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Concentrations of Uranium and Thorium Isotopes in Uranium Millers' and Miners' Tissues

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THE CONCENTRATIONS OF URANIUM AND THORIUM ISOTOPES
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EXECUTIVE SUMMARY

The concentrations of alpha-emitting isotopes of uranium (U-238, U-235, and U-234 and thorium (Th-232, Th-230 and Th-228) were determined in the lungs of 14 former uranium miners and in the soft tissues and bones of three former uranium miners and two former uranium millers. Soft tissues included lung, liver, kidney, spleen, heart, and gonads. Bones included vertebrae, ribs, and sternum. However, all soft tissues and bones could not be procured from each miner and miller. Uranium and thorium isotopes were also determined in soft tissues and bones of seven normal healthy persons from the general population who had died suddenly.

Uranium and thorium were determined by radiochemical procedures developed in this laboratory. The methods consist of tissue digestion, separation of uranium and thorium from each other and also from other elements by solvent extraction, electrodeposition, and solid-state alpha-spectrometric measurements using Si(Li) surface barrier detectors.

The concentrations of U-238, U-234 and Th-230 measured in 17 former uranium miners lungs are as follows: U-238 ranged from 1.6 to 311 pCi/kg with an average of 75 pCi/kg, U-234 ranged from 2.5 to 325 pCi/kg with an average of 80 pCi/kg and Th-230 ranged from 3.7 to 306 pCi/kg with an average of 79 PCi/kg wet weight. The concentrations of U-238 and U-234 in individual lungs were close to equilibrium with an average ratio of U-238/U-234 being 0.92 and range of 0.64 to 1.06. The mean ratio of Th-230/U-234 was 1.04 with a range of 0.33 to 3.54. The near equilibrium between Th-230 and U-238,234 indicates that the elimination rates of uranium and thorium are similar in former uranium miners' lungs.

The results of the determination of concentrations of uranium and thorium isotopes in soft tissues and bones of three former uranium miners suggest that the concentrations of U-238 and U-234 were very high in lung as compared to all other soft tissues as well as bone. However, the concentration of Th-230 in bones was either higher or comparable to its concentration in lung but much higher than its concentration in all other soft tissues. These results, therefore, suggest that uranium cleared from lung does not deposit in bone as much as thorium and/or uranium may also clear from the bone with a much shorter biological half life. Similar results were obtained in soft tissues and bones of two former uranium millers.

The concentrations of uranium and thorium isotopes were measured in the soft tissues and bones of seven individuals from general populations, mostly from the state of Colorado. These results give the background concentrations of these isotopes in man without any occupational exposure. The higher concentrations of U-238, U-234 and Th-230 in uranium miners and millers tissues can mainly be attributed to the occupational exposure, since the

concentrations of these isotopes in soft tissues and bones of general population are very low.

In contrast to the results obtained from animals exposed to uranium ore dust where Th-230/U-238 ratios in lung increase with time, our results on uranium miners' and millers' lungs show the average Th-230/U-234 and Th-230/U-238 ratios to be near equilibrium, suggesting that the mobilization of uranium and thorium from the lung occurs at approximately the same rate. To determine whether analytical problems are responsible for the different results in human and animal studies, an interlaboratory comparison on the determinations of U-238, U-234 and Th-230 in eight dog lungs exposed to uranium ore dust at Pacific Northwest Laboratory was undertaken. The results obtained from three different laboratories including Los Almos National Laboratory, Pacific Northwest Laboratory and University of Utah Radiobiology Laboratory, indicate generally good agreement between laboratories. The activity ratios from all three laboratories indicated that a fairly large disequilibrium between uranium isotopes and Th-230 exist. The explanation of this difference between human and experimental animals remains to be resolved, but it is clearly not caused by a difference in radiochemical methods.

The concentration ratios of Th-230/U-234 in bone of uranium miners and millers measured in our laboratory have been compared with the results predicted by the ICRP 30 metabolic models for uranium and thorium. The results indicate that the ICRP metabolic models for thorium and uranium were only marginally successful in predicting the ratio of Th-230/U-234 in bones and differed greatly for soft tissues.

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CHAPTER 1

INTRODUCTION

In 1960, when the Atomic Energy Commission (AEC) adopted the uranium ore dust maximum permissible concentration (MPC), it was believed that the behavior of Th-230 in uranium ore dust in the lung would be similar to that of the prevalent uranium in the dust. They believed that Th-230 in ore dust should be assigned an MPC of 1×10^{-10} µCi/ml, rather than the value of 1×10^{-11} µCi/ml considered appropriate for insoluble Th-230 (RO60, FR60). Subsequently, evidence from animal studies showed that uranium and thorium in laboratory generated uranium ore aerosol are metabolized at different rates, with the Th-230 remaining in the lung for much longer periods than the U-238 and U-234 (Stu75).

The purpose of this study is to evaluate the concentrations of uranium and thorium isotopes in uranium mill workers' tissues in order to aid in the evaluation of the adequacy of the special ore dust limits. We found that most millers have also been involved in mining at some time in their lives, making it necessary to determine the concentration of these uranium and thorium isotopes in tissues from uranium miners for comparison with results from those who were both miners and millers. In addition, knowledge of background concentrations of uranium and thorium isotopes in human tissues from people who were not occupationally exposed is essential to properly evaluate the importance of the concentrations of uranium and thorium isotopes in tissues. Our earlier studies determined isotopic concentrations of thorium in tissues of the general population (Wr 81). As a result further study was necessary in which uranium and thorium isotopes were measured in the same tissue samples.

Battelle Pacific Northwest Laboratories conducted inhalation studies in animals in order to identify the levels of uranium mine air contaminants necessary to produce the lung cancers. Among these studies eight dogs were exposed to uranium ore dust. The lung tissues obtained from these dogs at autopsy were divided into three sections and distributed to three different laboratories; the Radiobiology Laboratory at the University of Utah (Dr. N.P. Singh), Los Alamos National Laboratory (Dr. James McInroy), and Battelle Pacific Northwest Laboratory (Dr. Peter Jackson). These tissues were analyzed by all three laboratories for the concentrations of uranium and thorium isotopes and an interlaboratory comparison was conducted. These results are reported in Chapter 2.

CHAPTER 2

MATERIALS AND METHODS

2.1 Collection of Tissues

We contacted a number of pathologists for their cooperation in collecting tissues from former uranium millers. Geno Saccomanno, M.D., Ph.D., of St. Mary's Hospital, Grand Junction, Colorado, was most helpful in providing tissue. Dr. Victor E. Archer, University of Utah, provided us with a few uranium miners' lungs. Dr. Steinbecker, Grand Junction, Colorado, also promised to provide us with tissue samples if possible. Dr. Samutz, Director, Uranium Epidemiological Study Cancer Research and Treatment Center, University of New Mexico, Albuquerque, New Mexico, was also asked for his cooperation in collecting tissues from former uranium millers and miners.

Dr. Bryce Breitenstein, former director of the Transuranium Registry in Richland, Washington, was contacted. They will undertake tissue collection, for which they are funded by the Department of Energy (DOE). In return we have agreed to provide the Registry with the results of our study.

We also contacted thirteen uranium mill operations requesting their cooperation. They were not able to provide us with access to the tissue samples required.

The State Medical Examiner's or Coroner's offices in the states of Utah, Wyoming, Colorado, and New Mexico were contacted and replied with assurances of cooperation.

In addition, a list of uranium millers was requested from Philip Landrigan, M.D., Chief, DSHEFS, CDC, PHS, DHHD, National Institute for Occupational Safety and Health, Cincinnati, Ohio, through Victor E. Archer, M.D. After resolving certain legal problems (as stipulated in Federal Register, Vol. 47, No. 104, p. 23577), we received a list containing over 2000 names of uranium millers from NIOSH, subject to the Privacy Act of 1974.

Whole lungs from eight uranium miners were collected at autopsy by Dr. Geno Saccomanno and sent to Dr. Oscar Auerbach at East Orange, New Jersey for detailed histological studies. The lung specimens were then sent to our laboratory for the determination of long-lived isotopes of uranium and thorium. Pathological status and weight of the tissue, occupational history, smoking habits, and the age at death of the subjects were noted. Pathological examinations revealed that four miners had squamous cell carcinoma, two had carcinoma in the upper left lobe, one had carcinoma of the ascending colon and carcinoma in situ in the lung and one had a tumor in the right pleural cavity with a drain to the pericardium. Information was unavailable on one of them. Lungs from six other former uranium miners were provided by Dr. Victor E. Archer. They were reported to have died of bronchogenic cancer.

We received three sets of tissues from former uranium miners and two sets from former uranium millers collected at autopsy by Dr. Geno Saccomanno at St. Mary's Hospital. One set of uranium miners tissues included lung, kidney,

spleen, heart, gonad, vertebrae, ribs, and sternum. Another included lung, liver, kidney, spleen, heart, gonad, vertebrae, ribs and sternum. A third included lung, gonad and vertebrae. The two sets of tissues obtained from former uranium mill workers included lung, lymph nodes, kidney, liver, gonad, heart and vertebrae.

Five sets of tissue samples obtained at autopsy from Colorado were collected for the determination of background concentrations of alpha-emitting isotopes of uranium and thorium. The tissues generally included lung, liver, kidney, spleen, gonad, ribs, vertebrae, and sternum.

2.2 Radiochemical Determination of Uranium and Thorium

Concentrations of uranium and thorium were determined using the procedures of Singh et al. (Si83a, 83b, 79a, 79b) and Wrenn et al. (Wr78).

Soft Tissues

Wet ashing. The weighed amount of tissue spiked with U-232 and Th-229 tracers was wet ashed by adding enough HNO_3 to immerse the tissues, followed by slow heating until frothing ceased. At this point, 20 ml of H_2SO_4 were added and the tissue was heated vigorously with occasional additions of 1 to 2 ml of HNO_3 and H_2O_2 until the evolution of brown fumes ceased, indicating the complete removal of organic materials. Most of the H_2SO_4 was then evaporated by continuous heating without going to complete dryness. The lung and lymph nodes require an additional heating with a mixture of 10-20 ml HNO_3 and 2-5 ml HF to dissolve insoluble materials, presumably silica. The HF was removed with continuous heating.

Co-precipitation. After wet ashing, the residue was dissolved in 100 ml each of 1:1 HNO_3 and 1:3 HCl. Then 0.2 ml of iron carrier was added to the clear solution, which was boiled for several minutes, and cooled. Uranium and thorium were co-precipitated by adding concentrated ammonium hydroxide, bringing the pH to between 10 and 11. The solution was then boiled for 15 minutes, cooled, and centrifuged. The supernatent was discarded and the precipitate dissolved in 25 ml of dilute HNO_3 (2-3 M). The sulphates were then reprecipitated with ammonium hydroxide, adjusting the pH to 10-11. This process was repeated until the supernatent was free of sulfate ions. The sulfate ions have been completely removed if barium chloride solution does not form any precipitate when added to an aliquot of the supernatent. Finally, the precipitate was dissolved in 30-40 ml of concentrated HCl and the acidity adjusted to 10 M.

Solvent Extraction. The tissue solution (40-50 ml) in 10 M HCl was transferred into a 250-ml polypropylene centrifuge tube. An equal volume (~ 50 ml) of 20% tri-lauryl amine (TLA) solution in xylene pre-equilibrated with 10 M HCl, was added to the same tube, shaken for ten minutes, centrifuged and transferred to a 250-ml separatory funnel. The aqueous phase was mixed with an equal volume of 20% TLA solution and the extraction repeated once more. Uranium was extracted into the organic phase, leaving thorium in the aqueous phase. Uranium was then back-extracted from the TLA phase by shaking with an equal volume of 0.1 M HCl for ten minutes.

Removal of iron. The aqueous phases obtained after the back-extraction of uranium also contained iron. When uranium was electrodeposited, iron was also co-electrodeposited, causing a massy plate which degraded the alpha-spectra of uranium. Therefore, it was essential to remove iron before electrodeposition.

The aqueous solutions (200-300 ml) containing uranium obtained after the two back-extractions were evaporated to dryness. However, just before the beakers were completely dry, 1-2 ml of HNO_3 and H_2O_2 were added to remove all organic matter. The residue was dissolved in a minimum volume (~ 20 ml) of concentrated HCl. The solution was transferred to a 50-ml polypropylene centrifuge tube. An equal volume (~ 20 ml) of isopropyl ether was added to the same tube and the tube was shaken for 2 min. The tube, with loosened cap, was warmed in a hot water bath for 2 to 3 min. The organic phase contained most of the iron, leaving uranium in the aqueous phase. To remove iron quantitatively, a minimum of 2 (~ 20 ml) extractions were required for uranium.

The aqueous phase left after the extraction of uranium, which contained most of the thorium, was evaporated to dryness without baking. The residue was dissolved in 4M HNO_3 and acidity adjusted to 4M. The solution was transferred to a 250-ml centrifuge tube. An equal volume of 20% TLA solution pre-equilibrated with 4M HNO_3 was added to the same centrifuge tube. The mixture was shaken for ten minutes, centrifuged and transferred into a 250-ml separatory funnel. The aqueous phase was mixed once again with an equal volume of 20% TLA solution pre-equilibrated with 4M HNO_3, shaken for ten minutes, centrifuged and transferred into the same separatory funnel. The aqueous phase was separated and discarded. The organic phase, which contained most of the thorium, was mixed with an equal volume of 10 M HCl, shaken for ten minutes, centrifuged and transferred into a 250-ml separatory funnel. The aqueous phase, which contained most of the thorium was transferred into a clean beaker. The back-extraction of thorium from the residual organic phase was repeated, and the aqueous phase added to that previously collected.

Electrodeposition. Five ml of 5% sodium bisulfate solution in 9 M H_2SO_4 was added to the (~ 20 ml) solutions containing uranium after the ether extraction of iron, and thorium. These solutions were then heated strongly with occasional additions of 1-2 ml HNO_3 to remove the last traces of organic matter, and evaporated to dryness. The residue was dissolved in 5 ml of 1 M $(NH_4)_2SO_4$ solution and transferred into the plating cell. The beaker was washed once with a 5 ml portion of 1 M $(NH_4)_2SO_4$ solution and transferred into the same plating cell. Concentrated ammonia was added drop by drop to a yellow endpoint, using thymol blue as an indicator. A few drops of 2 M H_2SO_4 were added until a salmon pink color was obtained. Uranium and thorium were electrodeposited onto platinum planchets at a constant current of 1.2 amp for 1 hr. The platinum planchets were then counted alpha-spectrometrically.

Bone

Wet ashing. Weighed bone samples were transferred into proper sized beakers (600-1000 ml) and spiked with 1-2 dpm of U-232 and Th-229 tracers. They were heated, but not boiled, on a hot plate to remove the fluid content of the bone. The beakers were transferred into a muffle furnace and heated overnight at a temperature of 550° C., obtained by gradual increments of

50° C. The beakers were cooled, and the bone ash dissolved in concentrated HNO_3 by heating, occasionally adding a few drops of HNO_3 and H_2O_2 until the evolution of brown fumes ceased. HNO_3 was removed by evaporation and the residue was dissolved in 100-200 ml of 1:3 HCl by heating.

Co-precipitation. Two hundred mg $SnCl_2$ and 25 ml HI were added to the bone solution obtained in 1:3 HCl with constant stirring and without boiling. Ammonium hydroxide was added slowly to adjust the pH so that precipitate formed but did not persist. One hundred ml of 10% oxalic acid solution was added and boiled for 15 minutes. The precipitate was cooled, filtered through a Buchner funnel, and washed two to three times with distilled water. The precipitate was transferred into a 400-600 ml beaker and heated overnight in a muffle furnace at 550° C., obtained by gradual increase of temperature in increments of 50° C. Calcium carbonate residue thus formed was dissolved in concentrated HNO_3 and heated with occasional additions of a few drops of HNO_3 and H_2O_2 until a clear, colorless solution was obtained. Nitric acid was evaporated and the residue was dissolved in 10 M HCl. The solution was transferred to a 250-ml polypropylene centrifuge tube.

