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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.

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

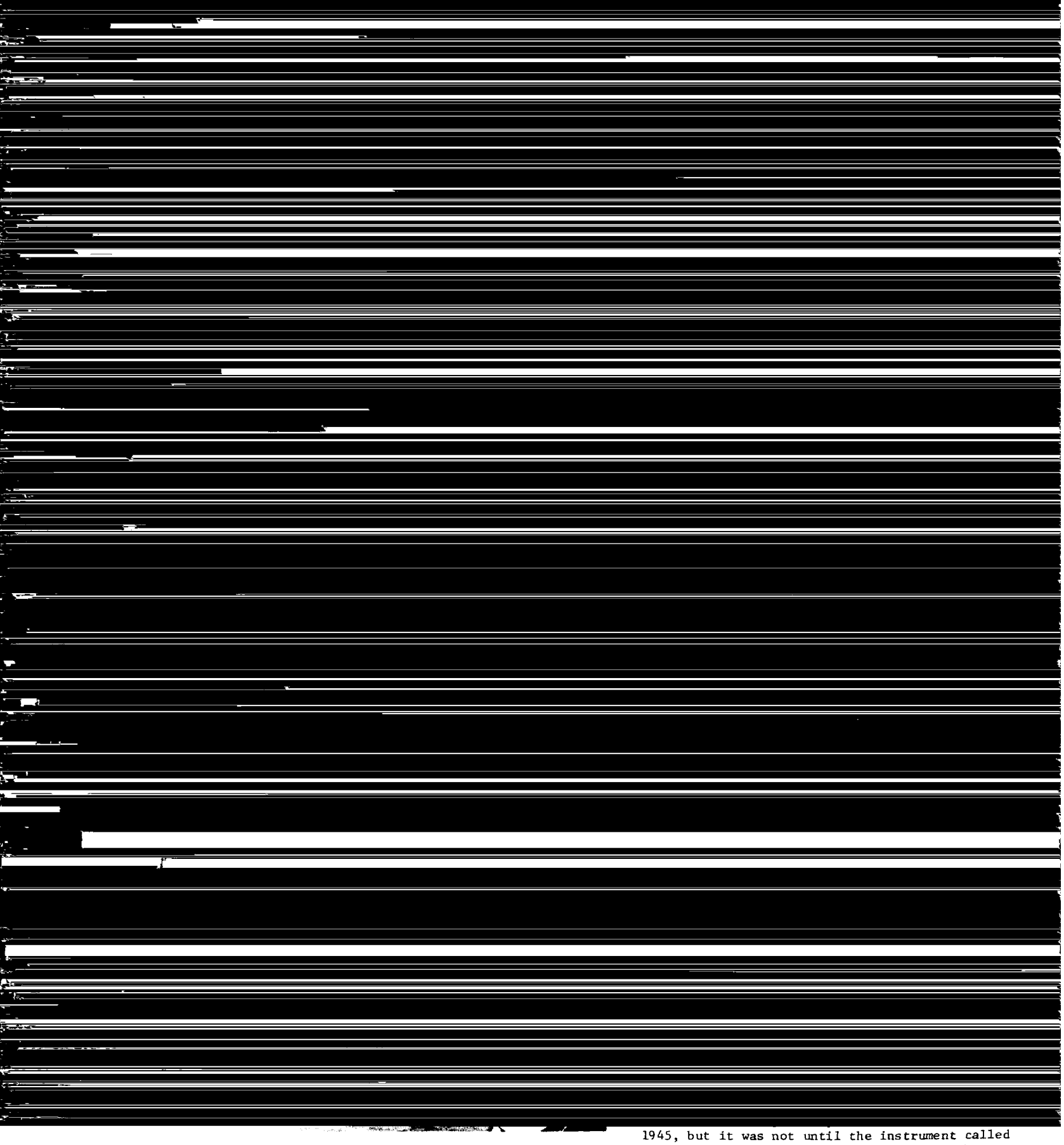

Fig. 1. The original wooden D Building which housed the chemists and metallurgists in CMR Division. Note that the chemical hoods of the laboratories on the first floor were vented individually, usually without filtration.

