

JUN 26 1987

EH-331

Financial Support for Project Indalo and Oak Ridge National Laboratory (ORNL) (Technical Support to Spain)

Neal Goldenberg, EH-33

U.S. funding support for the subject project has continued since the Hall-Otero Agreement letter (Attachment 1) was signed on February 25, 1966. Direct funding to the Spanish government (Project Indalo) provides supplemental assistance to their environmental surveillance program for Palomares and medical follow-up of area residents. ORNL funding provides project coordination, technical support, training for Spanish scientists, and scientific equipment. In FY 1986, the responsibility for this effort was transferred from the Office of Operational Safety (OOS) to the Office of Nuclear Safety (ONS).

Our current ONS budget summary continues to reflect a significant FY 1987 funding reduction for the subject projects. The ONS FY 1987 budget summary indicates \$120K has been appropriated for Project Indalo. \$100K of this funding was transferred to Spain in February, 1987, to supplement DOE's (OOS) FY 1986 funding support, bringing the total funding for Spain's 1986 fiscal year to \$240K (the original amount requested). This leaves \$20K in our FY 1987 budget to support Spain's FY 1987 request of \$500K. Please note that Spain's fiscal year runs from February 1st to the following January 31st.

The increase in Project Indalo funding from FY 1986 to FY 1987 is a result of several factors. First, Spain is interested in increasing their urine bioassay program considering a growing number of residents since 1983 (52 out of 664) have submitted urine samples that showed Pu-239 and Pu-240 activities greater than the detection limit (0.37 mBq/day). Also, recent Americium separations have shown some Am-241 positives in urine as well (included in totals above). Second, Spain desires the United States to increase its support for the followup effort from about 30 percent to 64 percent. Finally, the exchange rate of the dollar has dropped about 25 percent.

ORNL's funding request for FY 1987 is \$120K. The ONS budget summary reflects \$20K for this support. With only 3 months left in FY 1987, ORNL can only spend an additional \$10K for the purchase of a meteorological tower.

On September 15, 1986, Mr. Tommy McCraw, then with the Office of Nuclear Safety, provided a briefing to Mary Walker on the history, status, and funding needs of the Palomares followup project. At the conclusion of the meeting, Mr. McCraw indicated that Ms. Walker agreed that funding was to be provided. To date, such funding has not been made available.

The Department's continued silence on this issue will only exacerbate the situation. Without proper action, Spain may elect to go through other channels to acquire the requested funds. Such action would reflect poorly on

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EH and the Department. Accordingly, we recommend Spain be provided their FY 1987 requested funding level and that OKNL be provided an additional \$10K as indicated above.

This fall, ORNL will convene a meeting of DOE's advisory panel on Palomares matters. An EH representative will be in attendance. It is expected that the panel's report will provide a basis on which EH can base future funding decisions.

Original signed by
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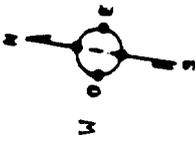
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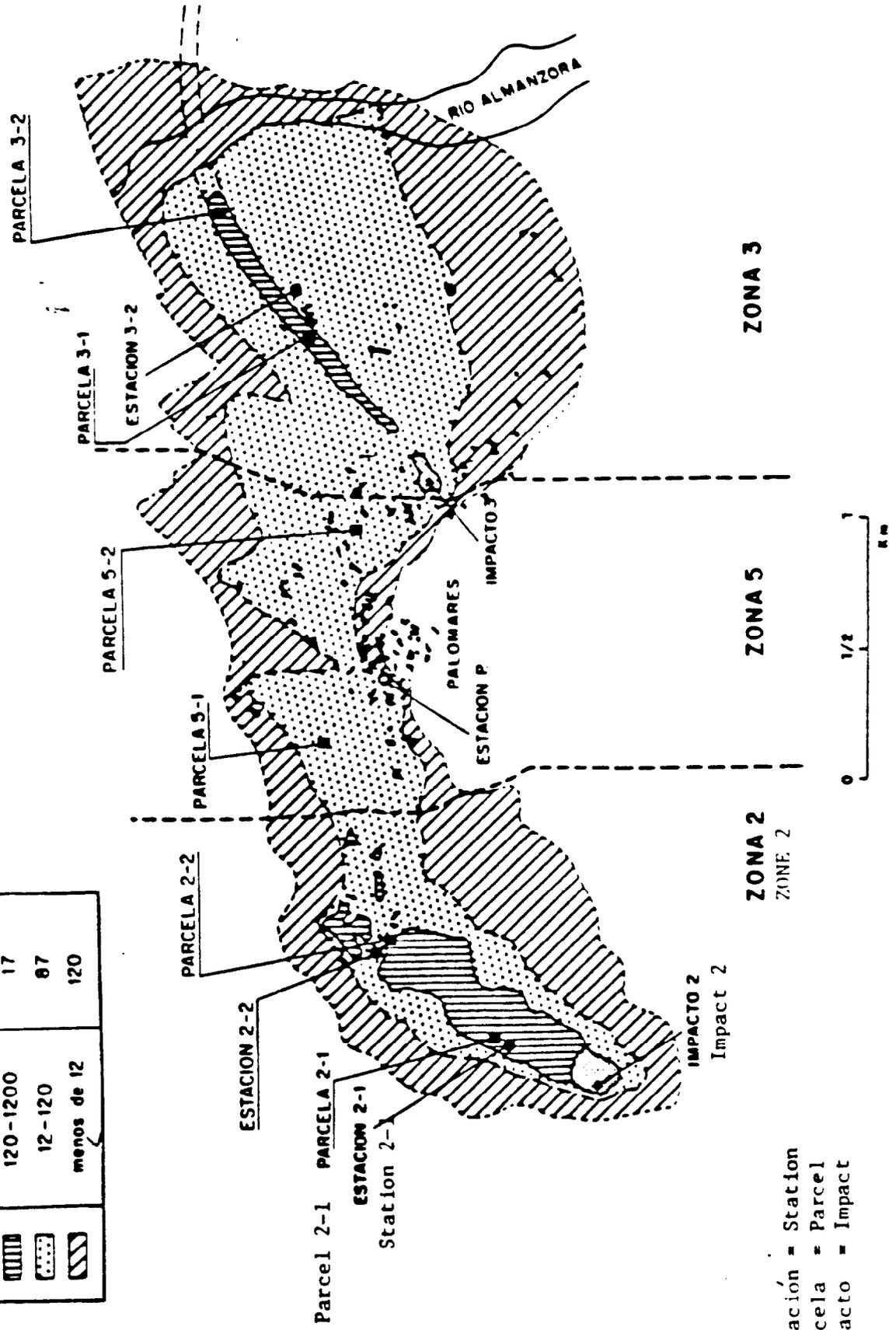
PROJECT INDALO BRIEFING

History of Accident at Palomares, Spain

- o On January 17, 1966, during a refueling operation, a U.S. Air Force KC-135 tanker and a B-52 bomber carrying four nuclear weapons collided and disintegrated over Palomares, Spain, a village of about 1500 people. One crew member survived.
- o Two nuclear weapons were recovered intact. Two others experienced detonation of the high explosives on impact. There was no nuclear yield and no one on the ground was seriously injured.
- o Two overlapping patterns of Plutonium-239 contamination were laid down over a predominantly agricultural area, but also including the village of Palomares.
- o Working cooperatively, representatives of the U.S. and Spanish Governments initiated a joint program for cleanup of the contaminated area. Agreement was obtained on cleanup methods and on cleanup radiological criteria.
- o Contaminated vegetation was collected for disposal. Contaminated soil was scraped into piles, loaded into trucks, and collected in a holding area where it was packaged for shipment to the United States for disposal. A large area was watered down and deep plowed.
- o The contaminated vegetation and soil from Palomares were transported by ship to Charleston harbor and transported to the Savannah River Plant for land burial.
- o In the Hall-Otero Agreement between the United States and Spain, the United States agreed to provide technical and financial assistance to Spain for radiological followup of Palomares residents and their environment. This assistance, Project Indalo, has continued for the past 20 years.
- o It was agreed that the findings for Indalo would be published by Spanish scientists. This has been done. No report on cleanup field operations appears to have been issued by U.S. participants. The fact that Palomares soil containing plutonium from U.S. weapons was buried at SRP has appeared in an ORNL report dated June 1985. Burial at SRP was also reported by Spanish scientists at an international symposium in Monaco in October 1966, and their report on this presentation contains this and other details on cleanup operations.
- o Dr. Emilio Iranzo, Director of Project Indalo in Spain, has submitted a report for publication in Health Physics entitled "Air Concentrations and Potential Radiation Doses to People Living Near Contaminated Areas in Palomares (Spain)." This should be published before the end of this year. This will be the first publication of exposure estimates for Palomares residents in a journal with a large readership. This will focus the attention of the radiation protection community (worldwide) on the accident at Palomares. We can anticipate requests for additional information.



