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Low-intensity Spectrometry of the Gamma Radiation Emitted by Human Beings

By C. E. Miller *

Equipment and techniques have been developed to study the metabolism of gamma-ray-emitting elements in the intact, healthy human body. With these techniques it has been possible to identify, locate and measure less than 10^{-9} c of gamma-ray emitters in the body and to follow the transport of these elements from one organ to another. As a result, accurate data have been obtained on the elimination rates from the whole body, on clearance rates for particular organs such as the lungs and on the fraction of the total inhaled quantity retained in the various organs.

Extensive data have been obtained by studying individuals who have accidentally inhaled quantities well below the Maximum Permissible Level (MPL) as set forth in Handbook No. 52, National Bureau of Standards.[1] These data obtained with healthy normal humans are valuable since they provide a direct check on the currently accepted elimination rates for man, which have been extrapolated from animal data.

The long-term effects ~~of radium in~~ humans [2] are under direct investigation by clinical studies on individuals who painted watch dials 30 to 40 years ago. A body content of 10^{-9} c of radium, approximately the quantity retained 30 years after an intake of the presently accepted MPL of 10^{-7} c, can be measured quickly with the facilities which will be discussed in this paper. It is thus possible to select for clinical study individuals who contained an amount about equivalent to a MPL 30 years ago.

EQUIPMENT

Detector

Since all normal unexposed humans contain at least two radioactive elements, a measurement which shows only the total number of gamma rays leaving the body per minute without information as to their energies (spectrum) is useless. The detector and associated electronic circuitry must provide some degree of energy discrimination for even a simple measurement of a subject's potassium content. The accuracy, sensitivity, and reliability of any measurement of the radioactivity of a man increases with improvement in energy discrimination (resolution) since interfering gamma rays are detected.

A sodium iodide (thallium activated) [NaI(Tl)] scintillation crystal, coupled to an appropriate photomultiplier tube, is used as the detector in the Argonne facility since it has spectrometric properties far superior to those of liquid and plastic scintillators of low atomic numbers. One advantage is that gamma rays of considerably lower energy can be measured with NaI(Tl) crystals without using special techniques to reduce the phototube noise since the light output per thousand electron volts of gamma-ray energy is considerably higher. When measuring the radioactivity of the human body, the sensitivity to low energies (less than 40 kev) is determined by the internal absorption in the human body and not by the properties of the crystal.

The physical dimensions of the crystal chosen for a particular study depend on the energies of the gamma rays to be measured and the resolution desired. Since the background increases approximately as the volume of the crystal, a crystal thickness is chosen which yields the highest ratio of signal to background at the photopeak. For example, since a crystal 5 mm thick totally absorbs 60-kev gamma rays, the use of a thicker crystal yields a higher background with no increase in signal. It is desirable to use a crystal of large diameter for all measurements in order to intercept as many gamma rays from the body as possible. Thin crystals of large diameter coupled to phototubes currently available are of questionable value if high resolution is required. The photons of light liberated in such a crystal are absorbed on the adjacent area of the photocathode surface and are not diffused uniformly over the entire surface. Thus the resolution of a thin crystal for an uncollimated source is limited by the nonuniformity of the photocathode surface of the photomultiplier tube. A number of small crystals, each coupled to its own small phototube (which possesses more uniform cathode response), will yield better results.

Large-diameter crystals may be used when measuring gamma rays of higher energy. As the thickness of the crystal is increased, the light quanta are scattered over larger and larger areas of the photocathode surface, and the resultant resolution for an uncollimated broad beam source becomes the

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average value for the entire photocathode surface. Resolutions are 12.5% for Cs^{137} and 9.5% for K^{40} with a crystal 20 cm in diameter and 10 cm thick when optically coupled to a single DuMont phototube of 12.5 cm diameter (Fig. 1). The resolution, when using a single 12.5-cm phototube, drops rapidly as the diameter of the crystal is increased beyond 20 cm. Several phototubes must be used on the larger crystals if resolutions are to be acceptable. However, this is done at the expense of convenience of calibration.

The spectra result from analyzing the voltage pulses at the plate of the photomultiplier tube by means of a 256-channel pulse-height analyzer.[3] Information on the selection of radioactively clean components and the mounting of the crystal has been published elsewhere.[4]

Measuring Facility

In order to achieve a good signal-to-background counting rate ratio, the detector and subject to be counted must be shielded from the gamma rays originating in concrete and brick building materials. In the absence of such shielding, placing a subject in front of the crystal decreases the net counting rate of a detector because of the shielding provided by the subject's body. The measurements under discussion were made in an iron-shielded room [4] with internal dimensions of $2.4 \times 2.2 \times 1.8$ meters. The walls, floor and ceiling are built up to a total thickness of 20 cm by iron plates 0.63 cm thick and of various widths staggered so that the joints do not coincide.

