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PLUTONIUM BIOASSAY RESULTS FOLLOWING DTPA TREATMENTS ON TWO CASES 7 YEARS AFTER ACCIDENTAL EXPOSURE

by

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ABSTRACT

Two employees were given a series of DTPA (diethylene-triaminepentaacetic acid) treatments 7 years after they were exposed to plutonium. Bioassay results show the amount of plutonium excreted during the course of treatment was 1.5% and 1.9% of the estimated body burdens of the two individuals.

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INTRODUCTION

Accidental plutonium exposures at this plant are treated with the chelating agent, DTPA (diethylenetriaminepentaacetic acid).[1] This treatment accelerates the excretion of plutonium in both the urine and the feces.[2] The degree of acceleration is dependent on the following:

- The chemical form and solubility of the plutonium
- The mode of entry into the body
- The promptness of treatment
- The method of administering the DTPA
- The amount of DTPA given per treatment and the frequency of treatments

This increased excretion is desirable in the treatment of the accident victim but it complicates the interpretation of the excretion data. At the present time, the plutonium excreted in the urine is used as an indication of the amount deposited in the body tissues and bones. A calculation of body burden is made using the Langham equation.[3] This equation relates urine excretion to body burden as a function of time. This equation, however, is based on the data obtained from an experiment which did not involve the use of chelating agents. Some data is available for plutonium excretion following DTPA treatments,[3-10] but more is needed before body burden calculations can be made on individuals receiving treatments.

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It is the purpose of this paper to add to the available data on excretion rates of plutonium following DTPA treatments.

TREATMENT

In 1964, DTPA treatments were given to two employees who had been accidentally exposed to plutonium in 1957. They received general body contamination, lacerations, and inhalation exposures. Both were given EDTA (ethylenediaminetetraacetic acid) treatments shortly after the accident. Their EDTA treatment schedule and bioassay results have been previously reported. (11)

In the recent series of treatments, employee A was given one DTPA treatment per week for a period of 12 weeks. Each treatment consisted of one gram of $CaNa_3$ DTPA in 250 cc. of normal saline solution administered intravenously by slow drip.

Employee B was given three treatments per week on a 3-weeks-on then 3-weeks-off schedule. This continued for 15 weeks with a total of 24 treatments given.

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BIOASSAY

Both employees have submitted 24-hour urine samples each month since the accident. Just prior to the recent DTPA treatments, employee A was excreting plutonium in the urine at the rate of 4 d/m/24 hours. Employee B was excreting 11 d/m/24 hours. These monthly assays have been used to estimate body burdens for each of the employees. These estimations indicate 3.5 and 8.6 times the MPBB (Maximum Permissible Body Burden) for Case A and Case B, respectively.

Nearly continuous urine sampling was conducted during the 1964 treatment on both individuals. Fecal samples were collected but not on a daily schedule. Blood samples were also assayed for plutonium. Prior to each DTPA treatment, a 30-ml sample of blood was taken for analysis.

For Case A, the urine, fecal, and blood assay results are shown in Tables I, II, and III, respectively. This information is also shown on semilogrithmic paper in Figure 1. The bioassay results for Case B are shown in Tables IV, V, VI, and Figure 2.

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DISCUSSION

During the course of treatment, nearly 13,000 d/m of plutonium was excreted by Case B compared to 1300 d/m in a similar period without treatment. This 10-fold increase represents, however, only 1.5% of the estimated body burden of the individual. Case A excreted 5000 d/m during his course of treatment. This represented a 15-fold increase but only 1.9% of his estimated body burden.

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TABLE I

AMOUNT OF PLUTONIUM EXCRETED IN URINE (CASE A)

Date (1964)	d/m-24 hrs.	Running Total (d/m)	Date (1964)	d/m-24 hrs.	Running Total (d/m)
8-12	88	88	9-27	71	2899
8-13	90	178	9-28	43	2942
8-14	49	227	9-29	273	3215
8-15	61	288	9-30	97	3312
8-16	50	338	10-1	101	3413
8-17	52	390	10-2	67	3480
8-18	57	447	10-3	63	3543
8-19	16	463	10-4	45	3588
8-20	7	470	10-5	16	3604
8-21	8	478	10-6	154	3758
8-22	5	483	10-7	100	3858
8-23	25	508	10-8	Lost	3858
8-24	39	547	10-9	25	3883
8-25	13	560	10-10	44	3927
8-26	1	561	10-11	29	3956
8-27	36	597	10-12	3	3959
8-28	67	664	10-13	Lost	3959
8-29	72	736	10-14	17	3976
8-30	52	788	10-15	113	4089
8-31	48	836	10-16	58	4147
9-1	232	1068	10-19	33	4180
9-2	100	1168	10-20	234	4414
9-4	108	1276	10-21	4	4418
9-8	86	1362	10-22	61	4479
9-9	65	1427	10-23	30	4509
9-10	37	1464	10-24	38	4547
9-11	51	1515	10-25	18	4565
9-12	38	1553	10-26	45	4610
9-14	34	1587	10-27	Lost	4610
9-15	126	1713	10-28	52	4662
9-16	70	1783	10-29	62	4724
9-17	90	1873	10-30	58	4782
9-18	99	1972	10-31	59	4841
9-21	59	2031	11-1	30	4871
9-22	376	2407	11-3	75	4946
9-23	138	2545	11-4	65	5011
9-24	96	2641	11-5	57	5068
9-25	99	2740	11-6	43	5111
9-26	88	2828			

TABLE II

AMOUNT OF PLUTONIUM EXCRETED IN FECES (CASE A)

Date (1964)	d/m-24 hrs.
8-9	0.74
8-27	20.
9-22	2.3
10-28	17.5

TABLE III

AMOUNT OF PLUTONIUM IN BLOOD ASSUMING A 6-LITER VOLUME (CASE A)

Date (1964)	d/m
8-11	1600
8-18	72
8-25	87
9-1	56
9-8	336
9-15	162
9-22	1440
9-29	186
10-6	Lost
10-13	1420
10-20	532
10-27	1140

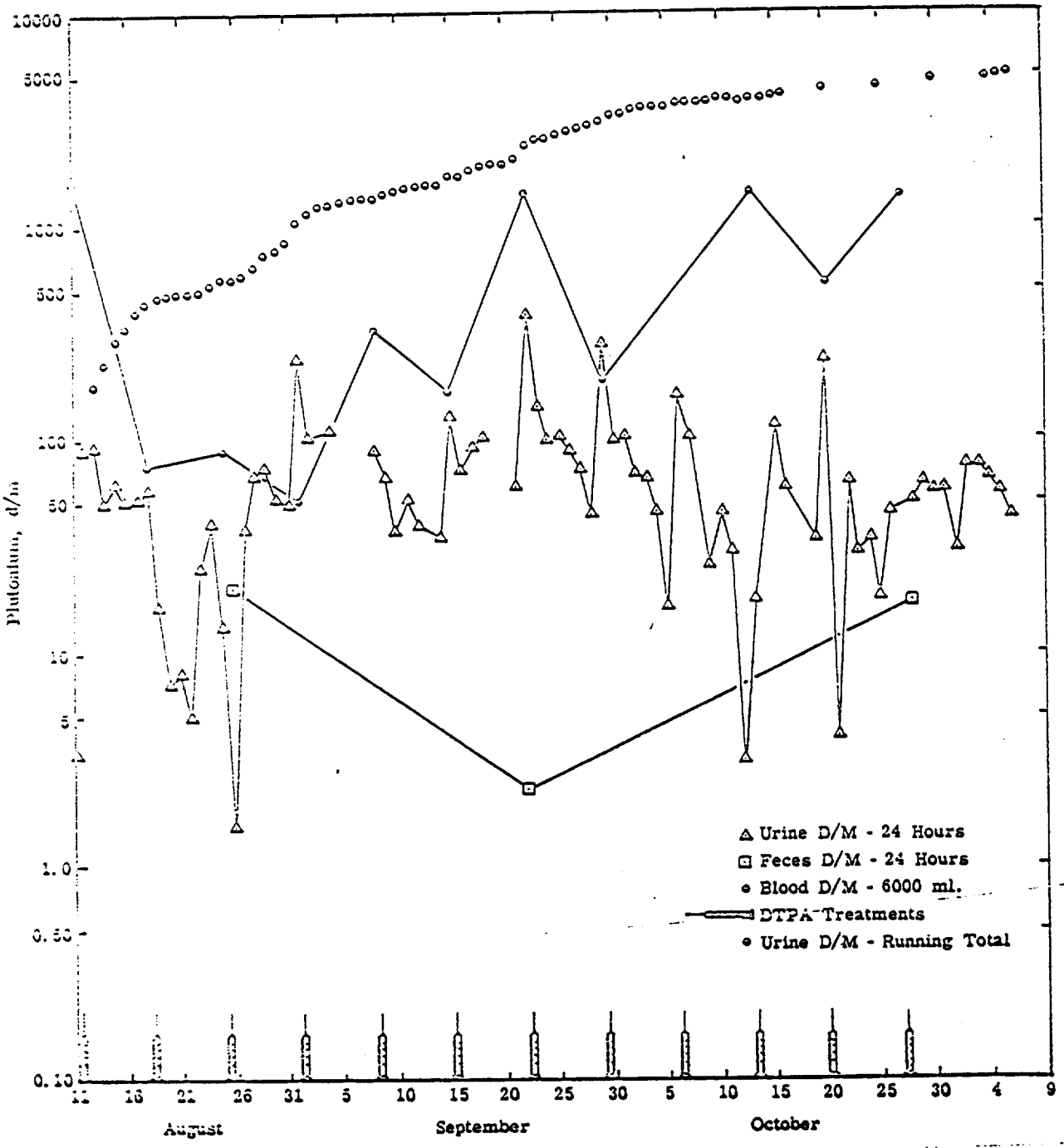

FIGURE 1

BIOASSAY RESULTS (CASE A)

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TABLE IV

AMOUNT OF PLUTONIUM EXCRETED IN URINE (CASE B)

Date (1964)	d/m-24 hrs.	Running Total (d/m)	Date (1964)	d/m-24 hrs.	Running Total (d/m)
7-7	568*	568	8-31	274*	9,256
7-8	578	1,146	9-1	169	9,425
7-9	508	1,654	9-2	357	9,782
7-10	675	2,329	9-3	226	10,008
7-11	333*	2,662	9-17	50	10,058
7-12	157	2,819	9-24	51*	10,109
7-13	305*	3,124	9-25	19*	10,128
7-14	509	3,633	9-28	286*	10,414
7-15	532	4,165	9-29	139	10,553
7-16	384	4,549	9-30	79	10,632
7-17	411	4,960	10-1	48	10,680
7-18	244	5,204	10-2	108	10,788
7-19	88	5,292	10-4	82*	10,870
7-20	450	5,742	10-5	495*	11,365
7-21	288	6,030	10-6	254*	11,619
7-22	420	6,450	10-7	254*	11,873
7-30	68*	6,518	10-8	215	12,088
8-6	50*	6,568	10-9	153	12,241
8-17	282*	6,850	10-12	150*	12,391
8-18	48*	6,898	10-13	87*	12,478
8-19	147	7,045	10-14	15	12,493
8-20	130	7,175	10-15	242*	12,735
8-21	246*	7,421	10-16	96	12,831
8-22	207*	7,628	10-17	130*	12,961
8-23	192*	7,820	10-18	95*	13,056
8-24	330*	8,150	10-19	65*	13,121
8-25	174	8,324	10-21	100*	13,221
8-26	285*	8,609	10-23	53*	13,274
8-27	179	8,788	10-27	38*	13,312
8-28	95	8,883	10-29	38*	13,350
8-29	99	8,982			

* extrapolated to 24 hrs from data obtained in more than 8 hrs.

TABLE V

AMOUNT OF PLUTONIUM EXCRETED IN FECES (CASE B)

Date (1964)	d/m-24 hrs.	Date (1964)	d/m-24 hrs.
7-8	15	8-26	16
7-9	70	8-28	128
7-11	22	9-2	82
7-12	9	9-3	8
7-13	20	9-17	3
7-14	22	9-25	--
7-15	38	9-29	--
7-16	101	9-30	1
7-17	41	10-1	--
7-19	104	10-2	--
7-20	65	10-6	47
7-22	23	10-8	Lost
7-30	21	10-12	2
8-6	124	10-14	8
8-17	6	10-21	19
8-18	9	10-23	14
8-19	24	10-29	9
8-24	2		

TABLE VI

AMOUNT OF PLUTONIUM IN BLOOD ASSUMING A 6-LITER VOLUME (CASE B)

Date (1964)	d/m	Date (1964)	d/m
7-7	168	8-24	156
7-8	2740	8-26	116
7-9	168	8-28	224
7-10	306	8-31	252
7-13	138	9-2	204
7-15	2016	9-4	120
7-17	720 & 138	9-17	300
7-20	96	9-28	660
7-22	284	9-30	426
7-29	972	10-2	792
8-17	822	10-7	182
8-19	96	10-27	1440
8-21	356	11-13	348

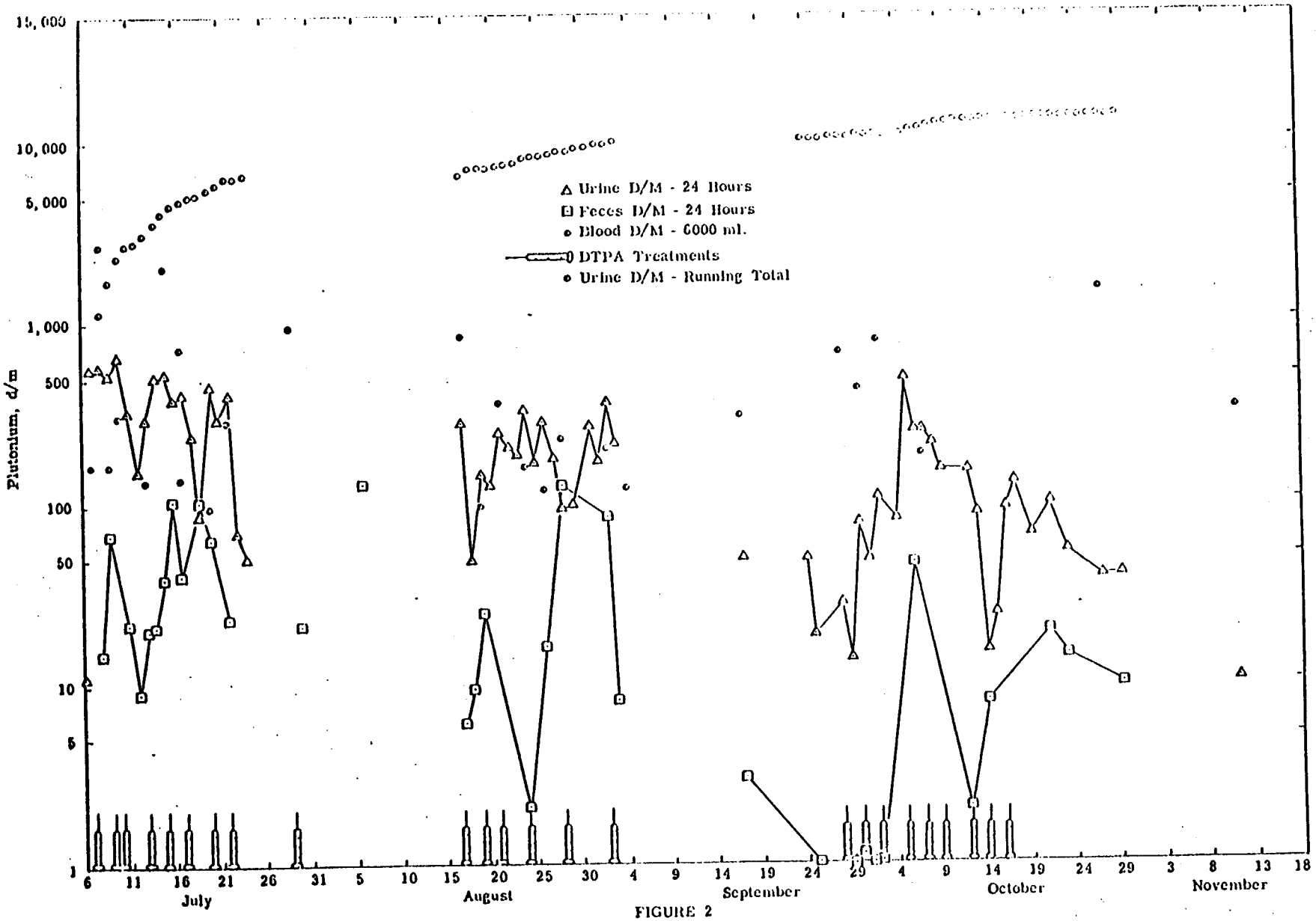

FIGURE 2

BIOASSAY RESULTS (CASE B)

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SM-56/4

EVALUATION AND TREATMENT OF AN ACUTE INTERNAL EXPOSURE TO PLUTONIUM

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1. Abstract

As a result of an explosion caused by burning plutonium which was accidentally dropped into carbon tetrachloride, an individual was exposed internally to large amounts of plutonium. The skin of his entire body was contaminated to milligram quantities. The tip of the second finger, left hand, was severed. The left thumb had a severe laceration on the anterior surface at the interphalangeal joint, a compound fracture, and dislocation. There were several small lacerations on other fingers and back of the left hand. Small pieces of metal were driven into the hand.

