PROJECT INDALO

WORK PERFORMED DURING 2002

SUMMARY

On September 15, 1997 a new agreement between the U.S. Department of Energy (DOE) and the Centro de Investigaciones Energéticas 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 Palomares, Spain, on January 17, 1966.

During 2002, environmental and personal radiological monitoring activities have been continued. Also, according to the work proposal for the year 2001, a research plan has been designed. The final goal of the research plan is to determine the most suitable options for environmental restoration; the scientific objective is to improve the knowledge on environmental and metabolic behavior of Pu and Am. The administrative process of ownership conveyance (from the current owners to an administrative body) of the lands, where the higher residual contamination levels from the 1966 accident remain, has not been completed at present. This process is essential in order to assure the implementation of further research activities in the more affected zones. However, during 2002, the preliminary phase of the research plan started. This phase mainly consists in updating and improving the radiological characterization of the global area of Palomares. Following the recommendations made by the expert panel in its 1998 report, other studies mainly related to soils have been carried out. Soil studies in 2002 were focussed on aspects such as the improvement of the characterization of zone 3, the investigation of the influence of the run-off in sub-zone 2-0, research on radioactive particles in soils, progress in isotopic composition of the bombs and variation of Pu solubility in soils.

The activities performed in 2002 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 (1), (2).

SOILS

Zone 3

In November 2001, a systematic surface soil sampling (top 2 cm) of zone 3 was carried out, following a square network and covering a total of 24 points from a surface of 2,465 m^2, located in the direction opposite to the original contamination plume of the bomb 3. The sampling also covered a small part of the zone 5, just close to zone 3. Measurements of Am have been made in the 24 samples collected; the obtained values range from 13 to 1,520 Bq/Kg, with an average of 336 Bq/Kg, which represents an average concentration of about 2 KBq/m^2 in the studied plot (excluding the points where fragments were founded). The Am

isoconcentration representation can be observed in Figure 1. Analyses of Pu are presently in progress.

