

PLUTONIUM CONTAMINATION TWENTY YEARS AFTER THE NUCLEAR WEAPONS ACCIDENT IN SPAIN

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INTRODUCTION

An accident involving two US Air Force planes engaged in a refueling operation occurred at 0922 GMT on January 17, 1966 over the town of Palomares near Cuevas de Almanzora within the province of Almeria in southeastern Spain. The mid-air collision (Figure 1) was followed by an explosion and pieces of the aircraft fell onto Palomares and neighboring Villaricos. Seven of the 11 crew members were killed and four thermonuclear weapons fell with the aircraft wreckage. Three of the bombs, one intact, were found on land, in or near Palomares, within 24 hours of the accident. Following an extensive search, the fourth was removed from the Mediterranean Sea on April 7. The parachutes of two of the bombs did not deploy resulting in the detonation of their conventional explosives and release of fissile material upon impact. Partial burning of the fissile material formed an aerosol that contaminated approximately 226 hectares (558 acres) of uncultivated, farmed, and urban land. Few people were working in the fields as most villagers were celebrating the festival of Saint Anthony, the patron saint of Palomares. Although no one was injured in Palomares, the explosions broke windows, cracked walls, and allegedly threw several people to the ground near impact point number 3 as shown in Figure 2.

The first weapon, only slightly damaged, was recovered southeast of Palomares, near the dry Almanzora riverbed, about 300 meters from the sea. This was designated as impact point number one. The second landed about one mile to the west of Palomares (impact point number 2). The third fell on the east edge of the village (impact point number 3). At the time of the accident a 30-knot wind was blowing from the west. The plutonium-bearing dust cloud from impact point 2 traveled across irrigated fields and the northern edge of the village. The cloud from impact point 3 traveled away from the village but across prime irrigated areas used for growing tomatoes, beans, and alfalfa. The last tomato crop of the season was just ready for harvest.

Personnel from the US and Spain began an assessment of the situation soon after the accident. Visible bomb fragments and pieces of the aircraft were collected and levels of alpha contamination established. The isopleths describing the surface Pu contamination were used as the basis for the cleanup operation. PAC-1S alpha detectors were used. Most of the houses in the village were not contaminated. Some were contaminated at low levels and

washed with high pressure water and detergents. Residual contamination was removed by hand and some houses were white washed.

The highest contamination levels were found on uncultivated land lying between low hills about 1500 meters SW of the village. Table I shows the remedial action taken following negotiations between the two governments. Soil was removed if the surface alpha concentration was more than 1200 kBq/square meter (32.4 uCi/square meter) and sent to the US for burial. Cultivated and natural vegetation with surface contamination greater than 1 kBq/square meter (27 nCi/square meter) was treated as radioactive waste. Arable land with levels below 1200 kBq/square meter (32.4 uCi/square meter) were wet down, plowed to a 30 cm depth, harrowed, and mixed. On rocky hillsides in area 2, where plowing was not possible, soil with Pu levels above 120 kBq/square meter (3.24 uCi/square meter) was removed by hand tools.

After the decontamination operations were completed, a program to study the effectiveness of the remedial action was established. The objectives of the program are shown in Table 2. Six 50 X 50 m study plots were established to allow the long-term investigation of residual Pu in soil and vegetation. Samples are taken periodically from nine points along a diagonal, equidistant from each other. These were designated as 2-1, 2-2, 5-1, 5-2, 3-1, and 3-2. Four air sampling stations were also constructed at locations near the study plots at 2-1, 2-2, 3-2, and in the village.

A meteorological station including one of the air sampling stations mentioned above was established in the village. In addition, a modern radiochemistry laboratory, including large radiation detectors for measuring low-energy photons in vivo, was set up in Madrid. Studies have been made since the accident on Pu levels in air, soil, vegetation and people. I shall summarize the results in each category.

AIR:

Table 3 shows the average air concentrations for four sampling stations. Stations P and 2-2 operated for the entire 15 year period; stations 2-1 and 3-2 were discontinued in late 1969. The calculated effective dose equivalent for people assumed to live at these locations are given. It is assumed that all the material is inhaled as one um diameter particles. The contribution of the committed dose equivalent in the five organs considered to the committed effective dose equivalent to people during the 15 year period was 0.0544 mSv (5.44 mrem) in the village and 0.5234 mSv (52.34 mrem) in area 2-2 corresponding to the critical group. These values are 0.07% and 0.7%, respectively, of the 75 mSv (7500 mrem) dose for the 15 year period that is calculated on the basis of a of 5 mSv/y (500 mrem/y) dose limit. During the same period of time a person would receive an effective dose equivalent of about 27 mSv (2700 mrem) from natural sources.

