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INTRODUCTION

The human gamma-ray spectroscopy group, consisting of Charles E. Miller and supporting staff, was transferred from the Radiological Physics Division to the Health Division on January 1, 1964. The principal purpose of this transfer was to consolidate the management of the study of long-term effects of radium in man, currently under investigation as a cooperative project of Asher J. Finkel and Charles E. Miller (Health Division) and Robert J. Hasterlik (Argonne Cancer Research Hospital). In addition to this study, which is its principal effort, the human gamma-ray spectroscopy group will continue to concern itself with a number of other problems involving the measurement of internally deposited, gamma-ray emitting isotopes in human subjects, as well as with related studies involving experimental animals.

The results of some of these studies are summarized in this first semiannual report of the research activities of the Health Division. Some of the reports included here will include data in a form that is too detailed and extensive for journal publication but which may be of interest to other investigators working with similar problems.

The human gamma-ray spectroscopy group consists of Charles E. Miller and supporting staff (administrative assistant plus one technician). Mr. Ramesh K. Hukkoo of the Atomic Energy Establishment, Trombay, India participated in some of the work of the group from April through December 1964, first as a participant of the Institute of Nuclear Science and Engineering at Argonne National Laboratory, and later as a Resident Research Associate in the Health Division.

In addition to Dr. Robert J. Hasterlik (ACRH) whose participation in the radium studies is noted above, collaborative studies have also been done with Dr. Alexander P. Remenchik (Loyola University, Chicago) and Dr. Wayne V. Kessler (Purdue University, West Lafayette, Indiana).

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LONG-TERM EFFECTS OF RADIUM DEPOSITION IN MAN:
PROGRESS REPORT

A. J. Finkel, C. E. Miller, and R. J. Hasterlik*

The current set of studies has been under way since 1957, during which time approximately 250 patients have been examined. Of these 82% are former radium dial painters, 9% were given radium therapeutically by personal physicians, 7% were Elgin State Hospital (Illinois) patients, and 2% were radium chemists (Table 1). Basic studies have consisted of acquiring a medical, family, and occupational history; whole-body measurement of radium by gamma-ray spectroscopy; radiography of the entire skeleton; and blood counts. In the case of patients who are sufficiently interesting, especially those with a radium burden of over $0.1 \mu\text{Ci}$, these studies have been repeated at intervals of several years.

TABLE 1

Long-term effects of radium deposition in man

Body content, μCi	No. of subjects	Type of exposure to radium			
		Dial painters	Medical		Chemists
			Private physicians	State hospital	
A. Body content measured					
<0.001	30	30	0	0	0
0.001-0.01	42	42	0	0	0
0.01-0.1	105	98	5	0	2
0.1-1.0	65	47	7	9	2
>1.0	45	22	13	10	0
Total	287	239	25	19	4
B. Body content measured and skeleton radiographed					
<0.001	18	18	0	0	0
0.001-0.01	31	31	0	0	0
0.01-0.1	89	83	4	0	2
0.1-1.0	64	46	7	9	2
>1.0	45	22	13	10	2
Total	247	200	24	19	4

*Argonne Cancer Research Hospital, The University of Chicago

More recently, the use of a new sodium iodide crystal and a newly devised system of multiple crystal positions (described in the next paper of this report) has enlarged the scope of the whole-body measurements of radium, principally by yielding information on the gross radium distribution along the long axis of the body.

Skeletal radiographs, taken for the most part in the Health Division of Argonne National Laboratory, have been evaluated for changes typical of radium deposition by use of a set of criteria that enables the investigators to grade the severity of the changes in each bone by a semiquantitative technique (Table 2). Coding criteria have been worked out in cooperation with the group at the Massachusetts Institute of Technology, and they have been found to be useful by both groups for scoring the extent of skeletal damage as shown by radiography.

TABLE 2

Long-term effects of radium deposition in man:
Criteria for coding radiographic changes

0	No departure from normal.
±	Questionable, barely detectable changes, such as slight coarsening of trabeculation.
+	Minimal areas of bone resorption (equal to or less than 10 mm in longest diameter) in the cortex of long or flat bones, or Isolated areas of patchy bone sclerosis, or Definite coarsening of trabeculation.
++	Large punched-out areas of bone resorption (greater than 10 mm in longest diameter) in flat or long bones, or Lesions of + severity plus (a) some areas of bone sclerosis or (b) definite coarsening of trabeculation.
+++	Extensive changes of ++ severity plus (a) changes in trabecular pattern or (b) areas of bone sclerosis.
++++	Large areas of aseptic necrosis and/or pathological fracture.
++++	Malignant change.

Table 3 summarizes the present situation with respect to the correlation of the degree of radiographic change with the contemporary radium content of the patients. This table clearly demonstrates the increase in pathological findings when the current radium content exceeds $0.32 \mu\text{Ci}$, and especially when it exceeds $1.0 \mu\text{Ci}$.

TABLE 3

Summary of long-term effects of radium deposition in man
Correlation of clinical and radiographic findings
with current body burdens of Ra^{226}

Body content, μCi	Number measured	Number X-ray	Radiographic changes in skeleton attributable to radium deposition					
			None	Minimal	Mild	Moderate	Advanced	Malignant
<0.001	30	18	16	2	0	0	0	
0.001-0.0031	9	7	6	1	0	0	0	
0.0032-0.0099	33	24	23	1	0	0	0	
0.01-0.031	70	59	54	5	0	0	0	
0.032-0.099	35	30	25	4	0	1*	0	
0.10-0.316	32	32	22	5	1	2**	2†	(1)
0.32-0.99	33	32	4	8	8	5	7	(5)
1.0-3.16	34	34	2	2	5	4	21	(11)
3.2-5.5	11	11	0	0	0	1	10††	(6)
Total	287	247						

*Further studies pending.

** MsTh present in each case.

†Severe tooth changes only in one case.

††Based on films taken elsewhere in two cases.

The presence of minimal radiographic change attributable to radium deposition at very low levels of body radium, including two cases at the lower limit of detectability, suggests that the films are being overinterpreted. Certainly there are no changes of clinical importance in these patients with minimal radiographic changes nor even in those with mild changes. The boundary of clinical significance lies somewhere between the mild and the moderate changes as seen in the skeletal radiographs. With one doubtful exception that requires further study, radiographic changes of moderate severity have not been seen below $0.1 \mu\text{Ci Ra}^{226}$. The incidence of moderate and advanced changes, however, increases sharply when the current body burdens exceed $0.32 \mu\text{Ci Ra}^{226}$.

These data may be considered to provide an approximate long-term dose-response curve for Ra^{226} deposition, especially since there has been no trace of mesothorium except for two cases each of which has a complex medical history. The data in Table 3 are subject to further refinement as we acquire new data from continued study of these patients and as we gain an increased understanding of long-term radium retention patterns in man.

Preliminary reports of these findings were made at the 1959 meeting of the Radiation Research Society, at the Second International Congress of Radiation Research in 1962, at the U. S. Congressional Hearings in 1962 and at a New York Academy of Sciences symposium in 1963.

One of the most significant observations that has been made in connection with these studies in the past few years has been the rapidly

increasing incidence of epithelial tumors of the mastoids and of the paranasal accessory sinuses (Table 4). The development of these malignant neoplasms in some of our patients during the time that they have been under our periodic observations has prompted us to set up a prospective study of patients whose radium burden exceeds $0.5 \mu\text{Ci}$. Such persons are being followed by special radiographs of the skull and laminographs of the mastoids in a cooperative study with Drs. Nels M. Strandjord and Galdino E. Valvasori of the Department of Radiology, The University of Chicago. Periodic reexaminations of these patients are planned.

TABLE 4

Analysis of malignancies attributable to radium deposition

Case No	Current or terminal radium burden, μCi	Source of radium	Type of malignancy	Living or dead
03-685	0 167	occup	Carcinoma of maxilla	living
03-216	0 49	med	Osteoblastic osteogenic sarcoma, left femur	dead
03-455	0 81	occup	Fibrosarcoma, left radius	living
03-141	0 88	med	Epidermoid carcinoma, left mastoid	dead
03-417	0 89	occup	Squamous cell carcinoma, right maxillary antrum	living
03-402	1 2	occup	Fibrosarcoma, proximal left femur Epidermoid carcinoma, right mastoid	living
03-209	1 2	med	Fibrosarcoma, right scapula	dead
03-649	1 3	occup	Osteosarcoma of left ischium	dead
03-212	1 3	med	Osteosarcoma of left foot	dead
03-210	1 35	med	Osteogenic sarcoma, left calcaneus	dead
03-407	1 4	occup	Epidermoid carcinoma, right mastoid	dead
03-214	1 44	med	Carcinoma of left mastoid	living
03-619	1 6	occup	Osteosarcoma, left leg	dead
03-401	2 45	occup	Pleomorphic sarcoma, left leg	dead
03-648	2 5	occup	Mixed carcinosarcoma of right mastoid Osteosarcomas of femur and humerus	dead
03-105	2 6	med	Carcinoma of ethmoid or sphenoid sinuses	dead
03-201	3 2	med	Osteosarcoma, right humerus	dead
03-120	4 7	med	Carcinoma of mastoid	dead
03-584	6 0	occup	Osteogenic sarcoma, right pelvis	dead
03-213	6 8	med	Fibrosarcoma of lumbosacral spine	dead
03-675	-	occup	Rhabdomyosarcoma, right maxillary sinus	dead

In addition to the work in the Chicago area, assistance has been given to Dr. Pierre Wenger, Centre de Radioactivite Medicale, Geneva, Switzerland in setting up and calibrating a whole-body counter, and in measuring a series of 70 patients. A series of skeletal radiographs taken in Switzerland was also critically reviewed by our group.

At Argonne National Laboratory and at the Argonne Cancer Research Hospital, the current program, which is expected to be in effect for at least the next five years, basically consists of:

- 1) Continued search for hitherto unstudied subjects, and continued attempts to induce the participation in these studies of the many persons who are known to us but who have been uncooperative in the past.
- 2) Periodic restudy of those previously studied patients whose body content exceeds $0.1 \mu\text{Ci}$ at the present time. A systematic program for continuing study of these patients is of importance in view of the high incidence of pathological fractures, of aseptic necrosis of bone, and of malignant tumors of bone and epithelial linings of the mastoid air cells in patients who have current body burdens in excess of $0.5 \mu\text{Ci Ra}^{226}$.
- 3) Extension of whole-body measurement techniques by application of the newly developed single-crystal, multiple-position procedure (reported by Charles E. Miller at the May, 1964 International Atomic Energy Agency meeting at Heidelberg) to selected patients in order to correlate the extent of gross nonuniformity of radium deposition with the findings of skeletal radiography.
- 4) Assembly of the data collected during the past seven years for a series of reports.

In addition, plans have been made to cooperate with Henry F. Lucas (Radiological Physics Division) in short-term studies of selected radium patients to acquire metabolic data such as radon breath values, and blood, urine and fecal radium values. Bone specimens from radium patients have been made available to Robert E. Rowland, Richard B. Holtzman and Henry F. Lucas of the Radiological Physics Division for the special studies in which they are interested.

MULTIPLE-CRYSTAL-POSITION TECHNIQUE FOR WHOLE-BODY MEASUREMENTS

C. E. Miller

Whole-body counters have been traditionally used to measure radioactivity within persons without reference to internal distribution. In the case of bone seeking radioisotopes, such as radium, it is important to have some knowledge of at least the gross distribution within the skeleton in order to study the nature of long-term dose response. The techniques described here were developed to detect substantial nonuniformity in distribution of radium in patients who cannot or will not lie still for more than one hour. The method has also been applied to the study of other internal emitters such as Cs^{137} .⁽¹⁾

As an extension of the single-position, single-crystal technique, a single-crystal, multiple-position procedure was devised to reconstruct the gamma-ray profile along the long axis of the body without the use of a collimated crystal, which would require heavy shielding. The gamma-ray profile was simulated by computing the values for seven equally spaced point sources in the body that would give the same readings as those observed when the crystal is successively moved to seven positions along the body. The magnitude of each calculated source is equal to the amount of radioactivity in a corresponding one-seventh segment of the body, and the sum of the seven point sources is equal to the total-body content.

This procedure assumes that for each arbitrary segment of the body the content of the gamma-ray emitting radionuclide is the same as if it were concentrated in a point at the middle of the segment. In order to calculate the magnitude of these point sources in the body from the observed measurements, a standard source is measured in a set of Presdwood phantoms, and the two sets of counts obtained for selected photopeaks are entered into a set of simultaneous equations. These equations are solved by an electronic computer by use of a program for matrix inversion and solution of linear equations. These calculations provide a series of point sources that would give the same gamma-ray profile as that of the patient, and hence the amount in each segment of the body. Since the sum of the magnitudes of the gamma-ray emitting isotope in each of the seven segments of the body is equal to the patient's total content of that isotope, this procedure provides a whole-body count as well as an approximate description of the extent of uniformity of distribution of the isotope in the body.

Crystal Position and Calibration

The patient is placed either supine or prone on an unpadded Lucite slab and centered lengthwise under the crystal track. A large uncollimated NaI(Tl) crystal is suspended with the face of the crystal 30 cm above the surface of the slab. The patient's gamma-ray spectra are then recorded with the crystal placed in turn at seven equally spaced positions 24.5 cm apart along the patient's longitudinal axis. The crystal positions are numbered one to seven starting from the head. The fourth or center crystal position is directly over the center position of the slab or the same distance from the top of the patient's head and the soles of his feet.

The calibration spectra required for the analysis of these measurements are obtained as follows: Presdwood sheets 24.5 cm wide by 30 cm long are piled in seven contiguous stacks of the appropriate heights to approximate the longitudinal cross section of the patient (Figure 1). A calibrated source is then placed in the center of the first stack and the gamma-ray spectra are recorded with the crystal placed in turn at each of the seven positions. The source is then placed in turn in the center of each of the other six stacks and the corresponding seven spectra recorded for each source position.

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Figure 1. Diagram of the arrangement of Presdwood segments used as a phantom in calibration of the single-crystal, multiple-position system

Computations

The count rates under selected photopeaks in the gamma-ray spectra obtained from the seven measurements of the patient and from the 49 measurements of the phantom are substituted into the equations below and solved with the aid of an electronic computer.

$$\begin{aligned}
C_1 &= R_1 X_{11} + R_2 X_{12} + R_3 X_{13} + R_4 X_{14} + \dots + R_7 X_{17} \\
C_2 &= R_1 X_{21} + R_2 X_{22} + R_3 X_{23} + \dots + R_7 X_{27} \\
&\vdots \\
&\vdots \\
&\vdots \\
C_7 &= R_1 X_{71} + R_2 X_{72} + \dots + R_7 X_{77}
\end{aligned} \tag{1}$$

where

$C_1, C_2, \dots$ are the actual count rates obtained from the patient at the seven crystal positions denoted by the subscripts;

$X_{11}, X_{12}, \dots$ are the count rates obtained from the phantom measurements, with the first digit of the subscript denoting the position of the crystal and the second the location of the source; and

$R_1, R_2, \dots$ are the quantities of radionuclide at each of the seven positions that would yield the same radioactive profile as that observed in the patient.

The quantity R_1 then is the amount of activity in the one-seventh segment of the body corresponding to the head, R_2 the amount in the chest, etc. The sum of the seven R terms is consequently equal to the total-body content of the isotope.

Construction of the Calibration Phantom

The thickness of the Presdwood phantom used at all positions except position 2 does not influence the calculated values (R) greatly. If the face of the crystal is 30 cm above the slab and the source is always located in the center of the phantom, the count rate under the photopeak will increase about 10% as the phantom thickness is increased from 8 cm to 22 cm. With this crystal-to-slab spacing, the higher absorption present when the source is buried deeper in the thicker phantom is more than offset by the fact that the source is closer to the crystal and thus subtends a larger solid angle of the crystal.

The count rates (X_{34}, X_{35}) that are contributed from a source located in Presdwood sections next to and one beyond that underneath the crystal are 45% and 10%, respectively, of the count rate (X_{33}) with the source directly under the crystal at position 3. The presence of a phantom under the crystal that is thicker than the human subject will somewhat reduce this contribution from side positions but again does not introduce a significantly large error,

since it is a small fraction of the 45% or 10% contributions from the side segments. The thickness of the phantom sections would become important only if this technique were used to measure the amount of activity present in one segment of the body when the adjacent section contained perhaps ten times as much activity.

The construction of the phantom used in position 2 to represent the lung segment will not appreciably affect the total-body content determination but it will alter the apparent distribution along the body. If a 17-cm thick solid phantom is used instead of a lung-equivalent phantom in position 2 to determine the term X_{22} in the equations, the calculated value for R_2 will be about 20% high, while the calculated values for R_1 and R_3 will be about 10% low. The total-body content found by the sum of the seven calculated terms will be about the same under either condition. The interdependence of the terms in the equation is such that with the solid phantom the calculated contributions from R_2 which are subtracted from positions 1 and 3 are too high, and those for R_1 and R_3 are low by about one-half the amount that R_2 is high.

Crystal Height

The success of this technique depends upon the fact that the crystal is close to the body so that the count rate falls rapidly as the crystal is moved along the body away from the location of the radioactive deposit. The crystal-to-slab distance of 30 cm was arbitrarily selected as a distance that would accommodate all but the most obese patients. The 10% increase in count rate observed when the phantom thickness was increased from 8 to 22 cm might be reduced if the crystal were raised slightly. However, the crystal should not be raised too high or the count rate will become independent of crystal position and then any combination of sources in the phantom equal in magnitude to the total-body content will satisfy the equations.

Crystal Spacing

Ideally, a crystal spacing equal to one-seventh of the patient's height should be used when measuring a patient. However, this will entail the remeasurement of a phantom consisting of stacks of Presdwood of the same width as the crystal spacing. For routine operation, the use of a crystal spacing of 22.5 cm for patients ranging in height from 4 ft 11 in. (150 cm) to 5 ft 5 in. (165 cm) and a crystal spacing of 25.0 cm for patients ranging in height from 5 ft 5 in. (165 cm) to 6 ft 1 in. (185 cm) would provide satisfactory results. With the shorter set of crystal positions, the values found for R_1 and R_7 will be slightly low for patients shorter than 5 ft 2 in. (157.5 cm) since they do not reach the entire span of crystal positions and slightly high for those taller than this midpoint height since they project

slightly beyond the first and seventh crystal positions. Similar distortions will obtain with the longer crystal spacing for patients below or above 5 ft 8 in. (175 cm) in height.

Number of Crystal Positions

The number of crystal positions which can be used feasibly with this technique is determined by the change in count rate when the crystal is moved to an adjacent position. When the detector crystal is a squat right cylinder 8 in. in diameter by 4 in. thick, with the axis aligned vertically, the count rate does not decrease greatly if it is moved 12.25 cm to either side of the source. This is the position spacing that would be used with a 13-crystal-position technique. Under these conditions, small variations in the count rates obtained from the patient would introduce significant differences in the calculated values for each of the 13 calculated points. Consequently, seven or eight crystal positions are about optimum when the 8 in. by 4 in. crystal is used. A greater number of positions could be used if a crystal with a small diameter and a long axis were placed transversely to the long axis of the subject.

Crystal Size and Shape

Although any shape of crystal can be used in the 7-crystal-position technique, more accurate results can be obtained with a crystal of optimum shape. The optimum crystal would be a long, right cylindrical crystal with a relatively small diameter, that, when placed crosswise with respect to the body spans the width but not much of the length of the body. The count rate observed with this log-shaped, small-diameter crystal will vary rapidly as the crystal is moved a relatively short distance from a position directly over the source of the activity. Since the cross section of such a crystal as viewed from any point along the length of the body is constant, the relative shape of the spectrum is the same whether the activity is directly under the crystal or at some distance from the crystal along the body. This property is important if it is necessary to strip high energy gamma-rays from the spectrum in order to determine the distribution of a low energy emitter within the body.

A low-background NaI(Tl) crystal was especially designed to meet these optimum conditions. In order to provide good light collection and high energy resolution, a 6 in. diameter by 8 in. long crystal was chosen. Photomultiplier tubes were optically coupled to each end of the crystal by means of 6 in. diameter by 3 in. thick light pipes of pure sodium iodide. These blocks of pure sodium iodide absorb the gamma rays originating in the photomultiplier tubes and effect a low background. This crystal has been found to have a very good energy resolution and its gamma-ray background does not increase when photomultiplier tubes that contain large amounts of radioactivity are attached.

This log-shaped compound crystal has been found to be very advantageous when used with the subject in the standard tilting chair. This advantage derives from the fact that each part of the body is viewed by an identical cross section of crystal, whereas with the squat right cylinder the head is viewed by the side of the crystal and the abdomen and thighs by the face of the detector. In practice, the log-shaped crystal has been used both with the tilting chair and with the multiple-position techniques in the study of Ra^{226} -, Th^{232} -, and Cs^{137} - containing patients.

