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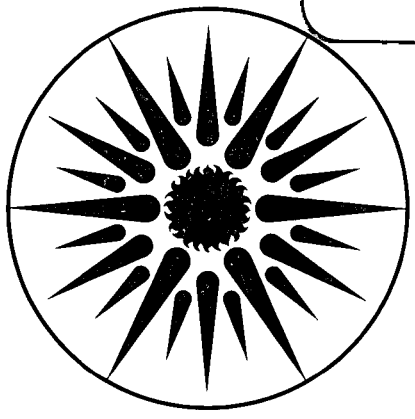
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October 1988

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The Performance of Charcoal-based Radon Detection Under Time-Varying Radon Conditions: Experimental and Theoretical Results

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ABSTRACT

Radon adsorption by charcoal is a widely used technique for measuring indoor radon concentrations, particularly when short-term results are desired. There are several different devices available, ranging from permeable envelopes filled with charcoal and open-face charcoal-filled canisters to devices incorporating diffusion limiting features to reduce losses of radon due to desorption. However, the integration characteristics of these samplers are not well understood, particularly under conditions of highly varying radon concentrations. A model for predicting the response of various types of charcoal-based detectors to time-variant radon concentrations has been developed; the model predictions compare well with results from chamber experiments. Both the experimental and theoretical results have also been compared with integrated continuous-sampling measurements. The implications of these comparisons for use of charcoal for screening measurements is discussed.

INTRODUCTION

Radon adsorption by charcoal is the basis for passive sampling devices that provide an easy and inexpensive method of obtaining quick diagnostic measurements of ^{222}Rn concentrations. This method is well suited to γ -ray counting and exposure times of only a few days provide sufficient activity for analysis. This feature facilitates measurements over a short period of time, however Rn can also desorb from charcoal under conditions of decreasing airborne concentrations. Thus, the response of charcoal-based passive samplers to time-variant Rn levels is an important consideration, particularly in light of the diurnal changes in radon concentrations that have been observed in some houses (1).

Several different charcoal-based passive samplers are available, of which there are two main types; devices in which the charcoal is exposed directly to the air (open faced canisters and permeable bags filled with charcoal are two common examples) and devices that incorporate a diffusion barrier that limits the rate of radon accumulation by the device, and at the same time, restricts the back diffusion rate of radon out of the sampler. A model for predicting the behavior of several of these devices has been developed, and is described briefly in this paper. A set of experiments has been conducted to test the model, and to examine the performance of both the model and the charcoal devices in comparison with data acquired with continuous radon monitoring instrumentation.

THEORETICAL DESCRIPTION

Models describing radon adsorption and desorption by charcoal canisters, both open-face and diffusion-limited, have been assembled, and a detailed discussion of the modeling is provided in reference (2). In general, the models are based on solving the one-dimensional diffusion equation (3),

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

where the charcoal bed depth is l cm and $0 \leq x \leq l$ is the height above the bottom of the canister, $y(x, t)$ the concentration of ^{222}Rn in the charcoal in pCi/cm^3 , t the time in seconds, D is the diffusion constant for radon in charcoal, and λ is the radioactive decay constant for ^{222}Rn . A sketch of the open-face canister is shown in Figure 1a.

Assuming that the lid is taken off at time zero, and that the canister had no ^{222}Rn in it beforehand, the initial condition is then $y = 0$ for all x . The boundary conditions at the bottom of the canister and when the top is exposed to the Rn in the environment are $\frac{\partial y}{\partial x} = 0$ at $x = 0$, and $y = k\rho C(t)$ at $x = l$. Here k is the adsorption coefficient of charcoal, ρ the measured bulk density of charcoal (which includes porosity), and $C(t)$ is the concentration of Rn in the air as a function of time.

It is not possible 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. Using this technique, along with separation of variables, one arrives at a series solution to $y(x, t)$. Hence, knowing $y(x, t)$ for all t , the total amount of ^{222}Rn in the canister at any exposure time T_0 can be found by integrating

$$\text{Rn in Canister at time } T_0 = \int_0^l y(x, T_0) dx. \quad (2)$$

In the case of the diffusion-limited devices, the picture of the diffusion barrier was simplified somewhat in order to utilize the one-dimensional solution derived for the open-face device, described above. By assuming air in the immediate space above the charcoal bed is well mixed and that the diffusion barrier connecting this space with the outside air can be described as an open cylinder (see Figure 1b), an iterative process is used to deduce the concentration of Rn in this well-mixed air for different times during the exposure. The main differential equation which describes the concentration of Rn in that layer of air is,

$$\frac{\partial C_i}{\partial t} = \frac{1}{V} \left[\frac{D_{air} A_{db}}{L_{db}} (C_o - C_i) - \frac{\partial Q_{Rn}}{\partial t} \right] - \lambda C_i, \quad (3)$$

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 $0.12 \text{ cm}^2 \text{ s}^{-1}$ at room temperature; L_{db} is the length of the diffusion barrier; 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. (3) 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 and the differential equation can again be solved.