The values in Table 3 also correspond to 5.44% and 52.34% of the effective dose equivalent limit of 1 mSv (0.1 rem) per year established by the

International Commission on Radiological Protection for continuously exposed populations ; that is 0.36% and 3.48% of the total effective dose equivalent limit for a 15-year period.

For comparison, Table 4 gives the Pu concentrations in air for New York City, northwestern Italy and two locations in Palomares for the nine year period beginning in 1966. There is rather good agreement among all locations except area 2-2 which for which Pu concentrations are higher. Also of interest is the increase at all locations in 1974. Data have been collected continuously since 1966 for the two locations in Palomares. The data for the period 1966 - 1980 has been summarized in a 1987 publication.

VEGETATION:

Vegetation was measured to determine the extent of contamination, especially in the edible portions, of plants from the study plots. After 1978 samples were taken from the entire area. Pu concentrations for plant samples with a minimum weight of 3 kg are shown in Tables 5 and 6. The data suggest that contamination is for the most part external. It is also clear that levels are lowest in the edible portions of the crops. In Spain, the annual limit for ingestion by the public of Pu oxides is 200 kBq (5.4 uCi). Thus, one would need to ingest over 7 million kg of tomatos or 235 thousand kg of barley grain per year to reach this limit. Food levels reported for New York city in 1972 were 0.00014 Bq Pu/kg (0.0037 pCi /kg). Pu intake via ingestion clearly is not a public health problem. Samples are now being measured for Americium 241.

Wild vegetation in the hilly area near impact point 2 (close to study plot 2-1) where Pu was removed with hand tools, is usually contaminated. Hemp grass (Esparto) average 575 Bq Pu/kg (155 pCi/kg). The maximum value for Esparto grass was found to be 26 kBq Pu/kg (0.70 uCi/kg).

SOIL:

Samples were taken from each study plot. Each was 30 mm diameter and 45 cm deep divided into five sections (0-5, 5-15, 15-25, 25-35, and 35-45 cm). Table 7 shows the concentrations of Pu and Am for the six study plots. The data represent 2160 radiochemical analyses of samples taken periodically since 1966 to a depth of 45 cm. All study plots except 2-1 are in areas used for cultivation. Pu is only found below 15 cm at locations where cultivation is practiced. The values in Table 7 for plot 2-1 are for the first 5 cm layer. At some locations close to impact point 2, values on the order of 20-50 Bq/g (0.54-1.35 nCi/g) have been found.

The distribution of Pu and Am concentrations are heterogeneous.;however, it has been observed that, because of farming practices over the years, homogeneity is increasing.

Values reported by Bennett for the northeast US in the early 1970's were 0.0016 Bq/g (0.05 pCi/g). A recent ORNL report contains a compilation of Pu

levels in air and soil from nuclear weapons testing for various locations around the world.

Particle size analysis of the soil indicates that less than 15% of the residual plutonium and americium contamination is associated with particles less than 10 μm diameter. Pu and Am content and soil particle size are directly related. Maximum activity concentrations correspond to size fractions ranging between 63 and 250 μm diameter.

PEOPLE:

Some residents of Palomares were measured for contamination just after the accident. The highest external contamination level was about 3 kBq/square meter (81 nCi/square meter). The early urine samples obtained from people and military personnel were often contaminated during sample collection. Measurements of Pu in urine and the body of Palomares residents were started in 1967 at the JEN in Madrid have continued uninterrupted since 1975. In addition to medical examinations, measurements are made to detect Pu and, more recently, Am in the body and in urine. To date 646 people, about 84% of the residents, have been measured at least once (the 1978 census of Palomares listed 767 people).

Chest measurements using a sophisticated phoswich detector indicate no Pu above the minimum detectable activity (MDA) of 814 Bq (22 nCi). The counting facility was included in an international calibration study several years ago.

