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WHOLE BODY COUNTING OF RADON DAUGHTERS

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ABSTRACT

Five adult males were exposed for one hour to radon and radon daughter products in an exposure chamber and were subsequently measured for radon daughter product activity in the chest region by whole body counting methods. The gamma-ray detection rate appeared to diminish as a single exponential with 35 minute half period, a form that is consistent with the physical decay of a mixture of RaB and RaC. About half of the deposited activity was associated with internal deposition and half with external deposition on clothing, skin and hair. The average counting rate from radon daughters on clothing was 10 times the average from skin and hair. These two components together contributed 51.2% of the total counting rate. A substantial contribution was made to the counting rate from deposits on under as well as outer clothing. A strong but statistically non significant correlation was found between internal and external deposition indicating that total activity provides an index of internal deposition.

INTRODUCTION

The quantitative uptake of radon daughter products by the lung is normally estimated from mathematical models of aerosol and unattached daughter product deposition on the mucous membranes of the bronchial tree and alveolar sacs. However, the technology and methods for experimental observation of radon daughter uptake have long existed, due to interest in the health effects of radium deposition in the body. Both ^{222}Rn and ^{226}Ra give rise to gamma-ray emitting decay chains of radon daughter products whose emissions can be easily detected by standard radiation detectors. The earliest applications of this method for radon daughter product detection employed electroscopes, and later Geiger-Muller tubes, without spectral resolution. Current applications are based on sodium iodide and semiconductor detectors to achieve moderate or high spectral resolution, respectively, of the emitted gamma-rays, and permit the separate measurement of RaB and RaC. An excellent review of the history of radium, i.e. radon daughter product, detection in the body and of the current use of sodium iodide systems has been published by Toohey et al. (1).

Our laboratory has employed whole body counting methods for radon daughter detection for several decades while investigating radium and radon gas retention in humans. The systems in use by us (1) are stationary and mobile whole body counters that employ thallium activated sodium-iodide crystals (designated NaI(Tl)) of large size, in the shape of right circular cylinders. The stationary whole body counters are subterranean and shielded by concrete, steel, and lead to reduce terrestrial gamma-ray and cosmic-ray background, and are located in rooms supplied with radon-free air to reduce background from airborne radon daughter products. Our mobile detector is contained in a trailer which can be hauled to field locations. Gamma-ray backgrounds and background variability are higher in the mobile counter than in the stationary counters due to the use of thinner shielding and untreated air in the mobile counter.

During the course of field investigations of gamma-ray emission from former radium workers in Bloomsburg, Pennsylvania, we observed easily measurable levels of radon daughter products in the bodies of some subjects (2). The radon daughters were first attributed to radium retained from occupational exposure but later identified as radon daughters directly deposited from indoor air or as the daughters of radon dissolved in body tissues from breathing indoor air. An immediate question for the interpretation of these data, many of which were collected with the subjects in their street clothes, was whether or not the detected gamma rays were due primarily to radon daughters on the clothing, body surfaces and hair, or to radon daughters in the body. A corollary was whether or not total daughter product activity was correlated with internally deposited activity. If so, total activity would be an index of internally deposited activity and could be used as an index of lung deposition. This question has been investigated by experimental exposures to radon and radon daughters in an exposure chamber. The initial work in this series of experiments is reported in this paper.

EXPERIMENTAL DESIGN AND METHODS

The subjects in this study were exposed to radon and radon daughter products and measured for daughter product activity in our stationary whole body counter in order to determine the time course of daughter product retention. The exposure chamber was a defunct walk-in freezer that had been converted to use for the calibration of radon daughter and radon gas measurement instruments. The chamber's interior dimensions are large enough to comfortably accommodate a chair, measurement equipment, and a radon source, with space to spare. Ventilation is by air seepage through the joints of the structure. Chilled water flow through the heat exchanger of the old refrigeration unit is used for temperature control. The bones of people with elevated occupational exposure to radium were used as the source of radon gas for the chamber. The radon emanation percentage from bone *in vitro* is generally several percent to several tens of percent.

