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y Tecnológicas

PROJECT INDALO

WORK PERFORMED DURING 2000

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SUMMARY

On September 15, 1997 a new agreement between the U.S. Department of Energy (DOE) and the Centro de Investigaciones Energeticas Medioambientales y Tecnológicas (CIEMAT) was signed to establish a framework for scientific and technological cooperation by the Parties in radiological studies resulting from the accidental release of fissionable materials that occurred in Palomar-es, Spain, on January 17, 1966.

During 2000, the activities related to the environmental and personal radiological monitoring have been continued. Also, according to the work proposal for the year 2000 and following the recommendations made by the expert panel in its 1998 report, soil studies have been carried out and calculational assessments and risk studies are in progress. Soil studies are focussed in Zone 2.0, where the highest levels of contaminants exist, but also in Zone 3, cultivated area under different conditions, including greenhouses, to investigate potential differences. A risk assessment in the area has been started, to estimate the current annual doses under the present conditions and under potential future use of the area. Also, specific research concerning the study of plutonium (Pu) hot particles in soils has been initiated in 2000.

The activities performed in 2000 are detailed in the two semi-annual reports sent to the Spanish national regulatory body, Consejo de Seguridad Nuclear (CSN). These two reports are attached.

Soils

Concerning the follow up of the measurements of soil contamination, a sampling of deep soils has been carried out in Zone 2.0 (9 core samples to a depth of 3 or 3.5 m). These 9 core samples have been divided in different deep sections (every 50 cm) for analysis, obtaining a total of 56 samples. Taking into account the high number of soil samples collected during 1999, this new sampling was the only decided to avoid the accumulation of samples in the laboratory. Also, another deep sampling in Zone 2.0 was planned in order to further investigate the existence of a burial of wastes; however some problems appeared with the owner of the land that involved the cessation of the sampling.

In addition to these sampling activities, the following analyses have been performed:

- 122 analyses of Pu (radiochemistry followed by alpha spectrometry) surface samples collected in Zone 3 in 1999.
- 60 analyses of americium (Am) (gamma spectrometry) corresponding to 6 of the 9 core samples collected in Zone 2.0 in 1999.
- 30 analyses of Am (gamma spectrometry) belonging to the 122 surface samples collected in Zone 3 in 1999.

The summary of the results obtained from the analyses of Pu, related to the 122 surface soil samples collected in 1999 in Zone 3, is the following:

- Only 7 samples, which represent 5.7 % of the total, showed values below the lower limit of detection (LLD). The rest of the samples (115) showed positive values in a broad range between the higher (around 10,000 Bq kg⁻¹) and the lower (<1 Bq kg⁻¹). Only 2 values were found above 1,000 Bq kg⁻¹ and 11 values were in the range between 100 and 1,000 Bq kg⁻¹. The remaining results (109) were below 100 Bq kg⁻¹.
- The 1999 sampling was made with the same systematic approach used in the 1998 sampling for Zone 3 (51 surface soil samples). The 1999 sampling covered an immediately parallel adjacent area to the original one affected by the contamination plume derived from bomb number 3 and sampled in 1998 (see Table 1). Zone 3 is presently cultivated including the use of greenhouses; only a small part remains uncultivated because transit ways or embankments and slopes close to the bomb 3 impact point. Comparing both set of samples (1998 and 1999 samplings), it can be observed that concentrations of Pu were significantly higher in the part of the area directly affected by the plume:

Table 1
Zone 3

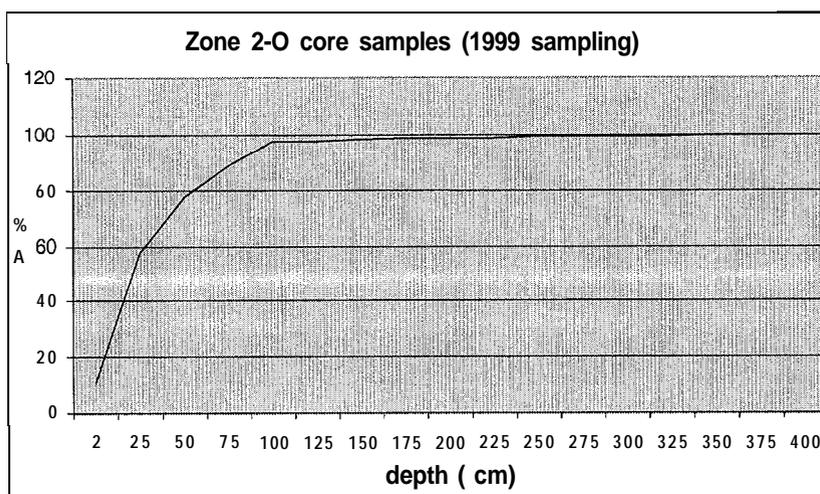
Plutonium (Bq kg⁻¹) in soil surface (2 cm depth)

