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● *Paper*

U.S. TRANSURANIUM REGISTRY REPORT ON THE ^{239}Pu DISTRIBUTION IN A HUMAN BODY

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Abstract—**The distribution of ^{239}Pu in a human whole body is reported. The body contained 246 Bq of ^{239}Pu of which 130 Bq (52.8%) was found in the lungs and associated lymph nodes. Of the remaining 116 Bq (47.2%) that constituted the systemic deposition, 51.2 Bq (44%) were in the skeleton, 48.6 Bq (42%) in the liver, and the remainder (14%) in the rest of the body exclusive of the lungs and associated lymph nodes. An unexpectedly high concentration was observed in the pituitary. The systemic distribution of Pu in this case, when combined with the exposure history, is suggestive of an initial partitioning ratio of ^{239}Pu between skeleton and liver of less than unity, and a tentative initial distribution from the transfer compartment of 25% to the skeleton, 50% to the liver, and 25% to the rest of the body and early excretion is proposed for this case. Older biokinetic models, when used with the available urinalysis data for this case, typically overestimated the deposition when compared with the tissue analysis results, but more recent models provided estimates in close agreement with the autopsy results.**

INTRODUCTION

SINCE its inception in August 1968, the United States Transuranium Registry (USTR) has been actively studying the behavior of the transuranic elements in humans and is currently the only research program in the United States actively studying internal depositions of Pu and the higher actinides in human tissues. The program has been described in detail elsewhere (Breitenstein 1981; Swint and Kathren 1984; Kathren 1989) and basically involves radiochemical analysis and subsequent evaluation of selected tissues and organs or the whole body obtained through voluntary donation by persons with known exposure to the transuranic elements.

To date, radiochemical analyses and preliminary evaluation of tissue distribution of actinides have been completed on six whole-body donations to the USTR (McInroy et al. 1989). One of these cases, a chemist who incurred an accidental occupational exposure to ^{241}Am via a wound, has been examined in detail and reported in a dedicated issue of *Health Physics* (Roessler 1985). The present paper describes the detailed evaluation of the second whole-body donation to the USTR and involves an individual with occupational exposure to ^{239}Pu.

CASE DESCRIPTION AND EXPOSURE HISTORY

The donor (USTR Case 193) was a Caucasian male who died at age 62 from respiratory failure associated with pneumonia. The donor had a clinical history of congestive heart failure that was consistent with the finding of left ventricular dilation observed at autopsy. Autopsy findings also revealed chronic lung congestion with changes typical of congestive heart failure and generalized moderate to severe atherosclerosis of the coronary arteries. There was multifocal fibrosis of the left ventricle, which was consistent with a clinical history of two previous myocardial infarctions. The clinical history of this case included a coronary occlusion in 1968 and a coronary insufficiency event in 1976, a history of hyperlipidemia, pneumonitis in 1974, and several surgical procedures. The subject was one of 26 male subjects exposed to Pu and medically followed for 37 y and is identified as Case 27 in that study (Hempelmann et al. 1973; Voelz et al. 1979, 1985).

Except for the period August 1946 to November 1949 when he was in college, the donor was continuously employed at the same site and involved in operations with potential for exposure to Pu from January 1945 until February 1982, approximately 10.5 mo prior to his death. Exposure potential was judged to be greatest in the early years of employment by the operational health physics staff at the place of employment. This was perhaps especially so in the first year and a half of his employment, when he was engaged in making Pu fluoride by fluorination of PuO in a chemical hood. Subsequent work was done in glove boxes, which would be expected to afford a greater measure of protection. After 1970, exposure potential was markedly reduced because of changes in job responsibilities that removed him from direct contact with Pu operations.

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Table 1. Radiourinalysis and nose wipe data.

Date	^{239}Pu in Urine mBq/24 h	^{239}Pu in Urine pCi/24 h	Other
27 Mar 1945			Nose wipes, 168/23 cpm[a]
20 Apr 1945	-3.7	-0.1	
12 Jun 1945			Nose wipes, 306/187 cpm[a]
14 Jun 1945			Nose wipes, 225/40 cpm[a]
6 Jul 1945			Nose wipes, 800/616 cpm[a]
16 Jul 1945			Nose wipe, 186 cpm[a]
31 Jul 1945			Nose wipes, 168/146 cpm[a]
1 Aug 1945			Nose wipes, 156/50 cpm[a]
3 Aug 1945			Nose wipes, 153/26 cpm[a]
12 Aug 1945			Nose wipes, 1562/156 cpm[a]
20 Aug 1945			Nose wipes, 7696/4598 cpm[a]
28 Aug 1945			Nose wipes, 124/106 cpm[a]
30 Aug 1945			Nose wipes, 374/88 cpm[a]
6 Sep 1945	102	2.75	
13 Feb 1946			Nose wipe, 224 cpm[a]
25 Mar 1946	102	2.75	
12 Jun 1946	28.9	0.78	
8 Aug 1946	93.6	2.53	
27 Apr 1950	-7.8	-0.21	
23 Sep 1951	-1.1	-0.03	
5 Nov 1951	-1.9	-0.05	
29 Jan 1952	-7.8	-0.21	
30 Jan 1952	-4.8	-0.13	
15 Mar 1952	-4.1	-0.11	
30 Apr 1952	13.7	0.37	
23 May 1952	-0.4	-0.01	
29 Aug 1952	-1.1	-0.03	
27 Oct 1952	-4.8	-0.13	
9 Dec 1952	-6.3	-0.17	
20 Jan 1953	4.8	0.13	
3 Jun 1953	4.8	0.13	
10 Aug 1953	5.2	0.14	
11 Feb 1954	-6.3	-0.17	
19 May 1954	5.2	0.14	
22 Jul 1954	3.7	0.10	
24 Aug 1954	3.3	0.09	
27 Sep 1954	-6.7	-0.18	
26 Oct 1954	5.6	0.15	
30 Nov 1954	-1.1	-0.03	
29 Dec 1954	1.1	0.03	
18 Feb 1955	6.7	0.18	
8 Mar 1955	-3.3	-0.09	
19 Apr 1955	4.8	0.13	
26 May 1955	12.2	0.33	
27 Jun 1955	-1.1	-0.03	
13 Jul 1955	3.3	0.09	
27 Jul 1955	-5.2	-0.14	

Table 1. (*Contd.*)

Date	^{239}Pu in Urine mBq/24 h	^{239}Pu in Urine pCi/24 h	Other
23 Aug 1955	-7.8	-0.21	
23 Sep 1955	-4.1	-0.11	
20 Oct 1955	3.3	0.09	
17 Nov 1955	4.1	0.11	
23 Dec 1955	-2.2	-0.06	
25 Jan 1956	-1.1	-0.03	
20 Feb 1956	-1.9	-0.05	
22 Mar 1956	-4.1	-0.11	
19 Apr 1956	2.2	0.06	
10 May 1956	4.1	0.11	
7 Jun 1956	-7.0	-0.19	
12 Jul 1956	10.7	0.29	
16 Aug 1956	-2.6	-0.07	
17 Sep 1956	-1.9	-0.05	
16 Oct 1956	-1.9	-0.05	
16 Nov 1956	-1.1	-0.03	
14 Dec 1956	-4.1	-0.11	
14 Jan 1957	-6.3	-0.17	
18 Feb 1957	3.0	0.08	
20 Mar 1957	3.0	0.08	
22 Apr 1957	3.7	0.10	
28 May 1957	1.9	0.05	
20 Jun 1957	2.6	0.07	
22 Jul 1957	2.6	0.07	
16 Aug 1957	2.6	0.07	Involved in acute accidental exposure incident; nasal wipe 7 Bq (186 pCi).
17 Aug 1957	7.8	0.21	
18 Aug 1957	0.0	0.00	Fecal sample contained 15 Bq (400 pCi).
23 Sep 1957	5.6	0.15	
24 Oct 1957	4.8	0.13	
26 Nov 1957	1.9	0.05	
6 Jan 1958	4.4	0.12	
17 Feb 1958	4.1	0.11	
28 Mar 1958	0.0	0.00	
14 May 1958	3.7	0.10	
23 Jun 1958	3.7	0.10	
28 Jul 1958	5.2	0.14	
10 Sep 1958	1.9	0.05	
3 Nov 1958	4.4	0.12	
18 Dec 1958	2.6	0.07	
26 Jan 1959	5.2	0.14	

Table 1. (*Contd.*)

