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MAN-MADE RADIOACTIVITY IN THE ALMANZORA GULCH AND BEACH EDGE OF PALOMARES, SPAIN

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Abstract — The $^{239+240}$Pu, ^{238}Pu and ^{137}Cs distribution along the Almanzora Gulch and beach edge have been studied. These areas were affected by the accident of 1966, in which a non-nuclear explosion of two thermonuclear bombs occurred. Fluvial sediments at 0–5 cm (in 1990) and 0–10, 10–20, 20–30 cm depth (in 1991) were sampled. The surface inventory (0–5 cm) of radionuclides ranges from 0.4 to 1.8 Bq.m^{-2} of $^{239+240}$Pu and from 1 to 60 Bq.m^{-2} of ^{137}Cs. The estimated total inventory (0–30 cm) shows values from 20 to 264 Bq.m^{-2} of $^{239+240}$Pu and from 328 to 1147 Bq.m^{-2} of ^{137}Cs. The ratio $^{239+240}$Pu/^{137}Cs changes significantly close to the river mouth, probably due to the different behaviour of both radionuclides. At one sampling station a contribution from the Palomares accident was noted based on the ratio ^{238}Pu/$^{239+240}$Pu. It seems evident that until now no influence from the Chernobyl accident has been detected in this area. The granulometry of the sediments manifests the river hydrodynamics, a dry gulch with periods of floods typical of certain Mediterranean rivers. An important input of terrigenous material containing radionuclides to the adjacent continental shelf occurs in these periods.

INTRODUCTION

The studies in the field of the environmental behaviour of radionuclides grew in Spain as a consequence of the development of the nuclear industry. Wide programmes of radionuclide monitoring were carried out at the beginning of the 1970s in areas close to the discharges from nuclear installations. The aims were to determine the radiological impact of their releases into the nearby ecosystems. Other zones, like Palomares, were selected to study in depth the radioecology of transuranics and the long-term consequences derived from the so-called 'Palomares accident'.

Palomares has received two different inputs of radionuclides: the fallout from nuclear weapons testing of the 1960s, as did other regions located at the same latitude band, and an extra input due to an aircraft crash in 1966 in which an aerosol of transuranics was accidentally released[1,2]. Even though efficient countermeasures were applied, part of the transuranic contamination remained in the region[3]. The radiological environmental surveillance was achieved by JEN the year of the accident and carried on by CIEMAT until the present time.

The study of the transuranics migration between the terrestrial and marine ecosystems at Palomares started in 1985. Marine sediments from the Vera Gulf were collected to study the depositional history of the translocated transuranics. The results were published in the literature[4–10], emphasising the importance of the terrigenous input of these elements by the Almanzora river to the Continental shelf off Palomares. This would mean a decrease of the measured transuranic concentrations along the river bed and beach edge, estimated as 1–200 kBq.m^{-2} of α emitters right after the accident[5]. The present distribution of radionuclides in both the Almanzora river bed and beach edge has been analysed, to extend knowledge of the behaviour of transuranics in Mediterranean rivers and to consider the exposure of the population due to particular utilisation of the land (sunbathing on the beach and use of beach sand in greenhouse agriculture). The Almanzora river is a gulch affected by semidesert weather: it seldom rains, but this zone reaches the maximum rainfall per day in Spain. Floods and other catastrophic situations are suffered by the local inhabitants. As a consequence of these heavy rains, big masses of ground soil are transported from land to sea.

Three aspects were considered appropriate to the study of the area:

(1) The hydrodynamic of the river could be described by the granulometry of the riverine sediments, being an important tool to explain the redistribution of the radionuclides after the periods of floods.
(2) The values of $^{239+240}$Pu, ^{238}Pu and ^{137}Cs activities along the river were expected to be very low except in the areas unaffected by the floods or the areas affected by the dry fallout of the dust from the resuspension of the Palomares soil.
(3) The different sources of plutonium could be defined by the ^{238}Pu/$^{239+240}$Pu ratio (0.018 accident, 0.04–0.06 fallout of weapons tests in this band of latitude). The $^{239+240}$Pu/^{137}Cs ratio could be also an indicator of the source term; but the distinct behaviour of these elements could have modified it.

