Radon and Its Daughters *in Vivo**

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Abstract - Prolonged exposure of a person to radon results in a reservoir of radon dissolved in body fat and fluids. If the subject then moves to an environment with a lower radon concentration, there is a net exhalation of radon, and the initial exhalation rate of radon depends on the radon concentration in the first environment.

This paper describes some aspects of the behaviour of radon and its short-lived daughters *in vivo* and demonstrates a relationship between the radon exhalation rate and the time after a meal. We observed a major but short-lived postprandial increase in the exhalation rate of radon produced in vivo from skeletally-deposited radium and can report a similar effect in the exhalation rate of environmental radon by persons containing no radium.

Persons who live in houses with elevated concentrations of radon may contain sufficient activity for it to be detectable by external gamma-ray counting. Some of the activity observed is due to inhaled daughter-products in the chest, and some to daughter-products associated with and produced by the decay of radon throughout the body.

It should be noted that a large amount of radon daughters in the chest will interfere in the investigation of possible internal contamination with plutonium or other actinides by external counting.

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Introduction

The retention of radon following inhalation has been reported as following a 5-component exponential function of time[1,2]. The half-times of the components range from about a minute or less, for a compartment considered to represent the lung, up to about 24 hours for a compartment thought to represent body fat. Consequently exposure of a person for a prolonged period to radon results in a reservoir of radon dissolved in the body, particularly in the fat. This reservoir only reaches equilibrium after several half-lives, i.e. several days. If the person then moves from an environment with a high radon concentration to one with a lower radon concentration, he (or she) will start to exhale the stored radon. The exhalation would proceed at a steadily decreasing rate as the reservoir depletes, but the initial exhalation rate should depend on the radon concentration to which the subject was exposed. That this is so is seen from the data plotted in Fig. 1, which shows the initial (i.e. on arrival at our laboratory, perhaps as much as 50 minutes after leaving home) exhalation rate of radon in pCi min^{-1} as a function of the radon concentration in pCi $litre^{-1}$ to which the subject was exposed in his/her house. When the breath was being collected, the subjects were breathing radon-free air. Several points in Fig. 1 are representative of typical low values (between 1 and 2 pCi $litre^{-1}$) of radon concentration in houses.

Postprandial Changes in Exhalation Rates of Radon

Some years ago in the course of our studies of all aspects of radium metabolism and toxicity, we discovered that the exhalation rate of radon produced from skeletally-deposited radium increases after a meal, and in fact doubles. Some results for seven persons are shown in Fig. 2[3]. One would expect the rate of exhalation of radon produced *in vivo* to be reasonably constant, and certainly not to decrease with time as in the case of radon inhaled from the environment, but for the subjects of Fig. 2 the radon exhalation rate decreased over the course of about two hours. For two of the seven subjects an increase is seen before the decrease; from a consideration of all the data available we can say categorically that there was an increase in the exhalation rate of radon by a factor of about two immediately after the meal (breakfast or lunch) followed by a decrease to "normal" levels. If the change in the blood flow during the process of digestion was related to this phenomenon, there might be a relationship between the exhalation rate of radon and the pulse.

The remarkable data in Fig. 3 show that in this case there is a very strong correlation between the exhalation rate of radon and pulse, in measurements made after breakfast. A reservoir of radon produced in the body must have existed; the increase in the blood flow in the torso during the digestive process flushed it into the blood, which carried it to the lung whence it was exhaled.

The question immediately arose, where is the reservoir? Also does the same reservoir exist in someone whose only source of radon is the air he or she breathes? We have looked for a similar effect in the exhalation rate of

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radon by persons containing no radium, and Fig. 4 shows the exhalation rate of radon by three persons as a function of time since leaving their houses. These are the three subjects for whom data are plotted in Fig. 1, and whose houses contained radon concentrations in excess of 4 pCi litre^{-1}. They came to Argonne without having eaten breakfast, which they brought with them. Two measurements were made, then the subjects ate their breakfast. It can be seen that, although the details are different in the three cases, each one shows an increase in the radon exhalation rate after breakfast, and a smaller one after lunch. Of course, it could be argued that the same thing would happen whether the subjects had breakfast or not before coming to Argonne, but what would happen if they had no breakfast at all? Would the increase be delayed until after lunch? Or would it not be demonstrable?

