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Health Physics Pergamon Press 1968. Vol. 14, pp. 439–447. Printed in Northern Ireland

FS 174
SFS 17.001

A DIRECT CALIBRATION USING GAMMA SPECTROMETRY FOR MEASURING RADIOACTIVITY IN HUMANS

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(*Received* 13 *July* 1967; *in revised form* 2 *October* 1967)

Abstract—Perhaps the major problem encountered in measuring radioactivity in humans is the fundamental calibration of the instrument. In this paper, a procedure is described by which the quantity of *in vivo* radioactivity can be evaluated entirely from theoretical calculations, eliminating the need for obtaining radioactive standards necessary in the usual empirical methods of calibration. Knowing the geometrical source-to-detector configuration and the mass absorption coefficient for NaI, the number of gamma-rays emitted from a source that are totally absorbed in a known volume of crystal can be calculated. Gamma-rays which are partially absorbed by the detector are corrected for by applying the appropriate factor from a peak-to-total ratio plot which has been prepared. Absorption of the gamma-rays in muscle and bone can be predicted by using the mass absorption coefficients provided in the paper. Thus, the gamma-rays found in the photopeak of the gamma spectrum can be measured and used to determine the number of gammas emitted by the source.

Although the technique can be applied to any gamma emitter, the reliability of the method is shown by the results of administering ^{40}K, ^{42}K, ^{41}Ar, ^{54}Mn, ^{133}Cs, ^{137}Cs, to human volunteers and measuring the response. The proposed procedure gave a smaller error than the conventional empirical method.

INTRODUCTION

PROCEDURES for measuring body radioactivity have evolved to include many counting configurations and detector arrangements. Each new technique[1–3] attempts to achieve a maximum sensitivity with a minimum of uncertainty. The fundamental calibration of the counter is the major problem when making quantitative *in vivo* radioactivity measurements. Three effective methods of calibration are currently being used. The first and perhaps most widely used is the empirical method[4] in which the net response of a subject is compared with the response from a simulated body-shaped standard. Another empirical method[5] involves the administration of known quantities of a radioactive material to a standard sized subject who is then counted by the adopted procedure. This empirical method has the obvious advantage of being a direct calibration, but has the limitation of not being applicable to all nuclides and of being subject to change with body assimilation and translocation. A major disadvantage in the empirical calibration technique is that some tracers which are used in a calibration are difficult to obtain. Even when the nuclide is available, the waiting period necessary to produce the activity is objectionable. A third technique utilizes accidental ingestion of radioactivity in personnel to calibrate the counter. Quantification of the ingested activity is accomplished by making measurements of the subject's body radioactivity between excretions, then by collecting and analyzing all excreta. A procedure is proposed to calculate directly from the gamma spectrum the quantity of body radioactivity. Radioactive standards are not used for calibration in the proposed method; therefore, there are no procurement or replacement problems.

EXPERIMENTAL

Theory

A known volume of thallium-activated sodium iodide crystal will attenuate gamma radiation in a constant and predictable manner. The mass absorption coefficiency curve has been

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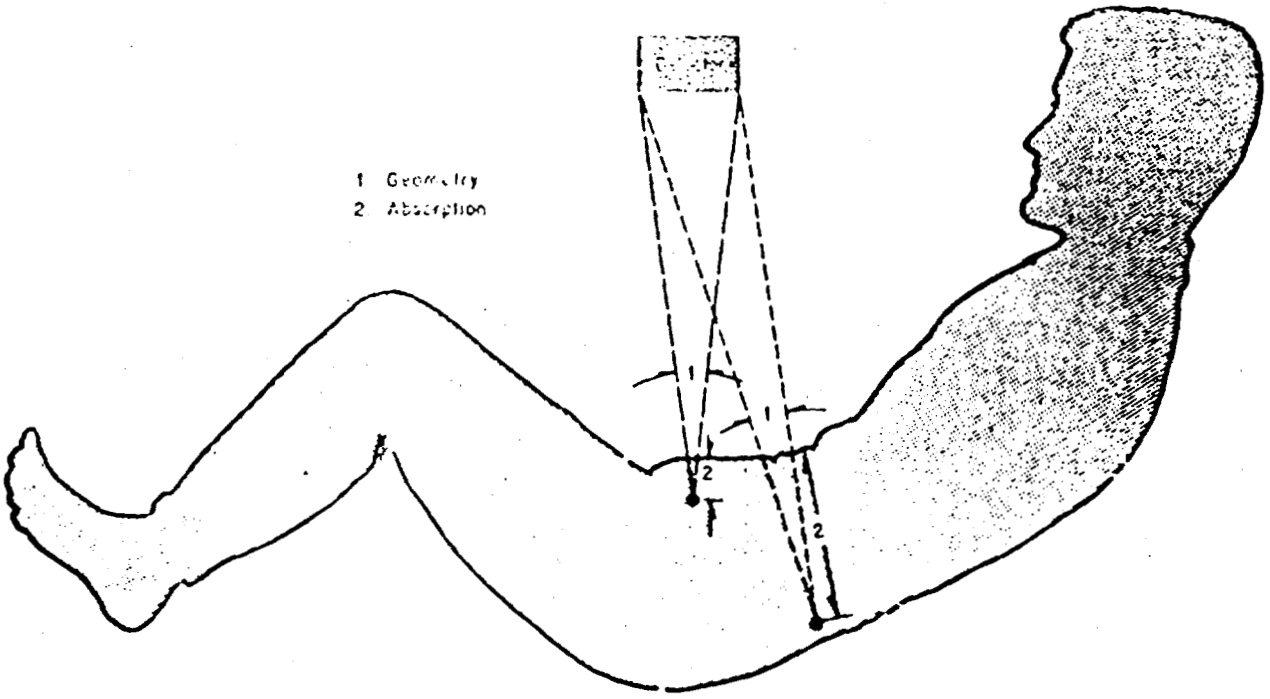