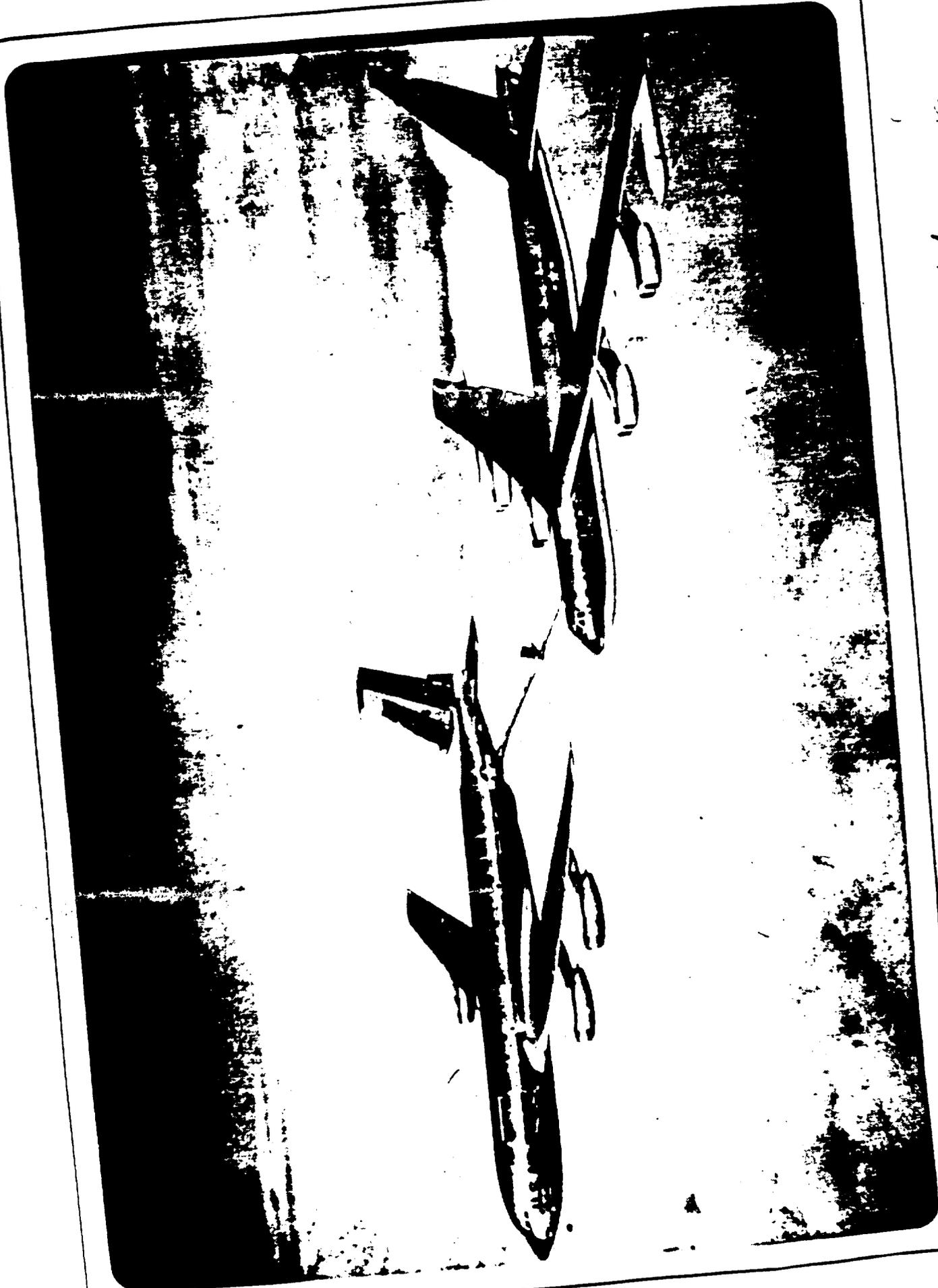
Bioassay procedures provide a more sensitive method to estimate internal contamination. However, most people have provided urine samples only once (216) or twice (247). Of the 1155 Pu bioassays obtained through 1985, ten people each supplied 8 samples and 21 people each supplied 6 samples. Of the 1155 assays, 1069 (92.3%) were less than the minimum detection level of 0.37 mBq/day (10 fCi/day).

By assuming a Gaussian distribution of the urinary excretion values the average excretion rate was found to be 13 mBq/day (350 fCi/day). By assuming chronic inhalation of Pu over the 18 year period and using the standard excretion function for Pu, one can derive an annual intake level of about 0.48 Bq/year (13 pCi/year). This quantity is equivalent to about 1% of the annual limit for the Spanish public. The average annual effective dose equivalent to the people in Palomares corresponding to this intake is calculated to be 0.042 mSv (4.2 mrem). This value is in good agreement with the integrated effective dose equivalent calculated for a person breathing Pu air concentrations measured at station 2-2 for 15 years.

