6

703486

REPOSITORY DOE - Forrestal
COLLECTION Markey Files
BOX No. 1 of 6
FOLDER FS-1 Plutonium

Reprinted from

Actinides in Man and Animals (M.E. Wrenn, Ed.) – RD Press

Proceedings of the Snowbird Actinide Workshop

October 15-17, 1979

pp. 191-197

1000073

MACRODISTRIBUTION OF PLUTONIUM IN THE HUMAN SKELETON

R.P. Larsen, R.D. Oldham and R.E. Toohey

ABSTRACT

The skeletal remains of two individuals who received plutonium by intravenous injection have been analyzed to establish the skeletal burden and its macrodistribution both amongst and within individual bones. The concentrations in most axial bones were factors of 2 to 4 higher than the average in the entire skeleton; the concentrations in the skull bones were about the same as the average, and the concentrations in the appendicular bones were a factor of 2.5 to 8 lower than the average.

Within any particular bone, the concentration in the trabecular portion was always higher than that in the cortical portion; amongst whole bones, plutonium concentration was not correlated with degree of trabecularity. The bone that could be readily taken at autopsy and whose plutonium concentration closely approximates the skeletal average is the clavicle.

INTRODUCTION

The burden and macrodistribution of plutonium in the skeletal remains of an 18-yr-old woman and a 65-yr-old man who received ^{239}Pu by injection in 1945 have been determined. At the time of the injections, the woman was suffering from Cushing's syndrome, hypertension, nephropathy with uremia, and osteoporosis; the man was suffering from muscular atrophy and dermatitis. They succumbed to their illnesses in 1947; the plutonium they received was tetravalent and in a citrate buffer medium. The subjects, referred to in the early literature as HP-4 and HP-9 (Lan50), have been assigned Center for Human Radiobiology (CHR) case numbers 40-010 and 40-015, respectively. The distribution amongst a few bones and in a long bone, the tibia, were reported previously (Lar76).

The remains of the woman at the time of exhumation were in excellent condition, considering the 26 years of interment. There was no evidence of severe decay. The skin was dehydrated, shriveled, and

---

Work performed under the auspices of the U.S. Department of Energy.

1000074

is colored, but the flesh was moist. Examination showed that a complete autopsy had been performed and all organs including the brain had been removed.

The skeleton was the only identifiable part of the man's remains. It was intact at the time of exhumation and was free of soft tissue. It was slightly moist with a thin coating of organic mud-like material. Some of the highly trabecular bones had begun to soften and decay. Radiograms showed that there was some postmortem lysis in these bones.

SAMPLING

40-010 - Prior to dissection, various tissue samples were taken: diaphragm, thigh muscle (quadriceps femoris) and skin, nerves (femoral), costal cartilage, trachea with larynx, scalp with hair, eyes, and teeth. At dissection, the flesh was carefully removed from the bones, leaving as much as possible of the periosteum intact.

With the exception of the tibia, the samples of bone analyzed were from the left half of the skeleton. For the paired bones, there were three types of samples: (1) groups of bones, such as the tarsals; (2) individual bones, such as the scapula; and (3) sections of individual bones, such as the tibia. The unpaired bones were cut in half, and there were two types of samples from these halves: (1) groups of sectioned bones, such as the processes of the cervical vertebrae; and (2) individual bones, such as the sacrum. In the sampling of the skull, the paranasal and mastoid sinuses were removed, and the remainder of the skull was cut in half along the mid-saggital plane and segmented. The samples from the skull were (1) half the frontal bone and a large fraction of the left parietal bone, (2) the remainder of the parietal bone and half the occipital bone, (3) the left temporal squamous bone, and (4) half the mandible. The samples from the paranasal and mastoid sinuses were not analyzed.

40-015 - The sampling was comparable to that used for 40-010, the principal exception being that some additional samples were taken to establish the symmetry of plutonium deposition. For the study of the relationship between the plutonium concentration and the bone type, the bone was ashed, the trabecular and cortical portions were separated, and each portion was analyzed. The trabecular portion was the material that could be removed from the inner surface of a bone with a minimum of force, and the cortical portion was the material that remained after the inner surface had been forcefully scraped.