FIG. 1. Uncertainties in measuring body radioactivity.

published[6] for sodium iodide as a function of gamma energy giving the total cross section and the fractional components due to Compton absorption, photoelectric absorption and pair production. The interaction of gamma rays with a crystal and the pulse amplitude spectrum obtained from the reaction are discussed by many authors.[7–10] Knowing the source-to-detector configuration and the mass absorption coefficient for NaI, the number of gamma-rays emitted from a source that are totally absorbed in a known volume of crystal can be calculated. Gamma-rays which are partially absorbed by the detector are corrected for by applying the appropriate factor from a peak-to-total ratio plot which has been prepared. Absorption of the gamma-rays in muscle and bone can be predicted by using the mass absorption coefficients provided in this paper. Thus, the number of gamma-rays found in the photopeak of the gamma spectrum can be measured and used to determine the number of gammas emitted by the source.

Two uncertainties which must be considered when measuring body radioactivity, counting geometry and absorption in body tissue, are illustrated in Fig. 1. The first and perhaps the easiest to control is counting geometry. A subject might have radioactivity anywhere throughout an average body thickness of 22 cm. By assuming that the radioactivity is located on the body centerline the maximum uncertainity is the distance from the center of the body to the surface or 11 cm. A source uniformly distributed in the muscle will effectively be equal to the distance from the detector to the body centerline. Even when the radioactivity is selectively absorbed into a particular organ, uncertainties will be small if the centerline of the body is assumed to be the location of the activity because most organs lie in the central region. When the exact location of radioactivity inside a body is not known, uncertainties can be minimized by having the body 50–100 cm from the detector. Another technique which can be used to minimize the geometric uncertainty is to revolve the detectors around the body during the count.[3]

The second area of uncertainty in counting gamma-rays emitted from human bodies is absorption and scattering. Scattering of the gammas from tissue has little effect on the total energy gamma peak (photopeak) in a simple spectrum. However, the shape of the Compton

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Table 1. *Calculated absolute detection efficiency* T(E) *of a* 8 in. × 4 in. NaI *detector*