CLAVE	CONTAMINACION SUPERFICIAL KBq /m ²	SUPERFICIE AFECTADA Ha
	> mos de 1200	2,2
	120 - 1200	17
	12 - 120	87
	menos de 12	120



Estación = Station
 Parcela = Parcel
 Impacto = Impact

Radiological Summary

- o 226 hectares (about 560 acres) of land was contaminated with Pu.
- o Radiological cleanup consisted of the following:
 - Removed all Pu contaminated cultivated and natural vegetation for disposal;
 - Removed 10 centimeters of soil with surface contamination greater than 1,200 kBq/m² (about 5.5 acres);
 - Deep plowed all arable land with surface contamination less than 1,200 kBq/m²; and
 - Contaminated surfaces of houses were washed with pressurized water and detergents.
- o Radiological Followup
 - DOE and its predecessors have been funding about 20 percent of the annual cost to monitor soil, air, vegetation, and people;
 - Recent monitoring of Pu in soil shows hot spots over an area of about 100,000 m²;
 - Soil samples from hot spots had Pu levels from 0.7 to 2.6kBq per gram (20,000 to 70,000 pCi/g);
 - Results of sampling Pu in air showed 100 percent of the results from three sampling stations had concentrations above the detection limit;
 - About 60 percent of vegetation sampled had Pu concentrations in the detectable range;
 - 146 residents of Palomares were taken to Madrid during 1985. 600 residents have been checked at least once for Pu internal contamination and given a medical examination;
 - Pu internal contamination has not been detected in 516 residents that have been checked;
 - Pu internal contamination has been detected in 52 residents;
 - The maximum committed effective dose equivalent derived from urine excretion of Pu were as follows:

22 persons	$\frac{\text{mrem}}{< 5,000}$
22 persons	5,000 to 10,000
8 persons	10,000 to 20,000

Risk estimates for the above exposures have not been published to date.

- o As a result of a technical review conducted for DOE in 1979, ORNL was requested to assume responsibility for coordinating the technical aspects of Indalo and to assist Spanish scientists. Funding for ORNL began in FY 1982.
- o Dr. Richmond, ORNL project manager, established a technical advisory group for the project that meets annually.
- o Dr. Emilio Iranzo has reported that he and his staff have expended much effort responding to the concerns of Palomares residents and their leaders. Their concern is a 20 year statute of limitations in Spain. The 20th anniversary of the accident was January 17, 1986. Statements have been made in the Spanish press that U.S. support will end now that the 20 years has passed.

Funding and Program Options

- o Starting in 1966, funding for Indalo was provided by the AEC Division of Biology and Medicine and, later, by the Office of Health and Environmental Research (OHER). When OHER was transferred to Energy Research (ER), Project Indalo was moved to the Division of Operational Safety under Environment. This was proper because Indalo is a health protection not a research program.
- o DOE funding for Project Indalo has been in two parts. There are funds to ORNL for project coordination, technical support, training for Spanish scientists, and scientific equipment. There is also funding directly to the Spanish government that amounts to about 20 percent of their costs for radiological monitoring of the Palomares residents and their environment.
- o Funding requested from the Office of Operational Safety (OOS) for Spain for FY 1986 (the Spanish fiscal year is from February to January) was 240K. For the first time in 20 years, an amount was put into the budget that was less than requested, i.e., 140K. Only 140K has been obligated.
- o By memorandum of June 13, 1986, the Director, OOS, stated that funding for Indalo more appropriately falls under the mission of the Office of Nuclear Safety (ONS) and that this was the final year of funding by OOS. There were no items in the ONS budget for ORNL and Spain for FY 1987. The amounts needed for FY 1987 are the same as for FY 1986.
- o The consequences of a reduction or termination of U.S. support are unknown but could be significant. Spain could seek U.S. funding to cover the entire cost of their followup operations.
- o Options:
 1. Extend the contract period to January 31, 1988, for Spain, fund Spain and ORNL for the full amount needed for FY 1987, i.e., 240K and 140K, and provide an additional 100K for Spain as was requested for support through January 31, 1987; or
 2. Provide interim funding until transfer of the program to the Office of Health and Environmental Research or to Defense Programs can be accomplished.
- o Advice:
 1. Do not reduce funding abruptly without advanced notice to the Spanish and to ORNL; and
 2. If support for the program is to be significantly altered, more attention must be given to the likely consequences and DOE management briefed on the impact.

● Paper

AIR CONCENTRATIONS OF ^{239}Pu AND ^{240}Pu AND POTENTIAL RADIATION DOSES TO PERSONS LIVING NEAR Pu-CONTAMINATED AREAS IN PALOMARES, SPAIN

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Abstract—On 17 January 1966, an accident during a refueling operation resulted in the destruction of an air force KC-135 tanker and a B-52 bomber carrying four thermonuclear weapons. Two weapons, whose parachutes opened, were found intact. The others experienced non-nuclear explosion with some burning and release of the fissile fuel at impact. Joint efforts by the United States and Spain resulted in remedial action and a long-term program to monitor the effectiveness of the cleanup.

Air concentrations of ^{239}Pu and ^{240}Pu have been continuously monitored since the accident. The average annual air concentration for each location was used to estimate committed dose equivalents for individuals living and working around the air sampling stations. The average annual ^{239}Pu and ^{240}Pu air concentrations during the 15-y period corresponding to 1966-1980 and the potential committed dose equivalents for various tissues due to the inhalation of the ^{239}Pu and ^{240}Pu average annual air concentration during this period are shown and discussed in the report.

1. INTRODUCTION

ON 17 January 1966, there was an aviation accident above the town of Palomares (Cuevas de Almanzora) in south-eastern Spain. As a consequence of this accident, four thermonuclear bombs carried by one of the planes fell, and at impact, the nuclear fuel in two of them partially ignited. This gave rise to the formation of an aerosol, which in varying degrees of magnitude, contaminated approximately a 226-hectare area of underbrush, farmland and urban areas.

The contaminated region and the full extent of the areas corresponding to the different levels of resulting α -radiation contamination measured in the period immediately following the accident are shown in Fig. 1. The points of impact of the two thermonuclear devices (which due to the numbering assigned them, correspond to bombs 2 and 3, respectively), are indicated in Fig. 1. The other two bombs were recovered intact, one in the dry river bed near the mouth of the Almanzora River and the other one in the sea.

Where the total soil surface α -contamination values were greater than $1.18 \text{ MBq} \times \text{m}^{-2}$ ($32 \mu\text{Ci} \times \text{m}^{-2}$), the contaminated vegetation and surface layer of soil (approximately 10 cm deep) were collected, separated and disposed of as radioactive waste. The rest of the contaminated area, devoted to agriculture, was irrigated thoroughly, plowed to a depth of approximately 30 cm, and subjected to a light homogenization to reduce the con-

centration of radionuclides contaminating the soil surface by diluting them in noncontaminated (below the surface) soil. This procedure reduced the α concentration to levels considered sufficiently safe, so there would not be any unacceptable existing Pu-isotope inhalation risks from resuspension to people living in this area or farming the land.

At the time the decontamination operations in the area were finished by removing the wastes and restoring the farmland, a research program was begun.

