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EXECUTIVE SUMMARY

A nuclear weapons accident occurred on January 17, 1966 over Palomares, Spain when a United States Air Force (USAF) B-52 bomber and an USAF KC-135 tanker aircraft collided. That accident led to the release of four thermonuclear weapons. The accident damaged two of the weapons with release of radioactive contamination, leading to a three-month response effort to identify, characterize, remove, and remediate the accident site. During the response effort, some personnel were exposed to airborne dust and debris contaminated with plutonium.

Radiation monitoring efforts during the response were limited to the evaluation of exposures and their possible effects on health using principles and methods accepted at that time. However, recent interest in radiation exposure to veterans and government employees, as well as the availability of improved technology for assessing doses led the Air Force to review the data for possible use in estimating radiation exposures.

Initial Exposure Evaluation

The response effort began on the evening of January 17. A base of operations (Camp Wilson) was established, and measurements for released plutonium began on January 18. The response force peaked at about 680 U.S. personnel on January 31, and then gradually fell until the effort ceased on April 11. Approximately 1,600 personnel participated during the operation.

Response personnel provided urine and nasal swab samples while on site to assess possible intakes of plutonium and the potential effects on health. The sample results were evaluated in terms of guidelines available at the time.

The assessment program concluded that of the nearly 1,600 participants, less than 20% showed levels of plutonium in their bodies that could be detected in urine samples. Only 26 personnel showed values of 7% to 67% of the upper limit for plutonium in the body (Odland 1968a). Those 26 were followed up for a period of 18 to 24 months following the accident. A 1968 Air Force review of the follow-up program concluded that no additional information could be gained from continued sampling and recommended that further sampling effort be suspended.

Exposure and Dose Updates

The evaluations conducted during 1966 through 1968 depended on the limited understanding of plutonium's behavior under field conditions. Since then, advances in that understanding and in methods for assessing dose provided an opportunity to reexamine the monitoring data. The approach uses the concept of Committed Effective Dose Equivalent (CEDE) - a cumulative dose, weighted for the contributions of individual organs, and summed over a 50-year period - as an indicator of possible risk from the exposure. Comparisons can also be made to the annual limit on intake (ALI) of 20,000 picocuries (NRC 2000), to the 21 rem from cumulative exposure to average background radiation over 70 years, or to the 50 rem guideline for cumulative dose to workers (1 rem per year over 50 years of work).

During the project, computer programs that perform the necessary intake and dose calculations were tested. Two programs (CINDY and LUDEP) were selected because intakes they estimated agreed to within a factor of two for the majority of the test cases. That agreement was judged reasonable and acceptable for this assessment.

Available Records

The initial urine sampling that began within three days of the accident experienced some problems such as sampling for less than the desired 24-hour period, possible sample contamination from dust spread by strong winds, and use of non-clinical sample containers. Follow-up sampling was conducted on personnel with initial urine results indicating retained plutonium at 10% of the maximum permissible body burden (MPBB) or more. This second phase was implemented to assess whether sample contamination may have produced spurious urine levels, indicating a false-positive exposure.

Most of the cases involved samples collected on site that were assayed once for gross alpha radioactivity. The remaining cases involving samples collected on site were either resampled, or reanalyzed using alpha spectrometry. Finally, 26 cases were resampled for 18 to 24 months.

Analysis of all the data produced the following four groups.

- A High 26 Cases Group that included the 26 individuals who were resampled for 18 to 24 months after the initial phase of sampling in 1966.
- A Repeat Analysis Cases Group that contained 54 individuals who either had submitted initial samples that were reanalyzed using more sensitive methods (alpha spectrometry), or who were resampled.
- A Contamination Cutoff Cases Group that included 313 individuals with results that were below an assumed cutoff level of 0.1 pCi per day.
- A Remaining Cases Group that contained 1,063 individuals with records that were not otherwise evaluated because their data indicated contamination from collection on site.

Environmental measurements obtained in the Palomares vicinity for over 15 years following the accident provided a basis for preparing independent estimates of intake and dose using representative scenarios for response force activities.

Results

The CEDEs estimated from urinary bioassay were judged unrealistically high when compared with estimates prepared for other plutonium exposure cases – persons residing in the Palomares vicinity and Manhattan Project workers. The estimates of plutonium intake and CEDE from inhalation using environmental data measured in Palomares ranged up to no more than about 0.2 rem. Consequently, the estimates from urine analyses are not useful as representative intakes and doses. The detailed evaluations performed for the High 26, Repeat Analysis and Contamination Cutoff Cases represent preliminary estimates that cannot be considered as definitive. Follow-up studies are required to develop credible estimates of dose that are compatible with those calculated from environmental data.

Conclusions

Preliminary results calculated for all 26 individuals in the High 26 Cases Group, the 54 individuals in the Repeat Analysis Cases Group, and the 313 individuals in the Contamination Cutoff Cases Group proved unrealistically high. They are inconsistent with those calculated from environmental data and when compared with the experience from exposed workers. Furthermore, the urine results are inconsistent with plutonium's known behavior and are inadequate by themselves to support meaningful intake and dose evaluations without confirmatory studies, such as analysis of urine samples now using very sensitive instrumentation,

detailed review of participant medical records, participant interviews, and comprehensive assessments based on sound environmental measurements.

Recommendations

Several future actions should be considered to further refine these initial estimates.

1. Additional effort is needed to reconcile the estimated intakes and doses derived from the urinary bioassay data with the estimates from environmental measurements. A targeted effort that includes participant activities, participant interviews, urine and other appropriate plutonium analyses using current techniques, medical records review, and modeling should be considered.
2. The results of this effort should be communicated to responders, veterans organizations, and other interested parties using appropriate information that clearly confirms the conclusions of the original medical evaluation program, recognizes the difficulties in preparing updated intake and dose estimates, and outlines the options for strengthening the estimates.
3. Further contacts with the Department of Energy for comparison with evaluations of their personnel who responded to this accident could provide useful data. The effort should be summarized in a companion document that conveys the details of the project and its potential effects on health in an easily understood manner. That document should be made available to any of the responders who desire a copy.

1 INTRODUCTION

LABAT-ANDERSON INCORPORATED was awarded TASK ORDER Number TO 799BG0031 under General Services Administration Contract GS-35F-4813G to provide services to the Air Force Medical Operations Agency for evaluating the radiation exposure records of personnel who responded to past nuclear weapons accidents and incidents for the purpose of updating dose estimates. The Task Order specified the following objectives:

- To identify, locate and review the records of the incident, radiation exposure assessments, and other information pertinent to the study.
- To evaluate current methods and models for estimating radiation doses and risks from the intake of radioactive materials contained in nuclear weapons.
- To recommend a methodology for conducting the re-evaluation of the available radiation exposure information.
- To evaluate any and all radiation exposure information, such as urine bioassays, nasal swabs, air sampling information, etc. for scientific soundness and possible use in updating the radiation records of the response personnel.
- To perform the update and prepare records for input to the Air Force Master Radiation Exposure Registry.

The Task Order did not specify extensive searches of personnel records, or efforts to locate and contact the personnel involved except on a limited basis where specific information might be useful or when individuals expressed interest in the project.

The Task Order also required that the effort should begin with the nuclear weapons accident of January 17, 1966 over Palomares, Spain involving a United States Air Force (USAF) B-52 bomber and a USAF KC-135 tanker aircraft. That accident involved a mid-air collision between the two aircraft, the release of four thermonuclear weapons, damage to two of the weapons with release of radioactive components, and a three-month response effort to identify, characterize, remove, and remediate the accident site. During the response effort, personnel were exposed to airborne dust and debris contaminated with plutonium.

Substantial response efforts provided a foundation for evaluating the potential radiation effects from the exposure using accepted principles and methods of the time. However, heightened interest in radiation exposure within the Department of Energy and veterans of the 1991 Gulf War led to this effort to review the data and update radiation exposures, wherever possible, using current methods and procedures.

This report provides the results of the efforts conducted under this Task Order and includes a review of the accident details and radiation assessment efforts and results in Section 2; and a summary of the environmental measurements and review of the radiation assessment data from 1966 through 1968, an evaluation of its accuracy and usefulness, and efforts to prepare the data for re-assessment of radiation doses in Section 3. Section 4 provides a summary of radiation effects and dosimetry methods. Section 5 discusses the methods and results of preparing estimates from environmental data. Section 6 summarizes the methods and results for preparing estimates from the urinary bioassay results. Section 7 discusses the results, assesses the

implications of the results on health, and Section 8 concludes with a summary and recommendations for further evaluations of the responders to this accident.

2 BACKGROUND

At 10:30 a.m. (local time), on January 17, 1966, a U.S. Air Force B-52 bomber and a USAF KC-135 tanker collided during a refueling operation at 9.44 km (31,000 ft.) over the southeastern coast of Spain (DNA 1975). The incident released four thermonuclear weapons that fell to earth near the small coastal hamlet of Palomares, Spain. Serious damage to two of the weapons caused dispersion of their contents over a limited area. Strong winds contributed to further spread of the material and contaminated aircraft debris to the village, surrounding lands, and agricultural crops (Odland 1968a). The response to the incident to find, safeguard, recover, and return weapons contents to the United States, and to assess and mitigate effects on the local populace required significant effort involving hundreds of personnel for almost three months.

Responding personnel encountered the contaminated debris, lands, village, and crops. Although emergency protection measures were followed, responders and local citizens were exposed to the plutonium dispersed from the two weapons. Extensive efforts assessed the effects of those exposures on US military and civilian responders during a program that went on for two years after the incident. Soon after the accident, the Government of Spain represented by the Spanish Junta de Energia Nuclear (JEN) and the Government of the United States, represented at the time by the Atomic Energy Commission (now the Department of Energy), agreed to cooperative programs for extensive follow-up studies of the site and surrounding areas (DOE 2001). Those studies have produced significant understanding about the characteristics of the residual plutonium, its environmental distribution, resuspension into the air, and migration through the soil and other pathways; as well as estimates of the radiation doses to the local populace and evaluations of their health condition.

This section provides additional details about the accident itself, discusses the nature of the response, reviews the methods, procedures and operation of the health and safety assessment program, and reviews the results and limitations of the assessment.

2.1 ACCIDENT SUMMARY

Both aircraft were destroyed in the air. Four thermonuclear weapons, 11 men (four survived), and hundreds of tons of debris fell to earth in and around the *barriada* (Hamlet) of Palomares. Parts of the aircraft were scattered over a wide area generally between Cuevas de Almanzora and Vera along the Mediterranean Sea between Puerto Rey and Villaricos. At that time Palomares had no telephones and did not appear on maps of the area. The population of the time was estimated to be about 1200.

The first of the four nuclear weapons was found intact with its primary chute deployed on the evening of January 17, 1966 just east of Palomares. A radiation survey showed that no radioactivity escaped the weapon. The area was designated impact point 1.

The primary chutes did not open for two other weapons whose chemical high explosives detonated. One weapon was found on the morning of January 18, 1966 about one mile west of the village (impact point 2). The third weapon was found about two hours later on the eastern edge of Palomares (impact point 3) with high explosive and radioactive material scattered by

impact and explosions. The fourth weapon was finally recovered intact from the Mediterranean Sea on April 7, 1966 (Odland 1968a).

The explosions and fires around impact points 2 and 3 produced airborne clouds of plutonium-containing dust that were carried over some distance by 30 knot winds. Eventually, a total of 558 acres of soil contaminated above 5.4 micrograms per square meter ($\mu\text{g}/\text{m}^2$) were remediated by removal or plowing. These levels provided many opportunities for responders to inhale or ingest the radioactive plutonium.

2.2 RESPONSE SUMMARY

The Guardia Civil, the first representatives of the Spanish government, arrived on site about one hour after the accident. They immediately took charge, secured the accident site and informed both Spanish and American authorities. The commander of the 16th Air Force headquartered at Torrejon Air Base near Madrid and the Strategic Air Command Headquarters at the Offutt Air Force Base in Omaha, Nebraska were notified and the "Broken Arrow " response system was initiated. The commander and three staff members surveyed the accident site from the air and arrived at San Javier (195 km from Palomares) at 1:30 p.m.

