

*GEOCHEMICAL DISTRIBUTION OF PLUTONIUM AND AMERICIUM
IN PALOMARES SOIL*

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Abstract. An aerial accident on January 17, 1966, over the Palomares (Almería) region, caused four thermonuclear bombs to be dropped, resulting in fractionation of two of them and ignition of part of their core components. The aerosol created by this accident contaminated 226 hectares of cultivated and unimproved calcareous-silicic land.

Soil samples from some chosen parcels were sampled fifteen years after the accident to study the geochemical distribution of plutonium and americium. The soil in each of the parcels of land has been divided into nine parts in order to analyze the distribution of actinides from particles of less than 5 microns to those of more than 1 mm in size, with the maximum build-up falling between 63 and 250 microns. In each of these divisions studies have been made of the chemical and mineral composition of the soil, and through radiographic techniques it has been determined that the minerals most responsible for actinide retention are iron oxides, carbonates and phyllosilicates.

In addition, studies were made to determine the electrochemical potential and chemical composition of equilibrium waters obtained by maceration of soils from each parcel. Values for these potentials are between 7.6 and 8.4 for pH, between 312 and 344 mV for Eh and between 484 and 2500 μ S for conductivity. The salinity usually found is partially conditioned by the brackish waters used for irrigation.

1. INTRODUCTION

On January 17, 1966, an aviation accident took place during a supply flight over the Palomares airspace in the municipality of Cuevas de Almanzora, province of Almeria. This accident resulted in four thermonuclear bombs being dropped; two of them had parachutes and were recovered intact, one in the dry riverbed of the mouth of the Almanzora River, the other in the sea. The other two bombs fell without parachutes and suffered fragmentation of fissionable fuel with ignition of part of it. The resulting aerosol was carried by the wind and contaminated, to differing degrees, an area of 226 hectares comprising brushlands, farmland and urban areas. Most of the urban area was not contaminated.

Figure 1 shows the contaminated regions defined according to the degrees of superficial alpha contamination measured immediately after the accident.

In places where superficial alpha contamination reached values exceeding $1.2 \text{ MBq} \times \text{m}^{-2}$, the surface layer was removed to a depth of approximately 10 cm. This layer of soil and contaminated vegetation was provisionally deposited in a site near impact number 2, packed in steel drums and later sent to the United States as radioactive wastes. The rest of the contaminated region used for agriculture was watered and plowed to a depth of approximately 30 cm in order to dilute the contaminated soil with clean soil and thus reduce the concentration of radionuclides on the surface.

This report deals with the studies that have been made to determine levels of correlation existing between Pu-239 + Pu-240 and Am-241 concentration and the granulometric, mineralogical and chemical composition of the soil. These aspects are important for evaluating the risks attending dispersion of such contaminants through atmospheric re-suspension, erosion and hydrological movement.

2. METHODOLOGY OF SAMPLING AND ANALYSIS

2.1. Sampling

The samples used in this study were taken from parcels 2-0, 2-2, 3-1, 3-2, and 5-3B. Their location is indicated in Figure 1.

Parcels 2-2, 3-1, 3-2, and 5-3B are usually cultivated; parcel 2-0 has been under cultivation for only one year. Parcel 5-3B is situated outside the area that was initially contaminated.

The samples were taken in 1981 from regular points distributed along diagonals on a squared area (50 x 50 m) of each parcel, to a depth of 15 cm.

2.2. Analyses

Analyses were made of aliquote parts obtained through splitting dry samples by means of a Centrifugal Sample Divider.

2.2.1. Granulometry

The samples were subjected to maceration and ultrasonic separation. From each sample, eight size fractions were obtained with the following size intervals: $\geq 1000-250-63-40-20-10-5 \geq \mu\text{m}$.

Fractional division of $> 63 \mu\text{m}$ particles, after leaching with water over a mesh of a prescribed size, was carried out by screening according to NLT 104/72 standards (1).