To maintain constant reference for the various proposed studies on air, vegetation, soil and persons in the Palomares area, the area contaminated during the accident has been subdivided into three zones. These zones correspond respectively to the two which were greatly influenced by contamination arising from each of the thermonuclear bombs that broke up and partially ignited and to the urban zone in which 90% of the population lives. The two zones corresponding to the impact points have been called 2 and 3, in accordance with the numbering of the bombs which fell in them; the zone containing the urban area has been labelled 5. In Fig. 1, the relative location of each one of these zones and its boundaries are shown.

The purposes of our study of air contamination in the area include:

- Measurement of existing ^{239}Pu and ^{240}Pu concentrations in the air of some zones of the area.

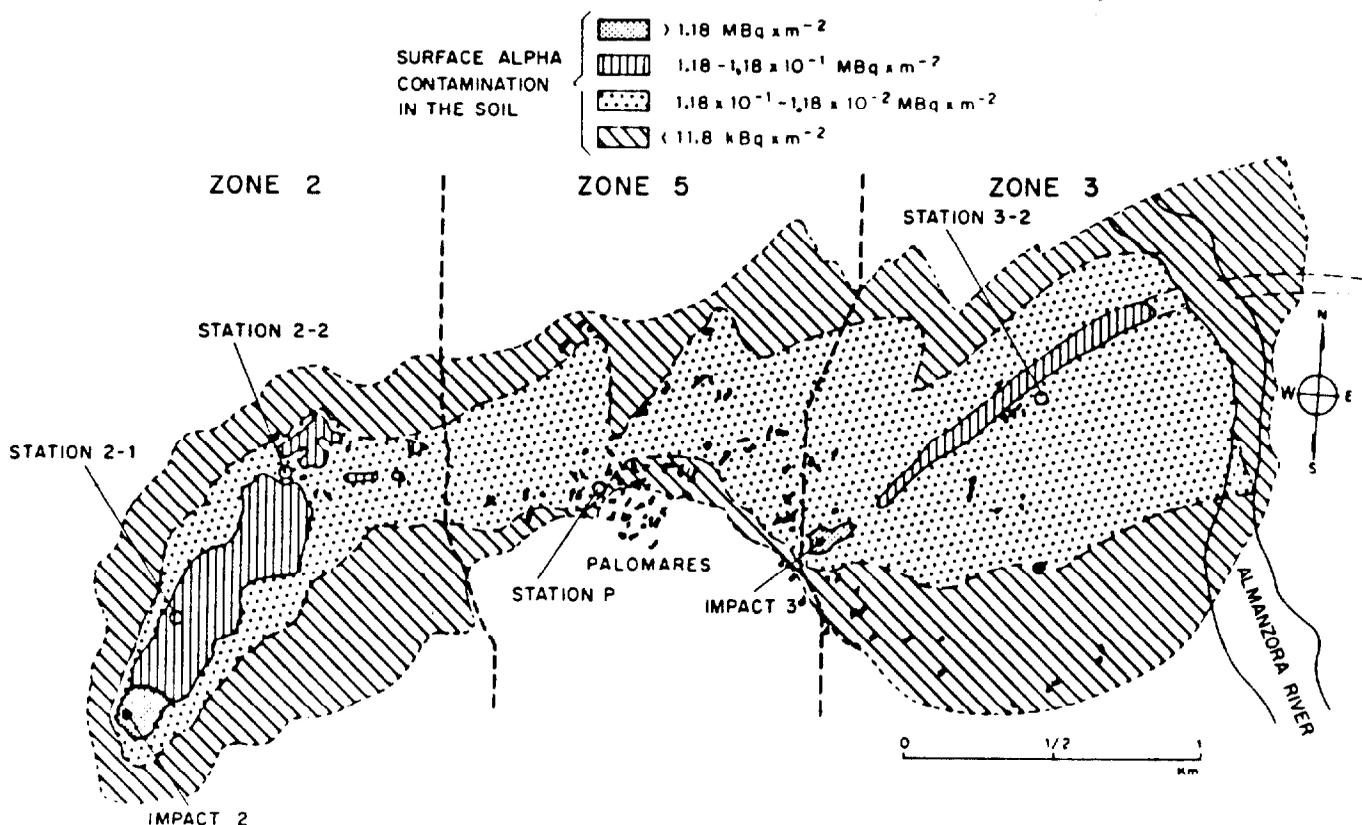


Fig. 1. Palomares area with original surface contamination levels and location of air sampling stations. Black spots show the location of houses.

- Determination of the inhalation risk for people living and farming in the area, and as a consequence, the internal radiation risk for permanent inhabitants, including children.

- Practical deduction about whether the decontamination and restoration actions in the area were correct, and consequently, whether the theoretical reasoning used to estimate the future residual contamination risk was adequate.

In this report the following matters are presented and discussed:

- Average annual ^{239}Pu and ^{240}Pu air concentrations during the 15-y period corresponding to 1966–1980.

- Potential committed dose equivalents for various tissues due to the inhalation of ^{239}Pu and ^{240}Pu average annual air concentration as an aerosol of $1\ \mu\text{m}$ (AMAD) by the ICRP Reference Man during the 15-y period 1966–1980.

2. SAMPLING AND ANALYTICAL PROCEDURES

2.1. Air sampling

Throughout the area, four sampling stations were set up—two in zone 2, because it is the most extensive and

has greater topographical variation, and one in zone 5, which corresponds to the town, and another in zone 3. In Fig. 1, the location of each is shown. The number and location of the air sampling stations was selected by taking into account the size of the area, its topographical characteristics and the dwelling house distribution.

Characteristics of the area surrounding the four sampling stations are as follows:

Station 2-1. The station was located in the small valley whose boundaries are the hills around the point of impact of bomb no. 2. The soil of the nearby surrounding area is rocky and has little brush or other wild plants. Because of these characteristics, plowing was not possible. The station was located in the center of an area in which the surface contamination levels originating from the accident ranged, at that time, from $1.18\ \text{MBq} \times \text{m}^{-2}$ ($32\ \mu\text{Ci} \times \text{m}^{-2}$) to $1.18 \times 10^{-1}\ \text{MBq} \times \text{m}^{-2}$ ($3.2\ \mu\text{Ci} \times \text{m}^{-2}$).

Station 2-2. This station is located on a parcel of land devoted to farming and near a small group of dwelling houses. In this parcel and its environs, surface contamination levels were between $1.18 \times 10^{-1}\ \text{MBq} \times \text{m}^{-2}$ ($3.2\ \mu\text{Ci} \times \text{m}^{-2}$) and $1.18 \times 10^{-2}\ \text{MBq} \times \text{m}^{-2}$ ($0.32\ \mu\text{Ci} \times \text{m}^{-2}$). However, surface contamination levels up to $1.18 \times 10^{-1}\ \text{MBq} \times \text{m}^{-2}$ ($3.2\ \mu\text{Ci} \times \text{m}^{-2}$) occurred in some nearby areas.

Station P. This station is located in the town and is considered representative of the air breathed by people living in the urban center, as it is in a place that is open to the prevailing winds, which come from the contaminated sectors. The surface contamination in the parcel in which it is located, and those around it, was less than $11.8 \text{ kBq} \times \text{m}^{-2}$ ($0.32 \text{ } \mu\text{Ci} \times \text{m}^{-2}$).

Station 3-2. This station was located in the center of a flat strip of cultivated land, which, if the surrounding land near the point of impact is not considered, would have greater contamination due to the impact of bomb no. 3. It is 5 m below the level at which the impact occurred. Surface α contamination of parcels near the sampling station ranged between $1.18 \text{ MBq} \times \text{m}^{-2}$ ($32 \text{ } \mu\text{Ci} \times \text{m}^{-2}$) and $11.8 \text{ kBq} \times \text{m}^{-2}$ ($0.32 \text{ } \mu\text{Ci} \times \text{m}^{-2}$).

At station 2-2 and P, air samples were taken daily throughout the period 1966–1980; at stations 2-1 and 3-1, samples were collected every day during the period from July 1966 to October 1969. Collecting samples from stations 2-1 and 3-1 were discontinued because of frequent disturbances in the wiring to their power supplies due to farming practices.

Air sampling is continuing and, since 1981, determination of ^{241}Am has been included in the analysis of the samples.

