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INFORMAL REPORT

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A Twenty-Seven Year Study of Selected Los Alamos Plutonium Workers

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A Twenty-Seven Year Study of Selected Los Alamos Plutonium Workers*

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PREFACE

This report of the health of war-time Los Alamos plutonium workers was inspired by Wright H. Langham and is dedicated to his memory. Wright's keen interest in plutonium is known to everyone and, as one of the if not *the* world's authority on plutonium biochemistry and toxicity, he was the first to be called upon by our government as well as those of other countries whenever plutonium problems arose. His ultimate concern, of course, was the toxicity of plutonium in man and, in this connection, he organized the UPPU Club composed of the subjects of this study. He kept in close touch with the Club members by periodic circulation of friendly UPPU Newsletters. It was undoubtedly the humor and enthusiasm expressed in these Newsletters that has been responsible for the excellent cooperation of the UPPU Club members in this study.

Wright's intense interest in plutonium continued until his untimely death. In fact, an early draft of this manuscript and a number of now declassified war-time reports on plutonium experiences were with him at the time of the fatal plane crash. Although Wright died before he could write his interpretation of the present data, he contributed enormously to the report in the course of discussions of the subject during the winter of 1971-1972. Although we are well aware that the study suffers greatly by not having Wright's direct participation, the authors have tried to write a report of which he would have been proud. In an effort to make the report into a personal living story, which it was to Wright, we have included many of the anecedotes that Wright loved to tell.

A TWENTY-SEVEN YEAR STUDY OF SELECTED LOS ALAMOS PLUTONIUM WORKERS

Compiled and Written by

L. H. Hempelmann, C. R. Richmond, and G. L. Voelz

ABSTRACT

Twenty-five male subjects who worked with plutonium during World War II under extraordinarily crude working conditions have been followed medically for a period of 27 years. Within the past year, 21 of these men have been examined at the Los Alamos Scientific Laboratory, and 3 more will be studied in 1973. In addition to physical examinations and laboratory studies (complete blood count, blood chemistry profile, and urinalysis), roentgenograms were taken of the chest, pelvis, knee, and teeth. The chromosomes of lymphocytes cultured from the peripheral blood and cells exfoliated from the pulmonary tract were also studied. Urine specimens assayed for plutonium gave a calculated current body burden (excluding the lungs) ranging from 0.005 to 0.42 μCi, and low-energy radiation emitted by internally deposited transuranic elements in the chest disclosed lung burdens probably of less than approximately 0.01 μCi. To date, none of the medical findings in the group can be attributed definitely to internally deposited plutonium. The bronchial cells of several of the subjects showed moderate to marked metaplastic change, but the significance of these changes is not clear. Diseases and physical changes characteristic of a male population entering its sixth decade were observed. Because of the small body burdens on the order of the maximum permissible level in these men so heavily exposed to plutonium compounds, we conclude that the body has protective mechanisms which are effective in discriminating against these materials following some types of occupational exposures. This is presumably explained by the insolubility of many of its compounds. Plutonium is more toxic than radium if deposited in certain body tissues, especially bone; however, from the practical point of view, plutonium seems to be less hazardous to handle.

I. INTRODUCTION

This is the story of how 25 young men were heavily exposed to plutonium at what is now the Los Alamos Scientific Laboratory (at Los Alamos, New Mexico) in the days of the Manhattan Project during World War II and of what has happened to them in the subsequent 27 years. All but a few of the subjects of this study were college science majors who were drafted into the Army and assigned to a Special Engineering Detachment of the Manhattan Project. All were sent to Los Alamos (Project Y) in 1944 or 1945 and given various technical jobs in the Chemistry and Metallurgy Research Division. In these capacities, they were engaged in processing plutonium prior to fabrication and testing of the first atomic bomb. Almost all of these subjects had body burdens of plutonium estimated from the urine assay for plutonium used at Los Alamos before 1950 that ranged from 0.1 to 1.2 μg[1] (0.006 to 0.08 μCi).

II. SAFETY PROBLEMS CONCERNED WITH PLUTONIUM PROCESSING

The potential danger of exposure to plutonium was recognized early in 1944 by its discoverer, Glenn Seaborg.[2] He was aware of the similarity of the radioactive properties of plutonium and radium and of the extreme toxicity of the latter element which caused bone cancer in man after deposition of microgram quantities in the body. In an effort to learn more about the biological and metabolic properties of plutonium (and, hopefully, thereby to avert another disaster such as happened in the radium dial painting industry in the 1920's), Seaborg gave about 10 mg of plutonium out of the first half gram produced to Joseph Hamilton of the Crocker Radiation Laboratory in Berkeley, California, for biological experimentation. Thanks to Seaborg's foresightedness, many basic facts related to the biology of plutonium

were known when the first milligram samples of the material arrived at Los Alamos in the spring of 1944.

However, to be aware of the potential biological hazards of plutonium and to protect against them were two entirely different matters. Safety regulations could be established on the basis of experience in the radium dial paint plants, but protection problems in the two cases differed by many orders of magnitude. (Milligrams of radium in watch plants were subjected to simple mechanical operations, whereas at Los Alamos kilogram quantities of plutonium were involved in complex chemical and metallurgical manipulations.) All work with plutonium was carried out in the wooden Chemistry and Metallurgy Building called "D Building"* (see Fig. 1). Stringent safety regulations put into effect included (a) a complete change of street clothing on entrance to the contaminated areas with two changes per day of freshly laundered coveralls, canvas bootees, and surgical caps (all persons showered on leaving the building); (b) use of surgical rubber gloves and respirators (Wilson 750) during all chemical procedures involving plutonium; and (c) whenever possible, use of closed systems (at first, homemade dryboxes with rubberized canvas sleeves attached to surgical gloves). Sometimes it was impractical to use a closed system, and chemical hoods had to be used (not made of stainless steel in the early days). All workers were fully apprised of the hazards of plutonium (as they were then known) and were required to sign a statement saying they would abide by the safety rules. With rare exceptions, the workers cooperated to the best of their ability, although during the tension and feverish activity of developing the first atomic weapons it was difficult to avoid some shortcuts in the observation and enforcement of safety rules.

When milligram quantities of plutonium first became available to Los Alamos chemists and metallurgists, efforts to live with what was considered to be safe contamination levels were hampered by the fact that portable alpha counters and continuous air samplers had not yet been developed. However, because of the urgency of the times, work with plutonium had to proceed, and improvised methods of monitoring and decontamination were unbelievably primitive by today's standards (see Appendix A). Because stationary counters were the only means of detecting alpha particles from plutonium,** it was necessary to make "swipes" with lightly oiled filter paper of the laboratory surfaces considered most likely to be contaminated and to bring these paper strips to the alpha proportional counter for counting. This was done systematically on a daily basis (or after an accident) in each laboratory containing plutonium (6 swipes per laboratory). Any "swiped" area (approximately 6 x 6 in.) having an activity of more than 500 counts per minute required

*It is a matter of interest to modern day plutonium workers that the air entering the D Building was cleansed of dust by electrostatic precipitation methods. However, air leaving the building was unfiltered for the most part, since few, if any, of the exhausts even from plutonium-containing hoods had filters.

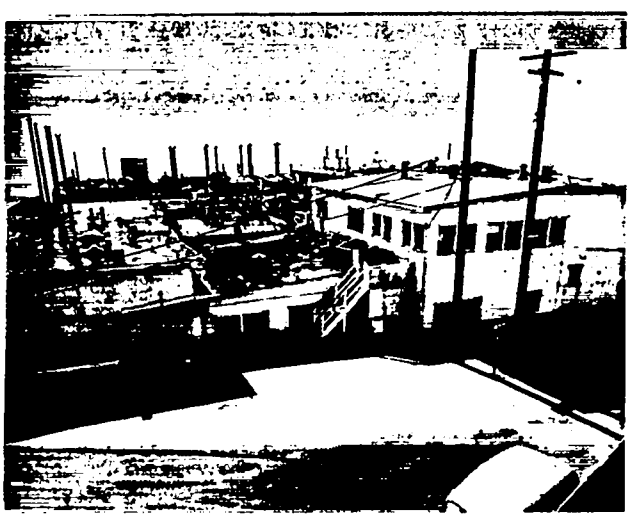

Fig. 1. The original wooden D Building which housed the chemists and metallurgists in CMR Division. Note that the chemical hoods of the laboratories on the first floor were vented individually, usually without filtration.

**There was a semi-portable counter called Pluto first available in March 1944, but it was very bulky and its limit of sensitivity was 5000 counts per minute. A more sensitive yet still cumbersome counter called Supersnoop came into use in March 1945, but it was not until the instrument called Poppy became available in June 1945 that the monitoring situation really was under control. Hand counters were not available until October 1944, when the first experimental models capable of detecting 200 counts per minute were installed in the locker rooms.

decontamination.*** It is difficult to believe but, because of the bulkiness and inconvenience of the early so-called portable counters, the swipe technique of monitoring was continued in some laboratories until June 1945. Similarly, in the absence of air samplers for radioactive dusts (the first became available in the fall of 1944), both nostrils of each plutonium worker were "swiped" routinely with rolled moistened strips of filter paper at the end of the working day or if accidental pulmonary exposure was suspected.**** These samples were also counted in a stationary alpha particle counter. If the nose swipe exceeded 50 counts per minute, the subject was questioned carefully about possible accidental inhalation or breakage of safety regulations (e.g., hand contamination of the nostrils).

In April 1945, kilogram quantities of plutonium from the newly operating Hanford piles began to arrive at Los Alamos for processing and fabrication into the first atomic bombs. Fortunately, by this time portable alpha counters (Fig. 2) and continuously operating air samplers were available to help with monitoring for contamination of certain laboratories. Operating equipment, including air lines in some laboratories and specially made positive-pressure masks, also had been improved. Although most procedures were carried out in dryboxes, some work had to be done in open hoods (Fig. 3). This was particularly true in the case of the Recovery Group, the activities of which were so diverse and unstandardized that closed systems were totally inoperable.[3] Also by March 1945, a

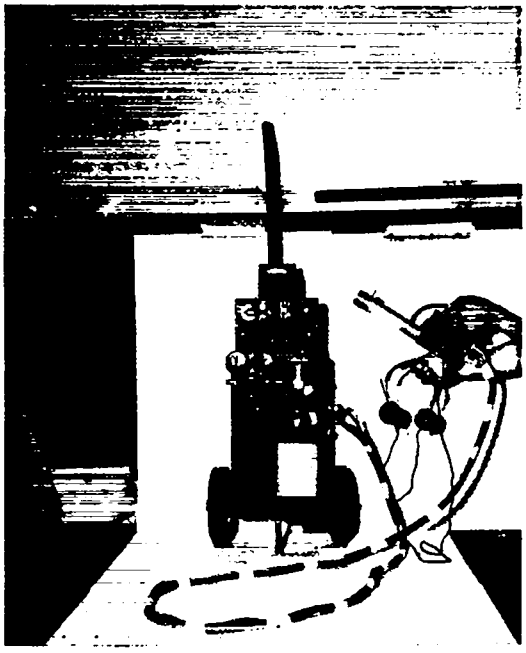

Fig. 2. A semi-portable alpha counter called "Supersnoop" used in early 1945.

urine assay method for plutonium[4] had been developed; this allowed crude estimates of the body burdens of plutonium to be made (not including insoluble compounds in the lung). However, even with improvements

Fig. 3. One of the steps in the purification operation performed in an open chemical hood. The operator is wearing the standard protective clothing and respirator (Wilson 750).

***The swipe technique measured only alpha particle-emitting materials not fixed to the surface. Oil on the filter paper used for surface swipes probably decreased somewhat the measurable alpha activity. The moistened paper strips used for nose counts were dried before counting; consequently, the measured activity was probably more accurate than that of the oiled swipes. The geometry of the stationary proportional counter was relatively good, approaching 50 percent. In contrast to poorly fixed radioactivity measured by the swipe technique, portable counters developed later measured fixed as well as unfixed radioactive contamination with a rather poor geometry (10 to 15 percent).

****The nose counts and air counts (at first one sampler in the most hazardous laboratory) were by no means accurate measures of exposure of personnel. At best, they were merely crude indications that permissible contamination levels had been exceeded or that safety regulations might have been violated.

in monitoring techniques, working conditions were deplorable by present-day standards. Fortunately, after initial bomb processing was completed in August 1946, all work with plutonium was sharply curtailed until the new and greatly improved facilities at DP Site in Los Alamos were opened in September 1946.

