ANL-8060
Part II

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RADIOLOGICAL AND ENVIRONMENTAL RESEARCH DIVISION ANNUAL REPORT

Center for Human Radiobiology

July 1972–June 1973

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FOREWORD

This is the fourth Annual Report to be published by the Center for Human Radiobiology. At the end of this volume is a complete list of 1346 radium cases with detailed exposure data, including body burdens, updated to December 31, 1972. The list includes 114 cases measured only by the New Jersey Radium Research Project which were added during the past year to our "measured cases" after examination of the analysis to date. To the best of our knowledge, the list now includes all exposed radium cases measured by one or more of the three principal earlier projects (ANL/ACRH, MIT, and NJRRP) and/or CHR.

From 1 July 1972 to 30 June 1973 medical examinations and radioactivity measurements of 302 radium patients were made at the Center; 83 of these were made at our satellite laboratory in MIT and 219 at Argonne.

During the past year the Center has added to its study two individuals who received plutonium internally, one in 1945 and the other in 1947. These individuals were studied in a hospital for two-week periods with complete excreta collection and medical examinations. The results of the excreta collections will be reported in the next issue of the Annual Report. In addition, one individual internally contaminated with the element berkelium has been added to our studied patients. A report in this issue gives the retention results obtained by in vivo radioactivity measurements and excreta studies of this individual over a period of approximately one year.

The best way of expressing and evaluating the toxicity of radium in man is a matter of considerable concern to us at this time. An often used method of expressing toxicity is a dose response function or a dose response plot which shows the number of malignancies in a given dose range as a function of dose. Evans and his co-workers have published dose response plots, using those cases deemed to be epidemiologically suitable in his analysis (see pp. 157-194 in Delayed Effects of Bone-Seeking Radionuclides, Ed. C. W. Mays, et al., Univ. Utah Press, Salt Lake City, 1969;

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pp. 431-468 in Radiobiology of Plutonium, Ed. B. J. Stover and W. S. S. Jee, J. W. Press, Salt Lake City, 1972). In previous issues of this Annual Report, we have published dose response curves derived by utilizing all of the radium cases with measured body burdens whether they were epidemiologically suitable or not (cf. ANL-7760, Part II, pp. 1-17; ANL-7860, Part II, pp. 1-8).

Both of these types of analysis show the same general pattern. A relatively high number of radiogenic tumors are induced at cumulative skeletal doses greater than about 1000 rads, but no tumors are yet seen in the great number of cases lying significantly below 1000 rads. Evans has interpreted these data as indicating the existence of a practical threshold or very sharp sigmoid configuration of the dose response function, whereas we have chosen to characterize the dose response as one in which the prevalence of malignancies increases approximately with the square of the dose in the low dose region.

We are now questioning these interpretations, for we have discovered that most of the low dose cases have been added to the study since 1957 by radioactivity measurements of living persons (for details see ANL-7960, Part II, pp. 6-22). While it is true that almost all of the cases have carried their radium for many years, those cases only recently measured for the first time may be considered to be the survivors from the group of persons who received radium years ago. Therefore, we ask the question: From observations mostly of cases that survived 20 years or more after exposure, is it proper to conclude that doses below 1000 rads have not induced malignancies? Could it not be that others who received the same quantity of radium at the same time died before development of recognizable signs of radiation damage and thus were eliminated from the study?

We are now searching for a means to evaluate properly the number of patient years at risk in each of the dose levels. Since only patients with measured burdens of radium are included in analyses of response vs. dose, one definition of patient years at risk is to consider a patient at risk only after we have identified the individual and measured his or her body burden.

Thus, those years the patients carried radium before entering the study would not be considered in our analysis of risk; only after they entered the study would they be considered to be at risk.

Dose response plots which take into account the number of patient years at risk in each dose category may well result in loss of some significance for the large number of cases with low doses, for they will be seen to represent a relatively small number of patient years at risk. As a much to be preferred alternative, increased emphasis will be placed on examination of medical records of those radium cases who died with no measurement of body burden and on exhumations in order to measure radium in the remains. Successful completion of the latter program would remove the implication that survival until measurement was a necessary criterion for entry into the radium studies.

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INTERFERENCE FROM INHALED RADON IN THE MEASUREMENT OF LOW LEVELS OF RADIUM IN VIVO

F. Markun and D. R. Huff

A case study is presented which illustrates the effect of high radon levels in air on the determination of ^{226}Ra body content. Questionable ^{226}Ra measurements of some low-level radium cases are considered.

Introduction

The ^{226}Ra content of a living person is obtained from separate determinations of the retained radon and the emanating ^{226}Ra. The former is measured by external gamma-ray counting of the ^{226}Ra daughter, ^{214}Bi (RaC), and the latter is determined from the radon exhalation rate.

Control subjects are measured under the same conditions as radium patients, and the results are used as "background" corrections. However, the apparent exhaled radon fraction exhibits much larger fluctuations than the retained radon for reasons which have been discussed in detail elsewhere.[1-3] A major cause of these fluctuations is the variability of the radon concentration in the air to which subjects were exposed prior to measurement. The following study illustrates discrepancies which may occur in the calculation of the ^{226}Ra body content when this natural radon level is significantly higher than the level considered to be the normal background based upon measurements of control subjects.

Case Study

Subjects for background measurements are chosen mainly from the RER Division, and they are assigned case numbers prefixed with 50-. Case 50-025 was expected to be a suitable control subject because he had worked at ANL for only a short time and had had no contact with radioactivity other than that from natural sources. He was first measured on the afternoon of 14 February 1973, according to the normal procedure, i.e., a whole-body gamma-ray count in the tilted chair for two 20-min periods, followed immediately by the collection of radon from the subject's exhaled breath.

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There appeared to be a large discrepancy between the gamma-ray and radon breath results (Table 1, line 1). The ratio of retained radon to total radium based on those results (0.10 ± 0.06) was quite different from values of about 0.3 to 0.4 found for high-level ^{226}Ra cases 20 years or more after intake.

A background correction based on 0.27 pCi/l Rn as the estimated "average" radon level of air is applied to all calculations of emanating ^{226}Ra.[1] It was suspected that this subject had been exposed to a much higher radon concentration prior to entering the vault for the above measurements. Consequently, air samples from a few strategic locations were analyzed and the analytical data are presented in Table 2. Apparently, when the subject was at home, he was inhaling radon at a concentration some 20 times that assumed in the calculation of the body content of emanating ^{226}Ra.

The following week, case 50-025 cooperated in a one-day deemanation study. He entered the vault at 0830, the first radon breath samples were collected after he had been breathing vault air for one hour, and a whole-body gamma-ray measurement was made immediately afterward. He remained in the vault the entire day, and radon breath collections followed by a gamma-ray measurement were repeated twice during the day. Table 1 (lines 2-4) shows the decrease in his exhaled radon, and thus the equivalent decrease in emanating ^{226}Ra content, although the deemanation was not complete after 7 hours. Equilibration with the radon concentration of vault air at that rate would have taken more than 24 hours.

The retention of inhaled radon can be expressed by a sum of five exponential functions of time of breathing radon-free air, and the constants (coefficients and half-times) are given by Lucas and Markun.[1] To more fully represent the experimental situation, an expression for the radon exhalation rate as a function of time should consist of the sum of the exponential functions plus a constant term. The latter represents the exhalation rate of the radon in the air which the subject is breathing in the period during which the breath samples are being collected.

In Figure 1 the nine measurements (three groups of three) of the radon exhalation rate of subject 50-025 are shown as a function of time. The

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TABLE 1. ^{222}Rn and ^{226}Ra Measurements for Case 50-025[a]

Date	Time breathing vault air, hr	^{222}Rn exhalation rate, pCi/min	Emanating ^{226}Ra content, nCi	RaC content, nCi	Equivalent total ^{226}Ra content, nCi	Total ^{226}Ra content based on RaC only,[b] nCi
2/14/73	1	0.926 ± 0.069	7.4 ± 0.6	0.80 ± 0.49	8.2 ± 0.8	2.1 ± 1.3
2/22/73	1	0.848 ± 0.075	6.81 ± 0.60	0.60 ± 0.76	7.41 ± 0.97	1.6 ± 2.0
2/22/73	4	0.453 ± 0.024	3.64 ± 0.19	0.98 ± 0.48	4.62 ± 0.52	2.6 ± 1.3
2/22/73	7	0.353 ± 0.018	2.84 ± 0.15	0.20 ± 0.50	3.04 ± 0.52	0.5 ± 1.3

[a] Errors shown are standard deviations based on counting statistics and estimated systematic errors.

[b] Assume that total ^{226}Ra = RaC/(0.38 ± 0.06).

TABLE 2. Radon Concentration of Ambient Air Samples for Case 50-025

Date	Location	Rn concentration[a] pCi/l
2/14/73 afternoon	shower room outside vault	0.30 ± 0.07
2/14/73 afternoon	subject's office at ANL	0.11 ± 0.05
2/14/73 evening	subject's bedroom	6.9 ± 0.3
2/15/73 morning	"	5.0 ± 0.2
2/21/73 evening	"	6.3 ± 0.3
2/22/73 morning	"	4.1 ± 0.2

[a] Errors shown are standard deviations based on counting statistics and estimated systematic errors.

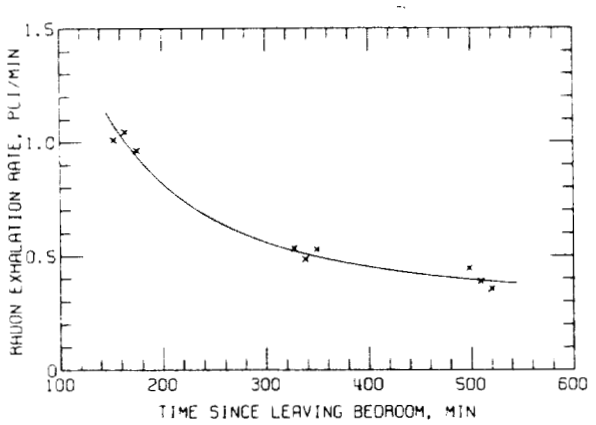

FIG. 1.--Radon exhalation rate of case 50-025 as a function of time after leaving a room containing radon at a concentration of 5.2 pCi/l. The curve represents the Lucas-Markun retention equation plus a constant (see text).

smooth curve represents the Lucas-Markun equation plus a constant, for which the intercept at zero time and the value of the constant were fitted to the data by least-squares analysis. The intercept obtained was 27 ± 3 pCi/min and the constant term was 0.24 ± 0.03 pCi/min. If the subject had been in equilibrium with the radon in the air of his room when he left it, we should expect a value for the intercept of 33.8 pCi/min from the product of the radon concentration of 5.2 pCi/l and the breathing rate of 6.5 l/min. The agreement is quite good.

Interpretation of the constant term is less obvious. The value of the constant we would predict for inhalation of the air in the underground vault, with a concentration of 0.02 pCi/l, is 0.13 pCi/min. However, the subject breathed aged tank air with a radon content of about 0.002 pCi/l for 10 min prior to and during each sampling sequence of three 10-min samples. Thus, radon from vault air would have been reduced by about a factor of four on the average for three consecutive samples. It seems most likely, then, that the constant term primarily served as an adjustable parameter to correct the standard deemanation curve applied to this subject. A similarly good fit was obtained by allowing the coefficient of the longest-lived component to vary.

Results on other control subjects on whom radon breath and/or whole-body measurements have been made are listed in Table 3. The values shown for emanating ^{226}Ra content, obtained from radon exhalation rates, were corrected for the effect of previous radon inhalation of 0.27 pCi/l. This value was determined empirically from the results of the first nine measurements listed.[1] Case 50-022 may contain a measurable amount of radium, despite no known exposure. The weighted mean of the emanating ^{226}Ra content for the other six measurements is 0.25 ± 0.36 nCi. Thus radon exhalation equivalent to the high emanating ^{226}Ra found for case 50-025 is atypical.

