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IN VIVO MEASUREMENTS OF BONE-
SEEKING RADIONUCLIDES

P R O G R E S S R E P O R T

1977 - 1980

Principal Investigator:
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MASTER

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SECTION I

Estimation of the Skeletal Burden of "Bone-Seeking" Radionuclides from In Vivo Scintillation Measurements of Their Content in the Skull: Contribution from Radionuclides in the Thoracic Skeleton to In Vivo Measurements of Activity in the Lung

Introduction

A primary factor in establishment of realistic procedures for health and exposure surveillance in an expanding nuclear economy is the ability to identify and determine the magnitude of personnel exposure. Furthermore, health protection considerations in planning job protocols and medical decisions regarding such prophylactic procedures as decorporation therapy depend on the ability to accurately determine the amount and the site of internally-deposited radioactive contamination.

The degree to which an exposure has occurred has historically been based on data obtained by analytical procedures for determining the nuclide content in excreta samples, usually the urine. Even after 30 years of research into the metabolic behavior of the actinides in animals

and man, the unambiguous interpretation of bioassay results, i.e., actinide concentrations in excreta, remains difficult with respect to their relationship to the magnitude and distribution of the body burden (1). Although measurements of excreta may indicate whether or not an exposure has occurred and, if the time of exposure is known, may permit an estimate to be made of the approximate exposure magnitude, they often do not establish the absolute magnitude of a systemic body burden, the deposition site and the amount of non-transportable material in the lung.

A nuclide which has been measured in the urine is assumed to be of systemic origin, having reached the kidney through the bloodstream. Material which appears in the feces, however, can be either systemic in origin, arriving via a biliary route, or present in the gastrointestinal tract directly from an ingestion or inhalation intake. Therefore, a relatively insoluble (i.e., non-transportable) contaminant which is present in the lung, the GI tract, or some body compartment which retains the nuclide tenaciously (e.g., the skeleton), may never be evident in the urine or present to only a small and unpredictable extent at any specific time. When a contaminant is systemic, the often poorly understood relationship between its rate of excretion and the body burden, exemplifies the significant advantages offered by *in vivo* counting techniques made with external scintillation-type detectors.

Since most occupational exposures occur by inhalation, estimates of the amount of material deposited in the lungs are usually performed by external *in vivo* measurements with scintillation detectors positioned in proximity to the lungs, i.e., over the thoracic cavity.

After the initiation of NaI(Tl), spectrometric methods for detecting photon-emitting radionuclides in man, it became evident that the high background count rates in the low energy regions of these detectors precluded *in vivo* measurement of many important bone-seeking nuclides, such as ^{238}Pu , $^{239}/^{240}\text{Pu}$, ^{241}Am , $^{242}/^{243}\text{Cm}$, ^{210}Pb , ^{234}Th , ^{235}U , etc., at levels of practical interest. To meet these needs a dual crystal, low energy sensitive NaI(Tl)-CsI(Tl) detector system was developed at this laboratory (2,3) which employed electronic discrimination against higher energy Compton scattering events, thereby lowering the background count rate and enabling the efficient detection of photon emissions occurring below 200 keV (Table 1). After modification of the original design to obtain still greater sensitivity (4), the detector was ultimately adopted by most major laboratories having research interests and surveillance responsibilities for actinides and alkaline earth radionuclides (Table 2). To date this *in vivo* system, in combination with the more routine bioassay procedures for measuring activity in excreta, remains the optimum methodology for the determination of internal exposure to low energy, photon-emitting

radionuclides. An example of this dilemma is the case of uranium, a "bone-seeker" for which one must consider both the chemical toxicity to the kidney for systemic material as well as a possible radiological hazard to the lung (and perhaps for high specific activity in the skeleton) as the limiting case for insoluble material deposited at this site.

Since most occupational exposures occur by inhalation, and often to long-lived, relatively insoluble Class Y-type materials, internal deposits must be quantitated with these detectors placed above the lung area (over the anterior thorax). Since many of the alpha-emitting nuclides, which can be measured by their low-energy photons, are "bone-seekers" (i.e., concentrate primarily within or on the surface of the skeleton), it is necessary to account for the contribution of material which has been transported from the lung and has subsequently deposited in or on the thoracic skeleton (5). Furthermore, since the skeleton (or the bone-tissue interphase) has also been demonstrated to be a sensitive and important locus for radiation-induced osteosarcoma (6,7) Table 3, there must be an adequate method available for the assessment of radiation dose to this structure.

The present report presents data which will be useful in the accomplishment of these goals and will extend the usefulness of current *in vivo* measuring techniques by accurately defining the physical (detection) and biological

parameters used in performing accurate external scintillation measurements.

Current practice is to estimate the activity detected in the lungs by comparing the counts detected over the thorax with "phantom" lungs (surrogate structures) containing known amounts of the nuclide usually distributed in a uniform fashion. The underlying assumption is that the activity is associated with nontransportable materials of a refractory nature and will be almost completely associated with the lung tissue. The major concession made to the presence of bone is the use of phantom structures containing embedded skeletons to account for the great differences in attenuation by soft and hard tissue, for low energy (i.e., < 150 keV) photons. The presence of unlabelled skeleton is insufficient, however, since it has been shown that no inhaled material is ever completely insoluble; and even highly insoluble materials rarely ever remain completely deposited in the lung (whether by biochemical transformation or dissolution (8), phagocytosis by alveolar macrophages (9), or some other extracellular physical transport mechanism) Table 4. Since most transuranic materials have been shown to deposit along bone surfaces, there is a high probability that a significant portion of radionuclide in the thorax will eventually be associated with rib and vertebral bone structures. Due to the very great attenuation of low energy photons by bone, it is evident that large errors will arise

if a significant portion of the thorax burden is deposited in or on bone and not considered during a routine *in vivo* "lung count". Furthermore, it can hardly be assumed from existing data that the lung is the one and only critical organ for the radiobiological effects of the long-lived alpha-emitting nuclides (10).

The skull represents a body structure of considerable bone mass [about 11-13% of the fresh skeletal weight (Table 5)], comprising 22 individual bones with relatively little overlying soft tissue cover, consisting primarily of scalp, hair, muscle and skin. The large bone mass and minimal overlying soft tissue make the head an ideal site for the *in vivo* measurement of "bone-seeking" radionuclides emitting photons of relatively low energy. Furthermore, there are several additional factors which contribute to the applicability and importance of head-site scintillation measurements, viz.:

- 1) For *in vivo* scintillation measurements, it is possible, by judicious use of lead and steel materials, to shield the head (skull) and adjacent detectors from emissions originating from other areas of the body.
- 2) There is no organ or soft tissue, associated with or in close proximity to the head (skull), that has been reported to significantly concentrate bone-seeking elements as might, for example, organs

such as liver, kidney, spleen and/or lung.*

- 3) Of several possible body measurement sites, the head (skull) is one of the least variable anatomical volumes (with respect to relative size and shape) among subjects of different heights and weights.
- 4) The percentage of the total skeleton represented by the skull has been well defined and represents the most concentrated area of available bone in the body (Table 3).
- 5) It is possible to comfortably surround the head structure almost entirely with detector units thereby optimizing geometry for *in vivo* assay and increasing both the accuracy and precision for any individual measurement (Figure 1).
- 6) The head is relatively easy to approximate with a surrogate structure or "phantoms" constructed of tissue equivalent materials; furthermore, actual human skulls are commercially available for purchase.

*Although the thyroid gland of the beagle has been noted to concentrate ^{241}Am to some extent (11), this same effect has not been noted to occur in other animal species. Furthermore, this concentration would not be significant with respect to the content of material associated with the skull.

- 7) In the experimental laboratory animal of our choice, i.e., the baboon, it has been a routine procedure to obtain biopsy samples from the frontal bones of the calvarium (by trefine techniques), which has enabled us to define the movement of the radionuclide ^{241}Am in the skull bone as a function of time (12,13).
- 8) Metabolic parameters of bone remodeling and resorption have been defined for the skull bones for comparison to other bone structures in the body (14).

Application

Several important nuclides which may be (or which have already been) measured in the skull bones with three dual-thin crystal detectors are:

- 1) ^{210}Pb , a bone, volume-seeking, calcium congener (alkaline, earth-like material), present in greater than normal levels in individuals who have been exposed to abnormally high concentrations of radon and radon daughters, i.e., uranium miners (15) and inhabitants of "uranium tailing homes" in Grand Junction, Colorado (16), as well as in individuals and experimental animals directly exposed to higher than average levels of ^{210}Pb , i.e., heavy cigarette smokers (17) and

adult baboons administered intravenous doses of ^{210}Pb (18).

- 2) ^{241}Am , a bone surface-seeking actinide (transuranic nuclide) whose deposition in bone approximates that of other transuranic nuclides by initially concentrating along periosteal and endosteal sites. *In vivo* measurements of this nuclide have been performed in workers engaged in nuclear fuel reprocessing procedures and in two individuals who became accidentally contaminated in their home during experiments with ^{241}Am for instrument development purposes (19,20).
- 3) $^{238}, ^{239}\text{Pu}$, bone surface-seeking transuranic nuclides whose deposition in bone is characteristic of concentration processes similar to those of thorium and americium. This nuclide has been measured by *in vivo* techniques in individuals industrially exposed during nuclear reprocessing, research, and reactor waste-handling operations.
- 4) ^{90}Sr , a bone volume-seeking alkaline-earth nuclide heavily used in research and industry and present in nuclear weapons fallout as well as in reactor waste products. In research concerned with the detection of this nuclide by measurement of bremsstrahlung radiation in the skull, it has been shown that this method is a feasible radiological protection monitoring procedure (21).

- 5) In addition to the radionuclides listed above, it is reasonable to consider the possibility of *in vivo* measurements of other important nuclides associated with bone tissue such as the gamma or X-ray emitting radioisotopes of uranium, radium, curium and thorium. Body burdens of individuals occupationally exposed to curium have already been reported (22) and measurements of depleted and enriched uranium burdens have been made at this laboratory (Section VI).

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TABLE 1

Some Important Radionuclides Detectable in Man by In Vivo,
Dual-Crystal Measurement Techniques

Nuclide	$T_{1/2}$ Radiol.	Photon Energy and Abundance	Average Major Energy Deposition in Man	Biological Characteristics (Major Sites of Deposition)	Importance
^{241}Am	433 y	60 keV, 35% 26 keV, 2.5% 18 keV Np L X ray	5.31 MeV α	Surface "Bone Seeker" Liver Lung (inhalation)	Nuclear waste; consumer products; commercial application
^{238}Pu	86.4 y	16 keV U L X ray, 11.5%	5.49 MeV α	Skeleton	Nuclear fuel cycle; waste
^{239}Pu	24,390 y	, 4.7%	5.10 MeV α	Liver	
^{240}Pu	6,580 y	, 10.8%	5.16 MeV α	Lung (inhalation)	
^{238}U	4.51×10^9 yr	15.5 keV Th L X rays	4.19 MeV α	Chemical toxicity Kidney Lung (inhalation)	Mining; milling; fabrication
↓ ^{234}Th	24 days	15.9 keV Pa L X rays	0.191 MeV β^-		
↓ $^{234\text{m}}\text{Pa}$	1.2 min.	16.4 keV U L X rays	2.29 MeV β^-		
^{235}U	7.1×10^8 yr	143 keV, 11% 185 keV, 54% 204 keV, 5%	4.41 MeV α	Chemical toxicity Kidney	Nuclear fuel cycle
^{244}Cm	17.6 yr	17 keV Pu L X rays	5.80 MeV α	Surface "Bone Seeker" Liver	Nuclear fuel cycle
^{210}Pb	22.2 yr	12.6 keV Bi L X rays 47 keV, 4%	0.061 MeV β^-	Skeleton Lungs	Uranium mining; milling
^{125}I	60.14 days	27-31 keV, $K_{\alpha,\beta}$ X ray, 100% 35.5 keV, γ , 6.7% ~55 keV, sum peak	0.022 MeV β^-	Thyroid	Nuclear medicine

TABLE 2

Some Major Research and Surveillance Laboratories

Employing Dual Crystal [NaI(Tl)-CsI(Tl)] In Vivo Detection Systems

United States Laboratories

Argonne National Laboratory, Argonne, Illinois

Battelle Pacific Northwest Laboratories (BNWL), Richland, Washington

ITRI (Lovelace Foundation), Albuquerque, New Mexico

Los Alamos Scientific Laboratory, Albuquerque, New Mexico

Mound Laboratories, Miamisburg, Ohio

New York University Medical Center, New York, New York

Rocky Flats, Division of Dow Chemical Company, Golden, Colorado

University of California, Lawrence Livermore Laboratories, Livermore,
California

University of Pittsburgh, Pittsburgh, Pennsylvania

Foreign Laboratories

Atomic Energy Establishment, Winfrith, England

Atomic Energy Research Establishment, Harwell, England

Centre d'Études Nucleaires, Fontenay-aux-Roses, France

Environmental Radioactivity Laboratory CNEN-CSN, Rome, Italy

Radiation Protection Division, Department of National Health and
Welfare, Ottawa, Canada

TABLE 3

Gross Distribution of ^{238}Pu in the Beagle
after Inhalation of $^{238}\text{PuO}_2$ (calcined
at 350°C). From Park, et al. 1976*

Time after Exposure (years)	% of Total Body Burden		Cause of Death
	Lungs	Skeleton	
1.9	4	64	Bone Fracture
3.0	32	32	R.I.
3.2	15	57	R.I.
4.5	34	41	Osteosarcoma
4.8	6	55	Osteosarcoma
5.0	7	43	Osteosarcoma
5.2	17	47	Osteosarcoma

*Park, J.F., A.C. Case, D.L. Catt, P.L. Hackett, J.E. Lung, G.J. Powers, H.A. Ragan and C.R. Watson. Late Effects of Inhaled $^{238}\text{PuO}_2$ in Beagles, Pacific Northwest Laboratory Annual Report for 1975, BNWL-2000, Part 1, UC-48 Biology and Medicine, p. 18 (1976).

TABLE 4

Gross Distribution of ^{241}Am and ^{244}Cm in the
Beagle After Inhalation of $^{241}\text{AmO}_2$ and $^{244}\text{CmO}_2$.

