NUREG/CR-1227
Thorium in Human Tissues
M.E. Wrenn, N.P. Singh, N. Cohen, S.A. Ibrahim, G. Saccomanno
September 1981

The content of Th-228, Th-230 and Th-232 was investigated in human tissue from the general public without known occupational exposure. Twenty-two sets of human tissues from Grand Junction, Colorado and ten sets from Washington, D.C. were analyzed. Organs included were lung, pulmonary lymph nodes, liver, kidney, spleen and bone. A radiochemical method was developed for the determination of environmental levels of alpha-emitting isotopes of thorium in tissue consisting of tissue digestion, separation of thorium isotopes by solvent extraction, electrodeposition, followed by alpha-spectrometric measurement using a surface barrier silicon diode. The method utilizes Th-229 as an internal tracer for radiochemical recoveries, and Th-228, Th-230, and Th232 are measured simultaneously. Concentrations of Th-230 and Th-232 decrease in the following order: lymph nodes > lung > bone > kidney > liver, and Th-228: lymph nodes > bone > lung > kidney> liver. As expected, Th-230 and Th-232 were distributed similarly with about 11-27% of the activity content in lung, and 55-70% in the skeleton. Th-228 was distributed differently with about 80% in the skeleton and only 5% in the lungs. Thorium-230 and 232 concentrations in bone were found to increase approximately linearly with age. The observed distribution of thorium among the organs is in good agreement with that recently adopted by the ICRP (ICRP 30, 1979). The total accumulation of thorium in the human body was equivalent to the average amount of natural thorium in 1 to 6 grams of soil. The highest median radiation dose rate for the groups studied from all 3 isotopes of Th was 5 and 0.6 mrad/year delivered to pulmonary lymph nodes and bone respectively.

Health Physics Library

NUREG/CR-1227

Thorium in Human Tissues

Prepared by M. E. Wrenn, N. P. Singh, N. Cohen, S. A. Ibrahim, G. Saccomanno

New York University Medical Center

Prepared for
U.S. Nuclear Regulatory
Commission

NOTICE

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for any third party's use, or the results of such use, of any information, apparatus product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights.

Available from

GPO Sales Program
Division of Technical Information and Document Control
U. S. Nuclear Regulatory Commission
Washington, D. C. 20555

Printed copy price: $4.75

and

National Technical Information Service
Springfield, Virginia 22161

NUREG/CR-1227
RH

Thorium in Human Tissues

Manuscript Completed: December 1980
Date Published: September 1981

Prepared by
M. E. Wrenn*, N. P. Singh*, N. Cohen, S. A. Ibrahim, G. Saccomanno

New York University Medical Center
Institute of Environmental Medicine
550 First Avenue
New York, NY 10001

*Currently with the Department of Pharmacology,
University of Utah, Salt Lake City, Utah 84112

Prepared for
Division of Health, Siting and Waste Management
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
NRC FIN B5756

PROPERTY OF U.S. GOVERNMENT

08704
5-86
4.75

TABLE OF CONTENTS

	Page
Executive Summary	iii
Acknowledgements	vi
List of Tables	vii
List of Figures	ix
1. Introduction and Experimental Approach	1
2. Review of Relevant Properties of Thorium	3
2.1 Thorium Distribution in the Environment	3
2.2 Radiological and Chemical Properties of Thorium	4
2.3 Distribution Pattern of Thorium among Tissues	7
2.4 Review of Thorium Measurements in Man	9
3. Experimental Plan	10
3.1 Tissue Collection	10
3.2 The Utilization of Thorium-229 as an Isotopic Tracer	11
3.3 Development of an Analytical Technique for the Determination of Alpha-Emitting Radionuclides of Thorium in Soft Tissues and Bone	16
3.4 Data Presentation and Associated Error	24
3.5 Critical Level for Detection of Net Activity	26
3.6 Analytical Quality Control	29
4. Stable Calcium Determination in Tissues	32
5. Thorium Concentrations Measured in Soil and Air	34
5.1 Results of Measurements of Thorium in Grand Junction Soil	34
5.2 Results of Thorium Measurements in Air Samples and Human Lungs from New York City	37
6. Results of Thorium Measurements in Human Tissues	41
6.1 Characteristics of the Poulations Sampled	41
6.2 Estimates of Central Tendency and the Appropriate Statistical Test	42
6.3 Concentration of Thorium in Tissues for the Cases Without Known Occupational Exposure	43
6.4 Thorium Concentration in Two Occupational-Exposure Cases	44
7. A Comparison of Thorium Concentration in Tissues from Grand Junction and Washington, D.C.	50
8. A Comparison of Thorium Concentration in Lungs from Smokers and Non-Smokers	55
9. The Distribution of Thorium in the Human Body	57

TABLE OF CONTENTS (continued)

	Page
10. Radiation Dose from Environmental Thorium to Major Organs	62
11. Accumulation of Thorium in Bone with Age	65
12. The Biological Half-Life (t_b) of Thorium in Lung	71
13. Summary and General Conclusions	76
Reference	81
Appendix A	87
Appendix B	97

INVESTIGATION OF THE THORIUM CONTENT IN HUMAN TISSUES

EXECUTIVE SUMMARY

The content of natural alpha-emitting isotopes of thorium (Th-228, Th-230, and Th-232) was determined in 22 sets of human tissue samples obtained at autopsy from Grand Junction, Colorado and 10 sets from Washington, D.C. Lung, pulmonary lymph nodes, liver, kidney and bone were obtained for each case. Personal data for each individual's age, sex, smoking history, and occupation were obtained in most cases.

A simple accurate analytical technique was developed for the determination of environmental levels of alpha-emitting isotopes of thorium using Th-229 as an internal tracer. The method consists of tissue digestion, separation of thorium from other elements by solvent extraction, electrodeposition, and solid-state alpha spectrometric measurements using Si(Li) surface barrier detectors.

Thorium-229 was successfully employed as an isotopic tracer to determine the analytical yield. This isotope has been chosen because of its long half-life and the ability to measure its alpha emission simultaneously with the natural alpha-emitting isotopes of thorium.

The included table summarizes median organ concentrations, derived distribution by organ, and calculated median radiation dose rates.

Thorium nuclides were measurable in the following order of decreasing concentration: lymph nodes > lung > bone > kidney > liver for Th-232 and Th-230, and lymph nodes > bone > kidney > liver for Th-228. The concentration of thorium nuclides were more than an order of magnitude higher in lymph nodes than in lung and the concentration of Th-230 was higher than Th-232 in all tissues, including bone, reflecting the relative geochemical abundances of the uranium and thorium series, respectively. On the other hand, the concentration of Th-228 was higher than Th-232 in bone. This suggests that a portion of Th-228 in bone is due to the intake of transportable Ra-228, which concentrates in bone and then decays to Th-228.

Thorium-230 was significantly higher in lung, lymph nodes, and bone of two occupational cases (a uranium miner and a hard-rock miner) from Grand Junction, Colorado. Thorium-230 in skeleton is apparently a good indicator of lifelong exposure to ore dust.

The median thorium content by isotope for the human body was estimated from the measured concentrations in organs, the reference weights of organs for men and women taken from IRCP Report 23, and weighted for the proportion of men and women in each population. Thorium-230 and Th-232 were distributed similarly (but not identically), as would be expected, with about 11-29% of the body content in the lung, and 55-75% in the skeleton. Thorium-228 was clearly distributed differently from Th-232 and Th-230, with about 80% in the skeleton and only 5% in the lungs. The highest total accumulation of the thorium isotopes was in the skeleton. Thorium-230 and Th-232 concentrations in bone were found to increase approximately linearly with age.

The data suggest that even though the non-mining residents from this area have lived in the vicinity of uranium mine tailings, they do not have clearly elevated Th-230 concentrations in their lungs, when compared to the residents of Washington D.C., who are not exposed to such tailings.

However, the concentration of Th-230 and 232 in the skeleton was statistically significantly higher in the Grand Junction subjects compared to Washington D.C. subjects which may be attributed to the higher median age of 67 years for the Grand Junction population compared to 33 years for the Washington D.C. subjects. It was shown that Th-230 and Th-232 concentrations in the skeleton increase with age. For Th-230 and Th-232 after adjustment for the age differences, the median age adjusted concentration in bone for the Grand Junction population was higher but not statistically significantly different.

The liver contained about 3 to 4% of the total body burden of the three isotopes, in agreement with the distribution assumed in ICRP 30 of ~ 4%; the latter was based on distribution studies of injected Th-228 in the beagle. The results reported here are the only sufficiently detailed study in man to date which allow assessment of the distribution among the organs. Thus the liver is not a major organ of storage for thorium isotopes accumulated from environmental sources.

The total median accumulation of thorium in the human body inferred from the measurements of tissue was equivalent to the average natural thorium content in 1 to 6 grams of soil. The highest radiation dose from all of the thorium isotopes was delivered to the lymph nodes and ranged from 1 to 5 mrads/year. Median doses averaged over bone, lung and liver were 0.41 and 0.23, 0.06 and 0.44, 0.19 and 0.07 mrad/year respectively, in the Grand Junction and Washington populations sampled, assuming complete equilibrium of the short lived daughters of Th-228.

Isotopes	Organs	Median Conc. pCi/kg Grand Junction	Median Conc. pCi/kg Wash. D.C.	Organ Distribution % Grand Junction	Organ Distribution % Wash. D.C.	Median Radiation Dose Rate mrad/yr Grand Junction	Median Radiation Dose Rate mrad/yr Wash. D.C.
Th-228	Lung	0.28	0.24	5.0	3.7	0.028, 0.17*	0.024, 0.14*
	Lymph Nodes	5.1	2.6	1.6	0.70	0.51, 3.0	0.26, 0.16
	Liver	0.07	0.09	2.6	2.6	0.0071, 0.042	0.0091, 0.054
	Bone	0.54	0.66	80.2	82.5	0.054, 0.32	0.067, 0.40
	Total Body			5.05 pCi	5.70 pCi		
Th-230	Lung	0.84	0.31	8.8	11.0	0.073	0.027
	Lymph Nodes	11.0	4.6	1.8	2.1	0.96	0.40
	Liver	0.15	0.15	2.7	5.8	0.013	0.013
	Bone	0.92	0.32	5.4	68.7	0.080	0.028
	Total Body			9.14 pCi	3.27 pCi		
Th-232	Lung	0.58	0.32	23.4	23.3	0.043	0.024
	Lymph Nodes	7.8	2.8	5.6	4.7	0.58	0.21
	Liver	0.03	0.05	2.3	6.2	0.0022	0.0037
	Bone	0.16	0.10	56.1	55.0	0.012	0.0074
	Total Body			2.14 pCi	1.29 pCi		

* For Th-228 and daughters in equilibrium.

Acknowledgements

The authors gratefully acknowledge the support of Dr. Merril Eisenbud, Director of Labortory for Environmental Studies, Drs. T. Kneip and B. Cohen for providing New York City air filters and autopsy lung specimens; J. Neton and H. Ford, for their technical help; Mr. Claude sill (E.G. & G, Idaho) for his initial assitance in the development of analytical techniques which got us off to a quick and well directed analytical start; Dr. L. Woodbury of the University of Utah for his assistance with the statistical analysis; Dr. R. Riddick for providing the autopsy specimens and pathological results.

Sincere thanks are due to Dr. Judith Foulk, the contract officer from NRC, for her cooperation and administrative support of the research project.

Finally, we would like to thank Eleanor Clemm, Francine Lupino and Deedre Miller (University of Utah) for the contribution to typing the manuscript several times.

This study was funded by contract No. NRC-0476-356 from the U.S. Nuclear Regulatory Commission and is part of center programs supported by Grant ES-00260 from the National Institute of Environmental Health Sciences and by Grant CA-13343 from the National Cancer Institute.

LIST OF TABLES

Table #	Title	Page
2.1	Half-Lives and Energies of Thorium Isotopes of Interest to this Study	6
3.1	Mean Weights of the Samples Analyzed from Grand Junction and Washington, D.C.	11
3.2	Recovery (%) of Thorium in Different Organs	24
3.3	Mean Counts for Net Reagent Blank and Counter Background Over Counting Time of 2500 min	30
3.4	Results of Replicate Analyses of Thorium-228 Content in Beef Liver	31
4.1	Median and Range of Calcium Concentration Measured in Human Tissues	33
5.1	Thorium Isotopic Concentrations in Soil and Tailings Samples from Grand Junction	38
5.2	Thorium Isotopic Concentrations in New York City Air	39
5.3	Thorium Concentration in Three Lung Samples Analyzed from the New York Area	40
6.1	Concentration of Thorium-228 in Autopsy Samples	46
6.2	Concentration of Thorium-230 in Autopsy Samples	47
6.3	Concentration of Thorium-232 in Autopsy Samples	48
6.4	Thorium Isotopic Concentration in Two Occupational Cases and the General Population of Grand Junction, Colorado	49
7.1	Wilcoxon Rank Sum Test for Significance of Thorium Concentration in tissues: Grand Jucntion vs. Washington, D.C.	52
7.2	The Median Concentrations of Thorium Isotopes in Autopsy Samples from Grand Junction, Colorado, and Washington, D.C.	53
7.3	Median and Mean of Th-230/Th-232 Ratio in Organs from Grand Junction and Washington, D.C. and in a Uranium Miner	54
8.1	Natural Alpha-Emitting Radionuclides in Cigarette Tobacco	56
8.2	The Median Concentration of Th-228, Th-230, and Th-232 in Lungs of Smokers and Non-Smokers	56

TABLE OF CONTENTS (continued)

Table #	Title	Page
9.1	Organ Weights of Reference Man	59
9.2	Median Organ-burden Estiamtes for the Thorium Isotopes (pCi/Organ)	60
9.3	Median Derived Organ Distribution for the Thorium Isotopes (percent)	61
10.1	The Effective Alpha Energies for the Thorium Isotopes	63
10.2	Median Dose Rate in mrad/yr for Thorium Isotopes in Tissues	64
13.1	Likely Route of Entry and Origin of Thorium Isotopes in Human Tissue	78

LIST OF FIGURES

Figure #	Title	Page
2.1	Mode of Decay of Thorium and Uranium Series	5
3.1	Alpha Spectra of Mixture of Th-228, 229 and 230 Eelctrodeposited from Aqueous Solution	14
3.2	Alpha Spectra of Th-229 Tracer	15
3.3	Schematic of the Alpha Spectrometry System	20
3.4	Schematic Flow Sheet of the Determination of Thorium in Soft Tissues	21
3.5	Schematic Flow Sheet for the Determination of Thorium in Bone	22
3.6	Alpha Spectra of a Human Lung Containing Th-228, Th-230, Th-232, and the Added Th-229 Tracer	23
5.1	Location of Soil Sampling Sites at Grand Junction, Colorado	35
5.2	Descriptive Map of the Tailings Pile	36
11.1	Variation of Thorium-232 Concentrations in Human Vertebrae with Age	67
11.2	Variation of Thorium-230 Concentrations in Human Vertebrae with Age	68
11.3	Variation of Thorium-228 Concentrations in Human Vertebrae with Age	69
12.1	ICRP Task Group Lung and Clearance Model for Plutonium	74
13.1	50th Percentile Concentration of Thorium-Autopsy Samples from Grand Junction and Washington, D.C.	79
13.2	Principal Pathways of Thorium Transport to Man	80

THORIUM IN HUMAN TISSUES

CHAPTER 1

Introduction and Experimental Approach

Although thorium is a widely distributed element in the environment, its occurrence and distribution in the human body has not been studied adequately, and it is certainly incompletely studied compared with the other natural radionuclides such as radium. This study was designed to determine the amount and distribution of the major alpha-emitting isotopes of thorium (i.e., Th-232, Th-230, and Th-228) in the important organs of the human body, so that the radiation doses associated with their intake can be evaluated. Because thorium has many chemical and biological properties similar to plutonium, a study of the distribution of thorium in man should provide useful information for inferring the potential for the long-term accumulation of plutonium from the environment. Also, it is of interest to know whether the people who live in towns that contain uranium mill tailings exhibit levels of Th-230 in their tissues that are above the levels of non-exposed individuals of the general population.

In the past decade, a variety of industrial applications for thorium have been developed. Thorium is used in refractories; as an alloying agent for magnesium and nickel to which it imparts temperature-resistant qualities. It is also used as an alloy with tungsten for welding electrodes and electrical filaments, is used in optical lenses, the manufacture of gas mantle tubes, and as a catalytic agent in the chemical industry. In addition, the former practice of injection of colloidal thorium preparation (thorotrast) for medical diagnostic purposes has led to a number of cases of liver cancer in man (Al66).

Thorium may become an important source of nuclear energy through the conversion of Th-232, a fertile material, to fissile U-233 in thorium cycle reactors. In order to analyze the potential consequences of accidental releases of thorium and uranium isotopes we need a better understanding of the behavior of thorium isotopes in the environment.

In vivo measurements are not sufficiently sensitive to provide accurate estimates of the thorium content in normal human tissues (Jo71); radiochemcial analysis of the tissues is required. Heretofore, no method has been available for the determination of the multiple isotopes of thorium in biological tissues. For this reason, a new and accurate analytical technique was developed for the determination of the alpha-emitting isotopes of thorium that would be sensitive to the low levels found in the soft tissues and bone of humans. The method uses Th-229 as a tracer to provide a measure of the radiochemical yield for the simultaneous spectrometric measurements of the alpha-emitting thorium nuclides (Th-232, Th-230 and Th-228).

Human tissue samples were collected at autopsy from 32 subjects, most of whom were accident victims. Each set includes samples of lung, liver, kidney, cortical bone, and spleen. The subjects were selected from populations from

two different geographic locations: 22 sets from a western town near a uranium tailings pile, and 10 from an eastern city in which no elevated source of thorium is known to exist. In addition, three lungs were collected from the New York City area.

The analytical determination of the alpha-emitting isotopes of thorium was accomplished by using processes of complete digestion of the tissue, solvent extraction and electrodeposition, which were then followed by alpha-spectrometric measurements. A limited number of air and soil samples were also analyzed for Th-232, 230, and 228 concentrations. The isotopic ratio of thorium nuclides in the tissues, soil, and air can be used to infer information about the source and likely route of entry of these isotopes.

CHAPTER 2

Review of Relevant Properties of Thorium

2.1 Thorium Distribution in the Environment

Thorium is fairly abundant in the earth's crust and occurs in varying amounts in more than 100 minerals, in which it is usually found with uranium. The thorium content reported by Faul (Fa54) indicates a range of 8.1 to 33 ppm for igneous rocks, with a mean value of 12 ppm. The radioactivity of the soil is determined by the rock from which the soil is derived. However, disequilibrium may arise from the differential migration of the uranium and thorium-series radionuclides via water.

An extensive study, using *in situ* gamma-spectral measurements at 200 U.S. locations, showed that the average concentration of natural thorium in U.S. soil is about 1 pCi/g Th-232 which is equivalent to 9.2 ppm thorium (Lo64).

An estimate of the average concentration in soil on a worldwide basis has been reported in NCRP Report # 50 (NC 50) as 0.7 pCi/g (6.4 ppm). Typical values for Th-232 concentrations in soil are reported in the literature to range from 0.2 (1.8 ppm) to 1.9 pCi/g (17.5 ppm).

Thorium may become airborne as soil or dust particles. Limited information on the concentration of thorium in air exists. Sedlet et al. (Se73) reported an average annual concentration of 30, 45, and 30 aCi/m^3 for Th-228, Th-230, and Th-232 in air in the vicinity of Argonne National Laboratory. For a reference atmosphere with 100 μg/m^3 of suspended material averaging 1 pCi/g the concentrations of all three thorium isotopes would be about 100 aCi/m^3 (Ha 79).

In addition to the natural thorium background, there is an additional source for these nuclides due to conventional power production. Eisenbud and Petrow (Ei64) concluded that electricity generation from the combustion of fossil fuels can cause a discharge of natural radioactivity (Ra-226, Ra-228, and Th-228) into the atmosphere. Martin et al. (Ma69) found elevated levels of Th-232 in air (300 aCi/m^3) near a coalfired power plant. Beck, et al. (Be78) have shown that certain more volatile uranium and thorium daughters concentrate on the fly ash particles. At the present time, there is not enough information to evaluate the contribution from this source to "normal" body contents.

2.2 Radiological and Chemical Properties of Thorium

There are 13 isotopes of thorium. Th-228, Th-230 and Th-232 are alpha-emitting isotopes and, because of their long half-lives, have industrial and environmental importance as potential health hazards. Th-232 and Th-228 occur in the thorium minerals. Th-230 is found in association with natural uranium. Th-227, another alpha-emitting thorium isotope, is present in uranium minerals as a member of the U-235 decay chain. However, Th-227 is relatively short-lived and is present in the environment in small amounts (4.5 % by activity of Th-230 when the uranium chain daughters are in equilibrium with the long lived parents). Because of the lesser activity and short half-life (18d), Th-227 should not be present in significant amounts relative to the other alpha-emitting isotopes in tissue (NC 76). Figure 2.1 shows the decay chains that contain the thorium isotopes; their alpha energies and half-lives are noted in Table 2.1. The physical constants are taken from Lederer, et al., 1978.

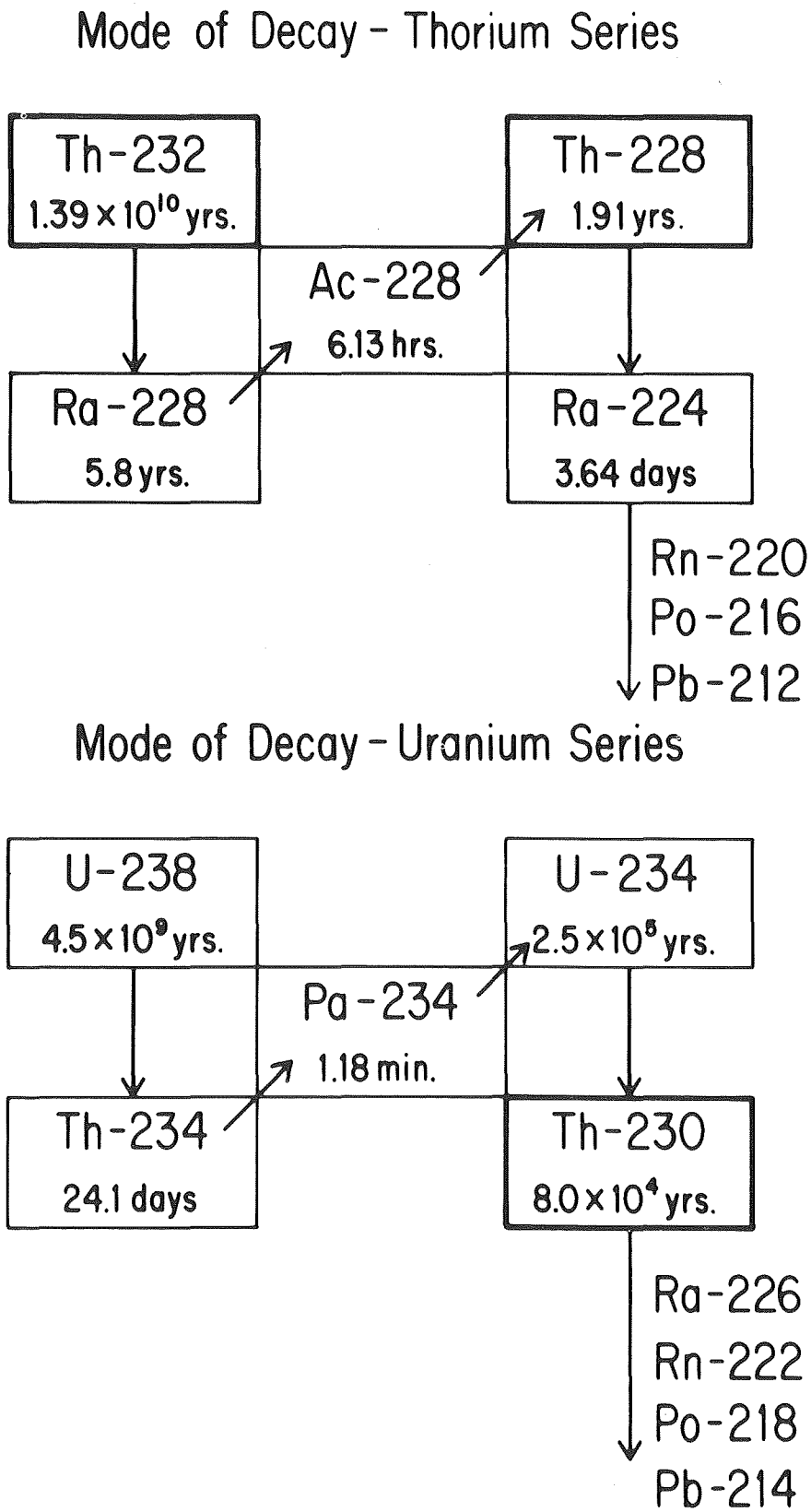
Thorium metal is highly electropositive and has a high melting point (1750°C). Dilute hydrofluoric, nitric, and sulfuric acids and concentrated phosphoric and perchloric acids dissolve thorium.

Because thorium exists in solution as a small highly charged cation, it interacts with water and with many anions. One simplification in the aqueous chemistry of thorium is that it has only one oxidation state and, hence, oxidation-reduction reactions do not occur. The water-soluble salts of thorium include the nitrate, the sulfate, the chloride, and the perchlorate.

The common insoluble compounds of thorium suggest a number of precipitates that may be suitable for the removal of tracer amounts of thorium from solution.

Thorium hydroxide is a highly insoluble compound; when alkali is added to an aqueous solution of Th^{+4}, a gelatinous precipitate is formed. Thorium hydroxide dissolves in aqueous solutions that contain such ions as the citrate or the carbonate which complex the thorium ion. Tracer amounts of thorium will coprecipitate quantitatively with a wide variety of the insoluble hydroxides, such as lanthanum hydroxide, ferric hydroxide and zirconium hydroxide. Because the hydroxide carrier precipitates are non-specific, they are usually used only to remove the thorium from a simple mixture of contaminants or as a group separation that is followed by more specific chemical purification procedures (Ha36).

FIGURE 2.1: Mode of Decay of Thorium and Uranium Series



The extractability of thorium from aqueous solutions and its concentration by organic solvents has been studied for many solvents. Most studies were done with hydrochloric acid and nitric acid systems.

Organic solutions of the high molecular weight amines and amine salts have been shown to be excellent extractants for removing thorium from the acid solutions (Ka57; Hy60).

TABLE 2.1

Half-Lives and Energies of Thorium Isotopes of Interest to this Study (Le 78)

Thorium Isotopes of Interest	Half-Life	Alpha Energy (MeV)	Abundance (%)
Th-227	18.7 d	6.04	24.5
		5.98	23.4
		5.76	20.3
		5.72	14.0
Th-228	1.91 y	5.43	72.7
		5.34	26.7
Th-229	7340 y	5.05	7.2
		4.97	10.0
		4.90	10.8
		4.84	56.2
		4.81	13.2
Th-230	8.0×10^4 y	4.68	76.0
		4.62	24.0
Th-232	1.40×10^{10} y	4.01	77.0
		3.95	23.0
Th-234	24.1 d	β^-	

2.3 Distribution Pattern of Thorium among Tissues

The distribution of Th-232 citrate in tissues was studied in rats by Boone (Bo58). The level of deposition in tissues varied slightly according to the amount administered; an average of 70-80% of the i.v. injected dose was retained at the time of death, 25-30% in bone and 50-60% in soft tissues. Half of the latter was found in the liver. Radioautography showed that the deposition of Th-232 in the liver, spleen, and bone was associated with the reticuloendothelial cells. Scott (Sc52) administered thorium intravenously to the rat as a sulfate salt and found 45-68% in the liver and 6-20% in the skeleton.

When thorium was injected intravenously into adult beagles as a citrate complex (St62), 60-93% was found in the skeleton and 3-4% in the liver. This distribution pattern is similar to that found by Boecker (Bo63) in the rat for Th-234 inhaled as the chloride; the absorbed thorium was deposited almost completely in the skeleton.

The relationship between the mass of intravenously injected thorium and the deposition in the skeleton was studied by Stover (St60) in adult beagles and by Thomas (Th63) in rats. For thorium administered intravenously in the range of 10^{-10} to 10^{-15} grams per kg body weight, about 80% of the body burden is deposited in the skeleton and 20% in the soft tissues, principally in the liver and kidney. When the thorium administered is > 10^{-3} g/kg body weight, the amount in the bone decreases by a factor of about 10, and the amount in the liver increases by the corresponding amount.

This mass effect is dependent on the concentration of the thorium atoms. Because thorium is readily hydrolyzed, the injection of soluble salts in high concentration will result in the formation of colloids that are circulated in the blood and removed by the reticuloendothelial system (liver, spleen, and bone marrow). When small amounts of thorium are present in the circulation, the thorium should be in monomeric (ionic) form and, as such, is deposited primarily on the skeletal surfaces in a manner similar to plutonium (St60).

Relatively few inhalation experiments have been performed with thorium, but some aspects of this behavior in the lung are reasonably well established: thorium oxide in the lung behaves as an insoluble material, and the rate of loss is very slow. When it is removed, it appears predominantly in the feces, a result of ciliary clearance.

Hodge et al. (Ho60) have shown that in repeated exposures of dogs to ThO_2 particles which have a median mass diameter of 1 μm in a concentration of 5 mg/m^3 of air, the lung burden rises steadily with continuing exposure and was more than six times greater after 14 months than after one month. During this same period, the amount of thorium in the liver, kidney, and spleen did not change significantly.

Once deposited in the lung, the persistence of ThO_2 is great, as indicated by the data from three dogs that were exposed for two weeks to 45 mg of ThO_2 per cubic meter of air. The concentration of thorium in the pulmonary lymph nodes was 50 times higher than in lung tissue seven years post-exposure; only about 2% of the lung burden was translocated to the skeleton.

The build-up of thorium in the lungs of rats, rabbits, and guinea pigs exposed to 5 mg ThO_2/m^3 for 14 months was found to be similar to that of dogs (Ho58).

Thomas (Th57) studied the metabolism of Th-230 (in the hydroxide form) administered to the rat by intratracheal injection in doses of 4 mg/kg and found only small amounts in all tissues except the lung. Practically all the Th-230 that does enter the circulation from the respiratory tract is translocated to the skeleton.

The Absorption of Thorium from the Gastrointestinal Tract

Maletskos et al. (Ma69) conducted experiments in which normal elderly subjects ingested radium dial paint that contained Th-228, in addition to Ra-226, as sulfate compounds. The results on six subjects yielded an average absorption of 0.02% for thorium. Experiments on rats and guinea pigs yielded thorium absorption factors equal to or lower than those found in man (Sa51; Sc52). Transport across the GI tract of adult rats, guinea pigs, and dogs was recently measured for soluble nitrate and insoluble oxide compounds of the actinide elements (Su79a). Th-228 absorption was lower than absorption of most of the other actinides studied. Absorption of Th-228 was 0.03%, a value slightly higher than Maletskos found for man using Th-234 $(SO_4)_2$. There was little variation among species; however, solubility and particle size are important factors in determining absorption. Larger fractions of the actinide compounds are absorbed from the GI tract of neonatal rats and guinea pigs (Su79).