Particles $< 63 \mu\text{m}$ in size were fractionated by the "British Rema" air classifier; mean density was considered to be 2.7 gm/cm^3 .

Granulometric distribution of components $< 5 \mu\text{m}$ in size was found with

a "Coulter TA II" particle counter.

Sherard methodology was used to determine the dispersion index (2).

2.2.2. Americium

The presence of Am-241 was measured by analysis of 15 gr samples through low-energy photon spectrometry with intrinsic germanium detectors.

2.2.3. Plutonium

The measurement of Pu-239 and Pu-240 was carried out on the same samples as were previously used for measuring Am-241, by alpha spectrometry using silicon block semiconductor detectors. The process calls for prior acid extraction with 1M HF and 12 N HNO₃, separation of plutonium with DOWEX AG-1X2 (50-100 mesh) resin, and electrodeposit of the evaporated resin eluate residue dissolved in HCl and ammonium oxalate. The chemical yield is measured by addition of Pu-236 as a tracer.

2.2.4. Mineralogical Composition

The mineralogical analysis was carried out through optic microscopy and X.R diffraction. The second technique was used to analyze each of the granulometric fractions. The quantitative analysis of the mineral species was based on the intensity of the most important reflections. The value of the intensities, I, represents arbitrary units, being 100 the value of I for the highest reflection intensity in the different granulometric fractions.

2.2.5. Autoradiographies

They were obtained on plastic, type CR-39 (oxydi-2, 1-ethanedyl di-2-

propenyl diester of carbonic acid), placed over thin polished sections of the granulometric fractions during 32 days.

2.2.6. Chemical Composition and Physico-chemical Parameters

The cationic and anionic macroconstituents were determined on solid sample and in water obtained from maceration and leaching of each of the samples. The techniques used were: Emission spectrometry with plasma source, ionic chromatography and specific ion electrode, turbidimetry, C-S analyzer and fluorimetry.

The Eh, pH and conductivity were determined from soil elutriation waters.

3. RESULTS AND DISCUSSION

The results of the activity concentration of $^{239}\text{Pu} + ^{240}\text{Pu}$ and ^{241}Am in the granulometric fractions corresponding to the soil of the selected parcels and their correlation with the mineralogic and chemical composition of those soils are discussed in the following paragraphs.

3.1. Pu and Am concentration

Table I shows the concentration and the activity ratios of $^{239}\text{Pu} + ^{240}\text{Pu}$ and ^{241}Am in each of the parcels, obtained as the mean value of all the measures carried out in the granulometric fractions. From these values we can deduce:

- The existence of a notable difference between the contamination levels of parcels 2 and the other lots.

- The Pu/Am ratio in samples from zone 2-2, with values >6 , and from zones 3-2 and 5-3B seem to indicate that a fractioning due to a higher

leaching of Am has occurred, given that the theoretical ratio calculated from the desintegration of ^{241}Pu content in 1986 was 3.05. In lots, 2-0 and 3-1, the experimental ratio is more or less similar to the theoretical one.

- The Pu/Am ratio is almost constant in the different size fractions, with a coefficient of correlation above 0.98 except in the 5-3B sample where no correlation is found. Nevertheless, Table II shows that in the very small sizes ($< 10 \mu\text{m}$) this ratio always increases.

Tables III and IV include the activity concentrations of Pu and Am for each of the size fractions. These values indicate that:

- Plutonium and americium concentration is maximal in the fractions from 63 to 250 μm , except in sample 5-3B where maximum concentration occurs in the portion smaller than 5 μm . A second maximum concentration in the 20 to 40 μm group was found in samples 3-1 and 3-2.

- Concentration of plutonium and americium in fractions larger than 250 μm is much lower than for all the remaining fractions and almost negligible in comparison.

- In fractions smaller than 250 μm , the coefficient of correlation between both actinides and the size of each fraction is greater than +0.75 except in sample 5-3B where it is -0.5.

