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June 16 1992

Mr. C. Rick Jones, Director
Office of Health Physics and
Ind.Hyg.Programs
U.S.D.O.E. EH-41
Washington D.C. 20585

Dear Rick:

My visit to CIEMET went very smoothly, and I was able to obtain the information I needed in one day, rather than two days, as we had originally planned. I appreciate your role in making the arrangements for me.

My trip report is attached. As I noted in the report, I did not attempt to summarize all the information they now have, as they will have their detailed status report ready in another few weeks. They are planning to have it in your hands by the end of July.

I am of the opinion that the information from Palomares can be useful in the Rocky Flats case, but the decision will ultimately be made by the attorneys. I have told Kirkland and Ellis that if they plan to pursue the matter further, they should arrange for their experts to meet with the CIEMET group when they are in Washington. Either they or I will be in touch with you if they should so decide.

I continue to be impressed with the quality of the CIEMET team. I do think, as noted in my report, that they should estimate the amount of Pu and Am present in the contaminated area, and was told they could do so in a few months. I hope this will be on your agenda when you meet with them.

Once again, my thanks to you and Tom Bell for your help in arranging for my visit.

Sincerely,

cc: Robert Morris, Madrid Embassy

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June 16 1992

HIGHLIGHTS OF THE STUDIES BEING CONDUCTED IN PALOMARES, SPAIN

NOTE: This report is based on a review of the published reports of Emilio Irenzo and his associates (1 - 6), and conferences with them in Madrid on September 8-9 1988 and June 8 1992. I have drawn heavily on the report I prepared for NASA in 1988 (7), since much of the background material is directly applicable to the question of whether the Palomares data are relevent to Rocky Flats. The Palomares investigators are currently preparing a project summary that will be delivered to DOE by the end of July.

INTRODUCTION

In January 1966, two U.S. Air Force aircraft crashed during a refueling exercise over the village of Palomares, near Cuevos de Almonzora on the southeastern coast of Spain. Of four thermonuclear weapons that fell with the wreckage, one was recovered intact from the Mediterranean Sea after a three month search, one was recovered intact from nearby fields where it landed by parachute, and two were destroyed by ignition of the chemical explosives contained in the weapons. Plutonium 239 was scattered by the explosion. The plutonium ignited, resulting in a cloud of plutonium oxide fume that was dispersed by a 30 knot wind from the west, and resulted in contamination of 226 hectares (about 0.87 square miles) of residential areas, farm land, and woods. The accident occurred in an area where the fields were farmed intensively, primarily for tomatoes, the last seasonal crop of which was ready for harvest.

Initial Countermeasures The initial levels of plutonium contamination, after removal of visible bomb fragments is shown in Figure 1. U.S. Airforce and Spanish personnel cooperated in the cleanup, and following radiological surveys of the area, it was decided to proceed according to the following criteria:

1. Soil contaminated above 32.4 uCi per square meter was removed, barreled, and shipped to the United States for burial at Savannah River. The area requiring treatment totalied 2.2 hectares, from which six thousand 250 liter drums of soil were removed.

2. Arable land below 32.4 uCi per square meter was mixed by plowing and harrowing to a depth of 30 cm. Seventeen hectares were treated in this way.

3. On rocky hillsides, where plowing was not practical, and the deposition was greater than 3.24 uCi per square meter, the soil was removed with hand tools and drummed for shipment to the

United States.

When these decontamination objectives were achieved, a surveillance program was established to monitor the air, soil, vegetation, farm animals, and people for plutonium. One hundred and fifty residents per year have been taken to Madrid for medical examinations, whole body counting, and urine analysis for plutonium. The program has been costing about $1,000,000 per year, of which $200,000 was provided, until recently, by the Department of Energy. At the time of my 1988 visit I learned that the DOE contribution had been discontinued without explanation, but upon my return I was told by DOE that it was a temporary problem that would soon be resolved. This proved not to be the case. During preliminary discussions with DOE preparatory to my recent visit I learned that the interruption in support continued until the Fall of 1991, at which time a $500,000 contribution by DOE to the program was made, and that continuity of support seems to be assured for the immediate future.

The surveillance program has now been in effect for about twenty-five years, and the findings have been summarized in a number of publications(1-6). Dr. Emilio Irenzo, of the Centro de Investgationes Energeticas Medioambioambientales y Technologicas (CIEMAT) directed the investigations from their inception until he retired about two years ago. His position is now held by Dr. Jose Gutierrez. Drs. Gutierrez and Irenzo were both present during my visit of June 8.

DOE has requested that a summary of the project findings to date be prepared in English and submitted to the Office of Environment, Safety, and Health this summer. At the time of my visit I learned that the report is expected to be ready in July, and will be hand delivered in Washington by the end of the month.

