continuously desorbing. This rate of radon desorption is also governed by the difference in radon concentration between the ambient air and the carbon bed. The rate of desorption is greater when the concentration of radon in the bed is high and that in the ambient air is low. This condition may exist when the ambient radon concentration in a home suddenly decreases, perhaps in response to increased ventilation or changing atmospheric conditions. A significant fraction of the radon that had adsorbed onto the carbon would then desorb back into the air. This has been well described by Prichard (Pr85). After the carbon bed is sealed or capped, equilibrium between the small volume of air above the bed and the bed will be reached, and desorption will effectively cease. The results of an experiment to determine the rate of radon desorption under various conditions is described in Section 3.

The third process ongoing in the carbon bed is radioactive decay. The number of radon atoms that remain in the carbon bed long enough to be sealed in it will decrease due to radioactive decay.

The crucial factor of interest to the EPA is how well the results of a measurement with this technique represent the radon concentrations during the measurement period. To test this, as well as our theories about the dynamics of radon in the charcoal bed, we conducted additional experiments in which the radon concentrations were varied. The results of these experiments are discussed in Section 5.

In addition, it has been well documented that temperature affects the rate of adsorption of gases onto carbon (Ad59; St78; Ko78; Si72), with decreasing temperature generally increasing the radon collection efficiency. Data illustrating this temperature dependence is presented in Section 6 of this report.

II. Materials

A. Activated Carbon Canisters

The passive radon charcoal canister used by the EPA consists of the following materials: (1) 8 ounce metal can with lid (4 inch diameter by 1 1/8 inch deep), (2) 70 ± 2 grams of 6 X 16 mesh PCB activated charcoal, (3) metal screen with an openness of at least 30 to 50 percent, (4) removable, internally expanding retaining ring, (5) pad material attached to the inner surface of the lid, and (6) a 13 inch strip of pliant vinyl tape. The materials are assembled as seen in Figure 1.

B. Counting System

The following components make up the counting system used by the EPA's Eastern Environmental Radiation Facility: (1) 3 inch by 3 inch sodium-iodide detector and photomultiplier tube inside counting shield, (2) high voltage power supply, (3) pre-amplifier and amplifier, (4) single channel analyzer and scaler, (5) timer, (6) time of day clock, and (7) printer.

Four sodium iodide detectors are served by a single high voltage power supply, timer, printer, and time of day clock. Each detector is used in conjunction with its own pre-amplifier, amplifier-single channel analyzer, and scaler. Four detectors are arranged together inside a shield which has steel walls approximately 8 inches thick. Within the shield the detectors are separated from each other by 4 inches of lead. A wooden jig is used on each detector to assure consistent counting geometry for the canisters on the detector.

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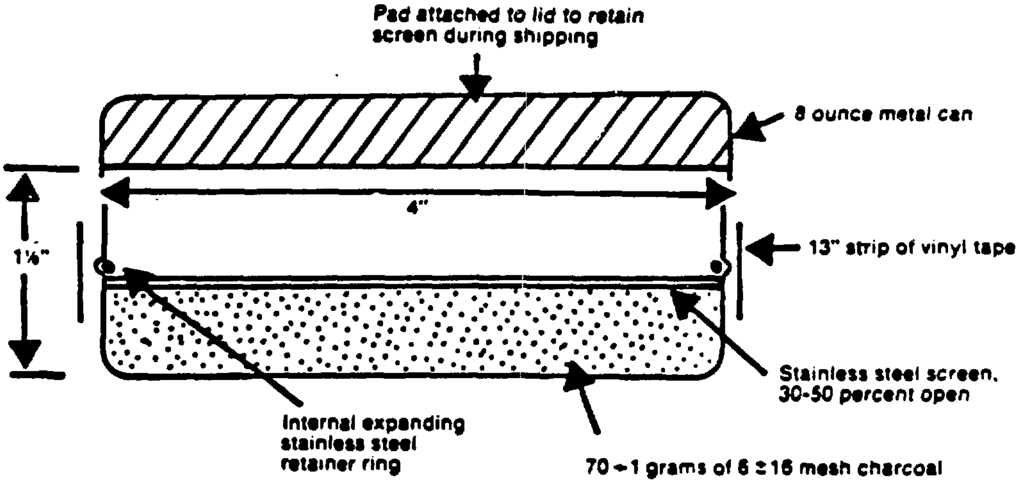

FIGURE 1
Charcoal canister assembly

The counting system is operated in the single channel mode with a lower discriminator setting of 270 KeV and an upper discriminator setting of 720 KeV. All counts in this region are summed during the counting period.

C. Radon Calibration Chamber

The EERF uses 2 radon calibration chambers to expose charcoal canisters and other instruments to known concentrations of radon and radon decay products in controlled environmental conditions. Radium-226 sources provide radon which continuously flows through the chamber. In the chamber, controlled conditions include relative humidity (10 to 90 percent), temperature (0° to 40°C), condensation nuclei (10^3 to 10^6/ml), radon concentration (10 to 1,000 pCi/L), and radon decay product concentration or working level (0.01 to 5 WL).

The temperature, humidity, radon concentration, radon decay product concentration, and condensation nuclei concentration are monitored and recorded each hour. Radon concentrations are monitored using a 0.5 liter flow through scintillation cell, and decay product concentrations are monitored by counting the alpha activity from decay products collected on a filter with a surface barrier detector. Grab sampling, for determination of radon and decay product concentrations in the chambers, is used to calibrate the hourly readouts.

III. Rate of Radon Desorption

If the concentration of radon in the air above the carbon bed is less than the concentration in the carbon bed, some radon will diffuse out of the carbon into the air. The rate of this back diffusion, as termed by Prichard (Pr85), is dependent on the difference in concentrations in the bed and in the air above the bed.

To determine the rate of this back diffusion, four charcoal canisters were exposed in a 100 pCi/L radon environment for 72 hours. The relative humidity during this exposure varied from 20-25 percent, and the temperature varied from 19 to 24°C (66-75°F). After exposure, the canisters were exposed in a relatively low radon concentration of 10 pCi/L, with approximately the same relative humidities and temperature. The charcoal canisters remained in that environment for 3 days, during which time they were periodically analyzed. Each successive count was made with the same analytic equipment, so that differences in counting efficiencies between detectors did not have to be accounted for. The daily change in the efficiency of any analysis system varied less than 3 percent and was considered negligible.

The entire sequence was repeated with four new canisters a second time, after altering the relative humidity in the chamber to 50 percent.

The results of the periodic analyses of the unsealed charcoal canisters conducted during the three days after exposure are summarized in Tables 1 and 2. The parameter pCi/g per pCi/L was calculated by determining the activity in pCi/g of the carbon bed at the time of each analysis, then dividing by the pCi/L of radon in the air to which the canister was exposed. This parameter is useful for understanding the decrease in the amount of radon in the carbon. A decay correction to the first count after exposure was made to eliminate the decrease in counts due to radioactive decay. The averages of the four replicate canisters are shown in Tables 1 and 2 and Figure 2.

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Table 1: Decrease in Radon in Carbon with Time in a 10 pCi/L Environment after a 72-Hour Exposure to 100 pCi/L at 20 Percent Relative Humidity

Time Delay From First Count (Hours)	pCi/g / pCi/L
0	3.31
4	3.02
6.2	2.84
8.2	2.55
10.2	2.33
20.9	1.48
27.2	1.14
33.2	0.93
45.0	0.68
50.9	0.57
56.9	0.51
69.6	0.42

Table 2: Decrease in Radon in Carbon with Time in a 10 pCi/L Environment after a 72 Hour Exposure to 100 pCi/L at 50 Percent Relative Humidity

Time Delay From First Count (Hours)	pCi/g / pCi/L
0	2.75
1.83	2.59
3.91	2.29
5.91	2.07
8.16	1.83
20.41	0.97
26.16	0.73

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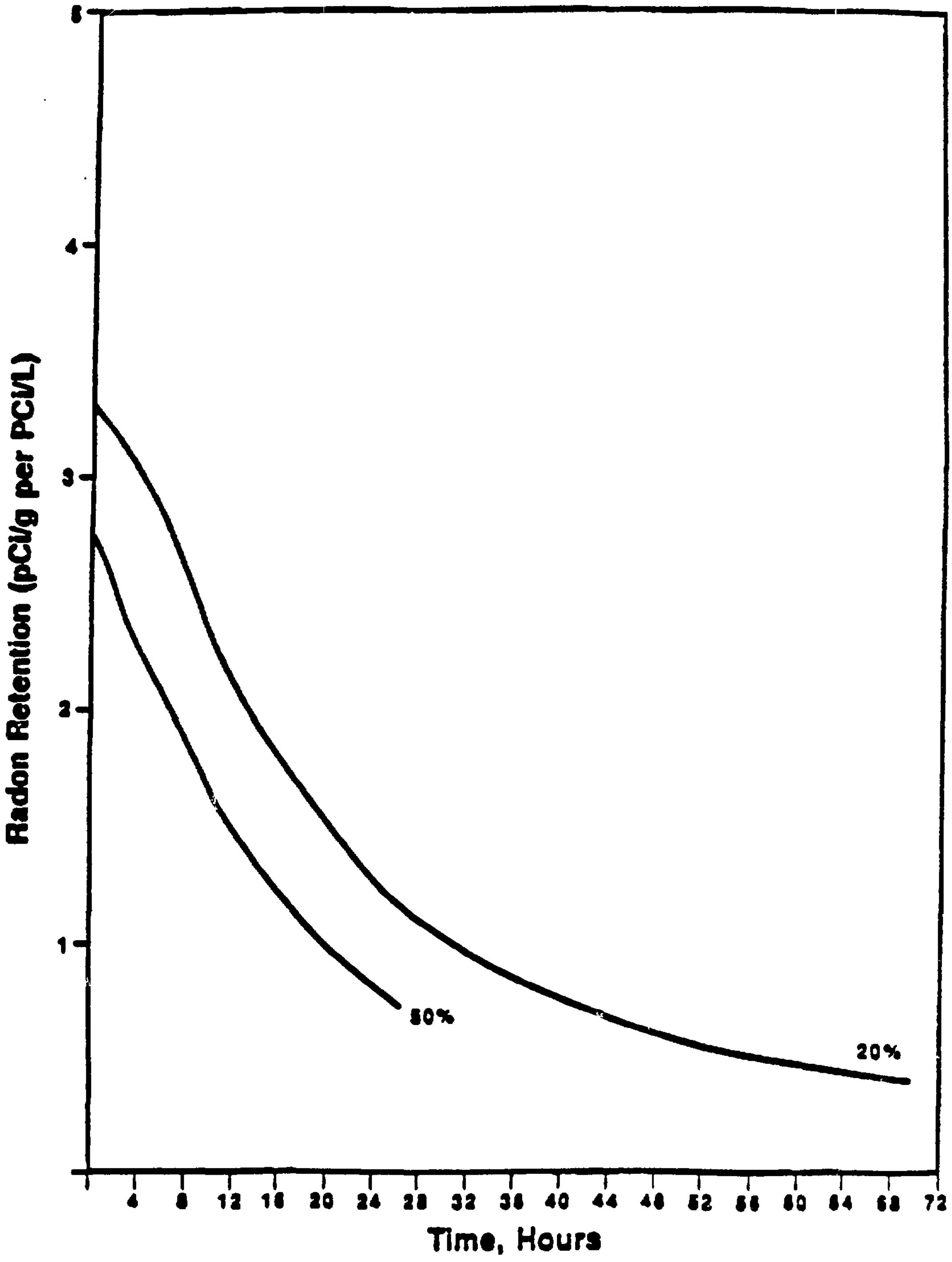
Figure 2: Radon Desorption from Activated Charcoal as a Function of Time and Relative Humidity



IV. Rate of Radon Adsorption

The rate of adsorption of radon onto the carbon bed is dependent upon both humidity and temperature. In this section, data illustrating the dependence of adsorption on humidity is discussed.

Activated carbon adsorbs or desorbs water if the humidity of the air to which the canister is exposed during the measurement period is different than the humidity of the air in the environment where the carbon was packed into the canister. For these canisters, the relative humidity of the air where the canisters were packed was about 38 percent. Therefore, at a relative humidity less than 38 percent there will usually be a loss in moisture in the carbon, and at any relative humidity greater than 38 percent there will be a gain in moisture in the carbon. Because of this moisture gain, there is a decrease in collection efficiency with increasing relative humidity. In addition, saturation of the activation sites in the carbon bed is reached more quickly at higher humidities.

To determine the rate of adsorption's dependence on humidity for these canisters, the following experiments were conducted. The radon chamber at the EERF was stabilized to the following conditions: (1) relative humidity of 20 percent, (2) temperature of 21°C (70°F), and (3) radon concentration of 100 pCi/L. A set of 30 canisters were put into the chamber and exposed in the following manner: a group of 5 each for 1, 2, 3, 4, 5, and 6 days respectively. The canisters are opened inside the chamber and the exact start times were recorded.

At the end of each 1 day period specified above, a group of 5 canisters were removed for counting. The lids were placed on the canisters while still inside the chamber and the canisters were removed, taped and the exact time was recorded.

The canisters were each counted for ten minutes on the sodium-iodide detectors described previously. This data was converted to pCi/g per pCi/L values as described in Section 3.

This exposure sequence was repeated twice more, at a humidity of both 50 and 80 percent. Temperature and radon concentration were kept as constant as possible during all exposure sequences. Additional exposures were performed at 38, 60, 70, and 100 percent relative humidities for a period of 1 to 4 days. The results of these experiments, tabulated in Table 3 and plotted in Figure 3, form the basis of the calibration factors used for the EPA canisters.

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Table 3: Radon Retention on Activated Carbon as a Function of Exposure Time

Exposure Period (Days)	Percent Humidity	pCi/g / pCi/L
1	20	2.58
2	20	3.60
3	20	4.32
4	20	4.48
5	20	4.67
6	20	4.69
1	38	2.42
2	38	3.43
3	38	3.74
4	38	3.88
1	50	2.53
2	50	3.15
3	50	3.28
4	50	3.24
5	50	3.20
6	50	3.12
1	60	2.36
2	60	3.02
3	60	3.19
4	60	3.11
1	70	2.41
2	70	2.75
3	70	2.59
4	70	2.27
1	80	2.24
2	80	2.54
3	80	2.14
4	80	1.83
5	80	1.46
6	80	1.18
1	100	2.06
2	100	2.32
3	100	1.85
4	100	1.30

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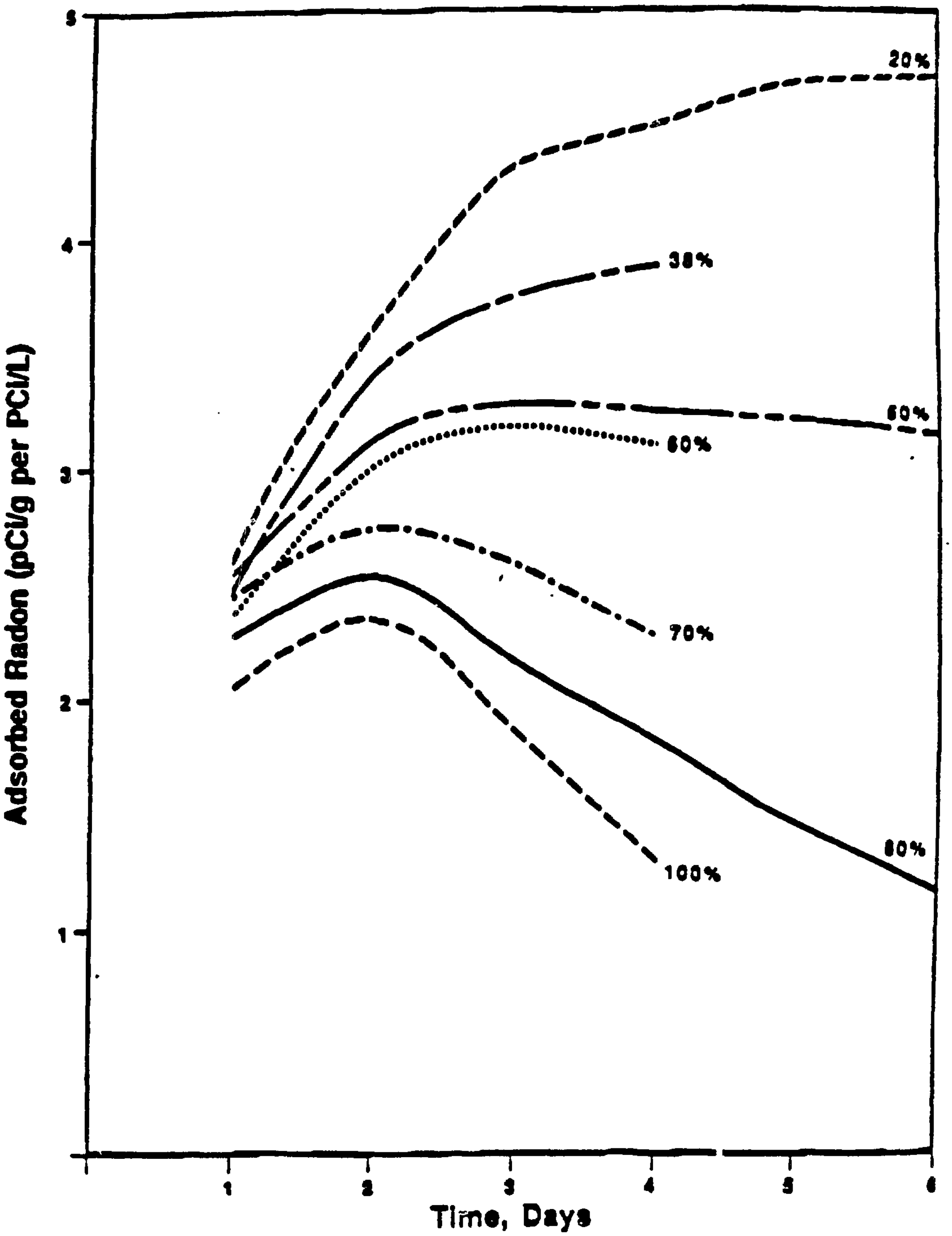
Figure 3: Radon Adsorption on Activated Charcoal as a Function of Exposure Time and Relative Humidity



V. Exposures to Two Different Concentrations

Radon concentrations in homes can vary significantly during a two to four day period (St79; Wi86). Large variations can be caused by extreme changes in ventilation rates or atmospheric conditions. To determine how EPA charcoal canisters respond to two different concentrations during the exposure period, the following experiments were conducted.

Duplicate sets of charcoal canisters were exposed to two radon concentrations with a relative humidity of 50 percent and a temperature between 19 and 24°C (66-75°F). Total exposure times were 48 and 96 hours. The ratios of the two concentrations ranged from 2:1 to 10:1, as well as the reverse. The radon concentration in the chamber ranged from 100 to 10 pCi/L.

The results of these exposures are tabulated in Table 4. In addition to the times and concentrations, the table shows the average radon concentration during the exposure period. The final column shows the difference, in percent, between the canister result and the average radon concentration to which it was exposed.

Table 4: Difference Between Canister Result and Average Radon Concentration During Exposure to Two Concentrations

Ratio of Concentrations $[C_1:C_2]$	t_1 (hrs)	C_1 (pCi/L)	t_2 (hrs)	C_2 (pCi/L)	Average C (pCi/L)	Canister Result (pCi/L)	Difference* (Percent)
2:1	48	19	48	8	13	4	- 72
1:2	48	8	48	19	13	15	+ 22
3:1	24	18	24	6	11	6	- 51
1:3	24	6	24	17	11	13	+ 21
6:1	24	54	24	9	31	15	- 50
1:6	24	8	24	49	28	37	+ 31
10:1	24	100	24	9	52	24	- 54
1:10	24	9	24	97	53	70	+ 32
10:1	48	98	48	9	53	13	- 75
1:10	48	9	48	100	55	90	+ 64

*Between canister result and average concentration during exposure.

Table 5: Effect of Temperature on Canister Calibration

Dry Bulb Temperature (°F)	Percent Humidity	Wet Bulb Temperature (°)	Chamber Concentration (pCi/L)	Canister Result (pCi/L)	Difference (Percent)
50	50	43	108	115	+ 7
50	80	47	104	125	+ 20
60	50	50	114	109	- 4
60	80	57	112	113	+ 1
80	50	67	112	89	- 21
80	80	75	107	82	- 23

*Between chamber concentration and canister result.

VI. Effect of Temperature

Since temperature as well as relative humidity affects the collection efficiency, tests were conducted at the EERF to determine the extent of the temperature dependence on collection efficiency.

The calibration factors used by the EPA for the carbon canister were generated at 21°C (70°F). To test the broader range of temperatures possible in the average home, the following sets of experiments were conducted. A set of three canisters were exposed in the radon chamber for a period of 48 hours at approximately 100 pCi/L, 10°C (50°F), and 50 percent relative humidity. The radon adsorbed was determined by analyzing the canisters for 10 minutes on sodium-iodide detectors. A new set of three canisters were subsequently exposed at 10°C (50°F) to a relative humidity of 80 percent. The entire series was repeated at 16°C (60°F) and 27°C (80°F) for both 50 percent and 80 percent relative humidities.

The results of these experiments are tabulated in Table 5. The last column in the table lists the difference, in percent, between the calculated radon concentration and the known chamber concentration during each exposure sequence.

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VII. Conclusions

A. Rate of Radon Desorption

As illustrated in Figure 2, the rate of radon desorption from a carbon bed is approximately the same for 20 and 50 percent relative humidities. For an exposure in a 20 to 50 percent relative humidity environment, one half of the radon initially in the carbon bed was desorbed in approximately 20 to 25 hours.

It should be noted that the desorption half-life will change, depending on the ratio of the initially high radon concentration to which the canister was exposed, and the low radon concentration in which the canisters were left, unsealed, where the radon desorbed. The ratio of the two radon concentrations used here was approximately 10:1. If the radon concentration decreases by a greater amount, the rate of radon desorption will increase and the desorption halflife will be shorter. If the radon concentration does not decrease so substantially, so that the ratio is 2:1, the rate of radon desorption will not be as fast, and the desorption halflife will be longer.

B. Rate of Radon Adsorption

The rate at which the carbon bed adsorbs radon is dependent on the relative humidity, or dewpoint, of the air to which the carbon was exposed. Figure 3 illustrates that for exposure times of three days or longer, there is a decrease in radon adsorption efficiency when the relative humidity is 50

(continued on next page)

percent or greater. For relative humidities greater than 70 percent, there is actually a decrease in the amount of radon on the carbon after an exposure period of 2 days. Both water and radon atoms are bound to the carbon by weak van der Waal's forces (Ch78), and since water is polar, this attraction is strong. When a high proportion of water molecules become trapped in the carbon, they compete with the radon atoms for sites on the carbon, resulting in the decrease in radon activity in the charcoal as shown in Figure 3 for high humidities and exposure times longer than 2 days.