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.

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.

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).

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

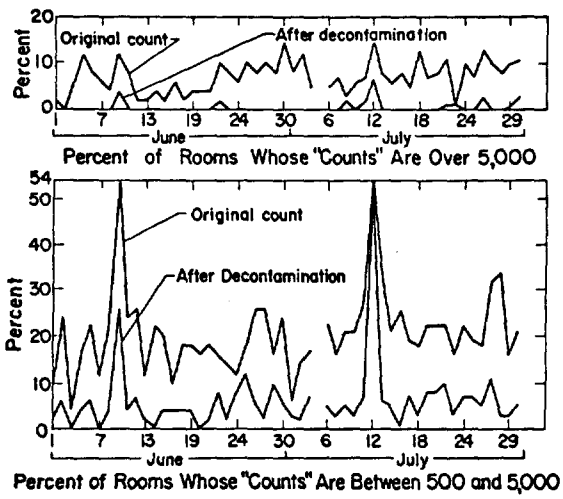

Fig. 4. Chart showing the percent of laboratories in D Building in June and July 1945 with "hot" spots requiring decontamination. The lower line indicates the counts after decontamination. Note that about 50 percent of the laboratories had contamination in excess of 500 counts per minute on two occasions.

*****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								

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. [redacted] 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

[redacted]

****** 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.

[redacted] 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]
Subject No. 5	
4-17-44[b]	175
28	199
7-19-44	207
10-12-44	-144
11- 7-44	188
12-29-44	207
4-20-45	717-408
5- 8-45	174-113
15	73-27
16	196-172
18	421-41
19	122-107
21	138-129
22	62-22
29	101-67
31	250-87
6- 4-45	930-82
5	272-174
6	163-43
7	267-131
9	177-118
14	170-165
15	504-416
16	246-514
18	720-501
19	2559-3111
20	199-150
23	320-200
25	126-78
26	608-94
27	68-50
7- 2-45	217-182
4	336-316
5	394-182
	324-182[c]
6	218-122
7	486-152
9	128-108
10	92-56
11	520-394
	1096-329[c]
12	64-80
13	100-80
14	100-60
	148-130[c]
16	96-76
17	100-96
23	70-66
25	64-52
8-17-45	696-90
20	144-48
23	88-26
24	76-60

Date	Nose Counts[a]
Subject No. 8	
6- 2-45	88-26
4	216-100
5	231-213
6	20-60
7	291-201
9	286-89
11	1181-323
12	454-385
13	1107-1292
14	509-229
15	623-560
16	1208-680
18	146-92
19	928-615
20	1506-1009
22	1898-1100
23	90-23
7- 2-45	400-274
4	164-170
	236-97[c]
5	324-126
	198-118[c]
6	338-278
7	268-72
9	1016-694
10	484-304
11	250-247
12	198-166
14	356-328
16	364-328
17	462-410
18	752-545
	179-183[c]
19	941-243
23	898-816
24	322-160
25	166-154
26	768-652
8-13-45	124-56
20	202-100
27	300-136
9-12-45	106-78
14	165-107
17	136-72
5- 8-46	52-825
6-17-46	168-401

Date	Nose Counts[a]
Subject No. 9	
6-28-45	372-146
29	562-98
30	698-664
7- 2-45	70-64
3	208-140
6	112-80
9	1164-608
	608-626[c]
10	57-50
11	879-706
23	838-842
	1296-306[c]
25	164-408
30	3040-464
31	2086-482
8- 2-45	8976-5180
3	660-204
4	674-530
6	3922-1752

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

[b] Only one nostril swabbed.

[c] Accidental exposure suspected.