	1998 sampling central line	1999 sampling side line
mean	2054	138
median	61	10
max	48,597	9,989
min	3.2	0.2
100-1000 Bq kg ⁻¹	40%	10%
>1000Bq kg ⁻¹	24%	1%
number	51	122

- The higher values in central line correspond to an uncultivated area, closer to impact point 3. For the cultivated area in Zone 3, values around 100 times lower were obtained.
- In general, for the side line, soils in open air cultivated zones show higher values than greenhouse's soils. This fact corresponds to the expected result, since soils in greenhouses contain sandy soils from external areas to Palomares, added to its original composition.

The results from the 60 analyses of Am, corresponding to 6 of the 9 core samples collected in Zone 2.0 in 1999, enable a preliminary estimation of the averaged distribution in depth according to Figure 1. Also, with the available data, the estimated averaged concentration of Am would be about 1.6 MBq m⁻² in the 0.5 hectares covered by this sampling. However, these estimates have to be confirmed after completion of the full set of measurements.

Figure 1



The results from the analyses of Am corresponding to 30 of the 122 surface samples collected in Zone 3 in 1999 will be summarized after completion of all the 122 measurements.

Air

In relation to the air monitoring, the sampling of air particles at stations 2-1, 2-2 and P have continued, changing the filter on a weekly basis. The air volume collected is about 10,000 m³ per sample in average. The air sampler in station 2-O has been out of service from December 1999 due to break of the air aspiration pump. In two occasions, it was transported to Madrid for repairing and then back to Palomares; however the equipment failed again soon after restarting and it was decided the replacement of the air pump by a new one which is presently being tested in the CIEMAT laboratory. The old station 2-1 has been replaced by a new one in April 14, 2000 placing it in a more adequate Zone 2-1 site. Station P restarted in March 17, 2000 after it was repaired; two small stop periods (from 2807.00 to 18.08.00 and from 01.09.00 to 15.09.00) were produced, both of them due to electrical supply problems. In total 124 air samples were collected during 2000.

Samples from 1999 have been analyzed and measured for Pu-239+240. Values of Pu ranged, during 1999, from 1.0 to 24.3 $\mu\text{Bq m}^{-3}$ in station 2-0, from 0.5 to 19.5 $\mu\text{Bq m}^{-3}$ in station 2-2, from 0.2 to 1.7 $\mu\text{Bq m}^{-3}$ in station 2-1 and from lower than LLD to 0.8 $\mu\text{Bq m}^{-3}$ in station P. In relation to Am, only the samples corresponding to the first half of 1999 in station 2-2 have been measured; the range obtained was between lower than LLD and 0.28 $\mu\text{Bq/m}^3$.

In general, these values are in the low range of the historical data series. Also, in agreement with the general trend, seasonal variability is higher than the annual one, as it could be expected considering seasonal weather changes and agricultural practices timing. Dose estimates due to the inhalation pathway are analyzed below (risk assessment studies)

Vegetables

For vegetables, 25 samples of different crops have been collected in 2000. These samples were further divided in different parts (leaves, edible, etc.) and pretreated for analyzing separately in order to know the type of contamination (external or internal) and its distribution.

