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On the Exhalation Rate of Radon by Man*

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Abstract

This paper describes some aspects of the exhalation rate of radon by man which may be relevant to its internal dosimetry and, therefore, to possible radiobiological consequences. Prolonged exposure of a person to radon results in a reservoir of radon dissolved in body fat and fluids. If the person then moves to an environment with a lower radon concentration, there is a net exhalation of radon and the initial exhalation rate depends on the radon concentration in the first environment. This is demonstrated for seven persons whose houses contained radon at concentrations varying from 10 Bq m^{-3} to almost 1000 Bq m^{-3}. About one hour after leaving the house, the subjects' average exhalation rate of radon, expressed as the equivalent volume of house air per unit time, was 236 mL min^{-1}.

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In general, the exhalation rate declined in a manner that seemed to be predictable from the integral of the equation that describes the retention of a single inhalation exposure to radon. However, the behavior is complicated by a major but short-lived postprandial increase in the exhalation rate of radon by persons whose only source of radon was in the air of their homes. The phenomenon was studied in seven subjects, who showed initial exhalation rates ranging from 3 mBq min^{-1} to 200 mBq min^{-1}. The excess radon exhaled amounted to ~4 L to ~15 L, when expressed as the equivalent volume of house air. This radon must have come from a reservoir somewhere in the body; the possible dosimetric and radiobiological consequences of this phenomenon are unknown.

Introduction

The current concern about the possible health effects of indoor radon necessitates the investigation of all aspects of radon in man, i.e., physical as well as biological factors. In this paper we present some observations on the rate of exhalation of radon inhaled in the indoor environment.

It has been known for many years that the retention in the human body of radon that has been inhaled can be described mathematically by the sum of five exponential functions of time (Harley et al., 1951; Lucas and Markun, 1972). Each function is considered to represent retention in a particular compartment in the body. Thus, the compartment that clears very rapidly (half-time 23 s) presumably represents the lung, while the slowest compartment (half-time about 18 hours) is identified as fat. The other compartments have half-times of about 4.5 min, 41 min, and 120 min. Consequently, prolonged inhalation of radon results in a pool in the body, the size of which is determined both by the radon concentration in the air and by the duration of the exposure. In general, an equilibrium will be reached in exceptional cases only (i.e., cases of continuous exposure for several days).

When a person moves to an environment with a lower radon concentration, there is a net exhalation of radon and the exhalation rate is determined by the same factors. If the parameters of the retention equation were fairly similar for all persons, there should be a linear relationship between the initial exhalation rate and the concentration in the house for exposures of similar durations.

Our observation of a postprandial peak in the rate of exhalation of radon by persons with long-standing burdens of radium (Rundo et al., 1978) raised the question of the origin of the "excess" radon exhaled during the period of digestion. If it were the result of a transient change in the fraction of radon lost from bone, the dosimetric consequences would be trivial. On the other hand, if it were the result of the flushing of a reservoir of radon dissolved in soft tissue, it could be important to identify the reservoir, which might be the recipient of chronic irradiation at levels greater than many soft tissues. This question would be resolved by the demonstration of a postprandial peak in the exhalation of radon, the only source of which was inhalation in the home environment.

We therefore decided to determine serially the exhalation rate of radon in the breath of ourselves and some colleagues, paying particular attention to the period following a meal. In this way we could explore the relationship between a "high" environmental radon concentration and the initial exhalation rate after leaving that environment, as well as seek evidence for a postprandial peak in the exhalation rate.

Experimental Methods

Samples of air were collected from the houses of seven subjects to determine the concentration of radon to which they had been exposed for periods of about 14 h. Data on the subjects are shown in Table 1.

Five of the subjects brought a breakfast of choice and went to the underground laboratory ("vault"), which is ventilated with radon-free air, as soon as possible after arriving at Argonne. One or two 10-min samples of breath were taken for determination of the initial exhalation rate of radon, and the subject then ate breakfast. Breath sampling was started immediately after the end of the meal and at first was almost continuous: serial 10-min collections were made during a period of 1-2 h, with a 2- or 3-min interval between each collection while the radon trap (charcoal) was being changed. After this time less frequent collections were made.