2.4 Review of Thorium Measurements in Man

What little information which is available about thorium in human tissues has been reviewed recently (Wr77). Most work has been on human bone samples.

Hill (Hi62) reported preliminary results of spectrometric measurements that were carried out on the natural levels of α-activity in humans and their environment. The spectral resolution in his work, using the pulse-counting ion chamber, was not sufficient to separate Th-230 completely from other interfering alpha emitters. The range he reported for the Th-232 values in nine lung samples was between 0.2 and 1 pCi/kg wet tissue.

Petrow and Strehlow (Pe67) measured the thorium content in a composite sample of ash from the skeletons of several adult individuals and found a value of 10 ng/g ash. Picer and Strohal (Pi68) reported a value of 23 ng/g Th-232 in bone ash for an individual of unknown age. The most extensive series of measurements of thorium in bone were reported by Lucas et al. (Lu70), who analyzed thorium from 38 human rib bones and found values between 0.1 and 72 ng/g ash. They observed that the concentration of thorium in bone increased approximately linearly with age at a rate of 0.16 ng/g of bone ash per year (the specific activity of Th-232 is 1.09×10^{-7} Ci/g).

Clifton et al. (Cl71) reported on the concentration of Th-232 in bones of unexposed persons in the United Kingdom. The concentrations in rib ash reported in this study ranged from 0.8 to 164 ng/g, with a mean of 28.7 ± 13.1 ng/g.

Inhalation is probably the main route of thorium uptake by man. The evidence is summarized in section 2.3.

CHAPTER 3

Experimental Plan

3.1 Tissue Collection

Twenty-two sets of tissues were obtained at autopsy from persons in the Grand Junction, Colorado area (a town in the western United States with a uranium tailings pile), and ten sets were collected from persons in Washington, D.C., where no elevated natural source of thorium is known to exist. In addition, three lung samples were obtained from persons in the New York area.

Samples taken at autopsy were generally from accidental victims, and subjects were generally long-time residents, as the table below shows. (See section 6.1 for further characterization of the subjects.)

Residence Time of the Subjects Autopsied
From Grand Junction and Washington, D.C.

	% of cases where residence time is known	mean residence time in years
Grand Junction	65	45
Washington, D.C.	80	15

From each case, the following organs were collected by a pathologist: the right lung, a kidney, the spleen, approximately one-half of the liver, and approximately 200 grams of a vertebral wedge. From a few cases other samples were obtained, such as the thyroid and the gonads.

At autopsy, specimens were weighed after a short period to allow drainage of the excess fluids. Prior to the chemical analyses, the weights were rechecked and recorded.

For each subject, the tissue samples were placed in individual plastic bags and then packaged in separate labeled containers. The specimens were frozen after autopsy and transported to our laboratory packed in dry ice.

The age range of the subjects was between 1.5 and 86 years. Most were victims of automobile accidents in which death occurred immediately or soon after arrival at the hospital. No subject had a chronic, debilitating disease or a metabolic disease known to affect liver or skeletal metabolism.

At each location all the autopsies were performed by or under the direction of a single pathologist, who examined all organs for degeneration or impairment and for evidence of tumors or carcinomas; any such evidence was

recorded. The cause of death was determined either by gross inspection or by histological examination; in addition, some microscopic sections were taken to determine whether any structural changes not readily apparent could be ascertained and co-related with the data from the chemical analyses.

The date of death, age at death, sex, race, residential histories, smoking and drug history, past medical history, and occupational data were obtained for each case.

The bronchopulmonary lymph nodes were carefully dissected to remove all visually identifiable lymph nodes along the large and smaller bronchi in the regions of bifurcation.

The number and the mean weights of the samples analyzed are reported in Table 3.1.

TABLE 3.1

Mean Weights of the Samples Analyzed from Grand Junction and Washington, D.C.

Organ	Number of Samples	Range of Sample Weights in grams	Mean of Sample Weights in grams
Lung	30	53 - 880	420
Lymph nodes	24	1 - 15	6.5
Liver	26	149 - 850	410
Kidney	25	45 - 210	130
Bone	23	20 - 295	150
Spleen	14	45 - 360	140

Further information about the populations measured is given in Chapter 6.1.

3.2 The Utilization of Thorium-229 as an Isotopic Tracer

Th-229 has been successfully tested and used at this laboratory as a suitable tracer to determine the analytical yield for the radiochemical determination of natural thorium isotopes (Wr78).

Th-234 has traditionally been employed as an isotopic tracer to determine the extraction efficiency and the total analytical yield for the radiochemical determination of "natural" thorium (Pe74; Si77). Accordingly, a number of methods have been reported for its preparation (Si77). Also, because Th-234 has a relatively short half-life of 24.1 days, new batches must be prepared frequently. Furthermore, Th-234 is a β-emitter, whereas most other "natural"

thorium nuclides emit α-particles. In contrast, Th-229 is an α emitter, and has a long half-life of 7340 years.

For this reason, the tracer (Th-229) and the nuclides being determined (Th-232, 230, 228) are deposited in an identical fashion on a single planchet (i.e., same thickness and uniformity of deposit); hence, they may be counted simultaneously with the same detector. With the Th-234 tracer, small changes in the distribution of deposited activity can cause profound changes in counting efficiency and hence the accuracy of the measurements (Si77); therefore, great attention to these details is necessary if the Th-234 tracer is to be used accurately.

The increase in accuracy, simplified counting procedures and freedom from preparation of fresh tracer periodically, in order of importance, are reasons why Th-229 is an improved tracer for most applications. Since the sample count rate in the Th-228 region includes some small contribution from Th-228 contamination in the Th-229 tracer, it was necessary to make corrections for this impurity when Th-228 is determined in the sample.

Th-227, another α-emitting radionuclide, was also considered as a possible isotopic tracer. However, because Th-227 is a decay product of U-235, Th-227 could be present naturally in some biological samples. But more importantly, in addition to Th-227 having a relatively short half-life (18.2 days), its α energies (Table 3.1) are too close to the decay products of Th-228 (Ra-224, α = 5.68 MeV and Bi-212, α = 6.05 MeV) to be easily resolved. For these reasons, Th-227 was not suitable as a tracer.

Experimental Materials: The isotopes Th-228 and Th-229 were obtained from the Oak Ridge National Laboratory, and Th-230 from Isotope Laboratory Products. Trilaurylamine (TLA) was purchased from Matheson Coleman and Bell Manufacturing Chemicals (MC/B); a 25% solution was prepared in xylene and shaken for 10 min. before use with 4 M HNO_3 in the ratio of 3:1. Other reagents and equipment included methyl red indicator, platinum electrodes and planchets, nickel discs, electrolytic cells (HASL manual specification), and electrolytic analyzer (Sargent Welch).

Experimental Procedures: Individual α spectra for Th-228, Th-229, and Th-230 were obtained from the standard solutions, both by direct transfer and evaporation onto platinum planchets and by electrodeposition. In addition, α-spectra were obtained for mixtures of these isotopes, that were prepared in aqueous solution and from a pre-spiked 200 g sample of beef liver which had been digested with HNO_3, followed by a mixture of HNO_3 and H_2SO_4. In this latter determination, the thorium was extracted into 25% TLA in xylene, back-extracted with 10 M HCl, converted to a sulfate with H_2SO_4, and finally electrodeposited onto a platinum planchet.

Results and Discussion

The energies of the thorium isotopes (232, 230, 229, and 228) were listed in Table 2.1. Because the resolution of the silicon surface barrier detector was 45-50 keV for a standard source at a source-to-detector distance of 1 mm, inspection of the α energies show that all four isotopes should be resolved

without any appreciable interference. Both the Th-228 and Th-229 have short-lived daughters that emit α particles; however, the energies of each of their daughters are higher than 5.68 MeV, so that no spectral interference can occur.

Figure 3.1, which shows the α spectrum obtained from a mixture of Th-228,229,230 electrodeposited on a platinum planchet, demonstrates that, as expected, the α energy peaks are well resolved for these three isotopes of thorium.

The radiochemical purity of Th-229 obtained from Oak Ridge National Laboratory contained about 6%, by activity, Th-228, and 0.3% Th-230 (as of Feb. 1977). The α spectrum of this tracer is shown in Figure 3.2. In spite of the presence of Th-228, the tracer is useful, provided that the amount of Th-229 added is such that the adventitiously added Th-228 does not exceed the Th-228 level in the sample to be measured.

The spectra in Figures 3.1 and 3.2 were obtained at 6.7% counting efficiency. A further increase in the counting efficiency to 24% did not cause any resolution problems.

At this time only two disadvantages exist for the use of Th-229. The first is the contamination of the available Th-229 with approximately 6% Th-228; this results in a small increase in the lower limit of detection for this isotope. Second, after long term operation, a recoil problem may cause contamination of the diode. This contamination, however, occurs in the energy region 5.68 to 8.78 MeV and does not interfere with the spectra of the thorium isotopes under consideration (Table 2.1). Furthermore, this recoil contamination problem can be reduced by using the procedures reported by Sill and Olson (Si70) and also by good laboratory techniques; namely, by removing the source of possible contamination from the chamber while not in use, and by avoiding the unnecessary use of overly active sources. The experience after two years is that no contamination which would interfere with "natural" thorium measurements is evident.

In conclusion, the advantages of Th-229 as an isotopic tracer for the radiochemical measurement of other isotopes of thorium are the following:

1. The increase in accuracy associated with simultaneous α-spectrometric measurement of the tracer (Th-229) and the nuclides being determined (Th-232,230,228).

2. Th-229 has a long half-life of 7340 years, and therefore unlike Th-234, requires neither repeated preparation and the associated time-consuming processes of purification and standardization, nor decay corrections.

3. Th-229 is an α-emitter as are the naturally-occurring isotopes of thorium which are of interest; accordingly, the α-spectrum provides, as part of the measurement, the determination of yield, thereby saving time and minimizing handling and separate β measuring techniques.

FIGURE 3.1: Alpha Spectrum of Mixture of Th-228, 229 and 230 Electrodeposited from Aqueous Solution

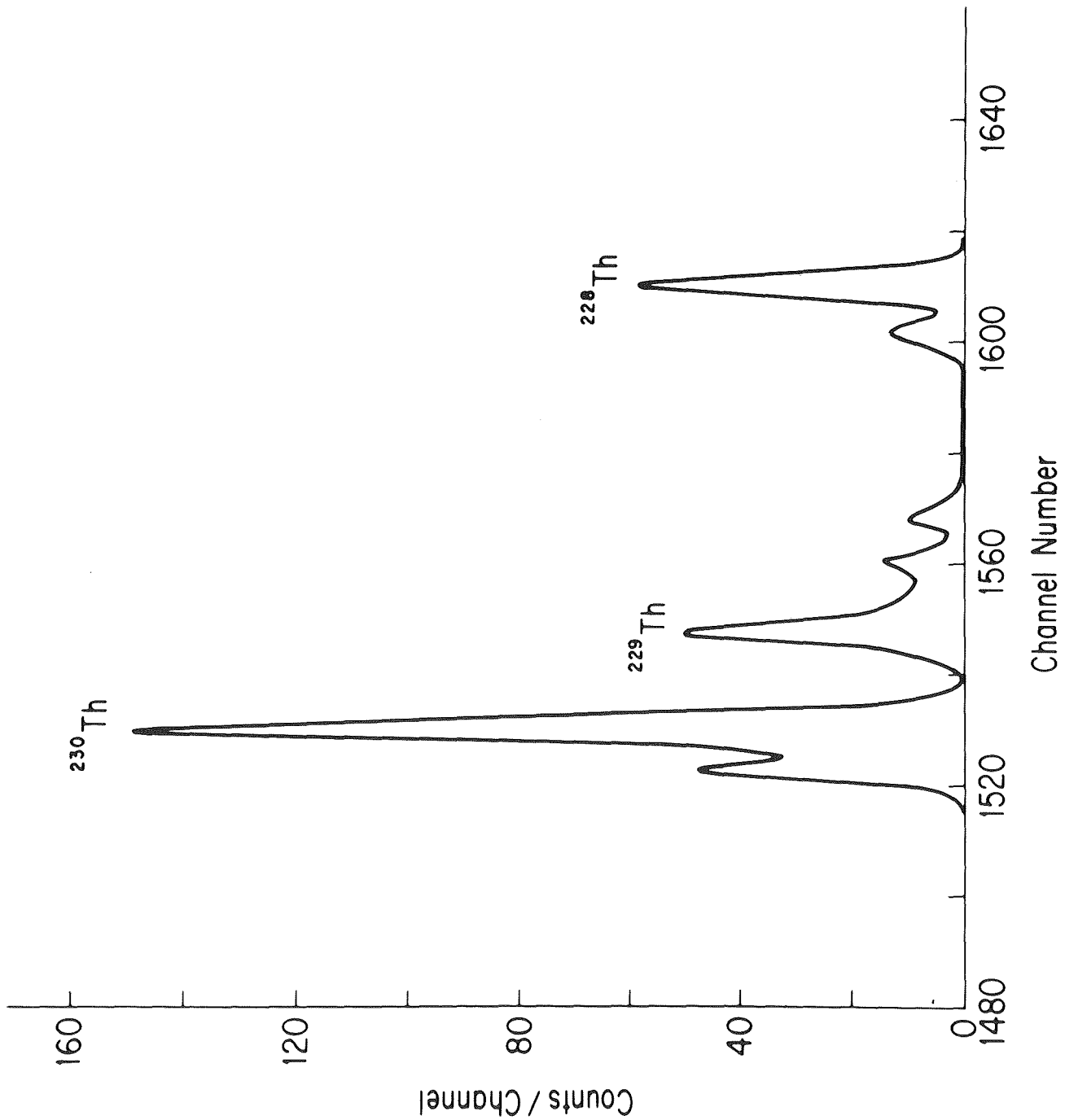
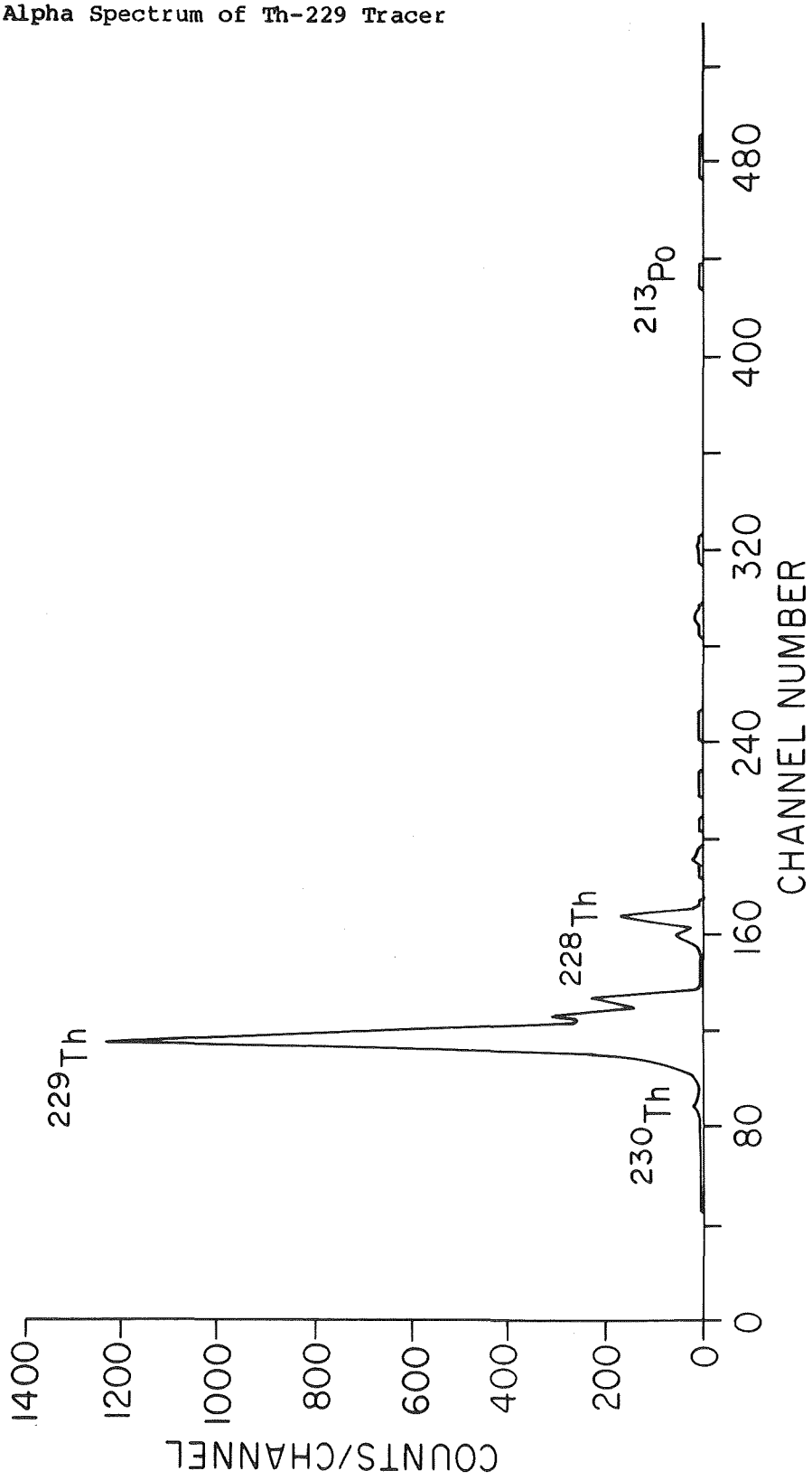


FIGURE 3.2: Alpha Spectrum of Th-229 Tracer



3.3 Development of an Analytical Technique for the Determination of Alpha-Emitting Radionuclides of Thorium in Soft Tissues and Bone

Introduction

To assess the content of the thorium nuclides Th-228, 230 and 232 in human tissues, a new analytical technique has been developed. The methods available for the determination of thorium in tissues are limited. Petrow, et al. (Pe64) described an indirect technique for determining Th-228 which takes a considerable amount of time and is limited to only one isotope of thorium. Another procedure described by Petrow and Strehlow (Pe67) is a colorimetric method for total thorium and again does not measure the isotopic composition. Sill (Si74; Si77) has reported techniques for determining thorium in soil and ore samples and Percival and Martin (Pe74) developed a method for assay of thorium isotopes in environmental and process waste samples. No method has been available, however, for the multiple isotopic determination of thorium in biological tissues. Therefore, a simple, precise and sensitive analytical technique has been developed in this laboratory for the determination of all α-emitting thorium isotopes in tissues (Si79a).

Experimental
Reagents and Apparatus

All the reagents used were of analytical grade. A 25% solution of tri-laurylamine (TLA), [Matheson, Coleman and Bell (MC/B)] was prepared in xylene and equilibrated with 4 M HNO_3 prior to use. A 10% oxalic acid solution was prepared in distilled, deionized water. 2 M H_2SO_4, 4 M HNO_3 and and 10 M HCl dilute acid were prepared by dilution of concentrated acids with deionized, distilled water. Th-229 tracer, (having ~ 6% Th-228 on an activity basis) and methyl red indicator were used. Apparatus used included: platinum planchets, nickel discs, electrolytic cell, electrolytic analyzer (Sargent Welch) and muffle furnace.

Sample Preparation

Weighed amounts of tissue were transferred into proper size beakers and 1-2 dpm of Th-229 tracers were added to each beaker. Concentrated nitric acid was added in sufficient quantity to immerse each tissue. Individual beakers were then covered with a watch glass and heated gently on a hot plate until all frothing had ceased. The temperature was raised slowly and heated continuously without boiling the nitric acid. For bone samples, vigorous heating and digestion with nitric acid was accomplished until most of the organics had been removed. All tissues were heated further with nitric acid and occasional additions of H_2O_2 until the evolution of brown fumes had ceased, indicating almost quantitative removal of organic materials.

For all other tissues, a 200 ml mixture of 1:1 HNO_3 and H_2SO_4 was added and heated vigorously until all the nitric acid had been driven off. A few drops of HNO_3 and H_2O_2 were added occasionally with constant heating until the evolution of brown fumes had ceased and a clear colorless solution remained. Most of the sulfuric acid was removed by evaporation before proceeding

further. For lung and lymph nodes, where significant amounts of silica may be present, the residues of the tissue disolution were heated further with HF and HNO_3 until the tissues were completely dissolved and then the HF was removed by continuous heating.

Procedures

Bone

After the bone sample was wet ashed with HNO_3 and H_2O_2, 200 ml of 1:3 HCl was added to the solution. Ammonia was added carefully and neutralized until precipitate formation started (around pH 4). This solution was heated to boiling and a calculated amount of 10% oxalic acid solution was added with constant stirring to precipitate ~ 10% of calcium available in solution, as calcium oxalate. The precipitate was filtered and washed two to three times with 1% oxalic acid solution and heated slowly in a muffle furnace by raising the temperature in short intervals to 550°C.

The resultant calcium carbonate was dissolved in a 1:1 HNO_3 and the acidity of the solution was adjusted to 4 M by titrating 100 µl aliquot with a 0.1 N NaOH and adding the required amount of nitric acid of appropriate concentration. Once the solution was finally prepared, the solvent extractions were carried out as described later in this section.

Soft Tissues

After the tissue was wet ashed and most of the sulfuric acid was removed, 200 ml of 1:3 HCl was added to the solution. This solution was boiled for several minutes then allowed to cool. Ten milligrams of iron carrier (as 1 ml $FeCl_3$) was added and swirled in the beaker. Concentrated ammonium hydroxide was added gently until the $Fe(OH)_3$ was completely precipitated. The precipitate was boiled for a few minutes and allowed to settle overnight. The supernatant was removed by centrifuging the precipitate in a 40 ml or 250 ml centrifuge tube depending upon the volume size. The precipitate was dissolved in a minimum volume of concentrated HNO_3, $Fe(OH)_3$ was reprecipitated with ammonia, and a large excess of distilled water was added to a maximum volume of the tube to remove the sulfate ions. The precipitate was centrifuged and the supernatent was discarded. The precipitation and dissolution was repeated until the supernatant was free of sulfate ions. The iron hydroxide precipitate, which coprecipitated plutonium and thorium hydroxide, was dissolved in 1:1 HNO_3. The acidity of the solution was adjusted to 4 M as described earlier. The solvent extractions of thorium were then carried out from this solution as described as follows.

Solvent Extraction

Thorium was extracted into 25% TLA solution in xylene (pre-equilibrated with 4 M HNO_3) by shaking for ten minutes (aqueous: organic 1:1 volume). The aqueous phase was removed into another separatory funnel and extracted once again with an equal volume of 25% TLA solution. The aqueous phase was discarded. The organic phases from both extractions were mixed into one large volume separatory funnel. An equal volume of 4 M HNO_3 was added and shaken

for ten minutes. The aqueous phase was discarded. Thorium was backextracted from the TLA phase by shaking with an equal volume of 10 M HCl for ten minutes. The aqueous phase was collected in a clean beaker. The back-extraction of thorium was repeated and the aqueous phase contained all the thorium. The aqueous solution was evaporated, 5 ml of 1:1 H_2SO_4 was added and the solution was heated. Concentrated HNO_3 and 30% H_2O_2 were added dropwise along the side of the beaker once the organics entrained with HCl were observed floating on the surface in order to completely decompose the organics. Following removal of all organics, the solution was ready for electrodeposition of thorium. In the case of bone samples, if large amounts of calcium are precipitated as oxalate, a white mass on the electrodeposited platinum planchet (presumably calcium sulfate) was observed. However, electrodeposition carried out in hydrochloric acid media resulted in cleaner planchets. Nevertheless, in both cases, larger amounts of calcium caused an increase in the mass of the planchet which diminished the resolution.

Electrodeposition

Apparatus

The electrodeposition apparatus consists of an elongated 22 mm plastic cap which holds an inverted, one-ounce polyethylene bottle with the bottom removed. The cap has been designed to accommodate an 18 mm diameter platinum disc supported by a nickel disc. The polyethylene bottle is firmly screwed into the cap holding the discs, thus forming a leak-proof plating cell. A threaded brass bushing is molded into the underside of the cap and establishes an electrical contact with the platinum disc cathode by clip leads. The entire cell is supported by a heavy brass base which is designed to anchor the cell in the ice water bath. The anode consisted of a 1.6 mm platinum-irridium rod, four inches long with a half-inch diameter platinum disc riveted at one end. This disc is provided with a number of 0.3 cm holes. However, these dimensions for the anode are not critical. The anode rod is connected to a constant speed stirrer and to the positive outlet of the power supply, which furnishes a constant current in the range from 0-6 amperes and a voltage ranging from 0-10 volts.

Procedure

The final solution, obtained in the solvent extraction step, was evaporated to dryness. One ml 2 M H_2SO_4 was added and heated gently at a low temperature on a hot plate. The solution was transferred to the plating cell. The beaker, which had contained the solution, was washed two times with 1 ml portions of 2 M H_2SO_4. The washings were transferred to the plating cell. One drop of methyl red indicator was added and the solution was titrated with 1:1 ammonia, by adding dropwise, until a yellow-colored end point was reached. Extreme caution was exercised not to add an excess of ammonia. Then 2 M H_2SO_4 was added drop by drop until the red color was obtained and 2-3 drops extra were added to achieve the required pH of the plating solution. Electrodeposition of thorium was carried out at an initial current of 1.2 amperes for one hour. The electrolyte was quenched with 3-4 drops of ammonium hydroxide at the end of one hour. The cell was dismantled and the platinum disc was rinsed with water and then followed by alcohol. The

disc was flamed to red hot over a burner. The recovery and the isotopic composition of the thorium was determined by counting the platinum planchet in an alpha spectrometer with a solid state surface barrier silicon diode detector.

Alpha Spectrometry

Once the sample had been electrodeposited, thorium isotopes were quantitated using state-of-the-art alpha spectrometry equipment. The detector systems employed were "ruggedized" 300 mm^2 silicon, surface barrier detectors (Ortec, Inc.) which possess a typical resolution of 50 KeV (FWHM) with a counting efficiency of 25-30%. The background count rate in the thorium energy regions of interest averaged less than one count per 1000 minutes of counting time. The data reduction and acquisition systems used were designed for long-term alpha counting and consist of NIM bin electronics incorporating a 4096 channel multi-channel analyzer with dual floppy disc storage and readout capability. The detector electronics are built into a dual vacuum chamber module which incorporates a preamplifier, power supply and linear amplifier into a single NIM module. The system also incorporates a built-in pulse generator for internal electronic stability checks. A simplified schematic of the counting system is given in Figure 3.3.

Results and Discussion

This solvent extraction technique for the determination of thorium was successfully employed for all organs except for bone where difficulties were encountered due to the presence of calcium and phosphate ions. However, once the phosphate ions were removed by calcium oxalate precipitation, coprecipitating thorium and leaving phosphates in solution, the method was as good as for the other organs. The schematic diagrams for soft tissues and bone are given in Figures 3.4 and 3.5, respectively. Figure 3.6 shows alpha spectrum of a human lung containing Th-228, Th-230, Th-232 and added Th-229 tracer. The radiochemical recoveries for thorium in each organ are given in Table 3.2. There were no significant differences in the radiochemcial yields between organs. There is no spectral interference from one isotope to another nor from any other element.

The alpha energies for Po-210 (5.30 MeV) and Th-228 (5.43 MeV) are very close and therefore polonium must be separated during the chemical analysis. The extraction of polonium from nitric acid at moderate concentrations (up to 8 M), using high molecular weight amines, has been reported to be poor (Fi61). In this method, the discrimination against polonuim exceeded 5×10^3 as determined from the addition of known amounts of Po-208 (alpha energy of 5.11 MeV, and half-life of 2.93 Y). The electrodeposition of polonium from sulfuric acid was poor (~1.7%) due to its low solubility in this media.

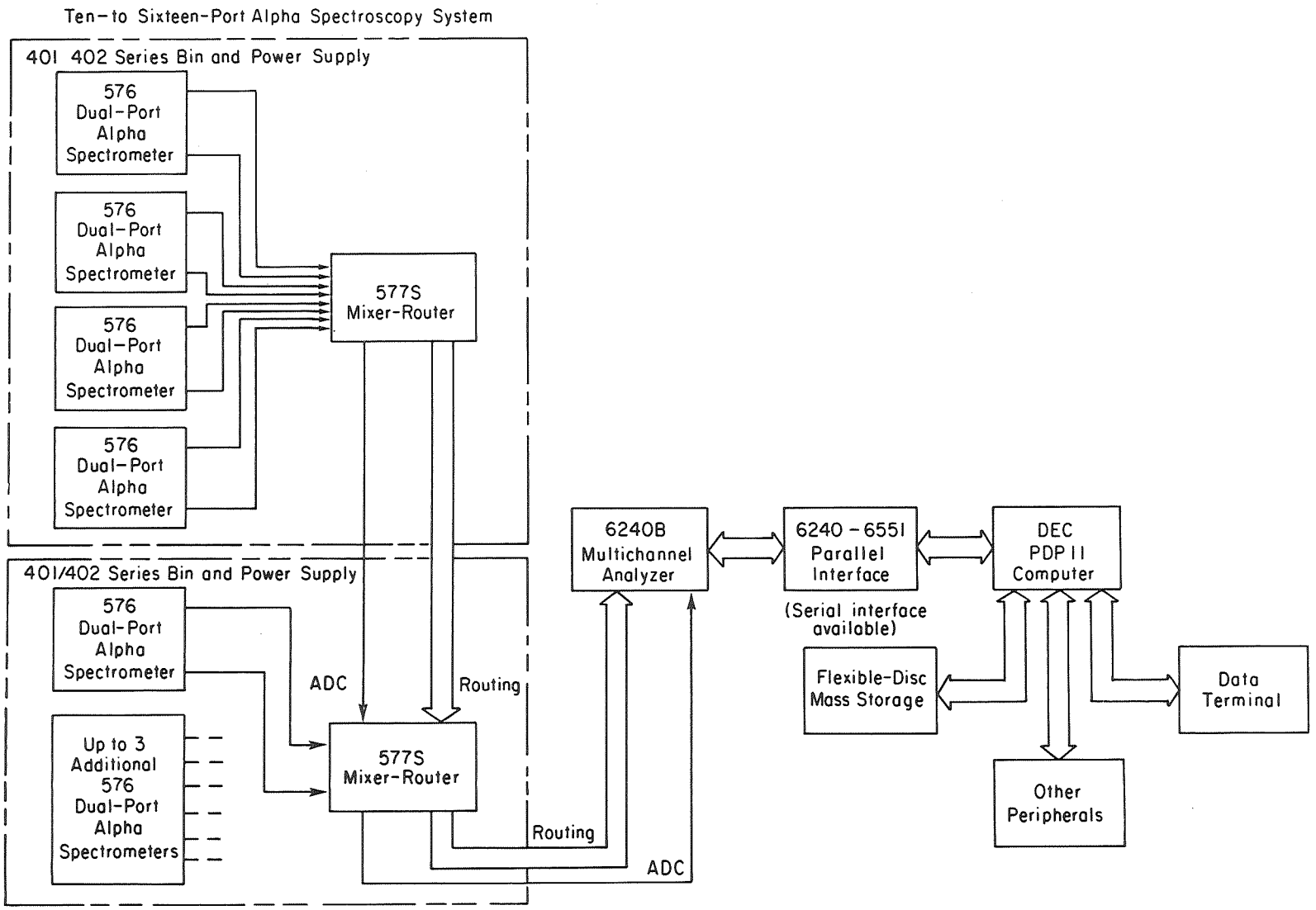


FIGURE 3.3: Schematic of the Alpha Spectrometry System

FIGURE 3.4. SCHEMATIC FLOWSHEET FOR THE DETERMINATION OF THORIUM IN SOFT TISSUES.

