

LA-TR-74-10

ENVIRONMENTAL CONTAMINATION BY RADIOACTIVE MATERIALS,
p. 529-40. 1969

RG	US DOE ARCHIVES 326 U.S. ATOMIC ENERGY COMMISSION
Collection	1320
Box	17-Salmonares
Folder	3-Spanish Publications

MEASURES TO DETERMINE THE RISK TO WHICH A POPULATION
CAN BE SUBJECTED AS A RESULT OF A NUCLEAR ACCIDENT

[Determinacion del Riesgo a que ha estado sometida la poblacion como
consecuencia de un accidente nuclear generador de un aerosol radiactivo]

By

E. Iranzo and E. Ramos

Translated by:

Ralph McElroy Co., Inc.

for

Los Alamos Scientific Laboratory

Los Alamos, New Mexico

1974

E. Iranzo* and E. Ramos*: Measures to Determine the Risk to Which a Population Can Be Subjected as a Result of a Nuclear Accident [Determinacion del Riesgo a que ha estado sometida la poblacion como consecuencia de un accidente nuclear generador de un aerosol radiactivo].

Reprint from "Environmental Contamination by Radioactive Materials", International Atomic Energy Agency, Vienna (1969).

* Nuclear Energy Committee, Madrid, Spain

DOE ARCHIVES

Translated from Spanish by the Ralph McElroy Co., Custom Division
2102 Rio Grande, Austin, Texas 78705 U.S.A.

MEASURES TO DETERMINE THE RISK TO WHICH A
POPULATION CAN BE SUBJECTED AS A RESULT OF A NUCLEAR ACCIDENT

This study describes the safety measures taken to protect people living in an area where a radioactive aerosol has been produced. The reasons to decide against the evacuation of the area and the way the personnel operated or co-operated to check the existence of internal contamination as a consequence of the inhalation of the generated aerosol are also given. In addition the paper gives a resume of the techniques used to determine the plutonium-239 burden in the lungs, the amount eliminated with the urine, and the results from 100 persons selected for this purpose.

On January 17, 1966, a B-52 and a tanker, both belonging to the United States Air Force, collided during refueling above the village of Palomares, located in the municipality of Cuevas de Almanzora in the province of Almeria. The wreckage of both planes was scattered over a wide area, mostly within the municipality of Cuevas de Almanzora, within the municipality of Vera, and along the Mediterranean Sea coast between Puerto Ray and Villaricos.

Palomares has a population of about 1,200 inhabitants and is located a short distance from the beach, near the mouth of the Almanzora River, on the right bank. The houses do not make up a definite town layout, but rather are irregularly distributed. Groups of houses, or isolated houses, are spread without pattern over an extensive area of irregular topography crossed by a highway, roads, and innumerable canals which irrigate neighboring orchards and fields.

DOE ARCHIVES

As a result of the accident, four thermonuclear bombs being transported by the B-52 were loosened and fell. Two, with their respective parachutes, were recovered intact, one in the bed of the Almanzora River, near its mouth, the other in the sea. The two remaining bombs fell without parachutes. Due to the violent contact with the ground, part of the conventional explosive detonated and caused a consecutive fragmentation and oxidation of the

uranium and plutonium sponge which constitutes the bomb. This generated aerosols of uranium and plutonium which were dispersed by the strong prevailing wind from southwest to northeast and contaminated the ground, plants, and buildings in a wide area. The most active spots in the area were the impact points. One bomb fell on a semi-barren hill about 1,500 meters southeast of Palomares, at the same altitude as the 72 marker on Algarrobinas hill. The other fell on some orchards in the same area, east of the village and about 200 meters from the two houses closest. Nearby were numerous metallic bomb fragments of different sizes.

When the condition of the two devices was confirmed, it was realized that a problem of α contamination had been generated. A rapid confirmation of the existence of such contamination was undertaken along with verification that serious β or γ radiation contamination did not exist.

These measures confirmed a contamination of a total surface area of approximately 226 hectares. Of these, the 2.2 with an α contamination greater than 700,000 dis/min per 100 sq cm were near the points of impact of the bombs, which were 2,600 meters apart, measured in a straight line. Approximately 17 hectares showed contamination between 700,000 and 70,000 dis/min per 100 sq cm and the remainder, about 207 hectares, showed contamination below 70,000 dis/min per 100 sq cm (slightly more than half showing less than 7,000).