To illustrate the degree of contamination of laboratories in D Building in 1944 and 1945, we will present a few almost random experiences taken from various reports issued at the time. Table I shows the maximum and minimum "swipe" counts in all laboratories in D Building in March 1944 [500 counts per minute correspond to 0.007 μg or 0.0004 μCi of plutonium isotope mixture of that time; note the 2,500,000 count per minute (or 35 μg) swipe in D-117 (used by the Recovery Group*****)]. Figure 4 shows

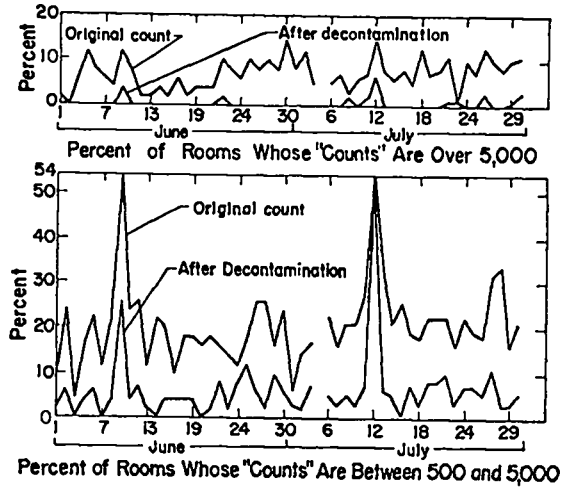

Fig. 4. Chart showing the percent of laboratories in D Building in June and July 1945 with "hot" spots requiring decontamination. The lower line indicates the counts after decontamination. Note that about 50 percent of the laboratories had contamination in excess of 500 counts per minute on two occasions.

*****During the war years, when plutonium was the primary concern of the radiochemists, the unit of measure of plutonium was usually considered to be its weight (i.e., micrograms, grams, etc.). In the 1950's, presumably because of the influence of the health physicists (and because chemists became more involved with physics), radioactivity (i.e., microcuries, etc.) became the unit of measure. Both units are given throughout this paper.

TABLE I

MAXIMUM AND MINIMUM SWIPE COUNTS IN D BUILDING (MARCH 1944)

Room No.	101	102	103	104	108	109	110	112	113	114
First	16	158	110	399	338	177	321	807	308	10
Maximum	274	3,319	739	14,560	4,652	4,078	15,176	17,450	561	172
Minimum	0	32	24	12	60	50	59	27	17	4
Last	18	642	122	150	826	3,547	59	51	57	31
Room No.	115	116	117	118	119	120	121	122	123	124
First	33	9	8	244	10	3	0	12	90	7
Maximum	70	58	2,500,000	6,000	1,387	816	81	128	130	176
Minimum	0	9	0	1	10	1	0	0	12	5
Last	21	37	1,198	2,368	48	22	42	24	64	46
Room No.	125	126	127	128	129	130	132	133	134	136
First	40	73	44	52	83	42	6	41	58	6
Maximum	4,592	17,832	363	94	83	3,173	5,796	610	68	3,712
Minimum	16	19	2	0	0	10	1	3	0	0
Last	506	28	22	46	6	71	85	13	25	91
Room No.	137	138	139	140	141	142	143	144	145	146
First	93	7	23	81	0	51	8	0	0	338
Maximum	2,928	448	23	81	15	51	15	0	114	338
Minimum	0	6	7	12	0	20	8	0	0	5
Last	833	33	12	12	2	20	8	30	126	16
Room No.	148	151	152	201	202	203	204	205	207	209
First	30	30	0	23	9	23	0	21	45	73
Maximum	30	79	104	23	9	23	0	21	45	73
Minimum	20	18	0	23	9	23	0	21	45	73
Last	20	36	42	23	9	23	0	21	45	73
Room No.	210	211								
First	19	22								
Maximum	19	22								
Minimum	19	22								
Last	19	22								

the percent of laboratories in June and July 1944 in which radioactivity levels (as measured by swipes) were above 5000 counts per minute or were between 500 and 5000 counts per minute. Table II shows the high nose counts (over 50 counts per minute) of three chemists in the Recovery Group (other nose counts of these men not included were consistently positive but below 50 counts per minute). In April 1945, 1243 "hot" (over 500 counts per minute) spots, mostly detected by portable counters, were decontaminated. In June, July, and August 1945, the number of decontamination procedures carried out were 1980, 3489 (of which 760 exceeded 30,000 counts per minute), and 5347, respectively. Lest the reader think that the operations were lax or careless, he should be advised that a large, well-trained monitoring staff of approximately 41 persons worked overtime to maintain the safety standards with the crude methods available. Pressures to build the bomb were so great that work had to proceed using the best, although admittedly unsatisfactory, safety measures of the times.

III. PLUTONIUM OPERATIONS CAUSING HEAVY EXPOSURES

Twenty-three of the 25 living exposed subjects****** worked in four operating groups: Plutonium Purification (wet chemistry), Plutonium Fluorination (dry chemistry), Plutonium Reduction (to metal), and Plutonium Recovery. Because the last operation was by far the most hazardous, it will be described first. Monthly reports of the Chemistry and Metallurgy Research and Health Divisions submitted in 1944 and 1945 (many still classified), as well as references 3 and 5, are the source of much of the material in this section. Extensive interviews with the subjects and their supervisors, as well as with the health monitors, provided supplementary information.

A. Recovery Operation

Fourteen of the 25 subjects with measurable body burdens of plutonium worked in the recovery operation between March 1944 and September 1946. This group was charged with the following responsibilities: (a) recovery of the then priceless plutonium which had been either left behind in any of the experimental or operational procedures or spilled accidentally or lost as contamination, and (b) converting the recovered plutonium to the +4 valence state suitable for fluorination and subsequent reduction to plutonium metal.

In 1944, the Recovery Group dealt first with milligram and later with gram amounts of plutonium; however, in March 1945, as a result of increased quantities being processed by the Chemistry and Metallurgy Division, the Laboratory began to handle kilogram quantities of plutonium. During peak work periods, the staff worked 12-hour shifts 7 days a week. Fortunately, by this time new laboratory facilities were in use in an annex of the old D Building. A suite of three laboratories (maintained under reduced pressure) had replaced the single laboratory D-117. These laboratories had open stainless steel hoods instead of the ordinary chemical hoods previously used in D-117. Although two of these laboratories were equipped with air lines, the commercially available positive-pressure masks were not entirely satisfactory in protecting personnel from airborne contamination. Not until July 1945, when a specially made mask (called the Kennedy-Hinch mask,******* after its designers) was developed, was there comparatively good protection against airborne radioactivity.

The types of plutonium-containing materials submitted to the Recovery Group for processing included analytical residues; washings (supernatant solutions) from various steps in the wet purification of plutonium; metallic scraps, shavings, and trimmings; crucibles composed of various materials used in the reduction of PuF_4; absorbent materials used to wipe up accidental spills and other contaminated materials; graphite tubes used in oxygen analysis; and drybox and chemical hood sweepings.

After considerable experimentation, the following basic procedure was adapted for recovery operations (see Fig. 5)********: (a) soluble salts of

******These men were the charter members of the UPPU Club formed by Wright Langham when this study began in the early 1950's. In essence, the acronym represents the phrase "you excrete plutonium." The name was originally IPPU, but Langham decided this was too personal.

*******The late Joseph W. Kennedy was the leader of CMR Division.

********It should be mentioned that not all of the recovered plutonium went through the entire procedure. Depending upon the chemical nature and purity of the plutonium being recovered, various steps of the total procedure were omitted.

TABLE II

EXAMPLES OF SUBJECTS WITH NOSE COUNTS ABOVE 50 COUNTS PER MINUTE

Date	Nose Counts[a]	Date	Nose Counts[a]	Date	Nose Counts[a]
Subject No. 5		Subject No. 8		Subject No. 9	
4-17-44[b]	175	6- 2-45	88-26	6-28-45	372-146
28	199	4	216-100	29	562-98
7-19-44	207	5	231-213	30	698-664
10-12-44	-144	6	20-60	7- 2-45	70-64
11- 7-44	188	7	291-201	3	208-140
12-29-44	207	9	286-89	6	112-80
4-20-45	717-408	11	1181-323	9	1164-608
5- 8-45	174-113	12	454-385		608-626[c]
15	73-27	13	1107-1292	10	57-50
16	196-172	14	509-229	11	879-706
18	421-41	15	623-560	23	838-842
19	122-107	16	1208-680		1296-306[c]
21	138-129	18	146-92	25	164-408
22	62-22	19	928-615	30	3040-464
29	101-67	20	1506-1009	31	2086-482
31 .	250-87	22	1898-1100	8- 2-45	8976-5180
6- 4-45	930-82	23	90-23	3	660-204
5	272-174	7- 2-45	400-274	4	674-530
6	163-43	4	164-170	6	3922-1752
7	267-131		236-97[c]		
9	177-118	5	324-126		
14	170-165		198-118[c]		
15	504-416	6	338-278		
16	246-514	7	268-72		
18	720-501	9	1016-694		
19	2559-3111	10	484-304		
20	199-150	11	250-247		
23	320-200	12	198-166		
25	126-78	14	356-328		
26	608-94	16	364-328		
27	68-50	17	462-410		
7- 2-45	217-182	18	752-545		
4	336-316		179-183[c]		
5	394-182	19	941-243		
	324-182[c]	23	898-816		
6	218-122	24	322-160		
7	486-152	25	166-154		
9	128-108	26	768-652		
10	92-56	8-13-45	124-56		
11	520-394	20	202-100		
	1096-329[c]	27	300-136		
12	64-80	9-12-45	106-78		
13	100-80	14	165-107		
14	100-60	17	136-72		
	148-130[c]	5- 8-46	52-825		
16	96-76	6-17-46	168-401		
17	100-96				
23	70-66				
25	64-52				
8-17-45	696-90				
20	144-48				
23	88-26				
24	76-60				

[a] Counts per minute of activity swiped from each nostril.

[b] Only one nostril swabbed.

[c] Accidental exposure suspected.

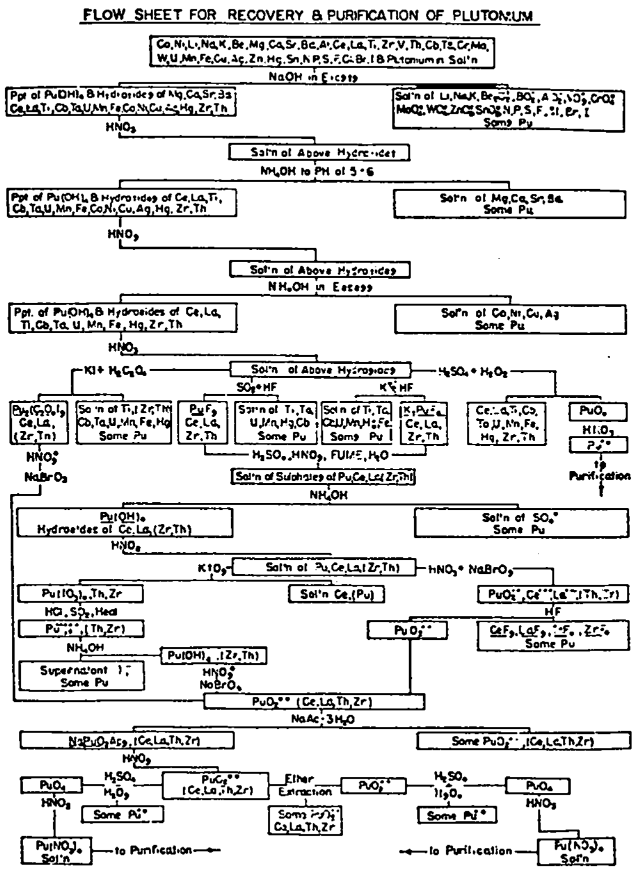

Fig. 5. Flow sheet for the recovery operation.[3] This gives the general outline of the procedures used, but each individual recovery operation required special conditions.

plutonium were dissolved directly in HCl; insoluble salts (oxides or carbides) also were dissolved in acid after persulfate or carbonate fusion; (b) plutonium was precipitated with NaOH or NH_4OH; (c) plutonium hydroxide was dissolved in HNO_3 and precipitated as the oxalate in the presence of reducing agents; (d) plutonium oxalate was redissolved in HNO_3 and oxidized to the hexavalent state by treatment with $NaBrO_3$; (e) ether extraction similar to that used in the purification process described later (steps d and e), which involved extraction from acid solution (in the presence of ammonium nitrate) and subsequent reduction with iodide and precipitation as the oxalate; and (f) after April 1945, the plutonium was extracted into ether and reduced to the tetravalent state and precipitated as the peroxide by addition of H_2O_2. The plutonium oxide was converted to nitrate (4 valence state) and was given to the Dry Chemistry Group for fluorination. Late in 1944, the dilute supernatant solutions were reduced with SO_2 and precipitated by treatment with $Al(OH)_3$. In December 1944, this process was turned over to another group (CM-10).