Results

The exhalation rates for the first 10-min samples of breath are plotted as a function of the indoor radon concentrations in Figure 1. There is an altogether respectable correlation between the two, the straight line having a slope of 236 mL min^{-1}, when radioactivity is expressed in units of the equivalent volume of room air (EVRA; Lucas and Stehney, 1956). The relatively slight spread of the points (in an admittedly small sample) indicates that the retention equations for the seven subjects must have been similar.

Other factors being equal, one would expect the radon exhalation rate to decline monotonically. In Figure 2 the serial results for subject 50-150 (for whom the initial exhalation rate plotted in Figure 1 was just over 100 mBq min^{-1}) show that "other factors are far from equal." The curve labelled "Integrated retention equation" is a plot of the retention function for a single intake of radon, integrated for continuous exposure for 14 h, and then normalized to pass through the early experimental points. It should be noted that statistical uncertainties are not indicated because, for all points, error bars ($\pm 1\sigma$) would be no greater than the heights of the symbols; the variability was therefore due to biological factors. After breakfast (eaten at the time indicated by the letter "B"), an initial decrease in the radon exhalation rate was followed by a substantial increase to a peak some 70-90 min after breakfast, and then a gradual decline to a "normal" or "expected" level some 3 h after the meal. The subject also ate his lunch (at the time indicated by the letter "L") in the underground laboratory, and serial breath collections and analyses for radon were continued. The results suggested that a second postprandial peak was present, but the exhalation rate 3 h after lunch was considerably higher than expected on the basis of the smooth curve representing the integrated retention function.

A similar series of breath collections and radon analyses was made for the two other subjects whose houses showed radon concentrations in excess of 150 Bq m^{-3}. The results are plotted in Figure 3 together with the results for subject 50-150.

In all three cases, there was a pronounced postprandial peak after breakfast, although the increase started sooner after the meal for subject 50-148. The dashed lines drawn under the first peaks represent assumed exponential baselines to permit the calculation of the "excess" radon exhaled during the period of the peak. The subjective nature of this procedure is self-evident and should be borne in mind. The evidence for a peak after lunch was less conclusive for subjects 50-026 and 50-148 than for subject 50-150, although still suggestive.

We wished to determine if the postprandial peak could be deferred until after lunch. Accordingly, two subjects ate no breakfast; the exhalation rate of radon was determined at intervals during the morning and continued more frequently after lunch, which was eaten in the underground vault. The results are plotted in Figure 4. Error bars ($\pm 1\sigma$) are again within the heights of the symbols. The postprandial peaks are quite unambiguous in both cases, but the shapes are very different. For subject 50-109, the dashed line represents the extrapolation of a least squares fit of an exponential function to the data from 143 to 295 min, and it was used as the baseline for the peak. This procedure was not possible for subject 50-070 because the data showed no well-defined decrease before lunch; the results also suggest that sampling of the breath was discontinued before the exhalation rate of radon had recovered from the elevated values.

The total amount of radon exhaled during the period of the peak was calculated for each subject. The amount that was exhaled during the time between the collections of successive pairs of samples was assumed to be at a rate equal to the average of the rates before and after. These quantities were calculated and added to the sum of the amounts found in the sampling periods. With one exception, the estimated amount of exhaled radon which was not collected ranged among the subjects from 30 to 41% of the total. The exception was the first test on subject 50-009, where it was 69% of the total; this was an exploratory test, and gamma-ray measurements were also being made in vivo during the course of the morning.

For each subject, we assumed that had there been no postprandial peak, the rate of exhalation of radon would have followed an exponential decline as exemplified by the dashed lines in Figures 3 and 4. The total area under the baseline was determined by integration of the exponential function between appropriate limits. This quantity was then subtracted from the total radon exhaled during the period of the peak; the results are set out in Table 2.

The uncertainties shown are based on the statistical errors of counting; however, in the calculation of the error on the total amount exhaled, the estimated error on the amount exhaled between successive pairs of samples was doubled before propagation with those for the samples, to allow for some biological variability. For each subject, the uncertainty on the area under the baseline was derived from the error on the ratio of the exhalation rates at the beginning and end of the period of the peak.

In order to compare the data for subjects who had been exposed to widely differing concentrations of radon (but for comparable periods of about 14 to 16 h), the net area in each peak was converted from activity to the equivalent volume of room air by dividing by the appropriate concentration in the house air. The results are in the penultimate column of Table 2. The range of values for the peak areas is reduced from about 70:1 to about 4:1. As a measure of the relative effect, the ratio of the net peak area to the area under the baseline was calculated, and the results are given in the final column of Table 2. They show a range of about 3:1.