DOE ARCHIVES

Simultaneous with operations to determine the existence and magnitude of the contamination, measurements were undertaken to judge whether, given the contents of the bombs, the actual risk of inhalation of radioactive aerosols justified a partial or total evacuation of the population of Palomares. For this purpose, aerosol samples were taken in various parts of the village and surrounding countryside, preferably among the houses near the impact (the east end of the village), and in those places where wind direction created the greatest probability of atmospheric contamination by radionuclides deposited on the ground and plants.

For this purpose the following was available:

- A mobile laboratory from the Division of Medicine and Protection, type 22 Volkswagen, equipped with materials from the "Frieeseke und Hoepfner (Erlagen-Bruck)" Co., and provided with, among other things, a portable collector of aerosol samples (model FH 422 - intake capacity $30 \text{ m}^3/\text{h}$), and a methane gas proportional flow counter (model FH 407 A). Generators in the rear of the van supplied the electrical energy.
- Portable aerosol sampling equipment (Staplex, type TF 1A - intake capacity $\sim 30 \text{ m}^3/\text{h}$), run by groups of AC generators.

The mobile laboratory, due to its great maneuverability, was used to take air samples in various places near the urban nucleus of Palomares and of the village of Villaricos, and in other places within a working radius of 6 km. The Staplex equipment was set up in the most strategic places, which interested us, of the center of the town, and in areas near the two points of impact of the thermonuclear devices. Varying the location of the equipment at each sample, within a radius of 100 meters each, the taking of samples began in 30 minutes.

α activity was measured first at the time when a sample was taken, in order to get a quick idea if it was much above the natural radioactivity of the area, and again 24 hours later, to confirm the concentration in the air of radionucleides with long half-lives.

Radioactivity counts did not show α concentrations that justified evacuation. Except at points less than 100 meters from the impact points, in the direction of a strong wind from the southwest which blew between 3:00 and 5:00 p.m. on January 20 (before any decontamination or land humidification), radionucleide concentration values were 10^{-13} microcuries/cc or less, being, therefore, under the 6×10^{-13} microcuries/cc maximum values permitted by the International Organization of Protection against Radiation (I.O.P.R.) for plutonium-239.

DOE ARCHIVES

At the indicated points, and under the previously specified special conditions, this value was exceeded by a factor on the order of 100.

These initial measurements indicated the magnitude of the problem caused by the accident, allowing us to draw the following conclusions:

- External radiation of the inhabitants was not a problem.
- The most pressing problem was possible internal contamination of the inhabitants of Palomares who inhaled the aerosols immediately after the accident.
- A problem of external contamination of the clothing, skin, and hair of the inhabitants of Palomares could cause later internal contamination if not eliminated as soon as possible. This external contamination could be due to:
 - a) direct deposit of the aerosol
 - b) direct contact with the bomb fragments and with wreckage of the airplanes, crops, soil, and other materials which could have been contaminated.
- The wind could resuspend radioactive nucleides that had contaminated the surface, thus prolonging the risk that inhalation could internally contaminate the inhabitants.
- Gathering the fruit crop, which was in full season, could cause internal and external contamination of the inhabitants. Ingestion of externally contaminated fruits and foods could cause external contamination, even though the nature of the radioactive nucleides and, consequently, their minimal intestinal absorption mean that an important risk is not involved.
- The contamination could be spread outside the affected area by contaminated inhabitants, material and vegetable products.
- Besides those who were in Palomares immediately after the accident, those of the village of the Villaricos could have been affected, as well as any of the curious who came to the towns located in that area and went through the various places where there were remains of the airplanes and the materials they carried. Many persons picked up small pieces and kept them as souvenirs.

DOE ARCHIVES

As a result of these conclusions the following plan of action was organized.

As far as the inhabitants were concerned, which is what interests us in this communication, we initially had to acquire a basic idea of the possible contamination of persons who were in Palomares when the accident took place. To do so, we first determined the external contamination of all the inhabitants of the houses close to impact point number 3, of those who did not live there but were in the area, and of those who lived in the houses closest to impact point number 2, even though the houses were a kilometer away. Relatively unimportant contamination was found, with high readings on the order of 2,000 dis/min per 100 sq cm corresponding principally to the shoes, lower part of the pants, and sleeves of dresses, jerseys, and jackets.

At the same time, partial urine samples were taken from all these persons to determine the excreted amount of α emitting radionuclides in general and of plutonium-239, in particular. Also, the urine samples would give a quick idea of the extent of internal contamination, if it existed. These tests, based on the experiments of Dr. Langham and colleagues at Los Alamos for Plutonium-239 calculation, adopted the model for absorption and elimination of plutonium-239 proposed by him [1].