In order to evaluate the attenuation of various amounts of shielding, the walls, floor and ceiling were built up uniformly to various thicknesses, and gamma ray spectra were obtained (Fig. 2) with a NaI(Tl) crystal 20 cm diameter by 10 cm thick. To demonstrate the pattern of attenuation, the total background expressed in counts per minute in a given band in the neighborhood of the K^{40} photopeak (1.326- to 1.575-Mev band) is plotted as a function of wall thickness (solid line in Fig. 3). This curve is apparently composed of two components, the most intense of which is considered to arise from natural activity originating entirely outside the walls. This fraction should decrease exponentially as the thickness of the walls is increased. The second component results from the activity in the detector, detector mount, inner plates of the room and cosmic rays. This component should not change appreciably as iron is added to the outside of the room. Subtracting 34 c/m from the background at each wall thickness yields the straight dashed line, a result consistent with the hypothesis that, within this band, 34 c/m originate from cosmic rays and internal activity within the confines of the room and less than 1 c/m originates outside the room. A similar analysis performed across the spectrum indicates that, with 20-cm iron walls, about 1 c/m per 50-kev band at the low energies and 0.2 c/m 5per 0-kev band at the higher energies originate outside the room. The straight lines obtained in the same way at other energy bands had slopes practically identical to the one above. The mass absorption coefficient calculated from the slopes is 0.045, which is the mass absorption coefficient of iron for gamma rays in the region between 1.5 and 2 Mev. The reason for finding the same mass absorption coefficient for the low energy part of the spectrum is not immediately apparent. It could mean that most of the low-energy counts observed result from the degraded high-energy rays from radium and thorium in the surrounding concrete.

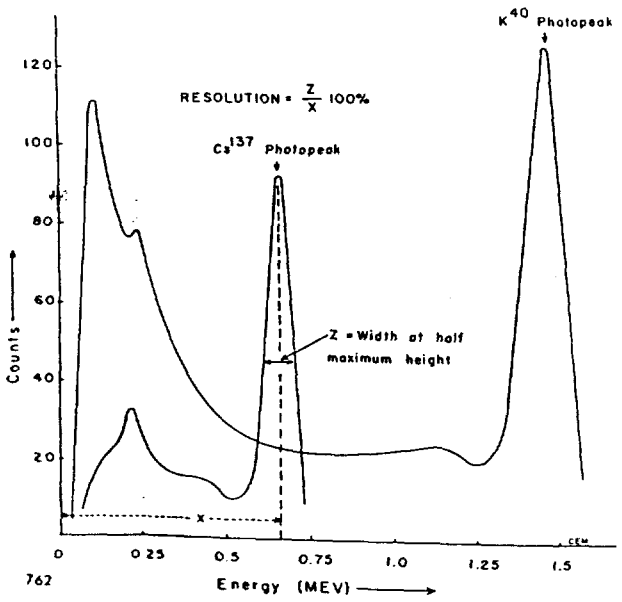

Figure 1. Spectra for K^{40} and Cs^{137}, demonstrating the resolution that may be realized with a single large crystal and the difference in amplitude at the lower energies, with respect to the amplitude of the photopeak, due to the quantity of internal scattering of a large (1 kg) K source vs. a small (1 g) Cs source

From the above studies, it seems evident that 95% of the background inside the present 20-cm iron room is indigenous to the room. Therefore, any practical amount of additional shielding on the outside will not reduce the background significantly. The low-energy background between 75 and 225 kev was reduced 40% by lining the inside surfaces of the room with 3 mm of lead free of radioactivity. The low-energy rays that are absorbed by the lead apparently originate within the iron. It has not been demonstrated whether these counts are due to the interaction of cosmic rays or to contamination of the iron plates.

The data given above demonstrate that it is illogical to build walls thicker than 13 cm until the radioactive content of the detector is reduced. It has been found that the radium and potassium contents of the photomultiplier tube envelope account for about 35% of the residual background and that the potassium content of the crystal (10 ppm) accounts for an additional 20%. Thus the background in the room will be cut in half when phototubes and crystals that are free of radioactivity become available. The realistic improvement which will result is discussed later.

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GENERAL TECHNIQUES OF TOTAL BODY MEASUREMENTS

Calibration

The techniques of analyzing gamma-ray spectra obtained from man, such as the standard errors involved, all obey the well-known laws of counting radioactive materials. The difference between measuring the radioactivity in the human body and in any inanimate sample is in the method of calibrating the sensitivity of the detector. The physical shape and size of the ordinary sample can be altered to yield a specified geometry with respect to the detector, and the sample can be mixed so that the activity is distributed throughout a uniform density. Thus the counter can be calibrated to yield absolute disintegration rates by using samples prepared to contain a known quantity of the radioelement in the desired mass.

The size and shape of the human cannot be altered; the radioactivity may be distributed throughout the body or concentrated in organs of relatively low density (such as the lungs) or of high density (such as the skeleton). As a result, the internal absorption and scattering varies not only because of the size of the subject but as a function of the location of the radioelement within the body.

The absolute quantity of any gamma-emitting element present in the body can be determined by administering an additional known quantity of the same radioelement to the human. The original quantity present is thus found by comparing the original counting rate with the increase from the known amount. The administered material must be in equilibrium (located in the same organs) with the original elements at the time of measurement, and the human must assume the same relative position with respect to the crystal in both counting situations.

This technique is applicable only to those radioelements which have a very short physical half life (K^{42} for example) or a short biological half time in the body. The calibration determined for one material cannot be used for other elements in the body unless the two elements are metabolized in identical fashion and the gamma-ray energies are very nearly equal. The calibration determined for a particular element can, however, be used for the same element in another individual of the same body build.

In the case of elements that have a long biological half time in the body, the gamma-ray spectrum from a human must be determined with some geometrical arrangement which can be calibrated without administering radioisotopes to the subject if quantitative answers are to result. In addition, it is desirable that the amplitude and shape of the spectrum be independent of the subject's build so that the system need not be recalibrated for each individual.

