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## MEASUREMENT OF THORIUM ISOTOPES AND $^{228}\text{Ra}$ IN SOFT TISSUES AND BONES OF A DECEASED THOROTRAST PATIENT

James F. McInroy, Edward R. Gonzales, and John J. Miglio\*

**Abstract**—The whole body of an individual injected with Thorotrast 36 y prior to her death was analyzed for  $^{232}\text{Th}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ , and  $^{230}\text{Th}$ . Measurement of these isotopes in all tissues of the body will provide data necessary to calculate the radiation dose to individual tissues and to evaluate the risk potential associated with deposition of thorium and progeny in humans. The tissues were ashed, dissolved in acid, and the thorium isolated by ion exchange and electrodeposition. The thorium activity was measured by alpha spectrometry. The  $^{228}\text{Ra}$  was determined by measuring the 0.991-MeV gamma rays associated with decay of the  $^{228}\text{Ac}$  daughter. It was estimated that almost all of the  $^{232}\text{Th}$  from the original injection was retained in the body, mostly in the tissues of the reticuloendothelial system. A total of 28 kBq (0.76  $\mu\text{Ci}$ ) of  $^{232}\text{Th}$  was measured in the soft tissues and bones. The body also contained 13 kBq  $^{228}\text{Ra}$ , 12 kBq  $^{228}\text{Th}$ , and 3.9 kBq  $^{230}\text{Th}$ . A Thorotrastoma contained about 3.5% of the total activity. Excluding the Thorotrastoma, approximately 45% of all the activity ( $^{232}\text{Th}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ , and  $^{230}\text{Th}$ ) was retained in the liver, 13% in the spleen, 2% in muscle, 1% in skin, slightly less than 1% in the respiratory tract, 4% in all other soft tissues, and 33% in the skeleton (bone and bone marrow). Sixty to 80% of the thorium activity in bones containing red marrow was located in the marrow. Bones containing yellow marrow had less than 40% of the thorium activity in the marrow. Highest concentrations were found in the hepatic and other abdominal lymph nodes, spleen, hilar lymph nodes, liver, trachea, and bone. Approximately 60% of the  $^{228}\text{Ra}$  formed from the decay of the  $^{232}\text{Th}$  had been excreted from the body. The  $^{228}\text{Ra}$  and  $^{228}\text{Th}$  were in approximate equilibrium throughout the body.

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Key words: Thorotrast; thorium;  $^{228}\text{Ra}$ ; leukemia

### INTRODUCTION

THE FIRST studies to measure the activity concentrations of  $^{232}\text{Th}$  and daughters in organs and tissues of Thorotrast patients were carried out in Germany between 1946 and 1949 by Schaefer at the Kaiser Wilhelm Institute in Frankfurt, Germany (Muth 1989). Many studies since have analyzed selected tissues taken at

autopsy in an effort to characterize the retention and distribution of these radionuclides (Rotblat and Ward 1953; Hursh et al. 1957; Goldin et al. 1972; Lucas et al. 1972; Dudley 1978). In these studies, only major deposition sites, liver, spleen, bone, and bone marrow were most typically collected and analyzed. This study, to the best of our knowledge, is the first time the distribution of  $^{232}\text{Th}$  and daughters has been measured in all soft tissues and bones of the whole body.

### MATERIALS AND METHODS

An autopsy was performed on U.S. Uranium Registry (USUR) Case 1001 within 2 h following death on 8 June 1989. Details of the autopsy and pathological findings are given by Graham et al. (1992). To measure the short-lived daughters of  $^{232}\text{Th}$  in various organs and tissues of interest, the pathology team had been alerted of the necessity to begin the autopsy as soon as possible after death. Tissues of special interest were processed for immediate gamma counting at the National Institute of Health (NIH) and the National Institute of Standards and Technology (NIST). The liver, spleen, pancreas, kidneys, esophagus, larynx, breasts (all of one breast and approximately one-half of the other), eyes, blood, and a rib were immediately removed and weighed. A subsample of approximately 200 g was removed from the whole liver for this part of the study. All tissues except the eyes and the rib were separately homogenized in a Waring blender, reweighed, and placed in Teflon<sup>®</sup> jars for counting. Liver, kidney, and breast were divided into roughly equal portions for simultaneous measurement at NIH and NIST. The eyes were placed in a jar and measured intact. The rib was cut into several small sections and placed in a Teflon jar. The blood sample was centrifuged and the plasma decanted. The red blood cell fraction and the plasma was each placed in separate Teflon jars. Details and results of the gamma counting of these samples are given by Mays et al. (1992).

From this point on, the body was handled in accordance with the standard USUR protocol for whole-body donations (Breitenstein 1981; Kathren

<sup>®</sup>Teflon, E. I. duPont de Nemours & Co. Inc., 1007 Market Street, Wilmington, DE 19898.

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1989). All remaining organs, including the brain, were weighed, individually packaged in plastic bags, and placed in Styrofoam<sup>†</sup> shipping containers. Dry ice was added and the specimens were shipped by air freight to Los Alamos National Laboratory (LANL) for analysis.

The eviscerated body was packed in ice and sent by air freight to the USUR facilities in Richland, WA. The body was gamma-counted in the whole-body counter at the Pacific Northwest Laboratory In-Vivo Radioassay Research Laboratory to determine the external radiation distribution (Mays et al. 1992).

Following these measurements, the remaining soft tissues and bones were separated, weighed, individually packaged, frozen, and sent to LANL for radiochemical analysis. The protocol used for the dissection was developed by the USTR/USUR and is explained in detail by McInroy et al. (1985). Briefly, the skin, including adipose tissue, was removed in sections from general anatomic areas, i.e., the head, upper arm, forearm, hand, upper thorax (front and back), trunk (front and back), thigh, calf, and foot. Each section was immediately weighed, individually packaged, and placed in a freezer. Similarly, the muscle was removed from the same anatomic areas, weighed, individually packaged, and frozen.

The entire skeleton was then disarticulated. The bones were freed of as much of the remaining soft tissue as possible and weighed whole. The following subdivisions of bones were made with a bone saw: The odd-numbered vertebrae (except for the atlas) were divided into bodies and arches; the long bones of the right side were divided into ends and shafts; the skull was divided into left and right halves; and the right half of the skull was further divided into facial and cranial portions. The wet weights of parts of the divided bones were measured, and the pieces were individually packaged and frozen. All samples were packed in insulated containers with dry ice and shipped to Los Alamos by air freight. The specimens were stored in a freezer until analyzed.

Marrow was separated from selected bones containing red or yellow marrow. Two vertebral bodies (thoracic-10 and lumbar-4) were cut into sections 4–5-mm-thick using a small band saw. Likewise, sections were cut from the left ilium. The left fibula was first divided into the proximal end, proximal shaft, distal shaft, and distal end. The left rib-8 and each section of the fibula was cut longitudinally to expose the marrow cavity. No red marrow was apparent in any of the fibula sections. Each section was rinsed with a stream of distilled water to remove adherent bone dust from the cutting, blotted dry, and weighed. The marrow was removed from the bone marrow cavities by the forceful impingement of a stream of distilled water (Goldin et al. 1972; McInroy and Kathren 1990). When no remaining marrow was visible, a jet of compressed air was blown through the bone to remove excess water, the bone was blotted dry and reweighed. As reported

by Goldin et al. (1972), the empty trabecular honeycomb-like cells that held the marrow were clearly visible after the cleaning. The marrow weight was determined by the weight difference. No calcium measurements were made on the marrow fraction to determine if any bone fragments were removed along with the marrow.

Although this procedure for removing marrow has been reported in the literature for many years, it should be noted that there is no guarantee that the fraction removed contains only marrow. As mentioned, bone fragments may be also washed out of the marrow cavities and, although we think it unlikely, unknown quantities of other organic material such as the endosteum may be removed along with the marrow. We know of no reported study that has examined this specific problem.