Treatment to reduce surface contamination consisted of shaving off hair, including eyebrows, and initial and daily washings of the body with sodium hypochlorite until contamination was reduced to acceptable levels.

Evaluations of deposition in the thumb were done by measurement of 17-kev x-rays. These evaluations led to the decision to amputate the thumb and finger.

The individual was given DTPA treatment within 1 hour after injury and was placed on a subsequent schedule of periodic treatments. Rapid evaluation of plutonium in urine and observations of the effectiveness of DTPA were accomplished by measurement of 17-kev x-rays from the raw urine. The limit of detection by this method is 14,200 d/m per 24-hr. urine sample. This limit can be extended by an evaporation and dissolution technique to 570 d/m per 24-hr. urine sample. Accumulated urine excretion for the first 60 days was 7.3 μc.

The individual was counted in the Rocky Flats Body Counter. Counts were made in the liver region, lung region, and left hand following amputation. Using a plastic human phantom for calibration, evaluations of lung and liver deposition were obtained. Evaluation of residual hand deposition was accomplished using a paraffin phantom for calibration and a positioning jig for the hand. Advantages of utilizing each of several available x- and gamma-ray energies were determined.

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2. Introduction

On June 12, 1964, at the Rocky Flats Division of The Dow Chemical Company, a male employee working in a glove box accidentally dropped about 1500 grams of burning plutonium into a degreasing bath. The burning plutonium reacted with the carbon tetrachloride bath to produce an explosion having a force approximately equivalent to about 1½ lbs of detonated nitroglycerine. Some of the windows of the glove box were shattered, and plutonium dust and chips were ejected into the room. The employee's gloved left hand, immediately over the bath, received multiple injuries. The tip of the second finger was severed, and the thumb was severely lacerated, fractured, and dislocated. The other fingers and the back of the hand received small lacerations. In addition, the employee was contaminated externally and exposed to high airborne concentrations of plutonium.

This paper will describe the treatment and recuperation of the employee with particular regard to his internal plutonium contamination.

3. Decontamination of the Skin

After the accident, the employee was transported to the medical center. He was conscious but dazed. The first evaluation of plutonium contamination on the exposed portions of the employee's body indicated greater than 1.5 uc/100 cm^2, the upper limit of the alpha detector. Lesser quantities were measured on other portions of his body. A plastic bag was placed over the injured hand and taped to his arm. He was placed on the decontamination table and washed with large amounts of water and dilute sodium hypochlorite. His hair was clipped short. No attempt at decontamination of the left hand was made at this time. At this point the main intent was to decontaminate the patient sufficiently so that DTPA (diethylenetriaminepentaacetic acid) could be administered. This was accomplished within one hour of the accident. Except for the left hand, skin contamination remaining at this time ranged from 0.1 to 1 uc/100 cm^2 about his head and shoulders with lesser amounts on other parts of his body.

Daily showers and local applications of dilute sodium hypochlorite gradually reduced the skin contamination. Reduction of skin contamination levels relative to time after the accident is shown in Table I. It was necessary to shave the patient's eyebrows to decontaminate that area.

4. Evaluation and Treatment of the Contaminated Hand

The left hand was too highly contaminated to measure surface contamination quantitatively. Small particles of plutonium appeared to have been driven into the palmar surface of the hand. Measurements of L x-rays using a scintillation detector indicated at least 40 μc of plutonium distributed throughout the thumb. These measurements were made with

(a) A probe with a ¾-inch diameter by 1 mm thick NaI crystal canned in 0.001 inch of aluminum, and

(b) A 1½ inch by 1 mm thick NaI crystal canned in 0.001 inch of aluminum.

Both detectors feed into the same analyzer and scaler system set to record pulses from photon energies between 10 and 20 kev.

At approximately 7½ hours post accident, a surgeon began repairing the hand. Under local anesthetic the hand was first scrubbed with dilute sodium hypochlorite. The collected washings assayed about 1 μc of plutonium. The second finger was debrided, amputated proximal to the wound and closed. The thumb (or first finger) was debrided, the fracture reduced and the wound closed. The decision to save the thumb, at least temporarily, was based on the following considerations:

(a) The injury to the thumb from the blast was not severe enough to warrant amputation.

(b) The amputation could be performed at a later date if it were deemed necessary, and the delay would not have harmful consequences. This judgement relied on the ability of DTPA to chelate the plutonium entering the blood, causing the plutonium to be excreted rather than deposited in body tissues and bones.

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TABLE I

Plutonium Contamination of Body and Arms (c/m)

Date	Face	Right Arm	Trunk	Legs
6-15-64	30,000-300,000	15,000-25,000	5000-10,000	1,000-10,000
6-16-64	4,000- 40,000	600- 6,000	Bkgd.-8,000	Bkgd.- 3,000
6-17-64	500- 2,500	250- 3,000	Bkgd.- 750	Bkgd.- 2,000
6-19-64	250- 2,000	Bkgd.- 1,000	Bkgd.- 500	Background
6-22-64	250- 1,500	Background	Background	Background
6-24-64	Bkgd.- 800	Background	Background	Background

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(c) There was much uncertainty in the measurement of the amount of plutonium in the thumb. It was not known, for example, what fraction of the measured plutonium was on the surface. It was also possible that there were small chips of metal present which could be removed easily at a later date.

(d) If the thumb were to be amputated at this time it would be difficult to prevent surface contamination from entering the surgical field.

Four days after initial surgery and repair, the left hand was carefully scrubbed with dilute sodium hypochlorite. Several microcuries remained on the surface of the thumb. Skin contamination on other parts of the hand was reduced to less than 6 nc per alpha detector area (67 cm^2). At this time x-ray detector measurements indicated at least 15 to 30 µc within the thumb. Later measurements of the 384-kev gamma ray from Pu^{239} indicated about 125 µc in the thumb. The latter measurement was accepted since absorption of these gammas within the thumb was negligible. X-ray measurements of the second finger showed about 0.35 µc remaining at the sutured tip. Another measurement revealed about 0.07 µc remaining in a laceration at the knuckle of the 3rd finger. On the following day, measurements of small black spots on the palm of the hand revealed two containing about 2 µc each and 3 containing about 0.2 µc each. The hand was washed daily to reduce surface contamination.

On the 12th day after the accident, following a review of the medical and radiological data, the injured first (thumb) and second fingers were amputated. For better functioning of the hand, it was decided to remove both fingers at the metacarpal phalangeal joints. The five previously-mentioned black spots and three others were excised at this time. The activity from these eight excisions totaled about 9.2 µc. It was assumed at the time that little or no plutonium remained above the metacarpal phalangeal joint of the thumb. Later measurements disproved this assumption.

Skin contamination on the left hand was reduced to insignificant amounts within 1 month post accident. Decontamination progress of the left hand is summarized in Table II.

5. Excretion and Blood Analyses and DTPA Treatment

After initial skin decontamination and approximately 1 hour post accident, the first DTPA was administered. One gram of DTPA in 250 cc of normal saline was given by slow intravenous drip. This treatment was repeated 3½ hours later. At 6½ hours post accident, the first urine sample was collected. An aliquot of the sample was evaporated and counted for alpha activity. The results extrapolated to the total sample showed that 0.46 μc had been excreted. A blood sample taken just prior to the first DTPA treatment assayed 6.7 d/m/ml. Just prior to the second treatment the blood level measured 16.0 d/m/ml. On the second day the blood level rose to 30.3 d/m/ml and on the third day decreased to 25.0 d/m/ml.

Fecal samples collected on the second and third days showed results of 0.64 μc and 5.5 μc, respectively. All excreta were collected for 45 days following the accident. Subsequently all urine and periodic fecal samples were collected. DTPA was administered according to the following schedule:

2 grams per day for 3 days
1 gram per day for 18 days
1 gram, 3 times per week, for 4 weeks
1 gram per week for 4 weeks
1 gram, 3 times per week, for 5 weeks
1/3 gram, 3 times per week, for 5 weeks

Blood, urine and fecal activity levels along with periods of DTPA treatment are displayed as Figure 1. Integrated urine excretion relative to time after the accident are displayed as Figure 2. In the interest of determining the effectiveness of different schedules of DTPA treatment, urine excretions were integrated on a weekly basis.

TABLE II

Plutonium Contamination of the Fingers - Left Hand (c/m)

Date	Thumb	Second	Third	Fourth	Fifth	Wrist
6-16-64	∞	300,000	200,000	100,000	40,000	500,000
6-17-64	650,000-∞	30,000	15,000	4,000	4,500	4,000
6-18-64	350,000-∞	15,000	7,000	3,000	4,000	4,000
7- 7-64	-----	-----	1,000	1,000	1,000	1,000
7-16-64	-----	-----	500	500	500	500
7-27-64	-----	-----	Bkgd.	Bkgd.	Bkgd.	Bkgd.

∞ above 10^6 α c/m limit of alpha survey meter

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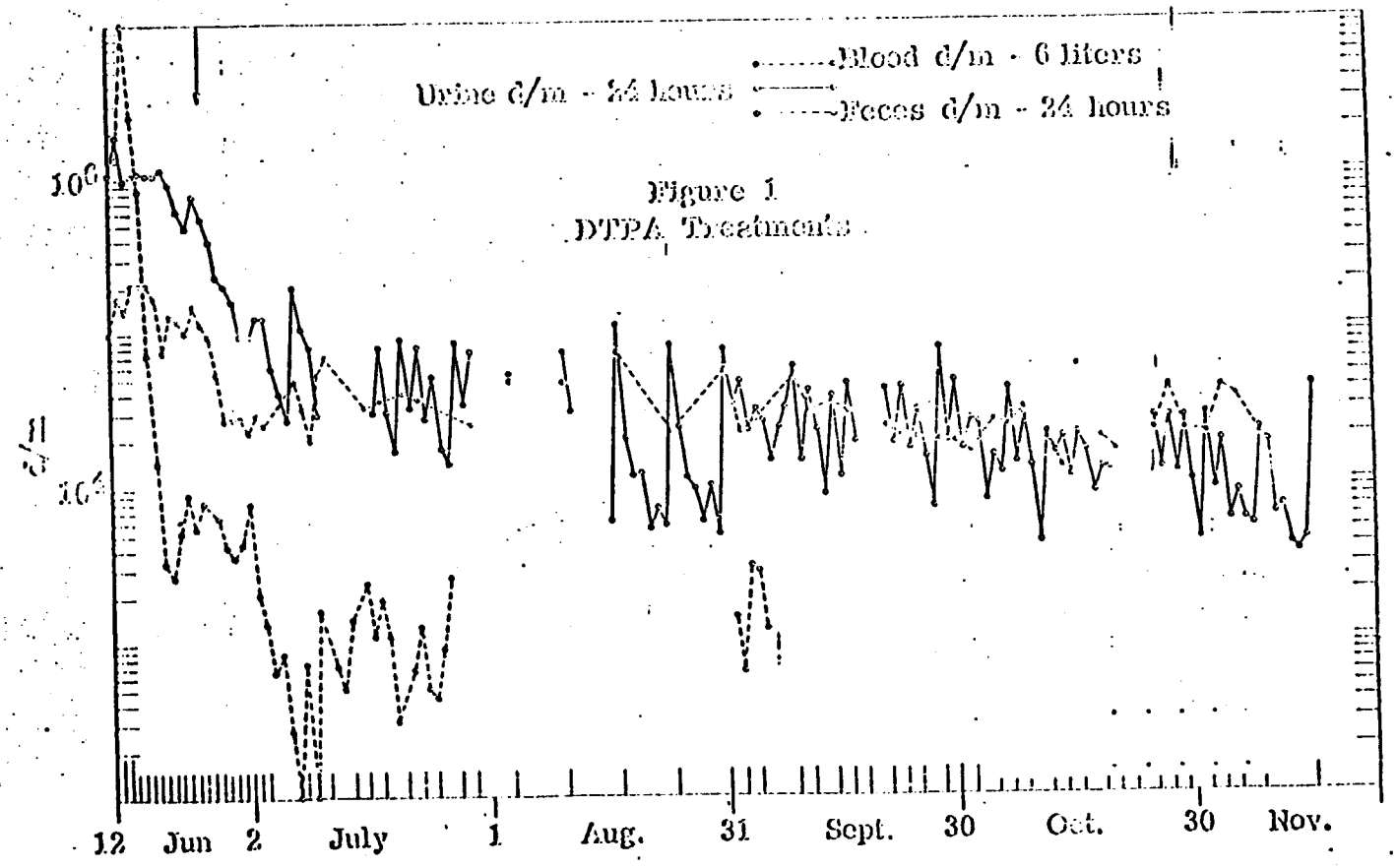