In retrospect, it may be reasoned that the greatest exposures occurred in step e when H_2O_2 was added (with vigorous stirring) to high concentrations of plutonyl nitrate (1 to 40 grams per liter). Although this procedure was carried out in an open hood in 3-liter cooled beakers, there was considerable fizzing and discharge of droplets into the air outside the hood. In April and May 1945, this procedure was often carried out by workers wearing respirators. (The number of high nose counts increased sharply when the peroxide step was added to the recovery procedure, but this also coincided in time with the increasing amounts of plutonium being handled.) The addition of NaOH to the large vats of dilute supernatant solution (0.1 to 1.0 gram per liter) also caused heavy fogging and occasionally spattering. Because of the size of the 50-liter containers, this procedure was carried out in the open laboratories without benefit of hoods. Other chemical steps which caused airborne contamination were concentrating dilute solutions by boiling; ashing combustible materials; and fusion procedures, especially those involving sodium peroxide plus carbon or persulfate.

As might be expected, in the atmosphere of excitement, tension, and fatigue that prevailed during the closing days of the war, the Recovery Group had its share of accidents which undoubtedly contributed to the exposures. The most dramatic of these occurred in May 1944 when the first 8-gram batch (the world's entire supply at that time) of purified plutonium was being processed. An initial 3.6-gram aliquot of plutonyl nitrate in solution, placed in a glass thimble prior to Soxhlet extraction, was heated in an oven. As it was being removed, the glass tray supporting the thimble broke and the concentrated solution spilled on the floor. The spilled material was aspirated by mechanical suction, and the remaining plutonium was recovered from the contaminated asphalt tile slabs torn up from the floor. When recovered and purified, this

material was combined with more newly purified plutonium, and the 8-gram sample, in the form of the hydroxide, was placed in a centrifuge tube. While being centrifuged, the tube broke,********* spilling the sample into the centrifuge cup and into the centrifuge itself. The plutonium was recovered once again by repeatedly washing the inside of the centrifuge with a dilute acid solution.********** Surprisingly, very little plutonium was lost in these two accidents.

In addition to these spectacular accidents, there were also occasional spills of large volumes (up to 50 liters) of dilute plutonium solution (5 to 10 mg) onto the floor. (The smaller concentrated solutions were handled in stainless steel hoods.) Four of the 14 subjects cut their hands with contaminated objects; fortunately, in each case, the excised tissues contained little radioactivity. One other man burned his hand while carrying out the carbon-sodium peroxide fusion procedure.

As might be expected, exposures during the late spring and summer caused great concern about the health of these workers. The following excerpt is taken from the Monthly Progress Report of the Chemistry and Metallurgy Division for May 1945 (issued June 1, 1945):

"The health situation in the recovery laboratories has become much worse in the past month due to the great increase in amount of product which is being processed. The trend in 24-hour urine counts is definitely upward. Because of this situation and because shutdown of recovery will essentially shut down purification, as well as increase hold-up of product, every effort is being made to improve the health situation. Forced air respirators have been put into use, although they have not materially altered the nose counts yet. Contamination of the air lines was suspected, but an investigation by CM-12 has failed to show appreciable contamination. The air pressure is being increased. Better respirators must be obtained quickly. Improvements in the handling of product are being made, but it will be some time before the hazard can be appreciably reduced except by the use of better protective devices. CM-1 has helped with a number of improvements such as installation of a new hood, studies on improving the ventilation, installation of foot-pedal sinks, etc. The situation is still critical, however."

All 14 subjects were removed in the summer of 1945 from this type of work because urine assays indicated that their body burdens were approaching or had exceeded the then maximum permissible levels (7 counts per minute per 24-hour urine specimen, suggesting a body burden of 1 μg or 0.06 μCi).

B. Purification Operation

Three of our subjects worked in the Purification Group in the Chemistry and Metallurgy Research Division. This group was given the responsibility of purifying by wet chemical methods the plutonium produced by the piles in Oak Ridge and later in Hanford. Because it was believed that nonmetallic impurities could best be removed by subsequent dry chemistry procedures (see next section), this group concentrated on removing contaminating metals.

The flow sheet used in the purification operation[3] until July 1945 is shown in Fig. 6. Plutonium nitrate slurry received in 160-gram lots in metal containers from Hanford was dissolved, diluted (cut), assayed, and transferred (sometimes with considerable difficulty) into a closed system. On the average, these preliminary steps took about 3.3 days. The following procedures were then carried out: (a) reducing Pu^{IV} (or Pu^{VI}) nitrate with iodide; (b) precipitating Pu^{III} oxalate and dissolving as Pu^{VI} with bromate and nitric acid; (c) precipitating plutonium as sodium plutonyl acetate and dissolving in HNO_3; (d) ether extracting plutonyl nitrate from the acid solution containing ammonium nitrate; and (e) reducing plutonyl nitrate with iodide and precipitating as the oxalate. This product, recovered in about 97 percent yield, was given to the Fluorination Group. Purification of plutonium in this form continued to

*********The reason for the accidents presumably was the unsuspected radiation-induced fragility of the glass vessels used in these procedures. It was believed that alpha particles alone would not affect the glass, but irradiation by neutrons arising from interaction of alpha particles with boron in Pyrex glass had not been anticipated.

**********There is a story that great difficulty was encountered in the fluorination of this ill-fated batch of plutonium. We have not been able to verify this.

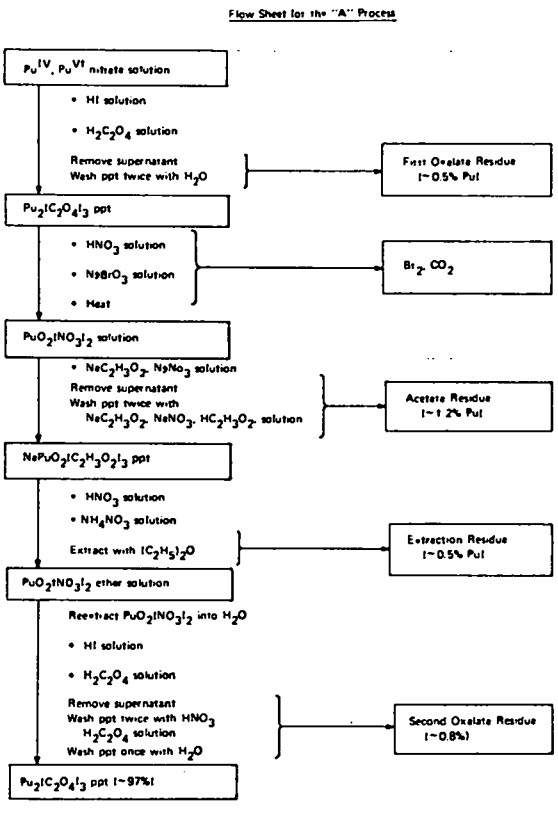

Fig. 6. Flow sheet for the purification operation.[3]

be carried out in D Building until September 1945.

Although this group handled 160-gram batches of plutonium totaling 4.9 kg in May 1945, 5.69 kg in June, 6.13 kg in July, and 9.2 kg in August, the hazards to personnel were not as great as those in the Recovery Group. The nature of the operations (mainly decanting supernatant solutions from precipitates) was such that they were quickly standardized and carried out in relatively simple closed systems. The body burdens of the 3 subjects in this group were consequently lower than those in the Recovery Group. Nevertheless, accidental exposures did occur. In August 1944, a vial containing 10 mg of plutonium chloride in the +4 state exploded while being opened and heavily contaminated the face and mouth of a young chemist. Although the skin of the face was thoroughly scrubbed and the mouth thoroughly washed out, heavy contamination of the face (estimated to be 1 μg) persisted for several days. From the ionization of expired air, it was estimated that the level of contamination in the mouth was of the order of 10 μg. By blowing at an open-faced ionization chamber across the room, the subject could cause the needle to go off-scale. This man is well today and did not have a body burden of plutonium as measured by older urine assay methods.**********

C. Fluorination Operation

Three of the subjects with measurable body burdens of plutonium worked on this operation. Their job was to convert plutonium oxalate (prepared by the Purification Group) to oxide and then to fluoride prior to reduction to the metallic state. In the spring and summer of 1945, these men handled plutonium in batches of 160 grams and worked in shifts around the clock.

Slurries of plutonium oxalate (from step 5 or 6 in the wet purification procedure) were poured into platinum boats and oxidized by baking in a tube furnace within a hood. The uncovered boats, containing oxide in the form of a light green powder, were carried across the room to an ordinary analytical balance also in a hood. After being weighed, these boats were carried back to a tube furnace in one of two large hoods with sliding glass doors and fired in the presence of hydrogen fluoride gas and oxygen. The plutonium fluoride was weighed and, if the conversion was not satisfactory, the fluorination procedure was repeated. Plutonium tetrafluoride powder finally was poured into a glass container, covered, and passed on to the Reduction Group.

The 3 men working in the fluorination operation could have been exposed to plutonium dust in the form of oxalate, oxide, or fluoride. Some exposure presumably occurred while they were carrying or weighing the powdered plutonium even though respirators were always worn. Further exposure could have occurred when the wooden hoods were decontaminated (i.e., by scrubbing with scouring agents, painting, etc.). Fortunately, there were no serious accidents such as dropping a boat in the open laboratory.

D. Reduction Operation

Three of the subjects worked in the metal reduction operation. They developed the methods of reducing plutonium fluoride to the metal, which was

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then fabricated into the shapes needed in the early atomic bombs.

Before plutonium became available in quantities, preliminary experiments were carried out using uranium and other metallic "stand-ins." The operators soon learned that reduction of plutonium and other metallic halides with alkali or alkaline earth metals was relatively easy but that, when operating on a small scale, the reduced metal tended to remain mixed with the cindery slag rather than to form well consolidated "buttons." (Since the melting point of plutonium was unknown at this time, the reaction vessels were considerably overheated, causing reactions with the refractories.) While experiments were carried out to enable plutonium buttons to be produced on a larger scale in stationary bombs, the separation of plutonium from the slag was first successfully achieved by the use of centrifugal force to throw the molten metal into the tip of a cone-shaped container. This was called the "centrifugal bomb" method. $PuCl_3$ or PuF_4, mixed with lithium in a refractory cone-shaped beryllium oxide liner, was placed in an atmosphere of argon in a steel bomb about 1 in. in diameter and 1-1/2 in. in height. This was welded shut and placed in an all-graphite centrifuge, heated electrically to 1100°C while being rotated. Good cohesive buttons were soon obtained. The first 500-mg metallic button of almost pure plutonium[5] was made by this method on May 26, 1944 (Fig. 7).

Although loading and unloading of the bombs were supposed to have been carried out in dryboxes, the welded steel bomb was difficult to saw open in such a confined space. On at least two occasions, certain procedures were carried out in the open laboratory. On one of these occasions, the nose count of one operator was 11,900 counts per minute, and the laboratory in which the work was carried on was heavily contaminated. This metallurgist and his assistant both have measurable body burdens of plutonium.********** In September 1944, this method was discontinued in favor of the "stationary bomb" method, which had been developed to the point where it was easier, quicker, and gave better results than the centrifugal method.

The principle employed in the stationary bomb method was to pack a mixture of PuF_4 and metallic calcium in refractory liners made of electrically fused MgO contained in argon-filled steel bombs. On heating the bomb a strongly exothermic/thermic reduction reaction occurred, and the plutonium metal separated clearly from the fluid CaF_2 slag. Addition of iodine to the charge helped ignition and improved collection efficiency. This became the standard method of reducing plutonium during the war. All loading and unloading operations were carried out in dryboxes. Because the lid was bolted on, disassembly of the bomb was easier than in the case of the welded bomb. Moreover, the well fused reaction products were less dusty than those in earlier small-scale experiments. Nevertheless, the metallurgist who developed this method does have a measurable body burden of plutonium.