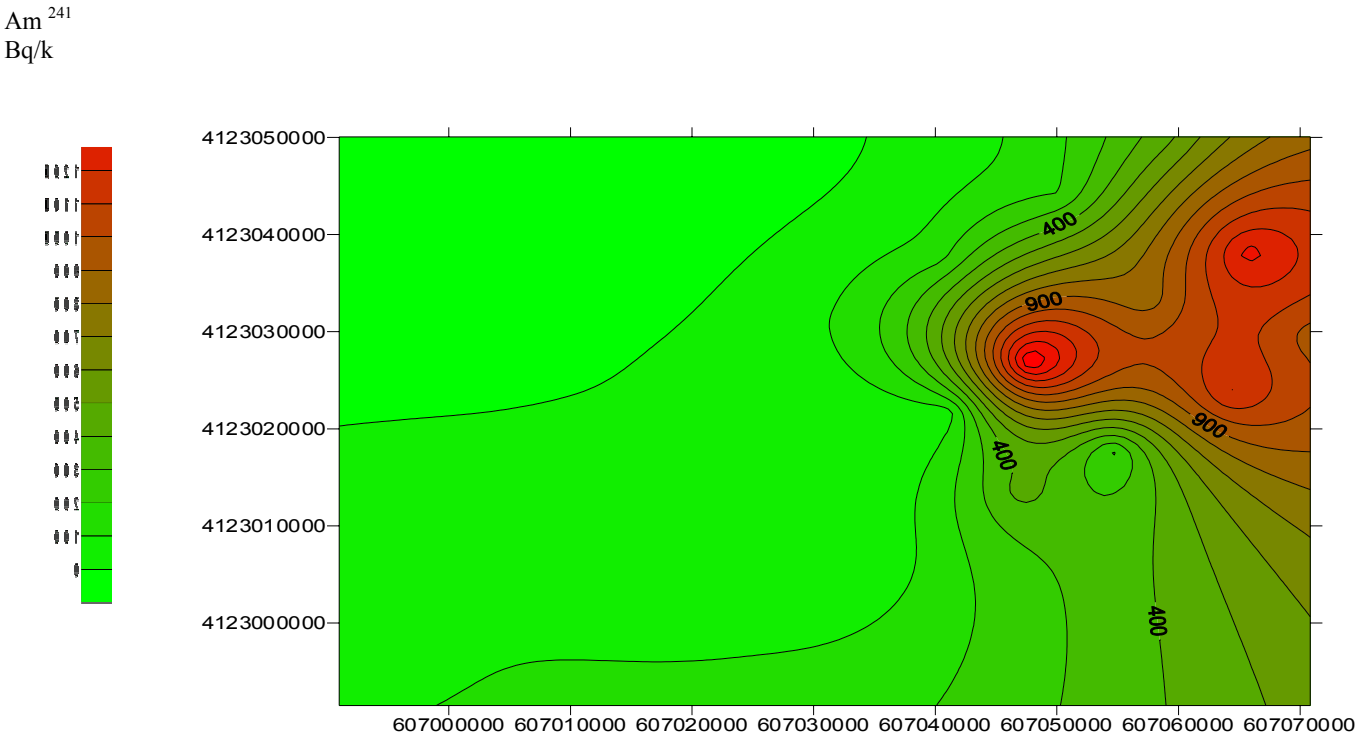


Figure 1. Representation of americium iso-concentration in zone 3-5

Simultaneously to this sampling, a foot-radiometric survey was performed covering the total surface mentioned earlier. As a result of this survey, seven new highly radioactive fragments were found and collected. These fragments are being treated in order to separate the soil dust and radioactive particles easily removed. From the smaller of these fragments, the adhered soil was separated in successive fractions by ultrasonic techniques. Then, Pu and Am were analyzed and measured in the soil fractions and one radioactive particle (named PC 21) was isolated, containing the most of the total soil activity. In this particle, and in the soil fractions mentioned, the isotopic Pu and Am composition was studied, giving the values reported in table 1. From these values, a preliminary estimate of the $^{239+240}$Pu/ ^{238}Pu and $^{239+240}$Pu/^{241}Am isotopic composition of bomb 3 fuel can be made, resulting in about 59.8 ± 2.7 and 4.2 ± 0.8 respectively. Further refinements of this estimate will be made as data became available.

Table 1. Isotopic composition in soil samples and one radiactive particle from zone 3

Sample Reference	$^{239+240}$Pu (Total Bq)	^{238}Pu (Total Bq)	^{241}Am (Total Bq)	$^{239+240}$ Pu/ ^{238}Pu	$^{239+240}$Pu/^{241}Am
E-7	66 ± 7	1.14 ± 0,12	12.8 ± 1.3	58.0 ± 1.6	5.2 ± 0.8
E-8	16.4 ± 1.7	0.26 ± 0.03	3.3 ± 0.3	64.0 ± 2.3	5.0 ± 0.7
E-9	190 ± 20	3.0 ± 0.4	47 ± 5	63 ± 4	4.1 ± 0.6
E-10	290 ± 30	4.9 ± 0.5	97 ± 10	60.0 ± 0.9	3.0 ± 0.4
E-11	94 ± 10	1.60 ± 0.17	28 ± 3	58.9 ± 1.7	3.4 ± 0.5
E-12	57 ± 6	0.98 ± 0.10	11.7 ± 1.2	58.3 ± 0.8	4.9 ± 0.3
PC 21	1,667±175			56.7±5.3	3.7±0.6
Mean				59.8 ± 2.7(σ)	4.2 ± 0.8(σ)

The presence of some fragments deeper than 10 cm in depth cannot be ruled out, since the zone has been ploughed several times.

Also, another foot survey, covering a circle 50 m diameter with its center in the impact point 3, was carried out to investigate the existence of metallic fragments in this zone located less than 10 cm in depth. The survey did not detect the presence of fragments, but one point with an exposure rate higher than in the rest of the surveyed area was identified. Therefore, the existence of some radioactive particles in this point cannot be ruled out.