METHODOLOGY

Sampling and analysis

Two sampling campaigns were carried out. The first one was done in 1990 to determine the plutonium activity levels on the surface riverine sediments, which was expected to be low after the floods occurred in 1970, 1973, and 1982. In this campaign, five sampling transversal lines were marked out onto the river bed and three stations were selected for each line. The sampling network is shown in Figure 1. Three areas were traced on the beach in order to obtain representative samples. Five clusters were chosen and forty-four surface areas $10 \times 10 \times 5\ cm^3$ were selected as representative backgrounds. The second campaign, in 1991, was intended to describe the redistribution of radionuclides in vertical profiles. Six stations were selected where plutonium and caesium had been determined previously. Note that it was impossible to get samples close to the river mouth (Stations 5 and 6) as it was completely inaccessible at that time because a bridge was being build. Because of the energetic transport in the flood periods, the downstream sediments are mainly stones and gravels and two different sampling techniques had to be tested: core and trench. The latter technique was selected as the more appropriate for this kind of sediment. Sediment trenches (0–10, 10–20 and 20–30 cm depth were retrieved in each station.

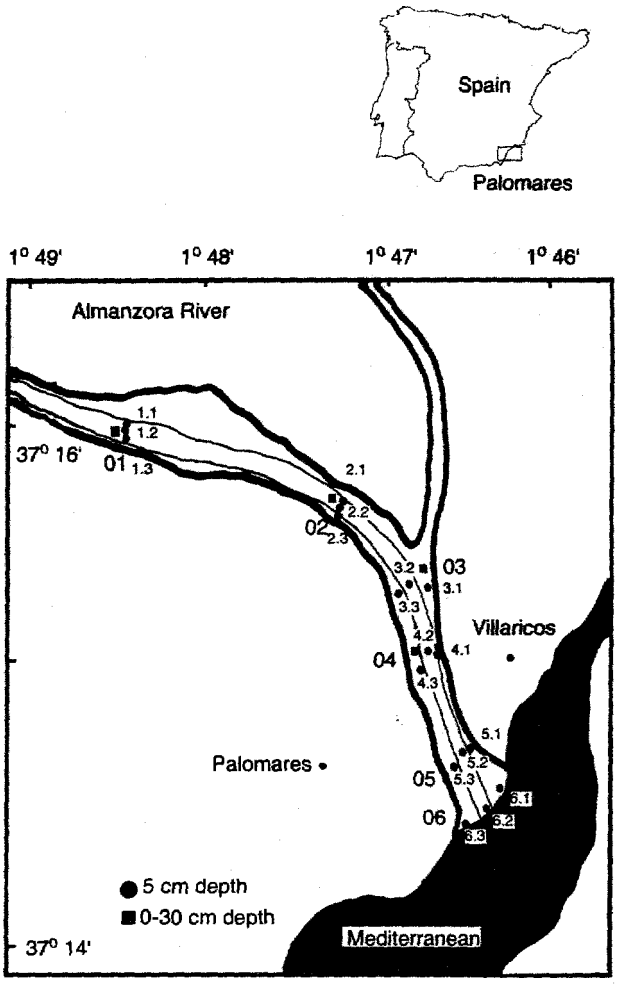

Figure 1. Sampling stations in the Almanzora river and beach edge, Palomares, Spain.

Pretreatment

Two kilograms of sediment were dried at 50°C for 5 days to constant weight. The dry samples were sieved through $1 \times 1\ mm^2$ mesh size. The gravels and stones were rejected as there is evidence that the majority of the activity is associated with fine sediment particles. The weight per unit area ($g.m^{-2}$) was determined for each sample.