To investigate this, we tested two persons who ate no breakfast, with the results presented in Fig. 5. It can be seen that the behaviour is different for the two subjects, but both show the same general feature - an increase after lunch. It is clear that the flushing of the radon reservoir can indeed be delayed, but it is also clear that even after two meals the body still contains radon because of the continued exhalation shown in Fig. 4. The next questions that arise concern the detectability of radon and/or its daughter-products by means other than measurements in the exhaled breath. Is the body's content of radon large enough to be detected by external measurements of gamma radiation from the short-lived decay products of radon? Is there an even bigger deposit of those decay products (unsupported by radon) in the lung?

External Counting of Radon Daughters

Fig. 6 shows spectra taken with a large sodium-iodide gamma-ray spectrometer. The lower curve is the background spectrum, while the upper one is the gamma-ray spectrum observed when the detector was presented to the chest of a subject. (This is the subject whose house contained radon at the unusually high concentration of 26 pCi litre^{-1}. It should be noted that such houses are the exception rather than the rule.) In Fig. 6 it can be seen that the principal peak is at 1.46 MeV due to ^{40}K, but there are peaks due to radon daughter-products, especially at 0.61 MeV, 1.1 MeV and 1.76 MeV, all associated with the decay of ^{214}Bi (RaC). A series of measurements was made over a period of time. The counting rates above background in the peak at 1.76 MeV were recorded for two detectors, one in front of the chest, the other behind it.

Fig. 7 shows the decay curves we obtained in this fashion. Initially the counting rate for the detector behind the back was about twenty percent higher than that for the detector in front of the chest, but later on the two counting rates were in essential agreement. In each set of data the curve drawn through the points represents the readily predictable decay of short-lived radon daughter-products plus an exponential function. We interpret this as follows: each detector was first observing the decay of unsupported short-lived daughter-products of radon deposited in the thorax. When this was complete (~200 minutes) the detector was responding to short-lived daughter-products in equilibrium with and supported by radon dissolved in body fat and fluids and declining only slowly but exponentially as a result of exhalation of the radon. The counting rates corresponded to several nCi of unsupported radon daughters in the chest plus a few nCi of radon dissolved in the body.

The dose equivalent rates to lung tissue due to the unsupported daughters, assumed to be distributed uniformly throughout the lung, amounted to several rem per year (QF=20). Actual annual dose equivalents would probably be lower because the occupancy factor would be less than 1.0 and the indoor radon concentration (averaged over a period as long as a year) might very well be less than during the period of exposure of the subject just prior to our measurements. Nevertheless, it is seen that in some houses radon can result in substantial doses to the lung. What we do not know yet is how many people in the population are being irradiated at this kind of level.

Finally, it should be mentioned that these findings may be of considerable importance in radiological protection measurements of workers with potential exposure to transuranic elements such as plutonium which are commonly only detectable by virtue of the emission of low-energy photons (such as characteristic L x rays) at low intensities. The presence of short-lived radon daughters in the thorax may influence the response of a detector being used in the investigation of possible internal contamination with isotopes of plutonium or other heavy elements by external counting.

Fig. 8 shows how this is possible. It shows counting-rates in a rather broad energy band (1-95 keV) as a function of time for a xenon-filled proportional counter presented to the chest as if we were testing for inhaled plutonium in the subject (a woman). The counting rates 35-40 minutes after she left her home corresponded to about 10 times the maximum permissible lung burden of plutonium, i.e. about 160 nCi of ^{239}Pu. The half-life for the decay of that counting rate was about 46 minutes which is what is typically observed when the activity of radon daughters is being measured. This woman washed her hair before she left her home that morning, then dried it with a hot hair dryer. This is an efficient way to collect (by electrostatic attraction) radon daughters in the hair. Her hair was spread out behind her head on the bed and the proportional counter was presented to it. The counting rates observed decreased much more rapidly than that from the chest, for reasons which are not immediately obvious. I will defer to the discussion an explanation of the difference. That concludes this presentation.

