# THE MEASUREMENT AND MANAGEMENT OF INSOLUBLE PLUTONIUM–AMERICIUM INHALATION IN MAN*

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**Abstract**—A drybox explosion on January 17, 1966, exposed three individuals to inhalation of dust containing a mixture of $^{239}$Pu and $^{241}$Am. Preliminary measurements at the University of Pittsburgh whole body counter 27 hr after the incident, using a 1 mm thick, 2 in. D, NaI detector indicated a possible lung burden as high as 0.4 $\mu$Ci $^{239}$Pu in one of the individuals. Although a considerable fraction of the contamination was believed to be $^{241}$Am, upper-limit estimates of $^{239}$Pu in the lung were still as high as 0.24 $\mu$Ci on day 4 (post-exposure), so the decision was made to administer 1 g/day DTPA intravenously for the next three days. On day 5, a hundredfold increase appeared in the count-rate and changes occurred in the spectral shapes indicating a sudden appearance of new surface contamination on the anterior chest. This activity, as well as its probable source (another spot of contamination found on the forehead) was removed. Subsequent spectral shapes indicated that further measurements were indicative of lung radioactivity. Isotopic analyses of contamination and air samples showed that the major fraction of the activity was $^{241}$Am. Interim estimates of lung burden were then: $7 \times 10^{-3}$ $\mu$Ci of $^{241}$Am on day 4; $4 \times 10^{-3}$ $\mu$Ci on day 11; $6 \times 10^{-4}$ $\mu$Ci on day 28; and $4 \times 10^{-4}$ $\mu$Ci on day 57. These values are consistent with the elimination of about 36,600 d/min of $^{241}$Am and 54 d/min of $^{239}$Pu in the first fecal sample. Urine excretion rates, initially less than 0.4 dpm/24 hr, increased 50–100 times between days 5–8, suggesting the efficacy of DTPA in removing insoluble $^{241}$Am from the lung.†

## I. INTRODUCTION

In this paper, measurements and final evaluations of internal and external $^{239}$Pu and $^{241}$Am on a glovebox operator following a contamination-releasing explosion will be summarized, showing how a single 2 in. D $\times$ 1 mm sodium-iodide detector may be used together with other bioassay and survey data in the medical management of inhalation accidents involving X-ray emitting nuclides. Also, the possible effi-

* Supported in part by a grant from the Health Research and Services Foundation, Pittsburgh, Pennsylvania. An interim report of this work was presented at the annual meeting of the Health Physics Society in Houston, Texas, June 1966.

† There was an error in the abstract in the *Health Physics* journal. The surprising aspect of this case was that the DTPA seemed to remove what initially behaved like "insoluble" $AmO_2$.

cacy of DTPA administration in removing inhaled plutonium and americium oxides is indicated.

## II. DESCRIPTION OF THE INCIDENT

At 2:05 p.m., Monday, January 17, 1966, an explosion occurred in a glovebox when a technician attempted to ignite a propane torch. The torch had apparently leaked after a new cylinder was attached. The explosion blew out the gloves and knocked the operator to the floor. Within seconds the operator proceeded to the change room and within one minute the plant evacuation alarm was sounded.

Hot gases from the open glove ports had singed the operator's eyebrows and produced minor first degree facial burns. He was also contaminated over his face, hair and chest with alpha activity up to several hundred thousand disintegrations per minute per 100 $cm^2$. Nose swipes read 100,000 counts/min for the right nostril. The glovebox operator showered until all external alpha contamination had been removed, except for one spot reading 1200 counts/min on the right front chest. His nasal contamination was reduced below 1000 counts/min per smear by irrigating with water. Urine samples were collected from all persons involved in the incident and fecal samples were also collected from those who entered the area immediately after the explosion.

Air samples indicated alpha air concentrations of up to $1.1 \times 10^{-7}\mu Ci/cc$ in the room where the accident had occurred, averaged over 10–20 min after the accident. Floor contamination levels in the vicinity of the incident were up to 300,000 counts/min per 100 $cm^2$, and contamination was spread throughout the entire plant. However, the glovebox operator involved had the highest contamination and was the only one who was found later to have measurable internal activity. Thus only his measurements will be summarized here.

## III. EVALUATION AND MEDICAL

On the day after the accident, the company health physicist learned that our new whole body counter facility* had just obtained a thin-window 2 in. D × 1 mm NaI detector.