DOE ARCHIVES

These samples were sent by air to the Division of Medicine and protection laboratories of the Nuclear Energy Committee in Madrid. Analysis of the samples and the 24-hour samples taken the next day from the same persons, indicated to us that the quantity of plutonium-239 inhaled did not present any grave immediate danger which would require the administration of chelating agents. The values were lower than or only slightly higher than those corresponding to the I.O.P.R.'s maximum permissible charge (0.04 micricuries for bone as a critical organ and 0.4 microcuries for the entire organism as a critical organ). On the other hand, it was necessary to take into account that during collection, the urine could be easily contaminated by the external environment, which could lead to an erroneous or excessive evaluation of the

internal contamination of the persons involved. An analysis of externally decontaminated persons maintained in a rigorously controlled environment totally free from contamination confirmed this. One of these persons who gave a higher value was transferred to Madrid and submitted to daily control for 10 consecutive days. Urinary elimination of plutonium-239 was such that, according to the previously mentioned Langham formula, its charge could not be greater than 80% of the maximum permissible, considering bone as the critical organ. Consequently, and since the person was 75 years old, external contamination of urine during testing did not give cause for serious worry.

Once external contamination had been confirmed for those persons whom it was considered could have been most affected, Civil Guard personnel who had helped in the contaminated zones and all residents of Palomares, Villaricos, and the surrounding towns who had, for various reasons, visited the place of the accident, in the first hours were tested for external contamination in almost continuous shifts of about 16 hours per day.

They were examined for three principal reasons:

- Tranquilizing psychological effect upon seeing that they were not externally contaminated, since in the first days the totally false news which circulated created a certain suspicion among the area's simple folk and could have caused unjustified general panic. **DOE ARCHIVES**
- Detection of external contamination on persons and its amount, in order to, as a function thereof, determine those who, even though they were away from the places where they could have logically inhaled the radioactive aerosol, could have been exposed later to inhalation risk due to direct contact with earth, vegetable produce, or contaminated material and whose internal contamination should, immediately or later, be determined.
- Detection of the contamination of clothing, skin, and hair, in order to, in case of positive results, give adequate norms for their decontamination or, if the contamination were high, collect the contaminated pieces to eliminate sources of later internal contamination.

External contamination was initially measured *in situ* with portable α activity detection equipment (Nuclear Chicago Corporation model 2672). After the second day two fixed detection devices (Intégrateur Portatif α β I.P.A.B. 2B, "Nardeux") were installed at a site in the village, equipped for this purpose and were operated by two crews of two technicians each from the Division of Medicine and Protection of the Nuclear Energy Committee.

These tests did not reveal, in the 1,950 persons examined, any important contamination. Only a small number showed contamination of 2,000 dis/min per 100 sq cm and another group, even smaller, showed contamination of 20,000 dis/min per 100 sq cm on isolated spots on their clothing. This last group was Civil Guard personnel on guard duty at the most contaminated spots.

To avoid possible new external contamination, reduce the risk of inhalation due to being in contaminated places, and to avoid the spread of contamination to areas distant from the accident, the following measures were taken.

- Prohibition against entry into cultivated areas around Palomares until the contaminated areas were defined.
 - Prohibition against nearing or browsing around the wreckage of the planes until the existence or non-existence of contamination was confirmed.
 - Prohibition against harvesting tomatoes and lima beans, which were in full harvest, until those which could be harvested without danger to the gatherer or the user could be determined.
- DOE ARCHIVES**
- Prohibition against entry into the urban area of Palomares by the many curious visitors from other areas until the existence or non-existence of very low contamination values was confirmed.
 - Prohibition against sending to outside markets the tomatoes which were already in store-houses and which had been gathered since the 17th of January, until the existence or non-existence of contamination on them was confirmed.

- Confirmation of the existence of contamination in foods stored in the town.
- Control of the entry and exit of vehicles from the area.
- Covering the immediate area of the impact points of the two fragmented thermonuclear devices with a layer of mineral oil, and sprinkling with water, to keep constantly damp, the other areas of greatest contamination, to eliminate the risk of resuspension in the air of the contaminating radioactive nucleides.

This emergency plan was established to avoid a subsequent spread of contamination and to evaluate the extent of the inhabitant's possible internal contamination in the first moments and day, both with respect to the approximate number of persons affected as well as its amount in those who showed greatest potential risk of exposure. Procedures were thus established for follow-up in the coming months and years, in order to observe the inhabitants of the region and deduce the consequences of the accident itself and of the solutions used.