There have been three phases to this work. In the first phase we determined the distribution of radon daughter products between the exterior and interior of the body during and after exposure in street clothes. In the second phase we established a calibration factor for the quantitative determination of radon daughter deposition in the chest. In the third phase we compared the results of combined internal and external exposure, pure internal exposure, and pure external exposure in males and females.

The first phase, which is the subject of this paper, involved the exposures of five males, all associated through their employment at Argonne, with this or related projects. The subjects were middle-aged non smokers of average weight and height (Table 1). Following a measurement in the whole body counter to establish baseline whole body radioactivity, the subjects sat for one hour in the exposure chamber, dressed in street clothes, and breathed exposure room air. The chamber air temperatures were in the mid to high 20's, centigrade, and relative humidities were 40 to 60%. Working level values, measured by an EDA Instruments model RDA-400 continuous monitor, set for 10 minute data collection intervals, are presented in Table 1. Working level ratios, based on data taken earlier, were probably in the range 0.1 - 0.3 during the exposure periods.

After exposure, the subjects were measured in the whole body counter while lying supine with a NaI(Tl) crystal 10 cm high and 29 cm diameter positioned over their chests to measure the gamma-ray emission from the upper body. Subjects then changed to pajamas, had two more whole body measurements, took a shower and shampoo and had another two whole body measurements. In total, six whole body measurements were made on each subject, one for baseline, one in street clothes after exposure, two in clean clothes before shower and shampoo, and two in clean clothes after shower and shampoo. The protocol with timing is given in Table 2. These measurements permitted the estimation of the distribution of radioactivity between clothes, unwashed skin and hair, and body interior, and washed skin and hair, and body interior. Throughout this work, we have assumed that washing removes all residual radon daughter products from the skin and hair.

Table 1. Age, weight, height, and working level exposure of subjects.

ID	Age (y)	Weight (kg)	Height (cm)	RnD (WL)
50-032	45	90	184	0.18
50-040	61	91	184	0.22
50-109	46	89	173	0.14
50-128	60	76	170	0.17
50-166	53	83	183	0.20

Table 2. Exposure and measurement protocol.

Elapsed Time (min)	Allotted Time (min)	Activity
0	10	Don clean pajamas
10	30	First (baseline) whole body measurement
40	10	Don street clothes
50	60	Exposure to radon and radon daughters
110	10	Set up whole body measurement
120	5	Second whole body measurement (in radon daughter contaminated street clothing)
125	10	Don clean pajamas
135	15	Third whole body measurement
150	2	Reset instruments
152	15	Fourth whole body measurement
167	20	Shower, shampoo, and don clean pajamas
187	15	Fifth whole body measurement
202	2	Reset instruments
204	15	Sixth whole body measurement
209	10	Don street clothes

GAMMA-RAY DATA

For illustration, the gamma-ray spectrum of radon daughter products is shown in Figure 1. Some gamma-ray spectrum peaks originating from the decay of RaB or RaC are identified. In general, the data between the left-hand border of the graph and the RaB marker are mostly from RaB, with contributions from the Compton scattering of RaC gamma rays.