The equipment was immediately set up and the exposed technician was brought in for his first interim examination 18 hr after the incident. In Fig. 1 the technician is shown lying on a cloth cot, with the 1 mm NaI crystal detector

* See J. O. MEHL and K. R. BECK (Editors), *Directory of Whole-Body Radioactivity Monitors*, IAEA, 1964. In the first case study from the whole body counter, the authors would like to acknowledge the contributions of their former associate, Dr. Francis J. Bradley, toward the design and specifications of the facility.

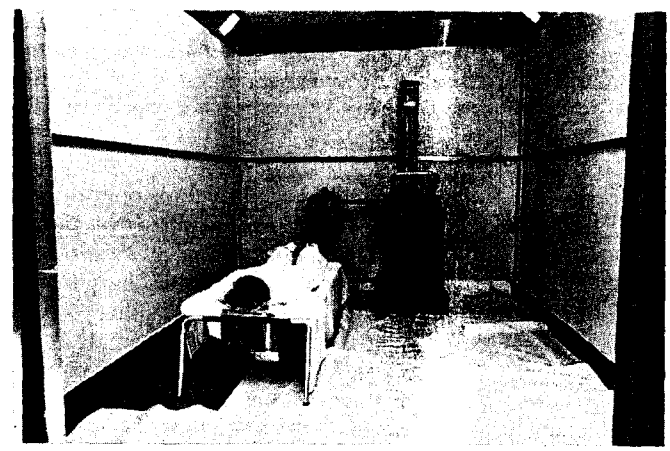

FIG. 1. View of simple apparatus used for emergency detection, 2 in. D × 1 mm NaI crystal with 0.005 in. Al window beneath cloth cot.

placed about 2 cm underneath his chest. This detector was purchased from Harshaw with a 0.005 in. aluminium window, which transmits about 96% of 17 keV photons. This detector is similar to that originally developed by Roesch and Baum for wound monitoring.[1] Twenty minute counts at a given position were repeatable within a factor of two. Many 20-min counts were subsequently taken near different body positions, and spectral shapes and intensities were utilized together with geometry considerations to discriminate between internal and external radioactivity and its location in the body. Only a brief illustration of the measurements, lines of reasoning, and resulting medical actions will be given here.

All spectra were recorded on a Northern Scientific Company 256-channel analyzer and a teletype printer, with all settings remaining fixed during the series of examinations. Only half the memory was used for each count so that spectral shapes could be inter-compared on a log scale. This turned out to be a valuable procedure in the detective work that followed. With so many variables to consider during such an emergency, it is most important to fix those parameters that can be fixed and constantly inter-compare data with given standards. In this way, confusion and inadvertent errors may be minimized.

All spectra connected with the evaluations were photographed with a polaroid camera and kept in a notebook in order of date.

Figure 2 shows a comparison of the chest spectra of the exposed technician (on the right) with that of one of the unexposed technicians (on the left) as first observed 18 hr after the accident. Due to time considerations in the early period of the emergency, the gain was fixed to that of the preceding measurement with the 17 keV peak (of $^{239}$Pu or $^{241}$Am) in only channel 3 and the 60 keV peak of $^{241}$Am in channels 21–25. This turned out to be a fortunate operational decision since the printer began to print out zeros above channel 30 several days after the accident. Although a more expanded spectra would have been clearer, the highest count of the 17 keV peak remained consistently in channel 3 for an unattenuated source during the critical weeks of measurement. Thus, ratios of counts in channels 21–25 could be used to study body absorption. The low relative height of the 17 keV peak, as in Fig. 2, compared to that of a smear of contamination from the incident was initially indicative of a possible internal burden of plutonium or americium.