The sampling stations consist of a filter holder, a 1425 rpm, one-third horsepower suction pump, a gas meter for measuring the air volume, and a voltage regulator. Except for the filter holder, this equipment is inside a small wooden booth which protects it from the weather. The filter holder was placed at a height of 1.70 m above the ground.

Filters used are of a cellulose type having a 47-mm diameter and 0.8- μm mesh; the daily aerosol sample taken corresponds to an average air volume of 90 m^3 , for the 24-h period.

2.2. Analyses

Analyses were performed on composite samples, corresponding to a period of 10 d, for each of the sampling stations. This means that the spectrometric measurement of each compound sample corresponds to an air volume of about 900 m^3 .

The radioanalytical technique used to determine the concentration of ^{239}Pu and ^{240}Pu in the aerosol samples basically includes Pu dissolution using nitric acid, separation by ionic exchange, electroplating on stainless steel planchets and α -spectrometric measurement of the α activity.

The analytical technique is as follows:

Filters corresponding to the aerosol samples for each 10-d period are subjected to concentrated nitric acid wet digestion at temperatures of 250°C and 300°C until a white-colored residue is obtained; homogeneous heating is carried out in Al blocks. To calculate the efficiency of the procedures for each sample, 1 mL of standard ^{236}Pu solution, in the early years, and later a ^{242}Pu solution which contains 50 mBq (1.35 pCi) was added.

The residue is dissolved in 50 mL of 8 M nitric acid solution, and then the solution is passed through ion exchange resin* (50–80 mesh) in chloride form, at a flow rate on the order of 1 mL/min. During this phase of the analyses, the Pu and Th are retained in the resin.

To eliminate the small amount of U that the resin might have retained, washing is carried out several times with 8 M nitric acid until a total 150-mL volume is obtained. To eliminate the Th retained in the resin, the column is washed once with 12 N HCl and the wash effluent is discarded.

Some grains of hydroxylammonium-chloride are added to the resin column and the Pu fixed in the resin is eluted by means of three washes with 5 mL of 0.5 N HCl. The effluents are dried at a temperature not to exceed 80°C , dissolved again in the concentrated HCl and are dried again at the same temperature as before. These dried residues are dissolved in 1 N hydrochloric acid.

Four percent ammonium oxalate is added to the 1 N hydrochloric acid solution and it is subjected to electrolysis. The Pu is electroplated onto the 14-mm-diameter stainless steel planchet at a power of 20 V and a current intensity of 200 mA for 3 h.

The planchets are measured by α spectrometry with Si barrier detectors having an active area of 300 mm^2 and a multichannel pulse height analyzer. The counting efficiency of the equipment used is on the order of 31%.

The detection limit of the method is $1.8 \text{ } \mu\text{Bq} \times \text{m}^{-3}$ ($5 \times 10^{-2} \text{ fCi} \times \text{m}^{-3}$), which is equivalent to 10^{-5} of the derived air concentration (DAC) for ^{239}Pu and ^{240}Pu compounds of Class Y to radiation workers.

3. RESULTS AND DISCUSSIONS

The results of ^{239}Pu and ^{240}Pu concentrations in samples taken at the air sampling stations and the potential committed dose equivalents to various tissues in the scenario of continuous exposure to inhalation of the average annual concentrations by the dwellers of Palomares are discussed below.

3.1. Air concentration of ^{239}Pu and ^{240}Pu

In accordance with the annual limit on intake (ALI), the DACs for ^{239}Pu and ^{240}Pu recommended by the International Commission on Radiological Protection (ICRP) in its *Publication No. 30*, for radiation workers exposed 2000 h per working year are the following:

Class Y compounds: $200 \text{ mBq} \times \text{m}^{-3}$ ($5.4 \text{ pCi} \times \text{m}^{-3}$)

Class W compounds: $80 \text{ mBq} \times \text{m}^{-3}$ ($2.16 \text{ pCi} \times \text{m}^{-3}$).

As the ICRP still has not established ALI and DAC values for the public and has suggested the use of a fiftieth or a hundredth of the values for established radiation workers, we have decided to use values that could be more

* Dowex AG-1X-2, Bio. Rad Laboratories, 32nd & Griffin, Richmond, CA.

or less equivalent to those that might be recommended in the future by the ICRP.

People in the Palomares area should be considered individually among the continuously exposed public, that is, $8,760 \text{ h y}^{-1}$.

Taking into consideration the ratio between annual dose limits recommended for workers and members of the public by the ICRP, 10:1, and the continuous exposure of the public in the area (an adult Reference Man breathing volume = $23 \text{ m}^3 \times \text{d}^{-1}$), it has been calculated that the DACs of ^{239}Pu and ^{240}Pu for members of the public are the following:

$$\text{Class Y: } 5.9 \text{ mBq} \times \text{m}^{-3} \text{ (0.16 pCi} \times \text{m}^{-3}\text{)}$$

$$\text{Class W: } 2.4 \text{ mBq} \times \text{m}^{-3} \text{ (0.06 pCi} \times \text{m}^{-3}\text{)}$$

Because we are dealing with people living in an urban zone and some of them will continue to live there for their lifetime in agricultural work, we applied the ICRP recommendation that, in such cases, the annual effective dose equivalent limit should be reduced so that the effective dose equivalent throughout their lives corresponds, at a maximum, to the value resulting from receiving a 1-mSv (0.1 rem) mean annual effective dose equivalent rate. Consequently, we adopted the following values for the DAC, which we designate DAC (general public) in this report.

$$\text{Class Y: } 1.2 \text{ mBq} \times \text{m}^{-3} \text{ (32 fCi} \times \text{m}^{-3}\text{)}$$

$$\text{Class W: } 0.5 \text{ mBq} \times \text{m}^{-3} \text{ (13 fCi} \times \text{m}^{-3}\text{)}$$

We believe that the Pu aerosols are found as oxides, PuO_2 , such as were formed during the burning of the thermonuclear bombs at impact. Consequently, the conclusions, insofar as real existing risk is concerned, will be those derived from the concentration of ^{239}Pu and ^{240}Pu and the DAC for Class Y compounds.

A comparative study to consider the presence of Class W compounds was conducted to assure ourselves that maximum protection was being provided.

The analyses of the values obtained for the ^{239}Pu and ^{240}Pu concentrations in the air samples taken in the Palomares area during the period 1966–1980 and their comparison with the DAC limits set forth in the preceding paragraphs allow us to make the following observations and conclusions:

(a) In the air samples taken at the four sampling stations, the frequency of ^{239}Pu and ^{240}Pu particles in concentrations exceeding the detection limit, $1.8 \mu\text{Bq} \times \text{m}^{-3}$ ($0.05 \text{ fCi} \times \text{m}^{-3}$), are shown in Fig. 2. From the values shown in Table 1, it can be concluded that:

- Plutonium-239 and ^{240}Pu particles are resuspended in the air. The presence of such particles has been established in samples taken at the four stations.

- The frequency of the existence of Pu has been varying as a function of time for each one of the sampling stations.

- The sampling station location with regard to residual contamination, taken as the average annual percentage of concentrations greater than the detection limit during the period 1966–1969, was 37.0 in the urban zone