<u>Summary of Environmental Studies.</u>

Residual Contamination of Soil.

The soils of Palomares are derived from sandstone with a clay content (muscovite-illite) of 39%. The sand content is 48%, and the organic carbon content is 0.27% of which about half is present as humic acids, which are known to complex with plutonium and increase its mobilization rate. Studies of the the plutonium content of the soil have been underway since 1966 in the six study plots (50 m.x 50 m.) shown in Figure 1. Vegetation from the plots are also analyzed to determine the extent of plutonium uptake as well as the amount of contamination on the surface of the plants.

A total of 2160 samples of soils have been collected and analyzed since 1966. The samples were collected to a depth of 45 cm. and separated by particle size into eight fractions which were analyzed separately. Aliquots are digested in HF and HNO_3 prior to plutonium separation, electrodeposition, and counting by alpha spectrometry. Because Americium 239 builds into Plutonium 239, measurements of that nuclide were also made so that the comparative behavior of the two actinide elements could be studied. The Americium results will not be discussed in this report.

After more than twenty years, more than 99% of the plutonium in untilled soils has remained within a depth of five cm (4). If, as was likely, the particles were originally formed as a fume from the burning metal, and would therefore be introduced into the environment as fine particles (< 1um), they would have soon attached themselves to larger particles that would thereafter determine their aerodynamic behavior. Using autoradiographic methods, it has been found that most of the plutonium is associated with soil particles too large to be respirable, the largest fraction being in the particle size range of 63 - 250 um. About 9% of the plutonium is associated with particles < 5um in heavily cultivated soils, and < 3% in more lightly cultivated soils. This should greatly reduce resuspension, since the aerodynamic behavior of the Pu will be determined by the size and mass of the host Particles.

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The mean rainfall in Palomares is only 20 cm/yr, which is not greatly different from Rocky Flats (33 cm/yr).

Considerable effort has been expended in studying the influnce of mineralogical factors on the distribution of Pu and Am. The concentrations are greatest with fragments of iron hydroxide, lessor amounts with carbonates, and the lowest amounts with the micaceous fraction. This seems inconsistent with the earlier conclusions that the strongest associations were with clay minerals (5). This question will no doubt be resolved in the report now in preparation.

I did not take the time to study the considerable amount of data that exist on the relative separations of Pu and Am.

At the time of my first visit I noted the paucity of information on the total amount of Pu and Am present in the Palomares soil. At that time Dr. Irenzo estimated that the total Pu content to be less than 40 Ci, but he noted that there were few data to support his estimate. He has, however, undertaken detailed analysis of the soils at Station 2, which is the most contaminated part of Palomares. On the basis of these measurements he estimates that about 25 Ci are present on a 2500 sq.m. tract. I discussed with him the importance of extending these measurements to the entire contaminated tract, since this would greatly add to the value of whatever models for environmental transport are developed from the Palomares studies. He stated that this could be done in a few months, but that additional funding might be needed.

Contamination of Vegetation

Tomatoes, barley, and alfalfa have been the main crops since the time of the accident, with much lower production of melons, peppers, corn, and other vegetables. It is well known that absorbtion of the actinide elements by plants is minimal, and the analytical procedures are difficult because of the large sample sizes (5 to 10 kg) required. It is also necessary to differentiate between plutonium that has been absorbed metabolically into the plants from that present as surface contamination due to dusting or rain splatter. This is accomplished by thorough washing of the vegetables or restricting the analyses to the parts that are eaten. Tomatoes can be peeled, for example, or beans can be removed from the pods.

Typical data on the plutonium content of vegetables grown in Palomares have been published by the Spanish investigators, and are given in Table 1 and 2 (3). These are very low values, the dosimetric implications of which are nil. Nevertheless, the concentration factors (Pu/gm soil/ Pu/gm vegetable are higher than the values reported by other investigators. The Palomares investigators are aware of this and will discuss the reasons in the report now being prepared.

Air Studies

The extent to which plutonium resuspends into the atmosphere as a result of wind or mechanical action is generally accepted as the main index of potential risk in an area contaminated by plutonium. Six air sampling stations were originally established, using 47 mm diameter cellulose filters through which air is drawn at a rate of 62.5 l/m. The filter head is located 1.7 m above the ground, and the filters are normally changed every 24 hours. Because of the low concentrations of plutonium encountered, the filters from each of the sampling stations are composited every ten days for analysis by methods that include chemical separation, electrochemocal deposition, and alpha spectrometry. Plutonium 239/240 recoveries are estimated by adding Pu-242 as a tracer.

For various reasons, not all of the stations have been maintained during the entire period of the study. However, there is a full record for two of the stations, indicated as Stations 2-1 and P in Figure 1. Station P is located in the small village of Polomares, the population of which is currently about 760. Station 2-1 is located in a valley, the hills of which surround the region of maximum contamination produced by one of the bombs that was destroyed. The station is located about one kilometer NE of the point of impact. Only the data from these two stations will be discussed here.