Solvent extraction and electrodeposition were carried out in the same manner as discussed for soft tissues. The platinum planchets were then counted alpha-spectrometrically.

CHAPTER 3

URANIUM AND THORIUM MEASUREMENTS IN URANIUM MINERS' LUNGS

3.1 Collection of tissues

Eight lungs collected at autopsy by Dr. Geno Saccomanno were sent to Dr. Oscar Auerbach at East Orange, New Jersey for detailed histological studies. After these studies were completed, the lungs, preserved in formalin and packed in plastic bags were returned to us. An additional three lungs as well as other soft tissues and bone were sent directly to us packed in dry ice and another six lungs were provided by Dr. Victor E. Archer. Lung tissues were collected in accordance to the description given in Section 2.1. Additional information such as dates of birth, death, and retirement, smoking habits and occupational histories are included along with medical histories in Table 3.1.

3.2 Results

The concentrations of U-238, U-234 and Th-230 measured in uranium miners' lungs are given in Table 3.2 and typical alpha spectra of uranium and thorium isotopes in uranium miners' lungs are given in Figures 3.1 and 3.2. The concentration of U-238 ranged from 1.6 to 311 pCi/kg wet weight with an average of 75 pCi/kg. The concentration of U-234 ranged from 2.5 to 325 pCi/kg wet weight with an average of 80 pCi/kg.

The concentration of Th-230 ranged from 3.7 to 306 pCi/kg wet weight with an average of 79 pCi/kg, which is about 65 times higher than the concentration of Th-230 found in the lungs of the general population from a western mining region (Wr 81). The concentration of Th-230 in lungs of this non-occupationally exposed population of Grand Junction, Colorado ranged from 0.10 to 6.0 pCi/kg with a mean of 1.4 pCi/kg. The concentrations of Th-228 and Th-232 in uranium miners' lungs were as low as in the general population, with a mean concentration of 1.1 pCi/kg Th-228 and 1.2 pCi/kg Th-232.

3.3 Discussion

The concentrations of U-238 and U-234 in individual lungs were close to equilibrium with an average ratio of U-238/U-234 being 0.92, and ranging between 0.64 and 1.06. The mean ratio of Th-230/U-234 was 1.04 with a range between 0.33 and 3.54. The equilibrium between Th-230 and U-238,234 indicates that the elimination rates of uranium and thorium in lung are similar. This differs from results reported for beagles which chronically inhaled carnotite ore, where the Th-230/U-234 ratio was reported to range from 5.4 to 7.4, with an average of 6.3.

Stuart and Jackson (Stu75) reported that a rapid in vivo separation of thorium from uranium occurs in lungs, tracheobronchial lymph nodes and systemic organs when inhaled as constituents of uranium ore. This pattern has been found with two types of ores in three species of mammals, and with a variety of concentrations and exposure schedules.

Clearly, the results reported for animals differ from those in uranium miners tissues in that Th-230 exceeds the uranium precursor in the animals, but on the average does not in humans. Materials inhaled, particle size, temporal patterns of exposure, radiochemical techniques, and/or dissolution rates between man and experimental animals could explain these differences.

Fisher et al (Fi83) made measurements of uranium and thorium in the urine and feces of active uranium mill crusher operators, retired crusher operators, and a control group not occupationally exposed to uranium ore dust. They found that U-238/Th-230 concentration ratio in urine to be ~ 2 in the control group, ~ 0.7 in the retired group, and ~ 23 in the active group, because more uranium than thorium is excreted in the urine of the control group and the workers, but it does not necessarily mean that uranium is dissolved faster than thorium in the lungs of these workers, as inferred by the authors. Increased urinary excretion could also be due to larger intake of uranium in these workers, presumably from other sources such as yellow cake dust from direct exposure near the precipitation, drying, packaging areas of the mill, or from inhalation of yellow cake dust released during dryer stack emissions, as the authors themselves point out.

TABLE 3.1
Occupational, Smoking, Medical and Personal Histories of Former Uranium Miners

Sample No.	Date of Birth	Date of Death	Age at Death	Date of Retirement	Occupational History	Smoking Habits	Cause of Death	Miscellaneous
U-214	11-28-15	10-20-70	55	1963	5549 WLM 17 yrs UGU	1 ppd x 30 yrs quit in 1956	Bronchogenic cancer	
U-215	07-07-08	10-25-70	60		3177 WLM 18 yrs UGU	1 ppd x 45 yrs	Bronchogenic cancer	Some mill experience
U-225	09-04-14	08-18-71	57	1962	2576 WLM 15 yrs UGU	1 ppd x 30 yrs quit in 1969	Bronchogenic cancer	No mill work
U-241	04-15-23	10-29-71	48		2057 WLM		bronchogenic cancer	Oil shale refining 4-47, 11-48
U-242	01-29-15	10-21-71	56	August 1966	3087 WLM 28 yrs UGU 2 yrs HRUG	1 ppd x 42 yrs	Bronchogenic cancer	No mill work
U-243	08-26-08	11-20-71	63	1965	1951 WLM 20 yrs UGU 3 yrs HRUG	2 ppd x 44 yrs quit in 1967	Bronchogenic cancer	
U-246	00-00-11	12-00-71	60		Heavy equipment operator			9/71—right pneumonectomy 12/71—autopsy: metastatic carcinoma to liver, adrenal gland, bone, and retroperitoneal soft tissue
U-337	00-00-23	04-01-75	52	$\frac{1}{2}$ ppd x 30 yrs	Never miner, mineral & oil driller			Squamous cell carcinoma (WHO-1A), emphysema, acute bronchitis, pulmonary fibrosis, all lobes of lung
U-342	10-10-15	05-18-75	60		10 yrs UGU and mill 10 yrs HRUG	1 ppd x 30 yrs	Oat cell carcinoma	Metastatic carcinoma to liver, spleen and adrenal glands

TABLE 3.1 (continued)

Sample No.	Date of Birth	Date of Death	Age at Death	Date of Retirement	Occupational History	Smoking Habits	Cause of Death	Miscellaneous
U-343			78				Broncho-pneumonia	Autopsy—Carcinoma of lung, history of "long" mining activity, no smoking history
U-344	09-06-01	06-01-75	74		30 yrs UGU No coal/HR	2-3 ppd x 50 yrs quit in 1963	Coronary thrombosis	1/71—diagnosed CIS lung 6/74—excision of lesion of right forearm At death-squamous cell carcinoma (WHO-1B) metastatic to liver, spindle cell carcinoma of right forearm
U-350	09-02-10	05-00-75	65	1970	25 yrs UGU 25 yrs HRUG	1-2 ppd x 50 yrs quit in 1968	Broncho-pneumonia	9-26-74—Upper right lobe of lung removed (squamous cell carcinoma)
U-367	04-01-00	03-18-77	77	1958	6 yrs UGU 18 mos HR	1-1 1/2 ppd x 62 yrs 1/2 ppd x 5 yrs until death	Aortic aneurysm	At death—carcinoma of ascending colon, CIS lung (WHO-1A)
U-383	01-01-13	12/00/78	65	1959	21 yrs UGU 3 yrs HRUG 1 yr coal	1 ppd x 47 yrs		At death—squamous cell carcinoma (right upper lobe)
U-384	07-07-18	08-01-79	61		18 yrs UGU	1 ppd x 37 yrs quit in 1979		
U-522	00-00-23	12-03-82	59		10 yrs UGU	1 ppd x 40 yrs quit in 4/1982		Oat cell carcinoma
U-536	08-10-37	06-04-82	45	1982		"heavy" smoker		
83-A-7[illegible]	09-20-25	07-14-83	58		0 yrs UGU mill mechanic 33 yrs; around yellow cake U	3 ppd x 8 yrs quit in 1951		Recent surgery: decortication of tumor of right pleural cavity with drain to pericardium Autopsy: mesothelioma (right pleural space), right pyothorax and hemocardium with mesothelioma extension

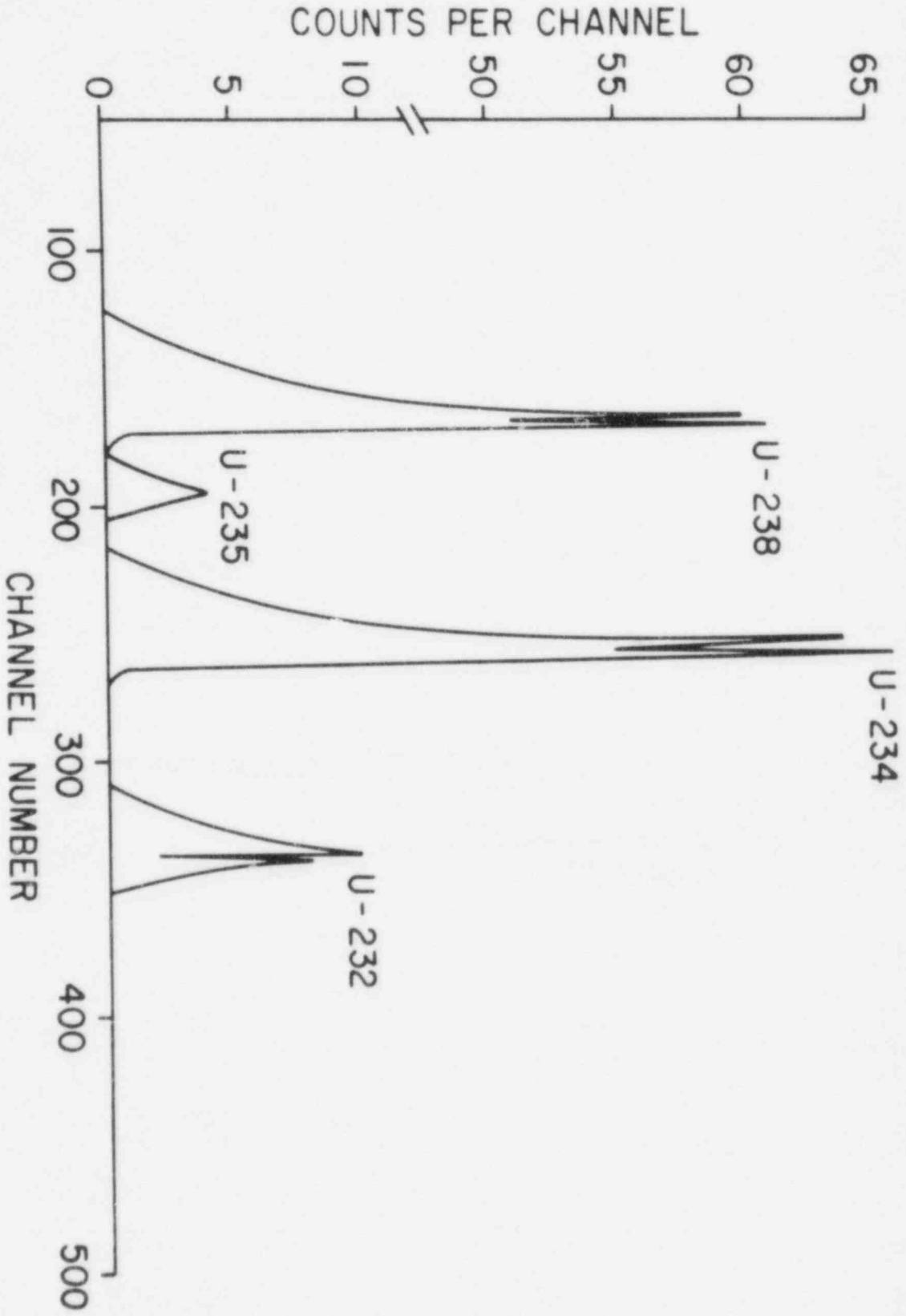
Table 3.2

Uranium and Thorium Isotope Concentrations in Uranium Miners' Lungs and Activity Ratios (pCi/kg wet weight)

Sample Number	^{238}U	^{234}U	^{230}Th	$^{238}U/^{234}U$	$^{230}Th/^{234}U$	$^{238}U/^{230}Th$
U-214	311 ± 5	325 ± 5	213 ± 4	0.96 ± 0.02	0.66 ± 0.02	1.46 ± 0.02
U-215	197 ± 5	212 ± 3	306 ± 3	0.93 ± 0.02	1.44 ± 0.02	0.64 ± 0.02
U-241	110 ± 5	120 ± 6	148 ± 4	0.92 ± 0.07	1.23 ± 0.06	0.75 ± 0.05
U-242	110 ± 2	104 ± 2	106 ± 4	1.06 ± 0.03	1.02 ± 0.04	1.04 ± 0.04
U-243	72 ± 3	92 ± 3	107 ± 3	0.77 ± 0.05	1.16 ± 0.04	0.67 ± 0.05
U-246	6.1 ± 0.4	6 ± 0.5	17 ± 1	0.95 ± 0.10	2.57 ± 0.01	0.37 ± 0.09
U-367*	54 ± 3	54 ± 3	35 ± 4	1.00 ± 0.08	0.65 ± 0.13	1.52 ± 0.1
U-337	51 ± 2	56 ± 2	———	0.91 ± 0.05	——	——
U-342	94 ± 2	102 ± 2	34 ± 1	0.92 ± 0.03	0.33 ± 0.04	2.79 ± 0.04
U-343	16 ± 0.7	17 ± 1	51 ± 5	0.98 ± 0.06	3.08 ± 0.1	0.32 ± 0.11
U-344	43 ± 2	42 ± 2	54 ± 2	1.02 ± 0.07	1.27 ± 0.06	0.80 ± 0.06
U-350**	63 ± 2	66 ± 2	39 ± 1	0.96 ° 0.04	0.60 ± 0.04	1.61 ± 0.04
U-348	33 ± 1	40 ± 1	22 ± 3	0.80 ± 0.04	0.54 ± 0.14	1.50 ± 0.1
A-78-16	———	———	54 ± 0.8	——	——	——
U-522	29 ± 0.4	32 ± 0.4	57 ± 2	0.91 ± 0.02	1.80 ± 0.02	0.51 ± 0.04
U-531	1.6 ± 0.1	2.5 ± 0.2	3.7 ± 0.4	0.64 ± 0.10	1.50 ± 0.1	0.43 ± 0.12
81-A-31	5.1 ± 0.3	5.2 ± 0.2	18 ± 0.4	0.98 ± 0.07	3.54 ± 0.04	0.28 ± 0.06
Average Range	75 (1.6 - 311)	779 (2/5 - 325)	79 (3.7 - 306)	0.92	1.4	0.98
Ratio of Averages				0.94	0.99	0.95

* Retired 1958, died 3/77.
** Retired 1970, died 5/75.

Figure 3.1: Alpha-spectrum of uranium isotopes in the lung of uranium miner U-522.



