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Human Biokinetics of Plutonium: A Compilation of Experimental Data

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ABSTRACT

This report is an updated version of HPA-RPD-017 published in 2006. It has been prepared to give a detailed compilation of all the available data from the UK plutonium human volunteer studies except where they are fully documented elsewhere. It is not intended to provide a review of the results but rather to bring available data together in one publication. References are given to published work in which results are analysed. As further measurements may be made or samples analysed for some of the studies, the report may be updated further as more data become available.

Data from three studies are included. Two of the studies were initiated by the National Radiological Protection Board (NRPB), now part of the Centre for Radiation, Chemical and Environmental Hazards (CRCE), Public Health England (PHE). The third was initiated by AEA Technology, Harwell (AEAT) and subsequently continued by Middlesex University (MU).

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1 INTRODUCTION

There have been three long-term human volunteer studies involving plutonium in the UK. The first was initiated by the National Radiological Protection Board (NRPB), now part of the Centre for Radiation, Chemical and Environmental Hazards (CRCE), Public Health England (PHE). This study was designed to measure the absorption of ingested plutonium as well as its long-term urinary excretion after intravenous administration to males. The second study was initiated at AEA Technology, Harwell (AEAT) and subsequently continued by Middlesex University (MU). In this study, blood and liver content and urinary and faecal excretion were measured in females after intravenous administration of plutonium citrate. The third study was led by NRPB (now CRCE), with the administration of plutonium to the volunteers carried out under sub-contract by AEA Technology (AEAT). This study was performed to investigate the absorption from the respiratory tract of inhaled plutonium nitrate in humans, and to evaluate absorption parameter values for comparison with values determined for the same aerosol inhaled by other mammalian species. Each of the studies is described separately below and the results obtained to date are given for all three studies. All the studies received ethics committee approval and involved very low radiation doses approved by the Department of Health Administration of Radioactive Substances Advisory Committee. The isotopes used were ^{237}Pu, which has a half-life of 45.3 days (ICRP, 1983; NEA, 2005) and so is suitable for short-term studies, and ^{244}Pu, which has a half-life of 8.26×10^{7} years (ICRP, 1983) and is suitable for long-term studies because there will be virtually no radioactive decay over the lifetime of the volunteers. ^{237}Pu decays by electron capture and is thus quantifiable in bioassay samples and *in vivo* by measurement of its K X-ray emissions, while the long half-life of ^{244}Pu means that it is quantifiable in bioassay samples by mass spectrometry.

This report is an updated version of HPA-RPD-017 published in 2006 (Etherington et al., 2006).

2 THE CRCE STUDY OF INGESTION AND INTRAVENOUS INJECTION OF ^{244}Pu IN MALES

2.1 Outline of methodology

The gastrointestinal absorption and urinary excretion of ^{244}Pu were measured in five healthy adult males in a two-stage study. First, the volunteers ingested about 10^{14} atoms (~ 4×10^{-8} g) of ^{244}Pu as Pu(IV) citrate in solution with a mid-day meal and urinary excretion was measured for the following 7 - 9 days (Table 1). After a period of at least six months, the same volunteers were given an intravenous injection of about 2 x 10^{12} atoms (~ 8×10^{-10} g) of ^{244}Pu as Pu(IV) citrate in solution. Urinary excretion was then measured for the following 7 - 9 days and subsequently at intervals over periods currently up to 15 years after injection (Table 2).

Each sample was analysed by adding ^{242}Pu tracer and using established radiochemical methods to isolate the plutonium. In the earlier years of the experiment, measurements were made by thermal ionisation mass spectrometry at the Atomic Weapons Establishment, Aldermaston, UK. Since 1996, measurements have been made by resonance ion mass spectrometry at the Johannes Gutenberg University at Mainz in Germany. The latter

technique was used for samples collected after day no. 1600 for volunteer 1, after day no. 1200 for volunteers 2 and 3 and for all samples for volunteers 4 & 5; these are indicated by bold type in Table 2.

More complete descriptions of the study have been published (Popplewell et al., 1994; Ham and Harrison, 2000).

2.2 Uncertainties

The uncertainty on individual measurements resulting from counting statistics, expressed as a calculated standard deviation, was between 2 and 5% of the measured values. Other sources of uncertainty are more difficult to quantify. However, the change from thermal ionisation to resonance ion mass spectrometry did not produce a discontinuity in the excretion curve of the existing volunteers or a significant difference between results at similar post-injection times measured by different techniques. This strongly suggests that there is no bias between the different methods and that uncertainties from the particular measurement techniques are not significant.

The systematic uncertainty from the ^{242}Pu tracer used was about 2% (at one sigma, 1σ) as it was prepared by gravimetric dilution of an NPL certified tracer solution. The tracer purity was confirmed on both mass spectrometers by analysis of pure tracer samples. As the same tracer solution was used for the calibration of the injection solution and the measurement of the urine samples, its accuracy does not affect the measure of percentage excreted. Reagent blanks were analysed alongside all samples. For most analyses the ^{244}Pu content was below the limit of detection, and for the few cases where ^{244}Pu was measurable the blank values were subtracted from the analytical result.

It can be concluded that analytical uncertainties are small and of little importance in comparison to biological variability. Differences between volunteers are attributable to differences in biokinetic behaviour.

2.3 Results

Table 1 Gastrointestinal absorption of ^{244}Pu in male volunteers

Volunteer (Vol)	1	2	3	4	5
Age at ingestion, years	58	40	54	64	36
Atoms ingested	1.44×10^{14}	1.33×10^{14}	1.18×10^{14}	1.20×10^{14}	1.15×10^{14}
Urine collection period, days	8	9	9	7	8
Urine collection, atoms ^{244}Pu	2.32×10^{9}	8.02×10^{8}	2.12×10^{9}	2.97×10^{9}	4.48×10^{8}
Amount excreted by t=8d (%)[a]	1.82	2.43	2.23	2.14[b]	3.02
Total absorbed, atoms ^{244}Pu	1.27×10^{11}	3.21×10^{10}	9.21×10^{10}	1.39×10^{11}	1.48×10^{10}
Fractional absorption, f_1	8.9×10^{-4}	2.4×10^{-4}	7.8×10^{-4}	1.2×10^{-4}	1.3×10^{-4}

Notes

a. Expressed as a percentage of the amount absorbed after intravenous injection (from Table 2).

b. t=7d for Volunteer 4.

Table 2 Urinary excretion of plutonium after intravenous injection in male volunteers

Day	Measured Atoms					Proportion of initial intake excreted per day				
	Vol. 1	Vol. 2	Vol. 3	Vol. 4	Vol. 5	Vol. 1	Vol. 2	Vol. 3	Vol. 4	Vol. 5
	Atoms Intravenously Injected									
	2.35E+12	*1.97E+12*	*1.92E+12*	***2.17E+12***	***2.12E+12***					
1	2.08E+10	2.70E+10	2.25E+10	**3.13E+10**	**4.11E+10**	0.8835%	1.3706%	1.1712%	**1.4411%**	**1.9405%**
2	7.42E+09	5.44E+09	5.18E+09	**5.50E+09**	**7.27E+09**	0.3157%	0.2760%	0.2695%	**0.2532%**	**0.3432%**
3	3.26E+09	4.51E+09	4.62E+09	**3.03E+09**	**3.52E+09**	0.1389%	0.2287%	0.2404%	**0.1395%**	**0.1662%**
4	3.18E+09	3.41E+09	3.10E+09	**2.07E+09**	**3.92E+09**	0.1351%	0.1731%	0.1610%	**0.0953%**	**0.1851%**
5	2.23E+09	2.51E+09	2.18E+09	**1.78E+09**	**1.76E+09**	0.0948%	0.1275%	0.1134%	**0.0820%**	**0.0831%**
6	1.94E+09	1.74E+09	1.88E+09	**1.56E+09**	**2.70E+09**	0.0825%	0.0881%	0.0978%	**0.0718%**	**0.1275%**
7	2.51E+09	2.00E+09	1.78E+09	**1.15E+09**	**1.90E+09**	0.1069%	0.1013%	0.0924%	**0.0529%**	**0.0897%**
8	1.39E+09	1.33E+09	1.56E+09		**1.84E+09**	0.0591%	0.0676%	0.0809%		**0.0869%**
9		1.29E+09	1.37E+09		**1.56E+09**		0.0653%	0.0713%		**0.0737%**
9.5	1.08E+09					0.0460%				
11		5.78E+08					0.0293%			
13.5		7.57E+08					0.0384%			
14					**7.37E+08**					**0.0348%**
15.5	6.07E+08					0.0258%				
18.5		6.34E+08					0.0322%			
19.5	4.39E+08					0.0187%				
20					**4.06E+08**					**0.0192%**
23					**3.46E+08**					**0.0163%**
26.5		5.23E+08					0.0265%			
27.5	4.13E+08					0.0176%				
30					**3.97E+08**					**0.0187%**
40		3.63E+08					0.0184%			
45	2.47E+08					0.0105%				
68		3.20E+08					0.0163%			
71					**2.49E+08**					**0.0117%**
75			1.69E+08					0.0088%		
78	1.62E+08					0.0069%				

Day	Measured Atoms					Proportion of initial intake excreted per day				
	Vol. 1	Vol. 2	Vol. 3	Vol. 4	Vol. 5	Vol. 1	Vol. 2	Vol. 3	Vol. 4	Vol. 5
84				**1.21E+08**					**0.0055%**	
105			2.72E+08					0.0142%		
109		2.62E+08					0.0133%			
122	1.55E+08					0.0066%				
146	1.44E+08					0.0061%				
157			1.81E+08					0.0094%		
160		2.10E+08					0.0106%			
174	1.36E+08					0.0058%				
193			1.51E+08					0.0079%		
205	1.20E+08					0.0051%				
216		1.68E+08					0.0085%			
254	1.09E+08					0.0046%				
263		1.85E+08	1.30E+08				0.0094%	0.0067%		
290				**3.10E+08**					**0.0143%**	
294					**7.00E+07**					**0.0033%**
325	1.11E+08					0.0047%				
378		1.31E+08					0.0066%			
382			1.23E+08					0.0064%		
470					**1.30E+08**					**0.0061%**
471				**2.63E+08**					**0.0121%**	
483		1.61E+08					0.0081%			
484			1.02E+08					0.0053%		
521	9.69E+07					0.0041%				
648		1.43E+08					0.0073%			
651			1.01E+08					0.0053%		
664	7.88E+07					0.0034%				
743					**4.27E+07**					**0.0020%**
853	5.60E+07					0.0024%				
1111			8.40E+07					0.0044%		
1119			9.62E+07					0.0050%		
1139		9.90E+07					0.0050%			
1155	6.03E+07					0.0026%				
1236				**4.86E+07**					**0.0022%**	
1256					**6.40E+07**					**0.0030%**
1583			**7.33E+07**					**0.0038%**		
1584		**8.50E+07**					**0.0043%**			
1798		**7.50E+07**					**0.0038%**			
1801			**8.45E+07**					**0.0044%**		
1943	**1.16E+08**					**0.0050%**				
1970		**1.22E+08**					**0.0062%**			
2197	**6.12E+07**					**0.0026%**				
2224			**1.97E+07**					**0.0010%**		
2459		**6.25E+07**					**0.0032%**			
2667	**4.55E+07**					**0.0019%**				
2730			**4.98E+07**					**0.0026%**		
2927		**7.20E+07**					**0.0037%**			
3190	**6.75E+07**					**0.0029%**				
3219				**4.71E+07**	**4.80E+07**				**0.0022%**	**0.0023%**
4715			**5.10E+07**					**0.0027%**		

Day	Measured Atoms					Proportion of initial intake excreted per day				
	Vol. 1	Vol. 2	Vol. 3	Vol. 4	Vol. 5	Vol. 1	Vol. 2	Vol. 3	Vol. 4	Vol. 5
5426		**5.14E+07**					**0.0026%**			

3 THE AEAT/MU STUDY OF INTRAVENOUS INJECTION OF ^{237}Pu AND ^{244}Pu IN FEMALES

3.1 Outline of methodology

This work was initiated by AEAT but was subsequently continued by MU. It followed earlier work by AEAT in which six healthy men had received injections of ^{237}Pu (Talbot et al., 1993, 1997; Talbot and Newton, 1994; Warner et al., 1994; Newton et al., 1998).