#### Analytical procedure

Soft tissues, organs, and bones were either analyzed whole or subdivided for convenience of analyses as well as to ascertain possible differences in radionuclide distribution or variation throughout the larger specimens (McInroy et al. 1985). The basic radiochemical procedures were detailed by Gonzales (1990) and Boyd et al. (1987). Briefly, specimens were oven-dried, alternately dry- and wet-ashed until all visible carbonaceous material was destroyed, and dissolved in 8 M HCl. (Water was added to some samples to expedite dissolution. No attempt was made to determine if colloid formation occurred.) An appropriate aliquot was selected,  $^{234}\text{Th}$  tracer was added, and the sample was heated to dryness and redissolved in the minimum amount of 8-M  $\text{HNO}_3$  to obtain a concentration not to exceed 25 mg ash cm<sup>-3</sup>.

The solution was passed through an ion exchange column containing Bio-Rad anion exchange resin<sup>‡</sup> (AG 1-X4; 100–200 mesh). The thorium retained on the resin was eluted with 9 M HCl-Cl<sub>2</sub>. The eluate was evaporated to dryness, 9 M H<sub>2</sub>SO<sub>4</sub> was added, and the sample was heated until the H<sub>2</sub>SO<sub>4</sub> fumed. The pH was adjusted to 2.3, using thymol-blue indicator and NH<sub>3</sub> gas, and the solution was transferred to an electrodeposition cell. The thorium was electroplated for 2 h at 0.55 A onto a 1.27-cm-diameter electropolished stainless steel disk using a modification of Talvitie's procedure (Talvitie 1972). The disk was counted in an alpha-pulse height-detector system using a 300 mm<sup>2</sup> Si surface-barrier detector. The background for each detector was measured for 70,000 s and compared to a running average of 10 consecutive backgrounds measured prior to the current measurement. If the measurement fell outside the control limits (three standard deviations of the average efficiency), the detector chamber was cleaned and a new background determined. If the background remained outside the control limits, the detector was replaced. The average of the mean backgrounds in the 16 detectors was  $1.3 \pm 0.7$ ,  $7.8 \pm 1.9$ , and  $2.6 \pm 0.6$

<sup>†</sup> Styrofoam, Dow Chemical Company, 3020 Willard, N. Low Center, Midland, MI 48674.

<sup>‡</sup> Analytical Grade Anion Exchange Resin, Bio-Rad Laboratory, 414 Harbour Way, Richmond, CA 94804.

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counts in 70,000 s for  $^{232}\text{Th}$ ,  $^{234}\text{Th}$ , and  $^{230}\text{Th}$ , respectively. The background average was updated weekly.

The efficiency of each detector was determined weekly by a 1,020-s count using a secondary standard of  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ , and  $^{242}\text{Pu}$ . This secondary standard was periodically calibrated with a NIST  $^{239}\text{Pu}$  standard using a gas-flow proportional counter. The result for each detector was compared with a running average efficiency that represented at least 10 determinations. If the detector efficiency fell outside the control limits (three standard deviations of the average efficiency), the detector was replaced. The detector efficiency was updated weekly. An estimate of the minimum detectable activity (MDA) was made using the methods of Currie (1968, 1984). The MDA was based on the instrument background counts (in the energy areas of the thorium isotopes) obtained from the analysis of chemical reagent blank-quality control samples, the percent tracer recovery, and the counting efficiency of the system. The MDA for a sample was approximately 1 mBq ( $0.08$  disintegrations  $\text{min}^{-1}$ ) per sample aliquot at the 0.05 confidence level. The calculation assumes a mean recovery of the tracer of 60%, a mean background of three counts, a 70,000-s count length, and a counting efficiency of 20%. Any sample with a tracer recovery less than 60% was reanalyzed.

Two thorium isotopes were evaluated for use as a tracer in these analyses.  $^{232}\text{Th}$ , an alpha emitter, decays with several energies, producing a broad spectrum that was judged unacceptable for these measurements because of the resolution of our alpha detectors. The alpha energies (4.8–5.0 MeV) were too close to the 4.7-MeV alpha energy of  $^{230}\text{Th}$ , one of the thorium isotopes of interest in this study, for the computer code used to integrate the counts under each peak satisfactorily. Therefore,  $^{234}\text{Th}$ , a beta-emitting isotope with a 24.1-d half-life, was used. Approximately 33 Bq ( $2,000$  disintegrations  $\text{min}^{-1}$ ) of  $^{234}\text{Th}$  were added to the dissolved sample at the time of aliquoting. The samples were run through the ion exchange and electroplated as soon as possible because of the short half-life of the tracer. Chemical losses were corrected relative to the recovery of the added  $^{234}\text{Th}$  tracer as determined on the basis of total beta counts using a gas-flow proportional counter. Thorium was separated from unspiked (no tracer added) tissue solutions and counted for beta activity. The indigenous beta activity accounted for less than 5% of the activity added as a tracer but, consequently, could have increased the apparent chemical recovery of the sample by this amount. This adds about 5% to the total uncertainty of the thorium measurements. Only sample aliquots that contained  $<0.1$  Bq ( $\sim 25$   $\mu\text{g}$ )  $^{232}\text{Th}$  were used because, at higher activities, the mass of the  $^{232}\text{Th}$  attenuated the emitted alpha particles to a different degree than the beta particles on the size plates used, thus negating the validity of  $^{234}\text{Th}$  as a tracer.

Reagent blanks were spiked with  $^{234}\text{Th}$  and processed through each step of the analytical procedure with every set of samples analyzed. The average  $^{232}\text{Th}$  back-

ground ( $n = 13$ ) was  $0.25$  mBq ( $\sigma = 0.23$  mBq);  $^{234}\text{Th} = 0.27$  mBq ( $\sigma = 0.38$  mBq); and  $^{230}\text{Th} = 0.23$  mBq ( $\sigma = 0.23$  mBq). Twenty-six reagent quality-control samples spiked with  $^{232}\text{Th}$  were also analyzed concurrently with the tissue samples. The average recovery of the  $^{232}\text{Th}$  was 104% ( $\sigma = 7.5\%$ ). The average recovery of the  $^{234}\text{Th}$  tracer in 310 tissue samples was 83% ( $\sigma = 26.5\%$ ). Any sample with a tracer recovery less than 60% was reanalyzed.

The  $^{226}\text{Ra}$  was determined by measuring the 0.911-MeV gamma rays emitted during the decay of the  $^{226}\text{Ac}$  daughter. Aliquots of the dissolved tissue between 100  $\text{cm}^3$  and 500  $\text{cm}^3$ , in increments of 50  $\text{cm}^3$ , were placed in polyethylene bottles. The counting system consisted of a 10.2-cm  $\times$  15.2-cm ( $4'' \times 6''$ ) Bicon NaI(Tl) well counter<sup>1</sup> coupled to a 7.6-cm ( $3''$ ) photomultiplier tube with a 1.9-cm (0.75 in.) quartz optical coupler. The energy calibration was determined using  $^{137}\text{Cs}$ - $^{60}\text{Co}$  sources. Standard solutions of  $^{226}\text{Ra}$  in 8 M HCl, prepared from NIST SRM 4339<sup>2</sup>, were used to determine the counting efficiencies for the sample volumes used. Two quality-control samples, also prepared from NIST SRM 4339 in 8 M HCl, were measured with each set of 18 samples. All quality-control samples, backgrounds, and most of the samples were counted for 7,200 s. A few samples that were expected to contain levels of activity near the detection limit of 1.2 Bq ( $70$  disintegrations  $\text{min}^{-1}$ ) were counted for 50,000 s. Forty-six quality-control standards containing 2.9 Bq ( $175$  disintegrations  $\text{min}^{-1}$ ) or higher were counted with an accuracy of  $\pm 10\%$ . Three quality-control standards were counted with an accuracy of  $\pm 15\%$ . All samples were analyzed over a 13-mo time period. No calculations were made to correct the results back to the time of death. Thirty-eight samples were recounted at the end of the study. Two samples had measured activity statistically higher than the initial counts. This phenomenon is being investigated and will be reported in a later paper. In addition, no samples were overspiked with  $^{226}\text{Ra}$  to determine if there was any attenuation of the counts due to the sample matrix, i.e., bone samples vs. soft tissue. This is also being investigated and will be reported at a later date.