ANALYTICAL METHOD

The method of analysis was alpha-spectrometric isotope-dilution. The bone was ashed at 600°C, weighed, and then a weight-aliquot was taken. This was dissolved in nitric acid; a known amount of ^{242}Pu and an excess of 9M hydrobromic acid were added and the solution was

1000075

evaporated to incipient dryness. This ensured isotopic exchange between the ^{239}Pu that was in the bone and the isotopic diluent, ^{242}Pu. The plutonium was separated from the other bone constituents, using the anion-exchange procedure of Larsen and Oldham (Lar75); it was electrodeposited, using the technique reported by Kressin (Kr77), and the deposit was assayed in an alpha spectrometer. The recovery of the plutonium ranged from 75% to 90%; the resolution (FWHM) of the ^{239}Pu and ^{242}Pu peaks in the spectra ranged from 45 to 60 keV. The relative error in any value obtained was less than 5% (1σ). As a quality-control procedure, one bone sample was analyzed repeatedly (10 times) during the period the determinations were made, and 25 other bone samples were analyzed in duplicate at separate times. The relative difference in the values obtained was, in all cases, less than 5% (1σ).

For a few bones it was necessary to estimate their plutonium contents because only portions or none was available for analysis. The estimates were made in one of two ways: (1) from the values obtained in the analysis of a similar bone(s) and the wet weights of the bones, and (2) from the values obtained in the analysis of other sections of the same bone and the wet weights of the sections. An example of the former is the value for the sinuses where the value used was the mean of those obtained for the frontal-parietal, occipital-parietal, and temporal squamous bones. An example of the latter is the value for the diaphysis of the humerus. The mean of the values obtained for three sections was used to calculate the values for the other four sections. The amount of plutonium in those bones for which estimates were made was 6% of the skeletal burden. The assigned relative error of the estimated values was ±25%.

RESULTS AND DISCUSSION

Distribution in the skeleton of 40-010 - The results for 40-010 are summarized in Table 1. The amount of plutonium in the entire skeleton was established by summing the amounts found in the unpaired bones and twice the amounts found in the paired bones. This is 54 ± 2% of the amount injected. The true error is probably larger than this because not all of the bones were analyzed, and lateral symmetry was assumed in the calculation of skeletal burden.

In considering which bone should be taken in autopsy as representative of the concentration of the skeleton, the clavicle would appear to be superior to any other. In the past, rib has been the bone-of-choice because it is readily available. This could continue to be the case, but as seen in Table 1, the mean concentration in a rib is twice that of the average concentration of the skeleton. A comparison of the values for the individual ribs showed that the specific rib taken is not very important; the relative concentrations ranged from 1.72 to 2.34.

Comparison of the distribution of 40-015 with that in 40-010 - The data in Table 2 show that the distribution of plutonium in the skeleton

1000076

TABLE 1: SUMMARY OF PLUTONIUM DISTRIBUTION IN THE SKELETON OF CHR 40-010.

Bones(s)	Percent of Total Pu	Percent of Total Ash		Relative Pu Concentration
Frontal*-Parietal	5.7	5.6		1.0
Occipital*-Parietal	3.6	3.0		1.2
Temporal squamous	1.6	1.7		0.9
Sinus*	5.4 ± 1.3	5.0		1.1
Mandible*	0.8	1.8		0.4
Skull*	17.1 ± 1.7	17.1	Mean	1.00
Innominate	23.4	7.2		3.3
Vertebrae*	20.7	6.2		3.3
Ribs	11.8	5.6		2.1
Scapula	4.7	2.5		1.9
Clavicle	1.1	1.0		1.1
Sacrum*	2.9	0.8		3.6
Sternum*	1.8	0.4		4.5
Axial	66.4	23.7	Mean	2.8
Femur	8.5	20.3		0.4
Patella	0.2	0.6		0.3
Tibia	2.0	13.7		0.15
Fibula	0.5	3.4		0.15
Foot	1.8	6.2		0.3
Humerus	2.0	7.2		0.3
Radius	0.4	2.2		0.2
Ulna	0.4	2.8		0.15
Hand	0.5	2.7		0.2
Appendicular	16.3	59.1	Mean	0.27

*Values given are for half of this (these) bone(s).

1000077

TABLE 2: SKELETAL DISTRIBUTION OF INJECTED PLUTONIUM IN SELECTED BONES OF 40-010 AND 40-015.

Bone	Relative Concentration, Bone/Humerus Midshaft	
	40-010	40-015
Humerus - distal end	0.93	1.09
Femur - head	2.75, 6.40*	2.11
Rib - 2	10.2	10.2
Rib - 8	11.2	8.9
Sternum	26.0	18.0
Vertebrae, cervical, body	19.2	19.3
Vertebrae, thoracic, body	23.6	29.8
Tarsals	1.42	1.09

*These values are for the right and left femur heads, respectively.

of 40-015 is very similar to that in 40-010. The concentrations are relative to those in the respective humerus midshafts. It had been thought that the distribution in 40-010 might be abnormal because of her illness or her age, or both. Cushing's syndrome is characterized in part by osteoporosis and her skeleton could still have been metabolically very active. These factors do not appear to have affected the distribution of plutonium significantly.