E_γ(MeV)	0.0622	0.105	0.152	0.212	0.332	0.566	1.10	2.04	3.80	5.50
h(cm)					Point source					
1.00	0.451	0.451	0.448	0.445	0.433	0.407	0.367	0.329	0.306	0.304
3.00	0.358	0.356	0.352	0.344	0.325	0.297	0.262	0.231	0.214	0.212
10.00	0.149	0.147	0.143	0.137	0.127	0.113	0.098	0.0867	0.0798	0.0790
20.00	0.0540	0.0534	0.0523	0.050	0.0472	0.0428	0.0376	0.0332	0.0306	0.0304
40.00	0.0153	0.0152	0.0150	0.0147	0.0141	0.0130	0.0116	0.0103	0.00956	0.00948
100.00	0.00256	0.00255	0.00254	0.00251	0.00245	0.00231	0.00209	0.00187	0.00174	0.00173
					Disc source with radius = 0.250 that of crystal					
1.00	0.450	0.450	0.447	0.443	0.431	0.404	0.364	0.326	0.303	0.300
3.00	0.356	0.353	0.349	0.341	0.322	0.293	0.258	0.228	0.211	0.209
10.00	0.147	0.145	0.141	0.135	0.125	0.112	0.0976	0.0858	0.0789	0.07814
20.00	0.0536	0.0530	0.0518	0.0501	0.0469	0.0426	0.0374	0.0331	0.0305	0.03019
40.00	0.0153	0.0152	0.0150	0.0147	0.0140	0.0130	0.0116	0.0103	0.00954	0.00945
100.00	0.00256	0.00255	0.0025	0.00251	0.00245	0.00231	0.00210	0.00187	0.00174	0.00172
					Line source with length/2 = 0.250 of crystal radius					
1.00	0.450	0.450	0.447	0.444	0.432	0.405	0.365	0.327	0.304	0.301
3.00	0.356	0.354	0.350	0.342	0.323	0.294	0.259	0.229	0.212	0.210
10.00	0.147	0.145	0.142	0.136	0.126	0.112	0.0980	0.0860	0.0792	0.0784
20.00	0.0537	0.0531	0.0520	0.0502	0.0470	0.0426	0.0375	0.0331	0.0305	0.0302
40.00	0.0153	0.0152	0.0150	0.0147	0.0140	0.0130	0.0116	0.0103	0.00954	0.00946
100.00	0.00256	0.00255	0.00253	0.00251	0.00245	0.00231	0.00209	0.00187	0.00174	0.00172
					Disc source with radius = 0.999 that of crystal					
0.50	0.432	0.430	0.424	0.413	0.391	0.360	0.322	0.290		0.267
1.50	0.360	0.356	0.350	0.339	0.319	0.292	0.260	0.233		0.214
3.00	0.282	0.279	0.274	0.265	0.248	0.226	0.200	0.179		0.164
10.00	0.118	0.116	0.114	0.110	0.103	0.0933	0.082	0.0729		0.0667
20.00	0.048	0.0473	0.0464	0.0450	0.0425	0.0388	0.0343	0.0305		0.0279

continuum is altered by scattering which complicates evaluation of complex spectra, but this alteration does not prohibit a quantitative appraisal. By assuming the location of the radioactivity is at the body centerline, an absorption correction can be made for each individual according to his body thickness. Thus, a proper absorption correction must be applied for each individual according to his body thickness. An empirical calibration normally does not permit individualized corrections.

Factors used in calculating body radioactivity

A. *Absolute detection efficiency.* Gamma-rays emitted from a radioactive source are detected if they emerge from the nucleus in the proper direction and interact with the detector. The probability of detecting a single gamma-ray can be predicted by considering the incident angle between the source and the detector and the mass absorption coefficient of the sodium iodide crystal for all the gamma reactions. A combination of these two parameters for all the gamma reactions has been calculated for the most used source-to-detector distances. The resultant absolute detection efficiency $T(E)$ is used to correct for the gamma rays emitted by the source which are not detected by the scintillation crystal. $T(E)$ for an 8 in. × 4 in. sodium iodide crystal including the energy range of gamma rays up to 5.5 MeV are given in Table 1 for various configurations. Efficiencies for the most-used configurations have been plotted in Figs. 2 and 3. The table lists values for various distances from the detector face to source of activity, h(cm), as a function of gamma-ray energy.

B. *Correction for angle of measurement.* The $T(E)$ values that are listed in Table 1 are correct when sources are on the perpendicular axis of the crystal. If the source is at some position not on the perpendicular axis of the crystal, a correction needs to be applied according to Fig. 4. These values were obtained by making measurements at points between 0° and 90° on the 34- and 40-cm arcs from the center of the