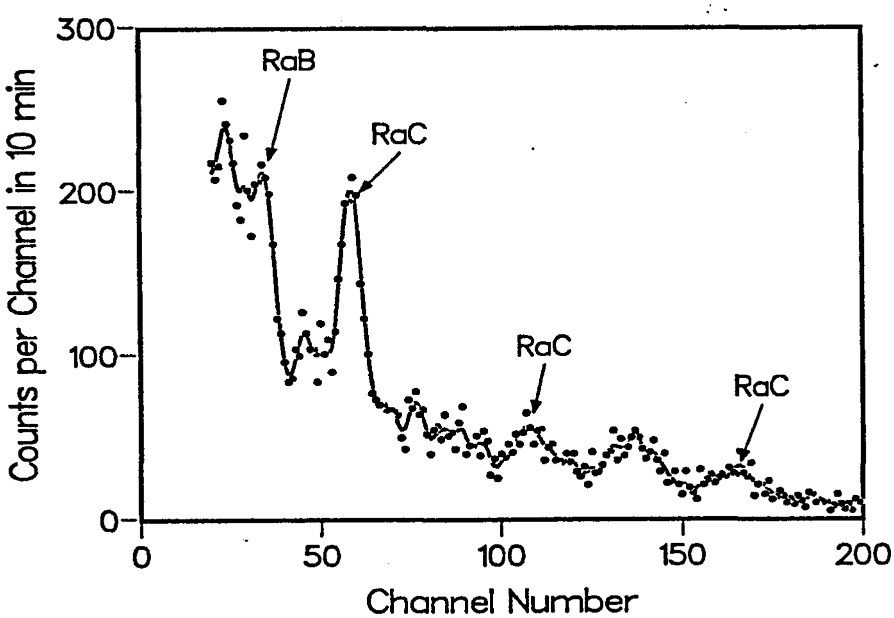

Figure 1. Gamma-ray spectrum of radon daughter products with the identification of some peaks associated with RaB and RaC decay.

The sum of all data in the gamma-ray spectrum for energies above 200 keV provides a simple measure of radon daughter product activity and was used, after conversion to counting rate and adjustment for baseline radioactivity, as the variable for analysis of the data from this experiment. The data for three subjects are presented in Table 3 and the data for the remaining two subjects appear in Figure 2.

The decline of radon daughter counting rate in Table 3 and Figure 2 is due to clothes change, shower and radioactive decay. The lines through the data in the figure represent a single exponential with 35 min half life, and help when judging how much of the decline is due to each source.

Table 3. Net radon daughter counting rates over the chests of three subjects.

Subject	Condition	Time Since Exposure (min)	Rate (min^{-1})
50-040	Contaminated clothing	12.5	2650
	After clothes change	32.5	1060
	After clothes change	49.5	768
	After shower	84.5	335
	After shower	101.5	266
50-109	Contaminated clothing	12.5	3750
	After clothes change	32.5	1200
	After clothes change	49.5	919
	After shower	84.5	410
	After shower	101.5	288
50-166	Contaminated clothing	12.5	3560
	After clothes change	32.5	1391
	After clothes change	49.5	1000
	After shower	84.5	429
	After shower	101.5	316

ANALYSIS AND RESULTS

The difference between the counting rate with contaminated clothing and the counting rate represented by the exponential with 35 minute half life provides an estimate of the portion of the rate due to radon daughters on the clothing. The balance of the radon daughters for the counting period centered at 12.5 min post exposure are deposited on the skin surface or in the body. Similarly, the difference between the counting rates after shower and shampoo and the rates represented by the exponential are due to the removal of skin and hair contamination. The radon daughter counting rates after shower and shampoo are due solely to radon daughters in the body.

The difference between the exponential line and the observed counting rate is an estimate of the radon daughters associated with clothing. As an example, the observed counting rate, with subject 50-032 dressed in contaminated clothing, was 1920 min^{-1} and the counting rate represented by the exponential at 12.5 min was 985 min^{-1}. Therefore, 100%(1920 - 985)/1920 = 48.6% of the observed activity was due to radon daughters deposited on the clothing, and 51.4% was due to radon daughters in the body and on skin and hair. The observed counting rates after shower and shampoo were 210 and 171 min^{-1} at 84.5 and 101.5 minutes respectively. The counting rates represented by the exponential line were 237 and 169 min^{-1} at those same times respectively. The percentage difference at 84.5 minutes is 100%(237 - 210)/237 = 11.4%. Using the same formula, the difference at 101.5 minutes is -1.2%, and the average for the two times is 5.1%. Re-expressed as a percentage of the total counting rate at 12.5 minute, this average becomes 5.1%(51.4%)/100% = 2.6%.