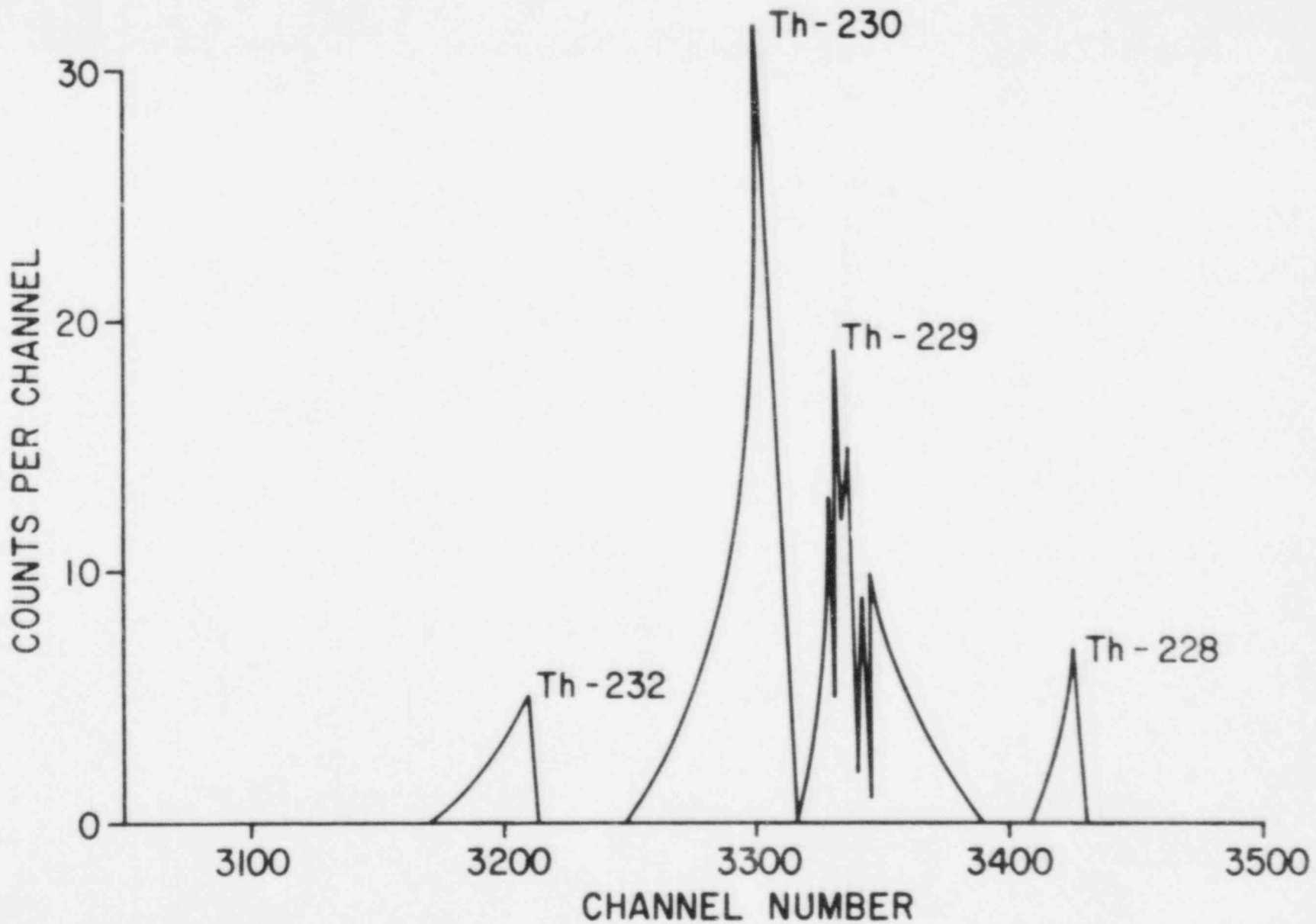


Figure 3.2: Alpha-spectrum of thorium isotopes in the lung of uranium miner U-522.

CHAPTER 4

URANIUM AND THORIUM CONCENTRATIONS IN URANIUM MINERS' TISSUES

4.1 Collection of Tissues

Tissue sets were collected on autopsy from three former uranium miners by Dr. Geno Saccomanno of St. Mary's Hospital, Grand Junction, Colorado and shipped to our laboratory for radiochemical determinations of uranium and thorium. One tissue set contained lung, kidney, spleen, heart, gonad, vertebrae, ribs and sternum; the second contained lung, kidney, liver, gonad, heart and vertebrae. The third set contained only lung, gonad and vertebrae.

4.2 Results

Concentrations of alpha-emitting particles of uranium and thorium were determined in soft tissues and bones of three former uranium miners and the results are given in Tables 4.1, 4.2, and 4.3. Typical alpha-spectra of uranium and thorium isotopes in one uranium miner's vertebrae are given in Figures 4.1 and 4.2, respectively. In one miner the concentrations of U-234 and U-238 were highest in lung (31.5 and 28.7 pCi/kg), followed by kidney (1.82 and 1.10 pCi/kg). In the other miner the concentrations of the two isotopes were highest in kidney (4.82 and 2.79 pCi/kg), followed by lung (2.47 and 1.60 pCi/kg). The concentrations of these two uranium isotopes among other soft tissues such as liver, gonad, heart, and spleen were comparable in both miners.

In uranium miner U-536, the concentration of U-234 was highest in kidney (4.82 ± 0.53 pCi/kg), followed by vertebrae (3.43 ± 0.65 pCi/kg), and lung (2.47 ± 0.17 pCi/kg). The concentrations of U-234 in liver, gonad and heart were similar, with a range of 1.52 to 1.67 pCi/kg.

The concentrations of U-238 followed the same pattern, the highest being kidney (2.79 ± 0.41 pCi/kg), followed by vertebrae (2.59 ± 0.56 pCi/kg), and lung (1.60 ± 0.13 pCi/kg). The concentrations of U-238 were 1.11 ± 0.06 pCi/kg in liver, 1.01 ± 0.09 pCi/kg in heart, and 0.73 ± 0.27 pCi/kg in gonad. The concentrations of U-238 were lower than the concentrations of U-234 in all organs.

The concentrations of U-235 were very low in all the organs, varying from 0.05 ± 0.02 pCi/kg in heart to 0.23 ± 0.12 pCi/kg in kidney.

The concentration of U-234 was highest in lung (0.65 ± 0.15 pCi/kg), followed by kidney (0.23 ± 0.13 pCi/kg) and vertebrae (0.17 ± 0.08 pCi/kg). The concentrations in liver, gonad, and heart were not detected (i.e., the counting error was equal to or greater than the values found).

The concentration of Th-230 was highest in vertebrae (10.6 ± 0.7 pCi/kg) followed by lung (3.74 ± 0.36 pCi/kg). All other tissues contained lower concentrations of Th-230 ranging from 0.76 ± 0.15 pCi/kg in liver to 1.33 ± 0.37 pCi/kg in gonad.

As expected, the concentration of Th-228 was highest in bone (vertebrae, 1.71 ± 0.32 pCi/kg). Th-228 was not detectable in the soft tissues.

For uranium miner U-522, the concentration of U-238 was highest in lung (28.7 ± 0.35 pCi/kg), followed by gonad (1.22 ± 0.50 pCi/kg), and kidney (1.10 ± 0.28 pCi/kg). Other soft tissues contained very small concentrations of U-238. Among bones, the concentration of U-238 in ribs (0.87 ± 0.12 pCi/kg) was more than twice the concentrations in vertebrae (0.36 ± 0.05 pCi/kg) and sternum (0.37 ± 0.08 pCi/kg).

Similarly, the concentration of U-234 was highest in lung (31.5 ± 0.36 pCi/kg), followed by gonad (3.22 ± 0.80 pCi/kg), and kidney (1.82 ± 0.36 pCi/kg). Among bones, the concentration of U-234 in ribs (1.60 ± 0.17 pCi/kg) was more than twice the concentration of U-234 in vertebrae (0.61 ± 0.06 pCi/kg) and sternum (0.65 ± 0.10 pCi/kg).

The concentration of U-235 in lung was 1.01 ± 0.16 pCi/kg. In all other soft tissues and bones, the concentrations of U-235 were undetectable.

The concentration of Th-230, daughter product of the U-238 series, was highest in lung (56.6 ± 2.01 pCi/kg), followed by gonad (15.8 ± 0.61 pCi/kg). The concentration of Th-230 was highest in ribs (49.2 ± 1.65 pCi/kg), followed by vertebrae (33.1 ± 1.38 pCi/kg) and sternum (29.2 ± 1.25 pCi/kg).

The concentrations of Th-232 and Th-228 from the thorium series, were much lower than the concentrations of Th-230. The concentration of Th-232 was highest in lung (4.50 ± 0.57 pCi/kg), followed by spleen (1.76 ± 1.04 pCi/kg), and gonad (0.21 ± 0.07 pCi/kg). Kidney and heart did not contain detectable amounts of Th-232. The relative concentration of Th-232 among bones was similar to concentrations of Th-230, U-238, and U-234. The concentration was highest in ribs (0.45 ± 0.17 pCi/kg), followed by sternum (0.28 ± 0.14 pCi/kg) and vertebrae (0.16 ± 0.12 pCi/kg).

The concentration of Th-228 among soft tissues was highest in spleen (2.15 ± 1.38 pCi/kg), followed by gonad (1.99 ± 0.25 pCi/kg), lung (1.79 ± 0.35 pCi/kg) and kidney (0.78 ± 0.26 pCi/kg). The heart did not contain appreciable amounts of Th-228. Among bones, the concentration was highest in sternum (2.93 ± 0.42 pCi/kg), followed by ribs (2.26 ± 0.39 pCi/kg) and vertebrae (1.50 ± 0.30 pCi/kg).

4.3 Discussion

The results obtained from the measurements of uranium and thorium in soft tissues and bones of three former uranium miners reveal that lung contained the highest concentrations of U-238 and U-234. Other soft tissues and bone contained much lower concentrations of these two isotopes. On the other hand, bone contained the highest concentration of Th-230; comparable to that in lung but much higher than its concentrations in all other soft tissues. These results strongly suggest that uranium cleared from the lung either does not deposit in bone as strongly as does thorium or is cleared from the bone with a much shorter biological halflife.

TABLE 4.1

Concentrations (pCi/kg wet weight) of Alpha-Emitting Isotopes of Uranium and Thorium in Uranium Miner's Tissues (U-522)

Tissue	Sample Size (g)	^{238}U	^{235}U	^{234}U	^{228}Th	^{230}Th	^{232}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Lung	799	28.7 ± 0.4	1.01 ± 0.2	31.5 ± 0.4	1.8 ± 0.4	56.6 ± 2.0	4.5 ± 0.6	1.10	1.80
Kidney	137	1.1 ± 0.3	0.005 ± 0.03	1.8 ± 0.4	0.78 ± 0.26	2.1 ± 0.4	−0.08 ± 0.07	1.65	1.16
Spleen	106	0.95 ± 0.52	0.02 ± 0.02	0.86 ± 0.52	2.2 ± 1.4	2.6 ± 1.3	1.8 ± 1.0	0.91	3.02
Heart	253	0.15 ± 0.05	0.01 ± 0.02	0.28 ± 0.07	−0.25 ± 0.31	1.8 ± 0.4	−0.003 ± 0.08	1.87	6.46
Gonad	45	1.2 ± 0.5	−0.37 ± 0.26	3.2 ± 0.8	2.0 ± 0.3	15.8 ± 0.6	0.21 ± 0.07	2.74	4.91
Vertebrae	313	0.36 ± 0.05	0.01 ± 0.01	0.61 ± 0.06	1.5 ± 0.3	33.1 ± 1.4	0.16 ± 0.12	1.69	54.3
Ribs	67	0.87 ± 0.12	0.07 ± 0.04	1.6 ± 0.2	2.3 ± 0.4	49.2 ± 1.7	0.45 ± 0.17	1.84	30.8
Sternum	71	0.37 ± 0.08	0.03 ± 0.03	0.65 ± 0.10	2.9 ± 0.4	29.2 ± 1.3	0.28 ± 0.14	1.76	44.9

TABLE 4.2

Concentrations (pCi/kg wet weight) of Alpha-Emitting Isotopes of Uranium and Thorium in One Set of Uranium Miner's Tissue (U-536)

Tissue	Sample Size (g)	^{234}U	^{235}U	^{238}U	^{228}Th	^{230}Th	^{232}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Lung	375	2.5 ± 0.2	0.11 ± 0.03	1.6 ± 0.12	0.12 ± 0.12	3.7 ± 0.4	0.65 ± 0.15	1.6 ± 0.2	1.5 ± 0.1
Kidney	39	4.8 ± 0.5	0.23 ± 0.12	2.8 ± 0.4	0.28 ± 0.41	1.2 ± 0.3	0.23 ± 0.13	1.7 ± 0.3	0.25 ± 0.07
Liver	205	1.5 ± 0.07	0.06 ± 0.01	1.1 ± 0.1	−0.56 ± 0.07	0.76 ± 0.15	0.07 ± 0.04	1.4 ± 0.1	0.50 ± 0.10
Gonad	16	1.7 ± 0.4	−0.01 ± 0.12	0.73 ± 0.27	1.4 ± 0.8	1.3 ± 0.4	0.05 ± 0.09	2.3 ± 1.0	0.76 ± 0.29
Heart	100	1.6 ± 0.1	0.05 ± 0.02	1.0 ± 0.09	−0.50 ± 0.09	1.1 ± 0.2	0.05 ± 0.04	1.5 ± 0.2	0.69 ± 0.13
Vert.	115	3.4 ± 0.6	0.22 ± 0.17	2.6 ± 0.6	1.7 ± 0.3	10.6 ± 0.7	0.17 ± 0.08	1.3 ± 0.4	3.1 ± 0.6

TABLE 4.3

Concentrations (pCi/kg wet weight) of Alpha-Emitting Isotopes of Uranium and Thorium in Uranium Miner's Tissues (81-A-31)

Tissue	Sample Size (g)	^{238}U	^{235}U	^{234}U	^{228}Th	^{230}Th	^{232}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Lung	403	5.1 ± 0.2	0.25 ± 0.04	5.2 ± 0.2	0.27 ± 0.04	18.4 ± 0.4	0.48 ± 0.06	1.02 ± 0.06	3.5 ± 0.2
Gonad	21	0.86 ± 0.27	0.26 ± 0.15	1.7 ± 0.3	0.88 ± 0.21	8.7 ± 2.8	0.99 ± 0.23	1.98 ± 0.8	5.1 ± 2.0
Vertebrae	122	1.6 ± 0.2	0.09 ± 0.06	1.8 ± 0.3	0.23 ± 0.22	171 ± 2.4	0.13 ± 0.07	1.14 ± 0.07	96.3 ± 15