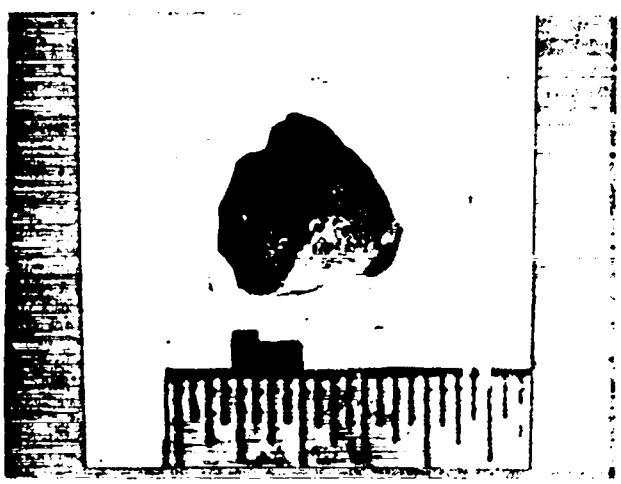

Fig. 7. A 500-mg metallic button of almost pure plutonium.

IV. EXPOSURE OF THE SUBJECTS

By March 1945, the plutonium urinary assay method[6] had been developed to the point where it could be applied to the plutonium workers. The assay system had to be extremely sensitive, as data

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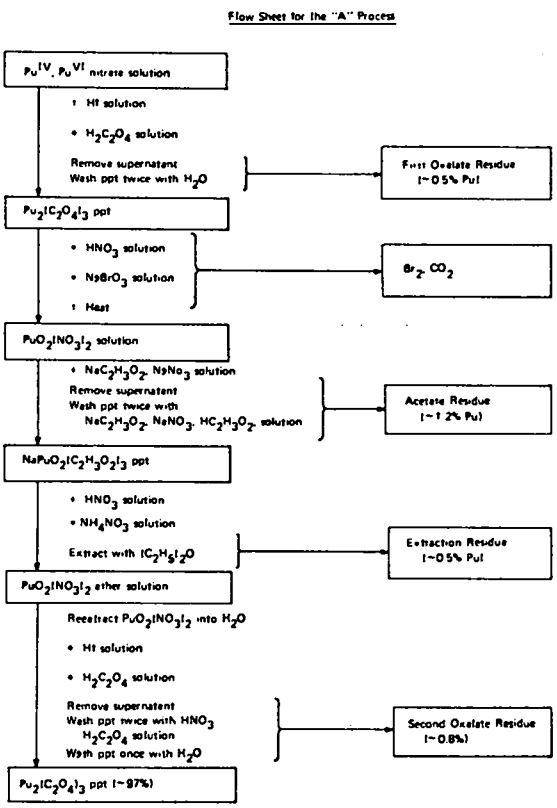

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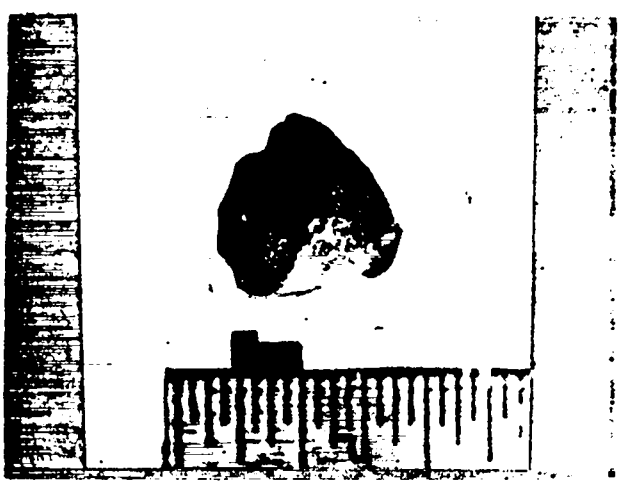

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from animal experiments suggested that a person with a maximum permissible body burden (at that time 1 μg) would excrete only 5×10^{-5} μg (50 pg) of plutonium per day (7 alpha counts per minute or 50 pg per 24-hour specimen). Nevertheless, the assay method gave us our first hope of estimating how much plutonium had actually been deposited in the bodies of our workers.

Beginning on March 11, 1944, urine samples of members of the Recovery Group were assayed monthly or oftener. To the consternation of everyone, the radioactivity of the initial 24-hour urine samples was of the order of hundreds of counts per minute, suggesting that the body burdens of plutonium were many times the acceptable levels. When a system of urine collection designed to minimize contamination of the urine specimens had been perfected (see Appendix B), the radioactivity of the urine samples decreased precipitously. Table III shows early estimates of body burdens of plutonium based on urine assays, as well as the number of high nose swipes (over 50 counts per minute) and total activity of all high nose counts for each subject.[6] Although the correlation between the number of high nose counts and body burden is not good, 8 of the 11 persons with more than 20 high nose counts

TABLE III

RESULTS OF URINE ASSAYS AND NOSE SWAB COUNTS CONDUCTED ON LOS ALAMOS PLUTONIUM OPERATORS SHOWING POSITIVE EXPOSURE

Subject Number[a]	Average Date of Exposure	Estimated Body Burden (μg ± 50%)	Total Number High Nose Swabs	Total Activity in[b] High Nose Swabs (cpm)
1	Late 1944	0.5 - 1.0	1[c]	11,606
2	Late 1944	0.1 - 0.5	3[c]	290
3	May 1945	1.2	37	4,267
4	June 1945	1.2	24	14,968
5	June 1945	1.2	55	27,246
6	June 1945	1.0	32	8,859
7	June 1945	1.0	28	15,699
8	June 1945	0.7	60	36,407
9	July 1945	1.0	22	39,778
10	July 1945	0.8	24	5,334
11	July 1946	0.4	6	
12	July 1945	0.4	23	8,607
13	July 1945	0.3	8	2,016
16	July 1945	0.1	4	5,403
17	August 1945	0.7	14	7,762
18	August 1945	0.6	9	6,429
19	August 1945	0.5	8	2,266
20	August 1945	0.3	6	2,417
21	August 1945	0.3	28	7,470
22	August 1945	0.3	22	11,688
23	September 1945	0.3	8	2,541
24	September 1945	0.1	8	5,107
25	September 1945	0.1	8	4,984
26	October 1945	0.3	3	478
27	October 1945	0.3	11	18,342

[a] Subject Nos. 14 and 15 were dropped because of the death of one subject of coronary heart disease and the low body burden of the other as determined by modern assay techniques.

[b] This column was added to the table given in reference 6 and represents the sum of all high nose counts (both nostrils).

[c] Incomplete records were available for these cases.

had a body burden of 0.5 μg or more compared with 3 in 11 persons with lower levels. Furthermore, 6 of the 8 persons with total nose counts exceeding 10,000 counts per minute were in the former group.

In Fig. 8 the urine radioactivity, the number of high nose counts, and the total activity of the high nose counts per month of exposure of 8 subjects in the Recovery Group are correlated with the amount of plutonium processed and protective measures used. There was a sharp rise in plutonium excretion in 6 subjects (Nos. 3, 4, 5, 6, 9, and 17); in each case, the rise was preceded by a high activity of nose swipes, suggesting considerable exposure to airborne plutonium. There is no correlation between the level of body burden of plutonium as indicated by the urine radioactivity and total activity in the nose, but this is not surprising in view of the crudeness of the nose count technique and the inevitable false positives due to contamination.

V. CLINICAL, LABORATORY, AND RADIOACTIVITY OBSERVATIONS

A. Medical Observations

In the early 1950's it was thought that 29 plutonium workers had measurable body burdens according to the assay methods of the time, but 3 have been dropped from the series as a result of the use of more reliable methodology, and one other died. In 1953, a program for periodic examination of these men (financed by the U. S. Atomic Energy Commission) was established. At first, a very thorough study every 2 years was planned. This included interval history and physical examination, complete blood count, blood calcium, phosphorus and alkaline phosphatase, urinalysis, and stool examination for occult blood. The following roentgenograms were taken: lateral skull, PA chest, AP pelvis, AP of knee and elbow, lateral of foot, AP of foot, and dental films of right teeth.

In 1953 and again in 1955, 22 and 25 subjects

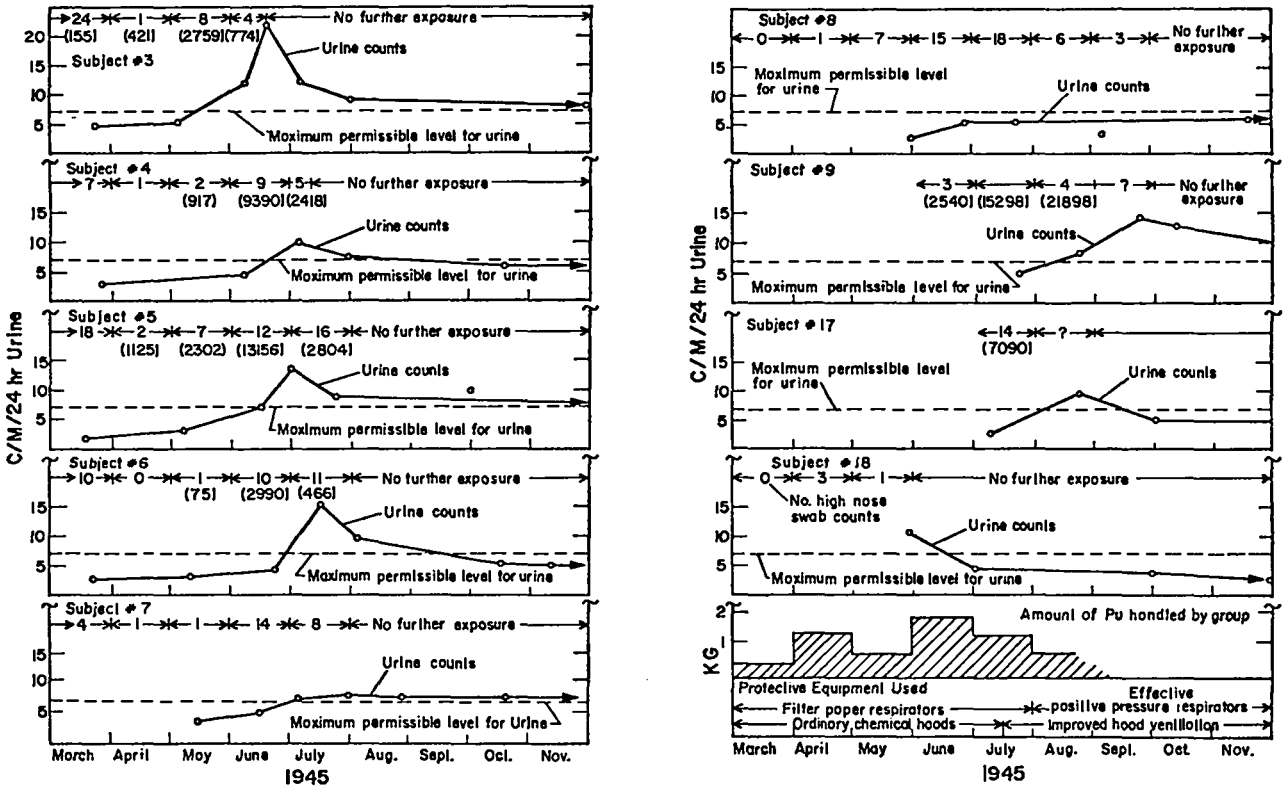

Fig. 8. Graphs for 9 subjects showing the urine count, number of high nose counts per month, and total radioactivity of nasal swipes per month (parenthetical figures) from March to November 1945. The amount of plutonium handled by the Recovery Group and the protective measures used are shown in the last graph of the second chart.

of the series, respectively, were examined by physicians associated with the U. S. Atomic Energy Commission. There were no significant medical findings, but some concern was expressed about what appeared to be an excess of bone islands in the roentgenograms. However, after careful comparison with normal male subjects of the same age, this observation was considered to be within normal limits. After reviewing the experimental program in 1956, it was decided to increase the interval between examinations to 5 years and, on the advice of Dr. William Christiansen of the University of Utah, to reduce the number of x-rays. All 25 of the men were examined by their family physicians in 1960, 17 in 1966, and 24 in 1970.

In late 1971 and early 1972, 21 of the 25 subjects came to Los Alamos for a complete study including urine assays for plutonium, *in vivo* measurements for ^{239}Pu in the chest, pulmonary cytology, and chromosome analyses. The medical examination consisted of the following procedures: complete history and physical, complete blood count, urinalysis and blood chemistry profiles (alkaline phosphatase, cholesterol, total bilirubin, total protein, albumin, globulin, A/G ratio, total lipid, SGOT, LDH, creatinine, glucose BUN, and urea). Roentgenograms were also taken of the chest, pelvis, and teeth.

