

CONF-810153--4

DE83 008616

Some Aspects of Radon and Its Daughter-Products  
in Man and His Environment\*

by

**MASTER**

J. Rundo, F. Markun, N. J. Plondke and J. Y. Sha

Center for Human Radiobiology  
Argonne National Laboratory  
9700 S. Cass Avenue  
Argonne, IL 60439, U.S.A.

**DISCLAIMER**

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

**NOTICE**

**PORTIONS OF THIS REPORT ARE ILLEGIBLE.**

It has been reproduced from the best available copy to permit the broadest possible availability.

*JHP* DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

## SUMMARY

In this paper we report on three different aspects of radon and its short-lived daughter-products in man and his environment. The term radon is to be understood as referring to 3.82-day  $^{222}\text{Rn}$ , the daughter of  $^{226}\text{Ra}$ . The impetus for the studies derived partly from the study in this Center of the late biological effects in persons contaminated internally with  $^{226}\text{Ra}$  due to occupational or iatrogenic exposure. Such persons exhale substantial amounts of radon, and it used to be our practice to determine the exhalation rate as part of the determination of the body content of radium, although much variability in the exhalation rate has long been recognized. (1,2) We observed a major but short-lived postprandial increase in the exhalation rate; (3) we report here a similar effect in the exhalation rate of radon by persons containing no radium.

The second aspect to be described arose from the studies just mentioned, and it concerns the concentrations of radon and its short-lived daughter-products in houses. We observed unusually high ( $>5-10 \text{ pCi l}^{-1}$ ,  $>185-370 \text{ Bq m}^{-3}$ ) concentrations of radon in some houses with unpaved crawl spaces, (4) and subsequently in some houses with concrete basements. We have also determined the concentrations of the short-lived daughter-products in some of these houses. The results suggested an equilibrium factor of only 0.1, in disagreement with the results of others.

Finally, we report observation by external counting of radon daughter-products in the residents of one of the houses with a high level of radon. We also note the relevance of this to radiological protection, in that there may be interference with the assay of plutonium in the lung of persons who live in houses with high concentrations of radon.

### I. EXHALATION OF RADON

#### Introduction

Our observation of a postprandial peak in the rate of exhalation of radon by persons with long-standing burdens of radium, (3) raised the question as to 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, but 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 which had been inhaled previously. We therefore made serial determinations of the exhalation rate of radon by ourselves and some colleagues, paying particular attention to the period following a meal. The radon had been inhaled in the subject's home, where it builds up a "reservoir" in body fat and fluids.

Subjects brought a breakfast of choice and went to the underground low background laboratory ("vault") as soon as possible after arriving at Argonne. One or two 10-minute 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-minute collections were made during a period of one to two hours, with a two- or three-minute interval between each collection while the charcoal radon trap was being changed. After this time less frequent collections were made. The amount of radon exhaled in each 10-minute period was determined and the exhalation rate was plotted as a function of time after leaving home. The results were somewhat variable, but the common features were that a postprandial peak was present and that it was superimposed on a declining baseline. The most detailed results, obtained on three subjects who were tested for up to seven hours and who ate lunch as well as breakfast in the underground vault, are plotted in the left-hand half of Fig. 1 .

These were the subjects whose houses had the highest concentrations of radon (between 4 and 26 pCi  $\ell^{-1}$ ); the exhalation rates reflected this. Error bars ( $\pm 1\sigma$ ) are not shown because they were no greater than the heights of the symbols.

In addition to a pronounced postprandial peak after breakfast, there was a definite peak after lunch for two subjects and slight evidence for it for the third subject.

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 vault. The results are plotted in the right-hand half of Fig. 1. Error bars ( $\pm 1\sigma$ ) are again within the heights

of the symbols. The postprandial peaks are quite unambiguous in both cases, but the details are very different. For subject 50-109, the straight line represents the results of a least squares fit of an exponential function to the data from 143 minutes to 295 minutes, and the dashed extrapolation was used as the baseline for the peak. This procedure was not possible for subject 50-070; the results suggest strongly 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 case. The amount which was exhaled during the time between successive pairs of samples was assumed to be at a rate equal to the average of the rates before and after. These amounts were calculated and added to the sum of the amounts found in the sampling periods.