The propagation of error associated with counting statistics, addition of tracer, background, and counting efficiency variations in the thorium measurements averaged 3–5% of the measured activity. The overall uncertainty of the thorium measurements was about 10–15%. The radium measurements also had an uncertainty ranging from 10–15% of the measured activity. Thus, with a lower limit of detection of 1 mBq and an uncertainty of 10–15%, the concentration of thorium may be measured in a small sample with reasonable accuracy. However, the radium procedure, with a

<sup>1</sup>Bicon 5"  $\times$  4" NaI(Tl) Well Counter, Bicon Corporation, 12345 Kinsman Road, Newbury, OH 44065.

<sup>2</sup> $^{226}\text{Ra}$  Standard Solution, NIST SRM 4339, National Institute of Standards and Technology, Quince Orchard and Clopper Roads, Gaithersburg, MD 20899.

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Lower limit of detection of 1.2 Bq (1,200 times higher than the thorium) and an uncertainty of 10–15%, was unable to determine the radium (actinium) content in some of the smaller samples.

Four soft tissues and two bones were selected for confirmatory analysis for thorium. Liver, spleen, stomach, skin, ilium, and the distal shaft of the radius were picked as representative of samples having a considerable range of activity and were among those most difficult to analyze. Solutions containing the dissolved samples were sent to another laboratory at LANL for measurement using inductively coupled plasma-mass spectrometry (ICP-MS). The mass measurements obtained by ICP-MS were converted to activities using the specific activity of <sup>232</sup>Th (4.11 kBq g<sup>-1</sup>).

RESULTS

The results of the interlaboratory comparison of sample analyses using alpha spectrometry and ICP-MS are shown in Table 1. The results agreed to within 10% for the liver, spleen, stomach, and skin. The ICP-MS analysis for the ilium was about 15% lower than the alpha spectrometry measurement. The calcium concentration of the radius shaft interfered with the ICP-MS analysis. At the dilution required to prevent the calcium salts from plugging the orifice of the instrument, the concentration of thorium was below the detection limit. However, the overall results gave confidence in the accuracy of the radiochemical procedures used.

A total of approximately 28 kBq (0.76 μCi) of <sup>232</sup>Th was measured in the tissues of USUR Case 1001 some 36 y following the initial injection of Thorotrast in this individual. This is almost 20% more than the 23 kBq estimated to have been injected in 1953 (Mays et al. 1992) but may be reasonable in light of the lack of documentation as to the actual amount administered and the uncertainties associated with our measurements. The biokinetic modeling and dosimetry resulting from the injection of Thorotrast in this individual are discussed in detail by Kathren and Hill (1992).

Table 1. Comparison of thorium measurements using alpha-pulse-height spectrometry and inductively coupled plasma mass spectrometry (ICP-MS).

	Mass (g)		<sup>232</sup> Th content (Bq kg <sup>-1</sup> ) <sup>a</sup>		Difference (%)
	Wet	Ash	Alpha-spec	ICP-MS	
Liver	1,033		12,200	12,800	5.2
Spleen	25		144,000	144,000	0.0
Stomach	196		220	241	9.5
Skin, head	585		100	107	6.8
Ilium crest	27	13	11,400	9,727	14.7
Radius, distal shaft	13	6.8	33	<DL <sup>b</sup>	—

<sup>a</sup>Soft tissue sample concentrations per kilogram wet mass; bone samples per kilogram ash.

<sup>b</sup>DL = detection limit.

A 25-cm<sup>3</sup> vial of Thorotrast obtained from the U.S. Transuranium and Uranium Registries was submitted to the Analytical Chemistry Group, Chemical and Laser Sciences Division (LANL) for thorium analysis using thermal ionization mass spectrometry. The vial contained 6.13 g <sup>232</sup>Th (0.68 μCi), 9.5 × 10<sup>-10</sup> g <sup>228</sup>Th (0.78 μCi), and 2.1 × 10<sup>-5</sup> g <sup>230</sup>Th (0.43 μCi). The estimated reported accuracy of the analysis was 20% at 1σ.

Following is a brief summary of the analytical results.

Table 2. Distribution of thorium isotopes and <sup>228</sup>Ra in the whole body of a Thorotrast patient 36 y postinjection.

Tissue	Thorium and <sup>228</sup> Ra content (Bq)			
	<sup>232</sup> Th	<sup>228</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th
Respiratory tract <sup>a</sup>	232	89	82	29
Liver	12,433	5,727	5,392	1,839
Kidneys	11	2	5	2
Spleen	3,607	1,521	1,481	443
Smooth muscle organs <sup>b</sup>	114	44	43	13
Striated muscle	453	199	162	54
Other muscle <sup>c</sup>	14	5	5	2
Skin (including adipose tissue)	362	132	122	46
Other soft tissues <sup>d</sup>	832	522	438	108
Bones and teeth	9,568	4,253	3,954	1,265
Total	27,626	12,494	11,683	3,800
Thorotrastoma	994	415	390	118
Total body	28,620	12,909	12,073	3,918

<sup>a</sup>Lung, trachea, larynx, hilar, and peritracheal lymph nodes.

<sup>b</sup>Stomach, small intestine, large intestine, and urinary bladder.

<sup>c</sup>Heart, tongue, and diaphragm.

<sup>d</sup>Adrenals, pericardium, pancreas, gall bladder, bile, mesentery, epidura, brain, hair, eyes, breast, thyroid, pituitary, abdominal lymph nodes, aorta, carotid artery, and esophagus.

Table 3. Wet weight, thorium isotopes, and <sup>228</sup>Ra content in dissected Thorotrastoma and carotid artery.

Tissue	Wet weight (g)	Sample content (Bq)				Total
		<sup>232</sup> Th	<sup>228</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th	
Carotid artery	7.8	106	44	41	14	205
Carotid artery and Thorotrastoma	32.4	770	317	297	90	1,474
Thorotrastoma	12.4	118	54	52	14	238
Total	52.6	994	415	390	118	1,917

Table 4. Activity ratios of thorium isotopes and <sup>228</sup>Ra in dissected Thorotrastoma and carotid artery.

Tissue	<sup>228</sup> Th: <sup>232</sup> Th	<sup>228</sup> Ra: <sup>232</sup> Th	<sup>228</sup> Th: <sup>228</sup> Ra	<sup>230</sup> Th: <sup>232</sup> Th
Carotid artery	0.39	0.42	0.93	0.13
Carotid artery and Thorotrastoma	0.39	0.41	0.94	0.12
Thorotrastoma	0.44	0.46	0.96	0.12

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Table 5. Weights and content of thorium isotopes in bone and marrow.