Six healthy women (Table 3) received intravenous injections of ^{237}Pu and ^{244}Pu as Pu(IV) citrate; other details of the volunteers, including haematology, are given elsewhere (Talbot et al., 1997). Early patterns of excretion, retention in blood and uptake by liver were determined by measurements of the radioactivity of ^{237}Pu (Talbot et al., 1997; Newton et al., 1998). Later samples of excreta up to 8-9 years were analysed for ^{244}Pu by inductively-coupled plasma mass spectrometry (ICPMS), using the Micromass PlasmaTrace 2 double-focussing instrument operated by AEAT, or by accelerator mass spectrometry (AMS) performed by the Australian National University (ANU), Canberra (Priest et al., 2001). Further samples have extended the observations to 15 years. It was clear that ICPMS would be too insensitive for determining ^{244}Pu in blood in the long term, and only AMS was used.

3.2 Uncertainties

The uncertainties quoted in Tables 4-6 are those arising from the statistics of recorded events only. Other sources of potential error are as follows.

3.2.1 Stability of machine operation

With both ICPMS and AMS, the mass of ^{244}Pu present was determined relative to that of the ^{242}Pu, added to each sample in known quantity prior to processing. With either method, collection of the relevant ions of ^{244}Pu and ^{242}Pu did not occur simultaneously; typically there would be a series of cycles with alternating collections of each isotope. The validity of this procedure depends on how stable the ion currents are during measurement of a sample. Unstable operation could produce errors in estimates of ^{244}Pu in individual samples but these would be random in size and direction and so should not affect averaged results or trends (Newton et al., 2005) from sampling over a long period.

3.2.2 Isobaric interferences

AMS for ^{244}Pu is not prone to isobaric interferences, but with ICPMS complications can arise from lead/chlorine or thorium/carbon species formed in the plasma, with masses summing to

244; the requirements for chemical processing of the samples are consequently more stringent. It was therefore important to confirm, through ICPMS analysis of analytical blanks, that the separation had been effective; these samples were ^{244}Pu-free solutions with ^{242}Pu as the recovery tracer, which had been subjected to the same procedures as applied to the excretion samples using the same contemporary stock of reagents. The 85 excretion samples analysed by ICPMS (Tables 4 and 5) were processed in five batches at various times over a 9-months period. The blank test solutions processed with each batch all gave a positive "^{244}Pu" estimate. For two of the 41 urine samples this "background" amounted to >20% of the indicated sample content; this was true for 14 of the 44 faecal samples, which generally contained less ^{244}Pu than was present in contemporary urine.

The indicated contents of the relevant blank solutions were subtracted from the measured ^{244}Pu contents from ICPMS before the excretion rates in Tables 4 and 5 were calculated, and in most cases the result is assumed to be valid. Supporting evidence may be seen in Table 7, where averaged results by ICPMS for two subjects during the period 1½ - 3 years after the injection are compared with those obtained by AMS (where investigations showed no evidence for "background" effects attributable to the chemical content of excretion samples). However, the ICPMS blanks prepared for one particular batch of samples showed greater, and more variable, "^{244}Pu" contents than had their predecessors. The reason for this could not be established, but conceivably contamination with adventitious lead occurred between the ion-exchange removal of lead and preparation of the final solution for ICPMS. In this case the affected data, identified in Tables 4 and 5, could contain errors of unknown size and direction. However, in Table 4 none of the potentially affected urine samples shows a result grossly different from those found in others from the subject taken at about the same time; any such differences as may be discerned in Table 5 should be viewed in the context of the day-to-day variability of faecal excretion evident from unaffected results, both those based on ICPMS and AMS. For this reason, and because the potentially affected samples are relatively few in number and are confined to the first year after injection, their retention in analyses of the long-term trends in plutonium excretion by these subjects (Newton et al., 2005) would not change the outcome.

3.2.3 Contamination effects in AMS determinations

Positive estimates of ^{244}Pu in blank samples were obtained but, compared with those recorded with ICPMS, they were at levels much lower relative to the content of excretion samples, and were not due to other constituents of the sample. They arose instead from residual ^{244}Pu in the vicinity of the ion source, left after periodic checks of the efficiency of ion production in which a test sample, of much greater ^{244}Pu content than those under investigation, was used.

Under the operating conditions employed, analysis of four samples containing ^{242}Pu, but with no ^{244}Pu, gave estimated "^{244}Pu" contents of 0.3 ± 0.1 (Standard error on the mean, SEM) fg, and this value was subtracted from the indicated ^{244}Pu contents from AMS before the excretion rates or concentrations in blood were calculated. Contributions from the contamination were unimportant in relation to amounts present in excretion samples. The measured content of ^{244}Pu in faecal samples was never < 6 fg, and in only five of the 45 samples in Table 5 was it <10 fg. For urine (Table 4) the smallest value determined was 14 fg.

The situation is less satisfactory for some of the later blood samples, with ^{244}Pu contents much smaller than in contemporary excretion collections. The inferred interference level (0.3 fg) is

not strictly applicable to all samples examined during the intervals of several hours between performance checks, because the ^{244}Pu contamination around the ion source would reduce during that time. Consequently, the uncertainties displayed in Table 6, based solely on the statistics of recorded events, will in some cases underestimate the true values; samples for which the assumed interference (0.3 fg) amounts to >20% of the estimated ^{244}Pu content are identified in the table.

3.2.4 Indications of possible systematic error

Table 4 includes estimates of the fractional ^{244}Pu excretion by Subject J on six occasions between 20 and 84 d. These may be compared with the corresponding results for ^{237}Pu excretion (Talbot et al., 1997) determined by x-ray counting of the same samples. The mean ratio (^{244}Pu/^{237}Pu) was 1.02 ± 0.06 (SEM), i.e. as was to be expected, the isotopic ratio in urine was consistent with that in the injected mixture. By contrast, Table 8 compares concentrations in blood, sampled at 14-15 days, as determined by the two methods; here a mean ratio of 0.86 ± 0.03 is found.

An explanation may lie in the fact that the ^{242}Pu solutions used to spike the excretion collections and the blood samples were of different origins. The excretion collections were spiked at Harwell with ^{242}Pu calibrated by traceable reference to a solution provided by the UK National Physical Laboratory, while the blood samples were spiked by MU with a solution which, although derived from one with National Bureau of Standards certification, had undergone dilution elsewhere. The assumed calibration of the two solutions, if inconsistent, could account for the low ratios seen in Table 8, and the possibility should be borne in mind that all of the concentrations given in Table 6 underestimate the true values by ~16%.

3.3 Results

Tables 4-6 show amounts of ^{244}Pu in individual samples, measured by mass spectrometry; the data are expressed relative to the administered quantity, either as daily excretion rates or as concentrations in blood. Data based on x-ray counting of ^{237}Pu in samples taken during the first few months are tabulated elsewhere (Talbot et al., 1997).

Table 3 Female subjects, amounts of ^{244}Pu administered and periods of study

Subject	Age (y)	Injection (ng)	Period of study (y)
G	53	0.98	10.9
H	47	0.98	10.9
I	36	1.21	10.3
J	57	1.18	10.3
K	35	1.12	9.7
L	51	1.13	4.5

Table 4 ^{244}Pu in urine collections of females G - L (percent of injection per 24h) measured by AMS (Method A) or by ICPMS (Method P). Quoted uncertainties (±1σ) relate to statistics of recorded events.

Time d	Duration h	Content % d^{-1} ± 1σ	Method	Time d	Duration h	Content % d^{-1} ± 1σ	Method
		Female G				**Female H**	
172*	***24***	***0.0083 ± 0.0003***	***P***	157	21	0.0057 ± 0.0002	P
173	24	0.0062 ± 0.0003	P	158	17	0.0045 ± 0.0002	P
292	24	0.0050 ± 0.0003	P	159	28	0.0039 ± 0.0003	A
293	24	0.0057 ± 0.0003	A	286	24	0.0038 ± 0.0002	P
514	24	0.0034 ± 0.0004	A	288	24	0.0047 ± 0.0002	P
516	24	0.0022 ± 0.0002	A	508	26	0.0021 ± 0.0001	P
517	24	0.0032 ± 0.0001	P	509	23	0.0039 ± 0.0003	A
518	24	0.0034 ± 0.0002	P	510	24	0.0034 ± 0.0001	P
727	24	0.0025 ± 0.0002	P	511	24	0.0026 ± 0.0002	A
728	24	0.0034 ± 0.0002	P	705	24	0.0029 ± 0.0001	P
729	24	0.0031 ± 0.0001	P	706	24	0.0030 ± 0.0001	P
942	24	0.0033 ± 0.0001	P	707	24	0.0035 ± 0.0001	P
943	24	0.0036 ± 0.0002	P	708	24	0.0028 ± 0.0001	P
944	24	0.0032 ± 0.0002	P	709	24	0.0039 ± 0.0002	P
945	24	0.0036 ± 0.0002	P	921	24	0.0046 ± 0.0001	P
1196	24	0.0034 ± 0.0004	A	922	24	0.0036 ± 0.0004	A
1197	24	0.0039 ± 0.0003	A	924	17	0.0038 ± 0.0003	P
1616	24	0.0030 ± 0.0002	A	1153	24	0.0033 ± 0.0003	A
3221	72	0.0016 ± 0.0001	A	1154	24	0.0041 ± 0.0003	A
3644	71	0.0010 ± 0.0001	A	1657	72	0.0016 ± 0.0001	A
3994	72	0.0011 ± 0.0001	A	3204	104	0.0017 ± 0.0001	A
				3616	103	0.0013 ± 0.0001	A
				3978	97	0.0038± 0.0002	A
		Female I				**Female J**	
155	24	0.0078 ± 0.0002	P	20.5	24	0.0439 ± 0.0022	A
156	24	0.0099 ± 0.0007	A	21.5	22	0.0370 ± 0.0012	A
157*	***24***	***0.0085 ± 0.0002***	***P***	22.4	23	0.0407 ± 0.0020	A
306	24	0.0037 ± 0.0002	P	39	24	0.0183 ± 0.0008	A
307	24	0.0043 ± 0.0002	P	83	25	0.0076 ± 0.0007	A

Time d	Duration h	Content % d^{-1} ± 1σ	Method	Time d	Duration h	Content % d^{-1} ± 1σ	Method
308	22	0.0045 ± 0.0004	A	84	26	0.0097 ± 0.0002	P
563	24	0.0040 ± 0.0004	A	157	23	0.0063 ± 0.0002	P
564	26	0.0051 ± 0.0001	P	159	24	0.0050 ± 0.0002	P
764	24	0.0050 ± 0.0003	A	160	23	0.0055 ± 0.0005	A
765	24	0.0041 ± 0.0003	A	303	24	0.0042 ± 0.0003	A
1033	24	0.0034 ± 0.0004	A	305	23	0.0042 ± 0.0002	P
1034	24	0.0043 ± 0.0005	A	306	25	0.0038 ± 0.0002	P
1278	73	0.0024 ± 0.0001	A	552	24	0.0036 ± 0.0003	A
1723	73	0.0014 ± 0.0001	A	553	25	0.0028 ± 0.0001	P
2897	71	0.0019 ± 0.0001	A	799	25	0.0068 ± 0.0003	A
3390	88	0.0020 ± 0.0002	A	800	24	0.0044 ± 0.0003	A
3753	70	0.0021 ± 0.0002	A	801	24	0.0044 ± 0.0004	A
				1065	25	0.0044 ± 0.0004	A
				1066	24	0.0042 ± 0.0003	A
				1333	73	0.0024 ± 0.0001	A
				1863	72	0.0023 ± 0.0001	A
				3743	79	0.0018 ± 0.0002	A
		Female K				**Female L**	
188	26	0.0038 ± 0.0004	A	***179****	***27***	***0.0071 ± 0.0003***	***P***
189*	***22***	***0.0042 ± 0.0001***	***P***	***180****	***23***	***0.0065 ± 0.0002***	***P***
190	24	0.0051 ± 0.0001	P	333	23	0.0041 ± 0.0003	A
353	25	0.0031 ± 0.0003	A	334	27	0.0039 ± 0.0002	A
355	25	0.0069 ± 0.0002	P	593	24	0.0069 ± 0.0047	A
588	24	0.0031 ± 0.0004	A	594	23	0.0041 ± 0.0006	A
589	24	0.0023 ± 0.0003	A	796	24	0.0027 ± 0.0003	A
797	24	0.0024 ± 0.0003	A	797	22	0.0026 ± 0.0004	A
798	24	0.0013 ± 0.0002	A	1139	72	0.0019 ± 0.0001	A
1090	71	0.0015 ± 0.0001	A	1651	71	0.0036 ± 0.0002	A
1651	77	0.0018 ± 0.0001	A				
2805	47	0.0018 ± 0.0001	A				
3183	72	0.0013 ± 0.0001	A				
3538	49	0.0014 ± 0.0001	A				

Time d	Duration h	Content % d^{-1} ± 1σ	Method	Time d	Duration h	Content % d^{-1} ± 1σ	Method

**** Result in bold italics possibly affected by contamination with lead (see text)***

Table 5 ^{244}Pu in faeces collections of females G – L (percent of injection per 24h) measured by AMS (Method A) or by ICPMS (Method P). Quoted uncertainties (±1σ) relate to statistics of recorded events.