Date	^{239}Pu in Urine mBq/24 h	^{239}Pu in Urine pCi/24 h	Other
3 Feb 1959			Involved in acute inhalation exposure while wearing respirator; nasal wipe contained 0.7 Bq (18 dpm)
9 Feb 1959	7.8	0.21	
24 Feb 1959	2.6	0.07	
8 Apr 1959	3.7	0.10	
21 May 1959	3.0	0.08	
29 Jun 1959	4.1	0.11	
15 Jul 1959			Involved in possible actue inhalation exposure; no record of nasal wipe(s).
20 Jul 1959	3.0	0.08	
11 Sep 1959	1.5	0.04	
19 Oct 1959	3.0	0.08	
30 Nov 1959	3.0	0.08	
25 Jan 1960	2.2	0.06	
2 Mar 1960	3.7	0.10	
15 Apr 1960	2.6	0.07	
26 May 1960	2.2	0.06	
12 Jul 1960	0.0	0.00	
26 Aug 1960	3.3	0.09	
6 Oct 1960	3.3	0.09	
16 Nov 1960	0.4	0.01	
17 Jan 1961	3.3	0.09	
17 Feb 1961	0.4	0.01	
13 Apr 1961	2.2	0.06	
29 May 1961	1.9	0.05	
17 Jul 1961	1.2	0.03	
5 Sep 1961	1.9	0.05	
19 Oct 1961	3.0	0.08	
7 Dec 1961	2.6	0.07	
26 Jan 1962	3.3	0.09	
15 Mar 1962	1.9	0.05	
25 Apr 1962	2.6	0.07	
13 Jun 1962	3.0	0.08	
16 Jul 1962	0.7	0.02	
31 Aug 1962	0.0	0.00	
17 Oct 1962	0.7	0.02	
23 Jan 1963	0.0	0.00	
13 Mar 1963	3.3	0.09	
26 Apr 1963	2.6	0.07	
18 Jun 1963	4.1	0.11	
2 Aug 1963	1.9	0.05	
23 Sep 1963	1.1	0.03	
21 Nov 1963	4.1	0.11	
13 Jan 1964	3.0	0.08	

Table 1. (*Contd.*)

Date	^{239}Pu in Urine mBq/24 h	^{239}Pu in Urine pCi/24 h	Other
5 Feb 1964	4.8	0.13	
1 Apr 1964	3.3	0.09	
25 May 1964	4.1	0.11	
6 Jul 1964	0.0	0.00	
31 Aug 1964	3.7	0.10	
24 Sep 1964	1.9	0.05	
21 Oct 1964			High room air count; possible acute inhalation exposure.
26 Oct 1964	3.0	0.08	
17 Dec 1964	1.9	0.05	
25 Jan 1965	2.2	0.06	
8 Mar 1965	3.3	0.09	
22 Apr 1965	3.3	0.09	
19 Jul 1965	2.2	0.06	
18 Oct 1965	1.1	0.03	
20 Jan 1966	0.4	0.01	
18 Apr 1966	4.4	0.12	
29 Nov 1966	4.4	0.12	
22 May 1967	3.3	0.09	
30 Nov 1967	3.3	0.09	
6 Jun 1968	3.3	0.09	
2 Dec 1968	6.7	0.18	
12 Jun 1969	0.0	0.00	
11Dec 1969	5.6	0.15	
11 Jun 1970	8.9	0.24	
12 Nov 1970	3.7	0.10	
18 Jan 1971	1.9	0.05	
19 Jul 1971	5.9	0.16	
6 Jan 1972	5.2	0.14	
6 Jan 1972	4.4	0.12	
6 Jan 1972	4.8	0.13	
18 Jan 1972	4.4	0.12	
8 Jun 1973	4.8	0.13	
18 Jul 1973	4.1	0.11	
7 Jan 1974	3.3	0.09	
10 Jun 1974	3.3	0.09	
11 Sep 1974	1.5	0.04	
5 Dec 1974	5.6	0.15	
5 Mar 1975	5.2	0.14	
9 Jun 1975	4.8	0.13	
5 Sep 1975	1.5	0.04	
8 Dec 1975	3.3	0.09	
12 Mar 1976	3.7	0.10	
14 Jun 1976	4.1	0.11	
7 Sep 1976	1.9	0.05	

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Table 1. (*Contd.*)

Date	^{239}Pu in Urine mBq/24 h	^{239}Pu in Urine pCi/24 h	Other
6 Dec 1976	1.5	0.04	
10 Mar 1977	1.9	0.05	
10 Jun 1977	2.2	0.06	
12 Sep 1977	3.0	0.08	
13 Jul 1978	3.0	0.08	
12 Oct 1978	2.2	0.06	
8 Dec 1978	1.1	0.03	
14 Mar 1979	4.1	0.11	
8 Jun 1979	2.2	0.06	
18 Sep 1979	1.9	0.05	
12 Dec 1979	1.5	0.04	
19 Mar 1980	3.3	0.09	
11 Jun 1980	4.4	0.12	
10 Sep 1980	1.1	0.03	
5 Jan 1981	2.2	0.06	
8 Mar 1981	1.9	0.05	
12 Jun 1981	3.0	0.08	
2 Sep 1981	4.1	0.11	
7 Dec 1982	2.2	0.06	
13 Jan 1982	2.2	0.06	

[a]cpm = counts per minute; efficiency and other calibration factors unknown.

Over the course of his working lifetime, the donor had been credited with a documented external exposure of 132 mSv (13.2 rem) to the skin of the whole body and 93 mSv (9.3 rem) of whole body penetrating radiation, including 38 mSv (3.8 rem) from neutrons. Various bioassay measurements were made over the course of his employment including 178 radiourinalyses for Pu, *in-vivo* counting on 12 occasions subsequent to 1970, and nasal wipes on a routine basis during the early months of employment and following possible acute accidental inhalation exposures. The Pu urinalysis data from 1945–1977 have been reported elsewhere (Voelz et al. 1979) but are reproduced for convenience and completeness in Table 1 along with subsequent urine data and nasal wipe results.

Not all nasal wipe data are given in the table; nasal wipes were taken more or less routinely as frequently as several times daily, depending on the work, from about mid-February 1945 to mid-May 1946 and the results were recorded in counts per minute. Typically, nasal wipe levels were on the order of a few—less than 10—counts per minute, but in some instances significantly higher counts were recorded. Only those instances in which the levels for either nostril exceeded 100 counts per minute are included in Table 1. Exact interpretation of the nasal wipe results cannot be made, as counting techniques, geometries, efficiencies, background level, and other necessary data are lacking, including particle-size distributions, solubility, respirator usage, and whether the individual in question was a mouth breather. They are, however, indicative of potential exposure to airborne Pu during 1945–1946, particularly during June to August 1945 when significant activity was noted on 11 different occasions. During these months, the typical nasal wipe count was also elevated by about an order of magnitude, running a few tens of counts per minute. Subsequently, the counts dropped back to a few counts per minute, and then to near zero shortly before the program was terminated. Air monitoring data for this period are not available.

A crude order of magnitude estimate of intake during this period, based on the limited data available and in accordance with operational health physics experience and practice, indicates that several hundred becquerels, and perhaps as much as 1 kBq, may have reached the T-B (tracheobronchial) and P (pulmonary) regions of the respiratory tract. For the purposes of this estimate, the counting efficiency was assumed to be 25%, and the particle-size distribution was such that 90% of the inhaled activity was deposited in the nose and subsequently removed by the nasal wipe. These assumptions may or may not be valid.

counting results were positive, indicating approximately 13–19 Bq (0.35–0.5 nCi) of ^{241}Am. The lower detection limit for ^{241}Am chest counting during this time was 13 Bq (0.34 nCi). The chest counts in recent years were made using twin 127-mm (5-inch) diameter phoswich detectors and a counting time of 2000 s. At the 95% confidence level, the lower level of detection for this system is approximately 1 kBq (26 nCi) for ^{238}Pu and 2 kBq (55 nCi) for ^{239}Pu. The LLD for ^{239}Pu is greater than the activity measured in the respiratory tract (130 Bq) by postmortem radiochemical analysis, and it is thus not surprising that this nuclide was not detected by chest counting. A count of the hands was made with this same detector system in 1981 and revealed no detectable ^{239}Pu activity [LLD-40 Bq (1 nCi)]. Other *in-vivo* counts were made with a hyperpure germanium detector and included a liver count, in which the detector was positioned over the organ. No activity above the LLD for ^{239}Pu of approximately 40 Bq (1 nCi) was detected.