The RaC results in Table 3 were obtained after subtraction of background counting rates observed in the absence of human subjects and phantoms. The residual values shown are not significant in individual cases,

TABLE 3. Measurements of ^{226}Ra in Unexposed Subjects[a]

Case No.	Emanating ^{226}Ra content, nCi	RaC content, nCi
50-001	0.33 ± 0.56	0.66 ± 0.47
003	0.94 ± 0.38	1.0 ± 0.50
007	-0.13 ± 0.48	-0.42 ± 0.60
008	0.17 ± 0.42	0.78 ± 0.50
009	0.19 ± 0.66	-0.17 ± 0.50
010	-0.38 ± 0.45	0.07 ± 0.48
011	-0.48 ± 0.47	0.31 ± 0.51
012	-0.34 ± 0.49	0.99 ± 0.57
013	-0.39 ± 0.50	-0.30 ± 0.46
014	---	0.81 ± 0.67
015	-0.26 ± 0.51	0.54 ± 0.76
016	-1.30 ± 0.90	0.60 ± 0.47
017	1.01 ± 0.55	0.58 ± 0.49
018	-0.04 ± 0.50	-0.75 ± 0.62
019	---	0.87 ± 0.47
020	-0.84 ± 0.72	-0.22 ± 0.47
021	1.15 ± 0.44	0.47 ± 0.49
022	2.08 ± 0.44	1.52 ± 0.55
Mean[b]	0.11 ± 0.13	0.35 ± 0.13

[a] Errors shown are standard deviations based on counting statistics and estimated systematic errors.

[b] Excluding case 50-022.

but the positive value for the average indicates the presence of additional "background" of unknown origin when subjects are being counted.

Relevance to the Radium Studies

The above control study emphasizes the importance of proper background corrections in the calculation of ^{226}Ra body contents at very low levels. The best procedure, usually impractical, would be to have a control subject from the same house as the patient and to measure the two consecutively. An air sample from the subject's environment would be useful, and the feasibility of obtaining such samples for cases of usually low ratios of retained-to-emanating radon is being considered.

An examination of the measurements made on radium patients at Argonne during the past year (1 March 1972 — 29 February 1973) revealed that 16 out of 233 cases (7%) showed unusually high values for emanating ^{226}Ra. These "suspicious" cases are listed in Table 4, where the RaC values have been corrected for the background determined from measurements of control subjects.

Air samples were obtained from the home of one patient (case 10-313) one month after she was measured; the average radon content was 3.2 pCi/l. The subject's measurement may also have been complicated by a slower elimination of radon than normal because she was obese (weight: 98 kg; height: 153 cm); it is thought that radon is more slowly deemanated from fat than from other parts of the body.[3] Compare case 50-025, who weighed 76 kg and was 171 cm in height.

Although the observed fraction of cases with low ratios of RaC to total

TABLE 4. Low-Level Radium Cases with Apparently Excessive Content of Emanating ^{226}Ra[a]

Case No.	Emanating ^{226}Ra content, nCi	RaC content, nCi	Total ^{226}Ra content, nCi	RaC/ Total ^{226}Ra	^{226}Ra content based on RaC only, nCi[b]
03-409	7.2 ± 0.6	1.1 ± 0.6	8.3 ± 0.9	0.13 ± 0.06	2.9 ± 1.6
03-537	4.9 ± 0.5	1.2 ± 0.6	6.1 ± 0.8	0.20 ± 0.08	3.2 ± 1.7
03-570	3.1 ± 0.6	0.77 ± 0.61	3.9 ± 0.8	0.20 ± 0.13	2.0 ± 1.6
03-695	5.8 ± 0.6	1.3 ± 0.6	7.1 ± 0.9	0.18 ± 0.07	3.4 ± 1.7
03-788	4.6 ± 0.5	0.17 ± 0.76	4.8 ± 0.9	0.04 ± 0.17	0.4 ± 2.0
03-838	6.6 ± 0.4	0.77 ± 0.59	7.4 ± 0.7	0.10 ± 0.07	2.0 ± 1.6
10-081	4.5 ± 0.6	0.67 ± 0.59	5.2 ± 0.8	0.13 ± 0.10	1.8 ± 1.6
10-111	3.5 ± 0.5	0.86 ± 0.62	4.4 ± 0.8	0.20 ± 0.12	2.3 ± 1.7
10-145	13.3 ± 0.7	1.7 ± 0.8	15.0 ± 1.0	0.11 ± 0.05	4.5 ± 2.2
10-149	3.7 ± 0.6	0.67 ± 0.61	4.4 ± 0.9	0.15 ± 0.12	1.8 ± 1.6
10-206	5.2 ± 0.7	0.96 ± 0.60	6.2 ± 0.9	0.15 ± 0.08	2.5 ± 1.6
10-225	3.8 ± 0.6	0.57 ± 0.61	4.4 ± 0.9	0.13 ± 0.12	1.5 ± 1.6
10-291	4.0 ± 0.6	0.25 ± 0.58	4.3 ± 0.8	0.06 ± 0.13	0.7 ± 1.5
10-292	6.1 ± 0.5	0.25 ± 0.60	6.3 ± 0.8	0.04 ± 0.09	0.7 ± 1.6
10-313	8.8 ± 0.6	0.3 ± 1.0	9.1 ± 1.2	0.03 ± 0.10	0.8 ± 2.6
10-322	3.4 ± 0.6	-0.20 ± 0.64	3.2 ± 0.9	-----	-0.5 ± 1.7

[a] Errors shown are standard deviations based upon counting statistics and estimated systematic errors.

[b] Assume that total ^{226}Ra = RaC/(0.38 ± 0.06).

^{226}Ra is greater than would be expected from statistical fluctuations alone, it is not clear that time-consuming studies of previously absorbed radon for each case is warranted, although the data do suggest that an estimate of the total ^{226}Ra body content of a low-level case should not be accepted at face value. It is first necessary to assess the results of both the radon breath and the whole-body measurements. Because low values of radon retention could be the result of either recent exposure to ^{226}Ra or of high levels of environmental radon, consideration should be given to both possibilities.

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RETENTION AND ELIMINATION OF BERKELIUM-249-CALIFORNIUM-249 FOLLOWING ACUTE ACCIDENTAL INHALATION*

J. Rundo and J. Sedlet**

A case of accidental inhalation of a small quantity of an ignited mixture of ^{249}Bk and its decay product, ^{249}Cf, was studied by body radioactivity measurements (for ^{249}Cf) and excretion analysis (for both nuclides). The results obtained thus far cover the first year after intake. External measurements of the gamma rays from the ^{249}Cf indicated an approximate chest content of 3 nCi of this nuclide at the time of the first measurement (day 7). The 300-fold more abundant parent, ^{249}Bk, could not be detected. Subsequent measurements of the activity of the ^{249}Cf could be reasonably well described by the sum of two exponential components, 17% having a half-time of 25 days and the remainder a half-time of 1210 days. Except for an initial rapid clearance via the feces during the first 10 days, the urinary and fecal excretion rates of both nuclides increased with time until a maximum was reached 60 to 70 days after intake and thereafter declined. If the early fecal excretion was neglected, the results could be reasonably well described as the difference between two exponential components, one with half-times of 15 to 30 days representing the initial increase and one with half-times between 90 and 120 days representing the subsequent decrease in excretion rate. The increase suggests some change in the inhaled material after intake, possibly an increase in its rate of dissolution.

Introduction

This paper presents the results of the first year's study of an inhalation intake of ^{249}Bk and ^{249}Cf by a chemist during an experimental separation of the two nuclides on an ion-exchange column. A solution of the two nuclides was evaporated on a tantalum disc and heated in a gas flame in preparation for counting. Subsequently, activity on this disc became airborne while it was being handled by the chemist, as evidenced by the early detection of extensive surface contamination in the area. Fecal and urinary collections began immediately, and body radioactivity measurements were begun one week later. The circumstances of the incident strongly suggested an inhalation exposure.

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* A condensed version of this paper will appear in the Proceedings of the 3rd International Congress of the International Radiation Protection Association, September 9-14, 1973, Washington, D. C.

** Occupational Health and Safety Division.

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A smear of a contaminated surface was collected to obtain a sample of the airborne material. This activity was used to prepare standards for the body radioactivity and excretion measurements. The Bk/Cf ratio in this smear is assumed to be the same as in the inhaled material. The berkelium-californium preparation contained only traces of solid material as impurities and since it was ignited, probably existed as the finely-divided oxide. The possibility of its admixture with a small amount of tantalum oxide may exist since tantalum becomes slightly discolored when flamed.

Berkelium-249 decays with a 314-day half-life, principally by emitting a beta particle with an end-point energy of 0.125 MeV, to ^{249}Cf, an alpha emitter whose principal particles have energies of about 5.8 MeV. The californium isotope also emits 333- and 388-keV gamma rays in 16% and 72% of its decays, respectively.[1]

This case contains some unusual and interesting features. There is little reported information on the behavior of californium in the body, and ^{252}Cf may find wide use as a neutron source. If this occurs, the potential for human exposure exists. Since the active material in this incident was ignited before intake, it was presumably highly insoluble. The behavior of insoluble forms of the actinides in the body is of considerable interest, again because of the potential widespread use of these elements. Finally, the parent-daughter relationship of the two nuclides, coupled with the very long half-life of the daughter, presents some interesting problems in data interpretation.

Body Radioactivity Measurements

Seven days after the contamination incident, a measurement of the radioactivity in the subject was made with a crystal of NaI(Tl), 152 mm in diameter by 203 mm long suspended over the subject sitting in the tilted chair. In addition to the normal contributions from ^{40}K and ^{137}Cs at 1.46 MeV and 0.66 MeV, respectively, the gamma-ray spectrum revealed a small peak at about 0.39 MeV, which was attributed to ^{249}Cf.

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Because inhalation was the presumed mode of intake of the contaminant, further measurements were made with two detectors, each 292 mm in diameter by 102 mm thick, positioned above and below the supine subject and placed alternately at the levels of the mid-sternum and of the lower abdomen. The former pair of spectra showed the peak at 0.39 MeV, while the latter did not. In Figure 1 are plotted the spectra obtained with the crystal behind the subject's back, at the time of the first measurement (day 7). The spectra are similar in shape above about 0.48 MeV; below this energy the peak due to the gamma rays from ^{249}Cf is prominent in the spectrum from the chest region and not detectable in the other spectrum. The same pattern was observed in the spectra from both detectors on all subsequent occasions that the subject was examined. The spectra measured with the counter in the abdominal region were used, after appropriate normalization, as "backgrounds" for the spectra from the chest region.

