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BIOASSAY EXPERIENCES IN SUPPORT OF FIELD OPERATIONS ASSOCIATED WITH WIDESPREAD DISPERSION OF PLUTONIUM

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THE INCIDENT

During 1966 popular news periodicals such as *Newsweek* (1966a, b, c, d), *Time* (1966a, b, c), *Business Week* (1966a), *U.S. News and World Report* (1966a, b), *Life* (1966a, b) and *Saturday Evening Post* (1967) carried accounts of an aircraft accident near Spain involving planes of the United States Air Force. In addition, *Radiological Health and Data Reports* (1966), *Azarcot* (1967), *Hawkins* (1966), *Newsweek* (1966e, f), *U.S. News and World Report* (1966b, c), *Business Week* (1966b) and *Time* (1966c, d), described uncontrolled dispersion of four unarmed nuclear weapons over the south-eastern coastal area of that country. Hubbell (1966), Morris (1966) and Lewis (1967) each compiled book-length accounts of the accident and subsequent events.

According to the above accounts, a mid-air collision was followed by an explosion. Seven crew members were killed in the accident and four nuclear weapons dropped to earth. Three were quickly found, two of which experienced a non-nuclear explosion, and Hubbell (1966), Morris (1966), Lewis (1967), *Time* (1966b, c), and *Business Week* (1966b) described scattering of their contents over a wide area. The fourth weapon was not easily located, and as described by *Time* (1966e), *Business Week* (1966a, b, c), *Life* (1966a), *Newsweek* (1966e, f), *Commonweal* (1966), *Saturday Review* (1967) and *Hawkins* (1966), subsequent land- and sea-search efforts involved large numbers of military and civilian personnel. *U.S. News and World Report* (1966c) reported that on 7 April 1966 the missing bomb was removed from the Mediterranean Sea. The land search was climaxed by removal of topsoil, sealing it in drums and shipping it to the United States for burial. The amount removed varies with the reporting source. *Time* (1966b) reported 1,600 tons; *U.S. News and World Report* (1966b), 4,900 barrels; *Saturday Evening Post* (1966) and *Radiological Health and Data Reports* (1966) each quoted the figure of 1,500 cubic yards. According to *Radiological Health and Data Reports* (1966): 'the earth and vegetation contain only small quantities of radioactive material scattered when the nuclear weapons impacted'.

FIELD PROBLEMS

Violation of integrity of two devices permitted limited dispersal of contents, and strong winds over the area enabled material to be spread over a larger area. Land-search operations for the weapons were somewhat impeded by the necessity for taking certain precautions in the event that radioactive material would be encountered. Therefore, routine use of survey instruments and personnel protective procedures were necessary during all phases of the search operation. The manpower requirements for the program were quickly met by assigning

* The opinions expressed in this paper are those of the authors and in no way represent official views of the United States Air Force.

personnel from these troops had detection instruments increased, mainly to conduct the in this duty even new contingent

As with all in or minimize corrosion, skin, equipment each individual study at the USA but the logistics sible. Opportunity a wide area, includes, initial sar ones were procur environment was

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personnel from adjacent areas to temporary duty in Spain. Only a very small percentage of these troops had had any experience with incidents of this nature or use of radioactive detection instruments. Initially, equipment shortages were acute, and later, as the supply increased, malfunction and breakage was very serious as inexperienced personnel attempted to conduct the necessary surveys. In order to lessen personal hardships, troops were rotated in this duty every two weeks, thus, any experience gained on the job was soon lost when a new contingent reported for duty.

As with all incidents of this nature, standard USAF procedures were followed to prevent or minimize contamination of personnel by radioactive material. Decontamination of clothes, skin, equipment, etc., was done under field conditions, and prior to departing from the area each individual was isolated for 12 hrs., during which time all urine output was collected for study at the USAF Radiological Health Laboratory. A 24-hr. urine specimen was preferred, but the logistics of the operation were such that isolation for longer than 12 hrs. was impossible. Opportunities for sample contamination were frequent. Strong winds spread dust over a wide area, including the base camp, troops did not always follow decontamination procedures, initial samples were collected in make-shift containers, and when more acceptable ones were procured from sources in Europe and the United States, their storage in a dust-free environment was not always possible.