Analytical method

Plutonium was extracted from the sediment matrix using two leaching techniques[11,12]. For surface sediments, 15 g were attacked with a mixture of $HF/HNO_3/HCl$. For trenches, 75 g of sediment were analysed. The plutonium was leached in this case with 8N HNO_3 three times.

These solutions containing the transuranic were passed through anion exchange columns and finally electroplated onto stainless steel discs, according to Talvitie's procedure[13] for α spectrometry counting. ^{137}Cs was determined by gamma spectrometry using a hyperpure coaxial n-type germanium detector. The accuracy of the analytical procedure was checked with standard samples of sediments. Intercalibration exercises confirm the reliability of the data.

The grain size composition was studied by sieving the components with a grain size >0.062 mm, and the components <0.062 μm were measured with a Coulter TAII in a tube of 280 μm with isoton electrolyte.

RESULTS AND DISCUSSION

The activity concentrations of radionuclides in the surface sediments (5 cm depth) sampled in 1990, are shown in Table 1. These concentration levels ranged from 0.041 to 0.2 $Bq.kg^{-1}$ of $^{239+240}Pu$ and from 0.2 to 5.2 $Bq.kg^{-1}$ of ^{137}Cs. These values are in good agreement with those published in the literature for other Mediterranean areas[14–16]. Although few data from artificial radioactivity in Mediterranean river sediments are found in the Spanish literature, an excellent compilation of the radionuclide fallout levels after the Chernobyl accident in soils was done by the Consejo de Seguridad Nuclear[17] being the unique realistic data on the concentration of artificial radionuclides in Spanish territory.

The decreasing of the caesium concentration close to the Almanzora river mouth should be explained. The mean ratio between $^{239+240}Pu/^{137}Cs$ changes significantly in this area from 0.04 (average value of Stations 1.1 to

5.3) to 1.0 (average value of Stations 6.1, 6.2 and 6.3) and it could be attributed to a different behaviour of both radionuclides close to the estuary in the periods of floods. The decreasing activity and inventory of ^{137}Cs in the downstream sediments could be the result of the salinity differences encountered in the water since it starts with fresh water upstream and approaches the saline water of the Mediterranean. A similar difference was also noted in the Hudson River Estuary[18,19] where it was found that the magnitude of the ^{137}Cs Kd value could be expressed as an inverse function of the chloride ion water concentration. In others words, ^{137}Cs originally associated with fresh water sediments is released back to solution when salt water is encountered. The Kd is larger in a body of fresh water than in a salt water environment. Simpson[18,19] also noted that there are no differences in the $^{239+240}Pu$ Kd between fresh and brackish water. In our case, similar concentrations have been found in the river sediments between Stations 1.1 to 5.3 (2.6 ± 1.5 Bq.kg^{-1} of ^{137}Cs, average value), and a lower concentration at Stations 6.1, 6.2 and 6.3 (0.21 ± 0.06 Bq.kg^{-1} of ^{137}Cs) near the river mouth.

In contrast, the concentration of plutonium is higher at Station 6 (6.1, 6.2 and 6.3) than at the upstream stations. Much of the $^{239+240}Pu$ has migrated downstream (possibly in association with river-borne particles) and is now found accumulated in higher levels in the sediments near the river mouth. This is a noteworthy feature and demonstrates the mobility of plutonium in this particular environment.

Table 1. Inventories* and concentration activities in Almanzora river sediments (surface, 1990).