Distribution with bones - A general statement about the way in which plutonium is distributed in the skeleton has been that the highest concentrations of plutonium are found in those bones that are primarily trabecular. The results obtained in the analysis of 40-010 revealed exceptions to this generalization: The average concentration in the ribs, which are primarily cortical bone, was a factor of 2.1 higher than the average concentration for the entire skeleton and was the same as that of the femur head, which is primarily trabecular bone. In contrast, the concentration in the tarsals, which are primarily trabecular bone, was a factor of about four lower than the skeletal average and was about the same as the concentration for the femur diaphysis, which is primarily cortical bone.

The distribution of plutonium in the trabecular and cortical portions of selected bones of 40-015 is given in Table 3. The concentrations are relative to that in the cortical bone from the femur midshaft.

1000078

TABLE 3: PLUTONIUM CONCENTRATIONS IN THE CORTICAL AND TRABECULAR PORTIONS OF SELECTED BONES FROM CASE 40-015 RELATIVE TO THAT IN THE FEMUR SHAFT

Bone*		Relative Concentration		Concentration Ratio
		Trabecular	Cortical	Trabecular/Cortical
Femur shaft	(<10)	4.5	(1.00)	4.5
Femur head	(>75)	2.7	1.19	2.3
Humerus shaft	(<10)	2.9	1.12	2.6
Tarsal	(>90)	1.19	**	
Clavicle	(50)	3.3	0.92	3.6
Rib 2	(<10)	25	7.1	3.5
Rib 8	(<10)	29	6.1	4.8
Sternum	(>75)	39	9.1	4.3
Vertebral body				
Cervical	(>75)	31	13.3	2.3
Thoracic	(>75)	43	22.2	1.9

*Numbers in parentheses are the percentages of trabecular bone.
**No cortical portion could be isolated.

It is evident from these data that the generalization cited above (relating plutonium concentration to the degree of trabecularity) is valid only when comparing one part of a bone with another part of the same bone. The distribution of plutonium within the skeleton would appear to be related to the level of metabolic activity of individual bones, not to bone-to-bone differences in composition. For those bones that are the most active metabolically, the concentrations of plutonium in both the trabecular and cortical portions are high, relative to the skeletal average.

A comparison of the plutonium concentration with surface area, as determined by Lloyd and Hodges using computer analysis of microradiographs (Ll71), showed the relationship between the extent of plutonium deposition and area. The trabecular to cortical concentration ratios for the femur head and thoracic vertebrae were 2.3 and 1.9, respectively; while the corresponding area ratios were 3.3 and 4.0. For the ten bones we analyzed, the mean of the concentration ratios was 3.3 $\pm$ 1.0 ; for the four bones they analyzed, the mean of the area ratios was 4 $\pm$ 1.

Two general conclusions can be drawn from this part of the study of plutonium distribution: (1) the concentration of plutonium will always be higher in the trabecular portion of a particular bone than it is in the cortical portion because the surface area of the former is always higher than that of the latter; and (2) the highest concentrations of plutonium in both trabecular and cortical bone are in those bones that have the highest metabolic activity.

1000079

REFERENCES

Kr77 Kressin, I.K. 1977. Electrodeposition of plutonium and americium for high-resolution alpha spectrometry, Anal. Chem. 49: 842-845.

Lan50 Langham, W.H., Bassett, S.H., Harris, P.S., and Carter, R.E. 1950. Distribution and excretion of plutonium administered intravenously to man, Los Alamos Scientific Laboratory Report LASL-1151.

Lar75 Larsen, R.P., and Oldham, R.D. 1975. Anion-exchange separation of plutonium in hydrochloric-hydrobromic acid media, Talanta 22: 577-580.

Lar76 Larsen, R.P., Toohey, R.E., and Ilcewicz, F.H. 1976. Macrodistribution of plutonium in selected bones from an abnormal skeleton, Radiological and Environmental Research Division Annual Report, July 1975 - June 1976, ANL-76-88, Part II, pp. 42-44.

Ll71 Lloyd, E., and Hodges, D. 1971. Quantitative characterization of bone: a computer analysis of microradiographs, Clinical Orthopedics, No. 78, 230-250.

1000080