All samples were flown to the laboratory for analysis. As of 1 March 1967 over 1,900 had been received, none were lost in transit, and only seven arrived with contents partially or totally lost because of leaky or broken containers. Average transit time was six days, which could have been reduced with higher shipping priorities. In addition, a few nasal swipes, water, soil, vegetation and miscellaneous biota were submitted for study. A small wedge of lung tissue was obtained at time of necropsy from an individual who participated in the early phases of the search operation and later died (28 October 1966) of heart disease.

LABORATORY PROCEDURES

Urine samples were considered in one of two categories; initial or resample. The initial were those usually collected at the site of operations and represented the first sample from any given individual.

Resamples were those collected usually several months after duty at the site, in containers provided by the USAF Radiological Health Laboratory, at the individual's permanent base or station, and under close medical supervision, to ensure the sample represented a complete 24-hr. output.

The initial urine samples were analyzed for alpha-emitting radionuclides, using a gross alpha procedure, the essential steps of which were:

1. Wet ashing of an aliquot of the urine sample with concentrated nitric acid and hydrogen-peroxide to a white ash.
2. Solubilizing the white ash and coprecipitation of plutonium with bismuth salts.
3. Dissolution with hydrochloric acid followed by the addition of lanthanum carrier before hydrofluoric acid precipitation.
4. Direct mounting of the precipitate on a 2" steel planchet.
5. Counting for 120 min. in an internal proportional counter.

Pooled normal urine samples were spiked with ^{239}Pu and processed in a like manner, to obtain data on chemical recovery of the procedure.

Early in the operation it was discovered that the exterior surfaces of the sample containers were contaminated with an alpha-emitting radioisotope. This immediately discredited the assumption that the alpha activity in the urine sample had, in all cases, been cycled through

a metabolic system. It was decided to continue screening all initial samples for gross alpha activity, and to assume that this activity represented disintegration of ^{239}Pu atoms. In those samples where the activity found suggested a systemic body burden of 10% or more of the maximum permissible, as recommended in *National Bureau of Standards Handbook 69* (1959) for this nuclide, a resampling program was conducted at the period of 90-150 days after collection of the initial sample.

Systemic body burdens were calculated, using Langham's (1956) formula for conditions of single, acute exposure. Since there was no way of knowing precisely when the exposure(s) may have occurred, the elapsed time between possible inhalation of isotope and collection of sample was taken from the median day of duty at the operation site to the date of sample collection. In this manner the greatest probable error was usually no more than 7-10 days.

Twelve-hour urine samples required use of certain assumptions when body burdens were calculated, since Langham's (1956) expression was based on a 24-hr. output. When the 12-hr. volume was less than 1.2 l, calculations were so adjusted as to express the total activity had the output been 1.2 l. In other words, an average 24-hr. urine output of 1.2 l for each individual was assumed. When the volume exceeded 1.2 l, the actual value for calculating systemic body burden was used. The use of 1.2 l as an average value was supported as a reasonable estimate by results of the resampling program (Table III) when a 24-hr. output was obtained.