Figure 1
DTPA Treatments

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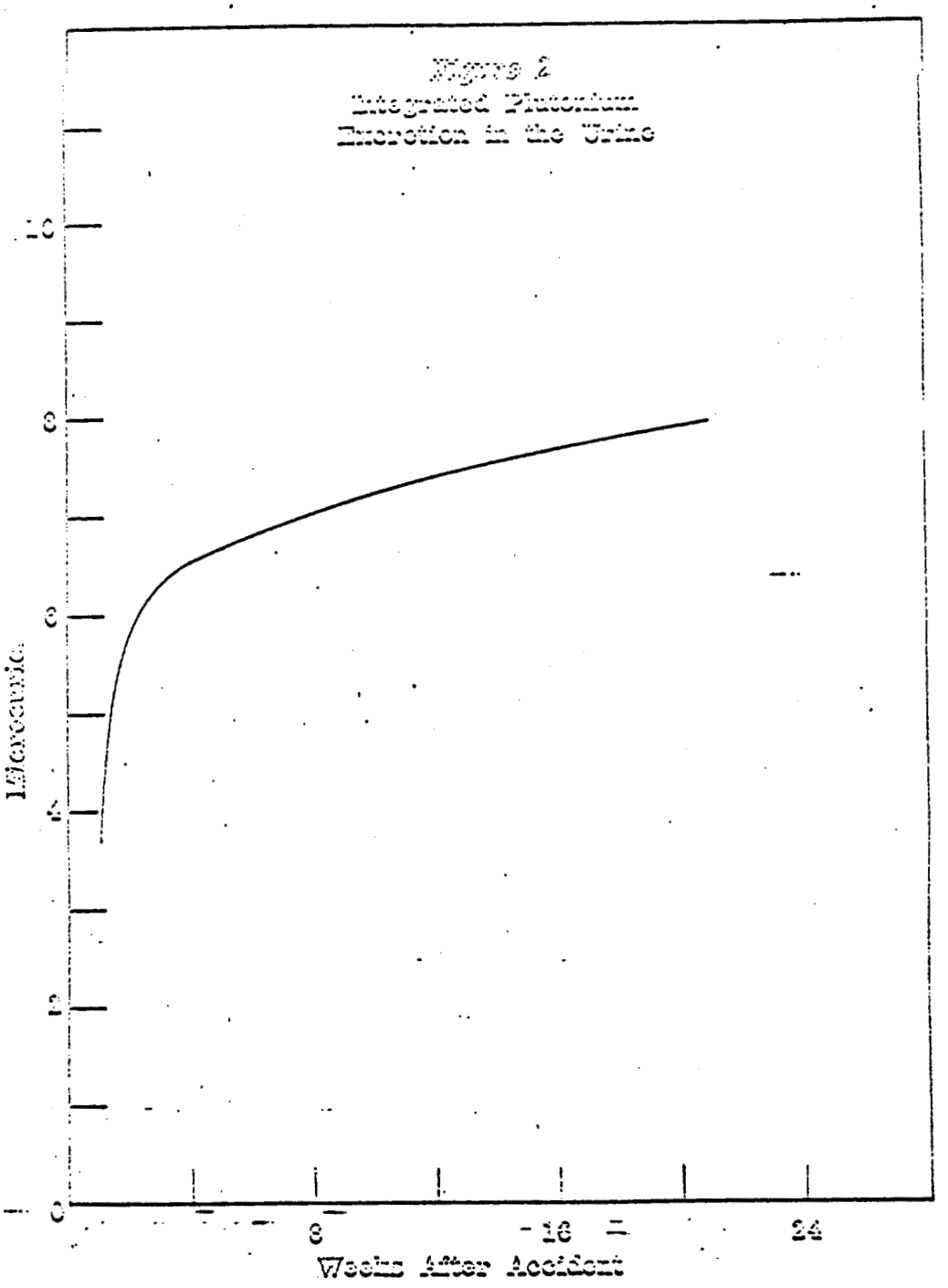
Figure 2
Integrated Plutonium
Excretion in the Urine
10
8
6
4
2
0
Micrograms
8
16
24
Weeks After Accident

Figure 3
Weekly Plutonium Excretion
in Urine as Affected by
DTPA Treatment Schedule
Microcuries
10
1.0
0.1
0.01
Daily 1 gram
3 Times Per
Week 1 gram
Weekly 1 gram
3 Times Per
Week 1 gram
3 Times Per
Week 1/3 gram
Weekly 1 gram
6
7
11
10
11
Weeks After Accident

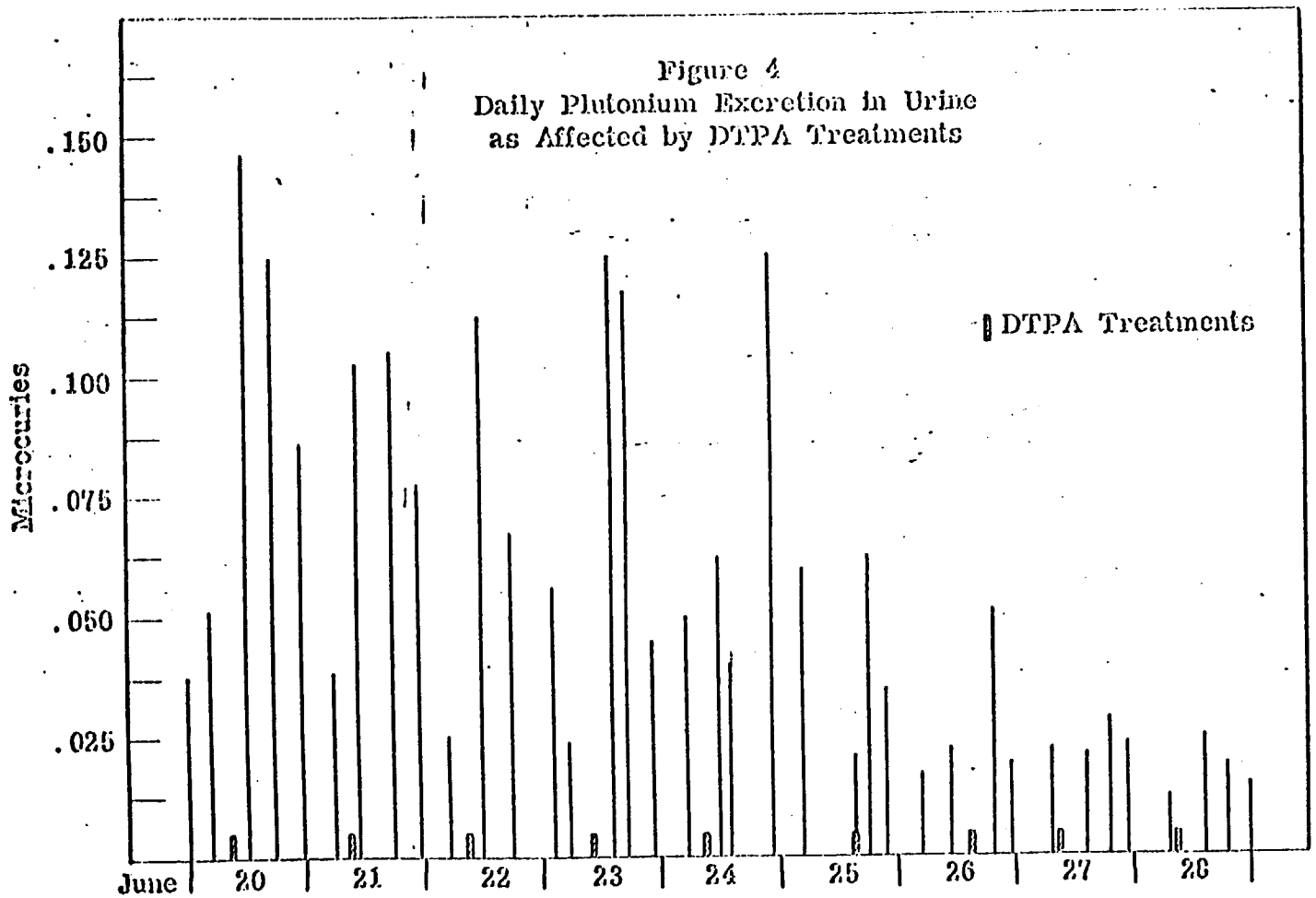
Figure 4
Daily Plutonium Excretion in Urine
as Affected by DTPA Treatments
DTPA Treatments
Microcuries
.150
.125
.100
.075
.050
.025
June
20
21
22
23
24
25
26
27
28

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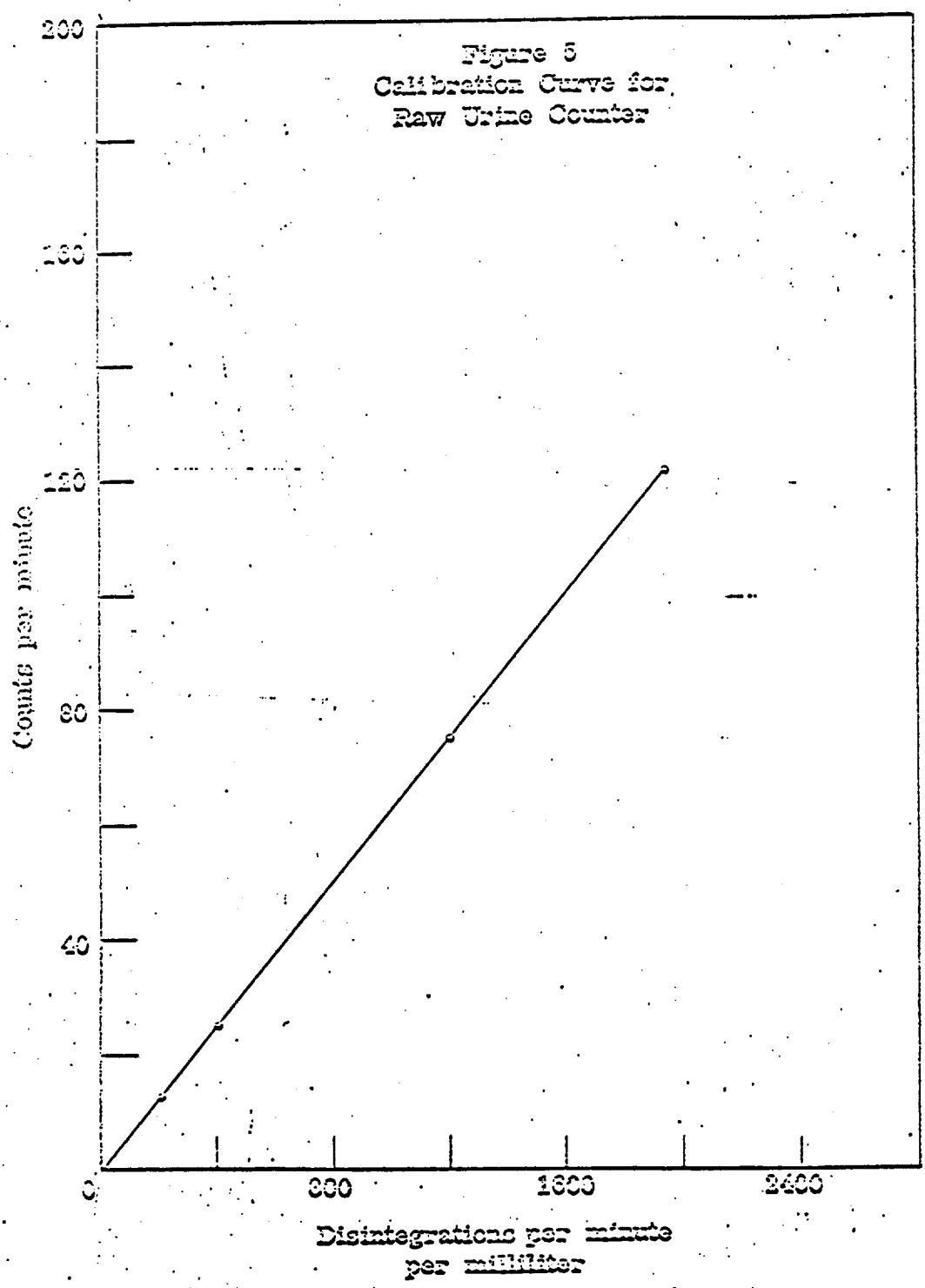
Figure 5
Calibration Curve for
Raw Urine Counter
200
160
120
80
40
0
Counts per minute
800
1600
2400
Disintegrations per minute
per milliliter

The corresponding drop in count rate made it impractical to maintain similar accuracy in a reasonable length of time. Thirty minutes was arbitrarily chosen as the maximum counting time. This 30-minute count provided better than ±20% error at the 95% confidence level for the next 4 weeks. After this time, however, the amount of plutonium in his urine had decreased to a level where it became necessary to improve the counting system.

Improvements were possible by increasing the counting time, obtaining a larger diameter crystal, or by concentrating the urine by evaporation. The concentration by evaporation method was selected for the following reasons:

(a) A factor of 10 increase in sensitivity could be easily obtained by this method.

(b) There was no longer a necessity for a rapid determination of the plutonium in the urine because the treatment was no longer determined on a day-to-day basis. Since all samples were being chemically analyzed for plutonium in the bioassay laboratory, this direct counting technique was now providing only a parallel study.

(c) The regular procedure for analyzing plutonium in the bioassay laboratory involved a step in which the urine was evaporated to dryness and then redissolved in 45 ml of 6 N HNO_3. The chemical procedure could be easily interrupted at this point for a count on this acid solution.

A 150-ml beaker was cut down to hold 45 ml. The crystal was covered with Saran Wrap* and the counting was done as before. This method provides an accuracy of about ±20% error at the 95% confidence level in 30 minutes from a urine sample which contains 1000 d/m or more of plutonium.

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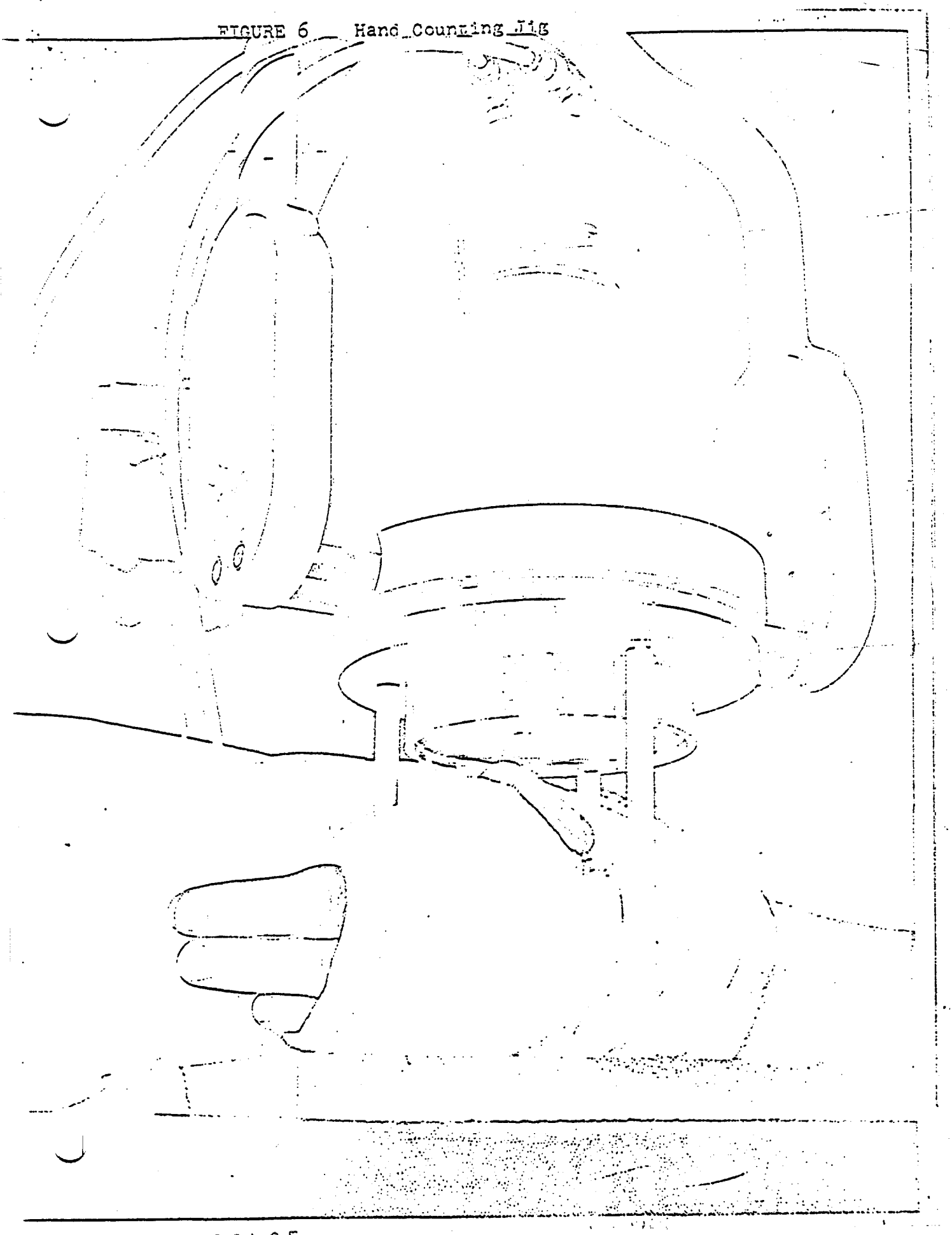
* Registered trademark of The Dow Chemical Company; Midland, Michigan

FIGURE 6 Hand Counting Jig



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Eventually, in order to minimize uncertainties because of tissue absorption, the 384-kev gamma ray from Pu^{239} was measured using the large-crystal detector. Figure 7 shows the quantity of plutonium remaining in the hand at various times after surgery. Most of the activity is located in the sutured region where the thumb was removed. The inconsistencies in the data collected up to the middle of August were probably caused by a fluctuating geometry in the hand-detector system. The initial swelling of the hand and its eventual return to normal may have influenced this. No known electronic malfunctions occurred during this period. The subsequent increase from August 17 to September 2 was due to a gradual change in the plutonium calibration. When this was discovered the system was recalibrated.