ANALYTICAL PROCEDURES

This whole-body donation was handled in accordance with the standard USTR protocol for whole-body donations (Breitenstein 1981; Kathren 1989). A standard autopsy was performed, and the internal organs were removed, frozen, and sent to Los Alamos National Laboratory (LANL) for subsequent radiochemical analysis. The body was shipped to the USTR facilities in Richland, WA, where the soft tissues and bones were separated, wet weighed, frozen, and sent to LANL for radiochemical analysis along with the internal organs. Tissue and organs were either analyzed whole or subdivided for convenience of analysis as well as to ascertain possible differences in actinide distribution or variation throughout the larger specimens.

The basic radiochemical procedure for Pu has been described in detail by Boyd and coworkers (1981) and McInroy et al. (1985). Oven-dried tissues are spiked with ^{242}Pu and alternately wet and dry ashed in a furnace until all visible carbonized material is destroyed, normally a process that requires 4 d. After treatment with $LiNO_3$-$NaNO_3$, the ash-salt mixture is dissolved in 7.8 M HNO_3; lung and lymph nodes are treated with HF before final dissolution. Samples that do not dissolve in the nitric acid are dissolved in HCl.

The acidic solution is treated with $NaNO_2$ to stabilize Pu (IV) and eluted through an anion ion exchange column using multiple washings with HNO_3, HCl, and HF. Samples that are insoluble in HNO_3 are run through a chloride anion exchange. Plutonium from the eluate is electrodeposited from acidic solution of H_2SO_4 and H_3PO_4 onto stainless steel planchets and quantified by α spectroscopy using silicon surface barrier detectors. The method has a detection limit of approximately 0.7 mBq (0.02 pCi) per aliquot analyzed.

Pu CONTENT OF TISSUES

The Pu content of the various tissues and organs, along with wet organ weights, are summarized in Table 2; complete data are given in tables in Appendices A and B. Specific data for individual bone samples have been previously presented and evaluated in terms of their con-

Table 2. Tissue weights and Pu contents.

Tissue	Wet weight (g)	^{239}Pu Content		Percent of total in body	Percent of systemic
		Total (Bq)	mBq g^{-1}		
Respiratory tract	1,337	130.0	97	52.8	—
Lungs	1,303	86.8	67	35.3	
Trachea	17.4	0.07	4.3	0.03	
TBLN	16.5	43.1	2,612	17.5	
Liver	1,863	48.6	26	19.7	41.8
Kidneys	326.3	0.09	0.28	0.03	0.1
Spleen	258.3	1.46	5.7	0.6	1.2
Smooth muscle[a]	1,972	0.59	0.30	0.2	0.5
Striated muscle	24,659	8.59	0.35	3.5	7.4
Misc. muscle[b]	945	0.29	0.31	0.1	0.3
Skin	19,688	2.57	0.13	1.0	2.2
Testes	14.4	0.01	0.69	—	0.0002
CNS	1,332	0.18	0.14	0.07	0.2
Other soft tissue[c]	420.1	2.80	6.6	1.2	2.6
Pituitary	0.54	0.01	28.8		
Gall bladder	26.5	0.11	4.0		
Prostate	24.2	0.10	4.1		
Skeleton[d]	8,691	51.2	5.9	20.8	43.9
Skeleton (ash)	2,681		19.1		

[a] GI tract + urinary bladder.
[b] Heart, tongue, diaphragm.
[c] Pancreas, gall bladder, prostate, peritesticular tissue, eyes, salivary glands, thyroid, pituitary.
[d] Including teeth.

The urinalysis data presented in Table 1 need to be carefully considered and evaluated for relevancy and accuracy with respect to exposure potential; serious questions can be raised with respect to the validity of the earlier measurements. In their history of the Pu bioassay program at Los Alamos from 1944 to 1972, Campbell and coworkers (1972) reported marked differences in the sensitivity and reliability of radiourinalyses performed in the early years when compared with those performed more recently. The specific urinalysis data for this case, up through 1977, were examined by J. N. P. Lawrence and included as Appendix I in the paper by Voelz et al. (1979). Lawrence notes that prior to 1957, urine assay results were recorded in terms of counts per minute per 24-h sample, without correction for chemical recovery, chemical reagent background, or counting geometry (Lawrence 1978; Voelz et al. 1979). Blank and chemical recovery procedures were performed periodically, but not on a regular basis (Lawrence 1978). Conversion of the count per minute values to activity was made on the basis of a statistical evaluation of the raw data recorded in laboratory notebooks (Lawrence 1978); these are the data that appear in Table 1.

Thus, the early urinalysis data, defined as that prior to 15 January 1957 but excluding 1945–1946, as noted below, are of questionable validity in that the detection limit at the 95% confidence level for $^{239+240}$Pu in a 24-h urine sample was greater than about 4 mBq (0.1 pCi). For urine samples collected and analyzed prior to this cutoff date, the majority of the results (29 of 52) are reported as negative values (Table 1, Voelz et al. 1979). The four high values observed during 1945–1946 are considered indicative of a valid intake of Pu during that time period and thus are consistent with the nasal wipe data for that same period. Subsequent to January 1957, the urine assay procedures were altered, and based on the discussions and evaluations by Campbell et al. (1972) and Lawrence (1978; Voelz et al. 1979), the urinalysis results reported in Table 1 from February 1957 onward are considered satisfactory and achieved at a detection limit of 1 mBq (0.03 pCi) per 24-h urine sample.

Additional insight into the exposure history can be obtained from Fig. 1, which is a plot of urinary excretion as a function of time. To smooth out variations in the individual data points, all individual urine values obtained during a given calendar year were averaged and then plotted against the year. Because of the uncertainties and negative values in the early data, no points are shown for the years prior to 1957 except for 1945 and 1946, when the highest and clearly positive individual values were obtained. Review of Fig. 1 and the more detailed urinary excretion data in Table 1 along with other information from the exposure history, such as nasal wipe data and potential involvement in known incidents, is suggestive of two periods during which most of his inhalation exposure is likely to have occurred—one of several months duration during 1945, and a longer period from about 1963–1970 to lower levels. Subsequent to 1970, exposure either ceased or was very much reduced. There may also

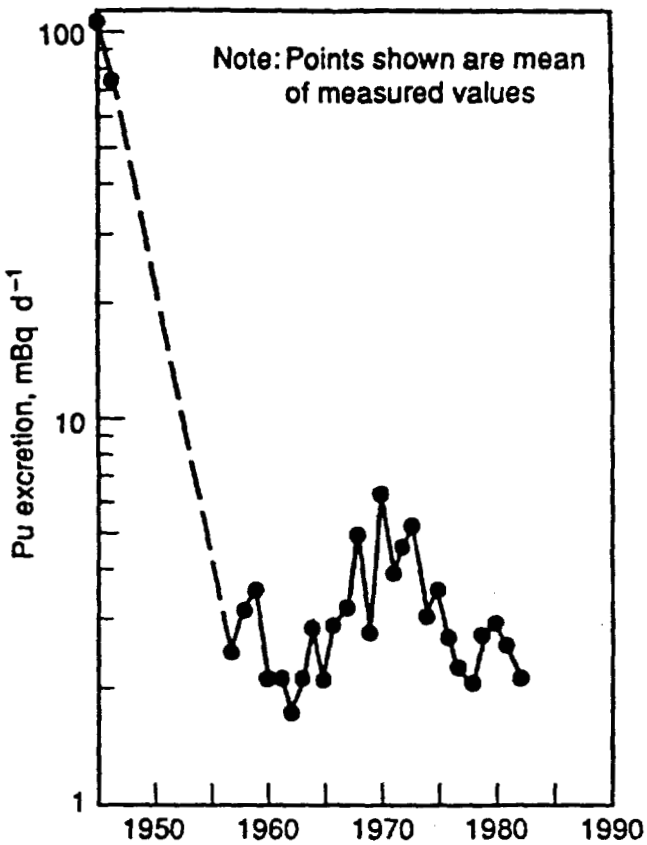

Fig. 1. Mean of measured daily urinary excretion values of by year for USTR Case 193.

have been some additional small exposures between 1949 and 1963. The data suggest that the intake during 1945 was somewhat greater, perhaps severalfold, than the intake during the subsequent period of exposure. During the early exposure period in 1945 and 1946, the chemical form was a mixture of Pu fluoride and Pu oxide; the later exposures were probably largely to oxides of Pu.