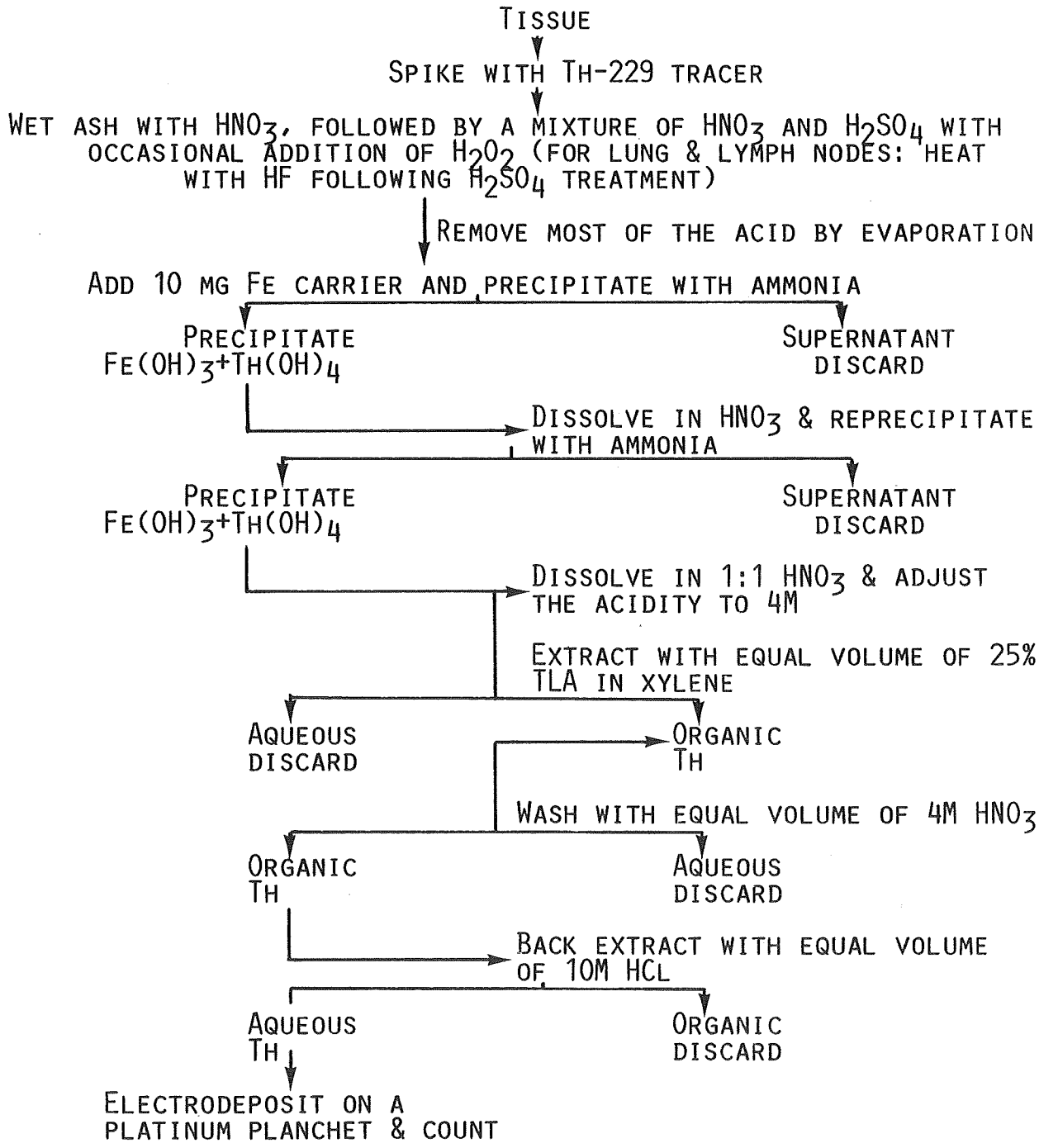


FIGURE 3.5. SCHEMATIC FLOWSHEET FOR THE DETERMINATION OF THORIUM IN BONE.

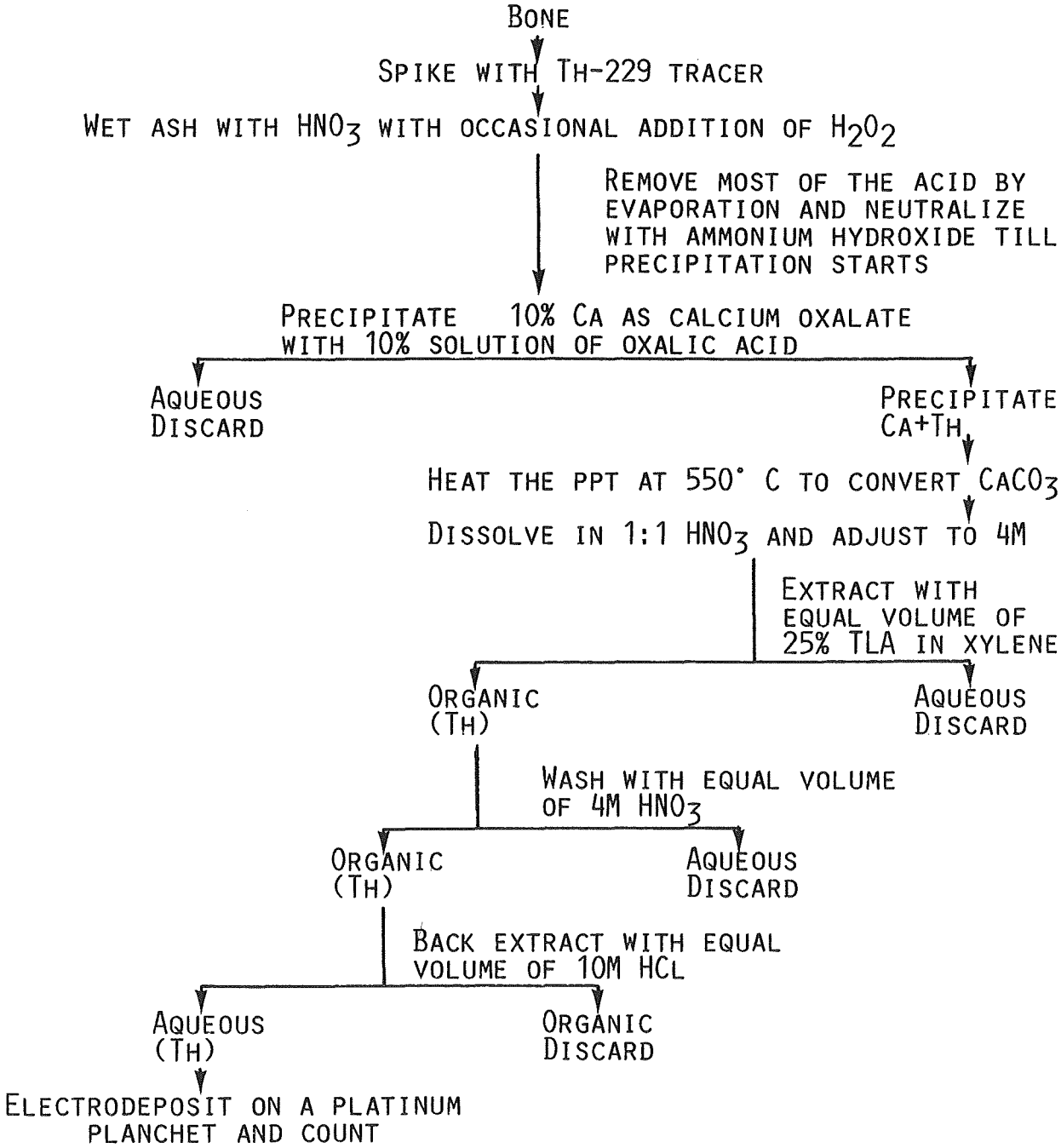
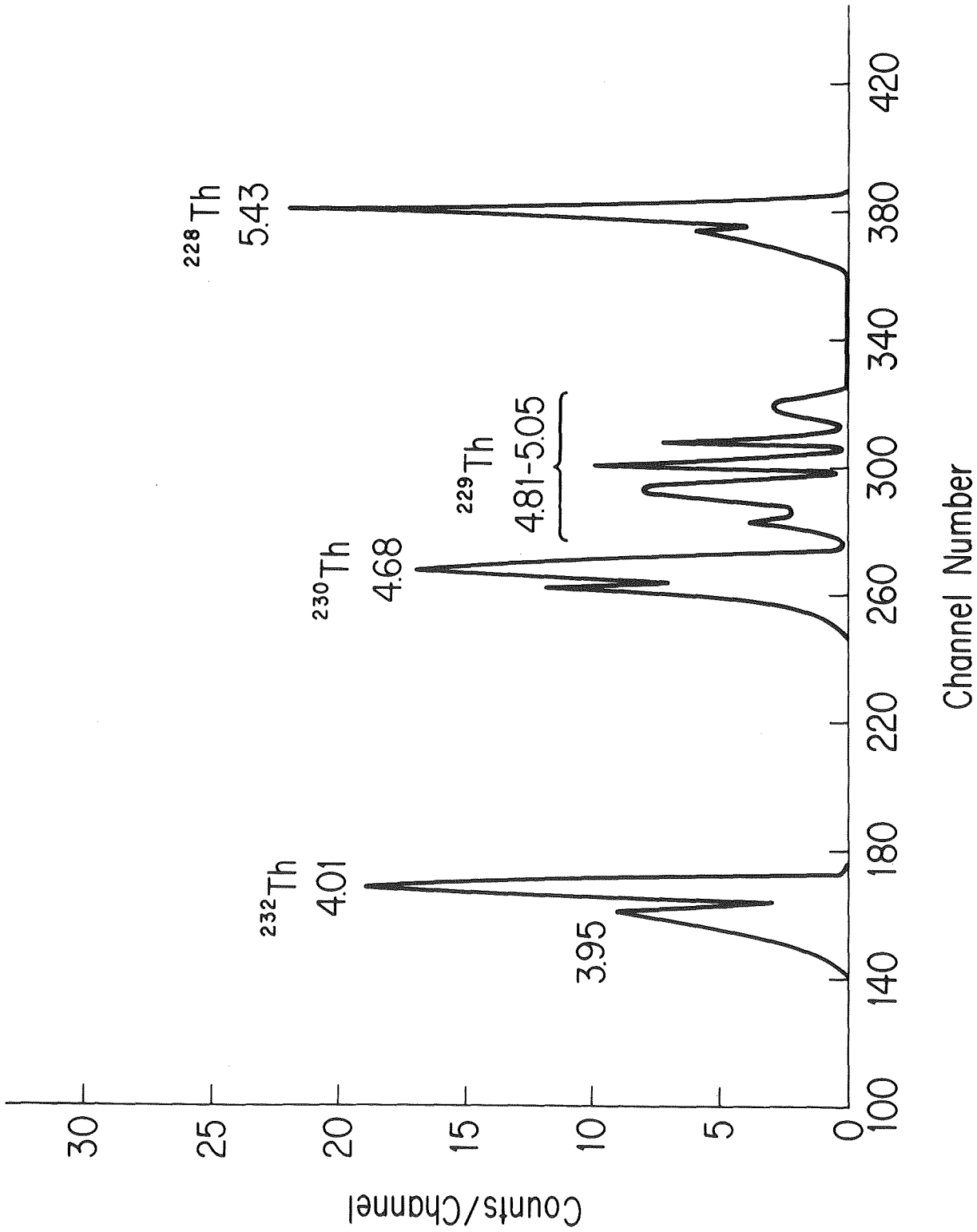


FIGURE 3.6: Alpha Spectrum of a Human Lung Containing Th-228, Th-230, Th-232, and the Added Th-229 Tracer



Recovery of Po-208 Tracer for Different Experimental Conditions

Experimental Condition	Po-208 in dpm Added	Recovered	% Recovery
(1) Direct transfer to a platinum disc and flamed over a burner	7.652	0.102	1.33
(2) Electrodeposited from sulfuric acid as described in the procedure	20.113	0.352	1.70
(3) Extracted from acid blank spiked with Th-229 as described in the general procedure	27.293	0.136	0.50

TABLE 3.2
Recovery (%) of Thorium in Different Organs

Organs	Thorium Mean ± 1 σ
Lung	41 ± 15 (29)*
LN	38 ± 16 (24)
Liver	39 ± 16 (26)
Kidney	53 ± 23 (25)
Bone (Vertebra)	43 ± 19 (23)
Spleen	48 ± 15 (14)
Gonads	64 ± 22 (4)

* The number in the bracket indicates the number of samples analyzed for each organ.

3.4 Data Presentation and Associated Error

The activity of each isotope of thorium was determined after integrating the counts in the appropriate energy regions of the nuclide of interest. Each energy region or channel spread was determined by counting a mixed standard source containing Pu-242, Pu-239,240, and Pu-238 which provided an energy calibration of the system. Identical procedures were used for determining counter background and reagent blank counts in the thorium and plutonium energy regions.

Counter background and reagent blank counting rates seen in the energy regions of interest were subtracted from the gross sample count rate before arriving at a net count-rate estimate. This sample count-rate estimate was then divided by the product of counter efficiency x chemical yield x sample mass in kilograms to arrive at an estimate of activity in the sample:

$$A = \gamma \frac{R}{EY} \tag{1}$$

where A = sample activity

R = net count rate (cpm in the energy region corresponding to the isotope being measured)

E = detector efficiency (cpm/dpm)

Y = radiochemical yield

γ = factor to convert dpm to activity.

The propagated random error in A can be obtained from Equation 1 by the standard techniques of propagating error (Be68) for products and quotients and is:

$$\frac{S^2A}{A^2} = \frac{S^2R}{R^2} + \frac{S^2(EY)}{(EY)^2} \tag{2}$$

Since the count rate (R) in the Th-228 region includes that contributed by Th-228 contamination of Th-229 tracer, which is added in order to make a radiochemical determination of yield (Th-228/Th-229 = 0.0814 on an activity basis) and also the background. Hence,

$$R = R_1 - R_2 - R_3 \tag{3}$$

where: R_1 = total count rate in the Th-228 region,
R_2 = count rate in the Th-228 region due to tracer, and
R_3 = count rate in the Th-228 region due to background.

The count rates of sample and background are simply:

$$R_1 = \frac{C_1}{t_1} \text{ and } R_3 = \frac{C_3}{t_3} \tag{4}$$

and the estimate of the standard deviation

$$s\ (R_1) = \sqrt{C_1}/t_1,\ s(R_3) = \sqrt{C_3}/t_3 \tag{5}$$

where C_1 is the total number of counts in the Th-228 region in counting time t_1.

$$R_2 = FR_0 \tag{6}$$

where F is the ratio Th-228/Th-229 established from measurement of the tracer ($F = 0.0814$ and $s(F) = 0.0038$ for this work), and R_0 is the count rate in the Th-229 region, and C_0 = total counts in the Th-229 region in time t_0.

Again propagating the error for Equation 6 gives:

$$\frac{s^2(R_2)}{R_2^{\,2}} = \frac{s^2(F)}{F^2} + \frac{s^2(R_0)}{R_0^{\,2}} \qquad (7)$$

R_3 is the background count in ~1000 min (the average counting time used for background); background counts are less than 1, so that $s^2(R_3)$ is of the order of 10^{-6} and can be neglected for our system.

By the standard rules for propagation of error for sums and differences, the random error associated with R from Equation 3 is:

$$s^2(R) = s^2(R_1) + s^2(R_2) + s^2(R_3) \qquad (8)$$

and $s^2(R)$ can now be readily calculated by combining Equations 5 and 7 with 8.

The cpm observed/dpm added is the product EY, so that if E is known, then Y may be calculated. In fact,

$$Y = \frac{R_0}{ET}$$

The error is propagated in the usual fashion for products and quotients.

In short, the error in A (Equation 2) consists of two terms, comprising the contributions from the error in observed count rate and the product of yield and efficiency. The relative importance of the two as sources of error depends on the activity in the sample and the amount of added tracer.

3.5 Critical Level for Detection of Net Activity

Measurements of environmental levels of radioactivity in human tissues are sometimes difficult because of the relatively low activity concentration and the small size of some organ of interest, e.g., lymph nodes, thyroid and gonads.

Considering samples whose activities produce low count rates, it is important to define a minimum detection limit (i.e., the count below which the sample activity is not distinguishably different from the background counting rate).

This level is what has been defined by Altshuler and Pasternack as the minimum significant measurable activity or critical level (L_c). The critical

level (L_c) is defined in terms of: 1) α, the probability of concluding activity is present when it is absent and 2) σ_s, the standard deviation of a net count when the true mean of the sample, μ_o, is zero. L_c is thus defined as:

$$L_c = K_\alpha \sigma_s$$

where the constant K_α is the upper percentile of the standardized normal distribution. The values for K_α can be found in tables of the normal distribution function (for a selected $\alpha = 0.05$, K_α is 1.645).

To calculate σ_s, the standard deviation of a net count when μ_o the true mean of the sample is zero, one must consider that for a zero activity sample the observed sample counts can be described as:

$$\mu_s = \mu_o + \mu_{BL} + \mu_{BK}$$

where: μ_s = the observed sample counts
μ_o = the mean of a zero activity sample
μ_{BL} = the mean of a net blank
μ_{BK} = the mean background.

The variance of μ_s can therefore be described as:

$$\sigma_s^2 = \sigma_o^2 + \sigma_{BL}^2 + \sigma_{BK}^2$$

where: σ_s^2 = the variance of the observed sample counts
σ_o^2 = the variance of the zero activity sample counts
σ_{BL}^2 = the variance of the blank counts
σ_{BK}^2 = the variance of the background counts.

Since four different radioelements were determined in this study, there are four separate critical levels which need to be calculated based on each nuclide's background and blank counts. Table 3.3 lists the mean counts plus or minus the variance for the background and blank measurements for each of the nuclides measured for a 2500 minute counting time.

From the values in Table 3.3, the total variance for each nuclide measurement can be calculated from:

$$S_s^2 = S_o^2 + S_{BK}^2 + S_{BL}^2.$$

For the inidividual nuclides this becomes:

Th-228 $S_s^2 = 5 + 2 + 1 = 8$ counts

Th-230 $S_s^2 = 2 + 1 + 1 = 4$ counts

Th-232 $S_s^2 = 2 + 1 + 1 = 4$ counts

For $\alpha = 0.05$, $k\alpha = 1.645$ and the critical level can be calculated to be:

Th-228 $Lc = 1.645\sqrt{8} = 4.7$ counts

Th-230 $Lc = 1.645\sqrt{4} = 3.3$ counts

Th-232 $Lc = 1.645\sqrt{4} = 3.3$ counts

If an estimate is made assuming:

1. an average chemical yield of 50% for thorium;
2. an average detector efficiency of 25%;
3. an average counting time of 2500 min.; and
4. the average count rate of the background and net blanks as listed in Table 3.3;

Then the Lc for each nuclide can be calculated as follows:

Th-228 $$Lc = \frac{9.4}{(2500)\ (.25)\ (.5)}$$

$$= 0.030 \text{ dpm/sample}$$
$$= 0.0136 \text{ pCi/sample}$$

Th-230 and Th-232

$$Lc = \frac{3.3}{(2500)\ (.25)\ (.5)}$$

$$= 0.010 \text{ dpm/sample}$$
$$= 0.005 \text{ pCi/sample}$$

Most of the samples analyzed in this study contained activity well above the critical level; the number of samples below that level are summarized in the following table and are marked with an asterisk in Appendix A. They adequately describe the distribution of each sample type, all measured values were reported regardless of their relationship to the critical level.

Number of Samples Below the Critical Level

Nuclide	Lung	LN	Liver	Kidney	Bone	Spleen	Gonads
Th-228	3/29	4/24	6/26	17/25	3/24	9/14	4/4
Th-230	-	1/24	-	2/25	1/24	-	1/4
Th-232	-	2/24	2/26	12/25	3/24	2/14	3/4

3.6 Analytical Quality Control

During the course of this study, an internal quality control program and participation in interlaboratory comparisons were in effect to document the validity of the analytical results. The data obtained for the quality control program included:

1. Blind Duplicates

A number of replicate analyses were performed on large beef liver samples. Each sample was dissolved in nitric acid, divided into three to seven portions, spiked with tracer, and submitted for analysis with different identification numbers. Since these samples are not carried through the analytical procedure together, they are not affected similarly by laboratory conditions, and hence give a good estimate of laboratory precision. The results on three different beef liver samples are summarized in Table 3.4. The precision of the analytical technique is seen in the results. The standard deviation for the mean of each pooled sample was within the error of an individual analysis.

2. Reagent Blank Samples

The main purpose of analyzing reagent blanks was to detect any external contamination. One blank was analyzed with each group of samples and counted for 300 minutes. A total of 15 reagent blanks were measured. The blanks gave average counts of 2 ± 2 for Th-228, 1 ± 1 for Th-230, and 1 ± 1 for Th-232.

3. Interlaboratory Comparisons

Internal quality control programs of replicate analyses gave the precision of the analytical technique; however, the accuracy of the method was determined by participation in interlaboratory comparison studies run by the U.S. Dept. of Energy Environmental Measurements Laboratory (EML).

In this program samples were prepared by the incorporation of carefully calibrated amounts of nuclides (at their environmental levels) and distributed among participating laboratories. The data from analyses are reported in a special EML publication annually.

The results for natural Th-228 concentration in a sample of animal bone ash reported for the quality control program by this laboratory was within 6% of the EML reference value.

TABLE 3.3

Mean* Counts for Net Reagent Blank and Counter Background Over Counting Time of 2500 min

(Counts ± S^2)

Radionuclide	Net Counts Reagent Blanks	Background Counts
Th-228	28 ± 5†	2 ± 1
Th-230	1 ± 1	1 ± 1
Th-232	1 ± 1	1 ± 1

* All reagent blanks and counter background counts represent the mean of 15 determinations. Reagent blanks were run with tracer.

† Th-228 counts contributed by the contamination of Th-228 (6%) resulted from the addition of 1-5 dpm Th-229 tracer.

TABLE 3.4

Results of Replicate Analyses of Thorium-228 Content in Beef Liver

Sample #	Percent Recovery Tracer Th-229	Total Th-228* (dpm)	Natural Th-228 in Liver (dpm)	Natural Th-228 pCi/kg Beef Liver
A-I	24±1	1.65±0.05	0.46±0.22	1.59±0.78
A-II	76±4	0.62±0.06	0.32±0.08	1.12±0.29
A-III	28±4	0.76±0.04	0.54±0.16	1.86±0.56
A-IV	75±6	0.51±0.06	0.38±0.08	1.61±0.33
A-V	93±4	0.50±0.01	0.27±0.06	1.14±0.25
A-VI	93±4	0.66±0.04	0.30±0.07	1.27±0.30
A-VII	81±3	0.76±0.05	0.29±0.08	1.20±0.35
	Mean 67%			Mean = 1.40±0.29
B-I	90±5	1.13±0.07	0.89±0.10	2.85±0.33
B-II	49±3	1.09±0.09	0.84±0.14	2.71±0.44
B-III	78±4	1.15±0.07	0.91±0.11	2.92±0.37
B-IV	59±4	1.38±0.10	1.14±0.16	3.65±0.50
	Mean 69%			Mean = 3.03±0.42
C-I	67±4	0.70±0.05	0.45±0.08	1.46±0.26
C-II	69±4	1.10±0.07	0.85±0.11	1.37±0.17
C-III	71±4	1.65±0.08	1.39±0.14	1.12±0.11
	Mean 69%			Mean = 1.32±0.18

* Total Th-228 represents that naturally occurring plus that added as a contaminant in the tracer.

CHAPTER 4

Stable Calcium Determination in Tissues

There have been some reports on the thorium content of human bone obtained from different anatomical bone samples (i.e., rib, vertebrae) (Lu70a, Hi62, Cl71). Some of these data were reported by wet weight, others by ash weight.

The ratio of calcium to ash weight among the various bone types has been shown not to vary greatly (Ho63). Thus the knowledge of the calcium concentration allows one to infer the ash weights from the wet weights with reasonable precision.

The calcium determination by atomic absorption is essentially that reported in EML manual (Ha78). The measurements of calcium were performed on 1% aliquots of the samples (lung, lymph nodes, liver, kidney, and bone) after they were wet ashed and solubilized. Each aliquot was diluted with 1% lanthanum solution (La_2O_3 in nitric acid). Lanthanum is used as a releasing agent to eliminate chemical interference of phosphorus, aluminum, silicon and sulfur and their anions in the analysis of calcium. Reagent blanks were also processed in the same manner. The instrument calibration was adjusted using a number of standard calcium solutions containing 0, 1, 2, 5, 8, 10 and 20 μg of calcium/mℓ. The mean absorbance value for each calcium standard was used to prepare a calibration curve by plotting absorbance vs. concentration of the standard.

The calcium concentrations of the tissue samples were obtained using the calibration curve corrected for dilutions, calcium in the blank, and the sample weight, and expressed as μgrams of calcium per gram of tissue fresh weight.

Results

The median concentrations for stable calcium in the lung, lymph node, liver, kidney, and vertebrae samples are summarized in Table 4.1. The highest calcium concentration was found in the vertebrae, followed by lymph nodes and lung. The calcium concentrations measured in this study are consistent with other published data (Fi79, IC75, An71, Ne72).

From this study, the wet vertebrae contains about 5.5% stable calcium, compared to 10% for the composite skeleton (ICRP75 and An71). However, on the basis of 14% dry ash in anatomical vertebrae sections (Bo74), the ratio of calcium to ash weight in the vertebrae obtained from this study was 0.39. This ratio is consistent with the following information compiled from the report of the task group on reference man:

	Wet Wt.(g)	Ash Wt.(g)	Calcium Contents (g)	(g) Calcium Per (g) Ash
Total skeleton	10,000	2,800	1,000	0.36
Mineralized bone	5,000	2,700	1,000	0.37
Cortical	4,000	2,200	800	0.36
Trabecular	1,000	500	200	0.40
Marrow	3,000	12	-	-
Cartilage and Connective Tissues	2,000	82	-	-

From this information, the calcium contents of the ash from different bones is reasonably invariant at about 37%. Mineralization of bone is a gradual process, and bone calcification is known to depend on age and diet (IC 75).

The concentrations of Th-228, Th-230, and Th-232 were expressed on the bases of the calcium concentrations in tissues (pCi/g Ca). The results indicate no obvious relationship between calcium and thorium concentrations in tissues.

TABLE 4.1
Median and Range of Calcium Concentration
Measured in Human Tissues
(μg calcium/g wet tissue)

Tissue Type	(n)	Median	Low	High
Lung	20	204	99	443
Lymph Nodes	10	782	156	1341
Liver	17	62	33	141
Kidney	19	162	95	351
Bone (vertebrae only)	16	58,000	7,200	119,000
Gonads	4	137	91	224
Spleen	9	132	17	617

CHAPTER 5

Thorium Concentrations Measured in Soil and Air

5.1 Results of Measurements of Thorium in Grand Junction Soil

Thorium isotopes were determined in six soil samples collected in 1978 from three different locations in Grand Junction. Two samples were obtained from each site; one from the surface (0-2 cm in depth) and the other at a depth of 10-15 cm. Each sample consisted of ten portions of a 1-2 ft^2 area taken with a small top soil cutter. The ten portions were composited to make a single sample weighing from 4 to 10 pounds. In addition, two samples of tailings were collected in 1979 from Grand Junction by the local DOE office. One was from the sand cover of the pile, and the other was taken from the slime. Figure 5.1 shows the location of the tailings pile and the soil sampling sites. Figure 5.2 shows a descriptive map of the tailings pile.

Before the analysis, samples were oven dried (90°), crushed and mixed to obtain uniformity. An aliquot of a few grams was weighed and spiked with Th-229 tracer. Duplicate samples were decomposed completely using the method described by Sill, et al. (Si74). The residue was dissolved in dilute hydrochloric acid and the thorium was then coprecipitated by the addition of ammonium hydroxide. The radiochemical analysis was performed using the method described earlier in the general procedure.

The results obtained from the analysis of the soil and tailings samples are summarized in Table 5.1. Mean concentrations were found to be 1.0, 1.1, and 1.2 pCi/g of soil for Th-228, Th-230, and Th-232, respectively. The values for Th-232 in the soil samples are similar to the average reported for thorium (1 pCi/kg) in soils from various locations in the United States (NC76). The variation of thorium concentration in soils is treated in more detail in Chapter 2. The Th-230/Th-232 ratio in soil was 1.1 with a range from 0.5 to 1.9. The Th-230 concentration in the slime sample taken from the pile was about 2000-fold higher than in the soil.