The body counter was calibrated with a human phantom, REMAB*, a plastic form containing a skeleton and cavities for the major organs. The liver cavity was first filled with a solution containing 0.06 μc of a known isotopic mixture including Pu^{239} and Am^{241}, the remainder of the phantom was filled with water, and the phantom was counted. Then the liver cavity was filled with 0.25 μc and counted again. Then the lung, filled with an epoxy lung equivalent material* containing 0.31 μc of the radioactive mixture, was counted. Finally, the liver region was counted with the lung cavity filled and then the lung region counted with the liver cavity filled.

Figure 8 shows a typical spectrum of the liver area of the phantom. The shape of the lung area spectrum is almost identical. The thin crystal detector proved directional under the conditions of this experiment; the spectrum in each case was unaltered by the presence or absence of the radioactive mixture in the other cavities.

The body counter was recalibrated several times with the human phantom. However, point sources of radioactive materials were used daily to check the counter for stability and sensitivity.

* Manufactured by Alderson Research Laboratories; Long Island City, New York.

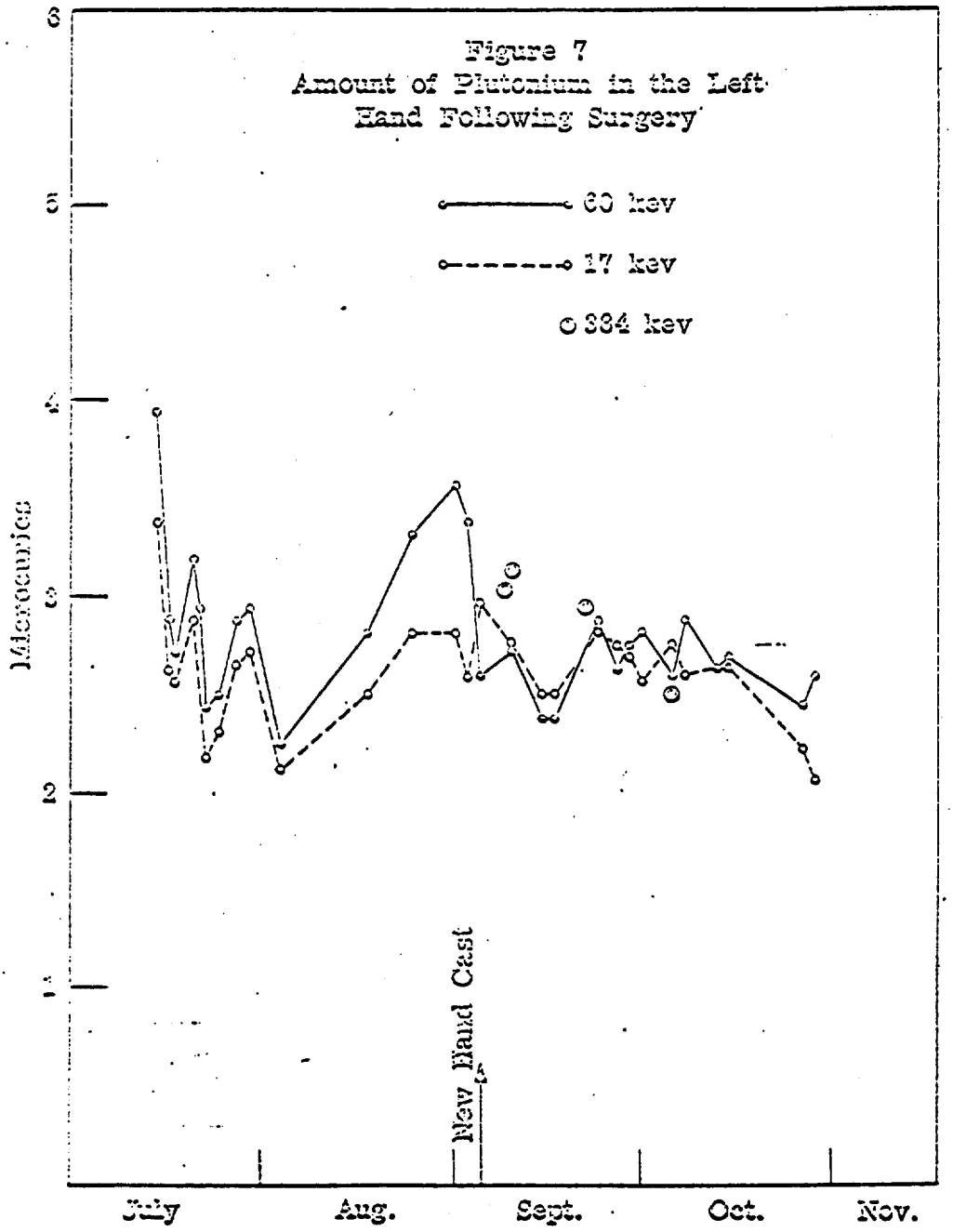
Figure 7
Amount of Plutonium in the Left Hand Following Surgery
60 kev
17 kev
384 kev
Microcuries
6
5
4
3
2
1
New Hand Cast
July
Aug.
Sept.
Oct.
Nov.

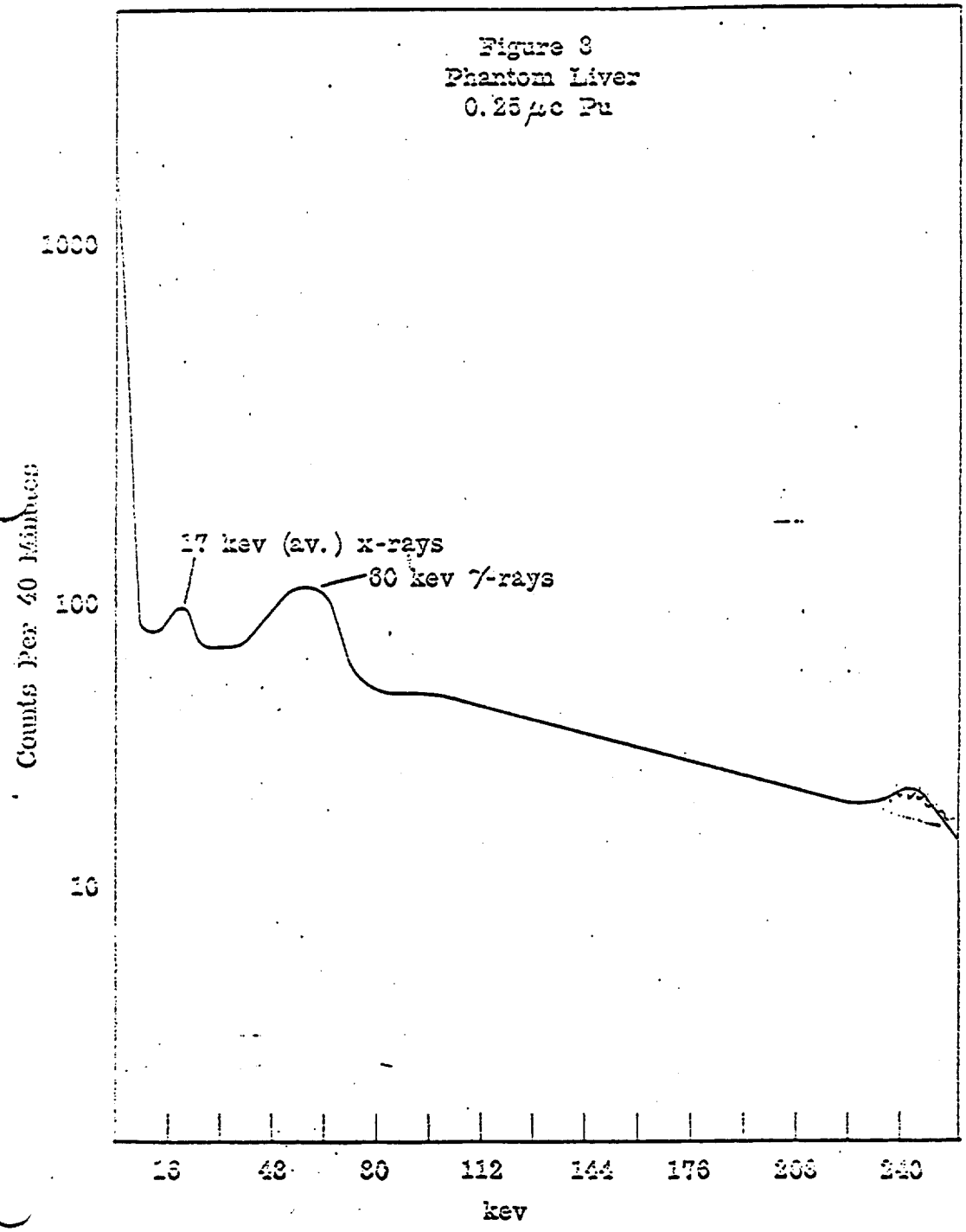
Figure 8
Phantom Liver
0.25 μc Pu
1000
100
10
Counts Per 40 Minutes
17 kev (av.) x-rays
80 kev γ-rays
16
48
80
112
144
176
208
240
kev

Figure 9 shows a spectrum with the thin-crystal detector positioned over the subject's liver. The prominent 60-kev peak was used for estimating the amount of radioactive material in the subject's liver and lungs. These estimates for various times after the accident are shown in Figures 10 and 11. No corrections were made to compensate for differences in body dimensions between the subject and the phantom.

7. DTPA Excretion Rate Studies

A decision was made to study the excretion rate of DTPA in order to help better establish a frequency for administrations. The method of Darbey [2] for the determination of ethylenediamine-tetraacetic acid was modified to a procedure for DTPA (diethylenetriaminepentaacetic acid) in urine. The principle of the method consists of complexing the DTPA with nickel and removing the excess nickel ion with dimethylglyoxime.

The Ni-DTPA complex is destroyed with strong acid and the nickel determined spectrophotometrically with dithioloxalic acid. The lower limit of detection in a 25-ml portion of urine is one milligram of DTPA. Figure 12 shows the correlation of DTPA excretion with plutonium excretion.

Most of the DTPA is excreted via the urine within 24-hours after administration. Table III shows the percent of DTPA excreted the first day after treatment during some of the treatment periods. Some variation in the rate of decrease after the first 24 hours has been observed. Table IV shows the percent of administered dose observed in the urine at various periods after administration. It will be noted that in some cases the DTPA is less than the detection level within 2 days and in another case observable up to 6 days post administration.

TABLE III

Amount of Administered DTPA
Excreted in First 24 hours

Date of Treatment	Percent DTPA
8- 3-64	92
8-17-64	76
9-11-64	96
9-14-64	81
9-16-64	68
9-21-64	115
9-23-64	127

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TABLE IV

Concentration of DTPA in Urine Following Treatment

Date of Administration	Time after Treatment (hrs.)	Percent of Initial Dose
8- 3-64	144	1.8
8-17-64	42	<0.1
9-11-64	53	<0.1
9-14-64	42	0.7
9-16-64	42	1.8
9-21-64	42	1.7
9-23-64	42	1.2

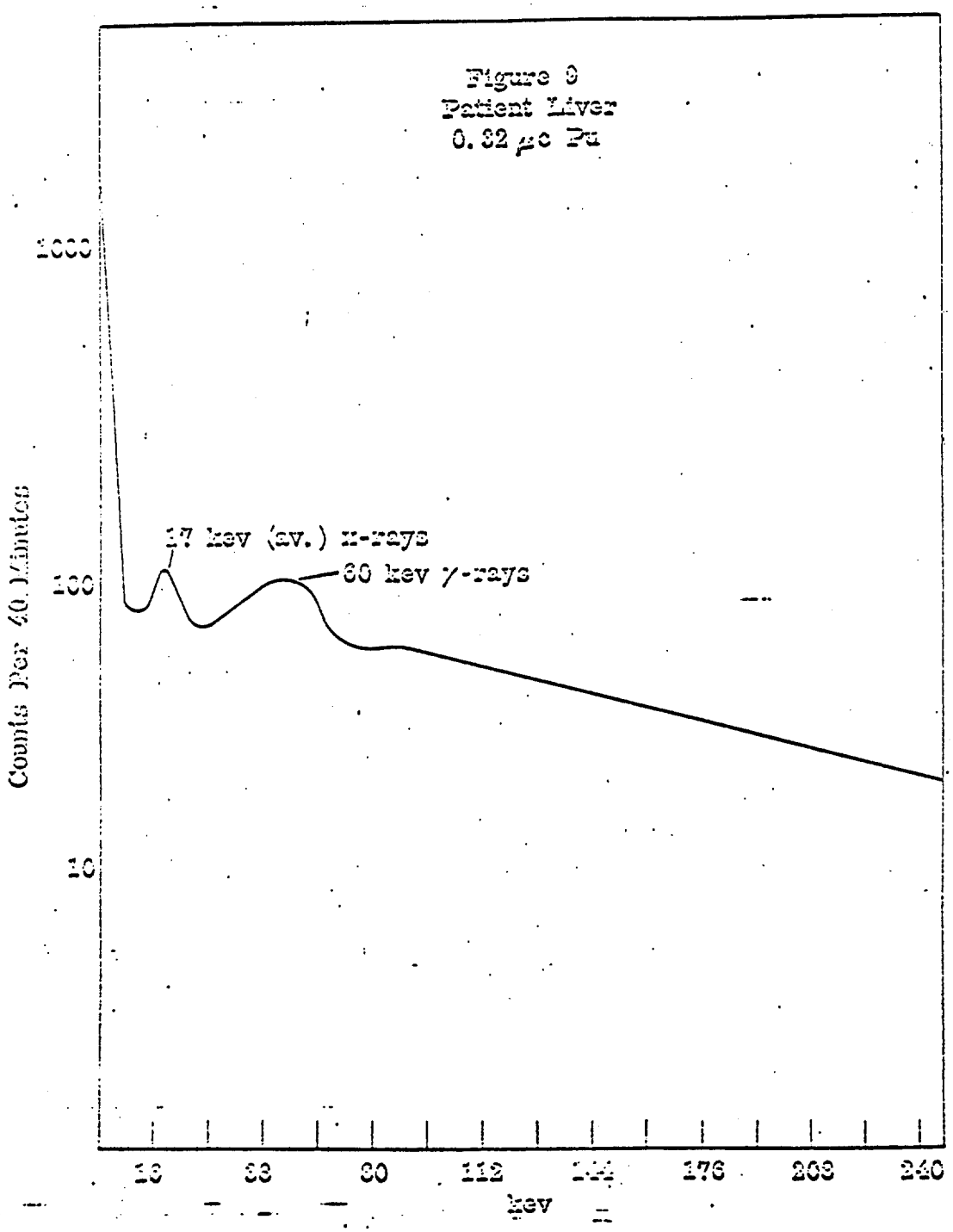
Figure 9
Patient Liver
0.32 μc Pu
1000
100
10
Counts Per 40 Minutes
17 kev (av.) x-rays
60 kev γ-rays
16
48
80
112
144
176
208
240
kev