From Craig, et al. 1976*

		(As % of Final Body Burden)			
		Days Post Exposure			
		<u>10</u>	<u>30</u>	<u>90</u>	<u>270</u>
<u>Am-241</u>					
Lung		78.5	55.4	29.4	13.8
Skeleton		5.1	10.9	18.2	45.1
<u>Cm-244</u>					
Lung		32.9	19.9	18.0	8.1
Skeleton		18.4	19.8	29.7	26.6

*Craig, K.K., W.C. Cannon, D.L. Catt, J.P. Herring, J.F. Park, G.J. Powers, and C.R. Watson. Distribution of ^{241}Am and ^{244}Cm in Dogs After Inhalation of the Oxides, Pacific Northwest Laboratory Annual Report for 1975, BNWL-2000, Part 1, UC-48 Biology and Medicine, p. 23 (1976).

TABLE 5

Relative Weights of Individual Bones of the Skeleton
(% of Whole Skeletal Weight).

Bone Type	Dry Fat Free (1)	Dry Fat Free (2)	Dry Wt. (3)	Dry Wt. (4)	Fresh Wt. (5)	Ash Wt. (6)	Fresh Wet Wt. (7)	Ash Wt. (7)	(Ash Wt./Fresh Wt.) (7)
1. Skull	17.1	18.9	18.4	13.1	11.5	15.3	13.0±1.64	18.7	31.4
2. Femur	32.0	31.0	8.9	9.2	--	--	15.3±1.46	18.0	24.0
3. Tibia			(1) 5.08	(1) 5.39	--	--	11.3±1.36	12.3	23.3
4. Fibula			(1) 1.25	(1) 1.16	--	--			
5. Humerus	11.9	10.1	(1) 3.33	(1) 3.62	--	--	(2) 5.3±0.49	(2) 6.1	24.4
6. Ulna			(1) 1.47	(1) 1.35	--	--	3.6±0.55	4.7	28.5
7. Radius			(1) 1.21	(1) 1.09	--	--			
8. Vertebrae	10.5	13.2	--	9.36	--	10.0	19.0±1.59	10.4	12.1
9. Ribs	6.3	6.3	(24) 4.4	6.6	--	5.7-6.5	7.0±0.42	7.8	24.2
10. Clavicle	--	--	(1) 0.50	0.54	--	--	(2) 0.8±0.13	(2) 0.8	23.1
11. Scapula	--	--	(1) 1.51	1.56	--	--	(2) 3.6±0.23	3.6	21.0
12. All other bones (i.e. pelvis, feet, hands, etc.)	22.2	20.6	--	--	--	--	--	--	--

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Numbers in parentheses within the table are the number of bones represented by the given value.

FIGURE 1

Subject-Detector Standard N.Y.U. Head Counting Geometry

I-20

SECTION II

Design and Optimization Characteristics of In Vivo Detection System

A method has been developed, employing dual crystal scintillation detectors and real time computer analysis, to improve the detection limits (MSMA) for low energy photons (less than 150 keV) in the presence of other higher energy photon emitters in the body. Computer modification of the low energy gamma spectrum derived from an *in vivo* measurement can improve the detection limits for isotopes of plutonium, americium, or curium, for example, assuming knowledge of the K-40 Compton scatter contribution to the low energy measurement.

Simultaneous *in vivo* measurement of low and high energy photon emitting radionuclides in man is accomplished in this laboratory by use of the dual crystal (NaI(Tl)/CsI(Tl)) scintillation detector. The dual crystal detector was originally designed to take advantage of the inherent difference in the natural phosphor decay of NaI(Tl) and CsI(Tl) and uses this feature to identify interactions which occur in either the thin (3 mm) NaI(Tl) crystal or the thicker (5 cm) CsI(Tl) crystal (23,24). Commercially available detector systems, which include pulse shaping electronics, are routinely used to perform *in vivo* measurements for internally-deposited, low-energy emitting radionuclides. For example, the reduction in detector background

provided by the electronic pulse shape discrimination employed with the present dual crystal detector is approximately 99% in the 50-67 keV energy region (Am-241) or the 14-25 keV energy region (Pu-238).

Measurement Technique

Another mode of operation for the dual crystal detector has now been developed which provides for a simultaneous measurement of high and low energy photons in a single counting period and in the same geometry. In particular, K-40 (1.46 MeV) can be measured at the same time as the low-energy emitting radionuclides. Without this new measurement capability it would be necessary to perform each analysis individually with different detectors, in order to measure both high and low energy events. Repositioning detectors add additional uncertainty to an *in vivo* counting procedure since errors associated with small changes in the subject-to-detector geometry are much greater in magnitude than statistical counting errors. Furthermore, the apportionment of counting time is most efficiently utilized by measuring high and low energy emitters simultaneously and provides the greatest degree of comfort to the subject being measured.

Since, as this study indicates, the Compton scatter events from K-40 are responsible for a significant portion of the counts in the low energy background spectrum (Table 1), it is possible to reduce background below 150 keV by

simultaneously measuring the K-40 and calculating, whether from a mathematical model or a direct proportionality, its contribution to the spectrum below 150 keV. In fact, the Compton scatter contribution from K-40 to the low energy spectrum can be determined in one measurement by using the CsI(Tl) crystal to detect the 1.46 MeV photon from K-40 while the low energy events are measured with the NaI(Tl) crystal. Furthermore, using the same hypothesis, this low energy background reduction technique can be applied when other high energy photon emitting radionuclides (e.g., Cs-137, C-60, etc.) are present.

Dual Crystal Scintillation Detectors

The dual crystal detectors used in our multi-detector *in vivo* measurements have been especially fabricated to provide the optimum efficiency with the lowest background. To insure that the energy response of each dual-crystal detector used in a multi-detector counting array is the same requires that the NaI(Tl) and CsI(Tl) in each detector be manufactured from the same ingot, respectively (i.e., each cut from a parent crystal). The energy response (i.e., light output) of each crystal was tested with a point source of radioactivity by the manufacturer* to verify its characteristics and insure that an array of

*Harshaw Chemical Company, Solon, Ohio

these crystals, with output signals connected in parallel, would have a matched response during a whole body counting procedure.

In the past, it was found that various dual crystal detectors varied in efficiency and resolution from unit to unit. When these detectors were subsequently employed in an array, the inherent difference in their response would introduce a nonuniform (i.e., "nonisotropic") counting geometry. Although the low energy response could be electronically balanced among detectors in the array, the response of the CsI(Tl) crystals could not be made isotropic without employing an extensive series of amplifiers. Specifying that the new detectors employ NaI(Tl) and CsI(Tl) crystals specially selected for matched output has avoided the elaborate electronic balancing required in the previous detector array and has, therefore, greatly simplified calibration procedures. There has also been a great improvement in system stability and lack of electronic gain shifting among detectors.

The major contribution to detector background in our whole body counting room (other than that of electronic origin) arises from cosmic radiation secondary and tertiary interactions with the graded-Z walls of the counting room and, to a lesser extent, with materials in the detector itself. (For this discussion the contribution from Rn-222 and its short-lived daughters have been neglected.)

Potassium-40 is a naturally radioactive material found in the glass envelope of the photomultiplier tube. Its presence adds to the total K-40 activity detected in a person during an *in vivo* measurement.

The other major radioactive contaminant of considerable importance is Cs-137, which we have found present in all the dual crystal NaI(Tl)/CsI(Tl) detectors. Its presence at 0.662 MeV is quite prominent in all spectra of room background obtained with the CsI(Tl) crystal (Figure 1). In addition, a 32 keV $K_{\alpha+\beta}$ barium X-ray peak appears in the low energy spectrum measured with the thin NaI(Tl) crystal. Table 2 gives the decay scheme for Cs-137 and Figure 2, for reference, shows the low energy and high energy measurement of a Cs-137 point source.

Experiments were performed to quantitate the amount of Cs-137 contamination in the CsI(Tl) crystal of each detector. Our detectors employ CsI(Tl) crystals 15.24 cm in diameter and 5.08 cm thick so that each detector contains approximately 4200 g of CsI (MW = 259.83 g = 4.51 g/cm³). It can then be calculated using the 0.662 MeV photopeak from Cs-137 that the total activity in each detector is approximately 1 pCi. The origin of this contamination remains unknown; but our discussions with the manufacturer (Harshaw Chemical Company) indicate that even though the cesium ore is obtained from a deep underground mine, the Cs-137 is suspected of originating from weapons testing fallout

[although its "natural" formation as a result of the proximity of Be to U in the cesium pollucite ore* is not completely ruled out (25,26,26a)].

The Cs-137 contamination in the detectors is counted in what is essentially a highly efficient 4π -type geometry since the activity is probably incorporated into the CsI crystal lattice. The barium 32 keV $K_{\alpha+\beta}$ X ray detected in the NaI(Tl) crystal originates from Cs-137 decay and is in close proximity to the NaI-CsI(Tl) crystal interface. The relative emission abundancies, attenuation coefficients and loci of Cs-137 deposition, in proximity to either the CsI(Tl) or the NaI(Tl), all account for the magnitudes of the count rates, obtained at 32 keV in the NaI(Tl) compared to 0.662 MeV in the CsI(Tl).

The 15.24 cm diameter dual crystal scintillation detectors employ a 3 mm NaI(Tl) crystal coupled to a 5.08 cm thick CsI(Tl) crystal. A 0.254 mm thick beryllium entrance window is epoxied to a stainless steel detector housing with no overlapping material (i.e., lip) to reduce the effective surface area. The epoxy is a white material with black dye added to prevent light leaks. The reflector material is HR 15-P which Harshaw Chemical Company has

*We have now obtained samples of high purity CsI and pollucite ($H_2O_2 \cdot 2Cs_2O \cdot 2Al_2O_3 \cdot 9SiO_2$) (which is 40.1% cesium and is the ore from which the cesium is refined) to determine possible Cs-137 contamination. This material was obtained from the Rare Metals Division of Kawecky Berylco Industries.

determined to have the lowest background and the maximum reflectance. A 12.7 cm, low-noise photomultiplier tube with a low-background, low-profile base is optically coupled to the crystals. Controls for high voltage and focus on the photomultiplier tube base are eliminated. Metal-foil, low-noise resistors are used in the voltage divider circuit for the photomultiplier tube.

In order to reduce the K-40 contribution to the overall background of the detector from the potassium in the glass envelope surrounding the PM tube, it was suggested that a pure NaI light pipe be inserted between the PM tube and the NaI(Tl) and CsI(Tl) crystals. Theoretically, the pure NaI light pipe would absorb the K-40 photons and not scintillate with a wavelength detectable by the photocathode on the PM tube. However, the natural fluorescence of pure NaI at room temperature detectable by the photocathode is not totally negligible and potentially could increase the background of the detector. The manufacturer could select the batch of pure NaI with the least fluorescence yield but could not guarantee that the detector background would be unaffected. It was therefore omitted from the design of the new detectors. Although it was not possible to find Cs-137 free CsI(Tl), the batch with the least amount of contamination was selected for the new detectors currently employed for the whole body *in vivo* counting.

Electronic Pulse Shape Discrimination System

The electronic pulse shape discrimination system, which operates in conjunction with the dual crystal detector, is diagrammatically shown in Figure 3. We employ two unique counting geometries which require either three detectors to surround the head or two detectors placed in close proximity to the lungs on the anterior thorax of the subject. Each scintillation detector has an individual preamplifier which shapes the signal from the detector and drives a 50-foot cable to the input of the sum amplifier. This configuration maintains the energy calibration for the system by essentially isolating the effective change in the input impedance to the amplifier when the number of detectors in a particular counting geometry is changed. Therefore, the amplifier gain is independent of the number of detectors used in an *in vivo* counting procedure.

Since there are no gain (high voltage) or focus controls on the detectors it is necessary to closely measure the high voltage applied to the divider circuit on each photomultiplier tube. Separate digital meters monitor the high voltage applied to each photomultiplier tube. The individual response of the three detectors are nearly identical, having been expressly fabricated for this application, so that the signal from the sum amplifier is directly driven into two spectroscopic amplifiers, one for NaI(Tl) events and the other for CsI(Tl) events.

Photon interactions which occur in the CsI(Tl) or NaI(Tl) crystal are identified by a pulse shape analyzer which produces a logic pulse whenever a preset pulse shape timing criteria, established for either crystal, has been met by the incoming pulse. That respective logic pulse will open an electronic linear gate to permit the delayed (approximately 4 μ sec) signal to pass into the multiplex input of a multichannel analyzer. Therefore, from a single dual-crystal detector two characteristic spectra are obtained; one ranging up to 150 keV, from the NaI(Tl) crystal, and the other ranging up to 1.7 MeV, from the CsI(Tl) crystal.

Computer Assisted Analysis

Our computer based, multichannel analyzer is completely interactive with a 16K bit NOVA computer. In fact all the mathematical and display functions evolve as a result of pre-programmed instructions which are entirely accessible as part of any user created programs. By virtue of the interactive computer-based operation, programs have been written to perform various routine mathematical operations such as determination of resolution, efficiency, and figure of merit (S^2/B).

A library of 25-50 unexposed control subjects* is now being assembled of individuals who are routinely being whole body counted over the head (skull) and chest (anterior thorax) positions. From these data it will be

*These volunteer control subjects were from local clubs and fire departments and, for the most part, represent male subjects between the ages of 17 and 60 years.

possible to determine specific relationships between background, body size (as determined by several anthropometric measurements), age, and the K-40 content of the subject. By employing cassette tape to extend the storage capacity of the computer, it will be possible to generate a background control spectrum for an *in vivo* measurement depending upon the exposed subject's anthropometric measurements and the amount of K-40 measured in the same geometry analyzed for the radioactive contaminant.

The computer will first be able to search for a control subject by finding a suitable individual similar in age, height, weight, gender, and K-40 content to the exposed subject. Once the control subject library is enlarged by completing an increased number of whole body counts of unexposed subjects, it will be possible to mathematically construct the contribution that K-40 has to the low energy spectrum in the exposed individual from a knowledge of the K-40 measurement and several anthropometric details to indicate body size. By removing the low energy, Compton-scattered component of the K-40 activity in the body from the low energy spectrum (i.e., live time spectrum stripping using the computer while data is being accumulated), the background signal is reduced and the exposed subject is effectively used as his own control.