The annual average concentrations for the two stations are summarized in Table 3 for the 15 year period, 1966-1980. More recent data will be included in the report now being prepared. For puposes of comparison, the data for the period 1966-1974 are listed in Table 4 with data collected at control stations in Northwest Italy and New York City. Airborne Plutonium 239 has been detectable worldwide in tropospheric air as a result of of nuclear weapons tests prior to the 1963 ban on nuclear weapons tests in the atmosphere. Most of that plutonium was injected into the stratosphere, from which slow transfer to the troposphere has been occurring. The following conclusions can be drawn from Tables 3 and 4:

1. The concentration of airborne plutonium has been elevated above "background" for all years at the station in the village of Palomares, and 11 out of 15 years at station 2-1.

2. The annual average airborne concentrations in New York and northwest Italy are comparable (1.5 and 1.7 uBq/m^3) during the 15 year period. The annual average concentrations for stations P and 2-1 were elevated by factors of 2.5 and 48 during that period.

3. During the two year period 1973-4, the concentrations in New York and Northwest Italy were 0.70 and 0.95 uBq/m^3. These values are lower than the 15 year average for the two stations, reflecting the diminution in the residual stratospheric inventory accumulated during the period of weapons testing. Durihg the same period Stations P and 2-1 were higher than the controls by factors of 3.9 and 6.8 respectively.

4. The values within the town of Palomares show a tendency to increase towards the end of the 15 year period. This might be the result of contamination of the streets with soil carried into town from the more heavily contaminated agricultural areas by farm equipment, people, and farm animals. The higher values at Station 2-1 are attributed by the Spanish investigators to be due to the opening of new land to cultivation.

5. Because the data are obtained from fixed stations, they do not include exposures of people working in the fields. Operators of farm equipment when the fields are dry are probably exposed more heavily than is indicated by the data collected at the fixed stations.

Human Measurements

For the purpose of risk assessment, the most meaningful measurements have been those concerned directly with evidence of human absorption of plutonium. These measurements have been of two types, urinanalysis and whole body counting. The data obtained by urinanalysis has made it possible to estimate the amounts of plutonium deposited in the bodies of the Palomares residents and the dose received.

Urinanalysis.

By 1988, twenty four hour urine samples from 93% of the Palomares residents had been collected and analyzed for plutonium at least once. As for other types of samples, the plutonium is separated chemically after adding Pu^{242} as a tracer. The lower limit of detection (LLD) is 0.37 mBq/day (10 fCi/day), which compares favorably with laboratories in the US, and corresponds to a 50 year committed effective dose equivalent of about 18 mSv (1.8 rem).

Of 1815 assays, 92% were below the lower limit of detection. Only

124 people were above the LLD. The estimated 50 year committed effective dose equivelent for these subjects are (6):

Estimated Dose mSv (rem)	Number of People
20-50 (2-5)	22
50-100 (5-10)	22
100-200 (10-20)	11

By the urinanalysis methods that have been in use, effective dose commitments as low as 1.8 rem can be estimated. The investigators are interested in using fission track analysis, which would greatly reduce the lower limit of dose estimation. They will no doubt discuss the matter when they visit Washington this summer. They understand that it will be necessary to rely on laboratories in the U.S. for the time being, but would like to send a technician to the States for a period of training, with the objective of eventually developing the ability to perform the enalyses in Madrid.

Whole Body Counting

Whole body counting of Pu^{239} is based on detection of either the soft penetrating radiation from its decay, or the slightly more penetrating radiation emitted by its americium daughter product. The Madrid laboratory is equipped with a phoswich whole body counting system with which they have been making chest measurements on about 100 Palomares residents per year. All measurements have been below the LLD of 814 Bq (22nCi).

Medical Followup

Each year 150 of the Palomares residents are taken to Madrid for medical examinations, urinanalysis, and whole body counting. The examinations are more extensive than needed and include complete batteries of biochemical studies that go far beyond what could possibly be useful in following a plutonium contaminated cohort. The project director knows this but does not believe it is desirable to reduce the scope of the examinations because it would be upsetting to the residents.

One weakness in the program results from the difficulty of obtaining samples of human tissue for radiochemical analysis. This is due primarily to objections to autopsies on religious grounds. Tissue samples might also be useful, and could possibly be obtained from residents who go to surgery. However, the population is small, and they go to several cities when surgery is required. The project director does not believe it would be possible to devise a practical system for obtaining samples.