	Wet weight (g)	Thorium content (Bq)			Ratio (sample content:total content) <sup>a</sup>		
		<sup>232</sup> Th	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th	<sup>228</sup> Th	<sup>230</sup> Th
<i>Vertebrae</i>							
Thoracic-10, body marrow (red)	2.9	36	13	4	0.80	0.75	0.73
bone	3.5	9	4	1	0.20	0.25	0.27
Lumbar-4, body marrow (red)	4.1	55	23	7	0.79	0.78	0.80
bone	6.3	14	7	2	0.21	0.22	0.20
Rib-8, left marrow (red)	2.0	18	8	2	0.74	0.72	0.45
bone	14.3	6	3	3	0.26	0.28	0.55
Ilium, left marrow (red)	11.9	120	49	13	0.68	0.63	0.63
bone	25.1	57	28	8	0.32	0.37	0.37
<i>Fibula-left</i>							
Proximal end marrow (yellow)	3.0	0.24	0.08	0.05	0.34	0.10	0.45
bone	10.5	0.46	0.71	0.06	0.66	0.90	0.55
Proximal shaft marrow (yellow)	0.7	0.28	0.11	0.04	0.37	0.10	0.32
bone	13.5	0.48	1.03	0.08	0.63	0.90	0.68
Distal shaft marrow (yellow)	1.5	0.10	0.05	0.02	0.22	0.06	0.22
bone	12.3	0.33	0.74	0.06	0.77	0.94	0.78
Distal end marrow (yellow)	1.8	0.09	0.06	0.01	0.16	0.07	0.14
bone	7.5	0.46	0.82	0.06	0.83	0.93	0.86

<sup>a</sup> Ratios may contain small differences due to rounding effects.

**Thorium isotope and radium contents in soft tissue**

The thorium and radium contents in various soft tissues and organs are shown in Table 2. Complete data, including organ weights, are given in Tables A-1 through A-3 in Appendix A. All concentrations for soft tissues are expressed in units of activity per kilogram of wet tissue analyzed (Bq kg<sup>-1</sup>). As reported by others, most of the <sup>232</sup>Th and daughters were retained in the reticuloendothelial system (RES) (Goldin et al. 1972; Kaul and Noffz 1978; van Kaick 1984). Highest concentrations were found in the hepatic and other abdominal lymph nodes, spleen, hilar lymph nodes, liver, and trachea. Approximately 45% of the retained <sup>232</sup>Th, <sup>228</sup>Ra, <sup>228</sup>Th, and <sup>230</sup>Th was found in the liver, 13% in the spleen, 2% in muscle, 1% in skin, slightly less than 1% in the respiratory tract, and 4% in all other soft tissues combined.

The activity ratio of <sup>228</sup>Ra to its parent <sup>232</sup>Th in soft tissues averaged about 0.40 (with a range of 0.20-4.37), indicating a state of disequilibrium in which approximately 60% of the <sup>228</sup>Ra had been excreted or translocated. A few tissues, such as the cerebrum and breast, had activity ratios >1, suggesting a preferential

uptake and retention of <sup>228</sup>Ra. The <sup>228</sup>Ra and <sup>232</sup>Th are close to being in equilibrium in the soft tissues with notable exceptions being the lungs, small intestine, and many of the muscle and skin specimens. Since the sensitivity of the analytical procedure was much greater for measuring thorium than for radium, the uncertainties associated with radium measurements make conclusions concerning equilibria of any of the isotopes with radium tenuous.

As reported by Graham et al. (1992), a Thorotrastoma was excised from the vicinity of the right carotid artery. A portion of the artery was removed with the adherent sample. The specimen was further subdivided at Los Alamos into three samples: the carotid artery, a well-defined piece of the Thorotrastoma, and a section of the artery fused to the adjacent Thorotrastoma. Each sample was analyzed for thorium and radium (Table 3). Highest concentrations were measured in the fused carotid artery-Thorotrastoma specimen, suggesting that this may have been the location of the point of injection. The calcified Thorotrastoma sample had the lowest concentration. The total activity measured was

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994 Bq of  $^{232}\text{Th}$ , 415 Bq of  $^{226}\text{Ra}$ , 390 Bq of  $^{228}\text{Th}$ , and 118 Bq of  $^{230}\text{Th}$ .

Isotopic activity ratios (Table 4) in the Thorotrastoma specimens are similar to those previously described for other soft tissues. Approximately 60% of the  $^{226}\text{Ra}$  associated with these tissues had been translocated or excreted and the  $^{226}\text{Ra}$  and  $^{228}\text{Th}$  were very close to equilibrium.

#### Thorium and radium analyses in bone

Thorium isotope concentrations in mineral bone and red and yellow bone marrow are found in Table 5. For those bones analyzed, 75–80% of the total thorium was in the red marrow of the vertebra and rib, 62–68% of all thorium was in the red marrow of the pelvis, and 16–37% of the  $^{232}\text{Th}$  was in the yellow marrow of the fibula. Ratios varied for the other isotopes (6–10% for  $^{226}\text{Th}$ ; 15–45% for  $^{230}\text{Th}$ ). The ratio of  $^{226}\text{Th}$ :  $^{232}\text{Th}$  activity in all marrow was similar to other soft tissues, ranging from 0.32–0.68 in yellow marrow and 0.36–0.44 in red marrow. Mineral bone from bones containing red marrow also had ratios <1 (0.44–0.50). However, bones containing yellow marrow had ratios ranging from 1.5–2.2, indicating an enhanced retention of  $^{226}\text{Ra}$ . For a more-detailed discussion of the isotopic ratios, distribution, retention, and excretion, see the report by Kathren and Hill (1992).

Wet weights, ash weights, thorium, and radium concentrations in the whole bones measured are shown in Appendix B. Thirty-four percent of the total whole-body content of thorium and  $^{226}\text{Ra}$  (excluding the Thorotrastoma) was found in the skeleton (Table 2). Highest concentrations were found in bones containing red marrow. Lowest concentrations were in bones with small marrow cavities or containing chiefly yellow marrow. We were unable to measure  $^{226}\text{Ra}$  concentrations in many of the smaller bone specimens. Isotopic activity ratios of  $^{226}\text{Ra}$ :  $^{232}\text{Th}$  were generally highest in the cortical bone and often times exceeded unity, indicating a preferential retention of the  $^{226}\text{Ra}$ .

#### SUMMARY

The whole body of an individual injected with Thorotrast 36 y prior to death was analyzed for  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Th}$ , and  $^{230}\text{Th}$ . From the available information concerning the amount of Thorotrast likely to have been injected into this individual (Mays et al. 1992), it was estimated that almost all of the  $^{232}\text{Th}$  was retained in the body, mostly in the reticuloendothelial system. A total of 28 kBq (0.76  $\mu\text{Ci}$ ) of  $^{232}\text{Th}$  was measured in the soft tissues and bones. The body also contained 13 kBq  $^{226}\text{Ra}$ , 12 kBq  $^{228}\text{Th}$ , and 3.9 kBq  $^{230}\text{Th}$ . A Thorotrastoma contained about 3.5% of the total activity. Excluding the Thorotrastoma, approximately 45% of all the activity was retained in the liver, 13% in the spleen, 2% in muscle, 1% in skin, slightly less than 1% in the respiratory tract, 4% in all other soft tissues, and 33% in the skeleton (including bone marrow). Sixty to

80% of the thorium activity in bones containing red marrow was located in the marrow. Bones containing yellow marrow had less than 40% of the thorium activity in the marrow. Highest concentrations were found in the hepatic and other abdominal lymph nodes, spleen, hilar lymph nodes, liver, trachea, and bone. Sixty percent of the  $^{226}\text{Ra}$  formed from the decay of the  $^{232}\text{Th}$  had been excreted from the body. The  $^{226}\text{Ra}$  and  $^{228}\text{Th}$  were in approximate equilibrium throughout the body.

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APPENDIX A  
 Tables of Data from the Analyses of Individual Soft Tissues and Organs  
 Table A-1. Wet weights, thorium isotopes, and <sup>228</sup>Ra concentrations of organs and specialized tissues.