EXPERIMENTAL DETAILS

Two examples of each of the two basic types of charcoal sampler were used for this study. For the open-face type devices, one of the samplers, referred to here as 'EPA', is an open-face canister 10 cm in diameter, containing approximately 70 g of charcoal in a bed 1.8 cm deep. A set of these canisters was obtained directly from the EPA Eastern Environmental Radiation Facility. The second open-face type sampler is a flow-through charcoal filter cartridge, designated here as 'FLT', often used for respirators. This device has essentially two open faces, and was modeled as the equivalent of two back-to-back open-face canisters each with half the measured bed thickness.

For the diffusion-limited samplers, one device, referred to here as 'COH', is a canister containing 25 g of charcoal in a bed 1.4 cm deep, covered by a lid containing a 1.9 cm diameter hole. This hole is covered by very fine mesh nylon screen, which serves as a diffusion barrier. These detectors, described in ref (4), were obtained from the University of Pittsburgh Radon project. A second diffusion-limited device, labeled 'LBL', is based on an EPA charcoal canister, covered by a lid containing a 1.4 cm-diameter hole and a 2.7 cm-long section of copper tube that serves as a diffusion barrier. Since the configuration of this device was designed to match that used in modeling the diffusion-limited samplers, it provided a means of checking the accuracy of the model assumptions. Three of the four device configurations used in these experiments are illustrated in Figure 1.

In order to determine empirically the radon diffusion coefficient in charcoal, $D_{charcoal}$, the charcoal was first removed from the EPA and COH canisters, then loaded with radon by spreading the charcoal into thin beds in a room-size chamber containing radon. After several hours the charcoal canisters were reassembled and placed on a 3 x 3 NaI detector in a low-background, low radon, lead-shielded enclosure through which air was circulated at a low flow rate. The lid (including the diffusion barrier in the COH device) was removed from the canisters and radon allowed to desorb and diffuse out of the charcoal bed. All the charcoal sampler experiments were done by counting the 609 keV peak from ^{214}Bi , a radon decay product. The effective diffusion coefficients for the EPA (and LBL, since it is based on the EPA canister) and COH devices were then directly determined by fitting the experimental data using the open-face model, for which $D_{charcoal}$ is the only free parameter.

A similar set of diffusion-out experiments in which the diffusion barriers were left in place for the LBL and COH devices was conducted to provide an empirical determination of the diffusion barrier parameters. In using the diffusion-limited model, the area and length are effectively the two parameters that can be adjusted to fit the diffusion-out data. In the case of the LBL device, no alterations to the measured diffusion barrier parameters were needed to adjust the model results to the measured data. For the COH device, the measured hole diameter was used, and the effective length of the diffusion barrier adjusted to fit the model results to the data. The modeling parameters determined from these experiments are summarized in Table 1.

Chamber experiments were then conducted in order to compare the response of these various charcoal sampler configurations with integrated data from continuous radon measurements under conditions of varying radon concentrations. In addition, the predicted and observed performance of the charcoal samplers could be compared, providing a test of the models. Several canisters of each type were placed in the two rooms of the chamber. One room had a high ^{222}Rn content, ranging between 50 and 250 pCi/L and the other room had a lower ^{222}Rn content, ranging between 5 and 50 pCi/L. The actual ^{222}Rn concentrations were measured over time using continuous radon monitors. The 4-day experiment was divided into four approximately equal exposure periods. At the end of each period, some of the canisters were switched from one room to the other to vary the ^{222}Rn levels to which the canisters were exposed. Other canisters were sealed and removed, in order to test the effect of varying the exposure times. Additional ^{222}Rn was supplied to the high concentration room at the beginning of the second, third and fourth periods. The actual radon concentrations in the two rooms and the exposure periods are illustrated in Figure 2.