References

[1] J. H. Harley, E. Jetter and N. Nelson, USAEC Report HASL-32, New York Operations Office, 1958.

[2] H. F. Lucas and F. Markun, Argonne National Laboratory Report ANL-7960, Part II, pp.136-140, 1972.

[3] J. Rundo, F. Markun and J. Y. Sha, Science, 199, 1211 (1978)

Captions for diagrams

Fig.1 The radon exhalation rates immediately after arrival at Argonne for seven subjects as a function of the radon concentrations in their homes.

Fig.2 The radon exhalation rates for seven subjects with long-standing skeletal deposits of radium, plotted as a function of time since breakfast or lunch. A clear postprandial effect is seen. Numbers by each set of data are the subject identification number (03-571, etc), the ages of the subjects at the time of these measurements and the year of first exposure to radium. The statistical errors of counting are small; errobars (± 1 S.E.) would be about the same size as the symbols.

Fig.3 Showing the correlation between pulse and radon exhalation rate after breakfast for a 71-year old female former radium dial painter first exposed to radium in 1922.

Fig.4 The rates of exhalation of environmental radon for the three subjects exposed to the highest levels (>4 pCi litre^{-1}) plotted in Fig.1, showing pronounced postprandial increases after breakfast (B) and less pronounced ones after lunch (L), instead of monotonically declining rates. Errorbars would again be about the same height as the symbols.

Fig.5 Demonstrating a postprandial effect in the exhalation rate of environmental radon for each of two subjects who ate no breakfast. L is the mid-point of the lunch period.

Fig.6 Background spectrum (lower curve) and spectrum observed from the chest of a subject 35 minutes after leaving her house which contained radon at a concentration of about 30 pCi litre^{-1}.

Fig.7 Decay of counting-rate in the 1.76-MeV peak for each of two detectors, placed above and below the chest of the subject of Fig.6. Each curve represents the decay of a mixture consisting initially of equal activities of ^{218}Po, ^{214}Pb, and ^{214}Bi (RaA, RaB and RaC), plus a single exponential decay. See text for explanation.

Fig.8 Decay of the counting rates observed in a broad energy band (1-95 keV) with a xenon/methane-filled proportional counter presented alternately to the chest (square symbols) and to the hair (triangular symbols) of the subject of Fig. 6. A half-life of 46 minutes is typical for the decay of a mixture of short-lived radon daughter-products, while 26 minutes is the half-life of ^{214}Pb (RaB).

Discussion

Question: Where is the reservoir of radon in the body?

Answer: Somewhere in body fat and fluids. I cannot be more precise, although the omentum is a distinct possibility.

Question: Why did the counting rates from the hair decrease more rapidly than those from the chest?

Answer: Because the detector was responding to different radiations in the two cases. When it presented to the chest, it responded to scattered and degraded radiation from x and gamma rays from both the RaB (^{214}Pb) and RaC (^{214}Bi), for which a typical half-life is about 45 minutes. There is essentially no scattering in the hair because its mass is so small, so the detector only responded to the L x rays of 26-minute ^{214}Pb, and not at all to the much higher energy K x rays and gamma rays from both this nuclide and its daughter ^{214}Bi.

Question: Would you say something about the behaviour of thoron daughters in vivo?