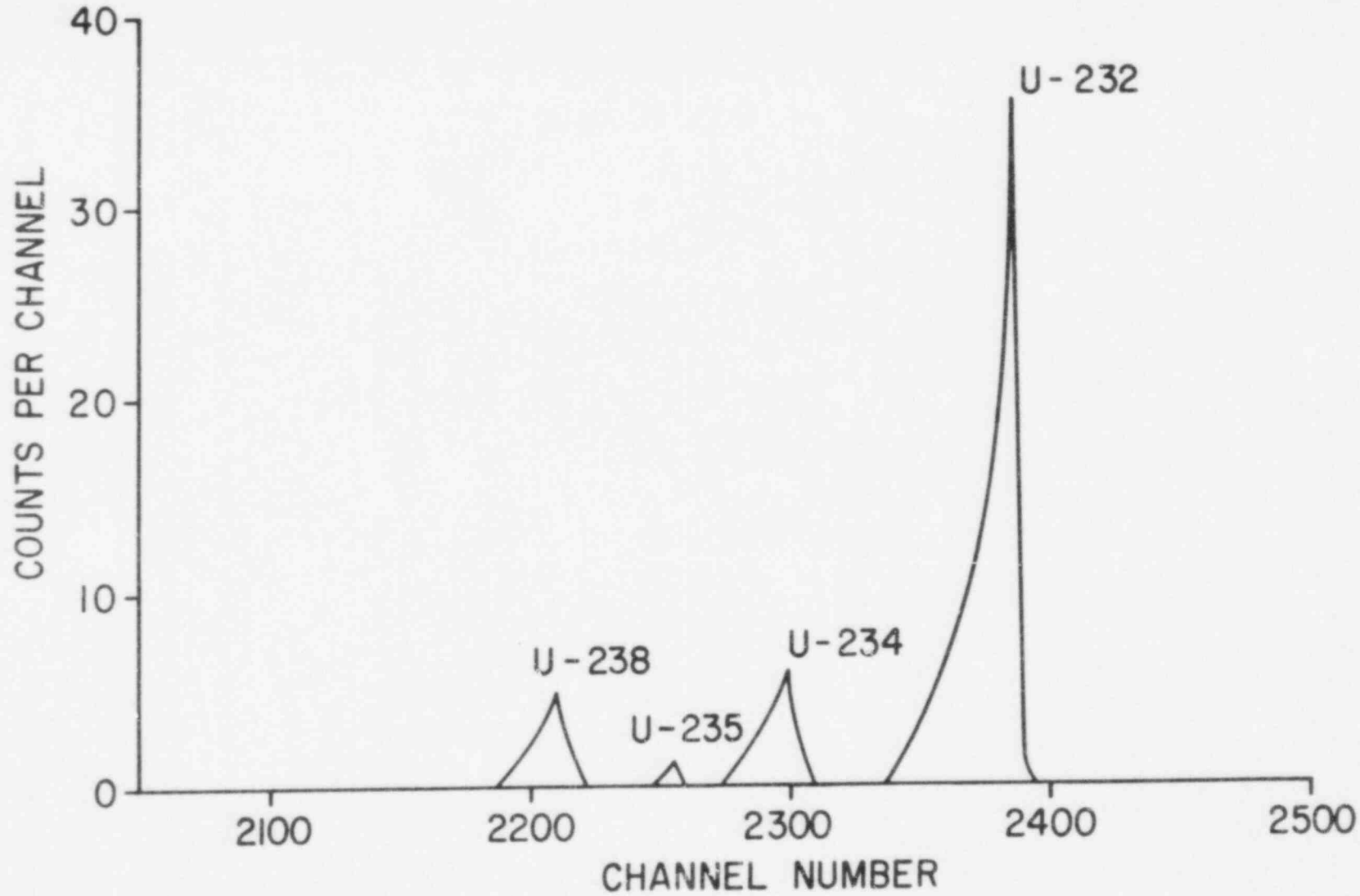
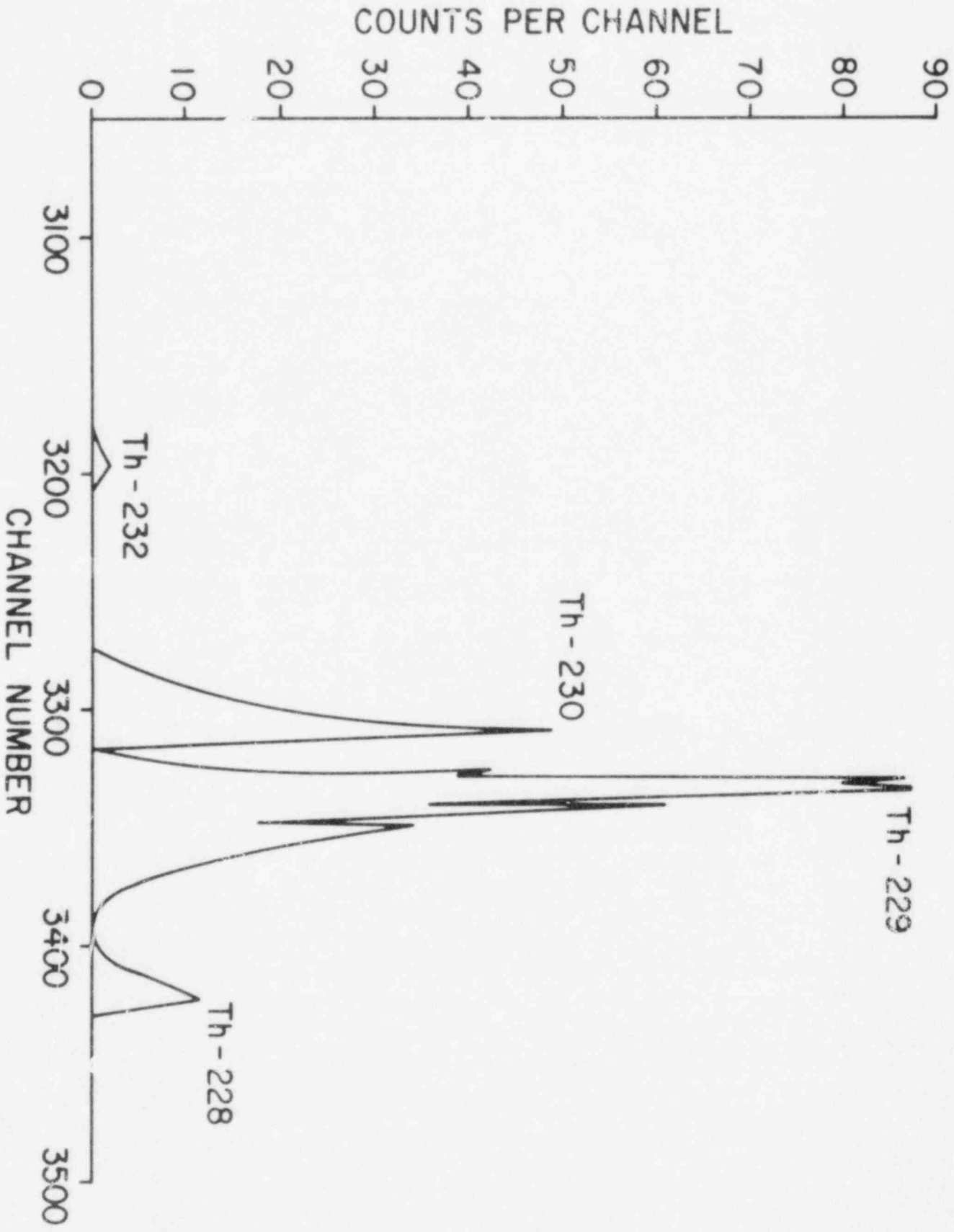


Figure 4.1: Alpha-spectrum of uranium isotopes in vertebrae of uranium miner U-522

Figure 4.2: Alpha-spectrum of thorium isotopes in vertebrae of uranium miner U-522



CHAPTER 5

URANIUM AND THORIUM CONCENTRATIONS IN TISSUES FROM URANIUM MILLERS

5.1 Collection of Tissues

Two sets of tissues were collected upon autopsy from former uranium millers by Dr. Geno Saccomanno of St. Mary's Hospital, Grand Junction, Colorado. Tissue sets included lung, lymph nodes, kidney, liver, gonad, heart and vertebrae. These tissues were shipped to our laboratory for radiochemical determinations of uranium and thorium.

5.2 Results

The results on the concentrations of uranium and thorium isotopes in soft tissues and bones of former uranium millers are given in Tables 5.1 and 5.2. Table 5.1 gives the results on the tissues of a former mechanic who had been employed in active uranium mills for 33 years. Alphaspectra of uranium and thorium isotopes in uranium millers' lung and lymph nodes are given in Figures 5.1 through 5.4.

The concentration of U-238 was highest in lymph nodes (1908 ± 79 pCi/kg), followed by lung (187.4 ± 3.3 pCi/kg). Other soft tissues, such as kidney, liver and gonad, contained almost equal concentrations varying from 1.91 ± 0.44 pCi/kg in gonad to 2.91 ± 0.42 pCi/kg in kidney; liver contained 2.62 ± 0.19 pCi/kg. The most surprising result was heart; the concentration of U-238 were 111.6 ± 3.0 pCi/kg wet weight. The concentration of U-238 in the vertebrae was 4.92 ± 0.77 pCi/kg wet weight, almost twice the concentration of U-238 in kidney, liver and gonad.

Similarly, the concentration of U-234 was highest in lymph nodes (2038 ± 81 pCi/kg wet weight) followed by lung (165.9 ± 1.4 pCi/kg). Kidney, liver and gonad contained very low concentrations of U-234 ranging from 1.96 ± 0.45 pCi/kg in gonad to 4.20 ± 0.50 pCi/kg in kidney; liver contained 3.33 ± 0.21 pCi/kg. Again, the concentration of U-234 was higher in heart (117 ± 3.1 pCi/kg). The concentration of U-234 in vertebrae was 6.53 ± 0.89 pCi/kg wet weight.

The concentration of U-235 was highest in lymph nodes (75.3 ± 15.8 pCi/kg wet weight), followed by lung (6.98 ± 0.64 pCi/kg), a pattern similar to that observed for the concentrations of U-238 and U-234. The concentrations of U-235 were 0.22 ± 0.13 pCi/kg in kidney, 0.16 ± 0.05 pCi/kg in liver, and undetectable in gonads. Heart contained 4.59 ± 0.61 pCi/kg. The concentration in vertebrae was 0.36 ± 0.21 pCi/kg.

The concentrations of Th-228 and Th-232, members of the thorium series, are very low compared to the concentration of Th-230, a member of the uranium series. The concentration of Th-230 was highest in lymph nodes (3066 ± 113 pCi/kg) followed by lung and heart with concentrations of 256 ± 4.61 pCi/kg and 168 ± 2.6 pCi/kg, respectively. The concentration in liver was 21.5 ± 0.69 pCi/kg, followed by gonad (8.38 ± 1.37 pCi/kg) and kidney (2.89 ± 0.74

pCi/kg). The concentration of Th-230 in vertebrae was 150.9 ± 3.20 pCi/kg, comparable to concentrations of Th-230 in heart.

The distribution of Th-232 was similar to Th-230 in soft tissues, but its concentration was lower. The concentration of Th-232 was highest in lymph nodes (63 ± 16 pCi/kg wet weight) followed by lung (3.63 ± 0.57 pCi/kg). Kidney, liver and gonad contained similar amounts from 0.12 ± 0.05 pCi/kg in liver to 0.31 ± 0.25 pCi/kg in kidney, and 0.22 ± 0.22 pCi/kg in gonads. The concentration of Th-232 was surprisingly higher in the heart (2.53 ± 0.32 pCi/kg). The concentration of Th-232 in vertebrae was very low (0.08 ± 0.09 pCi/kg).

The concentration of Th-228 was negligible in the soft tissues. The errors associated with the analytical results were almost as great as the results obtained. The concentration of Th-228 in the vertebrae was much higher (1.23 ± 0.06 pCi/kg) than the concentration of Th-232. Ra-228 ingested in food is well absorbed in bone and then decays to Th-228.

The ratio Th-230/Th-234 was 1.33 in lung, not too far from equilibrium. Th-230, U-238 and U-234 were dissolved and cleared from the lung at about the same rate. The ratio Th-230/U-234 in lymph nodes was 1.50. The ratio was lower than 1 (0.64) in kidney, which suggests a larger intake of U-238 and U-234 through the food chain than of Th-230. The ratio in the heart was 1.43. The ratios in gonad and liver were much higher, 4.3 and 6.5. These higher ratios suggest that the biological halflife of Th-230 in gonad and liver is much longer than that of uranium. The concentration ratio Th-230/U-234 was as high as 23 in vertebrae, possibly because the biological halflife of thorium in bone may be much longer than that of uranium, and because the transfer fraction from blood to bone may be much greater for thorium than for uranium.

We received another set of tissues from a former uranium miller including lung, lymph nodes, liver, kidney, spleen, gonad, heart and vertebrae. The concentrations of uranium and thorium were measured and the results are included in Table 5.2. The concentration of U-238 was highest in lymph nodes, followed by lung. The other soft tissues, excluding gonads contain lower concentrations of U-238, ranging from 0.16 to 0.58 pCi/kg wet weight. Gonad contained 1.21 ± 0.34 pCi/kg. The concentration of U-238 in bone (vertebrae) was 2.3 ± 0.16 pCi/kg.

The concentration of U-234 was highest in lymph nodes, followed by lung. The concentrations in liver, kidney, spleen and heart were very low, ranging from 0.20 ± 0.03 pCi/kg in liver to 0.82 ± 0.14 pCi/kg in spleen. The concentration of U-234 was slightly higher in gonad (1.56 ± 0.39 pCi/kg) comparatively, but much lower than the concentration of U-234 in lung (24.04 ± 0.39 pCi/kg) and lymph nodes (59.34 ± 4.48 pCi/kg). The concentration of U-234 in bone (vertebrae) was 7.37 ± 0.20 pCi/kg wet weight.

The concentration of U-235 was very low in all the soft tissues and bone.

The concentrations of Th-232 and Th-228 in all the soft tissues and bone were much lower than the concentrations of Th-230, because Th-232 and Th-228 are members of the thorium series, while Th-230 is a member of the uranium series in which a miller is exposed. The concentration of Th-232 was highest in lymph nodes, followed by lung and bone. Liver and kidney contained the lowest concentrations with 0.06 ± 0.02 pCi/kg and 0.03 ± 0.04 pCi/kg. Concentrations of Th-232 in gonad and spleen were higher than the concentrations in liver and kidney, but much lower than the concentration of Th-232 in lung, lymph nodes and bone.

The concentration of Th-228 was highest in lymph nodes, followed by spleen and gonad. The analytical errors associated with the results on the concentration of Th-228 in spleen and gonad were very large. Liver, kidney and heart contained the lowest concentration ranging from 0.08 to 0.21 pCi/kg. The concentration of Th-228 was slightly higher in bone (1.14 ± 0.15 pCi/kg) than in lung (0.97 ± 0.12 pCi/kg).

The concentration of Th-230 was highest in lymph nodes, followed by lung and bone. Liver, spleen, gonad, heart and kidney contained very low concentrations, ranging from 1.56 ± 0.13 pCi/kg in heart to 2.71 ± 0.22 pCi/kg in kidney.

The concentration ratio U-234/U-238 for all the soft tissues and bone is given in Table 5.2. The ratio ranged from 1.05 ± 0.03 in lung to 1.82 ± 0.48 in spleen, showing near equilibrium between the two isotopes of uranium. The concentration ratio of Th-230/U-234 for all soft tissues and bones is also given in the same table. The ratio was 1.07 ± 0.03 in the lung, suggesting that uranium and thorium in equilibrium in the lung translocate from the lung at the same rate. The concentration ratios of Th-230/U-234 in all other organs was not at unity, ranging from 0.54 ± 0.05 in kidney to 11.90 ± 1.92 in liver. The ratio in bone was 6.77 ± 0.44, suggesting a greater percentage of thorium being transferred to bone and having a longer biological half-life.

5.3 Discussion

The concentrations of uranium isotopes were higher in lymph nodes, followed by lung. Other soft tissues and bone contained much lower concentrations of these uranium isotopes. The concentration of Th-230 in bone was similar to its concentration in lung. The concentrations of uranium isotopes and Th-230 in lymph nodes was highest, suggesting that uranium bearing particles are quite soluble and are translocated to tracheobronchial lymph nodes. In addition, the concentration ratio of Th-230/U-234 in lung of both millers was close to 1, suggesting thereby that the elimination rates of uranium and thorium are similar in former uranium millers' lungs to those observed in uranium miners' lungs.