On at least four separate occasions, this case was involved in some manner in contamination incidents involving Pu. The dates of these incidents were: 16 August 1957, 3 February 1959, 15 July 1959, and 21 October 1964. The first two of these involved minor explosions that resulted in significant airborne Pu levels in the workroom. As indicated in Table 1, nasal contamination was detected in both instances. Fecal sampling following the first instance revealed a significant quantity of Pu, suggestive of inhalation or ingestion of insoluble material. These bioassay data are suggestive of a small intake of soluble Pu. The latter two incidents were apparently of minor significance, without recorded indication of intake.

In-vivo counts were taken at irregular intervals on 12 occasions, the last being in January 1982. The first count, taken in April 1970, showed no detectable activity, but the state of the art at that time renders this resu[l]t questionable. The next chest count, taken in August 19[7] indicated 20 Bq (0.55 nCi) of ^{241}Am. Subsequent ch[e]

centration and ash content (Kathren et al. 1987) and hence are not repeated in this work. The total body content of ^{239,240}Pu was determined to be 246 Bq (6.6 nCi), approximately equally divided between the respiratory tract and the remainder of the body. Plutonium concentrations were greatest in the lungs and associated lymph nodes. The tracheobronchial lymph nodes weighed 16.5 g and contained 43.1 Bq (1.2 nCi) or approximately 17.5% of the total Pu found in the body. The concentration in the tracheobronchial lymph nodes was 2.6 Bq g^{-1} (71 pCi g^{-1}), which is 25-fold greater than the concentration in the remainder of the lungs and two to four orders of magnitude greater than found in all other tissues. This distribution is consistent with the known exposure history of this individual as described above.

Of the Pu exclusive of that in the respiratory tract—i.e., the so-called systemic deposition—approximately equal amounts were found in the liver and skeleton (41.8% and 43.9%, respectively), with approximately 10% in the muscles and skin. About 1.2% of the systemic deposition was found in the spleen. Excluding the respiratory tract, concentrations were greatest in the liver and bone ash, being about two orders of magnitude greater than those in the remaining tissues. The fraction of the systemic Pu in the gonads, 1.95×10^{-4}, and the concentration relative to the concentration in the other soft tissue was in agreement with what has been observed in other mammals and previously reported by Richmond and Thomas (1975) for U.S. residents exposed via fallout.

Certain tissues or specific portions (samples) showed apparently elevated concentrations of Pu, including gall bladder, prostate, and pituitary, data for which have been included in Table 2. In particular, the pituitary showed a high concentration [28.8 mBq g^{-1} (0.789 pCi g^{-1})]. Although the quantity of Pu in this organ is clearly quite small because of its small size, a concentration of the magnitude observed would deliver a dose to this organ on the order of that delivered to bone or liver and hence could be of importance from a protection standpoint.

Elevated concentrations were also noted in the skin of the left hand [5.7 mBq g^{-1} (0.15 pCi g^{-1})] and a portion of the abdominal muscle and overlying fat and fascia (11.5 mBq g^{-1}). At least in the case of the hand, the possibility of an undetected wound appears to be a reasonable explanation; this might also be true for the abdominal muscle and fat, although less likely.

The activity content and distribution in specific bones and bone samples have previously been reported and were similar to those of the first whole-body case (Kathren et al. 1987). Additional analysis of the distribution of Pu in the skeleton of this case has been reported separately along with comparative data from three other USTR cases (Lynch et al. 1988).

COMPARISON WITH A RELATED CASE

USTR Case 60, also a male Caucasian, provides a direct comparison with this case. Case 60 has been previously identified as Case 16 in the series of Los Alamos workers reported by Hempelmann et al. (1973) and Voelz and coworkers (1979, 1985). From approximately June 1945 to June 1946, Case 60 and Case 193 had worked together in the same organizational component, essentially alongside one another in the laboratory on a daily basis if anecdotal accounts of other coworkers are accurate. In mid-1946, Case 60 moved to another part of the country and entered another line of employment that did not involve work with or exposure to Pu or other actinides. Following his death in 1975 at age 50 from injuries received in a traffic accident, tissue samples obtained at autopsy were donated to the USTR and radiochemically analyzed for Pu and Am. The relative distribution of activity in the tissues of the two cases was similar.

A total of six urine samples was obtained from Case 60 during 1945–1946, while he was involved in Pu work, with nine more taken some years later. With one exception, these samples were all positive for Pu and are indicative of an internal deposition. Like Case 193, urinalysis results obtained prior to 1957 may be of questionable validity. However, the post-1957 urinalysis data were used in conjunction with the Los Alamos PUQFUA3 code to obtain an estimated body burden of 670 Bq (18 nCi) at the time of death. (The value quoted in Voelz et al. (1985) was incorrectly stated as 7 nCi.)

The same PUQFUA3 code was also used to obtain an estimated body burden for Case 193. This was also calculated to be about 259 Bq, or 0.43, the value obtained for Case 60. Previous urinalysis-based estimates were made in 1972 and 1977 with earlier versions of PUQFUA; these indicated that Case 193 had 1.67 and 1.5 times, respectively, as much deposition as did Case 60. The estimated burdens for both cases made with the earlier versions of the PUQUFUA code were severalfold greater than those made with the latest version.

As has been noted, the two cases reportedly worked together during the period 1945–1946, but this does not mean they received equal depositions during this period. Case 60 received his only known exposure during 1945–1946. Large differences were observed in the ^{241}Am to ^{239}Pu activity ratio in comparable tissue samples [liver, lung, muscle, bone (three samples)] for these two cases. In Case 193, the activity ratio of ^{241}Am to ^{239}Pu was typically three to eight times greater than the activity ratio for comparable tissues in Case 60. Only a small part of this difference can be accounted for by additional ingrowth of ^{241}Am from the Pu exposure incurred during 1945–1946, since Case 193 lived approximately 7 y longer than Case 60. Thus, the additional ^{241}Am in USTR Case 193 is indicative of subsequent exposure to ^{241}Am or ^{239}Pu containing a higher percentage of ^{241}Am at some later time in his career. This is fully consistent with the occupational exposure history and urinalysis results available for both cases 60 and 193, as discussed above.

COMPARISON WITH REFERENCE MAN

As radionuclide intake limits are established using anatomical and physiological values of the Reference

Man, which in turn are derived from composite data representative of young (20–30 y old) Caucasian males (ICRP 1975), comparison of organ weights and other physical characteristics for Case 193 with those of Reference Man is of interest. As determined from medical records over a period of years, the USTR donor was 1.73 m (68 inches) tall and normally weighed 75 kg (165 lb). These values are approximately those of the Reference Man (1.70 m and 70 kg). Representative organ and tissue weights, obtained after receipt of the various specimens at the radiochemical laboratory, are given in Table 3 along with comparable values derived from Reference Man (ICRP 1975).

For most organs, weights were in close agreement with those reported for Reference Man (ICRP 1975), but in a few cases were significantly greater. For the smaller tissues and organs, this increase may in some measure be attributable to a small amount of adjacent tissue removed with the organ and included in the initial organ weight. Higher organ weights, particularly in the case of the lungs, also may be attributable to excess fluid content (edema) attributable to the terminal illness (congestive heart failure). The autopsy report noted some fibrinopurulent material adherent to the surfaces of the left lung in the area of the thoracotomy performed shortly before death. The weight of the liver, an important depot for ^{239}Pu in the body, was within a few percent of the Reference Man value.

The wet weight of the skeleton of this case was approximately 87% that of Reference Man and about 11.6% of his normal adult body weight, as compared with 14.2% for Reference Man. This difference is not considered significant. The ash fraction of the skeleton was slightly greater than that of Reference Man—30.9% as compared with 28%—with the weight of the ash, 2681 g, being only a few percent less than the Reference Man value of 2800 g and within the limits of variability reported by Trotter and Peterson (1962). The skeleton is thus unremarkable and typical of a normal 62-y-old male.

Table 3. Comparison of tissue and organ weights with Reference Man.