Stations	Activites $\pm 2\sigma$ (Bq.kg^{-1})		Inventories (Bq.m^{-2})		Ratios Pu/Cs
	$^{239+240}Pu$	^{137}Cs	$^{239+240}Pu$	^{137}Cs	
1.1	0.079 ± 0.018	5.2 ± 0.6	0.93	61.1	0.015
1.2	0.081 ± 0.016	1.4 ± 0.4	1.53	25.5	0.058
1.3	0.111 ± 0.028	4.9 ± 0.8	0.64	28.1	0.023
2.1	0.041 ± 0.016	1.4 ± 0.8	0.41	13.8	0.029
2.2	0.055 ± 0.016	3.4 ± 0.7	0.66	40.5	0.016
2.3	0.080 ± 0.022	5.5 ± 0.9	1.08	73.7	0.015
3.1	0.057 ± 0.012	1.5 ± 1.0	0.52	13.6	0.038
3.2	0.064 ± 0.020	2.7 ± 1.3	0.70	29.8	0.024
3.3	0.073 ± 0.026	1.5 ± 1.0	0.93	19.3	0.049
4.1	0.046 ± 0.026	3.2 ± 1.4	0.72	50.5	0.014
4.2	0.053 ± 0.020	2.0 ± 1.5	0.60	22.6	0.027
4.3	0.232 ± 0.060	1.5 ± 1.0	1.80	11.4	0.155
5.1	0.110 ± 0.026	2.6 ± 1.0	0.65	15.6	0.042
5.2	0.055 ± 0.014	1.3 ± 0.9	1.07	24.9	0.042
5.3	0.074 ± 0.030	1.6 ± 0.5	1.35	30.0	0.046
6.1	0.189 ± 0.040	0.23 ± 0.06	1.09	1.3	1.217
6.2	0.192 ± 0.080	0.19 ± 0.06	1.15	1.1	1.011
6.3	0.156 ± 0.040	0.21 ± 0.06	0.90	1.2	0.740

* 5 cm depth

In the few samples where the ^{238}Pu concentration was above the detection limit (0.014 Bq.kg^{-1}) the $^{238}Pu/^{239+240}Pu$ ratios (see Table 2) were at fallout levels (0.04–0.06 at this latitude). This leads to the conclusion that the origin of the Pu measured is due to global fallout from nuclear weapons testing.

The possible redistribution of radionuclides in vertical profiles was studied in 1991. The results of concentration activities, total inventories and radionuclides ratios are shown in Table 2. The trenches of Stations 1 to 4 show a clear homogenity on plutonium distribution; this is explained by the characteristics of these river sediments: during the period of floods the sediments are well homogenised and the transuranic distribution seems equal in the different transects. The distribution of caesium shows changes with the depth; these differences can be explained by the chemical exchangeability of the caesium in the soil column depending on the main components. The activity of plutonium found in the trench 3 is one order of magnitude higher than in the other trenches, indicating the existence of another input of Pu besides global fallout: it could be some plutonium from the original surface contamination caused by the 1966 accident that has not been washed down in the floods, or the plutonium resuspended from the adjacent zone by the winds and the later deposition on the river bed in the dry season. The $^{239+240}Pu/^{241}Am$ ratios also are indicative of the transuranics origin (accidents or nuclear weapons testing fallout). Due to the low chemical recovery of the radioanalytical method for americium and the low level activities in the 10 g samples, only in the areas influenced by the Palomares accident was the americium determined. This is the case in Station 03: the ^{241}Am concentration in the top layer (0–10 cm) was determined by γ spectrometry ($0.47 \pm 0.21(2\sigma)$ Bq.kg^{-1} of ^{241}Am). It should be noted that gamma measurements and alpha radioanalysis were performed in different aliquots of the sediment. Therefore, the Am/Pu ratios were not calculated as it was preferred to isolate the detected americium heterogeneity rather than determine the plutonium in this fraction.

The calculated low values for global inventories, 20 to 45 Bq.m^{-2} of fallout $^{239+240}Pu$, when compared with the 80 Bq.m^{-2} of $^{239+240}Pu$[20] estimated at this latitude, could be explained by the mean rainfall in Almeria (semidesert climate) and the occasional sweeping by the floods. The same explanation could be given for caesium. The inventories ranged from 328 to 1147 Bq.m^{-2} of ^{137}Cs and the estimated fallout is 4000 Bq.m^{-2} of ^{137}Cs[21].