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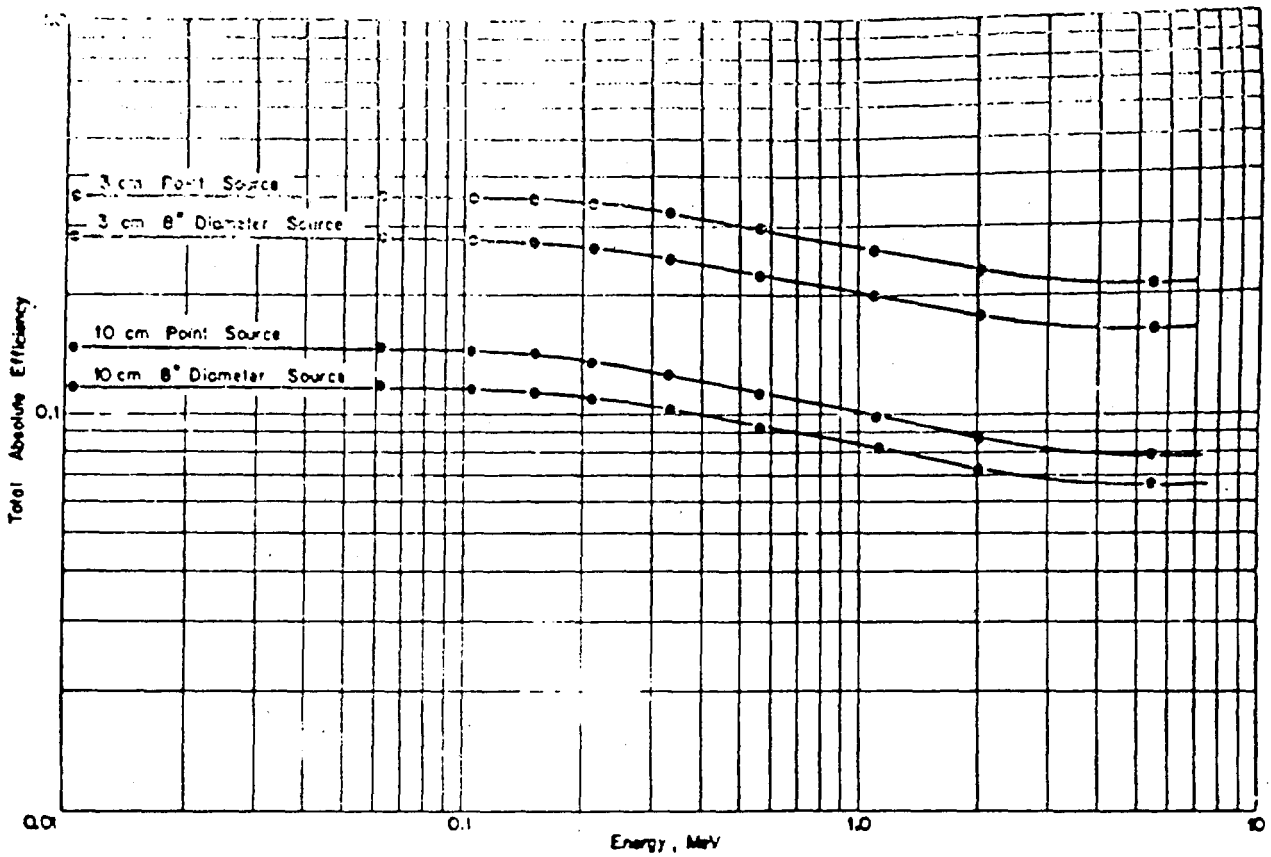

FIG. 2. Absolute detection efficiency for an 8 in. × 4 in. crystal.

crystal surface. A simple efficiency calculation is not easily made because of the variable thickness of the detector which is viewed by a point moving along an arc around the face of the crystal. Application of the correction is made according to the facts which are known. When the location of the activity is unknown, if the exposure was by inhalation it is assumed that the activity is located in the thorax region of the body. Those cases where ingestion is the mode of entry are calculated assuming the activity is in the abdominal region.

C. *Fraction of gammas in photopeak.* A gamma ray traversing a NaI(Tl) crystal having a given volume has a known probability of being annihilated and giving up its total energy. The total energy peak will contain a different fraction of the total gammas detected (peak-to-total ratio) as the volume of the crystal varies. Large crystals will be nearer unity. Low energy gamma-rays have a high probability of undergoing a photoelectric reaction in which the photon gives all its energy to a bound electron. As the gamma rays increase in energy, the photons give only partial energy to free electrons and continue at a different angle and escape from the detector. This Compton reaction is the predominant mode of interaction at 1 MeV. In large crystals, the original gamma-ray which has undergone one Compton reaction has a good probability for having a secondary or tertiary reaction and becoming annihilated thus being included in the total energy peak. Primary, secondary and tertiary reactions occur within such a short time interval that the light pulses are integrated and a single pulse emerges which is proportional to the sum of the two or three reactions. The final mode of interaction of a photon is pair production wherein a photon disappears in the field of an electron or a nucleus creating an electron-positron pair. Pair production starts at about 1 MeV and becomes the dominant mode of reaction at higher energies. The peak-to-total ratio determined in this study