In order to compare the data for subjects who had been exposed to widely differing concentrations of radon (but for comparable periods of about 14-16 hours), the area in each peak (above a baseline assumed to be declining exponentially) was converted from pCi to an equivalent volume of room air by dividing by the appropriate concentration in the house air. The range of values for the peak areas was reduced from about 70:1 (280 pCi to 4 pCi) to about 4:1 (15 litres to 4 litres).

### Discussion

The observation of the postprandial peak in the exhalation of radon from the environment proves conclusively 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. The identify of the reservoir remains a mystery, although one possibility is the capillary bed. More data are needed before any conclusion can be drawn.

The suggestions of "splitting" or asymmetry of the postprandial peak seen in Fig. 1 warrant further study. It is possible that this non-invasive technique may offer a tool to physiologists for the study of some aspects of the digestive process not otherwise amenable to investigation. However, it is clear that determination of the exhalation rate of radon is not an adequate method of assay of  $^{226}\text{Ra}$  *in vivo*. Indeed observation of exhalation rates in the range 4-6 pCi/min (subject 50-026, Fig. 1) would be interpreted as corresponding to 32-48 nCi of freely emanating radium in a person with prior potential exposure to that element.

## II. RADON AND DAUGHTERS IN HOUSES

### Introduction

It has already been mentioned that the radon concentrations in the houses of the subjects of the left-hand half of Fig. 1 were between 4 and 26 pCi  $\ell^{-1}$ . Investigation of the house (H-0001) of subject 50-026 showed that the source of the radon was the bare soil in the unpaved crawl space beneath part of the living area. As a result we determined radon concentrations in one or two samples of air from a number of houses with unpaved crawl spaces.<sup>(4)</sup> These studies have continued and have been expanded to include some determinations of the concentrations of the short-lived radon daughters. The results are presented here.

### Methods of Measurement

Spot determinations of radon in the air of houses are made by taking "grab" samples of air into previously evacuated radon counting bottles. With background counting rates of about  $0.1 \text{ min}^{-1}$ , these have a limit of detection of approximately  $0.1 \text{ pCi } \ell^{-1}$  for a counting time of 400 minutes. Continuous determinations of radon are made with a detector of the Spitz-Wrenn design;<sup>(5)</sup> counts are integrated over a period of 30-60 minutes. The limit of detection is about  $0.3 \text{ pCi } \ell^{-1}$  for a 60-minute observation.

The short-lived radon daughter-products can be determined serially with an "Environmental Working Level Monitor" (EWLM).<sup>(6)</sup> After collection on a filter of the activity in about 100 litres of air, simultaneous alpha-particle spectrometry and scintillation beta-particle counting permit the calculation in an internal computer of the concentrations of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ . The results are printed out as pCi  $\ell^{-1}$ , and also in units of Working Level. The EWLM has a limit of detection of about  $10^{-3}$  WL, corresponding to  $0.1 \text{ pCi } \ell^{-1}$  for each of the daughter-products.

### Results

A total of 73 houses has now been investigated and a summary of the results is set out in Table 1. These data include results for 25 houses in a group of about 40 in an area of about  $0.2 \text{ km}^2$  (50 acres) which are being investigated systematically. At least one sample of air was taken from each level in the house, during the months of August to October 1980. Some houses had windows open when the samples were taken, while in others ventilation with outside air was at a minimum, so that the results are not strictly comparable. However the data suggest that the distribution of the concentrations for each level is

approximately log-normal with geometric mean values of  $1.6 \text{ pCi } \ell^{-1}$ ,  $0.5 \text{ pCi } \ell^{-1}$  and  $0.7 \text{ pCi } \ell^{-1}$  for lower level (usually basement or crawl space), first floor (entrance level) and second floor respectively. The corresponding geometric standard deviations were 2.4, 3.3 and 2.4 respectively. The limited data on radon in earth-shielded houses suggest that the average concentration may be higher than in many conventional houses.