Tables V and VI show the distribution (per cent) of the plutonium and americium content of the soil. From this it is deduced that:

- In the parcels of land contaminated by the accident, concentrations of Pu-239 + Pu-240 and Am-241 activity associated with particles smaller than 10 μm are never greater than 15% of existing activity concentration in the soil; in parcels 3-1 and 3-2 (heavy cultivation) it is greater than 10% and in parcels 2-0 and 2-2 it is less than 10%.

In the most heavily cultivated lands, the activity associated with

fractions smaller than 5 μm is 8.74% maximum, while the less cultivated lands show a maximum value of 2.74%.

- For both Pu-239 + Pu-240 and Am-241 the percentages of activity found in fractions from 40 to 250 μm are greater than 75% in parcels 2-0, 2-2, and 3-2. In parcel 3-1, activity in the same size group comes to only about 50%, since an important proportion, about 30%, is found in the particles between 20 and 40 μm in size.

It is in the range from 63 to 250 μm where the greatest proportion is found, accounting for over 50% of total activity in the soil.

- In parcel 5-3B, where contamination has been the consequence of dispersion of contaminants during the 15 years that have elapsed since the accident, 57% of Pu-239 + Pu-240 activity in the soil is found in fractions smaller than 5 μm , while the same group shows only 7% of Am-241 activity.

In fractions between 40 and 250 μm only 9.83% of plutonium and 16.27% of americium are found.

- The percentage of Pu-239 + Pu-240 and Am-241 activity found in fractions larger than 250 μm is insignificant compared with the other fractions.

- An effort was made to obtain an idea of the distribution of plutonium and americium in particles smaller than 5 μm : using sample 2-0 the concentration of plutonium was estimated on the basis of α activity in fractions obtained through sieve analysis by sedimentation, assuming the aforementioned proportional distribution. The results are shown on Table VII and it can be deduced that only 0.4% of all the plutonium contained in the sample from parcel 2-0 is contained in fractions smaller than 0.6 μm .

3.2. Mineralogical Composition

The samples that were studied are composed of silty sand. The propor-

tion of sands ($t > 63 \mu\text{m}$) predominates with about 46% against 38% of silt ($63 \mu\text{m} > t > 5 \mu\text{m}$) and 14% of "clays" ($5 \mu\text{m} > t$).

Tables VIII and IX show the granulometric distributions of the total sample and the $< 5 \mu\text{m}$ fraction respectively. Fig. 2A shows the accumulated granulometric distribution of the samples and Fig. 2B shows the accumulated distribution of plutonium. From these figures, the marked relation between size and plutonium content becomes very evident.

According to the petrographic composition, the samples are classified as lithograywacke-phyллоarenite (3).

Mean composition, estimated by optic microscope is: 40% quartz, 29% muscovite-illite, 2% chlorite, 22% carbonates (calcite and dolomite), 3% iron oxides and opaque minerals, and 2% vegetal remains. These components represent more than 95% of the total composition.

Other components, estimated at less than 1%, are: garnet, plagioclase, hornblende, pyroxene, epidote, zoisite, tourmaline, zircon, rutile and apatite.

X-ray diffraction study of the mineralogical composition of each of the granulometric fractions are shown in Tables X to XIV.

The general tendency of variations in content in the size fractions, for each of the mineral components, is the following:

- Chlorite and muscovite-illite: irregular distribution, maximum values between 20 and 250 μm .

- Quartz: its content tends to diminish with grain size.

- Calcite: distribution reflects both textural forms; sparite: decreases as sizes approach 63 μm ; micrite: increases from 63 μm down to the finest sizes.

- Dolomite: irregular distribution; most frequent maximums found in sizes over 40 μm

In the study of the association between minerals, it was deduced that the most significant values for the linear coefficient of correlation were +0.71 for chlorite-muscovite and -0.64 for quartz-dolomite.

The coefficients of correlation between mineral components and plutonium are listed in Table XV.