OPERATION RECOVERY was initiated by deciding to bring personnel in from two Spanish bases, Moron, and Torrejon. Movement of personnel started at 0100Z on January 18 from Moron with a second convoy at 0310Z. 126 personnel were transported in six buses. The first of two convoys from Torrejon departed at 0137Z, the second at 0202Z, with 175 persons in six buses. Following a 12 to 14 hour drive to the southern coast, the first of buses arrived at 1300Z and the last arrived about 1700Z.

Another Disaster Control Team from Offutt Air Force Base in Omaha Nebraska arrived at the accident scene at 7:30 am on January 18. Members of the Joint Nuclear Accident Coordinating Center (JNACC), Sandia Corporation, and the Los Alamos Scientific Laboratory (LASL) left Albuquerque at 1800 GMT on January 17.

By the evening of January 17, 49 U.S. personnel were on site. Camp Wilson was established as a general headquarters, and measurements for released plutonium began on January 18. About 300 more airmen from the Moron and Torrejon air bases were on site by the evening of January 18. A maximum of about 680 U.S. personnel were at Camp Wilson on January 31.

By January 21, the camp moved to leveled, higher ground some 5.6 km east of the Garrucha where it remained until April 3. A helicopter pad, motor pool, and 75 tents were on firmer ground in less danger of flooding. The camp was moved again where it remained until closure on April 11.

Manning reached a peak by January 31, with 598 Air Force, 64 Army, and 19 Navy. All except some officers were housed at the camp. Those were quartered in two hotels close to the accident scene. Personnel involved with search, recovery, and decontamination generally rotated through the camp at two-week intervals. Population at the camp varied, but from the high on January 31, there was a gradual reduction until the camp closed on April 11. The first major reduction occurred on February 9 and 10 when about 50 of the clean-up personnel and the 40-man ordnance disposal team left. A slight upswing occurred from March 11 to 17 during the period of filling of 4,810 barrels with contaminated soil and crops. Other personnel at camp included 126

Guardia Civil and 39 Spanish personnel who worked in the cleanup and other activities. Overall, almost 1,600 personnel participated in the response effort at one time or another.

Response activities included performing radiation surveys, protection, and recovery of nuclear weapons, development of remediation plans, and decontamination of affected areas. These will not be discussed in this report. However, details of the efforts to assess and control radiation exposure are of vital importance to this effort and are described next.

2.3 SUMMARY OF HEALTH ASSESSMENT ACTIVITIES

This accident represented one of the first times that plutonium had been dispersed on and around civilian property outside the United States. Furthermore, the response placed a significant number of military and civilian personnel resources at risk. Procedures for assessing and controlling contamination from the materials in these weapons were available and used. However, there were many questions about the behavior of inhaled and ingested plutonium under field conditions.

2.3.1 On-Site Sampling

Urine sampling, recognized as a reasonable method for assessing exposure to plutonium, was begun within three days of the accident. Urine sample collection on site was subject to collection of less than the desired 24-hour specimen and possible sample contamination. Samples were shipped by the most expedient means to the USAF Radiological Health Laboratory (USAF RHL) at Wright-Patterson AFB, Ohio for analysis. Two sampling phases were used – an initial phase and a resample phase.

2.3.2 Interpretation of Urine Results

The results were evaluated in terms of the maximum permissible body burden (MPBB, see Appendix A) of ^{239}Pu as recommended by the National Bureau of Standards (NBS) in Handbook 69 (NBS 1959). The NBS recommendations were based in part on Publication 2 of the International Commission on Radiation Protection, *Recommendations of the International Commission on Radiological Protection, Report of Committee II on Permissible Dose for Internal Radiation*, published in 1959 (ICRP 1960). The MPBB for ^{239}Pu considers the bone as the “critical organ” or the organ that is most susceptible to radiation from plutonium and is the basis for developing protection limits. The body burden is defined as that portion of ^{239}Pu distributed by systemic circulation. It does not include that amount fixed in the lungs. The MPBB was 0.044 microcurie (μCi) of ^{239}Pu .

The MPBB was developed as an operational tool for limiting dose to a critical organ over a working lifetime. The dosimetry model used assumed uniform deposition of the radionuclide in the organ, energy emitted equals energy absorbed, and the characteristics of the model could be represented by “Standard Man” data. The concept was designed to provide adequate protection over a 50-year working lifetime and as such applied to continuous intake of radionuclides over the entire period. Thus for a material like plutonium, the limit would allow for continuous intake for 50 years while keeping the dose to the bone (the critical organ) below the limit.

An individual’s body burden was estimated from the measured urinary gross alpha radioactivity for initial samples. The following equation was used, taken from Langham (Langham 1956):

$$D_r = 435 U t^{0.76}$$

where:

- D_r = retained systemic body burden (pCi or Bq); meaning the amount retained in the body "t" days after exposure
 U = ^{239}Pu activity (pCi or Bq) in a 24-hour sample
 t = time in days from exposure to sampling

The analysis required assumptions about the type of exposure (acute or continuous), and about whether samples represented true 24-hour urine outputs. This calculation applies to a single acute exposure. The individuals responding to the incident were generally on site for two weeks, some more and some less. Others remained for almost the entire period of operations. The beginning date for the exposure was assumed as the midpoint of time an individual arrived on site until ceasing activities (departing). Odland (Odland 1968b) reported that "When the 12-hour volume was less than 1.2 L, calculations were so adjusted as to express the total activity had the output been 1.2 L. When the volume exceeded 1.2 L, the actual value for calculating systemic body burden was used."

2.3.3 Resampling Program

The Air Force conducted a resampling program at 90 to 150 days after collection of the initial sample. This resampling applied to individuals whose gross alpha results for initial samples suggested a systemic body burden of 10% or more of the MPBB.

The program established procedures to identify and quantify the isotope of interest in the urine – ^{239}Pu .

2.4 SUMMARY OF RESULTS

Odland reported that the USAF RHL processed almost 1600 urine samples during the initial phase (Odland 1968a). Table 1 gives the distribution of the samples in relation to the systemic body burdens they represent. Those results indicate that 20 individuals potentially exceeded the MPBB and 442 samples exceeded 10% of the MPBB and required resampling. However, the possibility for contamination of the initial samples collected on site introduced uncertainty about that conclusion. This potential for sample contamination in and around Palomares was also recognized by the Spanish Junta de Energia Nuclear (JEN), which transferred urine sample collection and medical examination of local residents from Palomares to Madrid in 1967 (Iranzo 1987).

Table 1. Initial Urine Samples (Percentage of one MPBB).

| | |
|----------------------|------|
| Number Analyzed | 1586 |
| BB greater than 100% | 20 |
| BB 9% to 99% | 422 |
| BB 0.9% to 9% | 537 |
| BB less than 0.9% | 607 |

A resampling program began shortly after on-site operations ended. Originally, samples were desired at two-month intervals; however, this became impractical and samples were collected primarily at the discretion of the individuals. Table 2 contains the results of the resampling program (Odland 1968a). The laboratory processed 422 samples during the resampling phase. Of those, only six exceeded 10% of the MPBB with slightly less than half of those resampled (203) showing results below 1% of the MPBB.

Table 2. Urine Resampling Program Results.

| | |
|---------------------|-----|
| BB greater than 10% | 6 |
| BB 1 to 10% | 213 |
| BB less than 1% | 39 |
| BB zero | 164 |
| Total | 422 |

A small specimen of lung tissue, obtained at time of necropsy from an early responder who died from heart disease, contained 2.8 pCi of ^{239}Pu ; or about 0.00034 microcuries (about 2% of estimated maximum permissible lung burden) when extrapolated to the total mass of the lung. Early urine analyses for the individual indicated a rapid decrease in gross alpha radioactivity that was attributed to contamination. However, early behavior of inhaled plutonium was not excluded as a possibility (Odland 1968a). Nevertheless, if correct, the quantity in the lung of this individual represents a small fraction of the MPBB after 9 months following exposure.

In summary, the assessment program indicated that of the nearly 1,600 participants, less than 20% indicated systemic body burdens of plutonium that could be detected by urine bioassay, and only 25 showed values in the range 7% to 67% of the MPBB guideline (Odland 1968a). Those 25 and one additional individual were followed up for a period of 18 to 24 months following the accident.

2.5 PLUTONIUM DEPOSITION REGISTRY BOARD

The Air Force recognized that the consequences of possible exposure to plutonium from the Palomares Broken Arrow required in-depth and credible assessment, provisions for long-term maintenance of the records, and possible follow-up of those exposed. To satisfy that need, representatives of the U.S. Air Force Medical Service met in Omaha, Nebraska in March 1966 and identified the need for a detailed and long-range program to provide follow-up and treatment, when required. The concept of a special board to satisfy those needs was developed into a Plutonium Deposition Registry and Board with the following purposes as stated in the proceedings of the first meeting (Odland 1966):

- (1) It would provide adequate follow-up of personnel with internal deposition of plutonium, in order that any possible biological injury would be detected at the earliest date, and it would provide, when required, the best possible treatment to reduce body burdens of Plutonium-239.
- (2) It would provide the government with complete factual data upon which to evaluate claims for compensation that might subsequently arise.

- (3) It would provide the medical profession with additional urgently needed data with which to manage medical problems at future Broken Arrows or laboratory accidents of a similar nature.

The Plutonium Deposition Registry Board met first on October 26-28, 1966 (Odland 1966) to establish the Board; to review progress to date, and to set policy for further follow-up. The Board reflected a tri-service nature as well as an interagency flavor with participation by the Atomic Energy Commission, the Veterans Administration, and the Defense Atomic Support Agency. Additionally, several recognized experts in plutonium medical effects participated as Board Members or as Consultants (Odland 1966). Board deliberations produced recommendations in the following areas:

- Samples should be collected from all that departed the accident scene without submitting a sample, or whose initial samples suggested a systemic body burden greater than 9%.
- No further sampling of individuals whose **initial** urine results suggested a systemic body burden of less than 9%.
- Sampling should be continued on members whose results on resampling were in the top 10% of the resampling group and showed systemic body burdens of 1-10%.

The Board also discussed the use of whole-body counting as an additional assessment tool and the use of ^{239}Pu to ^{241}Am ratios in the weapon components, soil and urine as possible method for determining ^{239}Pu in the lungs; however, no specific recommendations were developed.

On January 16, 1968, the Air Force Logistics Command Surgeon issued a letter report that reviewed progress of the follow-up effort (Wallace 1968). The report summarized the results of resampling of the 26 individuals whose initial urine samples showed the highest ^{239}Pu content suggesting systemic body burdens of 7% to 67%. The report concluded that little additional information could be gained from continuing the effort. Finally, the report announced that the Surgeon General of the Air Force had concurred with canceling the Board meeting scheduled for 1967 and that further activities would be limited to analyzing tissue specimens, as they became available. As a practical matter, this letter report suspended activities of the Board in the matter of the Palomares accident. Research during this project identified no evidence of additional testing efforts or results.

Our review of the urinary levels reported during the assessments conducted in 1966 and 1967 indicated that the initial intakes could exceed the current annual limit on intake (ALI) recommended by the ICRP (ICRP 1979). Consequently, a repeat evaluation of the urinary data seemed warranted to provide assessments using currently accepted methods for analysis and management of radiation risks. The remainder of this report discusses the detailed approach for performing those assessments.

3 ASSESSMENT OF AVAILABLE DATA

The response effort discussed in Section 2 above included a health evaluation program that generated records of the possible doses to those who responded to the accident. Locating those records involved contacts with the Air Force Medical Operations Agency (AFMOA) at Bolling AFB, DC and the Air Force Institute for Environmental, Safety and Occupational Health Risk Analysis (AFIERA) at Brooks AFB, TX. Those records required detailed review to understand

the data they contained and the processes that produced the data; an analysis of the consistency and reliability of the contents; and possible adjustments to estimate intake and dose equivalent.

