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HUMAN DATA DEMONSTRATING EXTRA LONG RETENTION OF PLUTONIUM IN THE LUNG

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Abstract--Case histories are presented of 10 humans with inhalation depositions of Pu in which the Pu is retained in the lung or pulmonary lymph system for extremely long periods. The retention half-times range from 1000 to at least 20,000 d for most (in some cases all) of the initial deposition. In no case was a clearance half-time in the hundreds of days observed. form of Pu involved in these cases is believed to be calcined Pu oxide.

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INTRODUCTION

This paper presents 10 case histories involving Hanford Site workers who received internal depositions via inhalation of Pu and whose cases demonstrate retention in the lung remarkably different from the retention predicted using the inhalation classes and lung model defined in Publication 30 of the International Commission on Radiological Protection (ICRP 1979). Specifically, the Pu is retained in the lung much more tenaciously than expected for class Y material described by the ICRP 30 lung model; hence, the Pu is referred to in this paper as "super" class Y Pu.

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The cases examined in this paper are merely those that have recently come to light and for which the intake histories are fairly straightforward. Other cases having similar characteristics but involving multiple intakes of various forms of Pu were rejected. A survey of all historical cases at Hanford was not done, so the cases examined are not meant to be all inclusive.

Based on information in the case histories and from general knowledge of Pu processing at Hanford, the authors believe that the form of Pu involved is Pu oxide that has been heated, probably to at least 500°C. In all cases, Pu oxide was indicated as the chemical form at the time of the intake. In four cases there is some uncertainty associated with the Pu oxide determination, because either the details of the intake scenario are scanty or, where better known, they show that other forms of Pu were present in the same operating area, such as a rupture in a glovebox in which metal is being converted to oxide.

The limitations of the case data and the case histories are described in the following subsections. The complete set of bioassay measurements is in the Appendix. The behavior of super class Y Pu in the lung is also discussed and some related studies that also indicate the existence of super class Y Pu are summarized.

LIMITATIONS OF THE DATA

The characteristics of super class Y Pu, as presented in the fourth section of this paper, were based on an overall perspective from all 10 cases. In no one case were the data sufficiently complete and certain to provide a complete picture of the behavior of Pu in and the removal of Pu from the lung.

The data span several decades and, therefore, reflect changing bioassay measurement techniques and associated detection levels. This limitation is most evident concerning chest counting for ^{241}Am; chest counting was nonexistent or crude at the time of intake for several of the cases. Also, the data were obtained from a program designed to demonstrate compliance with regulations applicable at the time. Decisions on type and frequency of bioassay measurements were made by various dosimetrists over the years, reflecting different compliance monitoring philosophies. And, of course, the data reflect limitations in dealing with human subjects. For instance, for one reason or another the worker did not always provide the samples he was requested to provide, and some workers changed jobs or retired before complete sets of data were obtained.

The medical histories of the 10 persons were reviewed to determine if the lung retention characteristics could be due to pulmonary disfunction. However, the cases represented a mixture of ages at intake, smoking habits, and respiratory health; and no pattern of pulmonary disfunction was observed.

For interpretation of the chest count data, it was assumed that ^{241}Am was a valid tracer for the behavior of Pu in the lung. The basis for this assumption was that the Am was buried as individual atoms in the Pu host matrix, having ingrown from ^{241}Pu (Stradling and Stather 1989).

DISCUSSION OF CASES

Some aspects of each case are summarized in Table 1 and are discussed in the following subsections. The case numbers were chosen at random and do not reflect a ranking by any particular parameter. The biological clearance rates

from the lung are approximate, sometimes because appropriate bioassay data shortly after the intake were lacking and often because important isotopic ratios, such as ^{241}Pu to ^{241}Am and ^{241}Pu to ^{239}Pu, were not obtained at intake.

HAN01

HAN01 had the most complete set of data. This case was first presented at the Snowbird Actinide Workshop in 1979 (Spitz and Robinson 1981), but considerably more data have been collected since then. Figure 1a shows the lung burden of ^{241}Am. Although primarily an intake of Pu, there was enough ^{241}Am to track the burden from the very first day. In this case, initial isotopic ratios were determined from samples of the material, so the ingrowth curve shown in Figure 1a provided a reasonably accurate biological clearance rate of 0.00011 d^{-1} (half-time of about 6,000 d). Figure 1a clearly shows that there is only a single clearance rate from the regions measured by the detectors (tracheal-bronchial, pulmonary, and pulmonary and tracheobronchial lymph nodes) starting even from the day of intake. (Recent measurements on the tracheobronchial lymph nodes did not detect any activity [Northcutt et al. 1988].)

Chelation therapy with diethylenetri-aminepentaccetic acid (DTPA) was given 5 times in the first 10 d; even so, only about 100 mBq (3 pCi) of Pu alpha activity was excreted via urine in the first 10 d. After the first 10 d, daily urinary excretion stayed below the detection level of about 0.8 mBq (0.02 pCi) until late 1983, when the detection level was decreased to 0.3 mBq (0.009 pCi) for ^{239}Pu. Since then, daily urinary excretion of ^{239}Pu

has fluctuated around the detection level. Total fecal excretion for the first 5 d after intake was about 100 Bq (3 nCi) of ^{239}Pu, but 99% of this occurred in the first voiding, indicating that most of this was clearance of large particles from upper respiratory passages or resulted from direct ingestion. Fecal samples were also obtained at 379, 609, and 2,261 d post intake. Only the first sample had slight activity above the detection level of 3 mBq (0.09 pCi). These values support the biological clearance rate determined from chest counting and do not support the ICRP 30 (1979) clearance half-time of 500 d.

HAN02

The first bioassay measurements for HAN02 were obtained about 24 h after the intake. No activity was detected in the first chest count (Figure 1b). The ^{241}Am detection level was not recorded at the time but is estimated to have been about 10 Bq (0.3 nCi). A single-void urine sample was collected at the same time as the chest count, and a 24-hour sample was collected 8 d after the intake. The first sample was analyzed by rush techniques (detection level of about 8 mBq [0.2 pCi]) and the second sample was analyzed normally. Neither sample contained detectable Pu alpha activity. No fecal samples were collected. Based on this information, it was initially assumed that an intake had not occurred.

The next chest count was not obtained until 2.5 y later; ^{241}Am was detected at that time, and all subsequent chest and head counts have shown a pattern of increasing ^{241}Am in the lung (Figure 1b). The ^{241}Pu-to-^{241}Am ratio at the time of intake was not measured, so the biological clearance half-time

of the Pu was estimated at >20,000 d, based on a general knowledge of the material and approximate curve-fitting. Despite the uncertainty in the calculation, a clearance half-time anywhere near 500 d is not appropriate to this case.

Twenty-four-hour urine samples have been collected at least annually since the intake. None have contained detectable Pu alpha activity.

HANO3

HANO3 was involved in two incidents in 1964, either or both of which could have resulted in an intake. HANO3 worked with or around Pu without further incident until 1967 and has not been a radiation worker since then.

The first chest count was performed on the worker in 1987 (Figure 1c). It and two confirmatory counts indicated an activity about 32 Bq (0.86 nCi) of ^{241}Am. Skeleton counts indicated no detectable activity, implying that the chest activity is in the lung. An interview with the worker and a complete review of all relevant radiation records indicated that the activity must have resulted from one or both of the incidents. Because the original ^{241}Pu-to-^{241}Am ratio is not known and because the data are too few to discriminate among various curves, it can only be deduced that the biological clearance half-time is very long. The ingrowth curve shown in Figure 1c is based on standard isotopic ratios used for the facility based on general experience.

None of the two urine samples collected shortly after the first incident and the five samples collected shortly after the second incident had detectable Pu. Only 2 of 21 subsequent urine samples collected between 1964

and 1975 had detectable Pu alpha activity, and they were probably false positives. No fecal samples were collected.