According to these coefficients, plutonium will be in some way associated with the muscovite-illite in samples 2-0, 3-1 and 3-2; with dolomite in samples 2-0 and 2-2; and with chlorite in sample 3-1. In sample 5-3B plutonium would be independent of the mineralogical composition, tending to an association with calcite.

Coefficients of correlation between americium and mineral components are analogous.

Fig. 3 present the concentration distributions of plutonium and americium and mineralogical proportions in the granulometric fractions of the sample corresponding to parcel 2-0. Here, variations in distribution and the existing correlation between the distribution of plutonium and americium and the distribution of muscovite-illite and dolomite can be seen in more detail.

The autoradiographic analyses yielded the findings seen in the photomicrographs presented in Figures 4 and 5. The results can be summarized as follows:

- There is an association between mineral components and plutonium and americium content of the soil.

- The main concentrations of actinides, estimated from the density of α traces, are found in some fragments of iron oxi-hydroxide; a lesser concentration is found in some carbonate fragments and weak concentrations

in micaceous aggregates. In all of the components indicated, there are fragments with high relative concentrations and others with weak or nil density of traces, but microscopic analysis did not reveal significant genetic differences among them.

- In the fine fractions ($< 63 \mu\text{m}$), high concentration impacts have been recorded without being able to identify the mineral support. The relation between the impact size and effective hydraulic size in the granulometric separation seems to make it necessary to discard the hypothesis of the existence of isolated plutonium oxide particles.

- Vegetal remains have been found with some actinide concentration.

3.3. Chemical Composition and Physical-Chemical Parameters

Tables XVI and XVII show the results of chemical analyses of the samples.

By chemical composition, the samples are classified within the graywacke-lithic group because of their SiO_2 and Al_2O_3 content, although K_2O and Na_2O contents are the inverse of the mean for this type of rock (3).

Then mean content of organic carbon, $0.27\% \pm 0.03$ can be considered to be within the limits of the graywacke sediment type and lower than the average values of soils that can support some kind of vegetation. Humic acids represent 47% of total organic carbon.

An analysis of maceration-elutriation waters is found in Table XVIII and values for Eh, pH and conductivity are included in Table XIX. In keeping with these results, the water from samples 3-1, 3-2, and 5-3B belongs to the chloro-sulphate group, that of 2-0 to the bicarbonate group, and that of 2-2 to the chloride group, according to their positions on the saline anionic classification diagram (4). From the point of view of cationic content, the water of samples 2-0 and 2-2 is sodic-potassic; that of 3-2 and 5-3B is calcic; and in 3-1 the relative proportions of Na+K, Ca and Mg cations are practically the same.

The total salinity and ionic content of sample 2-0 are significantly different than those of samples 3-1, 3-2, and 5-3B. This difference is probably due to the brackish character of the waters used for irrigation of the cultivated parcels. In parcel 2-2 the effects of cultivation and irrigation are less pronounced.

The Eh and pH values of the samples indicate an oxidizing and slightly alkaline environment. Eh varies from 312 to 344 mV and pH from 7.6 to 8.43. Figure 6 shows the region in which the samples are situated on the Eh-pH diagram in regard to plutonium ion species in solution, in equilibrium with $\text{PuO}_2(\text{s})$ at 25°C and 1 atmosphere (5).

According to the Eh-pH diagram it would seem that we are in the theoretical region of stability of PuO_2 , although the equilibrium concentration with PuO_2 shown in the above-mentioned concentration values is higher than the theoretical value. This seems to indicate the presence of other chemical species.

The content of plutonium in waters and the percentage leached during maceration and elutriation are shown in Table XX. The values for plutonium leaching are a function of the Eh and pH potentials. Greater leaching is achieved in the samples that are most oxidized and lower in pH.