In addition, the Government of Spain, in collaboration with the U.S. Department of Energy, has conducted extensive studies of the environmental characteristics of the residual contamination in the Palomares area. In particular, air sampling, particle size characteristics, and resuspension factors have been determined from data collected for more than 15 years. These data provide a valuable source for independent intake and dose estimates.

3.1 ENVIRONMENTAL DATA

Studies of the environment around Palomares have included air sampling at four locations, and estimates of the resuspension of plutonium particles from the surface into the air for subsequent inhalation by the local populace. Those studies used air samplers placed in four locations representing possible sources of plutonium. Samplers were located near the impact points of the two destroyed weapons, at another contaminated area, and in the town of Palomares. From 1966 to 1980, the highest annual average air concentration was measured at 11.9 fCi/m³ (442 μBq/m³) in 1967. The highest average for a weekly measurement period occurred in March 1967 with a concentration of 292 fCi/m³ (10.8 mBq/m³) (Iranzo 1987). Measurements during other periods were lower than these, but demonstrated some variation over time.

Studies at Palomares have also estimated the resuspension of plutonium at and around Palomares from the same air sampling data combined with knowledge of the plutonium surface contamination levels. Resuspension is a process that represents the air concentration of a material above a surface contaminated with the same material. The resuspension factor (in units of m⁻¹) is the ratio of the air concentration (expressed in units of pCi/ m³ or Bq/m³) to the surface contamination (in units of pCi/ m² or Bq/m²). The studies at Palomares indicate that the resuspension factors initially were 10⁻⁷ m⁻¹ initially, dropped to values on order of 10⁻⁹ m⁻¹ months later, and to 10⁻⁹ m⁻¹ to 10⁻¹⁰ m⁻¹ after several years (Iranzo 1994). The air concentrations were determined in areas where the surface contamination ranged from 3.2 μCi/m² (0.118 MBq/m²) to 32 μCi/m² (1.18 MBq/m²).

Both the air sampling and the resuspension results represent credible efforts that can be used as the basis for estimates of intake and dose.

3.2 AIR FORCE BIOASSAY DATA

During the initial contact, AFIERA and AFMOA provided records in the form of:

- Air Force Forms with laboratory analytical and exposure details of the nasal swipe and urine samples submitted and processed.
- Complete case files for the 26 individuals identified for follow-up in 1966 and commonly referred to as the "High 26".
- A Microsoft Excel spreadsheet prepared by AFIERA staff that contained the data from those Air Force Forms, and some data related specifically to the 26 individuals (referred to as the "High 26" who were considered as having the highest exposures).
- Copies of the accident response reports, USAF RHL documents on the evaluation of exposures by urinalysis, and selected publications from journals and conference proceedings.

Appendix B contains a detailed discussion of the information collected, an evaluation of the information's suitability for a dose evaluation, and adjustments made to the data for performing intake and dose calculations. The record prepared and maintained by the Air Force consisted of forms, computer spreadsheets, and written correspondence and reports of activities.

The data were evaluated to assess the availability of the elements required by the internal dosimetry models. Review indicated that the exposure date or dates, sample date, and results were not completely recorded for all cases. Substantial numbers of samples lacked one or more important pieces of data. Data forms for 115 individuals apparently represented a repeat analysis of a sample or a follow-up sample for an individual. Sample collection proceeded for only 12 hours for many samples collected at Camp Wilson, indicating a correction to 24 hours would be needed. Our review indicated that 12-hour samples were clearly designated in only 42 of the samples. Lacking any other recorded information, sample volumes were assumed to represent 24-hour output unless specifically designated as 12-hour samples.

Urine sampling, begun within three days of the accident, was subject to several compromises, including: collection limited to 12 hours or less for operational requirements; sample contamination from strong winds; non-uniform decontamination procedures; make-shift sample containers, and frequently contaminated storage areas.

Records for 122 nasal swab reviewed indicated that only 13 contained a result (8 were 0 pCi, 4 had values all below 1.5 pCi, and 1 was reported as NDA). Therefore, the nasal swab records were not used in this analysis.

The majority of available records contained results from the gross alpha method on samples collected on site. Most of the records for samples collected on site raised serious questions about estimates derived from them. Records for the 26 individuals in follow-up contained multiple samples collected up to two years after the incident. Unfortunately, the pattern of results for samples collected during the resampling phase often did not follow the pattern expected for Class Y (Type S) plutonium. However, treatment of the records for the 26 served as the model for the other cases. A second group of records contained repeated analyses using the more sensitive alpha spectrometry and provided a reasonably well-defined set of cases for analysis. These two groups were designated the High 26 Group and the Repeat Analysis Group, respectively. Appendix E provides additional details of the bioassay data evaluation and grouping of cases.

The remaining results generally fell into two categories: those with the results of some resampling; and those with one sample and often very high results. Careful review of the group of data indicated that processing all of the cases would produce unrealistic estimates that would be based on potentially contaminated samples. Gross alpha results from samples collected on site produced intake estimates and doses that seemed unreasonably high. Contamination of samples collected at the accident site continued to impact the evaluation as it did at the time of the accident. However, review of those data also indicated a substantial number of cases with urinary results that were essentially below the detection limit or were quite low. Their data were reviewed again to determine whether a reasonable lower cutoff could be determined. Analysis of the processes (Appendix E) supported a cutoff limit at 0.1 pCi/day for gross alpha activity. This was similar to the detection limit of 0.74 mBq/d (0.02 pCi/d) used in studies by the Government of Spain from 1966 to 1985 (Iranzo 1987). Consequently, 0.1 pCi/day was selected as a cutoff limit, and cases in that category were designated the Contamination Cutoff Group.

Applying a cutoff to urinary excretion to individual cases does not precisely impact all samples equally. A fixed value for the cutoff concentrations produces higher estimated intakes and correspondingly higher dose equivalent values for samples taken at longer times after the exposure, especially for Class Y (Type S) plutonium.

After applying the cutoff, 1,219 samples for 1,063 individuals had urine concentrations above 0.1 pCi/d that were classified in the Remaining Cases Group. These were not evaluated further.

4 RADIATION EFFECTS AND DOSIMETRY METHODS

Responders to the Palomares accident encountered sources of possible exposure from plutonium-contaminated aircraft debris, contaminated lands, and agricultural crops, and dust produced by winds. Evaluation of the potential radiation effects requires estimates of the exposure and associated radiation dose, and comparison with knowledge about the effects of radiation on human health. Furthermore, these evaluations must take into account current knowledge and apply accepted methods for estimating the radiation exposure and dose. The approach to accomplishing these estimates is guided by recommendations of both international and national scientific bodies concerned with radiological protection. These bodies, primarily the International Commission on Radiological Protection (ICRP) have published recommendations on the relevant guidelines for limiting radiation effects and exposure, and estimating doses from radioactive materials that may enter the body, as plutonium does.

This section summarizes the current understanding of radiation effects, in general, and plutonium, specifically, on health, and the guidelines to protect workers and the public from those effects. It also summarizes updated internal dosimetry methods relevant to evaluating plutonium exposures.

4.1 SUMMARY OF RADIATION EFFECTS

This study of exposure to plutonium at Palomares and calculation of possible doses to internal organs raises questions about the possible health effects that may be associated with them. This section provides a brief summary of our understanding of the possible health effects from ionizing radiation and plutonium in particular, some of the guidelines for limiting exposure to it, and some basic information about the possibility that a certain dose could cause some kind of effect on health.

4.1.1 *General Radiation Effects*

In discussing health effects relating to ionizing radiation, the term “dose” is used. “Dose” comes from the early medical use of x-rays, much as a dose of medicine is measured in grains or ounces. It refers to the amount of radiation energy absorbed by an organ, tissue, or cells, measured in rem (or Sv). Today, the average American receives a dose of 0.3 rem (0.003 Sv) every year from natural sources—radioactive materials in rocks and soil, cosmic radiation, radon, and radioactivity in our bodies. Over a 70-year lifetime, the cumulative background dose averages 21 rem (0.21 Sv). In some areas of the world, people receive much higher doses from background radiation. For example, in areas of India and Brazil the ground is covered with monazite sand, a radioactive ore. Radiation exposure rates there are many times the average background levels elsewhere. People who live in these areas receive doses of up to about 0.7 rem

(0.007 Sv) each year from the gamma radiation alone (NAS 1990). These levels combined with the other sources of background radiation (cosmic rays, radon, etc.), cause average doses that are about three times more than the U.S. average. Yet these people show no unusual rates of cancer or other diseases linked to radiation.

The effects of ionizing radiation can be categorized as either prompt or delayed, based on the time frame in which the effects are observed. Prompt effects, like rapid death, occur when high doses are received in a short period of hours to weeks. Delayed effects, such as cancer, can occur when the combination of dose and dose rate is too small to cause prompt effects. Both animal experiments and human exposures to high levels of radiation show that ionizing radiation can cause some cancers (NAS 1990). All of the observed effects of ionizing radiation in humans occur at relatively high doses. At the low doses that are of interest to radiation workers and the general public (that is, below a few rem), studies to date are inconclusive (NAS 1990). Although adverse health effects have not been observed at low doses, the carcinogenic nature of ionizing radiation makes it wise to limit the dose.

For low-doses, there are no conclusive data that relate dose to health effects or showing a threshold, or minimum, level for cancer. Because of this, experts who study radiation effects have decided that the results from high-dose, high-dose-rate studies must be used to control the low-dose, low-dose-rates experienced by workers and the public. A convenient way to do this is to assume that no effects occur at zero dose. In addition, since the rate at which effects occur is extrapolated from higher doses, it is also assumed that the effect increases linearly with dose. These two assumptions are known as the “linear-dose-response, non-threshold” (LNT) hypothesis. This implies that the same number of additional cancers would occur from exposing 100 persons to 100 rem (1 Sv), or 10 thousand persons to 1 rem (0.01 Sv), or 10 million persons to 0.001 rem (0.00001 Sv). No prompt effects have ever been reliably observed in humans below about 10 rem (0.1 Sv). Reports from the Japanese atomic bomb survivor studies conclude that the location and reality of such a threshold, if one does exist, are difficult to assess. Nevertheless, the Health Physics Society (HPS 1996) has stated that “Below 10 rem (which includes occupational and environmental exposures), risk of health effects are either too small to be observed or are non-existent.”

Within the first 30 years after the discovery of x-rays, standards were developed for the measurement of radiation. At about the same time, acceptable levels of dose were set. The first level, known as the ‘tolerance dose’, or that amount of radiation that could be tolerated, was set at one-tenth of a unit (about 0.1 rem (0.001 Sv) in today’s units) per day for 300 days a year, which amounts to 30 rem (0.3 Sv) in a year.

From World War II to the early 1980s, radiation dose limits were adjusted downward in response to increased concern about radiation effects, the increased uses of radiation, and because improved radiation protection technologies appeared. The National Council on Radiation Protection and Measurements (NCRP, established in the 1930s) developed the recommended changes for the United States. During that time, the dose limit was reduced from three-tenths of a rem in a six-day period in 1946 to 5 rem (0.05 Sv) per year in the mid-1950s. In addition, a limit for the public was set at one-tenth of the worker limit to provide an additional margin of safety.

Research does not show a clear threshold dose for cancers from radiation, so the small risk per person at low doses had to be considered in relation to the large number of workers who were receiving those doses (NCRP 1993b).

The NCRP adopted three radiation protection principles: (a) no practice shall be carried out unless it produces a positive net benefit (sometimes called justification); (b) all exposures shall be kept as low as reasonably achievable (ALARA), economic and social factors being taken into account (called optimization); and (c) the dose equivalent to individuals shall not exceed the recommended limits (called limitation). These principles work together to protect against both prompt and delayed effects in large groups of workers and the public.