At the time of the first measurement 18 hr after the accident, standard solutions of plutonium and americium were not yet prepared. Thus, a preliminary estimate was made of the maximum possible internal lung burden, as-

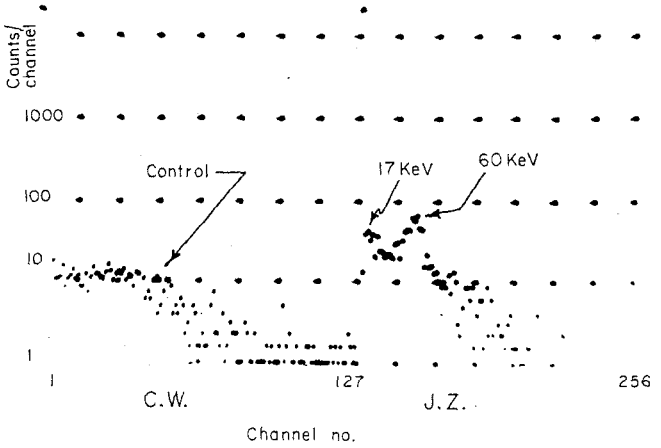


FIG. 2. Comparison of spectra of uncontaminated employee ("control" indicated on left) with spectrum of exposed technician 18 hr post-accident (right). Vertical counts channel scale is logarithmic.

*Table* 1. *Excretion Data Versus Time: Urine Data*

| Date | Total activities in sample* | |
|---|---|---|
| | dpm $^{238}$Pu | dpm $^{241}$Am |
| 1/18 (1st void) | 0.05 ± 0.05 | 0.3 ± 0.2 |
| 1/18 (24 hr) | 0.3 ± 0.2 | 0.5 ± 0.3 |
| 1/20 (24 hr) | 1.33 ± 0.16 | 0.6 ± 0.3 |
| 1/21 (24 hr) | 0.29 ± 0.06 | Lost Sample |
| 1/22 (*Preinfusion*) | | 0.08 ± 0.08 |
| 1/22 (*Post DTPA* 1) | 2.6 ± 0.33 | 4.4 ± 0.2 |
| 1/23 (*Post DTPA* 2) | 1.55 ± 0.31 | 31.4 ± 2.6 |
| 1/24 (*Post DTPA* 3) | 2.37 ± 0.24 | 28.5 ± 2.8 |
| 1/25 (24 hr) | 1.92 ± 0.13 | 7.5 ± 0.8 |
| 1/26 (24 hr) | 2.6 ± 0.7 | 36.7 ± 2.3 |
| 1/27 (24 hr) | 2.2 ± 0.2 | 25.4 ± 1.5 |
| 1/28 (24 hr) | 1.29 ± 0.16 | 17.6 ± 1.2 |
| 1/29 (24 hr) | 0.94 ± 0.12 | 10.1 ± 0.5 |
| 1/30 (24 hr) | 0.8 ± 0.1 | 17.4 ± 0.8 |
| 2/8 (24 hr) | 1.1 ± 0.1 | 0.5 ± 0.23 |
| 2/9 (24 hr) | 0.84 ± 0.15 | 4.9 ± 0.4 |
| 2/14 (24 hr) | 0.29 ± 0.12 | 1.3 ± 0.1 |
| 3/20 (24 hr) | 0.09 ± 0.08 | 10.9 ± 0.9 |

*Fecal Data.*

| Date | dpm $^{238}$Pu | dpm $^{241}$Am |
|---|---|---|
| 1/19 (83.7 g) | 57 ± 6 | 36,600 ± 1,100 |
| 1/21 (84.1 g) | 1.9 ± 0.8 | 8.8 ± 1.5 |
| 1/22 | 0.8 ± 0.9 | 2.8 ± 1.3 |
| 1/23 | 2.1 ± 0.9 | 16.9 ± 1.7 |
| 1/24 | 2.2 ± 0.9 | 15.3 ± 1.6 |
| 1/26 | 2.3 ± 0.5 | 26.6 ± 2.2 |
| 1/27 | | 18.6 ± 1.6 |
| 1/29 | | 15.1 ± 1.6 |
| 1/30 | | 11.3 ± 1.5 |
| 2/8 | 0.48 ± 0.15 | 1.5 ± 0.2 |
| 2/9 | 2.7 ± 0.7 | 12.8 ± 1.1 |
| 2/14 | 0.5 ± 1.1 | 4.7 ± 0.6 |
| 3/20 | 2.6 ± 0.7 | 1.5 ± 0.3 |

* Attempts were made to obtain 24-hr samples from the subject, but collection may be uncertain in some cases; also collections were separated between DTPA infusions in order to determine the effects of each treatment.