Interactive computer programming provides the functional capability to perform the background reduction while data is

being accumulated during a whole body counting procedure. In this manner, when a subject is being measured as a result of a known accidental exposure, his own K-40 background contribution can be removed from the measured spectrum and qualitative results from this preliminary background subtraction can be used to estimate an amount of low-energy emitting radioactivity present in the subject.

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TABLE 1

Estimated Stable Potassium and ^{40}K
Content of the Head and Total Body*

Specific activity = 853 pCi ^{40}K /g stable potassium

<u>Head Structure</u>	<u>Weight (g)</u>	<u>Stable K (g)</u>	<u>^{40}K (nCi)</u>	<u>Approx. % of Head Activity</u>
1. Skull with teeth	1200-1500	1.8	1.54	19.5
2. Teeth	46	--	--	--
3. Brain	1400	4.2	3.58	45.4
4. Face and neck muscles	803	2.4	2.05	26.0
5. Blood in Head	165	0.26	0.22	2.8
6. Hair	20	0.001	0.00085	0.01
7. Skin and scalp	162	0.38	0.32	4.1
8. Tongue	70	0.20	0.17	2.2
9. Eyes	15	--	--	--
10. Fat	--	--	--	--
11. Cervical vertebrae	--	--	--	--
Total head	3.9-4.2 kg	~9.25	~7.88	
Total body	70 kg	~140	~120	

*Determined from Reference Man data and personal communication, Dr. G. P. Howells (1972).

TABLE 2

CESIUM-137

Barium $K_{\alpha, \beta}$ X-rays $\bar{E} = 32.89 \text{ keV}$

FIGURE 1

WBC Room Background
3-NaI(Tl) CsI(Tl) Detectors
1000 Min.

FIGURE 2

Cs-137 Source $1.0\mu\text{Ci}$
NaI(Tl) CsI(Tl) Detectors

FIGURE 3. Schematic for modular electronics associated with three NaI(Tl)-CsI(Tl) detectors used for head measurement.

SECTION III

Development of a Calibration "Phantom" Structure for Determining Activity Deposited in the Thoracic Skeleton

Although there are many varieties of commercially available surrogate structures for the human torso (27-31), many of which incorporate actual human skeletons and removable tissue equivalent organs, only recently has consideration been given to the development of a calibration phantom for the specific application to *in vivo* measurements of low energy, photon-emitting nuclides (32-34). Consideration of the components to be used in a structure designed for measurement of these nuclides would incorporate the proper tissue equivalent materials chosen for their appropriate characteristics of low energy photon attenuation and absorption. The one phantom structure that has not yet been developed, however, has been for radioactive material deposited on the surface or within the skeletal structure.

It is now well established that alkaline earth materials act as chemical congeners of calcium, thereby replacing the latter and depositing within the appetite structure of the bone. In contrast, the actinide elements will deposit along the surfaces of the skeletal structures, probably in combination with the organic matrix (e.g., mucopolysaccharides, chondroitin sulphates, collagen and elastin),

eventually becoming more diffuse and burying within the bone due to normal bone resorption mechanisms. Therefore, the alkaline earths and the actinides can both be classified as "bone seekers" requiring the development of proper calibration structures for the *in vivo* measurement of material so deposited.

As previously discussed, this has already been accomplished, to an extent, for material deposited on the skull bones in a procedure whereby two matched skulls are painted with activity (i.e., Am-241), one on the inner and the other on the outer surfaces, so that by counting both structures it is possible to mathematically approximate, by using the geometric mean, the situation for a central line source or one which is uniformly distributed throughout the skull structures (35). Thus far, skull bone structures for the nuclides Am-241, Cm-243 and Pb-210 have been fabricated. Figures 1 and 2 are examples of the photon spectra obtained for the Am-241 phantom heads.

Development of a thoracic skeletal phantom is particularly difficult due to the convoluted structures of the rib cage and the many attendant differences among bone shape and types associated with the ribs, the scapulae, the sternum, the vertebrae and the cartilagenous material that comprises the outer regions of the ribs to the site of sternal attachment. It is extremely difficult, therefore, to construct such a phantom structure from a single material.

Furthermore, the assumption of a uniform distribution of the nuclide deposited therein may be grossly in error. In the present case, a skeletal phantom was achieved by fabrication procedures which are described in the following protocol:

1) Skeletal Considerations

The human skeleton was of an average-sized Caucasian male (estimated weight = 150-155 lbs.) of Spanish origin, approximately 30 years old at death. An additional 40 years elapsed before the skeleton was received in our laboratory. The skeleton, as a whole (without plastic replacements for cartilaginous structures), was in excellent condition with no visible defects. For our purpose, only the axial skeleton was utilized; limb bones were entirely removed. Thus, the boney portion of the phantom consisted of: skull (removable), complete vertebral column, complete rib cage including original cartilage and sternum, scapula, clavicle and pelvic bones. Since the upper trunk (i.e., neck to waist) was of interest, radioactivity was added only to those bones that would make a significant contribution (vis-à-vis our lung burden detection techniques) to a "chest" measurement (Figure 3). Activity was therefore applied only to the rib cage, scapulas, clavicles and the cervical, thoracic and lumbar portions of the vertebral column. The dimensions of this structure are given in Table 1.

2) Addition of Am-241

The radionuclide chosen for this initial phantom structure was Am-241 due to its importance as a nuclear reactor waste material and in various commercial applications (36). In addition, Am-241 emits Np L X rays having energies of approximately 17 keV (similar in energy to the L X rays emitted by plutonium, protactinium and uranium) and 59.5 keV gamma photons with an abundance of approximately 35%, thereby allowing for excellent detection efficiency with relatively small amounts of incorporated nuclide. As in the case of the skull bones, the skeletal structure was considered as having two surfaces and was accordingly treated with the activity by placing the material first on the inner structures (with a blue dye to define the surface over which the material had been placed), followed by coating the outer structures with a similar amount of material (incorporating a red dye).^{*} Again, by use of the dye material, it is possible to determine exactly which areas have been covered and to determine whether or not the particular surface has excessive dye

^{*}The difference between this thoracic skeleton phantom and the skull phantom is in the placement of the activity. In the present case the nuclide was actually placed in the marrow cavity to approximate endosteal surface deposition, whereas in the skull this was accomplished mathematically using two phantom structures.

concentrated in any specific area; by use of the blue dye on the inside structures and the red dye on the outside structures, it was also possible to determine areas of overlap, i.e., purple coloration between the two applications.

Approximately 0.5 μ Ci Am-241 was deposited on the inner structures (using a 25 x 5/8" needle and syringe) after drilling ~300 holes (diam. - 1 mm) uniformly distributed over all rib, scapulae and sternum structures, and within all cortical to lumbar vertebrae. By judicious use of the proper volume in which the activity was measured, it was possible to insure that the solution had indeed permeated and spread uniformly from one hole application to another. Each of these holes was then plugged using ordinary plaster of paris, after which it was possible to brush an identical total amount of activity on the outer surfaces of the skeleton. All excess material in the application procedures (e.g., activity remaining on the brushes, etc.) were determined by counting all tools and waste materials. The loss by this route amounted to 13% of the initial activity, resulting in a final deposition of 975 nCi on the thorax skeleton.

The distribution of activity among the various bones of the thoracic skeleton was modeled after that measured in several baboons who had been injected I.V. with Am-241 citrate 32 days prior to sacrifice and measurement of skeletal activity distribution (37). The baboon distribution

data in terms of percent of injected dosage is given in Table 2. Although the anatomical analogy between baboon and man has several specific differences with respect to skeletal structure, this is the only primate data for which detailed americium distribution values among bones are available. The baboon distribution values were consequently normalized to the scapulae content and equivalent amounts applied to the human skeleton. In the vertebral column, the baboon had an americium concentration gradient increasing by an order of magnitude when going from the cervical to the lumbar regions. This gradient was reproduced in the human thorax phantom by normalizing the baboon concentration values to the first three cervical vertebrae (Group 1). Table 3 lists the vertebrae groupings and normalized concentration factors with respect to Group 1. Use of these concentration factors enabled the same difference in magnitude for Am-241 concentration along the vertebral column of the human thorax phantom.

3) Chest Phantom Construction

After application of the Am-241, the entire structure was coated with a thin layer of polyethylene (by spray) to both physically fix the activity on the surface and to approximate the organic matrix which adheres to the surface of all bone structures. After application of the plastic spray, lungs with the composition determined for the Rando phantom (27) and of the approximate density, 0.25 g/cc,

were inserted into the skeletal structure. The heart and liver were fabricated from plastic bags filled with rice according to dimensions given in ICRP-23 Reference Man (38). The surrogate organs were secured in their proper anatomical positions with nylon fishing line. Finally, the muscle and skin were simulated by wrapping the entire structure with dental wax (density = 0.85) applied in layers of 0.12 cm thickness. The average thickness of the chest wall was determined by following the phantom design specifications of Dean, et al. (39), with an average chest wall thickness of 2.5 cm. Photographs and photon spectra for this phantom in various stages of fabrication are given as Figures 4, 5 and 6. The calibration for material so deposited on the surfaces of the thoracic skeleton for the various regions of interest are 20.87 cpm/nCi from 50-67 keV, i.e., the Am-241 60 keV region, and 4.19 cpm/nCi from 14-25 keV, i.e., the 17 keV X-ray region. Similar phantom structures are now being constructed for other size and shape skeletons so that it will ultimately be possible to derive the appropriate contribution factors regardless of the subject size.

From these values and the assumed relationship of uniform concentration in all bones of the skeleton, it is possible to derive the contribution from material deposited in the thoracic skeleton to that total measurement made over

the chest area. A mathematical formulation which expresses this relationship is given in Eq. 1 as:

$$M = \frac{AEF-DGJ}{EFL} \quad (\text{Eq. 1})$$

where:

M = activity deposited in the lungs, nCi

E } phantom constants { E=head (skull), cpm/nCi
J } { J=thoracic skeleton, cpm/nCi
L } { L=lung, cpm/nCi

A } detector measurements { A=total thorax, cpm
D } { D=head, cpm

F } anthropometric measurements { F=skull mass, g
G } { G=thoracic skeleton mass, g

and:

1. Subject head measurement = D cpm
2. Phantom head measurement = E cpm/nCi
3. Subject head activity = D/E nCi
4. Subject skull mass = F grams
5. Subject skull concentration = D/EF nCi/gram
6. Subject thoracic skeletal mass = G grams
7. Subject thoracic skeletal concentration = H/G = D/EF
8. Subject thoracic skeletal activity = H = DG/EF nCi
9. Phantom thoracic skeleton = J cpm/nCi
10. Subject thoracic skeletal contribution = HJ = B cpm
11. Subject total chest measurement = lung + thoracic skeletal contributions = A cpm = B cpm + C cpm
12. Subject lung contribution = A-B = C cpm
13. Phantom lung measurement = L cpm/nCi
14. Subject lung activity = C/L = M nCi.

Major assumptions:

- 1) Activity is uniformly deposited in the lungs
- 2) Concentration of activity is the same in all bones
- 3) Distribution of activity is the same in man as in the adult baboon one month after I.V. injection.

(Note that the thoracic bone contribution to the total thorax measurement is $\frac{DGJ}{EF}$).

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TABLE 1

Anthropometric Measurements of
Thoracic Skeletal Phantom

[Skull, Clavicles, Sternum, Scapulas, Ribs,
Vertebrae-25, Innominate Structure]

1. Date of Birth: ~1908
2. Sex: Male
3. Total Weight: 3.64 kg
4. Circumference: 74 cm (at 4th rib)
5. Diameter A/P: 19.5 cm
6. Diameter (left/right): 22 cm
7. Shoulder to shoulder: 39.5 cm

TABLE 2

Distribution of ^{241}Am in the Thoracic Skeleton of
an Adult Baboon Approximately One Month After IV Exposure¹

<u>Bone Type</u>	<u>Percent Injected Dose</u>	<u>Distribution Normalized to Scapula</u>
Clavicles (2)	0.19	0.10
Sternum (1)	0.52	0.30
Scapulas (2)	1.82	1.00
Ribs (24)	2.57	1.40
Vertebrae ² (25) ³	11.14	6.10

¹ Data from Guilmette, Cohen and Wrenn, 1975 (10).

² Includes cervical, thoracic and lumbar vertebrae only.

³ The baboon has one more lumbar vertebra than man.

TABLE 3
Distribution of ^{241}Am in the Vertebrae of an
Adult Baboon Approximately One Month After IV Exposure²

<u>Group</u>	<u>Vertebrae¹</u>	<u>Concentration Factor (Normalized to Group 1)</u>
1	Ce1-Ce3	1
2	Ce4-T7	3
3	T8-T12	7
4	L1-15	14

¹ Ce = Cervical
T = Thoracic
L = Lumbar

² Includes cervical, thoracic and lumbar vertebrae only.

FIGURE 1

FIGURE 2

FIGURE 3. Detector configuration and anatomical structures for anterior thorax measurement.

FIGURE 4. Am-241 labeled thoracic skeleton phantom - photon spectrum w.
NaI(Tl)-CsI(Tl) detectors in chest counting position - 5 min.

III-17

FIGURE 5. Thoracic skeleton prepared for Am-241 labeling on inner surfaces of all bones.

FIGURE 6. Thoracic skeleton labeled with Am-241 with surrogate lung and liver structures included.

SECTION IV

Computer Assisted In Vivo Measurements of Internally Deposited Radionuclides Using Dual-Crystal Scintillation Detectors

A channel specific modification of the low energy (i.e., less than 150 keV) photon spectrum from an *in vivo* measurement of internally deposited radionuclides is performed to remove the influence of naturally occurring K-40 in the body. Since K-40 makes a significant contribution to the background of a low energy spectrum as a result of Compton interactions by the 1.46 MeV photons, its influence might be obviated if the amount of K-40 in the exact counting geometry were known.* Measuring K-40 in the subject and calculating its contribution to the low energy spectrum of the same person might possibly reduce the need for a control subject, assuming that Compton scattered K-40 events and electronic "noise" comprise the majority of the background signal.