HAN04

In 1970 the worker was involved in an incident that was the most probable source of the intake, although HAN04 was also involved in several earlier incidents, each with potential for intake. Fecal data obtained after the 1970 incident confirmed an intake, whereas no fecal samples were obtained after the earlier incidents. Also, a chest count taken 2 months prior to the 1970 incident did not detect any activity, whereas all chest counts taken since the incident have shown activity in the chest (Figure 1d). The 1970 incident involved Pu oxide powder with a Pu alpha-to-^{241}Am ratio of 20:1.

The chest counts show a pattern of slightly increasing ^{241}Am activity, although the pattern is a bit rough because of changes in the state of the art of chest counting over the years. There is no measurable activity in the skeleton, so the chest activity is assumed to be in the lung. The ^{241}Pu-to-^{241}Am ratio at intake is not known, so it was approximated based on general knowledge of the facility. The ingrowth curve shown in Figure 1d indicates a biological clearance half-time on the order of 10,000 to 15,000 d. Chest counts starting on the day after the intake through the next 4 months gave no indication of any clearance from the lung, i.e., no early clearance.

Urine samples were taken from 1970 through 1971 and from 1981 through 1987. Only 2 out of 20 samples had detectable Pu alpha activity; these activities were not confirmed by prompt follow-up samples and were assumed to be false positives. Three fecal samples, collected on days 1, 2, and 3 after

the intake, contained a total of 6.8 Bq (0.18 nCi) of Pu alpha activity (with 93% of that occurring in the first sample); this early fecal clearance represents only about 4% of the estimated initial deposition in the lung. Three additional fecal samples, obtained at about 120 d post intake, indicated a clearance half-time via the gastrointestinal (GI) tract of about 12,000 d, which substantiated the clearance rate from chest counts and provided further evidence against a clearance half-time of around 500 d.

HAN05

A chest count performed on the day of the intake did not detect ^{241}Am. The next chest count, performed about 2.5 y later, and 10 out of 12 subsequent chest counts did detect ^{241}Am as shown in Figure 1e. No activity has been detected in the skeleton, so the activity is assumed to be in the lung. The ratio of ^{241}Pu to ^{241}Am was not determined directly but was estimated from general knowledge of the material and from the measured ratio of ^{239}Pu to $^{238}Pu+^{241}Am$ of 7:1, obtained on an air sample. The ingrowth curve shown in Figure 1e indicates a clearance half-time from the lung of about 6,000 d. No urine samples have had detectable Pu. The first urine sample was obtained 6 d after the intake, so a small early systemic clearance component could have been missed. No fecal samples were collected.

HAN06

This is the only case of the 10 that possibly shows two clearance rates. The date listed for this intake (3/7/66) is considered the most probable of

several possible dates in the period 1963 to 1966. Three out of five urine samples taken shortly after the 3/7/66 incident gave Pu alpha results that at the time were not considered positive but in retrospect are considered barely detectable. These three samples appear to support a small component with a systemic clearance half-time from the lung of about 50 d. No other urine samples taken through 1977 had detectable Pu alpha activity. Urine sampling was restarted in 1986 and most samples have shown low activity of ^{239}Pu. These sample results point to a slowly clearing component from the lung, with a systemic clearance half-time of about 5,000 d.

The worker transferred out of Pu work before chest counting was started at Hanford, and consequently HAN06 did not receive a chest count until 1987. That and all subsequent chest counts confirmed ^{241}Am activity in the chest (Figure 1f). No activity has been detected in the skeleton, so the activity is assumed to be in the lung. The initial isotopic ratios could only be estimated, so the ingrowth curve in Figure 1f is very approximate, but it is consistent with the clearance half-time observed through urine sampling. No fecal samples were collected.

HAN07

The intake date, 1969, is assumed based on an interview with the worker and because it is the only known incident involving this worker. The intake was not confirmed by urine samples taken at the time, and, in fact, Pu activity has not been detected in any urine sample to date.

Regular chest counts on this worker began in 1975 (Figure 1g). Chest counts since 1984 have consistently revealed ^{241}Am activity. No activity has

been detected in the skeleton. The isotopic ratios of the Pu involved in the incident were well documented, so the ingrowth curve shown in Figure 1h is a better estimate than for some of the other cases. The biological clearance half-time is calculated to be about 17,000 d.

HAN08

Of two possible intake dates, 1965 and 1970, the latter seems most likely. Both incidents involved Pu oxide. One of three urine samples taken shortly after the latter incident had barely detectable Pu alpha activity. All subsequent urine samples except one did not have detectable activity. The first chest count occurred in 1973 (Figure 1h). All chest counts from 1984 until 1986 (when the worker retired) indicated ^{241}Am activity. A skeleton count was not performed, but the lack of urinary Pu probably means little or no skeletal activity, i.e., the observed in vivo activity is in the lung. Nothing is known about isotopic ratios, and the lack of in vivo data from late 1970 to 1973 means that about all that can be said about this case is that the Pu presently in the chest has been there a long time.

HAN09

The intake for HAN09 resulted from the same incident in which HAN02 was involved in 1979. A next-day chest count did not detect any activity; nor did any chest counts until 1986 (Figure 1i). All subsequent chest counts have revealed ^{241}Am. Skeleton counts have not shown detectable activity, so the activity is assumed to be in the lung. Seventeen out of twenty urine samples

taken from just after the incident until 1990 have not had detectable Pu alpha activity. The estimated clearance half-time from the lung is about 20,000 d.

HAN10

The HAN10 intake resulted from an incident in 1969. The chest count on the third day after the intake was negative, as were all subsequent counts until 1986 (Figure 1j), when two successive, extra long counts revealed ^{241}Am. A skeleton count did not show any activity. The urine data generally show no detectable excretion, although the results for samples just after the intake are ambiguous. A single fecal sample was obtained at 2 d post intake, which contained 9.3 Bq (0.25 nCi) of Pu alpha (compared with an initial pulmonary deposition of about 20 Bq [0.6 nCi]).

Isotopic ratios of the inhaled material are not known. Based on assumed isotopic ratios from general knowledge of the facility, a clearance half-time from the lung greater than 20,000 d was estimated.

GENERAL DESCRIPTION OF THE BEHAVIOR OF SUPER CLASS Y PLUTONIUM IN THE LUNG

When the 10 cases are reviewed as a whole, they appear to provide the following information as compared with the ICRP 30 lung model for class Y material:

- Clearance from the lung to the blood shortly after an intake is much less than predicted. In fact, the rapid clearance may be so little that, from the perspective of applied internal dosimetry, Pu might not

be detectable in urine samples using state-of-the-art radiochemical techniques. This seems reasonable because material that is highly insoluble in the pulmonary region should also be highly insoluble in the upper respiratory regions.

- Gradual clearance to the blood, presumably from the pulmonary region and from the lymph system, is extremely slow. Observed clearance half-times were at least an order of magnitude longer than the nominal 500-d ICRP 30 class Y value.

- Gradual clearance from the lung to the GI tract was not accurately determined because in only two cases were fecal samples collected during the period after intake that would confirm or deny an approximate 500-d clearance half-time. However, in both of these cases the fecal excretion was too small to support a clearance half-time of 100s of days. Furthermore, in these two cases, chest counts also showed that there was no clearance from the lung, either to the blood or to the GI tract, represented by a half-time of 100s of days.

This information indicates that super class Y Pu clears from the lung/lymph system to the blood extremely slowly, beginning at the instant of intake; and, after the first few days, might clear extremely slowly to the GI tract as well. Not enough data were available to describe early GI clearance.

OTHER RELEVANT STUDIES

Studies that support the concept of very long retention of Pu in the lung/lymph system are described in this section.