Organ/tissue	Case 193 wet weight (g)	Reference man weight (g)	Case weight/ Ref. man weight
Lungs	1,303	1,000	1.30
[illegible]	1,863	1,800	1.04

SOME IMPLICATIONS FOR MODELING

Estimates of systemic and total body deposition from urinalysis

The movement and deposition of Pu in the tissues of the human body following intake is complex and dependent on many factors including the route of entry, physicochemical form, and time of entry. The earliest biokinetic studies were made during the days of the Manhattan District and were directed toward the practical health physics need of relating the urinary excretion of Pu to the intake and deposition of Pu. The initial attempts to develop a human biokinetic model were based on excretion data obtained from animal studies. These studies led to a series of experiments involving the injection of 18 hospitalized persons with soluble Pu during 1945 and 1946 (Russell and Nickson 1951; Durbin 1972).

From these experiments, Langham and his coworkers at Los Alamos, in conjunction with the Atomic Energy Project of the University of Rochester School of Medicine and Dentistry, derived an empirical power function excretion model for Pu in humans (Langham et al. 1950). This model was based on observations from 12 of the injection cases plus 6 y of experience with the control of occupational exposure associated with processing large amounts of Pu. The equation derived by Langham and his coworkers (1950) gives the percentage daily urinary excretion of an acute intake of Pu, $Y(u)$, as a function of the number of days t after intake as:

$$Y(u) = 0.2t^{-0.74}. \quad (1)$$

This model has been reevaluated and refined numerous times over the years by various investigators and has served as the basis for several models currently used in operational health physics practices to estimate the systemic burden of Pu.

Typically, estimates of systemic Pu deposition *in-vivo* are made from the measured Pu content of urine samples collected either on a routine basis or following a known or suspected acute exposure. These data are then used with a specific biokinetic model, generally based on that of Langham et al. (1950), to estimate the systemic deposition by dividing the fractional daily excretion on day t, i.e., the day the sample was collected, into the activity found in the urine. This deceptively simple procedure is fraught with great potential for error; some of the more obvious significant sources of error include the precision of the measurement of Pu in the urine sample, the

Rundo et al. (1976), Parkinson and Henley (1981), Leggett (1984), Jones (1985), and Leggett and Eckerman (1987). These estimates are given in Table 4. Certain assumptions are required in making estimates with these models, and for the purposes of these calculations, the simplifying assumption was made that the total exposure could be approximated by two acute exposure episodes, one in 1945 and the other in 1967, some 15 y prior to death. No consideration was given to continuous release of activity to the blood from a deposition in the lung of Class Y material, except in the case of the Healy modification to the Langham model.

Also included in Table 4 are three estimates of total body burden made with the PUQFUA technique developed at Los Alamos (Lawrence 1978, 1983; Voelz et al. 1985). This technique has been continually refined on the basis of experience. The higher estimates were obtained with the two earlier versions of the PUQFUA program, while the most recent version provided an estimated total body content very close to that observed at autopsy.

Deposition estimates made with biokinetic models using the observations of Pu in urine were typically greater than those obtained from the postmortem radiochemical analysis of the body. The original Langham et al. (1950) model provided the highest estimate and was approximately an order of magnitude greater than the autopsy result. The more recent models proposed by Rundo et al. (1976), Leggett (1984), and Leggett and Eckerman (1987) provided estimates approximately equal to the autopsy value. Excellent agreement with the autopsy estimate of systemic deposition was also obtained with the modified Langham equation, $Y_u(t) = 0.002(1 + 0.0008t)t^{-0.74}$, developed by Leggett and Eckerman (1987), which was further adjusted on the basis of limited observations for times beyond 10,000 d. The agreement between the autopsy results (116 Bq) and the estimate from this modified Langham equation (120 Bq) is remarkably close, probably fortuitously so since no consideration was given to continuous release to the blood from Class Y material retained in the lungs.

Although it is inappropriate to draw general conclusions from a single case, it is apparent that at least for this case the more recent biokinetic models provide reasonably good agreement with estimates of deposition based on postmortem tissue analysis for this case. The later models appear to be on the right track, and thus seem to be more suitable for *in-vivo* estimates of total or systemic burden based on urinalysis data.

Comparison of distribution with ICRP models

Perhaps the most widely accepted and used models for the distribution of Pu in humans are those put forth by the International Commission on Radiological Protection (ICRP). These models are dynamic in that there have been several revisions and refinements since the first model was published in 1959 (ICRP 1959). The most recent model is described in ICRP Publication 48 and notes that liver and bone are the principal deposition sites, accounting for about 80% of the Pu reaching the blood stream. ICRP Publication 48 further notes that although there is wide variation in the partitioning of Pu in individual cases, the most likely average initial deposition is 50% in the skeleton and 30% in the liver, with retention half-times of 50 and 20 y for these two organs, respectively (ICRP 1986). The previous ICRP model, described in ICRP Publication 30 (ICRP 1979), proposes initially equal distribution of 45% of the Pu reaching the blood stream to both the skeleton and liver, with retention half-times of 100 and 40 y, respectively. The partitioning of Pu between the skeleton and the liver in this case is more representative of the ICRP Publication 30 model, particularly when viewed in the context of when the exposures were received.

At any time t after deposition, the ratio, r, of activity in the skeleton to that in the liver can be calculated from:

$$r = \frac{f_{sk}e^{-\lambda_{sk}t}}{f_{liv}e^{-\lambda_{liv}t}}, \qquad (2)$$

Table 4. Estimated deposition by various biokinetic models.

Model/Reference	Estimated systemic deposition (Bq)	Comments
Autopsy–this study	116	
Langham (1950)	1000	
Healy (1956)	860	
Durbin (1972)	480	
Rundo et al. (1976)	110	
Parkinson and Henley (1981)	330	
Leggett (1984)	140	
Jones (1985)	210	
Leggett and Eckerman (1987)	220	
Revised Langham/Leggett and Eckerman (1987)	120	
PUQFUA1/Hempelmann et al. (1973); Voelz et al. (1985)	1850	Total body burden; should be compared with equivalent autopsy result of 246 Bq.
PUQFUA2/Voelz et al. (1979, 1985)	999	
PUQFUA3/Voelz et al. (1985)	259	

where f_{sk} and f_{liv} are the fractions initially deposited in the skeleton and liver, respectively, from the transfer compartment, and λ_{sk} and λ_{liv} are the effective net clearance constants for these two organs, respectively. Assuming 90% of the Pu in the tissues other than the lungs and associated lymph nodes came from the earlier exposure and the remaining 10% from the 1963–1970 period, the above equation can be used to compute the expected ratio of Pu in the skeleton to that in the liver using both the ICRP Publication 30 and Publication 48 constants. These data are presented in Table 5 for both the ICRP Publication 30 and Publication 48 models.

Similar partitioning of ^{239}Pu between skeleton and liver has been noted in 43 routine autopsy cases previously reported by the USTR (Kathren et al. 1988). These cases largely were individuals who had incurred chronic inhalation exposures to Pu over a period 10 or more years. Considering factors such as changes in radiation protection procedures, work assignments, and operations over the years, it is likely that these individuals received the bulk of their exposure early in their careers, similar to Case 193 reported in this study.

From the data in this case, and the apparent supporting data of the partial body cases, it appears that the initial partitioning ratio of ^{239}Pu between skeleton and liver is closer to the earlier ICRP Publication 30 value of 1 than the more recent ICRP Publication 48 value of 5/3 or the 7/3 proposed by Thomas et al. (1984). The available evidence is not inconsistent with and indeed supports an initial partitioning ratio between skeleton and liver of less than unity. If the effective organ clearance times reported in ICRP Publication 48 are more correct than those of ICRP Publication 30, then the data for this case and the previously reported partial body cases (Kathren et al. 1988) suggest rather strongly that this ratio must be less than unity; i.e., a smaller amount initially is deposited in the skeleton than in the liver. With time, the amount in the skeleton relative to that in the liver increases because the effective clearance time from the liver is more rapid than that from the skeleton. This is consistent with the observations in this case as well as the partial body results reported by Kathren et al. (1988).

Additional support for an initial partitioning ratio between skeleton and liver in humans of less than unity is derived from experiments in which monkeys were injected parenterally with ^{238}Pu (IV) citrate (Durbin et al. 1985). In these studies, the ratio of the initial distribution of ^{238}Pu between skeleton and liver was 0.47 (six adult monkeys). If this value is used with the ICRP 48 rate constants and the assumptions regarding exposure given above, the calculated ratio of the quantity of Pu in the skeleton relative to that in the liver at the time of death is 0.97, very close to the observed value of 1.05. While this close agreement is likely to be largely fortuitous considering the many unknowns in this case, it does lend support to the idea that the initial partitioning ratio of between skeleton and liver in humans is less than one. Given the observed soft tissue content in USTR Case 193, approximately 15% of the total systemic deposition, a provisional initial deposition from the transfer compartment of 25% to skeleton, 50% to liver, and 25% to other tissues and early excretion seems reasonable for this case. This assumes that the retention half-times in liver and the rest of the soft tissues are about equal and about half that in the skeleton.