Discussion

The observation of the postprandial peak in the exhalation of radon from the environment proves that the source of the similar peak in the exhalation of radon produced in vivo is a reservoir of radon dissolved in body fluids or soft tissue and not a change in the fraction released from radium in bone. It was hoped that the area of the peak, when expressed as the equivalent volume of room air, might give some clue as to the identity of the reservoir. However, there is no obvious correlation between the area of the peak and any of the variables in Table 1. The identity of the reservoir remains a mystery:

one possibility is the capillary bed; another, perhaps more likely, is the omentum, which may contain much adipose tissue. The higher solubility of radon in fatty media than in aqueous ones is well-known. More data are needed before any conclusion can be drawn concerning the dosimetric or radiobiological consequences.

The detailed behavior of the exhalation rate during the period of the peak merits comment. For subjects 50-026 (Figure 3) and 50-109 (Figure 4), there is a suggestion of a splitting or doubling of the peak. When first observed (in subject 50-109), this was thought merely to be biological variation, but its observation in subject 50-026 tended to confirm it as a real phenomenon, even though its existence depends on the validity of one low point in each case. The peaks for subjects 50-148 and 50-150 (Figures 2 and 3) do not show a doubling, but there is a marked asymmetry of the peak in each case. Because of variations in respiratory minute volume of subject 50-148, there was actually a small increase in the concentration of radon in the breath at 158 min. Similarly, the results for subject 50-070 (Figure 4) suggest that there may have been a "step" in the declining portion of the peak which was not observed because breath sampling was discontinued too early. These observations warrant further study, perhaps with a stable isotope of another of the noble gases, the behavior of which may be similar to that of radon. It is possible that this noninvasive technique may offer a tool to physiologists for the study of some aspects of the digestive process not otherwise amenable to investigation.

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Table 1. Details of seven subjects and the concentrations of airborne radon in their houses.

Subject No.	Sex	Height, m	Weight, kg	Age, yr.	Rn concentration[a] in house air, mBq L^{-1}
50-002	M	1.68	78.2	51	57 ± 4
50-009[b]	M	1.78	77.3	52	59 ± 4
50-009[b]	M	1.78	77.3	52	51 ± 4
50-026	F	1.58	94.5	29	962 ± 21
50-070	M	1.83	82.3	32	10 ± 3
50-109	M	1.71	84.1	38	46 ± 2
50-148	M	1.74	69.5	52	153 ± 6
50-150	M	1.73	79.3	30	518 ± 5

[a]Means of pairs of consistent results on samples taken the night before and on the morning of the test, except for subjects 50-026 and 50-070, where the house air samples were taken two days later.

[b]Subject 50-009 was tested on two occasions, a week apart.

Table 2. *Total, baseline, and net excess amounts of radon exhaled during the period of the postprandial peak.*

Subject No.	Total radon exhaled during postprandial peak, Bq	Area under baseline, Bq	Net peak area		
			Bq	EVRA[a] liter	Peak area / Baseline area
50-002	1.63 ± 0.01	1.25 ± 0.04	0.38 ± 0.04	6.7 ± 0.8	0.31
50-009	1.87 ± 0.03	1.59 ± 0.04	0.27 ± 0.05	4.7 ± 1.0	0.17
50-009	1.12 ± 0.01	0.93 ± 0.03	0.19 ± 0.03	3.7 ± 0.7	0.20
50-026	43.6 ± 0.3	32.9 ± 0.7	10.7 ± 0.7	11.1 ± 0.8	0.33
50-070[b]	0.57 - 0.63	0.43 - 0.44	0.14 - 0.19	13.6 - 17.8	0.33 - 0.42
50-109	1.32 ± 0.01	1.03 ± 0.05	0.28 ± 0.05	6.2 ± 1.2	0.28
50-148	3.32 ± 0.03	2.05 ± 0.05	1.27 ± 0.06	8.3 ± 0.5	0.62
50-150	12.3 ± 0.1	7.2 ± 0.2	5.1 ± 0.2	9.8 ± 0.4	0.71

[a]Equivalent Volume of Room Air - obtained by dividing the net peak area in mBq by the concentration in house air by mBq L^{-1} (Table 1).

[b]All values depend on starting and ending times assumed for peak and on how baseline is drawn. See Figure 4.