4. CONCLUSIONS

The following are the most significant deductions drawn from the material presented above:

1st. Plutonium and americium content and soil granulometry are directly associated (coefficient of correlation = 0.75). The maximum concentrations correspond to fractions ranging between 63 and 250 μm . Less than 15% of residual plutonium and americium contamination is associated with the fractions of particles smaller than 10 μm . The maximum Pu-239 + Pu-240 found in the portion smaller than 5 μm corresponds to 9% in heavily

cultivated soil and 3% in less cultivated soil. The maximum percentages for Am-241 in the same group smaller than 5 μ m are analogous in less cultivated soil and somewhat lower in the heavily cultivated soil.

In parcel 5-3B, which was contaminated well after the accident, Pu-239 + Pu-240 activity in the fraction smaller than 5 μ m was 57%.

2nd. The contaminants, plutonium and americium, have been found in some authigenetic iron oxides, carbonates (dolomite) and muscovite-illite. The maximum concentrations were found in the iron oxihydroxides.

3rd. The plutonium-americium ratio calculated for the different size fractions has a mean value of 3.0 ± 0.2 in parcels 2.0 and 3.1, while for parcel 2-2 it is 6.7 ± 0.5 and for parcel 3-2 it is 7.4 ± 0.6 . This would seem to indicate greater mobilization of americium in the latter three parcels which could probably be related to the Eh and pH potentials of the waters in "equilibrium" with the soil, which in parcels 2-2 and 3-2 create a slightly more reducing and alkaline environment.

4th. The potential leaching of plutonium estimated from the concentrations measured in the maceration-elutriation waters is found in the range of 2×10^{-4} to 21×10^{-4} per cent of the total content in the soil, with the greater values corresponding to the most oxidized and least alkaline soils.

5th. It is necessary to continue research aimed mainly at defining the mechanisms of plutonium and americium fixing and dispersion since even though the dispersion factor of clays in these soils is very low (0.23), mobilization of plutonium and americium colloids and complexes remains unknown.

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TABLE I. MEAN CONCENTRATION OF Pu AND Am IN THE SAMPLES

Radionuclide	Activity Concentration (Bq/g)				
	Parcels				
	2-0	2-2	3-1	3-2	5-3B
^{239}Pu ^{240}Pu	42.60	15.60	1.09	1.82	0.46
^{241}Am	13.86	2.54	0.34	0.32	0.02
Ratio Pu/Am	3.0 ± 0.2	6.7 ± 0.5	3.0 ± 0.2	7.4 ± 0.6	41.9 ± 27.8

TABLE II. ACTIVITY CONCENTRATION RATIO OF $^{239}\text{Pu} + ^{240}\text{Pu} / ^{241}\text{Am}$ IN SIZE FRACTIONS OF SOILS

Parcel \ Fraction μm	Fraction μm						
	< 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	2.99	3.90	3.22	2.21	2.93	3.21	2.80
2-2	9.21	5.83	5.08	6.14	6.09	6.10	8.33
3-1	4.08	3.28	2.05	3.39	2.29	3.18	2.85
3-2	8.60	9.33	7.38	6.08	6.50	5.22	8.89
5-3B	222	15.67	8.71	9.50	16.67	12.86	7.78

TABLE III. ACTIVITY CONCENTRATION OF $^{239}\text{Pu} + ^{240}\text{Pu}$ IN SIZE FRACTIONS OF SOILS (Bq/g)

Parcel \ Fraction μm	Fraction μm						
	< 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	8.55	24.57	19.83	15.48	87.28	97.13	0.98
2-2	3.13	3.09	4.68	9.33	29.16	49.88	0.05
3-1	0.53	0.59	0.43	2.24	0.80	2.54	0.02
3-2	0.86	1.12	0.59	1.46	0.39	5.48	0.08
5-3B	2.22	0.47	0.61	0.57	0.15	0.09	0.07

TABLE IV. ACTIVITY CONCENTRATION OF ^{241}Am IN SIZE FRACTIONS OF SOILS, (Bq/g)