[redacted]

ment 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

[redacted]

tion, 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 in tigation by CM-12 has failed to show appreciab contamination. The air pressure is being incr Better respirators must be obtained quickly. Improvements in the handling of product are be made, but it will be some time before the haza be appreciably reduced except by the use of be protective devices. CM-1 has helped with a nu of improvements such as installation of a new studies on improving the ventilation, installa of foot-pedal sinks, etc. The situation is st critical, however."

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

B. Purification Operation

Three of our subjects worked in the Puri tion Group in the Chemistry and Metallurgy Re Division. This group was given the responsib of purifying by wet chemical methods the plut produced by the piles in Oak Ridge and later Hanford. Because it was believed that nonmet impurities could best be removed by subsequen chemistry procedures (see next section), this concentrated on removing contaminating metals

The flow sheet used in the purification tion[3] until July 1945 is shown in Fig. 6. Pl nium nitrate slurry received in 160-gram lots metal containers from Hanford was dissolved, diluted (cut), assayed, and transferred (some with considerable difficulty) into a closed s On the average, these preliminary steps took 3.3 days. The following procedures were then ried out: (a) reducing Pu^{IV} (or Pu^{VI}) nitrat iodide; (b) precipitating Pu^{III} oxalate and d solving as Pu^{VI} with bromate and nitric acid; (c) precipitating plutonium as sodium plutony acetate and dissolving in HNO_3; (d) ether ext ing plutonyl nitrate from the acid solution c ing ammonium nitrate; and (e) reducing pluton nitrate with iodide and precipitating as the late. This product, recovered in about 97 pe yield, was given to the Fluorination Group. rification of plutonium in this form continue

*********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.

[redacted]

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

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

**********According to the most recent analysis by J. Lawrence, this man does have a measurable body burden of plutonium. He is not but should be a member of the UPPU Club.

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

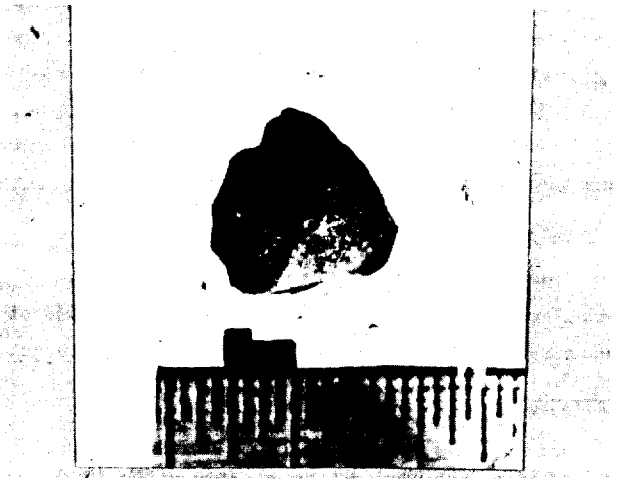

Fig. 7. A 500-mg metallic button of almost pure plutonium.

war. All loading and unloading operations wer
ried out in dryboxes. Because the lid was bol
on, disassembly of the bomb was easier than in
case of the welded bomb. Moreover, the well fu
reaction products were less dusty than those in
earlier small-scale experiments. Nevertheless,
metallurgist who developed this method does hav
measurable body burden of 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

********** The contamination occurred during p
aration of the metallic button mentioned above.
Unbeknown to their supervisors, these workers b
the reduction at midnight. The next morning th
plutonium button in an appropriate container wa
allegedly found on the desk of Dr. Cyril Smith,
Associate Leader of CMR Division, and extensive
contamination had occurred throughout the labor
and adjacent hallways. The workers were nowher
around but were finally located in the bar of t
La Fonda Hotel in Santa Fe, where they were cel
brating their success in reducing plutonium by
technique they had developed.