Human

McInroy et al. (1989) recently reported on the distribution of Pu in five whole bodies analyzed postmortem under the United States Transuranium Registry (USTR) program. In four of these cases the principal route of intake was inhalation. The exact time(s) of intake and the chemical forms of the Pu were not known. Probable and possible dates of intake for the four cases are listed in Table 2. Also listed in Table 2 is the percentage of Pu found in the respiratory tract relative to the total Pu in the whole body. All cases show remarkably high percentages of Pu in the respiratory tract compared with expectations based on an intake of class Y Pu at the midpoint of the probable period. Two cases show higher-than-expected percentages even if the worst-case assumption about date of intake is made. Although the chemical form(s) of the Pu for these intakes is not known, the data clearly show that some form of Pu is retained in the human respiratory tract to a much greater degree than predicted for class Y material (ICRP 1979).

Dog

Guilmette et al. (1984) exposed beagle dogs via pernasal inhalation to mono-disperse ^{239}Pu particles that had been oxidized at 1150°C. Three

particle sizes were used (0.72-, 1.4-, and 2.8-μm AMAD). For dogs studied through about 800 d post intake, lung retention (in terms of the percentage of the pulmonary burden at 4 d post intake) was described by two-component exponential decay with half-times in the 10s of days and between 1,000 to 2,000 d, depending on particle size. The retention in the thoracic lymph nodes, however, was described by an increasing power function. When added together, the retention was essentially stable from about 500 to 800 d at 64% of the initial long-term pulmonary burden.

Park (1987) gave beagle dogs inhalation exposure to poly-disperse (2.3-μm mean AMAD) ^{239}Pu particles calcined at 750°C. These dogs were allowed to live out their normal lifetimes, which in some cases extended to 15 y. The lung retention (in terms of the percentage of the lung burden at 14 and 30 d post intake) was described by a single exponential clearance with a half-time of 1,100 d. The thoracic lymph node retention grew rapidly for roughly the first 1,000 d then approached 39% asymptotically. The Park data indicate a combined lung/thoracic lymph node retention of about 75% at 800 d post intake, which, when the different starting points of 14/30 d post intake (Park 1987) and 4 d post intake (Guilmette et al. 1984) are accounted for, agree well with the Guilmette data.

The dog data indicate then that when activity in the thoracic lymph nodes is included the lung retention appears to become quite stable at a significant fraction of the initial long-term pulmonary burden. In addition, Guilmette et al. (1984) observed that only about 0.1% of the initial long-term pulmonary burden was found in the skeleton and 0.2% in the liver at 2 y post intake. This agrees well with the lack of detectable Pu in urine samples from the 10 Hanford human cases.

CONCLUSION

The Hanford and USTR human cases (McInroy et al. 1989) and the Guilmette et al. (1984) and Park (1987) dog studies all indicate that there is a form of Pu that, when inhaled, behaves considerably differently in the lung than described by the ICRP 30 lung model. Specifically, the Pu is more tenaciously retained in the lung/thoracic lymph nodes than predicted. The clearance to blood is characterized by extremely long clearance half-times, and the fraction of the initial long-term pulmonary deposition that clears to the GI tract may be less than predicted. The form of Pu involved seems to be Pu oxide that has been heated to at least several times ambient temperature, although the data do not imply this with certainty nor do they imply that Pu oxide always behaves this way.

The implications for the applied internal dosimetrist are that urine sampling, even when conducted immediately after a possible intake, may not be adequate to confirm a significant intake. In addition, if very little ^{241}Am is initially present in the material, chest counting may not be adequate to confirm a significant intake until several years later (Bihl et al. 1988).

REFERENCES

Bihl, D. E.; Lynch, T. P.; Carbaugh, E. H.; Sula, M. J. Methods to improve routine bioassay monitoring for freshly separated, poorly transported plutonium. Richland, WA: Pacific Northwest Laboratory; PNL-6695; 1988.

Guilmette, R. A.; Diel, J. H.; Muggenburg, B. A.; Mewhinney, J. A.; Boecker, B. B.; McClellan, R. O. Biokinetics of inhaled $^{239}PuO_2$ in the beagle dog: effect of aerosol particle size. Int. J. Radiation Bio. 45:563-581; 1984.

International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. Oxford: Pergamon Press; ICRP Publication 30; 1979.

International Commission on Radiological Protection. The metabolism of plutonium and related elements. Oxford: Pergamon Press; ICRP Publication 48; 1986.

McInroy, J. R.; Kathren, R. L.; Swint, M. J. Distribution of plutonium and americium in whole bodies donated to the United States Transuranium Registry. Rad. Prot. Dosim.; 26:151-158; 1989.

Northcutt, A. R.; Binney, S. E.; Palmer, H. E. In vivo counting of ^{241}Am in human lungs and tracheobronchial lymph nodes. Health Phys. 54:73-81; 1988.

Park, J. F. Inhaled plutonium oxide in dogs. In: Pacific Northwest Laboratory Annual Report for 1986 to the DOE Office of Energy Research, Part I, Biomedical Sciences. Richland, WA: Pacific Northwest Laboratory (PNL-6100 P.1) 1987:5-10.

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Spitz, H. B.; Robinson, R. Deposition of plutonium in the lungs of a worker following an accidental inhalation exposure. In: Wrenn, M.E., ed. Actinides in man and animals. Salt Lake City, UT: RD Press; 1981:115-135.

Stradling, G. N.; Stather, J. W. The use of animal experiments for assessing annual limits on intake and interpreting chest-monitoring data for workers exposed to industrial actinide-bearing dusts. Health Phys. 57, Sup.1:221-228; 1989.

TABLE CAPTIONS

Table 1. Summary of cases

Table 2. United States Transuranium Registry (USTR) case data

Case number	Intake date	Chemical form	Approximate clearance half-time[a] (d)	Confirmed by bioassay at time of intake?	Estimated initial pulmonary deposition [Bq (nCi)]
HAN01	5-23-78	oxide	6,000	yes	510 (14)
HAN02	3-5-79	oxide	>20,000	no	100 (3)
HAN03	3-26-64 or 6-8-64	oxide	>10,000	no	200 (5)
HAN04	8-11-70	oxide	13,000	yes	170 (4.6)
HAN05	3-6-70	oxide	6,000	no	200 (6)
HAN06	3-7-66	oxide	50 (20%)[b] 5,000 (80%)	no	30 (0.8) 100 (3)
HAN07	5-2-69	oxide	17,000	no	40 (1)
HAN08	5-22-70	oxide	6,000	no	80 (2)
HAN09	3-5-79	oxide	>20,000	no	40 (1)
HAN10	7-18-69	oxide	<1 (1-2%) >20,000 (98-99%)	no	20 (0.6)

[a] From lung and thoracic lymph nodes.
[b] Estimated % of initial deposition clearing by each clearance rate.

USTR case number	Probable period of intake	Possible period of intake	Died	% of Pu in respiratory tract[b]	% predicted by ICRP 30 lung model[a]	
					Probable[c]	Possible[d]
193	1945-57	1945-82	1982	53	9	89
208	1946-47	1946-47	1984	38	9	9
213	1946-67	1946-79	1984	19	8	41
242	1946-58	1946-72	1987	70	9	12

[a] Assuming 1-μm activity median aerodynamic diameter (AMAD), class Y particles and ICRP 48 (1986) systemic retention model.
[b] Percent of whole body content.
[c] Assuming single intake at midpoint of probable period.
[d] Assuming single intake at end of possible period.

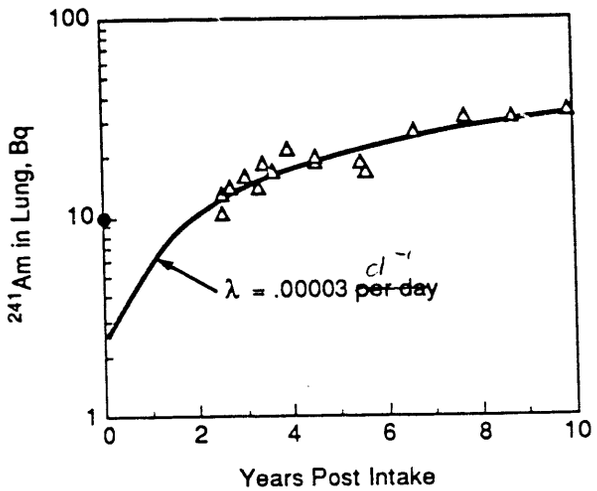

Fig. 1b. HAN02.