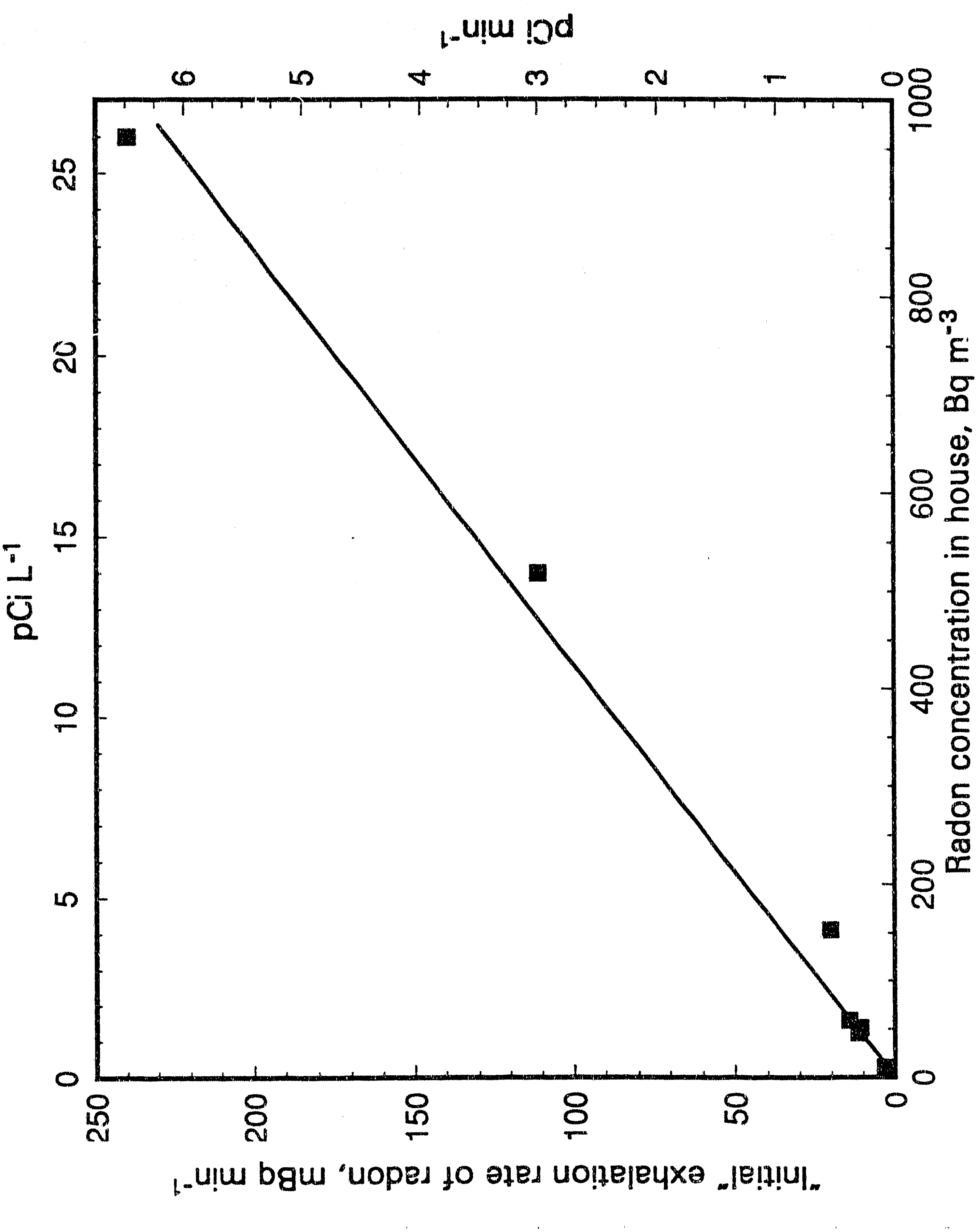
Figure 1. Exhalation rate of radon shortly after arrival at Argonne National Laboratory as a function of the radon concentration in the house for seven subjects.