Calibration measurements were made with a source of ^{249}Cf which had been standardized by alpha-particle counting. It was placed in turn in each of the six spaces of a lung phantom similar to that devised by Miller,[2] and the gamma-ray spectra were summed to give an average calibration spectrum from a simulated distributed source. For the energy band 295 to 485 keV, a calibration factor of 5.61 pCi/cpm was obtained when the responses from both detectors were combined, but there is some uncertainty about the reliability of this value. After subtraction of the subject background, the counting rates in this same energy band in the spectra from the subject 7 days after the exposure totalled (two detectors) 480 ± 15 min^{-1}. From the calibration factor above, the lung content on this occasion was 2700 ± 84 pCi. The ratio of the counting rates in the two detectors was 1.82 ± 0.13, with the detector below the back giving the higher rate. For the measurements of ^{249}Cf in the phantom, the ratio was 1.43, and it is this difference which casts doubts on the accuracy of the calibration. During the year following the incident, ten additional investigations were made of the subject's radioactive content. The experimental technique with the two large crystals presented alternately to the chest and to the abdomen was

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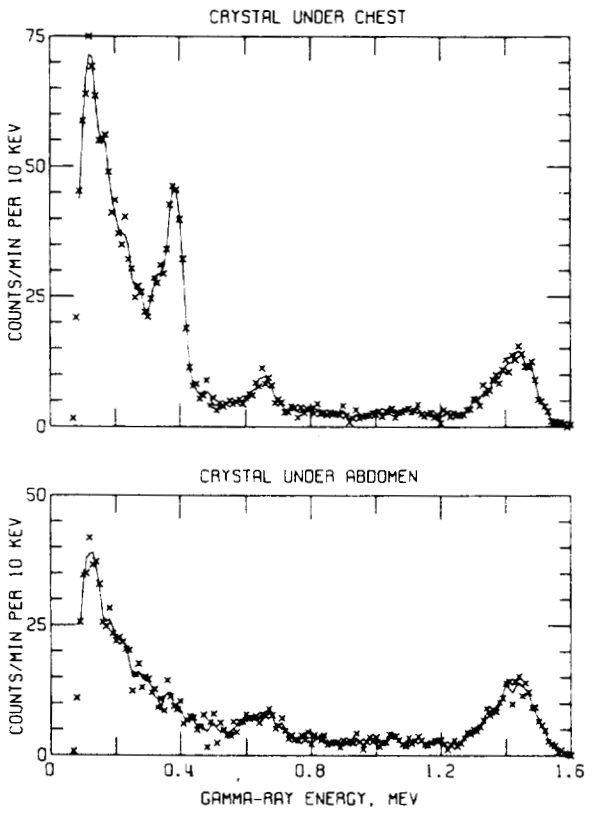

FIG. 1.--Gamma-ray spectra from the subject on the 7th day after intake.

used on each occasion. The total counting rates were corrected for "subject background" as described above before calculating the chest content of ^{249}Cf.

Radiochemistry

The smear sample was dissolved, and evaporated aliquots were standardized by counting in calibrated alpha- and beta-particle counters. The Bk/Cf activity ratio in the contaminant, measured two days after the incident, was 306, and the ratio at other times was calculated from this value and from the half-lives of the nuclides. The ratio of the latter, 418 for Cf/Bk, is similar to the initial activity ratio (Bk/Cf) in the contaminant, so that the complete decay of the original berkelium will nearly double the californium activity.

Samples of urine and feces were analyzed by specially developed radiochemical techniques and liquid scintillation counting of both nuclides simultaneously, as well as by separate alpha- and beta-particle counting in

low-background instruments. Details of the techniques will be described elsewhere, but one point must be mentioned. From five days after the inhalation, fecal samples contained sufficient natural thorium activity (ca. 2 to 4 pCi/sample) to require a californium-thorium separation. The latter was based on the extraction of both elements into di(2-ethylhexyl)phosphoric acid from dilute acid and the back-extraction into strong acid of the californium alone.

Results and Discussion

The results for the urinary excretion of both nuclides and for the fecal excretion of ^{249}Bk are plotted in Figures 2 and 3. The berkelium results have been corrected for radioactive decay from the time of exposure. The californium data have been corrected for the growth of ^{249}Cf from ^{249}Bk in vivo by methods to be described below in order to examine the excretion of inhaled californium only. In the interpretation of the results, a urine volume of 1500 ml (overnight samples were usually obtained — one requested 24-hr sample contained 1350 ml), and a single fecal sample are assumed to comprise the daily elimination. The analyses of the fecal samples for ^{249}Cf have not been completed, but the elimination pattern shown by the available data is similar to that of ^{249}Bk.

The excretion rate in the feces decreased by a factor of about 10^4 during the first week with a half-time of about 0.5 day. After about 10 days, an increase in the fecal excretion rate similar to that found for urine became apparent, and in the case of ^{249}Bk, for which the data are complete, the maximum occurs at the same time as for urine. The curves drawn through the excretion data points in Figures 2 and 3 are weighted least-squares fits to the data of the difference between two exponential terms, representing the initial increase and the subsequent decrease in excretion rates. The data for the fecal excretion of ^{249}Bk for the first 21 days were ignored in making the least-squares analysis. The parameters of the excretion equation,

$$E(\text{pCi/1500 ml or pCi/sample}) = -C_1 \exp(-\frac{0.693}{T_1}t) + C_2 \exp(-\frac{0.693}{T_2}t) \quad (1)$$

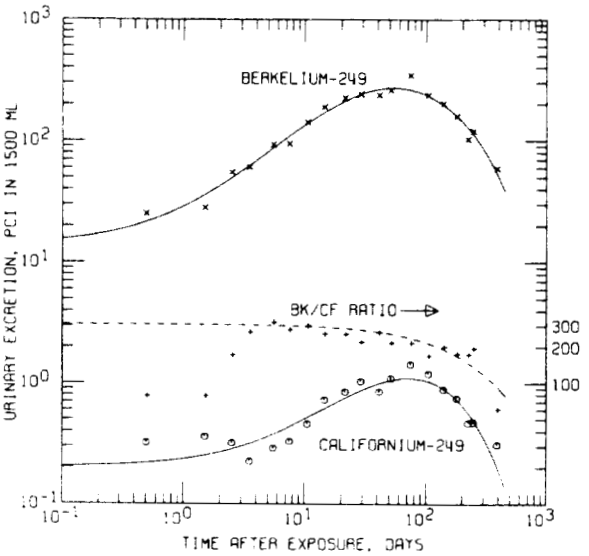

FIG. 2.--Urinary excretion of the two nuclides (x and o) and the curves fitted by least-squares analysis. The dashed curve shows the predicted behavior of the ratio of the activities of the nuclides in the contaminant; experimental values of the ratio in urine are shown as points (+) for comparison.

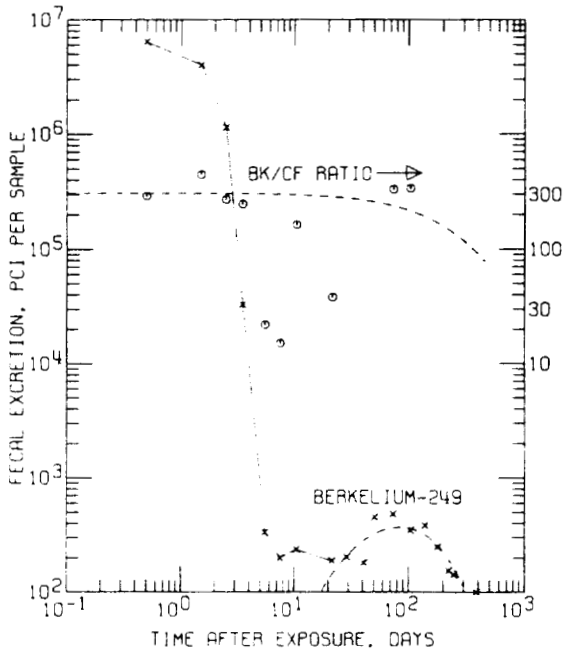

FIG. 3.--Fecal excretion of berkelium-249 (x); the lower dashed curve is the result of a least-squares fit to the points from 21.5 days onwards. The calculated $^{249}Bk/^{249}Cf$ activity ratio in the contaminant is plotted as in Fig. 2, and experimental values of the ratio in feces are included (o).

are given in Table 1. The symbols T_1 and T_2 are the half-time periods (in days) for the increasing and decreasing portions of the curve, respectively; C_1 and C_2 are the corresponding coefficients representing the value of each term (in the same units as E) at zero time, and t is the time since intake. Note that C_1 and C_2 should be equal to meet the criterion of zero excretion rate at zero time. In view of the analytical errors and the biological variation in excretion, the equations represent the data reasonably well. The C_1/C_2 ratios are all unity within the statistical errors, as required. The same processes appear to control the excretion of both nuclides. This is reassuring although not particularly surprising, since the nuclides were inhaled in the same form and have similar chemical properties.

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TABLE 1. Excretion Equation Parameters from Least-Squares Fitting[a]

Sample	C_1	T_1	C_2	T_2
^{249}Bk, urine	396 ± 40	16 ± 2	410 ± 42	132 ± 12
^{249}Bk, feces	1255 ± 300	13 ± 7	1100 ± 340	91 ± 13
^{249}Cf, urine	2.2 ± 1.0	30 ± 10	2.4 ± 1.0	109 ± 36

[a]See Eq. (1) for explanation of symbols.

The body radioactivity measurements also appear capable of representation by exponential terms. When the ^{249}Cf chest content was plotted against time, the trend of the points suggested a two-component exponential function, and a nonlinear weighted least-squares fit was made of such a function. The results are shown in the upper part of Figure 4, where the smooth curve drawn through the points has the equation

$$\text{Content (pCi)} = 480 \exp(-\frac{0.693}{25}t) + 2350 \exp(-\frac{0.693}{1210}t) .$$

The 25- and 1210-day components account for 17 and 83%, respectively, of the initial chest content. If this were a true representation of the retention in the chest, the first derivative of this equation (with a negative sign) would describe the excretion rate, and in the lower part of Figure 4 this is plotted as the broken curve. The points along the curve are the measured total californium excretion (uncorrected for growth) on those days for which measurements are complete. After the early massive fecal clearance is complete, there is respectable agreement between the predicted and observed excretion rates. This suggests that our concern over the reliability of the calibration factor was largely unfounded, although additional data may indicate that the agreement is due partly to a fortuitous combination of different half-times that result in agreement over a limited time interval.

However, this representation may be an over-simplification because of the growth of new ^{249}Cf from the ^{249}Bk. By the time of the last measure-

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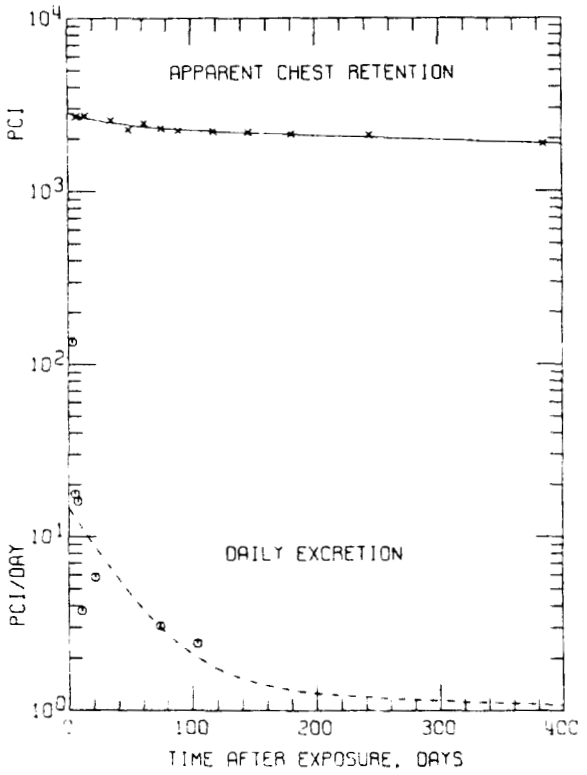

FIG. 4.--Apparent chest retention (x) of the ^{249}Cf, based on external gamma-ray counting; the regression equation is drawn as a smooth curve, and its first derivative is a dashed curve. For comparison with the latter, observed total excretion data are shown (o).

ment so far made (day 388) an additional 1.25 nCi of ^{249}Cf would have been formed and the question arises as to what extent this remained in the chest or was excreted preferentially. This question cannot be answered yet.

In order to investigate a possible change in the distribution of ^{249}Cf in the chest, the stability of the ratio of the counting rates from the front and back was studied. The mean value was 1.90, and the standard deviation of 0.16 may be compared with the statistical standard errors on the individual counting rate ratios; these ranged from 0.12 to 0.17. Thus, the ratio was constant (within statistics), which indicates that there was no major change in the distribution during the first year.