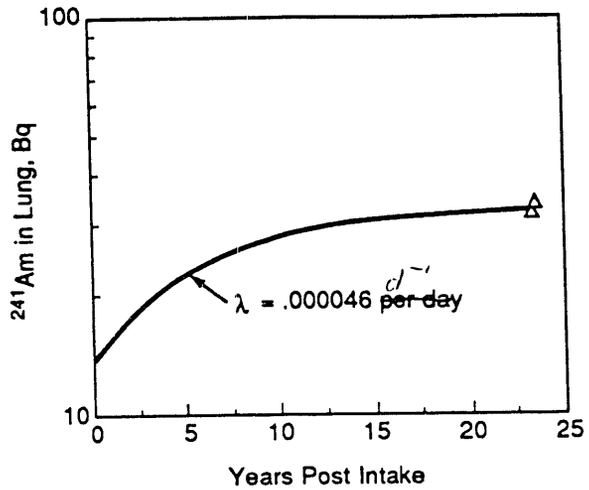

Fig. 1c. HAN03.

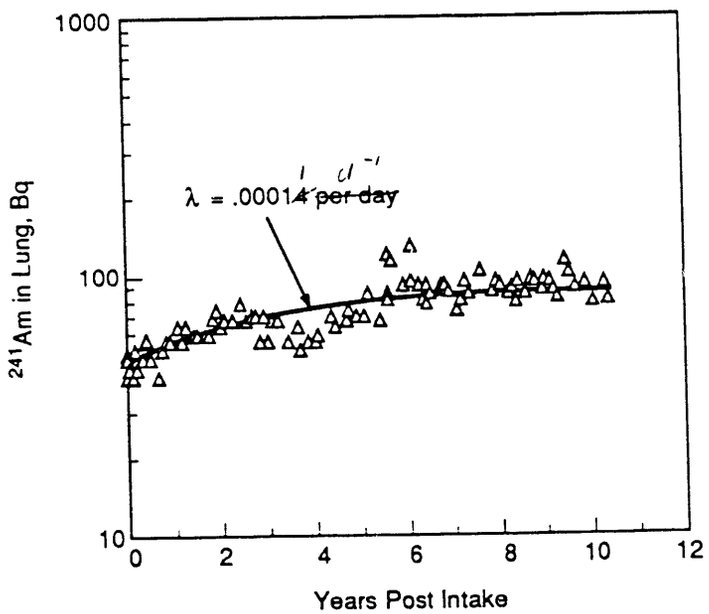

Fig. 1a. HAN01.

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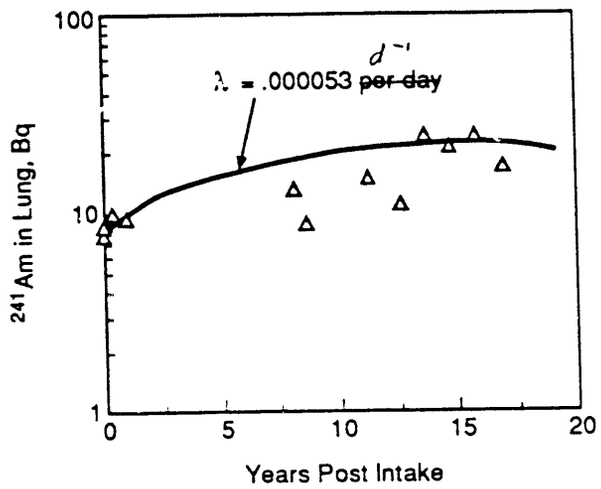

Fig. 1d. HAN04.

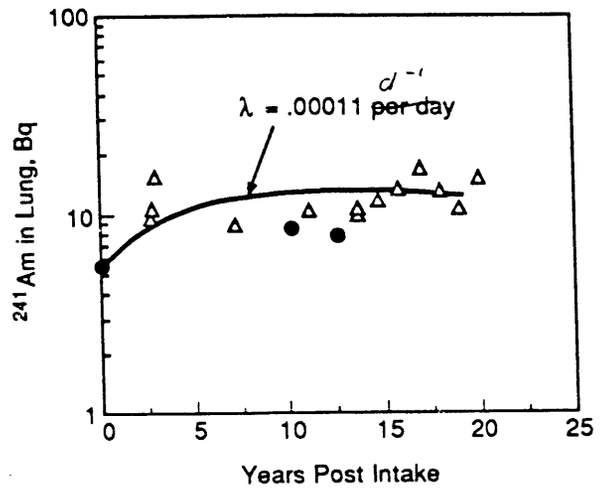

Fig. 1e. HAN05.

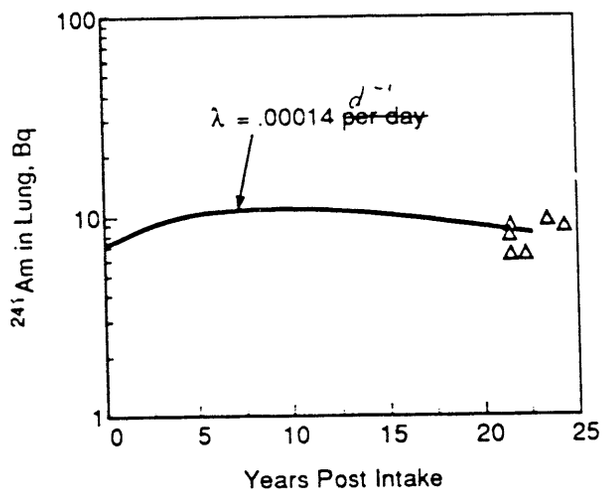

Fig. 1f. HAN06.

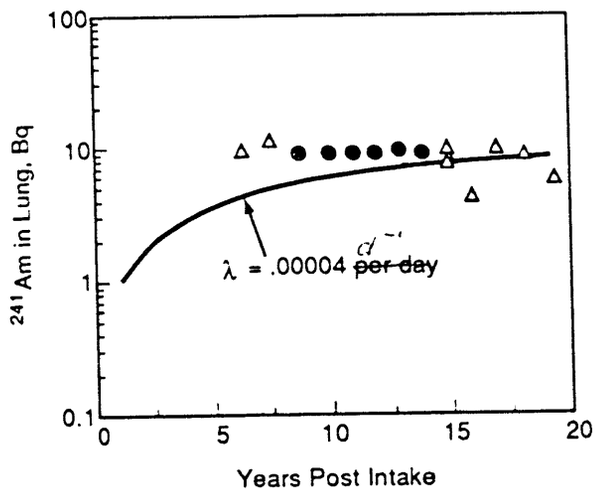

Fig. 1g. HAN07.

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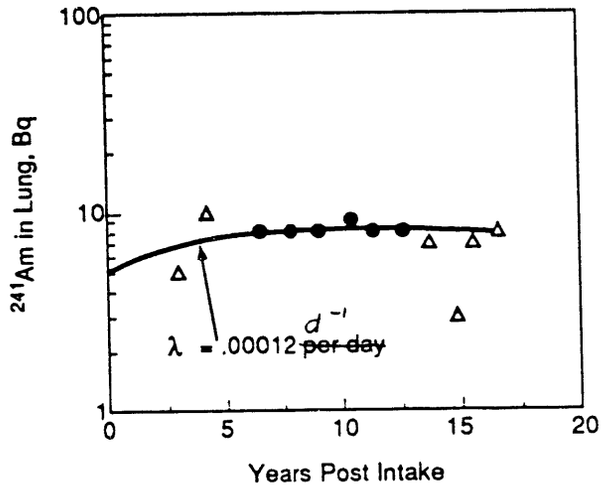

Fig. 1h. HAN08.