In 1993, the NCRP released a new set of national recommendations based on International Commission on Radiation Protection's 1990 recommendations. Those limits for non-threshold effects differ slightly from the earlier recommendations: 50 rem (0.5 Sv) per year to any tissue or organ and 15 rem (0.15 Sv) to the lens of the eye to avoid cataract formation. The recommended occupational limits on whole-body doses (total effective dose equivalent), first set at 5 rem (0.05 Sv) per year in 1958, are now set at no more than 5 rem (0.05 Sv) in any one year and a lifetime average of no more than 1 rem (0.01 Sv) per year (NCRP 1993).

Occupational radiation exposure limits for federal agencies are currently established in "Radiation Protection Guidance to Federal Agencies for Occupational Exposure," 52FR 1717, signed by President Reagan on January 20, 1987. The Nuclear Regulatory Commission implemented that guidance in its regulations on radiation protection (Title 10, Code of Federal Regulations, Part 20). These limits apply to all licensed uses of radioactive material under NRC's jurisdiction. Similarly, other Federal agencies as a matter of policy and directive, including the DoD in DODI 6055.8, Occupational Radiation Protection Program, also observe this guidance.

The current established protection standards are (NRC 1999):

- 5 rem in a year for workers (to protect against cancer).
- 50 rem in a year for workers to any organ (to protect against threshold effects, such as radiation burns, etc.).
- 50 rem in a year to the skin or to any extremity.
- 15 rem in a year to the lens of the eye (to protect against cataracts).
- 0.1 rem in a year (70-year lifetime) for members of the public.

These limits are in addition to the radiation doses a person normally receives from natural background, medical testing and treatment, and other sources.

The protection standards mentioned above provide regulatory guidelines to be used primarily for designing radiation protection programs and facilities. Their intent is to limit dose to a worker so that risk is limited to levels that are similar to so-called "safe industries." Limits for the public perform the same purpose but generally include additional margins of safety to account for a wider range of ages (childhood to aged), more diverse health condition, and individual sensitivities. Their primary purpose is to prevent exposures that are associated with risks exceeding the established guides.

These guidelines also offer usable comparisons for evaluating the possible effects of exposures. For example, the occupational limit of 5 rem (0.05 Sv) in a year provides one such value. Since 5 rem (0.05 Sv) represents an acceptable risk, any exposure below 5 rem (0.05 Sv) should be considered acceptable. NCRP recommends that the average dose equivalent per year for workers should not be more than 1 rem (0.01 Sv) a year over 50 years or work. That is the same as 50 rem (0.5 Sv) in 50 years. Therefore, 50 rem (0.5 Sv) provides a reasonable guide for an exposure

from radioactive materials in the body, such as plutonium. Since these guides are set with margins of safety, receiving a higher dose does not mean that one will be harmed. However, it would mean that further evaluation might be needed to determine whether the exposure was a one-time incident or one that could recur.

An alternate approach to evaluating the possible effects of an exposure considers the possibility that an exposure will lead to health effects, such as cancer or hereditary effects. The NCRP has provided risk factors for the probability that a certain dose equivalent from radiation will cause an effect. Those factors for workers are 0.0004 per rem (0.04 per Sv) for fatal cancer, 0.00008 per rem (0.008 per Sv) for non-fatal cancer, and 0.00008 per rem (0.008 per Sv) for hereditary disorders for a total of 0.00056 per rem (0.056 per Sv) (NCRP 1993a). For members of the entire population, these factors are 0.0005 per rem (0.05 per Sv) for fatal cancer, 0.0001 per rem (0.01 per Sv) for non-fatal cancer and 0.00013 per rem (0.013 per Sv) for hereditary disorders, for a total of 0.00073 per rem (0.073 per Sv).

4.1.2 Health Effects of Plutonium

Plutonium, discovered in 1941, is radioactive and can be dangerous when it gets into the human body. Some have even referred to plutonium as “the most toxic substance known to man”. Early concerns about the health risks of plutonium arose from knowledge of the effects of radium, discovered by Marie Curie in 1899. With its half-life of 1620 years, radium-226 presents an intense and constant radiation source for hundreds of years. Early uses of radium exposed workers to significant doses with acute cases ending in rapid death, and lower exposures leading to infections of the jawbones, pathological bone fractures, or cancers of the bone.

The National Bureau of Standards addressed radium’s dangers by developing an occupational standard for radium, adopted in May 1941, about two months before the discovery of plutonium. Scientists on the Manhattan Project then recognized the potential hazards of plutonium, which is similar to radium. They estimated that plutonium would be roughly as dangerous as radium when comparing equal masses.

Plutonium gives off alpha particles that produce heavy ionization and give up their energy more quickly than the lighter beta particles, or x-rays and gamma rays. In air, alphas travel only 3 to 5 centimeters and in living tissue only about 30 micrometers. That distance is less than the thinnest part of the dead layer of external skin cells (called the epidermis), or the thickness of a piece of paper (about 100 micrometers). Because of this low penetrating power, materials that give off alpha particles present no hazard when kept outside the body.

Unfortunately, when they get inside the body, alpha emitters come into very close contact with the body tissues and irradiate cells. Plutonium can be inhaled, ingested, or passed into the blood stream through a wound. When that happens, about 90 percent eventually goes to the lung, liver, or bones.

The half-life of plutonium-239 is 24,065 years. This half-life is short enough that 1 microgram of material will undergo more than 2000 decay events per second, but it is long enough to allow that microgram to decay at an approximately constant rate for thousands of years.

No one has ever died from an acute plutonium uptake. But, researchers have estimated lethal doses from studies on dogs, rats, and mice, which indicate that a few milligrams of plutonium per kilogram of tissue is a lethal dose. Extrapolated to humans, an intravenous injection of about

22 milligrams into an average human (70 kilograms; about 154 pounds) would be lethal within about 30 days to half the people exposed. Inhalation would require about four times more or 88 milligrams.

Recognizing the similarity of plutonium to radium, scientists worked to develop exposure standards that would limit the risks to workers, especially on the important war-time effort of developing a plutonium-implosion bomb. Beginning in 1945, those efforts have evolved into a set of radiation protection recommendations that have received international acceptance. In 1977, the ICRP published major revisions in those recommendations that based radiation protection for plutonium on dose rather than deposition in the body. Those recommendations, known as ICRP 30, have been largely adopted in the United States. In 1991, the ICRP published new recommendations (ICRP 60), which reduced the recommended annual occupational limit to 2 rem (20 millisieverts) per year. Thus far, these recommendations have not been adopted in the United States, however, they are considered in most radiation protection assessments.

Plutonium absorption in the body depends mainly on the plutonium compound and how it enters the body. The body generally absorbs the soluble forms (nitrates, citrates, and certain oxides) more readily than insoluble forms. Plutonium absorption through intact skin is usually quite low, but deposits in tissues through puncture wounds, cuts, and somewhat less through skin burns. Soluble plutonium begins movement throughout the body within minutes or hours of the uptake and may move to the lymph nodes near the wound; remaining for years. Some insoluble plutonium gets into the blood circulation quickly, but most remain at the site and are slowly redistributed over weeks and months. About 90 percent of the systemic burden deposits in the liver and bones. The kidneys excrete plutonium in urine that represents the concentration of the plutonium in the blood making plutonium measurements in urine a convenient indicator of plutonium in the body.

Ingesting plutonium is perhaps the least likely means for plutonium to enter the body. But even if plutonium is ingested, the gastrointestinal tract provides a natural barrier, and in adults only about 0.05 percent of the soluble plutonium compounds and a mere 0.001 percent of the insoluble ones enter the blood stream. The rest of the plutonium simply moves out of the body in feces.

Inhalation of plutonium dust provides the most likely entry route for plutonium. Particle size affects plutonium absorption. Smaller particles are more likely to be retained. Particles over 10 micrometers in diameter (considered large) are filtered out in the nose and upper respiratory region, swallowed, and eventually passed out through the gastrointestinal tract. Particles less than 10 micrometers in diameter (called respirable particles), deposit on the mucus layer of the bronchial tubes. Through a process, known as lung clearance, hair-like structures of the lining (called cilia) transport the mucus layer and dust particles up to the throat, removing much of the foreign material deposited in the bronchial tubes.

Smaller particles, especially those under 1 micrometer in diameter, are carried down into the tiniest airways of the lung and into alveoli (also known as air sacs). These structures have no effective lung-clearance mechanisms, but scavenger cells called phagocytes, engulf the inhaled plutonium particles, and transport them into lymph nodes or into lung tissues.

Autopsy studies reveal that, initially, plutonium is mostly deposited on the bone surfaces. Less than 5 percent of the plutonium is typically found within the bone marrow. Based on this this pattern of deposition, the primary carcinogenic risk from plutonium in the skeleton is bone

cancer. There is no conclusive evidence that plutonium increases the risk for leukemia, which is the unchecked proliferation of certain blood cells produced in the bone marrow.

Plutonium in the bone remains there for a very long time, gradually being redistributed throughout the bone. Current models (based on observation of exposed persons and autopsy data) estimate a half time of about 50 years for plutonium retention.

The plutonium deposited in the liver is eventually transformed from relatively soluble forms in hepatic cells into insoluble forms (hemosiderin deposits), which are sequestered in the cells that form the linings of liver ducts (reticuloendothelial cells). The retention half time for the plutonium deposited in the liver is approximately 20 years.

To date, there have been only few epidemiological studies of workers exposed to plutonium. Studies of workers at Los Alamos National Laboratory (Wiggs 1994) and Rocky Flats (Wilkinson 1987) are the only ones in the United States to have used quantitative measurements of plutonium exposures, but they involved few workers: 303 at Los Alamos and 1450 at Rocky Flats. These two studies showed no evidence of statistically increased rates of lung, liver, and bone cancers, which are shown in animal experiments to be the highest-risk cancers due to plutonium exposure. Another study (Reyes 1984) indicates that an increased brain-cancer rate in Rocky Flats workers was not caused by plutonium exposure or external radiation.

A study (Voelz 1983) involving 224 males exposed to plutonium between 1944 and 1974 who had plutonium deposition greater than 0.16 microgram (0.01 microcurie) found no cases of bone or liver cancer. By 1980, the final year of the study, only one person had died of lung cancer indicating risks were much lower than predicted by some nuclear-industry critics. Another study looked at 26 chemists, metallurgists, and technicians at Los Alamos, who were accidentally exposed to plutonium between 1944 and 1946. Their plutonium body burdens ranged from 50 Bq to 3,180 Bq when estimated by analysis of their urine (Voelz 1997). Interestingly, the mortality rate of these men has been lower than that of the population in general, and in 1996, 19 of them were still living.

Of those who are no longer alive, one died of lung cancer in 1989, at the age of 66, and two died of prostate cancer and congestive heart failure, respectively, but both had lung cancer at the time of death. All three men were very heavy smokers. Significantly, three cases of lung cancer are consistent with the national cancer incidence rate, over the same period, in U.S. white males of the same age. Another subject, who had an estimated plutonium deposition of 0.245 microgram, developed a rare bone cancer 43 years after exposure and died in 1990. This finding is statistically significant for the small group of 26, but in the Los Alamos study (Wiggs 1994) of 303 workers, this same individual remained the only one to have developed bone cancer. That one death from bone cancer in this larger group could well be due to chance and is not statistically significant. Finally, three more died of causes unrelated to cancer.

Overall, data from the several studies of persons exposed to low levels of plutonium radiation in the United States do not show a relationship between dose and effect. They merely indicate that such a relationship does not exist or cannot be confirmed. If plutonium is harmful at these low levels, its health risks are so small that, given the small number of workers involved, epidemiological methods cannot differentiate between effects triggered by plutonium radiation and variations in a group of people unexposed to such radiation.

Although studies on plutonium workers in the United States did not demonstrate the risk from plutonium radiation, there are data from much higher doses to which Russian plutonium workers have been exposed. Russian scientists have recently published two studies (Tokarskaya et al.1997, Koshurnikova et al.1998) of workers who had been exposed to plutonium at the Mayak Plant. The authors demonstrate that an increased risk for lung cancer is associated with higher exposures. Although both studies investigate this risk on many of the same workers, their conclusions about the relationship between dose and risk are different.