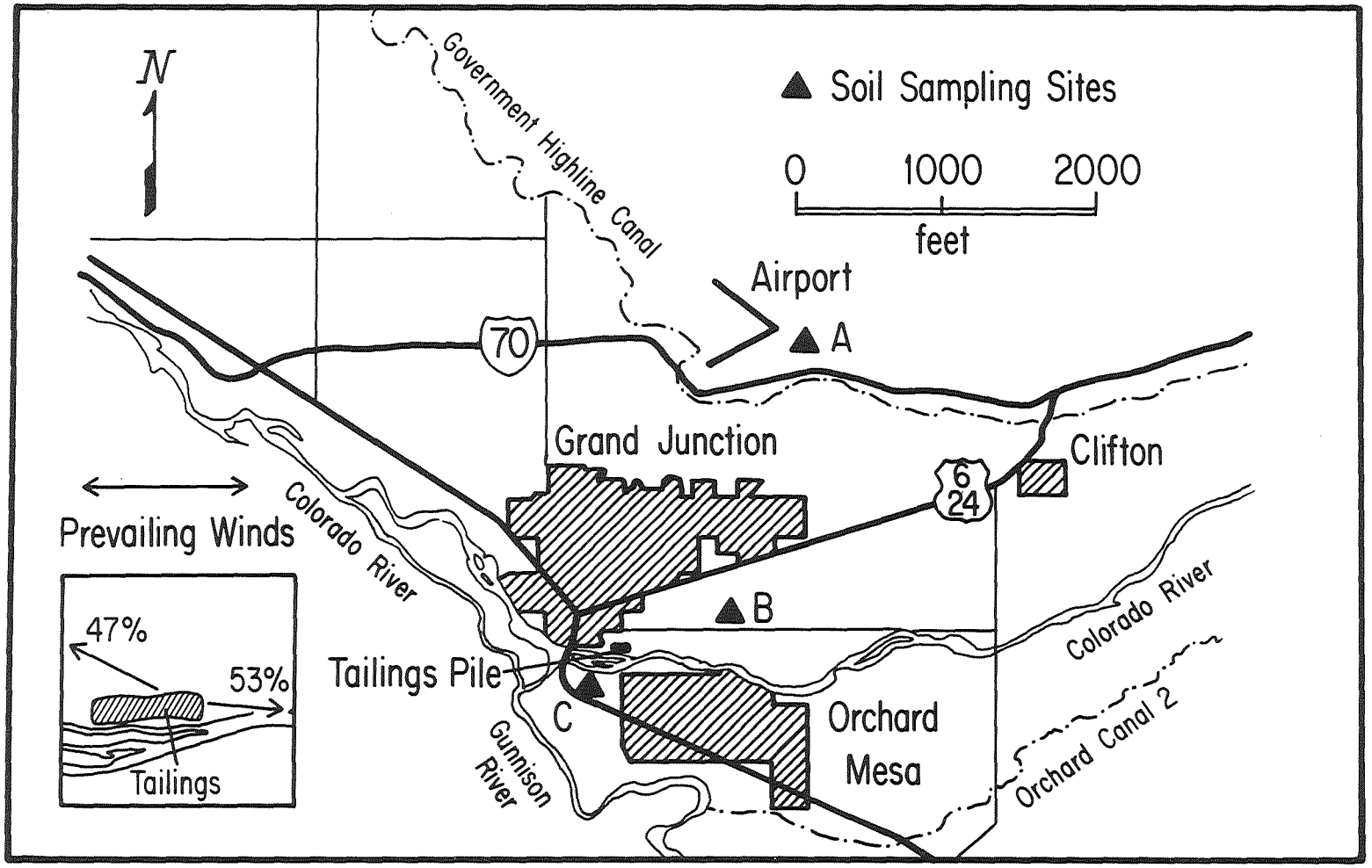
The error associated with Th-228 measurement in tailings was high because a large amount of Th-229 tracer (contaminated with about 6% Th-228) was required to measure the elevated Th-230 concentration. Note also that the error reported on the Th-232 concentration is also high because of the limited count rate that is associated with the small sample size and with the interference from the Th-230 in this sample.

The Th-230/Th-232 ratio in the tailings was about 400 (ranging from 265 to 1070) indicating that the thorium isotopic composition in the tailings pile is two to three orders of magnitude higher than normal in soil from Grand Junction. The ratio Th-230/Th-232 in the sand-cover sample from the uranium tailings pile over about 75 times higher than the normal soil.

Thus, Th-230 and Th-232 concentration and ratios in the 3 Grand Junction soils sampled appear in the range of normal U.S. reported values; the concentration of Th-230 is elevated in tailings 2000-fold and the ratio of Th-230/232 about 400-fold.

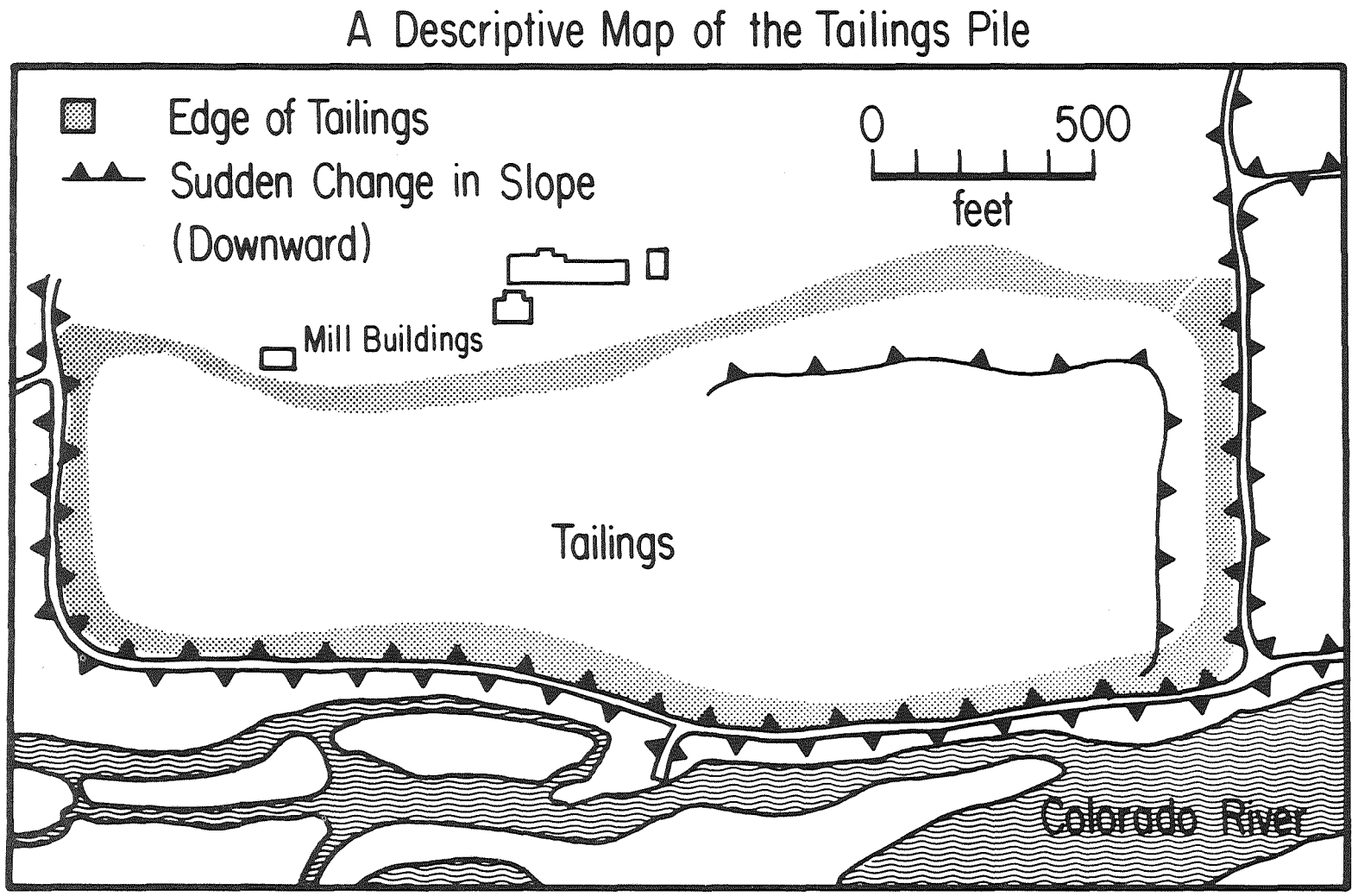
FIGURE 5.1: Location of Soil Sampling Sites in Grand Junction, Colorado

The Location of the Soil Sampling Sites at Grand Junction, Colorado



(Courtesy DOE Regional Office, Grand Junction, Colorado)

FIGURE 5.2: Descriptive Map of the Tailing Pile



5.2 Results of Thorium Measurements in Air Samples and Human Lungs from New York City

Three week long air samples were collected during the winter (December-February) of 1978-1979 from New York City. The total suspended particulate (TSP) was collected on 8 x 10" Gelman type A-E glass fiber filters mounted on a high-volume air sampler. The vacuum source was a roots pump designed to produce a nearly steady flow. The system included a pressure-sensing device (photohelic gage) calibrated to adjust flow to about 50 cfm. The average air volume sampled through each filter was about 5000 m^3. The sampler was placed on the roof (above the 14th floor) of New York University Medical Center, located in downtown Manhattan, 150 feet west of the East River. The sampling train and instrument calibration has been described in detail by Bernstein, et al. (Be76). The filters were wet ashed and leached with a mixture of concentrated nitric and hydrochloric acids (3:1); the leach solution was then analyzed as described in the radiochemical procedure. It was not possible to obtain air samples from Grand Junction and Washington, D.C. as part of this study.

The results of thorium isotopic concentration of the air samples are summarized in Table 5.2. The average concentrations of leachable thorium in air were 36 ± 14, 36 ± 22, and 37 ± 31 aCi/m^3 for Th-228, Th-230, and Th-232, respectively. The results are similar to that reported by Sedlet, et al. (Se73) for the annual average concentration near Chicago of 30 ± 15, 45 ± 23, 30 ± 15 for Th-228, Th-230, and Th-232, respectively.

The mean of the Th-230/Th-232 concentration ratios in suspended particles in New York City air was 1.0 ± .2; this ratio is similar to that found for three human lungs analyzed from the same area (0.9 ± .2) as seen in Table 5.3.

TABLE 5.1

Thorium Isotopic Concentrations in Soil and Tailings Samples from Grand Junction (in pCi/g)

Sample Identification and Location	Th-228	Th-230	Th-232	Ratio of 230/232
Soil A: surface	0.35 ± 0.02	0.63 ± 0.03	0.34 ± 0.02	1.9 ± 0.14
Soil A: at 15-cm depth	0.38 ± 0.02	0.46 ± 0.03	0.34 ± 0.03	1.3 ± 0.14
Soil B: surface	1.6 ± 0.07	1.0 ± 0.04	1.9 ± 0.06	0.5 ± 0.03
Soil B: at 15-cm depth	1.2 ± 0.04	0.72 ± 0.04	1.3 ± 0.05	0.6 ± 0.04
Soil C: surface	1.1 ± 0.05	2.6 ± 0.06	2.0 ± 0.05	1.3 ± 0.05
Soil C: at 15-cm depth	1.7 ± 0.07	1.6 ± 0.05	1.5 ± 0.05	1.0 ± 0.04
Average concentration ± 1σ	1.05 ± 0.57	1.1 ± 0.78	1.2 ± 0.73	1.1 ± 0.52
Sand cover of the tailings pile	2.00 ± 0.1	158 ± 10.3	2.1 ± 1.1	75 ± 39
Slime sample from the tailings pile	N.A.	2250 ± 19	5.3 ± 3.2	400 ± 256

TABLE 5.2

Thorium Isotopic Concentrations in New York City Air and a Blank Filter

Filter	Isotope	Concentration aCi/m^3	Ratio of Th-230/Th-232	Ratio of Th-232/Th-228
1	Th-228	25±9.9		
Collected	Th-230	22±3.8	1.0±0.2	0.9±0.4
Dec. 1978	Th-232	22±3.8		
2	Th-228	31±8.5		
Collected	Th-230	22±3.2	1.3±0.2	0.7±0.2
Jan. 1979	Th-232	17±2.7		
3	Th-228	52±22		
Collected	Th-230	64±17	0.9±0.3	1.2±0.5
Feb. 1979	Th-232	72±15		
Blank	Th-228	0.03±0.02 pCi/filter		
Filter	Th-230	0.02±0.01 pCi/filter		
	Th-232	0.01±0.01 pCi/filter		
Mean±1σ	Th-228	36±14		
	Th-230	36±22	1.0±0.2	1.0±0.3
	Th-232	37±31		

TABLE 5.3

Thorium Concentration in Three Lung Samples
Analyzed from the New York Area
(pCi/kg)

Sample Number	Isotope	Lung Concentration pCi/kg	Ratio of Th-230/Th-232	Ratio of Th-232/Th-228
N.Y. 1	Th-228	0.09±0.02		
age 68y	Th-230	0.17±0.07	0.6±0.4	3.0±0.6
F smoker	Th-232	0.27±0.09		
N.Y. 2	Th-228	0.33±0.04		
age 68 y	Th-230	0.37±0.03	0.9±0.1	1.2±0.2
M non-smoker	Th-232	0.40±0.04		
N.Y. 3	Th-228	1.00±0.17		
adult	Th-230	1.40±0.11		
male			1.1±0.1	1.3±0.2
	Th-232	1.30±0.11		
mean±1 σ	Th-228	0.50±0.50		
	Th-230	0.66±0.60	0.9±0.23	1.8±0.6
	Th-232	0.66±0.50		

CHAPTER 6

Results of Thorium Measurements in Human Tissues

Introduction

The main objective of this study was to determine the concentration of naturally-occurring thorium isotopes Th-228, Th-230, and Th-232 in human tissues. The tissues generally included lung, lymph nodes, liver, kidney, and bone. To establish this background concentration in the general population, two locations in the United States were selected: a western town (Grand Junction, Colorado) in which there is a uranium tailings pile associated with an inactive mill, and a location in the east (Washington, D.C.) where no unusual sources of thorium are believed to exist. The results of the tissue analysis for thorium obtained from Grand Junction will be compared with the results from the eastern group. As will be seen, the two groups are not comparable in age or occupation; thus the Washington, D.C. group is not an optimal control.

6.1 Characteristics of the Populations Sampled

The tissue samples were collected at autopsies from 32 cases during 1977, 1978, and 1979 from two locations in the U.S. Ideally, when two populations are compared, variables such as age, sex, occupation, and smoking history should be as nearly matched as possible. However, close matching was not practicable. The differences between the two groups are as follows: The age at death of the persons considered in this study was an uncontrolled variable. The individuals from Washington, D.C. were much younger; this may be due to the fact that they are largely from traumatic deaths which occur more frequently in younger males. The Grand Junction subjects were older than the Washington, D.C. subjects (median age 67 vs. 33 years); the number of smokers was 50% vs. 80% and contained a greater proportion of males (77% vs. 60%). The age distribution is particularly important because the concentration in human bone has been shown to increase with age (Lu70a).

Age Group	Grand Junction	Washington, D.C.
1-15	-	1
16-30	1	2
31-45	1	5
46-60	5	1
Over 60	15	1
Median Age	67	33
% Males	77	60
% Smokers	50	80

Information on the age, smoking, occupation, and residence time were not available in all cases. The urban population sampled from Washington, D.C. with a mean residence time of 15 years was a more transient one than that of Grand Junction, with a mean residence time of 45 years.

6.2 Estimates of Central Tendency and the Appropriate Statistical Test

For the purpose of comparing populations, the median (50th percentile) has been used to describe the central tendencies because it is not sensitive to extreme values (large or small). The geometric standard deviation (σg) is reported as a measure of the dispersion. In addition, the arithmetic average and the standard deviation are occasionally reported to show the degree of skewness of the distribution. The medians were used in the radiation dose calculations.

Samples from persons with an occupational history suspected to produce high Th-230 exposure (i.e., mining and/or milling) were excluded from the population under study.

Since most of the data was found to be adequately fit by a log normal distribution, statistical tests based on the normal distributions were applied to the log transformed data in some case as an adjunct to the non-parametric tests.

As an example, to test the hypothesis that the the ratio of Th-230/Th-232 in lymph-nodes in the subjects of Grand Junction and Washington, D.C. is different, two different statistical approaches were used:

a. A non-parametric method (Wilcoxon Rank Sum Test)
b. Normal-based statistical tests with logarithmic transformation of data. The "Student's" t test and the one factor analysis of variance were used.

As a result of the above testing, the hypothesis was rejected by both tests. Nonparametric procedures were used for most cases because of their independence of the population distribution.

The Wilcoxon Rank Sum Test is designed to test the hypothesis that two random samples were drawn from populations having identical distributions. The Rank Sum Test does not depend upon assumptions of normality or equal variances. The Rank Sum Test does not use the actual values of the observations, only their relative magnitude. In spite of this, theoretical investigations have established that even when normality and equal variance are obtained, the rank sum procedure has power only slightly less than that of the t Test (Re70).

6.3 Concentration of Thorium in Tissues for the Cases Without Known Occupational Exposure

The tissue samples were analyzed radiochemically. The method is discussed in detail in Chapter 3. The α-emitting thorium isotopic activity measured in each sample was converted to a concentration (pCi/kg), by dividing the activity by the weight of the fresh tissue. The detailed data for the individual tissue samples analyzed are given in Appendix A.

The concentration of thorium isotopes in the organs sampled was found to be adequately fit by a lognormal distribution, using the Kolmogorov-Smirnov Goodness of Fit Test (Ma51). The cumulative frequency distribution of the measured tissue concentrations is shown graphically in Appendix B.

The range, 50th percentile (median), and mean concentrations of Th-228, Th-230, and Th-232 by organ are given in Tables 6.1, 6.2, and 6.3. The 50th percentile concentration of the thorium isotopes in tissues in order of decreasing concentrations were:

Isotope	Location	Order of Decreasing Concentration
Th-228	Grand Junction	Lymph nodes>bone>lung>kidney>liver
	Washington, D.C.	Lymph nodes>bone>lung>kidney>liver
Th-230	Grand Junction	Lymph nodes>bone>lung>kidney>liver
	Washington, D.C.	Lymph nodes>lung>bone>liver>kidney
Th-232	Grand Junction	Lymph nodes>lung>bone>kidney>liver
	Washington, D.C.	Lymph nodes>lung>bone>liver>kidney

The concentration of Th-228 in the Grand Junction subjects was highest in the lymph nodes followed by bone and lung with medians of 5.1, 0.54, and 0.28 pCi/kg, respectively. In the Washington, D.C. subjects, the corresponding median concentrations were 2.6, 0.06, and 0.24 pCi/kg for the pulmonary lymph nodes, bone, and lung, respectively.

The concentration of Th-228, Th-230, and Th-232 in liver, kidney, spleen, and in four testicles appears to be smaller than that found in other tissues. On the other hand, the concentrations of Th-228, Th-230, and Th-232 in one thyroid sample from Grand Junction were 0.33±0.5, 0.82±0.37, and 0.65±0.33, respectively. The concentration of thorium isotopes in thyroid (based on one measurement) is higher than that found in other soft tissues except the lymph nodes and lung.

The data shows that thorium isotopes are concentrated in the pulmonary lymph nodes more highly than other tissues. The fact that thorium is highly discriminated against at the GI tract and that its isotopes are retained in the lymph nodes at a similar isotopic ratio to that found in air and soil

suggest that inhalation is the primary pathway of intake by man for Th-230 and Th-232.

The higher concentration of Th-228 in bone than lung is evidence of the ingrowth from ingested Ra-228. There is direct evidence that Th-228 and Th-232 in the air are in radioactive equilibrium (Se73 and this study). The ratio of Th-228/Th-232 concentration in human tissues would be expected to be similar to that found in air if inhalation were the only route of thorium uptake. This ratio, if elevated in human tissues, is likely to be due to the ingrowth of Th-228 from unsupported Ra-228.

The Th-228/Th232 concentration ratio has been calculated for the tissues from each individual, and the median of the ratios for both the Grand Junction and Washington, D.C. populations are:

Median of the Ratios of Th-228/232 in Tissues

Tissue	Washington, D.C.	Grand Junction
Lung	0.81	0.61
Lymph Nodes	0.86	0.53
Liver	1.9	1.5
Kidney	2.4	1.2
Bone	3.8	4.0
Spleen	NA	0.90

The median Th-228/Th-232 concentration ratio in lung and lymph nodes were less than one. The median ratio for liver, kidney and bone ranged from 1.5 to 4.0. The data suggests that Ra-228 from ingestion is contributing to the higher Th-228 concentration in all tissues except lung and lymph nodes. The difference in Th-228/Th-232 concentration ratio by organ between the Grand Junction and Washington, D.C. populations is not significant.

6.4 Thorium Concentration in Two Occupational-Exposure Cases

Two of the cases analyzed from the western location showed clearly elevated Th-230 concentrations in all tissues, compared to the rest of the subjects. The first was a hard-rock miner (age: 72 years); the concentration of Th-230 in his tissues was almost an order of magnitude higher than the median concentration of the other cases analyzed from the same location. The second case had been a uranium miner for 15 years (age: 59 years); his Th-230 concentration was more than two orders of magnitude higher than the median of the general population as shown in Table 6.4. In both cases, Th-228 and Th-232 concentrations were slightly elevated when compared to those of the rest of the subjects. The highly elevated Th-230 concentration is presumably caused by the higher exposures to uranium-bearing minerals. Nevertheless, the tissue distribution among organs for the thorium isotopes remains the same for

these cases as for the rest of the Grand Junction subjects; the highest concentration was in lymph nodes, followed by lung and bone.

The results obtained from these two cases were eliminated from the calculation of the 50th percentile concentrations previously because their exposures were clearly of an occupational origin. The elevated Th-230 concentration in both cases reflects the great relative abundance of Th-230 in their working environments.

It can be concluded that occupational exposure to uranium and to other mining activities can be a source in a significant elevation of Th-230 in tissues as compared to background amounts. When there was any evidence of occupational exposure from the autopsy information sheet, the case was classified as such and does not appear in statistical analysis of the data. Clearly, the inclusion of tissue from persons having had occupational exposures but unidentified as such can greatly change the average results in a group of samples.

TABLE 6.1

Concentration of Thorium-228 in Autopsy Samples
(pCi/kg wet weight)

Organ	# Samples Analyzed	Range	Mean	Stand. Dev. of Mean σ	Median	Geom. Stand Dev. σ_g
Grand Junction Subjects						
Lung	20	0.02-1.8	0.37	0.40	0.28	3.4
Lymph node	14	1.1-17.0	6.1	4.3	5.1	2.1
Liver	16	(-)0.01-0.51	0.13	0.14	0.07	3.0
Kidney	17	0.02-0.3	0.12	0.10	0.07	2.2
Bone	16	0.04-1.8	0.70	0.50	0.54	2.5
Testicles	4	(-)0.09-0.09	0.06	0.15	0.02	-
Thyroid	1	-	0.33	-	0.33	-
Spleen	14	(-)0.07-0.37	0.09	0.12	0.06	4.1
Washington D.C. Subjects						
Lung	10	0.04-0.40	0.23	0.13	0.24	2.3
Lymph node	10	(-)0.01-9.4	3.4	2.5	2.6	1.9
Liver	10	0.01-0.30	0.12	0.10	0.09	2.7
Kidney	8	0.03-0.36	0.12	0.10	0.09	2.0
Bone	7	0.20-1.4	0.77	0.40	0.66	1.5

TABLE 6.2

Concentration of Thorium-230 in Autopsy Samples

(pCi/Kg wet weight)

Organ	# Samples Analyzed	Range	Mean	Stand. Dev. of Mean σ	Median	Geom. Stand Dev. σ_g
Grand Junction Subjects						
Lung	19	0.10-6.0	1.4	1.4	0.8	2.6
Lymph node	14	5.5-25.0	12.0	5.3	11.0	2.6
Liver	16	0.02-0.57	0.20	0.18	0.15	2.8
Kidney	17	0.01-0.63	0.33	0.18	0.29	3.2
Bone	17	0.10-2.7	1.2	0.87	0.92	2.6
Testicles	4	0.02-0.15	0.088	0.046	0.09	-
Thyroid	1		0.82	-	0.82	
Spleen	14	0.05-0.43	0.28	0.30	0.13	2.1
Washington, D.C. Subjects						
Lung	10	0.08-0.71	0.36	0.20	0.36	1.9
Lymph node	10	0.35-11.0	5.6	2.5	4.6	1.9
Liver	10	0.03-0.70	0.24	0.23	0.11	2.8
Kidney	8	0.03-0.37	0.18	0.12	0.17	2.3
Bone	7	0.17-0.85	0.42	0.31	0.32	1.2

TABLE 6.3

Concentration of Thorium-232 in Autopsy Samples

(pCi/Kg wet weight)

Organ	# Samples Analyzed	Range	Mean	Stand. Dev. of Mean σ	Median	Geom. Stand. Dev. σ_g
Grand Junction Subjects						
Lung	19	0.02-2.8	0.60	0.60	0.58	3.3
Lymph node	14	2.0-23.0	9.7	5.9	7.8	1.9
Liver	16	0.01-0.38	0.08	0.11	0.03	3.6
Kidney	16	0.01-0.23	0.11	0.09	0.07	3.8
Bone	16	0.03-0.33	0.16	0.09	0.16	1.8
Testicles	4	0.02-0.07	0.05	0.02	0.05	-
Thyroid	1	-	0.65	-	0.65	-
Spleen	14	0.02-0.27	0.10	0.08	0.09	2.8
Washington, D.C. Subjects						
Lung	10	0.07-0.52	0.25	0.16	0.32	2.4
Lymph node	10	0.12-5.8	3.1	1.8	2.8	2.1
Liver	10	0.01-0.20	0.06	0.06	0.05	2.6
Kidney	8	0.01-0.12	0.05	0.04	0.03	2.0
Bone	7	0.00-0.20	0.097	0.064	0.10	3.0

TABLE 6.4

Thorium Isotopic Concentration in Two Occupational Cases and the General Population of Grand Junction, Colorado (pCi/kg)

	Lung			Lymph nodes			Liver		
*	Th-228	Th-230	Th-232	Th-228	Th-230	Th-232	Th-228	Th-230	Th-232
A	1.1 ±0.18	54.0±0.81	1.4 ±0.13	NA	NA	NA	0.25±0.04	32.0 ±0.36	0.12±0.2
B	0.70±0.24	12.0±0.79	0.61±0.18	12.0±2.4	37.0±3.9	4.6 ±1.4	0.05±0.01	0.82±0.07	0.06±0.02
C	0.21	0.88	0.37	4.8	13.0	8.1	0.08	0.13	0.07

	Spleen			Bone			Kidney		
*	Th-228	Th-230	Th-232	Th-228	Th-230	Th-232	Th-228	Th-230	Th-232
A	0.69±0.18	32.0±1.0	0.80±0.15	0.24±0.3	132.0±1.1	0.42±0.06	0.11±0.05	10.0±0.40	0.09±0.04
B	0.06±0.02	1.5±0.16	0.12±0.04	0.54±0.13	10.0±0.40	0.32±0.07	0.09±0.02	1.4±0.15	0.11±0.04
C	0.06	0.13	0.09	0.54	0.89	0.20	0.09	0.23	0.07

* A = Uranium miner.

B = Hard-rock miner.

C = 50th percentile for the general population.

CHAPTER 7

A Comparison of Thorium Concentration in Tissues from Grand Junction and Washington, D.C.

The median for Th-230 and Th-232 in Grand Junction tissues (see Chapter 6.1 for details) exceeded those from Washington, D.C. Hypotheses about the reasons for the variability of the concentration of thorium isotopes between tissue samples from the two populations can be tested with the observed data.

Hypothesis 1:

The concentrations of Th-230 and Th-232 in Grand Junction samples by organ are significantly different from those from Washington, D.C.

Test of Hypothesis 1:

The results of application of the Wilcoxon Rank Sum Test is summarized in Table 10.1. The hypothesis was accepted for Th-230 and Th-232 for lung, lymph nodes, kidney, and bone, and rejected for Th-228 for all tissues except lymph nodes at the P values specified in the table.

The median concentration of Th-230 and Th-232 in the lung, lymph nodes, kidney, and bone of Grand Junction subjects exceeded those of the Washington, D.C. subjects as indicated in Table 7.2.

One likely reason might be that the age distribution of the two groups is different; the median ages were 67 and 33 years for the Grand Junction and Washington, D.C. subjects, respectively. It will be shown in a latter section that both Th-232 and Th-230 in bone increase with age; thus, a higher concentration of both isotopes was expected in the Grand Junction samples than in those from Washington, D.C., particularly in bone. The concentration of Th-230 and Th-232 in bones from the subjects of both groups can be reasonably compared, even in the presence of age differences, by extrapolation based on a linear model of accumulation which is consistent with the information obtained in this study (see Section 11). For comparison, Washington, D.C. subjects had a median age of 33 and a median concentration of 0.32 pCi/kg, and 0.10 pCi/kg for Th-230 and Th-232 in bone, respectively. If the concentration in bone is extrapolated in a linear fashion to that expected at 67 years (the median age for Grand Junction subjects), a median concentration in bone of 0.64 and 0.2 pCi/kg for Th-230 and Th-232 would be expected. This estimate is comparable to that of Th-232 (0.16 pCi/kg) and slightly less for Th-230 (0.92 pCi/kg) observed in Grand Junction bones.

		Concentration in pCi/kg		
		Grand Junction	Washington, D.C.	
Organ	Isotope	Median Age 67 y	Median Age 33 y	Age Adj. at 67 y
Bone	Th-230	0.92	0.32	0.64
Bone	Th-232	0.16	0.10	0.20

The accumulation of thorium as a function of age or exposure to dust should not affect the ratio of Th-230/Th-232 in tissues, particularly in the integrating organs, if the source of exposure does not change throughout the lifetime.

Hypothesis 2:

The ratios of Th-230/Th-232 in organs are significantly different between Grand Junction and Washington, D.C.

Test of Hypothesis 2:

The ratio of Th-230/Th-232 concentrations in the tissue samples from the two groups was calculated for each individual sample. The ratio should be independent of biological variations between individuals, and should be age-independent, but will depend on the source of the exposure.

The Wilcoxon Rank Sum Test, performed using the Th-230/Th-232 concentration ratios for the individuals of the two groups, showed no significant difference in this ratio between the two groups in any tissues except in bone at $P \leq 0.1$ which indicates marginal significance at best. Table 7.3 summarizes the mean and median of the Th-230/Th-232 ratio in the tissues for both groups, and for the same ratio calculated from tissues of a uranium miner. The median ratio was 3.7, 8.4, and 310 in the bone samples from Washington, D.C., Grand Junction, and the uranium miner, respectively.

Because the skeleton should accumulate thorium throughout life, and the lung and lymph nodes reflect more recent exposures, the elevated ratio of Th-230/232 observed in the skeleton (but not lungs) of the Grand Junction subjects may be a result of past exposures to an elevated Th-230 source. The elevated Th-230/Th-232 ratio from tissues of the uranium miner are an example of a substantially elevated Th-230 source relative to Th-232.

The elevated Th-230/Th-232 ratio in Grand Junction bone samples may have been caused during the years of active uranium mining or before the tailings pile was stabilized (by the sand cover), or both. To explore this hypothesis further, the concentration ratio of Th-230/Th-232 was calculated for the bone and lung sample from three younger subjects (age 26, 38, and 47 years) from Grand Junction. The average ratios were 3.5 and 1.9 for the bone and lung, respectively. These ratios are comparable to those found in the Washington D.C. subjects who had similar ages (see Table 7.3); thus, this may be considered as evidence to support the hypothesis that a slightly elevated Th-230/Th-232 ratio found in bone samples from Grand Junction may have been caused by past exposure to a source with elevated Th-230/Th-232.