Data on the concentrations of the daughter-products have been collected in a few of the houses showing relatively high concentrations of radon, and in some cases the radon was determined simultaneously. Results from two houses are shown in Fig. 2. From these results, and those in Table 1, it is obvious that determination of radon in a single sample of air can not give a representative value for the concentration in a house. Much more data are needed to determine the reasons for the apparent seasonal differences in the results. From the data plotted in Fig. 2 the value for the equilibrium fraction, F (ratio of observed WL to maximum possible WL), is seen to be about 0.1. This is at variance with the value of 0.5 observed by others and the reason for this is still being investigated.

### III. RADON DAUGHTERS IN VIVO

#### Introduction

Inhalation of short-lived radon daughters at concentrations of several  $\text{pCi } \ell^{-1}$  should lead to a significant lung content of the daughters. We have made measurements of the body radioactivity of subjects 50-026 and 50-150, the residents of house H-0001, to investigate the possibility of detection of such a content.

#### Procedures

On arrival at ANL each of the subjects went immediately to the underground vault, quickly changed into clean clothing and was seated in the tilted chair. For both subjects, gamma-ray counting was started less than 1 hour after they had left the house. Several consecutive 10-minute observations were followed by consecutive 20-minute ones, with one-or-two-minute intervals for data readout, for about 1.5 hours. Less frequent 20-minute observations were continued for the next 6-7 hours. Background measurements were taken at intervals during the day.

#### Results

The gamma-ray spectra from both subjects showed easily measurable levels of

$^{214}\text{Pb}$  and  $^{214}\text{Bi}$ . Counts observed above background in the energy band covering the 1.75-MeV peak were used to estimate the body content of  $^{214}\text{Bi}$ . We used the average calibration factor that is used to calculate the skeletal content of this nuclide in radium-exposed subjects. This procedure gave values for  $^{214}\text{Bi}$  in soft tissues and blood which were approximately correct, but over-estimated a possible thoracic content by a factor of 1.6.

The estimates of body content of  $^{214}\text{Bi}$  are plotted on a logarithmic scale as a function of time on a linear scale in Fig. 3. An initial rapid decline was followed by a much slower one. We interpret this as a consequence of the decay of radon daughters unsupported by  $^{222}\text{Rn}$ , followed by the decay of daughters at a rate controlled by exhalation of radon transported to the lung from a supply stored in body fat and fluids. We made the assumptions that the unsupported daughters were all in the lung, and that at the time the subjects left the house they were present at equal activities. An equation describing the behaviour of the activity of  $^{214}\text{Bi}$  in a mixture consisting initially of equal activities of the three short-lived daughters, plus a single exponential term, was fitted to the data plotted in Fig. 3, by the method of least squares.

The fitted equation is represented by the continuous curves drawn through the data points in Fig. 3, and the extrapolations back to the time of leaving the house as dotted curves. We conclude that the subjects maintain a lung content of  $^{214}\text{Bi}$  in excess of 10 nCi, and a total body content of 2-5 nCi of dissolved radon, while they are in the house. The corresponding absorbed dose-rates to the lung from the alpha-particles were calculated as between 2 and 4 rad/yr, but these have to be reduced by the appropriate occupancy factors, and possibly by other factors to allow for the average annual concentrations of radon and daughters.

Finally, we may note that scattered gamma rays from  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  were easily detected by a plutonium lung counter. The interpretation of such a response as uranium L x rays from a lung burden of plutonium, may be the explanation for some of the poor reproducibility in assaying small but significant amounts of plutonium in vivo, and for some of the variability in the determination of "subject background" in those studies (see various papers in ref. 7).

#### References

1. Martin, E. J., A.M.A. Arch. Ind. Health, 15, 506 (1957).
2. Huff, D. R., Markun, F. and Essling, M. A., Argonne Natl. Lab. Report

ANL-75-60, part II, p. 120 (1975).