RESULTS AND DISCUSSION

The continuous radon concentration profile to which each canister was exposed was used in the appropriate model to predict the total radon activity in the canister. The predicted responses of two of the charcoal devices, EPA and LBL, are illustrated in Figures 3 and 4. As can be seen in Figure 3, the open-face device has a pronounced response to changes in the radon concentrations to which it is exposed. Radon activity in the canister increases rapidly upon initial exposure, but begins to decline as the room air concentration decreases. When the canister is moved into the low concentration environment, activity adsorbed in the canister

drops significantly due to desorption losses. In the case of the diffusion-limited sampler, the response to changing airborne radon concentrations is attenuated considerably, as can be seen in Figure 4. Similar simulations were run for the case where canisters were first exposed to low radon concentrations, followed by placement in the high concentration environment. These predictions of the radon concentration profiles in the canisters under different exposure conditions help demonstrate the fact that charcoal-based passive radon samplers "remember" the later exposure conditions better than the earlier conditions. This sampling memory is improved by use of a diffusion-barrier.

The predicted amount of radon in the samplers at the time the canister was closed and removed from the radon environment can be compared to the values measured for each sampler. These measured values can then be compared with the actual integrated exposures, based on data from the continuous radon monitors. The exposure conditions and the observed and predicted results are summarized in Table 2. As can be seen from this table, the model predictions for the two open face and the two diffusion barrier configurations compare quite well with the measured responses of these same charcoal devices. The last column in Table 2 compares the average radon concentrations inferred from the charcoal canister measurements with the actual integrated average concentrations based on the continuous data. The largest variations are found in the results for the open-face canisters where back-diffusion losses of ^{222}Rn are significant when the radon concentrations change from high to low. The responses of the diffusion-limited devices are damped and the differences between the average radon concentrations and those inferred from the sampler measurements are not as great. The fact that the devices do not appear to over-predict the average radon concentrations when exposed first to low then to high radon concentration environments is an artifact of the experimental conditions, since radon concentrations in each room were not held constant throughout the exposure period.

CONCLUSIONS

The models developed for both open-face and diffusion-limited devices appear to accurately simulate the behavior of charcoal-based passive samplers under conditions of varying radon concentrations. Overall, the model and experimental results indicate that the diffusion-limited charcoal detectors are better integrators than their open face counterparts. However, as noted earlier, results from charcoal-based detectors are heavily influenced by the most recent exposure conditions, although diffusion barriers improve this situation somewhat.

The comparison of actual average radon concentrations with those inferred from the charcoal samplers demonstrates the limitations in using charcoal canisters to obtain integrated readings over short periods of time when the actual ^{222}Rn concentrations are changing. It is possible, under the conditions of time-varying radon concentrations, that the results from charcoal sampler measurements could either under- or over-predict actual concentrations. The former is the potentially the more serious error that could arise in using these devices for screening measurements. Thus, in addition to the the uncertainties in using short-term measurements to predict long-term average radon concentrations, the charcoal sampling systems themselves are inherently uncertain, and quantitative comparisons of results obtained with such samplers with specific concentration guidelines are unwarranted without explicitly taking these uncertainties into account.

ACKNOWLEDGEMENTS

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Table 1: Modeling Parameters

Open-face Devices:

Canister	Bed Depth (cm)	D (cm ² /s)
EPA	1.8	1.6 x 10 ⁻⁵
COH	1.4	1.45 x 10 ⁻⁵ *
FLT	1.2	1.3 x 10 ⁻⁵ **

Diffusion Barrier Devices:

Parameter	COH	LBL
D_{air} (cm ² /s)	0.12	0.12
$D_{charcoal}$ (cm ² /s)	1.45 x 10 ⁻⁵	1.60 x 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.

** From ref. 4.

*** From ref. 5.

**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)]
			Measured (2)	Model Prediction	(1) (%)
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 2 for corresponding Rn concentration profiles.