If unknowingly slight occupational exposure may have occurred in some of the Grand Junction subjects, such an inclusion would contribute to a higher Th-230 concentration in their skeletons.

TABLE 7.1

Wilcoxon Rank Sum Test for Significance of Thorium Concentrations in Tissues: Grand Junction vs. Washington, D.C.

Isotope	Tissue Type	Results
Th-228	Lung	not significant, $P \geq 0.1$
	Lymph nodes	significant, $P \leq 0.1$
	Liver	not significant, $P \geq 0.1$
	Kidney	not significant, $P \geq 0.1$
	Bone	not significant, $P \geq 0.1$
Th-230	Lung	significant, $P \leq 0.1$
	Lymph nodes	significant, $P \leq 0.1$
	Liver	not significant, $P \geq 0.1$
	Kidney	significant, $P \leq 0.1$
	Bone	significant, $P \leq 0.1$
Th-232	Lung	significant, $P \leq 0.1$
	Lymph nodes	significant, $P \leq 0.1$
	Liver	not significant, $P \geq 0.1$
	Kidney	significant, $P \leq 0.1$
	Bone	significant, $P \leq 0.1$
Th-230/Th-232 ratio	Lung	not significant, $P \geq 0.5$
	Lymph nodes	not significant, $P \geq 0.5$
	Liver	not significant, $P \geq 0.5$
	Kidney	not significant, $P \geq 0.5$
	Bone	significant, $P \leq 0.5$

TABLE 7.2

The Median Concentrations of Thorium Isotopes
in Autopsy Samples from Grand Junction, Colorado, and Washington, D.C.
(in pCi/kg fresh weight)

Organ	# of Samples Analyzed	Grand Junction, Colorado			# of Samples Analyzed	Washington, D.C.		
		Th-228	Th-230	Th-232		Th-228	Th-230	Th-232
Lung	19	0.28	0.84	0.58	10	0.24	0.31	0.32
Lymph node	14	5.1	11.0	7.8	10	2.6	4.60	2.8
Liver	16	0.07	0.15	0.03	10	0.09	0.15	0.05
Kidney	17	0.07	0.29	0.07	8	0.09	0.17	0.03
Bone	16	0.54	0.92	0.16	7	0.66	0.32	0.10
Testicles	4	0.02	0.09	0.05	-	-	-	-
Spleen	14	0.06	0.13	0.09	-	-	-	-
Thyroid	1	0.33	0.82	0.65	-	-	-	-

TABLE 7.3

Median and Mean of Th-230/Th-232 Ratio in Organs from Grand Junction and Washington, D.C. and in a Uranium Miner

	Washington, D.C.			Grand Junction			
Organ	Median	Mean	S.D.	Median	Mean	S.D.	Uranium Miner
Lung	1.2	1.4	0.57	1.9	2.1	1.4	46*
Lymph nodes	1.8	1.9	0.65	1.4	1.5	0.77	
Liver	2.7	3.9	3.2	3.6	4.5	3.4	270
Kidney	5.8	5.8	3.5	3.1	3.9	3.0	110
Bone	3.7	4.2	2.4	8.4	7.5	4.5	310

* Lymph nodes were included in the lung sample.

CHAPTER 8

A Comparison of Thorium Concentration in Lungs from Smokers and Non-Smokers

Naturally-occurring α-activity in cigarette tobacco was first reported by Turner and Radley in 1960 (Tu60). Since then other investigators have measured the levels of Po-210, Ra-226, Th-228, Th-230, Th-232 and natural uranium in domestic and foreign tobacco (Tu64; Lu70; Ne79). Table 8.1 summarizes the results of these studies. Neton and Ibrahim (Ne78) have shown that Th-228 activity exceeds its parent Th-232 in tobacco by a factor of 10 to 20. This is presumably because Ra-228 is taken up from soil more avidly than Th-228 or Th-232 and subsequently Th-228 grows in from its immediate parent, Ra-228.

Various *in vivo* studies have been performed in man to determine the possibility of the presence of excess thorium in the lungs of cigarette smokers vs. those of non-smokers. To date, however, the data have been equivocal. Joyet (Jo71) reported measurable levels of thorium in the lungs of smokers, while the level of his non-smoking group was below his detection limit. Scott (Sc75) attempted to duplicate Joyet's findings, but he found no statistical differences in the levels of lung concentrations for thorium between smokers and non-smokers.

Kolber (Ko77) concluded that the normal variability among *in vivo* measurements for lung burdens of Pb-212 is too great to detect any difference between smokers and non-smokers.

To determine whether cigarette smoking effects the concentration of thorium in lung, the subjects were divided into two groups: first, the heavy smokers (which has been defined for the purpose of this analysis as those consuming at least one pack per day for ten years or more); and second, the group that included all known non-smokers. The data for the subjects from only Grand Junction were used in this test. Insufficient data was available to perform the statistical analysis for the Washington, D.C. subjects. Smoking habits were obtained from the individual information forms submitted by the pathologist with each autopsy case. The rank test was used to evaluate whether thorium concentrations found in the lungs of heavy smokers and of non-smokers were (statistically) significantly different.

Table 8.2 summarizes the median concentrations of Th-228, Th-230, and Th-232 in lungs of smokers and non-smokers. The median concentrations of these isotopes of thorium in lungs from non-smokers exceeded those for smokers, although the differences were not statistically significant ($p > 0.1$). The ratio of Th-228/Th-232 in lung was slightly higher for the non-smokers than the smokers.

On the basis of this limited information, cigarette smoking has no effect on the concentration of thorium isotopes in lung.

TABLE 8.1

Natural Alpha-Emitting Radionucides in Cigarette Tobacco

Nuclide	Mean (pCi/kg)	Range (pCi/kg)	Reference
Po-210	430	150-480	Ts64
Ra-226	210	59-390	Ts64
U (natural)	113	35-127	Lu69
Th-228	130	77-200	Ne78
Th-230	10	9-13	Ne78
Th-232	12	8-14	Ne78
Th-228/Th-232	11	9-15	Ne78

TABLE 8.2

The Median Concentration of Th-228, Th-230, and Th-232 in Lungs of Smokers and Non-Smokers (pCi/kg)

Nuclide	Smokers (n=7)	Non-smokers (n=7)
Th-228	0.22	0.37
Th-230	0.56	0.84
Th-232	0.43	0.60
Ratio of Th-228/ Th-232	0.51	0.62

CHAPTER 9

The Distribution of Thorium in the Human Body

The distribution of the thorium isotopes has not been measured in a complete human body; however the distribution can be constructed from thorium concentrations in autopsy samples, the "standard" organ weights in reference man, and data on relative tissue distribution in animals. Organ weights for reference man are summarized in Table 9.1. Based on animal studies the soft tissues most likely to contain thorium are lung, lymph nodes, liver and kidney. Stover et al. (St65) found around 10% of injected Th-228 was in muscle in the beagle. Therefore we assumed for man that 10% of the body burden would be found in soft tissues such as muscle, which we did not analyze.

The estimate of the thorium burden in the skeleton, based on the analytical results of a relatively small portion of the skeleton, is uncertain. In the absence of any information on the macro distribution of thorium (particularly at environmental levels) in different bones, it was considered reasonable although possibly incorrect to assume that thorium was distributed uniformly among all types of bones. Since we used vertebral wedge containing mineral bone and bone marrow in our analysis, the weight of the skeleton was taken as 8 kg for males (5 kg mineral, 3 kg marrow) and 5.8 kg for females.

The average population organ reference ($\bar{w}$) weight was calculated by:

$$\bar{W} = \frac{MW_1 + FW_2}{M + F}$$

where M is the number of males, F is the number of females, W_1 and W_2 are the ICRP-23 reference organ weights for males and females respectively (See Table 9.1). The total organ burden was then calculated by multiplying the median concentration by the average reference weight for the organ. The results are given in Table 9.2. The highest organ burden for all thorium isotopes was found in the skeleton followed by lungs. The body burden for both Th-230 and Th-232 was higher for the Grand Junction subjects than the Washington D.C. subjects. On the other hand, the total body burden for Th-228 in the Washington D.C. subjects was subsequently higher than that found in the Grand Junction subjects. This could be due to dietary differences in the Ra-228 concentrations between the two locations. The median total values of accumulation of thorium in the human adults were equivalent to the amount of thorium in 1 to 6 gms of normal soil.

The fractional distribution of the thorium isotopes in the different organs is presented in Table 9.3 for the Grand Junction and Washington, D.C. subjects.

Based on the animal work, an estimated fraction of 10% for the thorium was assumed to be in the body muscles (St60,65). Thorium-228 was present primarily in skeleton (83%); the remaining fraction was found in the pulmonary compartment (4-6%) and in liver (3%). Thorium-230 was distributed similarly; however, the skeletal fraction was lower (68-72%) and the fraction retained in

the pulmonary compartment was higher (13-15%) than that of Th-228. Thorium-232 was distributed to a lesser degree (58%) in the skeleton and to a higher degree (26%) in the pulmonary compartment.

The distribution of Th-230 and Th-232 between the skeleton and the pulmonary compartment, relative to that for Th-228, suggests that inhalation is the major route of uptake by man for these isotopes. On the other hand, the higher Th-228 fraction found in the skeleton, relative to that for Th-230 and Th-232, is presumably a result of ingrowth from Ra-228 taken up by man through diet.

The Th-232 percentage found in the lung and lymph nodes were higher than those of Th-230. Thorium-230 is a decay product of the uranium series, and the chemical and physical characteristics of its minerals may be considered different from those of thorium minerals which contain Th-232. Consequently, the differences in the extent of retention of Th-232 in the pulmonary compartment relative to Th-230 may be explained by: a) the selective *in vivo* solubilization of Th-230 (in the uranium minerals) is more rapid than natural thorium, and subsequently its preferential removal from the lungs and translocation to the skeleton; b) differences in the particle size distribution of the uranium and thorium minerals in the inhaled air. Larger and more dense particles tend to be removed or translocated to the lymph nodes, particularly in terms of insoluble materials, and this may be the case for the thorium minerals.

For the two occupational cases (the hard-rock miner and the uranium miner), although they had an elevated concentration of Th-230, their organ distribution patterns were similar to those of other non-occupational subjects.

The organ distribution pattern of thorium in man obtained in this study is in good agreement with the recommendations of ICRP Publication No. 30. This publication was based on the results of Stover, et al. (St60,65) in their study of the retention and distribution of injected thorium in adult beagle dogs (i.e., 80% in bone and 5% in liver). However, this is in contrast to the values suggested in ICRP Publication No. 19 based on the metabolic model for plutonium.

TABLE 9.1
Organ Weights of Reference Man*

Tissue or Organ	Organ Weight (grams)	
	Male	Female
Kidney	310	275
Liver	1,800	1,400
Lung	1,000	800
Lymph nodes	15	15
Spleen	180	145
Thyroid	20	20
Gonad-Testes (2)	35	-
Skeleton (wet with marrow)	10,000	6,800
(wet de-fatted)	8,000	5,800
(mineral bond)	5,000	3,400

* ICRP Publication No. 23 (1975).

TABLE 9.2

Median Organ-burden Estimates
for the Thorium Isotopes (pCi/Organ)

Organ	Th-228	Th-230	Th-232
Grand Junction Subjects			
Lung	0.25	0.80	0.50
Lymph nodes	0.08	0.16	0.12
Liver	0.13	0.25	0.05
Kidney	0.02	0.09	0.02
Spleen	0.01	0.02	0.02
Gonads (testicles)	0.0007	0.003	0.002
Thyroid	0.007	0.01	0.01
Skeleton	4.70	6.89	1.20
Washington, D.C. Subjects			
Lung	0.21	0.36	0.30
Lymph nodes	0.04	0.07	0.06
Liver	0.15	0.19	0.08
Kidney	0.03	0.04	0.01
Skeleton	4.70	2.28	0.71

TABLE 9.3

Median Derived Organ Distribution for the Thorium Isotopes (percent)

Organ	Th-228	Th-230	Th-232
Grand Junction Subjects			
Lung	5.0	8.8	23.4
Lymph nodes	1.6	1.8	5.6
Liver	2.6	2.7	2.3
Kidney	0.4	0.99	0.9
Spleen	0.2	0.21	0.9
Gonads (Testicles)	0.1	0.02	0.09
Thyroid	0.14	0.11	0.47
Muscle*	10.0	10.0	10.0
Skeleton	80.2	75.4	56.1
Total Thorium Contents of Body	(5.05 pCi)	(9.14 pCi)	(2.14 pCi)
Washington, D.C. Subjects			
Lung	3.7	11.0	23.3
Lymph nodes	0.70	2.1	4.7
Liver	2.6	5.8	6.2
Kidney	0.53	1.2	0.78
Muscle	10.0	10.0	10.0
Skeleton	82.5	68.7	55.0
Total Thorium Contents of Body	(5.70 pCi)	(3.27 pCi)	(1.29 pCi)

* Estimated fraction based on the animal studies (Stover et al., 1965).

CHAPTER 10

Radiation Dose from Environmental Thorium to Major Organs

The average dose rate to the soft tissues can be calculated in a relatively simple manner; Harley (Ha74) derived the following formula in convenient units for environmental work:

$$D = 18.7\ C\bar{E},$$

where D is the dose rate in mrad/yr, C is the activity concentration in tissue in pCi/g, and $\bar{E}$ is the energy absorbed per disintegration in MeV.

It is not possible to calculate the average dose rate accurately without some information about the degree of equilibrium of the Th-228 daughters. Accordingly, the calculations were made based on two extreme assumptions, namely, that no daughters were present, or that all the daughters were present in equilibrium. The effective energy absorbed per disintegration for α-emitting thorium isotopes are summarized in Table 10.1. The radiation dose rate from all thorium isotopes in the non-occupationally exposed subjects were calculated on the basis of the median concentration of thorium present in the analyzed tissues; the data are presented in Table 10.2.

The highest radiation dose from any thorium isotope was delivered to the lymph nodes, and was on the order of 1 to 5 mrads per year, assuming equilibrium of Th-228 and the daughters.

TABLE 10.1

The Effective Alpha Energies for the Thorium Isotopes (Le78)

Isotope	Energy (MeV)	Abundance (%)	Effective Energy (MeV)
Th-230	4.68	76*	3.56
	4.61	24	1.11
			4.67
Th-232	4.01	76†	3.04
	3.94	24†	0.94
			3.98
Th-228	5.34	28†	1.49
	5.42	72†	3.90
			5.39
Ra-224	5.19	0.4	0.02
	5.45	4.6	0.25
	5.68	95	5.40
Rn-220	6.28	100	6.28
Po-216	6.78	100	6.77
Bi-212	5.60	0.37	0.02
	5.62	0.05	0.003
	5.72	0.68	0.04
	6.05	23.8	1.44
	6.09	9.2	0.56
Po-212	8.78	66	5.79
Th-228 through Po-212			31.96 MeV

* Relative to total U-238 radiation.
† Relative to total Th-232 radiation.

TABLE 10.2

Median Dose Rate in mrad/yr for Thorium Isotopes in Tissue

Organ	Th-228 alone	Th-228 with Daughters in Equilibrium	Th-230	Th-232	Total Dose Rate Assuming No Th-228 Equilibrium	Total Dose Rate Assuming Equilibrium of Th-228 Daughters
In Grand Junction Subjects						
Lung	0.028	0.17	0.073	0.043	0.14	0.23
Lymph node	0.51	3.0	0.96	0.58	2.1	4.5
Liver	0.0071	0.042	0.013	0.0022	0.022	0.057
Kidney	0.0071	0.042	0.025	0.0052	0.037	0.072
Skeleton	0.054	0.32	0.080	0.012	0.15	0.41
In Washington, D.C. Subjects						
Lung	0.024	0.14	0.027	0.024	0.075	0.19
Lymph node	0.26	1.6	0.40	0.21	0.87	2.2
Liver	0.0091	0.054	0.013	0.0037	0.026	0.071
Kidney	0.0091	0.054	0.015	0.0022	0.026	0.071
Skeleton	0.067	0.40	0.028	0.0074	0.10	0.44

CHAPTER 11

Accumulation of Thorium in Bone with Age

The relationship between the thorium isotope content in bone samples and the ages of the subjects has been examined.

The primary mode of intake for Th-230 and Th-232 by man is most likely inhalation. According to the general actinide model given in ICRP No. 19 (IC72), the inhaled activity in the lung and lymph nodes should reach a state of equilibrium with the concentration in the atmosphere after a few years of normal intake and the concentration will not increase as a function of age. This is not the case, however, for the skeleton where the concentration is expected to increase as a function of age. The skeleton is considered the best integrating organ because of the long biological half-time of 8000 days (St60,62). For this reason, bone is the tissue that best reflects the accumulation of activity with time.

The concentration of Th-232 in bone has been measured in human rib samples by a few investigators (Lu70; Be67). In this study, the concentration of thorium isotopes was measured in samples which consisted of three or four vertebrae from the vertebrae column. The concentration of thorium isotopes in the bone samples analyzed were presented in Chapter 6.

Because measurements by previous investigators were reported in nanograms of Th-232 per gram of bone ash, their data were converted to pCi/g wet weight for comparison with the data obtained in this study. The ash weight of the human rib has been determined to be about 17% of the wet weight (La78). Using this factor and the specific activity for Th-232, the data from Lucas et al. (Lu70) and Petrow and Strehlow (Pe67) in pCi/Kg wet bone were:

Study	Bone Type	No. Samples	Median Age (Y)	Th-232 Conc. pCi/Kg wet
Lu70	Rib	38	54	0.12
Pe67	Composite Skeletons	1	unknown	0.19
This Study				
Grand Junction	Vertebrae	16	67	0.16
Washington D.C.	Vertebrae	7	33	0.10

Thus, the median concentrations of Th-232 in this study are similar to those reported by Lucas et al. and Petrow.

The Th-232, Th-230 and Th-228 concentrations in bone from the Grand Junction and Washington, D.C. samples were plotted against age at death and are shown in Figures 11.1, 11.2, and 11.3. The data obtained from occupationally-exposed individuals have been omitted from these plots. The scatter of thorium concentrations in bone according to age is observed from these figures; similar scatter was also noted by Lucas et al. The cause of this marked scattering may be explained by the physiological variations among the individual subjects and variations in the degree to which they were exposed to thorium inhalation in dust in their environments.

The lines shown in Figures 11.1, 11.2 and 11.3 have been obtained by standard least-squares analysis. The solid line in each graph represents the age-concentration relationship for bone for both the Grand Junction and Washington, D.C. samples combined, and the dashed line represents the Grand Junction samples only.

The linear accumulation of the Th-232 and Th-230 in the vertebrae is given by the following equation:

$$Y = AT + B$$

where Y = the nuclide concentration in pCi/Kg fresh bone weight

A = the slope of the regression line (pCi/kg/year)
T = age in years
B = the y-intercept at T = 0

The data for Th-232 and Th-230 show a generally increasing concentration with increase in age; if this relationship is assumed to be linear, then the Th-232 and Th-230 burdens are clearly very low at birth as indicated by the Y intercepts. The latter is reasonable based on the work of Sikov and Mahlum (Si68) which describes the placental discrimination of monomeric and polymeric plutonium; by analogy it is believed that the same discrimination may occur for the chemical congener thorium, and the newborn is portected against accumulation of Th-232 anmd Th-230 by this placental discrimination (Wr 77).

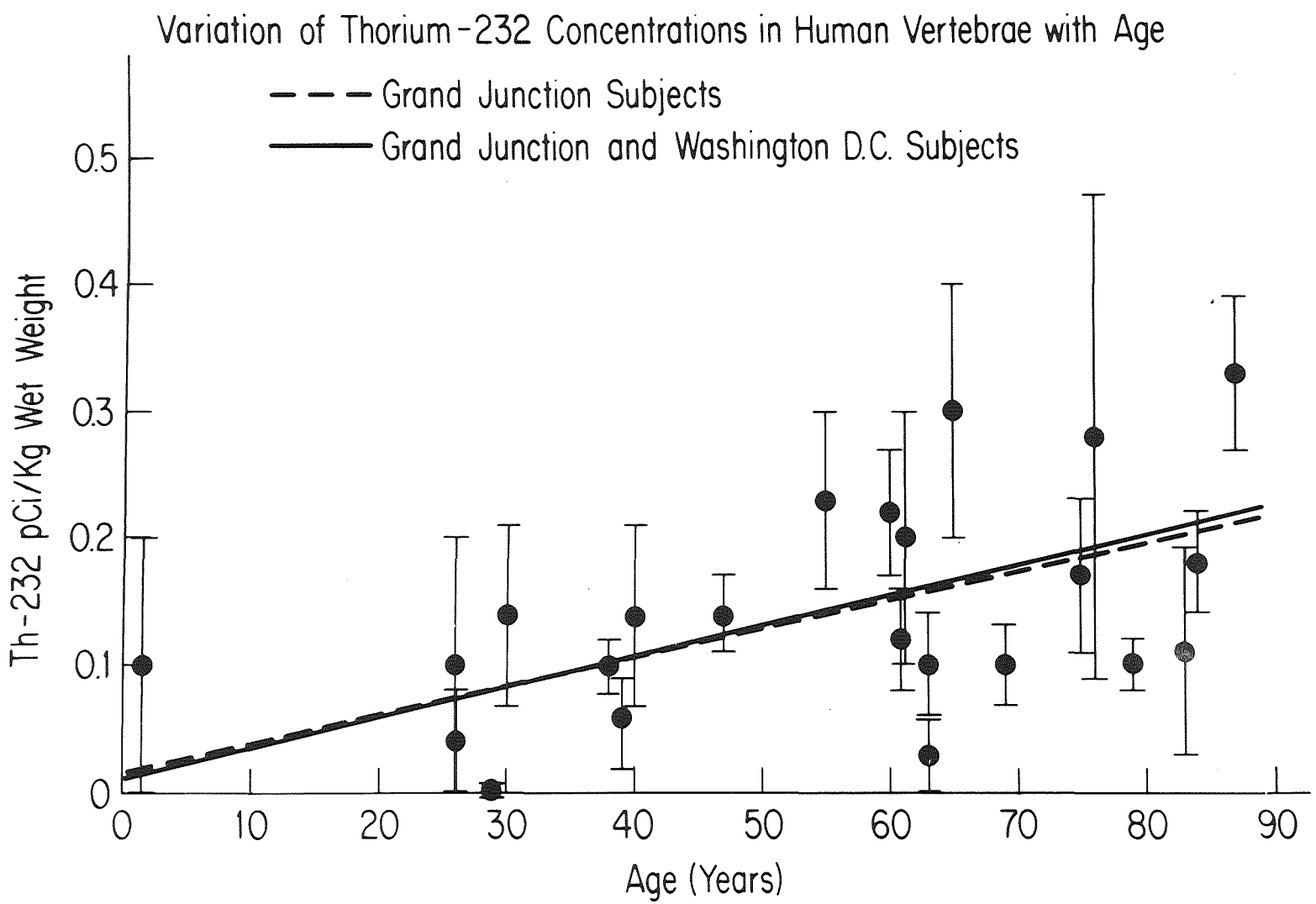


FIGURE 11.1: Variation of Thorium-232 Concentration in Human Vertebrae with Age

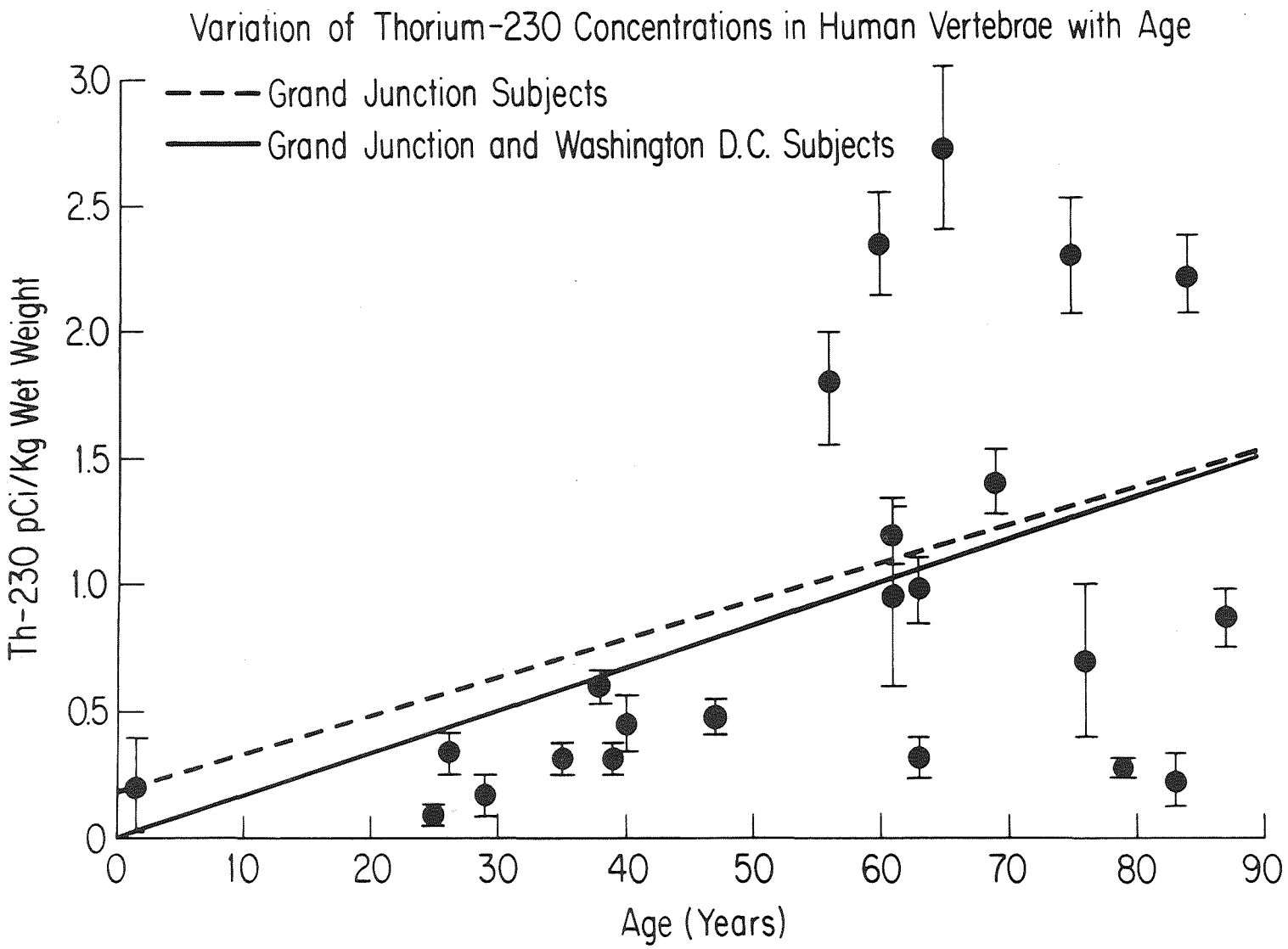


FIGURE 11.2: Variation of Thorium-230 Concentration in Human Vertebrae with Age

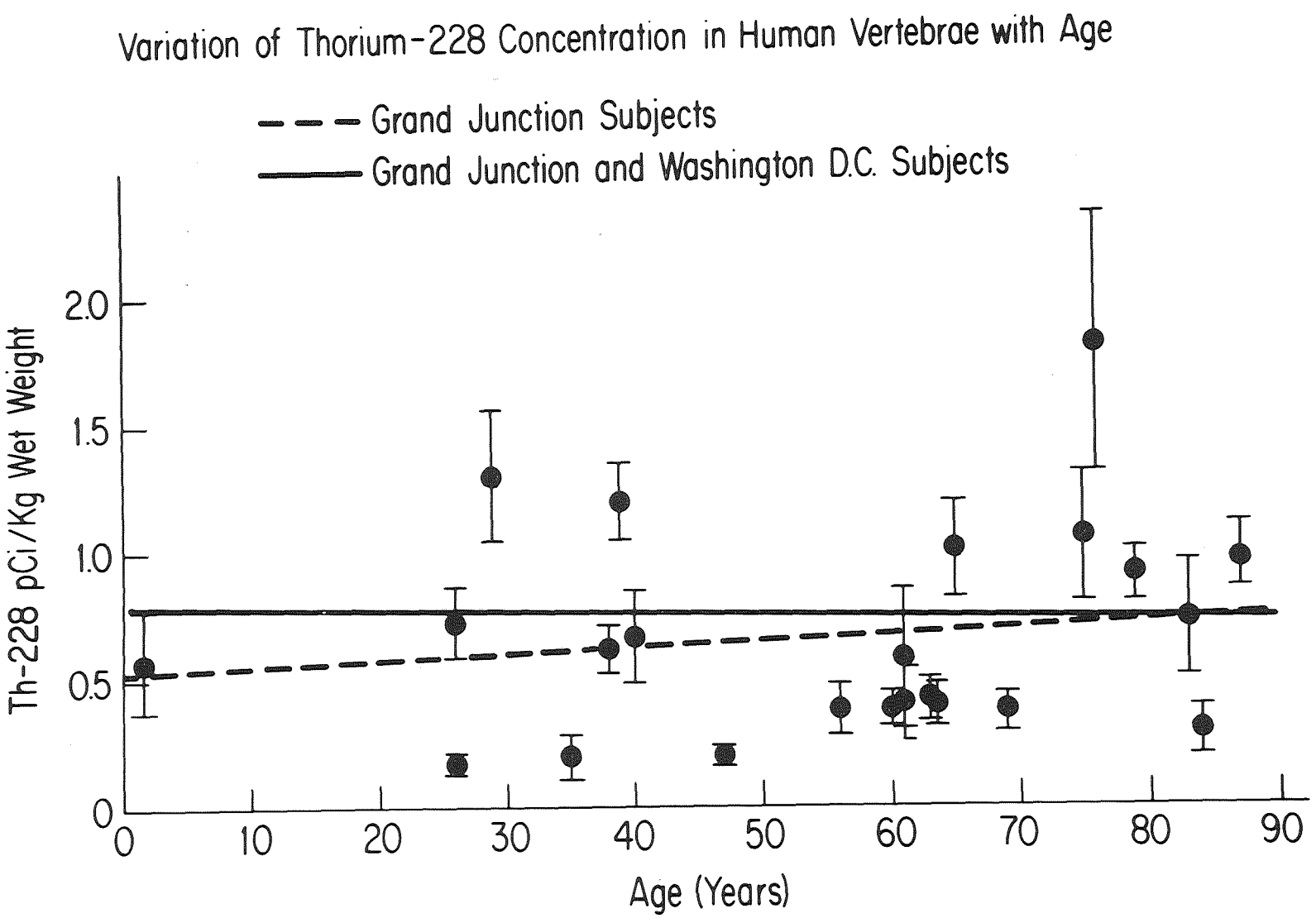


FIGURE 11.3: Variation of Thorium-228 Concentration in Human Vertebrae with Age

The annual rates of Th-230 and Th-232 increase represented by the slope of the regression line (A) and the Y-intercept (B) were:

Nuclide	Population	(A) Rate of Increase in vertebrae content (pCi/kg/year	(B) Concentration at T = 0 (pCi/kg)
Th-232	Grand Junction subjects only	0.0020±0.0009*	0.02
	Grand Junction & Washington, D.C. combined	0.0022±0.0006	0.02
Th-232	From Lucas et al.	0.0030±0.0004	0.00
Th-230	Grand Junction subjects only	0.0150±0.0090	0.20
	Grand Junction & Washington, D.C. combined	0.0170±0.0070	0.00
Th-228	Grand Junction subjects only	0.0002±0.0002	0.51
	Grand Junction & Washington, D.C. combined	-0.0008±0.0006	0.79

* The ± represents the standard error of the slope.