There is a need to develop a procedure for analysis of an *in vivo* measurement which is less sensitive to selection of a control subject than is currently employed. Even though "controls" (i.e., persons with no known occupational exposure to radioactivity) are selected on the basis of anthropometric parameters, the normal variability measured

*A significant contribution to the overall count rate from surface K-40 β^- radiation ($E_{\max} = 1.31$ MeV) may also be possible and is currently under investigation in this laboratory.

in our control population can exceed 15% depending upon the energy region under investigation.

Current practice is to select unexposed individuals who are of similar age, height, weight and sex as the exposed subject. For most cases a single control may be used, especially when the magnitude of activity being measured is sufficient to produce a prominent photopeak in the spectrum. However, if the presence of activity is uncertain and/or the identity of the nuclide is unknown, then the analysis depends solely upon the detection limits of the counting system. It is in the latter circumstance that the background spectrum from the control subject measurement is of critical consideration.

Dual-crystal scintillation detectors are employed to measure the presence of low-energy emitting radionuclides in the body. Usually a site-specific array (geometry or configuration) of detectors is positioned around the subject in close proximity to the organ or tissue (suspected of containing the contaminant) in order to obtain the maximum sensitivity. Although pulse shape discrimination used in conjunction with the dual crystal detector effectively reduces the detector background below 150 keV, the contribution to the low energy spectrum from Compton scattered photons (i.e., K-40: 1.46 MeV) cannot be removed electronically. Thus, K-40 in the site specific counting geometry will increase the detector background below 150 keV by

virtue of the Compton scattering process [as the K-40 photons exit the body and enter the detector(s)].

A method has been developed to take advantage of the CsI(Tl) crystal in the dual-crystal detector and use it to directly measure K-40 while the thin NaI(Tl) crystal (and pulse shape discrimination) measures only the low energy photons (40,41). The CsI(Tl) crystal measures K-40 events simultaneously as the thin NaI(Tl) crystal measures the low energy photons. In this manner, the amount of K-40 in the exact site-specific geometry can be known. Live-time, interactive computer analysis is used to determine the channel specific percentage contribution to the low energy spectrum, measured with the NaI(Tl) crystal, from the K-40 events detected with the CsI(Tl) crystal.

Our preliminary analysis assumes that the body content of K-40 is responsible for the majority of the control subject's background below 150 keV. Room background, which includes the cosmic radiation component and K-40 in the glass envelope of the photomultiplier tube, is considered in the potassium phantom which is used to determine the exact channel specific percent contribution to the low energy spectrum from K-40. Since the dual crystal detector simultaneously measures low and high energy nuclides, that amount of K-40 detected in the counting geometry can be used to determine what channel specific percentage of the low energy spectrum is due to K-40 Compton scattered

photons.

Several "phantom" structures have been designed from plastic cubitaners (cube-type containers) which are filled with distilled water and various amounts of KCl in order to approximate the concentration of K-40 that is measured in the head. The plastic container is wrapped with a plaster bandage material to simulate the attenuating properties of the skull. Figure 1 is a measurement of a typical potassium phantom and shows the 1.46 MeV photon from the K-40 decay and the low energy background contribution from a 10-min. count using 3 dual crystal detectors. For comparison, Figure 2 is the dual spectrum derived from a head count (60 minutes) of a control subject. Assuming the head (i.e., brain, skull, etc.) contains approximately 9.25 gm of stable potassium, this is equivalent to 7.88 nCi of K-40 (Table 1). The phantom contained 1.17 μ Ci of K-40 which indicated that the overall efficiency of the three detector geometry was 868.1 cpm/ μ Ci for K-40. The net counts measured in the low energy spectrum were due to K-40 Compton scattered photons. The net low energy count rate was 825.1 cpm or 705.2 cpm/ μ Ci derived from K-40 using an array of three dual-crystal detectors in the head counting geometry. These results indicate that the contribution from K-40 in the head to the low energy spectrum obtained from a "standard" control subject with 7.88 nCi K-40 would be approximately 5.56 cpm. Table 2 lists the results of the dual crystal scintillation measurement with the head phantom and a control subject.

Although the potassium phantom considers only K-40 deposited in the head, contributions from the remainder of the body cannot be neglected. There is considerable scatter into the counting geometry from 1.46 MeV photons originating at sites other than the head.

Although the head represents approximately 7% of the total body content of potassium this simple head phantom is acceptable in that it can be easily improved with the addition of cubitainers in sizes and with stable potassium concentrations to approximate the remainder of the body. The current work however attempts to account for the K-40 found only in the head. The live-time, interactive computer analysis will be able to reflect the additional contribution by simply changing the channel specific K-40 phantom calibration subroutine. All succeeding analyses will be unaffected.

Reduction of the low energy photon background spectrum is accomplished by removing a channel-specific percentage of a standardized K-40 spectrum whenever events are detected in the K-40 photopeak in the high energy spectrum. Since the multichannel analyzer is capable of interacting with the 16K bit minicomputer, this background reduction can be performed on a live-time basis so that the contribution from K-40 can be eliminated (and the resulting modified spectrum displayed) while counting is in progress. This live-time feature enables the observer to make qualitative judgments about the spectrum being accumulated and add or delete counting time accordingly.

A K-40 photoelectric interaction in the detector(s) is identified by the accumulation of counts in a region preselected to include the photopeak around 1.46 MeV. The region is chosen according to the maximum value of a figure of merit (i.e., S^2/B , where S is the signal and B is the background). Since the background signal in the CsI(Tl) detector is low, a wide region has been adopted to include essentially the entire Poisson-distributed photopeak.

During an *in vivo* measurement, computer analysis will occur at intervals preselected at the initiation of the live-time program (usually 10 seconds). The number of K-40 events accumulated during the period is determined by a computer reference to the photopeak region. A channel-specific percentage of the reference K-40 spectrum is removed from the low-energy spectrum depending upon the number of K-40 photoelectric interactions measured by the detectors during the counting interval. At completion of the analysis the spectrum displayed is upgraded to represent the modified data so that the results are immediately available to the observer.

The foundation of the analysis rests upon a reference K-40 spectrum obtained from the measurement of phantom structures. Initially, only the head is considered but no limitation is imposed by this decision since the phantom can be easily extended with the addition of other body extremities to account for the additional stable potassium.

The potassium phantom structure is counted in the same geometry as that used for a subject. Both high energy and low energy spectra are measured simultaneously so that the photoelectric interactions as well as the Compton scattered events can be determined in one counting position, the same as that used for human subjects. For an *in vivo* head count an array of three dual-crystal scintillation detectors are used with pulse-shape discrimination to provide individual signals from the thin (3 mm) NaI(Tl) crystals and the thicker (5 cm) CsI(Tl) crystals. The computer will generate (Appendix A) the channel-specific array, $F(X2)$, which is calculated from the ratio of the counts in each channel of the low energy spectrum to the total number of events in the K-40 photopeak. The dimension of $F(X2)$ depends upon the size of the low energy spectrum, usually 256 channels.

Live-time interactive analysis to reduce the low energy background during an *in vivo* measurement is accomplished by first initiating the computer with the K-40 phantom spectrum. This spectrum is stored on cassette tape. Additional input is then required to establish an interval of time ($T4$ seconds) between successive K-40 background subtractions. A background correction is performed every $T4$ seconds while the subject is being counted and will continue until the preset elapsed live time has been met. A truncated form of the corrected data is used for the display spectrum on the MCA but the actual data is

stored in memory (in array P(X5)) so that no information is lost. The main body of the program is listed in Appendix B.

The main concept of this background reduction program is a sequential spectrum stripping routine which is unique in that the magnitude of the channel specific percentage subtracted from the low energy spectrum is determined from the amount of counts accumulated simultaneously in the high energy region. At this stage no attempt has been made to perform a least square analysis with the measured spectrum nor to fit a modified gaussian function to the K-40 photopeak since these methods apply to a measurement from a single detector and would not easily adapt to the discontinuous spectrum (i.e., high energy and low energy) obtained from the dual-crystal detector. The head phantom measurement for K-40 provides an empirically derived standard spectrum which can be easily utilized in the background subtraction program rather than developing extensive mathematically derived relationships to decompose the spectrum. It is expected that the effect from more than one nuclide can eventually be incorporated into the "phantom" so that nuclides such as Cs-137 can be included in the background reduction for the low energy spectrum.

References

- (40) Shapiro, E.G. and A.L. Anderson. Dual Energy Analysis Using Phoswich Scintillation Detectors for Low-Level In-Vivo Counting, IEEE NS-21, p. 201 (1974).
- (41) Cohen, N., H.B. Spitz and M.E. Wrenn. Estimation of Skeletal Burden of "Bone-Seeking" Radionuclides in Man from In Vivo Scintillation Measurements of the Head, Health Phys. 33: 431-441 (1977).

TABLE 1

Estimated Stable Potassium and ^{40}K
Content of the Head and Total Body*

Specific activity = 853 pCi ^{40}K /g stable potassium

<u>Head Structure</u>	<u>Weight (g)</u>	<u>Stable K (g)</u>	<u>^{40}K (nCi)</u>	<u>Approx. % of Head Activity</u>
1. Skull with teeth	1200-1500	1.8	1.54	19.5
2. Teeth	46	--	--	--
3. Brain	1400	4.2	3.58	45.4
4. Face and neck muscles	803	2.4	2.05	26.0
5. Blood in Head	165	0.26	0.22	2.8
6. Hair	20	0.001	0.00085	0.01
7. Skin and scalp	162	0.38	0.32	4.1
8. Tongue	70	0.20	0.17	2.2
9. Eyes	15	--	--	--
10. Fat	--	--	--	--
11. Cervical vertebrae	--	--	--	--
Total head	3.9-4.2 kg	~9.25	~7.88	
Total body	70 kg	~140	~120	

*Determined from Reference Man data and personal communication, Dr. G. P. Howells (1972).

FIGURE 1

FIGURE 2

APPENDIX A

Statement Number

Statement

800	REM: SUBROUTINE TO REDUCE THE LOW ENERGY SPECTRUM BY THE CHANNEL
802	REM: SPECIFIC % AMOUNT OF TOTAL COUNTS IN K-40 REGION DURING
804	REM: TIME T4. ARRAY F(X5) IS CHANNEL SPECIFIC %. ARRAY P(X5)
806	REM: IS EXACT (UNTRUNCATED) DATA
810	DIM F(255), P(255)
812	FOR X5=0 TO 255
815	LET P1=D1*F(X5)
818	REM: SUBT. CHANNEL SPECIFIC % FROM LOW ENERGY SPECTRUM
820	LET P(X5)=DTA(X5)-P1
823	LET P3=INT(P(X5)+0.5)
825	CALL 3,X5,P3
827	NEXT X5
830	RETURN

APPENDIX B

Statement Number	Statement
550	REM: PROGRAM TO DETERMINE CHANNEL SPECIFIC CONTRIBUTION TO THE
551	REM: LOW ENERGY SPECTRUM AS A % OF THE TOTAL COUNT IN THE K-40
552	REM: REGION. THE K-40 SPECTRUM (LOW+HIGH) MUST BE IN QUAD B1+
553	REM: B2, RESPECTIVELY. K-40 ROI IS CHANNEL 440-497. (LINE 560)
554	DIM: A(255), F(255)
556	LET K1=0
558	REM: SUM K-40 REGION (HIGH ENERGY) INTO K1
560	FOR X1=440 TO 497
563	LET K1=K1+DTB(X1)
565	NEXT X1
567	REM: K2 IS CPM IN K-40 REGION
569	LET T1=LCN(122)
571	LET K2=K1/T1
573	PRINT K1; "COUNTS"; K2; "CPM"; T1; "SEC"
574	PRINT
575	FOR X2=0 TO 255
578	LET A(X2)=DTB(X2)
580	LET F(X2)=A(X2)/K1
582	NEXT X2
584	PRINT "DO YOU WANT ARRAY LISTED? (Y=1; N=0)"
586	INPUT Q8
588	IF Q8=0 GO TO 606
590	FOR X9=0 TO 255
592	LET X8=1200+X9
594	PRINT X8; "LET F(" ;X9;")=";F(X9)
596	NEXT X9
600	REM: LIVE TIME INTERACTIVE PGM TO MODIFY LOW ENERGY SPECTRUM
602	REM: BY A CHANNEL SPECIFIC % DEPENDING ON K-40 EVENTS IN THE
604	REM: HIGH ENERGY REGION
606	PRINT "READY TO START RUN. TYPE IN PRESET LIVE TIME (SEC)."
608	INPUT T9
610	REM: WRITE PRESET LIVE TIME INTO CORE AT LOC 106
612	CALL 1, 106, T9
614	PRINT "HOW MANY SECONDS BETWEEN EACH ANALYSIS?"
616	INPUT T4

APPENDIX B (Continued)

Statement Number	Statement
618	REM: CLEAR MEMØRY
620	CALL 4,4
622	CALL 5
624	REM: INITIATE DATA ACCUMULATION
626	CALL 4,-1
628	CALL 5
630	REM: SUM K-40 EVENTS INTO VARIABLE K2 TO SET REFERENCE BASE
632	REM: FOR SUBSEQUENT ANALYSIS
635	LET K2=0
637	FOR X3=440 TO 497
639	LET K2=K2+DTA(X3)
641	NEXT X3
643	REM: EXAMINE LT CLOCK AND WAIT T4 SEC.
645	LET T2=LCN(122)
647	PRINT T2; "SEC"
650	CALL 5
652	LET T3=LCN(122)
655	REM: TEST WHETHER COUNT HAS TERMINATED
658	IF (T3-LCN(106))=0 GO TO 709
660	IF (T3-T2)<T4 GO TO 650
662	PRINT T3; "SEC"
665	REM: PERFORM NEXT K-40 SUMMATION INTO VARIABLE K3
668	LET K3=0
670	FOR X4=440 TO 497
672	LET K3=K3+DTA(X4)
675	NEXT X4
678	PRINT K3; "COUNTS"
680	CALL 5
682	REM: SUBTRACT K-40 COUNTS IN K3 AND K2. STORE IN D1
685	LET D1=K3-K2
687	REM: IF D1=0, NO K-40 EVENTS. WAIT FOR MORE.
690	IF D1=0 GO TO 650
692	CALL 5
695	GOSUB 800
700	REM: RESET K2=K3 AS THE NEW K-40 REFERENCE BASE
702	LET K2=K3
705	LET GO TO 645
707	PRINT "DO YOU WANT EXACT (UNTRUNCATED) DATA TYPED?(Y=1; N=0)"