Finally, in December 2002, a survey by car was also performed at the SE part of zone 3 following a square network (cultivated area to the SE of the original contamination plume); exposure rats higher than normal were detected in several plots. Lettuces and surface soil were sampled in these plots. The analyses of Pu soil activity show a range from 17 to 690 Bq/Kg for 11 of the 12 collected samples. The remaining sample show a really higher value of about 113 KBq/Kg, probably due to the existence of radioactive particles, from the nuclear fuel of the bomb, in the sample. The collected lettuces were washed and treated to make up a unique composite sample for analysis, showing a Pu value of 0.5 Bq/fresh Kg.

Zone 2

The estimate of $^{239+240}$ Pu/ ^{238}Pu and $^{239+240}$Pu/^{241}Am isotopic composition in sub-zone 2-0 is progressing. Four hot particles, from the 18 previously isolated in this zone have been analyzed. The preliminary results are showed in table 2. Further refinements of this estimate will be made as data became available.

Table 2. Isotopic ratios in radioactive particles from sub-zone 2-0

Sample Reference	$^{239+240}$Pu (total Bq)	$^{239+240}$Pu / ^{238}Pu	$^{239+240}$Pu / ^{241}Am
PC-8	1,819 ± 175	65.8 ± 5.9	3.6 ± 0.5
PC-9	2,476 ± 260	58.8 ± 4.4	4.2 ± 0.6
PC-10	1,839 ± 193	59.9 ± 5.3	5.4 ± 0.8
PC-11	1,095 ± 115	59.8 ± 3.9	4.9 ± 0.8
Mean		61.1	4.5
Standard deviation		3.4	0.8

A survey by car was done in May 2002 in the part of zone 2, close to plot 2-2. The measured exposure rates were similar to the natural background and therefore lower than the measured in sub-zone 2-0.

On December 2002, surface (top 2 cm) soil samples were taken in a part of the sub-zone 2-2 (21 samples) never studied before. Also, 12 samples of surface soil were collected in the western part of the urban area, in a zone located close to the NE border of the zone 2 with the called zero line. Analyses of Pu and Am are currently in progress.

A radiological survey was carried out in the run-off area of impact point 2 in order to investigate the extension and magnitude of this process in the dispersion of the contamination. The run-off waters drain towards the south of the impact point to the called Jatico River. River sand samples (10) were collected from the almost dry bed along a distance of 900m in a straight line away from impact point 2. The measured exposure rates and the soil Pu concentrations obtained so far (from hundred to thousands of Bq/Kg) clearly indicate the spreading of contamination. by a run-off process. The analytical work is still continuing.

Zone 6

A survey by foot was done on the top of the Villaricos hills. Seven *in situ* gamma spectra were done with a NaI detector and the peak corresponding to ^{241}Am was identified in all of them. Seven soil samples were also collected in order to quantify Pu and Am concentrations and are now in progress.

Variation of Pu solubility in soils

In the last 14 years, an increase of the Pu temporal evolution in solubility has been observed, ranging between one and two order of magnitude, depending on the

situation of the sample and on the methodology used for estimation. The percentage of solubility in 1987 and 13 years later are shown in tables 3 and 4.

Table 3. Solubility in water solution of $Pu^{239+240}$ (1986).

	Percentage of $Pu^{239+240}$ dissolved					
Leaching solution	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Mean
Distilled water	0.0020	0.0024	0.0124	0.0087	0.0154	0.0082 ± 0.006

Table 4. Solubility in water solution of $Pu^{239+240}$ (2000-2001).

	Percentage of $Pu^{239+240}$ dissolved				
Leaching solution	139-99-(2-0)	8-01-(2-0)	27-01-(3)	43-01-(6)	Mean
Distilled water	0,09	0,15	0,24	0,15	0.16 ± 0.06

The increase of solubility can be due to two reasons:

- Hot particles are easily broken both in the ground and in the laboratory while being handled (Figures 2 and 3). In this respect in 1986, the highest specific activity was found in the granulometric fraction 125-250 μm, while a sample from the same location collected in 1999, the highest activity was found between 40-60 μm (Fig 4).
- Oxidation processes convert Pu(IV) to Pu(VI), which is more soluble.