Samples of vegetables (13) collected during 1999 have been processed for Pu, involving 47 analyses due to the above reason. These samples were collected in Zones 2, 3 and 5. With the exception of the samples from Zone 2, all the values were $\leq 1 \text{ Bq kg}^{-1}$; in general, values were higher for leaves than for fruits. The only samples cultivated and collected in Zone 2.0, were cauliflower, showing values around $50 - 70 \text{ Bq kg}^{-1}$ in leaves and around 1 Bq kg^{-1} in the edible part. Also 29 analyses from 14 samples collected during 2000 from different areas were carried out. As in the above case, the samples from Zone 2.0 (watermelons) showed the higher values, but always $\leq 1 \text{ Bq kg}^{-1}$.

Concerning Am-241, 75 samples collected during 1996, 1997 and 1998 in the Zones 2, 3 and 5 were measured (direct measurements by gamma spectrometry). In total 156 results were obtained, being the 81% lower than LLD ($<1 \text{ Bq kg}^{-1}$). Also, the higher values appear in the Zone 2.0 (leaves of watermelons and wheat grain), in the order of $25-30 \text{ Bq kg}^{-1}$. In addition, 5 samples collected during 1999 (Zones 3 and 2-2) were analyzed by radiochemistry followed by alpha spectrometry in order to reach a better detection capacity, obtaining values between 0.003 and 0.09 Bq kg^{-1} .

Other products

Concerning cow milk, 3 samples were collected, analyzed and measured for Pu (radiochemistry followed by alpha spectrometry) and Am (gamma spectrometry). Results for Pu were 2 mBq l^{-1} (similar than in previous years) in one of the samples and lower than LLD (1 mBq l^{-1}) in the another two. For Am, all the three results were lower than LLD. However the LLD reachable for Am by gamma spectrometry is not good, requiring that in the future milk analysis will be made by radiochemistry and alpha spectrometry.

A honey sample was also collected analyzed and measured for Pu; the result was $\leq 0.03 \text{ Bq kg}^{-1}$.

Snail samples from Zone 2.0 were collected and prepared for analysis, separating them in different parts (shell, meat, etc.). The analyses are presently in progress and the results are not yet available.

Due to the scarce rains during 1999, the rain water samples were not enough for analysis (a very small volume was collected). During 2000 three samples have been collected, corresponding, one to January (46.5 l m^{-2}) and two to October (44.1 and 90.6 l m^{-2} respectively); these samples will be analyzed later, jointly with samples to be collected during 2001. Given that rain samples do not offer significant information for monitoring and research and, in addition, they could be affected by external soil particle contamination, leading to errors, the continuation of this type of analyses should be reconsidered. However, the rain records will be

maintained, in any case, in order to relate relevant factors for data interpretation (washout, potential floods, etc.).

Urine

During 2000, 150 people from Palomares were transported to CIEMAT headquarters in Madrid for medical examinations and sampling of 24-hour urine collections and further bioassay analyses and internal dosimetric assessments. All those whose urine samples tested positive values, are requested to return to Madrid during the following year to confirm the existence and magnitude of their internal contamination. However, since the examinations are performed on those who agree to participate, not all those requested for follow-up examinations are re-evaluated during the following year. Some individuals choose not to participate for different (personal) reasons.

In relation to Pu, the measurements of the 150 people examined during 2000 were completed. Because two samples were spoiled during the analytical process, the related people will be requested to return to Madrid during 2001 for re-evaluation. From the remaining 148 samples, only one resulted in a positive value of 1.12 mBq in urine of 24 hours, corresponding to a woman 54 years old. This woman had been examined 12 times before, showing close positive values on three occasions. The estimated committed (50 years) effective dose is about 53 mSv.

The percentage of positive values for the year 2000 (0.67 %) is lower than the percentage for the period 1967-1999 which is of 4.2 % (142 positive values in a total of 3,398 measurements). The distribution by ranges of these results approach to a log-normal function; considering the median of the distribution as the most representative parameter for the more frequent committed (50 years) effective dose in the group of people having dose estimation, a value lower than 2mSv per year is obtained, The results obtained in 2000 do not modify the conclusions from the expert panel in its 1998 report.

Concerning Am, the urine analyses of the 150 people examined during 2000 also were completed. All of them showed values below the detection limit of 0.37 mBq in urine of 24 hours.

During 1999, no measurements in the whole body counter were made.