CONCLUSIONS

Several preliminary conclusions can be drawn from this analysis of the ^{239}Pu content of an adult male with known occupational exposure to Pu. First, the overall systemic distribution of Pu in the tissues was generally consistent with observations in animals and with the previous limited human exposure data, although the concentrations and content in the soft tissues generally were somewhat greater than predicted by most models.

The relative amounts of ^{239}Pu in the skeleton and liver were approximately equal, but given the shorter effective clearance time for this nuclide in the liver and the long period between the indicated exposure and death of this case, this observation is consistent with a smaller initial deposition in the skeleton than in liver. Based on the observation in this whole-body case, plus previous observations in 43 partial-body cases (Kathren et al. 1988) and experiments in monkeys performed with ^{238}Pu (Durbin et al. 1985), a tentative initial partitioning of 25% to the skeleton, 50% to the liver, and 25% to the rest of the body and early excretion is suggested for the Pu reaching the transfer compartment.

Estimates of systemic and total body burden made with earlier biokinetic models yielded values significantly greater than measured in the tissues at autopsy. The modified Langham equation recently proposed by Leggett and Eckerman (1987) and models proposed by Leggett (1984) and Rundo et al. (1976) gave values virtually identical

Table 5. Calculated and observed ratios of Pu in skeleton and liver.

	Initial deposition fraction		Effective clearance constant (y^{-1})		Pu in skeleton
Model/Reference	Skeleton	Liver	Skeleton	Liver	Pu in liver
This case (observed)					1.05
ICRP Publication 30	0.45	0.45	0.00693	0.01733	1.44
ICRP Publication 48	0.50	0.30	0.01386	0.03465	3.47

with the radioassay results, as did the PUQFUA3 program (Lawrence 1983) for estimating total body burden.

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Note in proof—It has come to our attention that low temperature firing of the nitric acid digests of bone does not always provide an accurate measure of the mineral content of a bone specimen (tends to overestimate the bone mineral content). The bone samples from this individual and subsequent whole bodies analyzed for the U.S. Transuranium Registry are being submitted for calcium analyses, the results of which will be reported separately.

General note to Tables in Appendices A and B: Slight differences between various comparable values may be attributable to rounding, or to slight changes in weight resulting from minor liquid or evaporative losses or bone dust lost in cutting.

APPENDIX A

Tables of data from the analysis of individual soft tissues and organs.

Table A-1. Plutonium-239 content, wet weight, and ^{239}Pu concentration of organs and specialized tissues.

Tissue	^{239}Pu content[a] (mBq ± 1 $\hat{\sigma}_D$)	Wet weight (g)	^{239}Pu concentration (mBq · g^{-1} wet weight)
Lung - left, sup. lobe	23924 ± 883	273	87.6
- left, inf. lobe	21510 ± 708	278	77.4
- right	41383 ± 2017	752	55.0
Trachea	74 ± 7	17	4.23
Lymph nodes - TBLN-1	5632 ± 181	1.8	3218
- TBLN-2	3573 ± 137	2.6	1390
- TBLN-3	29872 ± 931	5.0	5998
- TBLN-4	4015 ± 1000	7.2	558
Liver	48633 ± 2833	1863	26.1
Kidneys	86 ± 3	326	0.26
Spleen	1461 ± 59	258	5.65
Heart	76 ± 6	428	0.18
Pericardium	2250 ± 98	120	18.7
Stomach	460 ± 17	387	1.19
Intestine	108 ± 6	1459	0.07
Urinary bladder	26 ± 2	127	0.20
Brain	175 ± 8	1303	0.13
Testes	11 ± 1	14	0.78
Prostate	99 ± 4	24	4.09
Adrenals	23 ± 2	28	0.81
Pancreas	220 ± 17	131	1.70
Bile	8 ± 2	16	0.46
Gall bladder	106 ± 4	26	4.01
Thyroid	11 ± 1	13	0.84
Pituitary	12 ± 1	0.5	21.3

[a] $\hat{\sigma}_D$ is the 67% confidence interval of the sample count due to errors and uncertainties in counter backgrounds and counter efficiencies for measuring the sample and in the calibration of the ^{242}Pu tracer, as described by McInroy et al. 1979.

Table A-2. Dissected wet weights and ^{239}Pu contents and concentration in muscle removed in the dissection of the skeleton.

Tissue	^{239}Pu content (mBq ± 1 $\hat{\sigma}_D$)	Wet weight (g)	^{239}Pu concentration (mBq g^{-1} wet weight)
Combined soft tissue[a]			
Anatomical region			
Head	44 ± 27	167	0.27
Tongue	17 ± 2	90	0.18
Shoulder - left	83 ± 4	800	0.10
- right	55 ± 4	607	0.09
Arm, upper - left	85 ± 4	1047	0.08
- right	171 ± 8	996	0.17
lower - left	95 ± 5	647	0.15
- right	34 ± 3	615	0.06
Hand - left	50 ± 3	126	0.40
- right	17 ± 2	144	0.12
Thorax (1)	694 ± 23	1053	0.66
(2)	70 ± 5	1099	0.06
(3)	152 ± 7	1314	0.12
(4)	468 ± 17	889	0.53
Abdomen (1)	100 ± 6	1255	0.08
(2)	76 ± 8	1490	0.05
(3)	378 ± 17	2830	0.13
(4)	56 ± 4	199	0.28
Thigh - left	252 ± 11	2696	0.09
- right	273 ± 15	3160	0.09
Calf - left	152 ± 7	1284	0.12
- right	101 ± 7	1246	0.08
Foot - left	38 ± 3	174	0.22
- right	57 ± 3	297	0.19
Ears	8 ± 1	45	0.18
Eyes[b]	102 ± 4	14	7.29
Spinal Cord	6 ± 1	29	0.19
Nails - hand - right	1 ± 0.4	1.2	0.83
- foot - right (3)	2 ± 0.6	0.5	4.63
Hair	2 ± 0.5	21	0.10

[a]Combined skeletal muscle, connective tissue, blood vessels, tendons, ligaments.

[b]Fluid was lost from right eye.

Table A-3. Dissected wet weights and ^{239}Pu contents and concentration of skin removed in the dissection of the skeleton.

Tissue	^{239}Pu content (mBq ± 1 $\hat{\sigma}_D$)	Wet weight (g)	^{239}Pu concentration (mBq g^{-1} wet weight)
Anatomical region			
Head	114 ± 6	875	0.13
Arm, upper - left	454 ± 15	904	0.50
- right	54 ± 4	870	0.06
lower - left	36 ± 3	279	0.13
- right	30 ± 2	279	0.11
Hand - left	722 ± 23	127	5.69
- right	24 ± 2	133	0.18
Thorax (1)	60 ± 4	1190	0.01
(2)	385 ± 14	1605	0.24
(3)	66 ± 5	1298	0.05
(4)	48 ± 3	937	0.05
Abdomen (1)	32 ± 3	1073	0.03
(2)	30 ± 3	908	0.03
(3)	58 ± 5	1542	0.04
(4)	33 ± 3	1082	0.03
Thigh - left	116 ± 5	2567	0.04
- right	77 ± 5	1475	0.05
Calf - left	87 ± 4	648	0.13
- right	111 ± 6	1300	0.08
Foot - left	48 ± 3	342	0.14
- right	29 ± 2	254	0.11

APPENDIX B

Tables of data from the analysis of bones and parts of bones.

Table B-1. Weights and ^{239}Pu contents of bones of the head.