TABLE 5.1

Concentrations of Uranium and Thorium Isotopes in Tissues of Uranium Miller 83-A-79 (pCi/kg)

Tissue	Sample Size (g)	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Lung	518.9	187 ± 3	7.0 ± 0.6	192 ± 3	3.6 ± 0.6	256 ± 4.6	- 2.5 ± 1.3	1.03 ± 1.28	1.33 ± 0.02
Lymph nodes	1.30	1908 ± 79	75 ± 16	2038 ± 81	62 ± 16	3066 ± 113	334 ± 23	1.07 ± 0.06	1.52 ± 0.08
Kidney	164.6	2.9 ± 0.4	0.22 ± 0.13	4.2 ± 0.5	0.31 ± 0.25	2.9 ± 0.7	0.59 ± 0.54	1.45 ± 0.3	6.9 ± 0.84
Liver	404.4	2.6 ± 0.2	0.16 ± 0.05	3.3 ± 0.2	0.12 ± 0.05	21.5 ± 0.7	0.21 ± 0.11	1.27 ± 0.12	6.52 ± 0.45
Gonad	71.3	1.9 ± 0.4	- 0.05 ± 0.04	2.0 ± 0.5	0.22 ± 0.22	8.4 ± 1.4	0.75 ± 0.83	1.05 ± 0.34	4.2 ± 1.26
Heart	472.1	112 ± 3	4.6 ± 0.6	117 ± 3	2.5 ± 0.3	168 ± 3	1.1 ± 0.3	1.04 ± 0.04	1.44 ± 0.05
Vertebrae	130.3	4.9 ± 0.8	0.36 ± 0.21	6.5 ± 0.9	0.08 ± 0.09	151 ± 3	1.23 ± 0.06	1.33 ± 0.2	23.2 ± 3.25

TABLE 5.2

Concentrations of Uranium and Thorium Isotopes in Tissues of Uranium Miller 83-A-125 (pCi/kg)

Tissue	Sample Size (g)	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Lung	460.0	23.0 ± 0.4	1.1 ± 0.1	24.0 ± 0.5	1.1 ± 0.1	25.7 ± 0.6	0.97 ± 0.12	1.04 ± 0.03	1.07 ± 0.03
Lymph Nodes	15.2	431 ± 8.8	0.08 ± 0.56	59 ± 4.5	1.5 ± 1.0	32 ± 3	9.7 ± 2.1	1.4 ± 0.17	0.54 ± 0.07
Liver	608.0	0.16 ± 0.03	0.02 ± 0.01	0.20 ± 0.03	0.06 ± 0.02	2.4 ± 0.1	0.08 ± 0.04	1.25 ± 0.3	12.0 ±
Kidney	185.0	0.58 ± 0.11	-0.006 ± 0.026	0.61 ± 0.11	0.03 ± 0.04	2.7 ± 0.2	0.18 ± 0.08	1.05 ± 0.28	0.3 ± 0.14
Spleen	78.0	0.45 ± 0.09	0.05 ± 0.06	0.82 ± 0.14	0.39 ± 0.15	1.8 ± 0.3	1.9 ± 0.3	1.82 ± 0.48	2.2 ± 0.53
Gonad	55.0	1.2 ± 0.3	0.27 ± 0.17	1.6 ± 0.4	0.25 ± 0.14	1.8 ± 0.4	1.6 ± 0.4	1.33 ± 047	1.12 ± 0.38
Heart	315.0	0.21 ± 0.03	0.02 ± 0.02	0.33 ± 0.04	0.03 ± 0.02	1.6 ± 1	0.21 ± 0.06	1.57 ± 0.29	4.85 ± 3.09
Vertebrae	207.0	2.0 ± 0.2	0.17 ± 0.06	3.4 ± 0.2	0.53 ± 0.10	22.8 ± 0.6	1.1 ± 0.2	1.7 ± 0.2	6.7 ± 0.43

Figure 5.1: Alpha-spectrum of uranium isotopes in lung of uranium miller 83-A-77.

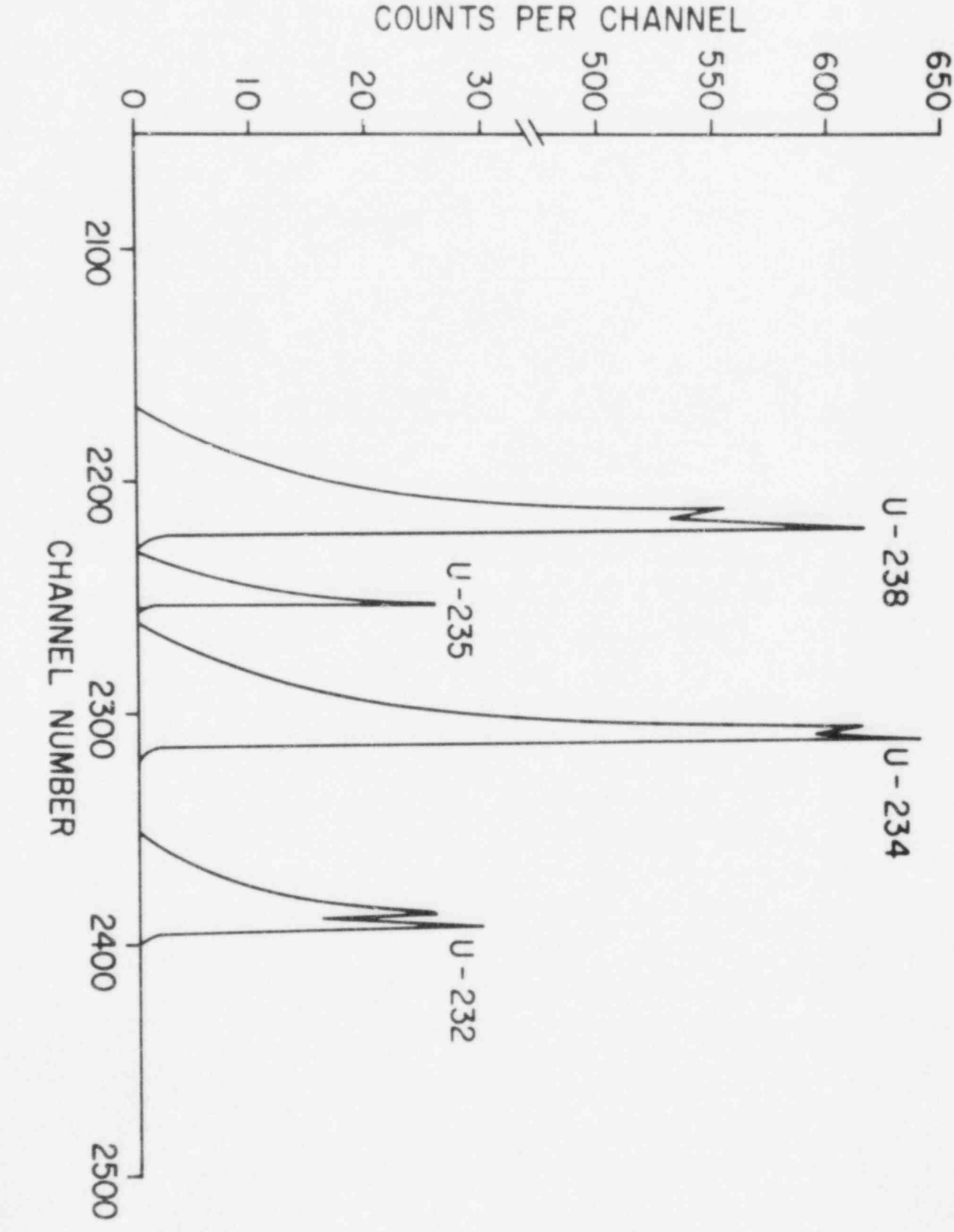
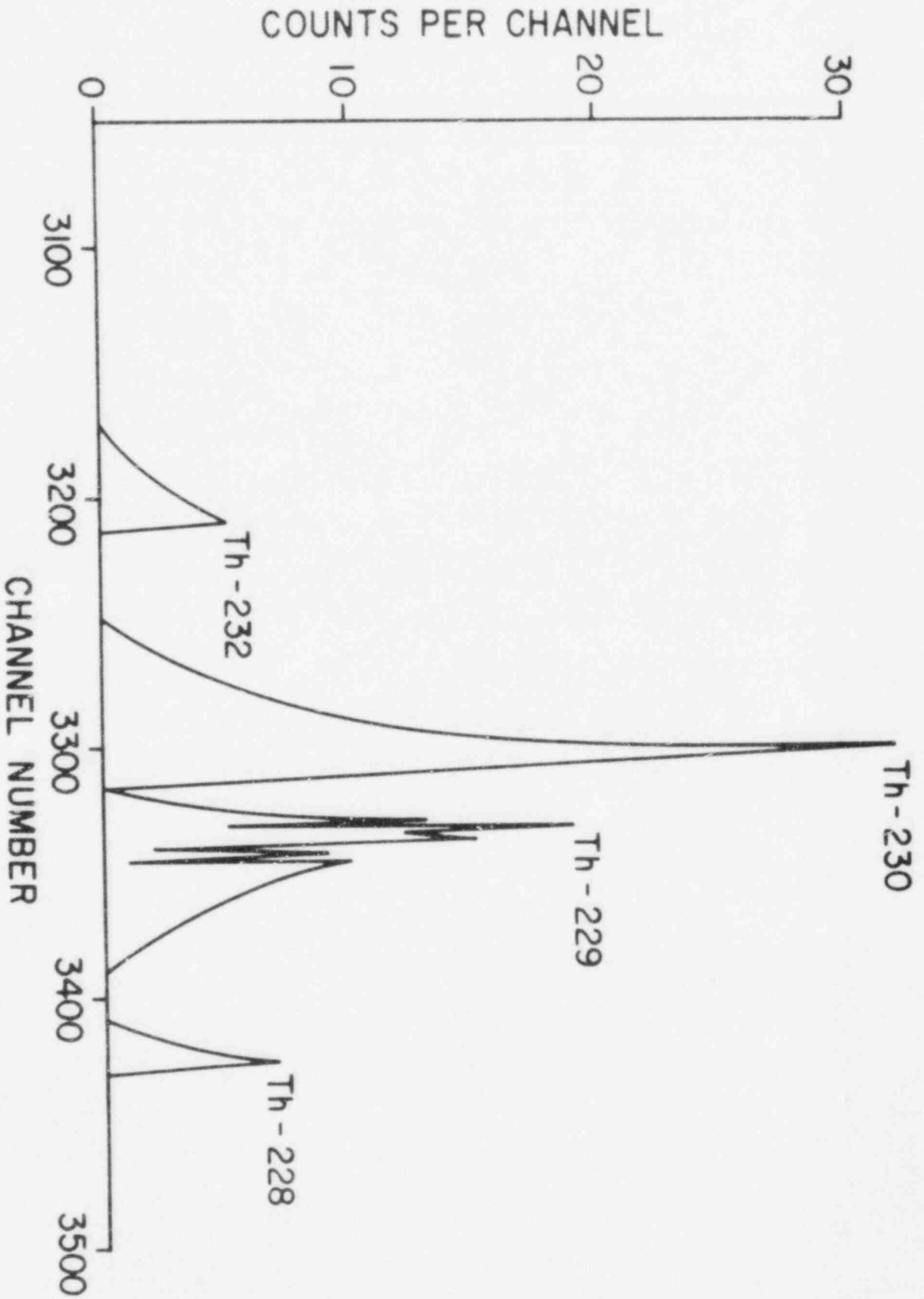


Figure 5.2: Concentration of uranium and thorium isotopes in uranium miller 83-A-125 (pCi/kg)



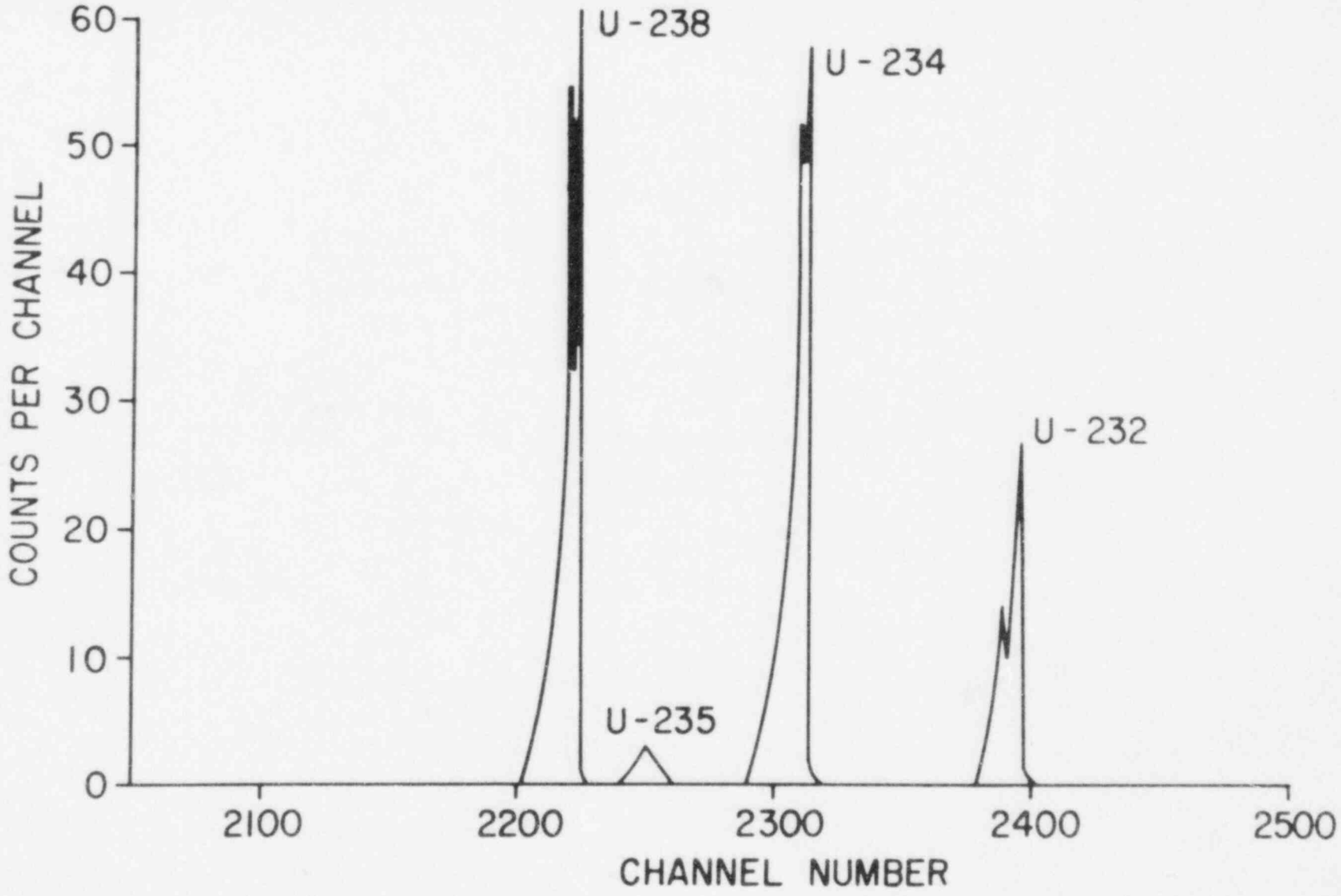
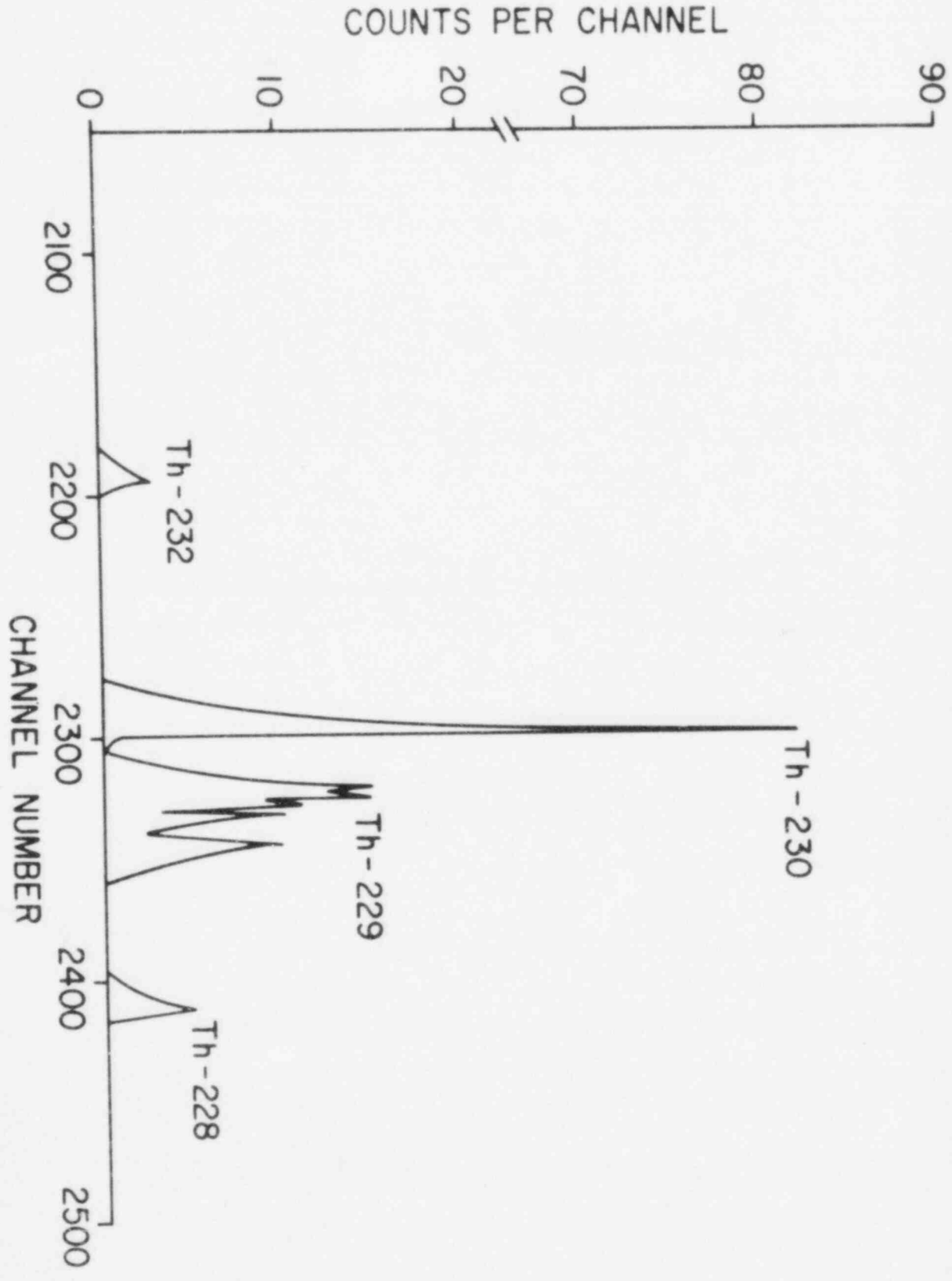