Answer: When thoron is inhaled, its short half-life means that its metabolism and solubility are in fact unimportant. The half-life of its granddaughter ^{212}Pb (ThB) is 10.6 hours, so what happens to inhaled thoron is really what happens to ^{212}Pb. When thoron is produced in the body, some may be exhaled, depending on the situation. Persons who may exhale thoron might be thorium workers who have internal deposits of thorium in the lung, or radium workers who have been exposed to ^{228}Ra which is deposited in the skeleton. For the latter the amount of thoron which is exhaled is a small fraction of the total produced because its half-life is short, but more may be exhaled if the site of production is the lung than if it is the skeleton, as in the case of the former group.

Question: Is there a similar effect in the exhalation rate of thoron after a meal?

Answer: No. Because the half-life is so short there is no possibility of building up a reservoir.

Question: There are some literature reports of higher lung cancer incidence for people who live in stone houses. Do you believe this?

Answer: If epidemiological studies of lung cancer related to type of residence have demonstrated an increased incidence in people who live in stone houses, certainly I believe them, although I am not familiar with such reports. However, the houses we have been investigating in the Chicago area in the U.S.A. are all of the wood-frame type. I think stone houses are very uncommon in the U.S.A., but house design and techniques of construction vary so much from one country to another that it would be both difficult and dangerous to generalize.

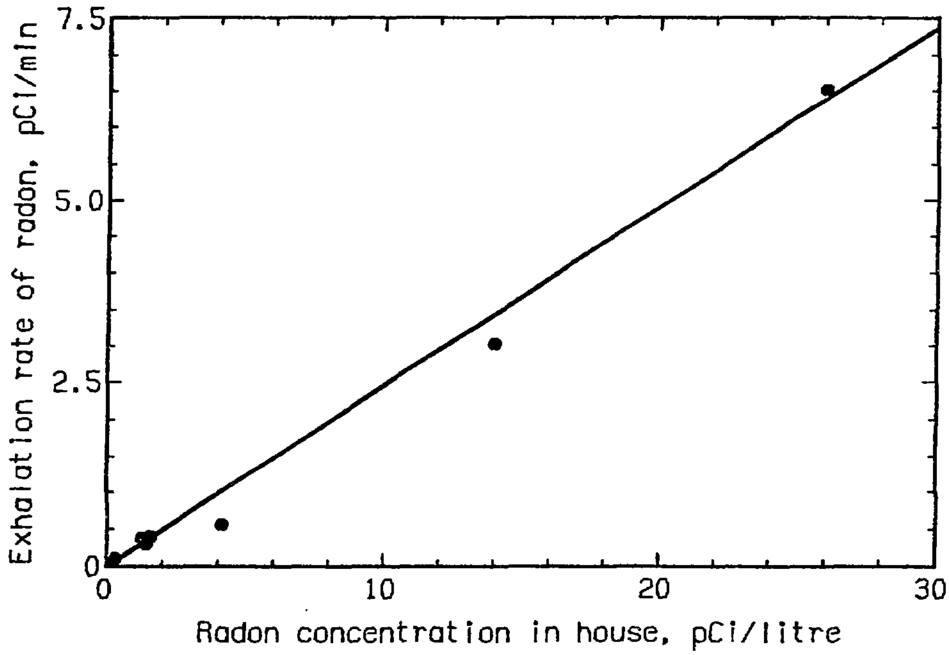
7.5
5.0
2.5
0
0
10
20
30
Exhalation rate of radon, pCi/min
Radon concentration in house, pCi/litre