Tissue	Wet weight (g)	Tissue concentrations (Bq kg <sup>-1</sup> )			
		<sup>232</sup> Th	<sup>228</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th
Lung	201	248	112	94	38
-left	364	181	79	66	20
-right	25	3,040	1,059	992	359
Trachea	25	483	*	164	59
Larynx	25				
Lymph nodes, pulmonary			2,260	1,745	624
-peritracheal	1.5	4,969		4,709	1,608
-hilar	1.7	13,540	5,037	5,218	1,780
Liver	1,033	12,000	5,543		
Kidney			18	19	4.8
-left	136	42	*	20	7.4
-right	122	47	*		
Spleen	25	143,900	60,670	59,090	17,670
Heart	283	17	5.6	5.5	2.0
Pericardium	38	57	*	22	5.9
Stomach	196	216	83	91	21
Intestine					
-small	570	76	32	24	9.8
-large	387	67	25	26	7.7
Urinary bladder	77	31	*	12	2.2
Adrenal, right	3.4	947	*	308	131
Pancreas	52	1,183	339	415	160
Gall bladder	13	122	*	42	14
Bile	58	0.06	*	0.18	0.02
Mesentery	249	40	20	16	5.4
Epidura	23	87	*	40	12
Cerebrum	783	1.2	5.5	0.80	0.14
Cerebellum	106	2.0	*	0.81	0.24
Eyes	18	6.6	*	6.9	0.82
Breast			*	9.7	3.9
-right	57	28	*	7.2	2.3
-left and some right	274	18	187		
Thyroid	5.2	1,226	*	466	128
Pituitary	0.4	422	*	160	50
Lymph nodes					
-hepatic	2.8	238,100	132,200	134,300	30,950
-aortic arch	0.9	1,906	*	662	228
-aorta, descend	0.5	59,900	25,130	22,960	7,622
-carotid, left	0.6	1,827	*	745	213
-mesenteric	20	989	376	339	95
-iliac, right	1.7	2,220	*	803	257
-iliac, left	1.9	1,525	*	570	185
Aortic arch	24	183	*	82	24
Aorta, descending	20	32	*	47	4.2
Aorta, abdominal	11	52	*	68	7.1
Carotid artery					
-left	5.3	107	*	40	14
Esophagus, proximal	8.8	570	*	201	69
Esophagus, distal	22	516	*	179	71
Blood plasma	104	0.16	*	0.21	0.03
Red blood cells	27	0.50	*	0.24	0.07

\* Activity less than minimum detection level (1.2 Bq per sample).

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Table A-2. Dissected wet weights, thorium isotopes, and radium concentrations in muscle removed in the dissection of the skeleton.

Tissue	Wet weight (g)	Tissue concentrations (Bq kg <sup>-1</sup> )			
		<sup>232</sup> Th	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th
Combined Soft Tissue <sup>a</sup>					
Anatomical region					
Head	400	381	148	139	42
Tongue	78	57	<sup>b</sup>	18	7.0
Arm, upper-left	575	6.8	7.2	2.4	0.7
-right	557	12	8.2	4.0	1.4
lower-left	320	8.7	<sup>b</sup>	3.1	1.1
-right	361	9.6	<sup>b</sup>	2.8	1.2
Hand-left	118	14	<sup>b</sup>	5.5	2.2
-right	102	14	<sup>b</sup>	5.6	1.5
Thorax, front-right (1)	396	151	59	55	16
(2)	206	31	17	11	4.0
-left (1)	360	36	13	12	4.5
(2)	319	28	12	9.3	3.6
back-right (1)	637	19	11	6.5	2.1
(2)	161	22	<sup>b</sup>	7.5	2.2
-left (1)	581	16	11	5.6	2.4
(2)	212	10	<sup>b</sup>	4.0	1.2
Abdomen, front-right (3)	304	21	<sup>b</sup>	7.6	2.3
(4)	556	6.8	5.9	2.5	0.8
-left (3)	277	8.2	<sup>b</sup>	3.0	0.9
(4)	768	16	<sup>b</sup>	4.9	2.1
back-right (3)	399	70	32	27	9.1
(4)	786	27	12	9.3	3.6
-left (3)	454	72	28	25	9.9
(4)	705	16	8.2	6.3	2.5
Thigh-left	2,662	5.2	4.3	1.8	0.6
-right	2,515	5.7	4.9	1.9	0.7
Calf-left	849	6.8	8.0	2.7	0.8
-right	855	6.7	8.2	2.6	0.8
Foot-left	192	12	<sup>b</sup>	4.7	1.3
-right	189	12	<sup>b</sup>	4.7	0.9
Ears	30	34	<sup>b</sup>	21	7.0
Eyes	18	6.6	<sup>b</sup>	6.9	0.8
Diaphragm	75	58	42	22	6.7

<sup>a</sup> Combined skeletal muscle, connective tissue, blood vessels, tendons, and ligaments.

<sup>b</sup> Activity less than minimum detection level (1.2 Bq per sample).

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Thorax, fi

<sup>b</sup>

Abdomen

Thigh-left  
-right

Calf-left  
-right

Foot-left  
-right

<sup>a</sup> Combined  
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Table A-3. Dissected wet weights, thorium isotopes, and radium concentrations in skin removed in the dissection of the skeleton.

Tissue	Wet weight (g)	Tissue concentrations ( $\text{Bq kg}^{-1}$ )			
		$^{232}\text{Th}$	$^{228}\text{Ra}$	$^{230}\text{Th}$	$^{230}\text{Th}$
Combined Soft Tissue*					
Anatomical region					
Head	585	100	41	47	14
Arm, upper-left	640	13	6.1	4.4	1.7
-right	655	28	9.6	8.7	3.7
lower-left	237	30	6.6	8.9	3.9
-right	274	29	9.4	9.9	4.9
Hand-left	100	56	<sup>b</sup>	19	7.3
-right	121	42	<sup>b</sup>	12	5.3
Thorax, front-right (1)	570	48	18	16	9.2
(2)	194	16	<sup>b</sup>	4.9	2.0
-left (1)	138	30	<sup>b</sup>	15	2.9
(2)	471	34	13	11	4.6
back-right (1)	465	144	48	50	13
(2)	604	8.3	4.3	2.8	1.2
-left (1)	306	26	11	10	3.0
(2)	281	17	<sup>b</sup>	5	2.7
Abdomen, front-right (3)	185	8.7	<sup>b</sup>	3.1	1.3
(4)	436	32	13	14	5.2
-left (3)	524	14	6.2	4.6	1.6
(4)	709	28	10	8.3	3.3
back-right (3)	531	10	4.8	3.6	1.1
(4)	662	8.5	<sup>b</sup>	2.9	1.1
-left (3)	466	7.2	<sup>b</sup>	2.4	1.0
(4)	980	16	<sup>b</sup>	5.3	2.0
Thigh-left	2,115	11	2.2	3.6	1.3
-right	2,215	10	2.1	3.4	1.3
Calf-left	648	15	7.9	4.8	1.7
-right	637	15	6.6	4.6	1.9
Foot-left	249	28	11	10	3.3
-right	217	37	18	11	4.4

\* Combined skin and adipose tissue.

<sup>b</sup> Activity less than minimum detectable level (1.2 Bq per sample).

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## APPENDIX B

## Tables of Data from the Analysis of Bones and Parts of Bones

Table B-1. Weights, thorium isotopes, and radium concentrations in bones of the head.