APPENDIX B (Continued)

<u>Statement Number</u>	<u>Statement</u>
709	INPUT Q1
711	IF Q1=0 GØ TØ 722
715	FØR Z1=0 TØ 255
717	PRINT P(Z1)
720	NEXT Z1
722	END
725	STØP
800	REM: SUBRØUTINE TØ REDUCE THE LØW ENERGY SPECTRUM BY THE CHANNEL
802	REM: SPECIFIC % AMØUNT ØF TØTAL CØUNTS IN K-40 REGIØN DURING
804	REM: TIME T4. ARRAY F(X5) IS CHANNEL SPECIFIC %. ARRAY P(X5)
806	REM: " IS EXACT (UNTRUNCATED) DATA
810	DIM : F(255), P(255)
812	FØR X5=0 TØ 255
815	LET P1=D1*F(X5)
818	REM: SUBT. CHANNEL SPECIFIC % FRØM LØW ENERGY SPECTRUM
820	LET P(X5)=DTA(X5)-P1
823	LET P3=INT(P(X5)+0.5)
825	CALL 3,X5,P3
827	NEXT X5
830	RETURN

TABLE 2

Measurement of KCl Head Phantom With
Three Dual-Crystal Scintillation Detectors

<u>Region</u>	<u>KCl Phantom¹</u>		<u>Control Subject²</u>
	<u>Gross (cpm)</u>	<u>Net³ (cpm)</u>	<u>(cpm)</u>
K-40	1016±10.0	1015.7±10.1	130.1±1.5
Low Energy ⁴	827.7± 9.1	825.1± 9.1	319.6±2.3

¹KCl phantom with 139.8 gm of stable potassium

²Randomly selected control subject

³Distilled water cubitainer phantom subtracted to account for electronic noise, cosmic radiation and photon scatter

⁴The low energy region encompasses most of the spectrum from 0 to approximately 150 keV

SECTION V

Low Energy, Photon Emitting Nuclides:

Reference Spectra Library

The dual crystal [NaI(Tl)-CsI(Tl)] scintillation detector has become increasingly popular in radiation measurement laboratories for several reasons:

- 1) it is an efficient detector for analyzing low energy (< 150 keV) photon radiation;
- 2) it has a large active surface area, permitting the efficient measurement of distributed sources, such as internal depositions of radio-nuclides in the human lung;
- 3) the design of the detector affords a reduction in the low energy background regions when the detector is used in conjunction with anti-coincidence electronics;
- 4) the response of the detector is directional, i.e., the efficiency of detection is highest for a source located along the radial axis; and
- 5) a single dual crystal detector can be used to generate a high energy photon spectrum simultaneously to, but independent of, the low energy photon spectrum.

A reference library consisting of the measured spectral response of the NaI(Tl)-CsI(Tl) detector to individual low energy photon-emitting nuclides in a point source geometry will undoubtedly be a valuable aid to the photon spectroscopist in a manner analogous to the Scintillation Spectroscopy Gamma-Ray Spectrum Catalogue (42), a collection of gamma spectra containing the response of NaI(Tl) detectors to

various high energy,¹ photon-emitting radionuclides. Identification of the spectral peaks should also facilitate the comparison with spectra generated by other types of detectors. The reference spectra collection being compiled at our Laboratory will be limited to radionuclides which emit photons in the energy range below 200 keV.

This section of the report contains a description of the source preparation, geometrical counting configuration, and the data handling involved in generating the reference spectra. The detector and associated electronics incorporated in the detection system are only briefly described in this section; a more detailed description of the spectrometer and low background counting facility is given in Section II.

Methods

Each source was prepared by evaporating purified solutions containing an uncalibrated quantity of an individual radionuclide on 1 inch diameter circles of Whatman #41 filter paper. The filters were dried under an infra-red heat lamp and subsequently enclosed in a ring and disk mount with a clear polyethylene cover, 0.9 mil thick.²

The counting facility at this Laboratory consists of a 2.44 m³ room with 15.24 cm thick steel walls lined with lead (5.08 mm), cadmium (0.7 mm) and copper (0.38 mm) which provide a "graded Z" shield designed to minimize natural background radiation. A low background relative to the

¹The designation "low and high" energy refers to (< 200 keV) and (> 200 keV < 3 MeV) photons respectively.

²The only exception to this method of preparation was for Xe-133,135 in which case a gaseous source, uniformly distributed about the detector, was employed.

standard NaI(Tl) detector is achieved by employing a dual thin NaI(Tl)-CsI(Tl) crystal in the anti-coincidence mode. In this manner, one can isolate photons which interact only with the NaI(Tl) crystal, i.e., predominantly low energy photon emissions (43).

In general, the pulse height distribution of each radionuclide was generated by counting the source in the above mentioned manner for a time interval suitable for demonstrating its characteristic and major photon emissions. In order that the system background should not influence the spectral shape, net spectra were obtained by subtracting a control room background spectrum (of a comparable time) for each radionuclide. The resulting spectrum was then plotted mechanically on one of two standardized rectangular grid forms, referred to as Types A and B, each having a different abscissa. The abscissa of Type A grid depicts the energy calibration associated with the pulse height distribution from the thin NaI(Tl) crystal only and is calibrated from 0-200 keV. The abscissa of the Type B grid is divided into halves; the first half is used to represent the NaI(Tl) output and is calibrated similarly to the Type A grid; the second half is used to depict the output from the CsI(Tl) crystal and is calibrated from 0.1 to 1.7 MeV. Thus, unless a particular nuclide has characteristic photon emissions above 200 keV, Type A grid is used.

Since in practice different sample-detector configura-

tions may not always yield the same spectral shape as shown here, the characteristic major photon peaks are labelled in each reference spectrum to facilitate rapid identification of unknown samples. All γ rays, L X rays and K X rays, when present, are therefore indicated. Wherever possible, daughter radiations are labelled as such. Compton and Rayleigh scatter radiation, escape peaks and bremsstrahlung continua are not labelled; in general, at photopeak energies below 80 keV, backscatter peaks are difficult to resolve from the photopeak due to the resolution of this detector design.

A list of the radionuclides analyzed in this section are presented in Table 1 and plotted in Figures 1-16. The nuclides in this table are listed alphabetically, and the individual isotopes are listed in ascending order of mass number for each element.

It should be re-emphasized at this time that the spectra given in this catalog are for point sources only and will appear somewhat different when obtained from sources present in a distributed configuration, i.e., as in a person's lung (and/or thoracic skeleton) or skull bones. Among other parameters, e.g., external bremsstrahlung, these differences are primarily due to attenuation and scattering phenomena.

References

- (42) Heath, R.L. Scintillation spectrometry gamma-ray spectrum catalogue. AEC Research and Development Report, TID-4500, 31st Edition, IDO-16880-1, 2nd Edition (1964).
- (43) Laurer, G.R. *In vivo* measurements of radionuclides emitting soft penetrating radiations. Ph.D. Thesis, New York University, May 1968.

TABLE 1

Low Energy Photon Emitting Nuclides:Reference Spectra Library

<u>Element</u>	<u>Isotope</u>	<u>Half-Life</u>	<u>Figure Number</u>
Control WBC Background	—	—	1
Americium	241	433 years	2
Americium	243	7.95×10^3 years	3
Cerium	144	284 days	4
Cesium	137	30.0 years	5
Cobalt	57	260 days	6
Curium	242	162.5 days	7
Curium	243,4	32 years, 17.6 years	8
Curium	244	17.6 years	9
Iodine	125	60.2 days	10
Lead	210	22.2 years	11
Plutonium	238	86.4 years	12
Plutonium	239	24,390 years	13
Technetium	99m	6.05 hours	14
Uranium	233	1.62×10^5 years	15
Uranium	235	7.1×10^8 years	16
Uranium	238	4.5×10^9 years	17
Xenon	133, 135	5.3 days, 9.1 hours	18

FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5

FIGURE 6

V-12

Counts

Co-57; 260 days

Point Source; Net Spectrum

NaI(Tl)/CsI(Tl) Detector

Diameter: 15.5 cm

Thickness: 3 mm / 5 cm

Entrance Window: 0.254 mm Be

FIGURE 7

Cm-242; 162.5 days
Point Source; Net Spectrum
NaI(Tl)/CsI(Tl) Detector
Diameter: 15.5cm
Thickness: 3mm / 5cm
Entrance Window: 0.254 mm Be

FIGURE 8

Cm-243; 32 yrs.
Cm-244; 17.6 yrs.
Point Source; Net Spectrum
NaI(Tl)/CsI(Tl) Detector
Diameter: 15.5 cm
Thickness: 3mm / 5cm
Entrance Window: 0.254 mm Be

FIGURE 9

Cm-244; 17.6 yrs.

Point Source; Net Spectrum

NaI(Tl)/CsI(Tl) Detector

Diameter: 15.5 cm

Thickness: 3 mm / 5 cm

Entrance Window: 0.254 mm Be

FIGURE 10

I-125; 60.2 days

Point Source; Net Spectrum

NaI(Tl)/CsI(Tl) Detector

Diameter: 15.5cm

Thickness: 3mm / 5cm

Entrance Window: 0.254mm Be

FIGURE 11

Pb-210; 22.2 yrs.
Point Source; Net Spectrum
NaI(Tl)/CsI(Tl) Detector
Diameter: 15.5 cm
Thickness: 3 mm / 5 cm
Entrance Window: 0.254 mm Be

FIGURE 12

Pu-238; 86.4 yrs.

Point Source; Net Spectrum

NaI(Tl)/CsI(Tl) Detector

Diameter: 15.5cm

Thickness: 3mm / 5cm

Entrance Window: 0.254mm Be

FIGURE 13

Pu-239; 24,390 yrs.
Point Source; Net Spectrum
NaI(Tl)/CsI(Tl) Detector
Diameter: 15.5 cm
Thickness: 3 mm / 5 cm
Entrance Window: 0.254 mm Be

FIGURE 14

Tc-99m; 6.05 hrs.
Point Source; Net Spectrum
NaI(Tl)/CsI(Tl) Detector
Diameter: 15.5 cm
Thickness: 3 mm / 5 cm
Entrance Window: 0.254 mm Be

FIGURE 15

U-233; 1.62×10^5 yrs.
Point Source; Net Spectrum
NaI(Tl)/CsI(Tl) Detector
Diameter: 15.5cm
Thickness: 3mm / 5cm
Entrance Window: 0.254 mm Be

FIGURE 16

FIGURE 17

U-238 (Th-234 - Pa-234m) $T_{1/2} = 4.51 \times 10^9$ yrs.
 Point Source; Net Spectrum
 NaI(Tl)/CsI(Tl) Detector
 Diameter: 15.5 cm
 Thickness: 3mm/5cm
 Entrance Window: 0.254 mm Be

FIGURE 18

SECTION VI

In Vivo Measurements of Exposed Individuals

Introduction

As an example of some of the actual in vivo measurements of exposed individuals made at this laboratory during the past three years, several studies will be briefly described in this section. In all cases the nuclides being measured can be classified as "bone seekers" and were measured with either two NaI(Tl)-CsI(Tl) detectors above the anterior portion of the thorax (with the detector edges resting on the clavicle and centered over the sternal notch), or with three NaI(Tl)-CsI(Tl) detectors surrounding the head (skull). For the purpose of this report, biological interpretation of this external measurement data will not be discussed.

A(1). Measurement of Occupational Contamination for ^{241}Am

During the past year measurements were performed to assess the internal body burden of a subject who came into contact with ^{241}Am during the course of a routine transfer procedure. As in the following case of the non-occupational contamination of a child and his father, the exposure occurred via inhalation so it was known that the initial deposition site was the lung. It was learned upon interview that this subject's exposure incident had occurred approximately

two years prior to whole body counting as was his only recorded exposure incident.

Upon arrival at the laboratory, routine anthropometric measurements were performed to establish such parameters as chest wall thickness and control subject matching characteristics. The various anthropometric measurements for this subject are given in Table 1. To determine the lung and skeletal burdens for this individual, measurements were performed over the anterior thorax and around the head using two and three phoswich detectors, respectively. Also, whole body measurements were performed to screen for high-energy contaminants using an 8x4 NaI(Tl) detector to preclude any possible interference from these nuclides. All measurements were performed in a graded-Z shielding, whole body counter. Details of the counting methodology and pulse shape coincident electronics have been reported in previous sections. The photon emission used to quantitate ^{241}Am was the 59.6 keV gamma which is emitted with a 36% abundance.

The estimated lung burden using the above counting methodologies was determined to be 4.01 nCi. This value is approximately 27% of the occupational lung burden limit of ~ 15 nCi for an adult. The skeletal burden estimate based on measurements of the skull is 1.93 nCi.

Using the model described in Section III, it was possible to conclude that the contribution of this subject's thoracic skeletal activity to the in vivo lung count was significant and would have increased the lung burden estimate by 15% if not taken into account.

A(2). Metabolic Studies of Am-241 in Man: Continued Follow-up of a Non-Occupational Contamination of a Child and His Father (44)

During the past 8 years, sequential measurements have been made of the internal Am-241 burdens of an adult male and his young son who are estimated to have received initial exposures within their home at the approximate ages of 50 and 4 years. It was discovered at this laboratory early in 1970, when these individuals had reached the ages of 56 and 10 years (45), that both had been internally contaminated with Am-241. From that time until the present, both subjects have been measured one or more times per year in a whole-body counting low background facility with 2 or 3 thin NaI(Tl)-CsI(Tl) "Laurer" detectors (46).

Americium-241 ($T_{1/2} = 433$ years) is an alpha particle-emitting actinide which also emits a 59.6 keV gamma ray in 36% of all decays. Although Am-241 also emits 26 keV gamma rays and 17 keV Np L X rays, only the count rate in the 59.6 keV gamma ray region was used to determine the magnitude of contamination.