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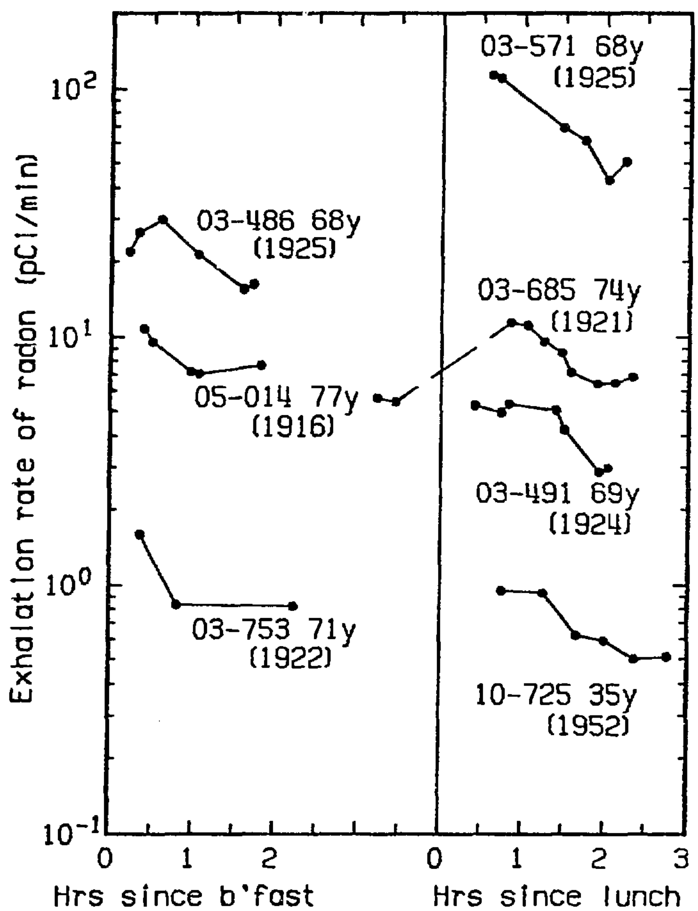
Fig. 1



Exhalation rate of radon (pCi/min)
10^2
10^1
10^0
10^{-1}
03-571 68y (1925)
03-486 68y (1925)
03-685 74y (1921)
05-014 77y (1916)
03-491 69y (1924)
03-753 71y (1922)
10-725 35y (1952)
0 1 2
0 1 2 3
Hrs since b'fast
Hrs since lunch

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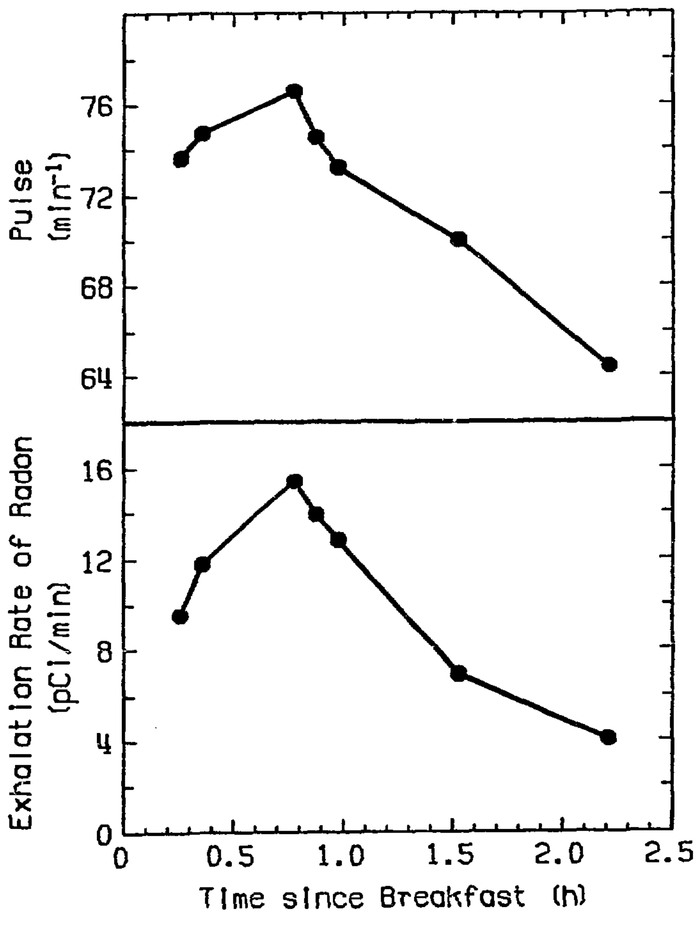
Fig. 2