The individual results of bioassay for each of the person who are annually examined, were registered and sent to each of them, jointly with the result of their medical examination. These individual data are confidential. No significant findings related to radiation exposure were reported concerning the medical examinations (150 people) performed during 2000.

Research on Pu hot particles in soils

During 2000, research for the study of hot particles in soils has been started and is presently in progress. A great effort is being put on this subject, because it should provide a better knowledge of the actual physical and chemical state of the residual contamination. This, in turn, will also help to predict its environmental and dosimetric behavior. Soils of Zone 2.0 were chosen for the study due to its

higher activity levels but more recently, the zone close to the impact point of bomb 3 also has been of interest.

Eighteen hot particles were separated from the fraction corresponding to the 125-250 μm in soil samples from Zone 2.0 with density greater than 3.3 g cm^{-3} and low magnetic susceptibility. This fraction was considered the most suitable given its high Pu concentration. These particles were obtained from a few grams of sample. All of them were analyzed by an electronic microscope (SEM), which confirmed that they are mainly formed by Pu and uranium (U) oxides, having a metallic luster and varying their color from silver white to light yellow. The samples, from which these particles were separated, were washed during the density separation and were subjected to centrifugation and ultrasound.

Another hot particle was isolated from an untreated soil sample of 0.4 g (Zone 2.0). Its color is dark and has the appearance of an oxidized metallic phase with soil aggregates.

Also, one particle, whose length is greater than 200 μm was isolated from a drill core sample at 3.5 m depth based on its high exposure rate. Its characterization and semi-quantitative chemical analysis has not been made yet.

More recently, a sampling was carried out in Zone 3, close to the impact point, in order to investigate the existence of hot particles from bomb 3. Close to impact point 3, but in the opposite direction to the original contamination plume, one soil sample, had an exposure rate significantly higher than the background level and was taken to analysis. After granulometric analysis, one piece observed in the $> 2 \mu\text{m}$ fraction, with fractures similar to the rock, was separated. Its weight is about 1 g and could not be characterized by direct gamma spectrometry because its activity blocked the system. This piece was measured with a germanium detector at a distance of 45 cm for 30 seconds and it confirmed the presence of Am-241, Pu-239 and U-235. In addition, a metallic object, with a weight of about 14 g, was found and separated from a soil sample collected in another point located to a distance of about 30 m SW from the above mentioned point. This last point was also selected because of its high exposure rate. The separated metallic piece measured an exposure rate on contact of about $100 \mu\text{Sv h}^{-1}$. Also, a stone, with fractures similar to the rock, was removed from a third soil sample collected at a distance of 3m of the preceding point. This stone also had an exposure rate significantly higher than the background level and was transported to CIEMAT's laboratory for analysis.

Risk assessment

Dose assessments for the population of Palomares was made along the duration of the Indalo Project and are referenced in the 1998 Scientific Review Report. Among them, annual internal dosimetry estimates were carried out by urine analyses for 150 persons and reported in another section of this document. Taking into account the progress in information from the monitoring program, the associated research in the area, and the potential new agricultural scenarios, mainly in Zone 2.0, a new risk assessment study was recommended and started in 2000. As it was stated in last year's document, "Annual Work Proposal", tasks will be addressed to estimate the current annual doses under the present conditions

and under potential future use of the area. The study is currently in progress, focussing, so far, on dose evaluations under present conditions for three main pathways (external irradiation, inhalation and ingestion). To do it, data from the last few years were reviewed and have been considered.

In relation to **External irradiation pathway**, Am-241 is currently the only radionuclide of potential concern. The data selected for the calculation were:

Results of Am activity measured in Zone 2.1 (plot FLL) from the first 1998 sampling (surface soil samples from NE side of Zone 2.1, that was affected by the original contamination plume and further cleaned by removal of the top 10 cm).

- Results of Am activity measured in Zone 2-1 (plot APM) from the second 1998 sampling. The sampling was made after a significant modification of the soil surface in order to extend the cultivated areas.

Results of Am activity measured in Zone 3 from the 1998 sampling (surface soil samples from the zone affected by the contamination plume of the bomb 3) and results of Am activity measured in Zone 2.0 from the 1999 sampling (6 of the 9 soil core samples).

The results of the measurements in the plot FLL, are summarized in Table 2.