In order to identify and quantitate the isotope of greatest interest, ^{239}Pu , a procedure specific for this nuclide, was adopted for analysis of all resamples. One-half of the total urine sample was adjusted to pH 2 with concentrated nitric acid. (The remaining one-half was saved until the analyses and counting were complete. Obtaining repeat samples from individuals several thousand miles removed, and with only passive interest in the problem, was a task far more difficult and uncertain in the event of laboratory error in processing, than retaining one-half as back-up should repeat or confirmatory studies be indicated.) A ^{236}Pu spike (approximately 4 dpm) was added to each sample in order to evaluate per cent recovery of the chemical procedure. The sample was then heated to boiling to break metabolic complex-bound plutonium. Coprecipitation of alkaline earth phosphates and plutonium was done by adjusting the urine sample to pH 10 with concentrated ammonium-hydroxide. The salts were dissolved in nitric acid and coprecipitated with radiochemically-pure cerium by adjusting to pH 4.5. This precipitate was dissolved in hydrochloric acid and passed through an anion-exchange (Bio-Rad AG2-X10) column which adsorbed plutonium. Interfering anions adsorbed on the column were removed by washing with hydrochloric acid. Hydriodic acid was used to elute the plutonium from the ion-exchange column. The evaporated column residue was heated in sulfuric acid to change the plutonium to the sulfate salt. After adjusting the pH of the solution to three, plutonium was electrodeposited on $1/2$ "-diameter steel planchets. A current of 300 milliamps was used for 180 min.

The lung tissue sample, after dissolution in 8N nitric acid, and addition of ^{236}Pu spike, was processed in a manner identical to the urine resamples.

Radioactive counting for initial samples was done with Nuclear Measurement Corporation Model PC-3A, windowless, gas-flow proportional counters. Daily checks were made on instrument performance by counting reference standards of ^{239}Pu , to ensure constancy of counting efficiency. Samples were counted for 120 min., and daily determinations of background radiation levels were made by counting for 720 min. These values ranged from 0.02 to 0.06 counts per min. Whenever the value approached 0.1 count per min., the chambers were vigorously cleansed.

Sample activity was calculated from the following expression:

$$\text{pCi/sample} = \frac{(\text{gross counts/gross ctg time}) - (\text{bkg counts/bkg ctg time})}{(\text{counting efficiency}) (2.22) (\text{procedural yield})}$$

Counting procedures for resamples involved use of solid-state surface-barrier detectors

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mounted in a vacuum chamber. Charge sensitive preamplifiers, designed and built by Mr. Robert L. Farr, of the Laboratory staff, were used to amplify signals from the detector. Output from the preamplifiers was fed to a Nuclear Data 130 AT multichannel analyser. 'Readout' from the analyser was in the form of typewriter printout.

Using an electroplated source containing known activities of ^{239}Pu and ^{236}Pu , instrument performance was checked each morning before beginning counting, and, normally, an additional time each afternoon. The performance check consisted of observing the peak channels for ^{239}Pu and ^{236}Pu , and adjusting the gain of the amplifier system, if necessary, to correct for any gain shifts. Additionally, the counting efficiency of the system was checked at the same time, to ensure constancy.

Background counts were made each night for 600 min. duration with a blank planchet in the counting chamber. The values ranged between 0.000 and 0.0025 counts per min. The daily background count also served as a check for any possible contamination in the counting chamber. Samples were routinely counted for 100 min.

The data was collected in an analyser memory of 127 storage positions. Total counts in two bands, centered on the peak channels of ^{239}Pu and ^{236}Pu , and each containing 11 storage locations, were totaled and used for the sample activity calculations. The same bands were used for both sample and background determinations. Sample activity was calculated from the following expression:

$$\text{pCi/sample} = \frac{(\text{net cpm in } ^{239}\text{Pu band}) \times (\text{dpm } ^{236}\text{Pu added})}{(\text{net cpm in } ^{236}\text{Pu band} \times (2.22))}$$

$$\text{where net cpm in } ^{239}\text{Pu band} = \left(\frac{\text{gross cts } ^{239}\text{Pu band}}{\text{gross ctg time}} - \frac{\text{bkg cts in } ^{239}\text{Pu band}}{\text{bkg ctg time}} \right)$$

$$\text{net cpm in } ^{236}\text{Pu band} = \left(\frac{\text{gross cts } ^{236}\text{Pu band}}{\text{gross ctg time}} - \frac{\text{bkg cts } ^{236}\text{Pu band}}{\text{bkg ctg time}} \right)$$

dpm ^{236}Pu added = activity of ^{236}Pu spike added to sample corrected for decay to date of count.