C. Exposure to Two Different Concentrations

The results in Table 4 illustrate two phenomena. First, the difference between the canister result and the average radon concentration during the exposure period was smaller for total exposure times of 48 hours. In other words, a 48 hour exposure yields a result that is more representative of the average radon concentration during the exposure period than does a 96 hour exposure. A 48 hour exposure to radon concentrations varying by a ratio of 10:1 underestimates the known result by 54 percent. This is in contrast to a 96 hour exposure to a similar concentration ratio which underestimates the results by 75 percent.

In addition, it is apparent that the result of the canister analysis will be greater than the average radon concentration during exposure when the carbon is exposed to the higher concentration in the latter part of the exposure period. Conversely, the canister result will be less than the average radon concentration during exposure when the initial concentration was higher than the second concentration to which the canister was exposed. This is due to both the desorption and radioactive decay of the radon that was adsorbed during the initial exposure period.

D. Effect of Temperature

The results in Table 5 illustrates the effects of temperature on the carbon's collection efficiency. As the temperature was increased, the collection efficiency decreased. An increase in temperature from 10°C (50°F) to 27°C (80°F) at a relative humidity of 50 percent resulted in a drop in collection efficiency of 26 percent. The decrease in collection efficiency with increasing temperature was greater in the higher humidity environment. At a relative humidity of 80 percent, the collection efficiency dropped by 40 percent when the temperature was increased from 10°C (50°F) to 27°C (80°F). These results illustrate the importance of conducting radon measurements during cooler months of the year when the collection efficiency of the carbon is greater, than during warmer temperatures.

Public health considerations make it important to ensure that measurements with EPA canisters estimate as accurately as possible the radon concentrations during the measurement period. It is equally important to ensure that radon concentrations are not underestimated. Based upon the results shown here, the measurement period for EPA canisters was reduced to 48 hours. This limits the effects of high humidities and changing exposure conditions.

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SECTION F

Recent Studies with Activated Carbon for Measuring Radon

A. C. George, U.S. Department of Energy

RECENT STUDIES WITH ACTIVATED CARBON FOR MEASURING RADON

A. C. George

Environmental Measurements Laboratory
United States Department of Energy

Activated carbon passive monitors for indoor radon measurements have been used by the Environmental Measurements Laboratory (EML) since 1979. The M-11 type canister described by George in 1984 was used for radon monitoring in several places in the New York City area, Damascus, Maryland, Allentown and Harrisburg, Pennsylvania, and Glen Ridge and Montclair, New Jersey, long before the discovery of high radon levels in the Reading Prong region.

Since 1985 there has been a proliferation of companies and individuals offering radon analyses using adaptations of these techniques. Alpha track monitoring, the major other technique for passive radon monitoring, is primarily under the control of a single company, Landauer, Incorporated (formerly Terradex). The activated carbon method is more popular with homeowners because of simplicity, cost, and the relatively short integration time of 1 - 7 days. Results are usually available to the homeowner within one to two weeks after a request has been made for a radon measurement.

The activated carbon method is designed for rapid screening for indoor radon only. Temperature and humidity variations are usually less indoors than outdoors, particularly when the U. S. Environmental Protection Agency protocols are followed that require closed house conditions for radon screening tests. Diurnal variability of the radon concentrations under these conditions usually is less than the typical factor of 2 - 3 found durin normal house use. Extreme variation in the indoor radon levels do occur during severe weather changes that affect the pressure differential inside and outside the building, and may occur during different seasons under normal house usage or during house remediation.

Experience at the EML shows that activated carbon devices of the open face type can be used successfully for radon screening purposes if the exposure period is less than 3 days. Addition of a diffusion barrier to slow diffusion into and out of the monitor, and to inhibit the diffusion of water vapor into the device, render the device less sensitive to radon for short integration periods. However the diffusion barrier improves the accuracy of an activated carbon monitor for longer integration times of up to one week. Diffusion barrier monitors are also less sensitive to moisture changes and to extreme changes in the radon concentration levels.

To select the best monitoring device for a particular type of radon measurement, the characteristics of the carbon used must be investigated over the intended integration time and the range of humidity expected. Selecting the dimensions of the open area of open face monitors make these devices less sensitive to humidity and to extreme variations in the radon concentrations. The geometry of the canister, the fraction of the screen open to radon diffusion, and the type of activated carbon employed, are very important parameters in the design of a monitor. These determine the rate of adsorption of radon as opposed to the deleterious effects of water vapor. Reduction of water adsorption can be accomplished by the use of a diffusion barrier or by reducing the air convection currents in the vicinity of open face monitors.

Figure 1 indicates the radon adsorption characteristics of different types and configurations of activated carbon monitors at the relative humidities typically found indoors when exposed to constant radon concentrations. All four types can measure the integrated radon exposure with reasonable accuracy if the total exposure time is less than four days.

Figure 2 shows the results provided by similar monitoring devices tested at higher relative humidities. Several of these monitors appear to collect radon efficiently for three to four days. However two types used by the EPA, which contain carbon that adsorbs both radon and water vapor very efficiently, begin to deteriorate after two days of exposure in a high relative humidity environment.

Figure 3 shows the magnitude of the errors determined for carbon monitors exposed to non-constant radon concentrations, when the radon concentration in the chamber varied by as much as a factor of 14 during the three day exposure at a relative humidity of 67%. The errors ranged from 2 to 17%, an acceptable margin of error for this type of measurement.

Regardless of whether one uses open face carbon monitors or diffusion barrier carbon monitors, all devices should be subjected to tests similar to those that generated the data shown in Figure 3. First, tests similar to those shown in Figures 1 and 2 should be performed to determine the optimum carbon type and canister configuration. If this is done single carbon monitors are usually capable of accurate measurement of environmental radon levels. Occasional exposures of duplicate monitors are recommended for quality assurance.

Questions concerning how well a single measurement of indoor radon, obtained with a carbon monitor that has an integration time of less than one week, accurately represents the true indoor radon concentration can be addressed by repeated measurements over a long period of time or by using more sophisticated instrumentation. The homeowner can usually save money by repeated measurements with a carbon monitor. It appears that less than 5% of the homes will fall into the category that would require several repeated measurements.

FIGURE 1. RADON ADSORPTION BY DIFFERENT TYPES OF CARBON AND CANISTER CONFIGURATIONS. WATER ADSOBRED IN GMS ON 4th DAY.

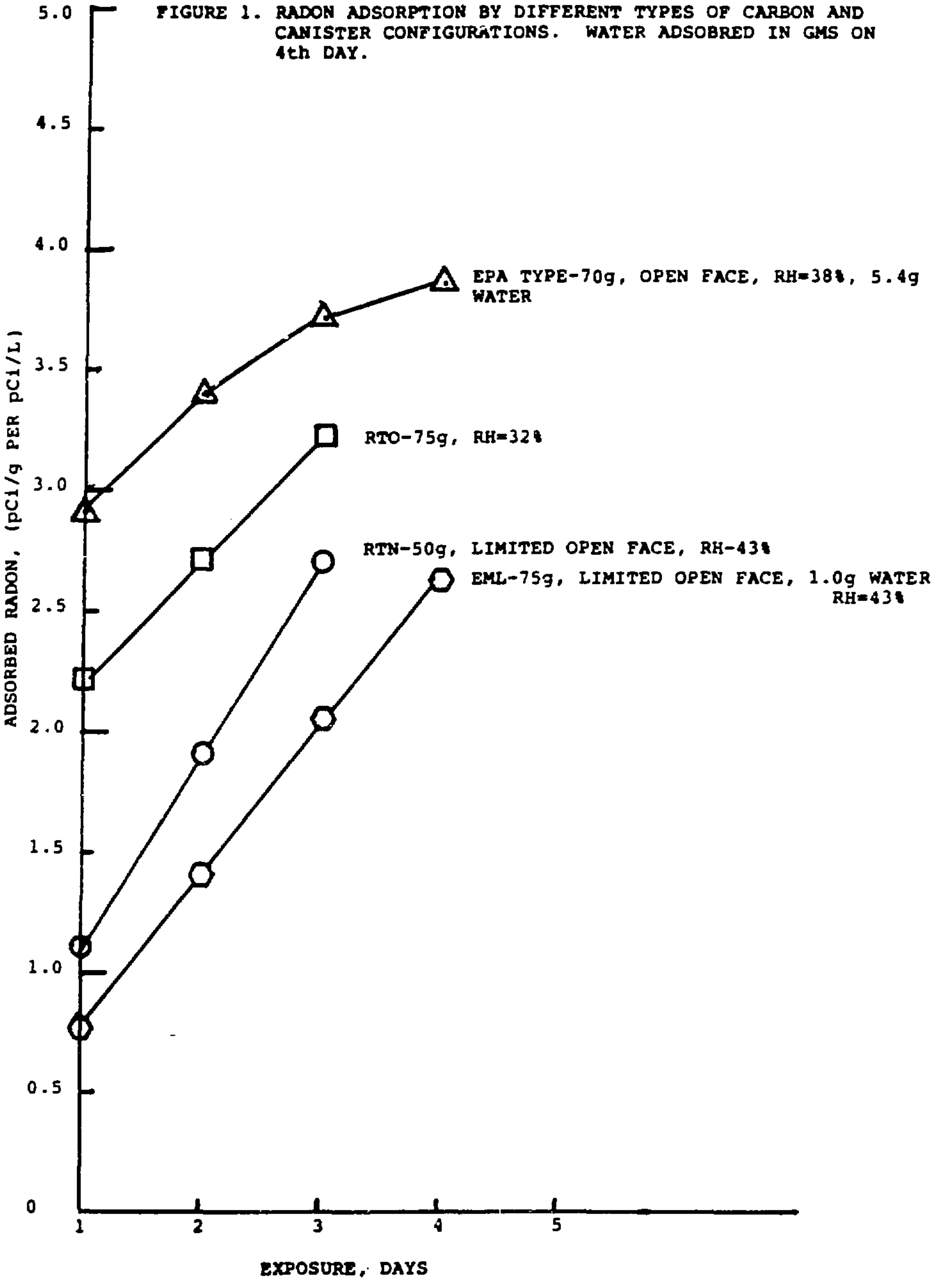
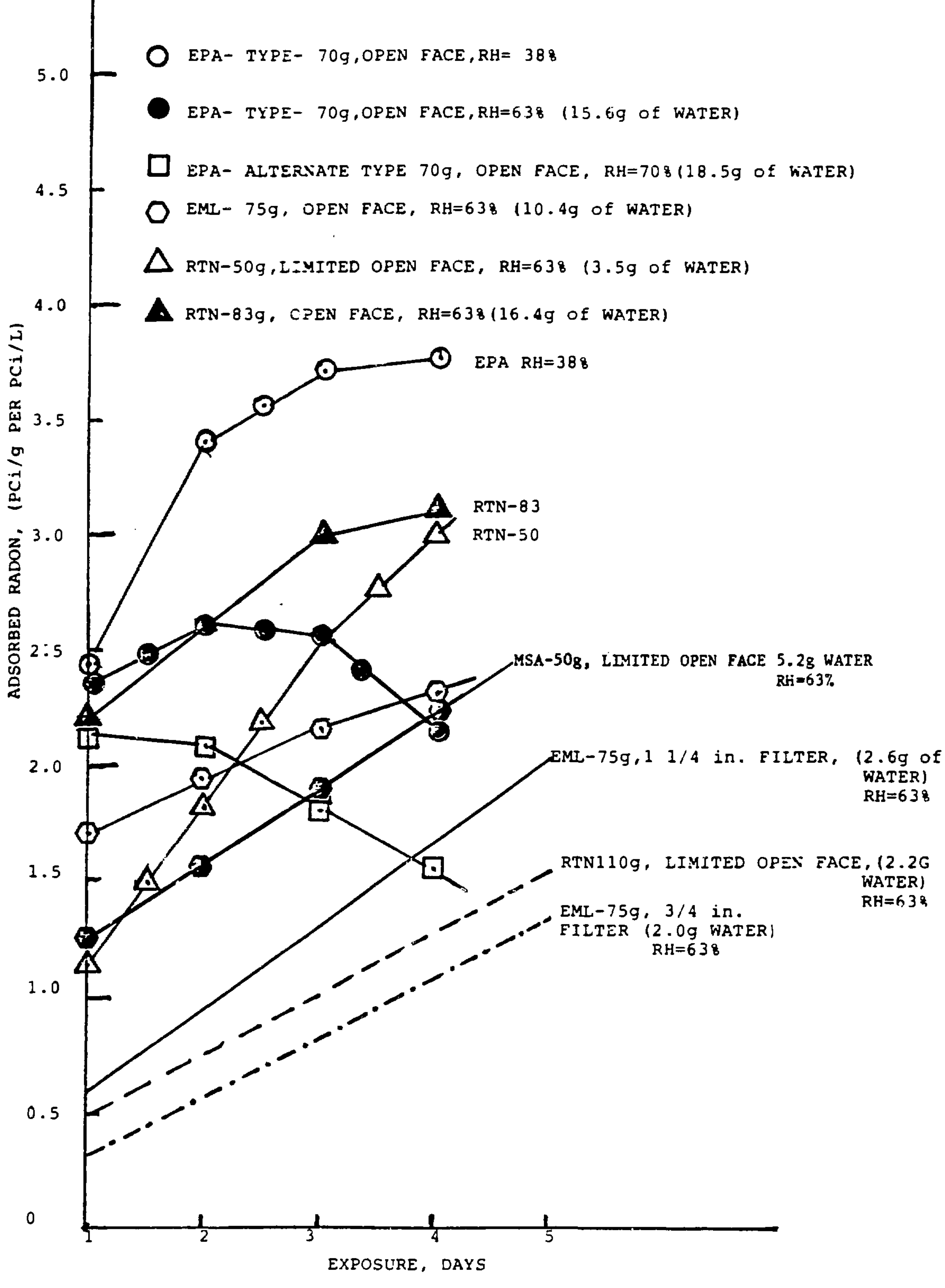
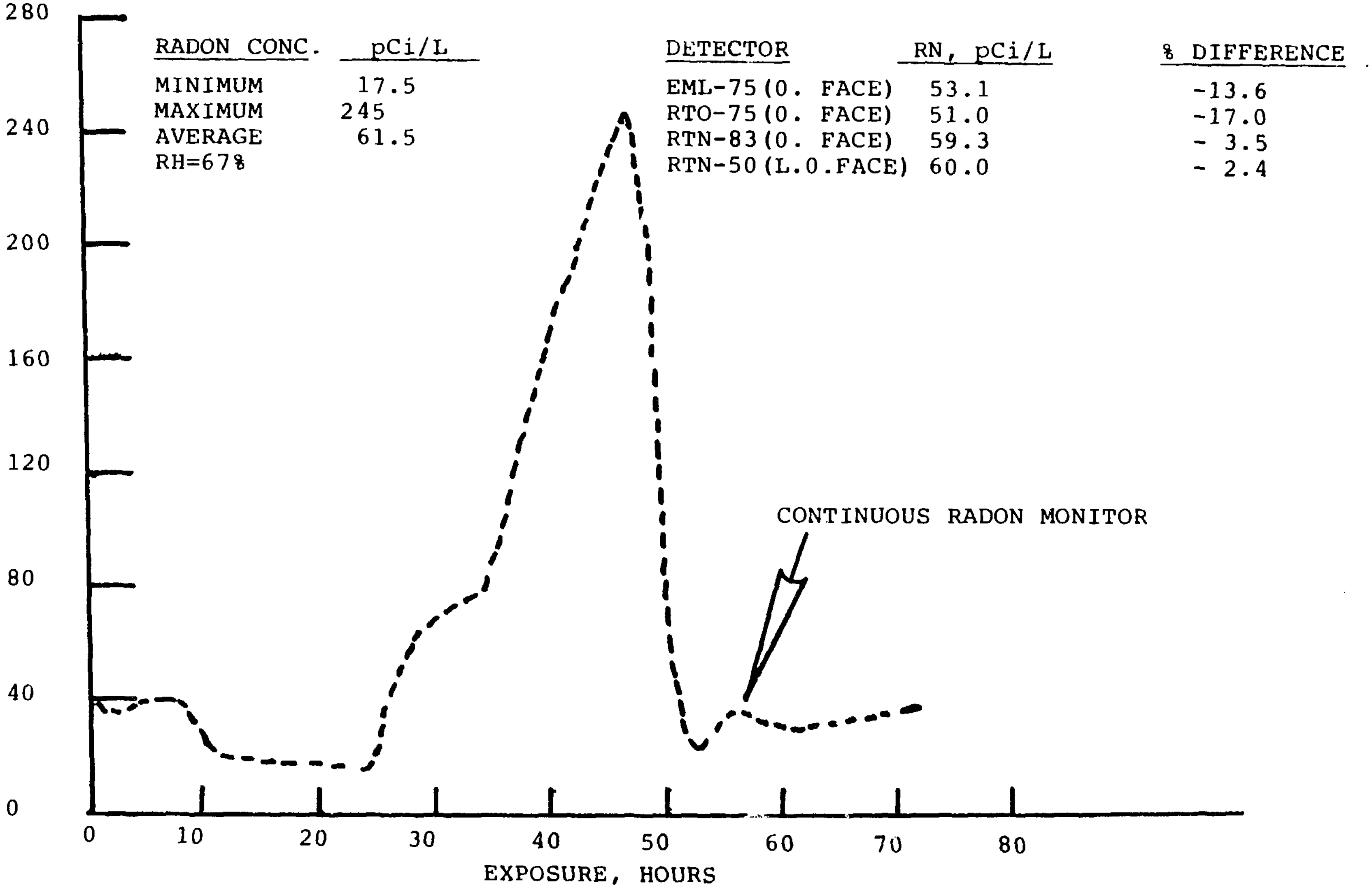


FIGURE 2. RADON ADSORPTION BY DIFFERENT TYPES OF CARBON AND CANISTER CONFIGURATIONS- WATER ADSORBED IN GMS ON THE 4th DAY. RADON CONCENTRATION = 35pC/L.





RADON CONC.	pCi/L
MINIMUM	17.5
MAXIMUM	245
AVERAGE	61.5
RH=67%	

DETECTOR	RN, pCi/L	% DIFFERENCE
EML-75(O. FACE)	53.1	-13.6
RTO-75(O. FACE)	51.0	-17.0
RTN-83(O. FACE)	59.3	- 3.5
RTN-50(L.O.FACE)	60.0	- 2.4

FIGURE 3. ACTIVATED CARBON MONITOR RESPONSE, WITH CHANGING RADON CONCENTRATION

SECTION G

Operational Experiences in Statewide Radon Surveys

Harry E. Rector and William A. Schoenborn,
GEOMET Technologies, Inc.

OPERATIONAL EXPERIENCES IN STATEWIDE RADON SURVEYS

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GEOMET Technologies, Inc., recently completed a study of radon concentrations in 6,500 homes throughout the State of Florida. The objective of this study was to identify all significant land areas in Florida where the State environmental radiation rule should be applied.

To accomplish this objective, two parallel approaches were used to collect data--a land-based survey keyed to inhabited land areas in each county and a population-based survey scaled to the number of occupied housing units in each county. The land-based survey was conducted by GEOMET technicians who deployed charcoal canisters and, in a subset of homes, indoor alpha-track detectors; both samplers were returned for analysis by the homeowner. In addition, measurements of soil radon and ambient gamma radiation were performed. The population-based survey included a mailout of charcoal canisters for deployment and return by homeowners. Both surveys were restricted to residences with slab-on-grade foundations. Indoor radon sampler (charcoal canister) return rates were 97.4 percent for the land-based survey and 82.4 percent for the population-based survey.

Quality control and assurance procedures included participation in the EPA's Radon Measurement Proficiency Program (RMP), training of field technicians by GEOMET staff, field blanks and duplicates, and transfer of technology from GEOMET's main office to a satellite laboratory established in Florida for analysis of charcoal canisters.

The accuracy and precision of the charcoal canister measurements in the EPA RMP program were generally within 5 percent, well within the EPA's guideline of 25 percent. The precision of colocated radon samplers (charcoal canisters) in homes with more than 2 pCi/L averaged ±5 percent in the land-based survey and ±8 percent in the population-based survey.

Indoor alpha track detectors were deployed in approximately 10 percent of the homes sampled in the land-based survey. The duration of deployment for these detectors was 30 days, compared with 72 hours for the charcoal canisters. The accuracy and precision of the alpha-track detectors, as measured in the RMP program, were both near 10 percent. The precision of the alpha track detectors used for field measurements was poor, averaging near ±40 percent, due to the relatively low concentrations measured and the short exposure period.

Charcoal canisters were colocated with indoor alpha track detectors in 232 homes in the study. The average measurement result for all homes with the charcoal canisters was 0.76 pCi/L and was 0.95 pCi/L for alpha track detectors. The correlation between the two sets of test results was 0.7.

SECTION H

Calibration and Experience with a Diffusion Barrier Charcoal Device and An Alternative Method for Charcoal Moisture Correction

A. S. Rood, Oak Ridge National Laboratory
(Grand Junction)
P. D. Kearney, Colorado State University

CALIBRATION AND EXPERIENCE WITH A DIFFUSION BARRIER CHARCOAL DEVICE AND AN ALTERNATIVE METHOD FOR CHARCOAL MOISTURE CORRECTION

A.S. Rood

Oak Ridge National Laboratory, Grand Junction Office

P. D. Kearney

Department of Physics

Colorado State University

CALIBRATION AND EXPERIENCE WITH A DIFFUSION BARRIER CHARCOAL DEVICE

A. KINETIC MODEL FOR RADON ABSORPTION ON CHARCOAL

$$R \dashrightarrow \boxed{\;Q\;} \dashrightarrow K$$

WHERE: Q = AMOUNT OF RADON IN CHARCOAL

R = RATE OF RADON INTAKE = Concentration (Ca) x sampling rate (r)

K = THE EFFECTIVE RATE CONSTANT ON RADON ON CHARCOAL

(Fig. 1)

DIFFERENTIAL EQUATION:

$$\frac{dQ}{dt} = R - K\,Q \qquad (1)$$

FINAL EQUATION:

$$Ca = \frac{C(Tc) \quad K \quad \exp(Kp\ Tc)}{Y\ r\ (1 - \exp[-K\ Ts])} \qquad (2)$$

WHERE:

Ca = THE RADON CONCENTRATION (PCi/l)

Y = THE COUNT YIELD (cpm/pCi)

C = THE NET COUNT RATE (cpm)

r = THE SAMPLING RATE (l/min)

Ts = THE EXPOSURE TIME

Tc = THE TIME BETWEEN EXPOSURE AND COUNT TIME

(Fig. 2)

B. DETERMINATION OF r, (the sampling rate)

1. PLACE DEVICE IN A KNOWN RADON CONCENTRATION AND SOLVE FOR r IN EQUATION 2.

$$r = \frac{C(Tc) \quad K \quad \exp(Kp\ Tc)}{Y\ Ca\ (1 - \exp[-K\ Ts])} \qquad (3)$$

WHERE Ca = THE KNOWN RADON CONCENTRATION

C. ADVANTAGES OF A DIFFUSION BARRIER:

1. REDUCES THE ERROR INTRODUCED BY FLUCTUATING RADON CONCENTRATION. (Fig. 3, 4, 5)

2. REDUCES THE MOISTURE UPTAKE (Fig. 6)

D. DISADVANTAGES OF A DIFFUSION BARRIER:

1. REDUCED SENSITIVITY OF DETECTOR TO RADON (Fig. 7)

AN ALTERNATIVE METHOD FOR MOISTURE CORRECTION

INTRODUCTION AND REVIEW

A. Moisture affects the radon collection efficiency on charcoal by occupying available space on the charcoal that would otherwise be occupied by radon.