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

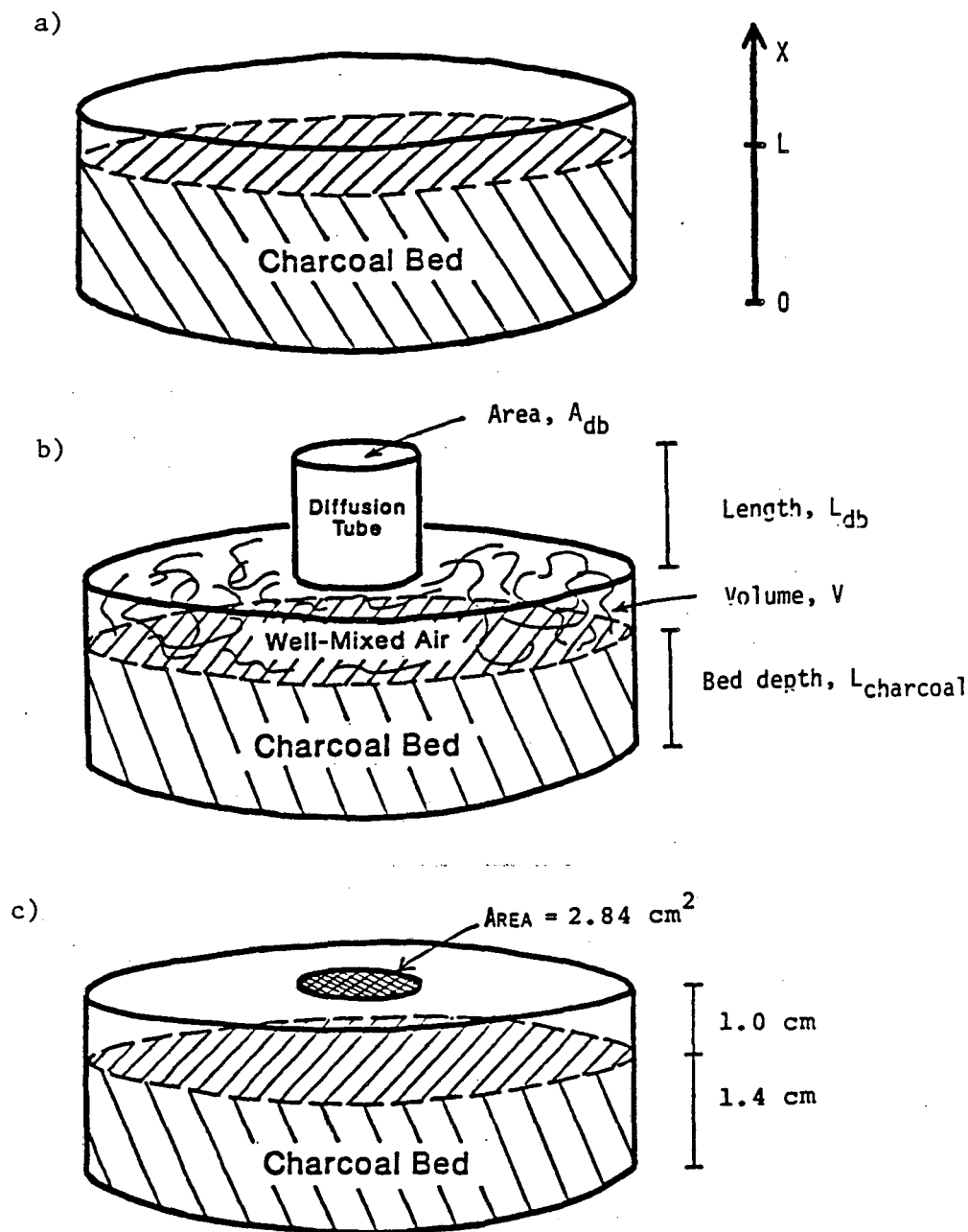
