

FIRST RESULTS FROM THE PROGRAMME OF ACTION FOLLOWING THE PALOMARES ACCIDENT

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INTRODUCTION

In reply to the kind invitation of the Fachverband für Strahlenschutz, we would like to give an idea of the work which is being carried out in connection with the wellknown Palomares accident, and give indications of some of the general results obtained, with the idea that in the Symposium on Agricultural and Health Aspects of Radioactive Contamination in Normal and Emergency Conditions which is going to be held in Cordoba (Spain), organized jointly by the I.A.E.A., F.A.O., and W.H.O., we shall present more detailed studies on each of the sections we shall briefly refer to here.

As a consequence of the air crash which caused, on the 17th January 1966, the explosion of part of the conventional explosive, which was a constituent, of two thermonuclear bombs, an aerosol of plutonium and uranium was created which caused the contamination of a total area of approximately 226 hectares, of which 2.2 hectares had an alfa contamination of more than 700,000 d.p.m./100 cm², being the areas closest to the points of impact of the two bombs, which were 2,600 m. apart in a line drawn through the air.

An area of approximately 17 hectares showed contamination between 700,000 and 70,000 d.p.m./100 cm², and the rest, some 207 hectares, showed contaminations of less than 70,000 d.p.m./100 cm², rather more than half of these being less than 7,000 d.p.m./100 cm².

A great proportion of the houses of the town were either well out of the contaminated area, or in the parts where contamination levels were lowest. The number of persons who were in the town at the time of the accident was approximately 1,000 - 1,200.

In another communication presented to this Symposium, it has been explained how the limits of the said zones were fixed, the decontamination operations carried out, and steps taken with people and vegetation at a time soon after the accident. Here we are going to deal with, although in a somewhat summarised form, the work plan devised for the vigilance of the people and the area, and the deduction of the consequences which might, in the future, result from such an accident and the measures which were taken.

Our greatest worry, as is logical, was to determine whether or not there was internal contamination by plutonium-239 of the persons present in the area during the moments following the accident, what possibilities there were of contamination during the period of time when the operations of limit-fixing and decontamination were being carried out, and whether there was any risk from the measures which it had been decided would be taken in the areas where contamination levels were lower than 700,000 d.p.m./100 cm².

This risk might be derived from:

a) inhalation of plutonium if, as a consequence of the tilling of the land, and the winds which are present, an aerosol, contaminated with the said element, is created, which reach a definite diffusion.

b) contamination as a consequence of tilling operations on ground where a certain quantity of plutonium-239 was left.

c) contamination by contact with or ingestion of vegetable products which are cultivated in the area, in the case of these showing external contamination.

d) contamination of vegetable products used as foodstuffs in the case of their producing, during their growth, an absorption of plutonium-239 or uranium-235.

We make the reservation that there is no risk of contamination through drinking water, since the water existing in the said area has such a high concentration of salts that it is completely undrinkable, and the drinking water which the public consumes is brought from a distance of 50 kilometres.

As a consequence of these considerations, we summarise below the deductions obtained during the first year of work in connection with the contamination of persons, ground, air and vegetation.

CONTAMINATION IN PERSONS

The study of the determination of possible internal contamination of the people of Palomares, both in the case of those who went there afterwards, and especially those who walked through contaminated areas during the first moments, was planned under the following two aspects:

1) Determination of the Plutonium-239 excreted in urine, in order to determine the fraction which could have passed the pulmonary and lymphatic limit.

2) Determination of the quantity of plutonium-239 present in the lungs.

To obtain this information, after the analysis of plutonium-239 and total alfa activity in urine had been carried out soon after the accident, on the persons who at that time were considered those most likely to have been internally contaminated, a selection was made from the people of Palomares, with respect to the following considerations:

1) Situation nearest to the points of impact of the fractional bombs, and the areas of greatest contamination at the moment when the accident occurred, therefore with the greatest possibilities of inhaling the aerosol created in the conventional explosions.

2) Stay in zones of highest contamination during the day of the accident and the following day.

3) Residence, at the time of the accident and the days following it, in houses and areas of the town where a higher contamination had been found, within the lower limits where contamination was produced in the town, as may be seen from the plan of the contaminated area.

4) Situation during the accident and following it, in houses and areas of the town where contamination was nil, so that they could serve as controls, and at the same time we could find out with certainty whether a mistake had been made in not considering evacuation of the people of that area as necessary in the operations of contamination limit fixing and decontamination.

Taking the above factors into account,¹⁾ a total number of 100 persons were selected,²⁾ of which 49 were males over 14 years of age, 32 were females over 14 years of age, 10 were males under 14 years, and 9 were females under 14 years.