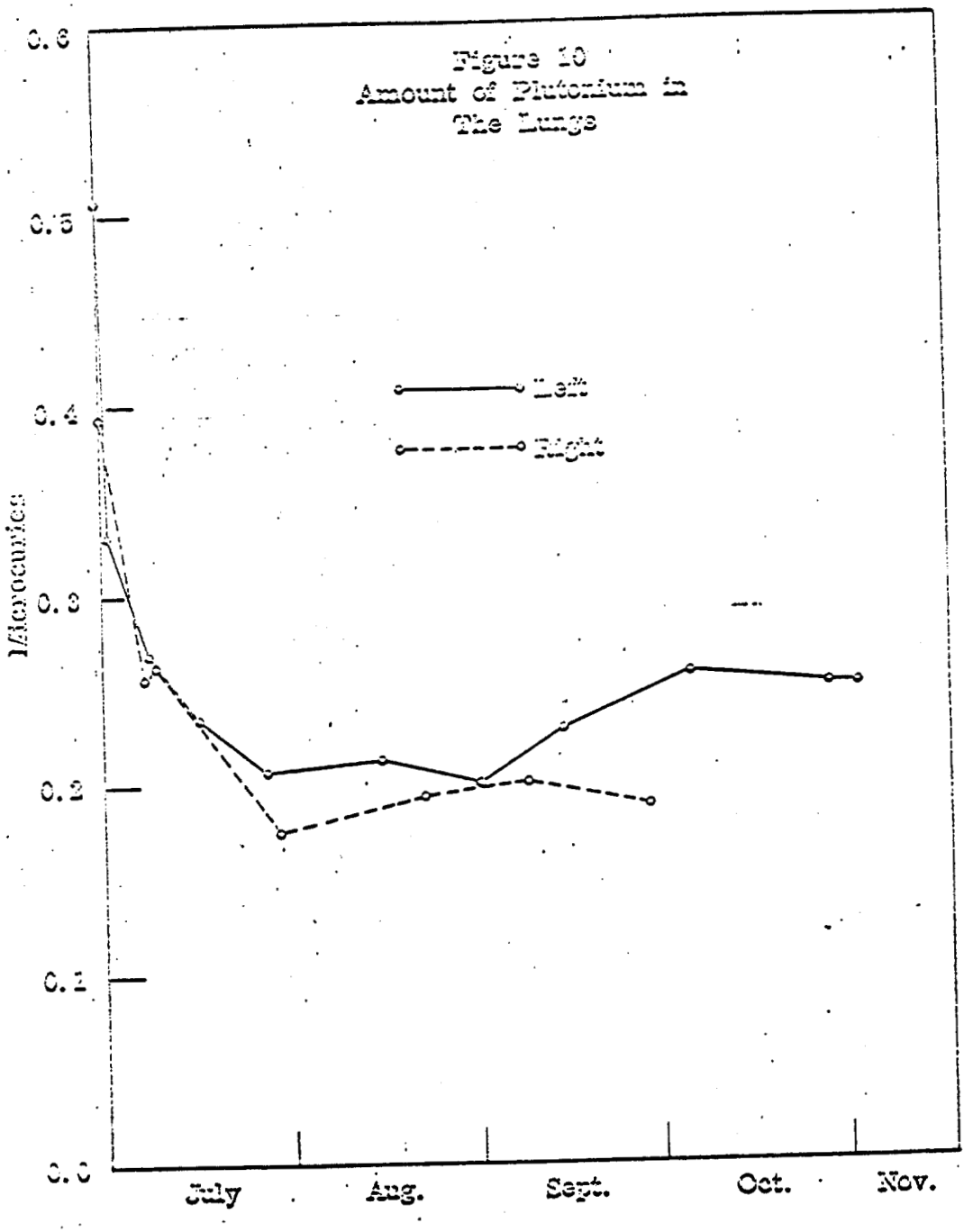
Figure 10
Amount of Plutonium in
The Lungs
Left
Right
Microcuries
0.6
0.5
0.4
0.3
0.2
0.1
0.0
July
Aug.
Sept.
Oct.
Nov.

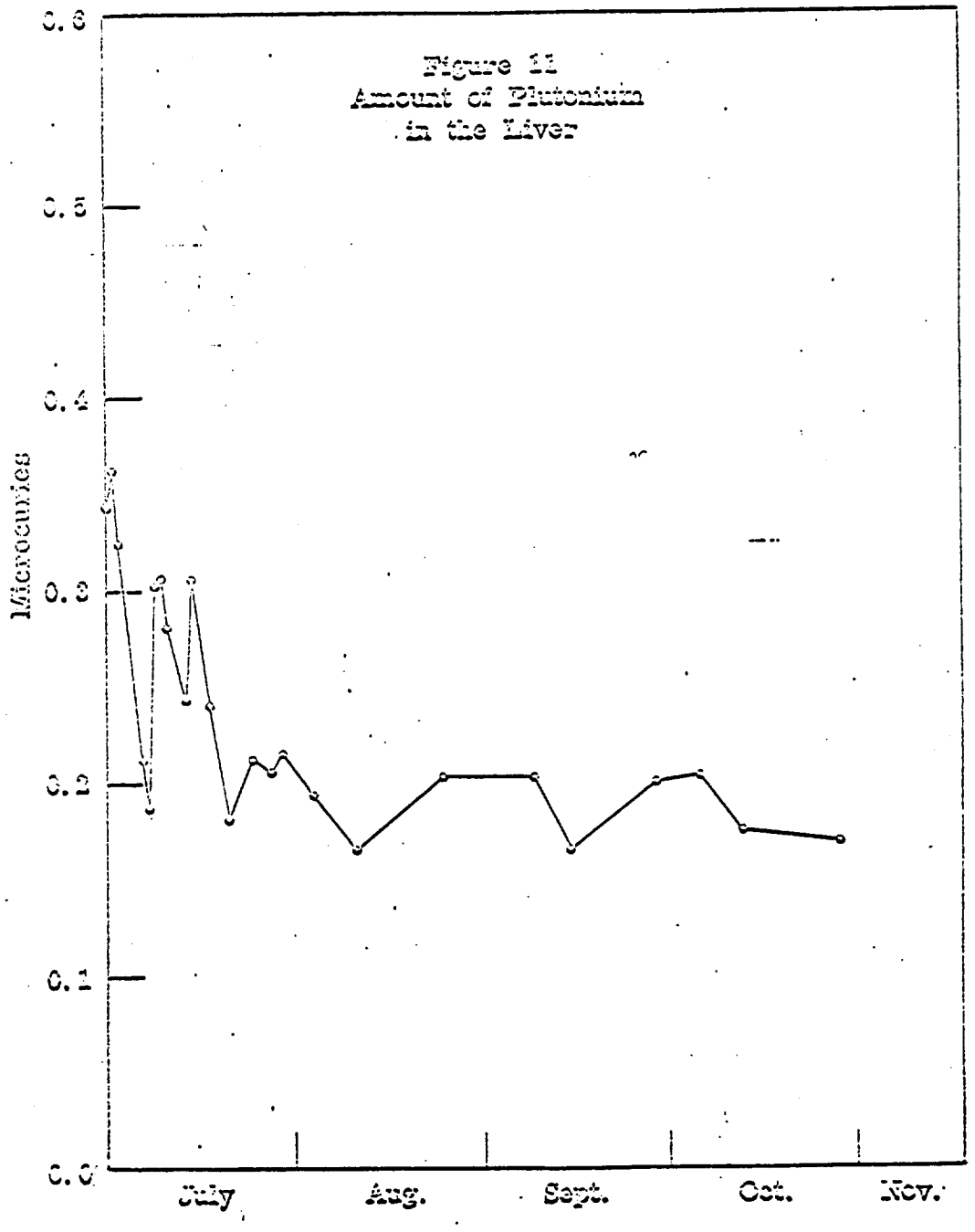
Figure 11
Amount of Plutonium
in the Liver
Microcuries
0.6
0.5
0.4
0.3
0.2
0.1
0.0
July
Aug.
Sept.
Oct.
Nov.

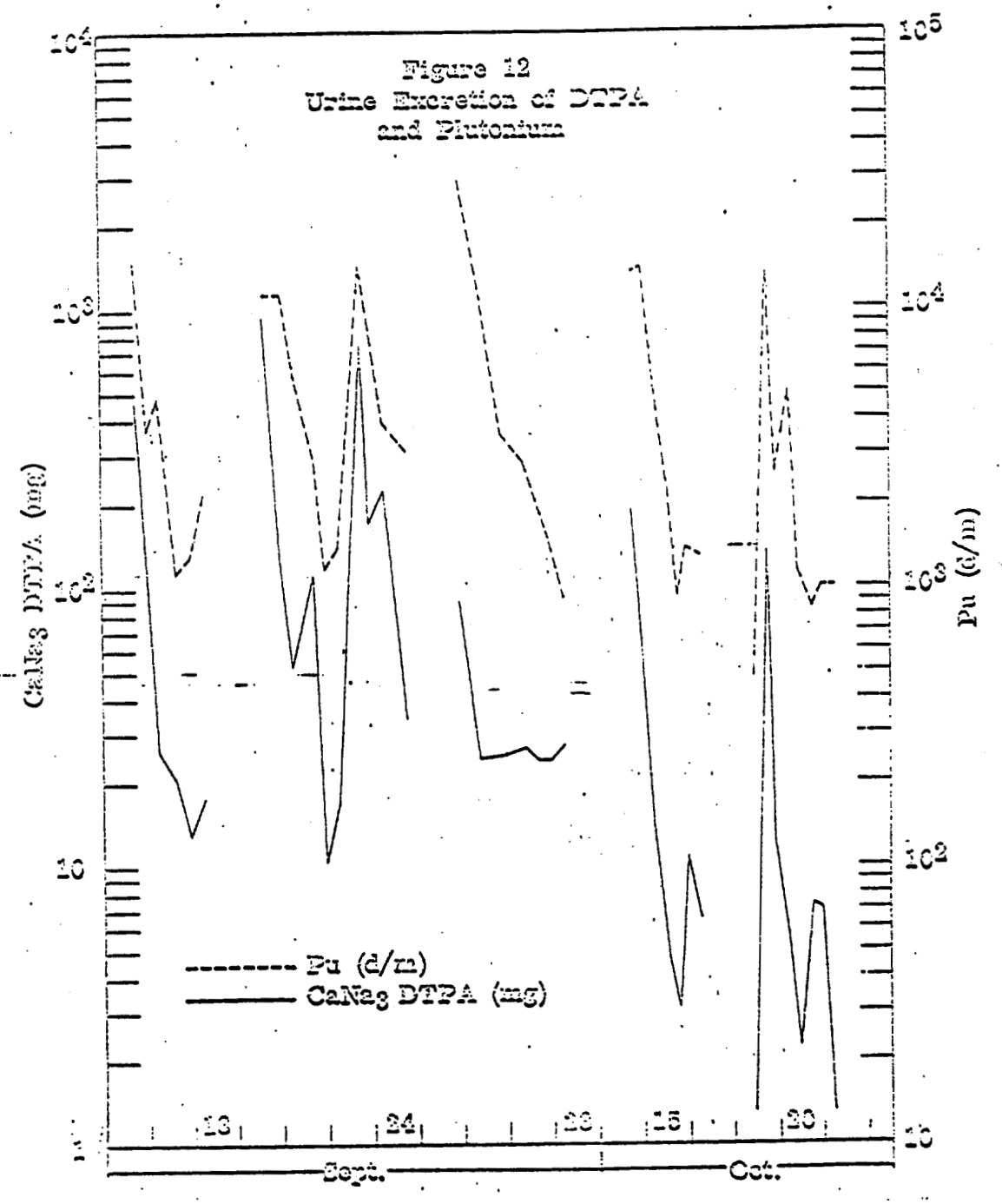
Figure 12
Urine Excretion of DTPA
and Plutonium
10^4
10^3
10^2
10
1
10^5
10^4
10^3
10^2
10
$CaNa_3$ DTPA (mg)
Pu (d/m)
Pu (d/m)
$CaNa_3$ DTPA (mg)
13
24
20
15
30
Sept.
Oct.

8. Measurement Capabilities of the Body Counter

Since the plutonium contained 473 ppm of Am^{241}, there was a sufficient number of 60-kev gammas available to use in evaluating lung and liver burden. It is difficult to establish a meaningful limit of detection for such a system. However, the following was observed and can be used as an index of the capability of the equipment. The background count rate in channels corresponding to 51 kev through 65 kev averages about 33 counts per minute for a 40-minute counting period. The count rate for 0.063 μc of plutonium mixture in the liver cavity of the phantom averages about 24 counts per minute over background for a standard 40-minute counting period. Assuming normal distribution standard error is ± 5.0%. The same amount of material in one lung will average about 16 counts per minute over background with a standard error of ± 7%.

Self-absorption of x-rays in the 13- to 20-kev region was too great for the quantities of interest. The 384-kev gamma ray of Pu^{239} lacks in abundance to make it useful in the region of interest for liver or lung burden determination.

9. Discussion

After an accidental exposure to plutonium, the burden of determining the magnitude of the exposure rests with the health physicist. Medical personnel require guidance in their course of action; management wants to know how serious the exposure is; and, most important, those exposed should be counseled as soon as possible. Counting raw urine by x-rays as soon as it is available provides a means of rapidly assessing the magnitude of the exposure and can at least sort out the serious from the not so serious. A second voiding may be required to get a reliable answer. In either case the answers are available in considerably less time as compared to time required for standard radiochemical analysis. Since the source of the x-rays is from Pu^{239}, Pu^{240} and Am^{241}, it is essential to know the relative amount of each to measure accurately the amount of activity in the urine.

Although the optimum size of DTPA dose was not determined, the data suggest that frequency of treatment may be important to obtain maximum enhancement of excretion.

The capability of the body counter was dependent upon the Am^{241} content in the plutonium. The results presented assume that Am^{241} translocates throughout the body in proportion to the plutonium. This may or may not be true. In the study presented here an increase in Am^{241} of almost 10% would occur in about a 2-month period. It would appear therefore that after a large exposure there is a changing sensitivity which may be affected by Am^{241} growth but may also be affected by a difference in translocation rates. More information in this area would be desirable.

10. Summary

An accident in which an individual received an internal exposure to plutonium through injury to his left hand and by inhalation and ingestion has been described. The course of treatment to the individual, excretion data, and body counter data is presented along with a discussion of methods and equipment used.

11. Bibliography

[1] M. F. Milligan, et al., "Analytical Procedures of the Industrial Hygiene Group, USAEC Report LA-1858 (2nd ed.) 1958 (Unclassified)

[2] Albert Darbey, "Colorimetric Determination of the Sodium Salts of Ethylenediaminetetraacetic Acid," Analytical Chemistry 24, 373 (1952).