A 12 in. long sodium iodide crystal would yield a higher count rate and consequently more accurate results than the crystal described above, but it would have the disadvantages of increased weight and cost.

Discussion

The 7-crystal-position technique has been particularly useful in two types of studies. It has provided a quick first measurement of accidentally contaminated persons in order to determine the location and magnitude of radioactivity present in or on the body, and it has been used subsequently to determine if the material was being redistributed within the body. In the study of long-term effects of radium in human patients this technique has been used extensively to determine both the total-body content and the approximate distribution of radium. An additional advantage is that this technique can be calibrated quite accurately with point sources placed in Presdwood phantoms.

Data accumulated to date indicate that when the radioactivity is not uniformly distributed throughout the body, the total-body content found with the 7-crystal-position technique is more accurate than that found with other whole-body counter techniques. In the future an improved version of this technique will undoubtedly find wide usage as a total-body measurement technique. The improved version will probably utilize four crystals, each connected to a separate input of a split memory, multichannel analyzer in order to reduce the total time required to count the patient and the calibration spectra.

Reference

1. Miller, C. E. Retention and Distribution of Cs^{137} after Accidental Inhalation, Health Phys. 10: 1065-1070 (1964).

DISTRIBUTION OF RADIUM IN HUMANS

C. E. Miller

The study of the long-term effects of radium deposition in man has provided whole-body radium measurements and skeletal x-rays for 280 patients. The overall changes in the skeleton attributable to radium can be correlated with current or terminal body burdens of radium for the construction of a preliminary dose-response table (Table 3). This type of correlation is based on total-body measurements of radium although the fact that radium is not uniformly distributed in the skeleton has been known since 1933.⁽¹⁾ Obviously, a more detailed study of dose response could be made if information were available on the amount of radium in each region of the body, if not for each bone.

Unfortunately, detailed information on the distribution of radium in most of these patients may never be obtained. None of the patients contain enough radium to permit a detailed scan of their bodies within any reasonable period of time. The patient with the largest amount of radium contains about 1 μCi of RaB-RaC distributed throughout 7000 grams of bone or 3500 cc of skeletal tissue, i.e., about 300 pCi of RaB-RaC per cc. Although microradiographs would be invaluable, it would be unfeasible to make them of all bones in the body, even if entire skeletons should become available.

In order to determine which patients contain gross nonuniformities of distribution of radium in the body, each patient who contained above 0.1 μCi of Ra^{226} (0.03 μCi of RaB-RaC) has been invited to return to the Laboratory for remeasurement with the 7-crystal-position technique discussed in the previous paper. The gamma-ray spectra were recorded at each position for either four or ten minutes, depending upon the amount of radium present and the length of time that the patient would lie still. Although it is desirable to measure each patient for at least ten minutes at each position, some of the patients were either physically unable or unwilling to lie still for more than one hour. The patients were also remeasured with the tilting-chair technique in order to acquire radium retention data for the last seven of the 33 to 40 years since the radium had been administered or ingested.

These studies were done in an attempt to bridge the gap somewhat between whole-body measurements of radium and microradiography and autoradiography of small samples of selected bones. Not only is general nonuniformity of radium distribution detectable by these segmental measurements, but it may be possible to correlate the pathological findings noted by skeletal radiography with the data on radium contained in regions of the skeleton.

Calibration and Internal Consistency

The seven-segment phantom discussed previously was used to calibrate the system for radium measurements. The radium standard was placed in the center of the segments located under positions 1 to 5 but only 2 cm deep in positions 6 and 7 in order to approximate the location of the long bones of the legs.

In order to provide an internal test of the procedure, to test the phantom spectra against the human spectra, and to determine the statistical accuracy of the various point sources, the analysis was performed with the count rates in five different energy bands. These energy bands were: 75-275, 275-375, 525-675, 775-1,275 and 1,575-2,575 keV. The gamma rays that fall in the first two bands are mainly those that are scattered in the body, while the unscattered primary gamma rays fall in the higher energy bands. The magnitude of the seven sources determined on the basis of each of the first two bands would be considerably different from those found with the higher energy bands if the radium were in fact located at a different effective distance in the patient than in the phantom. Since the magnitudes of the seven sources found with the five energy bands agreed quite well with each other, there is justification for assuming that the situation in the patient can be simulated by a point source in the phantom.

Corrections for K^{40} and Cs^{137}

In order to correct for the count rate contributed to each energy band at each crystal position by the K^{40} and Cs^{137} in the body, a normal female control subject was measured with the 7-crystal-position technique. For each energy band at each position the count rate of the control subject was subtracted from the net count rate of the radium patient. The K^{40} and Cs^{137} in the body of the unexposed female contributes a count rate that is equivalent to 0.008 μCi of whole-body RaB-RaC in the first energy band, to 0.006 μCi in the second energy band, to 0.017 μCi in the third energy band, to 0.009 μCi in the fourth energy band, and to 0.002 μCi in the fifth band. Although these quantities may represent a small fraction of the total-body RaB-RaC, omission of this correction can introduce a significant error in the determination of the quantity of RaB-RaC at a particular position. In one patient whose total-body RaB-RaC was 0.08 μCi , the Cs^{137} and K^{40} contributed about 40% of the observed count rate in position 3.

Results and Discussion

Forty-two radium-bearing patients have been measured with both the 7-crystal-position technique and the tilting-chair technique. The mean percentage of the total-body radium found in each segment of the body for

these 42 patients was calculated to determine the average distribution in this group of patients. These mean values, which are given in Table 5, are remarkably close to the percentage of the skeleton in the segment under the crystal at each of these positions. These results suggest that for a large group the average distribution of radium in the skeleton is uniform.

TABLE 5
Comparison of proportion of radium found in
body segments with skeletal mass in each segment

Segment of body	Radium in segment mean % $\pm$ S E	Calculated per cent of skeletal mass in segment		Portions of body in segment
		From data of Spiers & Burch ⁽²⁾	From data of Krogman ⁽³⁾	
1	16.2 $\pm$ 1.1	18.4	20.4	Skull mandible
2	16.5 $\pm$ 0.8	16.4	20.1	Ribs and sternum clavicle scapula cervical and thoracic spine proximal 1/3 of humerus
3	10.0 $\pm$ 0.7	6.0	6.8	Lumbar spine distal 1/3 of humerus proximal 1/3 of radius and ulna
4	18.1 $\pm$ 1.2	17.4	17.2	Pelvis sacrum proximal 1/5 of femur distal 2/3 of radius and ulna
5	12.4 $\pm$ 0.8	13.7	13.1	Middle 3/5 of femur hands and wrists
6	15.7 $\pm$ 1.0	13.1	13.3	Distal 1/5 of femur proximal 3/4 of tibia and fibula
7	11.2 $\pm$ 0.8	14.8	9.1	Distal 1/4 of tibia and fibula feet
Total	100.1	99.8	100.0	

The mean values and per cent of body radium content in each segment for eight selected patients are given in Table 6. The right-hand columns give the total-body radium estimated by the 7-crystal-position technique and the tilting-chair methods. The left-hand columns give the calculated mean RaB-RaC contents in microcuries for each one-seventh segment of the body as well as the percentage values.

TABLE 6
Summary of measurements of selected patients containing radium

Patient		Crystal Position							Sum of 7 positions	Tilting chair	Tilting chair/ 7 positions
		1	2	3	4	5	6	7			
03-571	μ Ci	0.0499	0.0399	0.0250	0.0454	0.0377	0.0490	0.0413	0.2883	0.296	1.028
	%	17.3	13.8	8.8	15.8	13.1	17.0	14.3	100.0		
03-139	μ Ci	0.0271	0.0103	0.0140	0.0165	0.0309	0.0235	0.0189	0.1412	0.146	1.033
	%	19.2	7.3	9.9	11.7	21.9	16.7	13.4	100.0		
03-206	μ Ci	0.1553	0.0694	0.0336	0.0545	0.1602	0.2966	0.1568	0.926	0.978	1.056
	%	16.8	7.5	3.6	5.9	17.2	32.0	16.9	100.0		
03-423	μ Ci	0.0181	0.0313	0.0089	0.0227	0.0236	0.0494	0.0291	0.1832	0.185	1.009
	%	9.9	17.1	4.9	12.4	12.9	27.0	15.9	100.0		
03-114	μ Ci	0.0198	0.0794	0.0591	0.1343	0.0363	0.0228	0.0134	0.3650	0.323	0.885
	%	5.4	21.8	16.2	36.8	9.9	6.2	3.7	100.0		
03-115	μ Ci	0.0476	0.0421	0.0349	0.0859	0.0233	0.0142	0.0203	0.2682	0.228	0.854
	%	17.7	15.7	13.0	32.0	8.7	5.3	7.6	100.0		
03-110	μ Ci	0.0772	0.0317	0.0202	0.0436	0.0254	0.0172	0.0188	0.2343	0.196	0.836
	%	32.9	13.6	8.6	18.6	10.9	7.3	8.1	100.0		
03-402	μ Ci	0.1166	0.0721	0.0527	0.0734	0.0315	0.0644	0.0298	0.4406	0.369	0.837
	%	26.5	16.4	12.0	16.7	7.2	14.6	6.8	100.0		

The seven calculated percentages for patient 03-571 compare fairly well with the average percentage found for the group of 42 patients. This finding suggests that the radium in this patient is distributed more or less uniformly throughout the various bones of the skeleton, but it does not rule out large variations in concentrations at the microscopic level within these bones. In contrast, data from some of the patients show a very nonuniform distribution. For example, patients 03-423 and 03-206 have 27% and 32%, respectively, of their total-body activity in segment 6, while patients 03-114 and 03-115 have 36.8% and 32% in segment 4, and patient 03-110 has 33% in segment 1. These percentages are roughly two times the average percentages for these segments. On the other hand, some patients contain only one-third as much in some segments as would be predicted from the average case, for example, segment 3 of patient 03-206 and segment 6 of patient 03-115.

These data demonstrate that in some patients portions of the skeleton are being irradiated at rates that are at least twice the average rate that would be calculated from the total-body measurement. However, it is highly unlikely that this high concentration of radium in the skull of patient 03-110 is uniformly distributed throughout this segment, but most probably it is concentrated in some particular small volume. It might be significant that both patients 03-110 and 03-402, who contain 32.95% and 26.5% in segment 1 have developed tumors of the mastoid air cells, while patient 03-206 who contains more total radium in segment 1 but does not exhibit the nonuniform distribution has not developed this neoplasm.

The total-body measurements obtained by the 7-position technique and by the tilting-chair technique are in very good agreement for the first four patients in Table 6. However, the tilting-chair measurement is only 85% of the 7-position value for the remaining three patients. This difference can be explained at least in the case of the last two patients (03-110, 03-402) on the basis of the distribution of radium in the body. Here, where 26% to 33% of the radium is in the head, the squat right cylindrical crystal used with the tilting chair has a different geometry with respect to the head than with the remainder of the body since the head faces the 4 in. wide side of the crystal. However, when dose-response calculations are made, an error of 15% or 20% in the total-body determination is not particularly significant in view of the fact that some portions of the body contain two or more times the average concentration of the whole body.

The detailed results of the 7-crystal-position measurements for each of the 42 patients are given in Tables 7 to 52 in the Appendix. The RaB-RaC content found for each seventh segment of the body for each of the five energy bands that were used are given in columns 2-8 in the tables. The means and standard errors of the five quantities found for each segment with the five energy bands are also given. The total-body content of RaB-RaC found by summing the values found for the seven segments, and the means and

standard errors of the five totals are given in column 9. In order to afford a more direct comparison of the distribution between patients, the mean quantity found for each segment has been expressed as a percentage of the mean total in the bottom line.

Two tables are given for patients 03-110, 03-115, 03-121 and 03-417. The quantities obtained when the log crystal was used with the 7-crystal-position technique can be compared with values when the squat, right-cylindrical 8 in. by 4 in. crystal was used. The percentage distributions found with the two differently shaped crystals, which were independently calibrated, agree fairly well.

The marked nonuniform distribution of radium within the body given in Table 6 may be due to a difference in the resorption rate of radium in the various sections of the body. On the other hand, the radium may have been deposited in this nonuniform fashion at the time of ingestion. If this non-uniform distribution existed from the time of ingestion, whole-body radium measurements made of some of these patients over the past 30 years contain individual errors that may be as large as 200%, depending upon the measurement technique used. An electrometer was employed in the early gamma-ray measurements made from 1929 through 1935. The electrometer, which was placed a few centimeters from the patient's spinal column midway between the head and pelvis, would have been very sensitive to radium located in the mid-section of the trunk, considerably less sensitive to radium in the head and pelvis, and probably relatively insensitive to radium located in the long bones of the legs since they are shielded from the detector by the patient's trunk. Many of the measurements made from 1935 until 1956 employed a G-M tube or a NaI(Tl) crystal, which was placed about 10 cm behind the patient's lower spine. A detector so placed would be very sensitive to activity in the pelvis and considerably less sensitive to activity in the head and long bones of the legs. By contrast, the tilting-chair technique employed since 1956 is slightly more sensitive to radium in the legs and slightly less sensitive to activity in the pelvis and head.

When the measurements by the 7-crystal-position technique are used as standards, the 1931-1932 electrometer measurements on patient 03-114, who in 1964 contained 36.8% of the body radium in the pelvis, were probably low; the 1951-1953 NaI(Tl) crystal measurements were about 200% high; and the 1959-1964 tilting-chair measurements about 12% low. Consequently, the various measurements on these patients will have to be corrected for the distribution of radium in the body if these measurements are to be used to determine a mathematical expression describing the retention of radium.

References

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2. Spiers, F. W. and P. R. J. Burch. Measurements of Body Radioactivity in a Radium Worker. Brit. J. Radiol. Suppl. 7, 81-89 (1957).
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APPENDIX

The following Tables (Nos. 7 through 52) provide the details of the results of measuring 42 radium patients by the 7-crystal-position technique. As noted in the text of the preceding report, four patients were measured by this method with each of two scintillation crystal detectors. For these four patients both sets of measurements are summarized in separate tables. The data in the tables are given in detail in this semi-annual report in order to make them available to interested investigators in a form that would otherwise be too elaborate for final publication in the open literature.

TABLE 7

Case No. 03-101

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0864	0.920	0.0333	0.0958	0.0514	0.0564	0.0046	0.4202
275-375	0.1292	0.1075	0.0307	0.0968	0.0986	0.0219	0.0322	0.5169
525-675	0.1314	0.1059	0.0284	0.0968	0.0860	0.0360	0.0230	0.5075
775-1,275	0.1209	0.1035	0.0376	0.0777	0.0934	0.0404	0.0144	0.4882
1,575-2,575	0.1276	0.1152	0.0267	0.0976	0.0891	0.0320	0.0280	0.5163
Mean	0.1191	0.1048	0.0313	0.0929	0.0837	0.0373	0.0204	0.4898
S.E. of mean	± 0.0084	± 0.0038	± 0.0019	± 0.0038	± 0.0083	± 0.0057	± 0.0049	± 0.0182
Per cent in segment	24.3	21.4	6.4	19.0	17.1	7.6	4.2	100.0

TABLE 8

Case No. 03-102

Energy band keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0376	0.0201	0.0037	0.0404	0.0387	0.0314	0.0557	0.2276
275-375	0.0355	0.0163	0.0036	0.0293	0.0311	0.0249	0.0464	0.1871
525-675	0.0389	0.0203	0.0049	0.0336	0.0344	0.0245	0.0536	0.2102
775-1,275	0.0365	0.0196	0.0052	0.0354	0.0324	0.0267	0.0529	0.2087
1,575-2,575	0.0394	0.0155	0.0087	0.0289	0.0343	0.0334	0.0491	0.2093
Mean	0.0376	0.0184	0.0052	0.0335	0.0342	0.0282	0.0515	0.2085
S.E. of mean	± 0.0007	± 0.0010	± 0.0009	± 0.0021	± 0.0013	± 0.0018	± 0.0017	± 0.0064
Per cent in segment	18.0	8.8	2.5	16.1	16.4	13.5	24.7	100.0

TABLE 9

Case No. 03-110 (8 in. x 4 in. crystal)

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0615	0.0255	0.0308	0.0431	0.0212	0.0242	0.0138	0.2201
275-375	0.0694	0.0262	0.0280	0.0350	0.0232	0.0197	0.0164	0.2179
525-675	0.0747	0.0298	0.0292	0.0386	0.0221	0.0227	0.0163	0.2333
775-1,275	0.0638	0.0275	0.0269	0.0343	0.0184	0.0212	0.0169	0.2090
1,575-2,575	0.0819	0.0300	0.0278	0.0386	0.0243	0.0241	0.0176	0.2443
Mean	0.0703	0.0278	0.0285	0.0379	0.0218	0.0224	0.0162	0.2249
S.E. of mean	± 0.0037	± 0.0009	± 0.0007	± 0.0016	± 0.0010	± 0.0009	± 0.0006	± 0.0062
Per cent in segment	31.3	12.4	12.7	16.9	9.7	10.0	7.2	100.2

TABLE 10

Case No. 03-110 (log crystal)

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0621	0.0255	0.0208	0.0483	0.0177	0.0201	0.0162	0.2107
275-375	0.0784	0.0322	0.0200	0.0418	0.0280	0.0173	0.0179	0.2356
525-675	0.0841	0.0281	0.0233	0.0405	0.0264	0.0158	0.0186	0.2366
775-1,275	0.0788	0.0333	0.0199	0.0430	0.0271	0.0176	0.0187	0.2384
1,575-2,575	0.0824	0.0396	0.0173	0.0447	0.0282	0.0151	0.0229	0.2501
Mean	0.0772	0.0317	0.0203	0.0437	0.0255	0.0172	0.0189	0.2343
S.E. of mean	± 0.0039	± 0.0024	± 0.0010	± 0.0014	± 0.0018	± 0.0009	± 0.0011	± 0.0064
Per cent in segment	32.9	13.5	8.7	18.7	10.9	7.3	8.1	100.1

TABLE 11

Case No. 03-114

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0157	0.0706	0.0606	0.1414	0.0337	0.0183	0.0137	0.3541
275-375	0.0205	0.0834	0.0548	0.1300	0.0426	0.0220	0.0134	0.3665
525-675	0.0220	0.0782	0.0601	0.1277	0.0380	0.0222	0.0144	0.3625
775-1,275	0.0160	0.0860	0.0568	0.1386	0.0286	0.0299	0.0096	0.3655
1,575-2,575	0.0248	0.0787	0.0630	0.1340	0.0386	0.0216	0.0159	0.3766
Mean	0.0198	0.0794	0.0591	0.1343	0.0363	0.0228	0.0134	0.3650
S.E. of mean	± 0.0018	± 0.0026	± 0.0014	± 0.0026	± 0.0024	± 0.0019	± 0.0010	± 0.0036
Per cent in segment	5.4	21.8	16.2	36.8	9.9	6.2	3.7	100.0

TABLE 12

Case No. 03-115 (8 in. x 4 in. crystal)

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0346	0.0439	0.0353	0.0983	0.0179	0.0210	0.0140	0.2650
275-375	0.0393	0.0428	0.0292	0.0794	0.0192	0.0179	0.0172	0.2450
525-675	0.0434	0.0472	0.0352	0.0848	0.0182	0.0241	0.0166	0.2695
775-1,275	0.0356	0.0414	0.0276	0.0816	0.0113	0.0223	0.0130	0.2328
1,575-2,575	0.0426	0.0480	0.0284	0.0925	0.0148	0.0198	0.0188	0.2650
Mean	0.0391	0.0447	0.0311	0.0873	0.0163	0.0210	0.0159	0.2554
S.E. of mean	± 0.0018	± 0.0013	± 0.0017	± 0.0035	± 0.0014	± 0.0011	± 0.0011	± 0.0071
Per cent in segment	15.3	17.5	12.2	34.2	6.4	8.2	6.2	100.0

TABLE 13

Case No. 03-115 (log crystal)

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0398	0.0377	0.0365	0.0906	0.0150	0.0167	0.0190	0.2554
275-375	0.0459	0.0459	0.0297	0.0863	0.0269	0.0087	0.0241	0.2675
525-675	0.0517	0.0462	0.0332	0.0888	0.0253	0.0126	0.0202	0.2780
775-1,275	0.0483	0.0390	0.0432	0.0763	0.0287	0.0143	0.0204	0.2703
1,575-2,575	0.0524	0.0417	0.0317	0.0875	0.0205	0.0186	0.0176	0.2699
Mean	0.0476	0.0421	0.0349	0.0859	0.0233	0.0142	0.0203	0.2682
S.E. of mean	± 0.0023	± 0.0017	± 0.0024	± 0.0025	± 0.0025	± 0.0017	± 0.0011	± 0.0037
Per cent in segment	17.7	15.7	13.0	32.0	8.7	5.3	7.6	100.0