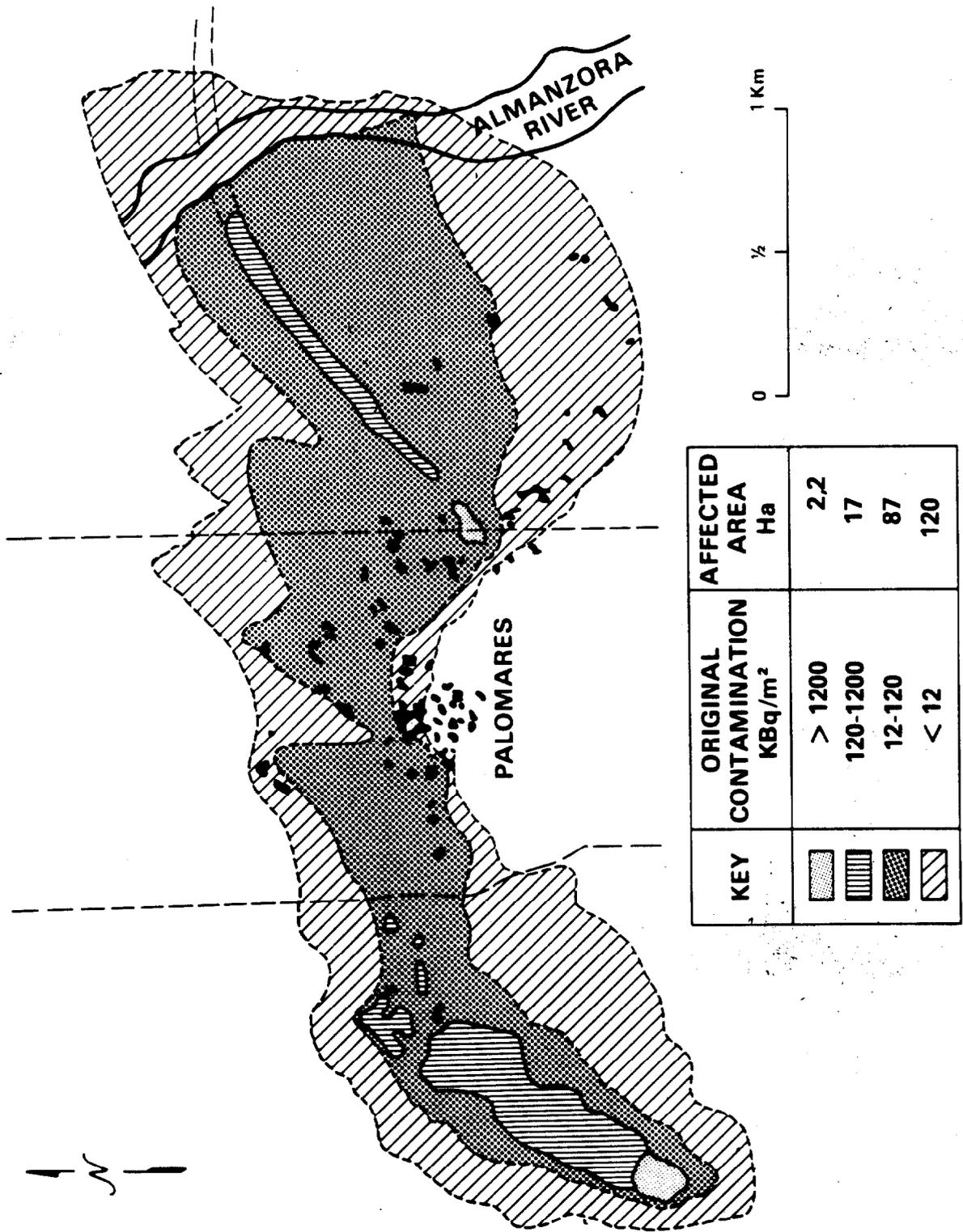
We have also calculated the committed effective dose equivalent for residents of Palomares based on the assumption of acute inhalation of Pu at the time of the accident. The intake of Pu by inhalation was estimated from the excretion data by using data tables developed at the Oak Ridge National Laboratory. These tables use a modified Langham excretion model and the ICRP inhalation

model..Committed dose equivalent for the main organs of interest and committed effective dose equivalent were calculated from ICRP factors for dose per intake (Sv/Bq) as given in ICRP publication 30. These data are given in Table 8. Of the total sample of 646 people, only 54, about 8%, are considered to be above the minimum detection level for the radiochemistry procedure. The remaining values are questionable as they are based on few data.