Pulse
(min⁻¹)
Exhalation Rate of Radon
(pCi/min)
Time since Breakfast (h)

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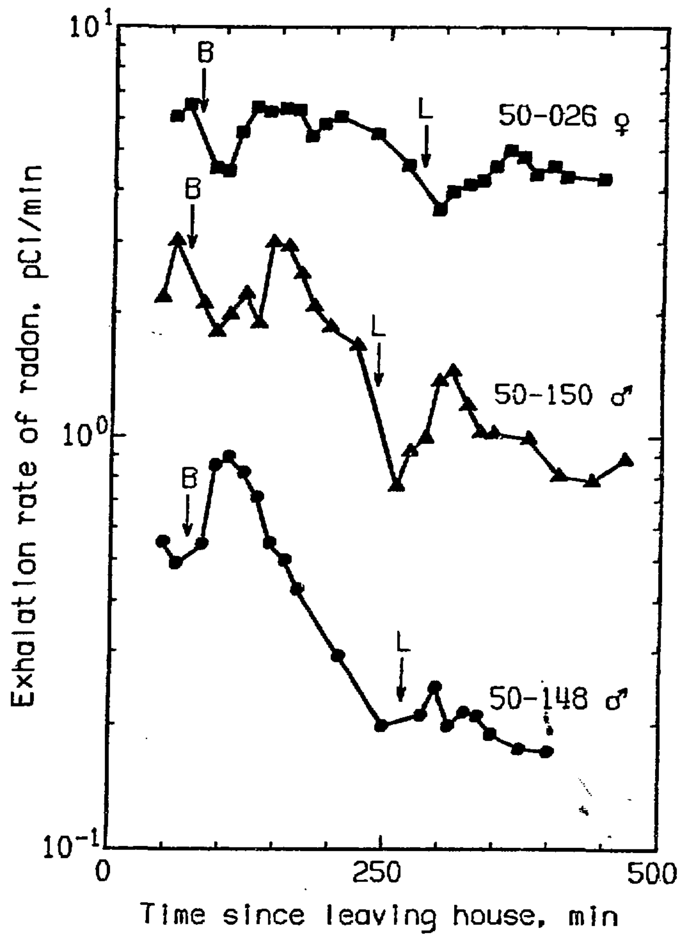
Fig. 3



Exhalation rate of radon, pCi/min
10^1
10^0
10^-1
0
250
500
Time since leaving house, min
B
L
50-026 ♀
50-150 ♂
50-148 ♂

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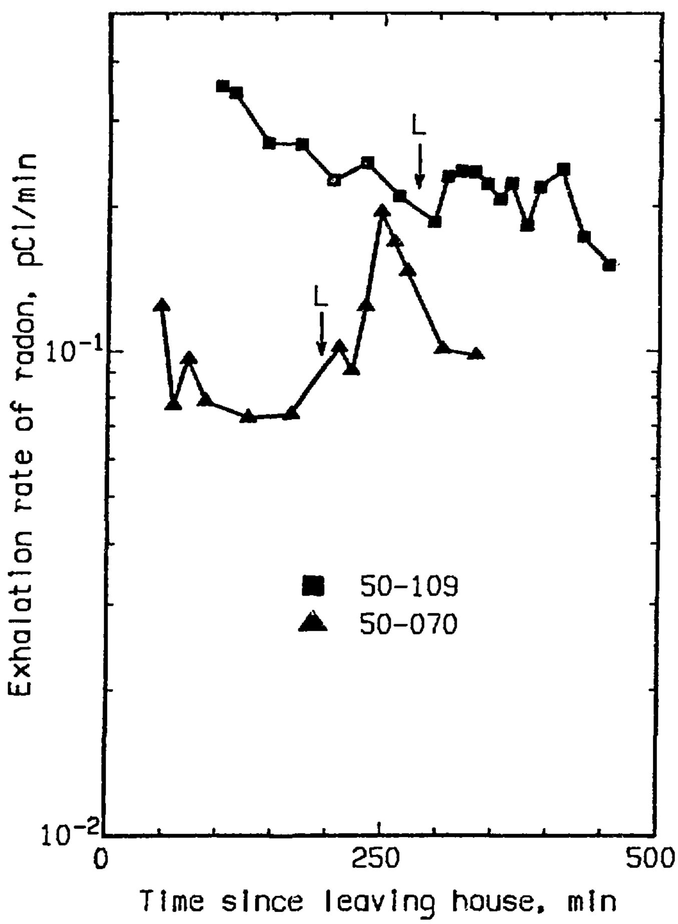
Fig. 4