Table 2.

Am-241 (Bq kg⁻¹) surface soils (2 cm) in plot FLL, 1998 sampling

	First sampling
mean	1.01E+02
median	7.74E+00
assymetry	4.62E+00
maximum	1.99E+03
minimum	1.62E+00

The samples in the second plot (APM) provide only a value over LLD, all measurements are lower than 9 Bq kg⁻¹

Results from area 3 from 1998 sampling

Table 3.

Am-241 Bq kg⁻¹, Area 3, 1998

mean	6.14E+02
median	8.59E+01
assymetry	4.61E+00
maximum	1.10E+04
min	2.75E+00

Results from the six core samples provide averaged values of for the concentration of Am in the two first horizons (O-2 and 2-25 cm depth) respectively of 5,300 and 2,000 Bq kg⁻¹

In all cases, the median of the distribution of values was calculated, using the positive values and the LLD when the result was lower than this limit (conservative calculation).

To evaluate individual doses from the surface soil results, a homogeneous activity concentration for the top 15 cm was assumed, considering an infinite extended source. For core soil results, the average of each equivalent section in depth was calculated and chosen.

Dose coefficients ($\text{Sv seg}^{-1}\text{Bq}^{-1} \text{m}^3$) for exposure to soil contaminated with Am to different depths (Federal Guidance Report N° 12, EPA-402-R-93-081), a soil density of $1,600 \text{ kg m}^{-3}$ and a permanency of 10 hours per day, during all the 365 days of the year (conservative assumption), were used for assessments.

The obtained results for external irradiation are the following:

- Annual Effective dose of $0.04 \mu\text{Sv y}^{-1}$ in Zone 2.1 that was affected by the original contamination plume and further cleaned by removal of the top 10 cm (plot FLL). The external dose in the other plot APM would be lower than $0.02 \mu\text{Sv y}^{-1}$
- Annual Effective dose of $0.4 \mu\text{Sv y}^{-1}$ in Zone 3 that was affected by the original contamination plume of the bomb 3.
- Annual Effective dose of $23 \mu\text{Sv y}^{-1}$ in Zone 2.0

Given the small significance of the obtained results, possible further refinements (use of Montecarlo statistical simulations to improve the distribution activity and topographical variations in the zone) in external dose calculation, under the present conditions of use in the zone, may be unnecessary.

Concerning the **Inhalation pathway**, Pu and Am were the radionuclides considered in the assessment. The data selected for the calculation basis related to the present situation were:

- For field practices, 1998 and 1999 Pu annual average activities and 1998 Am annual average activity in station 2.0 were chosen as representative. For urban stages, 1997 and 1998 Pu and Am annual average activities in station P were chosen as representative
- An aerosol solubility of 87% M class (medium) and 13% S class (slow), was assigned in agreement with results from biokinetic studies made in the framework of the European commission projects reported in previous years. The size aerosol particle used for the assessment was $1 \mu\text{m}$.
- Inhalation dose factors for adults and children 10 and 1 years old, specific breathing rates for each of these age groups and for each type of field or urban activity were selected from ICRP 72. Times of permanency were adapted to those typical in the zone (see Table 4 below). Shadowed values are for urban areas and the other ones are for the most contaminated field plot.

Table 4.
Parameters used to estimate intake by inhalation

		age			
		adults	10 y old	1 y old	
sleeping	$\text{m}^3 \text{h}^{-1}$	0.45	0.31	0.15	
	h day^{-1}	inside	8.50	10.00	14.00
rest	$\text{m}^3 \text{h}^{-1}$	0.54	0.38	0.22	
	h day^{-1}	inside	4.50	3.67	3.00
	h day^{-1}	outside	1.00	1.00	0.33
working and plying	$\text{m}^3 \text{h}^{-1}$	light exercise	1.50	1.12	0.35
	h day^{-1}	outside	9.00	7.33	6.00
playing	h day^{-1}	outside	0.75	2.00	0.67
	$\text{m}^3 \text{h}^{-1}$	heavy exercise	3.00	2.22	0.00
working and plying	h day^{-1}	outside	0.25		
	total hours in the field		10.00	2.00	0.00