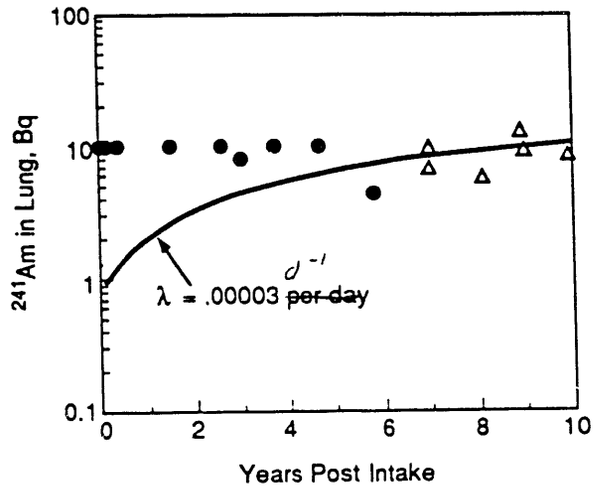

Fig. 1i. HAN09.

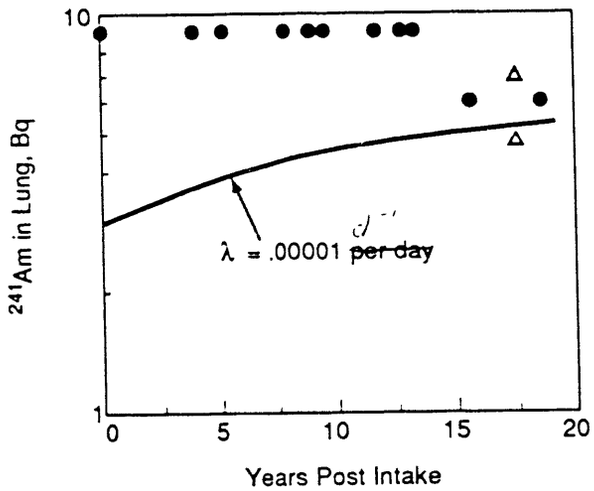

Fig. 1j. HAN010.

Fig. 1 a-j. Lung burden of ^{241}Am as a function of time post intake for each of the cases as determined by in vivo counting. The ● indicates counts where no americium was detected, plotted at the approximate detection level. The solid line shows the ingrowth of ^{241}Am from ^{241}Pu assuming a single rate of clearance from the lung as shown. In most cases the clearance rate is very approximate.

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APPENDIX
BIOASSAY DATA

HAN01

IN VIVO

The chest count data set for this case will appear in the proceedings of the Third International Workshop on Respiratory Tract Dosimetry in Radiation Protection Dosimetry ("Long-Term Follow-Up of HAN01, an Acute Plutonium Oxide Inhalation Case," Carbaugh et al.).

URINE

TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

(Note: DTPA was given on 5/23/78, 5/24/78, 5/25/78, 5/29/78, and 6/1/78.)

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
052378	4	100	1.6×10^{-2}		8.3×10^{-3}
052378	4	150	8.8×10^{-3}		•
052378	4	100	$<8.3\times10^{-3}$		•
052378	4	280	$<8.3\times10^{-3}$		•
052478	4	100	$<8.3\times10^{-3}$		•
052478	0	1500	5.1×10^{-2}		8.3×10^{-4}
052578	4	100	$<8.3\times10^{-3}$		8.3×10^{-3}
052578	0	900	1.2×10^{-2}		8.3×10^{-4}
052678	0	950	3.0×10^{-3}		•
052778	0	1250	$<8.4\times10^{-4}$		•
052878	0	700	2.8×10^{-3}		•
053078	0	700	1.7×10^{-3}		•
053178	0	1550	5.7×10^{-3}		•
060278	0	1200	3.7×10^{-3}		•
060378	0	1000	7.4×10^{-3}		•
060478	0	1200	1.6×10^{-3}		•
061078	0	1200	2.4×10^{-3}		•
061778	0	1100	1.8×10^{-3}		•
062478	0	1400	9.8×10^{-4}		•
070178	0	1100	$<8.3\times10^{-4}$		•
082278	0	1000	$<8.3\times10^{-4}$		•
091278	0	1100	6.2×10^{-4}		•
091478	0	650	$<8.3\times10^{-4}$		•
091978	0	800	•		•
021379	0	500	•		•
100479	0	1230	•		•
100979	0	1000	•		•
111579	0	1400	•		•
012280	0	1480	•		•
012980	0	500	•		•
040280	0	900	•		•
040281	0	900	•		•
042182	1	1300	0.0	9.0×10^{-4}	•
042083	1	1450	2.6×10^{-4}	1.4×10^{-4}	•
122183	1	1900	3.0×10^{-4}	1.5×10^{-4}	5.8×10^{-4}
092084	1	800	3.3×10^{-4}	9.3×10^{-5}	3.3×10^{-4}

URINE (cont'd)

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
071185	1	1315	7.7×10^{-4}	1.6×10^{-4}	•
070986	1	506	2.3×10^{-4}	7.9×10^{-5}	•
070887	1	1000	6.3×10^{-5}	7.1×10^{-5}	3.3×10^{-4}
071288	1	1680	1.2×10^{-4}	6.7×10^{-5}	•
071389	1	874	2.4×10^{-4}	1.3×10^{-4}	•

[a] 0 means not recorded, presumably a 24-h total or 24-h simulated sample; 1 means a 24-h simulated sample; 4 means a single void.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity (MDA) for the analysis procedure at the time.

FECES

Sample date	Sample mass (g)	Results (Bq)			
		^{239}Pu	^{238}Pu	^{241}Pu	^{241}Am
052478	106	$1.1\times10^{+2}$	$1.3\times10^{+1}$	$1.2\times10^{+3}$	6.2
052678	107	1.8×10^{-1}	7.2×10^{-2}	$1.1\times10^{+2}$	5.7×10^{-1}
052778	189	1.7×10^{-1}	7.5×10^{-2}	$<6.3\times10^{+1}$	$<5.5\times10^{-1}$
052878	158	2.5×10^{-1}	8.8×10^{-2}	$<5.3\times10^{+1}$	a
052978	155	1.6×10^{-1}	5.0×10^{-2}	$<5.3\times10+1$	a
022779	119	7.3×10^{-3}	1.3×10^{-3}	a	a
012280	100	$<3.3\times10^{-3}$	a	a	a
073184	22	1.0×10^{-4}[b]	1.0×10^{-4}[b]	a	a

[a] Not analyzed.
[b] One standard deviation of total propagated uncertainty was 3.5×10^{-4}.

HAN02

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
030679	Chest	< MDA	c			
082781	Chest	$1.0x10^{+1}$	$1.0x10^{+1}$			
090181	Chest	$1.3x10^{+1}$	$1.0x10^{+1}$			
090181	Chest	$1.1x10^{+1}$	$1.0x10^{+1}$			
111881	Chest	$1.4x10^{+1}$	$1.0x10^{+1}$			
030982	Chest	$1.6x10^{+1}$	$1.1x10^{+1}$			
071982	Chest	$1.4x10^{+1}$	$1.1x10^{+1}$			
080482	Chest	$1.9x10^{+1}$	$1.1x10^{+1}$			
100782	Chest	$1.7x10^{+1}$	$1.1x10^{+1}$			
012583	Chest	$2.1x10^{+1}$	$1.1x10^{+1}$			
081683	Chest	$2.0x10^{+1}$	$1.1x10^{+1}$			
081683	Chest	$1.9x10^{+1}$	3.7			
082384	Chest	$1.9x10^{+1}$	3.7			
092484	Chest	$1.7x10^{+1}$	3.7	Head	< MDA	7.8
100185	Chest	$2.7x10^{+1}$	3.7	Head	< MDA	8.9
111886	Chest	$3.2x10^{+1}$	3.7			
112387	Chest	$3.2x10^{+1}$	3.7	Head	< MDA	$1.2x10^{+1}$
012589				Head	< MDA	$1.3x10^{+1}$
012789	Chest	$3.4x10^{+1}$	2.7			
011190	Chest	$4.3x10^{+1}$	4.4			
011190	Chest	$5.6x10^{+1}$	4.1			

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.
[c] MDA was not recorded; authors estimate that it was about 10 Bq.