In one study, (Koshurnikova 1998) analyzed data from a cohort of 1479 workers who had been exposed to high doses of various types of radiation, including plutonium radiation, between 1948 and 1993. The control group was composed of 3333 other workers at Mayak who had also been exposed to radiation but within occupational limits. The study found a linear relationship between lung doses from 0.5 to 30 sieverts (or 50 to 3000 rem) and standardized mortality ratios. While this result found no threshold for effects, the trend of increasing rates with increasing dose is impressive.

The second study (Tokarskaya 1997) found a nonlinear threshold relationship between dose and lung cancer risk in a case-control study devoted to 162 plutonium workers who developed lung cancer between 1966 and 1991 and a control group of 338 Mayak workers who, during the same period, did not. The authors found no lung cancer risk up to a threshold dose of 16 sieverts, corresponding to about 1.6 micrograms of plutonium deposited. Above this threshold value, however, the risk rises rapidly. The two Russian studies are very different in the dose response relationships reported. However, the data demonstrate that lung cancer risk does indeed increase with higher doses.

A recently reported study to estimate the mortality risk per unit dose from exposure to plutonium produced results that compare well with estimates derived by other workers. This study developed the estimates using four independent approaches – epidemiologic studies of workers exposed to plutonium; epidemiologic studies of persons exposed to low-LET radiation combined with a relative biological effectiveness factor (RBE) for alpha particles appropriate to the cancer site; epidemiologic studies of persons exposed to alpha-emitting radionuclides other than plutonium; and controlled studies of animals exposed to plutonium and other alpha-emitting radionuclides extrapolated to humans (Grogan 2001). That work reported mortality risk per unit dose of 0.13 per Gy for lung, 0.057 per Gy for liver, 0.0013 per Gy for bone, and 0.013 per Gy for bone marrow (leukemia). Calculations of the risk for a unit intake compared well with estimates prepared by other workers.

It has been almost six decades since plutonium was first made. No doubt, the dangers of plutonium are real. However, plutonium has been handled in different chemical forms, fabricated as a metal, machined, and used successfully primarily because standards and procedures were established early. Because of this, there has been no instance of acute death from taking plutonium into the body.

4.2 REVIEW OF INTERNAL DOSIMETRY METHODS

Exposure to radiation can occur from sources of penetrating radiation outside the body, such as x-ray machines or industrial radiography sources, or from sources of radioactive materials, such as plutonium or uranium, that enter the body, locate in an internal organ or organs, and irradiate the tissues of those internal organs. The problem of calculating the dose depends on many factors

such as the shape of the organ, the type of radiation, the amount of the deposit, and the distribution of the deposit. Each of these individual factors is subject to considerable variability and difficulty in determining accurately. Once a dose is calculated, effectively communicating the possible effect of the dose on health requires additional skill and effort.

The current approach to limiting radiation exposure in the United States is derived from recommendations in ICRP Publications 26 and 30. The ICRP approach uses the concept of Committed Effective Dose Equivalent (CEDE) - a cumulative dose, weighted for the contributions of individual organs, and summed over a 50-year period for workers. Quantities derived from the CEDE such as the Annual Limit on Intake (ALI) and the Derived Air Concentration (DAC) provide operational limits for workers so that the overall guidelines will not be exceeded. The ALI is the activity of a radionuclide that would irradiate a person to the limit set by the ICRP for each year of occupational exposure. The DAC is found by dividing the ALI by the volume of air inhaled ($2,400 \text{ m}^3$) in a working year (2,000 hours) (ICRP 1979).

For internal exposures, determining the dose requires knowledge of the following questions:

- How does the material get into the body?
- Once in the body, how quickly does the material move to other organs?
- Does the material in the initial organ leave the organ or does some remain?
- Once in an organ, how does the material irradiate the organ and other organs?
- Once in an organ, how does the material move to other organs?
- Finally, how does is the material eliminated from the body if at all?

Answers to these provide the basis for developing an approach to calculate the dose to organs, the effective dose equivalent to the body, and interpreting the effects of the dose.

4.2.1 Internal Dosimetry Methods

The methods for estimating organ dose from internal radionuclides have evolved since radioactive materials were discovered and used. Until 1979, ICRP Publication 2 provided the guidelines and methodology. In 1979, ICRP Publications 26 and 30 changed the basic approach to limiting radiation, and for internal radionuclides in particular. ICRP Publications 54, 60 and 66 provided revised recommendations and updated models on the behavior of radionuclides in the body.

ICRP-2 assumed that a single organ could be considered the critical organ; that the organ retention could be represented by a single exponential term; that the physical characteristics, such as intake parameters, transfer functions, and tissue size and weight, could be represented by “Standard Man” data; that organs could be assumed to be spherical; and that scattered radiation could be ignored. Intakes of radionuclides were controlled by limiting “Maximum Permissible Concentration” (MPC) values in air and water for workers so that the annual dose limit to the critical organ would not be exceeded.

ICRP Publication 26 revised the system of dose limitation to one based on risk. This approach acknowledged the availability of sufficient information about the effects of radiation to estimate risk for fatal cancer from a unit dose equivalent in exposed people and in the risk of serious disease to offspring of exposed people. The basic recommendations addressed both stochastic

effects and non-stochastic effects. For stochastic effects, such as cancer and hereditary effects, risks are assumed to be directly related to dose equivalent with no threshold, meaning that the probability of the effect occurring, rather than the severity, is related to the dose equivalent. The severity of non-stochastic effects, such as cataracts and erythema, varies with dose, usually above a threshold or minimum dose.

ICRP Publication 30 provided revised dosimetry models that assume organ retention is represented by one or more exponential expressions, the critical organ concept no longer applies, the dose in an organ must consider radiation emitted by other organs in the body, and the physical characteristics are represented by "Reference Man" data in ICRP Publication 23 (ICRP 1975).

Under the revised system, dose equivalent limits are intended to prevent non-stochastic effects and to limit stochastic effects to acceptable levels. To meet this end, an annual occupational limit of 50 rem (0.5 Sv) to any organ was established (ICRP 1979). For stochastic effects, the limit on risk is the same whether the whole body is irradiated or organs are non-uniformly irradiated. This is accomplished by assigning organ weighting factors, w_T , that represent the ratio of the risk for the effect in an organ to the risk for whole body irradiation. The limit on risk to the whole body – called committed effective dose equivalent (CEDE) is then determined by summing the contributions for each irradiated organ and is limited to 5 rem (0.05 Sv). The committed dose equivalent (CDE) is the total dose equivalent averaged over a tissue (T) in the 50 years following intake and is limited to 50 rem (0.5 Sv). Table 3 contains the organ weighting factors from ICRP-30.

The dosimetry model calculates the absorbed dose averaged over the organ mass during 50 years following intake. It considers each radiation type and applies a radiation weighting factor, sometimes called the quality factor, which has the following value:

- Q=1 for beta particles, electrons and all electromagnetic radiation.
- Q=10 for fission neutrons emitted in spontaneous fission and protons.
- Q=20 for alpha particles from nuclear transformations, for heavy recoil particles, and for fission fragments.

Table 3. ICRP-30 Tissue weighting factors, w_T (ICRP 1979).

| Tissue | Weighting Factor, w_T |
|---|-------------------------|
| Gonads | 0.25 |
| Red Marrow | 0.12 |
| Lung | 0.12 |
| Breast | 0.15 |
| Thyroid | 0.03 |
| Bone Surface | 0.03 |
| Remainder | 0.30 |
| 0.06 for the organs with the five highest dose. | |

The ICRP further refined its basic recommendations and updated certain models for the respiratory tract and the biokinetics of deposited materials. The ICRP's 1990 recommendations

(ICRP 1991) provide weighting factors for tissues that were part of the remainder in the 1979 recommendations of ICRP-26 (ICRP 1979). Table 4 compares the tissue weighting factors of ICRP-26 and ICRP-60 and include a reduction in the bone surface and breast factors by three times, a 67 percent increase in the thyroid factor, and assignment of factors for additional organs, including the skin of the whole body.

Table 4. Tissue Weighting Factors (ICRP 1991).

| Tissue or organ | ICRP Recommendations | |
|-----------------|----------------------|------------------|
| | 1979 | 1990 |
| Gonads | 0.25 | 0.20 |
| Red Marrow | 0.12 | 0.12 |
| Colon | | 0.12 |
| Lung | 0.12 | 0.12 |
| Stomach | | 0.12 |
| Bladder | | 0.05 |
| Breast | 0.15 | 0.05 |
| Liver | | 0.05 |
| Esophagus | | 0.05 |
| Thyroid | 0.03 | 0.05 |
| Skin | | 0.01 |
| Bone Surface | 0.03 | 0.01 |
| Remainder | 30 ¹ | .05 ² |

¹ A value of 0.06 is applicable to each of the five remaining organs or tissues receiving the highest equivalent doses.

² The remainder is composed of the following tissues or organs: adrenals, brain, small intestine, kidney, muscle, pancreas, spleen, thymus and uterus.

The differences between the two ICRP models for the respiratory tract could be expected to produce differences in estimated doses. During development of the updated respiratory tract model, its performance was tested in detail to determine the affects of various parameters taken alone and in combination. Some examples of the performance of both systems provide useful information about likely differences in estimating both equivalent dose and effective dose equivalent.

One such evaluation, reported by James (James 1994) compared the lung dose equivalent and effective dose for several categories of radionuclides, including insoluble alpha emitters, such as plutonium at Palomares. In those illustrations, James compared doses for intakes of 1 μm activity median aerodynamic diameter (AMAD) particles although ICRP recommends 5 μm AMAD for workers. For 1 μm AMAD, Type S (Class Y) ²³⁹Pu, the ICRP-30 and ICRP-66 equivalent dose per unit intakes were 320 $\mu\text{Sv/Bq}$ and 84 $\mu\text{Sv/Bq}$, respectively. The ICRP-66 equivalent dose was lower by about a factor of 3.8. For 5 μm AMAD particles, ICRP-66 estimated 50 $\mu\text{Sv/Bq}$, or about 6 times lower. Calculating effective dose for the same conditions, ICRP-30 produced 60 $\mu\text{Sv/Bq}$ and ICRP-66 produced 16 $\mu\text{Sv/Bq}$ for 1 μm AMAD particles and 9.1 $\mu\text{Sv/Bq}$ for 5 μm AMAD particles, representing reductions of about 3.7 and 6.5, respectively. Thus, other factors being equal, the ICRP-66 respiratory tract model can produce equivalent doses that are roughly 3

to 6 times lower for the same intake than the ICRP-30 model. This difference, attributed to the modified model for lung deposition and clearance and revised tissue weighting factors must be recognized in evaluating methods for this project.

Determining the amount of material taken into the body during an exposure forms the basis for estimating the amount of material that is transferred to the blood stream and internal organs as well as the amount that clears from the body. Estimates of the organ dose from internal emitters generally follows from an intake assessment, which is usually based on measures of the material in the body or excreted from the body. Common methods include in-vitro bioassay of the amount of the material excreted, measurements of body or organ content, or estimates from air or water concentrations. For this case, estimates of the intakes from environmental plutonium concentrations provide the best available method for assessing the intake. The large collection of urinary analyses were evaluated and used to estimate intakes and doses; however, those were judged unrealistically high. The methods and models used for accomplishing the estimates from urine analyses are discussed in Appendix D.

4.2.2 Computer Models Investigated

Many computer programs have been developed and are available for performing the calculations of the models discussed above. Currently more programs implement the ICRP-30 system than the ICRP-66 model. This comes as no surprise since the ICRP-30 system remains the current system for regulation of the doses from radioactive materials in the United States. However, one objective for this project included the evaluation and recommendation of the best calculation method. Since ICRP provisions are usually adopted in the U.S., investigating at least one software program that implemented the most recent approach seemed reasonable. After some review of the available software, three programs were selected for further study – the Radiological Bioassay and Dosimetry Program (RBD) as modified for the Air Force, Code for Internal Dosimetry (CINDY), and Lung Dose Evaluation Program (LUDEP ver 2.06). Testing of program performance and selection for use are described in Appendix D.