"One-Meter-Arc" Technique. No single general technique can be used for all measurements *in vivo*. Various techniques that are applicable to particular types of measurements have been perfected and calibrated. One measuring technique, which may be calibrated with sources placed in a Presdwood phantom, is called the "one-meter-arc" technique. An empirical procedure of calibrating this method has been published elsewhere;[5] the "trial and error" method given here does not require any assumptions.

The subject lies on his side on a thin metal bed with his back or front touching the inner surface of

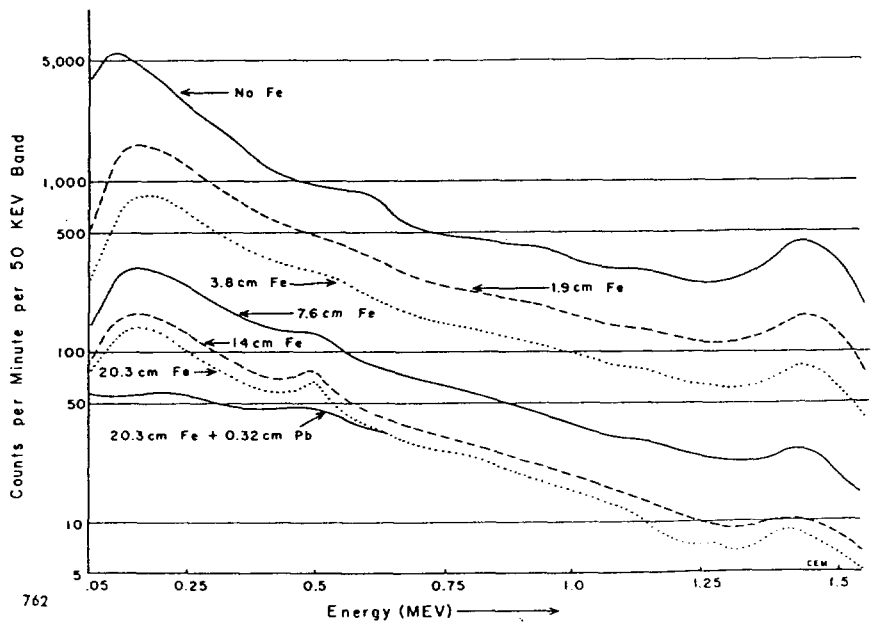

Figure 2. Reduction in gamma ray background of NaI (Tl) crystal 20 cm in diameter by 10 cm thick when the crystal is shielded with various thicknesses of iron and lead

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a thin form that has a radius of 1 meter. The crystal is placed at the center of curvature of the arc. The subject's gamma-ray spectra are obtained with the subject facing both toward and away from the crystal, and the radioelement present is identified by the energies of the photopeaks. A known quantity of the identified radioelement is then placed at a position equivalent to the center of the human, and various thicknesses of Presdwood are placed in front of and behind the source. The thicknesses of Presdwood on both sides of the source are varied independently until the shape of the phantom spectrum from the one side matches the spectrum obtained from the front of the human, and the phantom spectrum from the other side matches the subject's back spectrum. Agreement between the two pairs of spectra demonstrates the same scatter and absorption in the phantom as in the human. The quantity present in the human can thus be calculated.

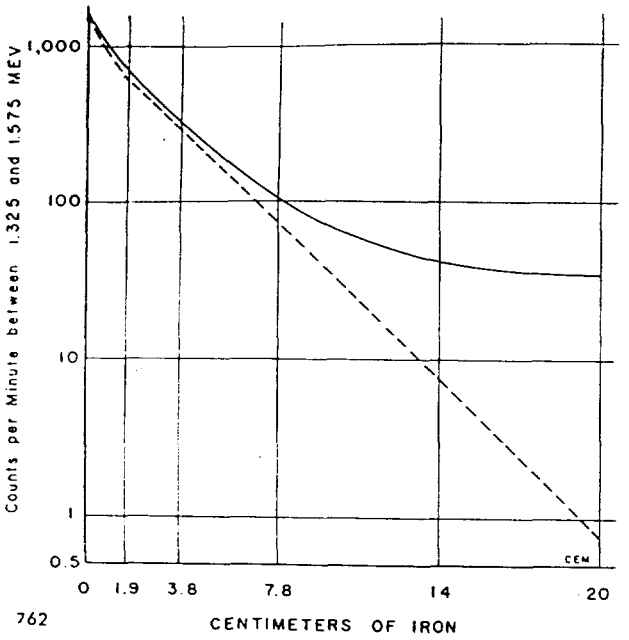

Figure 3. Plot (solid line) of the area under the gamma ray spectra of Fig. 2 from 1.325 to 1.575 Mev ; the straight dashed line resulted when 34 c/m was subtracted from the total at each thickness of shield

This technique may be used regardless of the location of the material in the body. Several trials may be required to match the spectrum if the front and back spectra are different, as is the case when the material is not uniformly distributed in the body. The sensitivity is poor since all volumes of the body are approximately 90 cm from the crystal and only a small percentage of the total gamma rays emitted are detected. These spectra-matching techniques will be expanded below.

Standard Chair Technique. In an attempt to find an arrangement that would yield higher counting rates and be relatively independent of body build, the following measurements were made. The gamma-ray spectra of 12 subjects of widely different builds were measured on the 1-meter arc and with the three crystal positions shown in Fig. 4. Each subject was then given 2 μc of 12.4-hr K^{42}, the excreta were collected for 24 hr, and the new gross spectrum was obtained with each configuration. The difference spectrum between the first and second runs is thus due to the presence of a known quantity of K^{42} distributed throughout the body. The amplitude of the spectrum above 500 kev was found to be less dependent on body stature if the crystal was placed at position 1, which is 42 cm from the seat and back of the chair, than when other positions were used. The counts per minute per microcurie in the photopeak for this position is practically independent of the height and weight of the subject. The total spread in counting rate from the smallest to the largest subject was only 5%. The counting rate from K^{42} when the crystal was in position 1 was 4.5 times the rate observed with the 1-meter arc.