Figure 1 DTPA Treatments

Figure 2 Integrated Plutonium Excretion in the Urine

Figure 3 Weekly Plutonium Excretion in Urine as Affected by DTPA Treatment Schedule

Figure 4 Daily Plutonium Excretion in Urine as Affected by DTPA Treatments

Figure 5 Calibration Curve for Raw Urine Counter

Figure 6 Hand Counting Cast

Figure 7 Amount of Plutonium in the Left Hand Following Surgery

Figure 8 Phantom Liver 0.25 μc Pu

Figure 9 Patient Liver 0.32 μc Pu

Figure 10 Amount of Plutonium in the Lungs

Figure 11 Amount of Plutonium in the Liver

Figure 12 Urine Excretion of DTPA and Plutonium

SUMMARY OF ROCKY FLATS EXPERIENCES WITH DTPA AS AN AGENT FOR INCREASING PLUTONIUM EXCRETION VIA THE URINE

by Health Physics

The Dow Chemical Company
Rocky Flats Division
Golden, Colorado

Since 1963 DTPA has been used at the Rocky Flats plant in 23 cases involving exposure to plutonium. Varying degrees of success have been noted based on measurement of plutonium in the urine. The effectiveness of DTPA seems to depend on the following:

1. The chemical form of the plutonium involved in the accident;

2. The length of time between exposure and treatment; and

3. The treatment schedule, dose, and method of administration.

In many cases, however, the degree of success cannot be anticipated. In these cases, DTPA is tried to see whether or not a significant increase in plutonium excretion is obtained.

Generally, our exposure cases have resulted from inhalation, puncture wounds, or acid burns. In three cases, however, exposures resulted from explosions where employees were exposed by both inhalation and puncture wounds.

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Inhalation Cases

Thirteen inhalation cases have been treated with DTPA and one with EDTA (RFP-128).

Eight of these cases resulted from a plutonium fire in October, 1965. Twenty-five persons received inhalation exposures with eight receiving DTPA treatments for a 5-day period. Generally speaking, the plutonium excretion in the urine increased slightly because of the treatments but the total amount excreted represented such a small fraction of the material present that the treatments were discontinued. The plutonium was found to be a very insoluble "high fired" oxide of small average particle size (0.3 micron). This incident is discussed in RFP-710 where several typical urine excretion curves are shown for both the treated and the untreated persons.

Five other inhalation cases were treated with DTPA after separate exposures. In each case, the treatments were suspended after a few days because it was judged that the amount of plutonium found in the urine represented only a small fraction of the material involved. Typical urine excretions for these cases were 1 to 20 dpm per 24 hours during the course of treatment followed by a rapid decrease to nearly pretreatment levels.

Puncture Wounds

Three cases of contaminated puncture wounds have been treated with DTPA and two with EDTA (RFP-128). The urine excretion of one of the cases that was treated with DTPA was reported in RFP-431. This case might be called

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typical of all the wound cases. The plutonium involved was a highly insoluble oxide. The material present at the wound site solubilized only very slowly in the body fluids. This small fraction would then find its way to the blood system where it would be nearly completely chelated by DTPA and excreted in the urine. The DTPA is effective in removing the plutonium from the blood but does not accelerate the solubilization process at the site of the wound. In all cases, urine excretions are higher following the treatment but in each case the treatment was suspended after a few days because only a small fraction of the material present in the wound had been excreted in the urine.

Burns

Four employees have been treated with DTPA following contaminated burns.

Three of these cases involved nitric acid burns. Two of these have been described in RFP-640 where their treatments were called "very successful." In one of these cases, a calculation shows that about 96 percent of the plutonium which entered the system was excreted in the urine. About 1½ times the maximum permissible body burden was excreted in the urine during the course of treatments. The other case reported in RFP-640 was probably equally successful but the smaller quantities of material involved did not permit a similar calculation.

A third employee received DTPA treatments for 5 days following a contaminated nitric acid burn. This case was very similar to the other two but it turned out that

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the treatments were not necessary because very little plutonium entered the body.

The fourth burn case was the result of a solvent burning in a contaminated atmosphere. In this case as well, the treatments were halted after a few days because very little plutonium was found in the urine.

Explosions

Two separate explosions have occurred in which three employees have been exposed to plutonium.

The first occurred in 1957 when two employees were exposed via inhalation, ingestion, and absorption through wounds caused by flying debris. The plutonium to which they were exposed was a mixture of oxides, hydroxides, and nitrates. These two employees received EDTA at the time of the accident. The effectiveness of these treatments can be seen in RFP-128. These two employees also received DTPA treatments 7 years after this incident. The results of these treatments have been summarized in RFP-577. This report shows that while urine excretion levels increased 10-fold, only about 1.5 and 1.9 percent of their calculated body burdens were removed during the course of treatments.

The second explosion occurred in 1963 and resulted in the exposure of an employee via inhalation, ingestion, and absorption from a massive wound of the left hand. The plutonium was a mixture of metal, oxides and chlorides. This case has been reported in RFP-445, -446, -510, and -709. The DTPA treatments in this case were found

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to be very effective. About 8 microcuries or 200 times the maximum permissible body burden was eliminated via the urine during the course of treatments. This not only dramatized the need for DTPA therapy in some accident cases but it also demonstrated the feasibility of orally administered DTPA in certain circumstances.

In general, our experience with DTPA has been very good. In some cases the effectiveness in removing plutonium was easily measurable. In other cases the treatments were used to demonstrate that little or no plutonium had gotten into the system. Even in the cases where little material benefit was derived, there was a good psychological effect.

March 20, 1967

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THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
GOLDEN, COLORADO

U. S. Atomic Energy Commission
Contract No. AT(29-1)-1106

EVALUATION OF LUNG BURDEN FOLLOWING ACUTE INHALATION EXPOSURE TO HIGHLY INSOLUBLE PuO_2

by

J. R. Mann
R. A. Kirchner

RECEIVED 22 August 1966

IN REVISED FORM, _____

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ABSTRACT

On October 15, 1965, a fire in a plutonium fabrication plant resulted in a large-scale spread of plutonium oxide. The Rocky Flats body counter was used to measure the plutonium in the lungs of all employees working in the area. Of approximately 400 employees counted, 25 were found to have enough plutonium in their lungs to deliver a dose of 15 rem per year or greater.

Data from each employee were obtained with two scintillation detectors in contact with the subject's chest. The 60-kev photon peak of Am^{241} was used in the measurements. The Am^{241} content of the plutonium released in the fire was determined and the plutonium quantity was then determined from calibrations done on a chest phantom with similar Am^{241} : Pu^{239} ratios.

The plutonium consisted of "high-fired" PuO_2. Particle size measurements of air samples collected after the fire indicated a 0.32-micron mass median diameter with a geometric deviation (σ_g) of 1.83 . Lung data so far show very slow clearance. This confirms the high degree of insolubility and the small particle size. On the average, 30 percent of material initially deposited was cleared in 2 to 3 months.

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The remaining material is clearing very slowly with little or no measurable absorption into the bloodstream.

INTRODUCTION

Since June, 1964, plutonium inhalation cases have been under study in the Rocky Flats body counter. [1,2] It is known that estimates of lung burdens of plutonium can be made using the body counter if the amount of Am^{241} present in the exposure material is known.

On October 15, 1965, during a maintenance operation in which an attempt was made to unplug an oil coolant line on a lathe, plutonium metal chips caught fire. The fire occurred outside of a glovebox in a 70,000-sq.ft. plutonium production area. About 400 employees were working in the room. Even though the fire was extinguished in a few minutes, airborne contamination spread quickly throughout the area. Air sampler filters indicated an activity range of greater than 1 $\mu c/M^3$ down to 10^{-8} $\mu c/M^3$ at the most remote locations.

Because the contamination spread so rapidly, many employees were in high airborne contamination areas without respiratory protection. Those showing high levels of contamination

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about the nose and mouth were scheduled for body counting first, followed by those with lower contamination levels.

As soon as a man was decontaminated externally, he went to the body counter. Within 2 hr. after the fire the first person was counted. Fourteen people were counted on the first day and evening of the fire. Seven of these showed significant amounts of plutonium in the chest. Eighteen additional exposures were found in the following weeks. DTPA treatments were given by the medical department to eight of the first positive cases. The treatments did not enhance plutonium excretion in the urine, so they were stopped. Subsequent positive cases did not receive DTPA treatments.

INITIAL SCREENING PROCEDURE

The Rocky Flats body counter has been described elsewhere. (3) Two 4-inch-diameter by 4-mm-thick NaI(Tl) detectors are used for chest counting, one on each side of the patient's sternum. Pulse-height spectra from each detector are displayed on 128-channel subgroups of the pulse-height analyzer. Using a calibration of 1 kev per channel, the 60-kev Am^{241} photopeak covers from channels 40 to 80.

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The normal time for routine counting is 40 minutes; however, 20-minute counts are adequate for initial screening purposes. This allows enough counts to accumulate in the 60-kev photopeak region to visually distinguish between a background spectrum and an Am^{241} photopeak on the readout oscilloscope.

Two criteria for initial screening purposes were used to decide if a person had a significant lung deposit:

(1) The presence of an Am^{241} 60-kev gamma ray photopeak, or

(2) A gross count rate of 46 c/m or greater when integrated over the 40- to 80-kev range.

The second criterion was based on Boss' study of the distribution of chest count data for 180 employees. These studies showed an average chest background of 32 c/m in the 60-kev photopeak with a 3σ value of 46 c/m. (4) It was therefore reasoned that a count over this value would be suspect. However, until the Am^{241} content of the exposure material could be determined, the degree of exposure to plutonium was not known.

Fourteen people were counted on Friday, the day of the fire. These were the employees that had been working on the equipment

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or that were in the immediate vicinity when the fire started. Seven of these had counts greater than 70 c/m and showed 60-kev photopeaks. The other seven had counts less than 46 c/m and none exhibited a 60-kev photopeak. After body counting, the plant physician administered 1 gram of DTPA intravenously to the exposed employees. On Saturday, Sunday, and Monday morning, the exposed employees received further DTPA treatments, 1 gram each day, and were given 40-minute counts in the body counter.

All urine and feces from the exposed employees were collected for laboratory analysis. Samples were also requested from all other employees checked in the body counter.

On Monday afternoon, 20-minute screenings were resumed for those employees that were at greater distances from the fire, but still having possibly inhaled plutonium from the fire. It was thought that all of the positive exposure cases had been counted on Friday; however, two more positive cases were found on Monday afternoon, one on Wednesday, and one on Thursday. The screenings continued through the second week following the fire. It was then realized that it would be necessary to count all personnel that were in the building at the time of the fire regardless of their proximity to the

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fire. It would also be desirable to accelerate the screening program in order to handle the number of people involved. It was suggested that two employees at a time might be counted using one detector for each. Experiments were conducted and it was found that a "hot" man could be counted with a "cold" man without increasing the cold man's chest count by placing the two men in seated positions in opposite corners of the room. One detector was placed over the left side of the chest of each of the two subjects. As a background monitor, a third detector was placed in the center of the room over a few bags of sugar. This procedure was begun on November 2, the third week after the fire. Three hundred eighty-eight employees in all were checked. The last positive case was found on December 23.

CALIBRATION

The body counter had been calibrated for plutonium chest measurements before the fire. This was done using a REMAB* phantom and a lung-equivalent compound*. A solution containing 0.3 μc of plutonium with 700-ppm Am^{241} was mixed with the compound and poured into the lung cavities in the

* Manufactured by Alderson Research Laboratories, Inc.; Long Island City, New York

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phantom. A series of measurements were then made using the two 4-inch by 4-mm detectors. A calibration chart was prepared giving the factors for converting counts per minute in the 60-kev photopeak to microcuries of plutonium for a range of Am^{241} concentrations.

CALCULATIONS

Since the measurement of plutonium in the chest is done by counting the 60-kev gamma rays from Am^{241}, it is necessary to have an analysis of the Am^{241} content of the material to which the person is exposed. Once the initial amount is determined, periodic corrections for americium growth are readily made.

Late in the day of the fire a value of 1830 ppm of Am^{241} was determined for the plutonium involved in the fire. Based on the previous calibration, with a detector positioned on either side of the sternum, this gave a value of 45 c/m net for one maximum permissible lung burden (MPLB) of 0.016 μc (see Appendix), or an average value of 22.5 c/m net for each detector.

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Once the Am^{241} content was known, and given the isotopic composition of the plutonium, the counts-per-minute data on each person could be converted to microcuries of plutonium. Most of the persons had not been counted before, so the previously determined average background of 32 c/m in each detector was assumed for each person. This number, when added to the MPLB net value of 22.5 c/m, gave 54.5 c/m as a gross count indicative of 1 MPLB. The previously chosen control point of 45 c/m used in the second criterion then turned out to be about one-half of the MPLB.

Each positive exposure case was immediately restricted from further work in a plutonium area and was put on a periodic 40-minute count schedule. Data from each were compared with previous unexposed employee data to find a person whose weight, Cs^{137} and K^{40} contents were closest to that of the exposed employee. The chest background of this "match" subject was then used as the background for the exposed employee in all subsequent measurements.

RESULTS

Figure 1 shows the data for the maximum exposure case. Blood, urine and fecal analysis data on this individual are also shown for comparison.

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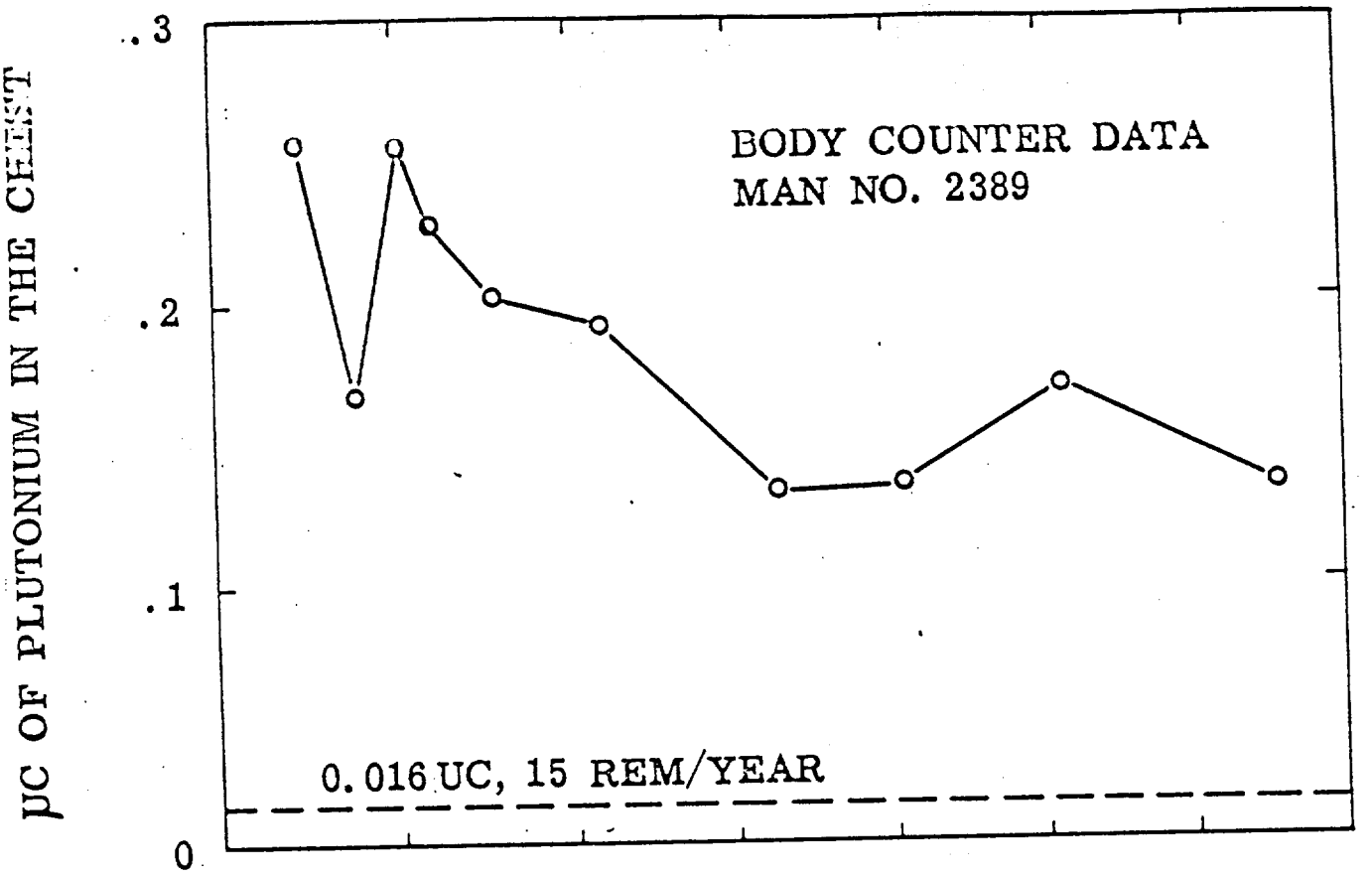