URINE
TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
030679	4	100	$< 8.3x10^{-3}$		$8.3x10^{-3}$
031379	1	800	$< 8.3x10^{-4}$		$8.3x10^{-4}$
071079	1	1000	$< 8.3x10^{-4}$		•
071279	1	900	$< 8.3x10^{-4}$		•
020780	1	800	$< 8.3x10^{-4}$		•
021281	1	1200	$< 8.3x10^{-4}$		•
082781	4	40	$< 8.3x10^{-3}$		$8.3x10^{-3}$
031782	1	600	$3.3x10^{-5}$	$1.3x10^{-4}$	$8.3x10^{-4}$
020283	1	800	$1.0x10^{-4}$	$1.0x10^{-4}$	•

URINE (cont'd)

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
022284	1	800	1.6×10^{-4}	6.8×10^{-5}	3.3×10^{-4}
042485	1	1050	3.3×10^{-5}	5.0×10^{-5}	•
031186	1	1400	1.6×10^{-4}	7.0×10^{-5}	•
031087	1	998	1.8×10^{-4}	7.7×10^{-5}	•
041587	1	596	9.8×10^{-5}	5.7×10^{-5}	•
041288	1	904	8.8×10^{-5}	5.5×10^{-5}	•

[a] 0 means not recorded, presumably a 24-h total or 24-h simulated sample; 1 means a 24-h simulated sample; 4 means a single void.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

^{238}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
022284	1	800	1.2×10^{-4}	5.0×10^{-5}	3.3×10^{-4}
042485	1	1050	1.3×10^{-4}	6.7×10^{-5}	•
031186	1	1400	2.8×10^{-4}	1.0×10^{-4}	•
031087	1	998	1.1×10^{-4}	5.7×10^{-5}	•
041587	1	596	2.3×10^{-4}	8.8×10^{-5}	•
041288	1	904	2.2×10^{-4}	7.5×10^{-5}	•

[a] 1 means a 24-h simulated sample.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

HAN03

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
101687	Chest	$3.2\times10^{+1}$	3.3	Head	<MDA	$1.4\times10^{+1}$
120287	Chest	$3.4\times10^{+1}$	3.2	Head	<MDA	$1.3\times10^{+1}$
120287	Chest	$2.9\times10^{+1}$	3.6			

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.

URINE

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
032964	0	600	$<8.3\times10^{-4}$		8.3×10^{-4}
040264	0	500	$<8.3\times10^{-4}$		"
061064	0	700	5.8×10^{-4}		"
061164	0	700	$<8.3\times10^{-4}$		"
061364	0	900	4.5×10^{-4}		"
061664	0	1600	$<8.3\times10^{-4}$		"
063064	0	900	$<8.3\times10^{-4}$		"
100864	0	500	$<8.3\times10^{-4}$		"
111964	0	500	4.6×10^{-4}		"
072265	0	500	5.0×10^{-4}		"
120965	0	500	1.2×10^{-3}		"
041266	0	600	$<8.3\times10^{-4}$		"
060266	0	650	5.2×10^{-4}		"
060766	0	600	$<8.3\times10^{-4}$		"
092066	0	700	5.2×10^{-4}		"
051967	0	680	1.2×10^{-3}		"
071167	0	850	8.3×10^{-4}		"
071367	0	580	$<8.3\times10^{-4}$		"
071867	0	700	$<8.3\times10^{-4}$		"
092167	0	900	$<8.3\times10^{-4}$		"
092667	0	500	$<8.3\times10^{-4}$		"
021568	0	600	$<8.3\times10^{-4}$		"
091868	0	1000	$<8.3\times10^{-4}$		"
020669	0	950	$<8.3\times10^{-4}$		"
122872	0	700	5.3×10^{-4}		"
120573	0	500	$<8.3\times10^{-4}$		"
121174	0	500	$<8.3\times10^{-4}$		"
071675	0	500	$<8.3\times10^{-4}$		"
102187	1	1305	2.2×10^{-4}	8.5×10^{-5}	3.3×10^{-4}
102787	1	740	2.2×10^{-4}	7.8×10^{-5}	"
102987	1	800	1.9×10^{-4}	8.4×10^{-4}	"

[a] 0 means not recorded, presumably a 24-h total or 24-h simulated sample, 1 means a 24-h simulated sample, 4 means a single void.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

FECES

Sample date	Sample mass (g)	Radionuclide	Resuk (Bq)[a]
102287	95	^{238}Pu	$(3.2 \pm 3.2)\times10^{-4}$
		^{239}Pu	$(18 \pm 8.8)\times10^{-4}$
		^{241}Am	$(8.3 \pm 3.7)\times10^{-4}$
111287	64	^{238}Pu	$(1.6 \pm 1.1)\times10^{-4}$
		^{239}Pu	$(2.7 \pm 1.6)\times10^{-4}$
		^{241}Am	$(8.2 \pm 2.2)\times10^{-4}$

[a] The MDA for these analyses was abou: 2×10^{-3} Bq.

HAN04

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
061170	Chest	<MDA	6.3			
081170	Chest	8.9	6.7			
081870	Chest	7.8	6.3			
092370	Chest	9.2	6.3			
120870	Chest	$1.0x10^{+1}$	6.3			
062571	Chest	9.6	6.3			
092571	Chest	$1.3x10^{+1}$	8.1			
033079	Chest	8.9	8.5			
102181	Chest	$1.5x10^{+1}$	8.5			
031683	Chest	$1.1x10^{+1}$	8.5			
030184	Chest	$2.4x10^{+1}$	5.2			
031385	Chest	$-2.2x10^{+1}$	3.3			
040986	Chest	$2.4x10^{+1}$	3.7			
071787	Chest	$1.7x10^{+1}$	3.3	Head	<MDA	$1.1x10^{+1}$

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.

URINE
TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
062370	0	1100	$<8.3x10^{-4}$		$8.3x10^{-4}$
081270	0	900	$<8.3x10^{-4}$		"
081370	0	800	$<8.3x10^{-4}$		"
081470	0	1200	$<8.3x10^{-4}$		"
092270	0	1100	$<8.3x10^{-4}$		"
092970	0	700	$<8.3x10^{-4}$		"
111370	0	1180	$6.8x10^{-4}$		"
120870	0	1300	$<8.3x10^{-4}$		"
120970	0	1300	$<8.3x10^{-4}$		"
121570	0	2000	$<8.3x10^{-4}$		"
021071	0	1150	$<8.3x10^{-4}$		"
051171	0	700	$<8.3x10^{-4}$		"
062571	2	400	$<8.3x10^{-4}$		"
102081	1	750	$-3.4x10^{-4}$	$1.0x10^{-2}$	"
020982	1	1200	$8.0x10^{-3}$	$7.0x10^{-3}$	"
020883	1	950	$8.4x10^{-3}$	$6.6x10^{-3}$	"

URINE (cont'd)

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
021484	1	850	6.9×10^{-3}	3.5×10^{-3}	3.3×10^{-4}
101684	1	833	2.5×10^{-3}	2.5×10^{-3}	"
042585	1	1049	6.9×10^{-3}	3.8×10^{-3}	"
050886	1	1091	9.5×10^{-3}	4.5×10^{-3}	"
050687	1	965	8.5×10^{-3}	3.8×10^{-3}	"

[a] 0 means not recorded, presumably a 24-h total or 24-h simulated sample; 1 means a 24-h simulated sample; 2 means a 12-h simulated sample; 4 means a single void.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

FECES

Sample date	Sample mass (g)	Radionuclide	Result (Bq)[a]
081270	210	Pu alpha	6.4
		Am-241	.6
081370	120	Pu alpha	.02
		Am-241	.6
081470	120	Pu alpha	.02
		Am-241	.6
120870	190	Pu alpha	.02
120970	not recorded	Pu alpha	.01

[a] The minimum detectable activity for these analyses at that time is not recorded.