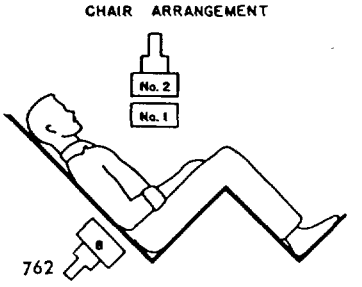

Figure 4. Relative position of NaI (Tl) crystal and subject under study ; B, early position used in many of our studies ; 1 and 2, latest positions found most independent of subject's anatomical configuration

However, once calibrated for a particular radioelement, the chair arrangement can be used to measure the body content of other individuals who contain the same element in the same organs of the body. The standard chair arrangement that meets the requirements of high sensitivity and lack of dependence of the calibration on body build cannot be calibrated with Presdwood phantoms but must be calibrated by measuring a person containing a known quantity of radioactive material. Furthermore, the calibration is dependent on the location of the radioelement in the body. The chair arrangement has been calibrated and is used routinely to measure body content of potassium, cesium, radium and thorium. Repeated measurements have demonstrated that quantitative determinations on new subjects using these calibrations are in error by not more than 5%.

Activity of the Normal Unexposed Man

Potassium Content. The potassium content of the 12 subjects was calculated from data obtained by the standard chair technique. The net K^{42} spectrum of each subject was normalized to make the area under the K^{42} photopeak equal to the area under the subject's K^{40} photopeak. When these manipulations were performed on spectra obtained in the spring of 1955, the normalized K^{42} spectra and K^{40} spectra were

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identical in shape and magnitude at all energies for most individuals, demonstrating that the K^{42} was in equilibrium with the subject's normal potassium pool (body content), that the procedure was valid, and that no significant quantity of any other isotope was present.

The potassium content of each subject was determined according to the following procedure. The gamma-ray spectra of a quantity of K^{42} equal to that in the subject diluted in 405 g of distilled H_2O in the bottle and of 405 g of ordinary KOH (containing 235 g of normal potassium) were obtained at a distance 40 cm from the crystal. The counting ratio for the K^{42} *in vivo* and the K^{42} in 405 g of H_2O must also apply to the K^{40} *in vivo* and to the K^{40} in the bottle. The subject's potassium content was then calculated from the equation

$$\text{Mass of K in body} = (235\ \text{g})\ \frac{V_{40}}{B_{40}}\ \frac{B_{42}}{V_{42}} \qquad (1)$$

where V_{40} and V_{42} represent the counting rates for K^{40} and K^{42}, respectively, in the body and B_{40} and B_{42} are the counting rates for the 235 g of natural potassium and for K^{42}, respectively, in the bottles. In each case the counting rate is the area in a 250-kev band under the photopeak.

Each subject's potassium content was calculated at all four positions, and agreement was excellent among all measurements on a particular subject. It is thus possible, with the data obtained with these 12 subjects in chair position 1, to estimate the potassium content of any other subject to within 3% regardless of his body build and without administering any radioactive tracer.

An accurate measurement of the body potassium content is of medical importance in studying various metabolic disturbances, for example, those following surgery or induced by disease. Also of medical interest is a determination of exchangeable body potassium. The fraction that is exchangeable can be determined easily. The subject's normal potassium content is measured, and then the subject is given a small amount of K^{42}. At any time after 14 hr, the amount of K^{42} present in the body and the amount of both K^{42} and normal potassium present in the urine are measured. The fraction that is exchangeable is then calculated from the equation

Fraction exchangeable

$$= \frac{\text{mass of normal K in urine}}{\text{mass of normal K } \textit{in vivo}} \times \frac{K^{42}\ \textit{in vivo}}{K^{42}\ \text{in urine}}. \qquad (2)$$

These measurements have been made on normal healthy individuals and are being published elsewhere.[6]

Cs^{137} Content. It was observed in March 1955 that some individuals contained about 10^{-9} c of some activity which emitted a gamma ray at about 660 kev. By September 1955, a photopeak at 661 kev could be discerned distinctly in all human spectra.[7] At this time, Dr. Jacob Sedlet (Head of the Bioassay Group, Industrial Hygiene and Safety Division, Argonne National Laboratory) chemically separated Cs^{137} from human urine.

Thirteen control subjects have been measured periodically since that time; the cesium content of eight of these is shown in Fig. 5. The cesium content is expressed as micromicrocuries of cesium per gram of body potassium to afford a direct comparison between subjects which is independent of musculature. The actual cesium spectra *in vivo* have been obtained for different body builds by subtracting the subjects' normalized K^{42} spectra, determined as mentioned above, from their total spectra *in vivo*.

ANALYSES OF TYPICAL *IN VIVO* SPECTRA

The subject's cesium and potassium spectra must be subtracted from the subject's gross spectrum, particularly if only a small quantity of the element under study is present. The *in vivo* cesium and potassium spectra obtained from the 12 control subjects have been used to analyze the spectra of other individuals whose normal spectra are unknown and who have accumulated a body content, either accidentally or medically, of other radioactive elements.