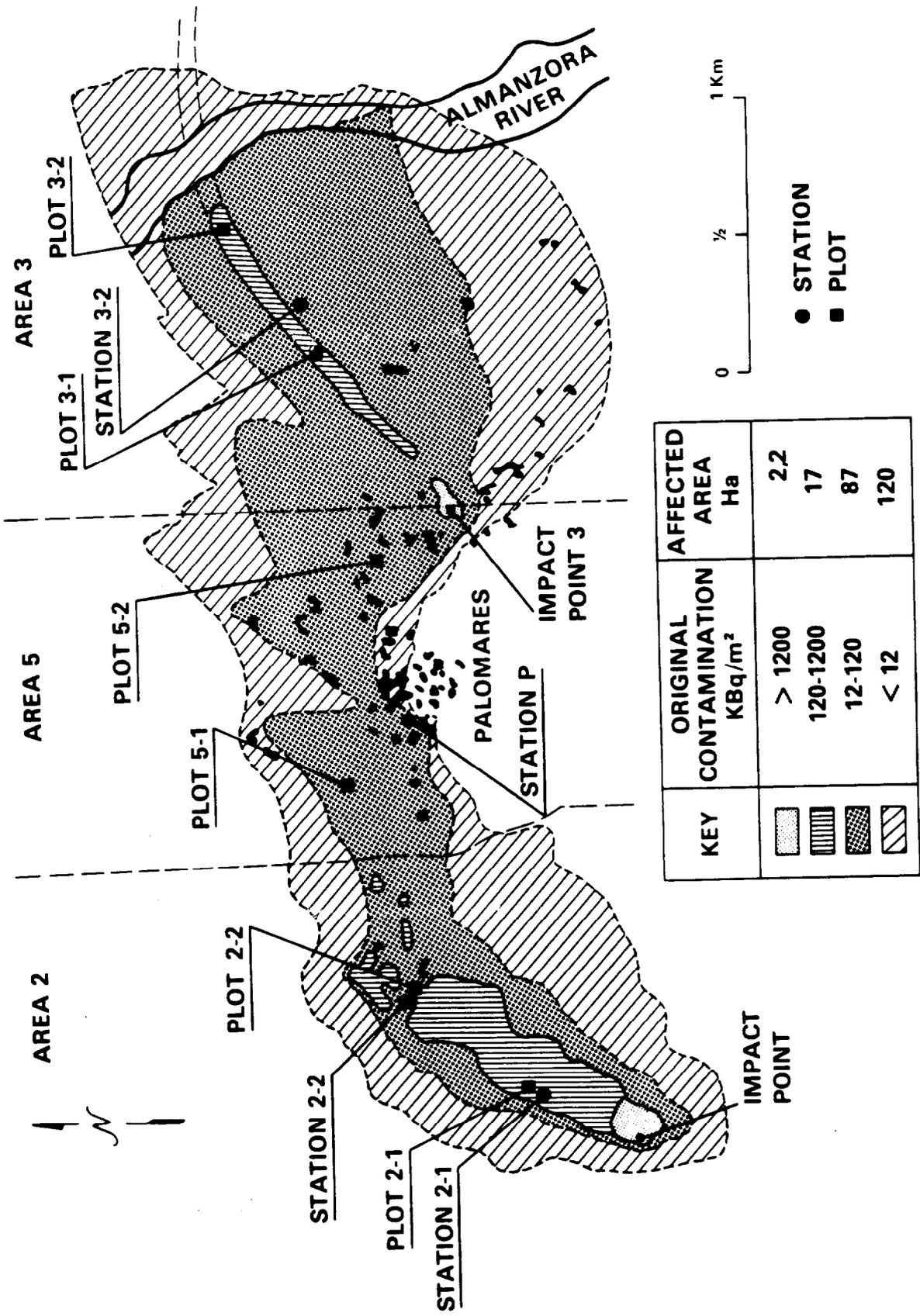
Jan. 16/10



PALOMARES AREA: ORIGINAL CONTAMINATION LEVELS



PALOMARES AREA: ORIGINAL CONTAMINATION LEVELS AND LOCATION OF SAMPLING STATIONS



**REMEDIAL ACTIONS TAKEN AT PALOMARES
FOLLOWING THE JANUARY, 1966, ACCIDENT**

Location	Hectares*	Acres	Remediation
Impact Point 2	1.6	4	Removed top 10 cm
Impact Point 3	0.6	1.5	Removed top 10 cm
Remainder	224	550	Plowed to 30 cm where possible
Total	226	558	

*259 hectares equal one square mile.