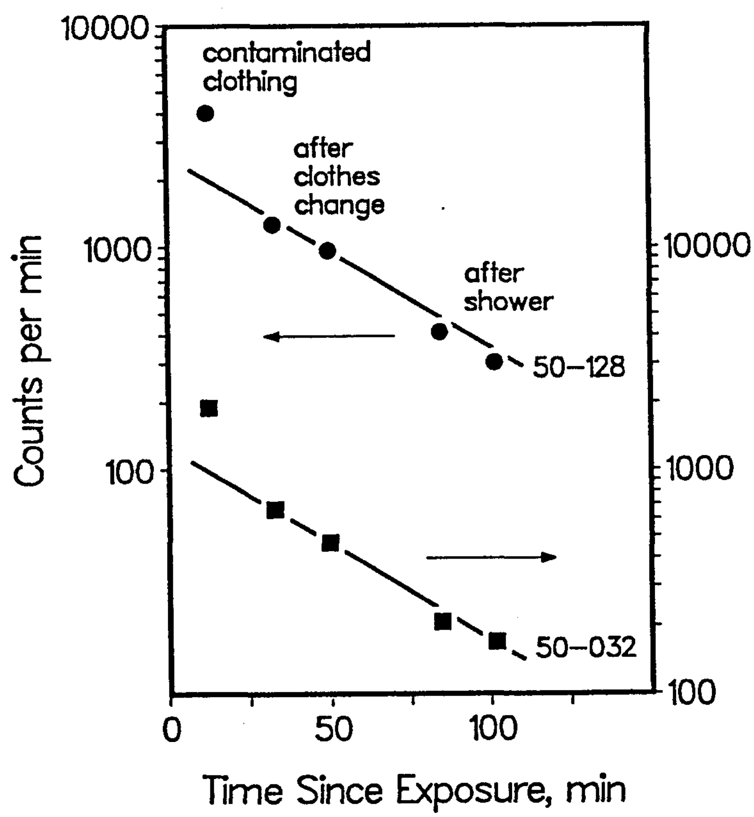

Figure 2. Gamma-ray counting rate for two subjects as a function of time since the end of exposure. The left-hand ordinate scale applies to 50-128, the right-hand scale to 50-032.

Percentages for the deposition in various compartments are presented in Table 4 for all subjects. The percentages for skin/hair are not statistically significant but the fact that all are positive lends support to the notion that there is a removable radon daughter deposit associated with skin and hair. Percentages for the body were obtained as the difference, e.g. the percentage in the body of subject 50-032 is 100% - (48.6% + 2.6%) = 48.8%.

Table 4. Percentage of radon daughter counting rate due to deposition on street clothing, on skin and hair, and in the body.

Subject	Clothing	Skin/Hair	Body
50-032	48.6	2.6	48.8
50-040	40.1	4.3	55.6
50-109	50.6	4.2	45.2
50-128	51.7	5.1	43.2
50-166	41.6	7.4	51.0

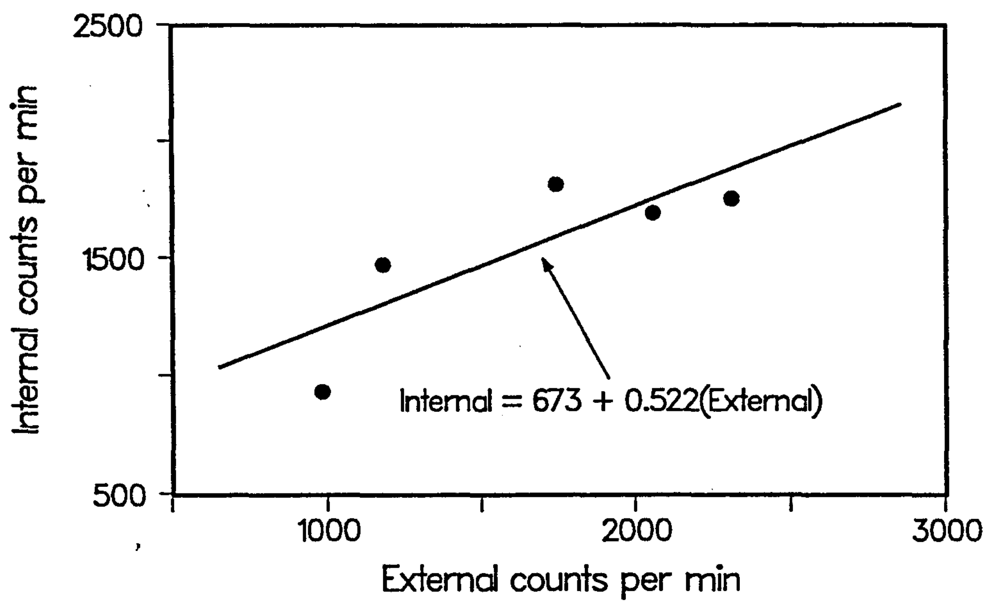

Figure 3. Counting rates from internal and external deposition of radon daughter products. The plot and equation of a regression line fitted to the data are shown.