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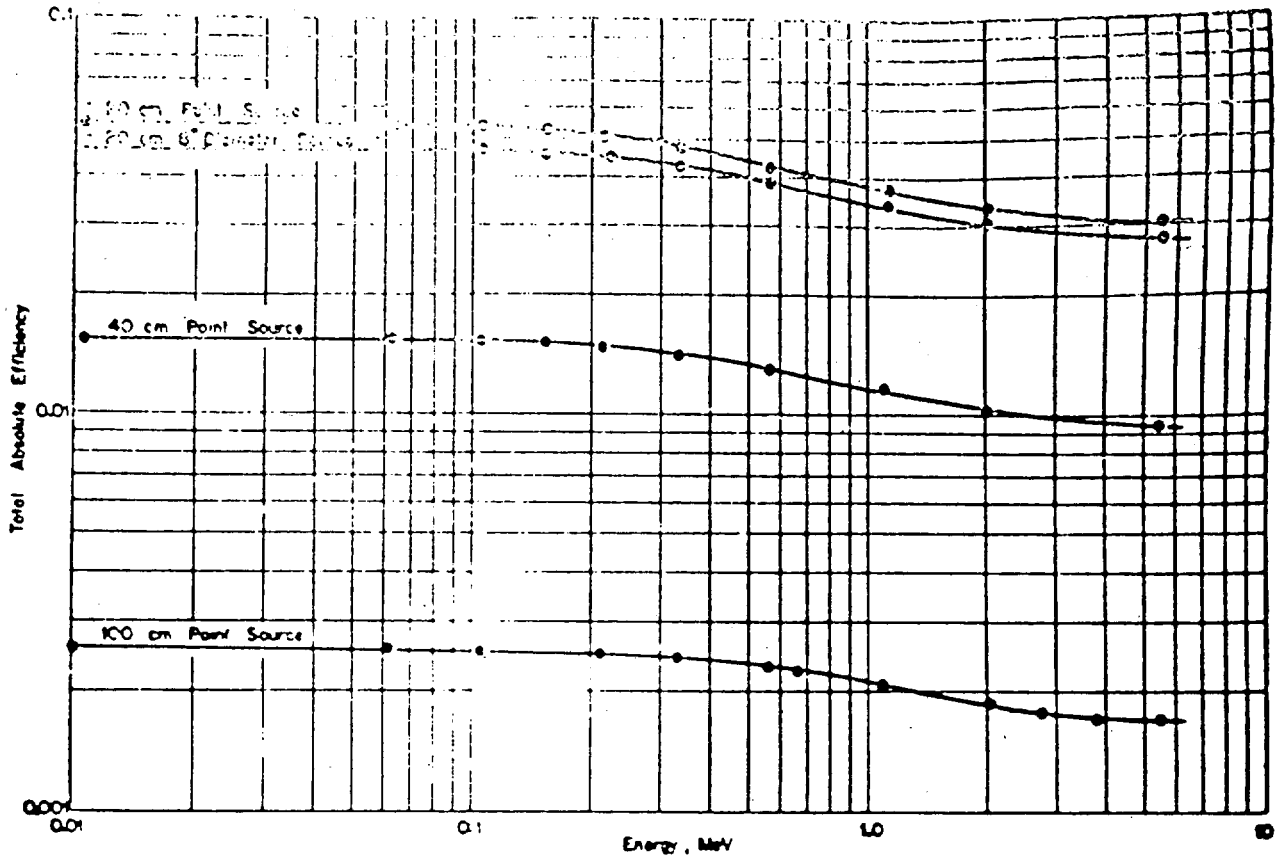

FIG. 3. Absolute detection efficiency for an 8 in. × 4 in. crystal.