There are some differences between the excretion behavior of the two nuclides that can be seen from the Bk/Cf ratios plotted in Figures 2 and 3. If the ^{249}Bk and ^{249}Cf, including the ^{249}Cf produced in the body by decay of ^{249}Bk, are excreted at the same rate, then the Bk/Cf ratio in each sample should be that calculated from the initial ratio in the smear sample. For the first three urine samples, the observed ratio was significantly lower than

that calculated, which indicates an excretion mechanism favoring californium; however, the ratio increased with time until the calculated value was reached about 5 days after intake. The ratio in the fecal samples showed a different behavior. The first few samples had a ratio close to the calculated one (except for a high value in the second sample, possibly due to analytical error), as expected if the excreted activity had not undergone any changes. Thereafter the ratio decreased very sharply, to indicate excretion favoring californium; then it increased to values somewhat greater than those calculated. The data are insufficient as yet to establish a pattern.

In order to correct for the contribution of ^{249}Cf from the decay of ^{249}Bk after intake, some assumptions are necessary since the excretion rate of the newly-generated ^{249}Cf is not independently known. If the Bk/Cf ratio in the excretions remained the same as that calculated from the half-lives, which is apparently the situation in the urine after 5 days, then it is reasonable to assume that all the californium is excreted at the same rate, and the fraction of californium produced in vivo in any sample can be calculated from the initial ratio. Another method of making this correction is to assume that in each sample the difference between the excreted ^{249}Bk activity and the amount that would have been excreted had it not decayed was actually excreted as ^{249}Cf. For the samples that required corrections in excess of 5% of the amount excreted (samples after 40 days), the differences between the two correction methods ranged from 0.5 to 12.5%. The biggest difference occurred in the last sample. The agreement between the two methods is considered good. Perfect agreement would be expected if the assumptions used in making the corrections were valid and if there were no measurement errors. The correction is not yet large, amounting to 25% of the excreted activity at one year, but it will increase with time.

Integration of the excretion equations gives that portion of the original body burden to be excreted with the biological half-times thus far evident in the excretion rates. The future appearange of longer-lived components would increase the amount calculated in this way, and it is possible that a

significant fraction may exist as a stationary pool in the body. However, if the exercise is carried out, the results in Table 2 are obtained. Also shown are the measured chest burden of ^{249}Cf and the ^{249}Bk chest burden calculated from this value. The integrated excretion accounts for 25% of the ^{249}Cf burden (2.8 nCi) and 20% of the calculated ^{249}Bk burden (870 nCi).

TABLE 2. Comparison of Total Excretion Based on Integrated Excretion Equations with Body Contents.

Sample		E(Tot.),[a] nCi	Body content, nCi
^{249}Bk, urine		69	
^{249}Bk, feces		106	
	Sum	175	(870)[c]
^{249}Cf, urine		0.28	
^{249}Cf, feces		(0.43)[b]	
	Sum	(0.71)	2.8

[a] Calculated from $\int_0^\infty E = -C_1/\lambda_1 + C_2/\lambda_2$.

[b] Calculated with the assumption that Bk(urine)/Bk(feces) = Cf(urine)/Cf(feces).

[c] Bk value based on Cf value and initial Bk/Cf ratio of 308.

This appears to be a reasonable fraction of an inhaled, insoluble material to be eliminated with half-times as short as 90 to 130 days.

The excretion pattern is consistent with the hypothesis that the activity was taken into the body in an "unexcretable" form and that, with the early clearance neglected, it required conversion to a second form before it could be excreted. If both the conversion and the excretion are exponential with rate constants, λ_1 and λ_2, respectively, then the observed excretion

rate, E, will be

$$E = \frac{\lambda_1\lambda_2}{\lambda_2-\lambda_1} A_0 \exp(-\lambda_1 t) - \frac{\lambda_1\lambda_2}{\lambda_2-\lambda_1} A_0 \exp(-\lambda_2 t) , \qquad (2)$$

where A_0 is the amount of unexcretable material inhaled. If a portion if the material inhaled (B_0) was originally in the excretable form, then the observed excretion becomes

$$E = \frac{\lambda_1\lambda_2}{\lambda_2-\lambda_1} A_0 \exp(-\lambda_1 t) - (\frac{\lambda_1\lambda_2}{\lambda_2-\lambda_1} A_0 - B_0) \exp(-\lambda_2 t) . \qquad (3)$$

The equations are analogous to those that describe two successive radioactive decays. This model is similar to that proposed by Healy[3] for the excretion of plutonium from insoluble deposits in the lung, and differs from his only in that he used a power function to describe the excretion portion rather than an exponential function. Since Eqs. (2) and (3) are in the same form as the excretion Eq. (1), the parameters A_0 and B_0 can be evaluated from the C and λ (or 0.693/T) values obtained from the least-squares fit (Table 1). The results obtained in this way are given in Table 3. The values of A_0 (or A_0 + B_0) should be the same as the integrated excretion values in Table 2, and they differ only because the average of C_1 and C_2 was used to evaluate A_0 from Eq. (2) and because only C_1 was used to evaluate A_0 from Eq. (3). The amount presumably inhaled in "excretable" form (B_0) is small compared to A_0, and within experimental error, it may be taken as zero. Although the validity of the model is not established by the data, it does account for the increase in excretion rate with time. Increases in excretion rate with time after inhalation of insoluble particulate material have also been observed by Wood and Sheehan[4] and by Campbell et al.[5] In these cases the contaminant was plutonium, and the increases, which continued up to several hundred days after intake, were attributed to the low solubility of the plutonium compound.

Evidence for the insolubility of the inhaled contaminant is obtained from the chemical behavior of the material excreted in the feces. In the

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TABLE 3. Evaluation of Excretion Parameters

Sample	Eq. (2) A_0, nCi	Eq. (3) A_0, nCi	Eq. (3) B_0, nCi
^{249}Bk, urine	67	66	0.014
^{249}Bk, feces	106	113	-0.16
^{249}Cf, urine	0.26	0.26	2×10^{-4}

first few fecal samples, a substantial portion (25% to 70%) of the activity could not be dissolved by our conventional nitric acid treatment, but it required additional vigorous treatment with hydrochloric acid. The fraction which was insoluble in nitric acid decreased in later samples and was never higher than 2% in the samples collected after day 10. Further, the activity in the nitric acid solutions of the early fecal samples was only poorly extracted into di(2-ethylhexyl)phosphoric acid, although it could be coprecipitated with nonisotopic carriers. At the same time, tracer solution added to aliquots of such samples was efficiently extracted. The activity could be rendered extractable by boiling with strong hydrochloric acid. Evidently the activity, although homogeneously distributed in the acid solution, was not in a simple ionic form that permitted extraction. Subsequent fecal samples contained activity that, based on its acid solubility and extraction behavior, had undergone a change to a more soluble form.

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PRELIMINARY ANALYSES OF SOME MEASUREMENTS OF ^{231}Pa IN VIVO

J. Rundo and C. J. Maletskos*

The radioactive content of a 67-year-old man was provisionally estimated to be 71 nCi ^{231}Pa, 51 nCi ^{227}Th, and 39 nCi ^{223}Ra 45 years after exposure to protactinium. Scans with an uncollimated counter suggested that the parent ^{231}Pa was confined to the lung.

Introduction

In this report we present preliminary analyses of some measurements made in the Radioactivity Center, Massachusetts Institute of Technology, of radioactivity in case 30-021, a 67-year-old man who had been exposed to ^{231}Pa some 45 years previously.

Protactinium-231 is the 32,500-yr daughter of ^{235}U; it emits 5-MeV α particles, and it heads the remainder of the actino-uranium, or 4n + 3, decay chain. The maximum permissible body burden for occupational exposure is 0.02 μCi, with bone as the organ of reference. Its toxicity is thus greater than that of the alpha-particle emitting isotopes of plutonium, but its assay in vivo is much simpler because it emits 300 keV-gamma rays albeit in only 6% of the disintegrations. A complicating factor is the emission of gamma rays by several members of the decay chain subsequent to the daughter, 21.6-yr ^{227}Ac. Not only are the intensities of these gamma rays similar to, or even greater than, those of the ^{231}Pa emissions, but their energies are not very different, being mainly in the range 230 to 400 keV. Difficulties can, therefore, be anticipated in the analysis of gamma-ray spectra of a mixture of the parent ^{231}Pa and the daughters of ^{227}Ac. These difficulties can be appreciated from a study of Figure 1, which shows the gamma-ray spectra of equal activities of ^{231}Pa unaccompanied by daughters, of its granddaughter ^{227}Th (18.2 days), and of an equilibrium mixture of 11.4-day ^{223}Ra with its descendants, all of which have much shorter half-

*Consultant, Radioactivity Center, Massachusetts Institute of Technology, Cambridge, Mass. 02139.

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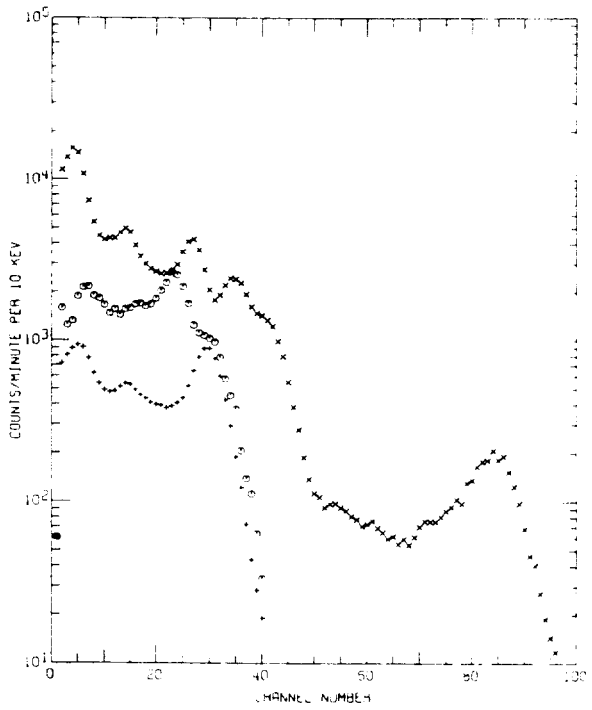

FIG. 1.--Gamma-ray spectra of equal activities of ^{231}Pa (+), ^{227}Th (Θ), and ^{223}Ra (X), demonstrating the difficulty of analyzing by gamma-ray spectrometry a non-equilibrium mixture of the three. The ^{223}Ra was accompanied by all its short-lived daughters in equilibrium.

lives. Least squares analysis is the only method capable of giving useable results for a non-equilibrium mixture at low levels of activity. We have used the versatile computer program GASP[1] in the analysis of the measurements reported here.

As will be seen, this case has some similarities to the case described by Newton, Rundo, and Sandalls,[2] but there are some important differences.

Experimental Measurements

As nothing was known about the level of activity in the subject or its distribution in vivo, a large number of measurements of the subject's radioactivity was made with an NaI(Tl) crystal, 11-1/2 inches in diameter by 4 inches thick, in the iron room at the Radioactivity Center, M.I.T. The following series of examinations were made:

a) Two measurements with the subject in arc geometry (radius 1.25 m) with the crystal at the arc center and the subject alternately facing toward and away from the detector.

b) a measurement with the subject seated in the tilted chair and

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the detector suspended above with the face of the detector 43 cm from the seat and 47 cm from the back of the chair, and

c) two 7-position scans with the detector above the long axis of the subject, lying alternately supine and prone; the positions were spaced at 25.4-cm intervals, and in position 4 the crystal was over the mid-point of the body.

The importance of the possible serious effects of small changes in the gain of the spectrometer was recognized, and a spectrum from a source of ^{226}Ra was accumulated whenever the detector was moved to a new position. In addition to measurements of the counter background, measurements were made in the same geometries of the response to an uncontaminated "background" subject of essentially the same height and weight.

Standardized sources of ^{231}Pa (free of daughters), ^{227}Ac (with all daughters in equilibrium), and ^{223}Ra (with all daughters in equilibrium) were supplied by R. Sjoblom of the ANL Chemistry Division. These sources were counted in a phantom in the geometries of series b) and c) above, and also behind the subject in the arc geometry of series a).