A scatter diagram of the internal (body) and external (clothing plus skin) counting rates, estimated with the aid of percentages given in Table 4, is presented in Figure 3. The slope of the regression line is not significant at the 5% level ($p = 0.089$) but the percentages in Table 4 may be biased estimates that render tests of significance invalid.

DISCUSSION

Street clothing accounts for a substantial fraction of the radon daughter activity associated with the body, based on the estimation procedure outlined above, which relies on a single exponential with 35 min half life for the back extrapolation of data. To test the plausibility of the extrapolation model, we fitted a sum of two exponentials, representing contributions from RaB and RaC, to a single exponential with 35 minute half life, i.e. the equation,

$$A(t) = b_o e^{-k_B t} + c_o e^{-k_C t} \qquad (1)$$

was fitted, with nonlinear regression techniques, to:

$$O(t) = e^{-t(ln2)/35} \qquad (2)$$

for times between 12.5 and 101.5 minutes; k_B and k_C are the decay constants for RaB and RaC, 0.0259 min^{-1} and 0.0352 min^{-1}, respectively. The fitted equation ($A(t)$) with $b_o = 2.141$ and $c_o = -1.223$ and the single exponential ($O(t)$) deviate by no more than 11% with an average absolute deviation of 2.9% and an average deviation of 1.8% Thus, a single exponential provides a plausible empirical representation of the gamma-ray emission from a RaB - RaC mixture whose activity is not significantly diminished by biological processes. The negative sign of c_o is consistent with an initial RaC activity that is less than the value at secular equilibrium. For clarification, the following equation gives the gamma-ray counting rate, $R(t)$, as a function of time after the RaA activity (3.05 min half life) has fallen to negligible levels. This most likely occurred before the subjects entered the whole body counter for the first time. The factors, f_B and f_C, are calibration constants that relate the counting rates for RaB and RaC to the activities associated with the subject. The counting rate has been normalized to B_o, the initial activity of RaB.

$$R(t)/B_o = (f_B + 3.77 f_C) e^{-0.0259t} + (C_o/B_o - 3.77) f_C e^{-0.0352t} \qquad (3)$$

Secular equilibrium occurs when the ratio of initial activities, C_o/B_o, is 3.77. A smaller ratio causes the coefficient of the second exponential term to be negative, verifying our interpretation of the negative sign of c_o.

During the course of this work we made direct measurements of clothing from the exposure chamber to verify radon daughter uptake and to supplement our inferences of substantial deposition on clothing based on observations of a reduction in counting rate after a clothes change. One of the most interesting aspects of these exploratory measurements was the observation of significant daughter product deposition in underclothing, following an exposure in loose fitting garments. The counting rates for gamma rays in the 609 keV peak of RaC were analyzed using the extrapolation techniques described above. We found that two-thirds of the 609 keV gamma rays associated with clothing originated in the underclothing. Although these experiments were not repeated, this one result suggests an important role for underclothing in radon daughter uptake. During field studies in which subjects are asked to change their clothes it is insufficient to remove only the outer clothing. Underclothing must also be removed to reduce external contamination to low levels.

The correlation apparent in Figure 3 provides another observation important for field studies. The linear trend in the data, emphasized by the regression line, suggests a strong correlation between externally and internally deposited radon daughter products even though the present data set is too small to establish a statistically significant correlation. The spread of the data in Table 4 is not large, suggesting that 50% internal deposition, 50% external deposition is a good working hypothesis. The small spread also suggests that total counting rate above the chest is a good index of radon daughter deposition in the lungs. This simplifies protocols for field measurements by eliminating the requirement for a clothes change, although data collected with a clothes change are preferable to those collected without one. It also permits a confident analysis of data collected from people dressed in street clothes, when information about internal deposition is desired.

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