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq ± 1 $\hat{\sigma}_D$)	(mBq g^{-1} ash)
Cranium	804			
Right half	387			
Left half	404			
Occipital	51	30.0	462 ± 19	15.4
Parietal-1	76	48.6	772 ± 32	15.9
Parietal-2	35	21.9	338 ± 14	15.4
Frontal-1	39	21.9	360 ± 17	16.4
Frontal-2	12	7.6	106 ± 5	13.9
Frontal-3	20	10.8	192 ± 9	17.8
Temporal-1	20	10.9	182 ± 9	16.7
Temporal-2	41	18.4	348 ± 16	18.9
Temporal-3	26	7.8	227 ± 8	29.1
Maxilla	24	9.5	246 ± 11	25.9
Mandible	83[a]			
Right	41			
Left	29	15.2	180 ± 8	11.8
Hyoid bone	2.9	0.57	21 ± 2	35.0
Teeth				
Lower right				
Incisor-1	0.64	0.49	5.2 ± 0.9	10.6
Incisor-2	0.79	0.32	4.9 ± 0.8	15.3
Canine	1.17	0.88	7.3 ± 1.0	8.3
Lower left				
Incisor-1	0.62	0.48	4.5 ± 0.8	9.4
Incisor-2	0.77	0.38	5.4 ± 0.8	14.2
Canine	1.30	0.82	7.9 ± 1.0	9.6

[a]Includes teeth.

Table B-2. Weights and ^{239}Pu content of bones and parts of bones of the spine and pelvis.

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq ± 1 $\dot{\sigma}_D$)	(mBq g^{-1} ash)
Vertebral column				
Cervical-1	21	7.44	183 ± 7	24.6
Cervical-2	26			
Cervical-3	17			
arch	8.8	2.56	75 ± 4	29.2
body	7.8	1.60	54 ± 3	33.8
Cervical-4	18			
Cervical-5	17			
arch	8.2	2.62	61 ± 3	23.3
body	8.5	1.92	57 ± 3	29.9
Cervical-6	20			
Cervical-7	23			
arch	13.1	3.92	96 ± 4	24.4
body	9.5	2.08	64 ± 3	30.6
Thoracic-1	32			
arch	19.5	5.66	145 ± 6	25.6
body	11.8	2.08	80 ± 5	38.3
Thoracic-2	29			
Thoracic-3	27			
arch	14.3	4.16	99 ± 4	23.8
body	12.5	1.91	70 ± 4	36.8
Thoracic-4	28			
Thoracic-5	30			
arch	14.8	3.58	101 ± 6	28.2
body	14.7	2.51	95 ± 4	37.9
Thoracic-6	34			
Thoracic-7	39			
arch	15.9	4.36	122 ± 5	28.0
body	20.5	2.45	100 ± 5	40.8
Thoracic-8	39			
Thoracic-9	43			
arch	20.7	4.98	145 ± 6	29.1
body	22.3	3.92	160 ± 6	40.8
Thoracic-10	57			
Thoracic-11	61			
arch	22.2	5.61	130 ± 5	23.1
body	37.7	5.48	200 ± 8	36.5
Thoracic-12	67			

Table B-2. (*Contd.*)

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq ± 1 $\hat{\sigma}_D$)	(mBq g^{-1} ash)
Vertebral column (cont'd)				
Lumbar-1	83			
arch	29.8	7.42	206 ± 8	27.6
body	49.8	6.46	259 ± 9	42.6
Lumbar-2	82			
Lumbar-3	98			
arch	35.3	9.56	308 ± 12	34.2
body	61.7	6.72	285 ± 11	42.4
Lumbar-4	98			
Lumbar-5	88			
arch	34.9	10.26	233 ± 10	22.7
body	52.1	8.77	273 ± 12	31.2
Sacrum	311	44.08	1554 ± 47	35.2
Coccyx		3.44	141 ± 9	41.0
Pelvis				
Left	448			
Right	452			
Ischium	232	56.84	1357 ± 43	23.9
Illium	214			
Chest	85	19.16	556 ± 19	29.0
Body	127	40.16	920 ± 37	22.9

Table B-3. Weights and ^{239}Pu contents of the analyzed bones and parts of bones of the shoulder and rib cage.

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq ± 1 $\hat{\sigma}_D$)	(mBq g^{-1} ash)
Clavicle				
Left	48.6			
Right	48.0			
Sternal end (SE)	12.0	2.66	55 ± 3	21.1
Shaft	20.5	9.88	134 ± 2	13.6
Acromial end	14.6	2.67	84 ± 4	31.7
Scapula				
Left	135.5			
Right	126.4			
Proximal end (PE)		12.5	148 ± 10	11.8
Spine		21.9	411 ± 16	18.8
Distal end (DE)		4.8	216 ± 9	45.0
Ribs				
1-Left	18.3	4.7	85 ± 4	18.0
Right	17.6			
Sternal end (SE)		0.74	37 ± 2	50.0
Middle shaft (MS)		1.58	32 ± 2	20.3
Vertebral end (VE)		1.06	20 ± 2	18.9
2-Left	16.0	5.1	92 ± 4	18.0
Right	16.8			
Sternal end		1.51	31 ± 1	20.5
Middle shaft		1.56	30 ± 2	19.2
Vertebral end		0.95	24 ± 2	25.3
3-Left	14.9	4.6	60 ± 3	13.0
Right	15.7			
Sternal end		1.22	28 ± 2	23.0
Middle shaft		1.68	34 ± 2	20.2
Vertebral end		1.74	39 ± 3	22.4
4-Left	21.9	6.0	109 ± 4	18.2
Right	23.7			
Sternal end		1.40	43 ± 3	30.7
Middle shaft		2.16	53 ± 3	24.5
Vertebral end		2.34	62 ± 3	26.5
5-Left	28.8	6.3	131 ± 5	20.8
Right	22.8			
Sternal end		1.56	51 ± 3	32.7
Middle shaft		2.42	59 ± 3	24.4
Vertebral end		3.36	71 ± 4	21.1

Table B-3. (*Contd.*)

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq ± 1 $\hat{\sigma}_D$)	(mBq g^{-1} ash)
6-Left	38.5	10.3	190 ± 7	18.5
Right	38.5			
Sternal end		2.48	58 ± 3	23.4
Middle shaft		3.40	84 ± 5	24.7
Vertebral end		3.92	92 ± 4	23.5
7-Left	37.1			
Right	33.0			
Sternal end		2.28	94 ± 5	41.2
Middle shaft		2.94	62 ± 3	21.1
Vertebral end		5.08	98 ± 4	19.3
8-Left	32.3	8.0	155 ± 6	19.4
Right	29.3			
Sternal end		2.23	246 ± 6	110.3
Middle shaft		3.48	56 ± 3	16.1
Vertebral end		3.98	78 ± 2	24.2
9-Left	30.9	8.0	156 ± 6	19.4
Right	24.2			
Sternal end		0.92	18 ± 2	19.6
Middle shaft		2.36	57 ± 3	24.2
Vertebral end		3.36	85 ± 4	25.1
10-Left	21.7	7.5	118 ± 5	15.7
Right	23.1			
Sternal end		0.78	27 ± 2	34.6
Middle shaft		2.56	40 ± 2	15.6
Vertebral end		2.86	59 ± 2	20.6
11-Left	13.3	4.0	59 ± 3	14.1
Right	12.9			
Sternal end		0.56	13 ± 1	23.2
Middle shaft		1.52	23 ± 2	15.1
Vertebral end		1.60	31 ± 2	19.4
12-Left	7.7	2.2	31 ± 2	13.9
Right	6.4			
Sternal end		0.28	6 ± 1	21.4
Middle shaft		0.88	16 ± 1	18.2
Vertebral end		0.86	24 ± 2	27.9
Costal cartilages				
Sternum	60.6	3.04	52 ± 3	17.1
Ribs				
Left	22.7			
Right	20.5	1.97	42 ± 2	21.3
Sternum	96.4	14.57	454 ± 8	31.2

Table B-4. Weights and ^{239}Pu contents of bones and parts of bones of the arms and hands.