The biological half times of many elements in the human body can be determined with small quantities (less than the MPL) of an appropriate short-lived isotope of the element. Since the amount remaining in the body can be measured accurately at any time, the true retention curve can be followed for as many as ten half times. The ultimate sensitivity of these techniques also permits data of this sort to be obtained by studying individuals who have accidentally accumulated very small quantities during routine operations. Consequently, these data do not suffer from the uncertainties involved under experimental conditions but represent typical operational-type exposures which will be encountered in the future.

Techniques When the Normal Cesium and Potassium Contributions Can Be Subtracted Directly

Two patients who had ingested tracer amounts of 65-day Sr^{85} (emitter of a 513-kev gamma ray) under the care of Dr. R. J. Hasterlik of the Argonne Cancer Research Hospital, were measured 300 days later. To obtain the net spectrum of Sr^{85} in the body of each subject, the combined spectrum due to K^{40} and Cs^{137} in his body was subtracted from his gross spectrum. Since the patient's normal spectrum had not been obtained previous to his ingestion of Sr^{85}, the net K^{40} and net Cs^{137} spectra of an unexposed normal subject of the same height and weight were used. The control subject's net K^{40} spectrum, normalized to yield the same area under the K^{40} photopeak as under the patient's K^{40} photopeak, was subtracted from the patient's gross spectrum. The net Cs^{137} and Sr^{85} spectrum of the patient resulted (Fig. 6). The net Cs^{137} spectrum of the control subject was normalized and subtracted in the same way to obtain the true *in vivo* Sr^{85} spectrum

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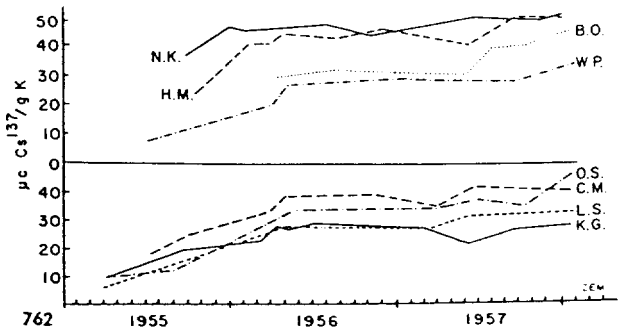

Figure 5. Cesium-137 content of eight individuals ; note equilibrium attained after spring 1956

of the patient. The absolute amount of Sr^{85} was calculated from this spectrum.

Subject RM had received 50 μc of Sr^{85} orally on Feb. 15, 1956 and was examined on Dec. 14, 1956, 302 days later. This subject contained 121 g of potassium, 5.5×10^{-9} c of Cs^{137}, and 9.4×10^{-8} c of Sr^{85}. Subject WSM, who had received 10 μc orally on Nov. 28, 1955 and was measured on Oct. 25, 1956, 332 days later, contained 121 g of potassium, 5.5×10^{-9} c of Cs^{137} and 1.0×10^{-8} c of Sr^{85}. The uncertainty in the Sr^{85} measurement when using a 50-min counting period would be 2×10^{-10} c if the subject's normal spectrum had been determined before ingesting the Sr^{85} tracer. Obtaining the normal spectrum before administering the tracer is particularly advantageous when the gamma-ray energy of the tracer is very close to the energy of either cesium or potassium. One advantage is that the net *in vivo* spectrum of the administered isotope resulting from a known quantity may be obtained, thus calibrating the detector.

Techniques When the Normal Cesium and Potassium Contributions Cannot Be Subtracted Directly

In some instances the subject's cesium or potassium spectrum cannot be subtracted as discussed above, since the gamma-ray energy of a foreign element may obscure the normal cesium or potassium photopeak. This situation occurred in the study of two subjects who had accidentally accumulated in the lung small amounts of Te^{129}, which emits three gamma rays of 0.465, 0.695 and 1.12 (low intensity) Mev, respectively. Although the potassium component could be subtracted, the 0.685-Mev gamma ray completely obscured the 0.661-Mev gamma ray from Cs^{137}. The assumption that these subjects contained an average amount of Cs^{137} would have introduced a relatively small quantitative error in the total amount of Te^{129}, yet would have invalidated the half-time determinations.

The subject's gamma-ray spectra were obtained weekly by the 1-meter-arc and the standard-chair techniques. The net spectral differences between weekly runs are the Te^{129} spectra *in vivo* corresponding to the fraction eliminated (Fig. 7). Two approaches may be used to determine the subject's normal spectrum from these difference spectra. In the "trial and error" method, the net Te^{129} difference spectrum was multiplied by an empirical factor and subtracted from the gross spectrum. The shape of the resultant spectrum at 465 kev was examined (dotted curve of Fig. 8), and the empirical factor was varied until the resultant energy spectrum showed neither a peak nor a dip at this energy. The final resultant spectrum for each subject was subtracted from his gross spectra taken on the various dates to yield the Te^{129} spectra *in vivo*. The biological half times can be determined from the areas under the Te^{129} curves without a knowledge of the quantity present. The trial and error method can be used to determine the biological half time regardless of the position used, but the total quantity present is not determined.

In the second or mathematical approach, a Te^{129} source was placed on the 1-meter arc and the physical arrangement of the Presdwood phantom enclosing it was varied until the phantom spectra matched the Te^{129} difference spectra, as discussed above. The quantity eliminated during each period could thus be deduced and an approximate biological half time calculated. With this information the total quantity present at the time of the first measurement could be calculated. Subtracting the corresponding spectrum from the gross spectrum gives the subject's normal spectrum.