TABLE 14

Case No. 03-116

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0450	0.1157	0.1147	0.1543	0.0358	0.0492	0.0302	0.5449
275-375	0.0401	0.1101	0.0810	0.1170	0.0382	0.0428	0.0244	0.4536
525-675	0.0444	0.1180	0.0932	0.1193	0.0449	0.0432	0.0289	0.4919
775-1,275	0.0373	0.1192	0.0894	0.1250	0.0365	0.0471	0.0259	0.4804
1,575-2,575	0.0464	0.1168	0.1034	0.1199	0.0452	0.0432	0.0297	0.5046
Mean	0.0426	0.1160	0.0963	0.1271	0.0401	0.0451	0.0278	0.4951
S.E. of mean	± 0.0017	± 0.0016	± 0.0058	± 0.0069	± 0.0021	± 0.0013	± 0.0011	± 0.0150
Per cent in segment	8.6	23.4	19.5	25.7	8.1	9.1	5.6	100.0

TABLE 15

Case No. 03-121 (8 in. x 4 in. crystal)

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0052	0.0193	0.0083	0.0420	0.0270	0.0134	0.0098	0.1248
275-375	0.0064	0.0202	0.0088	0.0345	0.0243	0.0134	0.0099	0.1178
525-675	0.0061	0.0228	0.0098	0.0370	0.0249	0.0157	0.0106	0.1269
775-1,275	0.0062	0.0175	0.0106	0.0341	0.0215	0.0109	0.0109	0.1117
1,575-2,575	0.0044	0.0229	0.0097	0.0422	0.0204	0.0192	0.0091	0.1279
Mean	0.0057	0.0205	0.0094	0.0380	0.0236	0.0145	0.0101	0.1218
S.E. of mean	± 0.0004	± 0.0010	± 0.0004	± 0.0039	± 0.0012	± 0.0014	± 0.0003	± 0.0031
Per cent in segment	4.7	16.8	7.7	31.2	19.4	11.9	8.3	100.0

TABLE 16

Case No. 03-121 (log crystal)

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0064	0.0185	0.0064	0.0410	0.0227	0.0147	0.0074	0.1172
275-375	0.0055	0.0266	0.0062	0.0377	0.0289	0.0150	0.0073	0.1272
525-675	0.0059	0.0269	0.0048	0.0364	0.0356	0.0028	0.0147	0.1272
775-1,275	0.0069	0.0224	0.0127	0.0330	0.0347	0.0122	0.0081	0.1300
1,575-2,575	0.0054	0.0287	0.0074	0.0417	0.0212	0.0261	0.0135	0.1441
Mean	0.0060	0.0246	0.0075	0.0380	0.0286	0.0142	0.0102	0.1291
S.E. of mean	± 0.0003	± 0.0018	± 0.0014	± 0.0016	± 0.0030	± 0.0037	± 0.0016	± 0.0043
Per cent in segment	4.6	19.1	5.8	29.4	22.2	11.0	7.9	100.0

TABLE 17

Case No. 03-125

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0650	0.0149	0.0267	0.0234	0.0488	0.0070	0.0402	0.2583
275-375	0.0783	0.0199	0.0230	0.0274	0.0528	0.0077	0.0383	0.2474
525-675	0.0843	0.0194	0.0262	0.0239	0.0515	0.0071	0.0399	0.2523
775-1,275	0.0775	0.0264	0.0212	0.0252	0.0529	0.0092	0.0435	0.2559
1,575-2,575	0.0860	0.0170	0.0278	0.0355	0.0280	0.0331	0.0309	0.2583
Mean	0.0782	0.0195	0.0250	0.0271	0.0468	0.0128	0.0386	0.2544
S.E. of mean	± 0.0037	± 0.0019	± 0.0012	± 0.0022	± 0.0048	± 0.0051	± 0.0021	± 0.0021
Per cent in segment	30.7	7.7	9.8	10.7	18.4	5.0	15.2	97.5

TABLE 18
Case No. 03-139

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0215	0.0090	0.0114	0.0166	0.0240	0.0229	0.0204	0.1259
275-375	0.0295	0.0116	0.0112	0.0225	0.0296	0.0274	0.0163	0.1482
525-675	0.0309	0.0093	0.0148	0.0169	0.0337	0.0192	0.0206	0.1454
775-1,275	0.0281	0.0094	0.0136	0.0180	0.0264	0.0273	0.0191	0.1420
1,575-2,575	0.0252	0.0121	0.0188	0.0085	0.0410	0.0207	0.0180	0.1444
Mean	0.0270	0.0103	0.0140	0.0165	0.0309	0.0235	0.0189	0.1412
S.E. of mean	± 0.0017	± 0.0006	± 0.0014	± 0.0023	± 0.0030	± 0.0017	± 0.0008	± 0.0039
Per cent in segment	19.1	7.3	9.9	11.7	21.9	16.6	13.4	99.9

TABLE 19
Case No. 03-141

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0295	0.0454	0.0239	0.0421	0.0365	0.0723	0.0242	0.2740
275-375	0.0387	0.0461	0.0277	0.0379	0.0393	0.0745	0.0269	0.2912
525-675	0.0414	0.0513	0.0267	0.0401	0.0399	0.0778	0.0297	0.3071
775-1,275	0.0414	0.0517	0.0277	0.0448	0.0364	0.0820	0.0289	0.3130
1,575-2,575	0.0397	0.0482	0.0304	0.0372	0.0417	0.0747	0.0323	0.3041
Mean	0.0381	0.0485	0.0273	0.0404	0.0388	0.0763	0.0284	0.2979
S.E. of mean	± 0.0022	± 0.0013	± 0.0010	± 0.0014	± 0.0010	± 0.0017	± 0.0014	± 0.0070
Per cent in segment	12.8	16.3	9.2	13.6	13.0	25.6	9.5	100.0

TABLE 20
Case No. 03-201

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0776	0.1364	0.2099	0.2574	0.0396	0.1360	0.0592	0.9161
275-375	0.0964	0.1454	0.1913	0.2267	0.0490	0.1286	0.0728	0.9102
525-675	0.1023	0.1460	0.1897	0.2288	0.0422	0.1343	0.0765	0.9177
775-1,275	0.0942	0.1399	0.2085	0.2412	0.0395	0.1324	0.0824	0.9381
1,575-2,575	0.0939	0.1522	0.1871	0.2270	0.0420	0.1325	0.0827	0.9174
Mean	0.0929	0.1440	0.1973	0.2362	0.0424	0.1328	0.0747	0.9199
S.E. of mean	± 0.0041	± 0.0027	± 0.0049	± 0.0059	± 0.0017	± 0.0012	± 0.0043	± 0.0047
Per cent in segment	10.1	15.7	21.4	25.7	4.6	14.4	8.1	100.0

TABLE 21
Case No 03-206

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0 1300	0 0718	0 0276	0 0619	0 1537	0 2871	0 1369	0 8688
275-375	0 1542	0 0581	0 0321	0 0529	0 1458	0 2854	0 1464	0 8749
525-675	0 1654	0 0697	0 0394	0 0535	0 1590	0 3013	0 1668	0 9552
775-1,275	0 1623	0 0775	0 0321	0 0567	0 1705	0 2982	0 1805	0 9780
1,575-2,575	0 1647	0 0700	0 0367	0 0477	0 1719	0 3110	0 1535	0 9554
Mean	0 1553	0 0694	0 0336	0 0545	0 1601	0 2966	0 1568	0 9264
S E of mean	+0 0067	±0 0032	+0 0021	±0 0023	+0 0050	±0 0047	+0 0077	+0 0227
Per cent in segment	16 8	7 5	3 6	5 9	17 2	32 0	16 9	99 9

TABLE 22
Case No 03-214

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0 0697	0 0451	0 0608	0 1139	0 0304	0 0643	0 0418	0 4261
275-375	0 0783	0 0475	0 0579	0 0887	0 0360	0 0523	0 0516	0 4122
525-675	0 0869	0 0459	0 0584	0 0904	0 0368	0 0522	0 0571	0 4274
775-1,275	0 0853	0 0448	0 0677	0 0957	0 0350	0 0511	0 0583	0 4378
1,575-2,575	0 0867	0 0517	0 0531	0 1055	0 0244	0 0636	0 0528	0 4380
Mean	0 0814	0 0470	0 0596	0 0988	0 0325	0 0567	0 0523	0 4283
S E of mean	+0 0033	+0 0013	±0 0024	±0 0048	±0 0023	+0 0030	+0 0029	±0 0048
Per cent in segment	19 0	11 0	13 9	23 1	7 6	13 2	12 2	100 0

TABLE 23
Case No 03-402

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0 0934	0 0639	0 0532	0 0795	0 0277	0 0638	0 0217	0 4033
275-375	0 1193	0 0703	0 0526	0 0706	0 0349	0 0573	0 0325	0 4376
525-675	0 1217	0 0766	0 0510	0 0739	0 0310	0 0625	0 0337	0 4496
775-1,275	0 1228	0 0687	0 0569	0 0713	0 0294	0 0724	0 0297	0 4513
1,575-2,575	0 1257	0 0812	0 0498	0 0727	0 0347	0 0661	0 0312	0 4613
Mean	0 1166	0 0721	0 0527	0 0736	0 0315	0 0644	0 0298	0 4406
S E of mean	±0 0059	±0 0030	±0 0012	±0 0016	±0 0014	±0 0024	±0 0021	±0 0101
Per cent in segment	26 5	16 4	12 0	16 7	7 1	14 6	6 8	100 1

TABLE 24

Case No. 03-404

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0090	0.0375	0.0288	0.0622	0.0373	0.0403	0.0096	0.2247
275-375	0.0156	0.0392	0.0299	0.0559	0.0387	0.0428	0.0119	0.2339
525-675	0.0145	0.0406	0.0298	0.0550	0.0337	0.0477	0.0116	0.2328
775-1,275	0.0120	0.0435	0.0292	0.0647	0.0341	0.0501	0.0087	0.2422
1,575-2,575	0.0121	0.0460	0.0247	0.0651	0.0357	0.0488	0.0097	0.2421
Mean	0.0126	0.0414	0.0285	0.0606	0.0359	0.0459	0.0103	0.2351
S.E. of mean	± 0.0011	± 0.0015	± 0.0010	± 0.0022	± 0.0009	± 0.0019	± 0.0006	± 0.0033
Per cent in segment	5.4	17.6	12.1	25.8	15.3	19.5	4.4	100.1

TABLE 25

Case No. 03-405

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0321	0.0387	0.0117	0.0324	0.0182	0.0328	0.0204	0.1863
275-375	0.0402	0.0373	0.0094	0.0290	0.0174	0.0320	0.0238	0.1891
525-675	0.0426	0.0344	0.0117	0.0227	0.0178	0.0314	0.0248	0.1854
775-1,275	0.0423	0.0354	0.0147	0.0314	0.0181	0.0342	0.0264	0.2027
1,575-2,575	0.0422	0.0422	0.0076	0.0340	0.0177	0.0344	0.0277	0.2058
Mean	0.0399	0.0376	0.0110	0.0299	0.0178	0.0330	0.0246	0.1939
S.E. of mean	± 0.0020	± 0.0014	± 0.0012	± 0.0020	± 0.0001	± 0.0006	± 0.0012	± 0.0043
Per cent in segment	20.6	19.4	5.7	15.4	9.2	17.0	12.7	100.0

TABLE 26

Case No. 03-412

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0048	0.0146	0.0080	0.0155	0.0066	0.0091	0.0038	0.0625
275-375	0.0067	0.0164	0.0062	0.0138	0.0071	0.0084	0.0051	0.0638
525-675	0.0054	0.0163	0.0059	0.0121	0.0036	0.0098	0.0051	0.0583
775-1,275	0.0041	0.0184	0.0074	0.0170	0.0055	0.0097	0.0060	0.0682
1,575-2,575	0.0038	0.0193	0.0081	0.0169	0.0051	0.0099	0.0072	0.0702
Mean	0.0049	0.0170	0.0071	0.0151	0.0056	0.0094	0.0054	0.0646
S.E. of mean	± 0.0005	± 0.0008	± 0.0004	± 0.0009	± 0.0006	± 0.0003	± 0.0006	± 0.0021
Per cent in segment	7.6	26.3	11.0	23.4	8.7	14.6	8.4	100.0

TABLE 27

Case No. 03-417 (8 in. x 4 in. crystal)

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0177	0.0163	0.0121	0.0218	0.0448	0.0436	0.0266	0.1828
275-375	0.0166	0.0160	0.0099	0.0194	0.0311	0.0371	0.0218	0.1520
525-675	0.0220	0.0198	0.0140	0.0227	0.0420	0.0427	0.0297	0.1928
775-1,275	0.0218	0.0194	0.0147	0.0262	0.0404	0.0441	0.0314	0.1979
1,575-2,575	0.0231	0.0179	0.0143	0.0212	0.0464	0.0461	0.0314	0.2006
Mean	0.0202	0.0179	0.0130	0.0223	0.0409	0.0427	0.0282	0.1852
S.E. of mean	± 0.0013	± 0.0008	± 0.0009	± 0.0011	± 0.0027	± 0.0015	± 0.0018	± 0.0088
Per cent in segment	10.9	9.7	7.0	12.0	22.1	23.1	15.2	100.0

TABLE 28

Case No. 03-417 (log crystal)

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0219	0.0154	0.0153	0.0177	0.0574	0.0378	0.0285	0.1940
275-375	0.0243	0.0167	0.0149	0.0191	0.0504	0.0278	0.0283	0.1815
525-675	0.0301	0.0173	0.0188	0.0233	0.0566	0.0364	0.0297	0.2123
775-1,275	0.0278	0.0193	0.0154	0.0249	0.0532	0.0366	0.0336	0.2107
1,575-2,575	0.0290	0.0186	0.0117	0.0271	0.0543	0.0404	0.0259	0.2069
Mean	0.0266	0.0175	0.0152	0.0224	0.0544	0.0358	0.0292	0.2011
S.E. of mean	± 0.0015	± 0.0007	± 0.0011	± 0.0018	± 0.0013	± 0.0021	± 0.0013	± 0.0059
Per cent in segment	13.2	8.7	7.6	11.1	27.1	17.8	14.5	100.0

TABLE 29

Case No. 03-419

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0226	0.0408	0.0159	0.0342	0.0198	0.0312	0.0309	0.1954
275-375	0.0288	0.0457	0.0143	0.0305	0.0248	0.0270	0.0391	0.2102
525-675	0.0313	0.0470	0.0149	0.0293	0.0211	0.0306	0.0416	0.2158
775-1,275	0.0291	0.0436	0.0150	0.0347	0.0191	0.0341	0.0378	0.2134
1,575-2,575	0.0303	0.0431	0.0206	0.0289	0.0237	0.0284	0.0424	0.2174
Mean	0.0284	0.0440	0.0161	0.0315	0.0217	0.0303	0.0384	0.2104
S.E. of mean	± 0.0015	± 0.0011	± 0.0011	± 0.0012	± 0.0011	± 0.0012	± 0.0020	± 0.0040
Per cent in segment	13.5	20.9	7.7	15.0	10.3	14.4	18.3	100.1

TABLE 30

Case No. 03-423

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0141	0.0277	0.0077	0.0237	0.0230	0.0496	0.0237	0.1694
275-375	0.0188	0.0312	0.0087	0.0217	0.0247	0.0463	0.0307	0.1820
525-675	0.0205	0.0309	0.0089	0.0204	0.0210	0.0514	0.0288	0.1821
775-1,275	0.0174	0.0342	0.0099	0.0231	0.0258	0.0484	0.0326	0.1915
1,575-2,575	0.0194	0.0327	0.0095	0.0244	0.0236	0.0517	0.0298	0.1913
Mean	0.0180	0.0313	0.0089	0.0227	0.0236	0.0495	0.0291	0.1833
S.E. of mean	± 0.0011	± 0.0011	± 0.0004	± 0.0007	± 0.0008	± 0.0010	± 0.0015	± 0.0040
Per cent in segment	9.8	17.1	4.9	12.4	12.9	27.0	15.9	100.0

TABLE 31

Case No. 03-428

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0209	0.0352	0.0234	0.0429	0.0125	0.0207	0.0155	0.1711
275-375	0.0281	0.0406	0.0222	0.0429	0.0133	0.0207	0.0194	0.1872
525-675	0.0309	0.0377	0.0268	0.0391	0.0126	0.0196	0.0217	0.1684
775-1,275	0.0291	0.0386	0.0301	0.0414	0.0157	0.0191	0.0234	0.1973
1,575-2,575	0.0236	0.0496	0.0207	0.0467	0.0130	0.0215	0.0236	0.1989
Mean	0.0265	0.0403	0.0246	0.0426	0.0134	0.0203	0.0207	0.1846
S.E. of mean	± 0.0018	± 0.0025	± 0.0017	± 0.0012	± 0.0006	± 0.0004	± 0.0015	± 0.0064
Per cent in segment	14.4	21.8	13.3	23.1	7.3	11.0	11.2	102.1

TABLE 32

Case No. 03-429

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0386	0.0660	0.0277	0.0578	0.0796	0.1399	0.0771	0.4867
275-375	0.0551	0.0731	0.0346	0.0581	0.0874	0.1487	0.0973	0.5543
525-675	0.0580	0.0771	0.0351	0.0540	0.0891	0.1606	0.0985	0.5724
775-1,275	0.0545	0.0772	0.0374	0.0585	0.0953	0.1518	0.1048	0.5795
1,575-2,575	0.0535	0.0862	0.0326	0.0610	0.0891	0.1680	0.0976	0.5880
Mean	0.0519	0.0759	0.0335	0.0579	0.0881	0.1538	0.0951	0.5562
S.E. of mean	± 0.0034	± 0.0033	± 0.0016	± 0.0011	± 0.0025	± 0.0049	± 0.0047	± 0.0182
Per cent in segment	9.3	13.6	6.0	10.4	15.8	27.7	17.1	99.9

TABLE 33

Case No. 03-431

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0590	0.0671	0.0638	0.0949	0.0304	0.0471	0.0194	0.3817
275-375	0.0789	0.0698	0.0523	0.0854	0.0354	0.0469	0.0254	0.3941
525-675	0.0810	0.0716	0.0514	0.0862	0.0322	0.0491	0.0254	0.3969
775-1,275	0.0781	0.0714	0.0694	0.0909	0.0357	0.0437	0.0323	0.4215
1,575-2,575	0.0764	0.0774	0.0593	0.0932	0.0314	0.0540	0.0249	0.4166
Mean	0.0747	0.0715	0.0592	0.0901	0.0330	0.0482	0.0255	0.4022
S.E. of mean	± 0.0040	± 0.0017	± 0.0034	± 0.0019	± 0.0011	± 0.0017	± 0.0020	± 0.0074
Per cent in segment	18.6	17.8	14.7	22.4	8.2	12.0	6.3	100.0

TABLE 34

Case No. 03-433

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0466	0.0569	0.0303	0.0479	0.0393	0.0497	0.0396	0.3102
275-375	0.0533	0.0538	0.0280	0.0351	0.0429	0.0439	0.0438	0.3007
525-675	0.0596	0.0562	0.0345	0.0362	0.0417	0.0494	0.0497	0.3273
775-1,275	0.0572	0.0592	0.0314	0.0418	0.0431	0.0436	0.0545	0.3308
1,575-2,575	0.0640	0.0640	0.0344	0.0428	0.0438	0.0583	0.0537	0.3610
Mean	0.0561	0.0580	0.0317	0.0408	0.0422	0.0490	0.0483	0.3260
S.E. of mean	± 0.0029	± 0.0017	± 0.0012	± 0.0023	± 0.0008	± 0.0027	± 0.0029	± 0.0103
Per cent in segment	17.2	17.8	9.7	12.5	13.0	15.0	14.8	100.0

TABLE 35

Case No. 03-445

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.1846	0.0836	0.0821	0.1634	0.0374	0.0843	0.0043	0.6397*
275-375	0.1911	0.0544	0.0540	0.1115	0.0407	0.0569	0.0058	0.5144
525-675	0.1442	0.0945	0.0440	0.1248	0.0356	0.0626	0.0079	0.5136
775-1,275	0.1326	0.1072	0.0374	0.1418	0.0236	0.0706	0.0011	0.5143
1,575-2,575	0.1424	0.0960	0.0519	0.1318	0.0304	0.0584	0.0093	0.5202
Mean	0.1590	0.0871	0.0539	0.1347	0.0335	0.0666	0.0057	0.5404
S.E. of mean	± 0.0120	± 0.0090	± 0.0076	± 0.0087	± 0.0030	± 0.0050	± 0.0014	± 0.0248
Per cent in segment	29.4	16.1	10.0	24.9	6.2	12.3	10.6	109.5

*The radium values as calculated from the count rate in the 75-275 keV band are high owing to electronic noise pulses introduced into the analyzer from the power line during this series of measurements.