Exhalation rate of radon, pCi/min
10^{-1}
10^{-2}
0
250
500
Time since leaving house, min
L
L
50-109
50-070

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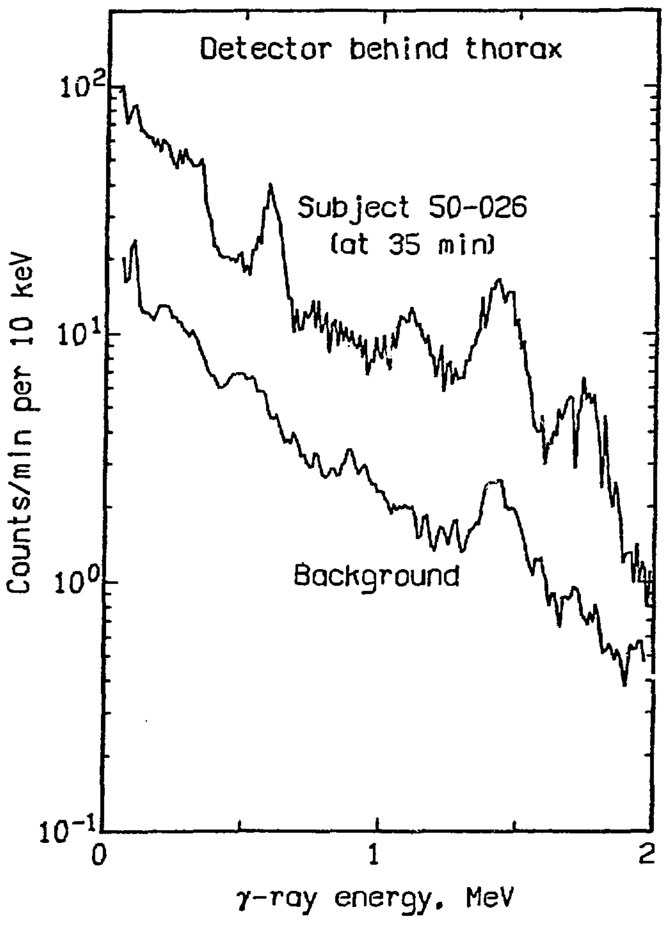
Fig. 5



Detector behind thorax
Subject 50-026
(at 35 min)
Background
10^2
10^1
10^0
10^{-1}
Counts/min per 10 keV
0
1
2
γ-ray energy, MeV