They were taken to Madrid for the said tests to be carried out in the laboratories of the Division de Medicina y Proteccion de la Junta de Energia Nuclear. They first underwent a complete medical examination, in order to find out their state of health and take it into account in the study and subsequent epidemiological control.

In order to determine the concentration of plutonium-239 excreted in urine, three complete 24-hour samples of urine were taken from each one of the said persons, on three consecutive days. These samples were submitted to a chemical treatment in order to determine the content of the said element, by alfa spectrometry, on the final electrodeposit following a separation by ion exchange resins.

The result of this analysis showed that in 71 per cent of cases there was no indication of the existence of plutonium-239, 18 per cent showed, in some analyses, contents of less than 0.1 disintegrations per minute in the urine of 24 hours, 9 per cent showed contents of between 0.2 and 0.1 disintegrations per minute over 24 hours, and finally, 2 per cent showed a content of between 0.2 and 1 d.p.m. in the urine of 24 hours.

Bearing in mind what these values represent on the alfa spectrum corresponding to the 1000-minute measurements taken, we have taken a deliberately pessimistic view, although in almost all cases they could be considered negative.

In order to determine the pulmonary content of plutonium-239, measurements were taken on all the previously-mentioned persons, with proportional pulmonary counters, considering the region between 10 KeV-28 KeV, where the XL rays of uranium are found of 13.6, 17.4 and 20.2 KeV in an abundance of 4% per disintegration. These counters consist of two gas flow proportional chambers, with a capacity of 14 litres and a sensitive detection surface of 17.4 x 30.1 cm each, which work with a mixture of argon (90%) and methane (10%) and which have been designed in the "Los Alamos National Laboratory" by Mr. P. Dean. The calibration of the said chambers was effected with a point source of plutonium-239, and two sheets of metyl metacrylate 2.54 cm thick to simulate the tissues of the thorax.

Taking into account the background of the chambers, the counting time and their efficiency, it was found that the minimum detectable value of plutonium-239 was 40×10^{-9} Ci. In the test made under these conditions, all the values obtained were on the order of background, which indicates that the amount of plutonium-239 which might be found in the lungs, if any, would be less than 40 nanocuries, a value which is quite encouraging, as the maximum permissible pulmonary burden is 16 nanocuries.

A certain modification was made in the chambers which permitted the lowest value detectable to be reduced to 16×10^{-9} Ci. The measurements taken under these conditions with some of that people showed that in no case did the values obtained exceed this figure.

CONTAMINATION OF THE GROUND

As has already been mentioned, the contamination of the ground surface was not homogeneous. In order to determine the degree of distribution, and what this distribution was on the surface, as well as its depth, after the operations of decontamination, and those planned to eliminate surface contamination in areas required, a study has been planned in order to find out which these are. We hope that it will permit us, as years go by, to discover the dynamics of plutonium-239 in cultivated land, both as regards its dynamics parameters,

and those introduced by tilling and cultivation methods.

For this purpose, six areas have been chosen of 50 m² each, two from each of the 3 zones into which the total contaminated area was divided, corresponding to the two areas where the bombs fell and broke into fragments, and the intermediate zone where the town is situated. These areas were chosen in places which had remained most contaminated after the removal of a layer of earth on the areas which had contaminations higher than 700,000 d.p.m./100 cm². Those in zones 2 and 3 comprise areas where total alfa contaminations were found to be between 700,000 d.p.m./100 cm²; area 2-1 belongs to a part where the upper layer of earth was also removed during decontamination operations. The areas in zone 5 showed contaminations of the order of 70,000 - 7,000 d.p.m./100 cm².

In order to find the average background of total alfa activity on the ground of the zone, two similar areas have been chosen, with similar geological characteristics and situated at 1,000 m. and 7,000 respectively from the zero line of the contaminated zone.

In each of the areas, and in accordance with its diagonals, during the first year, 1966, nine samples were taken which were divided into fragments, corresponding to depths of 0-5, 5-15, 15-25, 25-35 and 35-45 cm. Of the homogenised samples corresponding to each fragment, two fractions of 0.50 gm. were taken, which were submitted to chemical treatment and alfa counting, in order to determine the total alfa activity.

At present we are continuing to do this, and in successive years samples will continue to be taken at points in the same areas and following a preplanned distribution order in order to obtain complete knowledge of the dynamics of the contamination, as well as to deduce the contamination factors of the vegetable products which are cultivated in them. In 1967 the number of samples taken was higher, with a factor of two, than that of the previous year.