Water samples were processed and counted, using procedures described above for initial urine samples. Swipes taken from the surface of tomatoes were placed in the chamber of a Nuclear Measurement Corporation PC-3A windowless, gas-flow proportional counter, and gross alpha activity measured. Nasal swipes were taken with cotton wrapped around an applicator stick. The terminal 2" of the stick, containing the cotton, was cut off and directly dropped into a 19-ml solution of fluor.* Counting was accomplished in an automatic liquid scintillation counter (Packard Instrument Company Model 526). Results for all swipe samples were reported in terms of disintegrations per min., as control studies using known amounts of ^{239}Pu provided acceptable factors for efficiencies and self-absorption of alpha particles by the swipe.

Radioactivity in soil and on vegetation samples was readily detected by a survey instrument (Eberline Instrument Company Model PAC-1-S). Soil samples were sealed in polyethylene containers and a pulse height analysis of gamma emissions done, using a thin (0.005" beryllium) window NaI crystal and associated electronic equipment.

RESULTS

Chemical recovery of spiked samples was $75\% \pm 19$ (S.D.) using the gross alpha procedure; that for the resamples is shown in Table III.

* Prepared by dissolving 100 g naphthalene, 50 mg 1,4-bis-2 (5-phenyloxazolyl) benzene and 7 g 2,5-diphenyloxazole in 1 l of 1,4-p-dioxane.

Forty water samples were analysed, seven of which showed no detectable radioactivity.* In 33, the range of gross alpha activity was 0.1 to 633 picocuries per l. The median value for the series was 1.64 picocuries per l.

Seventy-eight swipes taken from the surface of as many tomatoes were processed, and, of this number, only 13 showed detectable alpha radioactivity. The range of results was from 0.1 to 4.3 picocuries per swipe.

No detectable radioactivity was found on 70 of 120 nasal swipes received. The range of results for the other 50 was 0.45 to 153 picocuries per swipe, the mean, standard deviation, and median value for this series was 11, 21.8 and 5.9 respectively.

In each of 23 soil samples, photopeaks at 16, 27 and 60 KeV energy were observed. In two of the group additional peaks at 110 and 185 keV appeared.

Vegetation samples showed high level of alpha activity with a survey meter. No further studies were accomplished.

Results of gross alpha analyses on initial urine samples are shown in Table I. In Table II the results of studies on resamples are displayed. Table III is a summary of statistics on factors of importance in relation to the results of Table II.

The lung tissue sample weighed 7.9 grams (wet), and contained 2.8 picocuries of ^{239}Pu . Total weight of the lungs was 950 grams. Table IV gives a complete history of bioassay studies on this individual prior to death.

TABLE I

Initial urine samples — Alpha activity (expressed as percentage of one systemic body burden)

| | Air Force | Army | Navy | Other | Total |
|---|-----------|------|------|-------|-------|
| Number analyzed | 1404 | 107 | 37 | 38 | 1586 |
| BB ¹ greater 100% ² | 19 | 1 | 0 | 0 | 20 |
| BB 0.99 to 0.09 | 375 | 33 | 5 | 8 | 422 |
| BB 0.09 to 0.009 | 487 | 23 | 20 | 7 | 537 |
| BB less than 0.009 | 522 | 50 | 12 | 23 | 607 |

¹ Systemic body burden, bone, critical organ — calculated on the basis of urinary excretion according to expression $D = 435 U t^{0.78}$ (where D = Systemic body burden; U = ^{239}Pu activity in 24-hr. sample; t = time in days from exposure to sampling).

² Value of 0.044 μCi ^{239}Pu for D represents one body burden or 100%.

DISCUSSION

Most of the water samples showing detectable levels of alpha activity were obtained from personnel decontamination shower effluent. The remainder were grab samples from the Mediterranean Sea.

Wipes from tomato surfaces showed less activity than suggested by reports in *Time* (1966c), *Life* (1966b), and *New York Times* (1966), if one assumes that the tomatoes wiped were a representative sample of the entire crop under condemnation.