B. Diffusion barriers inhibit moisture uptake in charcoal, however, many of the radon collection devices presently employed do not have diffusion barriers. Furthermore, the measurement of radon flux using charcoal requires a large surface area of charcoal exposed which is susceptible to large uptakes of moisture if sampling is performed in humid climates or over saturated soil.

C. The purpose of this study was to develop a method for moisture correction which is not dependent on the sampling time of the device.

KINETICS OF WATER ON CHARCOAL

A. Water absorption on charcoal can be modeled similar to that of radon on charcoal. That is, the model

$$R_w \dashrightarrow \boxed{Q_w} \dashrightarrow K_w$$

DESCRIBES THE SYSTEM WHERE ...

Q_w = THE AMOUNT OF WATER ON THE CHARCOAL AT TIME t

R_w = THE RATE OF WATER INPUT

K_w = THE FIRST ORDER RATE CONSTANT OF WATER ON CHARCOAL

THE DIFFERENTIAL EQUATION IS

$$\frac{dQ_w}{dt} = R_w - K_w Q_w \qquad (1)$$

WHICH HAS THE SOLUTION WITH THE BOUNDARY CONDITIONS $Q_w(t) = 0$ AT $t = 0$

$$Q_w(t) = \frac{R_w(1 - \exp(-K_w t)}{K_w} \qquad (2)$$

C. The radon collection efficiency at any time t is inversely related to the amount of water on the charcoal. Thus the collection efficiency decreases with increasing water content.

D. What is of interest is the average radon collection efficiency during a sampling period. This value at any time t is related to the average water content during the sampling period.

E. The average water content during the collection period is obtained by finding the mean value of eq(2). Thus:

$$I = \frac{Rw\ [Kw\ ts + \exp(Kw\ ts) - 1]}{ts\ Kw} \quad (3)$$

where I = the average integrated water content

ts = the sampling time

F. A plot of I vs E (the radon collection efficiency) and subsequent regression provides a means for determining the average collection efficiency based on the water content.

PRACTICAL CONSIDERATIONS

A. Determination of Kw

1. Kw can be determined empirically by placing two devices in an environment with a constant relative humidity and temperature. One of the devices has an initial amount of water placed on it ($Q1(0) > 0$) and the other has had no water placed on it ($Q2(0) = 0$)

2. Solving eq(1) with the boundary conditions $Q1(0) > 0$ and re-arrangement of eq(2) will yield an expression for K in terms of Q1, Q2, Q1(0) and t

$$Kw = \ln((Q1-Q2)/Q1(0))/-t \quad (4)$$

B. R may be determined using equation 2

RESULTS

A. The results presented here are for an MSA charcoal cartridge used to make flux measurements. The procedure would apply to an ambient radon collection device.

B. The build up of water on charcoal was found to follow the proposed model. This observation was also noted by Rogers, 1983.

(Fig. 8)

C. Kw was found to be temperature dependent.

(Fig. 9)

D. The presence of moisture on the charcoal resulted in a more erratic effective rate constant for radon.

(Fig. 10)

E. The plot of collection efficiency vs average integrated water content was fitted to an exponential curve in the form:

$$E = 0.69 \exp(-0.052 I)$$

$$r^2 = 0.94$$

(Fig. 11)

CONCLUSION

This method in combination with a similar model for radon collection allows for flexibility in making radon measurements. The method may be advantageous if a homeowner accidentally leaves the device out for a longer than prescribed time.

LIST OF FIGURES

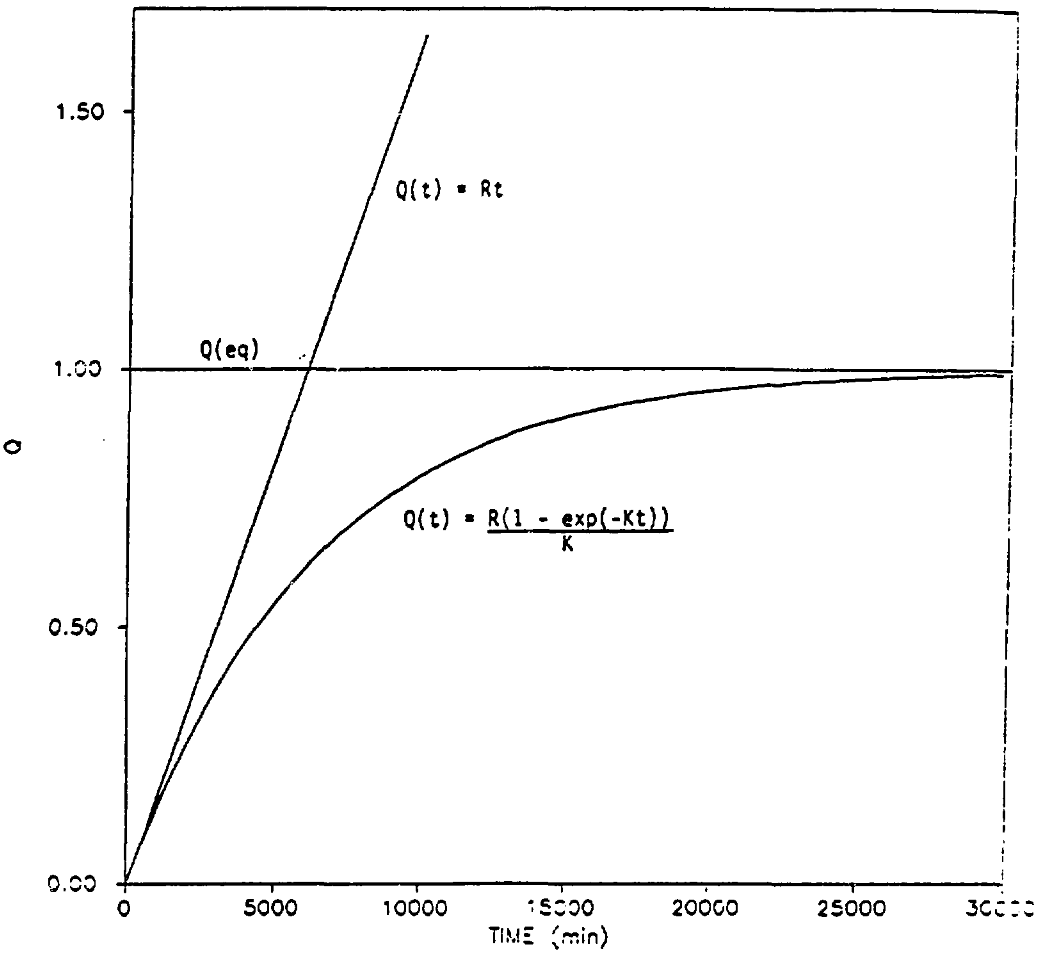

Figure 1. Hypothesized response of a charcoal collection device.

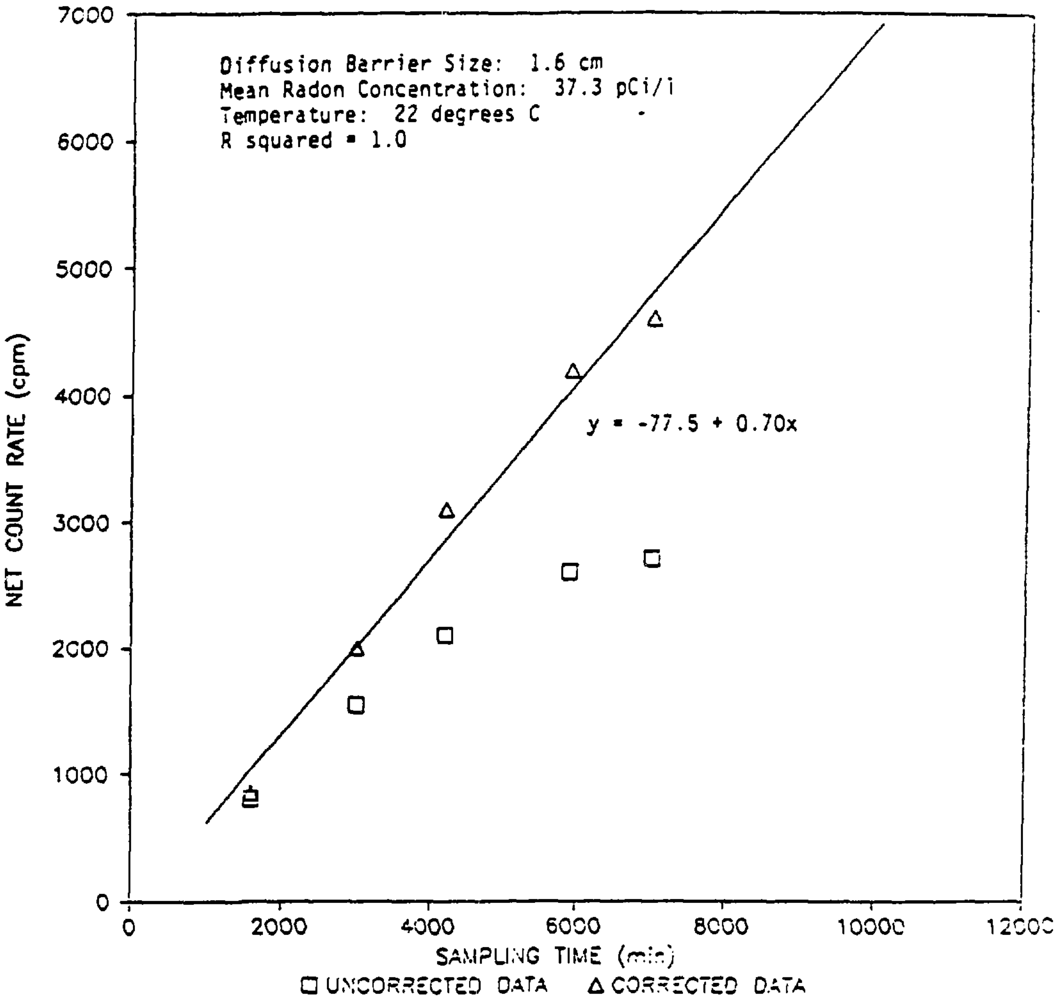

Figure 2. Detector count rate as a function of time. The squares represent data corrected only for decay after sampling and the triangles represent data corrected for decay and desorption during sampling.

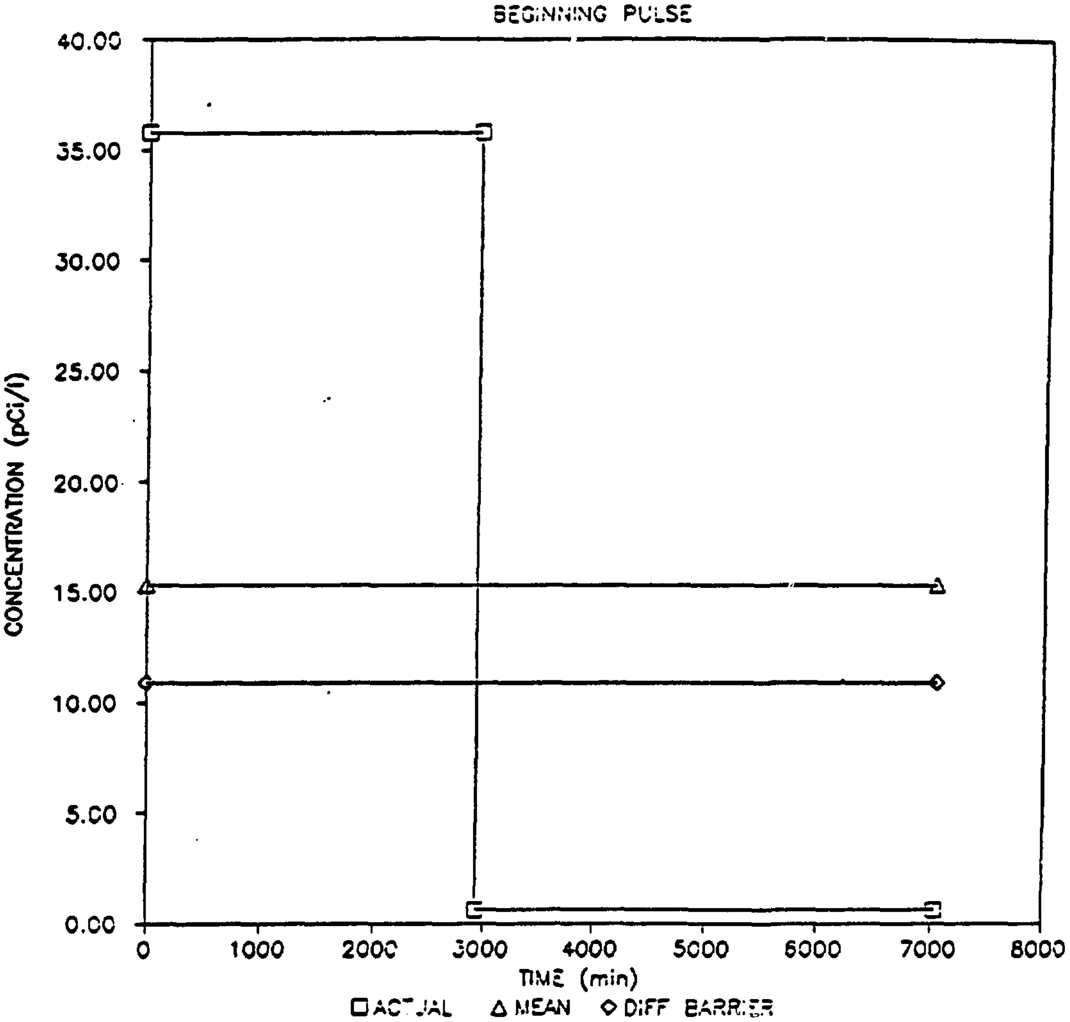

Figure 3. Measured and actual mean radon concentrations for a radon pulse at the beginning of exposure.

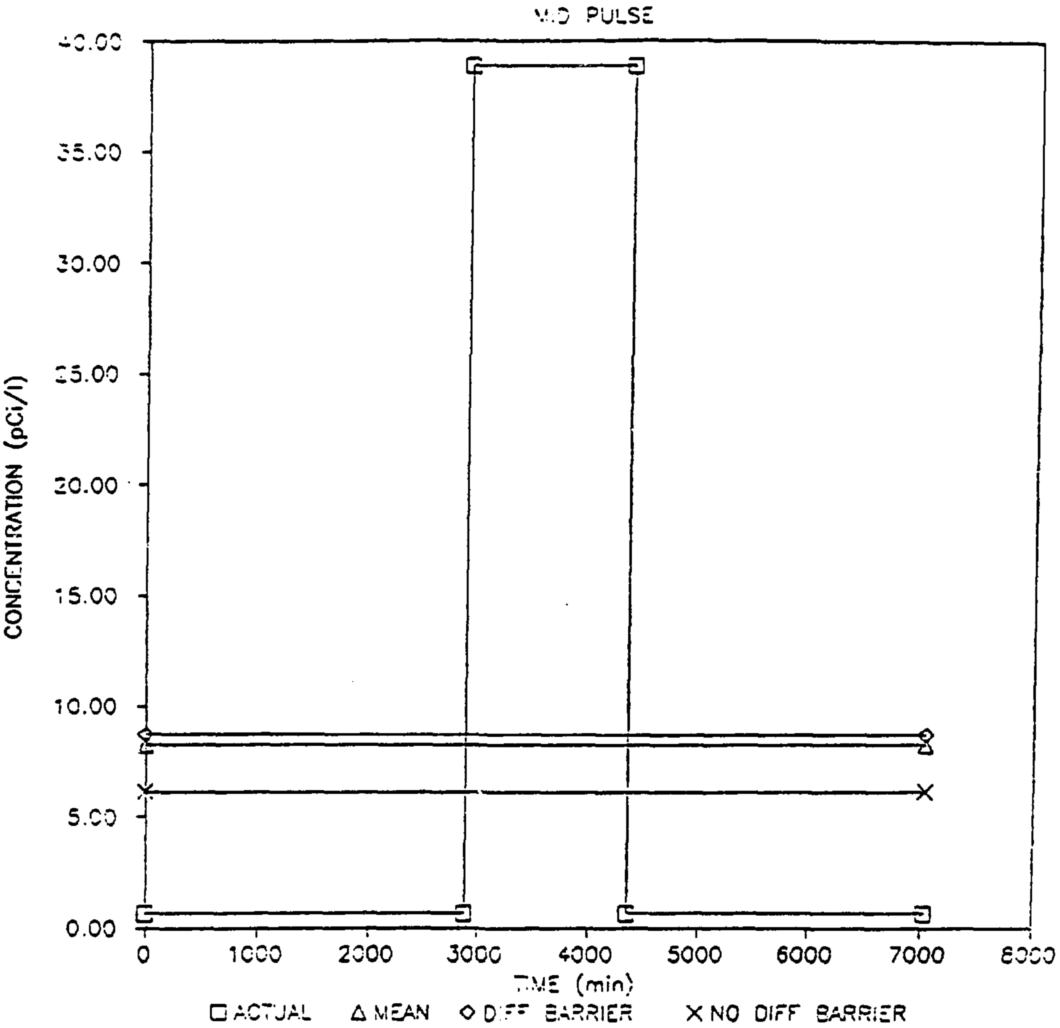

Figure 4. Measured and actual mean radon concentrations for a pulse at the middle of exposure.

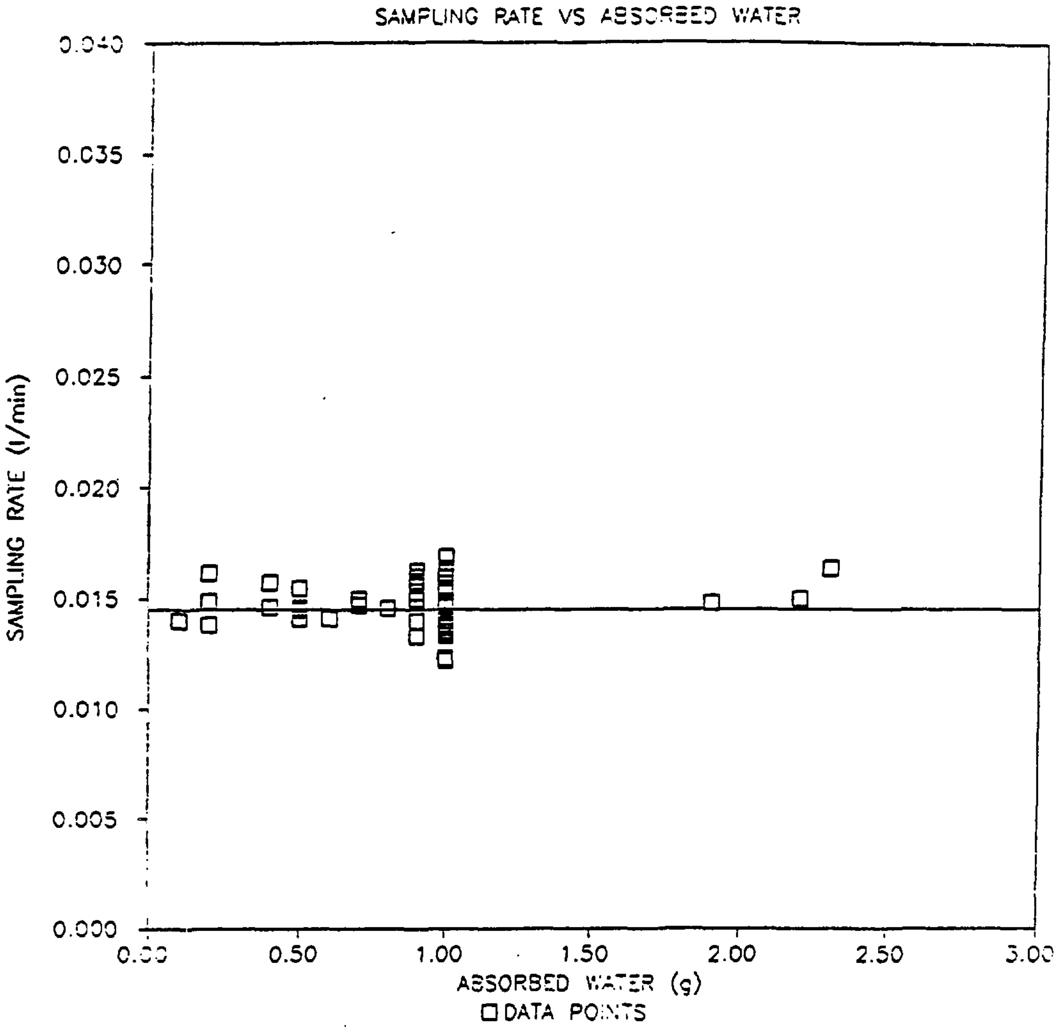

Figure 6. Sampling rate vs absorbed water for a diffusion barrier device.