TABLE 36

Case No 03-459

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0 0784	0 0502	0 0444	0 0721	0 0281	0 0281	0 0125	0 3139
275-375	0 0841	0 0405	0 0334	0 0554	0 0294	0 0236	0 0117	0 2783
525-675	0 0896	0 0451	0 0332	0 0664	0 0197	0 0359	0 0046	0 2944
775-1,275	0 0822	0 0534	0 0322	0 0596	0 0290	0 0254	0 0088	0 2907
1,575-2,575	0 0956	0 0419	0 0391	0 0450	0 0422	0 0187	0 0117	0 2941
Mean	0 0860	0 0462	0 0364	0 0597	0 0297	0 0263	0 0099	0 2943
S E of mean	$\pm 0 0030$	$\pm 0 0024$	$\pm 0 0023$	$\pm 0 0047$	$\pm 0 0036$	$\pm 0 0028$	$\pm 0 0014$	$\pm 0 0057$
Per cent in segment	29 2	15 7	12 4	20 3	10 1	8 9	3 4	100 0

TABLE 37

Case No 03-462

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0 0164	0 0203	0 0041	0 0144	0 0106	0 0210	0 0216	0 1084
275-375	0 0139	0 0166	0 0046	0 0147	0 0101	0 0256	0 0137	0 0993
525-675	0 0176	0 0164	0 0034	0 0129	0 0106	0 0204	0 0163	0 0978
775-1 275	0 0193	0 0164	0 0044	0 0112	0 0082	0 0208	0 0179	0 0983
1,575-2,575	0 0164	0 0203	0 0041	0 0144	0 0106	0 0210	0 0216	0 1084
Mean	0 0167	0 0180	0 0041	0 0135	0 0100	0 0218	0 0182	0 1024
S E of mean	$\pm 0 0009$	$\pm 0 0009$	$\pm 0 0002$	$\pm 0 0007$	$\pm 0 0004$	$\pm 0 0010$	$\pm 0 0015$	$\pm 0 0024$
Per cent in segment	16 3	17 6	4 0	13 2	9 8	21 3	17 8	100 0

TABLE 38

Case No 03-484

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0 0490	0 0977	0 0701	0 1748	0 0406	0 0349	0 0127	0 4800
275-375	0 0639	0 1085	0 0668	0 1528	0 0519	0 0297	0 0219	0 4955
525-675	0 0686	0 1157	0 0627	0 1581	0 0421	0 0357	0 0174	0 5004
775-1,275	0 0654	0 1104	0 0833	0 1677	0 0416	0 0344	0 0186	0 5214
1,575-2 575	0 0689	0 1143	0 0649	0 1703	0 0373	0 0400	0 0213	0 5171
Mean	0 0632	0 1093	0 0696	0 1647	0 0427	0 0349	0 0184	0 5029
S E of mean	$\pm 0 0037$	$\pm 0 0032$	$\pm 0 0036$	$\pm 0 0040$	$\pm 0 0024$	$\pm 0 0016$	$\pm 0 0016$	$\pm 0 0075$
Per cent in segment	12 6	21 7	13 8	32 8	8 5	6 9	3 7	100 0

TABLE 39

Case No. 03-499

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0219	0.0078	0.0099	0.0115	0.0194	0.0197	0.0111	0.1013
275-375	0.0182	0.0063	0.0078	0.0047	0.0183	0.0099	0.0120	0.0772
525-675	0.0212	0.0084	0.0081	0.0103	0.0173	0.0146	0.0107	0.0906
775-1,275	0.0196	0.0073	0.0096	0.0080	0.0152	0.0150	0.0115	0.0862
1,575-2,575	0.0191	0.0082	0.0051	0.0078	0.0168	0.0110	0.0130	0.0810
Mean	0.0200	0.0076	0.0081	0.0085	0.0174	0.0140	0.0117	0.0873
S.E. of mean	± 0.0007	± 0.0004	± 0.0009	± 0.0012	± 0.0007	± 0.0017	± 0.0004	± 0.0042
Per cent in segment	22.9	8.7	9.3	9.7	19.9	16.0	13.4	99.9

TABLE 40

Case No. 03-510

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0142	0.0289	0.0158	0.0356	0.0347	0.0552	0.0320	0.2164
275-375	0.0177	0.0304	0.0185	0.0337	0.0317	0.0523	0.0359	0.2203
525-675	0.0203	0.0306	0.0172	0.0347	0.0277	0.0556	0.0405	0.2267
775-1,275	0.0188	0.0321	0.0185	0.0383	0.0285	0.0606	0.0389	0.2357
1,575-2,575	0.0167	0.0349	0.0166	0.0365	0.0306	0.0601	0.0353	0.2308
Mean	0.0175	0.0314	0.0173	0.0358	0.0306	0.0568	0.0365	0.2260
S.E. of mean	± 0.0010	± 0.0010	± 0.0005	± 0.0008	± 0.0012	± 0.0016	± 0.0015	± 0.0035
Per cent in segment	7.7	13.9	7.7	15.8	13.5	25.1	16.2	99.9

TABLE 41

Case No. 03-522

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0068	0.0047	0.0035	0.0095	0.0053	0.0098	0.0037	0.0434
275-375	0.0066	0.0056	0.0029	0.0066	0.0076	0.0072	0.0052	0.0417
525-675	0.0080	0.0036	0.0040	0.0087	0.0016	0.0094	0.0041	0.0394
775-1,275	0.0078	0.0039	0.0062	0.0060	0.0072	0.0076	0.0067	0.0454
1,575-2,575	0.0067	0.0069	0.0037	0.0080	0.0067	0.0077	0.0068	0.0466
Mean	0.0072	0.0049	0.0041	0.0078	0.0057	0.0083	0.0053	0.0433
S.E. of mean	± 0.0003	± 0.0006	± 0.0006	± 0.0006	± 0.0011	± 0.0005	± 0.0006	± 0.0013
Per cent in segment	16.6	11.3	9.5	18.0	13.2	19.2	12.2	100.0

TABLE 42

Case No. 03-530

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0129	0.0207	0.0109	0.0206	0.0171	0.0302	0.0129	0.1253
275-375	0.0182	0.0236	0.0140	0.0216	0.0214	0.0291	0.0187	0.1466
525-675	0.0201	0.0246	0.0140	0.0184	0.0200	0.0337	0.0166	0.1476
775-1,275	0.0220	0.0231	0.0147	0.0198	0.0236	0.0291	0.0208	0.1531
1,575-2,575	0.0207	0.0287	0.0143	0.0215	0.0243	0.0347	0.0175	0.1617
Mean	0.0188	0.0241	0.0136	0.0204	0.0213	0.0314	0.0173	0.1469
S.E. of mean	± 0.0016	± 0.0013	± 0.0007	± 0.0006	± 0.0013	± 0.0012	± 0.0013	± 0.0060
Per cent in segment	12.8	16.4	9.3	13.9	14.5	21.4	11.8	100.1

TABLE 43

Case No. 03-535

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0172	0.0059	0.0004	0.0083	0.0149	0.0176	0.0066	0.0709
275-375	0.0157	0.0042	0.0015	0.0046	0.0130	0.0134	0.0076	0.0599
525-675	0.0187	0.0042	0.0017	0.0028	0.0165	0.0147	0.0090	0.0676
775-1,275	0.0193	0.0049	0.0036	0.0044	0.0158	0.0169	0.0086	0.0736
1,575-2,575	0.0208	0.0055	0.0045	0.0025	0.0194	0.0167	0.0109	0.0802
Mean	0.0183	0.0049	0.0023	0.0045	0.0159	0.0158	0.0085	0.0704
S.E. of mean	± 0.0009	± 0.0003	± 0.0007	± 0.0010	± 0.0010	± 0.0008	± 0.0007	± 0.0034
Per cent in segment	26.0	7.0	3.3	6.4	22.6	22.4	12.1	99.8

TABLE 44

Case No. 03-551

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0550	0.0771	0.0647	0.0871	0.0422	0.0544	0.0238	0.4044
275-375	0.0563	0.0913	0.0737	0.0784	0.0482	0.0560	0.0323	0.4363
525-675	0.0488	0.1014	0.0718	0.0830	0.0508	0.0574	0.0355	0.4487
775-1,275	0.0118	0.0853	0.0800	0.0706	0.0563	0.0471	0.0426	0.3937
1,575-2,575	0.0030	0.0984	0.0766	0.0789	0.0496	0.0666	0.0327	0.4058
Mean	0.0350	0.0907	0.0734	0.0796	0.0494	0.0563	0.0334	0.4178
S.E. of mean	± 0.0114	± 0.0044	± 0.0026	± 0.0027	± 0.0023	± 0.0031	± 0.0030	± 0.0105
Per cent in segment	8.4	21.7	17.6	19.0	11.8	13.5	8.0	100.0

TABLE 45

Case No. 03-571

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0504	0.0411	0.0247	0.0485	0.0376	0.0484	0.0448	0.2954
275-375	0.0420	0.0381	0.0245	0.0473	0.0366	0.0507	0.0330	0.2723
525-675	0.0527	0.0389	0.0248	0.0429	0.0382	0.0489	0.0399	0.2864
775-1,275	0.0541	0.0401	0.0263	0.0400	0.0386	0.0487	0.0441	0.2919
1,575-2,575	0.0504	0.0411	0.0247	0.0485	0.0376	0.0484	0.0448	0.2954
Mean	0.0499	0.0399	0.0250	0.0454	0.0377	0.0490	0.0413	0.2883
S.E. of mean	± 0.0021	± 0.0006	± 0.0003	± 0.0017	± 0.0003	± 0.0004	± 0.0023	± 0.0043
Per cent in segment	17.3	13.8	8.7	15.7	13.1	17.0	14.3	99.9

TABLE 46

Case No. 03-586

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0511	0.0588	0.0132	0.0438	0.0432	0.0514	0.0336	0.2952
275-375	0.0648	0.0582	0.0120	0.0378	0.0505	0.0463	0.0387	0.3083
525-675	0.0680	0.0642	0.0089	0.0464	0.0446	0.0516	0.0386	0.3225
775-1,275	0.0606	0.0654	0.0182	0.0371	0.0480	0.0549	0.0362	0.3204
1,575-2,575	0.0684	0.0626	0.0136	0.0431	0.0419	0.0472	0.0429	0.3197
Mean	0.0626	0.0618	0.0132	0.0416	0.0456	0.0503	0.0380	0.3132
S.E. of mean	± 0.0032	± 0.0014	± 0.0015	± 0.0018	± 0.0016	± 0.0016	± 0.0015	± 0.0051
Per cent in segment	20.0	19.7	4.2	13.3	14.6	16.1	12.1	100.0

TABLE 47

Case No. 03-588

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0067	0.0218	0.0095	0.0243	0.0134	0.0175	0.0094	0.1027
275-375	0.0051	0.0193	0.0084	0.0244	0.0119	0.0161	0.0072	0.0924
525-675	0.0059	0.0236	0.0060	0.0214	0.0147	0.0164	0.0086	0.0967
775-1,275	0.0081	0.0213	0.0070	0.0195	0.0124	0.0164	0.0089	0.0937
1,575-2,575	0.0067	0.0218	0.0095	0.0243	0.0134	0.0175	0.0094	0.1027
Mean	0.0065	0.0216	0.0081	0.0228	0.0132	0.0168	0.0087	0.0976
S.E. of mean	± 0.0005	± 0.0007	± 0.0007	± 0.0010	± 0.0005	± 0.0003	± 0.0004	± 0.0022
Per cent in segment	6.7	22.1	8.3	23.4	13.5	17.2	8.9	100.1

TABLE 48
Case No. 03-685

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0064	0.0064	0.0029	0.0044	0.0045	0.0097	0.0076	0.0420
275-375	0.0089	0.0067	0.0032	0.0051	0.0044	0.0110	0.0082	0.0474
525-675	0.0088	0.0072	0.0022	0.0031	0.0026	0.0120	0.0072	0.0431
775-1,275	0.0079	0.0087	0.0040	0.0039	0.0069	0.0097	0.0102	0.0513
1,575-2,575	0.0091	0.0118	0.0020	0.0063	0.0050	0.0118	0.0090	0.0551
Mean	0.0082	0.0082	0.0029	0.0046	0.0047	0.0108	0.0084	0.0478
S.E. of mean	± 0.0005	± 0.0010	± 0.0004	± 0.0005	± 0.0007	± 0.0005	± 0.0005	± 0.0024
Per cent in segment	17.2	17.2	6.1	9.6	9.8	22.6	17.6	100.1

TABLE 49
Case No. 5025

Energy band, keV	Crystal positions							Total
	1	2	3	4	5	6	7	
75-275	0.0055	0.0056	0.0046	0.0042	0.0066	0.0069	0.0052	0.0385
275-375	0.0073	0.0070	0.0042	0.0043	0.0073	0.0081	0.0059	0.0442
525-675	0.0074	0.0068	0.0050	0.0046	0.0069	0.0085	0.0063	0.0457
775-1,275	0.0078	0.0061	0.0053	0.0042	0.0074	0.0076	0.0072	0.0457
1,575-2,575	0.0076	0.0063	0.0048	0.0037	0.0066	0.0099	0.0045	0.0434
Mean	0.0071	0.0064	0.0048	0.0042	0.0070	0.0082	0.0058	0.0435
S.E. of mean	± 0.0004	± 0.0003	± 0.0002	± 0.0001	± 0.0002	± 0.0005	± 0.0005	± 0.0013
Per cent in segment	16.3	14.7	11.0	9.7	16.1	18.9	13.3	100.0

TABLE 50
Case No. 5215

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0964	0.0863	0.0946	0.1260	0.0195	0.0254	0.0170	0.4652
275-375	0.1184	0.0937	0.0888	0.1112	0.0212	0.0251	0.0215	0.4797
525-675	0.1298	0.0957	0.0903	0.1155	0.0191	0.0292	0.0221	0.5017
775-1,275	0.1249	0.0957	0.0976	0.1236	0.0174	0.0306	0.0214	0.5112
1,575-2,575	0.1231	0.1036	0.0890	0.1196	0.0218	0.0257	0.0234	0.5061
Mean	0.1185	0.0950	0.0921	0.1192	0.0198	0.0272	0.0211	0.4928
S.E. of mean	± 0.0058	± 0.0028	± 0.0017	± 0.0027	± 0.0008	± 0.0011	± 0.0011	± 0.0087
Per cent in segment	24.0	19.3	18.7	24.2	4.0	5.5	4.3	100.0

TABLE 51

Case No. 5281

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0119	0.0441	0.0116	0.0462	0.0070	0.0114	0.0144	0.1467
275-375	0.0167	0.0423	0.0116	0.0389	0.0097	0.0090	0.0191	0.1472
525-675	0.0178	0.0458	0.0121	0.0397	0.0117	0.0056	0.0216	0.1544
775-1,275	0.0162	0.0467	0.0161	0.0427	0.0089	0.0111	0.0206	0.1623
1,575-2,575	0.0169	0.0446	0.0116	0.0430	0.0054	0.0138	0.0175	0.1528
Mean	0.0159	0.0447	0.0126	0.0421	0.0085	0.0102	0.0186	0.1527
S.E. of mean	± 0.0010	± 0.0008	± 0.0009	± 0.0013	± 0.0011	± 0.0014	± 0.0013	± 0.0028
Per cent in segment	10.4	29.3	8.3	27.6	5.6	6.7	12.2	100.1

TABLE 52

Case No. 5284

Energy band, keV	Crystal position							Total
	1	2	3	4	5	6	7	
75-275	0.0122	0.0181	0.0080	0.0116	0.0120	0.0183	0.0075	0.0878
275-375	0.0170	0.0198	0.0087	0.0105	0.0136	0.0190	0.0101	0.0986
525-675	0.0173	0.0207	0.0088	0.0113	0.0133	0.0178	0.0115	0.1007
775-1,275	0.0166	0.0205	0.0106	0.0105	0.0149	0.0141	0.0204	0.1076
1,575-2,575	0.0174	0.0208	0.0083	0.0108	0.0151	0.0191	0.0088	0.1004
Mean	0.0161	0.0200	0.0088	0.0109	0.0138	0.0177	0.0117	0.0990
S.E. of mean	± 0.0010	± 0.0005	± 0.0005	± 0.0002	± 0.0006	± 0.0009	± 0.0023	± 0.0032
Per cent in segment	16.3	20.2	8.9	11.0	13.9	17.9	11.8	100.0

RADIUM RETENTION IN MICE.

I. AFTER SINGLE INTRAVENOUS INJECTION

C. E. Miller and A. J. Finkel

The retention of intravenously injected radium was studied in mice by means of whole-body gamma-ray spectroscopy. This study was done in order to acquire some understanding of the radium retention pattern in a laboratory mammal with the hope that such information could eventually be applied to the interpretation of data acquired in our studies with human patients who ingested or were injected with radium 30 to 40 years ago. In particular, we were interested in the validity of the power function proposed by Norris, Tyler, and Brues⁽¹⁾ as a description of radium retention after intravenous injection and in the possibilities of extrapolating from mouse data to the human situation. We were also interested in evaluating the pattern of retention of radon daughters with time and in ascertaining whether the situation in mice paralleled that found by Mays, et al.⁽²⁾ for rats, dogs and man.

Ra^{226} and the radon daughters that are present in small animals can be determined by external gamma-ray spectroscopy without injury to the animal. Measurements of the same animal can be made repeatedly during its lifetime. The retained radon, which does not emit gamma rays, is determined by quantitative measurement of its short-lived daughters, RaB and RaC. The Ra^{226} content is determined by stripping the RaB and RaC spectra from the gross gamma-ray spectrum of the animal.

This report deals with an intensive study of four young adult female mice after each had received an intravenous injection of Ra^{226} bromide. Preliminary results, reported at the Radiation Research Society in 1961,⁽³⁾ encouraged us to pursue these studies further, and to apply electronic computer techniques in the analyses of the data. The subsequent investigations that followed, dealing with other aspects of radium metabolism in mice, will be described in forthcoming reports.

Methods

Procedures and Counting Geometry

Each mouse received an injection of Ra^{226} bromide in a lateral tail vein. The amount injected varied from 0.112 μCi to 0.29 μCi , but all the results have been normalized to 1.0 μCi Ra^{226} . After the injection each mouse was caged separately with food and water always available. During the first few days after the injection the animal was transferred to a fresh cage several times in order to minimize contamination of the fur by the

wood-shaving bedding material. Also, during the first few days, the urine was manually expressed from the bladder before each measurement in order to eliminate the radium-containing urine from the whole-body counts of retained radioactivity.

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Figure 2. Diagram of counting arrangement with mouse in Lusteroid tube positioned under 8 in. by 4 in. NaI(Tl) crystal.

For these measurements the mouse was placed in a perforated Lusteroid tube, 2.6 cm diameter by 10 cm long, and the tube was corked and bolted into a plastic dish, 15 cm in diameter by 5 cm deep. This assembly was clamped against the beta filter under the face of an 8 in. diameter by 4 in. thick NaI crystal (see Figure 2). A mechanical fixture (not shown in the figure) accurately positioned the Lusteroid tube with respect to the 8 in. by 4 in. crystal. The gamma-ray spectra were recorded by means of a 400-channel analyzer with a punched paper tape output.

With this geometry the count rate is not dependent upon the position of the mouse in the tube, and

replicated measurements of the Ra^{226} content of the same mouse agreed within ± 25 picocuries of the average value.

Calibration Procedure

The counting system was calibrated for the separate measurement of Ra^{226} , RaB and RaC in mice by the following procedure. About $0.1 \mu\text{Ci}$ of Ra^{226} was injected into one of the lateral tail veins of a mouse, and a statistically accurate gamma-ray spectrum of the mouse was recorded about 400 minutes later. From work to be described later in this report it was known that by this time the RaB and RaC had decayed to about 0.2 per cent of the equilibrium value and only the 188-keV gamma ray emitted by Ra^{226} was readily discernible in the gamma-ray spectrum. The mouse was killed and sealed in a thin-walled glass tube of the same dimensions as the Lusteroid tube, and a statistically accurate gamma-ray spectrum was recorded immediately. The count rate of Ra^{226} under the 188-keV gamma photopeak of the two spectra demonstrated that the absorption by the glass tube was the same as that by the Lusteroid tube.

Since the Ra^{226} content of the sealed mouse remains constant, it is possible to obtain pure spectra for the RaB and RaC that grow in and reach equilibrium in 30 days. For this purpose the gamma-ray spectra of the

sealed mouse were recorded frequently during the next 30 days as the RaB and RaC grew into equilibrium with the Ra²²⁶. The original spectrum that had been obtained immediately after the mouse had been sealed, and which represented 99.8 per cent pure Ra²²⁶ and 0.2 per cent RaB-RaC, was subtracted from the spectrum obtained on the 30th day in order to obtain a net spectrum for RaB and RaC. This composite RaB and RaC spectrum in the band from 225 keV to 1.35 MeV was then normalized for the same count rate as that in the same band recorded immediately after the mouse had been sealed. This normalized RaB-RaC spectrum was then subtracted from the original spectrum at 400 minutes to correct for the small amount of RaB and RaC still present at that time and thereby to obtain a pure Ra²²⁶ spectrum (A in Figures 3 and 4). The normalized RaB-RaC spectrum was also added to the composite RaB-RaC spectrum to correct for the 0.2 per cent that had been subtracted along with the Ra²²⁶ component when the composite RaB-RaC spectrum was obtained. These manipulations provided pure calibration spectra for Ra²²⁶, and for RaB and RaC.