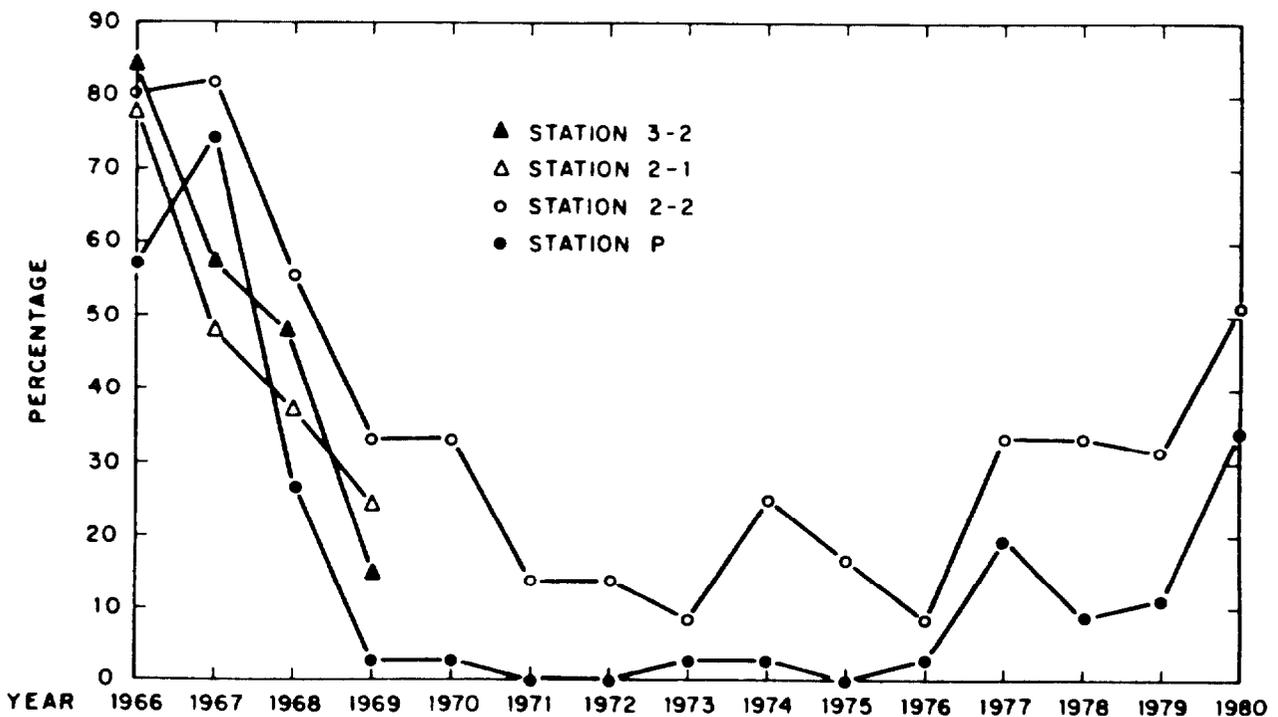


Fig. 2. Percentage of air samples with a ^{239}Pu and ^{240}Pu concentration exceeding the detection limit.

Table 1. Percentage of air samples with a ^{239}Pu and ^{240}Pu concentration exceeding the detection limit ($1.8 \mu\text{Bq} \times \text{m}^{-3}$) during the period 1966–1980

YEAR	AIR SAMPLING STATION			
	2-1	2-2	P	3-2
1966	77.8	80.0	57.1	84.2
1967	47.8	81.8	74.3	57.1
1968	37.1	55.6	26.5	48.0
1969	24.0	33.3	2.8	14.8
1970		33.3	2.8	
1971		13.9	0	
1972		13.9	0	
1973		8.3	2.8	
1974		25.0	2.8	
1975		17.1	0	
1976		8.3	2.8	
1977		33.3	19.4	
1978		33.3	8.8	
1979		31.4	11.1	
1980		51.4	34.3	
TOTAL	42.7	33.0	14.7	49.1
TOTAL 1966-1969	42.7	60.0	37.0	49.1

(station P), while at station 2-2, it was 60.0; values corresponding to these average annual percentages at stations 2-1 and 3-2 were 42.7 and 49.1, respectively.

• For samples taken at station 2-2 and station P during the period 1966–1980, the average annual percentage of concentrations greater than the detection limit was 33 at station 2-2, while at station P it was 14.7.

At station 2-2 and station P, the maximum values for the sampling percentage with a ^{239}Pu and ^{240}Pu content in excess of the detection limit occurred in 1967, with values of 81.8 and 74.3, respectively. At station 2-2, minimum percentages of samples having a ^{239}Pu and ^{240}Pu content in excess of the detection limit occurred in 1973 and 1976, with a value of 8.3 in both cases. As of 1977, an increase in values began and reached a value of 51.4% by 1980. This means that as of 1977, the resuspension of existing ^{239}Pu and ^{240}Pu in the soil of zone 2 increased, as evidenced by the fraction of measurements above the detection limit at station 2-2. The resuspension increase was, as can be proven, related to earth moving without previous watering on farmlands located in parts of zone 2, which either had not been cultivated since the accident or never had been cultivated, because it is hilly terrain covered with poor-quality, wild vegetation.

As of 1968, the number of samples at station P with a ^{239}Pu and ^{240}Pu content greater than the detection limit diminished considerably, so that between 1969 and 1976, percentages of samples with a content exceeding the de-

tection limit were between 2.8 and 0. As of 1977, and in the case of station 2-2, an increase in these percentages occurred, reaching a value of 34.3 in 1980, which we think was due to earth movement, especially in zones 2 and 5, in order to convert uncultivated land into cultivated land and to build greenhouses.

(b) Average annual concentrations of ^{239}Pu and ^{240}Pu have been assessed via the 10-d composite samples taken every year at each sampling station. The values for the average annual ^{239}Pu and ^{240}Pu air concentration during the 15-y period 1966–1980 for stations 2-2 and P are those shown in Table 2. Average annual concentrations for stations 2-1 and 3-2 during the period 1966–69 are also shown.

To be conservative, we assumed that all Pu is respirable with an AMAD of $1 \mu\text{m}$ and no account has been made for the Pu in the sample arising from nuclear fallout.

From the air concentration values shown in Table 2, we can arrive at the following:

- Most of the average annual concentrations of ^{239}Pu and ^{240}Pu in the air have been less than a tenth of the calculated DAC (general public) of Class Y Pu compounds [$1.2 \text{ mBq} \times \text{m}^{-3}$ ($32 \text{ fCi} \times \text{m}^{-3}$)], at all the sampling stations and in all years studied. The exceptions correspond to air sampling station 2-2 (1967 and 1969) and air sampling station 2-1 (1969).

- With the exception of 1 y (1967) at station 2-2, the average concentration values have also been less than the DAC (general public) values for Class W Pu compounds [$0.5 \text{ mBq} \times \text{m}^{-3}$ ($13 \text{ fCi} \times \text{m}^{-3}$)].

- At station P, whose values are representative of the exposure of the Palomares dwellers, the maximum

Table 2. Average annual concentrations of ^{239}Pu and ^{240}Pu in the air during the period 1966–1980

YEAR	CONCENTRATION AT THE STATION ($\mu\text{Bq} \times \text{m}^{-3}$)			
	2-1	2-2	P	3-2
1966	41.8	44.8	14.8	27.4
1967	15.2	441.8	4.1	13.0
1968	7.0	21.8	2.6	3.3
1969	161.0	142.1	2.6	14.1
1970		5.9	2.2	
1971		2.2	<1.8	
1972		10.4	<1.8	
1973		3.0	2.2	
1974		8.1	4.1	
1975		16.3	<1.8	
1976		4.4	<1.8	
1977		11.8	5.6	
1978		16.7	2.2	
1979		19.2	5.6	
1980		32.9	28.1	

average annual concentration was in 1966, $14.8 \mu\text{Bq} \times \text{m}^{-3}$ ($0.4 \text{fCi} \times \text{m}^{-3}$), and then a decline occurred, which for the period 1968 to 1976 remained more or less constant.

As of 1977, the annual concentration of activity increased, and in 1980 it reached the maximum values for the entire period, $28 \mu\text{Bq} \times \text{m}^{-3}$ ($0.76 \text{fCi} \times \text{m}^{-3}$). This increase was, as previously indicated, related to earth movement without previous watering to reshape a part of the hilly terrain with residual surface contamination corresponding to zone 2 and some tracts of uncultivated land corresponding to zone 5 into cultivable parcels.

Values from station 2-2 are representative of the double influence of working contaminated land (resuspension) and of some persistence of some of the original contamination in the hilly zone, because the slope and large number of rocks prevented plowing during the original remedial action activities. The maximum average annual concentration occurred in 1967, with a value of $442 \mu\text{Bq} \times \text{m}^{-3}$ ($11.9 \text{fCi} \times \text{m}^{-3}$), coinciding with renewed cultivation of the zone after the accident. Between 1970 and 1976 stabilization of the activity concentration occurred with relatively small fluctuation. Beginning in 1977, and as in the case of station P, the annual concentration of activity increased again, reaching a value of $33 \mu\text{Bq} \times \text{m}^{-3}$ ($0.89 \text{fCi} \times \text{m}^{-3}$) in the year 1980.