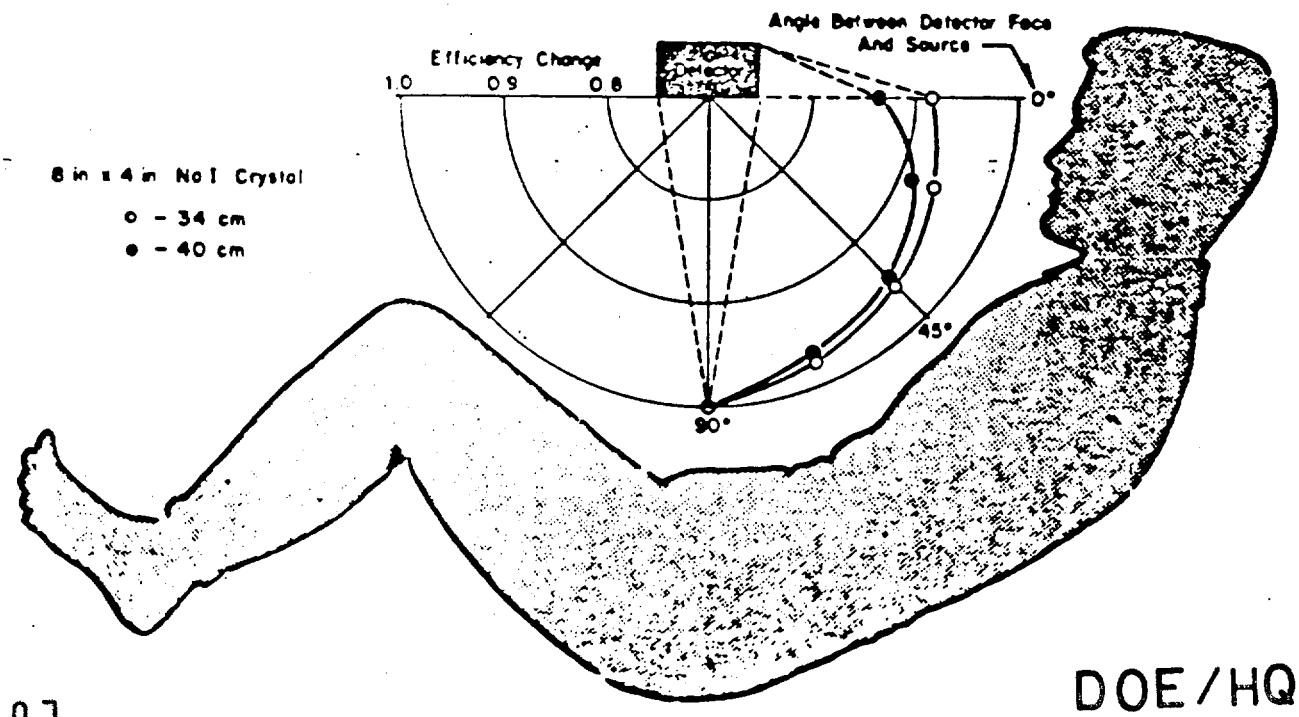

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FIG. 4. Correction for the detection efficiency as the detection angle varies.

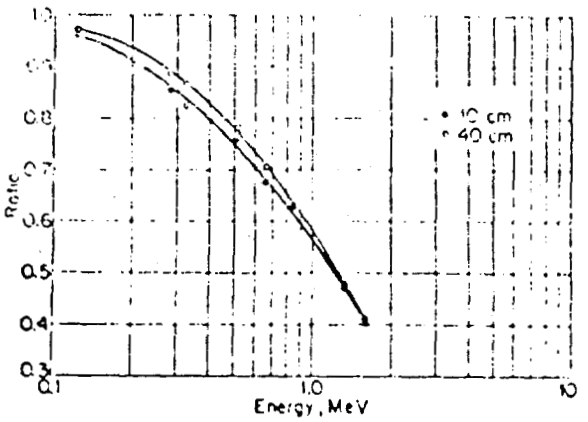

FIG. 5. Peak-to-total ratio for an 8 in. × 4 in. NaI detector.

was obtained using an 8 in. × 4 in. crystal for the gamma range from 0.1 to 1.0 MeV, and is plotted in Fig. 5. Standard sources of eight different nuclides were used to calculate the peak-to-total ratio using equation (2). A large shielded room with walls about 5 ft from the detector was used to minimize the error caused by gamma scattering occurring from the walls of the shield. The peak-to-total ratio is used in calculating the *in vivo* radioactivity by correcting for those events which are detected but are not included in the photopeak.

D. *Mass absorption coefficients for muscle and bone.* Gamma-rays are attenuated exponentially in matter when only the incident energy is considered. The intensity I_d transmitted through a thickness (d) in centimeters is given by the expression

$$I_d = I_0 \exp - (\mu/\rho) \cdot d\rho \qquad (1)$$

where I_0 is the incident intensity and μ/ρ is the mass absorption coefficient in cm²/g, and $d\rho$ is the surface density in g/cm². The density of muscle tissue was found to be slightly greater than 1 while the green bones had a density of about 1.2. Therefore, the density of body tissue was taken to be a 1.1 g/cm³. Green bones from beef cattle of relatively uniform thickness were used to measure the mass absorption coefficient for bone tissue. Absorption in muscle was measured using beef steaks stacked to give a variation of absorber thicknesses. Figure 6 is a plot of factors to correct for the absorption of gamma-rays in tissue and bones.