Figure 3. Diagram of gamma-ray spectrum of Ra²²⁶ and radon daughters, RaB and RaC. Cross-hatched portion, A, represents the contribution to the 188-keV photopeak from Ra²²⁶, while D represents the degraded gamma rays from RaB and RaC. Portion B represents primary gamma rays from RaB plus degraded gamma rays from RaC. Portion C represents gamma rays from RaC.

Figure 4

Diagram of net gamma-ray spectrum of Ra^{226} . The cross-hatched section, A, is the portion used to calculate the amount of Ra^{226} that is present.

In order to test the accuracy of the analysis, the new equilibrium RaB-RaC spectrum was normalized, in turn, for the spectra obtained during the intervening days between injection and equilibrium and subtracted from them. The resultant spectra obtained on various days should contain the identical count rate under the 188-keV photopeak for Ra^{226} . After several unsuccessful series of sealed mouse measurements, it was found that in order to obtain the same results for these intervening days, it was necessary to keep the mouse carcass frozen at 0°F (-14°C) so that radon did not diffuse from the carcass and RaB and RaC were not deposited on the walls of the glass tube. Although the RaB and RaC present on the glass walls were counted with almost the same efficiency as the RaB and RaC present in the carcass, when the sealed mouse was kept at room temperature the RaB and RaC count rates under the 188-keV Ra^{226} photopeak changed sufficiently to introduce at least a 10 per cent error in Ra^{226} as equilibrium was approached.

The composite equilibrium RaB-RaC spectrum was also separated into RaB and RaC spectra. Pure RaC was separated from a radium solution, injected into a mouse, and the RaC spectrum recorded. This spectrum was normalized to match the composite RaB-RaC spectrum from

425 keV to 1.35 MeV and subtracted to obtain a pure RaB spectrum. Pure gamma-ray spectra for the same quantity of Ra²²⁶, RaB and RaC, were thus obtained.

To simplify the calculations of Ra²²⁶, RaB and RaC, the count rates in only four energy bands have been used instead of the complete spectrum. The first band, from 128 keV to 225 keV, includes the 188 keV peak from Ra²²⁶ (Figure 3 A) and degraded gamma rays from RaB and RaC (Figure 3 D). The second band (Figure 3 B), from 225 keV to 425 keV, includes the primary gamma rays from RaB plus degraded gamma rays from RaC. The third energy band (Figure 3 C) and the fourth (not shown), 425 keV to 767 keV, and 767 keV to 1.35 MeV, respectively, include only the gamma rays emitted by RaC. The count rates contributed to all four energy bands by 1.0 μ Ci of RaC, the count rates contributed to the first two bands by 1.0 μ Ci RaB, and the count rate contributed to the first band by 1.0 μ Ci Ra²²⁶ were accordingly determined by appropriate manipulations of the spectra as outlined above. These calculations provided calibration constants for Ra²²⁶, RaB, and RaC for each energy band to which these radionuclides contributed gamma rays.

Analysis of Measurements

The spectra obtained during the first 400 minutes after the mice were injected, at which time the RaB and RaC were not in secular equilibrium, were analyzed as follows. The μ Ci of RaC present in the mouse were determined from the count rate in the two higher energy bands specified in the preceding paragraph. The count rate contributed by this amount of RaC was then subtracted from the second energy band. The net count in the second band now consisted entirely of the contribution from RaB. The separate count rates of the RaB and RaC were then subtracted from the first energy band to leave a net value for Ra²²⁶.

Since analysis of spectra obtained more than 400 minutes after injection had demonstrated that the RaB and RaC contents were in secular equilibrium, the analyses after 400 minutes were further simplified. For the sealed mouse, the count rate from RaB was added to the count rate from RaC in the second energy band to give a new calibration constant for RaB and RaC in secular equilibrium. This calibration constant was used to convert the second band count rate from the live mouse into a μ Ci value for RaB and RaC. The μ Ci values for RaC were derived from the third and fourth bands by the method given in the preceding paragraph. The quantities from the second, third, and fourth bands were averaged to give a mean value for RaB-RaC. Although any one energy band alone or the sum for all three energy bands could have been used to determine the amount of RaB and RaC present, the three individual values were always calculated to provide an internal check of the overall system. The mean RaB-RaC value was then subtracted from the first energy band to give the net Ra²²⁶ content.

The entire set of calculations, including the subtraction of the gamma-ray background of the whole-body counter, and the calculation of RaB, RaC and Ra²²⁶ values, were performed by the ANL-built computer, GEORGE. The analysis, including the time to read the program into GEORGE and to print the computer output required less than one minute of computer time for each mouse measurement, which contained data collected in 400 channels.

Results and Discussion

Retention of Ra²²⁶ and Radon Daughters

Two mice (Nos. 52 and 53) received intravenous injections of about 0.1 μ Ci Ra²²⁶ that had been drawn directly into a syringe from a rubber-capped vial of radium bromide solution maintained at room temperature. At the time of the first measurement, 20 minutes after injection, the RaB and RaC contained by the mouse was about 40 per cent of the equilibrium value with Ra²²⁶. These radon daughters decreased for the first 300 minutes after injection, and then slowly increased to a peak at about 27 days after which they gradually dropped in value (Figure 5). RaB reached a minimum at about 300 minutes, while RaC fell more slowly and reached its minimum at about 400 minutes, after which both isotopes were in equilibrium with each other. This first portion of the RaB retention curve falls along a straight line on a semilog plot, with intercept values at the time of injection of 62.0 per cent and 44.0 per cent, respectively, for Mouse 52 and Mouse 53. The rapid decline of the radon daughters is an expression of both physical decay and excretion. The minimum value of about 0.25 per cent of the equilibrium value with Ra²²⁶ represents the ability of the mouse to "de-emanate" the injected radium solution of the radon and its daughter products. The rise after 400 minutes presumably represents the increase in retained radon daughter products from radium deposited in the bone.

In order to test the ability of the mouse to rid itself of administered RaB and RaC, two additional mice (Nos. 54 and 55) were given an intravenous injection of a radium bromide solution that had been de-emanated by bubbling air through it overnight (Figure 6). At the time of the first measurement 20 minutes after injection, the RaC was 0.7 to 0.8 per cent, and RaB was 0.3 to 0.6 per cent, of the equilibrium value with Ra²²⁶. Both radon daughters then dropped to a minimum of about 0.18 per cent of the injected radium, this time at about 200 minutes, and the superiority of the mouse over simple physical procedures for purposes of de-emanating a radium solution was again demonstrated. For both mice a semilog plot of the RaB data for the first 200 minutes showed an intercept value of 1.1 per cent, which represents the percentage of radon daughters present in the solution at the time of injection.

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Figure 5. Double-log plot of retention of Ra^{226} and of RaB and RaC by Mouse 52 and Mouse 53 after single intravenous injection. Data normalized to an injection of $1.00 \mu\text{Ci } \text{Ra}^{226}$.

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Figure 6. Double-log plot of retention of Ra^{226} and of RaB and RaC by Mouse 54 and Mouse 55 after single intravenous injection of "de-emanated" solution. Data normalized to an injection of $1.00 \mu\text{Ci}$ Ra^{226} .

The values for retained Ra^{226} did not alter appreciably for the first 200 to 300 minutes, after which they dropped slowly but steadily (Figures 5 and 6). The detailed data are presented in the Appendix in Table 60 for the delectation of those members of the scientific community who enjoy re-working data collected by others.

Ra^{226} Retention Considered as a Power Function

Norris, Tyler and Brues,⁽¹⁾ and earlier Norris, Speckman and Gustafson,⁽⁴⁾ suggested that radium retention after the first day is best described by a power function, $R = at^b$, where R is retention, t is time, and a and b are parameters. The slope, b , is independent of time units although a , the value of R when $t = 1$, depends on the units of time that are used. The mouse data, illustrated in Figures 5 and 6, indicate that the Ra^{226} values do fall along a straight line starting at about 150 minutes after injection although there is a slight but definite break in the slope along this line at about 27 days.

The power function parameters for the data for each of the mice are given in Table 53. These values were obtained by a least-squares fit

TABLE 53

Power function description of radium retention by four mice.
Linear fit to logarithmic transformation of the data*

Mouse No.	Number of measurements	One-minute intercept	One-day intercept	Slope $\pm$ S.E.	Elapsed time to final measurement days
A. Single straight line from 150 minutes after injection to death					
52	41	3.607	0.6463	-0.2364 ± 0.00068	436
53	31	2.753	0.6619	-0.2053 ± 0.00055	253
54	37	4.346	0.5683	-0.2798 ± 0.00074	407
55	32	4.146	0.6004	-0.2656 ± 0.00084	199
Combined data	141	3.626	0.6067	-0.2458 ± 0.00039	
B. Straight line from 150 minutes to 39,000 minutes (27.08 days)					
52	25	2.623	0.6616	-0.1910 ± 0.00121	
53	17	2.313	0.6236	-0.1802 ± 0.00093	
54	25	3.448	0.5632	-0.2492 ± 0.00054	
55	24	3.498	0.5966	-0.2432 ± 0.00047	
Combined data	91	3.286	0.6050	-0.2327 ± 0.00059	
C. Straight line from 39,000 minutes (27.08 days) to death					
52	16	6.921	0.8287	-0.2919 ± 0.00332	436
53	14	3.456	0.6677	-0.2261 ± 0.00230	253
54	12	8.374	0.7230	-0.3368 ± 0.00346	407
55	8	10.705	0.8331	-0.3511 ± 0.00464	199
Combined data	50	7.384	0.7974	-0.3060 ± 0.00444	

*Data normalized to an injection of 1.00 μCi Ra^{226} .

of the straight line, $\log R = \log a + b \log t$, to the data normalized for an injection of $1.00 \mu\text{Ci}$. Both the one-minute and the one-day intercepts are given in actual values (not logarithms), and the value for the slope in each case is followed by its standard error. The parameters have been calculated for individual mice and again for the four mice combined.

The first portion of the table (Section A) gives the values for a single straight line, on a double-log plot, for the data from 150 minutes after injection until death of the mouse. The slope for all mice combined is -0.2458 ± 0.0004 , and the one-minute intercept, which is a mathematical construct since the curve does not become a straight line until after 150 minutes, is 3.626. The value for the one-day intercept implies that 61% of the injected dose is still retained by the mouse at the end of one day (1440 minutes after injection).

Since graphic analysis indicated a definite break in the log-log plot at 27 days, the data were recalculated for the two periods defined by this break. Section B of the table gives the data for the first portion of the curve, from 150 minutes after injection to 39,000 minutes. The parameters are slightly different so that radium retention over this range is given by $R = 3.286 t^{-0.2327}$, when time is measured in minutes, or $R = 0.6050 t^{-0.2327}$ when the time units are days. The parameters for retention after 39,000 minutes are based on somewhat fewer measurements (Section C) but the slope is appreciably steeper so that $R = 7.384 t^{-0.3060}$. The difference between these two slopes is highly significant statistically, with $P \ll 0.0001$.

The data for Mouse 54, which lived 407 days after injection, can be graphically divided into three discontinuous power functions. When overlapping points are used, the initial component from 217 minutes to 59,000 minutes (41 days) has a slope of -0.2531 while the final component from 93,500 minutes (65 days) until death has a slope of -0.2916 . While these are not very different from those calculated for a break at 27 days, they are connected by a short segment, from 44,620 to 113,760 minutes, that has a very steep slope, -0.4458 . While this type of manipulation gives a somewhat better fit than does a two-slope analysis, it is not quite as good a fit as that with multiple exponentials discussed in the next section. The data for the other long-surviving mouse, No. 52, are best described, on a log-log plot, in terms of two rather than three discontinuous power functions. The data for these two mice analyzed in this way are given in Figure 7.

The point has recently been made that the parameters of a power function may differ widely depending on whether a linear fit has been made to the logarithms of the data ("log plane") or whether a nonlinear fit to the function $y = at^b$ has been made directly to nontransformed data ("natural plane").⁽⁵⁾ The latter computation was made for our radium retention data by means of a Variable Metric Minimization program available in the Applied Mathematics Division at Argonne.⁽⁶⁾ The results, which

are given in Table 54, are essentially identical with those given in Table 53 for a linear fit to the logarithms of the data. The very close correspondence of both sets of computations in this investigation is undoubtedly a result of the fact that the observed values do in fact fall very close to the computed line so that there are no large deviations that might otherwise affect the computation of the slope.

194-25

Figure 7. Double-log plot of retention of Ra^{226} by Mouse 52 and Mouse 54 after single intravenous injection. Nonnormalized data are plotted as two discontinuous power functions for Mouse 52, and as three discontinuous power functions for Mouse 54.

TABLE 54

Power function description of radium retention by four mice
Variable metric minimization nonlinear fit to nontransformed data

Mouse No	Number of measurements	One-minute intercept	One-day intercept	Slope
A Single straight line from 150 minutes after injection to death				
52	41	3 298	0 6429	-0 2248
53	31	2 651	0 6154	-0 2008
54	37	3 931	0 5610	-0 2677
55	32	3 810	0 5947	-0 2554
Combined data	141	3 336	0 6062	-0 2345
B Straight line from 150 minutes to 39 000 minutes (27 08 days)				
52	25	2 560	0 6641	-0 1855
53	17	2 320	0 6239	-0 1806
54	25	3 506	0 5644	0 2512
55	24	3 498	0 5967	-0 2429
Combined data	91	3 144	0 6087	-0 2258
C Straight line from 39 000 minutes to death				
52	16	7 500	0 8562	-0 2984
53	14	3 586	0 6770	0 2292
54	12	9 080	0 7466	-0 3435
55	8	9 704	0 8043	-0 3424
Combined data	50	6 418	0 7694	-0 2917

*Calculations were based on nonnormalized data except for the combined data which were based on data normalized to an injection of 1 00 μCi Ra^{226}

Ra²²⁶ Retention Considered as a Sum of a Series of Exponentials

The parameters of a series of three, four and five continuous exponential terms that would best fit the radium retention data were determined by the use of a high-speed electronic computer and the Variable Metric Minimization program referred to in the preceding section. This program calculates the parameters of the exponential terms that minimize a specified error function between the measured and calculated values. In this case the error function was essentially chi square although no use of this function to test statistical significance was intended. In order to

avoid this implication, the error function $\sum \frac{(R_{\text{obs}} - R_{\text{calc}})^2}{R_{\text{calc}}}$ is arbitrarily referred to here as the "error sum." The parameters that were calculated when the sum of three, four and five exponential terms were employed to fit the data are given in Table 55 for Mouse 52 and Mouse 54, the two animals that survived long enough to make this analysis profitable.

TABLE 55

Radium retention in the mouse described by sums of exponentials*

Number of exponentials	Sum of three exponentials		Sum of four exponentials		Sum of five exponentials	
	a	T	a	T	a	T
Mouse 52 [44 measurements, starting at 20 minutes after injection to death at 627,840 minutes (436 days)]						
1	0.4534	783	0.1597	177	0.1614	184
2	0.3204	23,646	0.3691	1,500	0.3654	1,505
3	0.2301	848,327	0.2888	28,394	0.2862	27,898
4			0.2232	921,447	0.1406	571,898
5					0.0859	2,926,780
Total	1.0039		1.0408		1.0395	
Mouse 54 [38 measurements, starting at 28 minutes after injection to death at 586,080 minutes (407 days)]						
1	0.5800	614	0.4728	420	0.0307	160
2	0.2660	21,371	0.1986	3,136	0.4586	459
3	0.1678	690,039	0.2206	35,898	0.1874	3,416
4			0.1448	1,027,980	0.2181	36,688
5					0.1439	1,046,840
Total	1.0138		1.0368		1.0387	

*Retention at time $t = \sum_{i=1}^n a_n e^{-(0.693/T_n)t}$, where

a = intercept at $t = 0$, normalized for an injection of 1.00 μCi Ra²²⁶, and

T = half-time of the exponential term.

For each of these sets of computations, Table 55 includes values for a (the intercept at $t = 0$ normalized for an injection of $1.00 \mu\text{Ci Ra}^{226}$), and T (the half-time of the exponential term). The values of a , which in each set should total 1.00 but in fact actually exceed that value slightly, may be interpreted to indicate the proportion of the injected dose that enters a given exponential term. There is no intent here, or elsewhere, to attempt to equate exponential terms with physiological, biochemical, or even idealized compartments. The exponential terms are used here solely as a method of mathematical analysis.

When the data are described as the sum of three exponentials, both mice are very similar in that each has a large component with a half-time of 600 to 800 minutes, a medium-size component with a half-time in the neighborhood of 22,500 minutes (15.6 days), and a small component of about 770,000 minutes (534 days). The two mice differ from each other, however, when four or five exponentials are used, although for each mouse considered separately a set of five exponentials does little to change the set of four exponentials. For Mouse 52, the first three components are the same for both analyses and the fourth exponential in the set of four is divided into two further terms in the set of five. With Mouse 54 the last four terms of each set correspond very closely, and a very small component with a 160 minute half-time is added in the set of five.

Table 56 provides a comparison of the error sums when the data are fit with power functions and with sums of exponentials. The error sums for

TABLE 56

Comparison of error functions when radium retention is described as a sum of exponentials or as a set of discontinuous power functions*

	Error sum*	
	Mouse 52	Mouse 54
<u>A. Power functions</u>		
Single	73.716×10^{-4}	86.560×10^{-4}
Two discontinuous with break at 39,000 minutes	18.426×10^{-4}	17.598×10^{-4}
<u>B. Sums of exponentials</u>		
Sum of three exponentials	14.089×10^{-4}	36.242×10^{-4}
Sum of four exponentials	7.061×10^{-4}	4.651×10^{-4}
Sum of five exponentials	7.012×10^{-4}	4.954×10^{-4}

*Error sum = $\sum \frac{(R_0 - R_c)^2}{R_c}$ μCi has been calculated for the same span as that used for the power functions, starting with 250 minutes for both mice, although the parameters of the sets of exponentials were computed for all the data, starting immediately after injection.

both sets of analyses have been computed starting with measurements 250 minutes after injection because the earlier portions of the retention curve bend downward and are not along a straight line on a log-log plot. For both mice the error sum is one-fourth as large when two discontinuous power functions with a break at 39,000 minutes are used instead of a single straight line. For Mouse 52 the sum of three exponentials further reduces the error sum, while it doubles the best power function fit for Mouse 54. However, sums of four or of five exponentials lead to a decided improvement in both mice so that the best fit is accomplished with these summed terms. In one case five exponentials are slightly better than when four are used, and the situation is reversed with the other mouse. Moreover, when sums of exponentials are used to describe a retention curve they are based on measurements starting immediately after injection, while power function curves are required to start several hours after injection.

The measured values obtained for Mice 52 and 54 and the discontinuous power functions that best fit these data are plotted in Figure 7. The measured values for these mice and the curve fit by the sum of four exponentials are given in Figure 8. These plots demonstrate that the results can be fitted fairly well with either two or three discontinuous power functions and very well with the sum of four exponential terms.

194-13

Figure 8. Double-log plot of retention of Ra^{226} in Mouse 52 and Mouse 54 fit by a sum of four exponential terms for each mouse. Nonnormalized data are used.

Whether power functions or sums of exponentials are used to describe radium retention, it should be remembered that they are only convenient mathematical descriptions of the overall retention pattern. The extent to

which these descriptions are useful for prediction or retrospective extrapolation depends in each case on the purpose for which it is used. We have found the power function to be very useful in interpreting data on continuous ingestion of radium by mice. However, both the power function and sums of exponentials have been found to be helpful in interpreting radium retention patterns in human cases. These applications will be presented in detail in forthcoming reports.

Retention of Radon Daughters Considered as a Power Function

The radium deposited in the tissues of the body, presumably principally in the skeleton, gives rise to radon, which has a 3.85-day half-life. A substantial portion of this radon escapes from the organism, but the radioactive daughters of the retained radon, RaB and RaC, are readily measured by gamma-ray spectroscopy. It is of considerable interest to have some understanding of the pattern of retention of these radon daughters with time.

194-24

Figure 9. Double-log plot of per cent retention of radon daughters by Mouse 54. Simple ratio of RaB-RaC to Ra²²⁶ shown by X, corrected points shown by circles. See text for discussion of correction.

RaB-RaC present in the mouse was also divided by the amount of radon that had grown in from the time of injection. These corrected per cent radon retention data for Mouse 54 are given by the open circles in Figure 9. Detailed data are given in Table 61 in the Appendix.