* No detectable activity (NDA) means that the result of the analysis was below the minimum detectable activity (MDA) of the particular counting equipment utilized. (MDA is defined as the sample activity with an associated counting error at the 95% confidence level equal to 0.95 times the sample activity.) The exact value of the MDA is a function of the background counting level and can be obtained from the Laboratory upon request.

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Nasal wipes were not utilized to any significant degree, and over half of those submitted had no detectable activity. In theory, a wipe sample from the external nares, having radioactivity on subsequent analyses, provides excellent presumptive evidence of exposure to airborne nuclides. Depending on the amount of activity detected, applicable individuals could be promptly removed from hazardous areas. Urine samples from these individuals

TABLE II

Urine resampling program — ²³⁹Pu (expressed as percentage of one systemic body burden)

| | Air Force | Army | Navy | Other | Total |
|-----------------------------|-----------|------|------|-------|-------|
| BB ¹ greater 10% | 6 | 0 | 0 | 0 | 6 |
| BB 1 to 10% | 195 | 13 | 5 | 0 | 213 |
| BB less 1% | 26 | 11 | 1 | 1 | 39 |
| BB zero | 148 | 9 | 1 | 6 | 164 |
| Total | 375 | 33 | 7 | 7 | 422 |

¹ BB defined as systemic body burden, bone, critical organ. Calculations as explained in Table I.

TABLE III

Statistical review of selected factors for groups showing body burden of 1% and greater upon resampling (219 samples)

| | Mean | SD | Mode | Median | Range |
|---|------|-----|------|--------|---------|
| ²³⁹ Pu (curies × 10 ⁻¹⁵) | 93 | 114 | 29 | 66 | 11-1030 |
| ²³⁹ Pu spike (% recovery) | 77 | 15 | 61 | 76 | 43-113 |
| 24-hr. sample volume (liters) | 1.3 | 0.5 | 1.1 | 1.2 | 29-3.6 |
| Elapsed time (days) ¹ | 178 | 77 | 140 | 140 | 65-396 |
| BB (%) ² | 4 | 4 | 3 | 3 | 1-67 |

¹ Estimate of time between probable exposure and collection of sample.

² Body burden as defined in Table I.

TABLE IV

Bioassay and tissue studies - Case HPA 95-66

| Material | Date collected | Quantity | Procedure | Results (picocurie/sample) |
|----------|----------------|-------------|--------------------------------|----------------------------|
| Urine | ca 1/20/66 | 400 ml | gross alpha | 248 |
| Urine | ca 1/29/66 | 1650 ml | gross alpha | 9.9 |
| Urine | 1/31/66 | 4270 ml | gross alpha | 7.2 |
| Urine | 2/28/66 | 2520 ml | gross alpha | NDA |
| Lung | 10/28/66 | 7.8 g (wet) | ²³⁹ Pu (alpha spec) | 2.78 ± 0.16 |

Note: Probable date(s) of exposure 17-20 January 1966.

could then be obtained in a contamination-free environment. However, in practice, a nasal wipe is not always reliable. The procedure must be done properly to be of value. Often the cotton tip is not inserted into the nares sufficiently to contact all mucous surfaces. Many individuals are very sensitive to nasal probing, and if the corpsman is of lesser rank than the patient, and timid, the entire procedure is likely to be little more than a sham. A further complication is the natural tendency for individuals to insure a clean nose just prior to the wipe. Such blowing action may remove a significant amount of radioactivity.

Lacking information on the temporal relationship between collection of urine sample and taking the nasal swipe, it was not possible to obtain a significant correlation between activity on the swipe and that in the urine from the same individual.

Presence of radioactive material in soil and vegetation samples was established with ease. More detailed procedures to quantitate the radioactivity were not possible since there was no information as to how much surface area was involved with each type sample. Weight and volume factors would have no meaning since surface contamination was the point in question. The photopeaks in soil samples suggested the presence of ^{239}Pu , ^{241}Am and ^{235}U .