4.3 MODEL ADOPTION

RBD/AF, CINDY, and LUDEP all provide acceptable performance on estimating intake, calculating dose, and providing compatibility with the available data. LUDEP is somewhat less convenient for manipulating large numbers of cases and for generating outputs that can be used in other manipulations; however it implements the current ICRP respiratory tract model.

CINDY and RBD/AF implement the current regulatory system of the NRC and DOE for radiation protection, while LUDEP offers the alternative for applying the respiratory tract model and other features of recent ICRP recommendations. CINDY provides somewhat more flexibility in setup, estimating intakes, and reporting. Consequently, CINDY was chosen as the primary method for assessing the Palomares cases. LUDEP was retained as a reasonable alternate that provides complementary assessments for interesting cases and offers a much-needed point for comparison of results.

5 ESTIMATES FROM ENVIRONMENTAL MEASUREMENTS

5.1 METHODS

The environmental studies summarized in Section 3.1 reported values for the annual average air concentration and the highest weekly measurement obtained with air samplers located near the impact point for weapon number 2. These were selected as reasonable values for air concentrations that response force personnel could have experienced. Those values were combined with dose conversion factors for Type S plutonium calculated using LUDEP. Since breathing rates affect the intake – the more air one breathes in, the more plutonium that enters the lungs – the calculations were performed for standard workers (breathing rate of 1.2 m³ per hour) and for heavy workers (1.688 m³ per hour). Also, the calculations were performed for particle sizes of 1 micrometer and 5 micrometers AMAD. Smaller particle sizes tend to produce higher deposition in the lung and consequently higher doses. Previous recommendations of the ICRP (ICRP-30) recommended 1 micrometer AMAD; however, recent recommendations (ICRP-66) favor 5 micrometers AMAD as more representative of worker exposures.

5.2 RESULTS

Calculations of intake and dose were performed for three exposure scenarios. The first assumed that response force workers were on site for two weeks, and worked 6 days per week for 12 hours a day. This would represent many of the responders who rotated at two-week intervals. The second scenario used 4 weeks on site under the same work conditions to represent those who stayed somewhat longer. Finally, the last scenario assumed that responders could have been exposed for 11 weeks, which essentially represents the entire response effort; i.e. from just after the accident until March 31, 1966. Those estimates are shown in Table 5 and indicate that even the highest scenario produces much less than 1 rem whole body committed effective dose equivalent.

The resuspension factors described in Section 3.1 were used to calculate air concentrations, intakes, and doses (CEDE) for the same scenarios described above. The results listed in Table 6 indicate that even the highest dose (0.312 rem) is well below a significant amount. Furthermore, these estimates differ significantly from the intakes and dose estimates derived from urine analysis, and demonstrate the need to refine the analysis with follow-up studies.

6 RESULTS FROM URINALYSIS DATA

The response to the Palomares nuclear accident involved hundreds of personnel working toward the common purpose of recovering vital materials, protecting themselves and the local populace, and restoration of the accident scene to useable and safe conditions. The accident itself released plutonium during explosions and fires that followed the impact of two of the nuclear weapons with the ground. The plutonium was released primarily as airborne dust and as residues from fire that contaminated the ground. Since the fires essentially were out long before serious response efforts started, the main source of exposure arose from activities such as vehicle movement, handling debris during recovery, plowing fields to mix the contaminant into the soil, and vehicle movement. Persistent winds also contributed to the resuspension of contaminated soils from the

Table 5. Intake and dose estimates from air concentrations.

Average Air Concentration 0.000442 Bq/m³
 Maximum Air Concentration 0.0108 Bq/m³

| Scenario | Worker Type | Breathing Rate (m ³ /hr) | Particle Size (um) | Exposure Time (hours) | Dose Conversion Factor (Sv/Bq) | | Average Air Concentration | | | Maximum Air Concentration | | |
|--|-------------|-------------------------------------|--------------------|-----------------------|--------------------------------|-----------|---------------------------|--------------------------|-----------|---------------------------|--------------------------|-----------|
| | | | | | ICRP-26 | ICRP-60 | Intake (Bq) | CEDE (Sv)/ CEDE (rem) | | Intake (Bq) | CEDE (Sv)/ CEDE (rem) | |
| | | | | | | | | ICRP-26 | ICRP-60 | | ICRP-26 | ICRP-60 |
| 2 weeks 6 days per week 12 hours per day | Standard | 1.2 | 1 | 144 | 1.946E-05 | 1.531E-05 | 0.076 | 1.486E-06 | 1.169E-06 | 1.87 | 3.632E-05 | 2.857E-05 |
| | Standard | 1.2 | 5 | 144 | 1.084E-05 | 8.647E-06 | 0.076 | 8.279E-07 | 6.604E-07 | 1.87 | 2.023E-05 | 1.614E-05 |
| | Heavy | 1.688 | 1 | 144 | 1.975E-05 | 1.571E-05 | 0.107 | 2.122E-06 | 1.688E-06 | 2.63 | 5.185E-05 | 4.124E-05 |
| | Heavy | 1.688 | 5 | 144 | 1.227E-05 | 1.010E-05 | 0.107 | 1.318E-06 | 1.085E-06 | 2.63 | 3.221E-05 | 2.651E-05 |
| 4 weeks 6 days per week 12 hours per day | Standard | 1.2 | 1 | 288 | 1.946E-05 | 1.531E-05 | 0.153 | 2.973E-06 | 2.339E-06 | 3.73 | 7.263E-05 | 5.714E-05 |
| | Standard | 1.2 | 5 | 288 | 1.084E-05 | 8.647E-06 | 0.153 | 1.656E-06 | 1.321E-06 | 3.73 | 4.046E-05 | 3.227E-05 |
| | Heavy | 1.688 | 1 | 288 | 1.975E-05 | 1.571E-05 | 0.215 | 4.244E-06 | 3.376E-06 | 5.25 | 1.037E-04 | 8.248E-05 |
| | Heavy | 1.688 | 5 | 288 | 1.227E-05 | 1.010E-05 | 0.215 | 2.637E-06 | 2.170E-06 | 5.25 | 6.442E-05 | 5.303E-05 |
| Full Response 11 weeks 6 days per week 12 hours per day | Standard | 1.2 | 1 | 792 | 1.946E-05 | 1.531E-05 | 0.420 | 8.175E-06 | 6.431E-06 | 10.3 | 1.997E-04 | 1.571E-04 |
| | Standard | 1.2 | 5 | 792 | 1.084E-05 | 8.647E-06 | 0.420 | 4.554E-06 | 3.632E-06 | 10.3 | 1.113E-04 | 8.876E-05 |
| | Heavy | 1.688 | 1 | 792 | 1.975E-05 | 1.571E-05 | 0.591 | 1.167E-05 | 9.283E-06 | 14.4 | 2.852E-04 | 2.268E-04 |
| | Heavy | 1.688 | 5 | 792 | 1.227E-05 | 1.010E-05 | 0.591 | 7.250E-06 | 5.968E-06 | 14.4 | 1.772E-04 | 1.458E-04 |

Table 6. Intake and dose estimates from resuspension.

| | | | | | |
|-------------------------------|-----------|-------------------|-------------------------------|----------|-------------------|
| Minimum Resuspension Factor | 1.29E-09 | m ⁻¹ | Maximum Resuspension Factor | 1.00E-07 | m ⁻¹ |
| Minimum Surface Contamination | 1.18E+05 | Bq/m ² | Maximum Surface Contamination | 1.18E+06 | Bq/m ² |
| Minimum Air Concentration | 1.522E-04 | Bq/m ³ | Maximum Air Concentration | 0.118 | Bq/m ³ |

| Scenario | Worker Type | Breathing Rate (m ³ /hr) | Particle Size (um) | Exposure Time (hours) | Dose Conversion Factor (Sv/Bq) | | Minimum Air Concentration | | | Maximum Air Concentration | | |
|--|-------------|-------------------------------------|--------------------|-----------------------|--------------------------------|-----------|---------------------------|--------------------------|-----------|---------------------------|--------------------------|-----------|
| | | | | | ICRP-26 | ICRP-60 | Intake (Bq) | CEDE (Sv)/ CEDE (rem) | | Intake (Bq) | CEDE (Sv)/ CEDE (rem) | |
| | | | | | | | | ICRP-26 | ICRP-60 | | ICRP-26 | ICRP-60 |
| 2 weeks 6 days per week 12 hours per day | Standard | 1.2 | 1 | 144 | 1.946E-05 | 1.531E-05 | 0.026 | 5.119E-07 | 4.027E-07 | 20.4 | 3.968E-04 | 3.122E-04 |
| | Standard | 1.2 | 5 | 144 | 1.084E-05 | 8.647E-06 | 0.026 | 5.119E-05 | 4.027E-05 | 20.4 | 0.0397 | 0.0312 |
| | Heavy | 1.688 | 1 | 144 | 1.975E-05 | 1.571E-05 | 0.037 | 2.851E-07 | 2.274E-07 | 28.7 | 2.210E-04 | 1.763E-04 |
| | Heavy | 1.688 | 5 | 144 | 1.227E-05 | 1.010E-05 | 0.037 | 2.851E-05 | 2.274E-05 | 28.7 | 0.0221 | 0.0176 |
| | | | | | | | | 7.308E-07 | 5.813E-07 | | 5.665E-04 | 4.506E-04 |
| | | | | | | | | 7.308E-05 | 5.813E-05 | | 0.0566 | 0.0451 |
| | | | | | | | | 4.540E-07 | 3.737E-07 | | 3.519E-04 | 2.897E-04 |
| | | | | | | | | 4.540E-05 | 3.737E-05 | | 0.0352 | 0.0290 |
| 4 weeks 6 days per week 12 hours per day | Standard | 1.2 | 1 | 288 | 1.946E-05 | 1.531E-05 | 0.053 | 1.024E-06 | 8.054E-07 | 40.8 | 7.936E-04 | 6.244E-04 |
| | Standard | 1.2 | 5 | 288 | 1.084E-05 | 8.647E-06 | 0.053 | 1.024E-04 | 8.054E-05 | 40.8 | 0.0794 | 0.0624 |
| | Heavy | 1.688 | 1 | 288 | 1.975E-05 | 1.571E-05 | 0.074 | 5.703E-07 | 4.549E-07 | 57.4 | 4.421E-04 | 3.526E-04 |
| | Heavy | 1.688 | 5 | 288 | 1.227E-05 | 1.010E-05 | 0.074 | 5.703E-05 | 4.549E-05 | 57.4 | 0.0442 | 0.0353 |
| | | | | | | | | 1.462E-06 | 1.163E-06 | | 1.133E-03 | 9.012E-04 |
| | | | | | | | | 1.462E-04 | 1.163E-04 | | 0.1133 | 0.0901 |
| | | | | | | | | 9.080E-07 | 7.474E-07 | | 7.039E-04 | 5.794E-04 |
| | | | | | | | | 9.080E-05 | 7.474E-05 | | 0.0704 | 0.0579 |
| Full Response 11 weeks 6 days per week 12 hours per day | Standard | 1.2 | 1 | 792 | 1.946E-05 | 1.531E-05 | 0.145 | 2.815E-06 | 2.215E-06 | 112.1 | 2.182E-03 | 1.717E-03 |
| | Standard | 1.2 | 5 | 792 | 1.084E-05 | 8.647E-06 | 0.145 | 2.815E-04 | 2.215E-04 | 112.1 | 0.2182 | 0.1717 |
| | Heavy | 1.688 | 1 | 792 | 1.975E-05 | 1.571E-05 | 0.204 | 1.568E-06 | 1.251E-06 | 157.8 | 1.216E-03 | 9.697E-04 |
| | Heavy | 1.688 | 5 | 792 | 1.227E-05 | 1.010E-05 | 0.204 | 1.568E-04 | 1.251E-04 | 157.8 | 0.1216 | 0.0970 |
| | | | | | | | | 4.019E-06 | 3.197E-06 | | 3.116E-03 | 2.478E-03 |
| | | | | | | | | 4.019E-04 | 3.197E-04 | | 0.3116 | 0.2478 |
| | | | | | | | | 2.497E-06 | 2.055E-06 | | 1.936E-03 | 1.593E-03 |
| | | | | | | | | 2.497E-04 | 2.055E-04 | | 0.1936 | 0.1593 |

ground or contaminated dusts from the surfaces of accident debris, local buildings, or agricultural crops.