With these last samples, the tests have been not only to measure the total alfa activity, and consequently, through its relationship with the values obtained in the background sampling places, and the high specific activity of plutonium-239, deduce what amount is due to the said plutonium-239. Besides radiochemical separations are being carried out for each of the contaminating radionuclids.

The alfa activity of the soil in the region of Palomares (Almeria) is one of the highest in Spain, as has been shown in the comparison of results obtained from the background areas, situated far enough away to be certain that they were not contaminated, and those from other provinces and types of soil, as may be seen from Table I.

In order to give you an idea of the results we are obtaining, in Table II may be seen the average values of alfa activity from the samples taken in 1966 from each one of the zones, and in relation to depth.

From these values it may easily be deduced that:

- 1) In zone 2-1, where a 5 cm layer of soil was removed, the remaining contamination is nil.
- 2) As a result of ploughing and breaking up of the ground, a distribution of contaminating elements have been obtained to a depth of 30 cm. Generally highest contamination levels are found in layers between 15 and 25 cm down.

3) The maximum average value of alfa activity found in the areas studied is approximately 50 times higher than the minimum value of natural alfa activity found in the background soil of the zone, which in turn is two times higher than the minimums found in the areas studied.

The unhomogeneous distribution of the contaminating particles, even after the ploughing operations carried out in order to dilute the radioactive elements, plutonium and uranium, on a layer 30-35 cm deep, is perfectly clear from the values obtained, as might logically have been expected. Even results from different samples from the same point and fragment, after the greatest homogenisation possible in the laboratory, show quite a considerable dispersion, as may be seen from Table III. On this table may be seen the values from all the analyses corresponding to each one of the fractions of ground which gave the highest alfa activity values, in area 2-2, one of those of greatest contamination.

CONTAMINATION OF THE AIR

A network of aerosoles sampling has been set up in order to discover what possibilities there might be of internal contamination of the people who live in the zone and cultivate the fields, as an isolated or related consequence of the ploughing operations on the ground and the climatological characteristics of the zone, especially those related to the low rainfall and prevailing winds.

This network consists of four sampling stations, and two for the study of the speed and direction of the winds. The samples stations are situated in the places marked on the figure S-1, S-2, P and 3-1, which respectively correspond to the zones where the two fragmented bombs fell, and one point at the urban centre of Palomares. Those for measuring the characteristics of the winds are situated at S-1 and P. In each one of these stations, at a height of 1.70 m, continuous 24-hour samples are taken, with a volume of approximately 95 m³, every day of the year, on membrane filter paper. The samples are prepared for sending to the laboratories of the Medicine and Protection Division of JEN, where in principle, a minimum of one week after they are taken, a count is made of the total alfa activity due to radioactive elements with a long half life.

In Table IV, the average monthly values are shown for all the samples taken, during the first year of operation of the said network, at each one of the sampling stations. From the observation of the same, bearing in mind that the maximum permissible concentration in the air, for the public in general, of any mixture of alfa, beta and gamma emitting radionuclides is 4×10^{-14} microcuries/cm³, and for plutonium-239 itself is 6×10^{-14} microcuries/cm³, it may be deduced:

1. The average monthly values of airborne alfa activity in the area of Palomares has always been maintained below the M.P.C. Except on one occasion, in the month of August, the said average values have not exceeded one tenth of the M.P.C.
2. The maximum values of alfa activity have exceeded the M.P.C. on only seven occasions, never, however, reaching values higher than a factor of 10 over the M.P.C.
3. On the days when maximum values were found, the winds in the area had speeds of between 12 and 22 km/hr.
4. The average values of alfa activity in the district of Palomares, are comparable with those corresponding to the district of Madrid, where also,

on occasions, the maximum values found have been slightly over the M.P.C.

At present, because of the results obtained from the alfa count with the gas flow proportional counter, a radiochemical determination of plutonium in samples of air is now being carried out. For this the samples were kept, and at present are chemically treated in groups of ten days corresponding to each one of the stations. After their extraction with ion exchange resins and electro-deposition, they are measured by alfa spectrometry. In order to compare these results with those of the present background of plutonium from the contamination of the atmosphere with the said element, a sampling station was established in Madrid similar to those in Palomares.

CONTAMINATION OF THE VEGETATION

The work carried out with vegetables has tended to determine the possibilities of their external contamination, as a consequence of their cultivation in contaminated and surrounding areas, and to the study of the plants' absorption capacity, and its settling on fruits and seeds.