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq $\pm$ 1 σ_D)	(mBq g^{-1} ash)
Humerus				
Left	260.6			
Right	252.8			
Proximal end (PE)	80.8	15.1	501 ± 19	33.2
Proximal shaft (PS)	122.3	27.8	378 ± 17	13.6
Distal shaft (DS)		28.6	280 ± 12	9.8
Distal end (DE)	53.1	16.6	366 ± 13	22.0
Radius				
Left	70.8			
Right	72.1			
Proximal end	8.6	2.7	51 ± 1.4	18.9
Proximal shaft	43.5	12.7	102 ± 4.7	8.0
Distal shaft		9.6	55 ± 3.1	5.7
Distal end	18.3	4.6	86 ± 4.2	18.7
Ulna				
Left	86.3			
Right	84.0			
Proximal end	34.5	9.55	194 ± 7.6	20.3
Proximal shaft	43.7	43.5	101 ± 5.3	
Distal shaft			56 ± 3.2	
Distal end	6.7	1.39	35 ± 2.4	25.2
Carpals				
Left	28.1			
Right	28.0			
Scaphoid		1.3	34 ± 2.3	26.2
Lunate		0.6	19 ± 1.6	31.7
Triangular		1.0	28 ± 2.3	28.0
Pisiform		0.2	9 ± 1.1	45.0
Hamate		1.4	40 ± 2.5	28.6
Capitate		1.1	33 ± 2.2	30.0
Trapezoideum		0.5	15 ± 1.9	30.0
Trapezoium		0.8	23 ± 1.8	28.8

Table B-4. (*Contd.*)

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq $\pm$ 1 σ_D)	(mBq g^{-1} ash)
Metacarpals				
Left	42.5	13.2		
Right	43.7			
1		2.0	50 ± 3	25.0
2		3.4	69 ± 4	20.3
3		3.5	70 ± 3	20.0
4		1.9	47 ± 3	24.7
5		1.7	33 ± 2	19.4
Phalanges				
Left	37.3			
Right	35.5			
1-Proximal (P-1)		1.2	29 ± 2	24.2
Distal (D-1)		0.5	17 ± 1	34.0
2-Proximal (P-2)		1.9	29 ± 2	15.3
Middle (M-2)		0.6	14 ± 1	23.3
Distal (D-2)		0.2	7 ± 1	35.0
3-Proximal (P-3)		2.1	39 ± 2	18.6
Middle (M-3)		0.7	16 ± 2	22.9
Distal (D-3)		0.2	9 ± 1	18.0
4-Proximal (P-4)		1.6	31 ± 2	19.4
Middle (M-4)		0.9	14 ± 2	15.6
Distal (D-4)		0.2	10 ± 1	50.0
5-Proximal (P-5)		0.9	21 ± 2	23.3
Middle (M-5)		0.4	8 ± 1	20.0
Distal (D-5)		0.1	6 ± 1	60.0

Table B-5. Weights and ^{239}Pu contents of bones and parts of bones of the legs and feet.

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq ± 1 $\dot{\sigma}_D$)	(mBq g^{-1} ash)
Femur				
Left	800			
Right	908			
Proximal end (PE)	220	55.0	1469 ± 78	26.7
Proximal shaft (PS)	356	67.2	702 ± 32	10.4
Middle shaft (MS)		55.8	599 ± 26	10.7
Distal shaft (DS)		49.0	497 ± 22	10.1
Distal end (DE)	218	47.8	1118 ± 108	23.4
Tibia				
Left	446			
Right	442			
Proximal end	162	36.9	898 ± 84	24.3
Proximal shaft	222	58.4	605 ± 27	10.4
Distal shaft		49.4	441 ± 20	8.9
Distal end	53	14.9	307 ± 12	20.6
Fibula				
Left	83			
Right	76			
Proximal end	14.6	3.2	81 ± 4	25.3
Proximal shaft	48	15.3	140 ± 10	9.2
Distal shaft		11.9	73 ± 4	6.1
Distal end	16	5.1	89 ± 4	17.5
Patella				
Left	38			
Right	37	8.8	183 ± 7	20.8
Tarsals				
Left	194			
Right	193			
Talus		15.1	304 ± 11	20.1
Calcaneus		22.0	517 ± 17	23.5
Cuboid		3.4	94 ± 4	27.6
Navicular		4.7	105 ± 5	22.3
Cunieform, med		4.0	95 ± 4	23.8
int.		1.5	44 ± 3	29.3
lat.		1.7	44 ± 3	25.9

Table B-5. (*Contd.*)

	Weight (g)		^{239}Pu content	
	Wet	Ash	(mBq ± 1 $\hat{\sigma}_D$)	(mBq g^{-1} ash)
Metatarsals				
Left	61			
Right	62			
1		6.9	142 ± 6	20.6
2		4.2	76 ± 4	18.1
3		3.3	64 ± 3	19.4
4		3.3	64 ± 3	19.4
5		3.4	58 ± 3	17.1
Phalanges				
Left	23			
Right	24			
P-1		2.3	47 ± 3	20.4
D-1		0.9	28 ± 2	31.1
P-2		0.4	14 ± 1	35.0
M-2		0.5	7 ± 1	14.0
D-2		0.1	3 ± 1	30.0
P-3		0.4	14 ± 1	35.0
M-3		0.2	5 ± 1	25.0
D-3		0.1	6 ± 1	60.0
P-4		0.3	12 ± 1	40.0
M-4		0.1	5 ± 1	50.0
D-4		0.1	4 ± 1	40.0
P-5		0.4	11 ± 1	27.5
M-5		0.1	3 ± 1	30.0
D-5		0.1	3 ± 1	30.0

Table B-6. Wet and ash weights and ^{239}Pu content and concentration of bones and parts of bones of the entire skeleton, determined by method of McInroy et al. 1985.

Skeletal part	Weight (g)		^{239}Pu content	
	Wet[a]	Ash	(mBq)	(mBq g^{-1} ash)[b]
Skull				
Cranial bones				
Occipital	114.7	58.8	904	15.4
Parietal-1,2	248.8	138.0	2173	15.8
Temporal-1,2	135.9	57.2	1038	18.1
Frontal-1	86.7	42.9	715	16.4
Facial bones				
Maxilla	53.6	18.7	476	25.5
Frontal-2,3	71.3	36.1	582	16.1
Temporal-3	59.3	15.2	444	29.1
Mandible	77.6	36.3	432	11.9
Hyoid bone	2.9	0.57	21	36.8
Vertebral column				
Cervical (1-7) arches	84.1	26.5	680	25.7
bodies	56.5	12.1	385	31.7
Thoracic (1-12) arches	218.4	57.2	1502	26.3
bodies	261.4	39.5	1531	38.8
Lumbar (1-5) arches	166.7	45.4	1301	28.7
bodies	277.7	36.2	1387	38.3
Sacrum	237.0	44.1	1554	35.3
Coccyx	19.2	3.4	141	41.0
Pelvis				
Illium	423.2	118.2	2943	24.9
Ischium	462.9	113.3	2706	23.9
Clavicles				
Sternal end	24.2	5.4	113	21.1
Shaft	41.3	19.9	270	13.6
Acromial end	29.4	5.4	168	31.3
Scapulae				
Proximal end	74.0	24.1	417	17.3
Spine	130.1	42.3	794	18.8
Distal end	48.5	9.4	269	28.6

Table B-6. (*Contd.*)

Skeletal part	Weight (g)		^{239}Pu content	
	Wet[a]	Ash	(mBq)	(mBq g^{-1} ash)[b]
Ribs (1-12)	519.2	143.0	332	23.2
Costal cartilages	103.8	7.2	140	18.8
Sternum	96.4	14.6	454	31.2
Humerii				
Proximal end	159.2	29.8	985	33.0
Shaft	240.9	111.2	1297	11.7
Distal end	104.6	32.7	722	22.1
Radii				
Proximal end	17.4	5.5	103	19.0
Shaft	87.8	44.9	634	7.1
Distal end	36.9	9.3	173	18.6
Ulnae				
Proximal end	68.1	18.8	380	20.2
Shaft	86.2	42.7	310	7.3
Distal end	13.2	2.7	69	25.3
Hand bones				
Carpals	57.9	13.9	400	28.7
Metacarpals	86.1	26.7	545	20.4
Phalanges	74.2	22.5	483	21.4
Femora				
Proximal end	468.9	117.5	3139	26.7
Shaft	759.2	367.1	3836	10.5
Distal end	466.3	102.1	2384	23.4
Tibiae				
Proximal end	322.6	73.4	2888	24.4
Shaft	442.9	214.5	2101	9.8
Distal end	105.3	29.6	611	20.6
Fibulae				
Proximal end	28.0	6.2	154	24.8
Shaft	92.7	52.3	409	7.8
Distal end	29.7	9.8	170	17.3
Patellae	75.0	17.5	363	20.8
Foot bones				
Tarsals	387.3	104.5	2395	22.9
Metatarsals	123.6	42.8	814	19.0
Phalanges	46.7	12.0	335	27.8
Entire skeleton	8691	2681	51,138	19.1

[a]When wet weight is shown only for whole bone, subdivision was made at Los Alamos after a variable amount of drying had occurred.

[b]When replicated samples (e.g., arches of thoracic vertebrae (1-12) have been combined, the ^{239}Pu concentration shown is the grand average, Σ ^{239}Pu content/Σ ash weight, summed over all samples.