The granulometry of the sediments was determined. This granulometry indicates the dynamics of sedimentation along the river. This, at all stations in the river, is indicative of sand, except at Station 4.1 where it is a

Table 2. Activity concentration and inventories of radionuclides in Almanzora river sediments (trenches, 1991).

Stations	Depth (cm)	Activity concentration ± 2σ (Bq.kg^{-1})			Ratios (±2σ)		Inventories (Bq.m^{-2})	
		$^{239+240}$Pu	^{238}Pu	^{137}Cs	^{238}Pu/$^{239+240}$Pu	$^{239+240}$Pu/^{137}Cs	$^{239+240}$Pu	^{137}Cs
	0–10	0.147 ± 0.010	0.011 ± 0.010	0.97 ± 0.17	0.24 ± 0.21	0.049 ± 0.012	6.4	133
01	10–20	0.026 ± 0.014	0.005 ± 0.006	0.84 ± 0.08	0.21 ± 0.27	0.031 ± 0.017	7.7	240
	20–30	0.043 ± 0.012	0.004 ± 0.006	—	0.10 ± 0.14	—	5.2	—
	0–10	0.068 ± 0.020	0.009 ± 0.012	1.02 ± 0.18	0.13 ± 0.17	0.067 ± 0.024	10.9	157
02	10–20	0.098 ± 0.010	0.004 ± 0.002	2.80 ± 0.22	0.04 ± 0.02	0.035 ± 0.004	27.8	787
	20–30	0.028 ± 0.008	0.004 ± 0.003	0.82 ± 0.14	0.15 ± 0.14	0.034 ± 0.012	6.9	203
	0–10	1.898 ± 0.200	0.035 ± 0.012	1.17 ± 0.10	0.018 ± 0.007	1.62 ± 0.23	243	151
03	10–20	0.130 ± 0.022	0.025 ± 0.012	2.68 ± 0.18	0.19 ± 0.10	0.05 ± 0.01	13.9	346
	20–30	0.086 ± 0.016	0.020 ± 0.010	2.07 ± 0.16	0.23 ± 0.13	0.04 ± 0.01	7.4	190
	0–10	0.082 ± 0.012	0.014 ± 0.010	1.07 ± 0.11	0.16 ± 0.13	0.07 ± 0.02	11.3	228
04	10–20	0.006 ± 0.044	<LID	0.54 ± 0.06	—	0.10 ± 0.08	5.3	63
	20–30	0.054 ± 0.010	0.005 ± 0.002	0.41 ± 0.08	0.09 ± 0.07	0.13 ± 0.04	4.9	37

— Not determined (Loss of sample)