In contrast to Th-232 and Th-230, Th-228 shows no age dependence, which is reflected by its high initial concentration (0.5 and 0.8 pCi/kg in Grand Junction and the total population combined) and the very small slope of the regression. In fact this regression against the populations combined is negative which probably reflects the slightly higher content found in the Washington D.C. subjects.

That Th-228 should not depend strongly on age is reasonable, since Ra-228 which decays to Th-228 is ingested in food and water and is readily absorbed from the gastrointestinal tract. In addition little or no placental barrier is suggested for Radium, and the skeletal concentration of the membrane should reflect the endogenous Ra/Ca content of the mothers. An age independent model is consistent with the results obtained. The Th-228 concentration in bone of a 1.5 year old child from Washington, D.C. was 0.57 pCi/Kg fresh bone compared to a median concentration of 0.66 pCi/Kg for the Washington, D.C. subjects, with an average age of 33 years. An age independent model for Th-228 in skeletal was predicted by Wrenn (Wr 77).

CHAPTER 12

The Biological Half-Life (t_b) of Thorium in Lung

For known intake rates by inhalation, the ICRP lung model can be used to estimate the expected concentrations of thorium in lung and also skeleton if a metabolic model is used to describe transport to skeleton via blood.

There is no published experimental information on the clearance rate of thorium compounds from the lungs of man or animals.

Formerly ICRP used biological half-times (t_b) of 1 and 4 years for plutonium and thorium, respectively (IC59) to derive concentration limits in air. More recently ICRP has suggested that insoluble plutonium and thorium compounds be classified as Class Y with t_b = 500 days applicable to the long-term pulmonary region.

In this section, we estimate the biological half-life of thorium in the human lung using two approaches. First, from the data obtained from measurements of air samples and human lungs from New York City (presented in Section 5.2). The assumptions made are:

1) The radioactive decay for Th-230 and Th-232 may be neglected.
2) Inhalation is the main route of intake;
3) Equilibrium is established between the radionuclide in the inhaled air and in the lung; and
4) The intake rate does not vary.

Therefore, the rate of elimination is equal to the amount deposited per unit time, and the biological half-life may be expressed as:

$$t_b = \frac{0.693\ mC}{I\ f_a} \text{ (IC59)} \qquad (12.1)$$

in which:

t_b = biological half-life in years

m = mass of the organ of reference in grams (the lung in this case)

C = the average concentration of the radionuclide in pCi/g in the organ of reference (the lung)

I = average yearly intake of the nuclide in pCi

f_a = fraction of that taken into the body by inhalation that remains in the organ of reference (the lung).

Class Y parameters have been assumed because environmental thorium is most likely to be in insoluble form.

Let: D_5 stand for the fractional deposition in the pulmonary region. This is taken as 0.32 (Be74)

and $1-F_f$ stand for the portion of the deposition in the pulmonary compartment which clears with a long half-time. This is taken, per ICRP-30, as 0.6.

Then: $f_a = (1-F_f)\ D_5$ is the inhaled fraction which remains in the pulmonary compartment with slow elimination.

Let:

I stand for yearly intake by inhalation. This equals the product of average air concentration and volume breathed/year; the breathing rate is 20 m^3/d (7300 m^3/y) for Reference Man (ICRP-23) and the mean concentration measured in 3, week-long samples for both Th-230 and Th-232 was 4×10^{-5} pCi/m^3.

mC stands for the lung burden. The average of three New York lungs was 0.66 pCi/kg for both Th-230 and Th-232 including the amounts in lymph nodes, and the lung weight was assumed to be 1 kg.

Substituting these values into equation (12.1) leads to an estimate of 8 years for the biological half-time for Th-232 and Th-230, well in excess of any previous estimates. There is a large uncertainty in this estimate because of the small number of samples, both for lungs and air, and it is questionable whether the outdoor air samples were representative for long-term averages.

No analyses of the air from Grand Junction or Washington, D.C. were available; however, t_b can be inferred using human metabolic models, from the ratio of the amounts observed in lung and skeleton, without knowledge of Th-230 and Th-232 concentrations in air or the volume of air inhaled.

One can calculate the mean residence time in the lung by assuming intake by inhalation only and adopting reasonable values of transfer coefficients from lung to blood and to skeleton, in an appropriate metabolic model.

The ICRP lung model is adopted here except that the pulmonary retention time is considered unknown. In addition, we make the simplifying assumption that the mean residence time in the lymph nodes is the same as in the long-term pulmonary compartment. The amount measured in the lung was taken as equal to that in the pulmonary region plus lymph nodes or:

$$(mC)_\ell = If_aT_\ell(1-e^{-\lambda_\ell t}) \qquad (12.2)$$

at sufficiently long times $\lambda_\ell t >> 1$ and the equation simplifies to:

$$(mC)_\ell = If_aT_\ell. \qquad (12.3)$$

where T_ℓ is the effective mean life in lung.

Also, the amount in skeleton may be expressed as:

$$(mC)_s = If_sT_s\ (1-e^{-\lambda_s t}) \qquad (12.4)$$

where f_s is the fraction of the long term inhaled material deposited in skeleton.

Then by taking the ratio of equations (12.3) and (12.4) and assuming that the mean life in the skeleton is long compared to lifespan (i.e., $1-e^{-\lambda_s t} \simeq \lambda_s t$) we obtain the mean-life in lung, $T\ell$:

$$T_\ell = \frac{(mC)_\ell\ [D_3F_a + D_4F_c + D_5F_e]\ f'_{2s}t}{(mC)_s D_5(1-F_f)} \qquad (12.5)$$

The numerical values of D's and F's used are shown in Figure 12.1, f'_{2s}, the fractional transfer from blood to skeleton, is taken as 0.7 consistent with the distribution we observed and that adopted by the ICRP (IC 79).

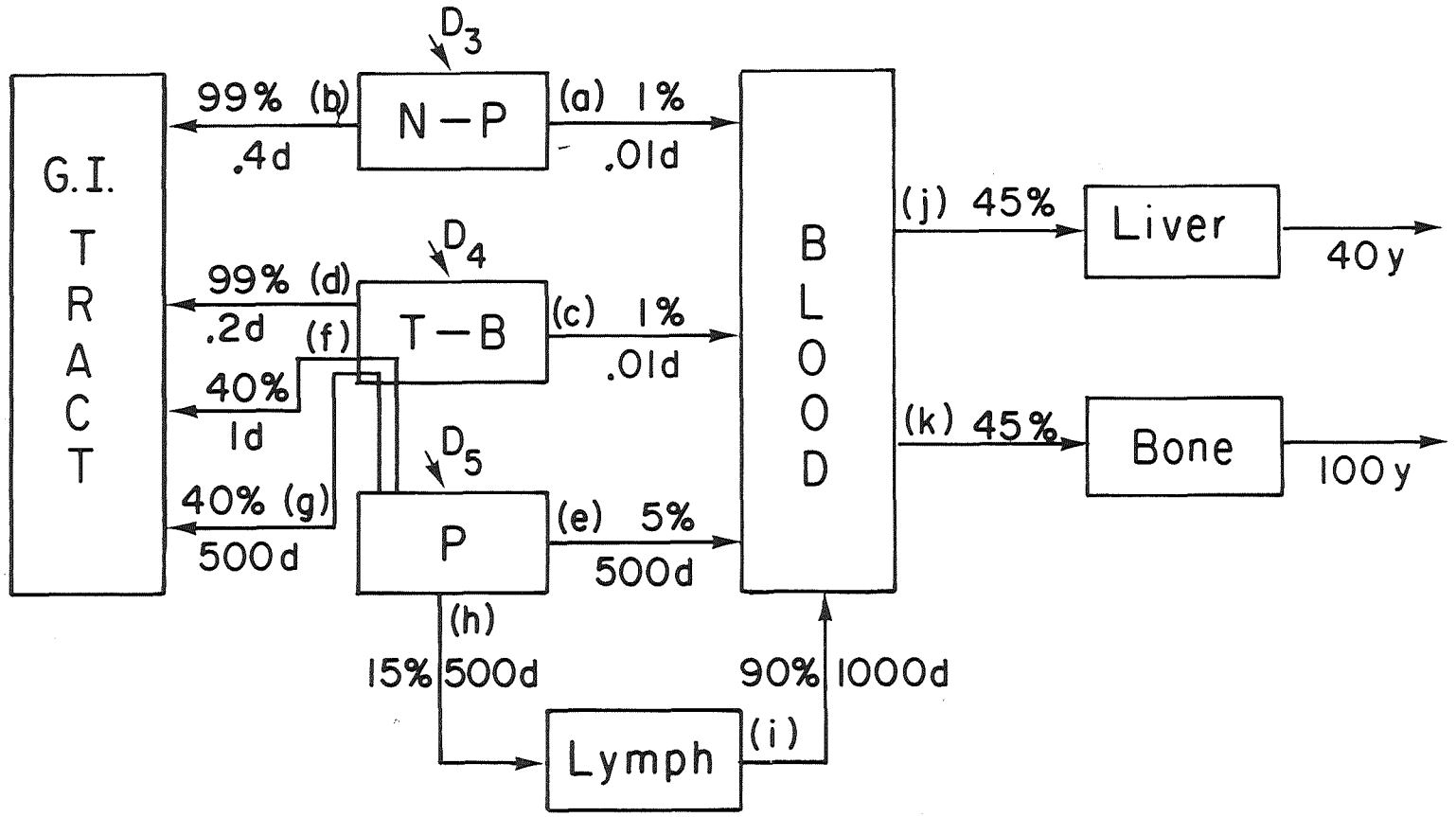
Now taking $(mC)_\ell$ = lung + lymph node burden, and $(mC)_s$ for skeleton as the means from Table 12.2, and t = 67 years for Grand Junction subjects and 33 years for Washington, D.C. subjects, we obtain estimates of t_b in lung as follows:

Half-Times (t_b) in Lung in Years

	Th-230	Th-232	Mean Age (y)
Grand Junction	1.4/1.0	5.3/3.9	67
Washington, D.C.	1.0/1.4	2.6/3.8	33

* The first value assumes infinite residence time in bone, and the second 100 years mean life in bone.

The estimated t_b for Th-232 is similar to the 4 years used in the early ICRP report (IC59). However, the estimated half-time for Th-232 is about 3 times greater than for Th-230. Because the physical and chemical nature of the material inhaled will influence the clearance rate from lung, this difference between the t_b values for Th-230 and Th-232 is probably real and may be due to the association of each isotope with a different type of matrix



Class Y Parameters

Particle Size: .4 μm

$D_3 = .10$

$D_4 = .08$

$D_5 = .32$

Figure 12.1 ICRP Task Group Lung and Clearance Model for Plutonium.

since Th-230 is found in uranium-bearing minerals and Th-232 is in thorium-bearing minerals.

Thus, the range of inferable long-term half-times in the lung is therefore 1 to 8 years. Since these estimates depend on other assumptions about the metabolism, i.e., effectively infinite retention in the skeleton, the range of estimates is not surprising, and the higher values may overestimate the half residence times in the lung.

CHAPTER 13

Summary and General Conclusions

The major objectives of this research were to:

1) Quantitatively establish the amount, distribution and age dependence of α-emitting, thorium isotopes in human tissues.

2) Investigate whether the tissues of subjects who lived in areas where uranium mill tailings were present exhibit a higher Th-230 concentration than in a control population.

3) Estimate the long-term potential for accumulation of plutonium from the environment by man using the information obtained on thorium metabolism.

To achieve these objectives, 32 sets of human tissues were collected at autopsy. Each set included: right lung, liver (one-half), spleen, kidney, and vertebrae. Pulmonary lymph nodes were separated from the lung and analyzed separately. Radiochemical analyses of the natural alpha-emitting isotopes of thorium were performed using tissue digestion, solvent extraction, and electrodeposition, followed by alpha spectrometric measurements. To do this we developed suitable radiochemical procedures which included:

a) Selection of a suitable tracer to determine the analytical yield and the radiochemical determination of natural thorium. Th-229, prepared by the Oak Ridge National Laboratory, was successfully tested as an isotopic tracer. Th-229 is a more advantageous tracer than Th-234, primarily because of its long half-life and the ability to measure its alpha emission simultaneously with the natural alpha-emitting thorium isotopes, Th-228, Th-230, and Th-232.

b) The determination of thorium isotopic contents in soft tissues and bone with sufficient sensitivity and specificity.

The general conclusions of this investigation are:

1) The thorium isotopic concentration, and the ratio of Th-230/Th-232 in Grand Junction soil appear in the range of normal U.S. reported values; however, the concentration of Th-230 is elevated in tailings 2000-fold and the ratio of Th-230/232 about 400-fold.

2) The ratio of the thorium isotopes in human lungs analyzed from New York is similar to that found in the air samples taken from the same location, suggesting equilibrium between the pulmonary system and inhaled air.

3) Thorium nuclides were measurable in lung, lymph nodes, liver, bone (vertebrae), kidney and spleen, in the following order of decreasing concentration:

Lymph nodes > lung > bone > kidney > liver for Th-232 and Th-230, and lymph nodes > bone > lung > kidney > liver for Th-228 (Fig. 13.1).

4) The concentration of Th-230 was higher than Th-232 in all tissues, including bone, reflecting the relative geochemical abundances of the uranium and thorium series.

5) The concentration of Th-228 was higher than Th-232 in bone. This suggests that the major portion of Th-228 in bone is due to the intake of transportable Ra-228, which concentrates in bone and then decays to Th-228.

6) The concnetration of all thorium nuclides are more than an order of magnitude higher in lymph nodes than in lung.

7) Two of the cases from Colorado showed clearly elevated levels of Th-230 caused by higher exposures to uranium-bearing minerals. In the hard rock miner, the concentration of this nuclide was almost 10 times higher than in the other cases. The second case had been a uranium miner; his Th-230 concentration was more than two orders of magnitude higher than the median of the general population.

8) The limited conclusions to date suggest that even though the residents from this area have lived in the vicinity of uranium tailings, they do not have clearly elevated Th-230 concentrations in their lungs, when compared to the residents of Washington, D.C., who are not exposed to such tailings. However, Th-230 concentration in the bone of Grand Junction subjects was slightly higher than that of Washington, D.C. subjects after suitable age adjustment. This increase may suggest an old exposure.

9) The relative distribution of all three isotopes among the organs was found to be different. For example, the lung including lymph nodes contained 5 to 7% of the Th-228, 10 to 13% of Th-230 and 28 to 29% of Th-232. Similarly, skeleton contained 80 to 82% of Th-228, 70 to 75% of Th-230 and only 55 to 56% of Th-232. The higher proportion of Th-228 in the skeleton may be due to intake of Ra-228 which decays to Th-228. Th-230 from the uranium series may have a more soluble mineral matrix than Th-232 or may be inhaled in smaller particles. Table 13.1 summarizes the likely routes of entry of thorium isotopes into man, and Figure 13.2 shows their suggested transport pathways.

10) The organ distribution pattern of thorium obtained in this study is in good agreement with that recently adopted in ICRP No. 30, based on the results of Stover et al. (St60,65) in beagles.

11) The total median accumulation of thorium in the human body (0.7 to 7 pCi for each nuclide) was equivalent to the amounts of thorium in 0.7 to 7 grams of soil.

12) The total dose rate from all three thorium isotopes in the occupationally-exposed former miner is between 4.8 and 10.5 mrads per year in lymph nodes, and 1.2 and 1.5 mrads per year in lungs. In non-occupationally exposed individuals from Grand Junction and Washington, D.C., the dose rate to lung ranged from 0.06 to 0.23 mrads per year, assuming equilibrium of the

Th-228 daughters. In the skeleton, the dose rate estimates ranged between 0.1 and 0.6 mrads per year; here the reasonable assumption of equilibrium is made. The highest dose rate from any of the thorium isotopes was delivered to the lymph nodes, and ranged from 1-5 mrads/year.

13) The highest total accumulation is in the skeleton. The skeleton is accordingly hypothesized to be a good integrator of the total lifetime input to the human body. The data for Th-232 and Th-230 show a general trend towards an increasing concentration with age, and this relation was fitted to a linear accumulation model.

14) The biological half-life of thorium in the lung was inferred from measurement. The data suggest a longer retention of Th-232 in the lung than Th-230. This may be related to the chemical and physical nature of the aerosols bearing both the thorium and uranium minerals, or to statistical variations due to the small sample size measured.

15) The half-times inferred for the long-term elimination from lung were different from the values accepted by ICRP Committee 2 for Class Y compounds. Although the ICRP model may be accurate for certain particle sizes and chemical forms, however, this data suggests that environmental Th isotopes do not behave strictly as Class Y but are probably retained with half times of a year or greater.

TABLE 13.1

Nuclide	Likely Route of Entry and Origin of Thorium Isotopes in Human Tissue
Th-232	Inhalation: soils, suspended particlates
Th-230	Inhalation: Tailings piles, soils
Th-228	Ingestion in food and water: ingrowth in body of Th-228 daughter from Ra-228 parent

FIGURE 13.1: 50th Percentile Concentration of Thorium-Autopsy Samples from Grand Junction and Washington, D.C.

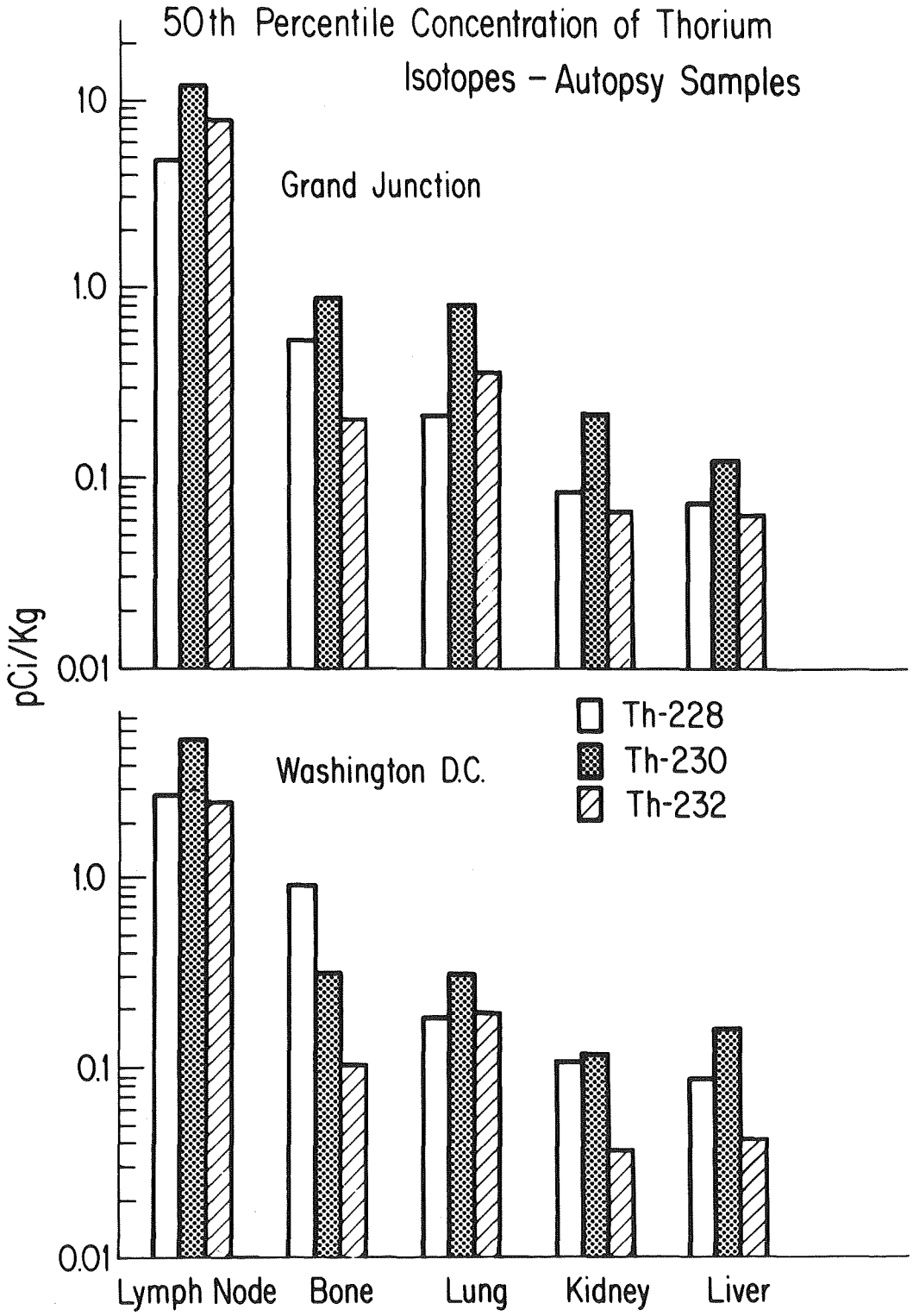
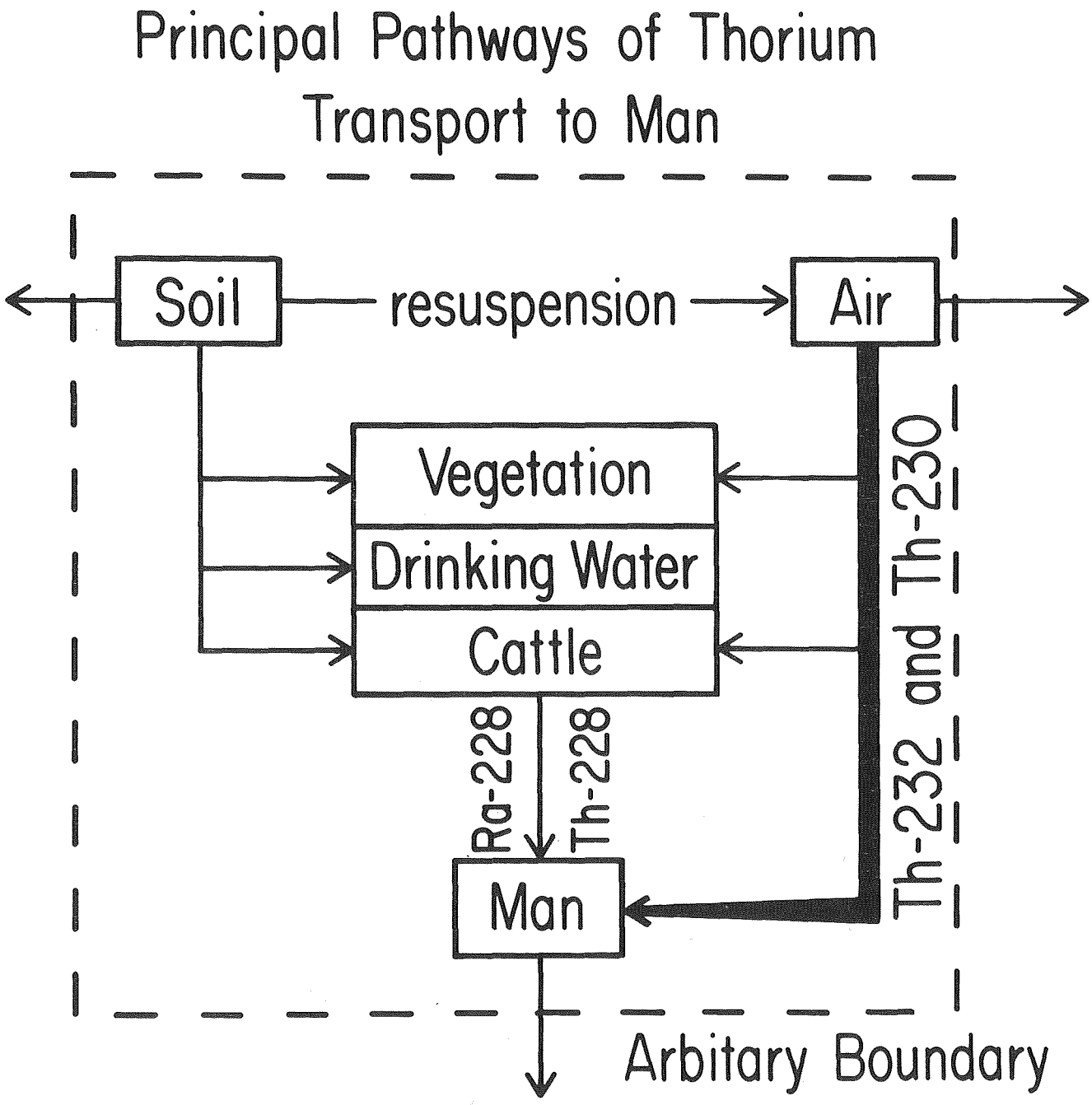


FIGURE 13.2: Principal Pathways of Thorium Transport to Man



REFERENCES

Al66 Albert, R.E. Thorium: Its Industrial Hygiene Aspects. Academic Press, New York. 1966.

Al63 Altshuler, B. and B. Pasternack. Statistical Measured of the Lower Limit of Detection of Radioactivity Counter. Health Phys. 9: 293, 1963.

An71 Anspaugh, L.R., W.L. Robinson, W.H. Martin and O.A. Rowe. Compilation of Published Information on Elemental Concentrations in Human Organs in Both Normal and Diseased States. University of California Report, 1971.

Ba70 Bair, W.J. Plutonium Inhalation Studies. Battelle Pacific Northwest Laboratories Report, BNWL-1221, p. 417, 1970.

Be74 Bennett, B.G. Fallout ^{239}Pu Dose to Man. U.S. Department of Energy, Fallout Program Report, Part I-41, HASL-278, January, 1974.

Be76 Bernstein, D.M., M.T. Kleinman, T.J. Kneip, T.L. Chan and M. Lippmann. A High Volume Sampler for the Determination of Particle Size Distributions in Ambient Air. J. Air Pollut. Control Assoc. 27(11): 1069, 1976.

Be78 Beck, H.L., C.V. Gogolak, K.M. Miller and W.M. Lowder. Perturbations on the Natural Radiation Environment Due to the Utilization of Coal as an Energy Source. In: Book of Summaries of the Natural Radiation Environment III, Houston, Texas, 1978.

Bo63 Boeker, B.B., R.G. Thomas and J.K. Scott. Thorium Distribution and Excersion Studies II. General Patterns Following Inhalation and the Effect of the Size of the Inhaled Dose. Health Phys. 9: 165, 1963.

Bo58 Boone, I.U., B.S. Rogers, D.C. White and P.C. Harris. Toxicity, Excretion and Tissue Distribution of Ionium (Th-230) in Rats. Am. Ind. Hyg. Assoc. J. 19: 285, 1958.

Bo74 Borisov, B.K. and A.N. Marei. Weight Parameters in Adult Human Skeleton. Health Phys. 27: 224, 1974.

Cl71 Clifton, R.J., M. Farrow and E.I. Hamilton. Measurements of Th-232 in Normal and Industrially Exposed Humans. Ann. Occup. Hyg. 14: 303, 1971.

Cu68 Currie, L.A. Limits for Quantitative Detection and Quantitative Determination. Anal. Chem. 40: 586, 1968.

Du76 Durbin, P.W. and N. Jeung. Reassessment of the Distribution of Plutonium in the Human Body Based on Experiments with Non-Human Primates. In: The Health Effects of Plutonium and Radium, (W.S.S. Jee, Ed.), The J.W. Press, University of Utah, Salt Lake City. 1976.

Ei64 Eisenbud, M. and H.G. Petrow. Radioactivity in the Atmospheric Effluents of Power Plants That Use Fossil Fuels. Science 144: 228, 1964.

EML78 Environmental Measurements Laboratory. Quarterly Report of the Department of Energy, Operational and Environmental Safety Division, Quality Assessment Program. EML-337, February, 1978.

EML79 Environmental Measurements Laboratory. Quarterly Report of the Department of Energy, Operational and Environmental Safety Division, Quality Assessment Program. EML-354, May, 1979.

Fa54 Faul, H. Nuclear Geology. John Wiley & Sons, Ind., New York. 1954.

Fi61 Figgins, P.E. The Radiochemistry of Polonium. Nuclear Science Series, National Research Council, National Academy of Sciences, NAS-NS # 3037. 1961.

Fi79 Fisenne, I.M. A model of the Distribution of Fallout Pu-239, 240 in New York City Residents Based on Measured Tissue Concentration. Doctoral Dissertation, New York University, Institute of Environmental Medicine, 1979.

Ha36 Hahn, O. Applied Radiochemistry. Cornell University Press, Ithaca, New York, p. 34, 1936.

Ha74 Harley, J.H. Calculation of Dose Factors for Internally Deposited Natural Radionuclides, HASL TM-74-2. U.S. Department of Energy, 1974.

Ha78 Harley, J.H. Environmental Measurements Laboratory Manual, U.S. Department of Energy, 1978.

Ha79 Harley, N.H. and B.S. Pasternack. Potential Carcinogenic Effects of Actinides in the Environment. Health Phys. 37: 291, 1979.

Hi62 Hill, O.R. Identification of α-Emitters in Normal Biological Materials. Health Phys. 8: 17, 1962.

Ho58 Hodge, H.C. and R.G. Thomas. Health Hazards from the Inhalation of Insoluble Uranium and Thorium Oxides. Proceedings of the 2nd International Conference on Peaceful Uses of Atomic Energy, Geneva 23: 302, 1958.

Ho60 Hodge, H.C., E.A. Maynard and L.J. Leach. The Chemical Toxicity of Thorium Dioxide Following the Inhalation by Laboratory Animals. Report UR-562, University of Rochester, Rochester, New York, January 1980.

Ho63 Holtzman, R.B. Measurement of the Natural Contents of RaD (Pb-210) and RaF (Po-210) in Human Bone - Estimates of Whole Body Burdens. Health Phys. 9: 385, 1963.