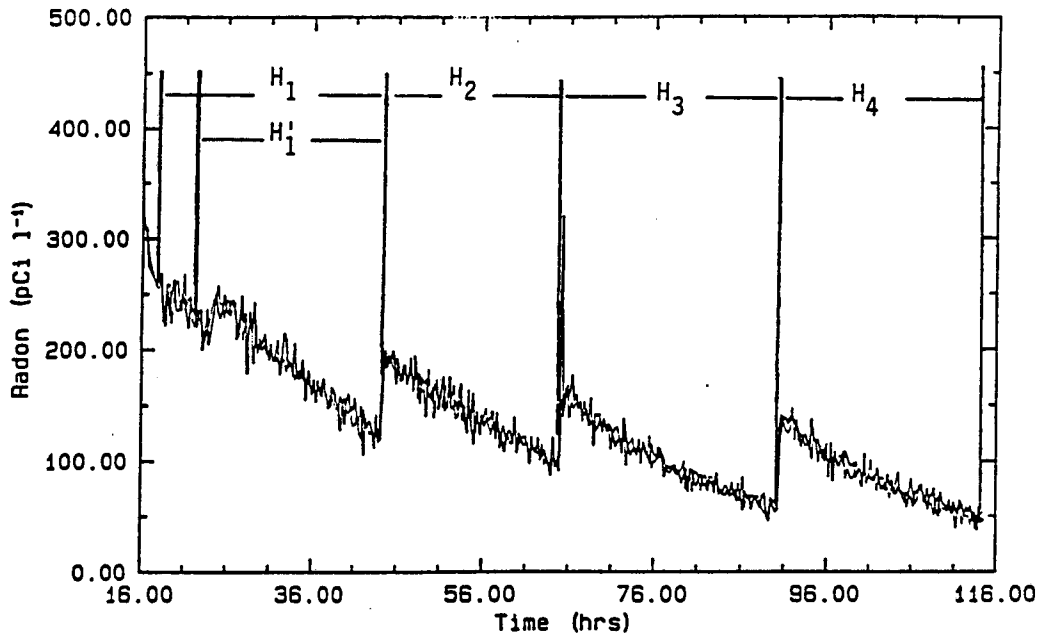
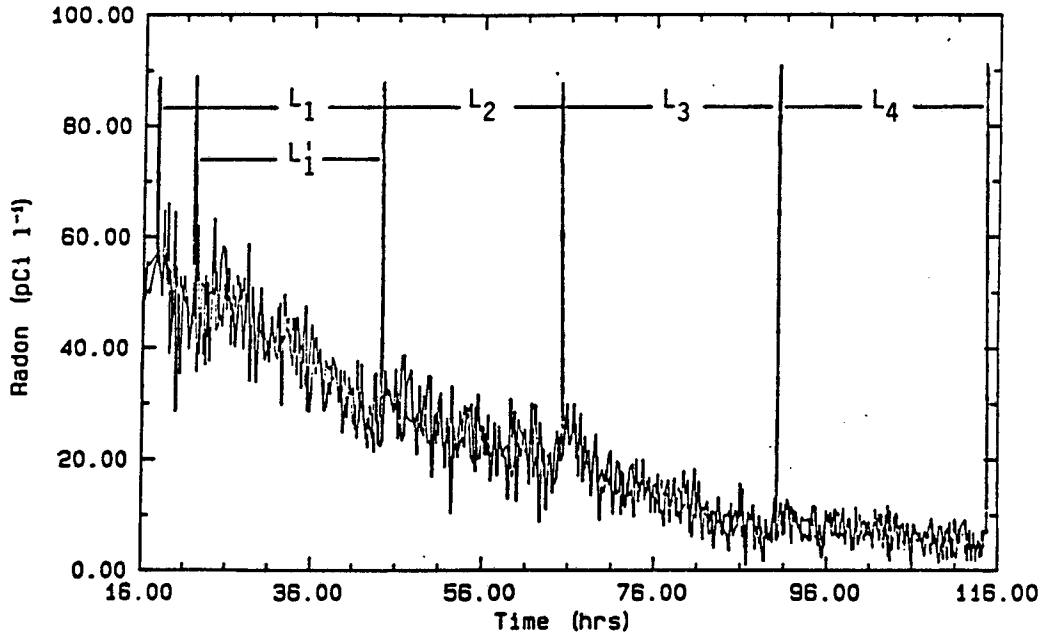