Figure 5.3: Alpha-spectrum of uranium isotopes in lymph nodes of uranium miller 83-A-71

Figure 5.4: Alpha-spectrum of thorium isotopes in lymph nodes of uranium miller 83-A-71.



CHAPTER 6

CONCENTRATIONS OF URANIUM AND THORIUM IN DOG LUNGS EXPOSED TO URANIUM ORE DUST: AN INTERLABORATORY COMPARISON

6.1 Experimental

The lung tissues from eight dogs exposed to uranium ore dust at PNL were divided into three sections and divided among three laboratories; the Radiobiology Division, University of Utah (Dr. N.P. Singh), Los Alamos Scientific Laboratory (Dr. James McInroy), and Battelle Pacific Northwest Laboratory (Dr. Peter Jackson). Dr. McInroy from Los Alamos National Laboratory was included in this study because of his involvement in radiochemical determinations of uranium in human tissues of persons occupationally exposed to uranium, as well as his experience with radiochemical determinatins of actinides in human tissues of the general population. The detailed radiochemical procedures utilized for the determinations of uranium and thorium in these dog lungs were discussed in Chapter 2.

6.2 Results

The concentrations of uranium and thorium obtained by these three laboratories are given in Tables 6.1 through 6.6.

The results obtained on the concentration of U-238 in these dogs are given in Table 6.1; the mean of these results and the standard deviations for each dog are given. The results obtained by the three laboratories are in agreement. Because the lungs were not homogenized to split samples, the concentrations are not known to be identical. In all samples except No. 1, the results obtained by the University of Utah Radiobiology Division were consistently higher than the other two participating laboratories; 5 of 8 samples analyzed by PNL showed the lowest concentrations of the three labs. Three of 8 Los Alamos samples showed unusually low concentrations. The largest percent deviation between the three results was almost 43% in sample No. 2; the smallest percent deviation was 12% in sample No. 1.

The results for U-234 were similar to those for U-238, as expected (Table 6.2).

The concentrations of Th-230 are given in Table 6.3. The results obtained by the University of Utah were highest in all but two samples. In sample No. 1, the University of Utah lab obtained the lowest concentration. In sample No. 5, the University of Utah lab obtained results higher than those of Los Alamos and lower than those obtained by PNL.

The differences between the three laboratories may be due to non-uniform distribution of radionuclides in the lung tissues. However, the concentration ratios U-234/U-238 and Th-230/U-234 should be similar among the three labs. The concentration ratios U-234/U-238 are given in Table 6.4. The mean concentration ratio U-234/U-238 and ranges for all eight dogs was 1.05 (1.02 -

1.12) for the University of Utah, 1.07 (1.00 - 1.12) for LASL and 1.02 (0.96 - 1.04) for PNL. These ratios that slightly exceed unity are not significantly different.

The concentration ratios Th-230/U-234 are given in Table 6.5. The ratios show disequilibrium between Th-230 and U-234; the results obtained by the three laboratories are consistent.

These results are expressed in a different form in Table 6.6. In order to compare the results of the three laboratories, the concentration ratio Th-230/U-234 was normalized to 1.0 for the University of Utah results.

The average of the ratios was 3% higher for LANL and 6% higher for BPNL than Utah. The difference is very small, considering that the 3 laboratories had not done any prior sample exchanges.

6.2 Discussion

The intercomparison conclusively demonstrates that the disequilibrium between Th-230 and U-234 in dog lungs reported by Battelle investigators is real and reproducible by radiochemical analysis of these tissues at 2 independent laboratories. This is in contrast to the results obtained in uranium miners' and millers' lungs which show the average Th-230/U-234 and Th-232/U-238 ratios to be near equilibrium, suggesting that the mobilization of uranium and thorium from the lung occurs at the same rate. The explanation of the difference between human and experimental animals remains to be resolved, but it clearly is not caused by a difference in radiochemical methods.

TABLE 6.1

^{238}U in Dog Lungs (pCi/kg wet weight)

Sample No.	University of Utah	Los Alamos Scientific Lab	Battelle Pacific Northwest Lab	$\bar{X} \pm 1$
1	3.93 ± 0.05	4.9 ± 212	4.26 ± 0.05	4.4 ± 0.5
2	12.6 ± 0.2	8.3 ± 333	5.22 ± 0.09	8.7 ± 3.7
3	9.8 ± 0.2	7.4 ± 293	8.3 ± 0.1	8.5 ± 1.2
4	10.0 ± 0.1	6.7 ± 276	8.0 ± 0.2	8.2 ± 1.7
5	11.1 ± 0.2	9.0 ± 342	8.3 ± 0.2	9.5 ± 1.4
6	13.1 ± 0.08	9.4 ± 392	7.5 ± 0.1	10.0 ± 2.8
7	6.15 ± 0.06	4.3 ± 184	4.5 ± 0.1	5.0 ± 1.0
8*	11.2 ± 0.07	6.8 ± 325	6.3 ± 0.1	8.1 ± 2.7

*Listed as Sample #9 by Los Alamos Scientific Laboratory

TABLE 6.2

^{234}U in Dog Lungs (pCi/kg wet weight)

Sample No.	University of Utah	Los Alamos Scientific Lab	Battelle Pacific Northwest Lab	$\bar{X} \pm 1$
1	4.42 ± 0.05	5.2 ± 0.2	4.4 ± 0.05	4.7 ± 0.4
2	12.9 ± 0.2	9.0 ± 0.4	5.4 ± 0.09	9.1 ± 3.8
3	10.1 ± 0.2	8.1 ± 0.3	8.5 ± 0.1	8.9 ± 1.1
4	10.7 ± 0.1	7.3 ± 0.3	8.3 ± 0.2	8.8 ± 1.8
5	11.9 ± 0.2	10.1 ± 0.4	8.6 ± 0.2	10.2 ± 1.6
6	13.51 ± 0.08	10.2 ± 0.4	7.7 ± 0.1	10.5 ± 2.9
7	6.31 ± 0.06	4.3 ± 0.2	4.4 ± 0.1	5.0 ± 1.2
8*	11.52 ± 0.07	7.2 ± 0.3	6.4 ± 0.1	8.4 ± 2.7

*Listed as Sample #9 by Los Alamos Scientific Laboratory.

TABLE 6.3

^{230}Th in Dog Lungs (pCi/kg wet weight)

Sample No.	University of Utah	Los Alamos Scientific Lab	Battelle Pacific Northwest Lab	$\bar{X} \pm 1\sigma$
1	25.3 ± 0.1	28.2 ± 0.8	25.4 ± 0.6	26.3 ± 2
2	106 ± 1	87 ± 2	24.8 ± 0.7	73 ± 43
3	82.4 ± 0.7	72 ± 2	73 ± 1	76 ± 6
4	123.4 ± 0.5	53 ± 2	106 ± 2	94 ± 36
5	172 ± 1	132 ± 4	179 ± 4	161 ± 25
6	144.7 ± 0.7	116 ± 4	113 ± 3	124 ± 18
7	45.5 ± 0.3	40 ± 1	31.6 ± 0.8	39 ± 7
8*	76.5 ± 0.5	51 ± 2	39.2 ± 0.8	56 ± 19

*Listed as Sample #9 by Los Alamos Scientific Laboratory.

TABLE 6.4

$^{234}U/^{238}U$ Ratio in Dog Lungs

Sample No.	University of Utah	Los Alamos Scientific Lab	Battelle Pacific Northwest Lab
1	1.12 ± 0.02	1.04 ± 0.06	1.03 ± 0.02
2	1.03 ± 0.02	1.09 ± 0.06	1.03 ± 0.02
3	1.03 ± 0.03	1.09 ± 0.06	1.03 ± 0.02
4	1.07 ± 0.01	1.08 ± 0.06	1.04 ± 0.04
5	1.07 ± 0.03	1.12 ± 0.06	1.04 ± 0.03
6	1.03 ± 0.01	1.09 ± 0.06	1.03 ± 0.02
7	1.03 ± 0.01	1.01 ± 0.07	0.95 ± 0.03
8*	1.03 ± 0.01	1.07 ± 0.06	1.03 ± 0.02
	$\bar{X} = 1.05$ $\sigma = 0.03$	$\bar{X} = 1.07$ $\sigma = 0.03$	$\bar{X} = 1.02$ $\sigma = 0.03$

*Listed as Sample #9 by Los Alamos Scientific Laboratory

TABLE 6.5

$^{230}Th/^{234}U$ Ratio in Dog Lungs

Sample No.	University of Utah	Los Alamos Scientific Lab	Battelle Pacific Northwest Lab	$\bar{X} \pm 1\sigma$
1	5.72 ± 0.07	5.46 ± 0.3	5.80 ± 0.2	5.7 ± 0.2
2	8.2 ± 0.1	9.66 ± 0.5	4.59 ± 0.2	7.5 ± 2.6
3	8.2 ± 0.2	8.94 ± 0.4	8.56 ± 0.2	8.6 ± 0.4
4	11.5 ± 0.1	7.33 ± 0.4	12.88 ± 0.3	10.6 ± 2.9
5	14.5 ± 0.3	13.12 ± 0.7	20.79 ± 0.7	16.1 ± 4.1
6	10.71 ± 0.08	11.42 ± 0.6	14.62 ± 0.4	12.3 ± 2.1
7	7.21 ± 0.08	9.29 ± 0.5	7.40 ± 0.3	8.0 ± 1.2
8*	6.64 ± 0.06	7.14 ± 0.4	6.11 ± 0.2	6.6 ± 0.5

*Listed as Sample #9 by Los Alamos Scientific Laboratory.

TABLE 6.6

Comparisons of $^{230}Th/^{234}U$ Ratios with U. of U. Values

Sample No.	University of Utah	Los Alamos Scientific Lab	Battelle Pacific Northwest Lab
1	1.00	0.94	1.0
2	1.00	1.18	0.56
3	1.00	1.10	1.0
4	1.00	0.63	1.1
5	1.00	0.90	1.4
6	1.00	1.1	1.4
7	1.00	1.3	1.0
8*	1.00	1.1	0.92
		$\bar{X} = 1.03$	$\bar{X} = 1.06$
		$\sigma = 0.20$	$\sigma = 0.27$

*Listed as Sample #9 by Los Alamos Scientific Laboratory

CHAPTER 7

CONCENTRATIONS OF THORIUM AND URANIUM IN HUMAN TISSUES OF GENERAL POPULATIONS

7.1 Collection of Tissues

Soft tissues and bones from five individuals of the general population from the State of Colorado were collected by pathologists upon autopsy. The tissue samples were provided by Dr. James F. McInroy of Los Alamos National Laboratory who received these tissues from the pathologist.

7.2 Results

The concentrations of uranium and thorium in the soft tissues and bones were determined by solvent extraction and alpha-spectrometry as reported in Chapter 2. Results are shown in Tables 7.1 through 7.5. The number of sets analyzed is too small to represent the distribution of concentrations expected from both elements in the Colorado population. However, our earlier work to determine separately the concentrations of uranium and thorium isotopes in other populations may serve the purpose of establishing the background concentrations of uranium and thorium. However, if one wants to find the concentration ratios U-234/Th-230 in different organs of the general population the concentrations of uranium and thorium should be measured in the same sets of human tissues, since the concentrations of these two radionuclides in the tissues of the general population may vary from place to place, and from person to person, depending on age, concentrations of uranium and thorium in the air, and other environmental factors.

The mean concentrations of uranium and thorium isotopes in different organs of five subjects from Colorado are reported in Table 7.6. Among soft tissues, the concentration of U-238 was highest in lymph nodes with a mean concentration of 1.5 ± 0.97 pCi/kg, followed by lung with 1.3 ± 0.36 pCi/kg, and kidney with 0.90 ± 0.63 pCi/kg. Liver, spleen, thyroid and gonad contained low concentrations of U-238 ranging from 0.001 pCi/kg in spleen to 0.20 ± 0.14 pCi/kg in liver.

The concentration of U-234 was again highest in lymph nodes (4.3 ± 3.4 pCi/kg), followed by lung (1.6 ± 0.52 pCi/kg) and kidney (1.1 ± 0.72 pCi/kg). Liver, spleen, thyroid, and gonad contained low concentrations of U-234, ranging from 0.03 pCi/kg in gonad to 0.31 ± 0.16 pCi/kg in liver.

The concentration of U-235 was very low in all organs.

Among three different bones, the mean concentrations of U-238 and U-234 were highest in sternum (2.22 ± 0.88 pCi/kg and 3.2 ± 1.9 pCi/kg, respectively). The concentrations of these two isotopes were similar in ribs with 1.1 ± 0.85 pCi/kg of U-238 and 1.8 ± 1.4 pCi/kg of U-234 and in vertebrae with 1.3 ± 0.85 pCi/kg of U-238 and 1.6 ± 1.0 pCi/kg of U-234. The concentration of U-235 was very low in all three bones.