MAJOR OBJECTIVES OF PROJECT INDALO

- Determine the magnitude of the risk from internal contamination of the area inhabitants immediately after the accident and during the emergency phase
- Determine the short, medium and long-term risk of internal contamination for the inhabitants of Palomares and its environs and to those who consume planet products cultivated in that area

**CONCENTRATION OF PLUTONIUM IN AIR AT
PALOMARES, AND RESULTING RADIATION DOSE,
1966-1980**

Station	X Concentration* $\mu\text{Bq}/\text{m}^3$	Committed Effective Dose Equivalent, mSv
P(town)	5.5	0.054
2-1	56	-
2-2	52	0.52
3-2	14	-

*Average value of the average annual concentration.

AVERAGE AIR CONCENTRATIONS OF PLUTONIUM AT VARIOUS LOCATIONS ($\mu\text{Bq}/\text{m}^3$)

Year	NYC,	Northwest	Town		Area 2-2
	USA	Italy	Palomares	Palomares	Palomares
1966	3.1	2.6	14.8		44.8
1967	1.3	1.6	4.1		441.8
1968	2.0	2.7	2.6		21.8
1969	1.5	1.8	2.6		142.1
1970	1.8	1.8	2.2		5.9
1971	1.5	1.7	1.8*		2.2
1972	0.7	0.8	1.8*		10.4
1973	0.3	0.5	2.2		3.0
1974	1.1	1.4	4.1		8.1

*Minimum detection level.

**PLUTONIUM CONCENTRATIONS IN CULTIVATED
CROPS FROM PALOMARES**

<u>Plant</u>	<u>No. Samples</u>	<u>Pu Concentration</u>		
<u>Species</u>	<u>Part</u>	<u>Total</u>	<u>% Positive</u>	<u>X, Bq/kg</u>
Barley	Grain	395	21.5	0.85
Barley	Stalk	403	33.7	2.61
Tomato	Fruit	236	9.7	0.03
Tomato	Plant	118	38.1	3.17
Corn	Grain	16	0	0*
Corn	Plant	48	18.8	0.97

*Zero, or below the detection level for the procedure.

**PLUTONIUM CONCENTRATIONS IN CULTIVATED
CROPS FROM PALOMARES**

<u>Plant</u>	<u>No. Samples</u>	<u>Pu Concentration</u>		
<u>Species</u>	<u>Part</u>	<u>Total</u>	<u>% Positive</u>	<u>X, Bq/kg</u>
Beans	Fruit	26	0	0*
Beans	Pod	26	7.7	0.01
Melon	Fruit	26	0	0*
Melon	Plant	14	28.6	0.27
Oranges	Fruit	6	0	0*
Figs	Fruit	24	8.3	0.03

*Zero, or below the detection level for the procedure.

**CONCENTRATION OF PLUTONIUM AND
AMERICIUM IN STUDY PLOTS AT
PALOMARES**

X Concentration Bq/g

Plot	Am-241	Pu-239,240
2-1	0.07	0.44*
2-2	0.55	2.06
3-1	0.24	1.10
3-2	0.49	1.79
5-1	0.04	0.13
5-2	0.09	0.29

*About 12 pCi/g.

**ESTIMATED COMMITTED EFFECTIVE DOSE
EQUIVALENT BASED ON URINARY EXCRETION DATA
AND ASSUMED ACUTE INHALATION OF PU AT TIME
OF ACCIDENT**

<u>Estimated Dose mSv (rem)</u>	<u>Number of People</u>
<20* (<2)	592
20-50 (2-5)	22
50-100 (5-10)	22
100-200 (10-20)	10

*Assumed to be negative.
MDA of radiochemical procedure is 0.37 mBq (~10 fCi)
corresponding to ~18 mSv (~1.8 rem).