Figure 1. Several charcoal canister configurations. The top sketch (a) is an open-face device, referred to in the text as 'EPA'. The middle diagram (b) shows a diffusion-limited sampler, illustrating the parameters used for the model. This configuration is also used by the device referred to as 'LBL' in the text. The bottom sketch (c) shows the diffusion-limited 'COH' sampler. The diffusion barrier consists of the hole in the canister lid covered by a fine mesh nylon screen. The dimensions shown in the figure are for the COH device.



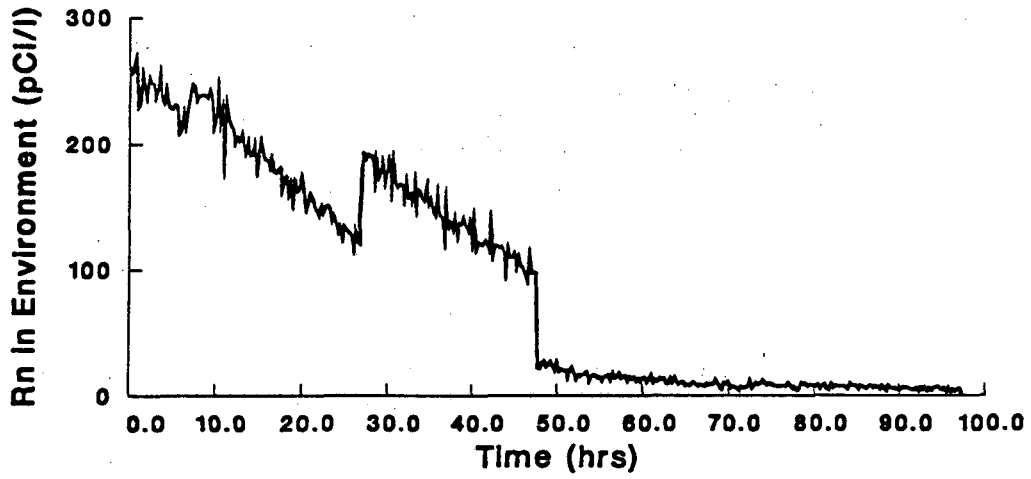
(a) High Conc. Room



(b) Low Conc. Room

Figure 2. Radon concentrations and exposure periods for the two-room chamber experiments.

Rn in Environment vs Time



Rn Activity in Canister vs Time

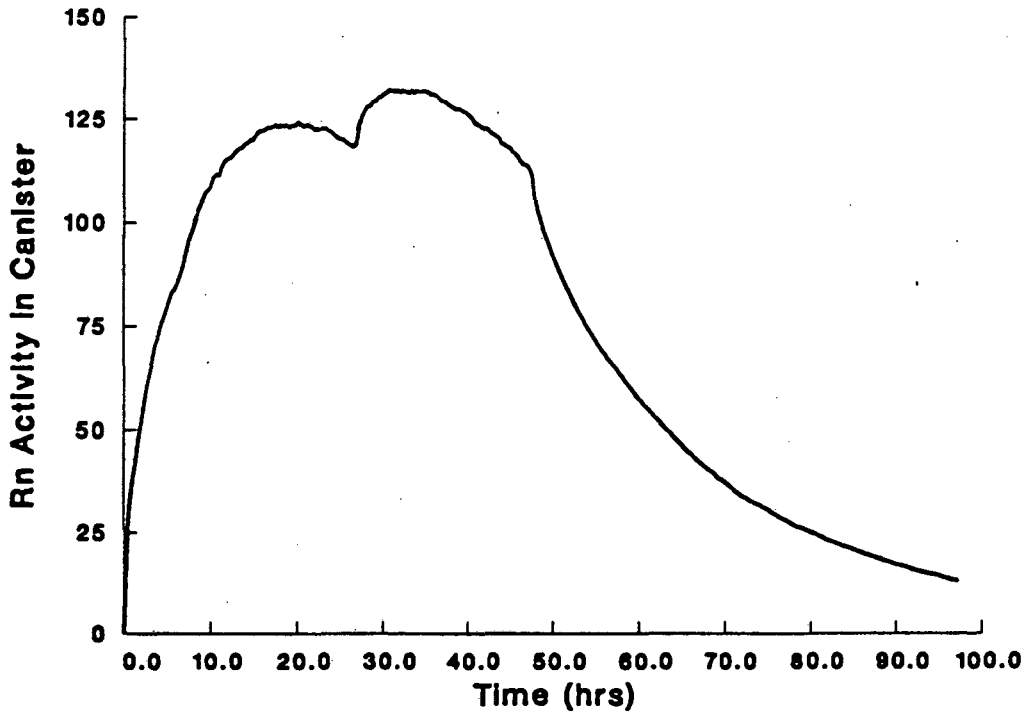


Figure 3. Predicted response of the EPA device to the radon concentration profile shown at the top of the figure. The exposure condition is summarized in Table 2 as H1,2L3,4.