	Weight (g)		Bone concentrations (Bq kg <sup>-1</sup> ash)			
	Wet	Ash	<sup>232</sup> Th	<sup>228</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th
Cranium	914					
Right half	474					
Left half	440					
Occipital	105	54.7	501	310	321	66
Parietal-1	50	28.0	590	302	334	76
Parietal-2	58	32.9	629	352	348	82
Frontal-1	46	25.7	254	205	234	34
Frontal-2	21	10.8	328	251	235	37
Frontal-3	23	11.0	611	297	400	90
Temporal-1	18	8.6	636	374	380	78
Temporal-2	58	21.7	2,486	1,039	934	261
Temporal-3	10	4.3	87	*	137	12
Maxilla	41	10.3	580	494	479	64
Mandible	108 <sup>b</sup>					
Right	34	17.4	452	*	395	50
Left	56 <sup>b</sup>					
Hyoid bone	3	0.70	7,152	*	3,110	990
Teeth						
Lower right						
Incisor-1	0.81	0.58	22	*	91	3.4
Incisor-2	1.21	0.82	29	*	89	11
Canine	0.61	0.34	27	*	111	4.2
Premolar-1	not analyzed <sup>c</sup>					
Premolar-2	1.71	1.13	35	*	107	3.8
Molar-1	1.04	0.73	20	*	78	2.8
Molar-2	1.55	1.07	33	*	119	5.4
Upper right						
Incisor-1	1.39	0.95		*		
Incisor-2	0.77	0.54		*		
Canine	1.54	1.00	22	*	101	2.9
Premolar-1	1.41	0.98		*		
Premolar-2	2.67	2.29	8.4	*	36	3.4
Molar-1	4.48	2.95	17	*	70	3.0
Molar-2	2.14	1.40	31	*	123	6.9

\* Activity less than minimum detection level (1.2 Bq per sample).

<sup>b</sup> Includes teeth.<sup>c</sup> Retained by pathologist.

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**Table B-2.** Weights, thorium isotopes, and radium concentrations in bones and parts of bones of the spine and pelvis.

	Weight (g)		Bone concentrations (Bq g <sup>-1</sup> ash)			
	Wet	Ash	$^{232}\text{Th}$	$^{228}\text{Ra}$	$^{230}\text{Th}$	$^{230}\text{Th}$
Vertebral column						
Cervical-1	16	5.59	7.7	2.2	2.9	1.0
Cervical-2	22					
Cervical-3	16					
arch	8.4	2.95	6.7	2.8	2.5	0.8
body	6.9	2.25	10.2	4.0	4.1	1.4
Cervical-4	19					
Cervical-5	20					
arch	10.6	3.00	10.5	4.5	4.6	1.4
body	9.1	3.20	17.2	2.4	5.9	2.1
Cervical-6	20					
Cervical-7	23					
arch	13.2	4.67	7.4	3.0	3.0	1.1
body	9.3	2.29	16.7	7.1	6.7	2.3
Thoracic-1	28					
arch	15.9	5.36	8.6	3.8	3.5	1.3
body	11.5	2.48	22.9	8.3	8.0	2.5
Thoracic-2	29					
Thoracic-3	26					
arch	13.9	4.34	9.9	4.3	4.0	1.2
body	11.6	2.34	31.4	13.1	11.3	3.6
Thoracic-4	26					
Thoracic-5	30					
arch	14.0	4.45	9.1	4.2	3.7	1.2
body	15.5	3.14	23.8	9.2	8.2	2.7
Thoracic-6	34					
Thoracic-7	36					
arch	14.6	4.64	10.3	4.0	4.2	1.0
body	21.1	4.13	27.0	10.2	10.6	4.4
Thoracic-8	42					
Thoracic-9	44					
arch	22.4	6.42	10.5	4.7	3.7	1.4
body	21.0	3.70	30.9	12.5	13.2	4.6
Thoracic-10	50					
Thoracic-11	*					
arch	28.1	7.87	10.7	4.5	4.0	1.5
body	*					
Thoracic-12	51					
Lumbar-1	66					
arch	26.2	8.60	6.4	2.4	2.6	1.0
body	39.0	9.81	8.8	11.7	3.1	1.2
Lumbar-2	77					
Lumbar-3	80					
arch	29.5	10.4	5.5	2.4	2.2	0.7
body	48.7	12.2	25.6	9.5	10.6	2.9
Lumbar-4	100					
Sacrum + Lumbar-5 <sup>b</sup>	232	45.9	15.1	6.6	6.2	2.0
Coccyx	12	1.7	1.4	<sup>c</sup>	0.8	0.2
Pelvis						
left	422					
right	426					
Ischium	159.0	45.8	9.7	4.4	4.1	1.1
Ilium						
crest	27.4	13.1	11.4	4.1	4.5	1.1
body	214.2	61.8	11.8	5.5	4.8	1.9

\* Thoracic vertebrae-11 vertebral body retained by pathologist.

<sup>b</sup> Lumbar vertebrae-5 fused to sacrum.<sup>c</sup> Activity less than minimum detection level (1.2 Bq per sample).

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**Table B-3.** Weights, thorium isotopes, and radium concentrations of the analyzed bones and parts of bones of the shoulder and rib cage.

	Weight (g)		Thorium and radium concentration (Bq g <sup>-1</sup> ash)			
	Wet	Ash	<sup>232</sup> Th	<sup>228</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th
Clavicle						
Left	23					
Right	33					
Sternal end (SE)	10.0	2.60	2.1	1.0	1.1	0.32
Shaft	12.7	5.87	1.5	0.30	0.64	0.16
Acromial end	10.3	1.98	10.5	4.3	4.2	1.2
Scapula						
Left	85					
Right	86					
Proximal end (PE)	17.7	5.84	1.0	0.57	0.74	0.09
Spine	38.2	13.87	4.7	2.7	1.9	0.85
Distal end (DE)	23.2	9.99	4.3	1.7	2.0	0.55
Ribs						
1-Left	18.2					
Right	18.2	5.00	7.3	3.3	2.8	0.93
2-Left	15.8					
Right	15.7	4.63	7.5	3.3	3.1	1.0
3-Left	18.5					
Right	17.7	5.06	9.5	3.7	4.0	1.3
4-Left	23.5					
Right	21.9	6.34	7.7	3.3	3.8	1.0
5-Left	25.6					
Right	30.4	8.59	7.9	3.5	3.0	0.87
6-Left	34.2					
Right	41.9	11.22	6.3	3.1	2.4	0.70
7-Left	13.1					
Right	34.7	9.24	6.2	3.0	2.4	0.72
8-Left	16.3					
Right	22.9	7.52	8.2	3.2	2.7	1.1
9-Left	16.6					
Right	17.2	5.66	7.2	3.2	2.7	0.95
10-Left	10.2					
Right	11.4	6.50	4.2	1.7	1.4	0.72
11-Left	27.4 <sup>a</sup>					
Right	7.8	2.4	9.8	4.0	3.1	1.2
12-Left	6.0					
Right	"					
Sternum	63.3	7.25	25.5	10.1	9.8	3.2

<sup>a</sup> Rib 11-L, 12-R.

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**Table B-4.** Weights, thorium isotopes, and radium concentrations of the analyzed bones and parts of bones of the arms and hands.