Time d	Duration h	Content % d^{-1} ± 1σ	Method	Time d	Duration h	Content % d^{-1} ± 1σ	Method
		Female G				**Female H**	
172	21	0.0089 ± 0.0005	P	157	24	0.0019 ± 0.0003	P
173	49	0.0044 ± 0.0006	A	***158****	***24***	***0.0023 ± 0.0001***	***P***
175	23	0.0089 ± 0.0004	P	159	48	0.0015 ± 0.0002	A
291	24	0.0025 ± 0.0004	A	285	24	0.0043 ± 0.0011	A
293	48	0.0025 ± 0.0002	P	286	24	0.0030 ± 0.0003	A
294*	***23***	***0.0045 ± 0.0002***	***P***	287	23	0.0016 ± 0.0003	P
296	47	0.0037 ± 0.0004	A	507	23	0.00076 ± 0.00038	A
517	25	0.0018 ± 0.0002	P	508	25	0.0015 ± 0.0002	P
519	48	0.0015 ± 0.0003	A	510	48	0.00080 ± 0.00004	P
520	23	0.0017 ± 0.0002	A	512	50	0.00069 ± 0.00007	A
521	25	0.0025 ± 0.0002	P	706	24	0.00053 ± 0.00017	P
727	24	0.0028 ± 0.0003	P	707	35	0.00057 ± 0.00005	P
728	24	0.00092 ± 0.00008	P	708	16	0.0021 ± 0.0002	P
729	24	0.0027 ± 0.0001	P	709	29	0.0011 ± 0.0001	P
730	24	0.0019 ± 0.0002	P	710	27	0.00092 ± 0.00018	P
944	24	0.0024 ± 0.0001	P	920	24	0.00017 ± 0.00007	P
945	24	0.0032 ± 0.0002	A	921	21	0.00081 ± 0.00012	P
946	24	0.0020 ± 0.0001	P	922	33	0.0014 ± 0.0002	A
1196	24	0.00056 ± 0.00018	A	923	13	0.0012 ± 0.0001	A
1624	70	0.00084 ± 0.00015	A	924	33	0.0020 ± 0.0002	P
3221	98	0.00078 ± 0.00018	A	1657	79	0.0004 ± 0.0004	A
3644	97	0.00054 ± 0.00019	A	3204	96	0.00066 ± 0.00015	A
3994	97	0.00050 ± 0.00014	A	3616	102	0.0010 ± 0.0001	A
				3978	91	0.0017± 0.0002	A
		Female I				**Female J**	

Time d	Duration h	Content % d^{-1} ± 1σ	Method	Time d	Duration h	Content % d^{-1} ± 1σ	Method
155	24	0.0069 ± 0.0002	P	82	36	0.0080 ± 0.0007	A
157	52	0.0030 ± 0.0002	P	***157****	***24***	***0.0036 ± 0.0002***	***P***
305	27	0.0035 ± 0.0002	P	158	36	0.0062 ± 0.0005	P
306	20	0.0061 ± 0.0002	A	159	11	0.0149 ± 0.0009	P
307	24	0.0049 ± 0.0007	A	302	45	0.00032 ± 0.00004	A
562	24	0.0047 ± 0.0003	A	304	33	0.0018 ± 0.0002	P
563	26	0.0085 ± 0.0003	A	306	39	0.0018 ± 0.0003	A
564	27	0.0033 ± 0.0002	P	307	24	0.0048 ± 0.0003	P
765	92	0.00076 ± 0.00039	A	554	13	0.0018 ± 0.0002	P
1033	76	0.0016 ± 0.0001	A	1066	69	0.00079 ± 0.00015	A
1279	71	0.0029 ± 0.0003	A	1332	58	0.0019 ± 0.0002	A
1720	123	0.00037 ± 0.00009	A	1863	72	0.0047 ± 0.0007	A
2897	69	0.0017 ± 0.0002	A	3743	58	0.00053 ± 0.00007	A
3390	74	0.0007 ± 0.0003	A				
3753	82	0.0009 ± 0.0001	A				
		Female K				**Female L**	
187	24	0.0028 ± 0.0002	P	180	23	0.0066 ± 0.0003	A
188	26	0.0035 ± 0.0002	P	***181****	***23***	***0.0035 ± 0.0002***	***P***
190*	***45***	***0.0026 ± 0.0002***	***P***	182	11	0.0075 ± 0.0002	P
354	22	0.0011 ± 0.0001	P	***333****	***24***	***0.0035 ± 0.0001***	***P***
355*	***23***	***0.0020 ± 0.0001***	***P***	334	24	0.0053 ± 0.0003	A
356	28	0.0013 ± 0.0001	P	335	25	0.0023 ± 0.0003	A
530	101	0.00074 ± 0.00011	A	552	74	0.00072 ± 0.00006	A
797	22	0.0018 ± 0.0004	A	797	44	0.0020 ± 0.0006	A
1091	70	0.00032 ± 0.00011	A	1138	102	0.00081 ± 0.00017	A
1651	95	0.00086 ± 0.00018	A	1651	72	0.0012 ± 0.0003	A
2805	96	0.00073 ± 0.00008	A				
3538	74	0.00100 ± 0.00016	A				

**** Result in bold italics possibly affected by contamination with lead (see text)***

Table 6 ^{244}Pu in blood from females G – L (percent of injection per kg whole blood) measured by AMS. Quoted uncertainties (1σ) relate to statistics of recorded events.

Time d	Mass of blood measured g	Concentration % kg^{-1} ± 1σ	Time d	Mass of blood measured g	Concentration % kg^{-1} ± 1σ
Female G			**Female H**		
14	21.8	0.737 ± 0.033	14	21.5	0.316 ± 0.019
21	21.1	0.583 ± 0.029	49	21.5	0.109 ± 0.015
49	21.5	0.192 ± 0.019	86	21.0	0.019 ± 0.007
87	21.2	0.099 ± 0.010	161	21.6	0.029 ± 0.007
151	21.4	0.062 ± 0.018	289	21.7	0.017 ± 0.008
326	21.6	0.013 ± 0.006	508	19.7	0.013 ± 0.002
543	21.0	0.022 ± 0.003	710	44.0	0.012 ± 0.002
710	43.6	0.015 ± 0.002	1157	41.6	0.009 ± 0.002
1208	42.9	0.011 ± 0.002	1868	44.6	0.010 ± 0.001
1705	36.3	0.019 ± 0.003	***3201****	***19.3***	***0.0043 ± 0.0026***
3202	17.8	0.010 ± 0.003	***3201**†*	***20.3***	***0.0026 ± 0.0020***
***3202**†*	***14.9***	***0.006 ± 0.003***			
Female I			**Female J**		
15	22.3	0.329 ± 0.015	15	21.3	0.186 ± 0.016
42	21.9	0.076 ± 0.006	44	21.4	0.077 ± 0.006
84	21.8	0.036 ± 0.004	78	21.5	0.027 ± 0.002
154	21.9	0.027 ± 0.003	305	21.2	0.018 ± 0.004
304	21.1	0.016 ± 0.003	569	21.0	0.039 ± 0.005
556	21.4	0.019 ± 0.005	826	21.2	0.010 ± 0.003
791	20.8	0.010 ± 0.006	1501	41.1	0.013 ± 0.002
1736	21.3	0.009 ± 0.002	1815	21.2	0.016 ± 0.006
2891*	***16.2***	***0.0072 ± 0.0026***			
***2891**†*	***17.4***	***0.0063 ± 0.0028***			
Female K			**Female L**		
14	21.4	0.387 ± 0.035	14	21.3	0.477 ± 0.025
44	21.8	0.095 ± 0.007	44	21.3	0.063 ± 0.004
106	21.1	0.032 ± 0.004	172	21.3	0.032 ± 0.007
604	21.4	0.0072 ± 0.029	336	21.9	0.015 ± 0.003

Time d	Mass of blood measured g	Concentration % kg^{-1} ± 1σ	Time d	Mass of blood measured g	Concentration % kg^{-1} ± 1σ
1172*	***19.6***	***0.0046 ± 0.0029***	1142	42.1	0.009 ± 0.002
1662	42.6	0.012 ± 0.003	1654	43.5	0.010 ± 0.002
2821*	***13.0***	***0.0056 ± 0.0027***			

**** Uncertainty possibly underestimated (see text)***
† Duplicate samples taken

Table 7 Mean 24-hour urinary excretion rates (percent of injection) in specified periods, showing consistency of analyses by ICPMS and AMS.

Subject	ICPMS				AMS			
	days*	n†	%	SEM	days*	n†	%	SEM
G	777 (517-945)	9	0.0032	0.0001	856 (514-1197)	4	0.0032	0.0003
H	711 (508-924)	9	0.0033	0.0002	850 (509-1154)	5	0.0035	0.0003

* mean and range
† number of samples in period

Table 8 Concentrations in early blood samples determined both by AMS for ^{244}Pu and by x-ray counting of ^{237}Pu in unprocessed samples: evidence of bias. Quoted uncertainties (1σ) relate to statistics of recorded events.

Subject	Days	Concentration (% injection kg^{-1})		Ratio (AMS/x rays)
		AMS	x rays*	
G	14	0.737 ± 0.033	0.99 ± 0.02	0.74 ± 0.04
H	14	0.316 ± 0.019	0.33 ± 0.02	0.96 ± 0.08
I	15	0.329 ± 0.015	0.41 ± 0.02	0.80 ± 0.05
J	15	0.186 ± 0.016	0.22 ± 0.04	0.84 ± 0.17
K	14	0.387 ± 0.035	0.43 ± 0.01	0.90 ± 0.09
L	14	0.477 ± 0.025	0.53 ± 0.03	0.90 ± 0.07
mean ± SEM				0.86 ± 0.03

* from Talbot et al. (1997)

4 THE CRCE STUDY OF INHALATION OF ^{237}Pu AND ^{244}Pu IN MALES

4.1 Outline of methodology

An outline of experimental methods is given here. Full details are presented by Puncher and Etherington (2017). A mixed ^{237}Pu/^{244}Pu nitrate aerosol was generated from a 0.5% sodium nitrate/0.01M nitric acid solution containing these tracers. The nebulised droplets dried rapidly to produce an aerosol of 1.1 μm mass median aerodynamic diameter and geometric standard deviation (σ_g) ~ 1.2. Two healthy male volunteers (Subjects C and D) inhaled the aerosol with a breathing pattern designed to maximise alveolar deposition. Initial lung deposits were approximately 8 kBq ^{237}Pu and 35 ng ^{244}Pu (Table 9).

Retention of ^{237}Pu in each lung and uptake by the liver were measured by external counting of the x-ray emissions from ^{237}Pu with 50 mm diameter semiconductor detectors mounted in pairs, up to about 120 d after inhalation. Measurements of ^{237}Pu in the head (to determine retention in the skeleton) were in all cases below the detection limit and are not discussed further in this report. Count times ranged from 10 minutes for the early measurements up to 30 minutes for the later measurements. Detectors were well-collimated, each having a field of view of about 16 cm diameter at a distance of 10 cm from the collimator face. Nevertheless, ^{237}Pu activity measured with detectors positioned over the lungs included a contribution from ^{237}Pu in the liver, and vice versa. A matrix method for correcting for activity measured from adjacent organs (Smith et al., 2008) was used to determine corrections to the measured ^{237}Pu lung and liver activities. The effectiveness of these corrections, and of the detector collimation, is demonstrated by the fact that the corrected liver activity extrapolated back to the time of inhalation is close to zero for both subjects. All *in vivo* measurements of ^{237}Pu were decay-corrected to the time of inhalation.