sumed to be $^{239}Pu$, on the basis of the assumption that about 4% of the alphas from $^{239}Pu$ are accompanied by $^{235}U$ $L$ X-rays in the 17 keV region. Using the reported attenuation of 17 keV X-rays in the tissue of dogs,[2] considering the half-value layers of about 0.6 cm from 17 keV X-rays in soft tissue and 0.03 cm in bone,[3] and a conservative factor of 3 to correct for rib absorption, it was estimated at 18 hr after the accident that the maximum possible lung burden of the exposed technician could be as high as 0.4 $\mu$Ci—about 10 times the amount that could be permitted to solubilize and enter the bone. The detection limit for $^{239}Pu$ with a 20-min count was estimated to be 0.03 $\mu$Ci, with a 5% chance of not detecting this amount if present in the lungs. This is only about twice the detection limit of Swinth, Griffin, and Park[2] of 0.016 $\mu$Ci in dogs using a 52-detector arrangement for total body counting. Since the single detector may be placed closer to a particular organ containing the activity, the use of more and more detectors may reach a point of diminishing returns when activity is located in a small region of the body.

Since the probable contamination was believed on the day after the incident to be insoluble plutonium oxide, it was decided to establish the fecal and urinary excretion for a few days and take further counts before deciding whether to administer DTPA. It was believed that any remaining plutonium in the lung at 18 hr would be solubilized by the body to a negligible degree in a few days, and that any soluble portion of the plutonium had probably already been transported to bone and could be partly removed by DTPA at a later time.[4, 5] The fecal and urinary excretion data may be followed in Table 1 and an abstract of the chest count data with the 2 in. D × 1 mm NaI detector is presented in Table 2. Counts over other areas of the body were also used in establishing the locations of internal and external contamination.

By day 4 post-accident, standard solutions were prepared containing 1 $\mu$Ci of $^{239}Pu$ and 0.044 $\mu$Ci of $^{241}Am$ in each of three solution volumes: 100 cc, 500 cc, and 1000 cc. The 1000 cc solution was used to simulate the lung spectrum and X-ray emission for comparison with the technician. The other solution gave some idea of the change in spectral shape with varying degrees of self-absorption, for use in discriminating between surface and internal contamination. The actual percentage of $^{241}Am$ in the contamination was initially believed to be smaller than this. However, the high $^{241}Am$ content of the fecal samples had indicated by day 4 that $^{241}Am$, which had previously been handled in the same glovebox, might actually be the major fraction of the contamination unless the body was preferentially eliminating the $^{241}Am$. (A paper in this conference[6] indicates that such preferential elimination may actually occur under some circumstances.) Thus,

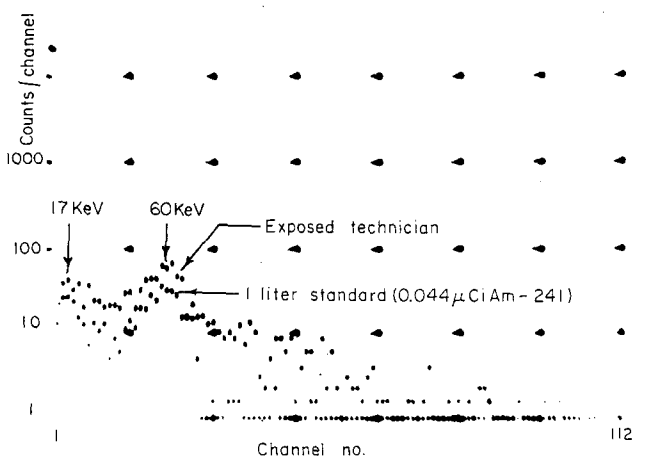


FIG. 3. Comparison of spectrum of exposed technician 4 days post-accident with spectrum of 1 $\mu$Ci $^{239}Pu$ and 0.044 $\mu$Ci $^{241}Am$ in 1 l. solution in polyethylene. Control counts of normal subject have not been subtracted from the technician's spectrum.