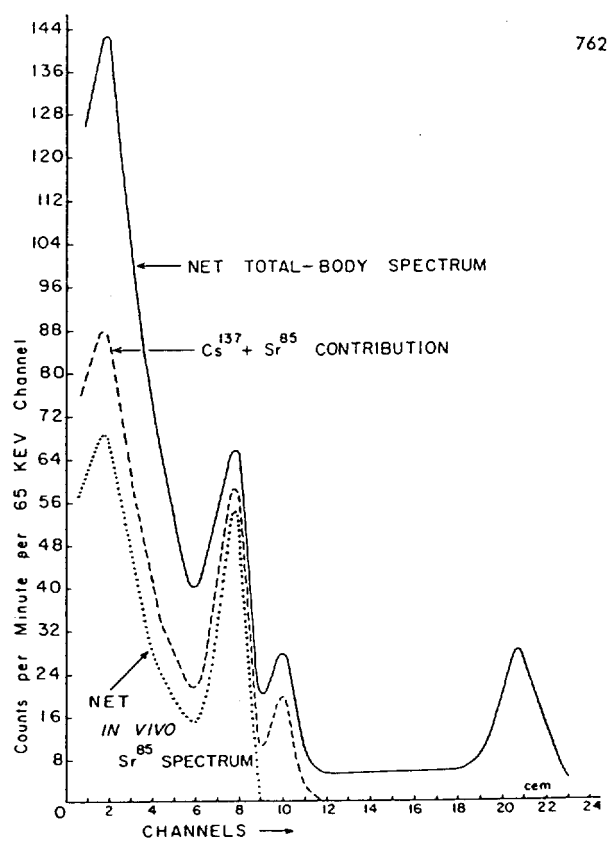

Figure 6. Analysis of gamma ray spectra of subject containing Sr^{85}

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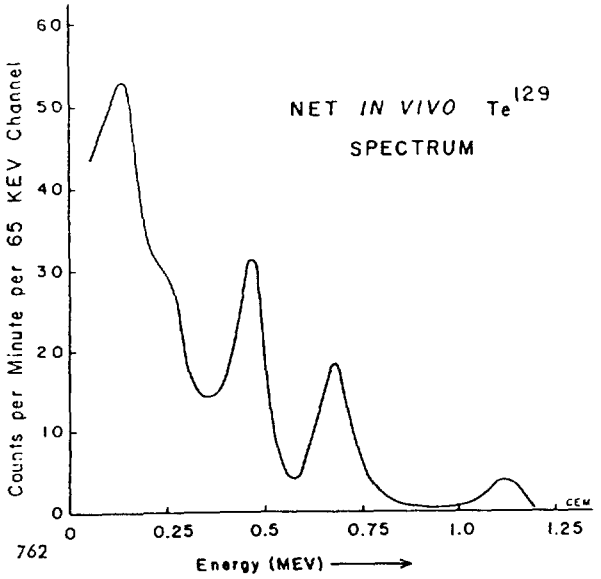

Figure 7. Net spectral difference between a subject's Te^{129} spectra on Nov. 8 and Dec. 14, 1956

Total body elimination rates are compared to excretion rates whenever possible. The two subjects submitted complete urine and fecal specimens from the 35th through the 39th day, and the activities contained were quantitatively determined by gamma-ray spectrometry. Approximately 1.25% of their total body content was eliminated each day, which is equivalent to a metabolic or biological half time of 55.1 days. This value combined with the decay constant of the isotope (half time=41 days) yields an effective half time *in vivo* of 23.5 days.

The body burdens of each subject, determined on the 32nd, 39th, 46th, 115th and 151st days, fell on straight lines on semilog plots with effective half times of 22 days for one subject and 22.5 days for the second. These two values are in fair agreement with the half time of 23.5 days estimated from the excretion rates on the 35th through the 39th days, and, as in previous tests, support the correctness of the measurements *in vivo*.

In these cases, the foreign material was inhaled and deposited in the lungs. Hence, the 22-day effective half life, constant from the 35th through 151st day, is representative of inhalation exposures only since the lungs acted as a reservoir and released the material into the rest of the body. There is reason to suspect that the inhaled material was not entirely an insoluble compound, since 60% of the amount excreted was present only in the feces.

Comparison of the spectra obtained on the arc with those obtained with the chair gave a chair-to-arc calibration factor. Thus the subject's body content was followed for at least two half times longer with the chair position than was possible with the arc. These Te^{129} spectra *in vivo* may be used in the future with the chair arrangement if the need arises.

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SPECIAL TECHNIQUES USED TO MEASURE THE AMOUNT OF ACTIVITY IN A PARTICULAR ORGAN

Techniques that may be used to determine the quantity of a radioactive element localized in a particular organ of the body have many applications. Measuring the radioactive content of the lungs is of great concern because of the possibility of inhalation of air-borne contaminants whenever radioactive materials are used. In many machining and processing operations, the individual may unknowingly inhale radioactive dust because of an improper operation on his part. It is impossible to calculate the quantity in the lungs of these individuals from excretion measurements since the fraction of the total lung content eliminated per day depends on many unknown factors. Some of these are the metabolism of the element, the chemical form, the particle size and the time elapsed between the intake and urine collection. In addition, when the body contains only a permissible quantity, the fraction of a permissible quantity excreted per day may be below detectable levels even if chemical separations are performed on the urine specimen. This is true in the case of Th^{232} since only 1 part in 10^6 of the burden is excreted per day.