Except for the ailments that one would expect in a group of men mostly in their early fifties, all subjects examined were in remarkably good health. One man had a coronary occlusion but had recovered and was well compensated, and another of the original group died in 1959 of a coronary at age 38. Another had a hamartoma of the lung surgically removed without complication in 1971 (see Appendix C and section V.C). A third had a melanoma of the chest wall (regional lymph nodes were negative). A fourth had a partial gastrectomy for a bleeding ulcer. Several had mild hypertension and moderate obesity, and one had gout. All men were actively working, most as successful executives.

No roentgenographic changes in the lungs or bones were apparent. The lamina dura of the jaws (which show the first changes in beagles given toxic doses of plutonium) were intact in all cases except in one edentulous subject.

B. Plutonium Body Burdens as Determined by Urine Assay

The estimates of body burden of plutonium as determined by assay of the urine of the subjects made during the 20-year observation period (1953-1972) are shown in Table IV. Because more knowledge has been gained and techniques have changed completely since 1953 when the first reasonably sophisticated estimates were made, we have presented in Appendix B a detailed description of the evolution of the methods of urine collection, radiochemical separation of plutonium, radiometric measurements, and the bases for estimation of the plutonium body burden. The current (1972) version of the PUQFUA (Plutonium Body Burden From Urine Analysis) code[7] was used to estimate what is considered to be the best value for the body burden of the

TABLE IV

PLUTONIUM BODY BURDEN ESTIMATES FOR UPPU SUBJECTS[a]

Case Code	1953[1]	1957[6]	1962[7]	1972
1	0.03 - 0.06	0.03 - 0.06	0.01	0.206
2	0.006 - 0.032	-	-	0.03
3	0.08	0.07	0.13	0.42
4	0.08	0.08	0.14	0.26
5	0.08	0.07	0.14	0.18
6	0.06	0.06	0.07	0.14
7	0.06	0.06	0.08	0.15
8	0.04	0.04	0.05	0.11
9	0.06	0.06	0.11	0.11
10	0.05	0.05	0.03	0.10
11	0.03	0.02	0.03	0.05
12	0.03	0.02	0.02	0.12
13	0.02	0.02	0.04	0.005
16	0.006	0.006	0.002	0.03
17	0.04	0.04	0.09	0.13
18	0.04	0.04	0.04	0.10
19	0.03	0.03	0.06	0.02
20	0.02	0.02	0.02	0.05
21	0.02	0.02	0.03	0.04
22	0.02	0.02	0.02	0.05
23	0.02	0.02	0.04	0.04
24	0.006	0.006	0.01	0.03
25	0.006	0.006	0.01	0.01
26	0.02	0.02	0.03	0.006
27	0.02	0.02	0.03	0.05

[a]Microcurie ± approximately 50 percent.

subjects. Assay of replicate urine samples was used as the basis for the values in Table IV. In the last column, all data acceptable to the PUQFUA code were used. In all but 2 cases, the current estimates of body burden are higher than those in 1953, usually by a factor of 2 to 3 and sometimes by a factor of 5 to 6. Since each figure is based on a number of assays ranging from 5 to 125 for different subjects, some values are considered to be firmer than others. There is uncertainty in these values, especially those based on relatively few assays, because of the inevitable errors in extrapolating the excretion curve back to the time of exposure many years before (see Appendix B).

The values shown in the last column of Table IV range from 0.005 to 0.42 μCi $^{239-240}$Pu, or from approximately 1/8 to 10 times the current maximum permissible body burden (0.04 μCi) for occupational workers. Eighteen of the 25 values listed for 1972 are equal to or exceed the 0.04 μCi value. In 1962, only 12 of the 25 subjects were thought to have body burdens exceeding 0.04 μCi. To test the validity of the values for body burden based on urine assay, we have compared the estimates based on urine assay of case No. 2 with that calculated from radioactivity of the rib removed prior to lobectomy. The latter estimate is crude, as it involves a large extrapolation and assumes uniformity of distribution of plutonium throughout the entire skeleton. For a 7000-gram skeleton, the plutonium content would be 11.27 nCi. If the liver content was of the same order of magnitude (not an unreasonable assumption), then the body burden in these two organ systems which contain most of the radioactivity (excluding lung) would be about 22 nCi. The estimated body burden for case No. 2, based on urine assay (Table IV), was 30 nCi. Most probably the excellent agreement is fortuitous.

Comparing the minute quantities of plutonium deposited in the body (excluding the lungs) with the large amounts to which the subjects had been exposed, we can only conclude that the gastrointestinal tract has a remarkable ability to exclude plutonium from entering the body. Had plutonium been as readily absorbed as radium, all subjects would unquestionably have lethal body burdens of radioactivity.

One of the first references to estimates of the body burdens of the subjects included in this report is found in the 1952 annual report of the Biomedical Research Group of the LASL's Health Division.[1] The basic information given in this report (with one minor change which may have been a transcriptional error) appeared in another report the following year,[6] together with information on nose swabs, as shown in Table III. In 1962, Lawrence[7] updated the Langham estimate of the body burdens on the basis of additional urinary assay data collected just prior in 1962. Lawrence used a computational model called PUQFUA[8] which, although based on the Langham metabolic model, was programmed to eliminate certain data points believed to be uncertain. On the average, the 1962 PUQFUA estimates of body burden for the 25 subjects were 1.6 times those estimated earlier by Langham. The differences among the values (shown in Table IV) are not surprising, as Lawrence's numbers were based upon more data collected over a longer time period, and his computer system was programmed to select the data to be used in the analysis (e.g., criteria for excluding data points are built into the PUQFUA code). Short discussions of the plutonium body burdens of the UPPU subjects also appeared in several other publications by Langham and colleagues.[9,10]

Some data for the early Los Alamos subjects appeared in 1969 in a document published by Langham.[11] The first 12 cases in Langham's table, shown in our Table V, are members of the UPPU Club. Of added interest is the fact that accumulated average organ doses included for bone, liver, and lung were estimated using the total radiation doses from time of exposure until the late 1960's. These dose estimates were based on metabolic models recommended by the International Commission on Radiological Protection. It is interesting to note that the estimated average accumulated doses in bone were less than 18 rads in all subjects. Since plutonium is concentrated on the periosteal and endosteal surfaces rather than uniformly distributed in bone, the dose to individual osteocytes may be considerably higher than this. The average liver dose was estimated to be up to 5 times higher than the bone dose.

To evaluate the possible consequences of bone doses of this magnitude, we must refer to animal data, particularly that in dogs given plutonium intravenously. The oldest and most extensive of

TABLE V

ESTIMATED ACCUMULATED AVERAGE ORGAN DOSES FOR PERSONS EXPOSED TO PLUTONIUM BY INHALATION[11]

Time since Exposure (years)	Estimated Exposure (MPL)[b]	Accumulated Average Organ Dose[a]		
		Bone (rads)	Liver (rads)	Lung (rads)
24	1.4	6.7	37	80
24	1.4	6.7	37	80
24	1.6	7.6	42	90
24	1.8	8.6	47	100
24	2.0	9.6	53	110
24	2.3	11.0	61	130
24	2.5	12.0	66	140
24	3.3	16.0	87	190
24	3.4	16.0	90	190
24	3.6	17.0	95	200
23	1.1	5.0	28	63
23	1.1	5.0	28	63

[a]These values are being brought up-to-date using the latest values for the body burden. This is a subject of a future report.

[b]One maximum permissible level (MPL) = 0.04 μCi.

these studies has been carried out at the University of Utah Medical School.[12] On December 1, 1952, the first group of 6 beagles were injected (IV) with ^{239}Pu citrate. Injected doses ranged from 0.016 to 2.9 μCi per kg body weight for 6 groups of about 12 dogs each. The first animal to die in July 1956 of an osteogenic sarcoma was in the highest dose group. In 1961, an osteosarcoma occurred in a beagle at the then lowest dose (0.016 μCi per kg), and since then several lower dose groups have been added to the experiment. The lowest dose group, 0.00064 μCi per kg, corresponds roughly to the maximum permissible occupational body burden (0.04 μCi) in man.

To date, the 0.016 μCi per kg dose level is of interest, as 4 dogs have developed osteosarcomas, giving a tumor incidence of 33 percent. The average time from injection to death for these animals was 9.9 years, and the average skeletal dose from injection to death was estimated to be about 80 rads[13] (a more recent estimate suggests about 50 rads). We do not know whether osteosarcomas will be produced at dose levels lower than 0.016 μCi/kg and can only await the outcome of this important experiment. However, we do know that the tumor incidence decreases and the time required for tumors to develop increases as the amount of plutonium injected is reduced. These observations have prompted numerous investigators to predict no adverse biological effects (e.g., practical threshold) below certain levels of injected plutonium. This point has not been accepted by everyone and has not been proven unequivocally for alpha-emitting radionuclides.[14]

C. Determination of Plutonium in the Body by *In Vivo* Measurements

1. Plutonium in the Chest Cavity. During the most recent (late 1971-early 1972) medical examinations at Los Alamos, estimates were made of the amount of plutonium in the lung and respiratory lymph nodes of each subject using an *in vivo* lung counter (see Appendix D). Figure 9 shows the pair of detectors in position for counting the low-energy x-rays emerging from the chest. Measurements also were made of the liver region in several subjects and of the hands of persons known to have had skin wounds contaminated by plutonium. The minimum detectable activity (MDA) for human subjects measured with the LASL detectors varies with several parameters such as body build and chest thickness. An average MDA for a 2000-second counting time is about 7 nCi if one uses the 95 percent confidence level. For the 68 percent confidence level and a similar

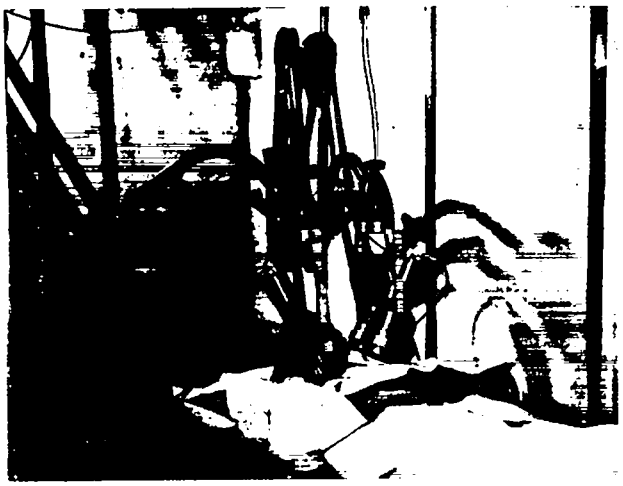

Fig. 9. The *in vivo* counter with detectors in position for counting the chest.

counting time, the comparable value is about 3.5 nCi.

Positive counts were obtained for 14 of 21 persons measured. These counts suggested chest burdens ranging from 3 to about 10 nCi. However, in no case did the estimated chest burden exceed the MDA at the 95 percent confidence level. Seven of the 14 subjects with positive chest counts had estimated chest burdens of 7 nCi or greater and may be considered (at the 68 percent level of confidence) to have statistically significant chest burdens of from 7 to 10 nCi. For reference, 10 nCi is about 2/3 of the maximum permissible lung burden for occupational workers (16 nCi). If maintained indefinitely, this burden will deliver about 15 rem per year to the lung, assuming uniform distribution of energy throughout the organ.

If one uses the 68 percent confidence level, certain qualitative statements can be made about the chest measurements. For example, the estimates of lung burdens of subject Nos. 1 and 2, who most likely received exposure to plutonium oxide, were approximately 10 nCi. Subject Nos. 4 and 9, who worked in the Recovery Group, each had a chest burden of about 8 nCi. These relatively small values are not surprising, in view of the known translocation of plutonium from the lung to other tissues as a function of time following inhalation.

The cumulative radiation doses to the lungs of some of our subjects have been estimated previously by Langham[11] (Table V). The values, all less than 200 rads, are below the lowest doses which caused adverse biological effects seen in experiments using beagles. For example, Bair and colleagues at Battelle-Northwest have not observed lung effects below about 2000 rads in dogs 10 to 12 years following a single inhalation exposure to $^{239}PuO_2$.[15] However, these animal data must be interpreted with caution, as lung tumor incidence is essentially 100 percent at the lowest plutonium exposure levels.