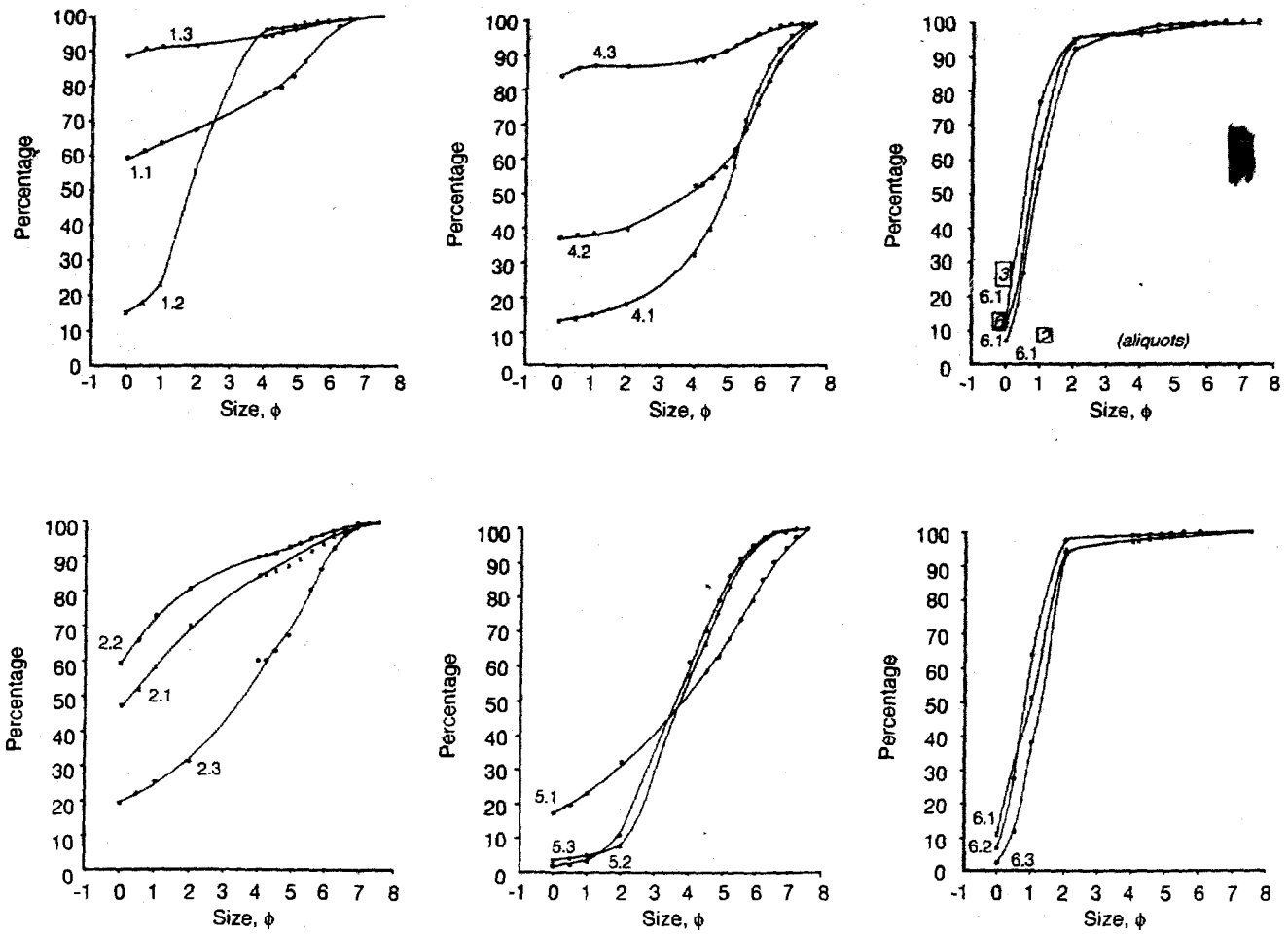

Figure 2. Accumulated granulometric curves. Distribution of percentage of grain size and ϕ where $\phi = -1$ g_2 diameter of particle.

sandy lime. The clay content, at 2.3%, is very low, the river sediments being depleted of this more chemically reactive component as usually there is an enrichment of transuranics in the clays fractions. In Figure 2, the curves of grain size distribution in the surface sediments (0–5 cm) are shown. Curve 1.2 corresponds to a well segregated sand, while in the margin of the river (Curves 1.1 and 1.3) the curves show more variation in energy and less selective sedimentation capacity. Similar profiles to these were obtained in most of the transects. At Stations 6.1, 6.2 and 6.3 (beach edge), the grain size is uniform, indicating the predominant influence of the marine media. Distributions such as those at Station 1.1 and Station 1.3, which show significant percentages of very large size particles (gravel), reflect the occasionally high hydrodynamic energy of the system in question, i.e. the flash floods which occur very infrequently.

CONCLUSIONS

The behaviour of global fallout ^{239}Pu and ^{137}Cs seems to be different along the Almanzora river bed: while the plutonium concentration increases near its mouth, the caesium concentration decreases. This loss of caesium close to the river mouth could be due to a better scavenging of this radionuclide in these particular estuarine conditions.