Bioassay (blood, urine and faeces) samples were collected regularly from both volunteers from the beginning of the experiment, but at increasing intervals as time progressed. Sample collections continued for 5.9 y and 13.4 y for Subjects C and D respectively. The blood sample volume at each time point was about 20 ml and the mass of each sample was also measured. Gamma spectrometry measurements of ^{237}Pu content were made on blood samples up to 12 days after inhalation (day 12) for Subject C and up to day 15 for Subject D. Individual urine samples were taken over the first 24 – 48 h, and continuous 24-h urine collections were then made until day 25 for Subject D and until day 32 for Subject C. Subsequently, bulked 3-day or 5-day samples were collected. Individual faeces samples were collected up to day 38 from both volunteers, after which bulked 3-day or 5-day samples were collected. Gamma spectrometry measurements of ^{237}Pu content were made on all urine and faeces samples up to day 101 and day 114 for Subject C and day 102 and day 108 for Subject D respectively. After these times, the ^{237}Pu was below the detection limit. All measurements of ^{237}Pu in blood, urine and faeces samples were decay-corrected to the time of inhalation.

Most blood samples and limited numbers of urine and faeces samples covering the entire time period of the experiments were selected for accelerator mass spectrometry (AMS) analysis. These included some samples previously measured by gamma spectrometry to provide a comparison of the results from the two measurement techniques. A known quantity of ^{242}Pu tracer solution was added to the samples for mass spectrometry analysis. For the blood and urine samples, the tracer was added before they were processed in the laboratory. For the

faeces samples, however, it was considered that the tracer might not distribute evenly throughout the sample if it was added to fresh material, and so tracer was usually added after the initial ashing of each sample. Processing of some of the faeces samples was suspended after the initial ashing and so some samples were stored without any tracer being added. No background subtraction was applied to the AMS results for blood, urine and faeces samples since no isobaric interferences are expected with AMS as noted earlier, and any background levels arising from contamination effects were expected to be insignificant compared with the levels of ^{244}Pu present in the samples.

It was not possible to measure all of the samples, partly due to the cost involved. To date, only a small number of faeces samples have been measured by mass spectrometry, because of the difficulty in effectively dissolving the faecal material. The majority of the samples were measured by AMS at the Australian National University (ANU). However, for comparison purposes, an aliquot of one sample was measured by resonance ion mass spectrometry at the Johannes Gutenberg University at Mainz, Germany and several samples were measured using a Multi-Collector ICPMS system at the British Geological Survey (BGS), Nottingham, UK.

Measurements of the Pu content of the 20 ml blood samples were used to estimate total blood content by multiplying the measured sample content by the ratio of estimated total blood mass to the measured sample mass. The total blood masses (4.2 kg and 4.3 kg for Subject C and Subject D respectively) were estimated for the two volunteers from their weights and heights (Talbot et al., 1997). However, it is possible that actual masses may differ substantially from such predictions (Nadler et al., 1962). The value (4.3 kg) adopted here for Subject D may be compared with two identical observations of 5.4 kg made 20 years previously using isotope dilution, one from injection of ^{131}I-labelled human serum albumin (HSA) and the other from re-injection of his red cells after incubation in ^{203}Pb. Blood mass tends to decline with age (Davy & Seals 1994), but over the relevant interval of 20 years the reduction is estimated to be only ~10% (Williams 1994). On that basis, an adjusted estimate of ~4.9 kg for Subject D may be indicated. Given the time delay between that assessment and the inhalation experiment, and since there was no assessment by isotopic dilution for Subject C, the estimates derived from weight and height data have been used here. Sufficient data are presented here to allow alternative total blood mass estimates to be applied.

Details of early analysis of *in vivo* measurements and bioassay samples up to 740 d after inhalation are given in Etherington et al. (2002) and Etherington et al. (2003). Results of what is currently expected to be the final analysis of the study are given in Puncher and Etherington (2017).

4.2 Uncertainties

The uncertainties in the *in vivo* measurements quoted in Tables 10 and 11 are those arising from counting statistics only. *In vivo* measurement uncertainties arising from variability in detector positioning are discussed in section 4.2.1. Uncertainties in sample bioassay measurements quoted in Tables 12 – 17 were obtained from assessments of total uncertainty (i.e. the uncertainty in the measured quantity arising from all known significant causes and determined by means of an uncertainty budget). Uncertainties in AMS measurements related to stability of machine operation, isobaric interferences and contamination effects are discussed in sections 3.2.1 – 3.2.3. Investigations of potential systematic errors in the mass

spectrometry and gamma spectrometry bioassay sample measurements are discussed in sections 4.2.2 and 4.2.3.

4.2.1 *In vivo* measurements of ^{237}Pu lung retention and liver uptake

In order to assess the intra-subject variability of individual measurements of retention and uptake, duplicate measurements separated by a short time interval were performed at later times, when the daily rate of change of retention or uptake was a relatively small fraction of the initial lung deposit (ILD). Between each measurement, the subject was allowed to reposition himself or remove himself from the detector system, after which the detector arrays and subject were repositioned in their standard positions. Differences between the duplicate measurements relative to the 1-sigma or 2-sigma confidence interval on the individual measurements provide an indication of the variability arising from variations in the positioning of the subject relative to the detectors. For Subject C, the duplicate lung retention measurements were performed at days 26, 65, 84, 104 and 119, while the duplicate liver uptake measurements were performed at days 26, 65, 104 (3 measurements) and 119 (Table 10). For Subject D, the duplicate lung retention and liver uptake measurements were performed at days 57, 85, 97 and 112 (Table 10).

For the lung measurements on Subjects C and D, differences were distributed within the range expected from counting statistics. Five out of the 10 pairs of duplicate measurements differed by less than the 1-sigma confidence interval on the individual measurements (±1.4% – ±2.3%, expressed in units of %ILD; measured lung retention at the times of the duplicate measurements was in the range 50 – 75 %ILD). The percentage of pairs of measurements that are expected to differ by less than the 1-sigma confidence interval where no difference is present is 53%*. Eight out of the 10 pairs of duplicate measurements differed by less than the 2-sigma confidence interval on the individual measurements. The percentage of pairs of measurements that are expected to differ by less than the 2-sigma confidence interval where no difference is present is 85% *. Thus, variability resulting from detector positioning over the lungs is judged to be not significant.

For the liver measurements on Subjects C and D, some differences exceeded the range expected from counting statistics. Only two out of the 9 pairs of duplicate measurements differed by less than the 1-sigma confidence interval on the individual measurements (±1.2% – ±2.6%, expressed in units of %ILD; measured liver uptake at the times of the duplicate measurements was in the range 16 – 35 %ILD). Six out of the 10 pairs of duplicate measurements differed by less than the 2-sigma confidence interval on the individual measurements. As noted above, the percentages of pairs of measurements that are expected to differ by less than the 1-sigma and 2-sigma confidence intervals where no difference is present are 53% and 85% respectively *. Thus, variability resulting from detector positioning over the liver was judged to be significant in some cases (specifically, day 26 for Subject C, and days 97 and 112 for Subject D).

* Determined by Monte Carlo simulation

4.2.2 Comparison of measurements made with alternative mass spectrometric techniques

A small number of measurements of ^{244}Pu in bioassay samples was performed using the Multi-Collector ICPMS (MC-ICPMS) system at the British Geological Survey (BGS), with the aim of providing comparisons and an independent confirmation of the AMS results using an alternative mass spectrometry system. BGS had not previously performed MC-ICPMS measurements of Pu and so CRCE provided five concentrated samples of ^{242}Pu for calibration purposes. MC-ICPMS measurements were made on aliquots from eight samples as well as on a single reagent blank sample. The reagent blank should not have contained Pu, but counts from "^{244}Pu" (i.e. mass number 244) and to a much lesser extent "^{242}Pu" (i.e. mass number 242) were detected. As indicated in section 3.2.2, the combined presence of thorium and carbon in the form of ^{232}Th and ^{12}C can cause an isobaric interference at mass number 244. The ^{232}Th content was therefore measured in the reagent blank and in the samples and corrections for the isobaric interference at mass number 244 applied, after which background subtractions were performed using the reagent blank. The ratio of the ^{244}Pu content to the ^{242}Pu tracer content in each sample was then determined. The uncertainty on the ^{244}Pu:^{242}Pu ratio was evaluated from the root mean square of the measurement uncertainty and the uncertainty on the reagent blank subtraction (the latter assumed by BGS to be 20%).

Aliquots of one Subject D urine sample were analysed independently at BGS and ANU. The ^{244}Pu masses estimated by BGS and ANU were $4.37 \times 10^{-11} \pm 0.11 \times 10^{-11}$g and $4.40 \times 10^{-11} \pm 0.23 \times 10^{-11}$g respectively, indicating good agreement. The remaining BGS results were consistent with the general trend in the ANU data with time after inhalation, and so the MC-ICPMS results for all eight samples were included together with the AMS results.

4.2.3 Indications of possible systematic error

Comparisons of the quoted ^{244}Pu:^{237}Pu isotopic ratios for the inhaled Pu nitrate aerosol (Table 9) with the isotopic ratios determined from mass spectrometry and gamma spectrometry measurements on blood, urine and faeces samples (decay-corrected to time of inhalation) could potentially indicate whether significant systematic errors are associated with one or other of the measurement techniques (see section 3.2.4). The comparison is of course limited to the small number of bioassay samples that were measured with both techniques.

For Subject C, the isotopic ratios measured in blood (nine samples), urine (four samples) and faeces (one sample) (mean ± standard deviation (1σ)) were $5.1\text{x}10^{-12} \pm 1.3\text{x}10^{-12}$ g Bq^{-1}, $4.7\text{x}10^{-12} \pm 1.80\text{x}10^{-12}$ g Bq^{-1} and $4.7\text{x}10^{-12}$ g Bq^{-1} respectively. For Subject D, the isotopic ratios measured in blood (10 samples), urine (6 samples) and faeces (one sample) (mean ± standard deviation (1σ)) were $4.4\text{x}10^{-12} \pm 1.1\text{x}10^{-12}$ g Bq^{-1}, $4.5\text{x}10^{-12} \pm 1.1\text{x}10^{-12}$ g Bq^{-1} and $4.2\text{x}10^{-12}$ g Bq^{-1} respectively. For blood and urine, these values are the means and standard deviations of the measured ratio for each sample. The standard deviations on the distributions of the measured ratios are greater (by a factor of 2 – 3) than would be expected from the evaluated uncertainties on each measurement. This may indicate that the total uncertainties associated with one or other (or both) of the measurement techniques have been underestimated, and for this reason SEM values are not quoted. Taking into account the relatively large observed variability in the measured ratio between bioassay samples, the mean values of the measured ratio are judged to be consistent with the quoted values for the

inhaled aerosol for both Subject C and Subject D shown in Table 9 (4.23×10^{-12} g Bq^{-1} and 4.77×10^{-12} g Bq^{-1} respectively).

4.3 Results

Table 9 provides information on the age of the subjects, the duration of the aerosol administration, the estimated ILD for each subject, the ^{244}Pu:^{237}Pu isotopic ratio at the time of inhalation, and the duration of each study (based on the dates of the final collections of blood, urine and faeces samples reported here). ILDs for Subjects C and D were determined from the intercepts at the time of the mid-point of the inhalation period (t=0) of a 3-component multi-exponential fit to the lung retention data. These ILD estimates are expected to be reliable, since good fits were obtained to each of the three initial lung retention measurements (at t = 0.5 h, 1.6 h and 3.9 h for Subject C, and t = 1.0 h, 1.8 h and 5.0 h for Subject D).

Tables 10 and 11 list the results of the ^{237}Pu gamma spectrometry measurements performed on the lungs and liver of subjects C and D respectively over the initial 3 – 4 months of the experiment. All ^{237}Pu measurements were decay-corrected to the time of inhalation.

Table 12, for Subject C, and Table 13, for Subject D, provide results for the blood content of Pu at different times up to the end of the study, determined from decay-corrected ^{237}Pu gamma spectrometry and/or ^{244}Pu mass spectrometry measurements made on weighed intravenous blood samples taken at each time point. The ^{237}Pu activity in total blood (3rd column) was estimated from the measured ^{237}Pu content of the 20 ml blood samples using the measured sample mass (2nd column) and the estimated total blood mass, as discussed in section 4.1. The number of ^{244}Pu atoms in selected samples was determined from mass spectrometric measurements (4th column), and the “equivalent ^{237}Pu activity” (5th column) was determined using the known ^{237}Pu:^{244}Pu ratio in the inhaled material (given in Table 9). Where both ^{237}Pu gamma spectrometry and ^{244}Pu mass spectrometry measurements were made on a particular sample, a weighted mean value of Pu content in total blood is given, in terms of the equivalent ^{237}Pu activity and as a percentage of the ILD (6th and 7th columns). Weighting factors were determined from the variances on each measurement.