Recently beagles have been exposed to lower initial alveolar depositions, but additional data on the dose-response function will not be available for many years. The estimated initial alveolar-deposited plutonium in beagles which developed lung tumors ranged from 0.2 to 3.3 μCi per total organ, or about 3 to 45 nCi per gram of bloodless dog lung. Park and his associates[16] estimate that this amount of plutonium is 100 to 1500 times the estimated maximum occupational lung burden in man (i.e., 0.016 μCi or about 0.03 nCi per gram of bloodless lung). As a conservative estimate based upon extrapolation of their data to the 15-year mean life span of beagle dogs, these authors feel that initial deposits of about 70 nCi in the lung (about 1 nCi per gram), which cause premature death in beagles, are about 30 times the concentration equivalent to the maximum permissible occupational lung burden for man. This conclusion, of course, is based on the assumption that lung tumor development is related to plutonium concentration and not to the total amount of plutonium in the lung. For example, if tumor induction is somehow related to the total number of cells at risk (therefore, the number and size of plutonium particles), then the relative sizes of human and dog lungs may be unimportant and the 70 nCi which causes premature death from lung cancer in the dog may be about 5 times the total amount of plutonium (70/16) considered to be the maximum occupational lung burden for man.[18]

The experimental data obtained from rodents are not as clear nor as encouraging. Moskalev[17] reports that the frequency of malignant neoplasms of the lungs of rats exposed to plutonium by inhalation is 2 or 3 times higher than in the control group at doses ranging from 41 to 234 rads. Moskalev states that, assuming the biological effect

is independent of the distribution of dose in time, we must accept a dose of 75 rads (accumulated over 50 years of occupational exposure) as inducing malignant tumors and pulmonary sclerosis.[17]

In man, Langham[11] estimated 100 rads to the lung as a completely empirical judgment as to a level "at or above which biological consequences may ensue in a small population of limited distribution."

Park and co-workers at Battelle-Northwest report that the plutonium content of the thoracic lymph nodes of beagles was about the same as that of the entire lung tissue about 5 years following a single inhalation exposure.[16] At this time, both lung tissue and thoracic lymph nodes contained about 30 percent of the initial alveolar-deposited $^{239}PuO_2$. At about 10 years following exposure, the lungs contained about 12 percent and the thoracic lymph nodes about 40 percent of the initial alveolar-deposited $^{239}PuO_2$. From these data, we might expect a significant fraction of the original lung burden of our human subjects to be present in the thoracic lymph nodes many years after exposure to plutonium oxide. However, one must exercise caution, as we do not know whether the translocation rates of plutonium oxide are the same for the beagle and man or whether the rates observed for beagles with large lung burdens would be realistic for *smaller* initial alveolar deposits in man. Although a single measurement of ^{239}Pu in the chest of subjects exposed 27 to 28 years ago is of little help in obtaining accurate estimates of the current chest burdens, similar periodic measurements on several recently exposed individuals have yielded data on temporal changes of plutonium within the chest. Had repeated, periodic measurements been made on the present subjects, we would have more confidence in our estimates of the chest burden.

The only direct measurement of plutonium in the lungs in this study was made on the operative specimens of subject No. 2. Table VI shows results of analyses for ^{239}Pu. The concentration of ^{239}Pu was approximately the same in the tumor and lung tissue, while the concentration of ^{239}Pu in bone was approximately half. The highest concentration, observed in the lymph node tissue, is consistent with the experimental findings in dogs exposed to PuO_2 by inhalation, as mentioned above. It is well established that the concentration in lymph nodes relative to that in lung tissue increases as a function of time following exposure. If one assumes a homogeneous distribution of ^{239}Pu throughout the lung and lymph nodes and a total lung weight of 1000 grams and respiratory lymph node weight of 20 grams, the total plutonium burden of the lungs and respiratory lymph nodes is approximately 8 nCi roughly equally divided between lung and lymph node. For reference, the total amount of plutonium in the lung of case No. 2 (estimated by tissue assay to be 3.85 nCi) is approximately 550 times contemporary total lung burdens in humans in the United States exposed to fallout resulting from plutonium dispersed by atmospheric weapons tests. Estimates of the chest burden of ^{239}Pu of subject No. 2, based on extrapolation from analysis of lung and lymph node tissue, are in reasonable agreement (a factor of about 2) with estimates based on chest-counting. Figure 10 shows an autoradiograph of a plutonium particle in a section of lymph node removed from subject No. 2. Observations of other plutonium particles in the lymph node tissue examined indicate a very nonuniform radiation dose-distribution from plutonium particulates.

2. Plutonium in Other Tissues. Because we have no reliable calibration system for plutonium

TABLE VI

PLUTONIUM-239 CONTENT OF TISSUES REMOVED FROM PATIENT NO. 2 IN MAY 1971[a]

Tissue	Wet Weight (grams)	Volume of Solution (ml)	Plutonium-239 (dpm per ml)	Plutonium-239 (dpm per gram)	Plutonium-239 (pCi per gram)
Lung	70.85	100	6.01	8.48	3.85
Lymph node	1.25	25	22.55	451.00	205.00
Hamartoma	0.77	25	0.23	7.47	3.40
Rib	20.00	100	0.71	3.55	1.61

[a]After ashing and dissolution of tissue.

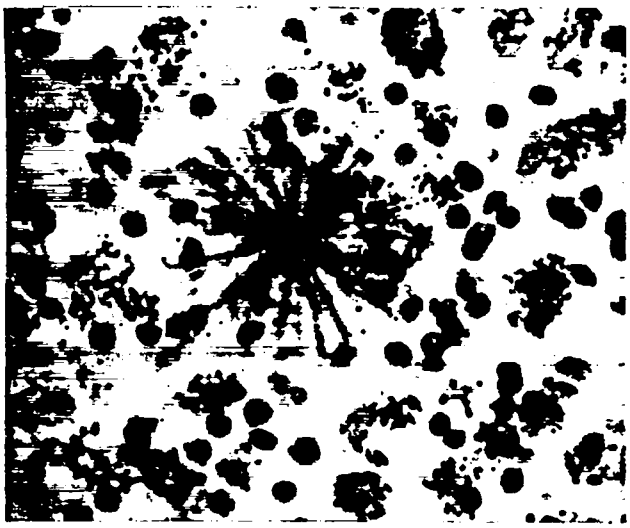

Fig. 10. Autoradiograph of lung section of subject No. 2 showing a radioactive particle.

in the liver of human subjects, we can make no quantitative statements concerning the observations in several persons who were measured. Plutonium within the bone cannot be measured by the *in vivo* counting technique. Traces of plutonium were detected by counting the hands of several subjects known to have hand wounds while working with plutonium in 1944 or 1945. The site was precisely located in one case (subject No. 1) and the amount estimated to be 5.3 ± 0.4 nCi. Of interest is the observation that the subject erroneously thought that the wound was on the other hand. At present, the skin overlying the active site appears to be grossly normal except for minimal scaling. We plan to make additional measurements on and observations of this deposit in the future.

D. Chromosome Analysis

During the 1971-1972 visit to the LASL, blood samples were obtained from the UPPU group for chromosome studies using standardized, established techniques. Although no abnormalities were found in these subjects, we plan to add newly developed chromosome banding techniques to our procedures when the group is next studied.

During recent years, considerable interest has been directed at the effects of various stressing agents on chromosomes of lymphocytes in the peripheral blood. The induction of chromosomal aberrations in human peripheral leucocytes has been studied in persons exposed to ionizing radiation as well as to a variety of exogenous agents such as cyclamates, caffeine, and LSD. More recently, the effects of elements such as cadmium that are environmental pollutants suspected of contributing to human disease[18] have been studied. Radiation-induced human chromosome aberration yields have been used to estimate radiation doses received by people involved in radiation accidents involving whole-body exposure to ^{60}Co.[19] Similar studies have also been performed on peripheral blood leucocytes obtained from subjects exposed years ago to Thorotrast;[20,21] the element ^{232}Th decays through 6 alpha and 4 beta disintegrations to ^{208}Pb (about 90 percent of the radiation energy is associated with high LET alpha rays). Radium daughter products deposit in bone, thoron is exhaled, and consequently both bone and lung tissue are irradiated. Finally, the chromosomes of blood lymphocytes of patients with body burdens of radium have been analyzed; many of these patients were exposed prior to World War II.[22,23] In both Thorotrast and radium patients, marked excesses of chromosome aberrations were reported.

Because of observations of chronic lymphopenia in dogs exposed to plutonium oxide aerosols, one might expect to observe chromosome damage in lymphocytes of exposed plutonium workers. This observation led Dolphin[24] to investigate the possibility of chromosomal aberrations in lymphocyte cultures obtained from workers in England known to have been exposed to plutonium. He compared the findings in 8 plutonium workers who had been exposed to plutonium plus 14 rads of external irradiation over a 7-year period with workers who had received external irradiation only and found that the dicentric yield of lymphocytes of the plutonium workers could be accounted for by the external radiation dose received by the workers. Dolphin[24] also cites another case in which a plutonium worker was found, by chest-counting, to have 10 to 20 times the permissible level of plutonium in the lung about 3 years after an inhalation accident. Chromosome analysis indicated minimal radiation damage to the lymphocyte series even at a high level of exposure of the patient. Other investigators are studying chromosomes obtained from uranium miners exposed to a variety of stressing agents, including radiation,

during their employment. However, one must use caution when interpreting quantitatively results obtained from chromosome analysis, as the number of aberrations cannot continue to increase with the accumulated dose because of the finite life span of the lymphocyte. For long periods of chronic exposure, one would expect a constant equilibrium level of aberrations.

E. Cytology of Exfoliated Bronchial Cells

Sputum cytology has been used by Saccomanno[25] as a possible indicator for detecting incipient or actual lung cancer in uranium miners exposed to radon daughters and other stressing agents in uranium mines. Because lung cancer has been observed experimentally in animals exposed to plutonium aerosols, we have added the cytological examination of bronchial cells to our periodic studies of the UPPU Club members. In a few subjects, moderate to severe dysplastic changes have been observed. The significance of these changes is not clear except in one man who was a heavy cigarette smoker (3 packages per day). We are now in the process of developing standardized procedures for collecting and preparing sputum samples. This presents a sizable problem, since our subjects live in different parts of the country often without easy access to large medical centers.

Since exposure to tobacco smoke and other toxic materials is known to alter the normal cytology of bronchial cells, it is difficult to interpret the observed effects. Primarily for this reason, we have asked all of our subjects to give up smoking. There is also a need to develop a standardized nomenclature to be used in reading and reporting sputum specimens. At periodic intervals, we will send bottles containing fixative for collecting sputum samples to each subject, who will then return the samples to us for analysis. If anyone shows more than a moderate cellular atypia, he will be asked to submit samples more frequently. We will also obtain sputum samples for cytological examination from local nonexposed personnel of the same age and smoking habits for purposes of comparison.

VI. CONCLUDING REMARKS

This report attempts to reconstruct the exposure conditions of Los Alamos plutonium workers during World War II and gives factual data on estimated body and lung burdens of plutonium, as well as medical and laboratory findings. A more comprehensive interpretation of these data will be the subject of a future report.

ACKNOWLEDGMENTS

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This report covers information and work that has been carried out over a period of many years and has involved many persons who have contributed to the success of the project. The special contributors have all played a prominent part in data collection or analysis in their particular specialty. The analytical urinalysis procedures were done by E. Campbell, M. Milligan, and W. Moss at the Occupational Health Laboratory, LASL. J. Lawrence calculated the current body burden values by means of the PUQFUA program. Roentgenographic studies were made by P. Lee, M.D., Los Alamos Medical Center, and W. Christiansen, M.D., Department of Radiology, University of Utah. *In vivo* measurements of plutonium within the lung were performed by P. Dean at the Health Research Laboratory, LASL. Pulmonary cytology studies were made by G. Saccomanno, M.D., Pathologist, St. Mary's Hospital, Grand Junction, Colorado, and Michael Stewart, M.D., Pathologist, Los Alamos Medical Center, Los Alamos, New Mexico. D. Petersen, Health Research Laboratory, LASL, assisted in procuring the samples for pulmonary cytology. The chromosome studies were performed by D. Petersen at the Health Research Laboratory, LASL. H. Whipple, M.D., Industrial Medical Group, LASL, directed the physical examination portion of the study, including clinical laboratory measurements. J. Healy, LASL Health Division, gave constructive criticisms in his review of the manuscript.

Many other persons too numerous to mention have contributed importantly to the work. A special acknowledgment is extended to J. Langham for photographic and autoradiographic work; H. Ide for

assistance in the *in vivo* chest measurements; P. Flynn, M.D., for medical examinations; O. Johnson for assistance in administrative arrangements; and D. Meyer for his help in recalling exposure conditions and health safety procedures in the early days.