The obtained results for the inhalation pathway can be seen in Table 5:

Table 5.
Committed effective dose by inhalation $\mu\text{Sv y}^{-1}$

Age group	Am-241	Pu-239+240
Adults	1.34E-01	1.98E+00
10 years	1.07E-01	1.21E+00
1 year	6.58E-02	6.45E-01

Also an assessment of the annual doses by Pu inhalation from 1966 to 1999 was made using the same parameters as in the above scenario. In this case, the data selected as a basis of calculation were the available Pu annual average activities from 1966 to 1999 in stations 2.0, 2.1, 2.2 and P. Stations 2.0, 2.1, 2.2, were considered as representative of field conditions work and station P as representative of urban conditions. For assessment, the maximum value of the annual average of stations 2.0, 2.1, 2.2 in each year, was chosen to represent field conditions.

The obtained results in this assessment can be seen in Table 6:

Table 6.
Committed effective dose from inhalation of Pu ($\mu\text{Sv y}^{-1}$)

year	adults	10 y old	1 y old
1999	1.7	0.4	0.1
1998	1.9	0.7	0.3
1997	35.1	6.4	1.0

1996	2.0	0.5	0.1
1995	2.8	0.5	0.1
1994	9.6	1.5	0.1
1993	14.4	2.5	0.3
1992	5.0	1.1	0.3
1991	1.9	0.5	0.2
1990	2.4	1.8	1.0
1989	5.3	1.3	0.4
1988	15.7	3.1	0.6
1987	68.3	12.5	2.0
1986	47.5	7.6	0.6
1985	17.1	3.6	0.8
1984	49.4	8.0	0.7
1983	21.5	5.7	1.8
1982	5.8	4.0	2.2
1981	10.6	5.0	2.4
1980	11.6	7.1	3.8
1979	5.5	1.9	0.8
1978	4.5	1.1	0.3
1977	3.7	1.6	0.8
1976	1.3	0.5	0.2
1975	4.4	1.0	0.2
1974	2.5	1.2	0.6
1973	1.1	0.8	0.4
1972	2.9	0.8	0.2
1971	0.8	0.5	0.2
1970	1.8	0.7	0.3
1969	41.5	6.4	0.4
1968	5.9	1.3	0.4
1967	113.5	16.8	0.6
1966	13.1	4.7	2.0

Under the assumption that measurements in the different stations are representative of the real activity concentration of Pu in the air inhaled by the population, the committed dose by Pu inhalation received by an individual would never be higher than 0.53 mSv from the time of the accident until 1999. However, further refinements have to be made in order to confirm such assumption.

A preliminary approach to the **Ingestion pathway** has been also produced, considering ingestion of crops (fruit and leafy vegetables as tomatoes, watermelons, cauliflower, lettuce, etc.) and other food products (milk and snails) locally produced. Results of Pu concentration (samplings 1995-1999), from eight types of vegetables, involving 100 samples analyzed, cow's milk (7 samples), goat's milk (3 samples) and meat of snails (4 samples), together with data of Palomares population's (adults) food consumption rates, have been considered to calculate the annual intake of Pu. Dose factors in Sv Bq⁻¹ from ICRP Publication 72 were used to obtain the committed effective dose by ingestion. A value of 7.1 μSv y⁻¹ was estimated. A further study is now in progress including Am estimates and considering different age groups.

ATTACHED DOCUMENTS

- Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear (Primer Semestre del Año 2000)
CIEMAT/DIAE/PPRI/51100/3/2000
- Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear (Segundo Semestre del Año 2000)
CIEMAT/DIAE/PPRI/51100/2/2001
- Espinosa Canal A., Aragón del Valle A., Martínez Serrano J., Fernández Amigot J.A. “Estimación de la dosis total recibida por la población de Palomares debida a la radiactividad natural y a la presencia de transuránidos”. VIII Congreso Nacional de la Sociedad Española de Protección Radiológica. Maspalomas 27-29 Septiembre 2000
- Aragón del Valle A. , Espinosa Canal A., “Estudio del contenido de plutonio y americio en gasterópodos”. VIII Congreso Nacional de la Sociedad Española de Protección Radiológica. Maspalomas 27-29 Septiembre 2000