Ingestion by hand to mouth transfer is a second possible route of entry. However, that route is very inefficient. Furthermore, the fraction of plutonium that enters the bloodstream from the intestines is very small (0.00001 for Type S). For reasons discussed in Appendices D and E, the ingestion route is not considered further.

The type of exposure was assumed a single acute exposure. This assumption accommodates the long time for removal of plutonium oxides from the human body. The response activity occurred from January 18, 1966 until April 3, 1966 when activities were moved from Camp Wilson to another location. Personnel on site reached a maximum in late January; tapered off during February, and then increased slightly in mid-March during the packaging of contaminated debris, soil and other wastes for disposal. Most departed the site by late March 1967. The nominal length of assignment was about two weeks. However, records indicate that some personnel stayed much longer.

6.1 HIGH 26 CASES

The responders were assigned to four groups of cases, as discussed above – the High 26 Cases Group, the Repeat Analysis Cases Group, the Contamination Cutoff Cases Group and the Remaining Cases Group. The High 26 Cases Group offered the best collection of urinary measurement data to develop an overall understanding of the relationship between the measurement results and possible intake of plutonium. Therefore, substantial effort was applied to evaluating these cases. Then, that understanding was applied to the remaining three groups of cases. As discussed above, however, the quality of the data set limited the preparation of reasonable estimates. This section describes the approach to evaluating each group, the results obtained, the relationship between the estimated dose equivalents and effects.

6.1.1 Methods and Results

The High 26 Cases Group represents the collected measurement data from 26 responders who were identified for follow-up after the initial phase of sampling in 1966. This group provided 127 urine samples during their on-site and resampling activities. Most of those samples (102 of 127) produced ^{239}Pu measurements from alpha spectrometry. Appendix E provides detailed discussions of the data evaluation, results of model fitting, and estimated intakes and doses.

6.1.1.1 Methods

Careful evaluation of the results revealed several difficulties with the reported results. These included differences in the reporting of confidence levels for the results. The reported errors for gross alpha measurements represented the 95% confidence level while the reported errors for alpha spectrometry measurements represented the 68% confidence level. Since the criterion for reporting a result as no detectable activity was based on the 95% confidence limit, some alpha spectrometry results may have been reported as positive when the estimated errors did not support that conclusion. In addition, some alpha spectrometry reports contained calculated values although the reported results indicated NDA. Those results were used in these estimates when recorded on the individual data cards.

Laboratory measurements experienced some difficulties with reproducibility also. In several samples with multiple analyses, differences in reported concentrations of two to three times were observed.

The urine analysis results for the High 26 Cases Group indicated that those cases with several measurements for samples collected over the entire initial and resampling efforts could provide the best data for testing. The data and CINDY and LUDEP program setup were varied in several ways. Assumptions were developed for the date of exposure, the use of gross alpha results and the use of NDA results. For the programs, the main adjustment involved the method for weighting results during intake assessment using CINDY and LUDEP. Generally, the date of exposure was assumed as the first day on site, gross alpha measurements were rejected, and values were developed for NDA reports. These variations are discussed in detail in Appendix E.

6.1.1.2 Results

For the 26 cases, the preliminary intake estimates varied from 34,000 pCi to 570,000 pCi from CINDY and 19,000 pCi to 2,600,000 pCi from LUDEP with the gross alpha results excluded in all the cases. Estimates of committed effective dose equivalent ranged from 10 rem to 170 rem (0.1 to 1.7 Sv) from CINDY and 1.3 to 180 rem (0.013 to 1.8 Sv) from LUDEP. LUDEP ranged from -83% to +150% of CINDY results. The range of differences between LUDEP results and CINDY results seems reasonable considering the variation in the data and the complexities of the assessment. In addition to the intakes and CEDE estimates, 50-year committed dose equivalents were calculated for organs using CINDY. Those results are discussed in Appendix E, however, when compared with independent estimates from environmental data and with the results of other exposure cases, these estimates seem unreasonably high.

6.2 REPEAT ANALYSIS CASES GROUP

Palomares responders were placed in the Repeat Analysis Cases Group if they met one or both of the following conditions:

- They submitted an initial urine sample while on site that was analyzed for gross alpha radioactivity and then reanalyzed by alpha spectrometry for ^{239}Pu ; or
- They submitted an initial sample while on site that was analyzed by gross alpha counting and then submitted one or more follow-up samples after returning to their base of assignment for analysis by alpha spectrometry.

6.2.1 Methods and Results

From January 17, 1966 to June 22, 1966, this group provided 82 urine samples from 54 individuals that produced usable results. The gross alpha and alpha spectrometry measurements are primarily greater than 0.1 pCi/d and the two types of measurements are interspersed among one another. Most of the samples were characterized by a gross alpha measurement followed by reanalysis by alpha spectrometry in an attempt to identify the radionuclide responsible for the gross alpha result. In most cases, the alpha spectrometry result was lower than the gross alpha measurement. Unfortunately, resampling was not accomplished for those in this group.

6.2.1.1 *Methods*

The Repeat Analysis Cases Group had exposure dates that extended over a broader range of dates than the High 26 Cases Group. However, many were among the initial responders who arrived in January 1966. Because the time on site seemed shorter and better recorded for this group, the exposure date was assumed as the midpoint of the time at Camp Wilson. In general, gross alpha results for samples collected on site were excluded from the analysis, gross alpha results reported as NDA were assigned a value of 0.009 pCi/d, numerical results recorded on alpha spectrometry records reported as NDA were used, and some alpha spectrometry results were excluded when they did not fit the expected urinary excretion pattern. Method details are provided in Appendix D.

6.2.1.2 *Results*

For the 54 cases, the estimated intakes varied from 2,900 pCi to 1,300,000 pCi from CINDY and 11,900 pCi to 5,240,000 pCi from LUDEP with the gross alpha results excluded in all the cases. Estimates of committed effective dose equivalent ranged from 0.9 rem to 400 rem (0.009 to 4.0 Sv) from CINDY and 0.8 to 367 rem (0.008 to 3.67 Sv) from LUDEP. LUDEP results ranged from -238% to +94% of CINDY results. In addition to the intakes and CEDE estimates, annual dose equivalents and committed dose equivalents were calculated for organs using both CINDY and LUDEP. Details of these results are discussed in Appendix E. As for the High 26 Group, these estimates are unrealistic when compared with the estimates from environmental measurements.

6.3 **CONTAMINATION CUTOFF CASES GROUP**

The Contamination Cutoff Cases Group of analyses was created to calculate estimated intake and dose equivalent for those whose urine measurement results indicated potentially contaminated samples collected at the accident site but were below a reasonable minimum level that did not represent unusually high exposures. While the data for this group were not especially robust, this approach offered an opportunity to evaluate additional cases. As discussed in Appendix E, a level of 0.1 pCi/d was adopted as reasonable maximum level for cases included in the Contamination Cutoff Cases Group.

6.3.1 **Methods and Results**

6.3.1.1 *Methods*

The procedures for analysis of the High 26 Cases Group were applied to the Contamination Cutoff Cases Group, except that the intakes and dose equivalents were calculated using only the CINDY program. The group had exposure dates that began over a similar range of dates to the Repeat Analysis Cases Group. Many of this group stayed on site for one to two weeks, with some up to a month. The exposure date was assumed as the midpoint of the time at Camp Wilson. See Appendix E for additional details of this group's analyses.

6.3.1.2 Results

For the 313 individuals in the Contamination Cutoff Cases Group, the estimated intakes varied from 1,500 pCi to 110,000 pCi. Estimates of committed effective dose equivalent ranged from 0.46 rem to 34 rem (0.0046 to 0.34 Sv). The higher estimated intake and dose were produced by a urine sample, taken at 25 days after the assumed exposure date, with a result of 0.099 pCi/d of gross alpha activity. According to the excretion function derived, the urinary content on day 25 represents approximately 9×10^{-7} of the inhalation intake. This case illustrates how urine concentrations that are even slightly above delectability can lead to sizeable estimated intakes and dose equivalents. This further illustrates the difficulty in obtaining realistic estimates from sparse data at or near the analytical methods detection limit.

6.4 REMAINING CASES GROUP

The cases that were not included in one of the previous three groups were placed in the Remaining Cases Group. These samples included those from individuals who submitted only one sample, or from cases where some follow-up was attempted but results were inadequate because of low or no chemical recovery or laboratory error. This group contains sample measurements on 1,063 individuals for 1,219 samples. For discussion purposed, the lowest and the highest urine results of 0 and 237.9 pCi/d of gross alpha radioactivity were input to CINDY, and produced estimated intakes of 75,000 pCi to 20,000,000 pCi corresponding to CEDEs of about 23 rem to 6,000 rem (0.23 to 60 Sv). These results are clearly unrealistic, not supported by the air concentrations observed at Palomares and require careful evaluation.

7 DISCUSSION

The preliminary intake and dose equivalent estimates for the Palomares response personnel used the available data to the best extent possible. The approach involved reasonable assumptions about the type of activities that the responders performed and about the length of time, they may have been exposed. Detailed assignment records on the personnel were not available, nor was any significant effort expended to determine the details. Written accounts of the accident and response, correspondence in the records of some High 26 Cases Group personnel, and personal conversations with some of these individuals provided a reasonable description of the situation during the response.

Results obtained in environmental characterization programs around Palomares for over 15 years following the accident provided an alternative route to assessing intakes and doses. Those estimates are much more realistic when compared with the estimated intakes and doses for other plutonium exposures to workers or members of the public.

7.1 RESULTS FROM ENVIRONMENTAL MEASUREMENTS

The estimated intakes and doses for three scenarios of worker activity indicate that the exposures are well below recommended limits for workers and a small fraction of the dose (10 rem) for which health effects have been reliably demonstrated in humans. The estimates are limited, however, because they represent evaluations using representative scenarios. They do not represent the exposures to any specific individual responder. Additional information on responder activities, time exposed, conditions of exposure, use of personal protective equipment,

and factors that influence intake are needed to develop case-specific assessments. Nevertheless, these estimates form serious concerns about the reliability of estimates from the urinary bioassay data. As a matter of fact, the difficulty in extrapolating urinary concentrations determined at the limits of detection of the analytical methods are well known and are most likely a major contributor to the disparity in the two approaches.

The estimates from the environmental data are very consistent with the results obtained for residents of the Palomares area and with results for Manhattan Project workers. These comparisons lend credibility to the bounds of estimates from the environmental data and support conclusions about the significance of the exposures reached in 1966 through 1968.

7.2 RESULTS FROM URINARY BIOASSAY

The estimated intakes and doses for all groups were unrealistically high as discussed above. Nevertheless the implications of these estimates for effects on health are included to provide some interpretation for what are likely to be upper bound estimates. Furthermore, comments on the analytical methods, case specific information, and other inconsistencies in the data are presented as background for possible reevaluations in the future.

7.2.1 Assessment of Possible Effects

Characterizing the preliminary estimates of intakes and dose are useful only to indicate that many individual cases represent significant to very serious situations when compared to accepted guidelines for management of radiation exposures. About half the estimates exceeded the cumulative dose that would be experienced by anyone in the United States from lifetime exposure to the average background dose (roughly 21 rem (0.21 Sv) over 70 years). Fortunately, the estimates derived from environmental data (Section 6.1.3.2 above), using very conservative scenarios and assumptions, provide upper bound estimates that are well below accepted guidelines and are more consistent with the exposure experience of the local populace on site at Palomares and of industrial plutonium workers. All, but the extreme cases of the estimates, are below the recommended average radiation exposure for members of the public in one year.