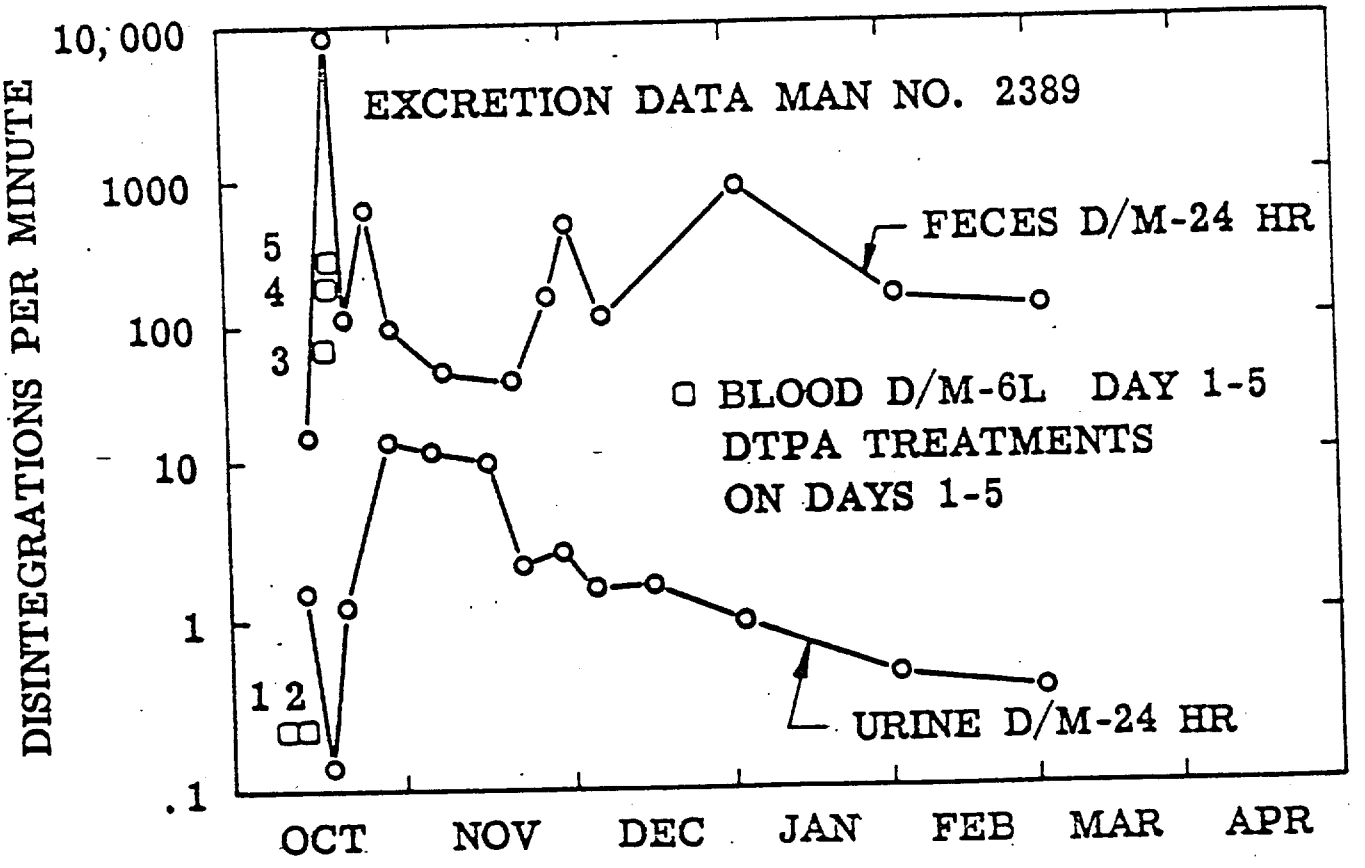

FIGURE 1 - BODY COUNTER AND EXCRETION ANALYSIS DATA ON THE HIGHEST EXPOSURE CASE, 17 LUNG BURDENS.

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The dose to the lung of this individual through December 31, 1965, was 42 rem.

Another typical case, of somewhat lower magnitude, is shown in Figure 2. The dose to the lung integrated through December, 1965, was 13 rem.

Table 1 illustrates the exposure ranges for the 25 cases.

Figure 3 shows body count and bio-assay data typical of eight cases in the 1-2 maximum permissible lung burden range who have been allowed to return to work in the plutonium areas since their exposure. Those cases whose lung burdens decreased to less than 1 MPLB, and appeared to be continuing in a downward trend would be allowed to return to a plutonium area. To date, calculations of retention time in the lung are inconclusive, but retention time appears to be a function of the magnitude of the exposure.

Air samples taken during the fire were analyzed for particle size. (5) The analytical procedure related the number of alpha particle tracks on Kodak Type A nuclear track film to the size of PuO_2 particles deposited on membrane filter paper. The three samples analyzed were obtained at distances of 15, 25 and 50 ft. from the fire. The results from these

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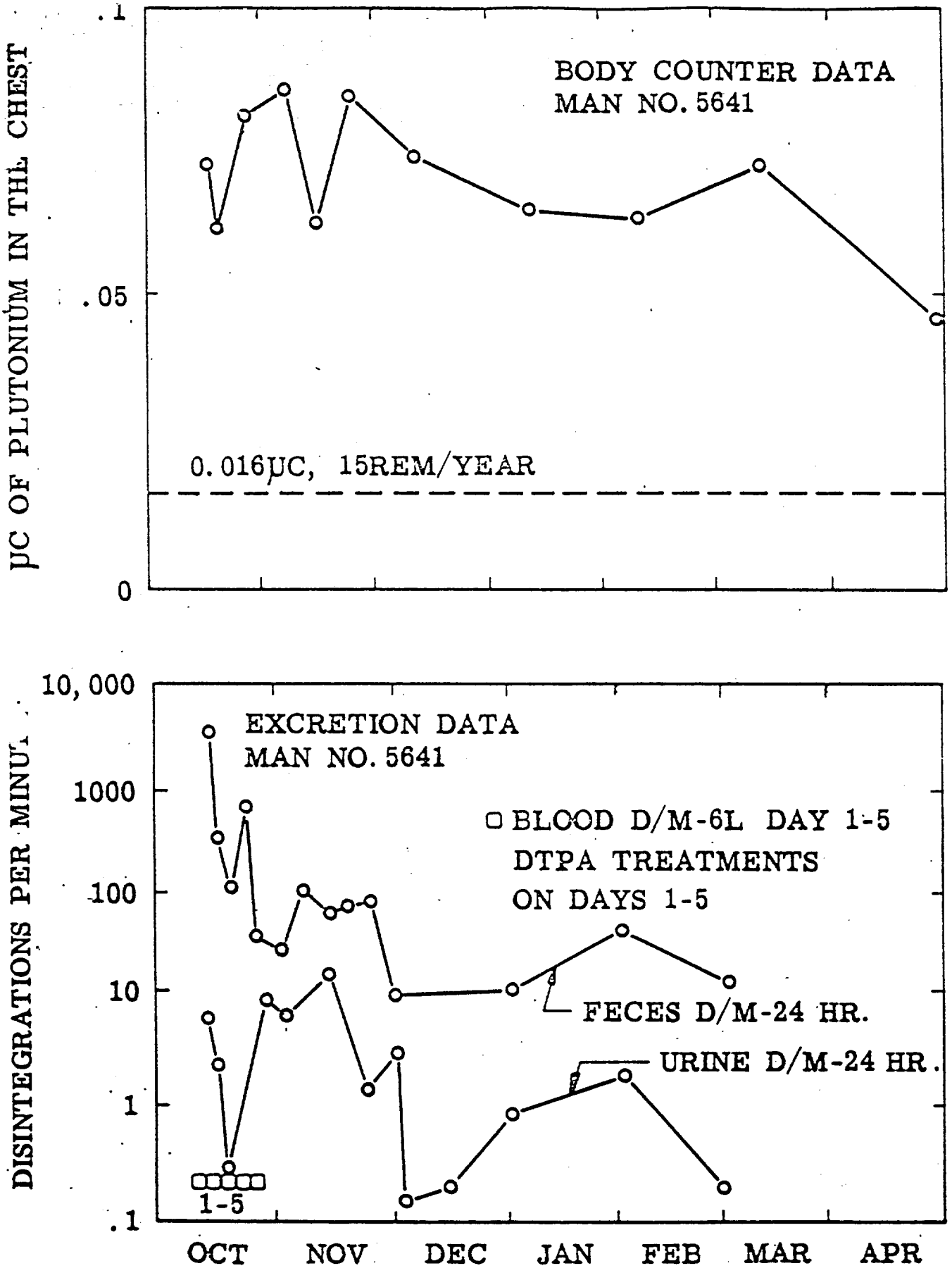

FIGURE 2 - BODY COUNTER AND EXCRETION ANALYSIS DATA ON A MEDIUM EXPOSURE CASE, 4 LUNG BURDENS.

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Table 1. Initial Exposure Range

Number of Maximum Permissible Lung Burdens	Number of Cases
17	1
10	1
7	1
3-5	3
1-2	19

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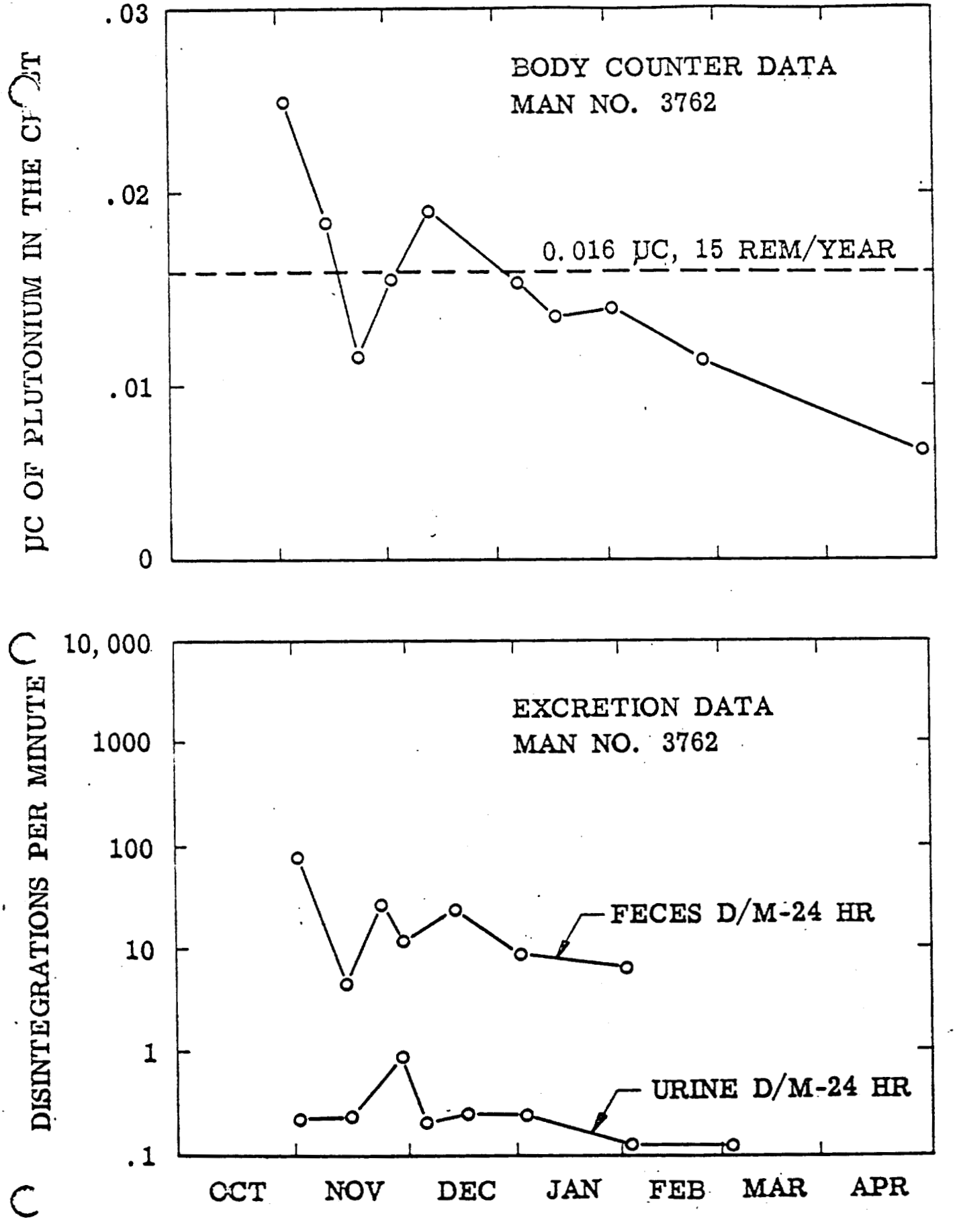

FIGURE 3 - TYPICAL BODY COUNTER AND EXCRETION ANALYSIS DATA ON THE LOWEST EXPOSURE CASES, 1 - 2 LUNG BURDENS.

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three samples and the results from an analysis of the composite of the three samples are shown in Table 2.

Figures 4 and 5 show the particle size distribution of the composite sample. A semiempirical relationship between particle size and particle deposition in the respiratory system is shown in Figure 6. This has proved to be accurate in estimating deposition for previous Rocky Flats plutonium exposure cases. For the size particles shown in Table 2, Figure 6 predicts that 35-40% of the total aerosol will be deposited in the respiratory tract and that 70-80% of the mass deposited will be in the pulmonary compartment.

The clearance of the material from the chest appears to follow two different rates. The initial clearance rate is 30% of the material measured in the chest within a few hours of the exposure in a period of 2-3 months. The final rate appears to be 50% in 1.5 years.

CONCLUSIONS

The DTPA treatments on the first exposure cases did not appreciably increase the amount of plutonium excreted in the urine, and the relatively low amounts in urine and blood indicate a highly insoluble form of plutonium.