Results

Measurements in Arc Geometry

The activity proved to be too low to measure in the arc geometry; counting rates were observed from the subject of 226 counts/min and 248 counts/min in excess of the counter background of 2150 counts/min. Approximately half of the net rate could be attributed to the subject's content of ^{40}K and ^{137}Cs, as determined from the measurement on the uncontaminated subject, for whom a net rate of 115 counts/min was observed. These results will not be considered further.

Measurement in Tilted Chair

After subtraction of the counter background, the gamma-ray spectrum of the subject's radioactivity as measured in the tilted chair was analyzed by the least squares method. Gamma-ray spectra of pure ^{231}Pa, of ^{227}Ac with all daughters, of ^{223}Ra with all daughters in a phantom, and of the

^{40}K and ^{137}Cs in the uncontaminated subject were used in the analysis. The computer program GASP determined the fractions of these spectra which, when combined in a linear manner, gave the best fit in the least squares sense to the spectrum from the contaminated subject. In the upper part of Figure 2 the measured and fitted spectra are plotted. In the lower part of Figure 2, the difference spectrum is plotted in units of statistical standard errors. Fitting was confined to channels 19 to 160. Relatively large fluctuations in the fitted spectrum above channel 46 are due to the fact that the spectrum which was used as the standard for ^{40}K and ^{137}Cs (i.e., that from the uncontaminated subject) was only measured for 30 min. It should be noted that use of the above-mentioned standards in the least squares fit is expected to indicate a negative contribution from ^{223}Ra if the level in vivo is lower than that of its parent, ^{227}Th. The true content of ^{223}Ra is, then, the algebraic sum of the computed contents of ^{227}Ac and ^{223}Ra, while the result for the ^{227}Ac is a measure of the content of ^{227}Th, since ^{227}Ac does not emit gamma rays. The true content of ^{227}Ac may be higher.

The analysis indicated contents of 71 ± 8 nCi ^{231}Pa, 51 ± 3 nCi ^{227}Th and 39 ± 10 nCi ^{223}Ra; the validity of these results depends strongly on how

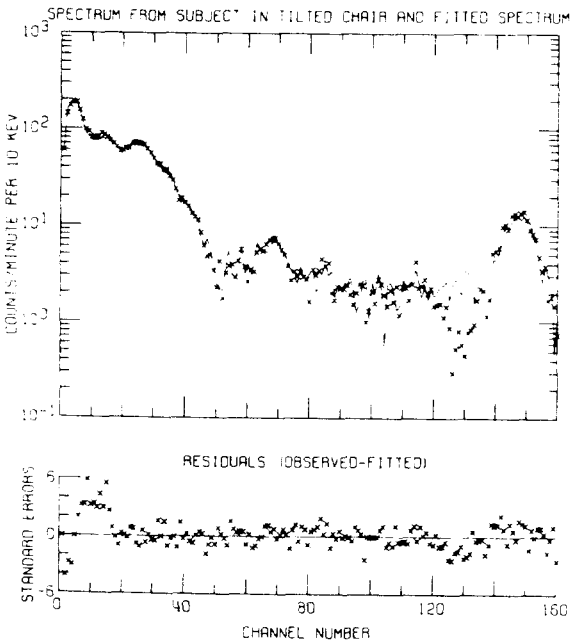

FIG. 2.--Gamma-ray spectrum from the contaminated subject in the tilted chair (experimental points) and the spectrum fitted by least squares analysis (curve). In the difference spectrum plotted in the lower part of the diagram, the counting rates are expressed in units of standard errors.

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well the actual geometry (i.e., the distribution in vivo) was matched in the measurements of the standards in the phantom. This will be discussed later.

Scan Measurements

The 7-position scan gives some information on the distribution of the activity in vivo, although the spatial resolution is not particularly good because the detector was not collimated. Even without analyzing the spectra for its components, we can obtain some information by plotting the total counting rates (counter background subtracted) as in Figure 3, which includes the results for the uncontaminated subject. The curves suggest a marked concentration in the region of position 2, and the asymmetry of each peak indicates that the center of activity is a little further down the body than the level of position 2. This region is identified as the thorax, although there is nothing to indicate whether the activity is in lung or in the tracheobronchial lymph nodes.

Before analysis of the 7 spectra observed in the scan, the gain of the spectrometer was determined from the measurements of the ^{226}Ra source at each of the 7 positions. Least squares fits were made to gaussians of the peaks at 0.609 MeV and 1.76 MeV, and the channel width was calculated from their positions. The results are of sufficient interest to warrant presentation here, and they are plotted in Figure 4. Because of (known) residual magnetism in the walls of the iron room,[3] the gain increased at the ends of the scan as the detector approached the walls, despite the fact that each of the seven photomultiplier tubes had a magnetic shield. The maximum change in gain was 2.6% (position 3 to position 7). The standardized sources and the background were measured in position 4, and the gains of the gamma-ray spectra taken in the other positions were changed to correspond to that at position 4 before subtraction of the background and analysis by least squares fitting.

The results of the analyses are shown in Table 1 and in Figure 5 where the standard errors of counting are shown. The totals of the entries in each column of Table 1 must be overestimates of the true content because no allowance has been made for the contributions to a spectrum in a given

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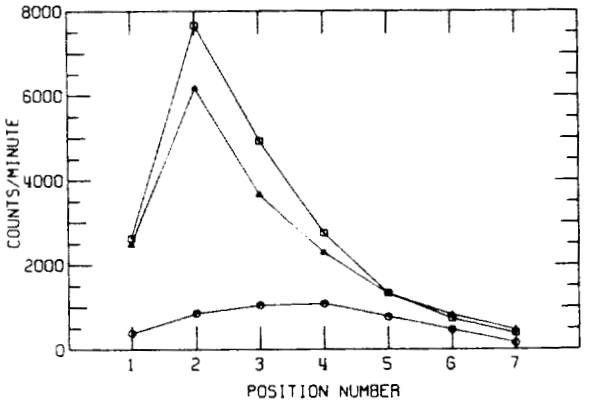

FIG. 3.--Counting rates observed (counter background subtracted) from the contaminated subject in the two 7-position scans compared with a scan of an uncontaminated subject in the supine position. Contaminated subject (prone) ⊡; Contaminated subject (supine) ▲; Normal subject ⦶.

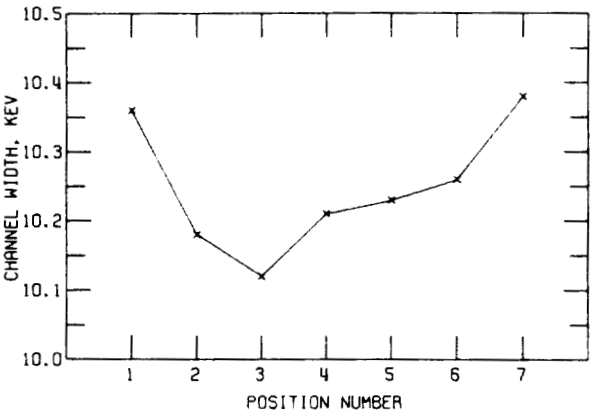

FIG. 4.--The channel width (inversely proportional to gain) of the gamma-ray spectrometer in the 7-position scan, showing the effects of residual magnetism in the end walls of the iron room.

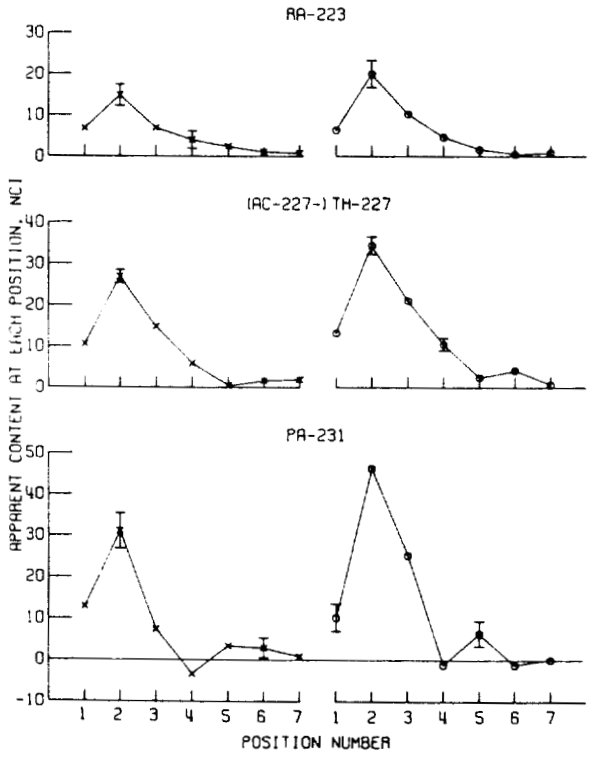

FIG. 5.--Results of analyses by the least squares method of the measurements made in the two 7-position scans. On the left, subject supine; on the right, subject prone.

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TABLE 1. Results (± Standard Errors) of Analyses by Least Squares of the 14 Spectra Accumulated in the Two 7-Position Scans

Scan position no.	Subject supine			Subject prone		
	^{231}Pa, nCi	^{227}Th, nCi	^{223}Ra, nCi	^{231}Pa, nCi	^{227}Th, nCi	^{223}Ra, nCi
1	12.9 ± 3.1	10.5 ± 1.2	6.8 ± 1.9	10.2 ± 3.3	13.2 ± 1.2	6.3 ± 2.0
2	31.1 ± 4.3	26.9 ± 1.6	14.8 ± 2.6	46.3 ± 5.5	34.4 ± 2.1	19.9 ± 3.3
3	7.4 ± 3.6	14.8 ± 1.4	6.9 ± 2.2	25.2 ± 4.7	21.1 ± 1.8	10.2 ± 2.9
4	-3.4 ± 3.3	5.9 ± 1.3	4.0 ± 2.1	-1.2 ± 4.1	10.5 ± 1.5	4.6 ± 2.5
5	3.4 ± 2.9	0.5 ± 1.1	2.4 ± 1.8	6.4 ± 3.1	2.5 ± 1.2	1.6 ± 2.0
6	2.9 ± 2.4	1.7 ± 0.9	1.1 ± 1.4	-1.2 ± 3.0	4.2 ± 1.1	0.5 ± 1.9
7	0.9 ± 2.3	2.0 ± 0.8	0.8 ± 1.4	0.0 ± 2.5	0.7 ± 0.9	0.9 ± 1.5
Total	55.2 ± 8.5	62.3 ± 3.2	36.8 ± 5.2	85.7 ± 10.2	86.6 ± 3.8	44.0 ± 6.3

position from activity in an adjacent one. In any event, the results for the scans with the subject supine and prone show the substantial differences that would be expected from the higher counting rates observed from the back (Figure 3). The means of the sums of the two sets of results may represent better estimates of the actual contents of the three nuclides. They are 70.4 ± 15.2 nCi for ^{231}Pa, 74.4 ± 12.2 nCi for ^{227}Th, and 40.4 ± 3.6 nCi for ^{223}Ra.

Discussion

The agreement between the results for ^{231}Pa and ^{223}Ra derived from the measurements in the tilted chair and the means of the totals from the scan measurements is perhaps largely fortuitous, but it does suggest that the true contents are not radically different from these. It should be noted that the ^{227}Th content cannot be greater than the ^{231}Pa content; indeed after growth from pure ^{231}Pa for 45 years, the activity of the ^{227}Ac, and therefore of ^{227}Th, could hardly exceed 0.75 of that of the parent, and this would require that no ^{227}Ac or ^{227}Th were excreted. The results from the

measurement in the chair geometry would, therefore, seem to be more reliable.

The results in Figure 5 and Table 1 indicate that there was little, if any, ^{231}Pa farther down the body than the level of position 4 and that the activity in the lower half of the body was due to ^{227}Th and ^{223}Ra, the former presumably being supported by ^{227}Ac in the same locations.