Scanning by a Collimated Crystal

Since its spectrometric properties permit the identification of primary rays as distinguished from the scattered rays, the collimated NaI(Tl) crystal is the obvious detector with which to determine the distribution of radioactivity in the intact body. Whereas the total counting rate without energy discrimination (as determined with a plastic scintilla-

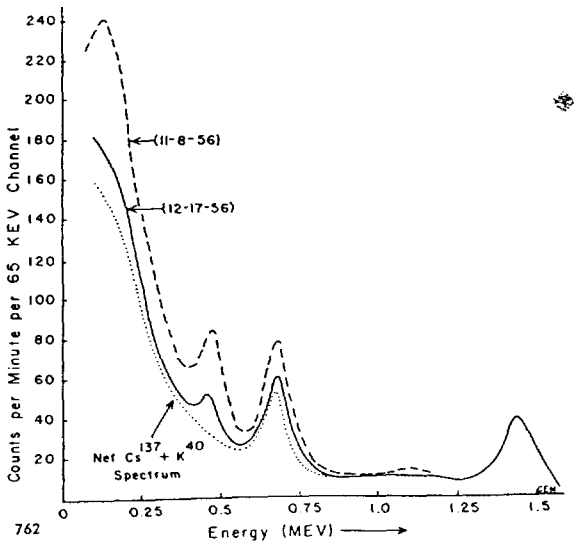

Figure 8. The spectrum of Fig. 7 was multiplied by the empirical factor 1.62 and subtracted from the spectrum of Nov. 8, 1956 ; subject's net Cs^{137} and K^{40} spectrum resulted (dotted line)

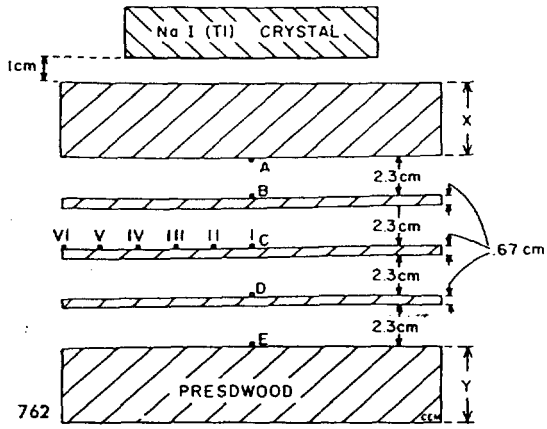

Figure 9. A NaI(Tl) crystal can be calibrated for measurement of radioactivity in the lungs with a Presdwood phantom using the above physical arrangement

tion counter) changes gradually as the collimated field of view is moved at right angles across the site of a source in a direction perpendicular to the line of sight, there is a rapid change in the counting rate under the photopeak (as measured with a NaI(Tl) crystal spectrometer). Thus the site of the element can be determined very accurately even though adjacent volumes of the body contain equal quantities of other gamma-ray emitters.

However, the collimated crystal method has two disadvantages. First, the actual sensitivity of the crystal is poor since only a small fraction of the total body content is measured at any one position unless, of course, the activity is concentrated in a very small area. Secondly, the absolute sensitivity must be determined with an appropriate phantom which, in practice, necessitates measurements with another technique in order to construct the phantom. The quantitative uncertainty of a measurement, while following the well-known laws of counting, may be restricted to less than 10^{-9} c of gamma activity if suitable counting times are used. Although the long time required to complete such a study is a momentary annoyance, valuable detailed data are obtained.

A machinist who inhaled a uranium compound containing U^{235} during a grinding operation has been measured on four occasions. On the first visit, the chest area of the subject was scanned with a collimated detector to determine whether the material was concentrated in any particular area or was uniformly distributed throughout the lungs. Nine points on a grid over the chest were measured. The horizontal grid lines were 34, 42 and 50 cm from the top of the head, and the vertical grid lines were located over the sternum and 8 cm to the right and to the left of the sternum. The highest readings were 8 cm to the right and left of the sternum, 34 cm from the top of the head. The values on either side of the sternum, 42 cm down, were about 20% less. The points 50 cm down and 8 cm right and left were about 25% of the top (32-cm line) values. The values along the sternum were somewhat lower than those on either side, showing that the material was present throughout the lungs. Distributions were essentially the same in those cases in which the individuals had inhaled Pa^{233}.

Large Uncollimated Crystal Close to the Chest

Although the total lung content can be determined by scanning with the collimated crystal scanner, the measurements require too much time to be used routinely. Hence, we investigated the feasibility of calibrating a large crystal for lung measurements. Spectra were obtained with the crystal positioned both 1 cm from the chest and 1 cm from the back of the U^{235} subject above, and similar measurements were made on an unexposed control subject of the same build. The control subject's spectra were subtracted from the patient's spectra to obtain the net spectra due to U^{235} in the lungs of the patient. A 100-mg uranium source was then placed in a phantom, which was arranged to have approximately the same density distribution as the subject's chest (Fig. 9). Thicknesses and the position of the source were varied until the front and back phantom spectra exactly matched those of the subject (Fig. 10). The fact that the spectra were identical at all energies proved that the absorption and scatter of the phantom were identical to those of the subject's thorax. From the known activity of the source, the lung content was determined. The total amount of uranium in the thorax calculated by this technique is given in Table I.