The straight line for RaB-RaC percentage retention on a log-log plot, shown in Figure 9, can be extended backward by this correction to as early as 629 minutes after injection. The slopes for this line and for similar lines for the other mice are given in Table 57. They range from 0.16 to

The amount of RaB and RaC in the mouse at any one time after intravenous injection has already been described [Figures 5 and 6; Table 60 (Appendix)]. When these retained daughter products are expressed as a percentage of the radium concomitantly present, they rise from a minimum at 200 to 400 minutes and fall along a straight line on a log-log plot after about 27 days (data for Mouse 54 shown in Figure 9). This increase from 400 minutes to 27 days is ascribable both to growth of radon in the bone as well as to an increase in the proportion of radon retained in the bone. In order to determine the actual percentage of radon retention and to eliminate the increase due to the growth of radon, the amount of

0.20 and all four mice combined give a slope of 0.18048. These values are very close to the value of 0.20 found by Mays et al.⁽²⁾ who plotted similar radon daughter retention data for rats, dogs, and man on a single graph. The pattern of retention of radon daughters is evidently quite constant in mammals that range in size from mouse to man, and is undoubtedly a function of bone-crystal chemistry rather than of size of the animal.

TABLE 57

Radon retention in mice after intravenous injection of Ra^{226} .
Linear fit to logarithmic transformation of the data

Mouse No.	Number of measurements	One-minute intercept	Slope $\pm$ S.E.
52	29	1.379	0.19165 ± 0.00077
53	25	1.232	0.20185 ± 0.00127
54	31	1.786	0.16923 ± 0.00036
55	29	1.871	0.16048 ± 0.00052
Combined data	114	1.559	0.18048 ± 0.00023

It is of amusing interest to compute what the percentage of radon daughter retention by the mouse might be if this tiny mammal were exceedingly long-lived. Table 58 presents the results of an extravagant extrapolation based on the power function $R = 1.559 t^{0.18048}$ that was found for the combined data for four mice. Thirty-day retention is 10.7 per cent, one year retention is 16.8 per cent, 25 year retention would be 30.0 per cent, while 100 year retention would be only 38.6 per cent. Even 10,000 years would not lead to more than 88.6 per cent, and it can be computed that only after 19,580.8 years would the retention of RaB-RaC be complete.

TABLE 58

Predicted radon retention percentage
in mice after intravenous injection*

Time	% Radon retention	Time	% Radon retention
1 minute	1.559	25 years	30.041
1 day	5.793	50 years	34.046
30 days	10.703	100 years	38.581
1 year	16.800	1000 years	58.459
10 years	25.462	10,000 years	88.579

*Calculations based on retention = $1.559 t^{0.18048}$

While these extrapolations are somewhat absurd, they are not entirely ridiculous or irrelevant. In Table 59 we have presented the radon daughter retention predicted from the mouse data along with actual recent measurements made on some of our human subjects. The striking correspondence of the retention percentages in mouse and man reinforces our conclusions that radon retention is independent of the size of the mammalian organism and that in this respect, at least, there is a sound basis for extrapolation from mouse to man. These results further imply that gamma-ray spectroscopy of human subjects for retained radium, which is calculated from the RaB and RaC that is present, must consider the time interval between administration of radium and subsequent whole-body count in order properly to convert the RaB and RaC measurements into an accurate value for Ra^{226} .

TABLE 59

Per cent radon retention predicted from
mouse data compared with measurements in man

Time since administration or exposure, years	Mice, %*	Human data, %**
29	30.86	8 State Hospital patients 31.2
35	31.92	6 Radium dial painters 34.4

*Retention values calculated from $R = 1.559 t^{0.18048}$.

**Patients remeasured in 1960.

We wish to thank Mr. Henry F. Lucas of the Radiological Physics Division for providing us with the results of radon breath analyses of the 14 patients summarized in Table 59. We also wish to acknowledge the assistance of Miss Dorice Czajka, Biological and Medical Research Division, in the preparation of a pure RaC solution. Mr. Merlin H. Dipert and Miss Carol A. Vancina, of the Biological and Medical Research Division, have generously assisted with some of the electronic computations, principally the linear curve fitting to logarithmic transformations of the data. Dr. Ronald C. Hamelink, Mr. Burton S. Garbow, and Mr. William J. Snow, all of the Applied Mathematics Division, helped with the applications of the Variable Metric Minimization program to our specific problems. Mr. John B. Corcoran of the Radiological Physics Division assisted with many of the gamma-ray spectroscopy measurements of the mice.

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APPENDIX

TABLE 60A

Radium and radon daughter data for Mouse 52 after intravenous injection.
Data normalized to injection of 1.00 $\mu\text{Ci Ra}^{226}$

Minutes after injection	$\mu\text{Ci Ra}^{226}$	$\mu\text{Ci RaB}$	Mean $\mu\text{Ci RaC}$	$\frac{\% \text{ RaC}^{**}}{\text{Ra}^{226}}$	Corrected† % radon retention
20.	1.0493	.418700	.471700	45.390	
34.	.9946	.272000	.376400	37.840	
52.	.9964	.176600	.309200	31.030	
82.	.9795	.080000	.176000	17.970	
107.	.9848	.041240	.105300	10.690	
* 147.	1.0170	.016340	.044290	4.354	
161.	.9464	.011460	.030920	3.267	
206.	.9259	.004937	.011120	1.201	
246.	.8929	.002866	.005312	.595	
266.	.8755	.001884	.003777	.431	
310.	.8857	.001741	.002420	.273	
346.	.8804	.002009	.002420	.274	
375.	.8330	.002187	.002411	.280	
* 424.	.7857	.002652	.002554	.330	6.343
464.	.8437	.002866	.002187	.286	5.039
553.	.8062	.003741	.003652	.456	6.788
630.	.8062	.003759	.004312	.512	6.714
686.	.7839	.003420	.003509	.444	5.371
699.	.7848	.003536	.003482	.446	5.297
756.	.7696	.003607	.003589	.468	4.800
811.	.7607	.003607	.004170	.523	5.398
* 1532.	.7277	.006509	.006562	.899	5.129
1571.	.7009	.006848	.007044	.996	5.551
3126.	.5625	.012770	.012990	2.295	7.058
3278.	.5575	.012230	.013230	2.316	6.853
6006.	.5062	.018870	.018440	3.670	6.920
6045.	.5107	.018840	.018400	3.631	6.817
8731.	.4562	.025280	.025240	5.534	8.301
8781.	.4643	.024840	.024680	5.328	7.967
* 13293.	.4202	.029800	.032300	7.490	9.222
* 13339.	.4194	.028290	.029930	7.007	8.615
17561.	.3964	.030040	.030440	7.644	8.586
28051.	.3571	.033620	.033280	9.350	9.632
40680.	.3312	.034520	.033790	10.274	10.336
51900.	.3000	.033370	.032900	11.018	11.018
69120.	.2696	.031010	.030970	11.490	11.490
112320.	.2244	.029150	.029290	13.036	13.036
118100.	.2123	.028460	.028960	13.559	13.559
138400.	.2054	.028380	.028320	13.802	13.802
148500.	.2094	.028760	.028280	13.576	13.576
178560.	.1988	.028210	.028210	14.196	14.196
213120.	.1948	.028470	.028020	14.461	14.461
233280.	.1971	.028610	.028260	14.393	14.393
299520.	.1759	.027680	.027910	15.814	15.814
364320.	.1715	.026300	.025710	15.107	15.107
364320.	.1736	.026290	.026390	15.185	15.185
406080.	.1565	.026950	.026290	16.937	16.937
502560.	.1521	.025860	.025610	16.900	16.900
597600.	.1391	.025670	.025860	18.552	18.552
627840.	.1460	.025880	.025900	17.737	17.737

* Error of about $\pm 5\%$ in Ra^{226} value because of known presence of Ar^{41} in whole-body counter atmosphere. Measurements excluded from analysis of data.

** The simple ratio of $\text{RaC}/\text{Ra}^{226}$ has been computed down to 346 minutes, after which the RaB values have been averaged in with the two RaC values so that starting with 375 minutes the numerator is a weighted mean of RaB and RaC .

† See text for discussion of this correction.

TABLE 60B

Radium and radon daughter data for Mouse 53 after intravenous injection.
Data normalized to injection of 1.00 $\mu\text{Ci Ra}^{226}$

Minutes after injection	$\mu\text{Ci Ra}^{226}$	$\mu\text{Ci RaB}$	Mean $\mu\text{Ci RaC}$	$\frac{\% \text{ RaC}^{**}}{\text{Ra}^{226}}$	Corrected† % radon retention
18.	.9913	.284000	.341000	34.410	
48.	.9727	.125400	.216500	29.010	
63.	.9974	.086240	.169400	16.980	
102.	.9197	.030800	.072490	7.880	
142.	.9537	.013080	.032330	3.053	
195.	.9275	.004473	.010240	1.105	
273.	.8669	.001687	.002509	.289	
327.	.7958	.001464	.002839	.357	
373.	.7754	.001651	.001902	.245	
386.	.7736	.001716	.001796	.232	
443.	.7732	.001902	.001790	.232	4.269
491.	.7527	.002241	.001723	.252	4.211
1212.	.6304	.004491	.004598	.725	5.126
1243.	.6286	.004437	.004446	.708	4.888
2762.	.5580	.013152	.011946	2.228	7.590
2846.	.5607	.010839	.011026	1.958	6.506
5946.	.5000	.017839	.017446	3.534	6.710
5978.	.4955	.017339	.017491	3.522	6.663
8415.	.4562	.021710	.021821	4.778	7.316
8480.	.4411	.021893	.021624	4.933	7.521
* 12966.	.4051	.028040	.029500	7.282	9.053
* 13020.	.4141	.027540	.027980	6.721	8.342
17580.	.3973	.029630	.030530	7.585	8.517
28080.	.3589	.033290	.033640	9.336	9.617
40341.	.3170	.033870	.033060	10.542	10.609
40700.	.3295	.034070	.033260	10.200	10.246
51800.	.2929	.032870	.031960	11.018	11.018
51860.	.2929	.033160	.032560	11.216	11.216
69100.	.2741	.031940	.031920	11.648	11.648
112300.	.2500	.031930	.031430	12.630	12.630
118086.	.2411	.031400	.031820	13.104	13.104
138246.	.2375	.032410	.033360	13.906	13.906
148300.	.2299	.032550	.032240	14.117	14.117
178560.	.2221	.031850	.032470	14.450	14.450
213120.	.2171	.031690	.031870	14.677	14.677
233280.	.2086	.032720	.032220	15.449	15.449
299520.	.2010	.031446	.031552	15.715	15.715
364320.	.1974	.030393	.030330	15.365	15.365

* Error of about $\pm 5\%$ in Ra^{226} value because of known presence of Ar^{41} in whole-body counter atmosphere. Measurements excluded from analysis of data.

** The simple ratio of $\text{RaC}/\text{Ra}^{226}$ has been computed down to 443 minutes, after which the RaB values have been averaged in with the two RaC values so that starting with 491 minutes the numerator is a weighted mean of RaB and RaC .

† See text for discussion of this correction.

TABLE 60C

Radium and radon daughter data for Mouse 54 after intravenous injection.
Data normalized to injection of 1.00 $\mu\text{Ci Ra}^{226}$

Minutes after injection	$\mu\text{Ci Ra}^{226}$	$\mu\text{Ci RaB}$	Mean $\mu\text{Ci RaC}$	$\frac{\% \text{ RaC}^{**}}{\text{Ra}^{226}}$	Corrected [†] % radon retention
28.	1.0263	.005710	.008034	.781	
77.	.9641	.002221	.003734	.387	
125.	.9393	.001748	.002383	.254	
* 217.	.9214	.001714	.001721	.187	
269.	.8552	.001559	.001486	.174	
321.	.8141	.002182	.002048	.252	
* 403.	.7366	.002262	.002044	.287	5.813
459.	.7714	.002514	.002300	.307	5.472
546.	.7334	.002644	.002655	.362	5.444
629.	.7090	.002859	.002703	.389	5.106
695.	.6910	.003241	.002990	.444	5.507
751.	.6590	.003448	.003210	.499	5.536
1512.	.5262	.005759	.005655	1.081	6.240
1817.	.5203	.007072	.007079	1.360	6.654
3254.	.4566	.010897	.010449	2.346	6.983
4731.	.4148	.013780	.013920	3.344	7.455
6221.	.3906	.017030	.017130	4.377	8.063
7781.	.3668	.018760	.018640	5.092	8.156
9093.	.3526	.020240	.019930	5.682	8.338
11900.	.3353	.022560	.022300	6.678	8.600
13080.	.3320	.022990	.023040	6.936	8.590
14781.	.3121	.023500	.023670	7.566	8.961
17425.	.3063	.025090	.025070	8.188	9.215
18900.	.2955	.024490	.024830	8.364	9.218
21900.	.2879	.026140	.025880	9.020	9.633
24720.	.2854	.026660	.026370	9.275	9.708
29140.	.2659	.026820	.026510	10.009	10.271
33412.	.2545	.026130	.025970	10.224	10.379
39240.	.2424	.026100	.026110	10.769	10.847
44620.	.2381	.025970	.026010	10.919	10.959
59000.	.2121	.025480	.025270	11.948	11.948
93600.	.1702	.021370	.020960	12.397	12.397
113760.	.1580	.020540	.020230	12.869	12.869
180000.	.1343	.018290	.018630	13.862	13.862
244800.	.1269	.018030	.018020	14.207	14.207
286560.	.1202	.017680	.017310	14.494	14.494
383040.	.1114	.017330	.017340	15.570	15.570
478080.	.1044	.017260	.017120	16.449	16.449
508320.	.1026	.017370	.017310	16.892	16.892
586080.	.0979	.017230	.017040	17.468	17.468

* Error of about $\pm 5\%$ in Ra^{226} value because of known presence of Ar^{41} in whole-body counter atmosphere. Measurements excluded from analysis of data.

** The simple ratio of $\text{RaC}/\text{Ra}^{226}$ has been computed down to 403 minutes, after which the RaB values have been averaged in with the two RaC values so that starting with 459 minutes the numerator is a weighted mean of RaB and RaC.

† See text for discussion of this correction.

TABLE 60D

Radium and radon daughter data for Mouse 55 after intravenous injection.
Data normalized to injection of 1.00 $\mu\text{Ci Ra}^{226}$

Minutes after injection	$\mu\text{Ci Ra}^{226}$	$\mu\text{Ci RaB}$	Mean* $\mu\text{Ci RaC}$	$\frac{\% \text{ RaC}^{**}}{\text{Ra}^{226}}$	Corrected† % radon retention
42.	.9804	.003951	.006026	.611	
90.	.9600	.002322	.003352	.349	
137.	.9537	.001933	.002074	.218	
227.	.9248	.001730	.001807	.195	
334.	.8148	.002050	.001867	.227	
422.	.8022	.002511	.002352	.293	5.670
473.	.7800	.002337	.002241	.291	5.037
561.	.7744	.002833	.002711	.355	5.214
642.	.7381	.003126	.003011	.413	5.320
706.	.7144	.003474	.003370	.476	5.604
762.	.7241	.003530	.003578	.492	5.383
1533.	.5730	.006419	.006256	1.101	6.279
1832.	.5689	.007333	.007296	1.284	6.208
3267.	.4905	.011300	.011500	2.331	6.897
4760.	.4389	.014340	.014260	3.256	7.225
6238.	.4160	.016580	.016590	3.986	7.331
7816.	.3955	.018620	.018830	4.743	7.576
9105.	.3788	.020030	.019940	5.492	8.053
11910.	.3494	.023660	.024690	6.971	8.987
13095.	.3466	.023490	.023260	6.734	8.339
14797.	.3414	.024840	.024790	7.266	8.606
17440.	.3280	.025720	.025590	7.815	8.795
18910.	.3207	.025220	.025110	7.839	8.640
21900.	.3099	.026380	.026190	8.473	9.048
24720.	.3013	.026600	.026610	8.830	9.243
29150.	.2866	.027090	.027000	9.430	9.676
33430.	.2758	.026760	.026400	9.613	9.759
39250.	.2601	.026240	.026310	10.107	10.179
44640.	.2550	.026870	.026610	10.470	10.507
59040.	.2295	.025520	.025640	11.320	11.320
93580.	.1870	.018740	.022600	12.140	12.140
113760.	.1769	.021930	.021510	12.240	12.240
180000.	.1499	.019950	.019820	13.247	13.247
244800.	.1374	.019300	.019240	14.021	14.021
286560.	.1340	.019110	.018770	14.089	14.089

* Error of about $\pm 5\%$ in Ra^{226} value because of known presence of Ar^{41} in whole-body counter atmosphere. Measurements excluded from analysis of data.

** The simple ratio of $\text{RaC}/\text{Ra}^{226}$ has been computed down to 422 minutes, after which the RaB values have been averaged in with the two RaC values so that starting with 473 minutes the numerator is a weighted mean of RaB and RaC .

† See text for discussion of this correction.

TABLE 61

Radon retention values for Mouse 54

Time		Per cent retention of radon	
Minutes	Days	Measured values	Calculated values*
629	0.437	5.106	5.304
695	0.483	5.507	5.394
751	0.522	5.536	5.466
1512	1.050	6.240	6.157
1817	1.262	6.654	6.352
3254	2.260	6.983	7.013
4731	3.285	7.455	7.474
6221	4.320	8.063	7.830
7781	5.403	8.156	8.133
9093	6.315	8.338	8.351
11,900	8.264	8.600	8.742
13,080	9.083	8.590	8.883
14,781	10.264	8.961	9.070
17,425	12.101	9.215	9.327
18,900	13.125	9.218	9.457
21,900	15.208	9.631	9.696
24,720	17.167	9.706	9.898
29,140	20.236	10.271	10.179
33,412	23.203	10.379	10.418
39,240	27.250	10.847	10.707
44,620	30.986	10.959	10.943
59,000	40.972	11.948	11.474
93,600	65.000	12.397	12.411
113,760	79.000	12.869	12.829
180,000	125.000	13.862	13.869
244,800	170.000	14.207	14.613
286,560	199.000	14.494	15.010
383,040	266.000	15.570	15.768
478,080	332.000	16.449	16.374
508,320	353.000	16.892	16.545
586,080	407.000	17.468	16.950

*Calculations based on Retention = $1.774 t^{0.1699}$. This curve was computed from nontransformed data by the Variable Metric Minimization method, and the parameters differ very slightly from that given in Table 57 computed by linear regression of the logarithms of the data.

SOME PROBLEMS OF ACCURATE MEASUREMENT OF POTASSIUM IN MAN

R. K. Hukkoo and C. E. Miller

Many scientific investigators are concerned with the accurate measurement of potassium in humans and animals. Clinical investigators are studying important interrelationships between the potassium content of the body and numerous physiologic processes. In animal husbandry the potassium content of the animal has been used as a guide in the selective breeding of domesticated animals. For these and many other reasons, large numbers of whole-body counters have been built principally to measure the potassium content of humans and animals.

The specifications for a whole-body counter that would satisfy each purpose vary since the accuracy required differs for each application. Some investigators are interested in measurement of the average potassium content of a large group of people and are not concerned with the absolute quantity present in any single individual. This type of study requires a short counting time and can be satisfied with a potassium measurement that contains an uncertainty of perhaps 10 per cent. The clinical investigator who is studying the depletion of potassium as a function of weight loss in obese patients, for example, may require a measurement that is accurate within 1 per cent. Other investigators are interested in measurements of the relative change in potassium levels over a period of time in an individual but may not be concerned with an absolute quantitative measurement.

Very little information has been published on the absolute accuracy of the various types of whole-body counters. Furthermore, it is very difficult to assess the absolute accuracy of potassium measurements made with a whole-body counter since there is no direct method of determining the amount of potassium present in the living subject. The accuracy of various whole-body counters can only be estimated by a comparison of the results obtained with each type.

Because the accuracy desired by some investigators may be beyond the capabilities of existing equipment, a study was undertaken to evaluate the accuracy of three whole-body counting techniques for measurement of potassium in man. This study was part of a cooperative effort made by groups from Argonne National Laboratory, Loyola University Medical School, and Purdue University. The potassium content of a group of patients* was measured with three different arrangements of NaI crystals, with a 4π liquid

*The patients were under the care of Dr. Alexander P. Remenchik, Loyola University Medical School and were hospitalized at Cook County Hospital, Chicago, Illinois. This phase of the program was supported by a U. S. Public Health Service grant, RH 00283-01.

counter, and by an isotope dilution method. The various whole-body spectrometry techniques were assessed for accuracy by an analysis of the internal consistency of the results. The measurements made at Argonne National Laboratory will be discussed in this report and will be compared with the results obtained by the isotope dilution method (Remenchik).