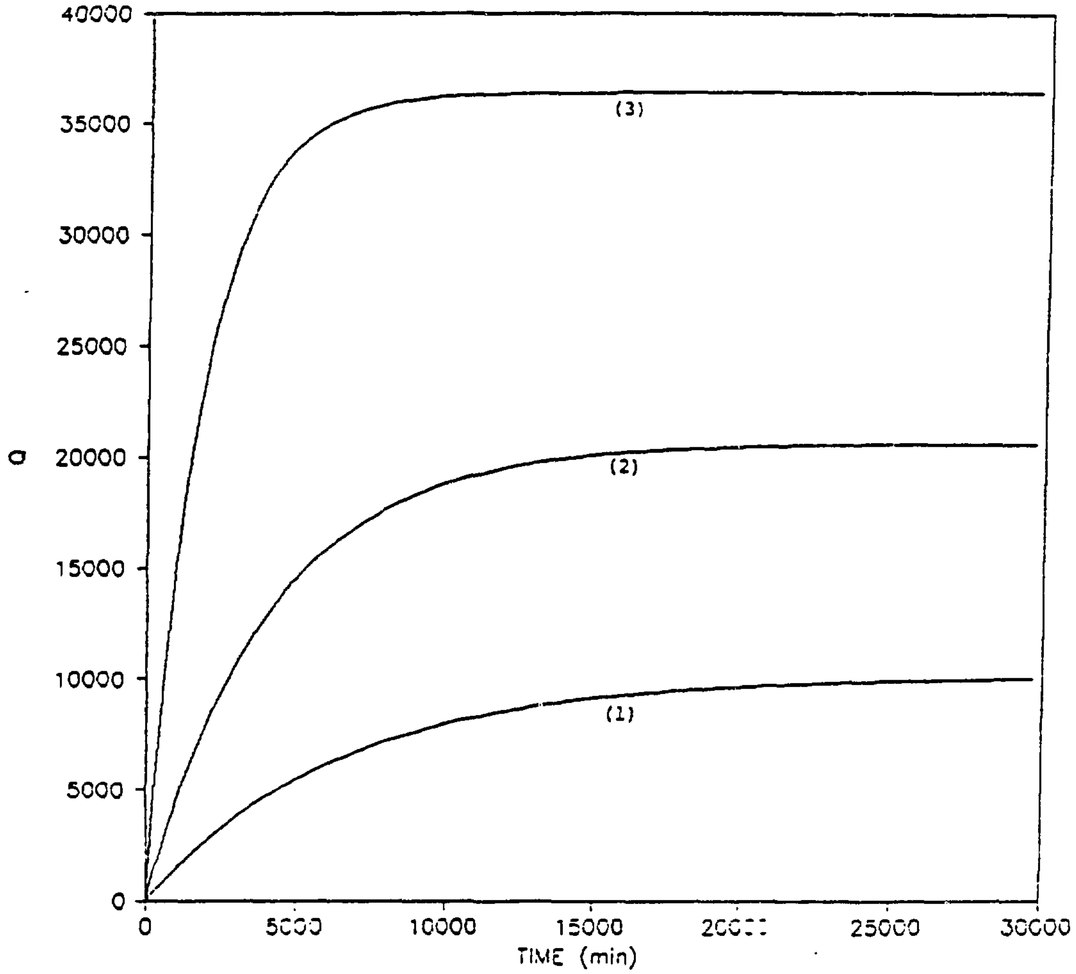

Figure 7. Hypothetical response of a charcoal device for various values of K, the effective rate constant. Curve 1, K = 1.58E-04; Curve 2, K = 2.48E-04; Curve 3, K = 5.5E-04.

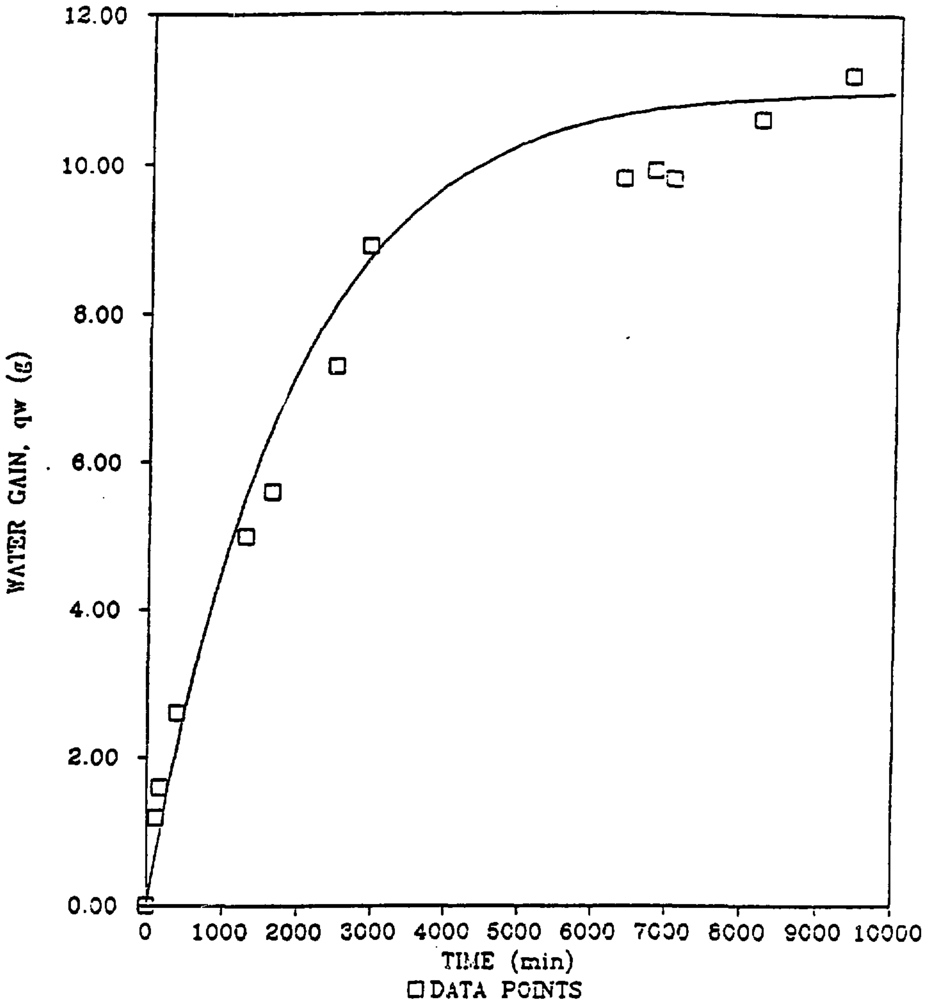

Figure 8. Build up of water on a charcoal device. The line represents the empirically determined Kw and Qw(eq).

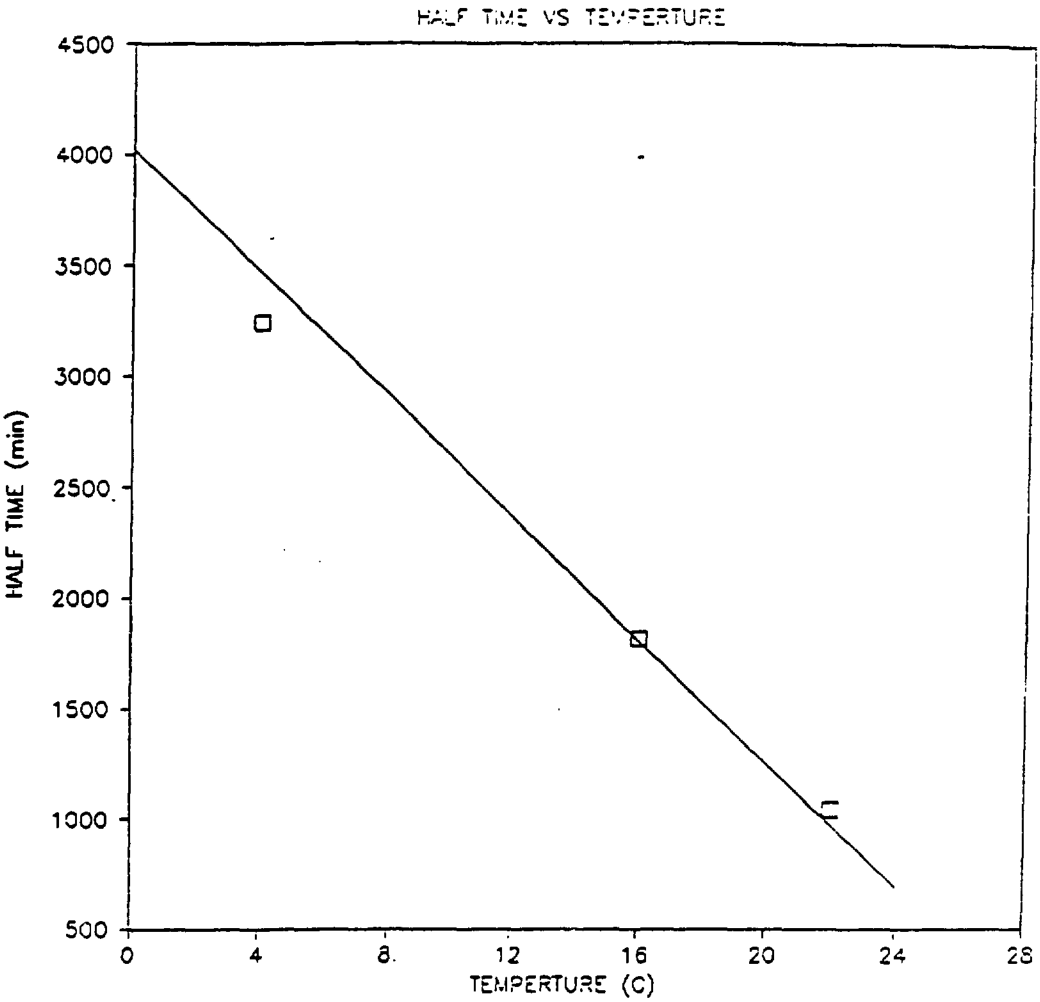

Figure 9. Half time of water on charcoal vs temperature.

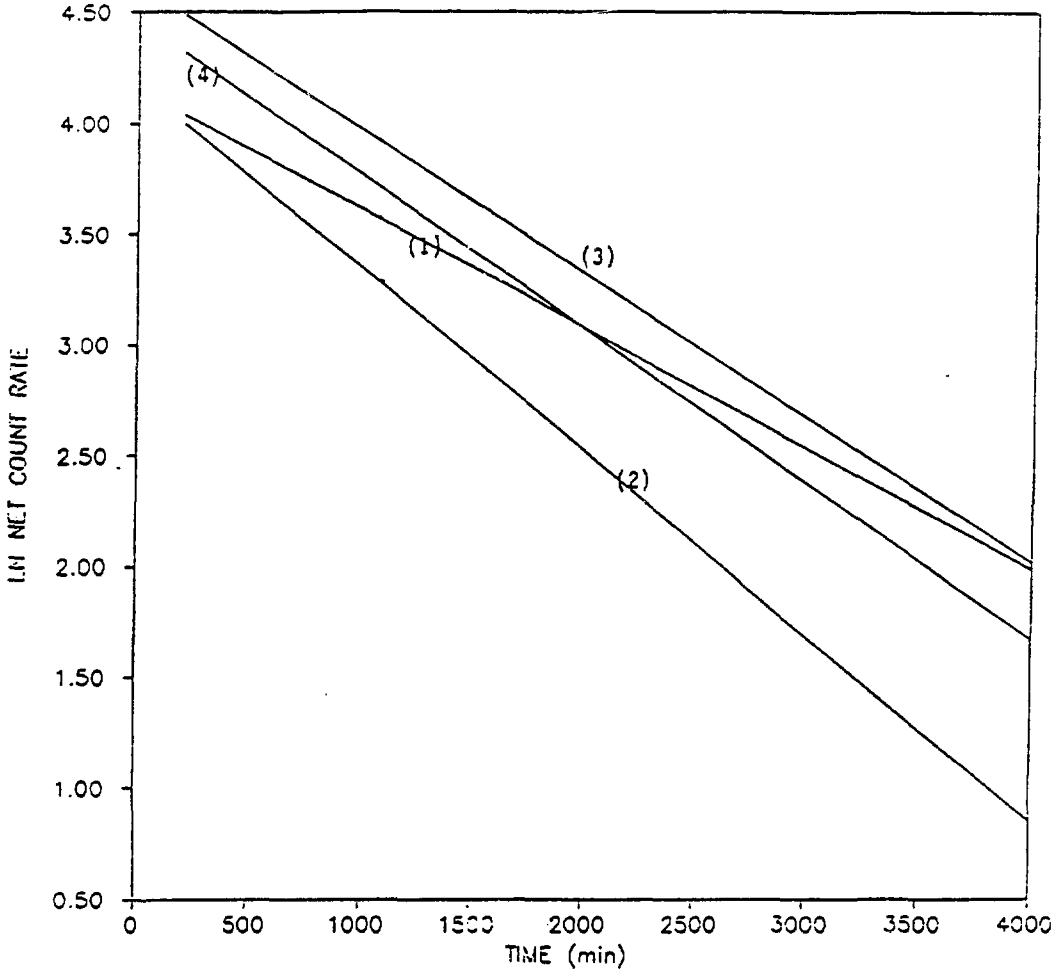

Figure 10. Natural log of the net count rate vs time for MSA cartridges with moisture (curve 1 and 2) and without moisture (curve 3 and 4).

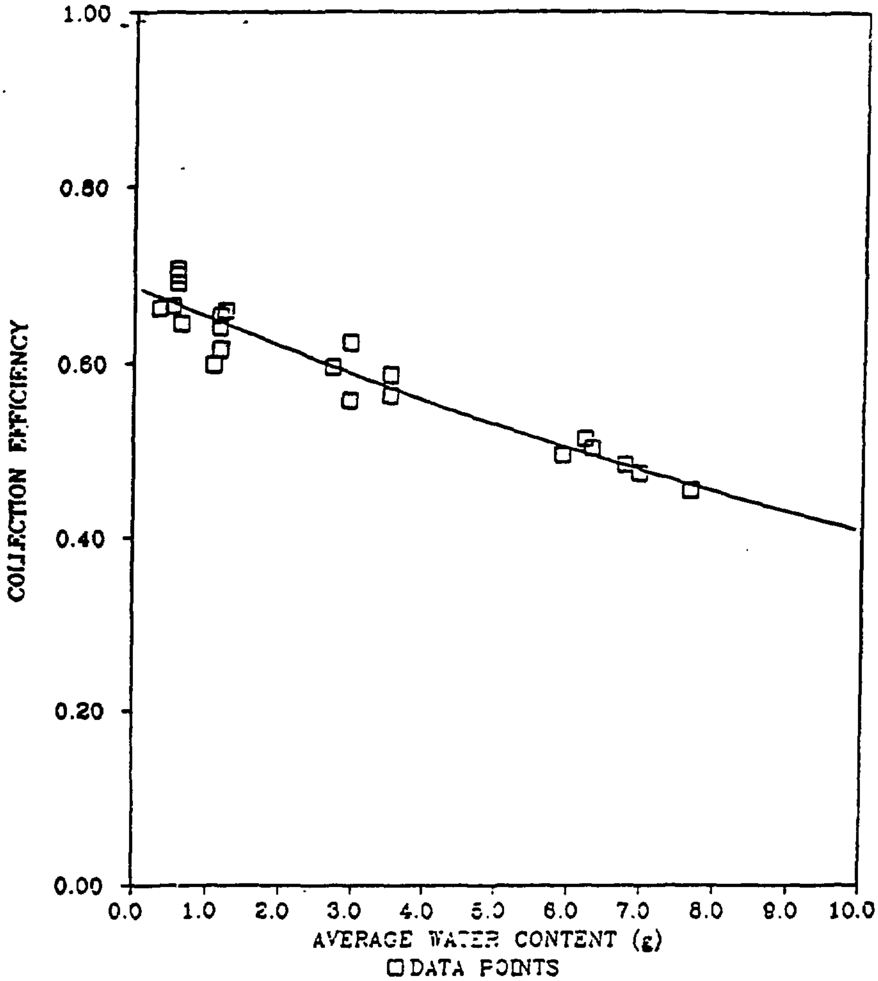

Figure 11. Radon collection efficiency vs average water content.

SECTION I

Moisture Insensitive Charcoal Canisters

Henry F. Lucas, Argonne National Laboratory

For Publication in: "Technical Exchange Meeting on Passive Radon Monitoring", Held in Grand Junction Colorado on September 21 and 22, 1987.

Moisture Insensitive Charcoal Canisters

Henry F. Lucas

Continuous monitoring of ^{222}Rn concentrations in the air in houses is the most appropriate approach for the real-time measurements, but this process requires complex and expensive instruments and is not practical for large studies. Activated carbon canisters have been used extensively for determining the average concentration over a period of a few days (Geo84). Because these "open face" charcoal detectors have an integration time constant of about 14 h, they are sensitive to short-term transient changes in the radon concentration. In addition, water uptake at high relative humidities reduces the radon uptake by the charcoal.

Cohen and Nason added a diffusion barrier and a nylon screen to a charcoal detector, producing a detector with an integration half-time ranging from 20 to 60 h and a reduced uptake of water at high humidities (Coh86). Other investigators have found that silicone rubber sheeting is relatively permeable to radon but impermeable to water vapor (Jen86). We decided to evaluate the effect of a silicone barrier on the charcoal canister radon collection device.

Experimental Procedure

The standard EPA-style open-face charcoal canisters* were modified by removing the screen and retaining ring. The edges of the screen were smoothed and taped. The filter paper was replaced with a sheet of silicone rubber,** after which the screen and the retaining ring were reinserted into the can. The ring was used to seal the silicone rubber against the side of the can except in the area of non-overlap.

Both styles of charcoal canisters were exposed to ^{222}Rn in our Radon Test Chamber at 23°C and 70% relative humidity for 4 to 7 days. The charcoal canisters were counted on a 4 x 4 NaI(Tl) detector in our underground counting room. where a continuous flow of radon-free air is used to purge the 100-cm-thick iron shield. The precision of the results for both styles of charcoal canisters was within counting error. The results obtained for a single four-day exposure are shown in Figure 1. Under these conditions, the uptake and retention of radon by the EPA-style open-face charcoal canister is nearly a factor of five greater than that by the canister with the rubber membrane. The background counting rate is 207 cpm so that the detection limit (3 sigma) for a 10-m count is 0.1 and 0.5 pCi/L for the open-face and modified charcoal canisters, respectively.

---

*Radon Collection Filter, Model RA40V, F and J Specialty Products, Inc., P.O. Box 660065, Miami Springs, FL 33166.

**Silicone rubber sheeting, vulcanized, non-reinforced, 0.005-inch-thick, Cat. # 500-1, Dow Corning, Box 997, Midland, Michigan 48640-4517.

The uptake of water was evaluated by placing both types of detectors in a 20-L can in which the humidity was maintained at 100%. When tested over a period of 20 days, the uptake of water by the canister with the silicone rubber membrane was about 20% of that by the open-face detector. Additional studies are in process with improved sealing of the membrane to the can.

Acknowledgement

The work described herein was performed under the auspices of the U. S. Department of Energy, under Contract W-31-109-ENG-38.

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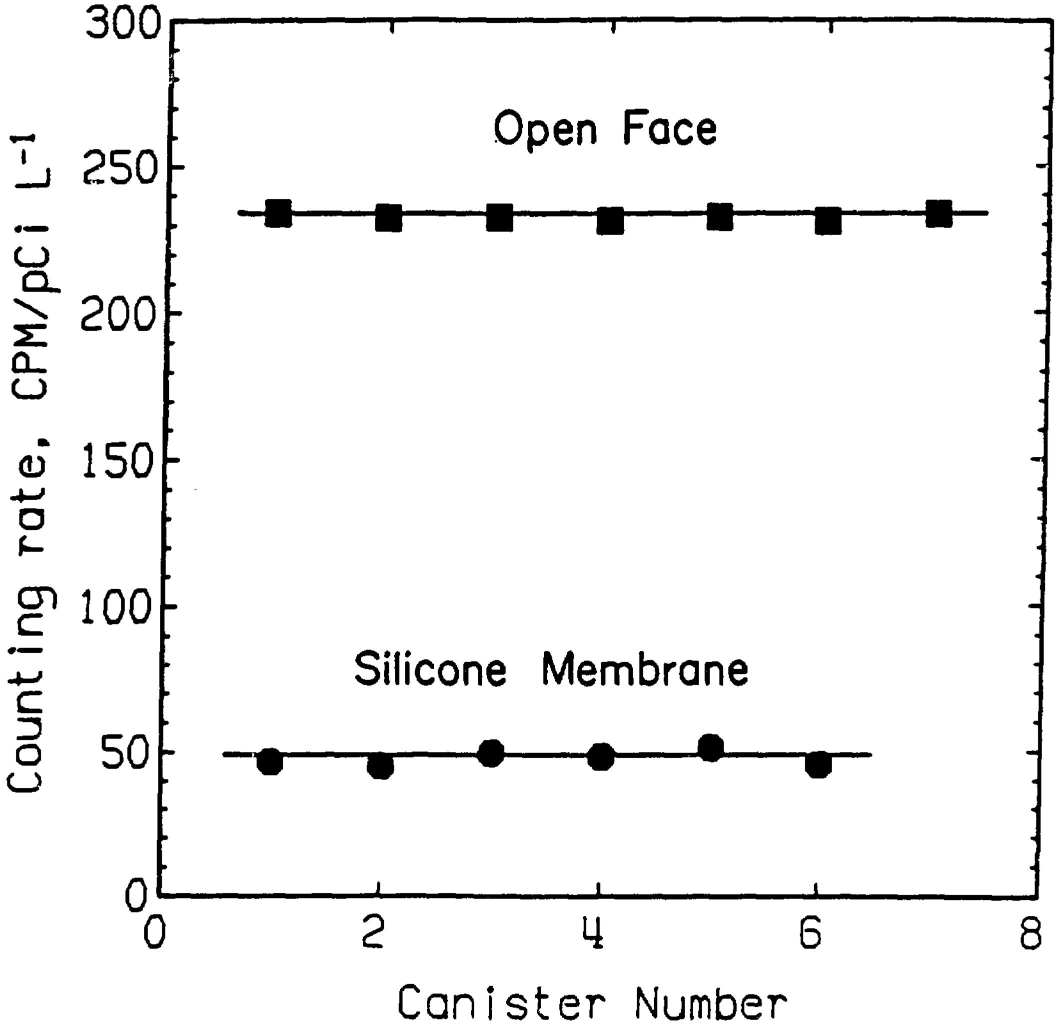
Geo86 A. C. George, Passive Integrated Measurement of Indoor Radon Using Activated Carbon, Health Phys. Vol. 46, pp. 867-872 (1984).

Jen86 Personal Communication.

Figure 1


Open Face
Silicone Membrane
Counting rate, CPM/pCi L^{-1}
300
250
200
150
100
50
0
0
2
4
6
8
Canister Number

SECTION J

A Mathematical Model for Radon Sampling by Charcoal Adsorption

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Illinois Institute of Technology
L. D. Michaels, V. B. Menon, and D. S. Ensor,
Research Triangle Institute

A MATHEMATICAL MODEL FOR RADON SAMPLING BY CHARCOAL ADSORPTION

by

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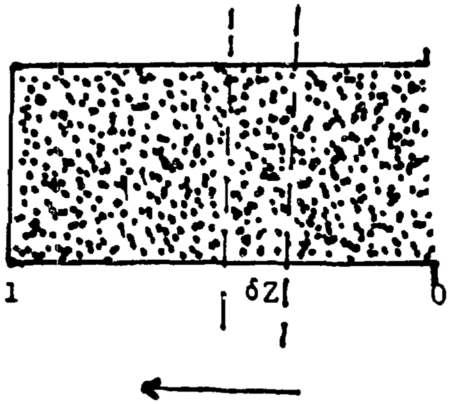
Abstract

An adsorption/diffusion model for the transport of radon in an activated charcoal porous bed is developed. Such a model is relevant to the charcoal canister sampling technique. The radon gas diffuses in the pores of the charcoal bed and is adsorbed on the surface of the granules. A mass balance over a differential volume equates the net accumulation to the net diffusional flux minus the decay. By posing appropriate boundary conditions (for example either constant or varying ambient radon concentration), the differential equation is solved numerically. The model predictions of the response of the canister for both constant as well as a periodic radon concentration is seen to agree well with experimental data obtained under controlled laboratory conditions. The results show that it is feasible to use computer simulations to test the performance of different types of canisters which makes it an effective tool to design experiments.