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Table 2. Particle Size Data

Air Head Number	Distance from Fire	Number of Particles Sized	Measured Mass Median Diameter
I- 2	50 ft.	435	0.285 microns (σ_g = 1.64)
I- 8	15 ft.	434	0.37 microns (σ_g = 1.92)
I-10	25 ft.	476	0.21 microns (σ_g = 1.72)
Composite		1,345	0.32 microns (σ_g = 1.83)

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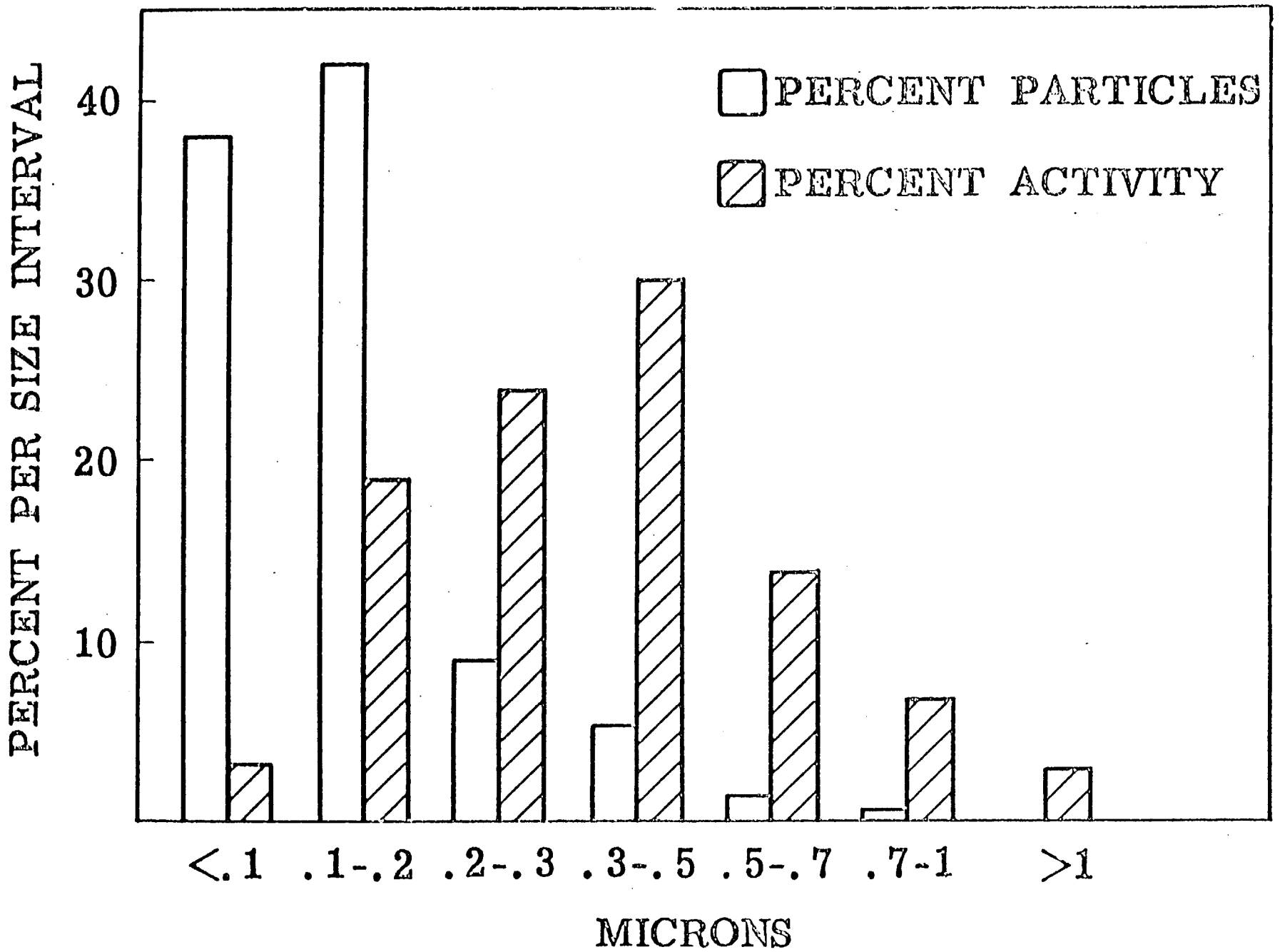

FIGURE 4 - COMPOSITE AIR SAMPLE RESULT (FIRE).

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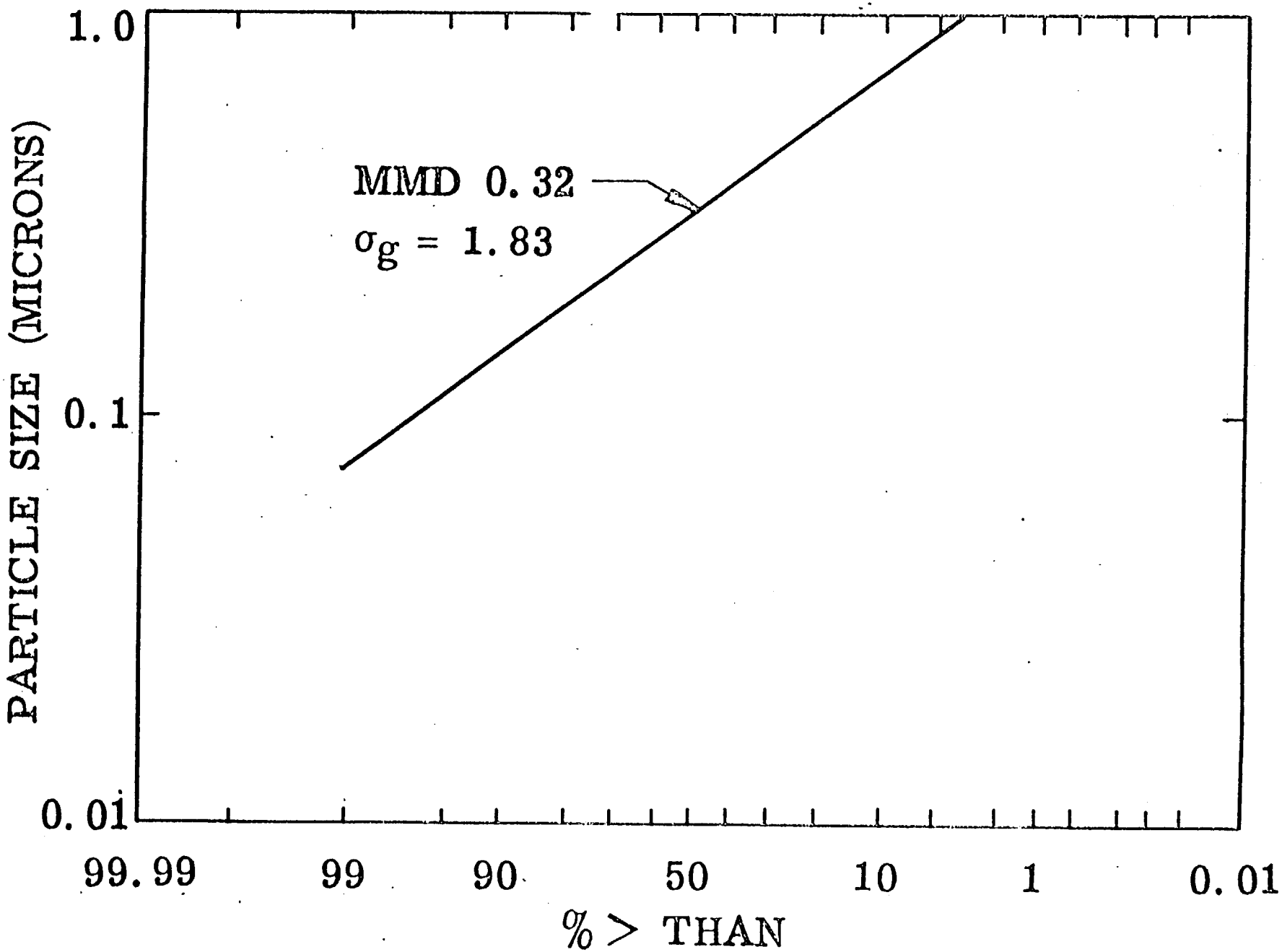

FIGURE 5 - DISTRIBUTION OF COMPOSITE AEROSOL FROM BLDG. 76/77 FIRE.

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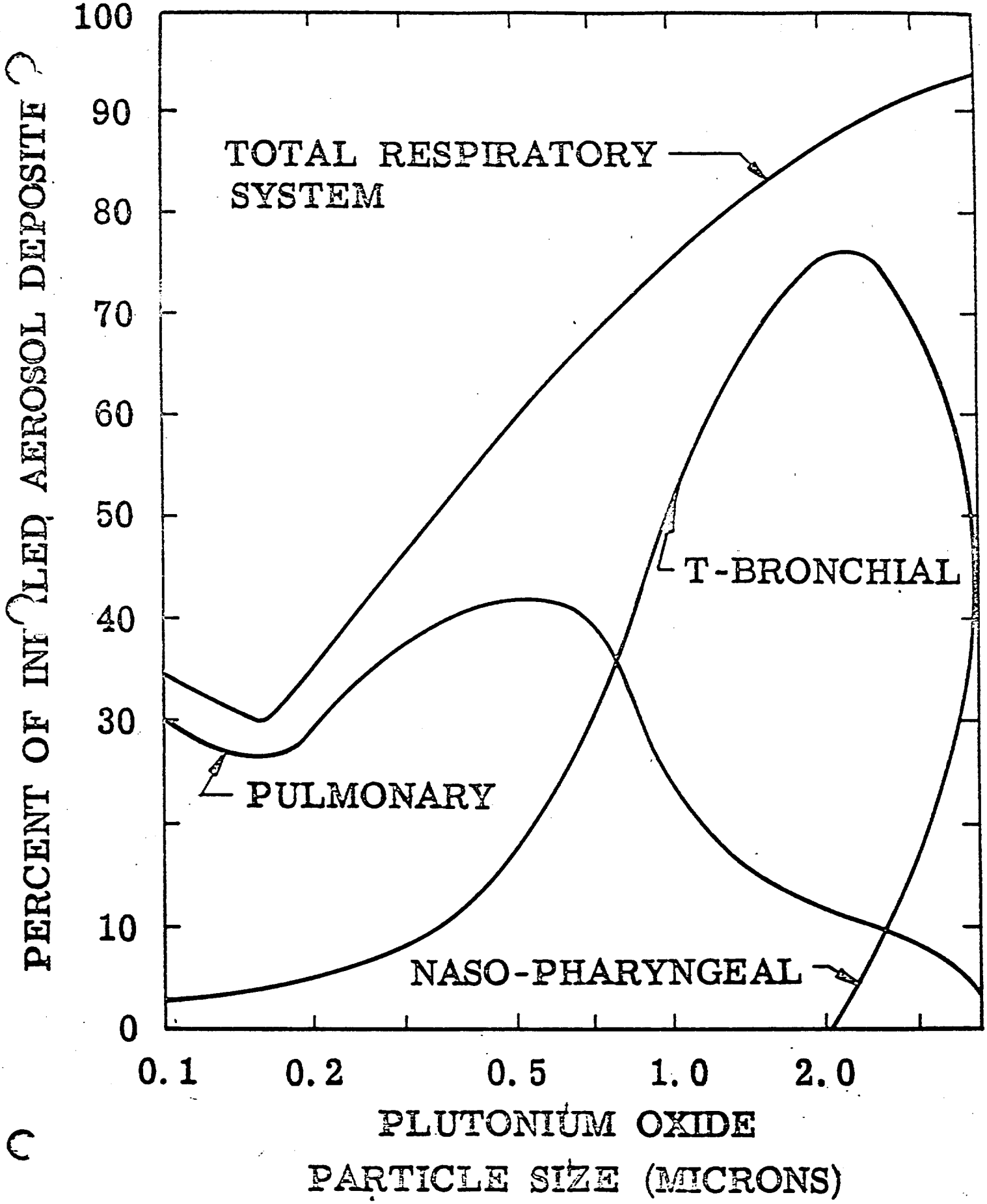

FIGURE 6 - REGIONAL AEROSOL DEPOSITION (PuO_2)

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The long half time of the material in the chest shown by the body counter data obtained so far indicates plutonium of small particle size.

The body counter is extremely valuable as a screening system for suspected plutonium inhalation exposures and in immediate determination of lung burden amounts.

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CAPTIONS FOR FIGURES AND TABLES

FIGURES

1. Body Counter and Excretion Analysis Data on the Highest Exposure Case, 17 Lung Burdens

2. Body Counter and Excretion Analysis Data on a Medium Exposure Case, 4 Lung Burdens

3. Typical Body Counter and Excretion Analysis Data on the Lowest Exposure Cases, 1-2 Lung Burdens

4. Composite Air Sample Result (Fire)

5. Distribution of Composite Aerosol from RFP Fire

6. Regional Aerosol Deposition (PuO_2)

TABLES

1. Initial Exposure Range

2. Particle Size Data

APPENDIX. METHODS WHICH CAN BE USED FOR ESTIMATING PERMISSIBLE LUNG BURDENS FOR PLUTONIUM

1. Using the formulation from the "Report of ICRP Committee II on Permissible Dose for Internal Radiation (1959)"

$$R = \frac{qf_3 \; e}{2.8 \times 10^{-3} \; m}$$

where R = rem/week
qf_3 = amount in organ of reference (μc)
m = mass of organ of reference (g)
e = $\Sigma EF(RBE)n$ (Mev).

using m = 1000 grams (from Standard Man Data)
e = 53 Mev (Report of ICRP Committee II, page 143)

$$R = \frac{qf_3 \; (53)}{2.8 \times 10^{-3} \times 10^3}$$

$$= 18.9 \; qf_3 \text{ rem/week}$$

if 1 μc Pu^{239} = 16.2 μg Pu^{239}, then

$$R = 1.17 \text{ rem/week/microgram } Pu^{239}.$$

2. Using 6.24×10^7 Mev/gram = 1 rad,

$$R = \frac{A_0 \times 2.22 \times 10^6 \text{ dpm/μc} \times 1440 \text{ min/day} \times \bar{E}}{m \times 6.24 \times 10^7 \text{ Mev/gram rad}}$$

where A_0 = μc
m = mass in grams
$\bar{E}$ = average energy, Mev per disintegration
R = rad/day.

using m = 1000 grams and
$\bar{E}$ = 5.15 Mev

$$R = 0.264 \; A_0 \text{ rad/day}$$
$$= 0.0164 \text{ rad/day/microgram.}$$

using a quality factor (QF) = 10,

$$R = 1.14 \text{ rem/week/microgram } Pu^{239}.$$

3. Using $(MPC)_a$ to calculate qf_a: Since $(MPC)_a$ for lung is given as 4×10^{-11} μc/cc in NBS Handbook 69, the ICRP report, and Title 10 CFR 20, some formulations can be worked backwards to give qf_a.

$$(MPC)_a = \frac{10^{-7}\ qf_a}{Tf_a\ (1-e^{-0.693\ t/T})} \quad (\mu c/cc)$$

where T = effective half life (days)

f_a = fraction inhaled which is retained in organ of reference

t = period considered for determining $(MPC)_a$

qf_a = permissible burden (μc).

Using a lung half life for plutonium of 300 days as recommended by most texts and $f_a = 0.125$ which is the basis for all $(MPC)_a$ calculations for plutonium, and substituting in the above

qf_a = 0.015 μc

= 0.242 micrograms.

4. Calculation of permissible dose to the lung from $(MPC)_a$:

$$(MPC)_a = \frac{4.1 \times 10^{-10}\ m\ R\ \lambda_o}{f_a\ (1-e^{-\lambda_o t})\ \Sigma EF\ (RBE)\ n}$$

where λ_o = effective decay constant ($days^{-1}$)
m = 1000 grams
f_a = 0.125
$\lambda_o = \frac{0.693}{300\ days}$

and all other terms are as previously defined.

R = 0.28 rem/week.

5. Checking on the assumption of a 300-day lung half life using the same formula as in part 4 and using R = 0.3 rem/week (".....R = 0.3 for all other organs [bone already discussed] except thyroid and skin,"..... ICRP report) as the allowable lung dose

$$(MPC)_a = \frac{4.1 \times 10^{-10} \times 10^3 \times 0.3 \times 2.31 \times 10^{-3}}{0.125 \times 1 \times 53}$$

$$= 4.3 \times 10^{-11} \ \mu c/cc.$$

The handbook value is 4.0×10^{-11} μc/cc confirming the assumption of a 300-day half life in the lungs.

Summary

a. All indications are that the permissible lung dose should be 15 rem/year.

b. The permissible body burden (qf_2) in the lungs is approximately 0.25 microgram.