The mean concentrations of thorium isotopes (Th-228, Th-230 and Th-232) in these tissue sets are also given in Table 7.6. Th-232 and Th-228 belong to the thorium series and Th-230 belongs to the uranium series. Therefore, their geochemical nature and abundance may differ, which may cause the concentration and distribution pattern of Th-232 and Th-228 to differ from that of Th-230 in human tissues. In Th-228, Th-228 from Ra-228 decay may contribute to the overall nuclide distribution.

The mean concentration of Th-232 was highest in lymph nodes among soft tissues (2.0 ± 0.20 pCi/kg), followed by lung (0.78 ± 0.15 pCi/kg) and gonad (0.82 pCi/kg). The concentrations in other soft tissues were low, ranging from 0.035 ± 0.010 pCi/kg in liver to 0.19 ± 0.16 pCi/kg in thyroid.

The concentration of Th-230 was also highest in lymph nodes (0.78 ± 0.15 pCi/kg), followed by lung (0.78 ± 0.15 pCi/kg), gonad (1.0 pCi/kg) and thyroid (0.81 ± 0.51 pCi/kg). The concentrations in liver and kidney were 0.13 ± 0.058 pCi/kg and 0.1 ± 0.050 pCi/kg respectively.

The mean concentration of Th-228 was highest in lung (0.67 ± 0.18 pCi/kg). All other soft tissues contained very low concentrations of Th-228.

Among bones, the concentrations of Th-228 was higher than those of both Th-230 and Th-232, probably because Ra-228 accumulates in bone and decays to Th-228. The mean concentration of Th-228 was highest in sternum (3.0 ± 1.3 pCi/kg) and almost equal in vertebrae and ribs with mean concentrations of 1.1 ± 0.93 pCi/kg and 1.3 ± 0.75 pCi/kg, respectively.

The mean concentrations of Th-230 in vertebrae (0.58 ± 0.16 pCi/kg) and ribs (0.71 ± 0.19 pCi/kg) were higher than the mean concentration of Th-232 in vertebrae (0.20 ± 0.13 pCi/kg) and ribs (0.27 ± 0.092 pCi/kg). The mean concentrations of Th-230 and Th-232 were almost equal in sternum (0.32 ± 0.10 pCi/kg and 0.35 ± 0.21 pCi/kg, respectively).

7.3 Discussion

The concentration ratios U-234/U-238 and Th-230/U-234 for each organ of the five subjects are given in Table 7.7, which also includes the mean of the ratios. The concentration ratios of U-234/U-238 in all the organs range from 1.2 to 1.6, except in liver (2.4 ± 0.48) and lymph nodes (2.5 ± 0.66). These two higher ratios are not real, but caused by large analytical errors associated with the determination of these two radioisotopes in liver and lymph nodes.

The concentration ratio Th-230/U-234 in lung ranges from 0.14 to 0.98 with a mean of 0.58 ± 0.22, which suggests that the intake of uranium is greater than that of Th-230 or that the clearance of uranium from the lung is slower than that of Th-230 It seems that subjects 80-A-22 and 32-110 inhaled more uranium than Th-230. In the other two subjects, the ratio was close to unity, suggesting that the clearance rates from lung for these two radionuclides are similar. In all other organs the ratio was close to unity, except in kidney where the concentration ratio Th-230/U-234 was 0.26 ± 0.18. This

ratio suggests that more uranium than Th-230 is retained in kidney. It is well-known that uranium is mostly cleared through the kidney. It is hard to explain the meaning of the concentration ratio Th-230/U-234 in all other organs, because the intakes of uranium and Th-230 are very different through food chains.

Table 7.1

Concentrations (pCi/kg wet weight) of Alpha-Emitting Isotopes of Uranium and Thorium in Tissues from General Population Residents of Colorado (Subject 80-A-22)

Tissue	Sample Size (g)	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Liver	528	0.10 ± 0.02	0.002± 0.01	0.20 ± 0.03	0.01 ± 0.02	0.08 ± 0.02	-0.06 ± ?	2.0 ± 0.1	0.40 ± 0.12
Lung	502	2.3 ± 0.1	0.11 ± 0.02	3.2 ± 0.1	0.84 ± 0.09	0.85 ± 0.09	0.77 ± 0.09	1.38 ± 0.07	0.27 ± 0.03
Kidney	147	0.27 ± 0.05	0.02 ± 0.01	0.46 ± 0.06	0.03 ± 0.03	0.06 ± 0.04	-0.08 ± ?	1.7 ± 0.4	0.13 ± 0.09
Vertebrae	378	0.48 ± 0.13	0.05 ± 0.05	0.79 ± 0.18	0.69 ± 0.07	0.76 ± 0.13	0.38 ± 0.12	1.6 ± 0.6	0.96 ± 0.27

Table 7.2

Concentrations (pCi/kg wet weight) of Alpha-Emitting Isotopes of Uranium and Thorium in Tissues from General Population Residents of Colorado (Subject 80-A-39)

Tissue	Sample Size (g)	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Liver	501	0.08 ± 0.02	0.04 ± 0.01	0.23 ± 0.03	0.04 ± 0.02	0.11 ± 0.03	-0.04 ± ?	2.9 ± 0.8	0.48 ± 0.15
Lung	504	0.81 ± 0.06	0.04 ± 0.02	1.04 ± 0.07	1.54 ± 0.18	0.95 ± 0.14	1.03 ± 0.2	1.3 ± 0.1	0.91 ± 0.15
Kidney	168	0.58 ± 0.07	0.03 ± 0.02	0.80 ± 0.09	0.03 ± 0.02	0.02 ± 0.03	-0.13 ± ?	1.4 ± 0.2	0.03 ± 0.038
Lymph nodes	2.52	2.45 ± 1.50	0.09 ± 0.90	7.72 ± 2.50	2.26 ± 1.37	4.06 ± 1.97	-3.38 ± ?	3.2 ± 2.1	0.53 ± 0.31
Thyroid	12.6	0.30 ± 0.21	-0.33 ± 0.18	0.48 ± 0.31	0.34 ± 0.27	0.38 ± 0.29	-1.38 ± ?	1.6 ± 1.5	0.79 ± 0.79
Vertebrae	220	0.56 ± 0.11	-0.01 ± 0.03	0.63 ± 0.13	0.11 ± 0.05	0.36 ± 0.11	-0.09 ± 0.1	1.1 ± 0.3	0.57 ± 0.21

TABLE 7.3

Concentrations (pCi/kg wet weight) of Alpha-Emitting Isotopes of Uranium and Thorium in Tissues of General Population Residents of Colorado (Subject 27-88)

Tissue	Sample Size (g)	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Liver	1075.2	0.02 ± 0.02	−0.004 ± 0.009	0.08 ± 0.03	0.06 ± 0.04	0.29 ± 0.08	0.002± 0.05	4.0 ± 4.3	3.6
Lung	526	0.34 ± 0.05	0.04 ± 0.01	0.28 ± 0.05	—	—	—	0.82 ± 0.19	—
Kidney	178.9	0.13 ± 0.04	0.01 ± 0.01	0.10 ± 0.04	0.06 ± 0.03	0.08 ± 0.04	0.37 ± 0.07	0.77 ± 0.39	0.8
Spleen	222.3	0.001± 0.02	−0.0003± 0.01	0.03 ± 0.02	—	—	—	—	—
Thyroid	16.4	0.66 ± 0.88	0.03 ± 0.41	0.81 ± 1.03	0.03 ± 0.23	1.4 ± 0.5	−16 ± 1	1.2 ± 2.3	1.7
Gonad	29.1	0.03 ± 0.17	0.06 ± 0.11	0.33 ± 0.24	0.82 ± 0.73	1.0 ± 1.1	5.8 ± 1.8	—	—
Lymph nodes	19.4	0.51 ± 0.38	0.20 ± 0.19	0.94 ± 0.48	1.9 ± 0.4	2.3 ± 0.5	− 0.36 ± 0.96	1.8 ± 1.7	2.5
Vertebrae	178.4	0.30 ± 0.06	0.03 ± 0.02	0.34 ± 0.07	0.14 ± 0.04	0.29 ± 0.06	4.8 ± 0.2	1.1 ± 0.3	0.85
Ribs	127.1	0.26 ± 0.09	0.04 ± 0.04	0.26 ± 0.10	0.28 ± 0.07	0.58 ± 0.10	2.8 ± 0.2	1.0 ± 0.5	2.25
Sternum	18.9	1.4 ± 0.5	−0.30 ± 0.22	1.6 ± 0.6	0.77 ± 0.41	0.28 ± 0.50	4.3 ± 0.9	1.2 ± 0.6	—

TABLE 7.4

Concentrations (pCi/kg wet weight) of Alpha-Emitting Isotopes of Uranium and Thorium in Tissues of General Population Residents of Colorado (Subject 27-88)

Tissue	Sample Size (g)	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Liver	278.7	0.05 ± 0.02	0.02 ± 0.01	0.09 ± 0.02	0.03 ± 0.02	0.02 ± 0.02	−0.17 ± 0.03	1.8 ± 0.8	0.22 ± 0.23
Lung	292.4	1.04 ± 0.07	0.06 ± 0.02	1.01 ± 0.07	1.2 ± 0.1	0.99 ± 0.12	0.72 ± 0.10	0.97 ± 0.09	0.98 ± 0.34
Kidney	112.7	0.12 ± 0.05	−0.03 ± 0.02	0.11 ± 0.06	—	—	—	0.92 ± 0.09	—
Vertebrae	274.1	0.26 ± 0.05	0.03 ± 0.02	0.38 ± 0.06	0.16 ± 0.04	0.35 ± 0.06	0.73 ± 0.09	1.5 ± 0.2	0.92 ± 0.21
Ribs	141.5	0.25 ± 0.09	0.04 ± 0.02	0.53 ± 0.12	0.11 ± 0.06	0.48 ± 0.11	0.71 ± 0.14	2.1 ± 0.9	0.91 ± 0.29
Sternum	28.7	1.3 ± 0.43	−0.004 ± 0.11	1.5 ± 0.4	0.16 ± 0.37	0.17 ± 0.37	4.2 ± 0.9	1.2 ± 0.5	2.9 ± 1.1

TABLE 7.5

Concentrations (pCi/kg wet weight) of Alpha-Emitting Isotopes of Uranium and Thorium in Tissues of the General Population (Subject 32-110)

Tissue	Size (g)	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th	$^{234}U/^{238}U$	$^{230}Th/^{234}U$
Liver	403.4	0.76 ± 0.07	0.03 ± 0.02	0.94 ± 0.08	—	—	—	1.2 ± 0.2	—
Lung	283.95	2.0 ± 0.1	0.06 ± 0.02	2.4 ± 0.1	0.34 ± 0.05	0.34 ± 0.05	0.17 ± 0.04	1.2 ± 0.08	0.14 ± 0.02
Kidney	163.1	3.4 ± 0.2	0.13 ± 0.004	3.9 ± 0.2	0.07 ± 0.03	0.28 ± 0.07	−0.22 ± 0.06	1.1 ± 0.08	0.07 ± 0.018
Vertebrae	56.9	4.7 ± 0.4	0.20 ± 0.10	5.8 ± 0.5	−0.07 ± 0.15	1.1 ± 0.3	−0.17 ± 0.39	1.2 ± 0.1	0.19 ± 0.05
Ribs	54.5	2.8 ± 0.4	0.08 ± 0.08	4.5 ± 0.5	0.43 ± 0.23	1.1 ± 0.3	0.49 ± 0.28	1.6 ± 0.3	0.24 ± 0.07
Sternum	29.8	4.0 ± 0.6	0.31 ± 0.12	6.5 ± 0.8	0.12 ± 0.11	0.51 ± 0.17	0.33 ± 0.18	1.6 ± 0.3	0.08 ± 0.027

Table 7.6

Mean Concentrations of Uranium and Thorium Isotopes in Human Tissues from Colorado

Tissue	No. of Samples	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th
Liver	5	0.20 ± 0.14	0.018 ± 0.0083	0.31 ± 0.16	0.035 ± 0.010	0.13 ± 0.058	0.067± 0.037
Lung	5	1.3 ± 0.36	0.062 ± 0.013	1.6 ± 0.52	0.98 ± 0.26	0.78 ± 0.15	0.67 ± 0.18
Lymph nodes	2	1.5 ± 0.97	0.15 ± 0.055	4.3 ± 3.4	2.0 ± 0.20	3.2 ± 0.86	−1.9 ± 1.5
Kidney	5	0.90 ± 0.63	0.032 ± 0.027	1.1 ± 0.72	0.048 ± 0.10	0.11 ± 0.056	−0.015± 0.13
Spleen	1	0.0010	−0.0030	0.030	—	—	—
Thyroid	2	0.48 ± 0.18	−0.15 ± 0.18	0.65 ± 0.17	0.19 ± 0.16	0.89 ± 0.51	−9.0 ± 7.6
Gonad	1	0.03	0.06	0.33	0.82	1.0	5.8
Vertebrae	5	1.3 ± 0.85	0.06 ± 0.036	1.6 ± 1.0	0.20 ± 0.13	0.58 ± 0.16	1.1 ± 0.93
Ribs	3	1.1 ± 0.85	0.13 ± 0.068	1.8 ± 1.4	0.27 ± 0.092	0.71 ± 0.19	1.3 ± 0.75
Sternum	3	2.2 ± 0.88	0.48 ±	3.2 ± 1.7	0.35 ± 0.21	0.32 ± 0.10	3.0 ± 1.3

* Range appears within parenthesis.

n.d. = not detected

TABLE 7.7

Concentration Ratios of $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ in Organs of the General Population

Tissue	$^{234}U/^{238}U$						$^{230}Th/^{234}U$					
	80-A-22	80-A-39	27-88	27-90	32-110	Mean (range)	80-A-22	80-A-39	27-88	27-90	32-110	Mean (range) or upper value
Liver	2.0±.1	2.9± .8	4.0 ±4.3	1.8 ±.8	1.2 ±.2	2.4 (1.2 -4.0)	0.40±.12	0.48 ±.15	0.22±.23	0.22±.23	—	(< 3.6)
Lung	1.4±.17	1.3± .1	0.82± .19	0.97±.09	1.20±.08	1.1 (0.82-1.39)	0.27±.03	0.91 ±.15	0.98±.15	0.98±.14	0.14 ±.02	(< 0.98)
Kidney	1.7±.4	1.4± .2	0.77± .38	—	1.14±.09	1.4 (0.77-1.2)	0.13±.09	0.025±.38	—	—	0.072±.018	(< 0.80)
Lymph nodes	—	3.2±2.1	1.8 ±1.7	—	—	—	—	0.53 ±.31	—	—	—	(< 2.5)
Thyroid	—	1.6±1.5	1.2 ±2.3	—	—	—	—	0.79 ±.79	—	—	—	—
Vertebrae	1.6±.6	1.1± .3	1.1 ± .3	1.5 ±.2	1.2 ±.1	1.3 (1.1 -1.6)	0.96±.27	0.57 ±.21	0.85±.25	0.92±.21	0.19 ±.05	0.70 (0.19-0.96)
Ribs	—	—	1.0 ± .9	2.1 ±.9	1.6 ±.3	(1.0 -2.1)	—	—	2.2 ±.9	0.91±.29	0.24 ±.29	(< 2.2)
Sternum	—	—	1.2 ± .5	1.2 ±.5	1.6 ±.3	(1.1 -1.6)	—	—	0.18±.33	2.9 ±1.1	0.078±.02	(< 2.9)

CHAPTER 8

COMPARISON OF RESULTS WITH PREDICTIONS OF ICRP 30 MODELS FOR URANIUM AND THORIUM

The measured ratios of Th-230/U-234 in bone of uranium miners and millers have been compared with the results predicted by the ICRP metabolic models for uranium and thorium. We were not able to compare the measured results in lung with the ICRP lung model because there was insufficient information available on the airborne concentrations of ore dust and mill atmospheres to which these uranium miners and millers were exposed. For the same reason, we cannot compare the measured results for either nuclide separately in lung, liver, kidney or bone with predicted values using the ICRP metabolic models.