TERMINOLOGY

C	Radon Concentration in Pores of Bed pCi/cm^3
$C_0(t)$	Surrounding Radon Concentration pCi/cm^3
q	Adsorbed Radon Concentration pCi/g
ε	Porosity based on Void Volume Fraction
ε_a	Porosity based on Cross Sectional Area Fraction
ρ_c	Density of Charcoal g/cm^3
D_{eff}	Effective Diffusivity cm^2/s
λ	Decay Constant $1/s$

ASSUMPTIONS

. Packed Bed is Macroporous
. Axial Diffusion only, Radial Diffusion Negligible
. Unsteady State Analysis
. Isothermal
. Humidity Effects Ignored
. Adsorption/Desorption is Instantaneous

TRANSPORT EQUATION

Material Balance

$$\varepsilon \frac{\partial C}{\partial t} + (1 - \varepsilon)\rho_C \frac{\partial q}{\partial t} = \varepsilon D_{eff} \frac{\partial^2 C}{\partial z^2} - \lambda(\varepsilon C + (1 - \varepsilon)\rho_C q)$$

Adsorption Isotherm

$$q = f(C)$$

$$\frac{\partial q}{\partial t} = \frac{dq}{dc} \frac{\partial C}{\partial t}$$

Initial and Boundary Conditions

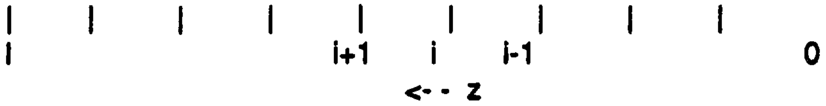
$$t = 0 \qquad 0 < z < l \qquad C = 0$$

$$z = 0 \qquad t > 0 \qquad C = C_0$$

$$z = l \qquad t > 0 \qquad -D_{eff} \frac{\partial C}{\partial z} = 0$$

NUMERICAL SOLUTION

Method of Finite Differences



$$\frac{\partial U}{\partial t} = \frac{\partial^2 U}{\partial z^2}$$

Backward (Implicit Method) Differencing

$$\frac{U_i^{n+1} - U_i^{n}}{\Delta t} = \frac{U_{i+1}^{n+1} - 2U_i^{n+1} + U_{i-1}^{n+1}}{\Delta z^2}$$

Solve Linear Algebraic Equations on a Computer

Adsorption Isotherm (Linear)

$$q = K_a C$$

Diffusivity

$$D_{eff} = D_{12}/\tau$$

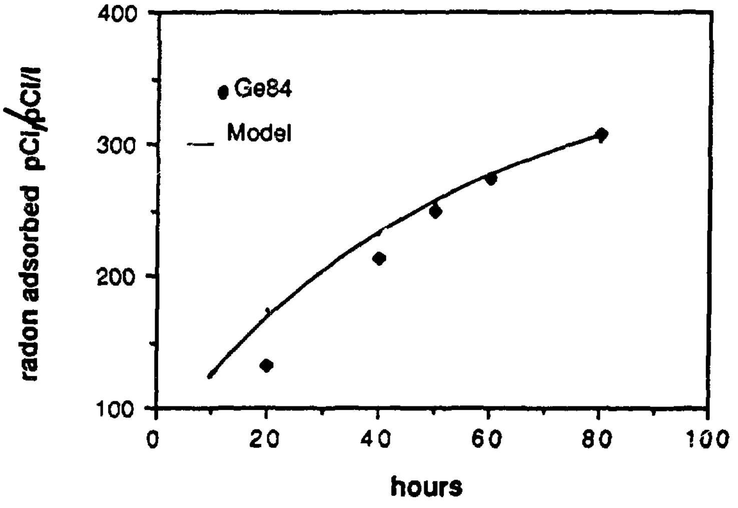
Typical Parameter Values

L	1.27cm
A	87 cm^2
D_{eff}	4.2 X 10^{-2} cm^2/sec
λ	2.1X 10^{-6} sec^{-1}
K_a	6.0 l/g
A	76 cm^2
ε	0.436
t	3
ρ_c	1.1 g/cm^3

CAUSE Constant Ambient Concentration

EFFECT

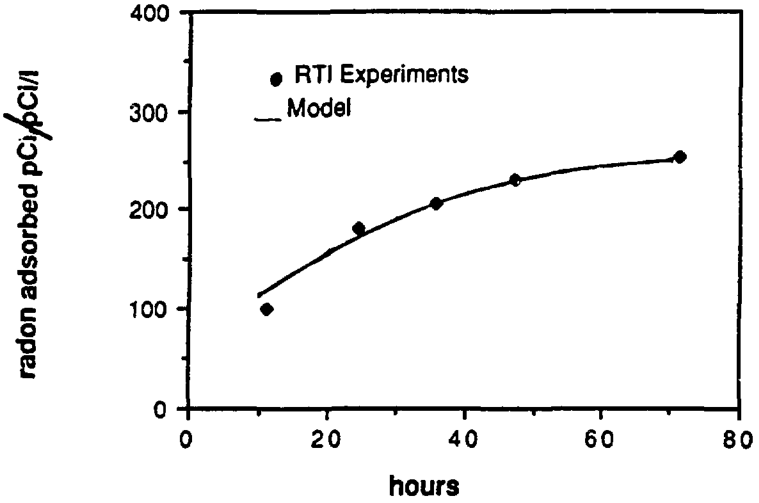
Radon adsorbed versus Time . Experiments - Ge 84

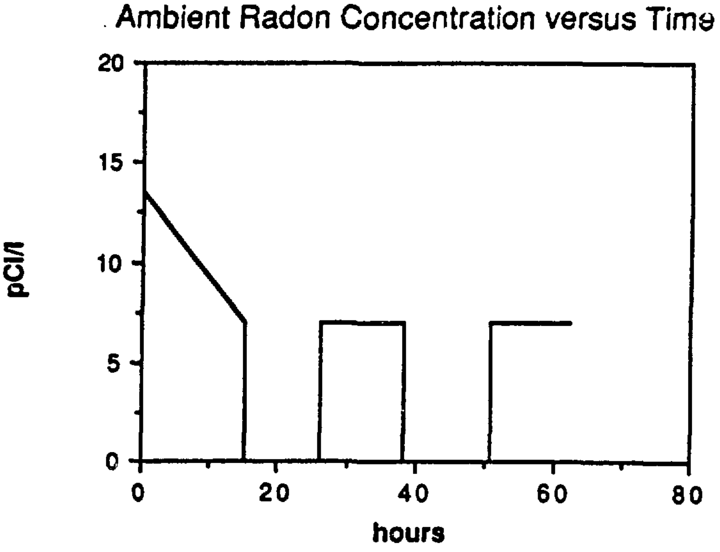


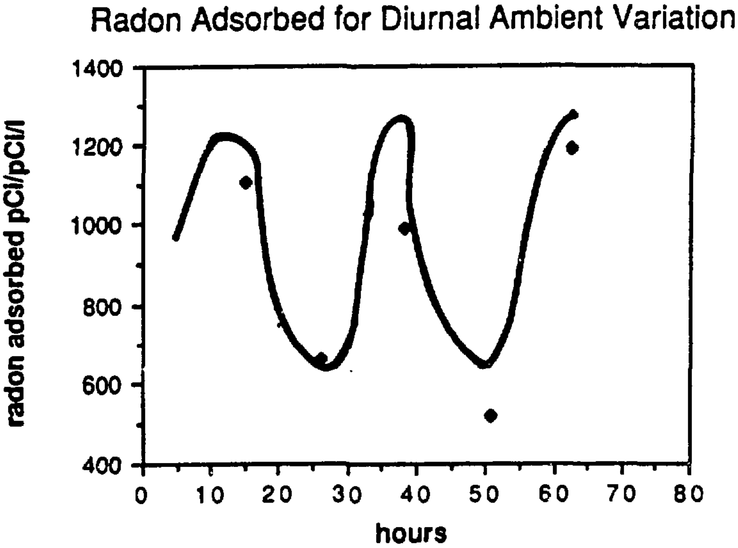
CAUSE Constant Ambient Concentration

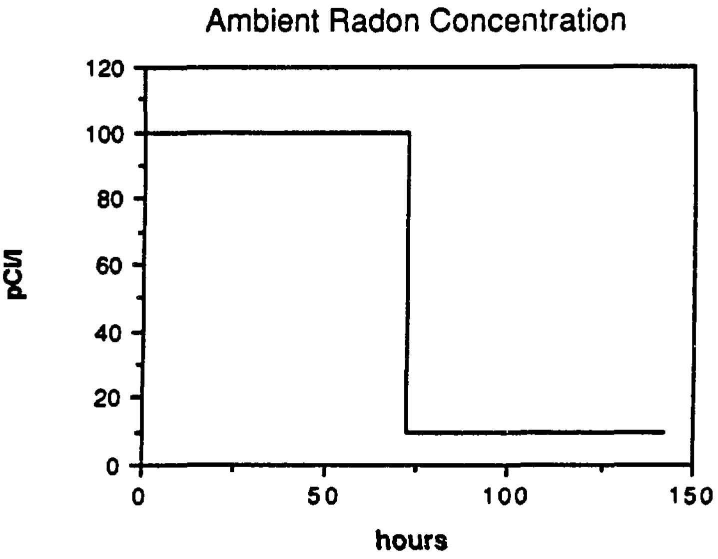
EFFECT

Radon Adsorbed versus Time Experiments -- RTI

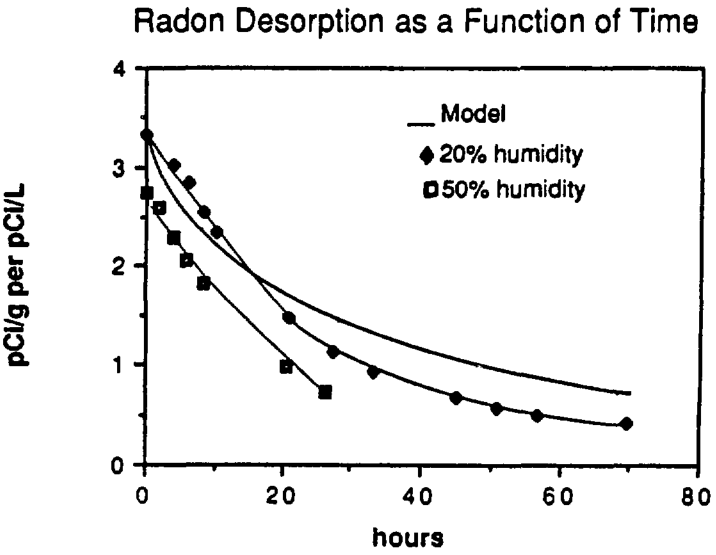



Ambient Radon Concentration versus Time
pCi/l
20
15
10
5
0
0
20
40
60
80
hours


Radon Adsorbed for Diurnal Ambient Variation
radon adsorbed pCi/pCi/l
1400
1200
1000
800
600
400
0
10
20
30
40
50
60
70
80
hours

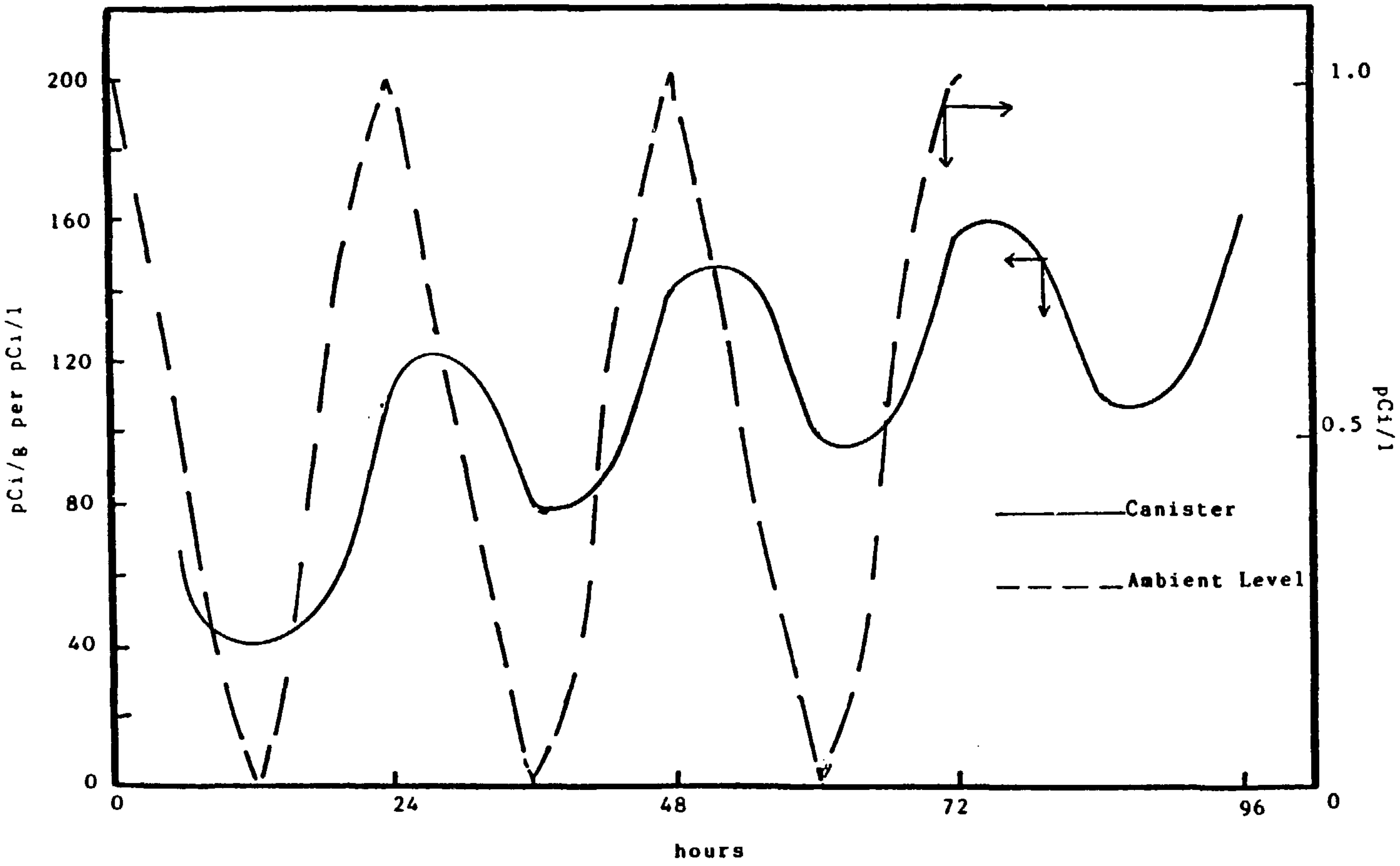


Ambient Radon Concentration



Radon Desorption as a Function of Time

Response of charcoal canister to a sinusoidal radon concentration variation



SECTION K

Charge Effects on Radon Alpha Track Distribution

Robert W. Pollock and Eugene V. Benton

Radon Environmental Monitoring, Inc.

Charge Effects on Radon Alpha Track Distribution

Robert W. Pollock

Eugene V. Benton

Work in Progress

The calibration factor for nuclear track detectors has been found to vary unpredictably with a standard deviation of 16%, presumably due to electric charge effects associated with use of plastic cups.

Bernard L. Cohen
Health Physics, 1986

Extensive experiments to show any correlation with ATD response and electric charge have shown no such correlation.

H.W. Alter
R.A. Oswald
R.V. Wheeler
Health Physics, 1987

Cohen attributes a 16% S.D. variation in calibration factors in other alpha track monitors to "electric charge effects associated with the use of plastic cups." This potential problem is recognized, and to solve this specific problem every Radtrak holder is made of electrically conducting plastic.

R.V. Wheeler
Radon – A Monitoring Strategy
Update, 1986

AT-100 Radon Monitor

- Hemispherical cup, 2.8 cm. diameter
- Electrically conductive plastic, 50 ohm-cm

Metalic conductors:	10^{-6} to 10^{-4} ohm-cm
Semi-conductors:	10^{-2} to 10^{7} ohm-cm
Conductive plastics:	10 to 10^{6} ohm-cm
Insulators:	10^{7} to 10^{20} ohm-cm

Conclusions

- Clumping not statistically significant with bare foils in AT-100 cup
- Higher sensitivity obtained with grounded foils
- Excess variability due to CR-39, as neutron exposures also show standard deviation of 16% at similar track densities.

The responses of diffusion chambers using CR-39 plastic treated in different ways.

	X-rayed (16.5 R)	Water Soaked	HCl Soaked	Aluminum Overlayer
Mean	0.0818	0.0845	0.0845	0.0872
S.D.	0.0031	0.0078	0.0078	0.0019

Response values are in tracks per square centimeter per pCi-hr/l

Exposure Conditions

— High Concentration, 1500 pCi/l

— Short Exposure Time, 45 minutes

— Low Track Density, 50 - 100 tracks per cm^2

— 3 Foil Types

Bare

Coated, 1000 A of Al

Coated and Grounded to Cup

	Tracks per square centimeter
Bare	54
Coated	68
Grounded	80

Quantifying the spatial track distribution

- Method developed by Hopkins & Skellam
- Measure distance to nearest neighbor
- Discard tracks closer to edge
- Compare distribution of distances to distribution from randomly generated points

	A	X	Z
Bare	0.909	0.524	+ 0.56
Coated	1.172	0.463	- 1.02
Grounded	0.884	0.531	+ 0.94

$$A = \frac{\text{sum of random distances squared}}{\text{sum of experimental distances squared}}$$

For n>50, the variate X has a normal distribution

$$X = \frac{1}{1 + A}$$

with a mean of 0.5

and a standard deviation $= \dfrac{1}{0.5\,(2N + 1)^{0.5}}$

SECTION L

A Comparison of Four Types of Alpha-Track Radon Monitors

Mark D. Pearson, UNC Geotech

A COMPARISON OF FOUR TYPES OF ALPHA-TRACK RADON MONITORS

Mark D. Pearson
UNC Geotech
Technical Measurements Center

The results of a series of 24 exposures of alpha-track radon monitors in the Department of Energy Grand Junction Projects Office Radon/Radon-Daughter Environmental Chamber conducted by the Technical Measurements Center over the past 18 months are presented. This test series was designed to evaluate the sensitivity of approximately 15 radon and radon-daughter monitoring devices to a number of environmental parameters, including temperature, relative humidity, different concentrations of condensation nuclei, and non-uniform radon concentrations.

The results obtained on four types of alpha-track radon monitors are reported: Terradex Track Etch® Type SF and Type F monitors, Glenwood Laboratories Radtrak® monitor, and the Radon Environmental Monitoring AT-100® monitor. The alpha-track radon monitors were exposed in groups of 6 - 8 to radon concentrations of about 40 pCi/l for three days. The integrated exposures ranged from 99 pCi-d/l to 165 pCi-d/l, with one low concentration test at 26 pCi-d/l. Two unexposed controls typically were returned to the manufacturers for processing along with the exposed devices. The reported radon concentrations are the net radon concentrations determined by subtracting the integrated exposures of the unexposed controls from the exposed monitors and dividing by the length of time in the chamber.

The manufacturers changed detector material batches over the course of the 14-month duration of the test series. Only one detector material batch of each type of monitor was in use during any one exposure. Track Etch® type SF, Track Etch® type F, and Radtrak® monitors were exposed throughout the entire study; AT-100® monitors were exposed for only six tests.

The radon concentrations in the chamber were measured by an Eberline RGM-2 continuous radon monitor with a readout of hourly average concentrations. Twice daily the calibration of this continuous monitor was checked with grab samples taken in 1/2-liter scintillation cells whose individual calibration constants are traceable to NBS radium solutions. The uncertainty in the average chamber radon concentration is ±3%.

None of the four types of alpha-track radon monitors showed any variations which could be correlated with changes in temperature or relative humidity over the ranges of 7°C to 45°C and 20% to 80%, respectively.

The results are presented in the form of frequency distributions of the ratio of the observed response of the alpha track monitors to the known exposure in the chamber. Some of the data sets appeared to fit normal probability plots quite nicely while other data sets appeared to be log normally distributed.

The frequency distribution and theoretical coefficient of variation for an absolutely perfect set of alpha-track detectors would depend only on the Poisson statistics associated with the purely random distribution of tracks on the detector. This theoretical coefficient of variation can be estimated from the net number of tracks recorded by each detector based on the simple 1/n relationship. At a typical exposure of 120 pCi-d/l, the Terradex Type SF monitors will record about 60 gross tracks for the area scanned, giving a theoretical coefficient of variation of 19% if Poisson statistics represents the only source of error. The Type F monitor will record about 150 gross tracks for the same exposure, yielding a theoretical coefficient of variation of 10%; the Radtrak® monitor would record roughly 80 gross tracks for a coefficient of variation of 18%; and the AT-100® monitor would record only about 25 gross tracks, for a coefficient of variation of 24%. These coefficients of variation represent the best that theoretically can be expected based on counting statistics. Any larger coefficient of variation than these theoretical minimum coefficients must be due to other factors.

The Technical Measurements Center (TMC) has adopted a 25% coefficient of variation as an acceptable figure for providing reasonable assurance that an annual average radon concentration measurement will be estimated by a monitor within ±50% of the actual value 95 percent of the time. This criterion was established several years ago by a committee with representatives from several agencies, and the approved monitoring techniques have been able to meet this criterion over the years. TMC tends to judge the performance of alpha-track monitors against this 25% coefficient of variation criterion.

Table 1 summarizes the results of exposures of the various types of alpha-track radon monitors. Detectors from the same detector material, as defined by the manufacturer, are grouped for comparison in Table 1. The expected coefficient of variation is calculated from the mean number of tracks for each type and batch of monitors. The mean ratio of reported radon to actual radon is the mean of the net radon concentration reported by each individual monitor divided by the actual chamber concentration.

Figures 1 - 7 are frequency distributions with normal curves for a number of monitor types and batches. The horizontal scale is the ratio of net reported to actual radon concentration while the vertical scale is the number of monitors reporting a given radon ratio. Figures 1 and 2 show the distribution for four batches of Terradex Track Etch® Type SF monitors. One point to notice about these is that of the four batches, only batch 24 comes close to meeting the theoretically expected frequency distribution. There are a number of monitors with high readings in each of batch 12 and 23. These two batches also possess means differing by almost a factor of 2, indicating a large difference in calibration.

Figures 3 and 4 present data regarding the Terradex Track Etch® Type F monitors. The three batches possess similar mean responses, though batches 12 and 23 in this case have narrower distributions. If the single high reading in each of batches 12 and 23 is discarded, the coefficients of variation are reduced to 25% and 21%, respectively. These fall within the TMC's reasonable assurance criterion but still exceed the theoretically expected value by a factor of two.