Tables 14 and 16 show results for the daily excretion of Pu in urine and faeces over the duration of the experiment for Subject C. Tables 15 and 17 show the same information for Subject D. The results are determined from decay-corrected ^{237}Pu gamma spectrometry and/or ^{244}Pu mass spectrometry measurements. Early excretion results are determined from measurements of the Pu content of single voidings. Subsequent results are determined from measurements of 24-hour collections whilst the later results are determined from measurements of bulk collections undertaken over a number of days. For each sample, the start, end and duration of the sample period is given (1st, 2nd and 3rd columns). Also shown are the measured ^{237}Pu content of each sample (4th column), the number of ^{244}Pu atoms measured in selected samples (5th column), the “equivalent ^{237}Pu activity” for those samples (6th column), and a weighted mean value of Pu content (7th column), determined as described above for blood sample measurements. The final column shows the daily excretion of Pu, expressed as a percentage of the ILD per day.

Table 9 Male subjects, date and duration of aerosol administration, amounts of ^{237}Pu and ^{244}Pu deposited, and duration of study

Subject	Age at administration (y)	Administration date	Duration of administration (minutes)	Calculated initial lung deposit[A]		Isotopic ratio of ^{244}Pu : ^{237}Pu on date of inhalation		Duration of study (y)
				^{237}Pu (Bq)	^{244}Pu (g)	g/Bq	atoms/Bq	
C	73	26/11/1997	27	7739.2	3.27E-08	4.23E-12	1.04E+10	5.9
D	60	4/12/1997	24	7578.8	3.62E-08	4.77E-12	1.18E+10	13.4

[A] Estimated from intercept at t=0 of fit to lung retention data (see text)

Table 10 Lung and liver content, Subject C, gamma spectrometry measurements. Amounts expressed as Bq of ^{237}Pu and as a percentage of the initial lung deposit of ^{237}Pu (ILD)[1]. Quoted uncertainties (±1σ) relate to counting statistics only.

Lungs			Liver		
Time (d)	Activity (Bq)	% of ILD[1]	Time (d)	Activity (Bq)	% of ILD[1]
0.03	7845±150	101.37±1.94	0.02	723±59	9.34±0.76
0.06	7974±150	103.03±1.94	0.07	270±33	3.49±0.43
0.19	7395±108	95.55±1.40	0.16	393±34	5.08±0.44
0.32	7269±100	93.92±1.29	NM [2]	NM	NM
0.35	7172±101	92.67±1.31	0.34	818±45	10.57±0.58
0.84	6936±99	89.62±1.28	0.85	1185±55	15.31±0.71
1.08	6969±102	90.05±1.32	1.07	1056±52	13.64±0.67
1.90	6427±97	83.04±1.25	1.89	916±53	11.84±0.68
3.92	6310±100	81.53±1.29	3.91	1231±61	15.91±0.79
4.90	6083±99	78.60±1.28	4.93	1404±64	18.14±0.83
6.85	6015±101	77.72±1.30	6.84	864±56	11.16±0.73
8.93	6310±124	81.53±1.61	8.94	1318±68	17.03±0.87
11.89	5858±119	75.69±1.54	11.90	1588±70	20.52±0.91
13.93	5646±137	72.95±1.76	13.94	1949±81	25.18±1.05
15.90	6010±108	77.66±1.39	15.89	1630±73	21.06±0.94
25.90	5866±110	75.80±1.42	25.89	2168±97	28.01±1.25
25.95	5571±109	71.98±1.41	25.95	1574±93	20.34±1.20
43.91	5391±118	69.66±1.52	43.89	2420±137	31.27±1.77
64.89	5143±118	66.45±1.53	64.88	2094±116	27.06±1.49
64.96	5392±120	69.67±1.55	64.97	1882±103	24.32±1.33
83.94	5018±127	64.84±1.65	83.89	2571±137	33.22±1.77
83.95	4782±126	61.79±1.63	NM	NM	NM
103.90	4965±145	64.15±1.88	103.89	2440±155	31.53±2.00
103.97	4960±152	64.09±1.97	103.98	2528±166	32.66±2.15
103.99	4891±152	63.20±1.97	104.02	2436±159	31.48±2.05
118.92	4662±169	60.24±2.18	118.90	2970±202	38.38±2.62
118.97	4575±174	59.11±2.24	118.99	2737±202	35.37±2.61

Notes
1. Initial lung deposit (ILD) for Subject C = 7739.2 Bq
2. NM = Not measured

Table 11 Lung and liver content, Subject D, gamma spectrometry measurements. Amounts expressed as Bq of ^{237}Pu and as a percentage of the initial lung deposit of ^{237}Pu (ILD)[1]. Quoted uncertainties (±1σ) relate to counting statistics only.

Lungs			Liver		
Time (d)	Activity (Bq)	%ILD[1]	Time (d)	Activity (Bq)	%ILD[1]
0.04	7457±147	98.39±1.94	NM[2]	NM	NM
0.08	7336±148	96.80±1.95	NM	NM	NM
0.21	7489±107	98.82±1.41	0.20	84±12	1.11±0.16
0.29	7099±104	93.67±1.37	0.30	225±23	2.97±0.31
0.38	6875±104	90.71±1.37	0.37	233±25	3.07±0.33
0.96	6585±101	86.89±1.33	0.95	255±29	3.36±0.38
1.20	6505±102	85.83±1.34	1.19	294±26	3.88±0.35
2.06	6316±98	83.34±1.29	2.05	445±30	5.87±0.39
3.00	6409±99	84.56±1.31	2.99	540±34	7.13±0.45
3.96	6120±98	80.75±1.29	3.95	523±41	6.90±0.54
5.96	6054±101	79.88±1.33	5.95	786±43	10.37±0.56
7.96	5817±100	76.75±1.32	7.95	919±49	12.13±0.65
10.97	5745±103	75.80±1.35	10.96	900±52	11.88±0.68
12.97	5270±107	69.54±1.41	12.94	973±54	12.84±0.71
18.14	5685±105	75.01±1.38	18.13	847±58	11.18±0.77
35.17	4895±167	64.59±2.20	35.15	1184±114	15.62±1.50
43.14	4922±117	64.94±1.54	43.13	1215±78	16.03±1.03
56.15	4469±126	58.97±1.67	56.14	1205±94	15.90±1.24
56.66	4312±124	56.90±1.64	57.14	1396±106	18.42±1.40
84.99	4061±125	53.58±1.66	84.97	1513±107	19.96±1.41
85.04	3944±137	52.04±1.80	85.05	1308±100	17.26±1.32
96.99	4078±142	53.81±1.88	97.00	1535±125	20.25±1.65
97.00	3940±127	51.99±1.67	97.03	1144±99	15.09±1.31
111.99	3962±146	52.28±1.92	111.97	1479±161	19.52±2.13
112.05	4094±158	54.02±2.09	112.06	870±115	11.48±1.52

Notes
1. Initial lung deposit (ILD) for Subject D = 7578.8 Bq
2. NM = Not measured

Table 12 Blood content, Subject C, gamma and mass spectrometry measurements. Amounts expressed as number of ^{244}Pu atoms, Bq of ^{237}Pu or as a percentage of the initial lung deposit of ^{237}Pu (ILD)[1]. Quoted uncertainties for ^{237}Pu measurements (±1σ) are total uncertainties.

Time	Sample mass	^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content in total blood (weighted mean of gamma and mass spectrometric measurements, when available)	
		^{237}Pu activity in total blood[2]	^{244}Pu atoms in sample[3,11]	Equivalent ^{237}Pu activity in total blood[2,3]	Equivalent ^{237}Pu activity	
(d)	(g)	(Bq)		(Bq)	(Bq)	% of ILD[1]
0.01	21.3	<22.76	1.199E+08	2.27±0.43	**2.27±0.43**	**0.0293±0.0056** [4]
0.03	21.6	<42.62	3.192E+08	5.95±1.13	**5.95±1.13**	**0.0768±0.0146**
0.05	21.5	<27.89	4.422E+08	8.26±1.57	**8.26±1.57**	**0.1067±0.0203**
0.11	21.5	20.19±5.66	SL[7]		20.19±5.66	0.2609±0.0732 [5]
0.25	21.5	43.28±11.24	2.986E+09	55.82±10.61	***49.91±7.71***	***0.6449±0.0997*** [6]
0.37	21.7	67.90±5.49	5.151E+09	95.33±18.11	***70.21±5.26***	***0.9072±0.0679***
1.25	21.4	145.63±10.60	7.540E+09	141.47±26.88	***145.07±9.86***	***1.8745±0.1274***
1.85	21.6	72.59±5.74	4.882E+09	91.02±17.29	***74.42±5.45***	***0.9616±0.0704***
3.89	21.6	71.73±9.36	3.825E+09	71.09±13.51	***71.52±7.69***	***0.9242±0.0994***
4.85	21.6	38.60±7.37	2.666E+09	49.60±9.42	***42.77±5.80***	***0.5527±0.0750***
6.81	21.2	63.57±11.53	2.681E+09	50.88±9.67	***56.12±7.41***	***0.7252±0.0957***
8.80	21.4	31.14±4.87	2.981E+09	56.00±10.64	***35.44±4.42***	***0.4579±0.0572***
11.80	21.3	24.41±7.39	1.291E+09	24.42±4.64	***24.42±3.93***	***0.3155±0.0508***
15.80	21.3	NA[8]	1.113E+09	21.06±4.00	**21.06±4.00**	**0.2721±0.0517**
25.91	23.5	NA	8.139E+08	13.95±2.65	**13.95±2.65**	**0.1802±0.0342**
44.02	21.8	NA	6.321E+08	11.66±2.21	**11.66±2.21**	**0.1506±0.0286**
64.81	21.3	NA	8.271E+08	15.62±2.97	**15.62±2.97**	**0.2018±0.0383**
84.00	21.2	NA	3.276E+08	6.22±1.18	**6.22±1.18**	**0.0803±0.0153**
103.80	21.9	NA	SNM[9]		SNM	
117.85	21.5	NA	4.834E+08	9.02±1.71	**9.02±1.71**	**0.1166±0.0221** [10]
299.87	23.2	NA	SNM		SNM	
481.85	20.0	NA	3.209E+08	6.45±1.23	**6.45±1.23**	**0.0834±0.0158** [10]
550.87	18.9	NA	SNM		SNM	
733.869	18.9	NA	1.410E+08	3.01±0.57	**3.01±0.57**	**0.0388±0.0074** [10]
733.871	12.5	NA	1.549E+08	4.98±0.95	**4.98±0.95**	**0.0643±0.0122**
828.91	22.4	NA	SNM		**SNM**	

Time	Sample mass	^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content in total blood (weighted mean of gamma and mass spectrometric measurements, when available)	
		^{237}Pu activity in total blood[2]	^{244}Pu atoms in sample[3,11]	Equivalent ^{237}Pu activity in total blood[2,3] (Bq)	Equivalent ^{237}Pu activity (Bq)	% of ILD[1]
(d)	(g)	(Bq)				
921.83	23.3	NA	1.575E+08	2.72±0.52	**2.72±0.52**	**0.0351±0.0067** [10]
1083.81	16.9	NA	SNM		**SNM**	
1332.12	19.7	NA	3.394E+08	6.94±1.32	**6.94±1.32**	**0.0896±0.0170**
1476.85	19.5	NA	8.666E+07	1.78±0.34	**1.78±0.34**	**0.0231±0.0044**
1700.87	18.3	NA	9.220E+07	2.03±0.39	**2.03±0.39**	**0.0262±0.0050**
1880.87	20.4	NA	6.688E+07	1.32±0.25	**1.32±0.25**	**0.0170±0.0032** [10]
2141.84	20.5	NA	8.524E+07	1.67±0.32	**1.67±0.32**	**0.0216±0.0041**

Notes
1. Initial lung deposit (ILD) for Subject C = 7739.2 Bq
2. Amounts in total blood estimated from the sample measurements, the sample weight and an estimated Subject C total blood mass = 4.2 kg (see text)
3. Uncertainties (1σ) on mass spectrometry measurements are estimated to be 19% of the measured value.
4. Bold, non-italic text entries - mass spectrometry measurements only
5. Plain text entries - gamma spectrometry measurements only
6. Bold, italic text entries - weighted mean of the gamma spectrometry and mass spectrometry measurement results. "Less than" values are not used to estimate mean values.
7. SL = Sample lost after gamma spectrometry measurement was made
8. NA = not available (sample not measured by gamma spectrometry)
9. SNM = sample available but not measured by mass spectrometry
10. These five results are the most recent mass spectrometry measurements (as of August 2016), and were not taken into account for the analysis published in Puncher and Etherington (2017)
11. For ^{244}Pu, 1E+08 atoms = 4.1E-14 g = 41 fg

Table 13 Blood content, Subject D, gamma and mass spectrometry measurements. Amounts expressed as number of ^{244}Pu atoms, Bq of ^{237}Pu or as a percentage of the initial lung deposit of ^{237}Pu (ILD)[1]. Quoted uncertainties for ^{237}Pu measurements (±1σ) are total uncertainties.