3. Rundo, J., Markun, F. and Sha, J. Y., *Science*, 199, 1211 (1978).
4. Rundo, J., Markun, F. and Plondke, N. J., *Health Phys.* 36(6), 729 (1979).
5. Wrenn, M. E., Spitz, H. B. and Cohen, N., *IEEE Trans Nucl. Sci.*, NS-22(1), 645 (1975).
6. Keefe, D. J., McDowell, W. P. and Groer, P. G., U.S.E.P.A. Report No. P7628C (1978).
7. Swinth, K. L. (ed.) Battelle Pacific Northwest Laboratories Report BNWL-2088 (1976).

Table 1 Summary of results of measurements of radon in houses

	No. of houses	Level	Range of Concentration of $^{222}\text{Rn}$ , pCi $\ell^{-1}$
All houses	65	Lower	0.19 - 51.9
	44	1	0.07 - 25.9
	26	2	0.11 - 54.5
Houses with unpaved crawl spaces	26	Lower	0.34 - 35.5
	8	1	0.29 - 19.2
	2	2	0.16 - 54.5
Earth-shielded houses	6	1	1.2 - 16.1

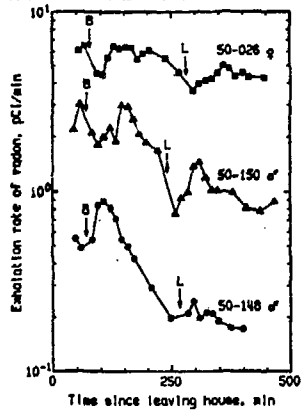


Fig. 1

Exhalation rate of radon by subjects containing no radium, showing prominent postprandial peaks. B - Breakfast; L - Lunch.

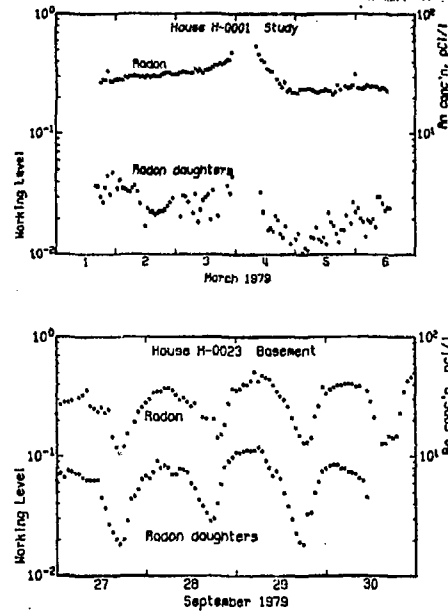
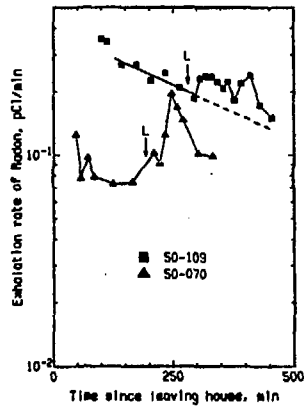


Fig. 2. Radon and radon daughters in two houses

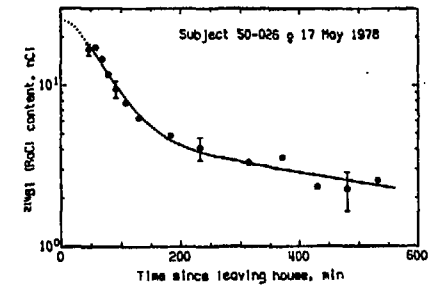
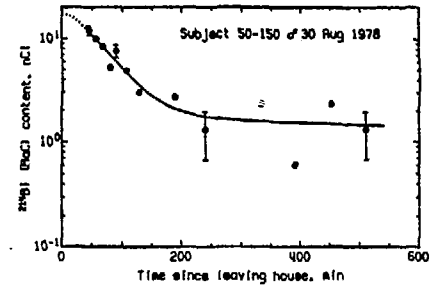


Fig. 3. Radon daughters measured in vivo