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 tonium workers had measurable body burdens acco to the assay methods of the time, but 3 have be dropped from the series as a result of the use more reliable methodology, and one other died. 1953, a program for periodic examination of the men (financed by the U. S. Atomic Energy Commis was established. At first, a very thorough stu every 2 years was planned. This included inter history and physical examination, complete bloo count, blood calcium, phosphorus and alkaline p phatase, urinalysis, and stool examination for cult blood. The following roentgenograms were taken: lateral skull, PA chest, AP pelvis, AP knee and elbow, lateral of foot, AP of foot, an dental films of right teeth.

In 1953 and again in 1955, 22 and 25 subje

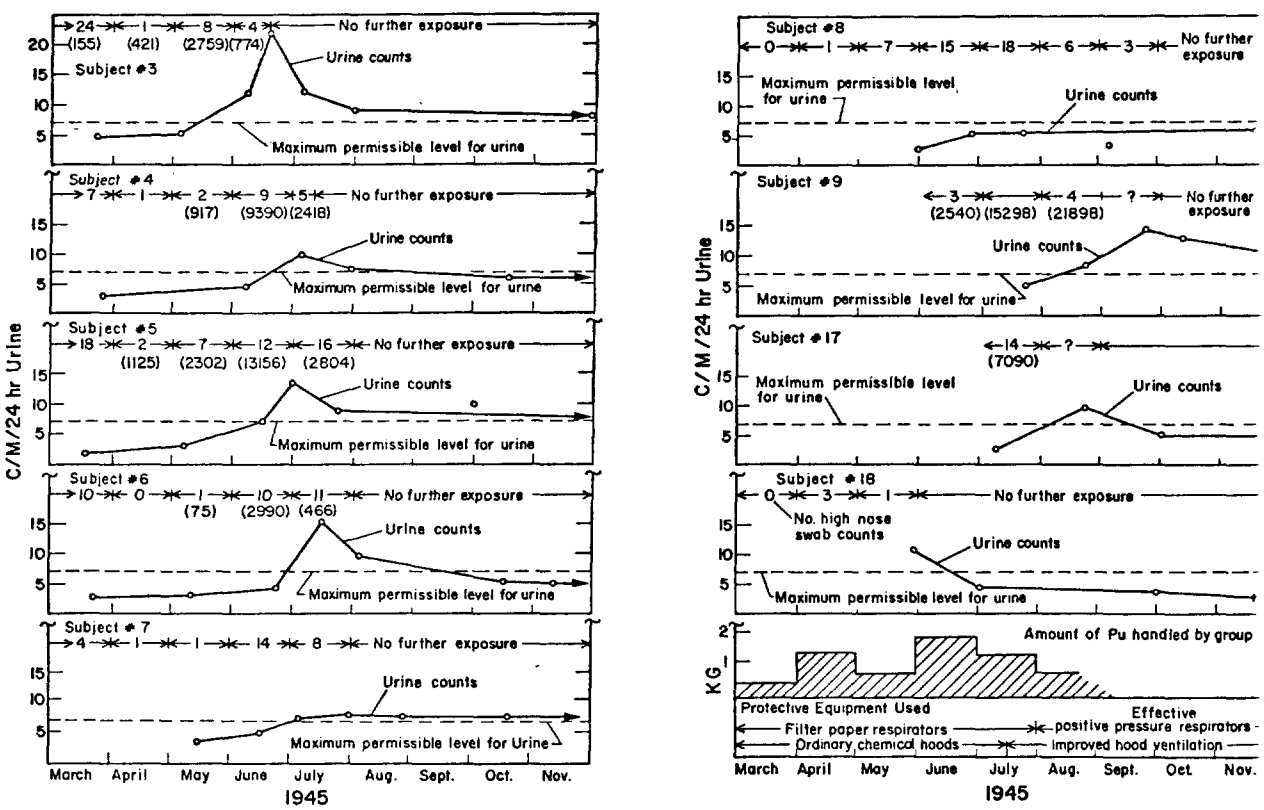

Fig. 8. Graphs for 9 subjects showing the urine count, number of high nose counts per month, and total radioactivity of nasal swipes per month (parenthetical figures) from March to November 1945. Th amount of plutonium handled by the Recovery Group and the protective measures used are shown in the last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

ments for 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.