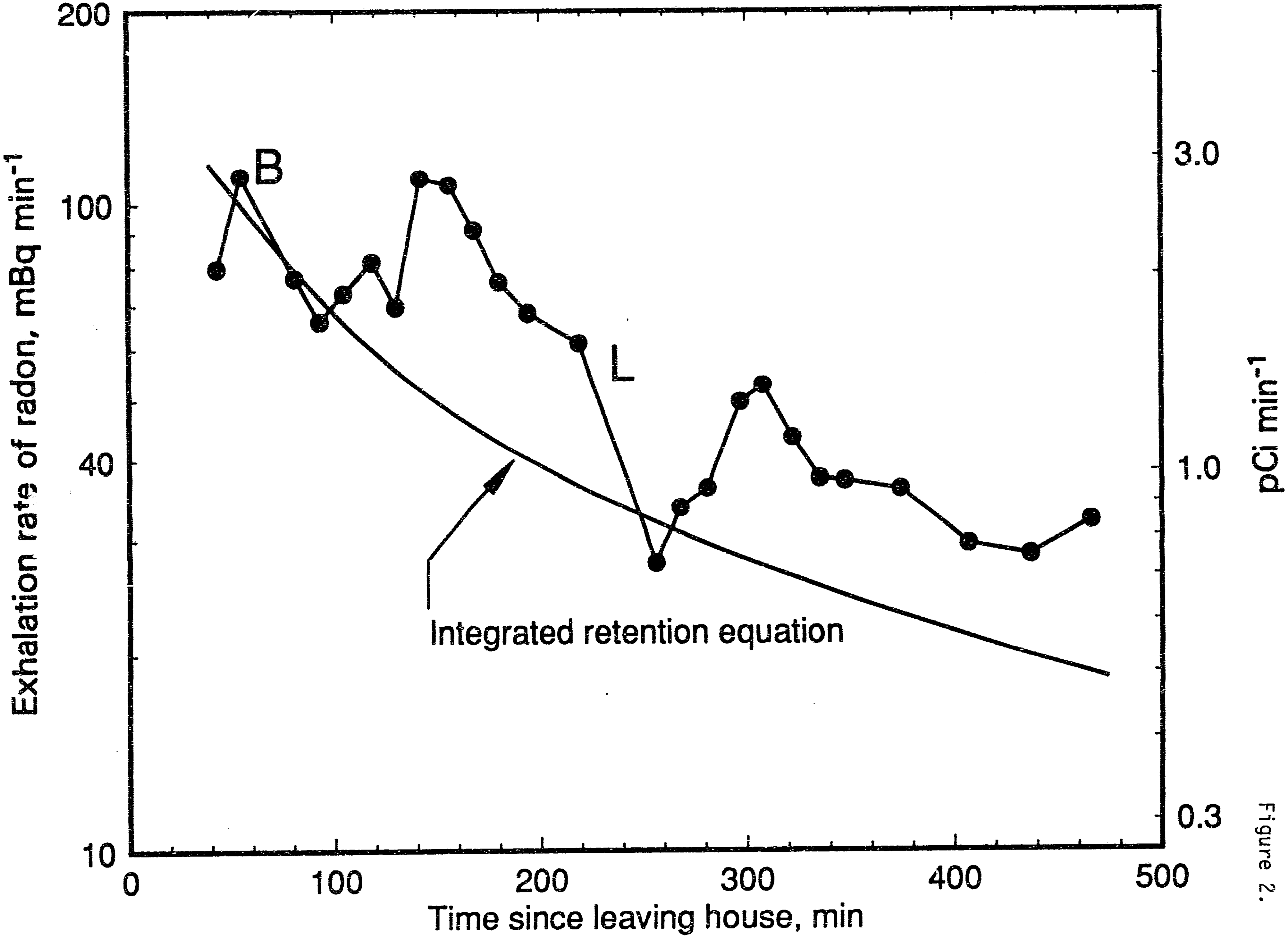
Figure 2. Exhalation rate of radon by subject 50-150 (experimental points) as a function of time since leaving home, compared with the rate predicted by the multi-exponential retention function, integrated for continuous exposure for 14 h and normalized by trial and error to the data (smooth curve). Letters indicate mid-points of meals (B - breakfast; L - lunch).

Figure 3. Exhalation rate of radon (logarithmic scale) for three subjects as a function of time since leaving home. The data plotted in Figure 2 are included. Letters again indicate mid-points of meals.

Figure 4. As for Figure 3, but for two subjects who ate no breakfast.