Hy60 Hyde, E.K. The Radiochemistry of Thorium. National Academy of Sciences, Nuclear Science Series, NAS-NS 3004, 1960.

IC59 International Commission on Radiological Protection. Recommendations of the International Commission on Radiological Proctection, Report of Committee II on Permissible Dose for Internal Radiation. ICRP Publication 2. Pergamon Press, Oxford. 1959. Also appears in Health Phys. 3:1, 1960.

IC72 International Commission on Radiological Protection. The Metabolism of Compounds of Plutonium and Other Actinides. ICRP Publication 19. Pergamon Press, Oxford. 1972.

IC75 International Commission on Radiological Protection. Report of the Task Group on Reference Man. ICRP Publication 23. Pergamon Press, Oxford.

IC79 International Commission on Radiological Protection. Limits of Intakes of Radionuclides by Workers. ICRP Publication 30. Pergamon Press, Oxford. 1979.

Je60 Jee, W.S.S. and T.H. Cochran. Microscopic Distribution of Plutonium-239 and Thorium-228 in Beagles. A Seminar on Thorium. University of Rochester. Atomic Energy Project, Rochester. 1960.

Jo71 Joyet, G. The Thorium Series in Cigarettes and in Lungs of Smokers. Experientia 27: 85, 1971.

Ka57 Katz, J.J. and G.T. Seaborg. The Chemistry of the Actinide Elements. John Wiley & Sons, Inc., New York. 1957.

Ko77 Kolber, A.B. In Vivo Measurements of Pb-212 in Smokers and Nonsmokers by Gamma Ray Spectroscopy. M.S. Thesis, New York University. October 1977.

La78 Larsen, R.P., R.D. Oldham, C.G. Cacic, J.E. Farnham and J.R. Schneider. Distribution of Injected Plutonium in the Skeleton and Certain Soft Tissues. Argonne National Laboratory Report No. ANL-78-65, Part II, Center of Human Radiobiology, Annual Report. July 1977 - June 1978.

Le78 Lederer, C.M., J.M. Hollander and I. Perlman. Table of Isotopes. 6th Edition. John Wiley and Sons, Inc., New York. 1978.

Lo64 Lowder, W.M., W.J. Condon and H.L. Beck. Field Spectormetric Investigations of Environmental Radiation in the U.S.A. In: Natural Radiation Environment (Eds., J.A.S. Adams and W.J. Lowder). University of Chicago Press, Chicago, Illinois, p. 597. 1964.

Lu70a Lucas, H.F., D.N. Edington and F. Markun. Natural Thorium in Human Bone. Health Phys. 19: 739, 1970.

Lu70b Lucas, H.F., Jr. and F. Markun. Thorium and Uranium in Blood, Urine and Cigarettes. Argonne National Laboratory Annual Report, ANL-7760, p. 47 (1969-1970). 1970.

Ma69 Maletskos, C.J., A.T. Keane, N.C. Telles and R.D. Evans. Retention and Absorption of R-224 and Th-234 and Some Dosimetric Considerations of R-224 in Human Beings. University of Utah Press, Salt Lake City, Utah. 1969.

Ma69 Martin, J.E., E.D. Harward and D.T. Oakley. Comparison of Radioactivity from Fossil Fuel and Nuclear Power Plants. In: Hearings Before the Joint Committee on Atomic Energy, Environmental Effects of Producing Electric Power, Part 1 U.S. Government Printing Office, Washington. p. 773. 1969.

Ma51 Massey, F.J., Jr. The Kolomogorov-Smirvov Test for Goodness of Fit. American Statistical Association Journal 46:68, 1951.

Mc76 McInroy, J.F. The Los Alamos Scientific Laboratory's Human Autopsy Tissue Analysis Study. The Health Effects of Plutonium and Radium. The J.W. Press, Salt Lake City, Utah, p. 249. 1976.

Mc79 McInroy, J.F., E.E. Campbell, W.D. Moss, G.I. Tietjen, B.C. Eutsler and H.A. Boyd. Plutonium in Autopsy Tissue: A Revision and Updating of Data Reported in LA-4875. Health Phys. 37:1, 1979.

Mo63 Moses, A. Analytical Chemistry of the Actinide Elements. Pergamon Press, New York. 1963.

NC76 National Council on Radiation Protection Measurements - Environmental Radiation Measurements. NCRP Report #50. 1976.

Ne72 Nelp, W.B., J.D. Denney, R. Murano, G.M. Hinn, J.L. Williams, T.G. Rudd and H.E. Palmer. Absolute Measurement of Total Body Calcium (Bone Mass) In Vivo. Journal of Laboratory and Clinical Medicine 79:430, 1972.

Ne79a Neton, J.W. The Determination of Pu-239,240 and Th-228, 230, 232 in U.S. Cigarette Tobacco. M.S. Thesis, New York University. June 1979.

Ne79b Neton, J.W. and S.A. Ibrahim. Alpha-Emittig Radionuclides in Cigarette Tobacco. Health Phys. 35: 922, 1979.

Pe74 Percival, P.R. and D.B. Martin. Sequential Determination of Radium-226, Radium-228, Actinium-227 and Thorium Isotopes in Environmental and Process Waste Samples. Anal. Chem. 46: 1742, 1974.

Pe64 Petrow, H.G., A. Cover, W. Schiessle and E. Parsons. Radiochemical Determination of Radium-228 and Thorium-228 in Biological and Mineral Samples. Anal. Chem. 36:1600, 1964.

Pe67 Petrow, H.G. and C.D. Strehlow. Spectrophotometric Determination of Thorium in Bone Ash Using Arsenato III. Anal. Chem. 39:265, 1967.

Pi68 Picer, M. and Strohol. Determination of Thorium on Uranium in Biological Materials. Anal. Chim. Acta 40:131, 1968.

Re70 Remington, R.D. and M.A. Schork. Statistics with Applications to the Biological and Health Sciences. Prentice-Hall, Inc., Englewood Cliffs, N.J. 1970.

Sa51 Salerno, P.R. and P.A. Mattis. Absorption and Distribution of Thorium Nitrate in the Rat. J. Pharmacol. Exptl. Therap. 101:31 (Abstract), 1951.

Sc52 Scott, J.K., W.F. Neuman and J.F. Bonner. The Distribution and Excretion of Thorium Sulfate. J. Pharmacol. Exp. Ther. 106:208, 1952.

Sc75 Scott, L.L. Comparison of Thorium Gamma Spectra in Smokers and Nonsmokers. Health Phys. 29:418, 1975.

Se73 Sedlet, J., N.W. Golchert and T.L. Duffy. Environmental Monitoring at Argonne National Laboratory - 1972. U.S. Atomic Energy Commission Report, Argonne National Laboratory, ANL-8007, Argonne, Illinois. 1973.

Si68 Sikov, M.R. and D.D. Mahlum. Cross Placental Transfer of Selected Actinides in the Rat. Health Phys. 14: 205, 1968.

Si69 Sill, C.W. and R.L. Williams. Radiochemical Determination of Uranium and Transuranium Elements in Process Solutions and Environmental Samples. Anal. Chem. 41: 1624, 1969.

Si70 Sill, C.W. and D.G. Olson. Sources and Prevention of Recoil Contamination of Solid-State Alpha Detectors. Anal. Chem. 42: 1596, 1970.

Si74 Sill, C.W., K.W. Puphal and F.D. Hindman. Simultaneous Determination of Alpha-Emitting Nuclides of Radium Through Californium in Soil. Anal. Chem. 46: 1725, 1974.

Si77 Sill, C.W. Determination of Thorium and Uranium Isotopes in Ores and Mill Tailings by Alpha Spectrometry. Anal. Chem. 49:618, 1977.

Si78a Singh, N.P., S.A. Ibrahim, N. Cohen, and M.E. Wrenn. Solvent Extraction for Determination of Plutonium in Soft Tissue, Anal. Chem. 50:357, 1978.

Si78b Singh, N.P., S.A. Ibrahim, N. Cohen and M.E. Wrenn. The Comparative Distribution of Thorium and Plutonium in Human Tissues. Twenty-Third Annual Meeting of the Health Physics Society (Abstract). June 1978.

Si79a Singh, N.P. S.A. Ibrahim, N. Cohen and M.E. Wrenn. Solvent Extraction Method for Determination of Thorium in Soft Tissues. Anal. Chem. 51; 207, 1979.

Si79b Singh, N.P., S.A. Ibrahim, N. Cohen and M.E. Wrenn. Simultaneous Determination of Alpha-Emitting Radionuclides of Thorium and Plutonium in Human Tissues Including Bone. Anal. Chem. 51:1978, 1979.

Si79c Singh, N.P., S.A. Ibrahim and M.E. Wrenn. Comparative Distribution of Plutonium and Natural Thorium in the Same Samples of Human Lung and Lymph Nodes and its Relevance on Pulmonary Clearance Classification for Lung Models. Twenty-Fourth Annual Meeting of the Health Physics Society (Abstract). July 1979.

St60 Stover, B. Summary of "Thorium Studies in Beagles." In: A Seminar on Thorium. University of Rochester Atomic Energy Project. 1960.

St62 Stover, B.J., D.R. Atherton and C.W. Mays. Studies of the Retention and Distribution of Ra-226, Pu-239 and Ra-228 ($MsTh_1$), Th-228 (RdTh) and Sr-90 in Adult Beagles. In: Some Aspects of Internal Irradiation. (Ed., T.F. Dougherty.) Pergamon Press, New York, p. 7. 1962.

St65 Stover, B.J., D.R. Atherton, D.S. Buster and F.W. Bruenger. The Th-228 Decay Series in Adult Beagles: Ra-224, Pb-212 and Bi-212 in Selected Bones and Soft Tissues. Radiat. Res. 26:132, 1965.

Su79a Sullivan, M.F. Absorption of Actinide Elements from the Gastrointestinal Tract of Rats, Guinea Pigs and Dogs. Health Phys. 38:159, 1979.

Su79b Sullivan, M.F. Absorption of Actinide Elements from the Gastrointestinal Tract of Neonatal Animals. Health Phys. 38:173, 1979.

Th57 Thomas, R.G. The Metabolism of Th-232 (Ionium) Administration by Intratracheal Injection to the Rat. Report UR-480, University of Rochester, Rochester, New York. January 1957.

Th63 Thomas, R.G., R.Lie and J.K. Scott. Thorium Distribution and Excretion Studies. I. Patterns Following Parenteral Administration. Health Phys. 9: 153, 1963.

Tu60 Turner, R.C. and J.M. Radley. Naturally Occurring Alpha Activity of Cigarette Tobaccos. Lancet 1:1197, 1960.

Vi59 Vinogradov, A.P. The Geochemistry of Rare and Dispersed Chemical Agents in Soils. Second Edition. Consultants Bureau, New York. 1959.

Wr77 Wrenn, M.E. Internal Dose Estimates. Proceedings of the International Symposium on Areas of High Natural Radioactivity. Brazilian Academy of Sciences, p. 131. 1977.

Wr78 Wrenn, M.E., N.P. Singh, S.A. Ibrahim and N. Cohen. Thorium-229 as an Isotopic Tracer for the Radiochemical Determination of Thorium Isotopes in biological Samples. Anal. Chem. 50: 1712, 1978.

APPENDIX A

Tabulation of history, organ mass, chemical yield, and concentration of Th-228, 230, and 232 in tissues of Grand Junction and Washington, D.C. subjects.

		Tissue Type	Mass (g)	Th-228 pCi/kg	Th-230 pCi/kg	Th-232 pCi/kg	Recovery %
Case No.	77-7	Lung	470	0.37±0.10	0.39±0.08	0.60±0.10	39.6
Location	G.J.						
Age (Y)	81	LN	5.0	6.9 ±1.3	12.0 ±1.7	8.7 ±1.5	60.5
Sex	M	Liver	420	0.22±0.06	0.26±0.05	0.32±0.06	18.8
Occupation	Construction						
Smoking	None	Kidney	100	0.31±0.10	0.63±0.16	0.20±0.09	36.5
Case No.	77-8	Lung	550	0.48±0.09	1.2 ±0.13	0.78±0.10	49.2
Location	G.J.						
Age (Y)	86	LN	10.5	12.5 ±3.5	12.5±3.4	17.9±4.0	33.7
Sex	M	Liver	590	0.04±0.01	0.15±0.04	0.03±0.02	41.2
Occupation	Farmer						
Smoking	1 pk/day for 50 years	Kidney	160	0.04±0.01	0.04±0.08	0.03±0.02	61.8
Case No.	77-11	Lung	520	0.22±0.04	0.20±0.04	0.24±0.05	29.1
Location	G.J.						
Age (Y)	83	LN	9.0	1.6±0.92	10.8 ±4.5	6.5 ±3.7	37.2
Sex	M	Bone	280	0.71±0.21	0.22±0.11	0.11±0.08	41.8
Occupation	Carpenter						
Smoking	Yes - unspecified	Spleen	125	0.37±0.08	0.23±0.07	0.10±0.05	54.5
Case No.	77-25	Lung	290	0.32±0.07	1.2 ±0.11	0.61±0.08	52.5
Location	G.J.						
Age (Y)	76	LN	3.5	6.9 ±2.0	7.6 ±3.4	7.4 ±3.6	54.0
Sex	F	Liver	435	0.15±0.6	0.19±0.07	0.11±0.05	12.1
Occupation	Housewife	Kidney	115	0.07±0.03	0.27±0.10	0.06±9,94	33.3
Smoking	None	Bone	70	1.8 ±0.50	0.70±0.31	0.28±0.19	43.0
Case No.	77-36	Lung	450	0.37±0.08	1.0 ±0.10	0.64±0.08	37.9
Location	G.J.						
Age (Y)	83	Liver	440	0.06±0.01	0.12±0.02	0.03±0.01	42.2
Sex	M	Kidney	120	0.03±0.01	0.11±0.04	0.09±0.04	81.2
Occupation	Unknown						
Smoking	Unknown						

		Tissue Type	Mass (g)	Th-228 pCi/kg	Th-230 pCi/kg	Th-232 pCi/kg	Recovery %
Case No.	77-44	Lung	430	0.36±0.05	2.0 ±0.10	0.43±0.04	25.8
Location	G.J.						
Age (Y)	63	LN	9.0	17.4 ±2.9	19.6 ±2.2	5.7 ±1.2	61.1
Sex	M	Liver	430	0.51±0.17	0.46±0.13	0.15±0.08	25.0
Occupation	Mining Engr.						
Smoking	2 pk/day for	Kidney	170	0.02±0.01	0.37±0.10	0.17±0.07	13.4
	25 years	Bone	112	0.04±0.01	0.98±0.12	0.02±0.01	60.9
Case No.	77-48	Lung	265	0.06±0.01	0.09±0.07	0.02±0.01	60.9
Location	G.J.						
Age (Y)	26	Liver	575	0.02±0.01	0.08±0.02	0.02±0.01	30.9
Sex	F	Kidney	130	0.03±0.03	0.05±0.02	0.03±0.01	66.0
Occupation	Housewife						
Smoking	Unknown	Bone	100	0.16±0.04	0.10±0.09	0.10±0.10	18.9
		Spleen	115	0.13±0.03	0.24±0.06	0.15±0.05	75.8
Case No.	78-8	Lung	502	0.06±0.13	2.7 ±0.17	1.1 ±0.10	29.5
Location	G.J.						
Age (Y)	57	Liver	570	0.01±0.004	0.04±0.01	0.05±0.05	30.4
Sex	M	Kidney	95	0.17±0.004	0.07±0.04	0.04±0.03	68.3
Occupation	Auto mechanic						
Smoking	None	Spleen	154	0.03±0.01	0.11±0.06	0.03±0.03	35.0

		Tissue Type	Mass (g)	Calcium µg/g ww	Th-228 pCi/kg	Th-230 pCi/kg	Th-232 pCi/kg	Thorium Recovery %
Case No.	78-12	Lung	530	99.4	0.15±0.03	0.56±0.06	0.25±0.04	39.4
Location	G.J.							
Age (Y)	56	Liver	850	NA	0.25±0.18	0.57±0.19	0.38±0.15	20.2
Sex	M	Kidney	201	NA	0.06±0.04	0.45±0.07	0.08±0.03	17.7
Occupation	Rancher							
Smoking	1 pk/day for	Bone	124	61500	0.36±0.10	1.8 ±0.20	0.23±0.07	46.3
	40 years	Spleen	160	NA	0.10±0.08	0.43±0.14	0.14±0.08	62.3
Case No.	78-14	Lung	620	202	0.25±0.01	1.1 ±0.16	0.75±0.13	38.3
Location	G.J.							
Age (Y)	87	LN	13.3	NA	4.5 ±1.1	7.6 ±1.4	12.3 ±1.7	20.2
Sex	M	Liver	490	NA	Lost	Lost	Lost	----
Occupation	Blacksmith							
Smoking	Pipe	Kidney	131	NA	0.20±0.15	0.45±0.07	0.08±0.03	27.7
		Bone	122	0.95±0.11	0.86±0.10	0.33±0.06	0.33±0.06	46.3
		Spleen	160	NA	0.12±0.07	0.17±0.06	0.14±0.08	33.2
Case No.	78-19	Lung	880	118	0.19±0.03	0.48±0.04	0.34±0.03	22.3
Location	G.J.							
Age (Y)	47	LN	9.5	NA	5.6 ±1.3	7.7 ±1.4	6.1 ±1.3	21.1
Sex	M	Kidney	125	160	0.06±0.04	0.29±0.10	0.06±0.04	23.1
Occupation	Farmer							
Smoking	1 pk/day for	Bone	295	NA	0.20±0.03	0.48±0.06	0.14±0.03	63.8
	25 years	Spleen	55	NA	Lost	Lost	Lost	----
		Gonads	52	91.3	0.08±0.03	0.15±0.13	0.04±0.02	44.4
Case No.	78-20	Liver	210	95.2	0.11±0.03	0.33±0.04	0.02±0.01	59.9
Location	G.J.							
Age (Y)	69	Kidney	123	237	0.07±0.05	0.45±0.12	0.09±0.03	46.8
Sex	M	Bone	240	NA	0.38±0.07	1.4 ±0.13	0.10±0.03	21.9
Occupation	Unknown							
Smoking	Pipe	Spleen	69	132	0.32±0.15	0.13±0.09	0.10±0.08	30.4

		Tissue Type	Mass (g)	Calcium µg/g ww	Th-228 pCi/kg	Th-230 pCi/kg	Th-232 pCi/kg	Thorium Recovery %
Case	78-23	Lung	385	175	0.02±0.01	0.22±0.03	0.13±0.02	41.0
Location	G.J.							
Age (Y)	63	LN	6.0	NA	1.1 ±0.82	5.5 ±1.1	2.0 ±0.64	34.1
Sex	N	Liver	520	100	0.16±0.07	0.06±0.03	0.06±0.03	48.0
Occupation	Machine Shop worker	Kidney	124	235	0.11±0.08	0.20±0.10	0.16±0.07	44.2
Smoking	None	Bone	260	7231	0.44±0.13	0.35±0.07	0.03±0.02	39.7
		Spleen	107	NA	-0.07±0.03	0.10±0.10	0.08±0.03	38.2
		Gonads	58	126	-0.09±0.04	0.09±0.04	0.07±0.03	82.7
		Heart	110	116	-0.04±0.03	0.15±0.04	0.08±0.03	37.0
		Blood	137	NA	-0.15±0.18	0.06±0.08	-0.02±0.07	17.0
Case No.	78-65	Lung	280	154	0.24±0.03	0.84±0.04	0.44±0.03	63.9
Location	G.J.							
Age (Y)	38	Liver	115	403	Lost	Lost	Lost	——
Sex	F	Bone	150	63000	0.64±0.08	0.60±0.06	0.10±0.02	21.9
Occupation	Teacher							
Smoking	None	Spleen	165	64.7	0.03±0.02	0.05±0.02	0.03±0.01	30.4
		Thyroid	10.0	550	0.33±0.50	0.82±0.37	0.65±0.33	43.5
Case No.	78.71	Lung	270	NA	1.8 ±0.09	6.0 ±0.15	2.87±0.11	64.9
Location	G.J.							
Age (Y)	60	LN	4.1	1340	7.2 ±3.3	24.6 ±6.2	22.9 ±6.1	8.1
Sex	F	Liver	440	62.1	0.06±0.02	0.50±0.05	0.06±0.02	32.9
Occupation	Unknown	Kidney	85	188	0.08±0.06	0.60±0.17	0.05±0.03	39.0
Smoking	Unknown	Bone	250	42800	0.37±0.08	2.35±0.18	0.22±0.05	58.8
		Spleen	60	122	0.12±0.08	0.30±0.07	0.13±0.05	65.3

		Tissue Type	Mass (g)	Calcium µg/g ww	Th-228 pCi/kg	Th-230 pCi/kg	Th-232 pCi/kg	Thorium Recovery %
Case No.	78-80	Lung	720	199	0.18±0.02	0.41±0.04	0.58±0.04	65.7
Location	G.J.							
Age (Y)	65	LN	8.8	NA	2.2 ±0.84	7.8 ±1.3	5.9 ±1.0	23.2
Sex	M	Liver	760	62.2	0.27±0.01	0.03±0.01	0.02±0.01	47.6
Occupation	Moving estimator	Kidney	210	199	0.11±0.03	0.09±0.02	0.04±0.01	69.8
Smoking	2 pk/day for 50 years	Bone	73	37400	1.03±0.18	2.71±0.30	0.30±0.10	22.2
		Spleen	165	NA	0.04±0.01	0.07±0.02	0.04±0.02	47.3
Case No.	78-110	Lung	367	248	0.41±0.08	4.15±0.18	0.10±0.08	21.4
Location	G.J.							
Age (Y)	84	LN	5.3	NA	4.6 ±4.0	15.7 ±3.1	8.7 ±2.7	13.8
Sex	M	Liver	708	46.3	0.02±0.01	0.14±0.01	0.02±0.01	65.5
Occupation	Welder							
		Kidney	124	352	0.09±0.10	0.33±0.10	0.01±0.01	19.6
Smoking	None	Bone	290	43800	0.29±0.08	2.21±0.14	0.18±0.04	26.0
		Spleen	98	149	0.04±0.06	0.12±0.04	0.27±0.07	46.2
		Gonads	61	225	-0.04±0.07	0.02±0.03	0.02±0.01	81.8
Case No.	78-125	Lung	630	280	0.77±0.07	1.5 ±0.07	0.78±0.05	33.7
Location	G.J.							
Age (Y)	75	LN	8.5	NA	4.7 ±1.6	14.0 ±2.3	8.9 ±1.9	43.0
Sex	M	Liver	450	81.0	0.05±0.02	0.02±0.01	0.02±0.01	31.1
Occupation	Coal Miner							
Smoking	None	Bone	81	65900	1.1 ±0.24	2.3 ±0.23	0.17±0.06	25.6
		Spleen	167	130	0.02±0.04	0.09±0.02	0.03±0.01	68.0

		Tissue Type	Mass (g)	Calcium µg/g ww	Th-228 pCi/kg	Th-230 pCi/kg	Th-232 pCi/kg	Thorium Recovery %
Case No.	79-1	Lung	487	269	0.02±0.01	0.29±0.03	0.05±0.01	36.8
Location	G.J.							
Age (Y)	61	LN	4.0	NA	3.6 ±3.2	12.4 ±6.4	6.5 ±3.6	16.4
Sex	M	Liver	533	NA	-0.01±0.01	0.05±0.01	0.01±0.01	55.3
Occupation	Salesman							
Smoking	Unknown	Kidney	157	162	0.23±0.27	0.27±0.10	0.17±0.08	58.0
		Bone	205	53200	0.40±0.14	1.2 ±0.14	0.12±0.04	18.6
		Spleen	170	NA	0.01±0.03	0.19±0.03	0.02±0.01	51.6
Case No.	79-2	Lung	579	213	0.45±0.10	0.67±0.08	0.86±0.09	32.7
Location	G.J.							
Age (Y)	79	LN	5.1	NA	6.9 ±2.9	8.3 ±2.6	8.3 ±2.6	35.9
Sex	M	Liver	283	141	0.07±0.03	0.19±0.02	0.03±0.01	50.0
Occupation	Electrician							
Smoking	None	Kidney	160	159	0.04±0.10	0.01±0.01	0.01±0.01	63.5
		Bone	285	56700	0.91±0.09	0.28±0.04	0.10±0.02	54.0
		Spleen	120	227	0.08±0.08	0.10±0.04	0.06±0.03	40.6
		Gonads	85	214	0.07±0.09	0.09±0.05	0.06±0.03	44.7

Case No.	1	Lung	270	175	0.40±0.04	0.71±0.04	0.52±0.04	66.6
Location	Wash., D.C.							
Age (Y)	26	Lymph nodes	3.3	1120	9.4 ±2.8	7.1 ±2.1	4.6 ±1.7	22.5
Sex	M	Liver	220	33.2	0.05±0.01	0.07±0.02	0.04±0.01	67.2
Occupation	Construction							
Smoking	Unknown	Kidney	120	122	0.08±0.01	0.03±0.01	0.02±0.01	91.2
		Bone	59	76300	0.87±0.12	0.34±0.07	0.04±0.04	61.6
Case No.	2	Lung	409	244	0.04±0.03	0.07±0.02	0.07±0.01	35.5
Location	Wash., D.C.							
Age (Y)	40	Lymph nodes	7.8	705	1.1 ±0.84	2.7 ±0.50	1.1 ±0.34	54.7
Sex	F	Liver	315	80.6	0.24±0.07	0.47±0.09	0.10±0.04	29.2
Occupation	Govt.							
	employee	Kidney	120	198	0.14±0.06	0.19±0.06	0.08±0.04	45.4
Smoking	Yes -	Bone	39	79200	0.66±0.18	0.45±0.11	0.14±0.07	79.8
	Unspecified							
Case No.	3	Lung	240	250	0.20±0.05	0.41±0.06	0.24±0.05	18.3
Location	Wash., D.C.							
Age (Y)	29	Lymph nodes	5.5	1000	5.3 ±1.3	11.0 ±1.4	5.8 ±1.0	45.2
Sex	F	Liver	250	37.0	0.30±0.07	0.45±0.09	0.19±0.06	71.6
Occupation	Bank clerk							
Smoking	Yes -	Bone	38	71100	1.3 ±0.25	0.17±0.09	-.002±0.06	65.4
	Unspecified							
Case No.	4	Lung	300	206	0.04±0.04	0.19±0.03	0.15±0.03	26.5
Location	Wash., D.C.							
Age (Y)	39	Lymph nodes	6.4	859	1.9 ±0.57	5.7 ±0.75	2.6 ±0.5	62.2
Sex	M	Liver	290	69.0	0.09±0.05	0.19±0.03	0.05±0.01	27.8
Occupation	Construction							
Smoking	Yes -	Kidney	115	127	0.05±0.02	0.14±0.03	0.02±0.01	79.0
	Unspecified	Bone	81	73900	1.2 ±0.15	0.29±0.06	0.06±0.03	85.8

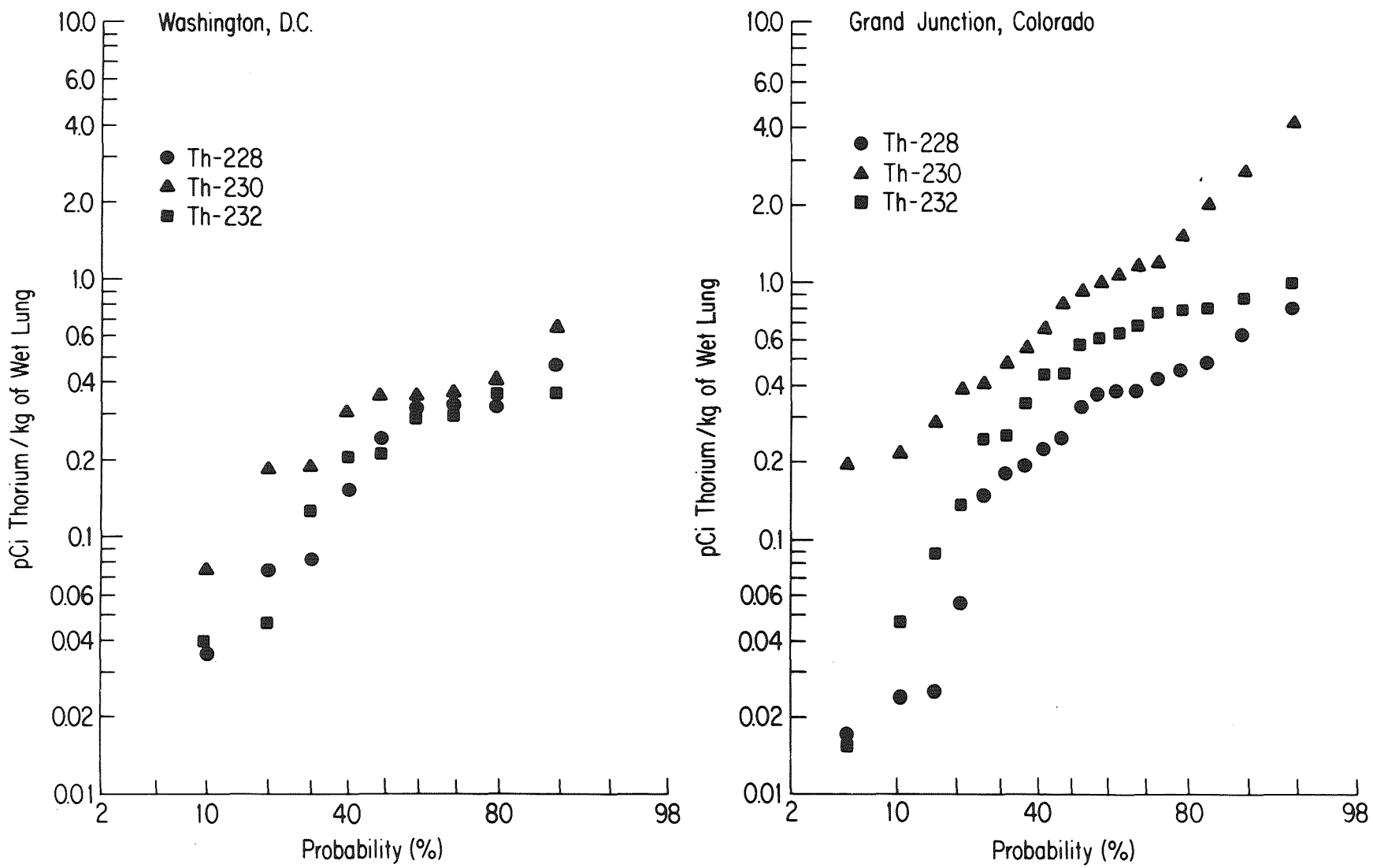
		Tissue Type	Mass (g)	Calcium µg/g ww	Th-223 pCi/kg	Th-230 pCi/kg	Th-232 pCi/kg	Thorium Recovery %
Case No.	5	Lung	395	202	0.28±0.05	0.36±0.04	0.32±0.03	36.7
Location	Wash., D.C.							
Age (Y)	32	Lymph nodes	12.5	440	2.4 ±0.76	4.8 ±0.79	3.0 ±0.62	46.6
Sex	M	Liver	225	50.2	0.03±0.01	0.05±0.01	0.02±0.01	63.9
Occupation	Construction	Kidney	135	95.0	0.10±0.03	0.10±0.02	0.02±0.01	40.7
Smoking	Yes - Unspecified							
Case No.	6	Lung	53	138	0.20±0.11	0.31±0.10	0.33±0.04	40.3
Location	Wash., D.C.							
Age (Y)	1.5	Lymph nodes	1.2	NA	-0.01±0.02	0.35±0.23	0.12±0.12	50.5
Sex	M	Liver	180	51.0	0.01±0.04	0.09±0.03	0.09±0.03	29.4
Smoking	None	Bone	20	36500	0.57±0.18	0.20±0.20	0.10±0.10	38.7
Case No.	7	Lung	340	443	0.29±0.03	0.66±0.03	0.31±0.02	75.5
Location	Wash., D.C.							
Age (Y)	61	Lymph nodes	3.9	365	1.6 ±2.1	1.6 ±2.1	0.66±0.66	30.7
Sex	F	Liver	128	71.1	0.18±0.05	0.11±0.03	0.07±0.02	32.4
Occupation	Housewife							
Smoking	Yes -	Kidney	70	130	0.36±0.27	0.37±0.21	0.12±0.12	72.0
	Unspecified	Bone	29	119000	0.56±0.33	0.85±0.36	0.20±0.20	48.6
Case No.	8	Lung	400	250	0.34±0.05	0.37±0.04	0.45±0.04	38.7
Location	Wash., D.C.							
Age (Y)	48	Lymph nodes	8.7	632	2.7 ±1.1	6.7 ±1.3	3.9 ±1.0	33.0
Sex	F	Liver	172	138	0.18±0.07	0.23±0.05	0.03±0.02	31.5
Occupation	Unknown							
Smoking	Yes - Unspecified	Kidney	69	186	0.10±0.09	0.09±0.03	0.01±0.03	84.9
Case No.	9	Lung	350	249	0.13±0.04	0.18±0.04	0.08±0.03	14.6
Location	Wash., D.C.							
Age (Y)	35	Lymph nodes	7.1	NA	2.8 ±0.81	4.4 ±0.74	2.6 ±0.57	45.0
Sex	M	Liver	305	NA	0.05±0.03	0.03±0.01	0.01±0.01	38.2
Occupation	Painter							
Smoking	None	Kidney	110	166	0.07±0.04	0.35±0.05	0.03±0.01	84.6
		Bone	49	36700	0.20±0.07	0.32±0.09	0.14±0.07	38.0