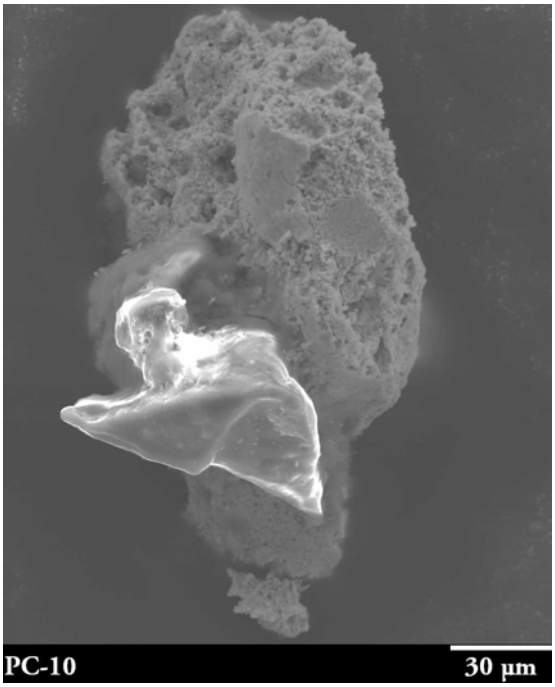
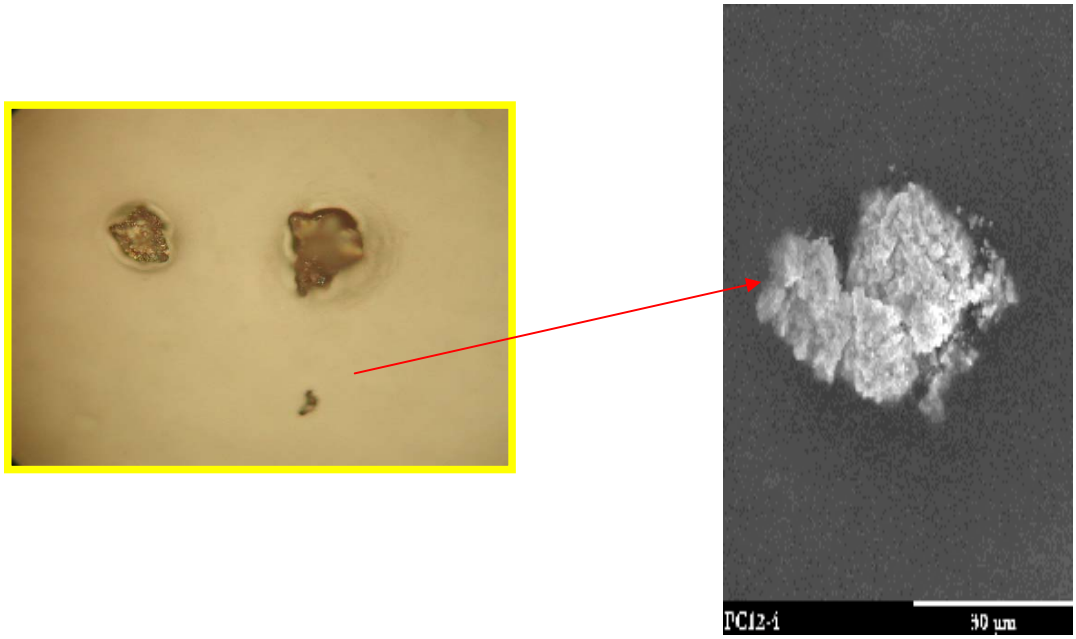


Fig. 2. Hot particle presenting a fissure at the bottom, observed in the SEM studies