*Table 2. Summary of Chest Count Data from Whole Body Counter*

| Date | Detector position | Net counts above control[a] per 20 minutes | | Estimated maximum quantity of $^{241}Am$ in lung ($\mu$Ci) |
|---|---|---|---|---|
| | | 17 keV peak | 60 keV peak | |
| 1/18/66 (1 day post-incident) | 1 in. below right front chest | 239 | 427 | 0.021 $\mu$Ci $^{241}Am$ or 0.46 $\mu$Ci $^{239}Pu$* |
| 1/21/66 (4 days post-incident) | Right front chest | 118 | 253 | 0.012 $\mu$Ci $^{241}Am$ or 0.27 $\mu$Ci $^{239}Pu$* |
| | Right front chest repeat | 28 | 126 | 0.006 $\mu$Ci $^{241}Am$ |
| | Right back chest | 13 | 56 | 0.003 |
| | Left front chest | 32 | 114 | 0.006 |
| 1/22/66 (5 days post-incident) (DTPA treatment at noon) | Right front chest, 5:20 pm (*Note*: High 17/60-keV peak ratios and comparison of front and back counts indicated the sudden appearance of contamination in chest region.) | 17,464 | 8,472 | † |
| | Right front chest, 5:55 pm | 22,731 | 13,704 | † |
| | Left front chest (*Note*: Contamination removed with undershirt, patient showered.) | 3,810 | 5,679 | † |
| | Right front chest, 10:00 pm | 249 | 372 | 0.018 |
| 1/23/66 (6 days post-incident) (DTPA treatment at noon) | Right front chest | 91 | 230 | ‡ |
| | Left front chest | 24 | 50 | |
| | Left back chest | 56 | 148 | |
| | Right back chest | 60 | 104 | |
| | Right front chest *Note:* Repeated after shower, spot of head contamination removed.) | 83 | 140 | 0·007 |
| 1/24/66 (7 days post-incident) (DTPA treatment at noon) | Right front chest | 11 | 52 | 0.003 |
| | | 8 | 17 | < 0.002 |
| 1/28/66 (11 days post-incident) | Average of 4 chest positions | 30 | 82 | 0.007 |
| 2/8/66 (22 days post-incident) | Average of 4 chest positions | 6 | 20 | < 0.002 |
| 2/14/66 (28 days post-incident) | Average of 4 chest positions | 9 | 12 | < 0.002 |

* Early interpretations assumed $^{239}Pu$ predominant and used a conservative factor of 3 to correct for rib absorption.

† Surface contamination.

‡ These counts were influenced partly by contamination found on the forehead.

(a) Control counts of 26 per 20 min in the 17-keV region and 69 per 20 min in the 60-keV region were subtracted from each 20 min patient count.

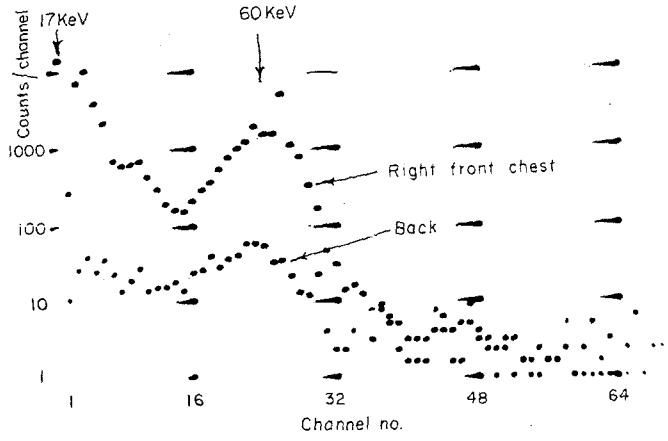


FIG. 4. Spectra of contamination discovered on chest, day 5 post-accident as viewed by crystal from front right chest (top) and through back (bottom). Oscilloscope *X*-axis was expanded, but analyzer gain was the same as in Figs. 2 and 3.

the presence of a known amount of $^{241}$Am in the standard was desirable.

Consequently, estimates of lung burden based on assumed $^{239}$Pu were made using only channels 2–4 in the region of the 17 keV peak. Estimates of $^{241}$Am were made using only the 60 keV peak in the region of channels 21–25 (see Figs. 2–6). Of course, $^{241}$Am also yields *L* X-rays (0.12 of 13.96 keV, 0.13 of 17.76 keV, and 0.03 of 20.80 keV per disintegration) as well as gamma-rays (0.36 of 59.57 keV and 0.025 of 26.36 keV per disintegration),[7] so it was recognized that if an appreciable quantity of $^{241}$Am were present, the $^{239}$Pu burden could be grossly overestimated. Later radiochemical analyses of air samples and contamination showed that more than 85 per cent of the activity was $^{241}$Am, so evaluations of $^{239}$Pu given in Table 2 are given only to illustrate the sequence of events that determined courses of action. Figures 2–6, as well as spectra of the contamination, also confirm that any additional $^{239}$Pu represented in the 17 keV peak can be considered to contribute only a fraction of the