Systemic body burdens were calculated from bioassay data to permit officials in the field a basis for decision-making. While reports in terms of disintegrations per min. per 24-hr. sample would have been of equal value, the body burden term was easier for line and certain medical service officers to understand, and it would suggest a degree of possible systemic deposition at whatever interval of time elapsed between exposure and date of sample collection; thus, if officials decided to use, for example, 100% as a value for action, a report of 50% body burden could be readily understood by all concerned. The limitations of assumptions upon which calculations of body burdens were based were well realized, but in an operational, real-world, urgent situation, some number had to be provided for responsible officials, and no other suggested procedure appeared to provide greater utility.

Personnel whose body burdens exceeded 100% on the basis of assay of initial urine sample (Table I) were immediately removed from the area and resampled in a rigidly-controlled environment. In all cases, subsequent studies gave much lower values or no detectable activity (NDA).

Sheehan's (1966) unpublished data from occupational inhalation exposures of plutonium suggests that urinary excretion of the isotope reaches a peak between 120-160 days post-exposure. The resampling program was so organized as to obtain a specimen during this period. Results shown in Table III indicate that this objective was met. Even though urinary excretion may reach its peak some 150 days after a single acute inhalation exposure, this does not suggest that earlier studies are without value. Yet, urine samples taken immediately after an acute inhalation exposure are unlikely to give reliable data other than the fact that plutonium is or is not present. Langham (1956) suggests that between 5-10% of an inhaled dose is rapidly solubilized and passes into the blood stream. Morrow *et al.* (1967) studying dogs, reached a similar conclusion and showed that excretion rates during the first one to three days post-exposure are virtually unrelated to administered dose and body burden. During field operations, urine sample collection should be deferred until probability of contamination is at a minimum. However, the nasal wipe, properly taken, may be used as a screening procedure, and urine sampling restricted to selected individuals during the operation, and required of all at conclusion of the incident. Fecal samples, while offering some advantages in assessing the magnitude of inhalation exposure of plutonium, present problems of collection and contamination, particularly from several hundred individuals working under field conditions, which loom as nearly insurmountable.

Based on results of the resampling program, a one-year follow-up program is in operation to resample at intervals of two months for one year, the 25 individuals showing the highest systemic body burdens. The values range from 7% to 67% of one permissible body burden in this group.

As of 1 March 1967, 21 individuals remain to be studied; 34 have not submitted an initial

sample, appropriate to submit a resample. The laboratory has many individuals who have left the area and are quite difficult to locate.

While the total body burden based on the entire population was due to contamination, it was not possible to support the data which have shown initial high values than a control.

The total body burden of ^{239}Pu is not valid if the man. Morrow's exposure, for a phragmatic area from ^{239}Pu occurred within five picocuries is about 0.15% of what Welfare (1967) reported.

Department of Health and Human Services, much pulmonary disease in Spain on the basis of correlation.

All information available for the bioassay.

Bioassay exposures were the many individuals were taken to the facility for an incident. 20% have been identified, or number, or Provisions and study.

Based on inhalation exposure demonstrated.

sample, apparently by-passing control procedures set up at the operation site: 16 have yet to submit a resample, and 11 more are being sought for a second sample as the first was lost in the laboratory during processing because of broken glassware and procedural errors. Since many individuals have been reassigned as often as three times subsequent to duty in Spain, or have left the service, the associated administrative problems in completing this program are quite difficult.

While the data available for correlating urinary excretion of ^{239}Pu and subsequent lung burden based on tissue analyses is very meager, it does provide reason for continued study of the entire problem. Initially, it was assumed that the higher urine value found in this case was due to contamination, and the greatly reduced levels in subsequent urine samples tended to support the thesis. However, as Morrow *et al.* (1967), Swanberg (1962) and Snyder (1962) have shown, urine excretion levels have little or no relation to body and lung burden. The initial high levels seen in this case could have represented the rapidly mobilized PuO_2 rather than a contaminant.