The basic point of all our worries has been as is logical, to determine 1) the existence or non-existence of internal plutonium-239 contamination of the persons who were in the area during the moments following the accident, 2) the possibilities of contamination which could have arisen during the period of delimitation and decontamination operations in the area, and 3) the existence or non-existence or risk derived from the measures which had been taken in the areas where contamination values were lower than 700,000 dis/min per 100 sq. cm. This risk derived from:

- inhalation of plutonium if, as a result of land-filling operations and existent winds, a plutonium aerosol was generated and diffused;
- contamination from filling the lands where a certain quantity of plutonium-239 had remained;
- contamination from contact with and ingestion of possibly contaminated vegetable products grown in the area.

DOE ARCHIVES

- contamination due to ingestion of vegetable products that had absorbed plutonium-239 or uranium-235 while they grew.

We mention the fact that there is no risk of contamination from drinking water, since the highly saline water available in this region is totally impotable, and drinking water for public use is brought from 50 kilometers away.

As far as direct measurement of possible internal contamination is concerned, the study was set up under the following aspects:

- Determination of plutonium-239 excreted in urine, in order to determine the fraction which could have passed to the pulmonary lymphatic region.
- Determination of the quantity of plutonium-239 in the lungs.

For these determinations and after the analyses of plutonium and total α activity in urine done in the first moments following the accident on those persons considered at that time to have the greatest probability of internal contamination, a selection was made of the population of Palomares according to the following factors:

- Nearest location to the impact points and the areas of greatest contamination at the exact moment of the accident, and, as a result, the greatest possibilities of inhaling the aerosol generated by the conventional explosions;
- Being in the areas of greatest contamination on the day of the accident and the day after; **DOE ARCHIVES**
- Staying, at the time of the accident and following days, in houses and areas of the town where greater contamination was found, within the low range found in the town;
- Location, during the accident and afterward, in houses and areas of the town where contamination was nil, to serve as controls, and at the same time so we could determine with certainty if any error had been committed

Page 10

when evacuation was considered unnecessary during operations of delimiting the contamination and decontamination.

At first, to determine plutonium 239 excreted in the urine, an initial sample of 69 persons was made from whom a 24-hour urine specimen per month for three consecutive months was taken *in situ*.

Later, to extend the investigation far from the Palomares region and determine by counting with proportional counters the pulmonary content of plutonium-239, the sample group was increased to 100 persons (49 males and 32 females over 14, 10 males and 9 females under 14).

All of these persons were taken to Madrid and tested at the laboratories of the Division of Medicine and Protection of the Nuclear Energy Committee.

Initially, they were submitted to a complete medical check-up to ascertain their sanitary condition and use it as a point of departure in the following epidemiological study and control.

In order to determine the plutonium-239 excreted in the urine, three complete 24-hour urine specimens were taken from each of these persons for three consecutive days.

Plutonium-239 was measured according to the technique of Campbell and Moss [2], with slight modifications. It consists, essentially, of co-precipitating with alkaline phosphates, fixating from solution in NO_3H 8N over Dowex AG 1 x 2 anionic resin, purifying with concentrated ClH to eliminate uranium and thorium ions and finally electrodepositing the eluide in ClH 0.5 N, using ClH 1 N-4% amonic oxalate as electrolyte, over stainless steel plates 13 mm in diameter, with a tension of 20 volts and a current intensity of 200 miliamperes.

α spectrometry measured electrodeposited plutonium-239. This allowed us, in addition, to determine the plutonium recovered in each analysis by measuring the plutonium-236 recovered from that previously added to each analyzed sample. **DOE ARCHIVES**

The results of the three specimens of urine belonging to each person, gathered *in situ*, showed on occasion a great dispersion, with an extreme case where even though two of the results were 0

and 0.2 dis/min in 24 hours, respectively, the third one reached as high as 113 dis/min in 24 hours. The persons were classified according to the maximum value of these three evaluations. Twenty-seven percent presented quantities of plutonium-239 lower than 0.1 dis/min in 24-hours, 41% 0.1 to 1 dis/min in 24-hours, 24% 1-10 dis/min in 24-hours, and 7% greater than 10 dis/min in 24-hours.

With respect to the urine specimens gathered during the stay of the previously mentioned persons in Madrid, with all precautions taken to eliminate possible specimen contamination the analysis showed that in 71% of the cases there existed no plutonium-239. In 18%, in some analyses, contents of less than 0.1 dis/min were found in the 24-hour urine, 9% showed contents between 0.2 and 0.1 dis/min for 24-hours and, finally, 2% showed a content between 0.2 and 1 dis/min in 24-hour urine.

The difference between the results for the two urine sample groups was very noticeable. Keeping in mind what the last values listed represent on the α spectrum corresponding to the 1,000-minute measurements done, we did not want to be over-optimistic, which, in almost all cases, could more exactly be considered negative.