		Tissue Type	Mass (g)	Calcium µg/g ww	Th-228 pCi/kg	Th-230 pCi/kg	Th-232 pCi/kg	Thorium Recovery %
Case No.	10	Lung	410	111	0.35±0.04	0.36±0.03	0.33±0.03	40.5
Location	Wash., D.C.							
Age (Y)	31	Lymph nodes	4.2	881	3.6 ±1.7	3.0 ±1.0	5.7 ±1.5	48.1
Sex	M	Liver	177	62.2	0.08±0.04	0.70±0.09	0.06±0.03	21.5
Occupation	Clerk							
Smoking	Yes - Unspecified							

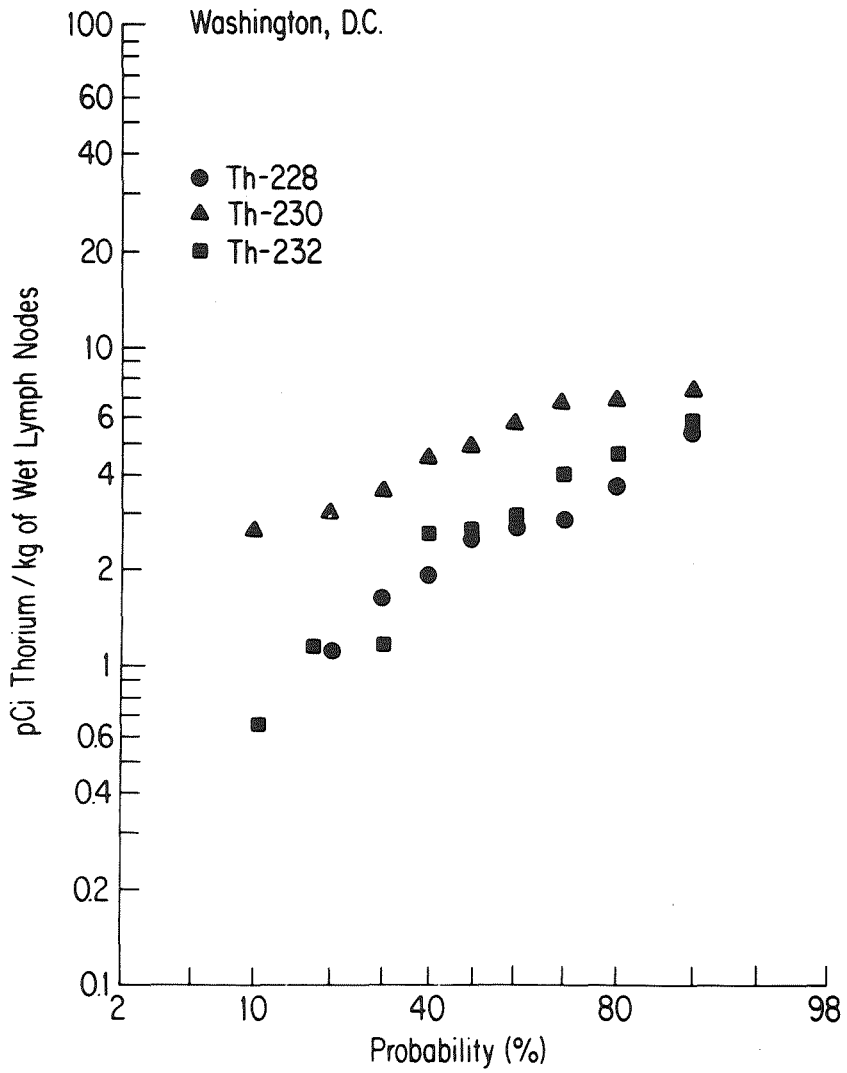
APPENDIX B

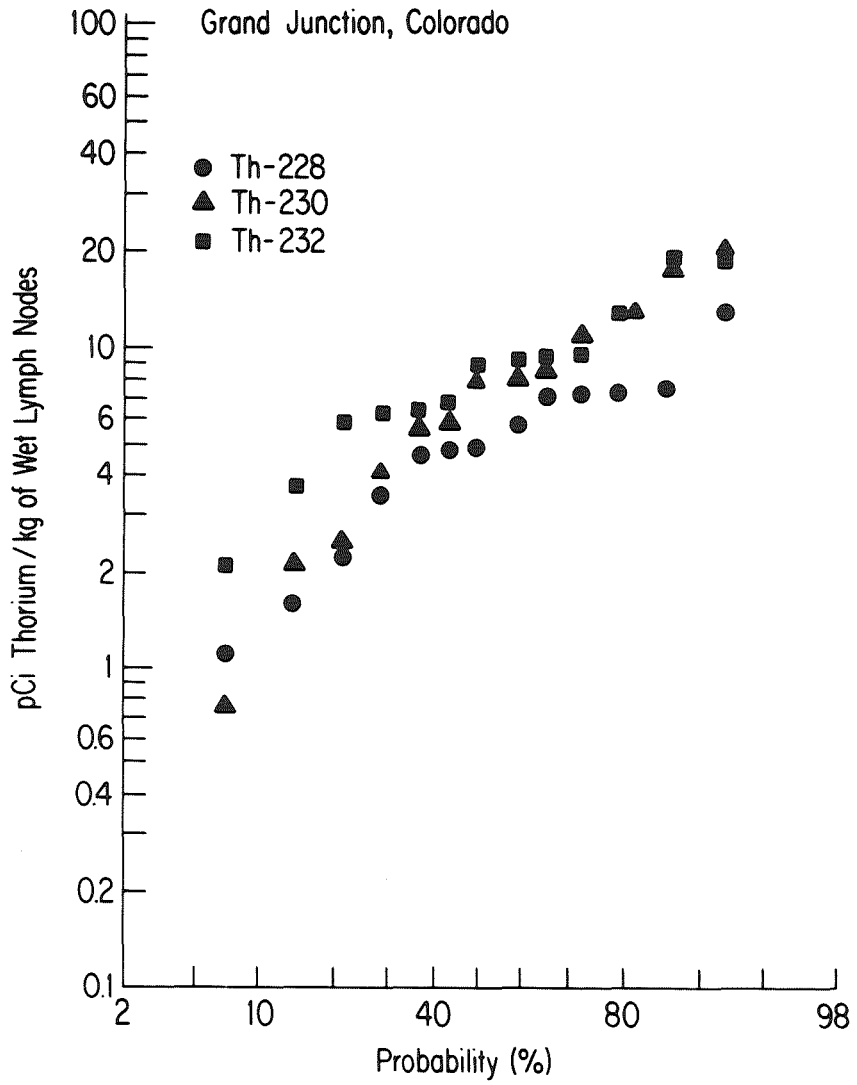
Cumulative frequency distribution for thorium isotopes in lung, lymph nodes, liver, kidney, spleen, and bone of Grand Junction and Washington, D.C. samples.

Cumulative Probability Distribution of Measured Thorium Concentration in Lung

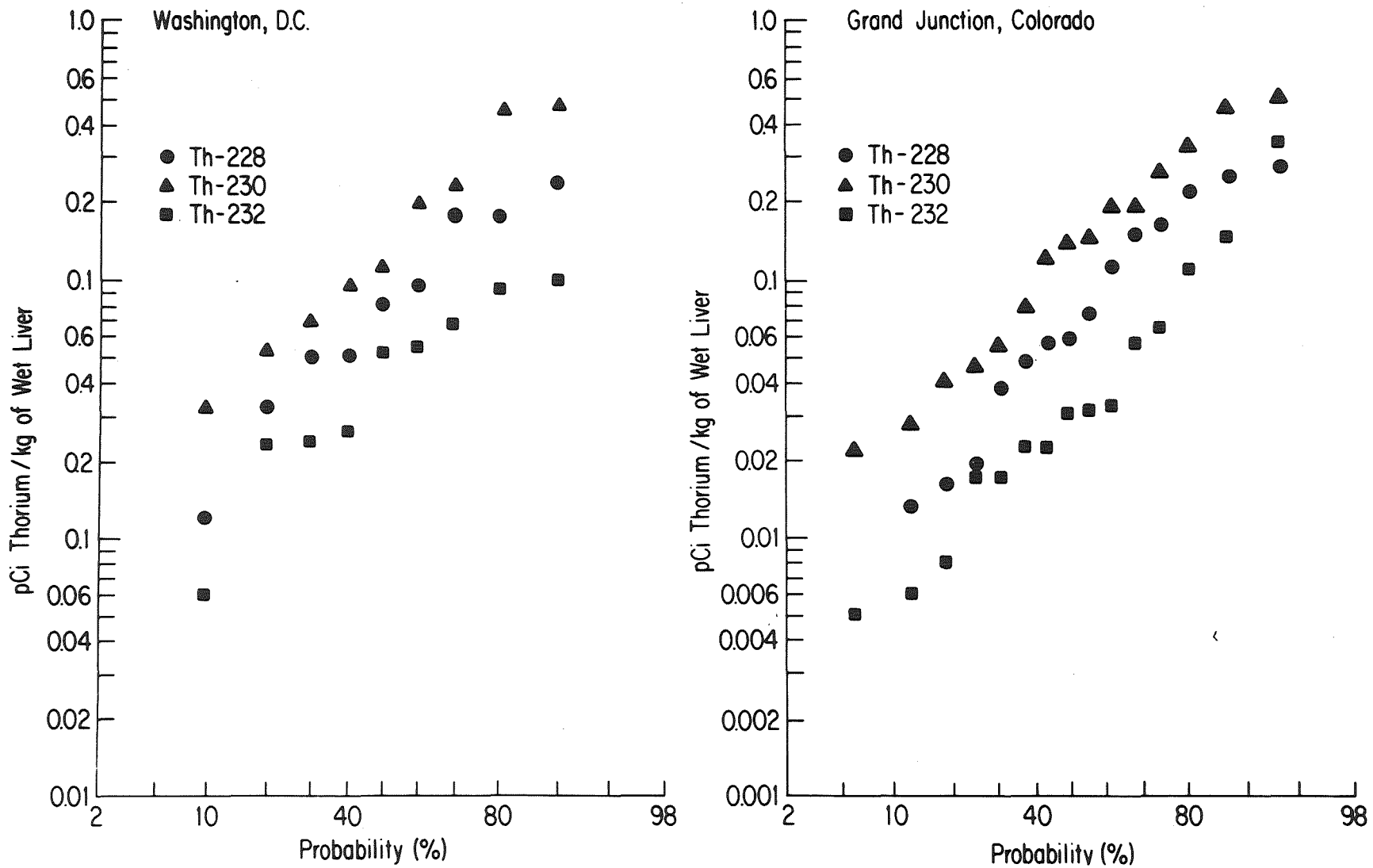


Cumulative Probability Distribution of Measured Thorium Concentration in Lymph Nodes

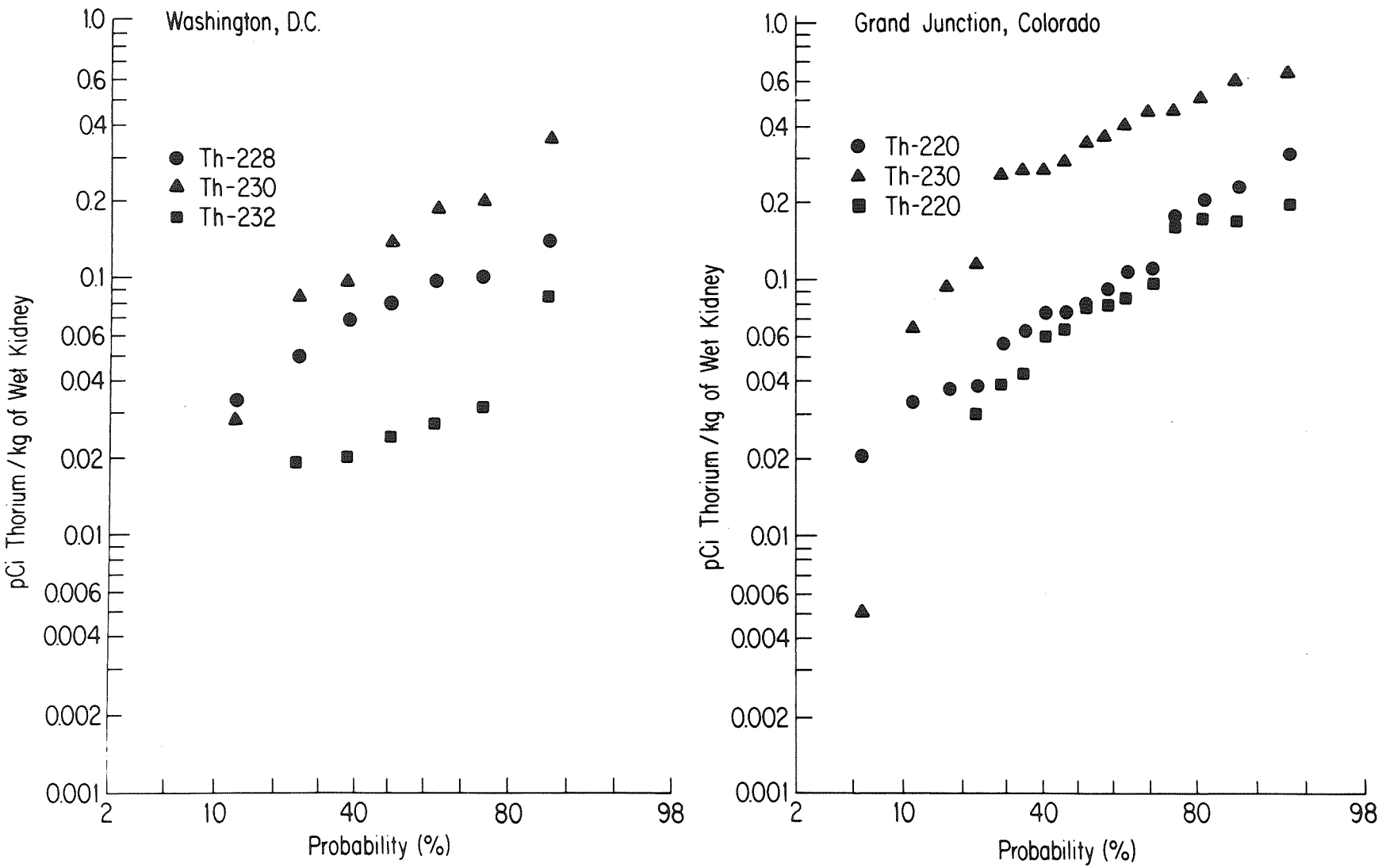


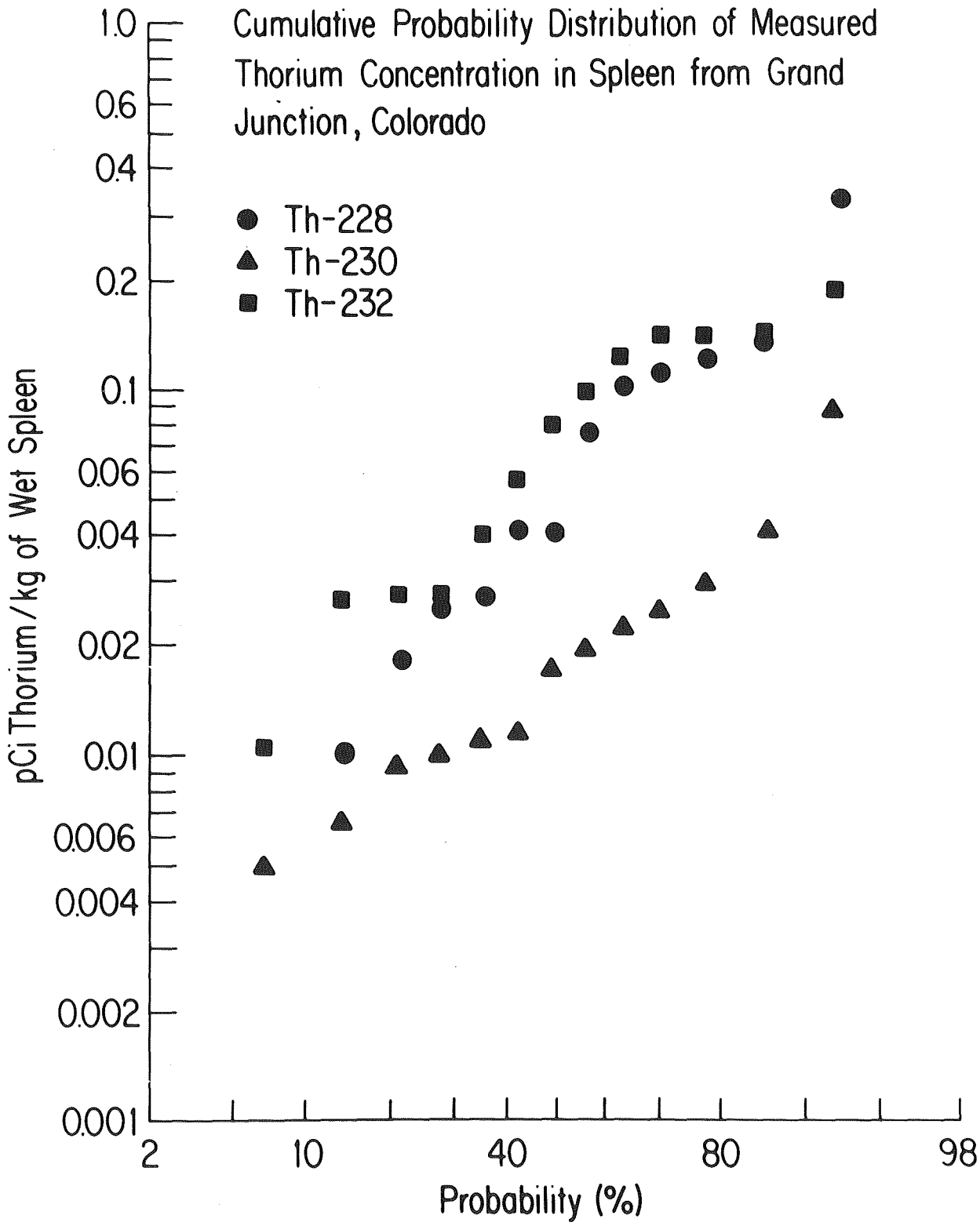


Cumulative Probability Distribution of Measured Thorium Concentration in Liver

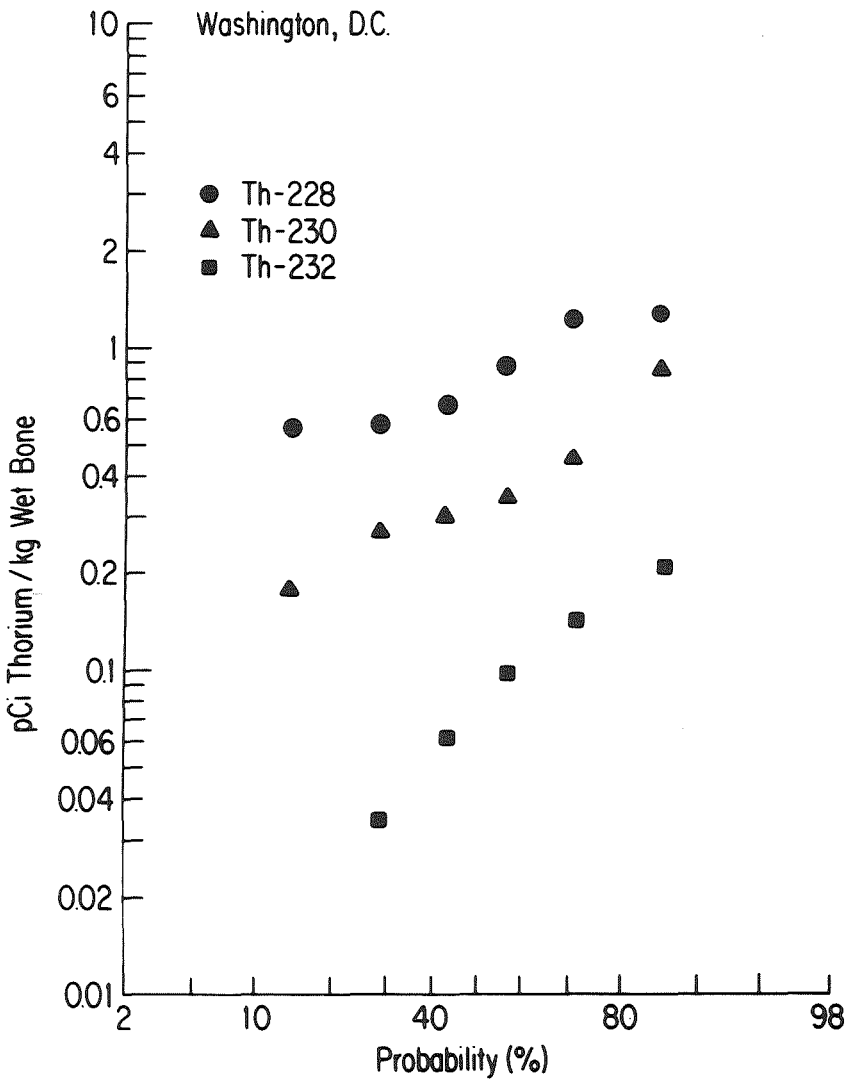


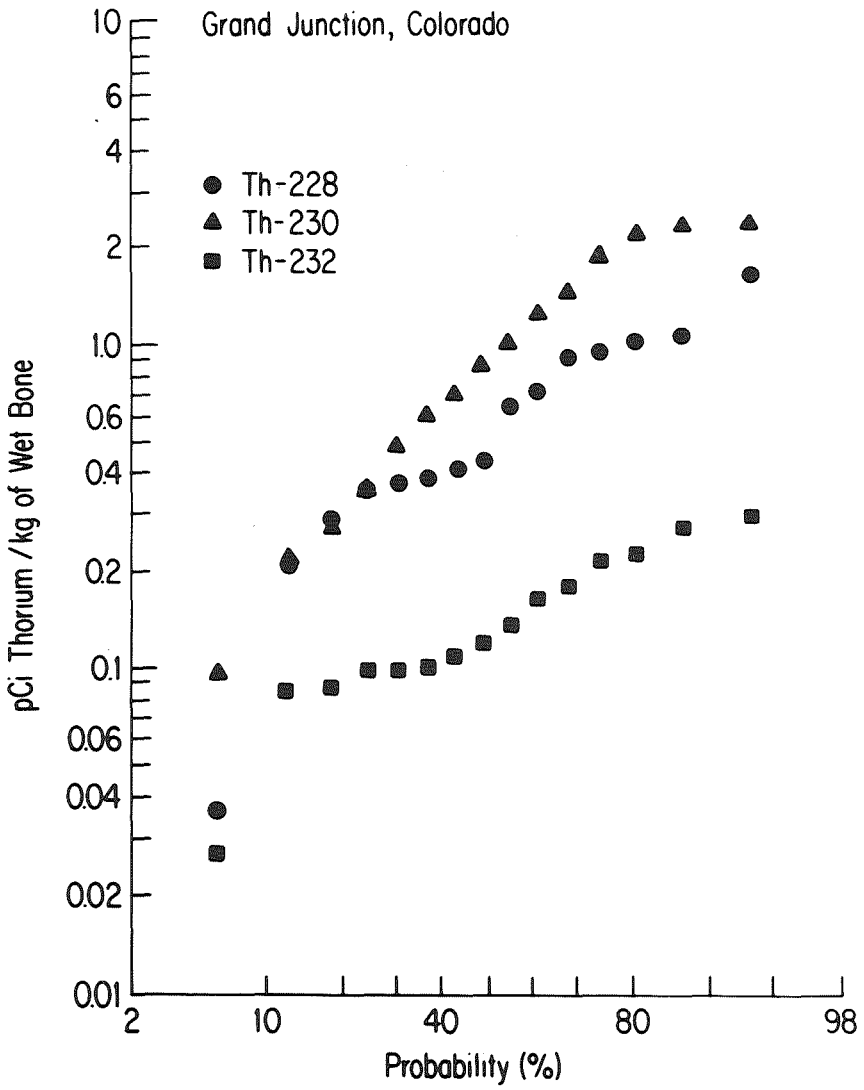
Cumulative Probability Distribution of Measured Thorium Concentration in Kidney




Cumulative Probability Distribution of Measured Thorium Concentration in Spleen from Grand Junction, Colorado
Th-228
Th-230
Th-232
pCi Thorium / kg of Wet Spleen
1.0
0.6
0.4
0.2
0.1
0.06
0.04
0.02
0.01
0.006
0.004
0.002
0.001
2
10
40
80
98
Probability (%)

Cumulative Probability Distribution of Measured Thorium Concentration in Bone





NRC FORM 335 (7-77)

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

1. REPORT NUMBER (Assigned by DDC): NUREG/CR-1227

4. TITLE AND SUBTITLE (Add Volume No., if appropriate): Thorium in Human Tissues

2. (Leave blank)

3. RECIPIENT'S ACCESSION NO.

7. AUTHOR(S): M.E. Wrenn, N.P. Singh, N. Cohen, S.A. Ibrahim, G. Saccomanno

5. DATE REPORT COMPLETED: MONTH December, YEAR 1980

9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
New York University Medical Center
Institute of Environmental Medicine
550 First Avenue
New York, NY 10001

DATE REPORT ISSUED: MONTH September, YEAR 1981

6. (Leave blank)

8. (Leave blank)

12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
Division of Health, Siting and Waste Management
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555

10. PROJECT/TASK/WORK UNIT NO.

11. CONTRACT NO.: FIN B5756

13. TYPE OF REPORT: Formal

PERIOD COVERED (Inclusive dates): 9/1/76 - 3/31/80

15. SUPPLEMENTARY NOTES

14. (Leave blank)

16. ABSTRACT (200 words or less)

The content of Th-228, Th-230 and Th-232 was investigated in human tissue from the general public without known occupational exposure. Twenty-two sets of human tissues from Grand Junction, Colorado and ten sets from Washington, D.C. were analyzed. Organs included were lung, pulmonary lymph nodes, liver, kidney, spleen and bone.

A radiochemical method was developed for the determination of environmental levels of alpha-emitting isotopes of thorium in tissue consisting of tissue digestion, separation of thorium isotopes by solvent extraction, electrodeposition, followed by alpha-spectrometric measurement using a surface barrier silicon diode. The method utilizes Th-229 as an internal tracer for radiochemical recoveries, and Th-228, Th-230, and Th232 are measured simulatneously.

Concentrations of Th-230 and Th-232 decrease in the following order: lymph nodes > lung > bone > kidney > liver, and Th-228: lymph nodes > bone > lung > kidney > liver. As expected, Th-230 and Th-232 were distributed similarly with about 11-27% of the body content in lung, and 55-70% in the skeleton. Th-228 was distributed differently with about 80% in the skeleton and only 5% in the lungs. Thorium-230 and 232 concentrations in bone were found to increase approximately linearly with age. The observed distribution of thorium among the organs is in good agreement with that recently adopted by the ICRP (ICRP 30, 1979).

The total accumulation of thorium in the human body was equivalent to the average amount of natural thorium in 1 to 6 gms of soil. The highest median radiation dose rate for the groups studied from all 3 isotopes of Th was 5 and 0.6 mrad/year delivered to pulmonary lymph nodes and bone respectively.

17. KEY WORDS AND DOCUMENT ANALYSIS

Thorium isotopes
Radiochemical procedure
Human Organs
Alpha-spectrometry
Radiation doses
Lung

17a. DESCRIPTORS

Bone
Liver
Lymph Nodes

17b. IDENTIFIERS/OPEN-ENDED TERMS

18. AVAILABILITY STATEMENT: Unlimited

19. SECURITY CLASS (This report): Unclassified

20. SECURITY CLASS (This page)

21. NO. OF PAGES

22. PRICE: $

NRC FORM 335 (7-77)

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

OFFICIAL BUSINESS
PENALTY FOR PRIVATE USE, $300

POSTAGE AND FEES PAID
U.S. NUCLEAR REGULATORY
COMMISSION

U.S. MAIL