The plotted results for ^{231}Pa shown in Figure 5 for the subject in supine and prone positions are consistent with activity distributed throughout the lung. Although by no means conclusive, the supporting evidence may be summarized as follows:

a) higher activity observed from the back than from the front,

b) maximum activity nearer the vertex when measured from the front than when measured from the back, and

c) a broader peak in the scan of the activity measured from the back.

These are all characteristics which can be predicted from a consideration of the size, shape, and positioning of the lungs in the thoracic cavity. If the concentration in this region were due to deposition in the tracheobronchial lumph nodes, we should not expect to observe these features. In this respect the present case differs radically from the one studied by Newton et al.[2]

If it is accepted that the ^{231}Pa content is confined to the lungs, then the counting geometry of the measurement of the standard in the phantom was a) reasonable from the point of view of the distribution, but b) bad from the point of view of absorption since the phantom was of unit density material. We should, therefore, not expect the shape of the spectrum from the standard to match that from the subject, especially at low energies. Somewhat similar arguments could apply to the standards of ^{227}Ac and ^{223}Ra; however, there appears to be activity due to these daughters in locations other than lung so that the counting geometry for these standards was poorer. As seen in the lower part of Figure 2, the synthesized spectrum did not match the observed spectrum well at low energies, below the band of channels (19-160) used for the least squares fit.

There is also a poor match in a region of the spectrum where none of the contaminants contributes, viz., between channels 120 and 140. This is

due to interference from ^{41}Ar from the reactor on the M.I.T. campus. A higher concentration was probably present in the air in the iron room when the background was measured than when the contaminated subject was measured, so that a negative peak appeared in the net spectrum. The opposite situation may have prevailed when the uncontaminated subject was measured, but to a smaller extent, so that the valley between the Compton edge of the 1.46-MeV quanta and the total absorption peak was filled. Because the difference due to ^{41}Ar is small compared to the contributions from the contaminants, its omission is not thought to have had a significant effect on the results of the analysis, but this needs confirmation.

From the upper two pairs of curves in Figure 5, it seems that the distributions of the ^{227}Th and the ^{223}Ra may have been similar. In this case, the estimates of the contents from the analysis of the measurement in the chair geometry are probably in about the correct ratio, even if the absolute values are in error. The ^{223}Ra content was 0.76 ± 0.20 of the ^{227}Th content, and therefore probably also of the ^{227}Ac content. This is very similar to the value of 0.7 found in the case studied by Newton et al.,[2] where the ^{227}Ac was concentrated in the liver and the skeleton. The body contents of 51 nCi ^{227}Th and 39 nCi ^{223}Ra can be used to calculate the apparent biological half-life of ^{223}Ra produced in vivo and the expected excretion rate of ^{223}Ra. The results are 37 days and 730 pCi/day, respectively.

Because the counting geometry for the standards of ^{227}Ac and ^{223}Ra was a poor match to the presumed distribution of these nuclides in vivo, their true contents must be somewhat higher than 51 nCi and 39 nCi respectively. It is a simple matter to calculate that, in the absence of excretion of either ^{231}Pa or ^{227}Ac would have grown in 45 years to 0.764 of the ^{231}Pa content, i.e., to 54 nCi. However, the provisional results of some analyses by F. Ilcewicz and R. B. Holtzman of one or two samples of urine and feces from this subject showed that there was definite excretion of both nuclides. The very low levels found indicate that the biological half-lives are very long (tens of years). Again, this is in agreement with the findings of Newton et al.[2]

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Further Work

From the results so far obtained, we can make some suggestions for future studies, which should enable us to re-analyze the data and place more faith in the results.

Measurements of the standardized sources in a more realistic phantom would be a relatively simple matter. Now that we know that the major part of the ^{231}Pa is in lung, we should use a phantom of suitably low density. For the ^{227}Ac and ^{223}Ra, the situation is not so clearcut; it is doubtful if the least-squares analysis could give sensible results if the program were supplied with two spectra for each standard, one measured in a phantom of unit density, the other in a lung phantom.

The conclusion that the ^{231}Pa is still in the lung even after 45 years is a startling one that needs experimental verification. When the subject becomes available for further measurements, these should include determination of two profile curves in the region of the thorax, i.e., scans with the detector fitted with a slit collimator, first down the length of the body with the slit perpendicular to the long axis, and then across the body at the point of maximum activity. The latter should show two peaks on either side of the midline if the activity is indeed in the lung. A single central peak would be evidence for concentration in the tracheobronchial lymph nodes.

Finally, complete collection of excreta for a period of about 7 days and careful analysis for ^{231}Pa, ^{227}Ac, ^{227}Th, and ^{223}Ra would yield most valuable information on the metabolism of these nuclides at very long times after intake.

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THE MEASUREMENT OF "MOCK PLUTONIUM" IN VIVO

J. Rundo, K. F. Eckerman,* R. E. Toohey and M. A. Essling

Three volunteers inhaled an insoluble aerosol labeled with ^{103}Pd and ^{51}Cr at Harwell, England, and they visited a number of laboratories in the U.S.A. and Europe. Our measurements of their radioactive content provided us with a preliminary calibration for the external counting of plutonium in lung. The method used to determine the ^{51}Cr content is described in detail, and some studies are presented on the effects of geometry and on the emission of the x and gamma rays from small (25 cm^2) areas of the chest of one subject.

Introduction

One of the three steel rooms in the underground vault is intended for the measurement of low energy (< 100 keV) photon emitters, and we have installed two detectors in it. One is a 7-in diameter proportional counter** filled to atmospheric pressure with a 90/10 mixture of xenon and methane and with a built-in anticoincidence section. The other detector is an 8-in diameter "phoswich," i.e., a composite of a 3-mm thick crystal of NaI(Tl) backed by a 51-mm thick crystal of CsI(Tl), the whole being viewed by a single 7-in diameter photomultiplier tube. The major use for these detectors will be the measurement in vivo of transuranium elements which emit only low energy electromagnetic radiation. The most important of these is, of course, plutonium, which is a (not very prolific) source of the characteristic L x rays of uranium. The energies of these quanta are approximately 13.6, 17.2, and 20.2 keV, and their attenuation in soft tissue from a source in vivo is severe; the half-value thickness is about 7 mm. Thus calibration for external counting of plutonium in the human lung is a challenging problem.

We were fortunate enough to be able to participate in the fall of 1972 in an I.A.E.A.-sponsored and funded experiment involving the measurement in vivo of "mock plutonium." This term was proposed in 1962 by Rundo and

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*Environmental Statement Project.

**Type CPX 180 M1, available from Compagnie Generale de Radiologie, Issy-les-Moulineaux, France.

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Taylor[1] to describe a radioactive aerosol with an x-ray output similar to that of plutonium, but suitable for inhalation by volunteers by virtue of the use of short-lived nuclides which decay by electron capture and emit characteristic K x rays. The radiation dose to the lung of a volunteer would, therefore, be acceptably small (less than 100 to 200 mrem). An isotope which has been successfully used in such work[2-4] is ^{103}Pd, which decays by electron capture with a 17-day half-life, emitting the characteristic K x rays of rhodium at 20.2 keV (K_α) and 22.8 keV (K_β) in approximately 80% of the disintegrations. In order to determine the amount of aerosol in the lung, a gamma-ray label is also necessary, and the 27.8-day electron-capture nuclide ^{51}Cr, which emits 320-keV quanta, was used.

The aerosol in the present experiment was similar to that used previously, viz., 5 μm-diameter microspheres of polystyrene, labeled with ^{103}Pd and ^{51}Cr. The amount of ^{103}Pd in the lung could be determined from a measurement by gamma-ray spectrometry of the ^{51}Cr content and application of the known ratio of activities. Thus, an external detector of the x rays from the ^{103}Pd could be calibrated; by calculation a calibration for plutonium could be determined. The aerosol was prepared and inhaled in the Health Physics and Medical Division of the Atomic Energy Research Establishment, Harwell, England. Three volunteers, representing small, medium, and large builds, visited a total of 14 laboratories (including ANL) in Europe and North America. Their vital statistics are shown in Table 1.

TABLE 1. Vital Statistics of the Three Volunteers

Subject	Age, yr	Height, cm	Weight, kg	Chest circumference, cm	Chest thickness at mid-sternum, cm
DN	35	169	60.8	91	21.4
JR	47	178	80.6	100	23.5
KB	50	174	89.9	111	26.1

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The amount of ^{51}Cr inhaled by each subject was estimated at Harwell, and sources of ^{51}Cr and of ^{51}Cr plus ^{103}Pd were calibrated at Harwell and were left at each laboratory visited. The participants were then to use their standard counting and calibration procedures as for a case of contamination by plutonium to determine the amount of Cr and Pd contained by each subject and to send the results to Harwell for intercomparison. However, since our counters for measuring lung burdens of plutonium were only recently installed, we did not have a standard calibration procedure. Consequently, we decided to use the results of this experiment as a preliminary calibration; that is, by determining the response of our detectors to a known amount of ^{103}Pd in the subjects' lungs, we could calculate the response to a given lung burden of plutonium. Nevertheless, we have made estimates of the ^{103}Pd contents for comparison with the values dictated by the ^{51}Cr contents and the known ratio of activities.

Measurements of ^{51}Cr

Large crystals of NaI(Tl) were used for estimating body contents of ^{51}Cr. Measurements were made in arc geometry (radius 1.5 m) with a 6-in diameter by 8-in long crystal. The subjects were counted in two positions, facing toward and away from the detector.

The body contents of ^{51}Cr were calculated from the equation

$$\text{Content } (\mu\text{Ci}) = \frac{\bar{R}\, e^{\mu d}}{f_{fs}} \times \left(\frac{d_e}{d_s}\right)^2 \times \frac{R_s}{C_s} , \qquad (1)$$

where $\bar{R}$ is the geometric mean of the photopeak counting rates observed from the subject in the two positions in the arc,

μ is the experimentally determined linear attenuation coefficient (cm^{-1}) for ^{51}Cr gamma rays,

d is the thickness (cm) of tissue which contributes to attenuation,

f_{fs} is a dimensionless factor to correct for the effect of forward scatter in the determination of the attenuation coefficient,

d_e is the effective distance (cm) from the activity in vivo to the

center of the detector,

d_s is the distance (cm) from the standardized source of ^{51}Cr to the center of the detector,

R_s is the activity (μCi) of the standardized source of ^{51}Cr,

C_s is the counting rate from the standardized source of ^{51}Cr.

When activity is distributed widely through the body (as in the measurement of ^{226}Ra), d and d_e are closely related. Thus, d is taken as half the average thickness of the whole body, and d_e is the distance from the arc bed to the detector center, less d. In the present instance, the activity is confined to the lungs, and d is made up of two components, d_1 and d_2. These are the average thickness of lung tissue of low density between the effective center of activity and the inside of the chest wall (d_1) and the thickness of the chest wall covering the lung (d_2). If we assume that the density of lung tissue in vivo is 0.25 g cm^{-3}, then the total equivalent thickness of tissue of unit density is

$$d = 0.25\, d_1 + d_2 .$$

We have made an indirect approach to the determination of d_1. We first calculated a value for d_2 from the empirical expression derived by Ramsden et al.[5] for the average thickness of the chest wall external to the rib cage:

$$d_2 = 15.3\, W/H - 0.01C - 3.55,$$

where W is the subject's weight in kg, H is the height in cm and C is the chest circumference in cm. By subtracting this value from half the chest thickness at mid-sternum, we obtain a value which we consider to be our best estimate of d_1. Thus, for subject JR, d_2 was 2.38 cm, and the chest thickness was 23.5 cm, so that

$$d = 0.25\,(11.75 - 2.38) + 2.38$$
$$= 4.72 \text{ cm} .$$

Constants in the expression for the content are the values of μ(0.0804 cm^{-1}), f_{fs} (1.076), d_s (150 cm) and R_s/C_s (0.00559 μCi/count/min). The various factors in the calculation and the results are set out in Table 2. In making these calculations, we have tacitly assumed that the use of the

TABLE 2. Data Used in the Calculation of the Lung Contents of ^{51}Cr for the Three Subjects and the Results

Subject	$\bar{R}$ c/m	d_2, cm	$e^{\mu d}$	d_e	$\left(\frac{d_e}{d_s}\right)^2$	^{51}Cr content, μCi	^{51}Cr content extrapolated from Harwell data, μCi
DN	288.9	1.04	1.320	140.3	0.875	1.74	1.86
JR	73.5	2.38	1.462	139.25	0.862	0.48	0.67
KB	233.3	3.25	1.580	138.0	0.846	1.62	1.88

geometric mean of the counting rates from either side of each subject correctly allows for the fact that the center of activity may not be in the midplane of the body. The counting rate from the back of each subject was higher than that from the front, and the data can be treated in an alternative way. From the ratio of the two counting rates, the depth of the effective center of activity was estimated,[6] and two values for the ^{51}Cr content were calculated from the individual counting rates from the front and from the back, with the appropriate values for d and d_e. For each subject, the two estimates were equal and the same as the values in Table 2, despite the fact that the counting-rate ratio ranged from 1.09 for JR to 1.225 for KB.