Time	Sample mass	^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content in total blood (weighted mean of gamma and mass spectrometric measurements, when available)	
		^{237}Pu activity in total blood[2]	^{244}Pu atoms in sample[3,11]	Equivalent ^{237}Pu activity in total blood[2,3]	Equivalent ^{237}Pu activity	
(d)	(g)	(Bq)		(Bq)	(Bq)	% of ILD[1]
0.01	21.7	<17.69	6.671E+07	1.12±0.21	**1.12±0.21**	**0.0148±0.0028** [4]
0.03	21.8	<29.32	2.942E+08	4.93±0.94	**4.93±0.94**	**0.0651±0.0124**
0.11	21.6	<25.57	1.509E+09	25.45±4.84	**25.45±4.84**	**0.3358±0.0638**
0.20	21.4	49.29±6.43	3.628E+09	61.97±11.77	***52.20±5.64***	***0.6887±0.0744*** [6]
0.35	21.5	120.92±11.21	4.717E+09	79.89±15.18	***106.44±9.02***	***1.4044±0.1190***
0.91	21.3	288.69±16.98	1.547E+10	264.60±50.27	***286.23±16.09***	***3.7767±0.2122***
1.15	21.1	305.83±17.75	1.808E+10	312.14±59.31	***306.35±17.00***	***4.0421±0.2243***
2.11	21.5	347.58±21.54	1.450E+10	246.08±46.76	***329.81±19.56***	***4.3518±0.2581***
2.95	21.5	263.22±16.41	1.875E+10	317.57±60.34	***266.96±15.83***	***3.5224±0.2089***
3.91	21.1	212.88±13.72	1.240E+10	214.46±40.75	***213.04±13.01***	***2.8110±0.1716***
5.91	21.3	144.65±9.19	9.384E+09	160.59±30.51	***145.98±8.80***	***1.9261±0.1161***
7.90	21.6	72.06±9.12	3.342E+09	56.35±10.71	***65.45±6.94***	***0.8637±0.0916***
10.88	21.7	80.13±12.23	2.794E+09	46.92±8.92	***58.44±7.21***	***0.7711±0.0951***
14.86	21.3	NA[8]	2.558E+09	43.75±8.31	**43.75±8.31**	**0.5773±0.1097**
19.10	21.5	NA	1.333E+09	22.67±4.31	**22.67±4.31**	**0.2992±0.0568**
35.89	21.8	NA	1.329E+09	22.19±4.22	**22.19±4.22**	**0.2928±0.0556**
55.92	22.8	NA	1.110E+09	17.79±3.38	**17.79±3.38**	**0.2348±0.0446**
84.89	21.2	NA	1.001E+09	17.27±3.28	**17.27±3.28**	**0.2278±0.0433**
101.96	21.5	NA	5.957E+08	10.10±1.92	**10.10±1.92**	**0.1333±0.0253**
115.93	21.4	NA	5.327E+08	9.06±1.72	**9.06±1.72**	**0.1196±0.0227**
288.01	21.8	NA	6.471E+08	10.84±2.06	**10.84±2.06**	**0.1431±0.0272**
473.01	21.2	NA	3.394E+08	5.85±1.11	**5.85±1.11**	**0.0772±0.0147**
552.01	21.0	NA	3.059E+08	5.31±1.01	**5.31±1.01**	**0.0700±0.0133**
740.01	21.2	NA	2.736E+08	4.70±0.89	**4.70±0.89**	**0.0620±0.0118**
817.01	21.8	NA	SNM[9]		SNM	

1072.01	21.4	NA	1.755E+08	2.99±0.57	**2.99±0.57**	**0.0395±0.0075** [10]
1335.96	21.3	NA	1.030E+08	1.76±0.33	**1.76±0.33**	**0.0233±0.0044**
1448.01	21.2	NA	1.290E+08	2.22±0.42	**2.22±0.42**	**0.0293±0.0056**
1729.02	22.6	NA	MI[7]	MI	**MI**	**MI**
1936.10	17.5	NA	8.560E+07	1.78±0.34	**1.78±0.34**	**0.0235±0.0045**
2076.01	16.8	NA	1.190E+08	2.58±0.49	**2.58±0.49**	**0.0340±0.0065**
2714.01	29.3	NA	1.510E+08	1.88±0.36	**1.88±0.36**	**0.0248±0.0047** [10]
3345.97	30.9	NA	1.508E+08	1.78±0.34	**1.78±0.34**	**0.0235±0.0045** [10]
4887.01	39.0	NA	1.552E+08	1.45±0.28	**1.45±028**	**0.0191±0.0036** [10]

Notes
1. Initial lung deposit (ILD) for Subject D = 7578.8 Bq
2. Amounts in total blood estimated from the sample measurements, the sample weight and an estimated Subject D total blood mass = 4.3 kg (see text)
3. Uncertainties (1σ) on mass spectrometry measurements are estimated to be 19% of the measured value.
4. Bold, non-italic text entries - mass spectrometry measurements only
5. -
6. Bold, italic text entries - weighted mean of the gamma spectrometry and mass spectrometry measurement results. “Less than” values are not used to estimate mean values.
7. MI = Measurement inconclusive
8. NA = not available (sample not measured by gamma spectrometry)
9. SNM = sample available but not measured by mass spectrometry
10. These four results are the most recent mass spectrometry measurements (as of August 2016), and were not taken into account for the analysis published in Puncher and Etherington (2017)
11. For ^{244}Pu, 1E+08 atoms = 4.1E-14 g = 41 fg

Table 14 Daily urine excretion, Subject C, gamma and mass spectrometry measurements. Amounts expressed as ^{237}Pu activity in the sample, and daily excretion as a percentage of the initial lung deposit of ^{237}Pu (ILD)[1]. Quoted uncertainties for ^{237}Pu measurements (±1σ) are total uncertainties.

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
-3.59	-2.59	1.00	<0.30 [2,3]	SNM[8]		<0.30	-
0.00	0.30	0.30	<0.96	6.972E+09	0.67±0.13	**0.67±0.13**	**0.0291±0.0055** [5,10]
0.30	0.40	0.10	1.57±0.12	SNM		1.57±0.12	0.1959±0.0155 [6]
0.40	0.69	0.29	<0.83	1.077E+10	1.03±0.20	**1.03±0.20**	**0.0457±0.0087** [10]
0.69	0.81	0.11	2.19±0.14	SNM		2.19±0.14	0.2467±0.0163
0.81	0.97	0.16	1.15±0.14	SNM		1.15±0.14	0.0929±0.0115
0.97	0.99	0.02	1.82±0.15	SNM		1.82±0.15	0.9944±0.0824
0.99	1.99	1.00	14.32±0.75	SNM		14.32±0.75	0.1851±0.0096
1.99	2.99	1.00	4.39±0.24	SNM		4.39±0.24	0.0567±0.0030
2.99	3.99	1.00	6.17±0.33	SNM		6.17±0.33	0.0797±0.0043
3.99	4.99	1.00	5.20±0.31	4.000E+10	3.83±0.73	***5.00±0.28***	***0.0646±0.0036*** [7,10]
4.99	5.99	1.00	2.46±0.17	SNM		2.46±0.17	0.0317±0.0022
5.99	6.99	1.00	2.71±0.17	SNM		2.71±0.17	0.0350±0.0021
6.99	7.99	1.00	2.04±0.19	SNM		2.04±0.19	0.0264±0.0024
7.99	8.99	1.00	1.76±0.18	SNM		1.76±0.18	0.0228±0.0023
8.99	9.99	1.00	1.57±0.15	SNM		1.57±0.15	0.0203±0.0020
9.99	10.99	1.00	2.07±0.18	SNM		2.07±0.18	0.0268±0.0023
10.99	11.99	1.00	2.13±0.15	SNM		2.13±0.15	0.0276±0.0019
11.99	12.99	1.00	2.08±0.17	SNM		2.08±0.17	0.0268±0.0022
12.99	13.99	1.00	1.59±0.13	SNM		1.59±0.13	0.0205±0.0017
13.99	14.99	1.00	1.83±0.13	SNM		1.83±0.13	0.0237±0.0016
14.99	15.99	1.00	1.81±0.15	SNM		1.81±0.15	0.0234±0.0019
22.99	23.99	1.00	1.34±0.15	SNM		1.34±0.15	0.0173±0.0019
23.99	24.99	1.00	1.53±0.15	1.763E+10	1.69±0.32	***1.56±0.14***	***0.0201±0.0018***
24.99	25.99	1.00	1.46±0.12	SNM		1.46±0.12	0.0189±0.0016
30.99	33.99	3.00	3.73±0.29	SNM		3.73±0.29	0.0161±0.0012

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
36.99	39.99	3.00	5.25±0.36	4.830E+10	4.62±0.88	***5.16±0.33***	***0.0222±0.0014***
43.99	44.99	1.00	1.16±0.09	SNM		1.16±0.09	0.0149±0.0011
44.99	46.99	2.00	2.19±0.16	SNM		2.19±0.16	0.0141±0.0010
50.99	53.99	3.00	4.55±0.29	SNM		4.55±0.29	0.0196±0.0013
56.99	61.99	5.00	5.05±0.53	SNM		5.05±0.53	0.0130±0.0014
65.99	68.99	3.00	4.92±0.63	SNM		4.92±0.63	0.0212±0.0027
71.99	74.99	3.00	6.58±0.50	SNM		6.58±0.50	0.0283±0.0022
78.99	83.99	5.00	10.42±0.68	1.850E+11	17.71±3.37	***10.70±0.66***	***0.0277±0.0017***
85.99	90.99	5.00	2.42±0.45	SNM		2.42±0.45	0.0062±0.0012
92.99	97.99	5.00	3.48±0.33	SNM		3.48±0.33	0.0090±0.0009
98.99	103.99	5.00	3.94±0.49	SNM		3.94±0.49	0.0102±0.0013
110.99	115.99	5.00	NA[9]	4.498E+10	4.31±0.82	**4.31±0.82**	**0.0111±0.0021** [10]
143.99	148.99	5.00	NA	SNM		SNM	
180.99	185.99	5.00	NA	SNM		SNM	
215.99	220.99	5.00	NA	1.050E+11	10.06±1.91	**10.06±1.91**	**0.0260±0.0049** [10]
244.99	249.99	5.00	NA	SNM		SNM	
278.99	283.99	5.00	NA	SNM		SNM	
305.99	310.99	5.00	NA	2.922E+10	2.80±0.53	**2.80±0.53**	**0.0072±0.0014** [10]
335.99	340.99	5.00	NA	SNM		SNM	
372.99	377.99	5.00	NA	SNM		SNM	
461.99	466.99	5.00	NA	3.140E+10	3.01±0.57	**3.01±0.57**	**0.0078±0.0015**
547.99	552.99	5.00	NA	SNM		SNM	
633.99	638.99	5.00	NA	2.390E+10	2.29±0.43	**2.29±0.43**	**0.0059±0.0011** [10]
729.99	734.99	5.00	NA	SNM		SNM	
824.99	829.99	5.00	NA	1.460E+10	1.40±0.27	**1.40±0.27**	**0.0036±0.0007**
917.99	922.99	5.00	NA	1.670E+10	1.60±0.30	**1.60±0.30**	**0.0041±0.0008**
1080.99	1085.99	5.00	NA	1.130E+10	1.08±0.21	**1.08±0.21**	**0.0028±0.0005**
1328.99	1333.99	5.00	NA	1.000E+10	0.96±0.18	**0.96±0.18**	**0.0025±0.0005**
1472.99	1477.99	5.00	NA	1.190E+10	1.14±0.22	**1.14±0.22**	**0.0029±0.0006**

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Duration (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
1697.99	1702.99	5.00	NA	9.640E+09	0.92±0.18	**0.92±0.18**	**0.0024±0.0005**
1876.99	1881.99	5.00	NA	9.570E+09	0.92±0.17	**0.92±0.17**	**0.0024±0.0004**
2138.99	2143.99	5.00	NA	7.780E+09	0.74±0.14	**0.74±0.14**	**0.0019±0.0004**

Notes

1. Initial lung deposit (ILD) for Subject C = 7739.2 Bq
2. Blank sample collected before the Pu inhalation
3. Limit of detection determined from the measured count rate in a region of interest centred on 101 keV (the energy of the 2nd of the three main gamma-ray spectrum peaks of ^{237}Pu).
4. Uncertainties (1σ) on mass spectrometry measurements are estimated to be 19% of the measured value.
5. Bold, non-italic text entries - mass spectrometry measurements only
6. Plain text entries - gamma spectrometry measurements only
7. Bold, italic text entries - weighted mean of the gamma spectrometry and mass spectrometry measurement results. “Less than” values are not used to estimate mean values
8. SNM = sample available but not measured by mass spectrometry
9. NA = not available (sample not measured by gamma spectrometry)
10. These seven results are the most recent mass spectrometry measurements (as of August 2016), and were not taken into account for the analysis published in Puncher and Etherington (2017)
11. For ^{244}Pu, 1E+08 atoms = 4.1E-14 g = 41 fg

Table 15 Daily urine excretion, Subject D, gamma and mass spectrometry measurements. Amounts expressed as ^{237}Pu activity in the sample, and daily excretion as a percentage of the initial lung deposit of ^{237}Pu (ILD)[1]. Quoted uncertainties for ^{237}Pu measurements (±1σ) are total uncertainties.