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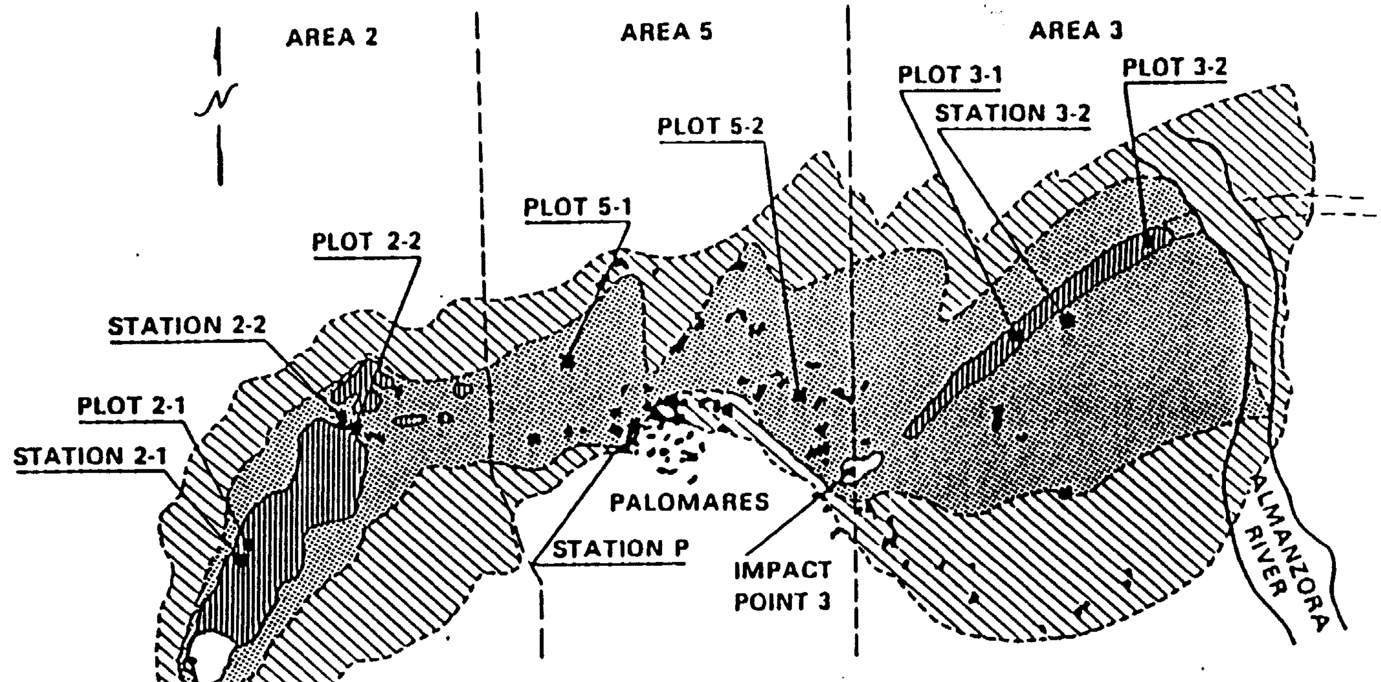
FIG 1

PALOMARES AREA: ORIGINAL CONTAMINATION LEVELS AND LOCATION OF SAMPLING STATIONS



KEY	ORIGINAL CONTAMINATION KBq/m²	AFFECTED AREA Ha
(blank)	> 1200	2.2
(vertical hatching)	120-1200	17
(stippled)	12-120	87
(diagonal hatching)	< 12	120

0 ½ 1 Km

● STATION
■ PLOT

REF 2

TABLE 1

PLUTONIUM CONCENTRATIONS IN CULTIVATED CROPS FROM PALOMARES.

PLANT		Nº SAMPLES		Pu Conc., $Bq \times Kg^{-1}$
Species	Part	Total	% Positive	$\bar{X}$ in positives
TOMATO	Fruit	159	28.3	0.22
TOMATO	Washed fruit	231	6.1	0.15
TOMATO	Plant	206	41.7	4.42
BARLEY	Grain	496	26.8	2.47
BARLEY	Stalk	496	37.0	5.87
BARLEY	Spicule	144	58.3	5.38
ALFALFA	Plant	112	39.0	3.33

REF 3

TABLE 2

- SOIL-PLANT CONCENTRATION RATIOS.

PLANT		CONCENTRATION RATIO
SPECIES	PART	
Tomatoes	Fruit	1.5×10^{-4}
"	Washed fruit	0.9×10^{-4}
"	Plant	2.3×10^{-3}
Barley	Grain	1.9×10^{-3}
"	Straw	5.0×10^{-3}
"	Spicule	6.2×10^{-3}
Alfalfa	Edible[1]	8.9×10^{-3}

REF 3

TABLE 3

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

Year	NYC, USA	Northwest Italy	Town Palomares	Area 2-2 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.