When plotted on a semilog graph, the first three of the above chest measurements fall almost on a straight line and indicate a biological half time in the lungs of 41.5 days. This is considerably shorter than the 125 days usually assumed. However, the values from the last two determinations (Dec. 10, 1957 and March 18, 1958) yield an average half time of 100 days, which approaches the accepted 125 days.

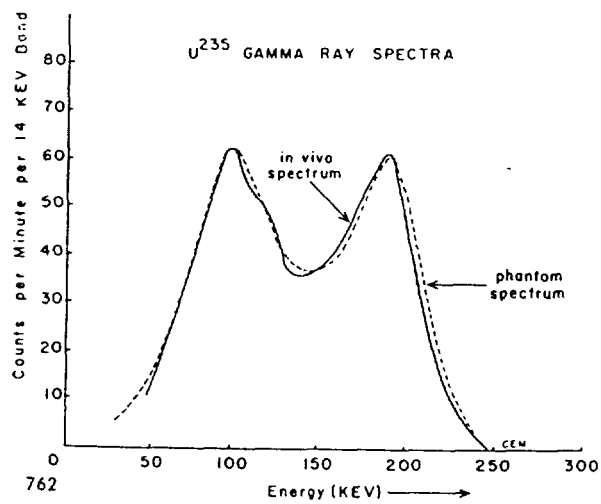

Figure 10. Comparison of *in vivo* and phantom spectra for U^{235}

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The subject's excreta were collected and analyzed by the employer's bioassay group. Both quantities measured in the excreta and the quantities leaving the lungs (as determined by these chest measurements) are given in Table 1. These quantities are in very good agreement, proving that the total quantity in the lungs as determined from the phantom calibration is, on the whole, correct. When the phantom representing the lungs was constructed by the trial-and-error technique, the arrangement of masses to match the U^{235} spectrum *in vivo* was identical with that to match the Te^{129} spectrum *in vivo* on the 1-meter arc.

The standard uncertainty has been calculated for a 10-min run used in measurements similar to the above and found to be equivalent to 30 μg of U^{235} in the lungs. This value is considerably below the MPL for the lungs. It would thus be practical to monitor all employees directly and thereby determine the total quantity present without all the above-mentioned uncertainties connected with excretion measurements. The simultaneous use of two crystals, one in front and one behind the subject, would allow further reduction of either the uncertainty or the counting time.

The biological half times in the lungs have been found for two other elements. The half time of Pa^{233}, calculated from data obtained with the techniques discussed above from two individuals, is 120 days during the period from the 30th through the 90th day after exposure. Although the activity could have been measured for several additional half times, this study was discontinued after the 90th day since the individuals being measured were employed several hundred miles away from our laboratory and had to be flown in for each measurement.

The biological half time of Te^{129}, based on measurements on two subjects discussed in the section entitled "Techniques When the Normal Cesium and Potassium Contributions Cannot Be Subtracted Directly," is 66 days during the period from the 30th through the 151st day after exposure. The short physical half life of Te^{129}, coupled with the small original content, discouraged additional measurements although they could have been continued for an additional 40 days.

Marinelli *et al.*[8] have studied a group who had inhaled $RaSO_4$. They found an average half time in the lungs of about 112 days for the period from 7 days until 700 days after the incident.

CONCLUSIONS

The extreme flexibility of the facility discussed in this paper permits many studies which cannot be carried out with other systems. By varying the position of the crystal with respect to the body or by using a collimated crystal, quantities larger than 5×10^{-10} c of gamma activity present in any volume of the body can be determined. On the other hand, 4π counters and those using fixed arrays of NaI(Tl) crystals can be used only to measure the total radioactivity present.

The time necessary to measure the total body activity can be decreased by using additional crystals. For example, an array of three or four crystals at the appropriate distance in front and a similar row behind the subject will undoubtedly yield a position-independent arrangement with which the subject's potassium and cesium contents can be determined with high accuracy in a short period of time. In addition, the array of several crystals will undoubtedly be very valuable for routine monitoring of large numbers of employees to determine, for example, the lung content of U^{235} as mentioned above. However, it has not been deemed desirable to use an array of crystals for the facility under discussion because of the increase in cost, the added complexity of energy calibration and the relatively limited advantage obtained.

A further decrease in the gamma-ray background, beyond the 50% reduction, which will be realized when phototubes free of radioactivity become available, will not result in a large improvement in sensitivity. Since the background spectrum in the Argonne facility is extremely stable and comparable in magnitude to a subject's normal spectrum, the uncertainty of an *in vivo* determination of the amount of any gamma-ray emitter whose energy is below 1.46 Mev results from the uncertainty of the human's own potassium cesium spectrum and not from the background uncertainty.

An individual's potassium content can be measured with a standard uncertainty of less than 5% by using

Table 1. Data on U^{235} Subject

Date of measurement	*Total amount of U^{235} in chest area (mg)*	*Decrease of U^{235} (mg)*	*Quantity of U^{235} in excreta during these periods (mg)*
November 2, 1957	2.75	0.7	0.74
November 19, 1957	2.05	0.58	0.48
December 10, 1957	1.47	0.77	0.78
March 18, 1958	0.7	—	—
Total	—	2.05	2.00

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only a 10-min run with one large NaI(Tl) crystal, 20 cm diameter by 10 cm thick. If the subject has a large body content of some other radioelement, the data from the one 10-min standard-chair run suffice to identify the element present and, in most cases, to determine the amount. Thus practically any desired measurement of the radioactive content of the human can be made.

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