Our estimates range from 72% to 94% of the Harwell estimates, and the reason for these differences is under investigation. It should be noted that if the calculation of the contents is repeated with no allowance for the low density of the lung, the absorption correction factors become much bigger. The contents would then be 2.98, 0.82, and 2.82 μCi for subjects DN, JR, and KB, respectively; these values are all about 70% greater than the results in column 7 of Table 2. They are also greater than the Harwell estimates but by a more variable amount (22% to 60%). Thus, it is very important to allow for the lower absorption in lung. However, we may not have estimated correctly the three values of d for Eq. (1). A change of 1 cm in

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the value of d, changes the results by 8.4%. Our estimates might well be incorrect by this amount, but it would not be enough to bring all the results into line with those of Harwell. While we should not necessarily expect perfect agreement, it would be comforting if the two estimates for each subject were in the same ratio. It is the fact that they are not which gives some cause for concern, and which demonstrates the need for further study.

Some measurements of the gamma rays from the ^{51}Cr were also made with the CsI(Tl) part of the phoswich detector. The electronic equipment was not optimized to give minimum background, but this was not important because of the high counting rates observed from the subjects. The counter was useful for simultaneous measurements of the x rays from the ^{103}Pd and the gamma rays from the ^{51}Cr.

The group at A.E.R.E., Harwell, has a phoswich which is nominally identical to ours. Measurements were made at both laboratories of the response to ^{51}Cr with the edge of the face of the detector 20 cm above the sternal notch of subject JR, in order a) to compare detector efficiencies, and b) to make an extended retention study possible. Provisional results are shown in Figure 1. A least squares fit was made of a single exponential function of time to the data (^{51}Cr peak counting rates), and the results indicated an effective half-life of 18.4 ± 0.5 days, which is considerably shorter than that indicated by the Harwell data alone. This may be a consequence of differences in the resolutions of the two counters, and further investigation is needed.

In the course of these measurements we noted a reduction, amounting to several percent, in the counting rate as the subject relaxed under the counter. On one occasion, a series of 100-sec measurements was made to study this, and the results are plotted, on an expanded scale, in Figure 2. The horizontal lines represent the mean ± one standard error for the last five observations. There is clearly a significant decrease in the counting rate, which is complete in about 10 min. Since the counter-body separation was 20 cm, it is apparent that the effect would be much more pronounced for the more usual arrangement of a short counter-to-body distance. The last five

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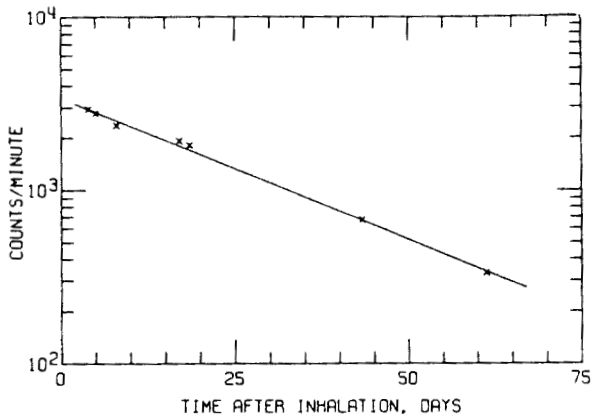

FIG. 1.--Retention of ^{51}Cr in subject JR as determined by the photopeak counting rate measured with two detectors. The first five points were measured at A.E.R.E., Harwell, the last two at ANL. The straight line corresponds to exponential decay with an effective half-life of 18.4 days. These data are provisional and subject to considerable revision.

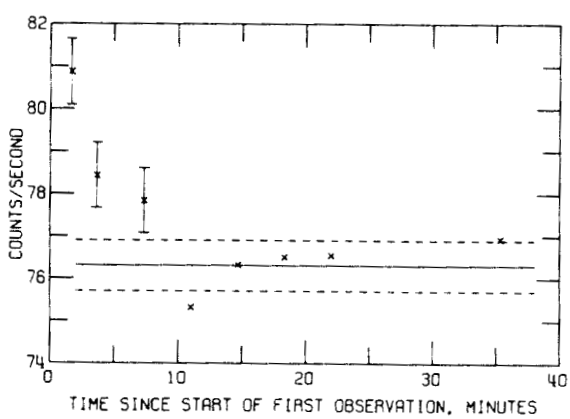

FIG. 2.--Effect of time spent under the counter on the observed counting rate for a detector with its edge 20 cm over the sternal notch. The horizontal solid line represents the mean of the last five observations; the dashed lines represent the levels of ± 1 standard error. (Subject JR.)

observations do not show significant deviations from the mean, but it is interesting to note the suggestion of an increase between 10 and 35 min.

Measurements of ^{103}Pd

The principal detector used for the measurement of the x rays from ^{103}Pd was the proportional counter with a 7-in diameter beryllium window 1 mm thick and operated with internal anticoincidence and pulse shape discrimination. Under these conditions, the counter background (15.6 to 23.9 keV) was 4.13 ± 0.09 count/min, while a typical subject gave an additional 1.6 counts/min. The energy resolution of this detector is illustrated by the spectrum in Figure 3 from the chest of the thinnest subject. The near resolution of the K_β line at 22.8 keV may be noted. Increased absorption in the heavier subjects has the effect of increasing the prominence of this line relative to the K_α peak. This spectrum was measured with the proportional counter

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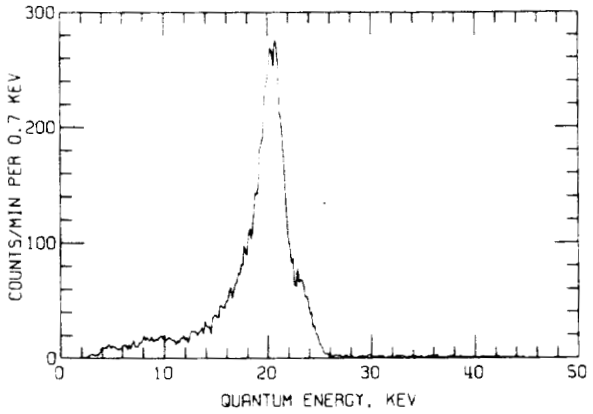

FIG. 3.--X-ray spectrum taken with the proportional counter centered over the sternum of subject DN. Note the very small contribution from the 320-keV quanta from ^{51}Cr.

centered over the sternum of the supine subject, a position which we have provisionally adopted as standard. Similar measurements on all three subjects were calibrated in the following manner.

The counting rate in the x-ray energy band was corrected for contributions from scattered and degraded gamma rays from ^{51}Cr, ^{40}K, and ^{137}Cs. The corrections for ^{51}Cr were determined from measurements of a source under various absorber thicknesses, while those for ^{40}K and ^{137}Cs were determined from measurements of the radiation from an uncontaminated subject of similar build.

Next, an effective tissue thickness was determined for each subject in the manner suggested by Rundo et al.,[3] and an experimental value was obtained for the attenuation coefficient for the x rays from ^{103}Pd (and one for those from ^{239}Pu), from measurements of their transmission through various thicknesses of tissue-equivalent material. The net counting rates from the subjects were then corrected for attenuation, and values for the ^{103}Pd contents were calculated from the response to a standardized source measured at a distance of 10 cm from the counter window. The results are set out in column 2 of Table 3, and they may be compared with the values in column 3 derived from our estimates of the ^{51}Cr contents and application of the known $^{103}Pd/^{51}Cr$ activity ratio. The agreement is only fair, but it is much better if we compare with the Harwell estimates of the ^{103}Pd content. In view of the rather crude method of calibration and of the limited experience

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we have had with the proportional counter, the results are gratifying.

We can derive provisional calibration factors for counting ^{239}Pu in subjects of the same sizes by calculation from the observed counting rates from ^{103}Pd in vivo. The reliability of the results depends on the accuracy of the intensities of the x rays emitted by ^{103}Pd and ^{239}Pu (we used values of 82%[7] and 4.6%[8]) and especially on the accuracy of the standardization of the ^{103}Pd sources. There is some question about the validity of the values used for the latter.[9] The results in column 5 of Table 3 should be assessed with these limitations in mind.

TABLE 3. Estimates of the ^{103}Pd Contents of the Three Subjects, and Approximate Counting Efficiency for ^{239}Pu, Derived from the Results in Column 3.

Subject	Estimated ^{103}Pd content, μCi	^{103}Pd content in μCi: ANL estimate of ^{51}Cr content	^{103}Pd content in μCi: Harwell ^{51}Cr estimate	Estimated counting efficiency for ^{239}Pu, c/m per μCi
DN	1.95	1.61	1.73	57
JR	0.69	0.49	0.67	22
KB	2.36	2.06	2.36	8.9

In calculating these results, we corrected for the different absorptions at the respective x-ray energies by interpolating in experimental attenuation curves at the appropriate effective tissue thicknesses, and for the higher counting efficiency for uranium L x rays.

Effects of Geometry

In the cases of subjects DN and KB, who were only available for limited times, we studied the effects of counter position and of arm position. The uncollimated proportional counter was centered over the mid-point of the sternum and the counting rate was observed alternately with the subject's

arms beside his body and with his hands under his head. Similar measurements were made with the counter displaced longitudinally 2 cm and 4 cm, both towards the vertex and towards the feet. The results are plotted in Figures 4 (DN) and 5 (KB), and there are some surprising differences between them. In the case of the lighter subject (DN), there are substantial (20 to 45%) increases in the counting rates at all positions, whereas the results for the heavier subject (KB) show only small increases at most positions and none at the mid-sternum. There are also striking differences between the shapes of the plots. For the lighter subject there is a peak in the response at mid-sternum, whereas the largest response in the case of the heavy subject is 4 cm nearer the head, and no maximum was observed.

Time permitted the determination of some "profile" curves with the proportional counter in the case of subject KB. The counter was shielded by two sheets of aluminum 5 cm from the window with a 2.5-cm wide gap between the sheets and along a diameter. A longitudinal scan (Figure 6) with the slit at right angles to the long axis of the body revealed a peak at 35 cm from the vertex, and a lateral scan (Figure 7) across the body with the slit parallel to the long axis showed the asymmetry expected from anatomical considerations.

Detailed Study of Emission from the Chest

In the case of subject JR, time permitted a detailed study of the emission of both the x rays and the gamma rays from the surface of the chest. The window of the phoswich was covered with lead 13 mm thick, which transmits less than 1% of 320-keV gamma rays, with a 5-cm square hole at the center. The counter was positioned 1 cm from the chest of the supine subject with the window parallel to the sternum. The first observation was made with the counter (and the hole) centered over the sternal notch, and no anticoincidence was employed, so that both x and gamma rays could be recorded. The counter was moved in steps of 5 cm to left and right of center, and this procedure was repeated at three other levels along the sternum, separated by 5 cm.