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APPENDIX A

DECONTAMINATION PROCEDURES AND RESPIRATORS USED AT LASL IN 1944

The following is an excerpt from a Secret letter written by L. H. Hempelmann to Dr. Robert S. Stone, Clinton Laboratories, Knoxville, Tennessee, dated July 20, 1944, which describes decontamination procedures and respirators:

"1. Decontamination Procedures:

A. Floors: A special corp of janitors assigned to these laboratories spend all their time on this job. They work under the supervision of a head janitor and a "laboratorian." In order to get good results and to keep the floor counts low, it is necessary for the floors to be covered with a smooth hard surface of wax. There does not seem to be much difference in the kinds of wax that we used so we have settled on the water-soluble floor wax supplied by the janitor service. Our present floors are made up of some asphalt tiles known by the trade name of "Mastic." In some new laboratories we plan to use a smooth asphalt surface with an MgO base.

1. Routine Cleansing: All floors are mopped once a day; floors of hot laboratories are mopped twice a day or oftener. They are wet mopped with a good lather of Ivory soap, followed closely with a dry (wrung out) mop. It is essential that the janitors be meticulously careful about covering the entire floor. The results are better if the wash and rinsing water be kept separate. By this method it has been possible to keep the floor counts in most labs below 100. In our "hottest" laboratories, the counts are always below 500. We are now using some detergents in an effort to obtain better results.

2. Decontamination after Spillage: This is done by a trained decontamination squad. The floor is first mopped with Ivory soap. The wax is then removed with boiling hot water, followed by kerosene and steel wool. The floor is then painted with two coats of any thick paint which will stick to the surface.

B. Desk Tops of Laboratory Benches: Every working surface is covered with a smooth surface preferably glass. Porous surfaces are covered with enamel paint before they are used for work. If they are contaminated by accident, an effort is made to seal them in the material by painting rather than to remove the contaminated part of the desk top. In the case of the lab benches made of "Kemrock" (the ordinary black surface made by the Kewaunne Mfg. Co., Adrian, Mich.) the surface responds to ordinary methods of decontamination although it appears to be porous. The same company puts out a surface paint of the same material, called "Kemrock Wipe Coat," which can be used to paint over contaminated surfaces.

1. Routine Cleaning: Smooth surfaces can be quite successfully decontaminated by wiping with a clean cloth wet with spindle oil. An excess of oil is used and this is followed by a clean dry cloth. The secret of the successful use of this method is to change cloths very frequently. The surface count is usually reduced below 50 per minute by this method.

2. Decontamination after Spillage (or after contamination from droplets produced by evaporation): First wipe the surface with 1 N HCl solution followed by dry rag, then clean with oiled rag as above.

C. Hands: Before starting work, spread thin coating of spindle oil over both hands; this must be done very carefully. Decontaminate gloves before removing them; remove gloves using surgical technique so as not to touch outside surface with hands. Apply coat of West's Sulfo soap to hands, moisten with water and lather, rinse; repeat with Sulfo soap and finally wash with Ivory soap.

The success of this method depends upon how well directions are followed particularly insofar as spindle oil is reapplied after each washing of the hands. It has been our experience that it is usually possible to reduce the hand counts to a few hundred or less by means of this method.

D. Gloves and Respirators: A 10 percent solution of the detergent "aerosol" is used for decontamination of rubber articles. Other detergents do not seem to work as well. The solution containing acetic acid formerly used by us has been discontinued because the acid tended to injure the rubber and was hard on the skin if not properly removed. The gloves are washed every day (after one wearing). They are rubbed and swished around in the solution until clean. This is quite time consuming as about two minutes is required for each glove (we hope to cut this down by getting each person to decontaminate his own gloves after wearing). The rubber respirator facepieces are cleaned once a week if their count is over 50.

E. Glassware: This is cleaned by running each piece of glassware through the following solutions:

1. water
2. Ivory soap water followed by rinsing
3. cleaning solution followed by rinsing
4. Ivory soap and water followed by rinsing
5. distilled water

Popham says that the results seem to be satisfactory but he will not guarantee them. Not very much work has been done on this problem.

F. Tools: Can be decontaminated with an oiled rag as described above. Use clean cloths and clean frequently.

2. Masks: We have used only Wilson respirators and positive pressure masks. Most of the respirators put out by this company have filter pads which are satisfactory for this type of dust. Their chief disadvantage is in their failure to fit properly but we have found that one of the following three types will fit just about any type of face: 200L, 5L and 750. We are careful to fit each mask to the person before letting them go into use. As far as the positive pressure masks are concerned we have not tried the A.M.S. type because they did not appear as satisfactory as the Wilson type as described in their catalogues. We have modified the masks by putting on softer tubing onto the facepiece since there were some complaints about the stiff tubing supplied by the company and by placing the filters on the wall rather than on the belts. The tubes are fastened to the belts to avoid the masks being jerked off the faces of the men if the tubing gets caught. We have still not been able to put the positive pressure masks in general usage because of the crowded conditions in our laboratories.

The Eastman masks which I described to you over the telephone appear to be the most satisfactory of all, but have not been well tested. If you see them and consider having some made up we would be very interested in placing an order at the same time."

Declassified by L. M. Redman (September 11, 1972)

APPENDIX B

URINE ASSAY METHOD AND ESTIMATION OF BODY BURDEN

URINE SAMPLE COLLECTION

Beginning in January 1944, spot urine samples were collected from plutonium workers in the hope that measurement of plutonium in the urine might yield results which could be related to exposure to and/or intake of plutonium. By March 1944, a procedure had been adopted whereby urine samples were collected on a 24-hour basis in so-called "clean" areas, but the data derived from analysis of urine samples suggested occasional massive artifactual contamination. To minimize the possibility of such contamination, a Health Pass Ward was established in the Los Alamos Hospital in the spring of 1945. Each employee heavily exposed to plutonium was granted 2 days off with full pay* prior to reporting to the hospital, where he stayed for 24 hours. During this time, an uncontaminated or minimally contaminated 24-hour urine sample was collected using all practical procedures to minimize contamination. Upon entering the Health Pass Ward, the men showered and changed into hospital pajamas, which were worn throughout the collection period. White mortician's gloves were used while collecting the urine specimens.**

Table I shows the rather striking differences recorded for urinary excretion of plutonium in 6 subjects who collected urine samples both at home and in the hospital. The data are shown as originally presented by Wright Langham at a plutonium conference in May 1945 and documented in a report in June 1945.[1] Unfortunately, this problem of sample contamination during collection has been forgotten periodically (e.g., operational exposures and accidents such as Palomares and Thule), and this has led to misinterpretation of data.

TABLE I

EFFECT OF METHOD OF COLLECTING SAMPLE ON COUNTS FOUND IN THE URINE

Subject	Place of Collection of Sample (cpm) At Home[a]	In Hospital[b]
6	10.1	2.2
3	41.6	4.3
4	16.1	3.4
c	2.8	0.1
c	17.8	-
5	30.6	2.2
Average	20.0	2.2

[a] Samples collected at home were 2 overnight voidings collected by the individual after thorough bathing and washing of hands.

[b] Samples collected in hospital were 24-hour samples collected under the rigorous hospital plan after a 2-day leave from the "Hill."

[c] Not in UPPU follow-up.

Also of interest is another table (Table II) from the same report,[1] which gives some information on the then current average urine blanks of 0.5 count per minute (per 24 hours) and recovery values for the urine assay procedure. In the early days, data were often presented as counts per minute. The efficiency of the proportional counter used at the time is uncertain but probably approached 50 percent.

In an effort to minimize costs, the procedure was modified in 1947 to eliminate the 2-day health pass period, but the employee still reported to the

* Employees, particularly those in military service, were forever grateful to Langham for the 2-day leave away from the "Hill," as the mesa was called.

** Langham's favorite story had to do with the enormous quantities of urine passed by many of the plutonium workers while in the hospital ward. The subjects, usually G. I.'s, were able to obtain beer from the nearby PX by some means and drank this in substantial quantities. Fortunately, plutonium output is independent of quantity of urine. According to Langham, the champion was a man nicknamed Piss Porter (not his real name).

TABLE II

RECOVERY OF KNOWN AMOUNTS OF PLUTONIUM FROM REGULAR AND MOCK URINE SAMPLES

Number of Determinations	Nature of Samples	Amount of Spike (cpm)	Recovery (%)	Spread (%)
24	Blanks (regular urine)	0.0	(average 0.5 cpm)	(0-1.2 cpm)
4	Mock urine solution	29.2	94	88-100
11	Mock urine solution	10.0	93	85-101
12	Regular urine	10.0	88	73-104
3	Regular urine	4.5	95	81-105

Health Pass Ward for sample collection. In a further effort to reduce costs, the Health Pass Ward was abolished in 1951, and employees were asked to collect at home 2 morning and 2 evening voidings. These samples were pooled and considered the "equivalent" 24-hour urine sample. A reusable metal kit was furnished the employee; each kit contained three 1-pint bottles, which were discarded after use. The metal kits proved eventually to be unsanitary, and the concept of collecting 4 voidings in three bottles proved unacceptable. Therefore, a disposable cardboard kit containing four 1-pint bottles was introduced in January 1958; a statistical review of the results obtained by analysis of samples collected by this means suggests that a "true" 24-hour sample is not necessarily collected. The pooled samples represent the urine voided over a period of 22 $\pm$ 4 hours when collected correctly. This collection procedure is used today.

Since 1969, occasional nonroutine samples have been collected in special containers with time marks on the lids in an effort to learn more about the constancy of plutonium urinary excretion. Information gained from analysis of such timed samples has made it possible to apply corrections by volume and specific gravity so that results expressed in disintegrations per minute per sample can be realistically corrected to disintegrations per minute per day.

RADIOCHEMICAL SEPARATION METHOD OF URINARY ASSAY FOR PLUTONIUM

To the best of our knowledge, as of January 1944, urine samples were ashed and the alpha activity of the ash was measured directly.[2] However, we know that in 1945 urine samples were ashed and the plutonium separated by co-extraction with an iron-cupferride complex. Existing data from that time suggest that plutonium recovery was 82 $\pm$ 19 percent associated with blanks of 0.69 $\pm$ 0.53 disintegration per minute. By 1949, the serial bismuth phosphate-lanthanum fluoride co-precipitation technique was in use, affording recoveries of 67 $\pm$ 20 percent with blanks of 0.15 $\pm$ 0.1 disintegration per minute. In 1955, an alkaline earth phosphate precipitation was used directly on urine samples to concentrate plutonium, eliminating the need to evaporate large volumes of urine. This change in procedure did not alter the measured recoveries or blanks.

The desire for greater sensitivity resulted in adoption in 1957 of the Hanford procedure. In this method, plutonium is separated by TTA extraction, and the final radiometric measurement is made by NTA (nuclear track alpha) film counting. The Hanford procedure afforded plutonium recoveries of 70 $\pm$ 17 percent with blanks of 0.007 $\pm$ 0.005 disintegration per minute. A somewhat simpler radiochemical separation procedure utilizing anion exchange was substituted in 1963. Introduction of electronic equipment capable of pulse-height discrimination made it possible in 1967 to include a step utilizing ^{236}Pu as a tracer or yield-determiner. In June 1972, ^{242}Pu was substituted for ^{236}Pu to reduce the background in the ^{238}Pu and $^{239-240}Pu$ regions. The procedure, in use today, affords an overall recovery of 80 $\pm$ 20 percent as determined by internal spiking with ^{236}Pu (or ^{242}Pu).

RADIOMETRIC MEASUREMENT

The measuring device used in 1944 was a gas flow proportional alpha counter of unknown efficiency

with a background of approximately 30 counts per minute. Simpson gas flow proportional counters, acquired in May 1945, were operated originally with an efficiency of 48 to 50 percent and a background of about 1 count per minute. Modifications of these counters, chiefly lining the counting chamber itself with electrolytic copper foil, reduced the background to 0.15 $\pm$ 0.1 count per minute. These counters, in continuous use until 1957, were supplemented in 1952 by four NMC gas flow proportional counters operating at 48 to 49 percent efficiency with backgrounds of about 0.1 count per minute. The NTA procedure, adopted in January 1957, involved the counting of microscopic tracks instead of electronic detection. This gave a background of 0.005 count per minute with an efficiency of 50 percent. The overall procedure had a sensitivity limit (50 percent confidence level) of about 0.05 disintegration per minute per sample. From April 1966 to January 1967, zinc sulfide scintillation counters were used; these devices were about 45 percent efficient with a background of 0.01 $\pm$ 0.006 count per minute. Acquisition of alpha spectrometric equipment in 1966 led to measurement of all samples by alpha spectrometry after January 1967; the procedure yields an efficiency of 27 percent and a background of 0.004 $\pm$ 0.003 count per minute. In use presently is an 8-detector system with electronic data processing capability, which allows one to measure simultaneously isotopes of ^{236}Pu, ^{238}Pu, $^{239-240}Pu$, and ^{242}Pu.