8.1 Thorium

The results of Stover et al. (Sto60, Sto65) on the retention of Th-228 in the beagle constitute the primary basis for the metabolic model adopted by the ICRP. A brief description of this model follows.

Of thorium entering the transfer compartment, 70% (f_{1Th}) is assumed to be translocated to bone, where it is retained with a biological half-life of 8000 days. Another 4% is assumed to be translocated to the liver where it is retained with a biological half-life of 700 days, while 16% is assumed to be uniformly distributed among all other organs and tissues of the body also with a biological half-life of 700 days. The remaining 10% is assumed to be eliminated in excreta.

The equation for thorium retention in bone $R_{Th\ bone}(t)$ in the ICRP model can be expressed as:

$$R_{Th\ bone}(t) = I\ f_{1Th} e^{-\lambda_{Th} t} \quad (1)$$

where I is the thorium intake to blood (or the transfer compartment). Inserting numerical values of individual coefficients in (1) gives:

$$R_{Th\ bone}(t) = I\ (0.7)\ e^{-\frac{0.693t}{8000\ d}} \quad (1')$$

8.2 Uranium

Of uranium entering the transfer compartment, 20% (f_{1U}) is retained in mineral bone with a 20 d half-life and 2.3% (f_{2U}) is retained with a 5000 d half-life. The ICRP model for retention can be expressed by the following equation:

$$R_{Ubone}(t) = I \left(f_{1U} e^{-\lambda_{1U} t} + f_{2U} e^{-\lambda_{2U} t} \right) \quad (2)$$

where I is the intake to blood which is presumed to be the same for U-234 and Th-230, since these radionuclides are close to equilibrium in ore and on the average were found to be in equilibrium in the lungs we measured. Equation (2) can also be written:

$$R_{bone}(t) = I\left[0.2e^{-\frac{0.693t}{20\ d}} + 0.023e^{-\frac{0.693t}{5000\ d}}\right] \qquad (2')$$

Using equations (1) and (2) the ratio of the concentration of thorium to uranium in bone at time t post instantaneous (or short term) exposure and deposition in bone can be calculated. Thus, for an instantaneous single exposure (t_2) the concentration ratio Th-230/U-234 in bone can be approximated by the following equation, which is valid whenever t greatly exceeds the mean residence time in lungs, a condition which holds for most of our samples.

$$F_{Th/U} = \frac{R_{Th\ bone}(t)}{R_{U\ bone}(t)}$$

$$= f_{1Th}\left[f_{1U}e^{-(\lambda_{1U}-\lambda_{Th})t} + f_{2U}e^{-(\lambda_{2U}-\lambda_{Th})t}\right]^{-1} \qquad (3)$$

We have calculated the concentration ratios expected from the occupational histories and periods of retirement for the specific uranium miners and millers we measured. For example, uranium miner U-522 retired after ten years of active mining and died two years later. If his exposure all occurred at the beginning of his first year of employment, then his postexposure time would be 12 years or 4380 days. By inserting 4380 for t in the above equation (3), the concentration ratio Th-230/U-234 is calculated to be 38.

Integrating equations (1) and (2) under the assumption that there was a continuous unvarying rate of intake of thorium and uranium during the miners' and millers' employment (i.e., from $t = 0$ to $t = t_1$), and that after retirement (i.e., from $t = t_1$ to $t = t_1 + t_2$) there was clearance at the rate predicted by the ICRP model functions, one obtains the following equations describing the time dependent accumulation in bone:

$$A_{Th}(t_1 + t_2) = \left[\int_0^{t_1} R_{Th\ bone}(t)dt\right] e^{-\lambda_{Th}(t_1 + t_2 - t_1)}$$

$$= \frac{I\ f_{1Th}}{\lambda_{Th}}\left(1 - e^{-\lambda_{Th}t_1}\right)e^{-\lambda_{Th}t_2} \qquad (4)$$

and

$$A_U(t_1 + t_2) = I \frac{f_{1U}}{\lambda_{1U}}\left(1 - e^{-\lambda_{1U}t_1}\right)e^{-\lambda_{1U}t_2} + \frac{f_{2U}}{\lambda_{2U}}\left(1 - e^{-\lambda_{2U}t_1}\right)e^{-\lambda_{2U}t_2} \qquad (5)$$

Thus the integrated retention ratio for thorium to uranium is obtained as the quotient of equations (4) and (5).

Considering again, for illustrative purposes, uranium miner U-522, one can substitute $t_1 = 3.65 \times 10^3$d and $t_2 = 7.3 \times 10^2$d in equation (6), and obtain a ratio A_{Th}/A_U equal to 32, which is within the measured range from 31 to 54 in vertebrae, ribs and sternum that we report in Table 8.1.

Table 8.1 compares the Th/U ratios predicted by equations (3) and (4)/(5) to the observed ratios of Th-230/U-234. Uranium miner 81-A-31 worked for 32 years. The predicted ratios a, b, and c which are listed in Table 8.1 were obtained by using either equations (3) or (4) and (5), according to the following assumptions:

a. using equation (3), and assuming that exposure occurred in a short period at the end of the last year of employment in mining and milling--that means $t = t_2$;
b. using equation (3), and assuming that exposure occurred at the beginning of the first year of employment in mining and milling, and elimination continued until death--that means $t = t_1 + t_2$; and
c. using equations (4) and (5), and assuming that continuous exposure occurred from the beginning to the end of employment in mining and milling--that means, integrating accumulation during exposure over $0 \leq t \leq t_1$ --and accounting for subsequent elimination during a time $t_1 \leq t \leq t_1 + t_2$ which corresponds to the retirement period.

Figure 8.1 illustrates the time periods used in calculation for exposure cases a, b, and c described above. The elimination periods in cases a and b start immediately after the exposure times, while in case c, there is elimination simultaneously with intake during the employment period, and only elimination during the retirement period.

Four important assumptions have been made; that the exposure from occupational employment is by inhalation, that the rate of exposure is constant during employment, that the dusts to which miners and millers are exposed contain U-234 and Th-230 in equilibrium, and that the exposures are sufficiently high that environmental exposure, particularly from ingestion of uranium can be ignored.

For the first and last cases the observed ratios fall within the extremes of the range predicted by the models. The predicted ratios for the second and third cases do not include the observed ratio within the range of predictions. For both of these cases the predictions would be more consistent with the observed ratio if the true elimination rate of U and Th isotopes from bone differed from those in the ICRP model. The origin of the difference would have to be:

Case 2 elimination of U was more rapid and/or elimination of Th less rapid than the ICRP model

Case 3 elimination of U was less rapid and/or elimination of Th more rapid than the ICRP model.

These 2 cases suggest opposite conclusions. The results from cases 1, 2, and 4 are consistent with the hypothysis that the rate of elimination of U from the long term compartment is really greater than that assumed by ICRP. There is in addition other informtion which usggests the long term half time assumed by ICRP is too long (Wr85).

We conclude that more measurements in the miners are required to better understand the rate of skeletal turnover for man for the long lived isotopes of U and Th.

Table 8.1

Comparison of the Concentration Ratios $^{230}Th/^{234}U$ in Bone of Uranium Miners and Millers as Predicted by ICRP 30 and Observed in Radiochemical Determinations

Subject ID	t_1 (years)	t_2 (years)	Values predicted employing ICRP 30 retention functions*			Observed Values†
			a	b	c	
U-522	10	2	31	38	35	31,45,54
81-A-31	32	0	3.1	56	37	96
81-A-125	4	28	52	56	54	6.8
83-A-79	33	0	3.1	57	32	23

* See also Figure 8.1.

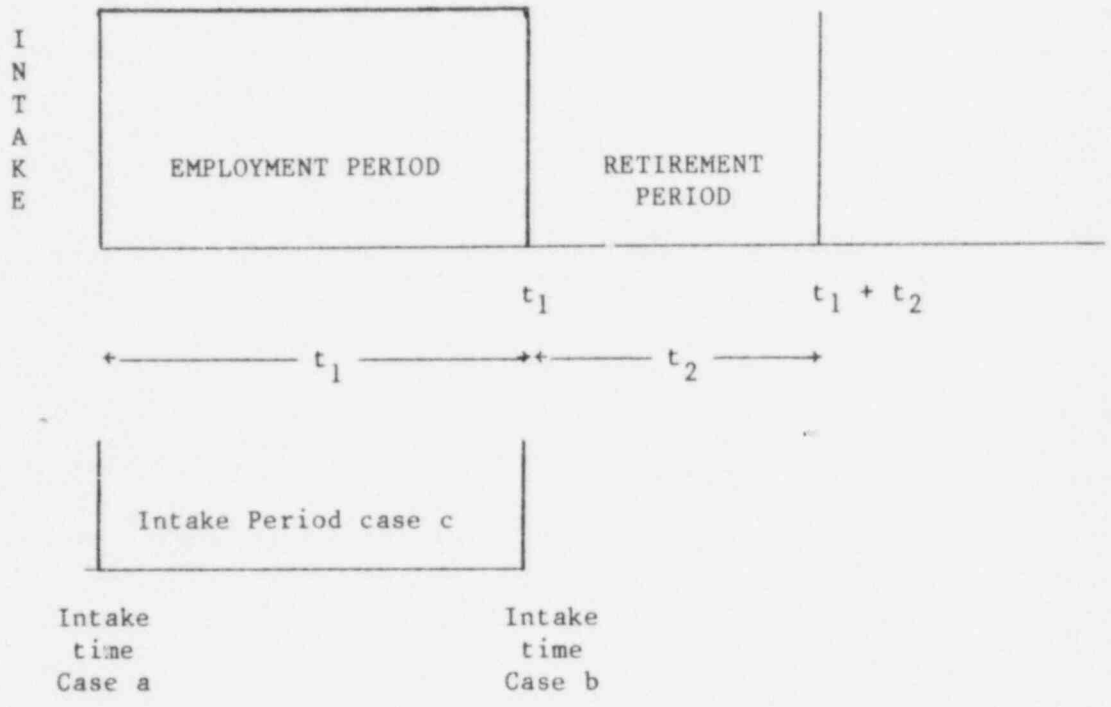
† Measured by radiochemical analyses of bone samples obtained at autopsy.

a Assuming that exposure occurred at the end of the last year of employment in mining or milling, and elimination occurred until death; calculated by equation (3) with $t = t_2$.

b. Assuming that exposure occurred at the beginning of the first year of employment in mining or milling, and elimination occurred until death; calculated by equation (3) with $t = t_1 + t_2$.

c. These subsequent numbers were obtained by using equation (6), where t_1 is the exposure time, and t_2 is the retirement period, which corresponds to the period of elimination after the end of exposure.

Figure 8.1 Illustration of exposure regimens a, b, and c described in the text and in the footnote of Table 8.1.



CHAPTER 9

GENERAL CONCLUSIONS

Among 17 sets of lungs, primarily from uranium miners who died of lung cancer, Th-230 was in equilibrium on the average with U-234 and U-238, suggesting no differential dissolution and retention in the lung of Th and U from retained particles of uranium ore.

The concentrations of Th-230 and U-234,238 were about 100 times higher than that found in tissues from the general population of Colorado.

It was possible to obtain samples of bone and soft tissues from three former miners. Of particular interest was the fact that the Th-230 concentration in bone exceeded that of U-234,238 by a factor exceeding 50 to 100 for two samples, and 3 for the third set of samples. The latter had very little U and Th isotopes in tissue, and appeared similar in content to the tissue for the general public.

An interlaboratory comparison among Los Alamos, the University of Utah and BPNL of radiochemical analyses of sectioned lungs from dogs which had previously inhaled laboratory generated aerosol from uranium ore showed that the ratios of Th-230/U-234 measured on 8 samples deviated on the average only 3% from the mean between laboratories. This indicates that the earlier work of Stuart and Jackson which reported disequilibrium between U-230 and U-234 in animals is a real biological result and that the radiochemical analyses are consistent with those determined by the two other laboratories.

It was possible to collect tissues at autopsy from only 2 former uranium millers. These showed elevated concentrations of U-234,238 and Th-230 in lung relative to the general population, close to radioactive equilibrium. U-234 in skeleton samples exceed Th-230, but both concentrations were low, about 1 pCi/kg.

The Th-230/U-234 ratio in tissues from 5 members of the general public was 1.1 for bone, and was lowest in kidney (0.3).

The ICRP metabolic models for Th and U were only marginally successful in predicting the ratio of Th-230/U-234 in bone of former miners and millers.

CHAPTER 10

RECOMMENDATIONS

1. We saw no evidence which suggested that the NRC special limit for long-lived activity in ore dust, particularly for Th-230, is not appropriate.

2. A long-term program should be established to measure and collect tissues from former millers and miners to better establish the amounts and ratios of long lived U and Th isotopes in their tissues and to test the ICRP models. Two sets of miller samples is insufficient, and experience with this program shows that a long-term program of measurements needs to be in existence in order to assure a sufficient number of samples will be measured to provide statistically and biologically significant information.

   There are two types of tissue collection programs which can furnish information.

   a. A willed autopsy program into which miners and millers enter by written agreement before death. A good program of this type is already being conducted on a continuing basis by the U.S. Uranium registry, operated by the Hanford Environmental Health Foundation, under a long-term contract with the DOE.

      We have an agreement with them to make radiochemical measurements of tissues which they collect but we have no long-term commitment from NRC or any other governmental agency. The U.S. Uranium Registry has no internal capability for radiochemical analysis of tissues.

   b. A program of tissue collection operated by pathologists and Medical Examiners, such as the one established under this contract. This program should be both continued and strengthened.

3. To better establish the distribution of concentrations of U-234,238 and Th-230 and their ratios in controls, further measurements of the same type are required on a larger number of sets of tissues from the generally unexposed public.

4. The ICRP and NCRP models for metabolism and bioassay of uranium and thorium isotopes should be tested against the data on tissues collected from either of these programs.

5. Any further work should be pursued which can shed light on the reasons for the observed fractionation of Th-230 and U-234,238 in animals exposed to laboratory generated ore dust and the lack of such fractionation in human lungs. In particular, the particle size distribution of residual particles in animal and human lungs should be measured to see whether the difference might be attributable to particle size.

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The alpha-emitting isotopes of uranium and thorium were determined in the lungs of 14 former uranium miners and in soft tissues and bones of three miners and two millers. These radionuclides were also determined in soft tissues and bones of seven normal controls. The average concentrations in pCi/kg wet weight in 17 former miners' lungs are as follows: U-238, 75; U-234, 80; Th-230, 79. Concentrations of each nuclide ranged from 2 to 325 pCi/kg. The average ratio of U-238/U-234 was 0.92, ranging from 0.64 to 1.06. The mean ratio of Th-230/U-234 was 1.04, ranging from 0.33 to 3.54. The near equilibrium between Th-230 and U-238,234 indicates that the rate of elimination of uranium and thorium from lungs is the same in former uranium miners. The concentrations of U-234 and U-238 were highest in lung; however, the concentration of Th-230 in bones was either higher than or comparable to its concentration in lung.

The concentration ratios of Th-230/U-234 in bone of uranium miners and millers measured in our laboratory have been compared with results predicted by ICRP-30 metabolic models. These results indicate that the ICRP metabolic models for thorium and uranium were only marginally successful in predicting the ratio of Th-230/U-234 in bones, and that effective release rate of uranium from skeleton may be more rapid than predicted by the ICRP model.

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