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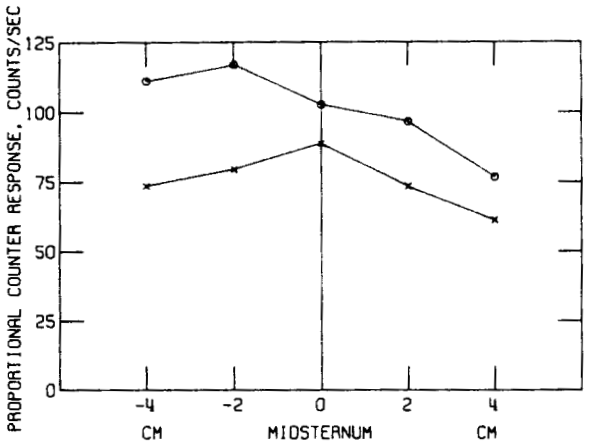

FIG. 4.--Longitudinal scans with the uncollimated proportional counter for subject DN with the arms beside the body (X) and with the hands under the head (O). The subject's head is to the left.

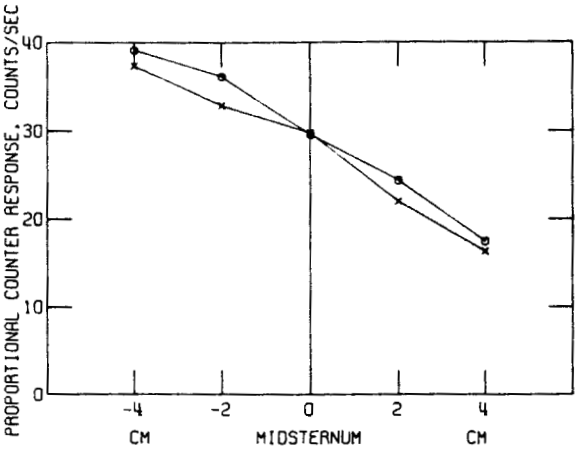

FIG. 5.--Longitudinal scans with the uncollimated proportional counter for subject KB with the arms beside the body (X) and with the hands under the head (O). The subject's head is to the left.

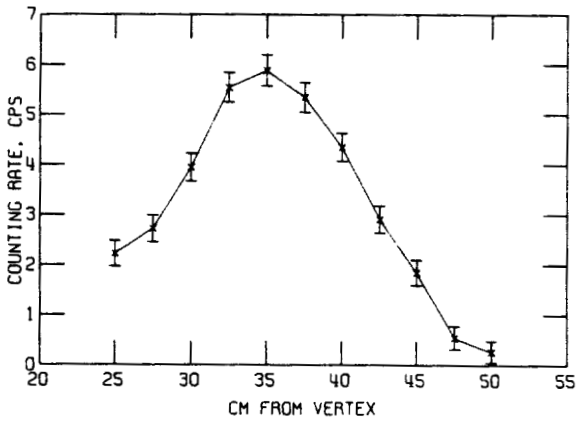

FIG. 6.--"Profile" curve for subject KB taken with the collimated proportional counter with a 1-in slit perpendicular to the long axis.

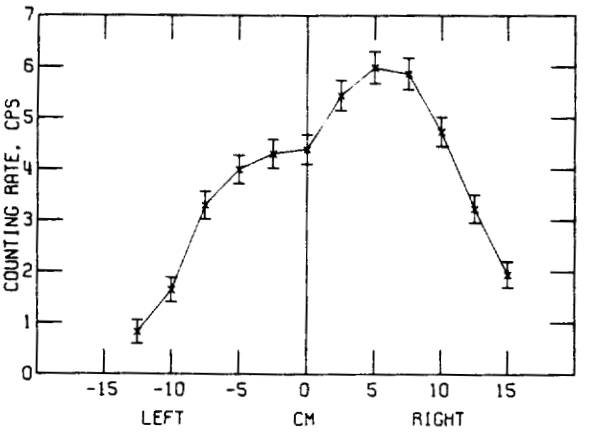

FIG. 7.--Lateral "profile" curve for subject KB at 35 cm from the vertex.

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The results of these measurements are presented in Figures 8 and 9 for the x rays and gamma rays. The data show some interesting similarities and differences. Thus, while the ^{51}Cr results show a broad but substantial peak at the level of the sternal notch, the corresponding scan for the ^{103}Pd is much flatter. The scans at the other three levels show asymmetry for both emitters, but it is more pronounced at the 5-cm level for the ^{51}Cr than for the ^{103}Pd.

The counting rate observed at any position is a function of both activity in the field of view and absorption in overlying tissue. The latter effect, is, of course, much more important for the ^{103}Pd than for the ^{51}Cr. To a first approximation, we can eliminate much of the effect of activity variations by examining the ratio of the counting rates from the two emitters. This ratio will have a strong dependence on absorption and a much smaller dependence on activity in the field of view. The results are plotted in Figure 10. In assessing this plot, we must bear in mind that the statistical uncertainty on each point is not negligible, amounting, as it does, to between 6 and 16%. The asymmetry in each of the plots of Figures 8 and 9 at the 5-, 10-, and 15-cm levels has largely disappeared and is replaced by a dip at 5 cm left of center over the heart, while the broad peak at the level of the sternal notch has become a broad depression. The most important conclusions we can draw from these plots are that the variations in counting rate per unit area (i.e., 25 cm^2) are not excessive over the 625 cm^2 studied, and that the variations in attenuation over this area are relatively small. It is of particular interest to note that, except over the heart, the increased counting rate observed from the right side, both in JR and KB (Figure 7), is due to variations in the amount of activity rather than in absorption. Thus, our choice of mid-sternum as a reference point for the placement of the 18-cm diameter propotional counter seems an entirely reasonable one.

We are grateful to the International Atomic Energy Agency and to the Health Physics and Medical Division of the Atomic Energy Research Establishmeth, Harwell, England, for the oportunity to participate in this experiment.

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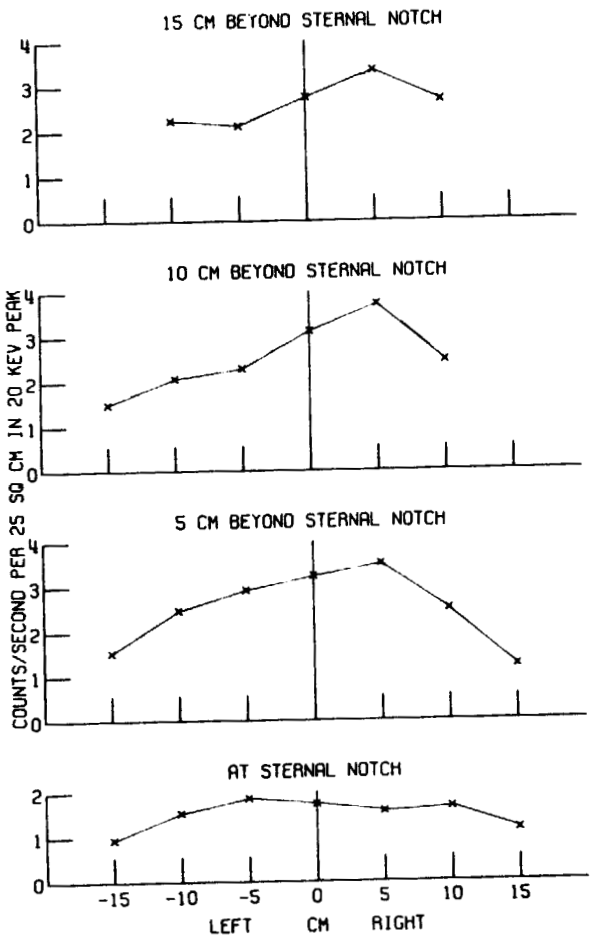

FIG. 8.--Area scan of ^{103}Pd emissions from chest of subject JR, taken with the phoswich shielded by 0.5 in of lead with a 5-cm square hole.

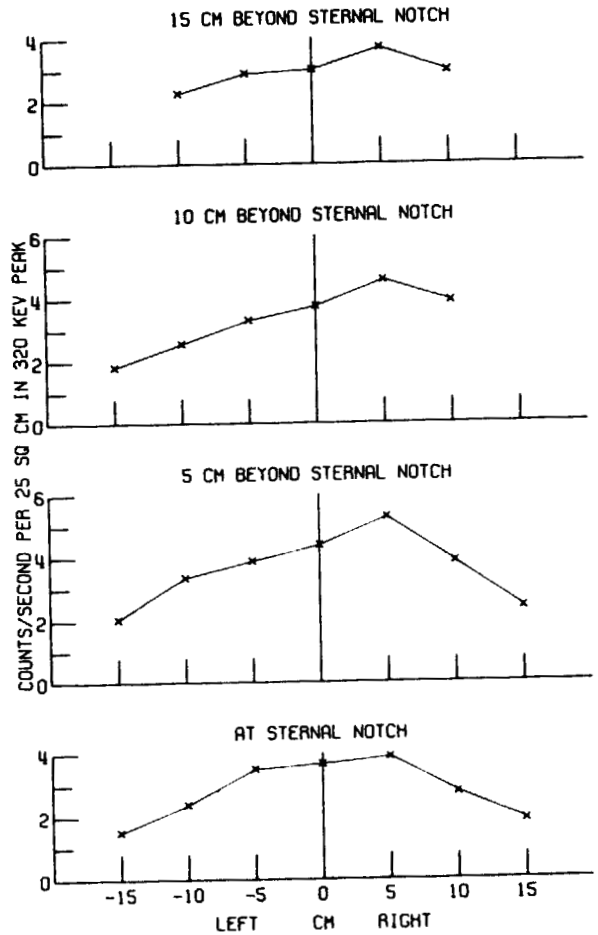

FIG. 9.--Area scan of ^{51}Cr gamma ray emissions from chest of subject JR, taken with the phoswich shielded by 0.5 in of lead with a 5-cm square hole.

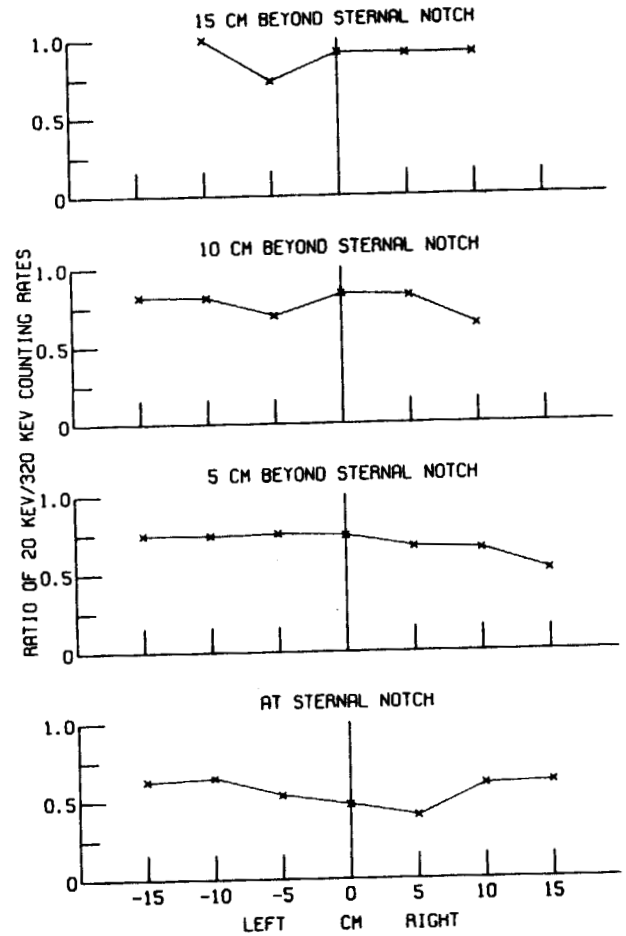

FIG. 10--The ratios of the counting rates from ^{103}Pd and ^{51}Cr taken from Figures 8 and 9. To a first approximation, the ratio varies inversely with the attenuation.

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