Figure 1.


pCi L^{-1}
0
5
10
15
20
25
pCi min^{-1}
0
1
2
3
4
5
6
Radon concentration in house, Bq m^{-3}
0
200
400
600
800
1000
"Initial" exhalation rate of radon, mBq min^{-1}
0
50
100
150
200
250



200
100
40
10
Exhalation rate of radon, mBq min-1
3.0
1.0
0.3
pCi min-1
B
L
Integrated retention equation
0
100
200
300
400
500
Time since leaving house, min

Figure 2.

Figure 3.

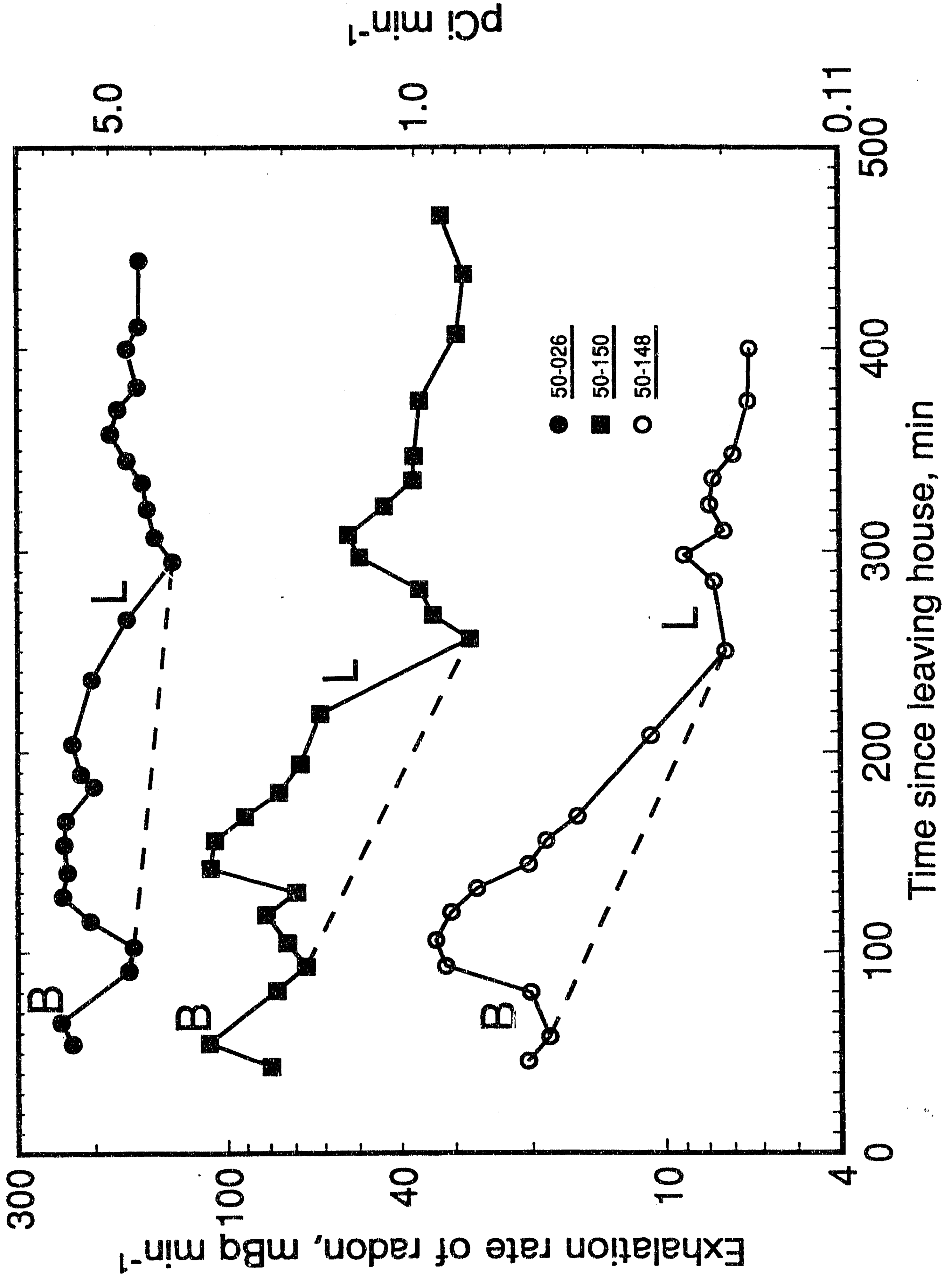