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
-10.14	-9.14	1.00	<0.67 [2,3]	SNM[8]		<0.67	-
0.00	0.11	0.11	<0.35	7.070E+09	0.60±0.11	**0.60±0.11**	**0.0689±0.0131** [5,10]
0.11	0.26	0.15	<0.21	SNM		SNM	
0.26	0.40	0.14	<0.29	1.370E+10	1.16±0.22	**1.16±0.22**	**0.1104±0.0210**[10]
0.40	0.51	0.11	0.66±0.07	SNM		0.66±0.07	0.0784±0.0089[6]
0.51	0.80	0.29	0.58±0.09	9.850E+09	0.84±0.16	***0.64±0.08***	***0.0289±0.0035*** [7,10]
0.80	0.87	0.06	2.43±0.15	SNM		2.43±0.15	0.4960±0.0309
0.87	1.00	0.13	0.65±0.13	SNM		0.65±0.13	0.0656±0.0132
1.00	1.00	0.002	<0.30	SNM		<0.30	-
1.00	1.80	0.80	7.45±0.40	1.450E+10	1.23±0.23	***2.81±0.20***	***0.0465±0.0033***[10]
1.80	2.00	0.20	2.31±0.14	SNM		2.31±0.14	0.1537±0.0094
2.00	3.00	1.00	10.56±0.55	SNM		10.56±0.55	0.1389±0.0072
3.00	3.99	0.99	6.75±0.37	8.970E+10	7.61±1.45	***6.81±0.36***	***0.0911±0.0048***
3.99	4.99	1.00	5.51±0.31	SNM		5.51±0.31	0.0727±0.0040
4.99	5.99	1.00	4.62±0.27	SNM		4.62±0.27	0.0609±0.0035
5.99	6.98	1.00	4.36±0.24	SNM		4.36±0.24	0.0578±0.0031
6.98	7.99	1.00	3.96±0.29	5.611E+10	4.76±0.90	***4.03±0.27***	***0.0530±0.0036***
7.99	8.99	1.00	2.89±0.18	SNM		2.89±0.18	0.0380±0.0023
8.99	9.98	0.99	3.45±0.21	SNM		3.45±0.21	0.0458±0.0028
9.98	11.00	1.01	3.24±0.19	SNM		3.24±0.19	0.0421±0.0024
11.00	11.97	0.98	2.30±0.14	SNM		2.30±0.14	0.0311±0.0019
11.97	13.00	1.02	3.67±0.20	SNM		3.67±0.20	0.0473±0.0026
13.00	14.00	1.00	3.31±0.19	SNM		3.31±0.19	0.0435±0.0025
17.06	18.06	1.00	1.59±0.13	SNM		1.59±0.13	0.0209±0.0017
18.06	19.07	1.01	2.39±0.15	SNM		2.39±0.15	0.0312±0.0019

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
19.07	20.07	1.00	2.61±0.16	3.046E+10	2.58±0.49	***2.60±0.16***	***0.0344±0.0021***
22.87	23.87	1.00	1.43±0.16	SNM		1.43±0.16	0.0188±0.0021
23.87	24.87	1.00	1.99±0.13	SNM		1.99±0.13	0.0263±0.0017
24.87	25.87	1.00	2.96±0.22	SNM		2.96±0.22	0.0392±0.0029
29.80	30.79	0.99	2.08±0.17	SNM		2.08±0.17	0.0276±0.0023
30.79	31.79	1.00	2.18±0.16	2.690E+10	2.28±0.43	***2.20±0.15***	***0.0291±0.0020***
31.79	32.80	1.01	2.23±0.17	SNM		2.23±0.17	0.0291±0.0022
36.91	39.91	3.00	6.73±0.36	5.640E+10	4.79±0.91	***6.47±0.33***	***0.0285±0.0015***
42.83	45.83	3.00	5.65±0.30	SNM		5.65±0.30	0.0249±0.0013
51.51	54.51	3.00	5.95±0.35	SNM		5.95±0.35	0.0262±0.0015
58.82	61.82	3.00	5.03±0.28	SNM		5.03±0.28	0.0221±0.0012
64.51	67.51	3.01	5.22±0.28	SNM		5.22±0.28	0.0229±0.0012
72.34	77.34	5.00	14.78±0.97	1.090E+11	9.25±1.76	***13.49±0.85***	***0.0356±0.0022***
77.34	79.32	1.98	NA[9]	SNM		SNM	
84.51	89.51	5.00	5.20±0.36	SNM		5.20±0.36	0.0137±0.0010
93.51	97.51	4.00	6.54±0.39	SNM		6.54±0.39	0.0216±0.0013
98.52	105.51	6.99	7.95±0.51	SNM		7.95±0.51	0.0150±0.0010
108.02	113.02	5.00	NA	7.982E+10	6.77±1.29	**6.77±1.29**	**0.0179±0.0034**
137.81	140.82	3.01	NA	3.150E+10	2.67±0.51	**2.67±0.51**	**0.0117±0.0022**
184.84	187.83	2.99	NA	4.963E+10	4.21±0.80	**4.21±0.80**	**0.0186±0.0035**
210.80	213.79	3.00	NA	6.254E+10	5.31±1.01	**5.31±1.01**	**0.0234±0.0044**
246.51	249.54	3.03	NA	5.827E+10	4.94±0.94	**4.94±0.94**	**0.0215±0.0041**
274.83	277.84	3.00	NA	4.837E+10	4.10±0.78	**4.10±0.78**	**0.0180±0.0034**
309.89	312.89	3.00	NA	3.121E+10	2.65±0.50	**2.65±0.50**	**0.0116±0.0022**
344.96	347.96	3.00	NA	2.982E+10	2.53±0.48	**2.53±0.48**	**0.0111±0.0021**
466.89	471.83	4.95	NA	3.107E+10	2.64±0.50	**2.64±0.50**	**0.0070±0.0013**
548.80	553.80	5.00	NA	4.130E+10	3.50±0.67	**3.50±0.67**	**0.0092±0.0018**
647.82	652.84	5.02	NA	2.038E+10	1.73±0.33	**1.73±0.33**	**0.0045±0.0009**

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
732.23	737.20	4.98	NA	1.690E+10	1.43±0.27	**1.43±0.27**	**0.0038±0.0007**
813.81	818.80	4.99	NA	1.527E+10	1.30±0.25	**1.30±0.25**	**0.0034±0.0007**
1071.92	1076.92	5.01	NA	SNM		SNM	
1097.81	1102.81	5.00	NA	1.242E+10	1.05±0.20	**1.05±0.20**	**0.0028±0.0005**
1319.81	1324.79	4.98	NA	SNM		SNM	
1380.89	1385.92	5.03	NA	1.121E+10	0.95±0.18	**0.95±0.18**	**0.0025±0.0005**
1465.80	1470.78	4.98	NA	8.251E+09	0.70±0.13	**0.70±0.13**	**0.0019±0.0004**
1695.04	1699.94	4.91	NA	1.079E+10	0.92±0.17	**0.92±0.17**	**0.0025±0.0005**
1872.84	1877.86	5.02	NA	8.002E+09	0.68±0.13	**0.68±0.13**	**0.0018±0.0003**
2130.82	2135.82	5.01	NA	1.068E+10	0.91±0.17	**0.91±0.17**	**0.0024±0.0005**
2587.90	2592.90	5.00	NA	8.005E+09	0.68±0.13	**0.68±0.13**	**0.0018±0.0003**
3288.84	3292.86	4.02	NA	7.896E+09	0.67±0.13	**0.67±0.13**	**0.0022±0.0004**
4901.90	4905.93	4.03	NA	4.574E+09	0.39±0.07	**0.39±0.14**	**0.0013±0.0005**[10]

Notes
1. Initial lung deposit (ILD) for Subject D = 7578.8 Bq
2. Blank sample collected before the Pu inhalation
3. Limit of detection determined from the measured count rate in a region of interest centred on 101 keV (the energy of the 2nd of the three main gamma-ray spectrum peaks of ^{237}Pu).
4. Uncertainties (1σ) on mass spectrometry measurements are estimated to be 19% of the measured value
5. Bold, non-italic text entries - mass spectrometry measurements only
6. Plain text entries - gamma spectrometry measurements only
7. Bold, italic text entries - weighted mean of the gamma spectrometry and mass spectrometry measurement results. “Less than” values are not used to estimate mean values.
8. SNM = sample available but not measured by mass spectrometry
9. NA = not available (sample not measured by gamma spectrometry)
10. These five results are the most recent mass spectrometry measurements (as of August 2016), and were not taken into account for the analysis published in Puncher and Etherington (2017)
11. For ^{244}Pu, 1E+08 atoms = 4.1E-14 g = 41 fg

Table 16 Daily faeces excretion, Subject C, gamma and mass spectrometry measurements. Amounts expressed as ^{237}Pu activity in the sample, and daily excretion as a percentage of the initial lung deposit of ^{237}Pu (ILD)[1]. Quoted uncertainties for ^{237}Pu measurements (±1σ) are total uncertainties.

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
-3.21	-2.21	1.00	<0.074[2,3]	SNM[8]		<0.074	
-0.19	0.81	1.00	49.12±2.50	SNM		49.12±2.50	0.6347±0.0324 [6]
0.81	1.79	0.98	474.79±23.81	SNM		474.79±23.81	6.2433±0.3130
1.79	2.77	0.98	310.36±15.57	SNM		310.36±15.57	4.0897±0.2051
2.77	3.76	1.00	87.26±4.43	SNM		87.26±4.43	1.1330±0.0575
3.76	4.98	1.22	29.79±1.50	SNM		29.79±1.50	0.3168±0.0159
4.98	5.81	0.83	17.03±0.86	SNM		17.03±0.86	0.2663±0.0135
5.81	6.81	1.00	15.09±0.81	SNM		15.09±0.81	0.1943±0.0104
6.81	7.78	0.97	5.37±0.28	SNM		5.37±0.28	0.0714±0.0037
7.78	8.76	0.98	11.73±0.61	SNM		11.73±0.61	0.1543±0.0080
8.76	9.82	1.06	10.51±0.54	SNM		10.51±0.54	0.1286±0.0066
9.82	10.82	1.00	11.52±0.60	SNM		11.52±0.60	0.1493±0.0077
10.82	11.92	1.10	7.43±0.38	SNM		7.43±0.38	0.0873±0.0045
11.92	12.85	0.93	21.54±1.09	SNM		21.54±1.09	0.2992±0.0151
12.85	13.76	0.91	8.27±0.42	SNM		8.27±0.42	0.1170±0.0060
13.76	14.79	1.02	6.78±0.35	SNM		6.78±0.35	0.0856±0.0044
14.79	15.83	1.05	9.71±0.50	SNM		9.71±0.50	0.1197±0.0062
15.83	16.95	1.11	5.17±0.30	SNM		5.17±0.30	0.0601±0.0035
16.95	17.81	0.86	7.03±0.43	SNM		7.03±0.43	0.1054±0.0065
17.81	18.78	0.98	10.78±0.56	SNM		10.78±0.56	0.1427±0.0074
18.78	19.75	0.97	7.32±0.38	SNM		7.32±0.38	0.0976±0.0051
22.78	23.78	1.00	2.40±0.15	SNM		2.40±0.15	0.0310±0.0019
23.78	24.82	1.04	5.18±0.28	SNM		5.18±0.28	0.0642±0.0034
24.82	25.83	1.01	7.56±0.40	SNM		7.56±0.40	0.0970±0.0052
30.77	31.77	1.00	14.92±0.81	SNM		14.92±0.81	0.1927±0.0105
31.77	32.78	1.01	7.91±0.43	SNM		7.91±0.43	0.1015±0.0056