The inventories of plutonium and caesium in the Almanzora sediments are lower than those estimated for this latitude band. The sparse rainfall index, along with the efficient transport or riverine material during the periods of flooding towards the adjacent continental shelf could be responsible for these reduced values.

The detection of a specific area (Station 3) containing an 'unpredicted high concentration' for both plutonium and americium points to the presence of an extra contribution of transuranics besides global fallout. The $^{238}Pu/^{239+240}Pu$ ratio confirmed the Palomares accident of 1966 as the source term of this inhomogeneity. This heterogeneity might have been transported from the nearby area (in which a residual transuranic contamination from the accident remains) by strong winds responsible for the resuspension phenomena existing in this zone. This heterogeneity, after being delivered by the air flows has been deposited at random on the dry river bed during the dry season.

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REFERENCES

1. Iranzo, E. *Geochemical Distribution of Plutonium and Americium in Palomares Soils.* In: Cycling of Long-lived Radionuclides in the Biosphere. Observations and Models. Report 2 (Madrid: CIEMAT) (1987).
2. UNSCEAR. United Nations Scientific Committee on the Effects of Atomic Radiation. *Report to the General Assembly.* New York: United Nations (1988).
3. Iranzo, E. *Air Concentrations of $^{239+240}Pu$ and Potential Radiation Doses to Persons living near Pu-Contaminated Areas in Palomares, Spain.* Health Phys. **52**, 453 (1987).
4. Iranzo, E., Gascó, C., Romero, L., Martínez, A., Mingarro, E. and Rivas, P. *Temporal Distribution of Pu and Am in the Marine Environment of Southern Coast of Spain.* Report CIEMAT 641 (Madrid: CIEMAT) (1989).
5. Gascó, C. *Estudio de la Distribución de Plutonio en el Ecosistema Marino de Palomares después de una Descarga Accidental de un Aerosol de Transuránidos.* Colección de la Universidad Complutense de Madrid, PhD.n° 104/91. M-11189-1991 (1990).
6. Romero, L. *Estudio del transporte tierra-mar de elementos transuránidos. Aplicación al accidente de Palomares (Almería) de 1966.* Colección de la Universidad Complutense de Madrid. PhD (1991).
7. Gascó, C., Iranzo, E. and Romero, L. *Transuranic Transfer in a Spanish Marine Ecosytem.* J. Radioanal. Nucl. Chem. **156**, 151–163 (1992).
8. Romero, L., Lobo, A., Holm, E. and Sánchez, J. A. *Transuranics Contribution off Palomares Coast: tracing History and Routes to the Marine Environment.* In: Radonuclides in the Study of Marine Processes. Eds P. J. Kersaw and D. S. Woodhead (Shannon: Elsevier Science) (1991).
9. Romero, L., Lobo, A. and Holm, E. *New Aspects on the Transuranics Transfer in the Palomares Marine Environment.* J. Radioanal. Nucl. Chem. **161**, 489–494 (1992).
10. Gascó, C., Romero, L., Mingarro, E. and Lobo M. A. *Geochemical Aspects and Distribution of Long-lived Radionuclides in Marine Sediments from Palomares.* J. Radioanal. Nucl. Chem. **161**, 389–400 (1992).
11. Wong, K. W. *Radiochemical Determination of Plutonium in Sea Water, Sediments and Marine Organisms.* Anal. Chim. Acta **56**, 355–364 (1971).
12. Holm, E., Fukai, R. and Ballestra, S. *A Method for Ion-Exchange Separation of Low Levels of Americium in Environmental Materials.* Talanta **26**, 791–794 (1979).
13. Talvitie, N. A. *Electrodeposition of Actinides for Alpha Spectrometric Determination.* Anal. Chem. **44**, 280–283 (1972).