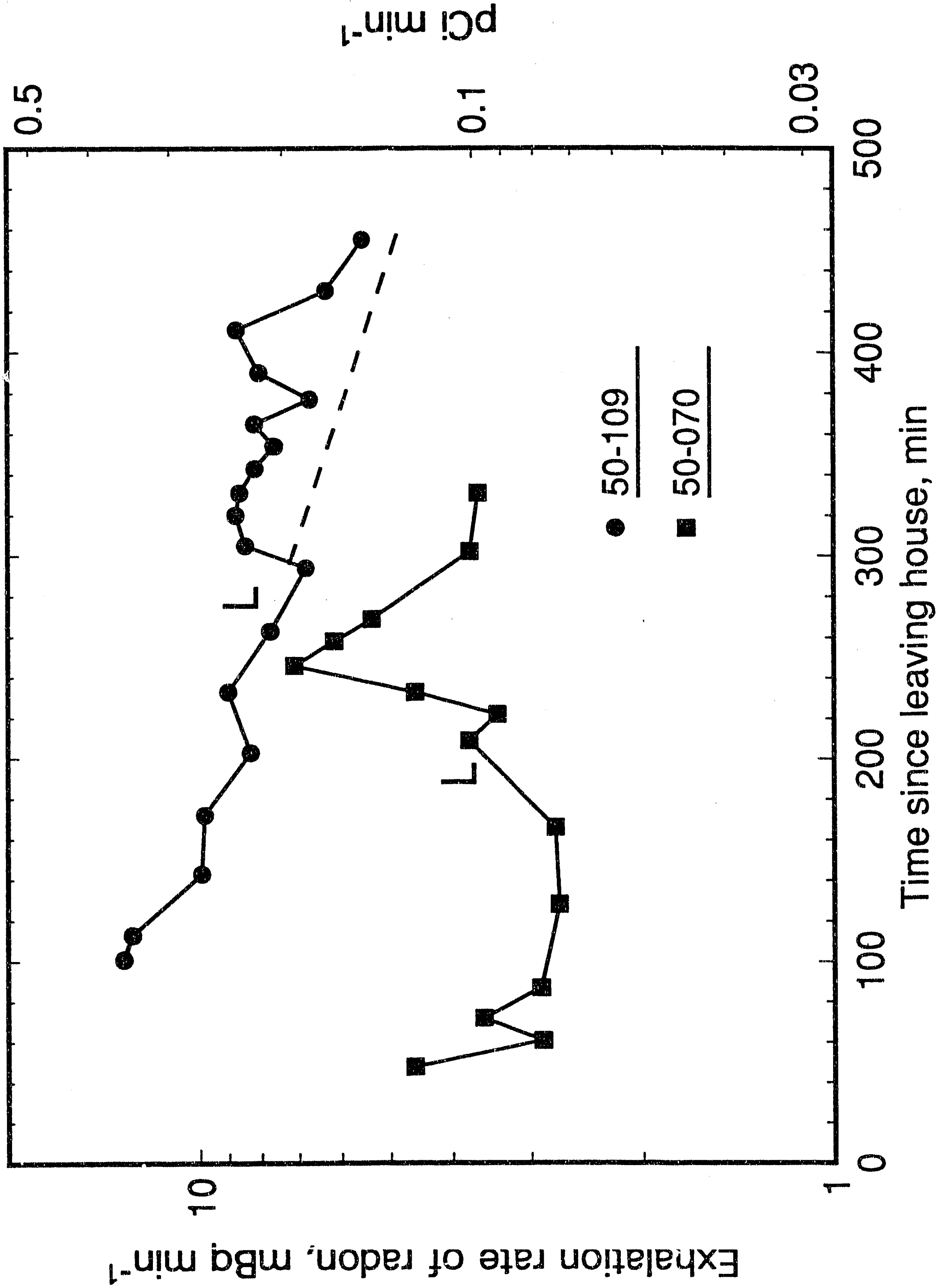

Exhalation rate of radon, mBq min-1
300
100
40
10
4
pCi min-1
5.0
1.0
0.11
Time since leaving house, min
0
100
200
300
400
500
B
L
50-026
50-150
50-148

Figure 4.



pCi min⁻¹
0.5
0.1
0.03
Exhalation rate of radon, mBq min⁻¹
10
1
Time since leaving house, min
0
100
200
300
400
500
L
L
50-109
50-070

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