Procedures

A group of patients whose weights ranged from 104 lbs (47.2 kg) to 336 lbs (152.5 kg) was selected for study. Patients of widely divergent weights were selected in order to determine if there was any relationship between total-body weight and accuracy of the measurement. The very obese people were of particular interest since they were entering a starvation program for weight reduction and were to be available for measurement prior to and after they had lost an appreciable amount of weight. Thus it was hoped that data would be obtained for the same individual at two different body weights.

Three counting geometries were evaluated in this study: The tilting-chair technique was used with 1) a squat right cylindrical crystal, 8 in. diameter by 4 in. thick, and 2) a log-type cylindrical crystal, 8 in. long and 6 in. diameter. Each crystal was positioned at a standard distance over the chair by means of a mechanical fixture attached to the arms of the chair. The log crystal was placed over the tilting chair with its axis horizontal and parallel to the planes of the seat and the back of the chair. The patient sat directly on the metal surface of the chair without padding in order to assure a fixed crystal-to-patient distance. The patient rested his knees against the arm rests of the chair and folded his hands in his lap. 3) Potassium content was also determined with the single-crystal, multiple-position technique described earlier in this report. The patient was placed in a supine position on a Lucite slab (bed) and the crystal was located with its face 30 cm above the bed for normal subjects and 37.7 cm for obese subjects. The patient's gamma-ray spectrum was obtained with the crystal placed in turn at each of seven preselected locations along the longitudinal mid-line of the body. At position 4 the crystal was at the patient's mid-height, and successive adjacent positions were 24.5 cm apart in each direction from the central position. The positions have been numbered 1 through 7 starting at the head of the patient.

The gamma-ray spectrum for each patient was counted for 40 minutes with each tilting-chair technique. The counting time at each position of the 7-crystal-position technique was 30 minutes, with a total counting time of 3.5 hours. Since it was uncomfortable for the patients to lie in one position for 3.5 hours without interruption, they were allowed to arise and relax each time after being counted at two positions and were then realigned when counting was resumed.

For each patient the normal gamma-ray spectrum was obtained first with each of the three techniques. The patients were then given K^{42} orally or intravenously and remeasured by all three techniques after a delay of 40 hours to allow the K^{42} to equilibrate with the body potassium. In order to determine the percentage of retention of K^{42} , the patient's urine was collected regularly after K^{42} administration until his new spectrum was recorded. The net K^{42} spectrum was obtained by subtracting the normal spectrum from the second spectrum.

The same volume of K^{42} that was administered to each subject was pipetted into a bottle, and the bottle was filled with distilled water to a convenient height to simulate the geometry of a similar bottle that contained a known amount of potassium salt (K standard). Both bottles were counted at an accurately calibrated position of 40 cm from each detector. All K^{42} counting rates were corrected for physical decay.

The potassium content of the subject is calculated by substituting the count rates into the following equation:

$$\text{grams K (subject)} = \text{c/m } K^{40} \text{ (subject)} \times \frac{\text{grams K (bottle)}}{\text{c/m } K^{40} \text{ (bottle)}} \times \frac{\text{c/m } K^{42} \text{ (bottle)}}{\text{c/m } K^{42} \text{ (subject)}} \quad (1)$$

The first term is the count rate from the patient's natural potassium spectrum. The second term consists of the grams of potassium in the standard salt bottle divided by the counts per minute observed from the bottle. The third term is the count rate observed from the K^{42} that had been pipetted into the bottle divided by the net K^{42} count rate from the patient corrected for physical decay and excretion from the body. In a previous paper,⁽¹⁾ the last term alone was designated the "g" (geometrical) factor. The magnitude of this last term will be different if the K^{40} and K^{42} bottles are counted at different distances from the crystal. The product of the second and third terms, which is actually the calibration factor for this technique, does not depend upon the distance of the bottles from the crystal since the two bottles are counted at the same distance from the crystal. In all of the discussion in this report, the expression "g factor" refers to the product of the second and third terms.

Only the count rates at the potassium photopeaks have been used for the calculation of potassium content. For this purpose a 250-keV wide band centered at each photopeak was used. No effort was made to correct the count rate from the patients due to the difference in absorption of the gamma rays for K^{40} and K^{42} , which is approximately 4 per cent for a 30-cm-thick tissue equivalent path. We realize that this error may be aggravated when obese subjects are measured. The accuracy of the results does not depend upon the isotopic abundance and the decay schemes of K^{40} and K^{42} but only on count statistics and the accuracy of K^{42} administration.

Analysis of Results

Tilting Chair with 8 in. by 4 in. NaI(Tl) Crystal

The g factors obtained for 20 subjects are plotted as a function of weight/height in Figure 10. For 14 normal subjects a regression line calculated by least squares had a slope of 0.051 and a standard error of estimate of 0.018. For six obese patients (seven points, since one was measured at two weights) the regression line had a slope of 0.440 and a standard error of estimate of 0.079. When all the points are combined, these values are 0.135 and 0.028, respectively.

Figure 10

The g factors obtained with 8 in. by 4 in. crystal for 20 subjects plotted against weight/height, with lines of regression obtained for normal (—), obese (---) and all (-.-) subjects

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The percentage differences between the actual g factor and the value given by the regression line for the normal subjects were calculated. Except for one subject all the points lie within ± 3.5 per cent of the regression line. If an arithmetic mean value of g is taken for the normal subjects, the maximum variation from this mean is found to be as much as ± 6.0 per cent, a value much larger than the ± 3.5 per cent observed when the g factors are corrected for weight/height. For obese patients, where the regression line is much steeper, the variation in g from this line is as much as ± 10 per cent. The large spread in g factors for these obese patients is probably ascribable to the greater variation in absorption and scatter of the gamma rays in these bodies. This large variation in g factors makes it necessary to administer K^{42} to each obese subject if accurate measurements are to be obtained.

Tilting Chair with the Log Crystal

The g factors obtained as a function of weight/height when the measurements were made with the log crystal over the tilting chair are given in Figure 11. The regression slopes with their respective standard errors of estimate for the three groups of patients are given in Table 62.

Figure 11

The g factors obtained with log crystal for 20 subjects plotted against weight/height, with lines of regression obtained for normal (—), obese (---) and all (-.-.-) subjects

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TABLE 62

Regression slopes for three groups of patients

Counting technique	All patients		Normal patients		Obese patients	
	Slope	$S_{y \cdot x}$	Slope	$S_{y \cdot x}$	Slope	$S_{y \cdot x}$
Tilting chair with 8 in. x 4 in. crystal	0.135	0.028	0.051	0.018	0.440	0.079
Tilting chair with log crystal	0.067	0.017	-0.008	0.012	0.208	0.048
Sum of 7-crystal positions	-	-	0.294	0.009	-	-

The g factors obtained for 85 per cent of the normal subjects fall within ± 2.3 per cent of the regression line. However, the same g factors for all the normal subjects fall within ± 2.5 per cent of their arithmetic mean. Thus, an average calibration factor could be used for this detector for subjects who weigh between 104 lbs to 268 lbs (47.2 kg to 121.7 kg) without introducing an error of more than ± 2.5 per cent in the measurements.

For obese patients the spread in g factors from the regression line is ± 8.0 per cent, which is somewhat less than that obtained with the 8 in. by 4 in. crystal. With this detector, too, the spread in g factors for obese patients suggests that each of these persons should be given K^{42} for accurate measurement of K^{40} . A comparison of the range of the ordinate ($= g$ factor) in Figures 10 and 11 indicates that the calibration factor is much smaller for the log crystal than with the 8 in. by 4 in. detector. This situation reflects a higher count rate per gram of potassium in the body with the log crystal than with the other crystal.

Single-crystal, Multiple-position Technique

The spectra obtained with the 8 in. by 4 in. crystal at each of the seven crystal positions were analyzed in two independent ways. First, the count rates in the K^{40} and K^{42} photopeaks, respectively, from the seven crystal positions were added and the sum was substituted into Equation 1 to calculate the total-body potassium in the patient. The calibration factors obtained in this way for nine patients who weighed from 104 lbs to 268 lbs are plotted as a function of weight/height in Figure 12. A regression line with a slope of 0.294 and standard error of estimate of 0.009 was found for these data. It can be seen that all of the g factors fall very close to the regression line. One patient, who weighed 279 lbs (126.7 kg), was so obese that she could not lie with her feet together and the activity in her legs could not be counted with the same sensitivity as in the other patients. Her g value fell far below the regression slope in Figure 12. The slope of the regression line found by the 7-crystal-position technique is much steeper than those found by the tilting-chair techniques.

Figure 12

The g factors obtained with sum of seven crystal positions for ten subjects plotted against weight/height. The point indicated by arrow is not included to obtain the line of regression.

The percentage differences between the actual g values and those given by the regression line, except for one patient, fall within ± 1.5 per cent of the regression line. Since the regression line is quite steep, the calibration factor used with a 7-crystal-position technique must contain a weight/height correction.

The crystal had to be raised to 37.7 cm above the bed in order to accommodate the four patients who weighed more than 279 lbs and were more than 30 cm thick. The number of subjects was too small to permit an analysis of the results obtained under these altered conditions.

In the second method of analyzing the spectra, the count rates observed with the crystal at each of the seven positions were substituted into Equation 1 and g factors were obtained for each position. Figure 13 is a

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Figure 13. The g factors obtained for ten subjects at successive crystal positions with 7-crystal-sum technique plotted as a function of weight/height. The point indicated by arrow is not included to obtain the line of regression.

plot of the g factors as a function of weight/height for each of the seven positions. Regression slopes and standard errors of estimate were calculated for the individual counting positions (Table 63). The maximum error in the measurements can be seen to be in positions 1, 7 and 6, in that order, since the count rate in these positions is dependent on height of the subject. On the other hand, the dependence of the g factor upon weight/height as evidenced by the greater slope is maximum for positions 3, 4 and 2. The seven graphs in Figure 13 indicate that the weight/height relationship observed with the tilting-chair techniques or with the 7-crystal-sum technique is due to differing absorptions in the trunks of the patients rather than in the heads and lower extremities.

TABLE 63

Regression slopes calculated for
individual counting positions

	Crystal positions for normal patients						
	1	2	3	4	5	6	7
Slope	0.038	0.218	0.377	0.298	0.096	0.073	0.153
$S_{y \cdot x}$	0.121	0.011	0.020	0.014	0.022	0.052	0.086

The variation in g factors for position 5 falls within 4 per cent of the average for this position, and compares quite favorably with the 3.4 per cent found with the tilting chair. The average g factor for position 5 is 2.18 as compared to 2.10 for the tilting chair so that the counting efficiency is only 3.7 per cent less when a single crystal is placed at position 5 than when the same crystal is placed over the subject in the tilting chair.

Five subjects were measured at position 5 with the crystal 25 cm above the bed to increase the sensitivity and to compare the results with the data presented in Figure 13. Since much greater variability was noted when the crystal was lowered to 25 cm, it was not possible to increase the sensitivity at this position by lowering the crystal and at the same time maintain the low variability of the g factor observed with the crystal at 30 cm. However, if a single crystal is placed at position 2 for the measurement and the g factors are corrected for the weight/height ratio of the subject, a much more accurate measurement of total-body potassium can be made than when the same crystal is used with the tilting chair (Tables 62 and 63).

Distribution of Potassium in the Body

The data obtained with the 7-crystal-position technique were also analyzed to obtain the distribution of potassium along the body. Seven stacks of Presdwood of varying heights, 24.5 cm wide by 30 cm long, were arranged to simulate the shape of the supine patient as shown in Figure 14. Two phantoms of this type were constructed, one with a cavity to simulate human thorax and another without a cavity. A K^{42} point source was placed in the geometrical center of the first phantom block and a K^{42} spectrum obtained with the crystal placed, in turn, at each of the seven crystal positions. The source was then placed in the geometrical center of each succeeding block and the spectrum obtained in the corresponding seven crystal positions. The count rate obtained from the subject and from the K^{42} in the phantom were substituted in Equation 1, p. 14 of this report. The solution vectors, R_1 , R_2 , R_3 , etc. should be equal to the amount of K^{42} present in the corresponding one-seventh segment of the subject's body.

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Figure 14. Schematic illustration of the arrangement of Presdwood as a phantom to simulate the supine patient

The amount of normal potassium present in each seventh segment of the body was found by substituting into the equation the patient's normal count rate at each of the seven positions and the count rate obtained when the K^{42} was placed in either phantom. For two subjects the distribution of potassium found by these methods is plotted in Figure 15. The distribution

Figure 15
 K^{40} distribution along the body for two subjects obtained by use of a solid phantom (---○---) and by a lung equivalent phantom (—○—)

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shown by the dotted line was found when the solid phantom was used and the distribution given by the solid line was obtained with the lung equivalent phantom. Earlier work with various thicknesses of phantoms had demonstrated that the distribution found by the seven simultaneous equations did not depend markedly on phantom thicknesses when solid phantoms were used at all positions. The differences in distribution in these two curves demonstrates that a phantom with a simulated lung cavity yields more realistic distributions of potassium along the body than the solid phantom.

K^{40}/K^{42} Ratios

In order to determine the K^{40}/K^{42} in any segment of the body, the K^{40} and K^{42} count rates in the body were independently substituted into the seven-equation matrix and the solution vectors were obtained. The relative ratio between the solution vectors for each segment is independent of the particular phantom used and should reflect the K^{40}/K^{42} ratio in that segment of the body.

The K^{40}/K^{42} ratio expressed in arbitrary units for the seven segments of the body for four men are given in Figure 16 and for five women in Figure 17. Some striking similarities and contrasts can be seen in these two figures. For the women, the K^{40}/K^{42} ratio is higher at position 1 than the average ratio for the whole body, a finding that indicates that the K^{42} has not equilibrated with the normal potassium in the head in the 40-hour period between administration and measurement. With the men, positions 1 and 7 have higher ratios than the average for the remainder of the body. The K^{40}/K^{42} ratios observed at positions 5, 6, and 7 vary inversely for men

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Figure 16. K^{40}/K^{42} ratios for seven segments of the body for four male subjects

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Figure 17. K^{40}/K^{42} ratios for seven segments of the body for five female subjects

and women, with the highest value in positions 5 and 7 and the lowest in position 6 for the men and with the reverse situation for the women. These variations are undoubtedly dependent upon the amount of muscle in these sectors of the body and upon the body build.

The distribution of K^{40} and K^{42} along the bodies of two women are given in Figure 18 and for two men in Figure 19. For each set the total-body K^{40} and K^{42} have been normalized to the same total-body potassium value. It will be noted from these four sets of curves that K^{42} is not distributed in the body in equilibrium with the normal potassium.

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139-3534

Figure 18. K^{40} and K^{42} distribution along the longitudinal axis of two female subjects

139-3542

139-3535

Figure 19. K^{40} and K^{42} distribution along the longitudinal axis of two male subjects

Obese Patients during Weight Loss

Four obese patients were measured with the 7-crystal-position technique with the crystal located at 37.7 cm above the surface of the bed. Since these patients are considerably thicker than normal persons, a new phantom similar to the one shown in Figure 14 was constructed of stacks of Presdwood 20, 33, 33, 31, 26, 14 and 10 cm thick, starting with position 1. The K^{40} and K^{42} count rates from these patients and the spectra obtained from the phantom were entered in the seven equation matrix (Equation 1, p. 14) and the K^{40}/K^{42} ratios were determined. The ratios for the four patients are given in Figure 20. It is quite obvious from the curves that the K^{40}/K^{42} ratio is not constant along the body. This ratio is particularly high for position 1 (head) for the two men and over the feet of all four subjects.

Figure 20
 K^{40}/K^{42} ratios for seven segments
of the body of four obese patients

139-3540

One of the obese patients lost 76 lbs (34.5 kg) and his total-body potassium decreased from 198 grams to 139 grams during a starvation period of 46 days. This patient was remeasured by all three techniques after the weight loss. The g factors obtained with the two tilting-chair techniques decreased about 10 per cent and are given before weight loss (point 1) and after weight loss (point 2) in both Figures 10 and 11. The two points demonstrate that the counting efficiency increases as the patient loses weight and as it does so it masks the change in body potassium: while the potassium in the body has decreased, the counting efficiency has increased, so that the measurements tend to yield the same count rate. The K^{40}/K^{42} ratio along the body before (solid line) and after (dotted line) weight loss is given in Figure 21. The curves are significantly different for positions 1, 6 and 7. The K^{42} has not equilibrated with the normal

potassium in the head and the feet for the first series of measurements and thus the K^{40}/K^{42} ratio was high for positions 1 and 7. This patient had been at bed rest during the 46-day starvation period, and his body was in negative calcium as well as nitrogen balance. It may be postulated that the K^{42} administered to the patient after the weight loss was absorbed by the skeleton and consequently the K^{40}/K^{42} ratio was considerably lower for positions 1 and 7 where the skeleton constitutes a larger proportion of the tissue mass than elsewhere. This patient retained practically all the K^{42} administered after the weight loss and did not excrete the 4.0 per cent to 6.0 per cent expected in the first 40 hours. Furthermore, the ratio of potassium as found by the isotope dilution technique to that found by the whole-body counter was 91.5 per cent at the time of the first measurement and 96 per cent after the weight loss. This increase would be expected if the K^{42} did not mix in the skeleton as thoroughly the first time as it did after starvation.

Figure 21

K^{40}/K^{42} ratios for seven segments of the body of an obese patient, before (—) and after weight loss of 76 lbs (---)

139-3537

Exchangeable Potassium

The grams of potassium found for each patient by the isotope dilution technique (Remenchik) are given in column 5 of Table 64. The ratios of grams of potassium found for each patient by the isotope dilution technique divided by that found by various whole-body counter techniques are given in columns 6, 7 and 8. The use of this ratio permits a direct comparison of the results from patient to patient even though the body weights and their respective potassium contents vary widely. The mean values and standard deviations of these ratios for each counting technique are given at the bottom of each column. The average value for exchangeable potassium obtained by each of the three techniques is 93 per cent and the three

average values agree within 1.0 per cent of each other. The coefficient of variation is about 5.0 per cent for the tilting-chair techniques and only 2.7 per cent for the 7-crystal-position technique. These indices of variability are in agreement with the spread in g factors given for the three techniques in Table 62.

TABLE 64
Potassium found for each patient
by various techniques

Subjects	Sex	Weight, lbs	Wt/ht lbs/in.	K by isotope dilution, g	Ratio of grams K by isotope dilution to that by various whole-body counters		
					Tilting chair		Sum of 7-crystal positions
					8 in. x 4 in.	Log crystal	
1-1	M	337.0	4.955	180.0	0.851	0.903	0.915
1-2	M	285.5	4.496	137.0	0.936	1.021	0.937
1-3	F	283.5	4.210	130.0	0.986	0.916	0.908
1-4	F	270.5	4.328	119.0	0.951	0.996	0.905
2-1	M	268.0	3.585	153.0	0.904	0.952	0.916
2-2	M	145.5	2.328	100.6	0.838	0.930	0.917
2-3	F	127.0	2.094	76.5	0.900	0.936	0.930
2-4	F	279.0	4.275	116.6	0.997	1.011	0.980
3-1	M	168.0	2.382	146.0	0.909	0.877	0.891
3-2	M	195.5	2.615	176.0	0.960	0.900	0.964
3-3	M	263.5	3.875	135.0	0.971	0.966	0.958
3-4	F	267.0	4.238	89.0	0.870	0.878	-
4-1	F	114.5	1.892	81.5	0.987	0.900	0.923
4-2	F	104.0	1.644	70.9	0.924	0.888	0.903
4-3	F	112.5	1.751	-	-	-	-
4-4	F	142.5	2.127	89.4	1.013	0.907	0.913
5-1	M	165.2	2.263	156.0	0.959	0.969	-
5-2	M	124.0	1.784	108.0	0.946	0.937	-
5-3	M	199.5	2.830	146.0	0.942	0.937	-
5-4	F	131.0	2.046	99.7	0.956	0.933	-
5-5	M	187.5	2.571	164.0	0.962	0.965	-
Mean					0.938	0.936	0.926
S. D.					0.047	0.045	0.025
Coefficient of variation					5.05%	4.84%	2.73%

The results with the obese patient who starved for 46 days suggest that the K^{42} was taken up by his skeleton. Before starvation his body potassium value (entry 1-1, Table 64) determined by isotope dilution averaged 89 per cent of that found by whole-body counting methods. After substantial weight loss (entry 3-3), the average ratio had increased to 96.5 per cent.

Discussion

As stated in the introduction, the accuracy of a potassium measurement with any technique can only be evaluated by comparison with the results obtained by other techniques. One of the tilting chair results listed in either column 6 or column 7 always agrees within 2.0 per cent of the value found with the 7-crystal-position technique listed in column 8, except for subject 1-4. The tilting chair value with the log crystal may agree with the 7-crystal-position technique for one patient while the 8 in. by 4 in. crystal over the tilting chair agrees with the 7-crystal-position technique for another patient. Analysis of the data presented in Table 62 suggests that a variability exists in the tilting chair measurement because of movement of the patient during measurement. Variations due to changes in background or energy calibration can be ruled out since this would have introduced variability into the 7-crystal-position technique measurement also.