Values for the ^{239}Pu and ^{240}Pu air concentration corresponding to the entire period, and especially the 4 y during the period 1977-1980, were generally greater at station 2-2 than at the others. These were related to the residual contamination in the hilly area and to proximity to the area in which earth movement occurred as of 1977.

The maximum value for a single 10-d ^{239}Pu and ^{240}Pu composite air concentration corresponds to station 2-2, $9.3 \text{mBq} \times \text{m}^{-3}$ ($0.25 \text{pCi} \times \text{m}^{-3}$); this value occurred during the month of August 1967, and was the only one exceeding the DAC for the public of Class Y Pu compounds [$5.9 \text{mBq} \times \text{m}^{-3}$ ($0.16 \text{pCi} \times \text{m}^{-3}$)].

Concentrations exceeding the DAC (general public) of Class Y Pu compounds [$1.2 \text{mBq} \times \text{m}^{-3}$ ($32 \text{fCi} \times \text{m}^{-3}$)] only have been produced during 1966-69 in four 10-d periods, three of which corresponded to station 2-2 and one to station 2-1. The latter occurred in 1969, and the others corresponding to station 2-2 in the years 1967 (two 10-d periods) and 1969 (one 10-d period). It should be recalled that the DAC (general public), as used in this paper is about 170 times below that used by ICRP for workers.

Concentrations exceeding the DAC (general public) of Class W Pu compounds occurred during the period 1966-69 in five 10-d periods. Of these, one corresponded to station 2-1, and four to station 2-2. During the period 1970-1980 only on one occasion, at station 2-2, was this DAC (general public) exceeded.

• The mean values for average annual concentrations of ^{239}Pu and ^{240}Pu in air for each of the sampling stations during the period 1966-69 were the following:

Station 2-1: $56 \mu\text{Bq} \times \text{m}^{-3}$ ($1.5 \text{fCi} \times \text{m}^{-3}$)

Station 2-2: $160 \mu\text{Bq} \times \text{m}^{-3}$ ($4.4 \text{fCi} \times \text{m}^{-3}$)

Station P: $6 \mu\text{Bq} \times \text{m}^{-3}$ ($0.18 \text{fCi} \times \text{m}^{-3}$)

Station 3-2: $14 \mu\text{Bq} \times \text{m}^{-3}$ ($0.39 \text{fCi} \times \text{m}^{-3}$).

The average air concentration values of ^{239}Pu and ^{240}Pu during the entire period 1966-1980 were the following:

Station 2-2: $52 \mu\text{Bq}$ ($1.4 \text{fCi} \times \text{m}^{-3}$)

Station P: $5.5 \mu\text{Bq}$ ($0.15 \text{fCi} \times \text{m}^{-3}$).

By comparing the average concentration values during each of the periods studied at each of the sampling stations with the DAC values, one can conclude that:

- In the urban area, P, the average concentration, both for the period 1966-69 as well as the period 1966-1980, has been below a hundredth of the DAC (general public) for Class Y Pu compounds and below one seventieth of the DAC (general public) for Class W Pu compounds.

- In the farming zone, 2-2, the average concentration for the period 1966-1980 was below a tenth of the DAC (general public) for Class Y Pu compounds and on the order of a tenth of the corresponding DAC (general public) for Class W Pu compounds. During the period 1966-69, the average concentration also was below the DAC (general public) for Class Y and Class W compounds.

- During the period 1966-69, the average concentration in sampling stations 2-1 and 3-2 was below a tenth of the DAC (general public) of Class Y Pu compounds and below the DAC (general public) of Class W Pu compounds.

3.2. Potential committed dose equivalents to people

For sampling stations P and 2-2, which are representative of the risk in the urban center and the most significant farming area (critical group zone), we have calculated the committed dose equivalents received by the assumed inhalation of the average annual ^{239}Pu and ^{240}Pu concentrations shown in Table 2 for the period 1966-1980. The committed dose equivalents to various organs and tissues have been calculated by means of the factors, Sv/Bq, for committed dose equivalents per unit activity of inhalation intake recommended by the International Commission on Radiological Protection (ICRP79b) and K. F. Eckerman (Ec81).

The potential committed dose equivalent for adults and a mean aerosol size (AMAD) of $1 \mu\text{m}$ are shown in Figs. 3 and 4 for each year. In Fig. 3 the committed dose equivalents to lungs, bone surface, liver, red bone marrow, and gonads of people in the urban zone are given. In Fig. 4 the values for people in the cultivated zone with the greatest potential risk, station 2-2, are shown.

From the discussion of values calculated for these organs the following has been deduced:

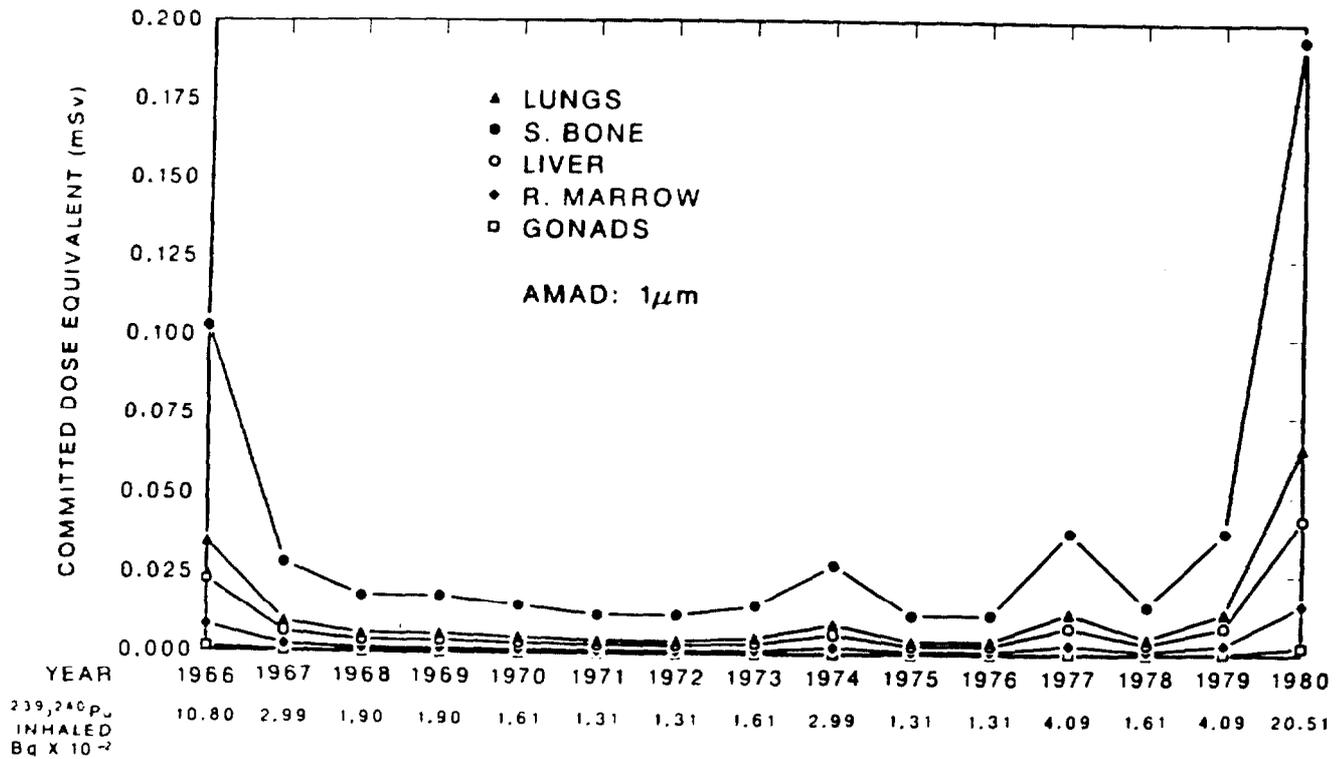


Fig. 3. Potential committed dose equivalents for adults in urban area from inhaling the average annual ^{239}Pu and ^{240}Pu concentration during the period 1966–1980.