The total lung burden extrapolated from this extremely small sample approaches 0.5 nanocurie, assuming a homogenous distribution. However, such an assumption is probably not valid if work on dogs is to be considered relevant to accidental inhalation exposures of man. Morrow *et al.* (1967), using 'lightly anesthetized dogs' during a controlled inhalation exposure, found that the right lung contained more of the isotope than the left, and diaphragmatic areas more than those more cephalad. There is no information regarding the area from which the eight-gram sample was taken.

^{239}Pu occurs naturally in the lungs and pulmonary lymph nodes of humans. Morrow (1965) related work of European observers who estimated that the standard man has inhaled about five picocuries to date, and that the highest alpha activity reported in the average human lung is about one picocurie per g. Pulmonary lymph nodes contain about three picocuries per g, 15% of which is ^{239}Pu . In the United States, the Department of Health, Education and Welfare (1966) found an average of 0.43 picocuries of plutonium per kg of lung in adults.

Department of Defense pathologists have been alerted to the desirability of collecting as much pulmonary and thoracic lymphatic tissue as possible from all individuals who worked in Spain on this project and have come to autopsy. This material will be studied in an attempt to correlate lung deposition with urinary excretion.

All information collected on this project has been placed on keypunch cards, and is readily available for recall and manipulation by electronic data-processing equipment. The file on the bioassay support of the Palomares incident is permanent.

SUMMARY

Bioassay experiences associated with the Palomares nuclear accident indicate that, in spite of the many handicaps of field operations, personnel protection and decontamination procedures were effective. The exercise demonstrated that modern communication and transportation facilities permit one well-equipped and staffed laboratory to provide adequate support for an incident of this nature anywhere in the world. Of nearly 1,600 participants, less than 20% have a systemic body burden of plutonium detectable by urinary bioassay, and of this number, only 25 showed a value in the range of 7-67% of one permissible body burden. Provisions have been made for long-term follow-up on the group of 25 as well as collection and study of autopsy material as it becomes available.

Based on available methods for estimation of systemic body burden of ^{239}Pu following an inhalation exposure, not one individual who participated in the Palomares operation has demonstrated systemic retention exceeding the maximum permissible amount.

REFERENCES

- AZANCOT, L. (1967): The day H-bombs fell on Palomares. *Saturday Review*, 50, 21.
Business Week (1966a): Navy dives deep for a lost H-bomb. 4, 72.
Business Week (1966b): The navy presses deep-sea probes. 4, 72.
Commonweal (1966): The missing bomb., 83, 549.
HAWKINS, P. (1966): How we recovered the missing H-bomb. *Family Weekly*, p. 5.
HUBBELL, J. G. (1966): The case of the missing H-bomb. *Reader's Digest*, 89, 239.
LANGHAM, W. H. (1956): Determination of internally deposited radioactive isotopes from excretion analysis. *Amer. industr. Hyg. Ass. Quart.*, 17, 305.
LEWIS, F. (1967): *One of our H-bombs is missing*. McGraw-Hill, New York, N.Y.
Life (1966a): Little subs big score: The H-bomb is found. 60, 34.
Life (1966b): The case of the missing H-bomb. 60, 106.
MORRIS, C. (1966): *The Day They Lost the H-bomb*. Coward McCann, Inc., New York, N.Y.
MORROW, P. E. (1965): Personal communication.
MORROW, P. E., GIBB, F. R., DAVIES, H., MITOLA, J., WOOD, D., WRAIGHT, N. and CAMPBELL, H. S. (1967): The retention and fate of inhaled plutonium dioxide in dogs. *Health Phys.*, 13, 113.
National Bureau of Standards Handbook 69 (1959): Maximum permissible body burdens and maximum permissible concentrations of radionuclides in air and in water for occupational exposure. Superintendent of Documents, U.S. Government Printing Office, Washington, D.C.
Newsweek (1966a): Still looking. 67, 33.
Newsweek (1966b): Enter the poodle. 67, 55.
Newsweek (1966c): Deep mystery. 67, 61.
Newsweek (1966d): Is it or isn't it? 67, 47.
Newsweek (1966e): An H-bomb is missing and the hunt goes on. 67, 55.
Newsweek (1966f): Sigh of relief. 67, 62.
New York Times, Magazine Section (1966): Palomares learns to love the bomb. p. 22.
Northeastern Radiological Health Laboratory (1967): Annual Report-Year 66. No. *NERHL 67-1*, Winchester, Mass.
Radiological Health and Data Reports (1966): Investigations of weapons carrier accident in Spain. 7, 205.
Saturday Evening Post (1967): Is poison safe? 239, 102.
SHEEHAN, W. (1966): Personal communication.
SWANBERG, F. JR. (1962): Comparison of urinary excretion data from selected plutonium exposure cases at Hanford. *Health Phys.*, 8, 761.
SNYDER, W. S. (1962): Major sources of error in interpreting urinalysis data to estimate the body burden of ²³⁹Pu; A preliminary study. *Health Phys.*, 8, 767.
Time (1966a): Dunderball. 87, 33.
Time (1966b): La bomba recuperada! 87, 27.
Time (1966c): Nuke fluke. 87, 37.
Time (1966d): How they found the bomb. 87, 53.
Time (1966e): Spain — Rough sea for Charlie. 87, 33.
U.S. News and World Report (1966a): Mystery of the missing H-bomb. 60, 10.
U.S. News and World Report (1966b): H-bombs on the loose — what about the dangers? 60, 66.
U.S. News and World Report (1966c): The rescue and unveiling of an H-bomb. 60, 11.