Since Am-241 is a trivalent actinide which, upon becoming systemic, deposits primarily in the liver and on skeletal surfaces (47), the detectors were positioned in configurations such that measurements could be made of Am-241 in the: 1) whole body, 2) thorax (lung), 3) liver, and 4) head (skull). Results of these measurements as determined from 1970 through 1977 are listed in Tables 1 and 2. For the whole body, when first measured in 1970, values for the man and his son represent 225 and 91%, respectively, of the occupational body burden limit of 40 nCi for an adult male (48).

The estimated lung burdens for Am-241 in the adult and his young son in 1970 were 14.9 and 7.8 nCi, respectively. These values are approximately 93% and 49% of the occupational lung burden limit of 15 nCi for an adult. Estimates for lung deposits were made from count rates obtained by placing two detectors over the anterior thorax and subtracting the contribution from activity associated with the rib cage structure, as derived by assuming a constant Am-241 concentration throughout the skeleton and the thoracic bone structure as an appropriate fraction of the skeletal burden (49).

Liver burden estimates were made from *in vivo* measurements performed over the liver area, subtracting the count rate from an adjacent and identical region on the left side

of the body (with appropriate correction for contributions from activity in the liver to the measurement over the left side). Resulting values obtained in 1970 were 3.0 and 10.7 nCi for the man and the boy, respectively.

The skeletal burden was derived from measurements made with two, and subsequently three, detectors surrounding the head as a configuration for determining the amount of activity associated with the skull bones. Then, by assuming that the concentration of activity in all bones is equivalent to that measured in the skull (49,50), the total 1970 Am-241 content in the father's skeleton was determined to be 72.1 nCi and 17.9 nCi for his young son (51). The percentages of the total body burden that these values represent can be calculated from Tables 1 and 2 as 80 and 49% for the father and his son, respectively.

Finally, the total body burden was measured with two detectors placed at one meter from the subject who was placed in an arc position. As noted earlier, these values were ~90 and ~36 nCi in 1970 for the adult and his son, respectively.

The only indication of any external contamination in 1970 was noted to be on the soles of the adult subject's feet due, presumably, to his walking barefoot on contaminated floors. This activity was, in fact, external, as

could be ascertained by the obvious presence of the 17 keV X ray. Due to its relatively large mass attenuation coefficient, it is less visible when material is deposited internally.

As the result of biological loss and the three courses of decorporation therapy, the last measured estimates in 1978 for activity present in the body of these individuals are 61.8% (whole body), 65.8% (lung), 50.5% (liver) and 61.4% (skeleton) for the adult, and 33.8% (whole body), 1.3% (lung), 15.9% (liver) and 58.7% (skeleton) for the adolescent, of the initial 1970 values.

The described incident is totally unique and extremely important for a variety of reasons. The most intriguing aspect of this case is the fact that, for the first time, a child has been exposed to this manmade nuclide and that the biological behavior of Am-241 could be followed over a long time period not only in a youngster, but in an adult similarly exposed. In this manner, a comparison of retention parameters and the efficacy of chelation therapy as a function of age can be made. As seen in Figures 1a, 1b and 1c of the sequential measurements, there is no doubt that decorporation procedures were more effective in the youth where considerable bone growth was still occurring.

Continued *in vivo* measurements of these individuals are in progress, as is further consideration of the interpretation of the Am-241 content of excreta samples.

B. Estimation of Employee Exposures to Compounds of Depleted Uranium

The U.S. Nuclear Regulatory Commission Guide 8.22, entitled "Bioassay at Uranium Mills," (52) suggests that "annual *in vivo* measurements should be performed using equipment capable of measuring 9 nCi or less of uranium in the lung." As used by the Commission, the term bioassay includes *in vivo* measurements as well as measurements of radioactive material in the urine. For the *in vivo* measurement of lung burdens of depleted uranium (DU), a maximum detection efficiency has been obtained by using thin NaI(Tl)-CsI(Tl) detectors in measuring the 63 and 93 keV gamma rays of Th-234, the first daughter product of U-238 alpha decay. As a result of purification, alloying and machining operations at various industries, employees have come into contact with DU in the form of the "green salt," UF_4 or U_3O_8 during their normal operating procedures. Upon interview of these subjects, it was immediately apparent that the possibility for external exposure should be an essential consideration for any estimate of internal lung burden. The two compounds of concern, U_3O_8 and UF_4 are both "fairly insoluble" to "insoluble" compounds classified as Category Y by the ICRP Lung Task Group. It can be expected, therefore, that the greatest percentage of any internal burden will still be associated with materials

deposited in the lung tissue. Depleted uranium consists of 99.7% U-238, 0.3% U-235 and 0.0034% U-234, and is therefore measured with greater sensitivity using the 63 and 93 keV gamma rays of Th-234 rather than the 185 keV photons from U-235.* It should be noted, however, that since *in vivo* analysis of uranium is routinely based on equilibrium between U-238 and Th-234, the assessment of uranium in the lungs of these men could possibly be different from an equilibrium situation, i.e., if exposures are to a material which has a uranium/thorium ratio different from that used in the phantom calibration torso, a significant difference might be expected when uranium is determined by the results of thorium measurements. Since Th-234 has a radiological half-life of 24.1 days, radiological equilibrium will be established in a matter of months. When material is "fresh," however, and the ratio of uranium to thorium is not that of radioactive equilibrium, an incorrect answer might result. Finally, whether or not radioactive equilibrium between

*Measurement of these higher energy gammas is, of course, indicated in the case of possible exposures to enriched uranium where U-235 is present in considerably higher percentages than in the case of the 0.7% natural uranium.

uranium and thorium has occurred, one should still consider possible differences in the biological retention of these two elements.

Each of the potentially exposed individuals were asked to undergo extensive personal hygiene preparation at home prior to whole-body assay. Before entering the whole body counting room, the subjects were required to shower at our laboratory and to wash their hair well with a strong shampoo. Subjects were then asked to wear freshly laundered, disposable, whole-body counting uniforms. In addition to whole-body counts in the standard chair geometry using an 8"x4" NaI(Tl) detector, dual crystal detectors were positioned above the anterior and posterior thorax as well as on either side and at the base of the head for measurement of low energy photons from these bone-seeking nuclides, which may have deposited in the skull bones. The dual crystal detectors were also placed over the kidney areas with the individual in the prone position. By examination of the ratio of the two photon peaks and by consideration of the relative count rate with detectors above either the anterior and posterior thorax, it was possible to determine the extent of external contamination for the subjects measured.

Thorium-234 has a half-life of 24.1 days and decays by beta emission with associated gammas of 63 keV (3.5%) and 93 keV (4%) abundance. For these measurements it was

assumed that Th-234 has come into secular equilibrium with its parent, U-238, and that these two nuclides do not translocate from each other in the body, but remain in *in situ* radioactive equilibrium at all times. The ratio of the X-ray peaks (approximately 15.5-16.5 keV) to the thorium gamma photon peaks can be used to determine whether material is deposited externally or present within the lung material. Finally, there are also photon peaks from Pa-234m occurring at 763 keV (0.3%) and 1.0 MeV (0.6%). However, their abundance is not as great as that of thorium gammas, thereby making their use less efficient than that of Th-234. The minimum significant measurable activity (MSMA) for our system utilizes the photons from Th-234 and is equivalent to 0.82 nCi of DU of this given composition or approximately 1-2 mg of uranium. Using the calibration factors derived from phantom construction employing the same material present at the site of the subject's employment, it was possible to obtain values for the activity in nCi and the number of mg of uranium present in each of the men measured.

Estimated results of internal and external DU exposure was given for each of the men measured and the difference between DU in a point source geometry (Figure 2) can be compared with that of a lung phantom containing this same material (Figure 3) compared to a control subject anterior thorax measurement obtained after a 30-minute count (Figure 4) and that of a subject (Figure 5) occupationally

exposed with a lung burden of 24.2 ± 0.5 nCi of DU. As noted in this last figure, some counts in the X-ray region still exist, indicating that the exposure was not totally internal and that in fact some material does remain externally deposited. This is certainly the case for similar measurements made over the head (Figure 6), where it can clearly be seen that there was a considerable contribution from material present externally and probably associated with the individual's hair.

C. In Vivo Measurement of Am-241 and Pu-239 in the Presence of Cs-137; Measurements of Residents of the Marshall Island Atolls (53)

A method has been developed for the measurement of Am-241 and Pu-239 in individuals contaminated with Cs-137. Dual crystal detectors are used for the measurement of low-energy photons in the presence of interference from both the 32 keV X ray from barium and scattering from the 0.662 MeV gamma from Ba-137m which both result from the beta decay of Cs-137. The MSMA of Am-241 in the head (skull) varies as the square root of the amount of Cs-137 in the body and equals 30 pCi for a subject with a Cs-137 burden of 200 nCi compared to 20 pCi subject with approximately 3 nCi of Cs-137 in their body. Three detectors were placed in a geometry which surrounds the head (skull). Pulse shape discrimination enabled measurements of low-energy photons from Am-241 and Pu-239 in the 3 mm NaI(Tl) crystal simultaneously with

higher energy events in the 5 cm CsI(Tl) detector. The low-energy spectrum can be corrected thereby for interference from an amount of Cs-137 measured in an identical configuration using the high energy spectrum.

Five individuals who spent different amounts of time on the Marshall Islands were measured for Am-241 and Pu-239. Two men were on Bikini Island and Majuro Atoll (approximately 400 miles south of Bikini) and resided there for four years after the period of testing. The first of these subjects had a Cs-137 body burden of 1.72 ± 0.004 μ Ci and the second 15.9 ± 0.5 nCi (Figure 7). A suitable Cs-137 phantom was subtracted to account for contribution of the 0.662 MeV gamma of Ba-137m and the 32 keV X-ray characteristic of barium. The resulting count rate in the Am-241 60 keV energy region, was not different from a mean control subject's spectra. It was concluded that there was no detectable Am-241 or Pu-239 in any of these subjects at the time of measurement. It has been calculated that with 1.72 μ Ci of Cs-137 present, our lower limit of detection for Am-241 in the skull is approximately 200 pCi.

D. Measurement of Occupational Contamination for Pb-210

In cooperation with the Canadian Radiation Protection Bureau, measurements have been performed on two individuals to estimate skeletal and lung burdens of Pb-210. One of the subjects (designated as IE-12) was of particular interest since he was suspected to have a significant burden of Pb-210 acquired 20-28 years ago during the preparation of RaD (Pb-210) sources in a laboratory.

Both subjects were measured in the whole body counting facility for lung, skeletal and whole body burdens for both low-and high-energy emitters. The various site specific arrays used for these measurements involved three detectors surrounding the head for skeletal burden estimates; two detectors positioned over the anterior thorax for lung burden estimates and an 8x4 NaI(Tl) detector placed in a modified meter arc position to quantitate whole body burdens.

The estimated skeletal burden for subject IE-12 is 1.8 nCi. This value is derived by assuming the concentration of activity in the skull is representative of activity associated with all bones. Since the Pb-210 activity associated with this subject's skull (~ 28 nCi) was well above our lower limit of detection of 300 pCi, a series of measurements were performed around the subject's head to evaluate individual

detector responses from the left side, right side and top sections of the cranium. By comparing these measurements to analogous measurements on our Pb-210 skull phantoms, it was possible to verify that the phantom structures accurately represent the average activity distribution in the head for each skull surface. By comparing these measurements to analogous measurements for our previously constructed Pb-210 skull phantoms, it was possible to verify that the phantom structures accurately represent the average activity distribution of Pb-210 in the human skull for each skull surface. The Pb-210 skeletal estimate for the other subject (IE-11) was inconclusive due to the presence of a higher than normal amount of Cs-137 contamination. Since the barium X-ray of Cs-137 emits a 33 keV photon, it was not possible to sufficiently resolve the 47 keV Pb-210 photon from this peak. Currently, multiple least squares regression analysis techniques are being attempted as a solution to this spectral overlap.

Anterior thorax measurements performed for both individuals revealed significant quantities of Pb-210 for subject IE-12 and Ra-226 for subject IE-13. Since most of subject IE-12's anterior thorax count rate was due to Pb-210 associated with the thoracic skeleton, it is necessary to determine a Pb-210 skeletal calibration factor. Future plans include the development of a Pb-210

skeletal phantom to accurately assess this activity, however, at this time we are attempting to calculate this subject's thoracic skeletal activity by using the already existing Am-241 skeletal phantom and applying attenuation and abundance correction factors.

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- (51) For the adolescent, the skull extrapolation technique did not agree with a "process of elimination" method (i.e., total body = liver + lung + skeleton). Since maximum confidence was placed in the whole body measurement and since bone growth was rapid in the youth, the latter method of calculation was used to obtain his skeletal estimates.
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TABLE 1

Physical Measurements for Subject IE-3

Height	1.71 m
Weight	80.7 kg
Age	39 yrs
Average Chest Wall Thickness*	27.9 mm
Time After Exposure	2 yrs
Chest Circumference	1.06 m

*Based on calculation of Dean, P.N., Estimate of Chest Wall Thickness in Lung Counting for Plutonium, Health Phys. 24: 440 (1973).

TABLE 2

Estimation and Sequential Measurements of ^{241}Am Distribution in an Adult Male⁽¹⁾

<u>Months Since 4/70</u>	^{241}Am Burden, nCi			
	<u>Lung</u>	<u>Liver</u>	<u>Skeleton⁽²⁾</u>	<u>Total Body⁽³⁾</u>
0	14.9	3.0	(70.1) 72.1	90.0
* 12	15.5	2.6	(75.9) 38.8	56.9
17	19.4	3.1	(74.7) 33.6	56.1
29	12.7	4.9	(70.1) 54.6	72.2
37	16.4	2.9	- 54.0	73.3
* 38	16.3	5.2	- 46.5	68.0
42	11.7	1.6	- 48.0	61.3
52	12.3	1.3	(40.2) 42.4	56.0
57	10.4	0.7	(34.5) 50.2	61.3
* 58	11.5	1.5	(40.2) 41.3	54.3
75	9.3	0.4	(48.4) 49.9	59.6
93	9.8	1.5	(45.3) 44.3	55.6

(1) No errors have been assigned to these values since it was determined that unknown positioning consistency, body size variability, detector refabrication and changing biological distribution of ^{241}Am will all account for errors which may be larger than counting statistics alone.