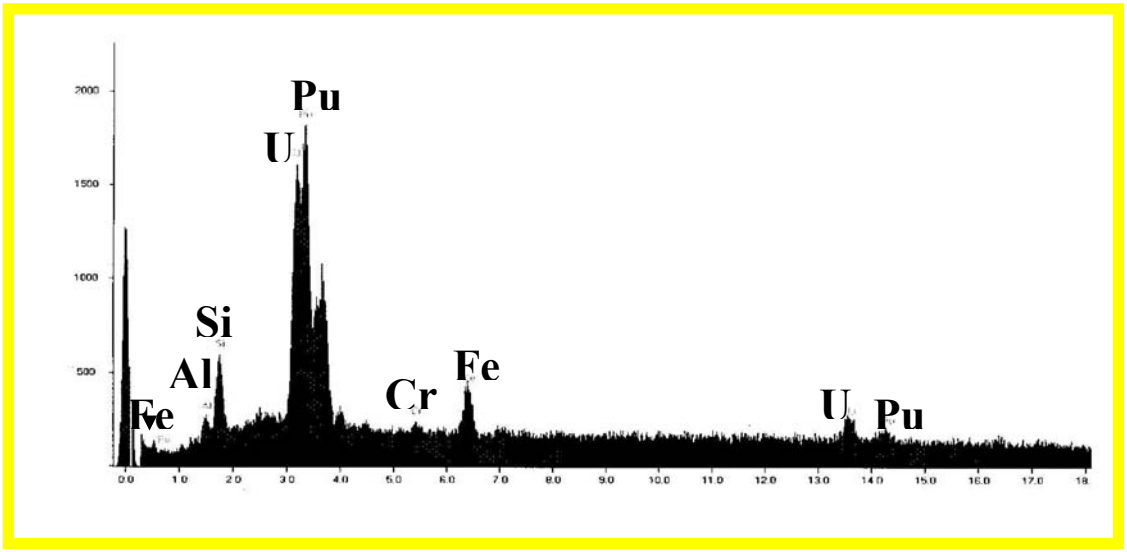


Fig. 3: Hot particle which was fissured during handling in the laboratory and its spectrum (electronic microscopy)

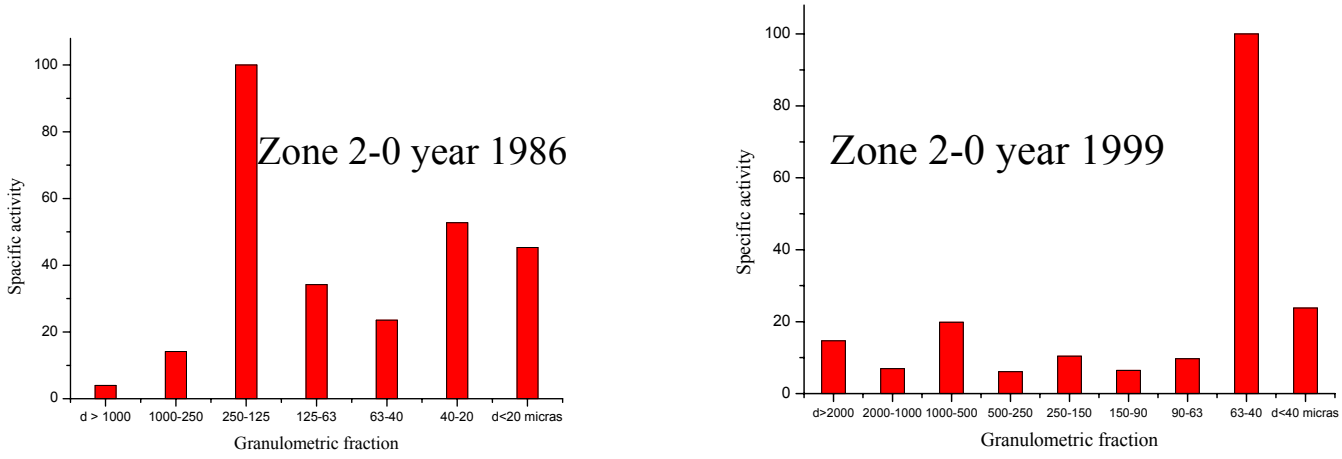


Fig 4. Variation of the specific activity of the different granulometric fractions in 1986 and 1999.