	Weight (g)		Thorium and radium concentration (Bq kg <sup>-1</sup> ash)			
	Wet	Ash	<sup>232</sup> Th	<sup>228</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th
<b>Humerus</b>						
Left	159.6					
Right	169.0					
Proximal end (PE)	58.4	12.0	3,949	1,611	1,690	483
Proximal shaft (PS)	36.2	17.4	1,604	537	218	189
Distal shaft (DS)	31.5	15.6	330	175	285	46
Distal end (DE)	37.6	12.2	173	196	339	32
<b>Radius</b>						
Left	43.3					
Right	43.3					
Proximal end	5.2	1.3	175	*	251	20
Proximal shaft	12.4	5.8	50	*	107	7.6
Distal shaft	13.2	6.8	33	*	89	3.7
Distal end	12.1	3.2	105	*	184	12
<b>Ulna</b>						
Left	51.5					
Right	54.2					
Proximal end	25.3	7.5	183	*	306	25
Proximal shaft	13.1	6.7	42	*	106	6.1
Distal shaft	11.5	5.6	42	*	100	3.9
Distal end	3.5	0.8	144	*	243	19
<b>Carpals</b>						
Left	18.2					
Right	16.9					
Scaphoid	2.6	0.77	81	*	200	12
Lunate	2.0	0.55	226	*	243	28
Triangular	1.8	0.45	236	*	350	23
Pisiform	0.8	0.24	141	*	374	15
Hamate	2.6	0.69	134	*	215	17
Capitate	2.3	0.64	202	*	293	13
Trapezoideum	1.4	0.41	208	*	293	16
Trapezium	3.4	0.99	136	*	277	5.9
<b>Metacarpals</b>						
Left	29.4					
Right	30.2					
1	5.7	1.77	183	*	236	19
2	8.2	2.91	166	*	202	9.7
3	7.6	2.72	176	*	227	13
4	4.4	1.34	200	*	232	10
5	4.3	1.40	69	*	150	17
<b>Phalanges</b>						
Left	25.7					
Right	25.0					
1-Proximal (P-1)	3.0	0.98	155	*	320	19
Distal (D-1)	1.1	0.35	234	*	375	18
2-Proximal (P-2)	3.8	1.34	119	*	212	13
Middle (M-2)	1.4	0.45	113	*	275	15
Distal (D-2)	0.6	0.18	115	*	446	28
3-Proximal (P-3)	4.5	1.67	143	*	211	14
Middle (M-3)	1.8	0.71	234	*	297	16
Distal (D-3)	0.8	0.23	193	*	399	27
4-Proximal (P-4)	3.0	1.03	192	*	223	12
Middle (M-4)	1.3	0.47	160	*	306	18
Distal (D-4)	0.6	0.16	144	*	412	29
5-Proximal (P-5)	2.0	0.61	32	*	277	13
Middle (M-5)	0.7	0.19	154	*	331	39
Distal (D-5)	0.4	0.11	268	*	385	12

\* Activity less than minimum detection level (1.2 Bq per sample).

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**Table B-5.** Weights, thorium isotopes, and radium concentrations in bones and parts of bones of the legs and feet.

	Weight (g)		Thorium and radium concentration (Bq kg <sup>-1</sup> ash)			
	Wet	Ash	<sup>232</sup> Th	<sup>228</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th
<b>Femur</b>						
Left	542					
Right	541					
Proximal end (PE)	161.7	48.1	8,254	3,160	3,006	1,055
Proximal shaft (PS)	64.3	33.5	1,556	717	725	182
Middle shaft (MS)	59.5	30.4	1,104	527	602	153
Distal shaft (DS)	78.9	29.9	211	154	190	37
Distal end (DE)	168.2	37.8	186	152	218	19
<b>Tibia</b>						
Left	316					
Right	332					
Proximal end	121.0	22.9	276	244	385	28
Proximal shaft	84.1	34.3	70	106	145	8.3
Distal shaft	66.2	31.3	47	50	129	5.7
Distal end	51.4	13.4	181	282	221	22
<b>Fibula</b>						
Left	65					
Right	66					
Proximal end	11.4	2.8	154		285	21
Proximal shaft	19.7	9.0	38	275	103	4.9
Distal shaft	19.7	9.9	44		124	7.0
Distal end	13.8	2.8	196		306	22
<b>Patella</b>						
Left	30					
right	31	7.0	146	*	331	29
<b>Tarsals</b>						
Left	139					
Right	135					
Talus	36.2	10.5	106	*	173	13
Calcaneus	59.7	14.6	133	*	207	15
Cuboid	10.7	2.6	105	*	207	13
Navicular	11.0	3.1	113	*	199	17
Cuneiform, med.	9.0	2.5	154	*	246	15
int.	3.5	0.96	100	*	199	10
lat.	4.9	1.33	90	*	181	8.0
<b>Metatarsals</b>						
Left	41					
Right	42					
1	14.8	4.6	110	*	196	11
2	7.5	2.5	97	*	219	8.4
3	6.1	1.9	120	*	234	14
4	6.0	1.8	106	*	232	15
5	7.4	2.4	127	*	261	25
<b>Phalanges</b>						
Left	17					
Right	16					
P-1	5.5	1.8	117	*	247	8.0
D-1	2.4	0.56	258	*	371	28
P-2	1.6	0.53	107	*	253	17
M-2	0.5	0.13	119	*	263	18
D-2	0.3	0.07	371	*	490	43
P-3	1.3	0.41	114	*	248	12
M-3	0.4	0.10	132	*	323	15
D-3	0.4	0.08	442	*	467	56
P-4	1.2	0.37	114	*	262	17
M-4	0.4	0.10	145	*	370	33
D-4	0.3	0.07	305	*	517	45
P-5	1.1	0.35	162	*	352	23
M-5	0.5	0.09	285	*	435	48
D-5	Fused with M-5					

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**Table B-6.** Weights, thorium isotopes, and radium concentrations of bones and parts of bones of the entire skeleton.

	Weight (g)		Thorium and radium concentration (Bq kg <sup>-1</sup> ash)			
	Wet	Ash	$^{232}\text{Th}$	$^{228}\text{Ra}$	$^{232}\text{Th}$	$^{230}\text{Th}$
Skull						
Cranial bones						
Occipital	202	106	501	310	321	66
Parietal-1,2	209	117	611	331	341	80
Temporal-1,2	145	58	1,961	850	777	209
Frontal-1	88	50	254	205	234	34
Facial bones						
Maxilla	79	20	580	494	479	64
Frontal-2,3	85	42	471	275	320	64
Temporal-3	19	8.3	87	<sup>b</sup>	137	12
Mandible	71	36	453	<sup>b</sup>	395	50
Teeth-upper	14 <sup>c</sup>	10	18	<sup>b</sup>	74	3.8
-lower	6.9 <sup>c</sup>	4.7	29	<sup>b</sup>	100	5.3
Hyoid bone	3.1	0.7	7,152	<sup>b</sup>	3,110	990
Vertebral column						
Cervical (1-7) arches	79	28	9,618	2,625	3,509	1,216
bodies	56	16	11,730	4,854	4,860	1,599
Thoracic (1-12) arches	245	74	9,791	4,192	3,780	1,277
bodies	177	46	20,670	8,982	7,952	2,829
Lumbar (1-5) arches	153	75	3,970	1,654	1,552	544
bodies	167	54	15,990	8,435	6,459	1,888
Sacrum	232	46	15,060	6,567	6,162	2,041
Coccyx	12	1.7	1,441	<sup>b</sup>	819	194
Pelvis						
Ilium	481	149	11,732	5,260	4,725	1,768
Ischium	317	91	9,671	4,431	4,077	1,143
Clavicles						
Sternal end	17	4.4	2,120	1,058	1,103	315
Shaft	22	10	1,463	295	642	158
Acromial end	18	3.4	10,540	4,293	4,184	1,199
Scapulae						
Proximal end	35	12	990	568	741	92
Spine	76	28	4,682	2,743	1,917	849
distal end	46	20	4,332	1,725	1,978	554
Ribs (1-12)	448	131	81,940	35,370	31,410	10,580
Sternum	63	7.3	25,460	10,100	9,753	3,157
Humeri						
Proximal end	73	24	173	196	339	32
Shaft	131	64	999	367	251	122
Distal end	114	23	3,949	1,611	1,690	483
Radii						
Proximal end	10	2.7	175	<sup>b</sup>	251	20
Shaft	51	25	41	<sup>b</sup>	98	5.5
Distal end	24	6.5	105	<sup>b</sup>	184	12
Ulnae						
Proximal end	49	15	183	<sup>b</sup>	306	25
Shaft	48	24	42	<sup>b</sup>	103	5.1
Distal end	6.8	1.6	144	<sup>b</sup>	243	19
Hand bones						
Carpals	35	9.8	162	<sup>b</sup>	267	15
Metacarpals	60	20	163	<sup>b</sup>	211	13
Phalanges	51	17	152	<sup>b</sup>	272	16
Femora						
Proximal end	324	96	8,254	3,160	3,007	1,055
Shaft	406	188	981	476	514	126
Distal end	337	76	186	152	218	19
Tibiae						
Proximal end	236	45	276	244	385	28
Shaft	293	128	59	79	137	7.1
Distal end	100	26	181	282	221	22
Fibulae						
Proximal end	23	5.5	154	<sup>b</sup>	285	21
Shaft	78	37	41	<sup>b</sup>	115	6.1
Distal end	27	5.5	196	<sup>b</sup>	306	22