Figures 5 and 6 give the results of exposures of Glenwood Laboratories Radtrak® monitors. Lot 1107B of detector material comprised some of the first commercially available Radtrak® monitors. It accounts for much of the variability and high reading monitors of all of the Radtrak® monitors. Later batches of material show an improved distribution, with the most recently exposed lot 1205 giving a distribution equivalent to that expected theoretically.

Figure 7 shows the distribution of Radon Environmental Monitoring AT-100® monitors. The AT-100® monitors were available for only six exposures towards the end of the study. As with the Radtrak® monitors, these were some of the first commercially available AT-100® monitors. When only the last four of the six total exposures are considered, these monitors give a distribution equivalent to the theoretically expected distribution.

Several conclusions may be drawn from this data. The alpha-track radon monitor occasionally performs to theoretical expectations, but more frequently shows much greater variation in response than that attributable solely to Poisson counting statistics. Additional work must be done in order to ascertain the sources and magnitudes of the various contributions to the errors observed. These sources may include errors in determination of the "actual" radon, errors introduced by handling, packaging, processing, and reading, errors in characterization of the detector material, or errors in reporting results.

Newer models of alpha-track radon monitors, such as the Radtrak® and Radon Environmental Monitoring devices, perform as well as older models and deserve further evaluation.

Finally, the data highlight the recommendation by TMC that any program employing alpha-track radon monitors should include a regular procedure of exposed and unexposed controls as well as duplicate detectors in order to monitor the quality of the results.

TABLE OF ALPHA-TRACK TYPE RADON MONITORS

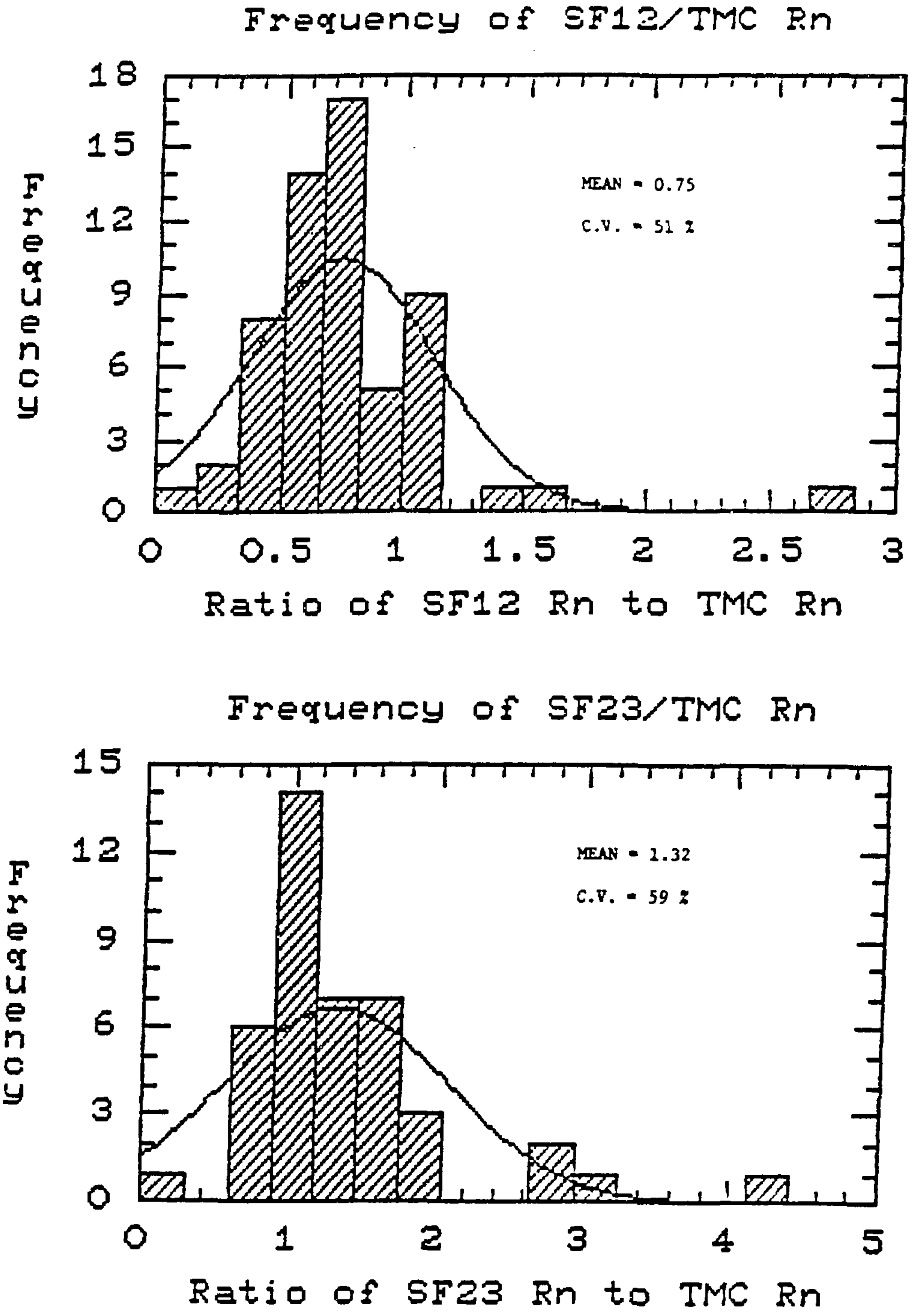
MONITOR TYPE & BATCH	# of Detectors	Mean Reported Radon / Actual Radon	Coefficient of Variation	
			observed	expected
Terradex SF 12	59	0.75	51 %	23 %
Terradex SF 23	42	1.32	59 %	16 %
Terradex SF 24	55	0.96	25 %	17 %
Terradex SF 25	18	0.67	47 %	20 %
Terradex F 12	50	0.81	31 %	11 %
Terradex F 23	33	0.96	36 %	10 %
Terradex F 24	50	0.91	52 %	9 %
Radtrak 1107B	51	0.95	65 %	21 %
Radtrak 1189A	32	1.27	40 %	20 %
Radtrak 1189B	29	1.07	18 %	17 %
Radtrak 1199A	10	0.92	17 %	17 %
Radtrak 1205	24	0.98	13 %	15 %
Radtrak - all batches	143	1.04	45 %	19 %
REM - all batches	46	1.15	36 %	24 %
REM - last 4 tests	30	0.94	24 %	24 %

* Actual radon exposure based on continuous 72-hour integrated average with an uncertainty of ±3%.

* TMC reasonable assurance criterion is a coefficient of variation less than 25%.

* "Expected" coefficient of variation based on the counting statistics of the net number of tracks from a 120 pCi-d/l exposure.

* Monitors were exposed in groups of 6 to 8. Only one batch of each type of monitor was in use during an exposure. Track Etch Type SF, Track Etch Type F, and Radtrak monitors were exposed throughout the entire study; REM AT-100 monitors were exposed for only six tests.


Frequency of SF12/TMC Rn

Frequency of SF23/TMC Rn

Figure 1. Frequency distributions of Terradex Track Etch Type SF monitors - Batch 12 and Batch 23.

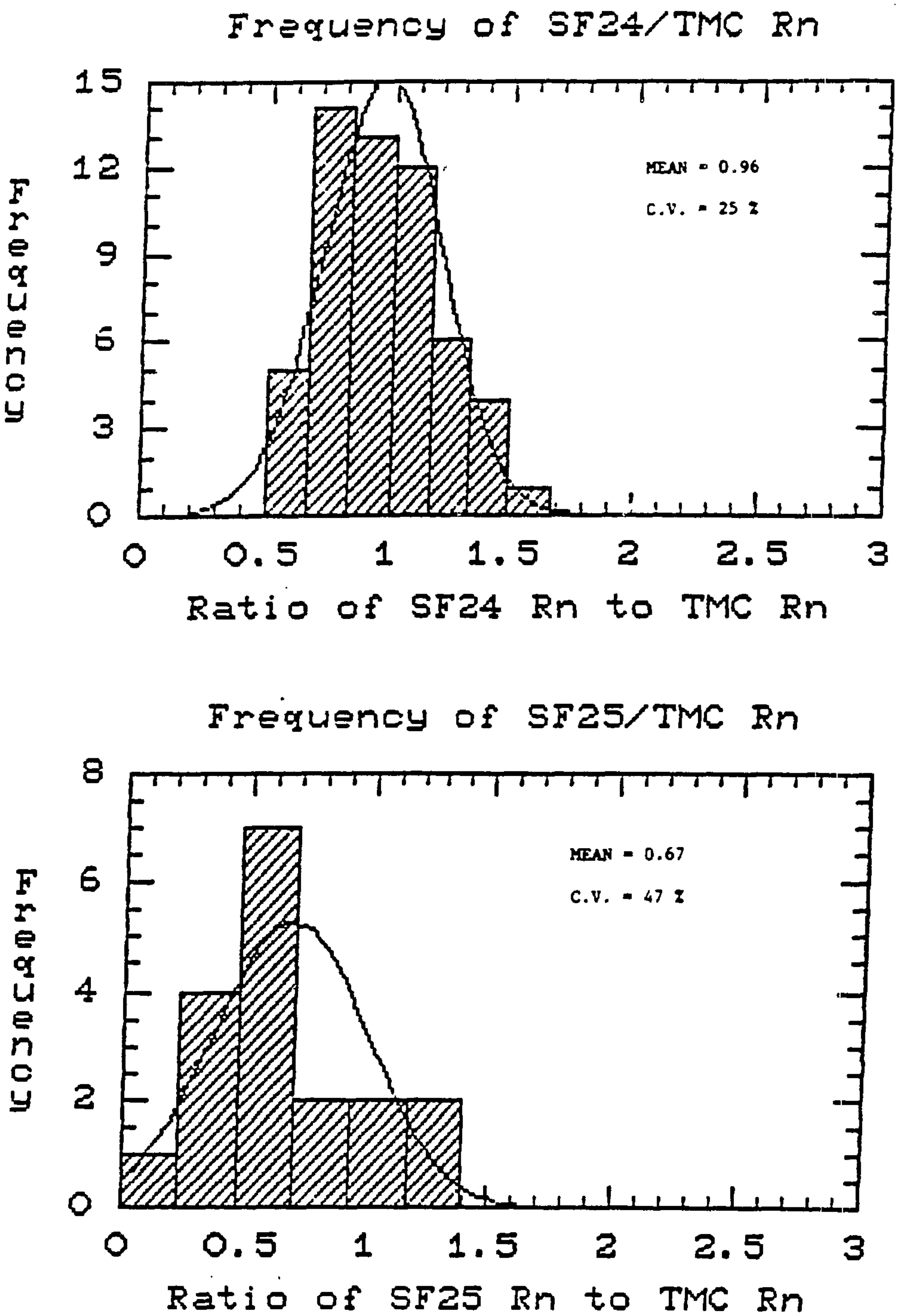


Figure 2. Frequency distributions of Terradex Track Etch Type SF monitors.- Batch 24 and Batch 25.

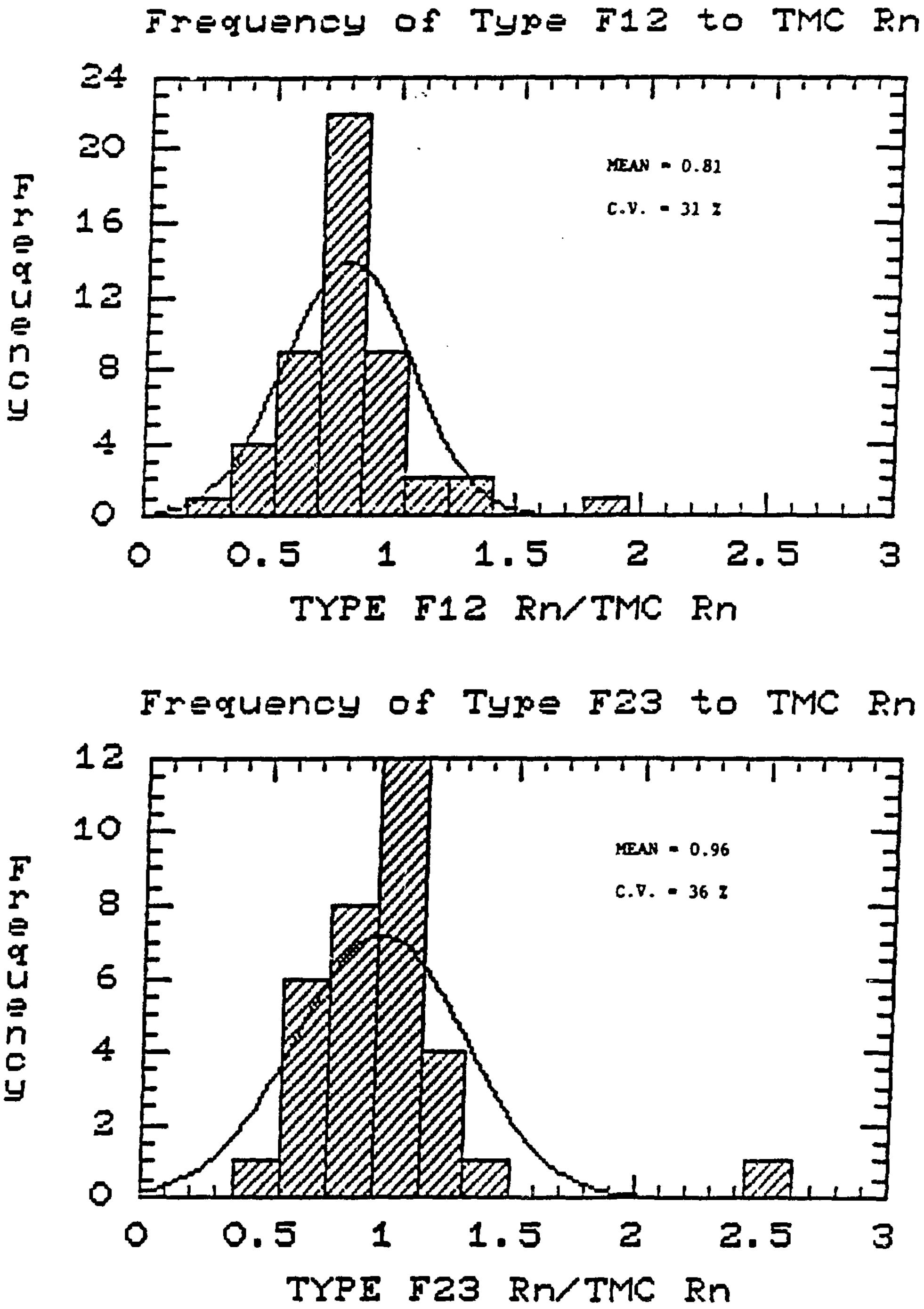


Figure 3. Frequency distributions of Terradex Track Etch Type F monitors - Batch 12 and Batch 23.

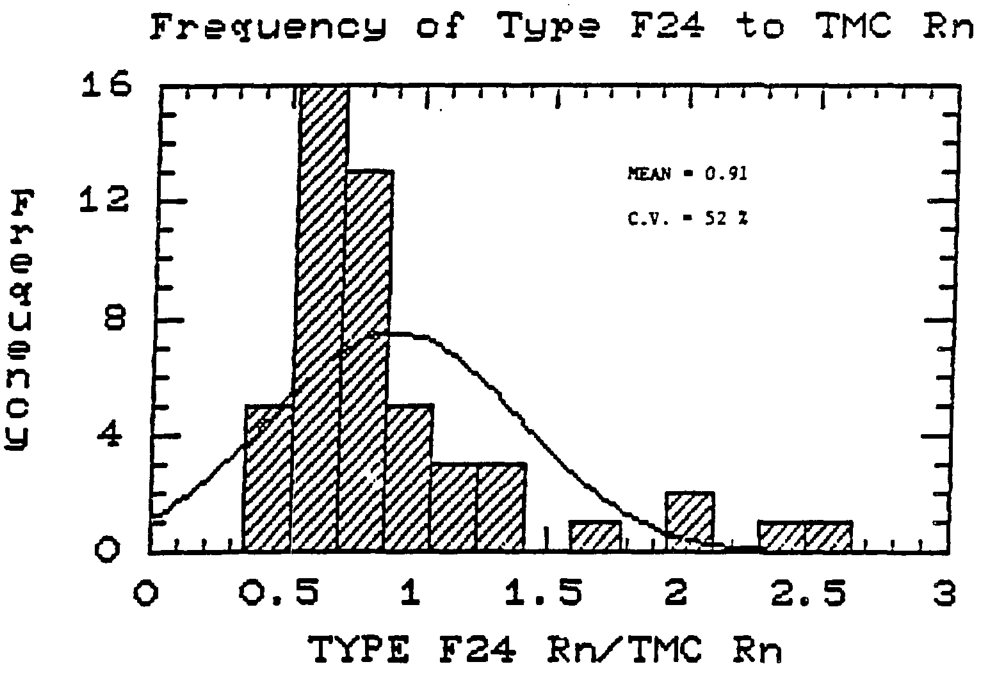
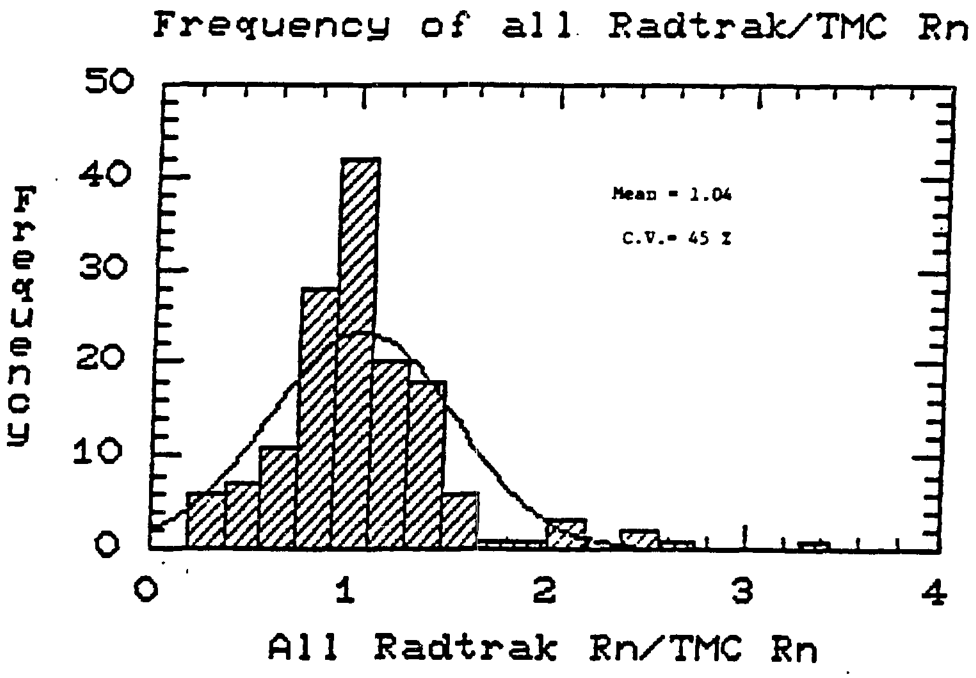


Figure 4. Frequency distribution of Terradex Track Etch Type F monitors - Batch 24.



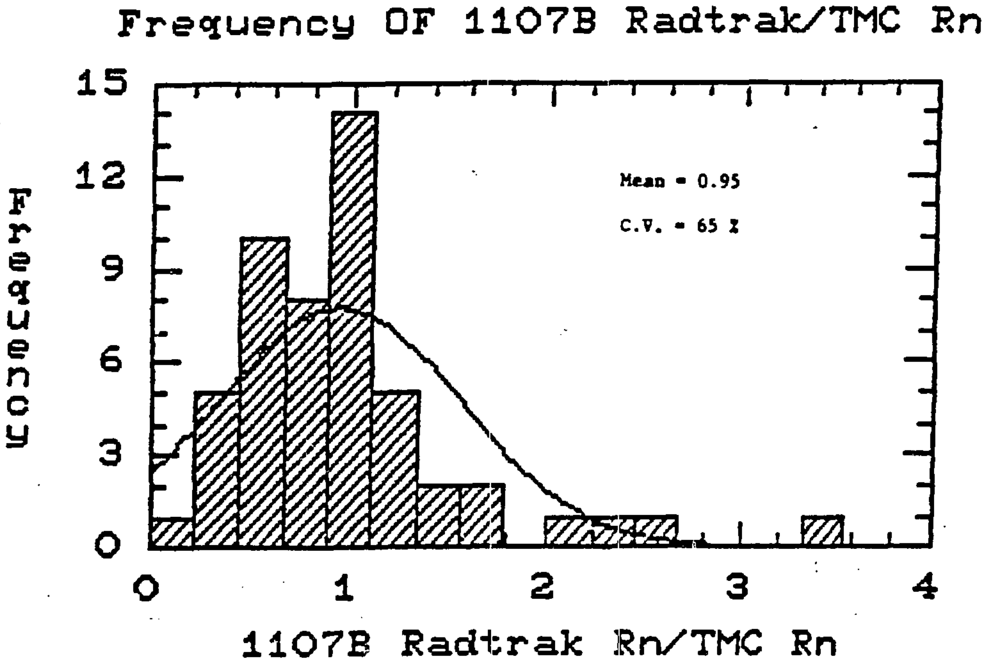


Figure 5. Frequency distributions of Glenwood Laboratories Radtrak monitors - all lots, and lot 1107B.