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 m^3 per sample in average. The air sampler in station 2-0 continues to be out of service from December 1999. At present a new air pump is ready to work but problems related to power supply appeared, voiding the operation. In addition, the owner of the parcel where the station 2-0 is located continues putting difficulties to operate the station. The station 2-1, failed on 24 May 2002, due to overheating of the air pump and restarted on 11 October 2002. Station 2-2 worked without interruption, with the exception of only one week (7 to 11 July) because of problems in the electrical power supply. Station P has been working during all the year 2001. In total 136 air samples were collected during 2002.

All the samples from 2001 have been analyzed and measured for Pu-239+240. Also all the samples from station P have been analyzed and measured for Am 241. Average monthly values of Pu ranged, during 2001, from 0.9 to 5.7 μBq m^{-3} in station 2-2, from 0.3 to 18.3 μBq m^{-3} in station 2-1 and from 0.3 to 35.2 μBq m^{-3} in station P. Monthly values of Am in station P, during 2001, are lower than the detection limit with the exception of the December value (2.6 μBq m^{-3}). In general, these values are in the range of the historical data series. However, the increase of Pu and Am in station P in December should be highlighted In agreement with the general trend, seasonal variability is higher than the annual one, as it could be expected considering seasonal weather changes and the timing

of agricultural practices. The results pertaining to the year 2001 are shown in the tables 5 and 6.

Table 5. Monthly concentration of $Pu^{239+240}$ in air. Year 2001

Period of sampling	ACTIVITY OF $^{239+240}$ Pu ($\mu Bq \cdot m^{-3}$)		
	Station 2-1	Station 2-2	Station P
De 29-12-00 al 03-02-01	0,68 ± 0,11	5,38 ± 0,60	2,54 ± 0,31
De 03-02-01 al 02-03-01	4,01 ± 0,56	3,28 ± 0,38	0,89 ± 0,11
De 02-03-01 al 30-03-01	18,28 ± 2,33	5,74 ± 0,65	0,69 ± 0,09
30-03-2001 al 27-04-2001	25,19 ± 2,65	3,10 ± 0,34	0,82 ± 0,11
27-04-2001 al 01-06-2001	(1) 2,99 ± 0,34	1,25 ± 0,17	1,25 ± 0,20
01-06-2001 al 29-06-2001		2,08 ± 0,24	12,87 ± 1,47
29-06-2001 al 03-08-2001		2,22 ± 0,26	0,77 ± 0,17
03-08-2001 al 01-09-2001		2,31 ± 0,26	0,71 ± 0,13
01-09-2001 al 28-09-2001		2,91 ± 0,35	0,30 ± 0,07
28-09-2001 al 02-11-2001	(2) 3,73 ± 0,42	1,71 ± 0,22	0,26 ± 0,09
02-11-2001 al 30-11-2001	(3) 0,34 ± 0,06	0,87 ± 0,12	0,77 ± 0,16
30-11-2001 al 28-12-2001	0,82 ± 0,10	1,73 ± 0,23	35,24 ± 3,76
Average	7,01	2,72	5,4
Standard deviation	9,38	1,51	11,2

Table 6 .- Monthly concentration of Am^{241} in air. Year 2001

PERIOD OF SAMPLING	Station P
	Activity of ^{241} Am μ Bq/m^3
De 03-02-01 al 02-03-01	≤ 0,02
De 02-03-01 al 30-03-01	≤ 0,02
30-03-2001 al 27-04-2001	≤ 0,02
27-04-2001 al 01-06-2001	≤ 0,02
01-06-2001 al 29-06-2001	≤ 0,02
29-06-2001 al 03-08-2001	≤ 0,02
03-08-2001 al 01-09-2001	≤ 0,02
01-09-2001 al 28-09-2001	≤ 0,02
28-09-2001 al 02-11-2001	≤ 0,02
02-11-2001 al 30-11-2001	≤ 0,02
30-11-2001 al 28-12-2001	2,60 ± 0,35

VEGETABLES

For vegetables, only 8 samples of different crops have been collected in 2002. The reason for this small number of samples collected was to compensate for the high number of samples from 2000 and 2001 waiting for analyisis. Samples are further divided in different parts (leaves, edible, etc.) and pretreated for analyzing separately in order to determine the type of contamination (external or internal) and its distribution.