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.

[redacted]

experiment. However, we do know that the tumor incidence decreases and the time required for tumors to develop increases as the amount of plutonium

[redacted]

7 nCi if one uses the 95 percent confidence level. For the 68 percent confidence level and a similar

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

1 nCi per gram), which cause premature death i beagles, are about 30 times the concentration alent to the maximum permissible occupational burden for man. This conclusion, of course, i based on the assumption that lung tumor develo is related to plutonium concentration and not t the total amount of plutonium in the lung. Fo example, if tumor induction is somehow related the total number of cells at risk (therefore, t number and size of plutonium particles), then relative sizes of human and dog lungs may be u important 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) c sidered to be the maximum occupational lung bu for man.[18]

The experimental data obtained from rodent are not as clear nor as encouraging. Moskalev[1] reports that the frequency of malignant neoplas of the lungs of rats exposed to plutonium by in halation is 2 or 3 times higher than in the con group at doses ranging from 41 to 234 rads. Mo alev states that, assuming the biological effec

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

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 contempo-

[redacted]

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.

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.

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

by chest-counting, to have 10 to 20 times the pe
missible level of plutonium in the lung about
3 years after an inhalation accident. Chromoson
analysis indicated minimal radiation damage to t
lymphocyte series even at a high level of exposu
of the patient. Other investigators are studyin
chromosomes obtained from uranium miners exposed
a variety of stressing agents, including radiati

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

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

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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. Ston Clinton Laboratories, Knoxville, Tennessee, dated July 20, 1944, which describes decontamination proced 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

TABLE I

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

Subject	Place of Collection of Sample (cp	
	At Home[a]	In Hospita
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 vo collected by the individual after thorough bat and washing of hands.

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

[c]Not in UPPU follow-up.

in June 1945.[1] Unfortunately, this problem of sample contamination during collection has beer forgotten periodically (e.g., operational expos and accidents such as Palomares and Thule), an has led to misinterpretation of data.

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

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

*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).

ted to the [redacted] flow proportional alpha counter of unknown efficiency

[redacted]

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

[redacted]

lowing exposure.

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

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

where U is the amount of plutonium measured in 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

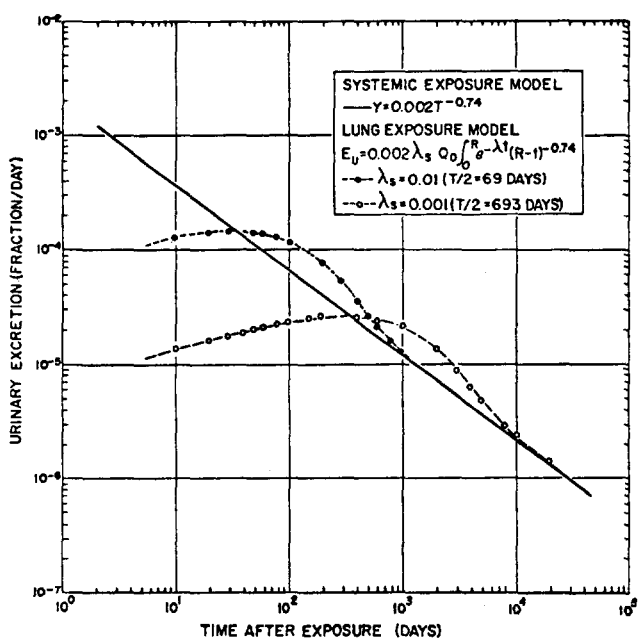

Fig. 1. Fractional urinary excretion as a function of time after acute exposure based on the systemic and lung exposure models.

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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diagnostic studies. During the following several days, he was subjected to a series of diagnostic

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 establi ing value judgments where inadequate data exist formulating risk evaluations.

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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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