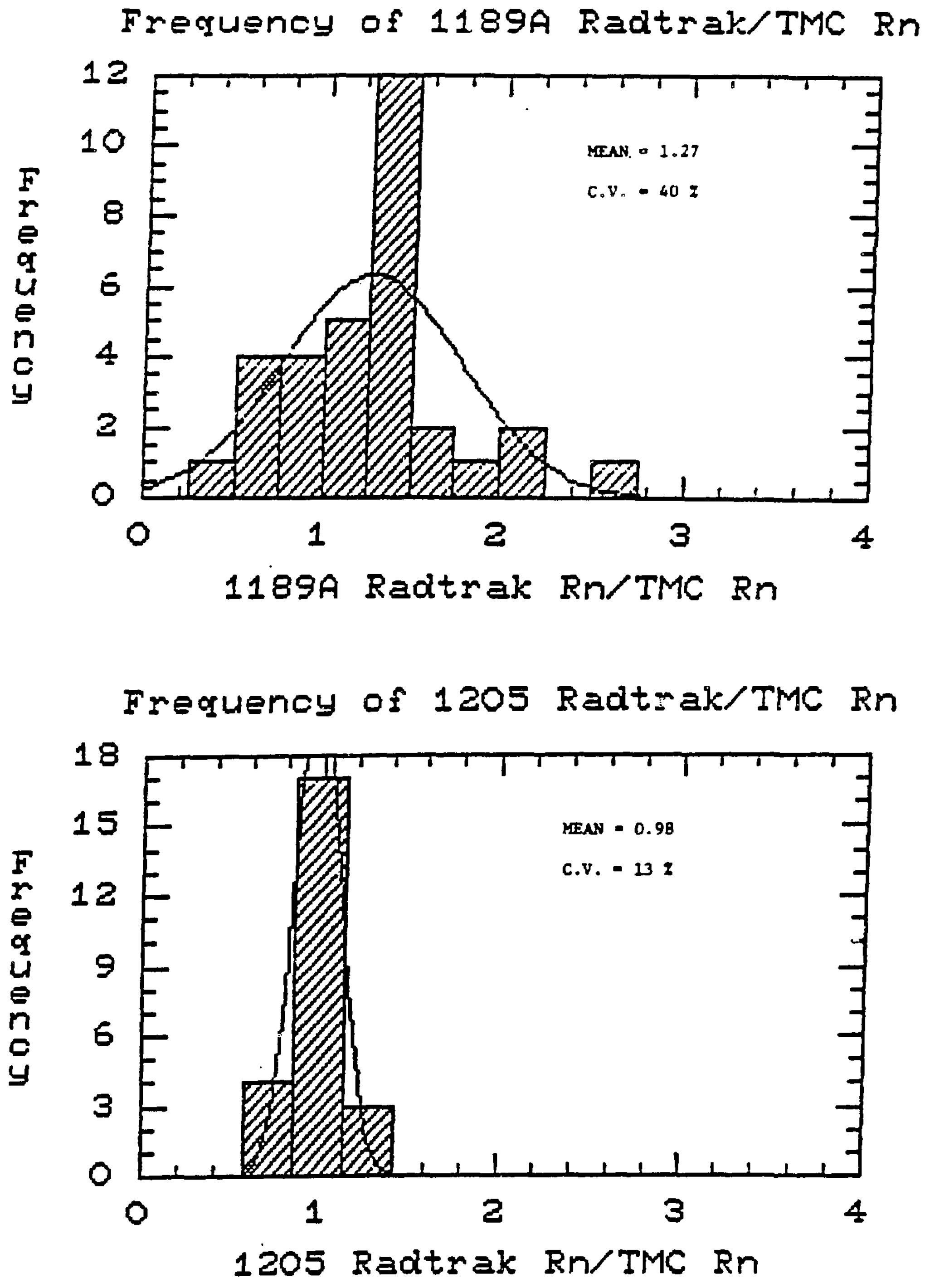


Figure 6. Frequency distributions of Glenwood Laboratories Radtrak monitors - lot 1189A and 1205.

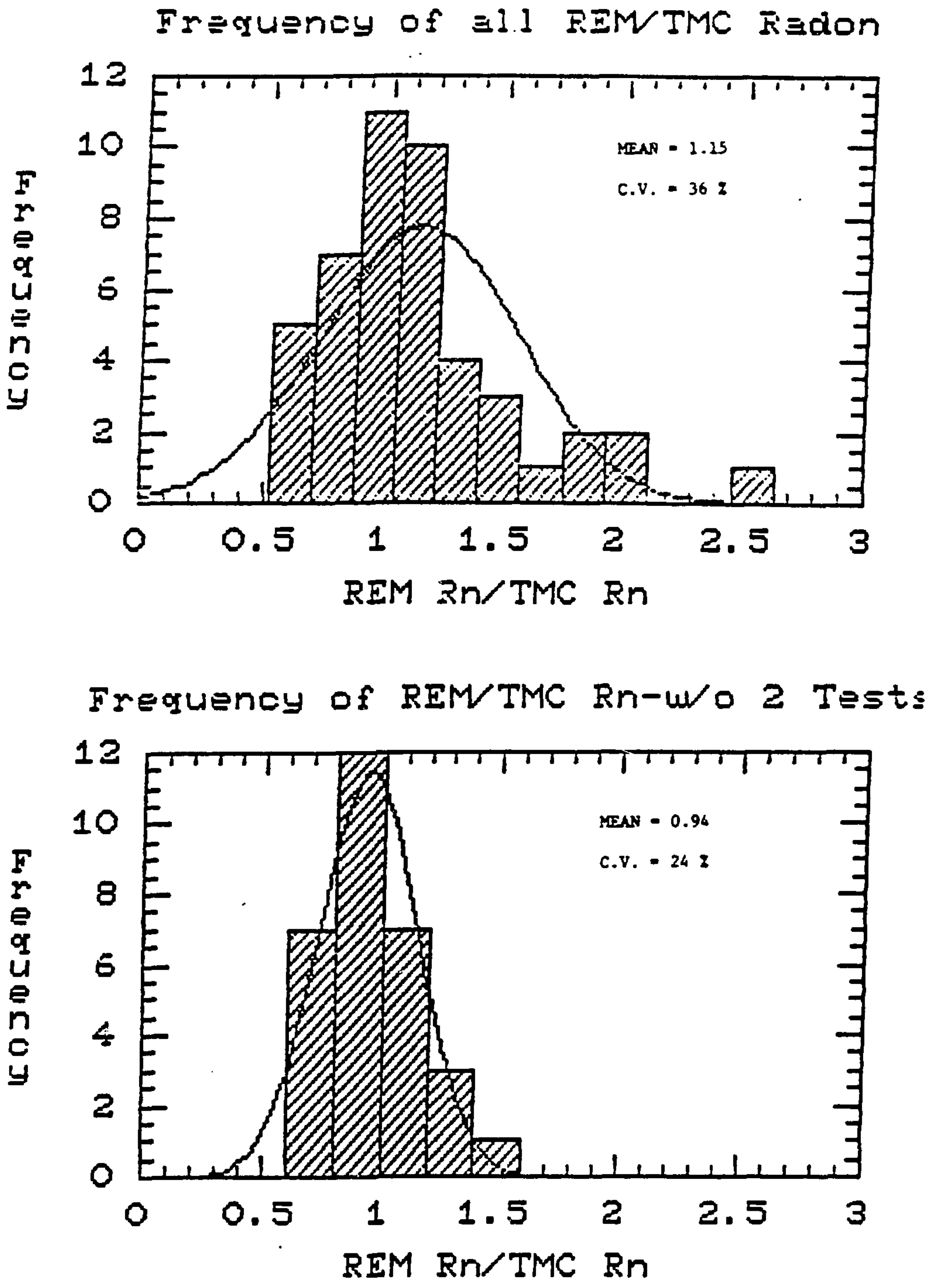


Figure 7. Frequency distributions of Radon Environmental Monitoring AT-100 monitors - all monitors and monitors from last 4 of 6 tests.

SECTION M

Comparison of Three Month Screening Measurements with Year-Long Measurements Using Track Etch® Detectors in the Reading Prong

Carl Granlund and Michelle Kaufman,
Pennsylvania Department of Environmental Resources

COMPARISON OF THREE MONTH SCREENING MEASUREMENTS WITH YEARLONG MEASUREMENTS USING TRACK ETCH DETECTORS IN THE READING PRONG.

By Carl Granlund and Michelle Kaufman *

Introduction

A large amount of data has been collected as part of the Bureau's Reading Prong Radon Screening Program. To date, close to 18,000 screening measurements have been made. As part of this program, yearlong follow-up measurements were made in a sizeable fraction of those homes that has screening levels above 4 pCi/l.

With yearlong measurement data becoming available early this summer (1987), the Bureau was interested in looking at four areas including:

1. The relationship between yearlong follow-up measurements in living areas and basement screening measurements using Track Etch detectors.
2. The number of people taking remedial action after a screening measurement and what technique was used.
3. The effect of remedial action on yearlong averages.
4. The possibility of using a three month basement screening measurement to estimate a yearlong first floor average.

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*Department of Environmental Resources, Bureau of Radiation Protection,
Harrisburg, PA 17120

BACKGROUND

Early in 1985 it was recognized that the house-to-house radon surveys being done in the Reading Prong by the Bureau would take years to complete. So, in October 1985, a mail campaign was organized to screen every home in the Reading Prong. Boundaries were drawn up encompassing the Reading Prong. Those homes within the boundaries were eligible for a free radon monitor. This monitor was a Terradex Type SF Track Etch Radon Detector.

The detectors were sent directly to the homeowner. Instructions asked the homeowner to, "post the detector in the lowest potential living space in the house, the basement if you have one." The instructions also directed the homeowner to, "expose the detector for at least three months."

The Bureau reported the screening results to the individual homeowner. Individual reports to homeowners were based on the screening result. If the screening level was less than 4 pCi/l the Bureau recommended that no further action was anticipated or suggested.

If the screening level was greater than 4 pCi/l, but less than 20 pCi/l the Bureau recommended further long term testing. This follow-up testing was to be in a area of the house where the family members spent most of their in-house time. A Track Etch detector was sent by the Bureau directly to the homeowner. The homeowner was directed to expose this detector for a period of one year.

If the screening level was above 20 pCi/l a house survey by one of the Bureau's radon survey teams was arranged. During the course of the survey a Track Etch detector was placed. A summary of the Bureau's recommendations based on screening levels is given below.

RESPONSE TO TRACK-ETCH SCREENING RESULT

RESULT =< 4.0 pCi/l	No further action anticipated
RESULT 4.1 to 20 pCi/l	Homeowner sent a second Track Etch detector and directed to place it for a period of one year in an area of the house where he spends the majority of his time.
RESULT > 20 pCi/l	House survey by one of the Bureau's survey teams. Survey included basement RDC measurement, placement of a charcoal canister in basement and first floor levels, obtain a water sample if home is supplied by a well, and placement of a Track Etch detector.

The Bureau also notified the individual homeowners of their follow-up radon measurements. There were two reporting forms. One was for those with an annual average greater than 4 pCi/l. This reporting form recommended the homeowner to undertake remedial action to reduce his exposure. No specific time frame for remedial action was mentioned, but it stated that the higher the radon level, the sooner the homeowner should take action.

The second report, for annual values less than 4 pCi/l stated that this type of measurement was a better estimate of what their true average annual exposure was and that no further action was anticipated.

Listed below are radon measurement summaries for both screening measurements and follow-up measurements. As can be seen, 60.9% had screening levels above 4 pCi/l and 12.2% had levels above 20 pCi/l. For the yearlong measurements the fraction of homes above 4 pCi/l fell to 21.4% and only 1.4% had yearlong measurements above 20 pCi/l.

PENNSYLVANIA READING PRONG MAIL PROGRAM
TRACK ETCH MEASUREMENT DATA

1. Three month screening results

Range (pCi/l)	#	Percentage	
<4	6681	39.0	
4.1-10	5688	33.2	* 60.9 > 4 pCi/l
10.1-20	2653	15.5	
20.1-100	1845	10.8	* 12.2 > 20 pCi/l
100.1-200	153	0.9	
>200	91	0.5	
	17111		

2. Yearlong follow-up measurement results

Range (pCi/l)	#	Percentage	
<4	1659	46.4	
4.1-10	515	14.4	* 21.4 > 4 pCi/l
10.1-20	199	5.6	
20.1-100	28	0.8	* 1.4 > 20 pCi/l
100.1-200	13	0.4	
>200	6	0.2	
	2420		

SAMPLE POPULATION

From the database of homeowners who had both a three month screening result and a yearlong measurement, a subgroup of 1500 homeowners was chosen. Each member of the study group was sent a questionnaire. The questionnaire asked the homeowner to indicate where the screening measurement and the follow-up measurement were made, if and when remediation work had been done, and what type of remediation was taken.

Responses from approximately 1100 homeowners were received. The questionnaire revealed that 13.6% had taken some sort of remedial action. The table below divides those who took remedial action after their screening measurements and those who did not take any action and the radon distribution for each group. The group with higher screening levels took remediation. For example, those who took remedial action, 20.3% had levels above 50 pCi/l, while of those who did not take remedial action, only 7.4% had levels above 50 pCi/l.

Screening measurement results

GROUP	10-20	21-50	51-199	>200	(pCi/l)
Remediation	36.4%	28.0%	16.1%	4.4%	
No Remediation	58.9%	29.2	7.0	0.4	

For the group that took remedial action, the techniques varied widely. The remedial techniques ranged from increased natural ventilation to subslab ventilation systems.

The questionnaire was not specific in determining the exact nature of increased ventilation or sealing. The table below lists each technique reported.

Remedial Techniques

#	Percentage	Remedial method
12	8.4	subslab ventilation (active and passive)
54	37.8	increased ventilation only
45	31.5	sealing only
19	13.3	sealing and increased ventilation
5	3.5	sump pump vents (active and passive)
4	2.8	air-to-air heat exchangers
2	1.4	area air filters
2	1.4	whole house electronic air filters

The vast majority of screening measurements were begun in November 1985. This corresponds to the start of the Mail Survey Program. The median exposure time for the screening measurement was 88 days. The median exposure period for the annual measurement was 365 days.

Both screening measurements and yearlong follow-up measurements showed approximately log normal distributions. The distribution for screening measurements in homes with no remediation can be seen in figure 1.

ANNUAL/SCREENING RATIOS

A. No remediation group.

To determine the relationship between the screening measurement and the yearlong measurement, the natural logarithm of (yearlong measurement divided by the three month measurement) was found. This distribution is approximately normal with a (log) average of -1.082, which corresponds an average of 0.34. This indicates, that on the average, a wintertime basement screening measurement overestimates the first floor annual average by a factor of three.

Figure 2 shows the distribution for those homes whose three month screening measurement was in the basement and whose yearlong measurement was taken on the first floor. No remediation was taken by this group.

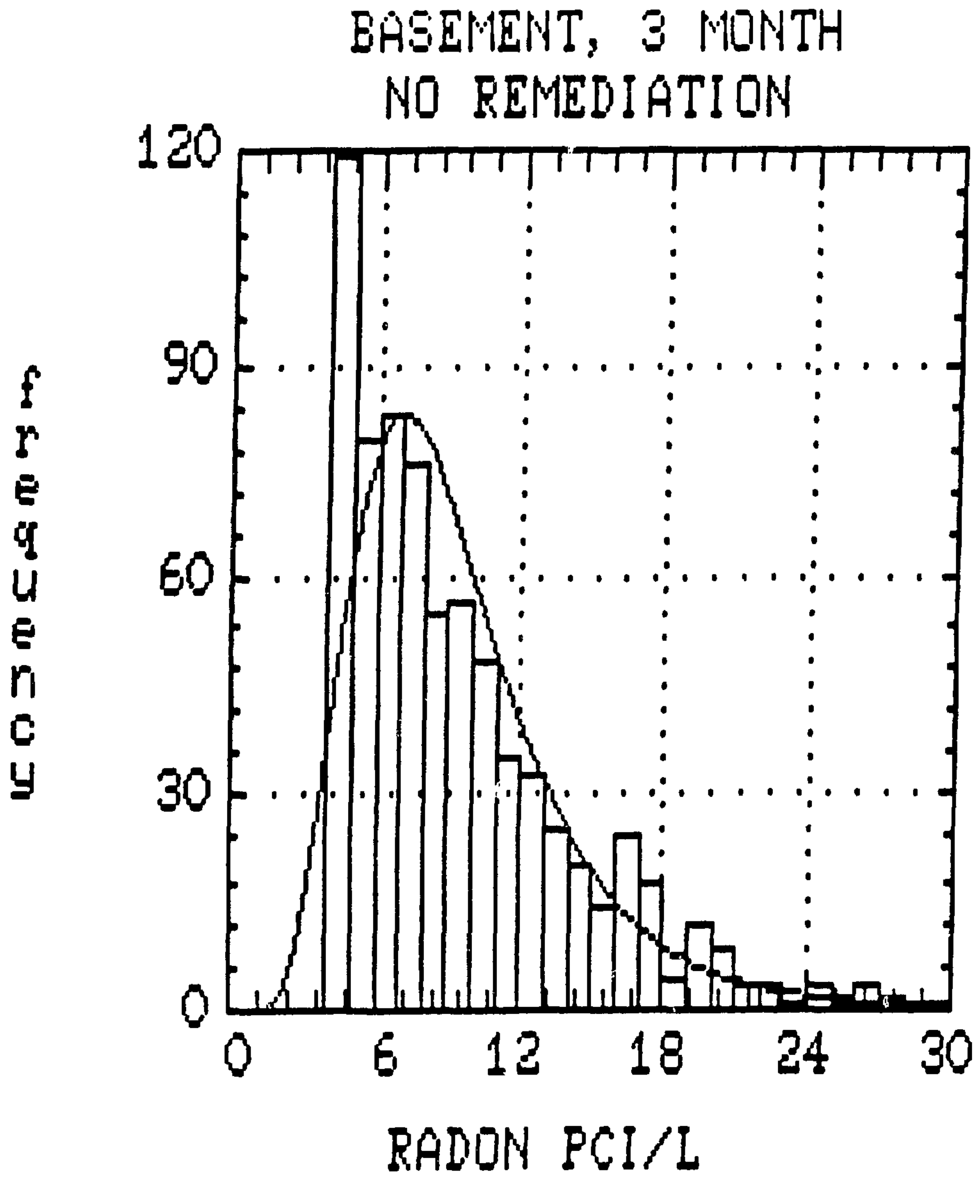
There was a small group of homeowners who made both screening and yearlong measurements in the basement (n=45). The average ratio, as found by the above method, was 0.73. This shows that a three month wintertime basement measurement overestimates a yearlong basement measurement by about 1.4 times.

A third group made both measurements on the first floor (n=54). The average ratio was 0.50. This shows that a three month wintertime first floor measurement overestimates an annual first floor measurement by a factor of 2.

B. Remedial action group.

To determine the effect of remedial action four groups were formed: 1. increased ventilation only, 2. sealing only, 3. increased ventilation and sealing, and 4. active systems which included subslab ventilation, air filtration, sump pit venting, and air-to-air heat exchangers.

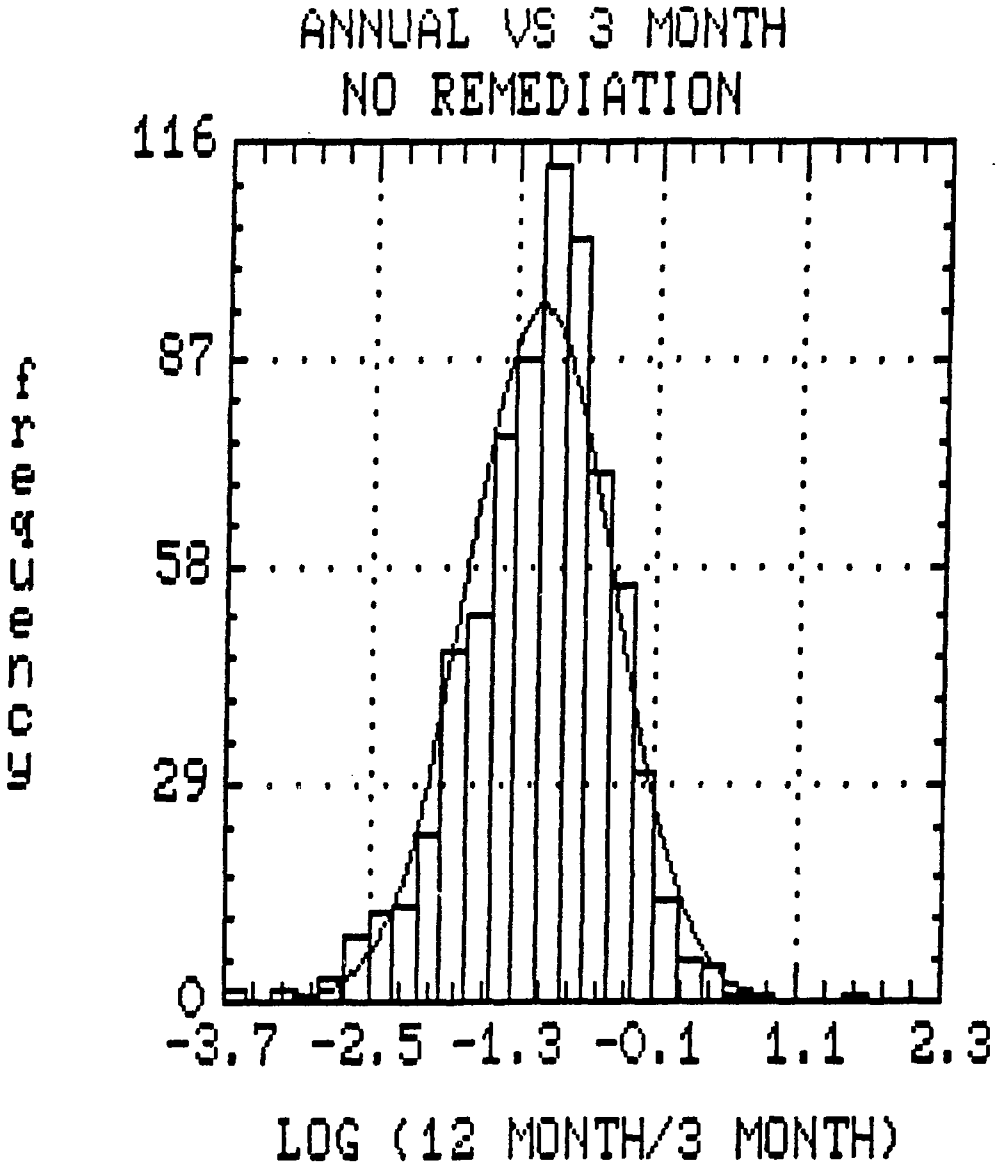
For the remedial group that increased ventilation only, the average ratio (annual 1st floor/basement screening) was 0.32. This is very close to the no remediation group average of 0.34. Increased ventilation did not produce a significant reduction as compared to the no remediation group.



BASEMENT, 3 MONTH
NO REMEDIATION

NUMBER OF OBSERVATIONS = 728
SAMPLE AVERAGE = 9.50165
SAMPLE VARIANCE = 21.4138
SAMPLE STANDARD DEVIATION = 4.62751
MEDIAN = 8.2

Figure 1



NUMBER OF OBSERVATIONS = 728
SAMPLE AVERAGE = -1.0823
SAMPLE VARIANCE = 0.38309
SAMPLE STANDARD DEVIATION = 0.61894
MEDIAN = -1.0453

Figure 2

For the remedial group that did sealing only, the average ratio was 0.29. There is no difference from the no remediation group at the 5% level of significance.

For the remedial group that increased ventilation and did sealing the average ratio was 0.23. This group showed a significant reduction from the no remediation group, although the sample size was small (n=17).

The remedial group that employed active systems had an average ratio of 0.14. This group showed a significant reduction from the no remediation group.

The following table shows the average ratios for both the remedial group and the no remediation group.

Average follow-up/screening ratios

A. No remediation group.

Screening Loc.	Follow-up Loc.	#	Ratio*
1. Basement	First floor	728	0.34
2. Basement	Basement	45	0.73
3. First floor	First floor	54	0.50

B. Remedial group

Technique	#	Ratio*
1. Increased ventilation	45	0.32
2. Sealing only	40	0.29
3. Sealing and ventilation	17	0.23
4. Active systems	21	0.14

* ratio of (yearlong follow-up/ 3 month screening measurement)

ESTIMATING YEARLONG AVERAGES

The Bureau's recommendations to homeowners who have screening results greater than 4 pCi/l, but less than 20 pCi/l is to perform a long term follow-up measurement. It was felt that the data set could give an indication if the above recommendation is realistic. For example, would a basement screening level of 20 pCi/l yield a first floor annual average greater than 4 pCi/l in a large fraction of homes.