For the purpose, in each one of the areas previously mentioned, and in the places where the soil samples are taken, samples have been taken of the existing crops. During the first year following the accident, not all the areas were cultivated, and for this reason it was not possible to take samples from areas 2-2 and 5-1. In area 2-1, and given that it was part of an uncultivated plot, of which the upper layer of soil had been carefully removed, making sure not to destroy the existing vegetation, the tomato plant samples given in Table V in its corresponding section were taken from nearby strips of cultivated land where the upper layer of soil had not been removed.

In Table V are given, classified by areas, the average values of total alfa activity corresponding to the various vegetables collected, establishing, in some cases, a distinction between washed and unwashed vegetables, in order to be able to determine the external contamination which is easily separable from the permanent or internally absorbed contamination. From these values, the following deductions can be made:

1. Maize cultivated in a contaminated area gives a value of alfa activity superior to that which grew in the blank area, with a factor of two for the plant and three for the seed.
2. The tomato plant cultivated on contaminated soil shows a separable and fixed alfa contamination higher than that growing on blank soil. As for the tomatoes, the edible part, no difference has been found. It is logical to suppose that the findings from the plant are of an external type, and found difficult to separate by washing, due to the leafiness of the plant.
3. Both bean plants and the beans fruit show a certain amount of external contamination, proportional to the contamination of the area under cultivation. The edible part of the beans, the seeds, however, do not show the least indication of such contamination, since in the two areas their alfa activity values are less than 6 d.p.m/kg of wet weight.
4. In the only area where alfalfa is cultivated, its alfa activity was only very slightly increased.
5. The greatest degree of contamination found was in wild plants (asparagus, esparto grass and other graminaceous plants), which may possibly have existed at the moment of the accident.

TABLE I: GROSS ALPHA ACTIVITY OF SEVERAL TYPES OF SOIL IN SPAIN

PROVINCE	TYPE OF SOIL	GROSS ALPHA ACTIVITY d.p.m/g.
PALOMARES (Almeria)	2-3B GypsumBurlap	20,8 \pm 1,3
	5-3B GypsumBurlap	19,6 \pm 0,6
OVIEDO	Calcarenite	11,1 \pm 1,7
MADRID	Akroses	12,6 \pm 2,0
MURCIA	Marl	10,8 \pm 2,2
BADAJOS	Sandstones	8,6 \pm 0,7
CORUNA	Granites	17,1 \pm 2,2
TENERIFE	Volcanic rocks	18,5 \pm 1,2

TABLE II: GROSS ALPHA ACTIVITY IN SOME AREAS OF PALOMARES

AREA	GROSS ALPHA ACTIVITY d.p.m/g.					d.p.m/g dry soil
	0-5 cm	5-15 cm	15-25 cm	25-35 cm	35-45 cm	TOTAL MEAN
2-3 B	24,5 \pm 2,8	21,6 \pm 3,2	22,3 \pm 1,8	18,7 \pm 2,8	16,9 \pm 1,3	20,8 \pm 1,3
5-3 B	20,0 \pm 1,2	19,9 \pm 1,3	20,1 \pm 1,5	20,6 \pm 1,3	17,3 \pm 1,5	19,6 \pm 0,6
2-1	11,3 \pm 2,2	9,0 \pm 1,1	10,6 \pm 1,3	11,7 \pm 1,6	9,6 \pm 1,0	10,4 \pm 0,5
2-2	472 \pm 190	376 \pm 296	851 \pm 827	10,4 \pm 1,3	9,1 \pm 0,9	344 \pm 178
5-1	48,6 \pm 35,8	28,2 \pm 16,8	90,1 \pm 72,4	12,3 \pm 1,6	11,8 \pm 1,1	38,2 \pm 14,6
5-2	13,7 \pm 4,9	12,8 \pm 1,5	11,3 \pm 1,4	16,3 \pm 6,5	13,8 \pm 2,1	13,6 \pm 0,8
3-1	24,6 \pm 2,4	30,6 \pm 6,4	13,0 \pm 1,7	14,3 \pm 3,1	11,8 \pm 1,8	18,9 \pm 3,7
3-2	215 \pm 103	182 \pm 100	1095 \pm 738	415 \pm 392	16,5 \pm 7,6	385 \pm 188

TABLE III: GROSS ALPHA ACTIVITY IN SOILS OF AREA 2-2.