(2) Values in parenthesis determined by extrapolation of skull measurement. Other values determined by: skeleton = total body-lung-liver; values so obtained for 12 and 17 months may be in error due to detector and calibration change.

(3) Maximum permissible occupational body burden = 40 nCi.

* Decorporation therapy administered during this period.

TABLE 3

Estimation and Sequential Measurements of ^{241}Am Distribution in an Adolescent Male⁽¹⁾

<u>Months Since 4/70</u>	<u>^{241}Am Burden, nCi</u>			
	<u>Lung</u>	<u>Liver</u>	<u>Skeleton⁽²⁾</u>	<u>Total Body⁽³⁾</u>
0	7.8	10.7	17.9	36.4
12	4.6	5.2	9.9	19.7
17	4.0	5.2	13.9	23.1
29	3.8	6.5	13.0	23.3
37	3.1	4.1	19.1	26.3
38	2.8	6.4	12.1	21.3
42	2.1	4.2	11.7	18.0
52	0.3	3.8	14.7	18.8
57	ND	2.6	15.9	18.5
58	0.3	1.8	10.1	12.2
75	ND	2.3	10.7	13.0
93	0.1	1.7	10.5	12.3

(1) No errors have been assigned to these values since it has been determined that unknown positioning consistency, body size variability, detector refabrication and changing biological distribution of ^{241}Am will all account for errors which may be larger than counting statistics alone.

(2) Skeletal estimates obtained from: skeleton = total body-lung-liver.

(3) Maximum permissible occupational body burden = 40 nCi.

* Decorporation therapy administered during this period.

FIGURE 1. Americium-241 content as a function of time and chelation therapy in a) liver, b) lung and c) total body.

FIGURE 2

Depleted Uranium U_3O_8 Net Spectra

60 minute count, dual detector B (NaI)

80 nCi ^{238}U 200 mgm

VI-22

ACTIVITY (Counts)

FIGURE 3

PHANTOM: 216.2 nCi D.U.

GEOMETRY: Anterior Thorax

FULL SCALE COUNTS: 2048

TIME: 30 min.

VI-23

FIGURE 4

CONTROL SUBJECT

GEOMETRY: Anterior Thorax

FULL SCALE COUNTS: 512

TIME: 30 min.

FIGURE 5

OCCUPATIONAL EXPOSED SUBJECT

GEOMETRY: Anterior Thorax

FULL SCALE COUNTS: 512

TIME: 30 min.

FIGURE 6

OCCUPATIONALLY EXPOSED SUBJECT

GEOMETRY: Head

FULL SCALE COUNTS: 512

TIME: 60 min.

FIGURE 7

Marshall Islands, Bikini Atoll Resident

Head Measurement; 60 Min.

2-NaI(Tl) CsI(Tl) Detectors

Body Burden = $1.72 \mu\text{Ci}$ Cs-137

$0.13 \mu\text{Ci}$ K-40

SECTION VII

Xenon-133,135 Interference with In Vivo

Low-Energy Measurements Systems

Xenon-133 and ^{135}Xe are abundantly produced gaseous fission products released in varying amounts during the normal operation of nuclear power and research reactors. A member of the family of noble gases, xenon is a chemically unreactive element, and consequently, its isotopes can be detected in the atmosphere at considerable distances from their production site (54). Furthermore, due to their inert character and relatively short half lives, i.e., 5.29 days for ^{133}Xe and 9.14 hours for ^{135}Xe , these nuclides will not accumulate in man or the environment to any appreciable extent and therefore receive minor consideration as a long-term radiological health problem. It was noted in one of the earliest reports describing the "crisis" at the Three Mile Island Nuclear Power Plant site that the radiation released from that facility, although undesirable, had not produced a serious health hazard. "Most of the escaping gas is xenon, mixed with a smaller amount of krypton. Both are inert, ... and ... will decay and dissipate in the atmosphere" (55).

Although dissipation and decay may indeed represent the fate for low levels of these beta/gamma-emitting nuclides, there is an increasing awareness that the presence of ^{133}Xe and ^{135}Xe released during normal reactor operation can represent a significant interference to in vivo low-energy, photon-emitting (lung and whole body) assay procedures. Xenon-133 has a characteristic photon spectrum with a cesium K X-ray photopeak at 31.6 keV and a gamma-ray photopeak at 81.0 keV (31% abundant), whereas ^{135}Xe emits gamma rays at 250 keV (91% abundant) (Figure 1). Their presence in the air of a whole-body counter may therefore contribute to the background count rate in the energy regions of interest for important radionuclides which emit photons below 300 keV. This is especially the case for radiation measurements made with dual thin crystal NaI(Tl)-CsI(Tl)-type scintillation detectors (56,57) for actinide nuclides including ^{235}U , ^{239}Pu , ^{238}Pu , ^{234}Th , $^{243,4}\text{Cm}$, ^{241}Am and the ^{238}U decay chain daughter, ^{210}Pb . Although the photopeaks for an ^{241}Am point source photon spectrum in the presence of ^{133}Xe do not overlap (Figure 2), this is not usually the case for the resolution associated with a distributed and shielded source. For a nuclide within a person, or for ^{133}Xe in the presence of a scattering mass,

significant increases in the lower limit of detection may result. Thus, for other nuclides or for higher concentrations of xenon in air, the contribution from ^{133}Xe and ^{135}Xe could become a primary consideration for determining the ability to detect regulatory organ and body burden limits.

With growing electronic sophistication, it is becoming increasingly possible to determine relatively low levels of internal radionuclide contamination in man. It is essential, therefore, to make gamma spectroscopists aware of the possible existence of ^{133}Xe and ^{135}Xe in the air of their whole body counting facilities. As has been previously noted, these gaseous fission products are released in significant quantities by boiling water reactors (54) and, to a lesser extent, by PWR reactors. Also, these nuclides are manufactured directly in significant amounts by reactors engaged in the production of radiopharmaceuticals. For whole body counting facilities with air supplies which do not have low-temperature activated charcoal traps for removal of radon (^{222}Rn) and other inert gases, the potential for ^{133}Xe and ^{135}Xe entering a facility and resulting in interference problems is a distinct possibility when in vivo counting laboratories are in meteorological proximity to nuclear reactors. In our whole body counting room* of approximate dimensions $(2.5\text{ m})^3$ with two 15 cm

*Steel, 15.24 cm; lead, 5.08 mm; cadmium, 0.7 mm; copper, 0.38 mm.

NaI(Tl)-CsI(Tl) detectors located at the midpoint of the room, measured detection efficiencies of 2.1 net cpm in the 45-75 keV ^{241}Am energy region (background = 6.15 cpm) and 8.0 net cpm in the 65-90 keV ^{133}Xe energy region (background = 7.0 cpm) have been observed for a concentration of 1 pCi of ^{133}Xe /liter whole body counting room air.

With increasing numbers of nuclear electrical generating plants, there will be a corresponding increase in the ambient level of ^{133}Xe and ^{135}Xe . Although these nuclides are released without danger of exceeding the MPC_a , they may be of concern for whole body counting facilities which routinely monitor low level lung and body burdens of the actinides.

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FIGURE 1. $^{133,5}\text{Xe}$ Photon Spectra; Distributed Source - NaI(Tl)/CsI(Tl) Detectors.

FIGURE 2. $^{133,5}\text{Xe}$ and ^{241}Am Photon Spectra; Distributed and Point Sources -
NaI(Tl)/CsI(Tl) Detectors.

SECTION VIII

Estimation of Background Corrections to Obtain Net Count Rates for In Vivo Measurements of Exposed Subjects

Dual crystal scintillation detectors are employed to measure the presence of low energy (< 200 keV) emitting radionuclides in the body. Usually a site specific array of detectors is positioned around the subject in close proximity to the organ or tissue suspected of containing the contaminant. Current practice for the measurement of internally-deposited radionuclides in man relies on the background subtraction of selected unexposed individuals who are similar in age, height, weight, and sex to the exposed subject. Even though control subjects are carefully selected on the basis of anthropometric parameters, the normal count-rate variability measured in the control population can exceed 15% depending on the energy region of interest. It is this variability in subtraction techniques that necessitates the development of a procedure for in vivo analysis that is independent of subtraction of a control subject's count-rate.

To develop an independent, subject-specific method for the determination of in vivo-deposited radioactivity based on a single measurement, it is necessary to precisely define the shape of the background spectrum for the region < 160 keV. The characteristic spectral shape of this

region is shown in Figure 1. It is apparent from this observation that the shape of this spectra can be closely approximated by two straight lines which intersect at approximately 80 keV. As can be seen from the figure, this is only true between the energies 8 and 160 keV. This is fortuitous, however, since many of the radionuclides of interest emit low energy photons that fall within this region (i.e., ^{241}Am , ^{234}Th , ^{239}Pu , ^{238}Pu , and ^{210}Pb).

To quantitatively determine if the 0-160 keV region can be approximated by two straight lines, seven control subject background spectra were analyzed using a least squares fit linear regression analysis for the 0-80 keV region and the 80-160 keV region. The linear correlation coefficient for each region was calculated using an interactive basic computer program so that data could be processed directly from the analyzer. All subject counts were routine chest counts using two dual crystal detectors positioned over the midline of the sternum and the detector housing adjacent to the clavicles. The analysis of the data is shown in Table 1. The subject mean linear correlation coefficient for the 0-80 keV region was 0.88 ± 0.08 and 0.89 ± 0.10 for the 80-160 keV region. Correlations of 0.98 and 0.99 were obtained for the chest phantom and room background spectra, respectively.

The inflection point for all curves was observed to be at approximately 80 keV; however, the inflection point tended to vary by approximately 5 keV and is not a sharp change. In Figure 1 it can be seen that it is really a more gradual curve than a sharp maximum. This maximum at approximately 80 keV represents a physical minimum in the absorption of photons in the body at this particular energy. Figure 2 shows the plot of the mass attenuation coefficient for gamma rays in water vs. photon energy. It can be seen from the graph that there is a minimum at approximately 80 keV in the total absorption (μ_a/ρ) of these photons which will result, therefore, in a greater transmission of these photons from the body. Thus, any photon in the body which is scattered to 80 keV will have an apparently greater detection efficiency.

Once it has been established that the two sides of the low energy spectra curve can be very closely approximated by two linear functions, the analysis of radio-nuclides within these regions can be performed by a simple technique which determines if there is significant deviation from linearity over the region of interest.

To illustrate this technique, let us examine the particular case of the determination of an ^{241}Am lung burden. For low level analysis we ordinarily wish to determine whether or not there is significant activity

above background in the region of interest. We also know from previous analysis of a number of control subjects that the background region from 12-80 keV is very closely approximated by a linear function; therefore, if we select two regions of interest on either side of the ^{241}Am photopeak region (45-75 keV) and determine the number of cpm per channel in these regions, we would expect that the average of these two bracketing regions would very closely approximate the cpm per channel in the intervening ^{241}Am photopeak region. This is true only if there really is no ^{241}Am in the subject. If one then calculates the observed cpm/channel in the ^{241}Am photopeak region and compares this to the extrapolated or expected cpm/channel in the region, one can determine if this ratio exceeds a calculated critical ratio which can be set any any desired level of significance. This will, in effect, allow a determination of whether or not the counts in the photopeak region represent random statistical fluctuation or are due to the presence of some radionuclide which emits a photon in that particular energy region. The major limitation of the technique lie in the fact that one must know in advance what the suspected contaminant is and also that there be only one contaminant.

The above described methodology has been applied to five individuals measured at this laboratory for possible

contamination of this region. These individuals lived nearby a uranium processing plant that deals with significant amounts of depleted uranium (DU) and, therefore, the potential for DU contamination of these individuals was a possibility. Some relevant physical parameters of these individuals is given in Table 2.

For the whole body counting measurements, the photons from ^{234}Th were used to determine the presence of ^{238}U . Thorium-234 has a half-life of 24.1 days and decays by beta emission with associated gammas of 63 keV (3.5% abundance) and 93 keV (4% abundance). For these measurements it was assumed that ^{234}Th has come into secular equilibrium with its parent ^{238}U and that these two nuclides do not translocate from each other but remain together at all times. This may not always be the case since it has been shown that ^{234}Th and ^{238}U are not always in equal proportions and that it may be possible to have either of uranium or thorium according to the manufacturer's process site from which samples are collected. For long term burdens, however, it can be assumed that the two nuclei will eventually achieve equilibrium and will remain together in the body.

Using the previously described methodology, the ratio of observed count rates to expected count rates for the photopeak region of 80-106 keV was calculated. This data

is tabulated in Table 3. From the table it can be seen that the ratio of observed to expected indicates no positive activity in any of the cases (i.e., there is no significant deviation from unity and we thus can conclude that there is no positive contamination in any of the subjects measured.

To facilitate spectral analysis of subject data an interactive computer program has been written which allows rapid data analysis using the above methodology. The program is flexible in that all that need be entered into the analyzer are the three desired regions of interest for the particular analysis. Figure 3 gives a listing of the program which calculates the observed to expected ratio, propagates the error on the analysis and reports a 95% confidence interval estimate on the ratio. If the ratio exceeds the 95% confidence interval estimate, then the subject is a possible positive and further measurements are necessary to determine the level of the contaminant in the individual. This would involve, using a subject recount for a longer period of time, subtraction of anthropometrically matched control and calibration using an appropriate phantom structure.

To summarize, a subject specific (i.e., the subject acts as their own control) method has been developed whereby a subject can be rapidly screened for positive contamination based on linear extrapolation of the low-energy phoswich spectrum. The major limitations

of the technique is that it is applicable only to situations where the possible contaminant is known and represents the only contaminant present. An interactive computer program has also been written to facilitate rapid data analysis. Future plans are to further define the shape of the low energy spectrum with more detailed mathematical considerations although for present the linear model appears to be a reasonable accurate fit approximation.