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Fig. 6

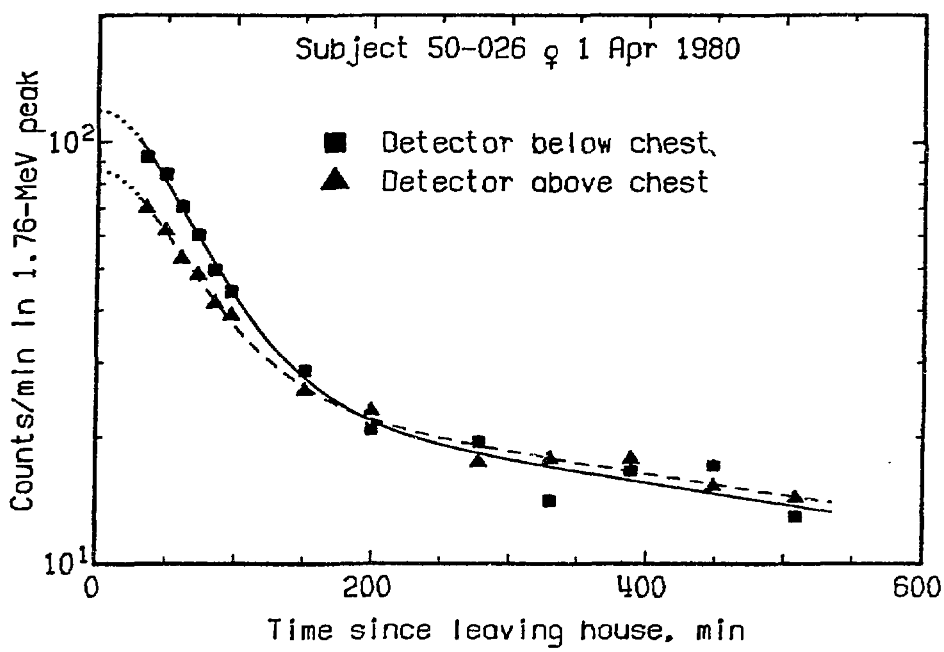
Subject 50-026 ♀ 1 Apr 1980
Detector below chest
Detector above chest
Counts/min in 1.76-MeV peak
10²
10¹
0
200
400
600
Time since leaving house, min

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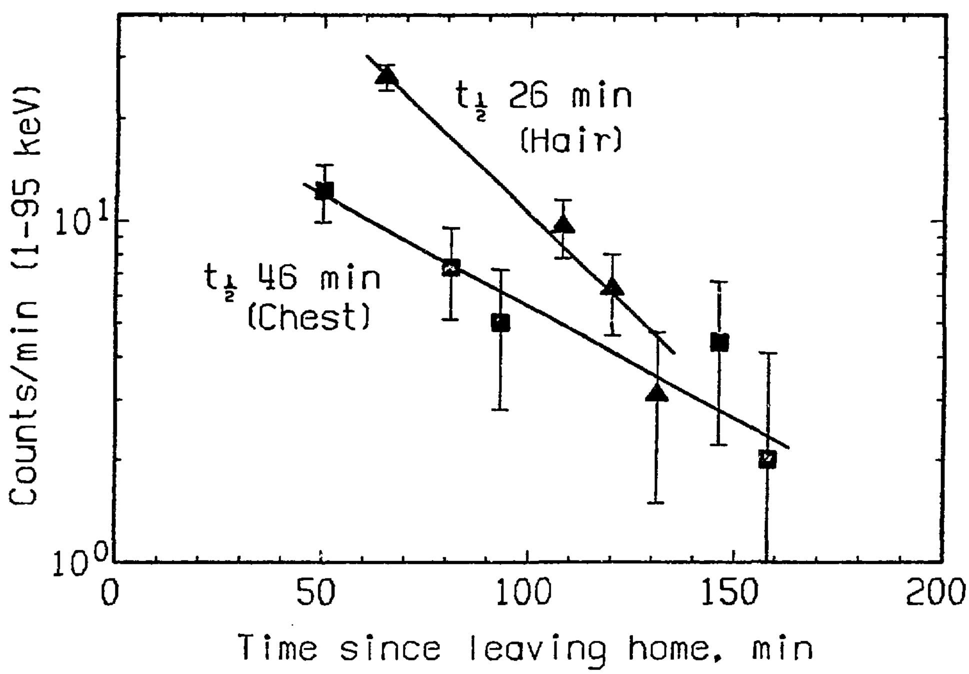
Fig. 7



t½ 26 min
(Hair)
t½ 46 min
(Chest)
Counts/min (1-95 keV)
10^1
10^0
0
50
100
150
200
Time since leaving home, min

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Fig. 8