HAN05

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
030670	Chest	<MDA	5.5			
112172	Chest	9.6	5.2			
120772	Chest	$1.1x10^{+1}$	5.2			
022373	Chest	$1.6x10^{+1}$[c]	4.8			
051877	Chest	8.9	7.8			
042380	Chest	<MDA	8.5			
041381	Chest	$1.0x10^{+1}$	7.8			
101882	Chest	<MDA	7.8			
102083	Chest	$1.0x10^{+1}$	7.8			
102483	Chest	$1.1x10^{+1}$	5.2			
111284	Chest	$1.2x10^{+1}$	4.1			
112685	Chest	$1.3x10^{+1}$	4.4	Head	<MDA	8.9
012987	Chest	$1.7x10^{+1}$	3.0	Head	<MDA	$1.3x10^{+1}$
012987				Liver	<MDA	3.7
011388	Chest	$1.3x10^{+1}$	4.1	Head	<MDA	$1.4x10^{+1}$
011889	Chest	$1.1x10^{+1}$	4.0	Head	<MDA	$1.3x10^{+1}$
011890	Chest	$1.5x10^{+1}$	4.7	Head	<MDA	$2.0x10^{+1}$

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.
[c] Considered faulty at the time of measurement; 11 was estimated to be the correct value.

URINE
TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
031270	1	1700	$<8.3x10^{-4}$		$8.3x10^{-4}$
031770	1	1400	"		"
031970	1	1750	"		"
041470	1	1550	"		"
081270	1	1150	"		"
100770	1	1100	"		"
011971	1	1700	"		"
012171	1	1550	"		"
071471	1	1500	"		"
011272	1	1100	"		"
112872	1	1300	"		"
020773	1	1400	"		"
022373	1	150	"		"
042578	1	1500	"		"
042979	1	1650	"		"
040280	1	2000	"		"
040281	1	1900	"		"
040682	1	1800	$-9.0x10^{-5}$	$9.0x10^{-5}$	"
041283	1	1500	$1.7x10^{-5}$	$7.8x10^{-5}$	"

URINE (cont'd)

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
041284	1	1600	0.0	4.0×10^{-5}	3.3×10^{-4}
040485	1	2449	1.6×10^{-4}	7.2×10^{-5}	•
040886	1	2162	5.2×10^{-5}	3.7×10^{-5}	•
040787	1	1188	1.1×10^{-5}	3.5×10^{-5}	•
040688	1	1934	3.2×10^{-5}	3.2×10^{-5}	•
040589	1	2401	3.8×10^{-5}	6.7×10^{-5}	•
020790	1	1852	5.5×10^{-5}	5.8×10^{-5}	•

[a] 1 means a 24-h simulated sample.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

HAN06

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
062487	Chest	8.1	6.6			
071587	Chest	8.9	4.8			
071587	Chest	6.3	4.4			
060188	Chest	6.3	4.4	Head	<MDA	$1.3x10^{+1}$
081689	Chest	9.6	3.3			
091980				Head	<MDA	$1.1x10^{+1}$
070690	Chest	8.9	5.2	Head	<MDA	9.6

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.

URINE
TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
030866	1	1450	$<8.3x10^{-4}$		$8.3x10^{-4}$
031066	1	1550	$<8.3x10^{-4}$		"
031566	1	1200	$1.2x10^{-3}$		"
032466	1	1300	$7.8x10^{-4}$		"
071366	1	600	$5.2x10^{-4}$		"
090866	1	1000	$<8.3x10^{-4}$		"
091366	1	1500	$<8.3x10^{-4}$		"
031167	1	1300	$<8.3x10^{-4}$		"
031368	1	700	$<8.3x10^{-4}$		"
091769	1	700	$<8.3x10^{-4}$		"
090970	1	1100	$<8.3x10^{-4}$		"
092171	1	1700	$<8.3x10^{-4}$		"
101172	1	800	$<8.3x10^{-4}$		"
112073	1	900	$<8.3x10^{-4}$		"
091074	1	1500	$<8.3x10^{-4}$		"
091675	1	1000	$<8.3x10^{-4}$		"
101276	1	1300	$<8.3x10^{-4}$		"
092077	1	1400	$<8.3x10^{-4}$		"
111286	1	1178	$4.4x10^{-4}$	$1.1x10^{-4}$	$3.3x10^{-4}$
123086	1	806	$2.4x10^{-4}$	$6.9x10^{-5}$	"
011387	1	1202	$3.4x10^{-5}$	$3.8x10^{-5}$	"
080487	1	990	$2.9x10^{-4}$	$9.3x10^{-5}$	"
111787	1	541	$2.8x10^{-4}$	$1.0x10^{-4}$	"
110888	1	1437	$9.5x10^{-5}$	$5.8x10^{-5}$	"
032190	1	1038	$1.7x10^{-4}$	$8.4x10^{-5}$	"

[a] 1 means a 24-h simulated sample.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

HAN07

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
082775	Chest	9.6	8.8			
111276	Chest	$1.1x10^{+1}$	9.2			
011978	Chest	<MDA	9.2			
033079	Chest	<MDA	9.2			
032680	Chest	<MDA	9.2			
022781	Chest	<MDA	9.2			
020882	Chest	<MDA	9.6			
021783	Chest	<MDA	9.2			
022084	Chest	$1.0x10^{+1}$	4.1			
022784	Chest	7.8	4.1			
031684	Chest	8.5	4.1			
031385	Chest	4.4	4.1			
041086				Head	<MDA	$1.2x10^{+1}$
041486	Chest	$1.0x10^{+1}$	4.1			
060287	Chest	8.9	4.8	Head	<MDA	$1.3x10^{+1}$
092188	Chest	5.9	4.4	Head	<MDA	$1.2x10^{+1}$
110389	Chest	9.6	5.2	Head	<MDA	$1.1x10^{+1}$

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.

URINE
TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
050669	0	1100	$<8.3x10^{-4}$		$8.3x10^{-4}$
050869	0	1100	$<8.3x10^{-4}$		"
051369	0	1500	$<8.3x10^{-4}$		"
061169	0	1150	$<8.3x10^{-4}$		"
091769	0	900	$<8.3x10^{-4}$		"
120269	0	1100	$<8.3x10^{-4}$		"
030470	0	800	$<8.3x10^{-4}$		"
060370	0	1000	$<8.3x10^{-4}$		"
090270	0	1200	$<8.3x10^{-4}$		"
120270	0	1200	$<8.3x10^{-4}$		"
031071	0	1200	$<8.3x10^{-4}$		"

URINE (cont'd)

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
022384	1	1550	1.5×10^{-4}	7.1×10^{-5}	3.3×10^{-4}
022884	1	1600	1.8×10^{-5}	3.2×10^{-5}	•
030184	1	1600	7.0×10^{-5}	3.5×10^{-5}	•
050185	1	1226	7.3×10^{-5}	5.3×10^{-5}	•
050186	1	1670	7.5×10^{-5}	4.3×10^{-5}	•
051288	1	1488	6.7×10^{-5}	4.7×10^{-5}	•
050289	1	1881	-2.8×10^{-5}	5.0×10^{-5}	•

[a] 0 means not recorded, presumably a 24-h total or 24-h simulated sample; 1 means a 24-h simulated sample.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

HAN08

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
051873	Chest	5.2	Not recorded			
090374	Chest	9.6	5.2			
120176	Chest	<MDA	8.1			
030778	Chest	<MDA	8.5			
051679	Chest	<MDA	7.8			
100680	Chest	<MDA	8.9			
090981	Chest	<MDA	8.1			
121382	Chest	<MDA	8.5			
020384	Chest	7.4	3.7			
020384	Chest	3.3	2.6			
112884	Chest	7.4	4.4			
010786	Chest	8.5	4.4			

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.