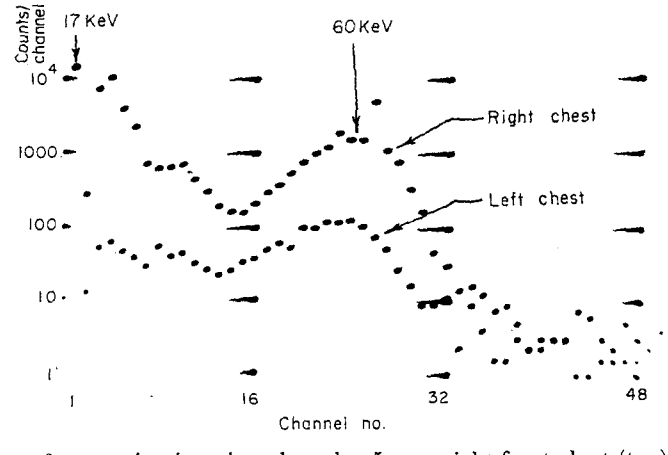


FIG. 5. Spectra of contamination viewed on day 5 near right front chest (top) and left front chest (bottom).

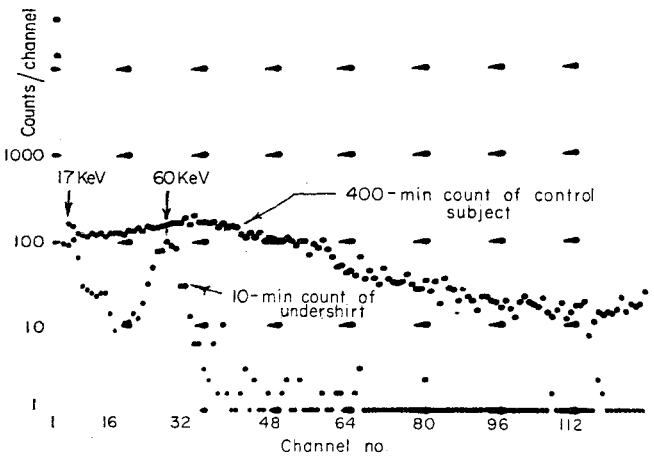


FIG. 6. Spectra of contaminated undershirt (bottom—10-min count) and unexposed subject (top—400 min-count).

exposure, since the permissible burdens of $^{239}$Pu and $^{241}$Am are about the same, and the proportion of $^{241}$Am/$^{239}$Pu may be expected to grow with time.[8]

On Friday, January 21 (day 4 post-incident), spectra were obtained similar in shape to that observed 18 hr post-incident, but somewhat lower in intensity (Table 2). Figure 3 shows one of the chest spectra taken on day 4 compared to the spectrum of the standard solution that was obtained by that day. The 1 l. standard contained 1 $\mu$Ci $^{239}$Pu and 0.044 $\mu$Ci $^{241}$Am. By 4 days post-incident, information from fecal analyses had indicated that the dust might be predominantly $^{241}$Am; yet the low 17 keV/60 keV peak ratio compared to that of the smear indicated a probable internal burden greater than 10% of the MPBB even for $^{241}$Am. Also, fecal excretion after the first day had decreased to very low levels. So the decision was made to administer 1 g DTPA intravenously on each of days 5, 6, and 7, and to follow the lung burden and excretion data daily to determine whether fecal and/or urinary levels would increase and whether the lung burden would subsequently decrease.

Suddenly, after the first DTPA treatment on day 5, an increase by up to a factor of 1000 was noted in the count-rate over the right chest area. Figure 4 shows the spectrum viewed with the crystal near the front right chest (top), and viewed through the back (bottom), with the horizontal oscillope scale expanded. The relatively high 17/60 keV peak ratio from the front of the chest (see Fig. 4) and the much lower intensity viewed through the back, indicated the presence of a new speck of contamination on the chest.

Figure 5 shows a spectrum near the right front chest (top) compared to that near the left front chest (bottom), localizing the contamination to the right front chest area. Late on the 5th day, the technician's undershirt was carefully removed and placed in a plastic bag. Then his chest was washed, the wash-rag was saved in a plastic bag, and the technician was sent in for another shower.