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Table B-6. Continued

	Weight (g)		Thorium and radium concentration (Bq kg <sup>-1</sup> ash)			
	Wet	Ash	<sup>232</sup> Th	<sup>228</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th
Patellae	61	14	146	<sup>b</sup>	331	29
Foot bones						
Tarsals	274	72	120	<sup>b</sup>	198	14
Metatarsals	83	26	112	<sup>b</sup>	223	14
Phalanges	33	9.5	152	<sup>b</sup>	291	17
Entire Skeleton	6,474	2,196	4,353	1,935	1,798	576

<sup>a</sup> When replicated samples (e.g., arches of thoracic vertebrae (1-12) have been combined, the thorium and radium concentration shown is the grand average,  $\Sigma$  radioactive content/ $\Sigma$  ash weight, summed over all samples.

<sup>b</sup> Activity less than minimum detection level (1.2 Bq per sample).

<sup>c</sup> Teeth from right side only.

### QUESTIONS AND COMMENTS

(Paper presented by J. F. McInroy, Los Alamos National Laboratory, Los Alamos, NM.)

**Q: J. Boice**

Was there detectable activity in the breast or eye tissue?

**McInroy:** Yes, but very little. The thorium measurements indicated that there were 6.6, 6.9, and 0.8 mBq of <sup>232</sup>Th, <sup>228</sup>Th, and <sup>230</sup>Th, respectively, in the eyes, and 10.9, 7.6, and 2.5 mBq of <sup>232</sup>Th, <sup>228</sup>Th, and <sup>230</sup>Th, respectively, in the breast. The <sup>228</sup>Ac measurements made at NIST showed 22 mBq g<sup>-1</sup> in the eyes and 5 mBq g<sup>-1</sup> in the breast.

**Q: N. Priest**

If you made reasonable assumptions about equilibrium conditions at the time of the thorium extraction, from what must have been a mixture of uranium and thorium, can you estimate what the equilibrium position should be between the <sup>232</sup>Th (half-life, 10<sup>10</sup> y) and the <sup>228</sup>Th (half-life, 1.9 y) at this time out from manufacture? I'm just wondering whether there's any evidence of diffusion or loss of radionuclides from the body.

**Kathren:** You won't get a clear picture about equilibrium since, when the thorium was separated years ago, the <sup>228</sup>Th present at that time would have decayed these 30-some years later.

**Priest:** The initial <sup>228</sup>Th would be gone; I accept that. But with the initial <sup>228</sup>Th, there wouldn't have been any radium or actinium, which are the parents of the <sup>228</sup>Th. So, now a number of years have passed that would allow for the equilibrium to re-establish. When in equilibrium, there should all be decays of one to one to one to one—i.e., <sup>232</sup>Th to <sup>228</sup>Ra to <sup>228</sup>Ac to <sup>228</sup>Th—but they are not. You've got a 1:3 ratio of <sup>228</sup>Th to <sup>232</sup>Th decays, and I'm just wondering what that ratio should be if it's growing in.

**Boice:** Interestingly, Goldin et al. (*Health Physics*, Vol. 22, pp. 471-482, 1972) published on two autopsy cases in 1972 and reported a similar 1:3 ratio (0.3) of <sup>228</sup>Th to <sup>232</sup>Th in tissue samples from lymph nodes, liver, and

cortical bone. They concluded that perhaps 65% of the <sup>228</sup>Ra (the first daughter of <sup>232</sup>Th) and also some <sup>228</sup>Th must be eliminated from the body.

**McInroy:** The uncertainty is that we don't know whether equilibrium existed at the time the injection was given to this patient. That is, we don't know the elapsed time between when the Thorotrast suspension was made originally and when it was injected into the patient. We don't know whether equilibrium would have been established.

**Priest:** But certainly, when looking at Thorotrast material and at the ratio of <sup>228</sup>Th to <sup>232</sup>Th activity, you can forget any <sup>228</sup>Th injected as this will have decayed away.

**Schima:** I would guess that the ratio of activities should be almost 95% for the equilibrium, if radium is not lost.

**Kathren:** That's the point I was trying to make, which suggests that there is some—

**Schima:** Loss of radium.

**Kathren:** Possibly due to various biological mechanisms.

**McInroy:** And this has been reported in the literature also, that there is disequilibrium between the various tissues because of preferential removal of the radium.

**Schima:** Because the radium has a 6-y half-life, there's plenty of opportunity for loss.

**Kathren:** You've got a secular equilibrium situation; after 30 y, it's going to show up.

**Kathren:** What you really have, if you ignore the actinium, which you could ignore, is true secular equilibrium. So I think the ratio should be 0.95. Why the 0.4 ratio is seen for Thorotrast, I don't know.

**Priest:** It should be one then, shouldn't it?

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**Kathren:** But you know, the reason you may get that 0.4 ratio is that, when you separate out the thorium, the radium may be leached out ahead of time. So what you're doing is removing the precursor of  $^{228}\text{Th}$ .

**Priest:** Perhaps what can be done is to obtain old batches of Thorotrast and then check the thorium activities in the solution to learn how much radium is present in the solution.

**Kathren:** That's probably not a bad idea, but I would expect it to be at equilibrium, sitting around for 30 y.

**McInroy:** I do have a bottle of Thorotrast, which Ron Kathren obtained for me. I've had it analyzed in another laboratory at Los Alamos because of the relatively large amount of radioactivity in the sample. They did a gravimetric analysis of the thorium content and found 2.45 g  $^{232}\text{Th}$ ,  $8.4 \times 10^{-6}$  g  $^{230}\text{Th}$ , and  $3.8 \times 10^{-10}$  g  $^{228}\text{Th}$ . We calculated the activities to be 271.9 nCi  $^{232}\text{Th}$ ,

1.734 nCi  $^{230}\text{Th}$ , and 311.9 nCi  $^{228}\text{Th}$ . Thus, the  $^{228}\text{Th}$ : $^{232}\text{Th}$  nCi activity ratio is about 0.87.

**Priest:** It would be important to know how much of that  $^{226}\text{Ra}$  is diffused away and similarly, at the next stage, how much of the  $^{220}\text{Rn}$  is diffusing away.

**Kathren:** It would be really neat if we could compare Nick Priest's three batches of Thorotrast with Jim McInroy's batch because I have a hunch that there will be significant differences in the thorium isotopic distribution. Depending on where it comes from and how much uranium is in association with it when it was separated, you get greater or lesser amounts of  $^{230}\text{Th}$ , an impurity in the Thorotrast preparation. It's conceivable that  $^{230}\text{Th}$  from an activity standpoint, with its 80,000-y half-life and relatively large specific activity, could become dominant, again depending on where the thorium was obtained from. ■ ■

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