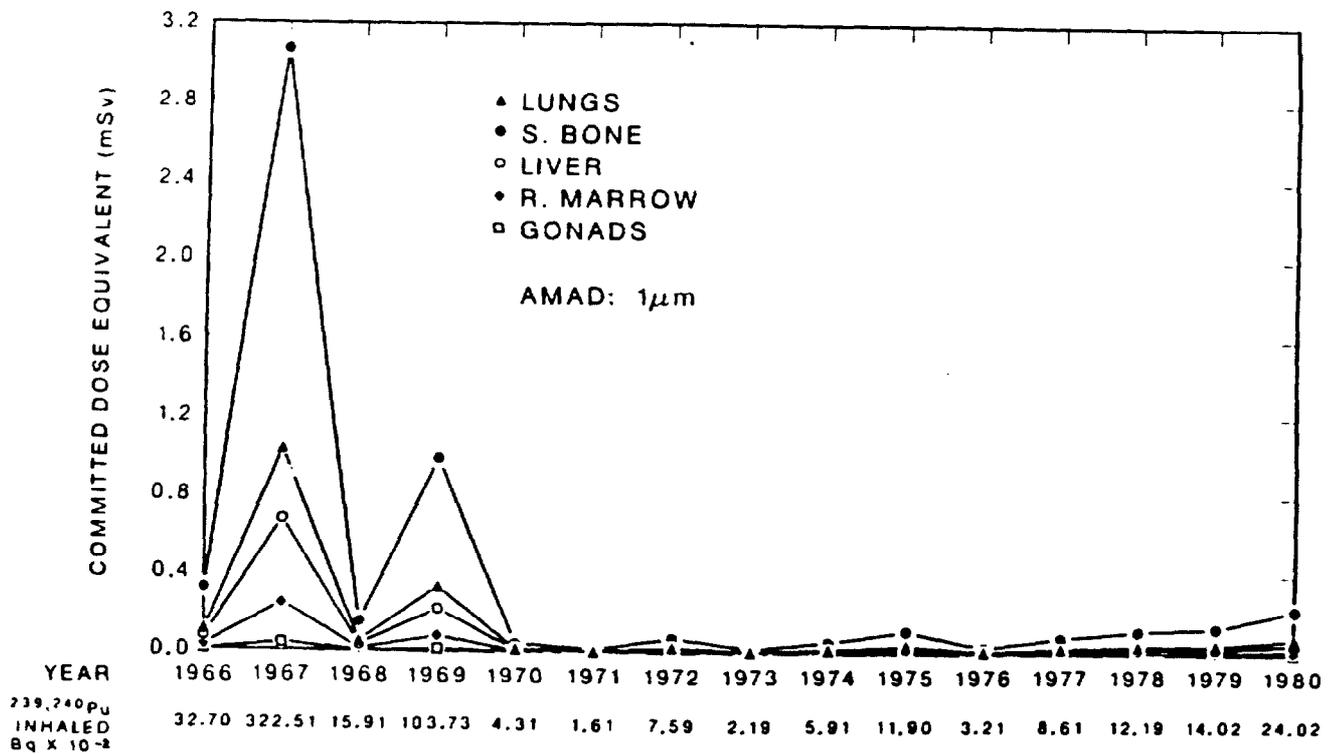


Fig. 4. Potential committed dose equivalents for adults in the "Critical Group" area from inhaling the average annual ^{239}Pu and ^{240}Pu concentration during the period 1966–1980.

• The potential committed dose equivalent to bone surfaces is the highest and the summation of the committed dose equivalents for each of the 15 y during the period 1966–1980 shows a value of 0.5637 mSv (56.37 mrem) for people in the urban zone and 5.4187 mSv (541.9 mrem) for people in the zone around station 2-2. Its contribution to the committed effective dose equivalent to people during the 15 y will be 0.0169 mSv (1.69 mrem) and 0.1625 mSv (16.25 mrem) at each zone, respectively.

The total potential committed dose equivalents for the other organs and their contribution to the committed effective dose equivalent during the period 1966–1980, at each zone, are shown in Tables 3 and 4 respectively.

• The ICRP has recommended a value of 1 mSv (0.1 rem) for the effective annual dose equivalent limit for individuals in the population who might be exposed over a long period of their lives. Bearing this in mind, the annual limiting dose equivalent value for each particular organ, considering its exposure to the exclusion of the others and keeping in mind the organ weighting factor for stochastic effects, is 33.33 mSv (3333 mrem) for the bone surface, 8.33 mSv (833 mrem) for the lungs, 16.67 mSv (1667 mrem) for the liver, 8.33 mSv (833 mrem) for red bone marrow and 4 mSv (400 mrem) for gonads.

On comparing the values of the committed dose equivalents calculated for each organ with those deduced and presented in the previous paragraph for the separate exposure of each organ, and in considering the exclusion of the others, we can observe that the total committed dose equivalent values for each organ during the 15-y period are less than the annual limits for the organ.

• The contribution of the committed dose equivalents in the five organs to the potential committed effective dose equivalent to people during the 15-y period 1966–1980 is 0.0544 mSv (5.44 mrem) in the urban area and 0.5224 mSv (52.24 mrem) in the area which corresponds to the critical group.

4. CONCLUSIONS

From the discussion of the results given in the previous sections, the following conclusions can be deduced:

(1) With the exception of the urban zone during 1971, 1972, and 1975, at all sampling stations and for all the years studied, ^{239}Pu and ^{240}Pu concentrations in the

Table 3. Total potential committed dose equivalents for organs during the period 1966–1980

ORGAN	TOTAL COMMITTED DOSE EQUIVALENT, mSv	
	URBAN ZONE (P)	STATION 2-2
BONE SURFACE	0.5637	5.4187
LUNGS	0.1902	1.8254
LIVER	0.1250	1.1978
RED BONE MARROW	0.0450	0.4336
GONADS	0.0071	0.0684

Table 4. Contribution of committed dose equivalents in organs to the total potential committed effective dose equivalent during the period 1966–1980

ORGAN	CONTRIBUTION TO COMMITTED EFFECTIVE DOSE, mSv	
	URBAN ZONE (P)	STATION 2-2
BONE SURFACE	0.0169	0.1625
LUNGS	0.0228	0.2190
LIVER	0.0075	0.0718
RED BONE MARROW	0.0054	0.0520
GONADS	0.0018	0.0171

air have been found on occasion to exceed the detection limit, $1.8 \mu\text{Bq} \times \text{m}^{-3}$ ($5 \times 10^{-2} \text{fCi} \times \text{m}^{-3}$). No corrections were made for the contribution to the measurements from worldwide nuclear weapons fallout.

The frequency of air samples containing ^{239}Pu and ^{240}Pu concentrations greater than the detection limit progressively diminished with time following the accident. During the period 1970–76, the frequency of measurements above the detection limit stabilized except for station 2-2 in 1974. During this period, agricultural operations were carried out normally throughout the zone, except for a hilly and uncultivated parcel near station 2-2; it was plowed and transformed into a cultivated parcel during 1974.

During 1977–1980, there was an increase at stations 2-2 and P in the frequency of samples with ^{239}Pu and ^{240}Pu concentrations in the air greater than the detection limit, especially at station 2-2. This increase was related to the major earth moving activities (leveling and terracing) of land that never had been cultivated (or at least, never after the accident) into cultivable land. This land corresponded to the hilly area in zone 2. Analogous changes were carried out during 1979 and 1980 on tracts of uncultivated land located in the zone near the urban center.

(2) The average annual ^{239}Pu and ^{240}Pu concentration in the air during the sampling periods corresponding to each of the stations has been below the DAC (general public) for Class Y Pu compounds [$1.2 \text{mBq} \times \text{m}^{-3}$ ($32 \text{fCi} \times \text{m}^{-3}$)]. Most of them have been below one tenth of the DAC (general public); the exceptions correspond to air sampling station 2-2 (1967 and 1969) and air sampling station 2-1 (1969).

(3) The average ^{239}Pu and ^{240}Pu concentration in air to which the public living in the urban zone has been exposed during the 15-y period (1966–1980) is less by a factor of 4.6×10^{-3} of the DAC (general public) of Class Y Pu compounds and by a factor of 1.1×10^{-2} for those of Class W.