In Fig. 6, a spectrum of the undershirt counted for 10 min (bottom) is shown together with the spectrum of an unexposed individual counted for 400 min. The spectrum from the undershirt, which offered little absorption of the 17 keV peak, again shows a high 17 keV/60 keV peak ratio.

Since the undershirt was contaminated, and the wash-rag was found uncontaminated, it was deduced that the observed contamination had suddenly appeared on the undershirt when it was pulled over the head that morning. On the 6th day, further alpha monitoring with an Eberline PAC-3A found some residual contamination of about 1000 dpm on the forehead,

indicating the probable source of the contamination found on the undershirt the previous day. After further showering the count near the head was reduced somewhat but not enough to indicate from geometry considerations that the contamination, while on the head, had appreciably affected lung measurements before day 5. Further chest spectra were consistent in shape and intensity, as viewed from front and back, and indicated true lung burdens.

After isotopic analyses confirmed that the contamination was more than 85% $^{241}$Am, evaluations of lung burden using the 60 keV peak then ranged from 0.021 $\mu$Ci $^{241}$Am after 1 day to less than 0.002 $\mu$Ci by day 7. In these evaluations only a factor of 2 was used to correct for rib absorption since experiments with a human skeleton showed this would be sufficiently conservative to account for the range of factors observed.*

Table 1 shows the high initial fecal excretion of 36,000 dpm $^{241}$Am (0.016 $\mu$Ci) and 57 $\pm$ 6 dpm $^{239}$Pu on the first day, with a negligible amount initially in urine. The fecal excretion dropped sharply on the 2nd and 3rd days and then both the fecal and urine excretion increased sharply following the DTPA administration. This striking effect of the DTPA was somewhat surprising since the inhaled material was mostly americium and plutonium oxides. No mathematical models can be fitted precisely to the data since the body processes were complicated by the DTPA administration, which apparently assisted in the rapid reduction of the internal portion of the contamination to below the sensitivity of the counter. (Table 2). Of course, there was no opportunity for a control experiment to determine whether the lung burden would have decreased even without DTPA administration. The primary medical consideration was to minimize the risk to the technician.

The initial amount of fecal excretion (0.016 $\mu$Ci), and the remaining lung burden of 0.02 $\mu$Ci after day 1 (Table 2) are considered within the range of ICRP models. This lends further confidence that the evaluated burdens in Table 2 are of the correct order of magnitude. The technician was followed for 4 months with no further appearance of detectable $^{241}$Am.

## IV. CONCLUSIONS

The following main points were emphasized in the above incident:

1. Although not conclusive, the correlation with time between the increase in excretion rates and the rapid disappearance of the lung burden following DTPA administration, suggests that DTPA may hasten the removal of even insoluble oxides of plutonium and americium from the human lung, although removal rates cannot be specified accurately from the data of this case.

2. A number of principles of emergency management of inhalation accidents were emphasized in this incident, including:

(a) Necessity for direct supervision of all measurements and actions at the whole body counter.
(b) Recording of all data and events with time and date as they occur.
(c) Necessity to keep exposed subject properly informed and briefed to develop an attitude of co-operation and trust, while avoiding alarming statements or divulging premature evaluations to unauthorized persons.
(d) Necessity to assign tasks quickly to all staff; everyone wants to help in an emergency, and their help must be constructive.

3. Multiple measurements and examination of spectral shapes with a 2 in. D $\times$ 1 mm NaI detector can be helpful in the detection of $^{241}$Am or $^{239}$Pu in the human lung, and in discriminating between internal and external body contamination. A 20-min count with the detector within a few centimeters of the chest can detect more than approximately 0.002 $\mu$Ci $^{241}$Am or 0.03 $\mu$Ci $^{239}$Pu. A 200-min count (or ten 20-min counts, which give the same statistical precision but better information on time dependence and fluctuations of measurement) can detect about 0.01 $\mu$Ci $^{239}$Pu or 0.0007 $\mu$Ci $^{241}$Am at the 5% level of significance, as defined by Roesch and Baum.[1]

* The authors are indebted to Mr. Andrew Bukovitz for carrying out the measurements.

**ACKNOWLEDGEMENTS**

The authors acknowledge the assistance of Mr. Robert Goempel, Mr. William Moore, Dr. Plinio Rey, and Mr. William C. Judd in setting up equipment and helping with the measurements during this incident.

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