AREA	PLACE	DEPTH	D.P.M./gr.						MEAN
2-2	1	0-5	681,4	34,8	150,1	8,9	77,3	6,9	159,9±106,6
"	1	5-15	11,7	94,3	7,6	7,8	14,3	6,9	23,8± 14,2
"	2	0-5	3.400,1	348,2	568,9	475,1	94,5	507,6	899,1±504,9
"	3	0-5	1.258,4	118,3	88,3	1.608,3	95,6	643,3	635,9±270,4
"	3	5-15	178,9	15,7	1.196,6	11,4	332,6	235,8	328,5±181,0
"	4	5-15	231,2	275,9	206,9	3.897,3	776,4	134,9	920,4±602,7
"	6	0-5	616,4	687,7	11,2	58,0	23,9	29,2	232,9±133,1
"	7	0-5	435,2	107,1	315,8	398,1	875,9	39,9	360,3±119,9
"	7	5-15	127,4	62,7	15,8	16,4	16,6	93,4	55,4± 19,4
"	8	5-15	23,3	20,0	5,8	23,3	21,7	35,5	18,1± 5,0
"	9	0-5	162,6	413,7	1.923,5	204,9	642,4	4.041,9	1.231,5±621,5
"	9	5-15	5.398,6	205,2	1.056,6	1.661,4	124,5	487,4	1.214,8±850,5

TABLE IV: GROSS ALPHA ACTIVITY IN AIRBORNE PARTICULATES

Mes	SAMPLING STATION. microcuries x 10 ⁻¹⁵ /cm ³														
	MADRID			Station 2-1			Station 2-2			Station P (in the town)			Station 3-2		
	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
June	8,6	0,7	2,1	1,8	0,7	0,8	4,3	0,7	1,0	5,3	0,7	1,1	1,0	0,7	0,7
July	160	0,7	8,5	20	0,7	2,7	7,8	0,7	3,6	9,8	0,7	2,6	10,0	0,7	2,4
August	7,3	0,7	2,7	7,2	0,7	2,7	140	0,7	8,6	250	0,7	10,1	8,2	0,7	2,4
Sept.	7,6	0,7	3,8	8,9	0,7	2,9	44	0,7	3,9	9,5	0,7	2,6	43,0	0,7	4,7
October	31,0	0,7	3,5	13,0	0,7	2,5	71	0,7	4,3	5,5	0,7	2,1	3,7	0,7	1,4
November	23,0	1,0	5,2	3,9	0,7	1,5	8,8	0,7	2,2	6,1	0,7	2,1	4,9	0,7	1,4
December	9,8	0,7	4,2	9,5	0,7	2,0	14,0	0,7	2,3	4,3	0,7	1,6	9,7	0,7	2,0
January	16,5	0,7	7,7	22,0	0,7	2,3	51,5	0,7	3,4	9,8	0,7	1,5	9,2	0,7	1,7
Febr.	7,7	0,7	2,3	48,0	0,7	4,4	24,7	0,7	3,4	8,1	0,7	2,2	69,8	0,7	3,6
March	8,5	0,7	2,3	2,8	0,7	1,2	11,0	0,7	3,3	4,7	0,7	1,2	3,9	0,7	1,2
April	8,2	1,0	2,6	6,8	0,7	2,1	273	0,7	2,7	3,4	0,7	1,6	3,8	0,7	1,4
May	6,4	1,0	3,0	4,8	0,7	1,8	10,5	0,7	2,8	6,6	0,7	1,3	4,6	0,7	1,5

TABLE V: GROSS ALPHA ACTIVITY IN THE VEGETATION

VEGETATION		AREA					
		2-3 B	5-3 B	2-1	5-2	3-1	3-2
Corn (Plant)	washed						
	d.p.m./kg. wet						
Corn (Seed)	no washed		450 \pm 130			454 \pm 200	933 \pm 475
	d.p.m./kg. dry		59			69 \pm 11	185 \pm 15
Tomato (Plant)	washed		224 \pm 75	8.472 \pm 7.346	96 \pm 30		
	d.p.m./kg. wet		407 \pm 100	17.338 \pm 5.817	1640 \pm 989		
Tomato (Fruit)	washed		19 \pm 3,4	25 \pm 12	18,5 \pm 6,5		
	d.p.m./kg. wet		26 \pm 6	27 \pm 3,5	19,5 \pm 1,5		
Bean (Plant)	washed				210 \pm 57		659 \pm 177
	d.p.m./kg. wet				177 \pm 5		664 \pm 134
Bean (Fruit)							
	d.p.m./kg wet				11,5 \pm 2		283 \pm 158
Alfalfa (Plant)	washed						
	d.p.m./kg. wet				1099 \pm 494		
Wild plants	washed	489 \pm 236			363 \pm 112		
	d.p.m./kg. wet	676 \pm 306		236.373 \pm 113.551			