Samples of vegetables collected during 2000 and 2001 and 2002 have been processed for Pu, involving 74 analyses. These samples were collected in Zones 2, 3, 5 (concretely 5-3B) and 6. With the exception of the inedible parts of some products (watermelon, tomato, olive and cauliflower), all the values were ≤ 1 Bq kg^{-1}. The exception was only one value for a sample of fruit of melon from zone 3 (8.1 Bq kg^{-1}). In general, values were higher for leaves than for fruits, even after having been washed, probably due to the wrinkled surface of the leaves.

An historical review has been performed on olive trees, since these have always been there (Table 7). As it can be observed, no clear evolutionary tendency is present, possibly because of anthropogenic reasons.

Table 7: Specific activity in olive fruit and leaves

Year	Olive fruit Bq · kg^{-1}	Nº samples	Nº positive	Olive leaves Bq · kg^{-1}	Nº samples	Nº positive
1979	1.38	2	2	2.26	2	2
1981	≥ LLD	2	0	4.29	2	2
1982	9.66	4	3	4.59	4	4
1983	0.36	2	1	Not sample		
1984	≥ LLD	1	0	3.5	1	1
1987	0.20	1	1	≥ LLD	1	0
1990	0.04	2	1	0.63	1	1
1992	0.05	1	1	≥ LLD	1	0
1993	0.05	1	1	2	1	1
1995	0.07	1	1	6.50	3	3
1997	0.10	2	2	2.87	2	2
1998	1.14	1	1	3.39	1	1
2001	0.03	1	1	13.4	1	1
2002	0.20	1	1	0.07	1	1

OTHER PRODUCTS

Concerning milk samples, 6 samples were collected, analyzed and measured for Pu and Am (radiochemistry followed by alpha spectrometry), 3 of them corresponding to cow milk and the other 3 corresponding to goat milk. The results

are shown in table 8 and present values lower than LLD or in the order of a few mBq/l. The exception was only one sample of goat milk presenting values in the order of Bq/l

Table 8. $^{239+240}$Pu and ^{241}Am in cow and goat milk samples

Reference	Sampling date	$^{239+240}$Pu Concentration mBq· l^{-1}	^{241}Am Concentration mBq· l^{-1}
1-cow-02	21-10-02	3.9 ± 0.9	≤ 0.46
2-goat-02	21-10-02	2.9 ± 0.7	≤ 0.44
3- cow -02	28-10-02	2.7 ± 0.7	≤ 1.26
4- goat -02	28-10-02	178.5 ± 19.4	57.3 ± 9.1
5- cow -02	04-11-02	23.0 ± 3.1	2.6 ± 1.2
6- goat -02	04-11-02	3.3 ± 0.7	≤ 0.44

In relation to snail samples, 1 sample of 467 g has been analyzed. Prior to their incineration, they were treated according to the normal cooking procedures. A total of 8 Pu analyses corresponding to the different parts of the sample and process have been made. The results are shown in Table 9. From these results a total rounded activity in snails of 26 Bq/Kg is obtained. The edible parts (soft tissue and boiling water) account for a rounded activity of 5 Bq/Kg, which represents less than 20 % of the total.

Table 9. $^{239+240}$ Pu in different parts of a snail sample

Reference	Sample description	$^{239+240}$ Pu Activity mBq in the total sample (467g of snails)
4 -C-00-2-0	Solid excrements	7248 ± 763
5 -C-00-2-0	Mucus	16 ± 2
3-C-00-2-0	Particles in water (∅ > 0.45 μm)	65 ± 7
2-C-00-2-0	Particles in water (0.22< ∅ <0.45 μm)	0.8 ± 0.2
1-C-00-2-0	Water from washing (filtered by 0.22 μm)	122 ± 13
6-C-00-2-0	Boiling water	127 ± 14
7-C-00-2-0	Shell	2472± 395
8-C-00-2-0	Soft tissue	2110 ± 226