RELIABILITY AND UTILITY OF THE METHOD

The reliability of the method was determined by calculating the body radioactivity of individuals who had ingested a known quantity of radioactivity. The best available information about the location of the nuclide inside the body was used to decide upon the detection efficiency and the correction factor for absorption as the cited ^{54}Mn case. When the activity was known to be in the lung, the distance from the crystal to the center of lung was approximated. Both ^{137}Cs and ^{40}K are distributed throughout the muscle tissue; and would effectively average to be at the body centerline. Therefore, those cases where the location of the activity was unknown the calculations were made assuming the activity was at the body centerline.

Cesium-132 experiment

A supply of ^{132}Cs was made by irradiating cesium nitrate in a linear accelerator, (^{133}Cs(γ, n)^{132}Cs). A 3.5 μCi source was administered

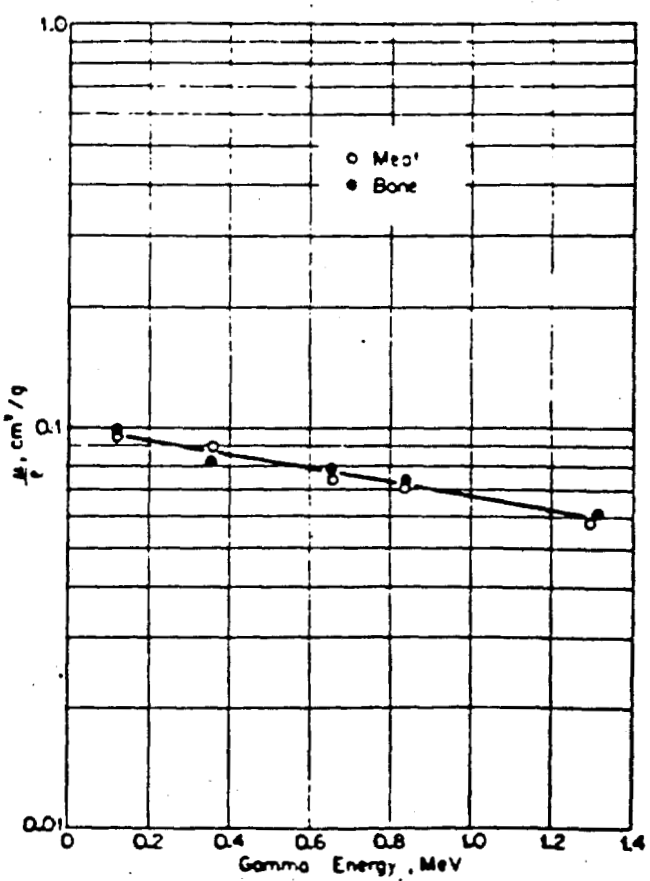

FIG. 6. Mass absorption coefficients for meat and bone.

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orally to a human volunteer. Prompt gamma-scans, delayed gamma-scans and excreta analyses were made during the week following the ingestion of the activity. A direct calculation of body radioactivity determined that 4.0 μCi of ^{137}Cs was present, which differed from the quantity taken by plus 14%.

Argon-41 experiment

Argon-41 was prepared by irradiating ^{40}Ar in the Materials Testing Reactor (^{40}Ar(n, γ)^{41}Ar). Calibrated sources of ^{41}Ar were inhaled and held in the lung while a 1-min gamma-scan was made, and the results are shown in Table 2.

Table 2. Quantity of ^{41}Ar taken and determined

Experiment	^{41}Ar taken (μCi)	^{41}Ar found (μCi)	Difference (%)
1	1.33	1.26	[illegible]
2	2.04	1.75	[illegible]
3	1.96	2.05	−5
4	2.25	2.62	−16

On the first two runs the exhaled gases were collected in balloons and analyzed. The first exhalation eliminated 60% of activity, the second eliminated an additional 20%. No detectable activity remained in the body after 5 min.

Potassium-42 experiment

Commercially available ^{42}K was calibrated and administered orally to a human volunteer. Both prompt and delayed whole-body measurements were made and the body radioactivity calculated assuming a distribution of the activity throughout the muscle tissue. The ingested 1.92 μCi source was calculated to be 1.84 μCi, indicating the result was low by 4%.

Cesium-137 and Potassium-40 experiment

Two individuals were counted as a part of an inter-calibration study of whole-body counters throughout the United States. One person had an elevated quantity of ^{137}Cs in his body whereas, the other had a normal body spectrum. A calculation was made to determine the ^{137}Cs and ^{40}K in each man and the results are shown in Table 3.