URINE
TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
061869	0	1300	$<8.3\times10^{-4}$		8.3×10^{-4}
052670	1	1500	7.2×10^{-4}		•
060270	1	1350	$<8.3\times10^{-4}$		•
060470	1	1500	$<8.3\times10^{-4}$		•
062570	0	1500	$<8.3\times10^{-4}$		•
060371	0	1500	$<8.3\times10^{-4}$		•
062872	0	1700	8.7×10^{-4}		•
091272	0	1400	$<8.3\times10^{-4}$		•
061174	0	600	$<8.3\times10^{-4}$		•
061075	0	1500	$<8.3\times10^{-4}$		•
061477	1	1700	$<8.3\times10^{-4}$		•
060678	1	1550	$<8.3\times10^{-4}$		•
061780	1	1400	$<8.3\times10^{-4}$		•
061681	1	1950	$<8.3\times10^{-4}$		•
060882	1	1500	-5.4×10^{-5}	7.2×10^{-5}	•
060783	1	1800	1.7×10^{-5}	7.8×10^{-5}	•
051584	1	1600	2.1×10^{-5}	4.7×10^{-5}	3.3×10^{-4}
061385	1	886	1.1×10^{-4}	7.7×10^{-5}	•
060486	1	2126	7.9×10^{-5}	4.9×10^{-5}	•

[a] 0 means not recorded, presumably a 24-h total or 24-h simulated sample; 1 means a 24-h simulated sample.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
030679	Chest	<MDA	$1.0x10^{+1}$			
042779	Chest	<MDA	$1.0x10^{+1}$			
072579	Chest	<MDA	$1.0x10^{+1}$			
090980	Chest	<MDA	9.6			
100281	Chest	<MDA	$1.0x10^{+1}$			
110182	Chest	<MDA	$1.0x10^{+1}$			
110783	Chest	<MDA	$1.0x10^{-1}$			
120384	Chest	<MDA	4.4			
022686	Chest	9.9	4.8			
030386	Chest	6.7	5.2			
042487	Chest	5.7	3.4			
020388	Chest	$1.3x10^{+1}$	4.8			
022588	Chest	9.1	3.6			
020889	Chest	8.5	3.3			

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.

URINE
TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
030679	4	110	$9.2x10^{-3}$		$8.3x10^{-3}$
031379	0	1000	$<8.3x10^{-4}$		$8.3x10^{-4}$
042779	0	400	$<8.3x10^{-4}$		•
052479	0	500	$1.0x10^{-3}$		•
071079	0	1000	$<8.3x10^{-4}$		•
100379	0	650	$<8.3x10^{-4}$		•
011381	1	700	$<8.3x10^{-4}$		•
011282	1	800	$-3.3x10^{-5}$	$1.3x10^{-4}$	•
012583	1	800	$5.7x10^{-5}$	$8.8x10^{-5}$	•
011084	1	1100	$5.7x10^{-5}$	$1.0x10^{-4}$	$3.3x10^{-4}$
051584	1	800	$6.8x10^{-5}$	$5.5x10^{-5}$	•
050785	1	1530	$1.5x10^{-4}$	$7.2x10^{-5}$	•
030686	1	1480	$7.8x10^{-5}$	$6.0x10^{-5}$	•
031286	1	1038	$2.9x10^{-5}$	$2.9x10^{-5}$	•
050886	1	1054	$1.2x10^{-4}$	$6.2x10^{-5}$	•
050687	1	1014	$-1.6x10^{-5}$	$1.6x10^{-5}$	•
050488	1	1401	$1.8x10^{-5}$	$5.8x10^{-5}$	•
050389	1	1381	$2.2x10^{-4}$	$9.1x10^{-5}$	•
071389	1	1235	$1.1x10^{-4}$	$7.9x10^{-5}$	•
071889	1	1150	$1.1x10^{-4}$	$6.3x10^{-5}$	•

[a] 0 means not recorded, presumably a 24-h total or 24-h simulated sample; 1 means a 24-h simulated sample; 4 means a single void.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

HAN10

IN VIVO
ALL RESULTS ARE FOR ^{241}Am

Measurement date	Measurement type	Result (Bq)[a]	MDA (Bq)[b]	Measurement type	Result (Bq)[a]	MDA (Bq)[b]
072169	Chest	<MDA	c			
060873	Chest	<MDA	c			
092774	Chest	<MDA	c			
052477	Chest	<MDA	8.8			
062978	Chest	<MDA	8.8			
012679	Chest	<MDA	8.6			
041080	Chest	<MDA	9.2			
041581	Chest	<MDA	9.2			
093082	Chest	<MDA	8.5			
021285	Chest	<MDA	5.6			
120486	Chest	4.8	4.1			
012087	Chest	7.0	3.0	Head	<MDA	$1.2 \times 10^{+1}$
120787	Chest	<MDA	6.3			

[a] Chest counts corrected for chest wall thickness using ultrasound measurements; results from head counts represent total skeletal activity.
[b] Minimum detectable activity calculated separately for each count as three standard deviations of counts in background channels.
[c] Recorded as 5.6 Bq, but authors believe that 8.8 Bq is more appropriate.

URINE

TOTAL PLUTONIUM ALPHA UNTIL 1984, THEN ^{239}Pu

Sample date	Sample type[a]	Sample volume (ml)	Result (Bq)	Uncertainty (Bq)[b]	MDA (Bq)[c]
071969	0	800	6.2×10^{-4}		8.3×10^{-4}
072169	0	800	$<8.3\times10^{-4}$		•
072369	0	1100	4.2×10^{-3}		•
081969	0	1300	$<8.3\times10^{-4}$		•
082669	0	900	•		•
082869	0	1100	•		•
090469	0	1400	•		•
120969	0	800	•		•
031070	0	500	•		•
022371	0	900	•		•
081171	0	850	•		•
020872	0	700	•		•
082372	0	500	•		•
021473	0	600	•		•
081473	0	800	•		•
021374	0	1000	•		•
022075	0	600	•		•
081375	0	700	•		•
020476	0	900	•		•
080476	0	800	•		•
020877	0	500	6.5×10^{-4}		•
081777	0	700	$<8.3\times10^{-4}$		•
020878	0	500	•		•
080978	0	600	•		•
022779	0	1000	•		•
021380	2	900	•		•
061081	1	800	•		•
060982	1	750	5.4×10^{-4}	7.2×10^{-5}	•
093082	2	500	2.5×10^{-5}	6.6×10^{-5}	•
021086	1	1042	1.0×10^{-5}	3.2×10^{-5}	3.3×10^{-4}
031787	1	1480	1.7×10^{-4}	1.2×10^{-4}	•
120987	1	1567	2.9×10^{-5}	2.9×10^{-5}	•

[a] 0 means not recorded, presumably a 24-h total or 24-h simulated sample; 1 means a 24-h simulated sample; 2 means a 12-h simulated sample.
[b] One standard deviation of the total propagated uncertainty, when known.
[c] Minimum detectable activity for the analysis procedure at the time.

FECES

Sample date	Sample mass (g)	Radionuclide	Result (Bq)[a]
071969	150	Pu alpha	9.3

DATE
FILMED
01/03/92