URINE

During 2002, 149 people from Palomares were transported to CIEMAT headquarters in Madrid for medical examinations and sampling of 24-hour urine collections and further bioassay analyses for internal dosimetric assessments. The number of 12-year old children, controlled for the first time, is 15. 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 only 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 149 people examined during 2002 were completed. Only 3 persons, corresponding to 3 women of different ages (64, 26 and 52 years old) resulted in positive values. The 64 years-old woman showed a value of 0.7 mBq in 24 h urine and she had been controlled in five occasions previously, always with negative results. The 26 years-old woman showed a value of 2.8 mBq in 24 h urine and she had been controlled in 6 occasions previously, 5 times with negative results and once with a positive value; based on the positive results, the related dosimetric assessment resulted in an committed effective dose (50 years) of 47 mSv. Finally, the 52 years-old woman showed a relative high value of 97.3 mBq in 24 h urine; she was controlled previously on five occasions, always with negative results; given the relative high value obtained, the possibility of external contamination of the sample cannot be ruled out and, therefore, she will be requested to return the next year for a new control. Based on the results of this future analysis, the related dosimetric assessment will be decided.

The percentage of positive values for the year 2002 (about 2 %) is lower than the percentage for the period 1967-2001 which is 3.9 % (144 positive values in a total of 3,693 measurements). The distribution by ranges of these results, for the total period, 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 2001 do not modify the conclusions from the expert panel in its 1998 report.

With respect to Am, the urine analyses of 80 from the 149 people examined during 2002 have been completed. 79 samples showed values below the detection limit of 0.37 mBq in urine of 24 hours. Only 1 positive result was obtained corresponding to the woman 52 years old mentioned before, with a anomalous value for Pu. The value obtained for Am was of 22.5 mBq in urine of 24 h, which support the suspicion of external contamination of the sample.

During 2002, no measurements in the whole body counter were made.

The individual results of bioassay for each annually examined person 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 (149 people) performed during 2002.

ATTACHED DOCUMENTS

(1) Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear. (Primer Semestre del Año 2002). CIEMAT/DIAE/PPRI/51100/03-2002).

(2) Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear. (Segundo Semestre del Año 2002). CIEMAT/DIAE/PPRI/51100/02-2003).

(3) Comportamiento ambiental de las partículas de combustible nuclear (fundamentalmente Pu) tras un accidente nuclear en un ecosistema de tipo mediterráneo. ESPINOSA, A. (Tesis doctoral, Noviembre de 2002).

(4) Location and isolation techniques of radioactive particles in Plutonium and Americium contaminated soil. . ESPINOSA, A.; ARAGÓN, A.; DE LA CRUZ, B.; FERNANDEZ, J.A. Pre-Conference Workshop Advanced techniques and radionuclide speciation within radioecology. IAEA-MEL, Monaco, August 2002

(5) Transuranics sequential extraction procedures: a comparison study. GASCÓ, C.; ANTÓN, M.P.; GONZÁLEZ, A.M.; MERAL, J. In International Conference on Radioactivity in the Environment. 1-5 September 2002. Mónaco.

(6) Distribution profiles of Pu, Am and Cs in margin sediments from the western mediterranean (spanish coast). GASCÓ, C.; ANTÓN, M.P.; GONZÁLEZ, A.M.; MERAL, J.; PAPUCCI, C.; DELFANTI, R. 2002. Journal of Environmental Radioactivity 59, 75-89.

(7) A new method to isolate americium from environmental samples using diphonex resins. GASCÓ, C.; ANTÓN, M.P.; NAVARRO, N.; GONZÁLEZ, A.M. 2002. Radioprotection 37, C1. Febrero 2002. Ed. F.Bréchignac. ISSN 0033-8451. ISBN 2-86883-596-1.

(8) Aplicación del programa GENIE-2000 al análisis de espectros alfa. ÁLVAREZ, A.; GASCÓ, C.; NAVARRO N. Y ANTÓN, M. P. Congreso de la SEPR. Bilbao. Mayo 2002.