Manganese-54 experiment

A 1.12 μCi source of ^{54}Mn was encapsulated in polyethylene and taken by a human volunteer. Several measurements were made while the capsule was moving along the gastro-intestinal tract. A calculation to determine the quantity of ^{54}Mn was made at many different body locations. The results are reported in Table 4.

UTILIZATION OF TECHNIQUE

An employee at the NRTS reported for a whole-body count just before the end of the day shift. His body was found to contain enough radioactivity that a one minute measurement gave sufficient counts to accumulate a desired spectrum. The spectrum showed that ^{141}Ce and/or ^{144}Ce was the major activity present with minor amounts of ^{95}Zr ^{95}Nb and trace quantities of ^{137}Cs, ^{131}I and $^{103-106}$Ru. The 146 keV ^{141}Ce and 134 keV ^{144}Ce photo peaks were not resolved in the body count so a sample

Table 3. A comparison of the ^{137}Cs and ^{40}K results

Proposed method			Empirical method	
Present (μCi)	Found (μCi)	Difference (%)	Found (μCi)	Difference (%)
		^{40}K		
0.140	0.155	+11	0.154	+10
0.132	0.134	+2	0.132	0
		^{137}Cs		
0.251	0.243	−3	0.239	−5
0.025	0.025	0	0.023	−8

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Table 4. Quantity of ^{54}Mn at body sites

Location	Empirical method (μCi)	Difference from known (%)	Proposed method (μCi)	Difference from known (%)
Stomach	1.14	+2	1.05	−6
Stomach	1.09	−3	1.10	−2
Stomach	1.14	+2	1.09	−3
Small intestine	0.62	−45	1.01	−10
Small intestine	0.52	−54	0.84	−25
Small intestine	0.93	−17	1.53	+36
Transverse colon	0.77	−31	1.26	+13
Descending colon	0.40	−64	1.14	+2
Descending colon	0.42	−63	1.18	+5
Descending colon	0.41	−63	1.16	+4

of excreta was used to determine the fractional part of the photopeak from each of the cerium isotopes. A calibration had not been made for all of the nuclides which were detected. The procedure proposed in this paper was used to determine the radioactivity in this individual which made it possible to have a preliminary value to report in a matter of minutes. An initial whole-body count showed a body burden of 36 μCi of ^{141}Ce, 27 μCi of ^{144}Ce and 13.5 μCi of ^{95}Zr ^{95}Nb. A rota-scan count of the individual showed the activity was lower in the lung region and seemed to be localized in the abdominal area. Thus, a lower bowel enema, a cathartic and a series of injections of diethylenetriamine pentaacetic acid were administered. At the close of the fourth day his body burden had dropped to 0.1 μCi of $^{141-144}$Ce and 0.1 μCi of ^{137}Cs. A calibration by the empirical technique could have been made in a few hours if the nuclides which were found in the individual had been available in the laboratory. However, if the nuclides were not available in the laboratory, the time required to procure the radioactivity and then make the calibration would have taken days, which would have been intolerable. This typical case shows how important it is to have the capability of calculating the quantity of activity which is detected in a body so that the problem can be assessed in the shortest possible time. Early assessment allows the physician to take the necessary steps to reduce the radiation dose received by the subject.

RESULTS AND CONCLUSIONS

Quantitative assessment of radioactivity in a human subject can be made by integrating the area under the total energy peak and calculating the disintegration rate from the following equation:

$$D = \frac{\Sigma}{(T)(P)(A)(B.R.)\,(\theta)}. \qquad (2)$$

D is the disintegration rate; Σ, the area under the total energy peak in events per minute; T, the absolute detection efficiency for the distance from the source to the detector; P, the peak-to-total ratio; A, the absorption correction for the person counted; $B.R.$, the ratio of the gamma branch for the nuclide being calculated; and θ, the correction for angular variance of the source from the perpendicular axis of the crystal. The proposed procedure gave a smaller error in comparison to that obtained using the conventional empirical method. This reduction in error is possible because more latitude is given to the operator for making corrections needed to reflect individual differences in body configuration. The advantages offered by this method of calculating body radioactivity should be considered by those seeking a method for calibrating a whole-body counter.

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Acknowledgments—Special thanks are given to Mr. C. W. [illegible], Chief, Analytical Chemistry Branch, for his encouragement and technical advice in the writing of the paper. The tables of absolute detection efficiency were made available by Mr. R. L. HEATH whose computer program was used to make the calculations.

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