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
32.78	33.77	0.99	6.46±0.37	SNM		6.46±0.37	0.0841±0.0048
36.77	39.77	3.00	22.23±1.20	SNM		22.23±1.20	0.0959±0.0052
43.76	46.72	2.95	23.90±1.27	SNM		23.90±1.27	0.1045±0.0056
56.78	61.78	5.00	21.22±1.08	SNM		21.22±1.08	0.0548±0.0028
71.78	74.78	3.00	14.73±0.82	SNM		14.73±0.82	0.0635±0.0035
86.72	87.72	1.00	9.61±0.53	SNM		9.61±0.53	0.1242±0.0069
87.72	88.74	1.02	6.05±0.42	SNM		6.05±0.42	0.0766±0.0053
88.74	89.82	1.08	3.46±0.24	SNM		3.46±0.24	0.0413±0.0029
98.75	101.76	3.01	14.05±1.01	1.620E+11	15.51±2.95	***14.20±0.95***	***0.0609±0.0041*** [7,10]
112.76	115.76	3.00	14.51±0.90	SNM		14.51±0.90	0.0625±0.0039
143.76	146.77	3.01	NA[9]	SNM			
180.74	183.75	3.01	NA	SNM			
215.75	218.75	3.00	NA	SNM			
246.78	249.78	3.00	NA	1.092E+11	10.45±1.99	**10.45±1.99**	**0.0450±0.0086** [5,10]
278.76	281.76	3.00	NA	SNM			
306.75	309.75	3.00	NA	SNM			
335.76	338.76	3.00	NA	SNM			
373.75	376.75	3.00	NA	5.493E+10	5.26±1.00	**5.26±1.00**	**0.0227±0.0043** [10]
462.74	464.74	2.00	NA	SNM			
548.77	550.76	1.99	NA	SNM			
635.76	637.76	2.00	NA	SNM			
731.77	733.75	1.98	NA	3.164E+10	3.03±0.58	**3.03±0.58**	**0.0198±0.0038** [10]
826.76	828.76	2.00	NA	SNM			
919.76	921.76	2.00	NA	SNM			
1081.78	1083.75	1.97	NA	SNM			
1330.74	1332.74	2.00	NA	1.006E+10	0.96±0.18	**0.96±0.18**	**0.0062±0.0012** [10]

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Duration (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
1474.74	1476.74	2.00	NA	SNM			
1699.74	1701.74	2.00	NA	SNM			
1878.74	1880.74	2.00	NA	SNM			
2140.74	2142.74	2.00	NA	2.384E+09	0.23±0.04	**0.23±0.04**	**0.0015±0.0003** [10]

Notes

1. Initial lung deposit (ILD) for Subject C = 7739.2 Bq
2. Blank sample collected before the Pu inhalation
3. Limit of detection determined from the measured count rate in a region of interest centred on 101 keV (the energy of the 2nd of the three main gamma-ray spectrum peaks of ^{237}Pu)
4. Uncertainties (1σ) on mass spectrometry measurements are estimated to be 19% of the measured value.
5. Bold, non-italic text entries - mass spectrometry measurements only
6. Plain text entries - gamma spectrometry measurements only
7. Bold, italic text entries - weighted mean of the gamma spectrometry and mass spectrometry measurement results. “Less than” values are not used to estimate mean values.
8. SNM = sample available but not measured by mass spectrometry
9. NA = not available (sample not measured by gamma spectrometry)
10. These six results are the most recent mass spectrometry measurements (as of August 2016), and were not taken into account for the analysis published in Puncher and Etherington (2017)
11. For ^{244}Pu, 1E+08 atoms = 4.1E-14 g = 41 fg

Table 17 Daily faeces excretion, Subject D, gamma and mass spectrometry measurements. Amounts expressed as ^{237}Pu activity in the sample, and daily excretion as a percentage of the initial lung deposit of ^{237}Pu (ILD)[1]. Quoted uncertainties for ^{237}Pu measurements (±1σ) are total uncertainties.

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
-11.17	-10.17	1.00	<0.17[2,3]	SNM[8]		<0.17	
-0.13	0.87	1.00	751.82±37.67	SNM		751.82±37.67	9.9200±0.4970 [6]
0.87	1.93	1.07	393.42±19.71	SNM		393.42±19.71	4.8698±0.2440
1.93	2.90	0.96	71.88±3.65	SNM		71.88±3.65	0.9861±0.0500
2.90	3.88	0.98	22.22±1.12	SNM		22.22±1.12	0.2994±0.0151
3.88	4.88	1.01	18.93±0.95	SNM		18.93±0.95	0.2481±0.0125
4.88	5.82	0.93	16.64±0.84	SNM		16.64±0.84	0.2351±0.0118
5.82	6.81	1.00	12.35±0.62	SNM		12.35±0.62	0.1635±0.0082
6.81	7.23	0.41	8.91±0.45	SNM		8.91±0.45	0.2847±0.0143
7.23	8.83	1.61	18.73±0.94	SNM		18.73±0.94	0.1538±0.0077
8.83	10.18	1.35	14.74±0.74	SNM		14.74±0.74	0.1444±0.0073
10.18	10.83	0.65	8.87±0.45	SNM		8.87±0.45	0.1802±0.0091
10.83	11.28	0.45	7.66±0.45	SNM		7.66±0.45	0.2255±0.0131
11.28	11.87	0.59	7.72±0.42	SNM		7.72±0.42	0.1715±0.0094
11.87	12.82	0.95	13.52±0.69	SNM		13.52±0.69	0.1882±0.0096
12.82	13.82	1.00	14.58±0.75	SNM		14.58±0.75	0.1917±0.0099
13.82	14.88	1.05	12.49±0.63	SNM		12.49±0.63	0.1567±0.0079
14.88	16.22	1.35	14.39±0.73	SNM		14.39±0.73	0.1409±0.0071
16.22	16.97	0.75	7.81±0.41	SNM		7.81±0.41	0.1381±0.0072
16.97	17.82	0.85	7.50±0.39	SNM		7.50±0.39	0.1169±0.0061
17.82	18.82	1.00	12.33±0.63	SNM		12.33±0.63	0.1627±0.0083
18.82	19.85	1.03	11.21±0.60	SNM		11.21±0.60	0.1430±0.0076
29.51	29.94	0.43	5.07±0.30	SNM		5.07±0.30	0.1540±0.0091
29.94	30.13	0.18	3.74±0.23	SNM		3.74±0.23	0.2733±0.0172
30.13	31.04	0.92	5.11±0.30	SNM		5.11±0.30	0.0736±0.0043
31.04	31.82	0.77	3.13±0.20	SNM		3.13±0.20	0.0534±0.0035

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Dur-ation (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
31.82	31.90	0.09	4.39±0.25	SNM		4.39±0.25	0.6673±0.0384
38.10	40.82	2.73	28.07±1.53	SNM		28.07±1.53	0.1359±0.0074
50.85	54.83	3.98	35.94±1.86	SNM		35.94±1.86	0.1192±0.0062
64.14	66.79	2.65	20.29±1.13	SNM		20.29±1.13	0.1009±0.0056
75.81	79.13	3.33	11.58±0.68	SNM		11.58±0.68	0.0459±0.0027
92.85	95.86	3.02	21.32±1.18	2.23E+11	18.91±3.59	***21.08±1.13***	***0.0922±0.0049*** [7,10]
106.91	109.85	2.94	15.87±1.14	SNM		15.87±1.14	0.0713±0.0051
136.82	139.82	3.00	NA[9]	SNM			
186.85	189.82	2.97	NA	SNM			
209.82	212.92	3.11	NA	SNM			
246.92	249.95	3.03	NA	7.80E+10	6.62±1.26	**6.62±1.26**	**0.0288±0.0055** [5,10]
273.83	276.88	3.06	NA	SNM			
308.82	311.82	3.00	NA	SNM			
343.82	346.86	3.04	NA	SNM			
466.85	470.83	3.98	NA	SNM			
550.82	553.95	3.14	NA	1.96E+10	1.66±0.32	**1.66±0.32**	**0.0070±0.0013** [10]
649.85	652.93	3.08	NA	SNM			
729.89	732.82	2.93	NA	SNM			
813.89	816.81	2.92	NA	SNM			
1070.82	1073.90	3.09	NA	5.29E+09	0.45±0.09	**0.45±0.09**	**0.0019±0.0004** [10]
1319.87	1322.92	3.05	NA	SNM			
1464.90	1467.85	2.95	NA	SNM			
1694.91	1697.99	3.07	NA	SNM			
1872.87	1876.90	4.03	NA	SNM			
2132.93	2135.89	2.96	NA	1.57E+09	0.13±0.03	**0.13±0.03**	**0.00059± 0.00011** [10]
2590.89	2593.89	3.00	NA	SNM			
3288.90	3291.91	3.01	NA	SNM			

Sample collection times			^{237}Pu gamma spectrometry measurement	^{244}Pu mass spectrometry measurement		Pu content (weighted mean when available)	Daily excretion
Start (d)	End (d)	Duration (d)	^{237}Pu activity in sample (Bq)	^{244}Pu atoms in sample[4,11]	Equivalent ^{237}Pu activity in sample[4] (Bq)	Equivalent ^{237}Pu activity in sample (Bq)	(%ILD[1] per day)
4900.93	4905.90	4.97	NA	2.09E+09	0.18±0.03	**0.18±0.03**	**0.00047± 0.00009** [10]

Notes

1. Initial lung deposit (ILD) for Subject D = 7578.8 Bq
2. Blank sample collected before the Pu inhalation
3. Limit of detection determined from the measured count rate in a region of interest centred on 101 keV (the energy of the 2nd of the three main gamma-ray spectrum peaks of ^{237}Pu)
4. Uncertainties (1σ) on mass spectrometry measurements are estimated to be 19% of the measured value.
5. Bold, non-italic text entries - mass spectrometry measurements only
6. Plain text entries - gamma spectrometry measurements only
7. Bold, italic text entries - weighted mean of the gamma spectrometry and mass spectrometry measurement results. “Less than” values are not used to estimate mean values.
8. SNM = sample available but not measured by mass spectrometry
9. NA = not available (sample not measured by gamma spectrometry)
10. These six results are the most recent mass spectrometry measurements (as of August 2016), and were not taken into account for the analysis published in Puncher and Etherington (2017)
11. For ^{244}Pu, 1E+08 atoms = 4.1E-14 g = 41 fg

5 DISCUSSION

The purpose of this report is to provide a compilation of available data, updated as new results become available. Analyses of the data can be found in peer reviewed journal papers, reports and conference proceedings for the:

- CRCE ingestion and intravenous injection study (Popplewell et al., 1994; Ham and Harrison, 2000)
- AEAT / MU intravenous injection study (Talbot et al., 1997; Newton et al., 1998, 2005), and
- CRCE inhalation study (Etherington et al., 2002, Etherington et al., 2003, Puncher and Etherington, 2017).

The results of these studies have provided important input into the validation and improvement of models used by the International Commission on Radiological Protection (ICRP) for the calculation of doses from intakes of isotopes of Pu and related elements (ICRP, 1989, 1993; Leggett, 2003; Leggett et al., 2005). They are also being used in the development of models for Pu biokinetics to be implemented in Part 4 of ICRP's Occupational Intakes of Radionuclides (OIR) report series (see ICRP, 2015). Reliable models are important in routine protection but also in the interpretation of health outcomes in exposed workers, as illustrated by current efforts to estimate doses and provide risk estimates for lung cancer and other radiation related diseases in workers from the Russian Mayak plutonium plant (Kreisheimer et al., 2003; Shilnikova et al., 2003; Gilbert et al., 2004; Leggett et al., 2005; Birchall et al., 2016).

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