In this urban zone the average annual ^{239}Pu and ^{240}Pu concentrations always were below those corresponding to the other sampling stations for the same year; maximum concentration was in 1980, with a value of $28 \mu\text{Bq} \times \text{m}^{-3}$ ($0.76 \text{fCi} \times \text{m}^{-3}$).

(4) At sampling station 2-2, located between cultivated parcels near the hilly area and which gave evidence of higher residual surface contamination, the highest average annual concentrations have occurred throughout the period, with the exception of 1969. The maximum concentration occurred in 1967, with a value of $442 \mu\text{Bq} \times \text{m}^{-3}$ ($11.9 \text{ fCi} \times \text{m}^{-3}$). Thus, most values of these average annual concentrations always were below a tenth of the DAC for the public of Class Y Pu compounds.

At this sampling station, the maximum Pu concentration in the air for the entire period was detected, and it had a value of $9.3 \text{ mBq} \times \text{m}^{-3}$ ($0.25 \text{ pCi} \times \text{m}^{-3}$), which corresponded to a 10-d sampling period during the month of August 1969.

(5) The contribution of the potential committed dose equivalents in the organs considered (lungs, bone

surface, liver, red marrow and gonads) to the potential committed effective dose equivalent received by people in the area by continuous inhalation during the period 1966–1980 is $5.44 \times 10^{-2} \text{ mSv}$ in the urban zone and $5.23 \times 10^{-1} \text{ mSv}$ (52.3 mrem) in the zone corresponding to the "critical group." These values correspond to 5.44% and 52.34% of the effective dose equivalent limit of 1 mSv (0.1 rem) per year, that is 0.36% and 3.48% of the total effective dose equivalent limit for a 15-y period.

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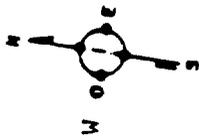
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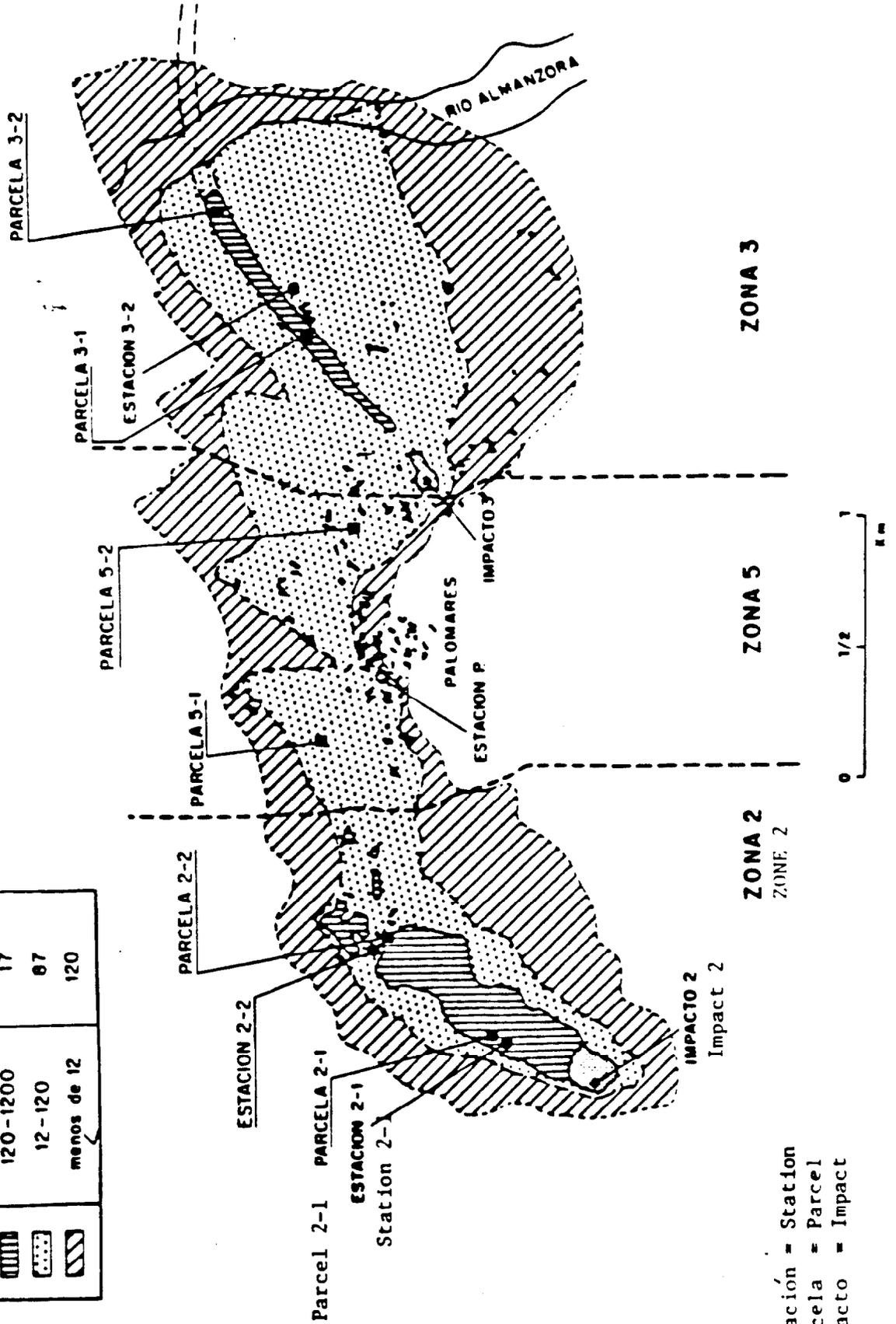
PROJECT INDALO BRIEFING

History of Accident at Palomares, Spain

- o On January 17, 1966, during a refueling operation, a U.S. Air Force KC-135 tanker and a B-52 bomber carrying four nuclear weapons collided and disintegrated over Palomares, Spain, a village of about 1500 people. One crew member survived.
- o Two nuclear weapons were recovered intact. Two others experienced detonation of the high explosives on impact. There was no nuclear yield and no one on the ground was seriously injured.
- o Two overlapping patterns of Plutonium-239 contamination were laid down over a predominantly agricultural area, but also including the village of Palomares.
- o Working cooperatively, representatives of the U.S. and Spanish Governments initiated a joint program for cleanup of the contaminated area. Agreement was obtained on cleanup methods and on cleanup radiological criteria.
- o Contaminated vegetation was collected for disposal. Contaminated soil was scraped into piles, loaded into trucks, and collected in a holding area where it was packaged for shipment to the United States for disposal. A large area was watered down and deep plowed.
- o The contaminated vegetation and soil from Palomares were transported by ship to Charleston harbor and transported to the Savannah River Plant for land burial.
- o In the Hall-Otero Agreement between the United States and Spain, the United States agreed to provide technical and financial assistance to Spain for radiological followup of Palomares residents and their environment. This assistance, Project Indalo, has continued for the past 20 years.
- o It was agreed that the findings for Indalo would be published by Spanish scientists. This has been done. No report on cleanup field operations appears to have been issued by U.S. participants. The fact that Palomares soil containing plutonium from U.S. weapons was buried at SRP has appeared in an ORNL report dated June 1985. Burial at SRP was also reported by Spanish scientists at an international symposium in Monaco in October 1966, and their report on this presentation contains this and other details on cleanup operations.
- o Dr. Emilio Iranzo, Director of Project Indalo in Spain, has submitted a report for publication in Health Physics entitled "Air Concentrations and Potential Radiation Doses to People Living Near Contaminated Areas in Palomares (Spain)." This should be published before the end of this year. This will be the first publication of exposure estimates for Palomares residents in a journal with a large readership. This will focus the attention of the radiation protection community (worldwide) on the accident at Palomares. We can anticipate requests for additional information.



CLAVE	CONTAMINACION SUPERFICIAL KBq / m ²	SUPERFICIE AFECTADA Mo
	> mos de 1200	2,2
	120 - 1200	17
	12 - 120	67
	menos de 12	120



Estacion = Station
 Parcela = Parcel
 Impacto = Impact