In addition to techniques such as alpha spectrometry and proportional counting, plutonium can be measured also in the urine by a track etch procedure following neutron irradiation. In the latter system, neutrons are used to produce fission fragments, usually in a thick plastic material supporting the plutonium-containing sample. The damaged areas of the plastic are then etched and can be quantitated by use of optical means or spark counters.[3] Although not in wide use at present, this system is said to offer a considerable improvement over others in terms of sensitivity.

ESTIMATES OF BODY BURDEN FROM BIOASSAY TECHNIQUES

Urine assay data are used as the basis for determining the body burden of internally deposited plutonium. In more recent times, computer programs have been written to relate urine assay data to the body content at a given time after contamination or at the time of contamination. However, whether one resorts to the convenience of computerized data reduction and analysis or to hand calculations as practiced in the early days, the procedures are based, at least in part, upon the systemic model of urinary elimination developed by Langham and colleagues.[4] This model was derived from data obtained from studies of human subjects of short life expectancy. These persons were given plutonium citrate by intravenous injection in 1945.[5] Langham used these data to derive a simple power function equation to relate urinary excretion rate to body content. The power function is often used to approximate a set of exponential functions characterized by both decreasing the amount and release rate from various compartments. More recent analyses of the data from the original patients and from industrial exposures suggest that either the power function or multiple exponential functions can be used to fit the observed excretion patterns.[6] Although most of the human data were obtained during the first 140 days after injection, some information is available for excretion periods up to 5 years. Use of power functions based on the latter data give the following specific expressions for rates of urinary and urinary plus fecal excretion in man given plutonium citrate over a period of 5 years:

$$ER_u = 0.002\ t^{-0.74} \qquad \text{(Eq. 1)}$$

$$ER_{u+f} = 0.008\ t^{-0.94}, \qquad \text{(Eq. 2)}$$

where ER_u and ER_{u+f} represent the *fraction* of the injected dose excreted per day and t the days following exposure.

Following a single exposure at a known time, one can estimate the body burden (Q) at time of exposure from measurement of a 24-hour urine specimen collected at a later time (t). Using Eq. 1 and the relation

$$ER_u = U/Q, \qquad \text{(Eq. 3)}$$

where U is the amount of plutonium measured in a 24-hour urine sample at time t, then

$$Q = 500\ Ut^{0.74}. \qquad \text{(Eq. 4)}$$

Substitution of known values for t and U allows one to estimate total-body burden at time of exposure in the same units that are used for U (i.e., count rate, mass, or activity). Similarly, the body burden (Q_R) at any time t following repeated exposure may be calculated by using the total elimination coefficient and assuming multiple inputs as a summation process.

A major difficulty in evaluating human exposures has been the problem of reservoirs of plutonium in various organs (not bone) that are slowly released to the circulatory system at rates that depend on many factors such as location within the body, particle size, and physiochemical form. This slowly translocated plutonium is subsequently deposited in other tissues and ultimately lost from the body via urinary and fecal excretion.

The concept of slow, continuous release of bound plutonium into the body fluids, first formalized by Healy,[7] has been built into models by several groups of investigators.[8,9] The rate of urinary or fecal excretion of plutonium in persons chronically exposed can be estimated from Eqs. 1 and 2 by summation of individual administrations. Healy's model regards relatively insoluble plutonium in the lung as a reservoir isolated from normal body metabolism yet continually releasing plutonium into the bloodstream. The model has no constraints regarding the position of plutonium in any portion of the lung or body; therefore, particles translocated from the lung to lymph nodes behave in the same manner as those in the lung, provided the rate of solution and entry into the systemic circulation is the same. The model assumes a constant fractional removal per unit time from the lung and then utilizes the systemic model developed by Langham.[10]

The quantity (Q) in the lung at time t following acute deposition after initial clearance of an amount (Q_0) of insoluble plutonium is:

$$Q = Q_0\ e^{-\lambda t}. \qquad \text{(Eq. 5)}$$

The overall elimination rate (λ) actually represents both solubilization and transfer to the systemic circulation and discharge from the lung by ciliary clearance mechanisms. One can then describe transfer to the systemic circulation as:

$$a = \lambda_s\ Q_0\ e^{-\lambda t}. \qquad \text{(Eq. 6)}$$

Assuming that each increment transferred is excreted according to the function shown in Eq. 1, the total excretion may be described as the sum of the excretion rates from each increment. Time (t) is relative to elapsed time since transfer from the lung rather than time since inhalation. Using R as the time urine samples are obtained after inhalation, the excretion rate (E_u) is:

$$E_u = 0.002\ \lambda_s\ Q_0 \int_0^R e^{-\lambda t}\ (R - t)^{-0.74}\ dt. \qquad \text{(Eq. 7)}$$

Unfortunately, Eq. 7 is not integrable and must be solved for individual values of λ, R, and t by expansion of the exponential term and solving until the series converges. The overall transfer rate (λ) was thought to be composed of two components: (1) rate of transfer into the systemic circulation (λ_s), and (2) rate of loss via ciliary clearance mechanisms (λ). Healy assumed that λ_s was the same as λ. Similar expressions were derived for fecal excretion and for the amount of plutonium in blood.

Figure 1 shows the calculated relation between

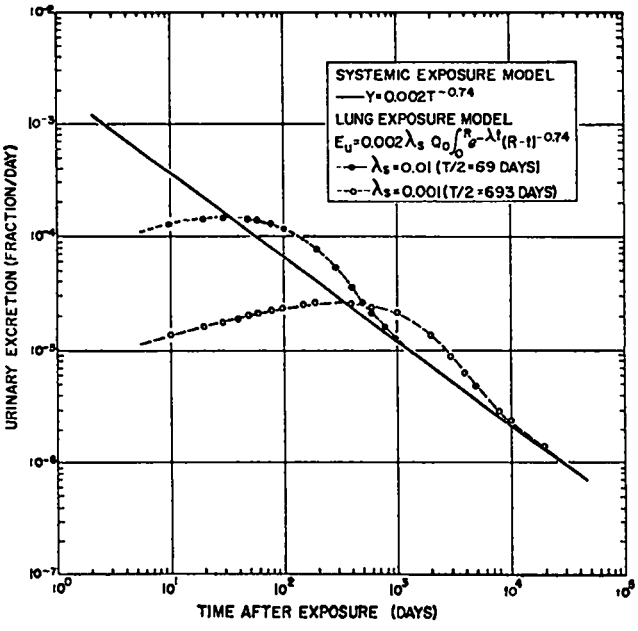

Fig. 1. Fractional urinary excretion as a function of time after acute exposure based on the systemic and lung exposure models.

predicted urinary excretion rate and time, assuming various rates of exponential clearance from a non-systemic reservoir.[11] The solid line represents the systemic exposure model (no transfer from the lung) which is the basis of the PUQFUA computational model.[12,13] Even for slow lung clearance times (e.g., 693-day half times), the lung model and the systemic model both yield similar urinary excretion values at 10^4 days, which corresponds to about 27 years following exposure. It is important to appreciate that these are idealized curves which are used as the bases for estimating plutonium body burdens. In real life, except for persons with industrial exposures who sometimes can be followed closely during employment, urine samples can be obtained only at relatively infrequent times. This is also true in the case of some UPPU Club members who, although exposed in 1944 or 1945, provided relatively few urine samples during the intervening 27 years. To date, the number of urine samples used to estimate body burdens for the subjects of this study have ranged from 5 to 125 for different individuals.

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APPENDIX C

MEDICAL FOLLOW-UP OF PATIENT NO. 2

Beginning in the spring of 1942, patient No. 2 was exposed to uranium halides while attempting to reduce uranium tetrafluoride to the metallic state. Patient No. 2, one of several nonmilitary members of the UPPU group, worked at Los Alamos from February 3, 1944, to August 16, 1944. He subsequently worked at other installations where he was not exposed to radioactive materials. However, late in 1946, he was exposed to beryllium oxide and beryllium chloride. In 1948, he was engaged in hot-pressing uranium powder with hardening agents. In 1952, he worked with catalytic agents but had no significant exposure to toxic chemicals. In 1954, he was exposed to sodium vapors as well as compounds of chlorine, arsenic, and antimony. Very often these materials were weighed in the open in crucibles that were evacuated and then fired. The crucibles were opened without the benefit of hoods or dryboxes. In 1960, he worked with cadmium selenite in a vacuum system but probably was not exposed to toxic materials. Since 1965, he has been involved in the manufacture of electronic instruments used to detect hydrocarbons.

During the medical examination in 1966, a small, coin-sized lesion was noted in the right lower lung. A previous chest roentgenogram made in 1960 had been normal. The follow-up examination in 1969 confirmed the lung abnormality which showed no detectable change in size. In a roentgenogram taken in March 1971, the lesion had nearly doubled in size and then had an outside diameter of about 5 cm. Following consultation between attending physicians, the individual concerned, and staff of the AEC's Division of Biology and Medicine, the patient was admitted on May 4, 1971, to a large hospital in the eastern United States for further diagnostic studies. During the following several days, he was subjected to a series of diagnostic tests including pulmonary function, sputum cytology, tantalum insufflation, bronchoscopy, and lung tomography. Cytological examinations based on sputum, bronchoscopic washings, and brush specimens showed no metaplasia in cells exfoliated from the bronchial tree. There were some indications of an inflammatory process which could be compatible with an underlying bronchial growth. Tomographic tests verified the previous findings of a well demarcated, simple lesion located in the right lower lung. Fiber-optic bronchoscopy indicated no abnormal changes in the lung within small bronchioles (to about 3 mm). Cytological study of specimens obtained by pinch biopsy was also negative. Results of the tantalum insufflation radiography indicated no intrusion on the bronchial tree or deposition defects.

Because of the demonstrable growth of the lesion since 1965, the attending physicians decided to perform a simple lobectomy, and on May 17, 1971, the right lower lobe was removed. Histological sections of the tumor and regional lymph nodes were made, and samples of the tumor, normal lung, regional lymph nodes, and bone were sent to several laboratories for radioactive assay and radiographic studies. Histologically, the neoplasm was a classical benign hamartoma (derived from residual embryonic cells) containing considerable adult cartilaginous and epithelial cells. Interestingly, the regional lymph nodes were perfectly normal histologically with no signs of cellular damage despite the relatively high radioactivity.

Patient No. 2 is currently in good health and returned to Los Alamos with other members of the UPPU group for additional study in November 1971. We believe it is worthwhile to point out that there is concern and interest for these subjects even though many years have passed since they worked with

plutonium. Perhaps patient No. 2 is a good example, as approximately 28 years have elapsed since his 6-month involvement with plutonium during the days of the Manhattan Project. This kind of information, even though very limited, is human experience of the most relevant kind for establishing value judgments where inadequate data exist for formulating risk evaluations.

APPENDIX D

RADIATION DETECTION EQUIPMENT FOR *IN VIVO* MEASUREMENT OF PLUTONIUM

The three following types of radiation detecting equipment are currently used to estimate the quantity of plutonium in the body: (1) proportional counters; (2) thin sodium iodide crystals and dual sodium iodide; and (3) cesium iodide crystals. Although these instruments are often called "lung" counters, they cannot distinguish between radioactive materials in the pulmonary and lymphatic tissue. In an effort to localize plutonium, particularly in the hilar lymph nodes, an intraesophogeal probe is being developed.[1] All of these instruments measure low-energy x-rays or gamma rays from plutonium or contaminant radionuclides such as ^{241}Am formed by decay of ^{241}Pu. The radiation energies of greatest interest are the 17- to 20-Kev uranium L x-rays from ^{241}Pu and the 60-Kev gamma ray from ^{241}Am. Low-energy x-rays also emitted by ^{241}Am must be differentiated from those given off by plutonium.

The system used at the Los Alamos Scientific Laboratory to measure *in vivo* deposits of plutonium in our subjects is comprised of two combined crystals, NaI backed by a 50-mm CsI crystal, each of which is about 125 mm in diameter and 3 mm in thickness. Each detector functions as an anticoincidence system, resulting in a suppression of background caused by Compton scatter from higher-energy radiations originating from within the body and in the local environment. The MDA at the 95 percent confidence level for this system averages about 7 nCi ^{239}Pu for a 30-minute counting period. Similar MDA values have been obtained at other facilities (for example, at the Lawrence Livermore Laboratory). To achieve MDA's in the range of roughly one-third the commonly accepted 16-nCi occupational lung burden, one must carefully estimate an individual's chest thickness. Ultrasonic techniques are used for this purpose at LASL. Measurements must be made also in "low-background" counting chambers constructed of massive iron shielding.

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