The probability that an annual first floor average will exceed a given value (4 pCi/l) can be obtained as a function of the (log) ratio of annual/screening values. This ratio yields a normal distribution. Using the distribution mean and standard deviation the probability that an annual average will exceed 4 pCi/l, as a function of a specified screening value can be found.

Figure 3 predicts that if the wintertime basement screening result is 11.8 pCi/l, there is a 50% chance that the yearlong first floor measurement will exceed 4 pCi/l. The solid black line is the theoretical curve and the hatched bars are the actual data set. The theoretical curve tends to predict the actual values very closely.

A second area the Bureau wanted to investigate was increasing the screening level. This would be possible if the theory that three month basement screening results were overestimating first floor annual averages by a wide margin was true. Judging from the data, this theory does not appear to be true. For example, raising the action guideline for a basement screening measurement to 8 pCi/l, still leaves 26% of the homes with an annual first floor average greater than 4 pCi/l.

CONCLUSIONS

- The radon problem in the Reading Prong is dramatic. For example: (from screening measurements)

  60.9% >4 pCi/l
  12.2% >20 pCi/l
  1.4% >100 pCi/l
  0.5% >200 pCi/l

- The follow-up measurement data shows a sizable fraction of the population with annual averages greater than 4 pCi/l. For example: (yearlong measurements)

  21.4% >4 pCi/l
  1.4% >20 pCi/l
  0.6% >100 pCi/l
  0.2% >200 pCi/l

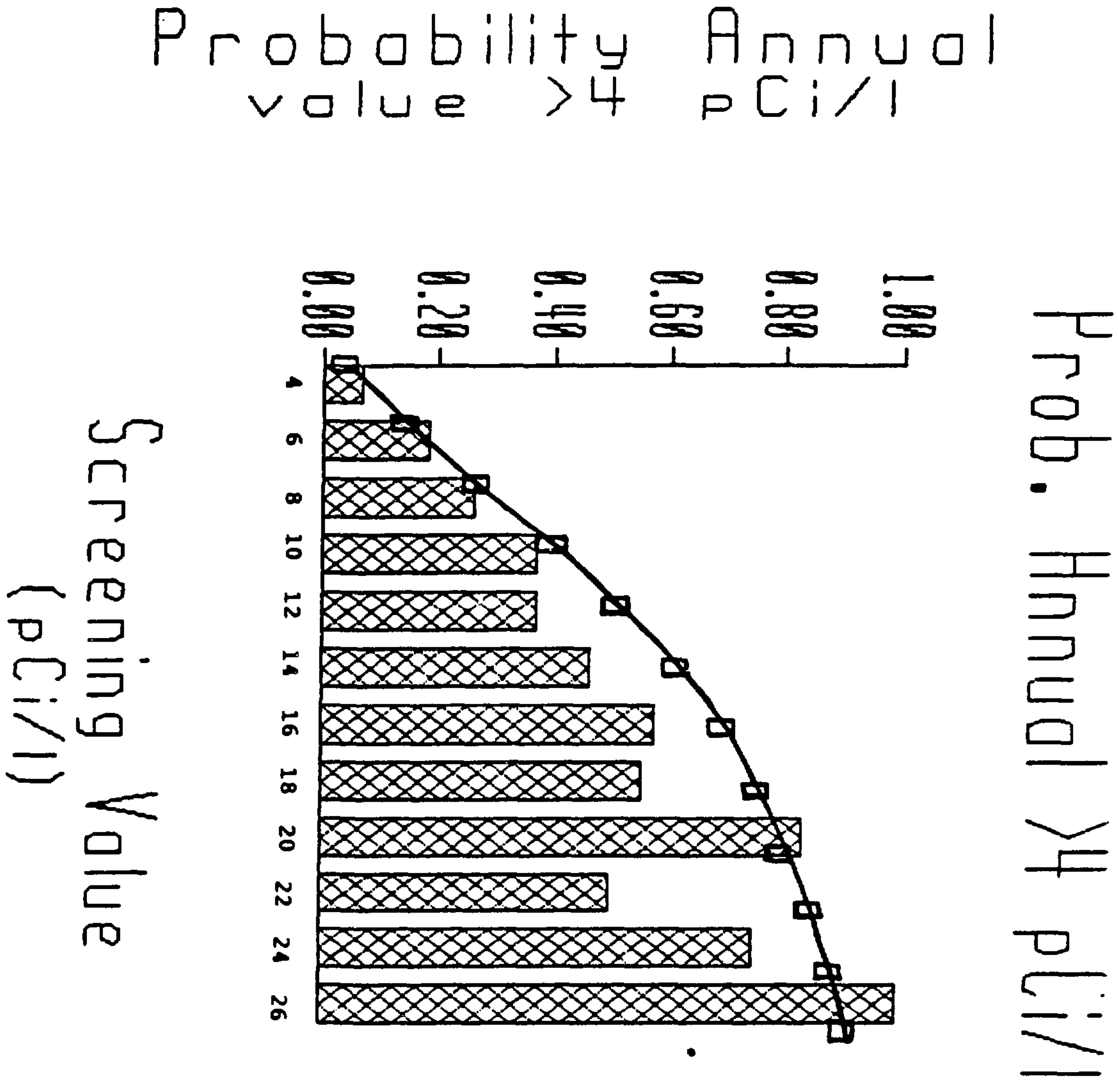

Probability Annual
value >4 pCi/l
Prob. Annual >4 pCi/l
0.00
0.20
0.40
0.60
0.80
1.00
4
6
8
10
12
14
16
18
20
22
24
26
Screening Value
(pCi/l)

Figure 3

- There was a sizable population that took remedial action after a single screening measurement (13.6%). The people who took remedial action were those with higher radon levels. The most common technique used was increased ventilation.

- Of those who took remediation, only those who performed sealing and increased ventilation or installed an active system showed a significant reduction in their annual averages, as compared to the no remediation group.

- On average, the probability of an annual first floor measurement being greater than 4 pCi/l when the basement screening result is 11.8 pCi/l, is 50%.

SECTION N

Comparison of Landauer Radtrak and Terradex Type SF Track Etch® Detectors for Radon Measurements in Residences

D. C. Landguth and Mary W. Smuin,
Oak Ridge National Laboratory, Oak Ridge, TN

COMPARISON OF LANDAUER RADTRAK AND TERRADEX TYPE SF TRACK ETCH^{R} DETECTORS FOR RADON MEASUREMENTS IN RESIDENCES*

D. C. Landguth and Mary W. Smuin**
Health and Safety Research Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831

ABSTRACT

Oak Ridge National Laboratory (ORNL) is the Inclusion Survey Contractor (ISC) for the Uranium Mill Tailings Remedial Action Project (UMTRA). As part of these surveys ORNL performs radon monitoring on vicinity properties involved in the UMTRA Project to determine if these properties exceed Environmental Protection Agency (EPA) standards for radon daughter concentration. A recommendation as to inclusion in or exclusion from the UMTRA Project is passed on to the DOE for a decision.

ORNL conducted radon monitoring on vicinity properties for one year using triplicates of both Terradex Type SF Track Etch^{R} and Landauer Radtrak detectors. Currently, Terradex Type SF Track Etch^{R} detectors are the only passive monitor approved for use by the DOE in the UMTRA Program. The purpose of this study was to determine if a significant correlation existed between the two types of radon monitoring devices. A linear regression was performed on the calculated working level results from both manufacturers on the vicinity properties and a regression coefficient of 0.95 was determined.

The conclusion of this study proposes that a more flexible, multi-vendor system be used where various detectors may be interchanged for use in the UMTRA Project.

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*Research performed by members of the Environmental Assessments Group of the Health and Safety Research Division at Oak Ridge National Laboratory under U.S. DOE contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

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SECTION O

Considerations in Assessing the Performance of Alpha Track Detectors

Richard A. Oswald, Terradex Corporation

CONSIDERATIONS IN ASSESSING THE PERFORMANCE OF ALPHA TRACK DETECTORS

Richard A. Oswald
Terradex Corporation

Editor's Note: At the Technical Exchange Meeting the author discussed the transfer of alpha track operations from Terradex to Landauer and the types of detectors available from the merged companies. Subsequently the author provided additional material not available at the time of the meeting related to topics presented by other authors and discussed extensively by the panel. This material is appended to this proceedings at the author's request.

Considerations in Assessing the Performance of Alpha Track Detectors

by

Richard A. Oswald

One component of variance in the use of alpha track detectors is the random fluctuation of track counts on detectors that are given the same radon exposure. This is frequently used as a basis against which to compare the performance of various detectors.

The ideal distribution of alpha track detector readings would simply be the Poisson distributed number of counts. Of course, a variety of other potential sources of variation do exist. These include electrostatic charge effects, etching conditions, variation of CR-39 sensitivity within a sheet, variations in background tracks, variations in counting equipment (including human) sensitivity, etc. The overall spread in observed results can be compared to the baseline Poisson distribution in order to get an idea of the relative significance of these other factors.

Since the U.S. Environmental Protection Agency has recently been performing proficiency testing of various radon detectors, a data base of results has accumulated. This data base represents a realistic opportunity to perform a retrospective analysis of alpha track results because it includes the complete process of manufacturing, packaging, delivering, exposing, returning, etching, counting and report generation. This opportunity is unique in that this process is tested over a protracted period of time and the detectors are handled by various users, as they normally would be. Another important feature of the EPA study is that many detectors of various kinds are exposed together, thereby providing for multiple cross-checking of the correctness of the delivered exposure.

The observed distributions of detector readings for both Radtrak and Terradex Type SF radon detectors in EPA Proficiency Round 4 are shown in Figures 1 and 2 respectively. The results have all been normalized by dividing the Terradex reported exposure by the EPA's delivered exposures. The bias and standard deviation of the distributions are 0.95 and 14.2% for Radtrak and 0.91 and 15.6% for Terradex Type SF. These results are in good agreement with the ideal values of 1.00 and 14.7% for 120 (pCi/l)-days. How the expected values are derived is described in the following section.

Statistical Errors

If we count N tracks on an exposed detector, these counts ideally follow a Poisson distribution. However, background considerations must also be taken into account when assessing the expected performance of alpha track detectors. Sheets of CR-39 have a intrinsic background of alpha particle tracks when received from the manufacturer. The process of fabricating and packaging alpha track detectors introduces further alpha particle tracks onto the detectors from environmental radon. This "in-house" background is subtracted from each detector we analyze. The fluctuation in this background is a source of random error that is always present and contributes to the observed variance in readings.

Another component of variance is introduced when the reading of an unexposed control detector is subtracted from an exposed detector. The purpose of such an unexposed control is to account for any radon exposure imparted to a detector in handling it after the completion of its intended exposure. Such control detectors can have a measurable radon exposure. The variance of the reported reading of such a detector is contributed to by both the random fluctuation of counts associated with the radon exposure it received and the fluctuation of the in-house background.

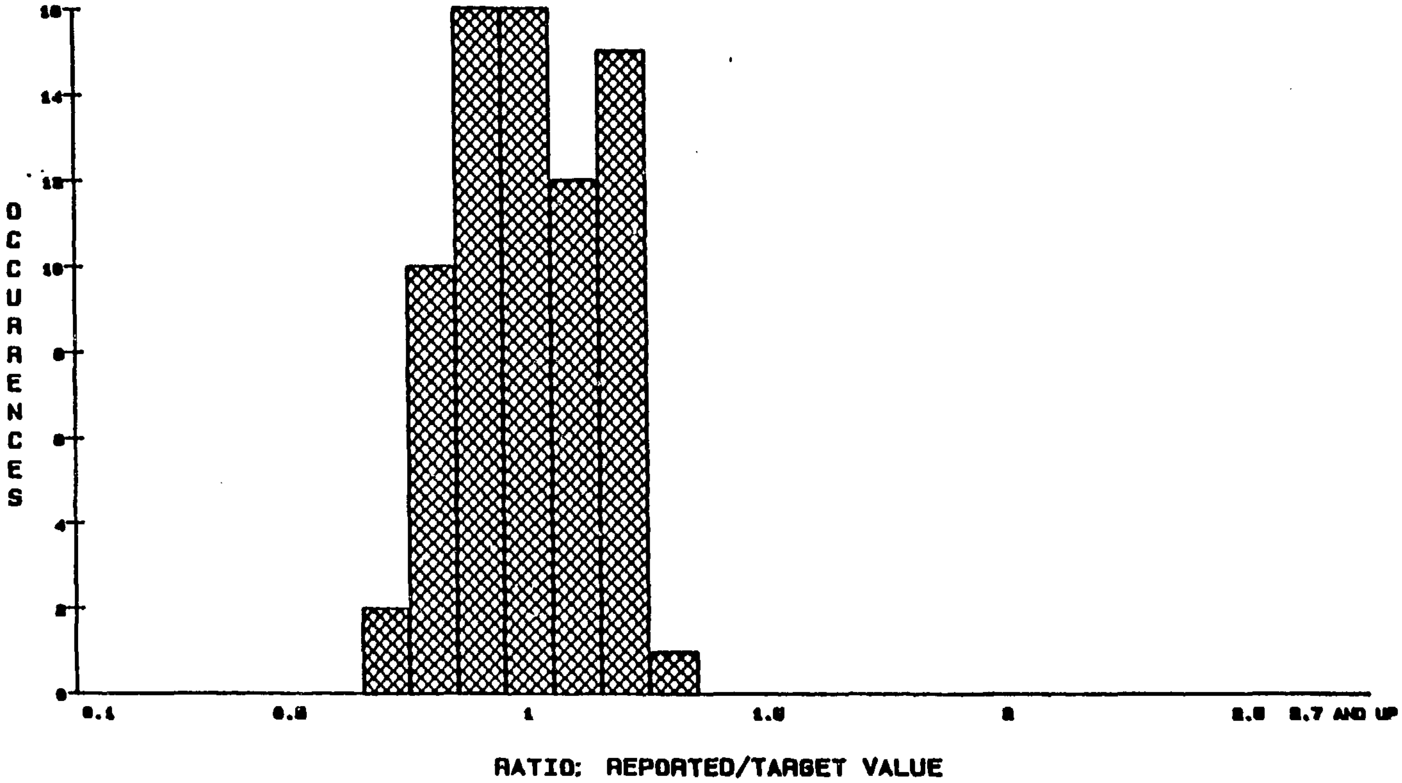
Finally, when this unexposed control reading is subtracted from the exposed detector reading, the uncertainties in each of the two detector readings combine quadratically to yield the final uncertainty in the background corrected result. Taking this into account, the table below shows expected uncertainties for various exposures for Radtrak detectors using our standard counting area. This table can also be used to compute the uncertainty in the difference between two readings using standard error propagation equations.

Radon Exposure (pCi/l)-days	Expected Uncertainty (Percent)
360	7.8%
180	11.5%
120	14.7%
60	23.1%

Conclusions

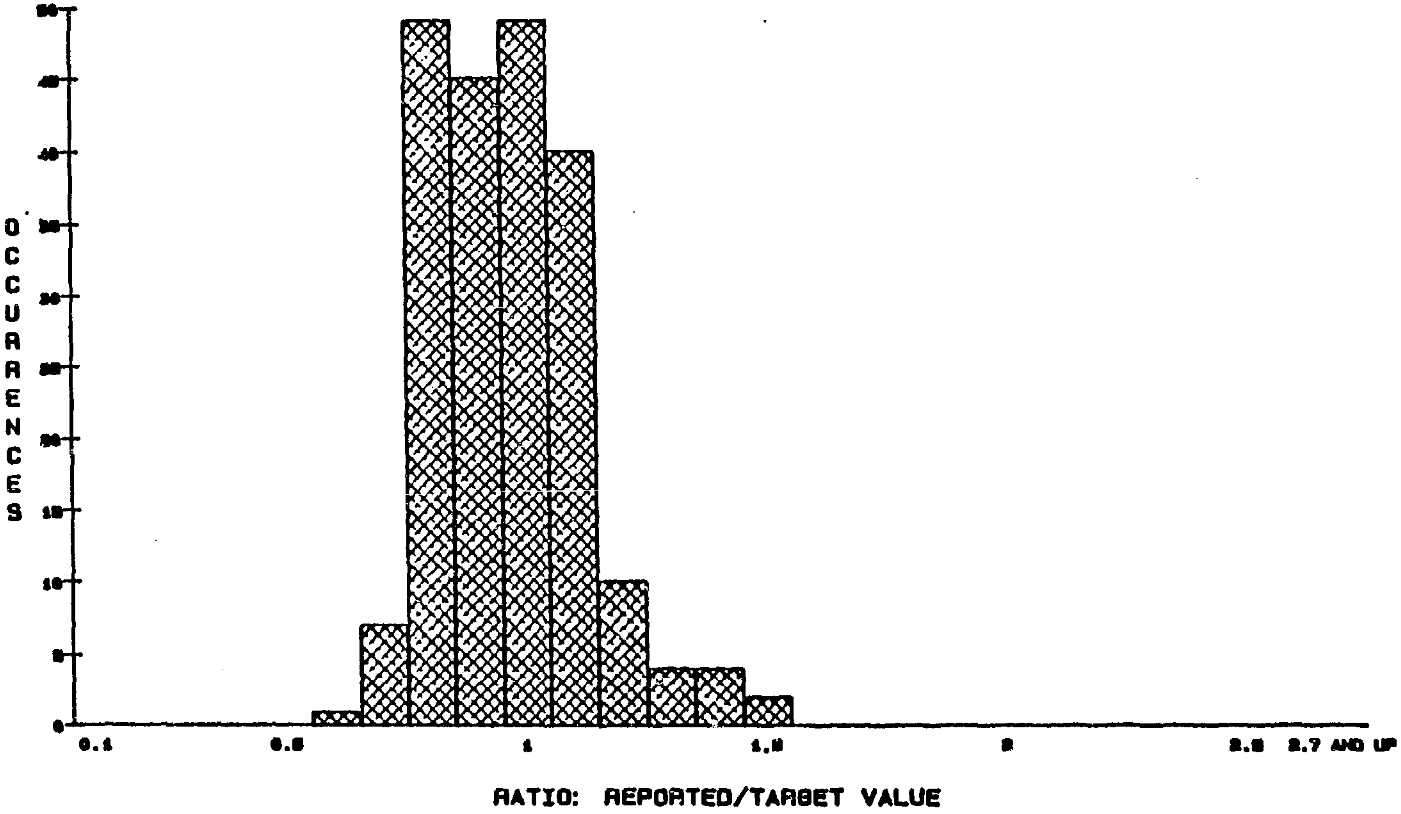
The coefficients of variation we observe in the EPA proficiency data for both Radtrak and Terradex Type SF detectors are in good agreement with the expected values tabulated above. We thus conclude that, for all practical purposes, the other potential sources of error are adequately eliminated by quality assurance procedures.

DISTRIBUTION OF RADTRACK MEASUREMENTS
FROM ROUND 4 OF THE EPA RMP



MEAN = 0.95
C.V. = 14.2%

DISTRIBUTION OF TRACK ETCH TYPE SF MEASUREMENTS
FROM ROUND 4 OF THE EPA RMP



MEAN = 0.91
C.V. = 15.6

SECTION P

Agenda

AGENDA

PASSIVE RADON MONITORING

Technical Measurements Center
UNC-Geotech
Department of Energy, Grand Junction Projects Office
September 21 - 22, 1987

Monday Morning, Sept. 21 - Rodeway Inn Compass Room
Session Chairman, Jack Duray, UNC Geotech

0830 Introduction
Leo Little, DOE-Grand Junction Projects Office

0900 "Studies of our Diffusion Barrier Charcoal Adsorption Detectors" - Bernard Cohen, University of Pittsburgh

0930 "A Comparison of Laboratory and Field Measurements of Radon - L. D. Michaels and A. S. Viner, Research Triangle Institute, T. Brennan, Cameoden Associates

1000 "The Response of Charcoal Canister Detectors to Time-Variant Radon-222 Concentrations" - Daniel D. Lee and Richard Sextro, Lawrence Berkeley Laboratory, University of California

1045 "Influence of Temporal Variations of Radon Concentrations on Passive Radon Monitoring" - Howard Pritchard, University of Texas

1115 "The Influence of Changing Exposure Conditions on Measurements of Radon Concentrations with the Charcoal Charcoal Adsorption Technique" - M. Ronca-Battista, U. S. Environmental Protection Agency, Washington, D. C. and David Gray, Jacobs Engineering Group

1145 "Recent Studies with Activated Carbon for Measuring Radon" - Andreas George, DOE Environmental Measurements Laboratory

Monday Afternoon, Sept. 21, Rodeway Inn Compass Room
Session Chairman, Dowell Martz, UNC Geotech

1330 "Operational Experiences in State-Wide Radon Surveys" - Harry Rector and William Schoenborn, Geomet Technology

1400 "Calibration and Experience with a Diffusion Barrier Charcoal Device, and An Alternative Method for Charcoal Correction" - Art Rood, Oak Ridge Associated Universities, Grand Junction Office, and Phil Kearney, Colorado State University

Monday Afternoon, Sept. 21 (Continued)

1430 "Moisture Insensitive Charcoal Canisters" - Henry Lucas, Argonne National Laboratory

1530 "Moisture Effects in Charcoal" - B. V. Alvarez, AirChek

1600 "Mathematical Modeling of Radon Uptake in Charcoal" - R. Nagarajan and D. T. Wasan, Illinois Institute of Technology, with L. D. Michaels, V. B. Menon and D. S. Ensor, Research Triangle Institute

Tuesday Morning, Sept. 22 - Rodeway Inn Compass Room
Session Chairman, G. Hal Langner, Jr., UNC Geotech

0900 "Charge Effects on Radon Alpha Track Distribution" - R. W. Pollock, Radon Environmental Monitoring, and Eugene Benton, University of San Francisco

0930 "Considerations in Assessing the Performance of Alpha Track Detectors" - Richard Oswald, Terradex Corporation

1030 "A Comparison of Four Types of Alpha-Track Radon Monitors" - Mark Pearson, UNC Geotech

1100 "Comparison of Three-Month Screening Measurements with Year-Long First Measurements Using Track Etch Detectors in the Reading Prong" - Carl Granlund and Michelle Kaufman, Pennsylvania Department of Environmental Resources

1130 "Comparison of Landauer RadTrak and Terradex Type SF Track Etch Detectors for Radon Measurements in Residences" - D. C. Landguth, Oak Ridge National Laboratory, and Mary Smuin, Oak Ridge Associated Universities

Tuesday Afternoon, Rodeway Inn Compass Room
Session Chairman, Dowell Martz, UNC Geotech

1330 Panel Discussion - "Current and Future Research Needs for Passive Radon Monitoring", Panel Members - Bernard Cohen, Henry Lucas, Richard Oswald, Linda Michaels, Carl Granlund, Moderator - Dowell Martz