TABLE 1

Linear Correlation Coefficients for the
Phoswich Low Energy Spectrum

Subject	8-80 keV Coefficient (r)	8-160 keV Coefficient (r)	Counting Time (sec)
AF	0.94	0.92	3800
AN	0.94	0.98	7200
MM	0.89	0.93	1800
TV	0.95	0.97	7200
JR	0.90	0.93	1800
JF	0.80	0.78	1200
WI	0.73	0.73	1800
$\bar{X} \pm 1 \sigma$	0.88 ± 0.08	0.89 ± 0.10	
Lung Phantom	0.98	0.99	60000
Room Background	0.98	0.99	100000

TABLE 2
Subject Physical Data

	<u>Subject</u>	<u>Date of Measurement</u>	<u>Birth Date</u>	<u>Sex</u>	<u>Height (cm)</u>	<u>Weight (kg)</u>
1)	TM	12/11/79	01/06/18	M	171.5	83.6
2)	MM	12/11/79	12/24/19	F	157.5	50.9
3)	PL	12/12/79	09/21/39	F	163.8	92.7
4)	JL	12/12/79	11/18/64	M	156.9	46.4
5)	MC	12/12/79	01/11/56	F	160.0	51.4
6)	GC	12/12/79	10/13/74	F	105.4	17.3
7)	Control (EC)	01/03/80	04/18/25	F	161.9	71.4
8)	Control (TM)	01/04/80	04/26/57	F	160.0	52.7
9)	Control (KM)	12/20/79	07/06/74	F	114.9	20.7
10)	Control (RD)	04/11/79	10/01/44	M	176.5	91.0

TABLE 3

Subject Count-Rate Data for Estimated Lung Burden
(Th-234 Region)

Subject	Region 1 = 77.5-80.6 keV (cpm/keV)	ROI = 80.0-101.0 keV (cpm/keV)	Region 3 = 102.9-106.0 keV (cpm/keV)	Calculated ROI	ROI Measured ROI Calculated $\pm 2\sigma$
1) TM	3.25	2.75	2.32	2.78	0.99 $\pm$ 0.10
2) MM	1.82	1.83	1.54	1.68	1.09 $\pm$ 0.14
3) PL	2.94	2.59	1.90	2.42	1.07 $\pm$ 0.13
4) JL	2.33	2.17	1.92	2.13	1.02 $\pm$ 0.11
5) MC	2.42	2.06	1.56	1.99	1.04 $\pm$ 0.14
6) GC	1.31	1.13	1.21	1.26	0.90 $\pm$ 0.18
7) Control (EC)	3.13	2.68	2.03	2.58	1.04 $\pm$ 0.13
8) Control (TM)	2.31	2.03	1.97	2.14	0.95 $\pm$ 0.14
9) Control (KM)	1.21	1.09	0.79	1.00	1.09 $\pm$ 0.26
10) Control (RD)	3.24	2.90	2.56	2.89	1.00 $\pm$ 0.10

VI-III-V

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*90 LIST
90 REM THIS PROGRAM CALCULATES THE OBSERVED TO EXPECTED RATIO
91 REM FOR A SELECTED REGION BASED ON LINEAR EXTRAPOLATION
92 REM OF TWO ADJACENT REGIONS
95 PRINT "ENTER REGIONS IN ANALYZER AS 1,2, AND 3."
6 PRINT "WHEN FINISHED ENTER 1"
97 PRINT " "
98 INPUT Q9
99 PRINT " "
100 CALL 4,18
110 LET X= ROI (1)
120 LET X1= LCN (157)
130 LET X2= LCN (158)
140 CALL 4,18
150 LET Y= ROI (2)
160 LET Y1= LCN (157)
170 LET Y2= LCN (158)
180 CALL 4,18
190 LET Z= ROI (3)
200 LET Z1= LCN (157)
210 LET Z2= LCN (158)
220 LET D1=X2-X1
230 LET D2=Y2-Y1
240 LET D3=Z2-Z1
250 LET C1=X/D1
260 LET C2=Y/D2
270 LET C3=Z/D3
280 LET A=(C1+C3)/2
290 LET R=C2/A
300 LET S1= SQR (X)
310 LET S2= SQR (Y)
320 LET S3= SQR (Z)
330 LET P1=S1/D1
340 LET P2=S2/D2
350 LET P3=S3/D3
351 LET J1=P1*P1
352 LET J2=P3*P3
353 LET J3=J1+J2
360 LET E7= SQR (J3)
361 LET E8=E7/2
370 LET I1=(E8/A)*2
371 LET I2=(P2/C2)*2
372 LET E9= SQR (I1+I2)
380 PRINT " "
381 LET E5=R*E9
384 PRINT "THE OBSERVED TO EXPECTED RATIO + OR - 2 SIGMA IS"
385 PRINT R,2*E5
386 PRINT " "
387 PRINT "INTEGRAL          SCHN          ECHN"
389 PRINT "-----          ----          ----"
400 PRINT X,X1,X2
410 PRINT Y,Y1,Y2
420 PRINT Z,Z1,Z2
421 PRINT " "
430 PRINT "THE COUNTING TIME WAS ", LCN (122)/60," MINUTES."
440 STOP

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FIGURE 1. Interactive Computer Program for Calculation of Significant Activity.

FIGURE 2

MASS ATTENUATION COEFFICIENTS FOR GAMMA RAYS IN WATER

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Hill Book Co., Inc.

FIGURE 3. Phantom Background Spectrum.

SECTION IX

Evaluation of Detector Positioning for Low-Energy In Vivo Gamma Spectrometry

The precision of any in vivo measurement depends to a great extent on the reproducibility of detector positioning over the site of a suspected contaminant. For example, the positioning of dual crystal detectors over the anterior thorax is subject dependent since the height and angle of crystal positioning depends on the specific chest contour of the subject. This problem is compounded further for some female subject measurements since the mass of breast tissue among females is highly variable.

Two methods are commonly utilized by laboratories performing in vivo monitoring to ensure that the detectors can be arranged in a reproducible geometry. These methods include 1) physically placing the detectors on the subject's chest and allowing the detectors to move freely with the subject's breathing pattern or 2) fixing the detectors over the chest at close proximity with no concession made for the normal rise and fall of the chest. The latter method of detector positioning has been adopted for our routine counting since it is believed that for longer counting times (≥ 30 minutes) an average detector-to-chest distance can be assumed. This arrangement also allows for more precise repositioning of the detectors since the mounting assemblies can be rigidly fixed during counting and the X, Y and Z coordinates of the assembly can be recorded. Precise detector repositioning is important for chest recounts if one desires to

monitor the clearance of an inhaled radioactive aerosol over time to determine the efficacy of chelation therapy treatment.

To ensure precision in detector positioning relative to a subject, two of our dual crystal detectors are mounted on independent assemblies each with six degrees of rotational freedom. This allows each detector to be positioned in X, Y and Z space coordinates as well as three degrees of angular rotation about these planes which are arbitrarily designated as θ , ϕ , and ψ . All of the possible degrees of movement are furnished with calibrated scale markings for reference purposes. Figure 1 shows a simplified drawing of the detector mounting assemblies.

The potential sources of error for an in vivo lung burden estimate are attributable to 1) variation in the area of the lung covered by the ribs, 2) contribution of skeletally deposited activity to the gross count, 3) chest wall thickness uncertainty, 4) non-uniformity of lung activity distribution, and 5) relative positioning of the detectors. We have investigated the relative detector positioning variability using the previously described positioning apparatus. To determine the variability of this parameter, a volunteer subject has been repetitively measured over the chest and around the head. In addition, an Am-241 lung phantom was measured over the chest in a similar manner. The detector positioning over the chest utilized two dual detectors positioned over the anterior thorax with the margins of the crystals adjacent to the clavicles and centered over the midline of the sternum. The head count employed three dual detectors positioned as shown in Figure 2. Figures 3 and 4 show a gross

phoswich transmission spectrum for a measurement taken over the chest and head, respectively.

The volunteer subject measurements were performed over a six-day period with a single measurement taken each day. The subject was placed in a supine position on a plexiglas whole-body counting table with the detectors positioned around the head and chest geometries as previously described. Prior to each day's count a standard calibrated Am-241 point source was measured in a reproducible geometry to insure that the efficiency of the detection system had not changed from the previous day.

This insured that any variability in the measured count rate would be due to detector repositioning. In addition, a room background count was performed to preclude the possibility of xenon gas interference since xenon is known to affect low-level, whole-body counting measurements. At the first counting session the X, Y and Z and the θ , ϕ , and ψ coordinates were recorded as a reference point for the next measurement. In between measurements the detector settings were returned to their normal "storage" position so that all subsequent counts required a complete resetting according to the original reference parameters.

The results of the chest and head repositioning measurements are shown in Tables 1 and 2. As can be seen for the individual regions of interest, the count rate variability was no greater than what would be expected from Poisson counting statistics at the 95% confidence level. This indicates that the

variability in detector repositioning for a single subject for both the head and chest geometry is reproducible for a given subject and does not introduce any significant error to the analysis.

The question arises, however, as to how well the above conclusion would hold for a subject with a lung burden. The apparent consistency of the uncontaminated subject chest measurement may not be a true indication of an exposure case since the background count rate of the chest may be uniform over a large area relative to an exposure deposition. To investigate this possibility, a Rando chest phantom was prepared with a uniform distribution of Am-241 in the lungs. The chest phantom was then repetitively measured in the same manner as the volunteer subject. Table 3 shows the results of the measurements. The variability is of the same order as for the uncontaminated subject measurement. Further experimentation is currently in progress to define the count rate variability (as a function of detector positioning) for non-uniform distributions of activity in the lung.

TABLE 1
Head Measurement

Count #	Am-241 ROI 45-75 keV (cpm)	Pu-239 ROI 11-25 keV (cpm)	Low Energy 13-184 keV (cpm)
1	55.7±3.4	29.4±2.4	270.0±7.3
2	68.2±3.7	30.0±2.4	298.4±7.7
3	69.1±3.7	26.8±2.3	295.1±7.7
4	65.1±3.6	26.4±2.3	290.0±7.7
5	64.8±3.6	29.1±2.4	297.0±7.7
6	62.2±3.5	29.5±2.4	279.1±7.5
$\bar{X} \pm 1\sigma$	64.2±4.4	28.5±1.40	288.3±10.38

TABLE 2
Chest Measurement

Count #	Am-241 ROI 45-75 keV (cpm)	Pu-239 ROI 11-25 keV (cpm)	Low Energy 13-184 keV (cpm)
1	87.2±3.4	21.3±1.7	337.7±6.7
2	87.2±3.4	21.7±1.7	336.5±6.7
3	87.4±3.4	21.0±1.7	335.0±6.7
4	90.5±3.5	22.6±1.7	346.3±6.8
5	90.3±3.5	23.0±1.8	347.3±6.8
$\bar{X} \pm 1 \sigma$	88.52±1.72	21.92±0.85	340.56±5.79

TABLE 3

Phantom Count Rate for Am-241 for Different Trials

Count #	Am-241 (cpm)
1	691.8±26.3
2	622.1±25.0
3	646.6±25.4
4	703.9±26.5
5	668.1±25.8
6	646.6±25.4
$\bar{X} \pm 1 \sigma$	663.18±30.79

FIGURE 1

FIGURE 2

Subject-Detector Standard N.Y.U. Head Counting Geometry

6-XI

FIGURE 3

Control Subject Chest Count

IX-10

ACTIVITY (Counts)

IX-11
COUNTS

NaI(Tl)

CsI(Tl)

Control Subject
Head Count
60 min.

0

(kev)

160

E N E R G Y

0.2

(MeV)

1.4

1.46 MeV
K-40

SECTION X

Modification of Existing Computing Capabilities

To upgrade the existing computing facilities for whole body counting data analysis, a system renovation has been undertaken which: 1) expands the existing storage and computational memory, 2) provides a multi-language computer-based system, and 3) increases data input/output speed.

The major addition to the existing system has been the installation of a General Datacom 212A modulate demodulate (modern) data set which now links the MCA interfaced 16K computer to both a CDC 6600 computer located at the NYU Courant Institute of Mathematical Sciences (CIMS) and to an IBM 1170 at the Health Science Computing Facility at Harvard University. With this addition, core space available for data storage and processing is virtually unlimited. The CDC 6600 includes under its Network Operating System (NOS) a FORTRAN compiler, an assembly language processor known as COMPASS as well as other language capabilities such as ALTRAN, APL, LISP, PASCAL, PLA and SPITBOL. Included in the NOS of the CDC 6600 are many preprogrammed scientific computational routines which are on file from the Los Alamos Scientific Laboratory, the Argonne National Laboratories and the Harwell Energy Research Establishment. In addition, graphic software routines are now available to obtain plotted output of data. It is planned that a permanent file of all exposure and control data will eventually be maintained for retrieval by this as well as any other laboratory with a computer

link to the CIMS. In this way, interlaboratory comparisons will be facilitated for the rapid exchange of calibration and count-rate data.

The General Datacom modem is interfaced with an IBM 3101 Model 12 display terminal which has a switch selectable BAUD rate of 110 to 9600 to regulate speed of data transmission. The 3101 provides a common display input/output control for both the NOVA 16K laboratory-based computer and the modem through a selectable EIA transfer switch. The 3101 terminal allows for fast file and program editing by virtue of its CRT capabilities.

To provide for hard copy input/output, an electronic teletype ASR 4330 (automatic send/receive) printer has been added. This peripheral unit provides faster output (300 BAUD) than the previous model 30 TTY. The ASR 4330 is linked to the CRT via an auxiliary printer port and provides output for either the NOVA or the modem. The ASR 4330 was chosen due to its additional ability to receive and send paper tape code. This was an important consideration since in addition to flexible disc and cassette tape some data here continues to be collected on paper tape. With this option, it is then possible to send data to a computer file without going through intermediate translational processes.

To complete the upgrade of the computing system, the single-drive TN-1002 cassette tape unit has been modified to a dual-drive system. This modification will allow for storage of a master program library on one drive unit while allowing for coincident intermediate spectral storage area in the other.

With this configuration, spectra can be processed before being placed on permanent file. Also, permanent files can be retrieved at some future date and modified in the analyzer without loss of the original data.

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