7.2.2 Comments on the Estimates

Substantial experience and useful observations arose from the attempts at preparing estimates of plutonium intake and dose from the urinary bioassay data. Those observations and comments are discussed below for each of the groups.

7.2.2.1 High 26 Cases

The intakes and doses discussed in the previous section represent conservative estimates of the intakes and dose equivalents for the High 26 Cases Group. Additional comments are required to put the estimates into perspective. Those comments address the quality of the urine bioassay measurements, assumptions about the type and duration of exposure, the class (type) of material involved and specific details of the duties performed by each individual. Without further details and possible confirmation, permanent assignment of these intakes and doses to the individuals may be premature.

The laboratory analyses performed in 1966 and 1967 represent a comprehensive effort to assess the possible exposure to plutonium. At the time, the urine results used the best available models for estimating body burden. However, methods for estimating intake and deposition of plutonium in the lungs were not well understood. Progress since then allows better estimates to be made now. In fact, deposition in the lungs and the associated dose is the major contributor to the annual dose in the first few years after the exposure. Unfortunately, a very small amount of plutonium in urine can be associated with an intake that produces sizeable doses.

For the cases evaluated, the amount of plutonium in the urine after about one month is more than one million times less than the amount of the intake. That fraction decreases slowly, but steadily, thereafter. The sensitivity of the analytical methods limit the ability to confirm the amount deposited. Samples were collected out to about 15 months following the accident yet the expected excretion curve implies that plutonium excretion would continue beyond that time for actual intakes. More sensitive techniques are now available that could provide new analyses of urine samples.

At 34 years after the accident, the amount excreted per day would be about two million times less than the initial intake. The feasibility of obtaining useful assessments of plutonium uptake by sampling urine now depends mainly on the sensitivity of the analytical techniques and on the ability of the available models to represent human excretion of the plutonium in urine.

Analytical techniques currently available that provide potentially adequate sensitivity include alpha spectrometry, neutron induced fission track analysis (FTA), and mass spectrometry (Wrenn 1994). Alpha spectrometry, which cannot distinguish between ^{239}Pu and ^{240}Pu has nominal sensitivity for both of about 0.02 pCi per sample. That is about the same level available during the resampling conducted in 1966 and 1967. Most mass spectrometry techniques provide about the same sensitivity as alpha spectrometry. Thermal Ionization Mass Spectrometry (TIMS) offers sensitivities of about 0.005 pCi per sample but is tedious and costly. Neutron induced FTA provides sensitivities of about 0.00003 pCi per sample, or about 1,000 times better than alpha spectrometry and routine mass spectrometry. However, FTA is performed at only one or two laboratories.

The biokinetics and urinary excretion models available in ICRP-30 and from Jones (Jones 1985) vary in their ability to model the available data on human excretion at long times after exposure. The Jones model corresponds quite well as recently discussed (Luciani 2000). At 34 years after exposure, the model predicts that the daily urinary excretion would be 10^{-5} of the amount transferred to the blood. As an example, a urine sample with a measured $^{239,240}\text{Pu}$ content of 0.00003 pCi/L would translate into an uptake of 4.2 pCi to the blood from the original inhalation intake. For Class Y plutonium, about 5 percent of the inhaled plutonium transfers to blood. Therefore, the intake would be 84 pCi, which is well below one ALI of 13,500 pCi. Follow-up sampling and analysis using the most sensitive techniques available today, offers a reasonable potential for obtaining useful information. A decision to use the approach should also consider other factors, such as cost, ability to locate and obtain cooperation of response personnel, and limited laboratory availability.

Assumptions were made concerning the type of exposure (single, acute inhalation) and dates of the exposure. For some individuals, this assumption may represent up to several weeks of difference in determining the elapsed time between exposure and collection of samples. The elapsed time is one of the primary parameters for estimating the intake.

The assessment also assumed that the plutonium was PuO₂ and represented by lung Class Y (Type S). All (100%) of the intake was assumed to be from this material. Limited tests were also performed using CINDY assuming a mixed material (50% Class W and 50% Class Y). Those attempts produced lower estimated intakes and doses, however, difficulties with reconciling the approach with experimental confirmation of typical plutonium at Palomares are problematic. In addition, as discussed in Section 3, the cases of mixed plutonium forms also demonstrate a long-term excretion component that is not observed for the data. Never the less, the estimates obtained with the 100% Class Y assumption are higher and therefore conservative.

Finally, these estimates were performed with limited information about the specific activities and times that the individuals were on the site. Efforts to perform a comprehensive search of all records and information, including interviews, were beyond the scope of this effort. Some additional refinement might be possible from an expanded search for more specific information. However, the cost of such an effort should be balanced with the possible benefits from confirmatory measurements of urinary content. Ultimately, credible estimates of intake and dose will depend on an expensive, multi-phased approach involving:

- Urinalysis of selected individuals using highly sensitive techniques to assess the presence of plutonium in their urine.
- Detailed interviews with individuals to develop the details of their exposure circumstances as well as they can recall them.
- Research and evaluation of all available information, especially that collected during the recovery and response phases of the incident, including records available at DOD's Defense Threat Reduction Agency, the Air Force Safety Agency, the Department of Energy, and possibly the appropriate representatives of the Government of Spain.

7.2.2.2 Contamination Cutoff Cases

The intakes and doses discussed in the previous section represent conservative estimates of the intakes and dose equivalents for the Contamination Cutoff Group. The estimates are considered conservative because the methods and data selected tend to overestimate the actual intakes and doses. The additional comments made regarding the High 26 Cases Group apply to these cases as well. Furthermore, confirmation of the possible exposures for this group are very important because this group did not have any measurements taken in late 1966 or 1967, when alpha spectrometry measurements were more commonly used.

7.2.2.3 Repeat Analysis Cases

The intakes and doses discussed in the previous section represent conservative estimates of the intakes and dose equivalents for the Repeat Analysis Cases Group. The estimates are considered conservative because the methods and data selected tend to overestimate the actual intakes and doses. The additional comments made regarding the High 26 Cases Group apply to these cases as well. Furthermore, confirmation of the possible exposures for this group are very important because this group did not have any measurements taken in late 1966 or 1967, when alpha spectrometry measurements were more commonly used.

7.3 COMPARISON OF INTAKES AND DOSES TO OTHER PLUTONIUM EXPOSURE CASES

The results can be evaluated for reasonableness by comparing them to other plutonium exposure situations. Two such reported cases are the evaluation of the citizens of Palomares by a Joint Spanish-United States effort since the accident, and the follow-up of Manhattan Project workers who received exposures to plutonium at Los Alamos. In addition, measurements of environmental plutonium at Palomares provide data for performing independent estimates of the intakes and doses for the accident response force.

7.3.1 *Dose Estimates for Residents of Palomares*

Since the accident, the Government of Spain has conducted a program to monitor the residual radioactivity at the accident site. That effort has included measurements of air concentrations of plutonium, soil contamination levels, and assessment of intakes and doses to the population.

During 1966, 59 people provided urine samples on three occasions. Those samples indicated the possibility of contamination (Iranzo 1997). In 1967 samples were collected in Madrid under controlled conditions. Of those, 23 exceeded the minimum detectable level of 0.02 pCi/ day. During the ensuing years, additional samples have been collected from a larger group of Palomares citizens and analyzed. The results indicate that 45 individuals who may have received intakes during the initial clean-up showed intakes that represented committed effective dose equivalents of 2 rem to 20 rem (0.02 to 0.2 Sv) (Iranzo 1987). That range includes the lower portion of the results obtained for responders. In addition, the early concerns for sample contamination and efforts to mitigate the possibility support similar concerns for the Air Force urine sampling. Although, the Air Force resample effort was conducted away from the accident site, the possibility exists that samples provided in mid to late 1966 and early 1967 may have been influenced by continued sample contamination.

7.3.2 *Manhattan Project Worker Evaluations*

During the Manhattan Project, 26 white, male adult workers received intakes of plutonium primarily by inhalation. Reports of follow-up studies of that group have indicated continuing refinement of the estimates of their plutonium deposition. A recent report provided the results of 50 years of follow-up. The report indicated that the depositions for the 26 individuals ranged from 1.35 nanocuries (50 Bq) to 85.86 nanocuries (3,180 Bq) (Voelz 1997). The corresponding effective doses ranged from 10 rem to 720 rem (0.1 to 7.2 Sv). If those exposures occurred by inhalation, the intake would have been approximately 20 times higher than the deposition or 27 nanocuries to 2.3 microcuries. Although the range of exposures is similar to the preliminary estimates for Palomares response personnel, the responders' exposures were unlikely to approach those of the Manhattan Project workers. Responders would have handled the much different (lower) quantities and forms of plutonium for much shorter times than the Manhattan Project workers. Those workers performed continuous, industrial operations on a daily basis over several years under what have been called "primitive conditions".

The results of follow-up of citizens of Palomares and Manhattan Project workers indicate the range of doses from exposures received under field conditions and those received in laboratory or industrial conditions. It seems reasonable to consider the results for the Palomares citizens as more representative of the kind of exposure conditions experienced by the response personnel

because both were exposed to the same or similar sources, while the Palomares residents were exposed for many years. Consequently, the results for responders that exceed even a fraction of the upper range of CEDE (20 rem/0.2 Sv), may well represent sample contamination or other artifacts. If that is the case, additional sampling and analysis of a carefully selected subset of the response force today offers an attractive approach to confirming the deposition and associated doses.

8 CONCLUSIONS AND RECOMMENDATIONS

Records of urinary ^{239}Pu and gross alpha radioactivity of samples collected from responders to the Palomares nuclear weapons accident were evaluated for possible use in calculating estimate radioactivity intakes and committed effective dose equivalent using accepted models. Data were reviewed and individuals assigned to four groups according to the amount and reliability of the data. The groups included:

- The High 26 Cases Group that included 26 individuals identified for resampling for 18 to 24 months after the initial phase of sampling in 1966.
- A Repeat Analysis Cases Group that included 54 individuals who either had submitted samples that were reanalyzed using more specific methods (alpha spectrometry), or who were resampled.
- A Contamination Cutoff Cases Group that included 313 individuals with results that were below a reasonable, assumed cutoff level of 0.1 pCi per day.
- A Remaining Cases Group that contained 1,063 records that were not otherwise evaluated and that were strongly suspected of contamination from collection on site.

Two current computer methods were tested and used to estimate intakes of plutonium by acute inhalation exposure. One method (CINDY) employed the ICRP-30 system for limiting internal dose. The other method (LUDEP) implemented the new respiratory tract model described in ICRP-66 and the organ/tissue weighting factors of ICRP-60.

Plutonium intake and dose values were estimated for all of the High 26 Cases Group, the 54 individuals in the Repeat Analysis Cases Group, and 313 individuals in the Contamination Cutoff Cases Group. The intakes and doses ranged from below annual occupational limits to more than the 50 rem (0.5 Sv) guideline for cumulative dose for workers. Some doses ranged as high as several hundred rem. However, when compared with estimates derived from environmental measurements, dose estimates for Palomares citizens, and dose estimates for Manhattan Project workers, these preliminary estimates seen unreasonably high in many cases. Additional efforts are needed to reconcile the results from the urine data with the levels that can be reasonably supported by the environmental data and experience with other exposed people.

Several future actions should be considered to further refine these initial estimates.

1. Additional effort is needed to reconcile the estimated intakes and doses derived from the urinary bioassay data with the estimates from environmental measurements. A targeted effort that includes participant activities, participant interviews, urine and other appropriate plutonium analyses using current techniques, medical records review, and modeling should be considered.

2. The results of this effort should be communicated to responders, veterans organizations, and other interested parties using appropriate information that clearly confirms the conclusions of the original medical evaluation program, recognizes the difficulties in preparing updated intake and dose estimates, and outlines the options for strengthening the estimates.

3. Further contacts with the Department of Energy for comparison with evaluations of their personnel who responded to this accident could provide useful data. The effort should be summarized in a companion document that conveys the details of the project and its potential effects on health in an easily understood manner. That document should be made available to any of the responders who desire a copy.

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