Previous measurements with the tilting-chair technique demonstrated that the sensitivity varied almost 10 per cent if the subject radically changed his position while in the chair. Although the patients had been instructed to assume and maintain the same position during the tilting chair measurements, they undoubtedly shifted position during the 40-minute measurement for normal potassium since they usually fell asleep. Therefore, the patients numbered 5-1 to 5-5 were counted in the chair with their knees loosely strapped to the arm rests of the chair. It will be noted that the 8 in. by 4 in. crystal data and log crystal data agree within 1.0 per cent for the four of the five cases. Thus the variability noted with the tilting-chair technique can probably be reduced considerably if the patient's legs and hands are comfortably immobilized.

The results presented in this paper indicate that in the normal human subject K^{42} does not completely exchange with potassium in bone. This finding is demonstrated by the fact that the K^{40}/K^{42} ratio is higher in the head and feet of the normal subjects. The total-body potassium as found by isotopic dilution techniques is definitely less than that found by whole-body counter techniques and averages about 93 per cent of the latter values.

The results also demonstrate that more accurate measurements of potassium content in the body can be made by a single-crystal, 7-crystal-position technique along the supine body than with the tilting-chair technique. However, the accuracy of the tilting-chair technique can be improved by immobilizing the patient so that he cannot shift his position radically even while asleep.

In a previous paper⁽¹⁾ a correlation between exchangeable potassium and total body weight was interpreted to imply that the whole-body counter measurement contained a systematic error as a function of weight. It is now believed that both the previous isotopic dilution measurement and the

whole-body counter measurements of potassium in the obese patients were probably correct. The observation that potassium was 98 per cent to 100 per cent exchangeable for the very obese patients included in the previous study was probably due to the fact that those subjects were hospitalized, clinically ill patients who were essentially at bed rest and thus in negative calcium and potassium balance.

The comparison of the results obtained by different types of techniques presented in this report demonstrates the difficulty of evaluating the accuracy of potassium measurements. For example, the potassium content found with the whole-body counter is not particularly dependent upon the location of K^{42} in the body, while the value found with the isotope-dilution technique is influenced greatly by the uptake of potassium in the skeleton under certain physiological conditions. The fact that the two techniques yield different amounts of potassium in the body may be more valuable as a diagnostic tool than as a test of the validity of one technique against another.

Reference

1. Miller, C. E. and A. P. Remenchik. Problems Involved in Accurately Measuring the K Content of the Human Body. Ann. N. Y. Acad Sci. 110, 175-188 (1963).

Cesium-137
Cs¹³⁷ TRENDS IN HUMANS FROM 1955 TO 1964

C. E. Miller

A group of Argonne National Laboratory employees have been measured periodically for Cs¹³⁷ and K⁴⁰ content in the whole-body counter since 1955. These employees were chosen because they did not work with radioactive isotopes, and consequently their cesium content should be typical of that found in residents of the Chicago area. These data from measurements made on the same persons from 1955 to the present time are given here in detail to permit other investigators to correlate these results with their cesium data.

The measurements during 1955 and 1956 were made with a 4 in. diameter by $1\frac{1}{2}$ in. thick crystal located 10 cm behind the small of the back (deck-chair technique). Since 1957 they have been made with an 8 in. diameter by 4 in. thick crystal located over the tilting chair (tilting-chair technique). Both crystal-subject geometries were calibrated for potassium measurements by administration of known amounts of K⁴² to the subjects. The tilting-chair technique was calibrated for Cs¹³⁷ measurements by administration of Cs¹³² to a separate group of patients. The deck-chair technique was then calibrated by use of both techniques and tilting-chair value. Each subject took a shower and donned radioactivity-free clothing before entering the whole-body counter for a 50-minute measurement.

The results of the individual measurements of five subjects from 1955 to 1964 and of five additional subjects from 1957 to 1964 are given in Tables 65 to 74. The average body weight and average body potassium content for each person are given at the top of each table. To facilitate comparison of these data with those obtained by other investigators, the cesium content is expressed in three ways: The Cs¹³⁷ content is given in mμCi in column 2, it is expressed as μμCi per kg of body weight in column 3, and as μμCi per gram of body potassium in column 4. The results of the Cs¹³² calibration demonstrated that the Cs¹³⁷ measurements given in column 2 contain an uncertainty of ±6 per cent because of the varying sensitivity of the counting system for subjects of different heights and weights. The quantities given in column 3 contain a still larger uncertainty because of fluctuations in body weight with time. The uncertainty in column 4 is greater than that in column 2, because it includes the variability in the potassium determination, but it is not as great as that in column 3.

Analyses of these measurements have demonstrated that the short-term variations of Cs¹³⁷ in an individual reflect his dietary habits for that period rather than the general fallout level. For example, the appreciable increase in cesium content of subject B from September, 1958 through June, 1959 was due to a sizeable increase in his consumption of milk during

TABLE 65

SUBJECT A Weight 81.5 kg
Body K 149.0 g
Sex Male

Date	$\mu\text{Ci Cs}^{137}$	$\mu\text{Ci Cs}^{137}$ kg body wt	$\mu\text{Ci Cs}^{137}/\text{g K}$
1955 7-6			8.0
12-5			15.4
1956 2-9			20.0
3-14			20.0
4-5			20.0
4-23			20.0
8-14			19.6
1957 2-18	6.50	79.75	29.5
6-17	5.00	61.35	27.8
9-25	5.05	61.96	28.8
1958 1-16	5.96	73.13	33.0
1-22	6.07	74.48	34.0
4-16	6.57	80.61	35.0
8-1	7.75	95.09	43.5
11-18	8.50	104.29	46.2
1959 3-18	9.92	121.72	53.5
6-16	9.86	120.98	59.0
9-8	9.43	115.71	54.7
12-8	9.21	113.01	53.6
1960 5-4	8.02	98.40	48.2
12-18	5.05	61.96	31.6
1961 6-23	4.04	49.57	25.4
10-2	4.81	59.02	31.5
10-6	4.76	58.40	31.3
1962 3-21	3.29	40.37	22.4
7-17	4.59	56.32	30.4
11-16	6.38	78.28	42.7
11-21	6.27	76.93	39.8
11-29	5.77	70.80	37.1
12-7	6.44	79.02	39.5
1963 2-12	5.00	61.35	34.5
5-21	6.40	78.53	43.4
9-16	9.07	111.29	61.4
12-9	12.14	148.96	84.3
1964 2-10	13.18	161.72	90.7
11-5	14.64	179.60	94.6

TABLE 66

SUBJECT B Weight 80 kg
Body K 142 g
Sex Male

Date	$\mu\text{Ci Cs}^{137}$	$\mu\text{Ci Cs}^{137}$ kg body wt	$\mu\text{Ci Cs}^{137}/\text{g K}$
1955 7-5			18.3
8-26			22.7
10-10			25.5
1956 3-27			33.7
4-24			38.5
11-30			39.0
1957 3-22	5.36	67.00	33.4
6-17	5.89	73.63	41.3
8-1	7.05	88.13	45.1
8-2	7.38	92.25	49.8
8-5	5.95	74.38	40.3
8-7	6.35	79.38	43.0
9-23	6.83	85.38	39.0
1958 1-20	5.77	72.13	39.0
4-25	7.05	88.13	44.1
8-1	6.35	79.38	43.0
11-20	8.47	105.88	58.0
11-25	8.87	110.88	58.7
12-31	9.28	116.00	66.0
1959 3-19	11.65	145.63	76.0
6-12	11.55	144.38	81.0
9-30	9.34	116.75	64.2
12-16	8.38	104.75	53.3
1960 5-23	8.61	107.63	55.6
6-2	7.75	96.88	50.5
12-21	4.81	60.13	33.9
1961 6-26	4.65	58.13	32.1
10-2	3.76	47.00	27.6
12-4	3.63	45.38	29.1
1962 3-7	2.64	33.00	28.2
7-16	5.41	67.63	39.2
10-11	6.92	86.50	48.6
11-21	8.50	106.25	59.2
11-29	7.90	98.75	55.3
1963 2-8	9.20	115.00	62.8
5-21	10.47	130.88	73.7
9-16	13.61	170.13	97.8
10-11	15.66	195.75	115.6
12-13	18.24	228.00	128.4
1964 2-7	19.55	244.40	136.1
5-6	22.62	282.80	153.4
7-16	21.49	268.60	165.9
9-30	17.89	223.60	143.5

TABLE 67

SUBJECT C Weight 55.8 kg
Body K 87 g
Sex Female

Date	$\mu\text{Ci Cs}^{137}$	$\mu\text{Ci Cs}^{137}$ kg body wt	$\mu\text{Ci Cs}^{137}/\text{g K}$	Date	$\mu\text{Ci Cs}^{137}$	$\mu\text{Ci Cs}^{137}$ kg body wt	$\mu\text{Ci Cs}^{137}/\text{g K}$
1957 6-20	3.39	60.75	32.6	1962 2-28	2.43	43.16	29.7
10-30	4.56	81.72	47.4	7-16	3.80	67.50	44.0
1958 1-20	3.69	66.13	31.9	10-11	4.59	81.53	51.8
4-18	4.63	82.97	42.3	11-21	4.87	86.50	52.3
7-28	5.62	100.72	51.7	11-29	4.15	73.71	48.9
11-18	5.84	104.66	52.0	1963 2-20	6.88	122.20	78.9
1959 3-25	6.04	108.24	59.0	5-22	6.77	120.25	81.9
6-16	5.51	98.75	54.6	10-1	9.26	164.48	104.9
9-4	5.05	90.50	54.5	12-6	11.28	200.36	124.9
1960 6-2	4.70	84.23	49.0	1964 2-7	12.14	217.56	132.3
12-14	4.39	78.67	44.6	5-6	15.36	276.27	181.3
1961 7-5	2.30	40.85	25.3	9-30	11.81	211.65	147.1
7-6	2.85	50.62	32.7				
10-6	2.69	47.78	29.4				

TABLE 68

SUBJECT E Weight 56 kg
Body K 91 g
Sex Female

Date	$\mu\text{mCi Cs}^{137}$	$\frac{\mu\text{mCi Cs}^{137}}{\text{kg body wt}}$	$\mu\text{mCi Cs}^{137}/\text{g K}$
1955 3-24 9-19			10.5 20.7
1956 1-6 2-9 3-2 4-6 4-11 5-4 7-9 8-15			22.5 16.7 23.3 27.0 23.3 27.2 28.3 28.3
1957 3-7 6-14 8-5 9-24	2.94 2.29 2.31 2.33	52.50 40.89 41.25 41.61	26.8 21.6 23.5 23.2
1958 1-20 4-18 7-25 11-28	2.70 3.18 4.01 3.74	48.21 56.79 71.61 66.79	26.6 33.2 38.0 36.0
1959 3-19 6-16 12-7	3.29 3.95 4.16	58.75 70.54 74.29	29.0 37.6 40.5
1961 5-24 10-5	2.17 2.59	38.75 46.25	22.6 24.9
1962 3-20	2.09	37.32	23.3
1963 2-21 5-21 10-8 12-10	4.10 5.05 7.64 5.31	73.21 90.18 136.43 94.82	42.2 53.2 86.9 62.0
1964 2-17 11-16 12-10	9.06 8.54 8.15	161.78 152.50 145.54	105.5 97.9 92.4

TABLE 69

SUBJECT F Weight 72 kg
Body K 134 g
Sex Male

Date	$\mu\text{mCi Cs}^{137}$	$\frac{\mu\text{mCi Cs}^{137}}{\text{kg body wt}}$	$\mu\text{mCi Cs}^{137}/\text{g K}$
1955 3-29 7-6 8-24			9.7 12.2 12.6
1956 1-5 2-10 3-14 4-6 5-3 6-1 7-6 7-26 8-16			22.4 26.8 32.2 30.0 32.4 34.0 34.0 31.5 31.5
1957 5-22 10-28	5.57 4.71	78.12 66.06	36.5 33.8
1958 1-16 4-17 7-23 11-18	6.38 6.45 7.10 7.61	89.48 90.46 99.58 106.73	44.0 43.0 48.4 51.0
1959 3-12 6-16 9-14 12-7	7.70 9.21 9.42 9.46	107.99 129.17 132.12 132.68	61.8 65.5 64.0 63.1
1960 5-4 12-20	7.19 5.36	100.84 75.18	46.6 37.3
1961 6-23 10-5 12-4	4.11 4.15 4.25	57.64 58.20 59.61	29.6 28.4 32.9
1962 3-20 7-17	3.73 4.06	52.31 56.94	28.2 28.9
1963 2-19 6-5 10-1 12-10	7.33 9.28 12.51 14.38	102.81 130.15 175.46 196.71	54.4 70.6 92.9 109.7
1964 2-10 9-29	15.47 14.61	211.62 199.86	113.5 114.6

TABLE 70

SUBJECT G Weight 74 kg
Body K 152 g
Sex Male

Date	$\mu\text{mCi Cs}^{137}$	$\frac{\mu\text{mCi Cs}^{137}}{\text{kg body wt}}$	$\mu\text{mCi Cs}^{137}/\text{g K}$	Date	$\mu\text{mCi Cs}^{137}$	$\frac{\mu\text{mCi Cs}^{137}}{\text{kg body wt}}$	$\mu\text{mCi Cs}^{137}/\text{g K}$
1957 6-17	7.24	97.84	45.0	1962 3-21 12-5	4.72 10.48	63.78 141.62	29.9 69.2
1958 1-16 4-17 7-25 11-18	9.00 10.30 10.94 11.59	121.62 139.19 147.84 156.62	54.0 62.0 64.0 69.5	1963 2-19 6-5 10-1 12-26	13.14 15.56 19.49 25.01	177.57 210.27 263.38 337.97	86.2 96.8 134.4 174.7
1959 6-19	12.98	175.41	77.0	1964 2-17 10-8	26.48 27.20	357.83 367.56	177.12 190.66
1960 5-24 12-19	11.08 8.91	149.73 120.41	71.0 57.2				
1961 5-24 10-2	6.88 5.53	92.97 74.73	44.2 36.9				

TABLE 71

SUBJECT H Weight 82.6 kg
Body K 163.0 g
Sex Male

Date	mμCi Cs137	μμCi Cs137 kg body wt	μμCi Cs137/g K
1957 6-14	11.44	138.50	31.0
8-8	10.69	129.42	39.5
10-28	11.25	136.20	39.5
1958 1-16	11.14	134.87	44.5
1-21	11.15	134.99	40.3
3-21	12.02	145.52	38.5
4-25	11.87	143.70	42.0
7-25	11.11	134.50	47.0
11-18	10.84	131.23	47.8
1959 3-18	10.68	129.30	54.5
6-17	10.64	128.81	57.2
9-8	10.88	131.72	45.5
12-8	11.30	136.80	42.2
1960 5-3	11.08	152.62	45.4
12-20	10.04	138.29	29.8
1961 6-23	10.09	111.57	30.7
10-5	9.94	136.91	22.4
1962 3-13	3.41	46.97	22.2
11-16	6.72	92.56	40.2
12-7	7.38	101.65	45.1
1963 2-14	8.67	119.42	53.4
6-5	11.87	163.50	72.2
9-17	14.80	203.86	91.4
12-9	18.50	254.82	113.6
1964 2-13	20.42	281.27	125.4
9-29	19.27	265.42	126.6

TABLE 72

SUBJECT I Weight 62.7 kg
Body K 91.5 g
Sex Female

Date	mμCi Cs137	μμCi Cs137 kg body wt	μμCi Cs137/g K
1957 6-20	3.05	48.26	31.0
8-5	3.07	48.58	29.6
9-23	2.53	40.03	23.0
1958 1-16	3.68	58.23	31.0
4-25	3.24	51.27	28.5
7-25	3.96	62.66	34.5
11-19	3.07	48.58	29.3
1959 3-23	4.56	72.15	38.7
9-8	4.31	68.20	40.2
12-8	4.39	69.46	39.9
1960 5-3	4.41	69.78	44.6
12-14	2.86	45.25	29.0
1961 5-24	2.22	35.13	23.9
11-10	1.86	29.43	21.8
1962 3-13	1.54	24.37	16.4
12-5	3.24	51.27	35.2
1963 2-12	2.26	35.76	28.2
5-21	4.00	63.29	41.2
10-1	7.53	119.15	82.3
12-13	7.61	120.41	81.0
1964 2-17	7.65	121.04	81.4
9-30	8.44	133.54	100.7

TABLE 73

SUBJECT J Weight 70.0 kg
Body K 136.0 g
Sex Male

Date	mμCi Cs137	μμCi Cs137 kg body wt	μμCi Cs137/g K	Date	mμCi Cs137	μμCi Cs137 kg body wt	μμCi Cs137/g K
1955 10-7			19.6	1960 5-23	8.92	127.43	64.9
10-10			28.6	12-15	7.53	107.57	55.9
1956 2-14			41.8	1961 7-5	5.56	79.43	39.9
4-5			34.7	10-2	4.66	66.57	35.8
5-7			45.0	1962 3-13	3.86	55.14	28.2
8-14			43.5	11-21	8.41	120.14	60.2
11-29			46.7	1963 2-14	10.62	151.71	77.8
1957 3-22	6.70	95.71	44.0	5-22	12.16	173.71	91.2
6-17	5.97	85.29	41.0	9-17	15.85	226.43	120.2
10-29	6.80	97.14	50.4	12-9	19.22	274.57	138.3
1958 1-16	7.26	103.71	50.5	1964 2-13	21.75	310.71	155.8
4-18	9.61	137.29	65.2	9-30	22.30	318.57	164.6
7-28	10.07	143.86	71.9				
11-20	9.57	136.71	68.5				
1959 3-19	10.93	156.14	75.2				
6-17	10.86	155.14	83.2				
9-30	9.58	136.86	68.2				
12-8	9.59	136.99	69.5				

TABLE 74

SUBJECT L Weight: 87.0 kg
Body K: 145.0 g
Sex: Male

Date	$\mu\text{Ci Cs}^{137}$	$\mu\text{Ci Cs}^{137}$ kg body wt	$\mu\text{Ci Cs}^{137}/\text{g K}$	Date	$\mu\text{Ci Cs}^{137}$	$\mu\text{Ci Cs}^{137}$ kg body wt	$\mu\text{Ci Cs}^{137}/\text{g K}$
1957 5-22	7.35	84.78	43.3	1962 2-28	3.80	47.56	26.9
5-24	6.48	74.74	40.5	7-13	4.50	56.32	30.9
10-29	6.03	69.55	38.4	10-11	6.20	77.60	42.6
1958 1-20	7.29	84.08	45.0	11-16	6.55	81.98	44.5
4-17	9.31	107.38	55.0	11-21	6.64	83.10	45.3
7-25	8.06	92.96	51.0	11-29	7.01	87.73	46.5
11-20	8.56	98.73	50.3	12-5	6.90	86.36	48.5
1959 6-17	9.24	106.57	57.3	12-7	6.32	79.10	43.8
9-14	10.0	115.34	61.7	1963 2-11	8.36	104.63	57.7
12-8	9.67	111.53	58.2	5-20	9.76	122.15	65.5
1960 5-4	8.97	103.46	53.1	9-16	13.79	172.59	93.6
12-20	6.68	77.05	42.7	10-11	14.70	183.98	103.7
1961 6-23	5.67	65.40	38.4	12-9	15.89	198.87	105.9
10-2	4.52	52.13	30.6	1964 2-10	19.41	242.92	123.3
10-30	3.46	39.91	23.0	9-29	16.71	209.13	130.7
11-10	4.63	53.40	34.4				
12-4	4.45	51.33	33.2				

that period (Figure 22). Conversely, it has been noted that Cs^{137} content as well as body potassium both decrease with a decrease in food intake and a corresponding weight loss. For example, the cesium content of subject A, who had been on a low-calorie diet since December, 1962, decreased from December, 1962 to February, 1963, while the cesium content of the other nine subjects increased during the same period. Furthermore, since subject A has remained on a low-calorie diet that does not include milk, his cesium content has not increased as rapidly during 1963-1964 as has that of subjects B, J or H.

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Figure 22. The cesium content in Subject B from July 1955 through September 1964

If the variations due to changes in diet are excluded, the secular trend of cesium in these ten human subjects follows a similar pattern. The average cesium content of the group increased from March, 1955, when its presence was first detected, to May, 1956; it remained rather constant from May, 1956 through May, 1957, after which time it increased and reached a maximum value in June, 1959. It then decreased from May, 1959, reached a minimum in about October, 1961, and remained constant until December, 1962. The cesium content increased during 1963 and reached a new maximum in mid 1964, which was twice that of mid 1959. The cesium content is now apparently decreasing. The average cesium content during 1961 was equal to that present during the fall of 1955.

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