DISCUSSION

BAIR: Are you suggesting the possibility that some individuals might have appreciable lung burdens?

ODLAND: There might be. We are looking for whole body counters to check some of these people. Our understanding is that what is in the urine does not reflect what is in the lung, so we made no mention of what is in the lung.

Certainly, the one case we have evidence of, an individual who died, had about 500 picocuries in the lung and in the last urine sample we had no detectable activity. Now this

may represent exposure to the excreted solubilized knowing.

DUNCAN: This occurs to very little the biological a series of based on.

ODLAND: I'm over the

DUNCAN: Yes

ODLAND: We field. The patient, but earth.

DUNCAN: On samples; useful.

ODLAND: I have amount of

POCHIN: At

WALD: I would levels and

ODLAND: Not other than laborator

may represent what Dr. Langham, Dr. Snyder and others mentioned - that in inhalation exposure you get rapidly solubilized fractions. It comes in pretty quick and then later on the excretion is reduced. What we thought was contamination, may have been the rapidly solubilized fraction we were looking at and really not contamination. There is no way of knowing.

DUNCAN: There is something that must be said in defense of urine analysis. One thought occurs to me about this follow up question. If the result is to mean anything at all, it is very little use getting people in every two months. I don't know what your experience in the biological variation of these results have been, but wouldn't it possibly be better to get a series of samples? I think there is a criticism of the interpretation of the urine analysis based on just one point.

ODLAND: I might say we don't get them in. They are located at 25 separate and distinct spots over the earth.

DUNCAN: You get the contents in.

ODLAND: We get the contents in. It is quite a job, though, to get the urine samples in the field. The physicians in the hospitals have difficulty getting urine samples from a bed patient, but try it with an individual who is free to go wherever he wants on the face of the earth.

DUNCAN: Our population is not quite as mobile as that, but we find difficulty in getting urine samples; getting five sequential samples even with longer sampling intervals is much more useful.

ODLAND: I agree. It would be much better. But there are other problems we in the military have that in civil life is not so burdensome. If we find anyone who excretes a constant amount that we consider significant, we will bring him in for more intense study.

POCHIN: Are there any other questions?

WALD: I wonder if you could relate these body burdens to field conditions of contamination levels and air concentrations?

ODLAND: No, absolutely not. I was not in Spain. I don't know any of the field conditions, other than what I read in *Time*, *Newsweek* and *Life*. All I know is what happened in the laboratory.