Pulmonary proportional counters measured pulmonary plutonium-239 content of all the previously mentioned persons. In the zone between 10 KeV-28KeV, XL rays of uranium of 13.6, 17.4, and 20.2 KeV were found in an abundance of 4% by disintegration. These counters consist of two proportional flow chambers with a capacity of 14 l and a sensitive detection of 17.4 x 30.0 cm each, which work with a mixture of argon (90%) and methane (10%) and which were designed at the National Laboratory at Los Alamos by Mr. P. Dean. These chambers were calibrated with a point source of plutonium-239, and the tissues of the thorax were simulated by two 2.54 cm thick polyethylene plates.

DOE ARCHIVES

As a function of the depth of the chambers of the counting time and of the efficiency of the chambers, it was determined that the minimum detectable value of plutonium-239 was 40×10^{-9} Ci.

Under these conditions, all of the values obtained were close to the minimum, which is an indication that the charge of plutonium-239 which could be found in the lungs, if it exists, would be lower than 40 nanocuries. This value is very comforting, since the maximum accepted pulmonary charge is 16 nanocuries.

Certain modifications to the chambers permitted the reduction of chamber depth, increasing their sensitivity to a minimum detection value of 16×10^{-9} Ci. Measurements taken under these conditions, with part of the persons, showed that in no case were the values obtained greater than the above figure.

Currently work is underway with thoracic models to achieve a better calibration of these chambers and to acquire a more exact idea of the possibilities available to determine pulmonary contamination by insoluble plutonium compounds. We are also up-to-date on any advances which would allow us to avoid the technical difficulties, with which we are now faced, in determining these pulmonary contaminations, since the Nuclear Energy Committee continues vigilance of the inhabitants to detect any abnormality which might arise and to confirm whether or not we are right that, according to data obtained up to now, no serious internal contamination was produced which could be the case, in the future, of radiological lesions.

References

- [1] W.H. Langham. Physiology and toxicology of plutonium-239 and its industrial medical control. Health Physics, 2, 172 (1959).
- [2] E.E. Campbell and W.D. Moss. Determination of plutonium in urine by anion exchange. Health Physics, 11, 737 (1965).

Discussion

DOE ARCHIVES

H. HOWELLS: Could you explain how you measured the α contamination on the ground due to the deposited plutonium from the damaged weapons? Did you detect the plutonium with x-ray detectors monitoring for the soft γ radiation emitted by the plutonium-239?

I should also like to ask, in the subsequent "clean up" operation, when there was a need to remove large quantities of soil, did you first treat the soil to reduce the possibility of resuspension of plutonium contamination into the air?

E. IRANZO: In answer to your first question, that type of detector was not used, but we measured the α activity of the plutonium-239 with proportional counters. This gave rise to great difficulties because of the need to keep them almost in contact with the ground, and the windows got perforated as a result of contact with small stones, vegetation, etc.

Regarding your second point, to avoid resuspension during the clean-up operation, we wetted the vicinity of the impact areas with mineral oil, and the remaining areas were sprayed with water to keep them moist.

J. TADMOR: I realize that you were not faced by a situation in which you had to evacuate populated areas. However, I assume that you discussed the emergency measures that might need to be taken in case the contamination of the atmosphere and soil had been severe. What were the action levels for radiation and contamination you decided upon, i.e. at which levels of contamination of soil and at which level of atmospheric contamination would you have taken measures in populated centers, such as total evacuation of the population, keeping the population indoors, restriction on the use of land, etc.?

DOE ARCHIVES

E. IRANZO: As you know, we are at present in process of trying to decide the radiation and contamination levels for emergency situations (some have already been established); and you are aware of the differences of opinion in this respect. Hence you can appreciate the responsibility involved in making a decision in this matter in face of a totally unforeseen situation.

But to answer your question briefly: We did not consider any radiation level since there was no risk of external radiation. If the environmental contamination values had been constant and greater than 100 times the MPC, we would have taken certain special steps with regard to the population. Given the type of

contamination, we considered that at all times except under very unusual conditions, resuspension attaining such values in the populated area would be virtually impossible.

As regards the ground, we considered that removal of the upper layers of the soil contaminated to the extent of more than 700,000 dis/min per 100 sq cm was adequate. On ground where the contamination levels were lower, we eliminated surface contamination by ploughing over the area, thereby diluting it considerably, and also eliminated subsequent risk of resuspension in the environment with concentrations of the order of the MPC; the values have been continually checked since that time by four aerosol sampling stations in the area, the α activity in general and the plutonium-239 and uranium-235 contents in particular being subsequently determined in our laboratories.

DOE ARCHIVES