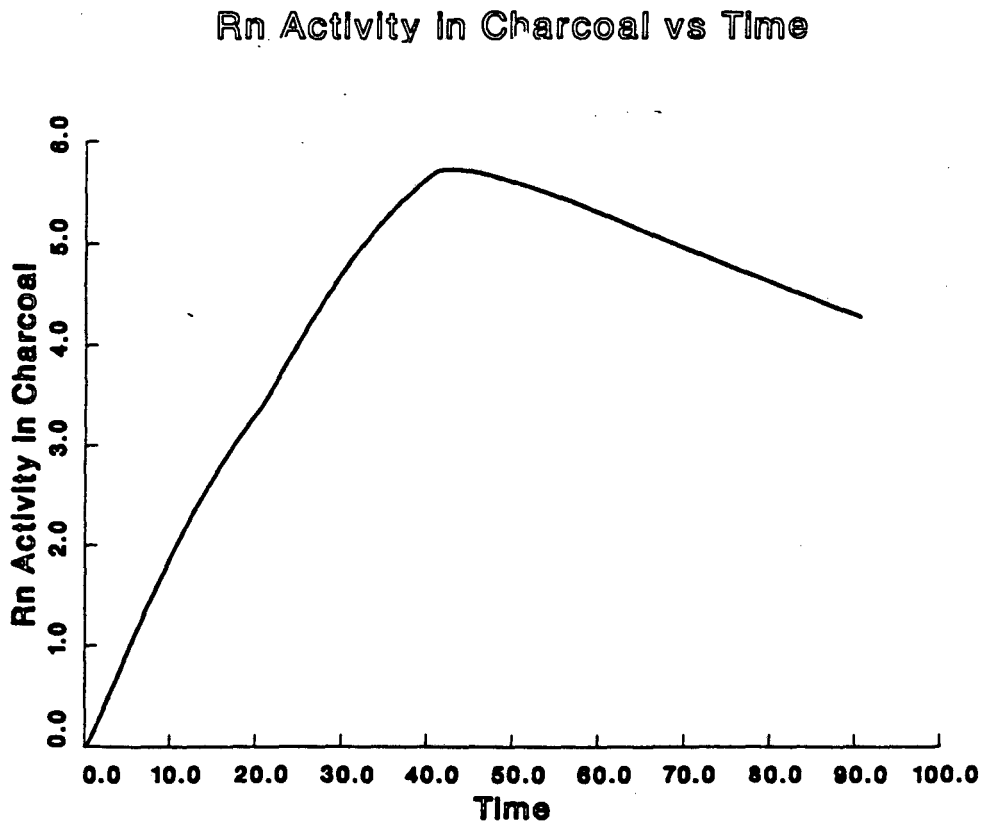
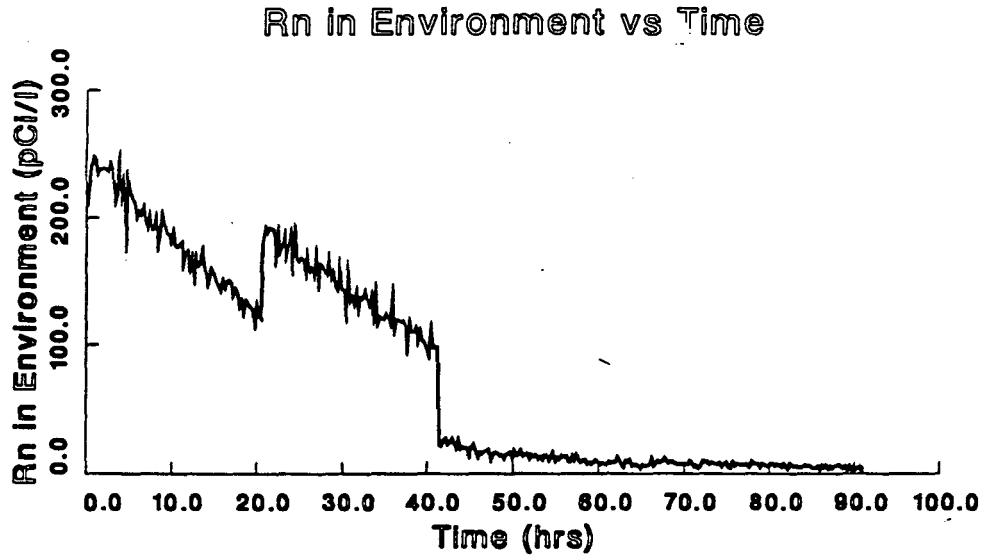


Figure 4. Predicted response of the LBL device to a radon concentration profile similar to that shown in Figure 3. The exposure condition, as summarized in Table 2, is H1',2L3,4.

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