Parcel	Fraction μm						
	< 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	2.86	6.30	6.15	7.00	29.77	30.26	0.35
2-2	0.34	0.53	0.92	1.52	4.79	8.18	0.00
3-1	0.13	0.18	0.21	0.66	0.35	0.80	0.007
3-2	0.10	0.12	0.08	0.21	0.06	1.05	0.009
5-3B	0.01	0.03	0.07	0.06	0.009	0.007	0.009

TABLE V. PERCENTAGE DISTRIBUTION OF $^{239}\text{Pu} + ^{240}\text{Pu}$ IN SIZE FRACTIONS OF SOILS

Parcel	Fraction μm						
	< 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	2.74	6.07	3.32	7.25	21.25	59.30	0.07
2-2	2.57	1.53	1.32	7.04	18.18	59.33	0.03
3-1	8.74	6.12	2.12	31.79	3.68	47.22	0.33
3-2	7.02	4.55	5.31	7.08	0.71	74.32	0.51
5-3B	56.98	6.05	5.62	20.30	2.88	6.95	1.22

TABLE VI. PER CENT DISTRIBUTION OF ^{241}Am IN SIZE FRACTIONS OF SOILS

Parcel	Fraction μm						
	< 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	2.82	4.79	3.17	10.07	22.28	56.80	0.07
2-2	1.68	1.61	1.59	7.03	18.32	69.75	0.02
3-1	6.82	5.97	3.34	30.17	5.25	47.94	0.51
3-2	4.55	2.78	3.95	6.46	0.60	81.34	0.32
5-3B	7.02	8.26	14.38	50.46	3.86	12.41	3.61

TABLE VII. DISTRIBUTION OF THE TOTAL α ACTIVITY IN FRACTIONS $< 5 \mu\text{m}$ OF SAMPLE 2-0

μm	Percentage	Activity Concentration (Bq/g)
5.00 - 2.10	6.70	10.59
2.10 - 1.80	0.76	2.84
1.80 - 0.87	1.27	18.17
0.87 - 0.79	0.28	16.07
0.79 - 0.60	0.54	4.13
0.60	4.13	4.00

TABLE VIII. PER CENT GRANULOMETRIC DISTRIBUTION OF THE SOILS

Parcel	Fraction μm							
	<5	5-10	10-20	20-40	40-63	63-250	250-1000	>1000
2-0	13.68	10.53	7.14	19.95	10.37	26.01	2.99	9.33
2-2	12.76	7.72	4.40	11.75	9.71	21.63	9.26	22.77
3-1	17.94	11.36	5.32	15.41	5.02	20.15	20.02	4.78
3-2	14.91	7.39	16.46	8.82	3.33	24.80	11.86	12.43
5-3B	11.80	5.86	4.20	16.23	3.81	35.85	8.65	8.90

TABLE IX. PER CENT GRANULOMETRIC DISTRIBUTION OF THE $< 5 \mu\text{m}$ FRACTION OF SOILS

Parcel	Fraction μm							
	1.0-1.3	1.3-1.6	1.6-2.0	2.0-2.6	2.6-3.2	3.2-4.1	4.1-5.1	5.1-6.5
2-0	7.57	9.34	10.87	13.11	15.65	15.53	15.20	12.73
2-2	13.91	14.01	17.55	14.13	10.93	10.04	10.16	9.27
3-1	28.81	17.52	7.02	7.66	9.53	10.07	10.62	8.77
3-2	26.95	10.42	7.82	8.50	10.98	11.66	12.59	11.08
5-3B	33.85	19.07	6.92	6.55	8.22	8.11	8.70	8.58

TABLE X. MINERALOGICAL COMPOSITION OF SIZE FRACTIONS

		Parcel 2-0				
I Size μm	Chlorite	Moscovite- Illite	Quartz	Calcite	Dolomite	
	$d(\text{\AA})-14$	$d(\text{\AA})-10$	$d(\text{\AA})-4.24$	$d(\text{\AA})-3.03$	$d(\text{\AA})-2.89$	
	100.00	50.70	49.89	69.35	20.48	
1000	41.30	33.80	100.00	73.99	19.58	
250	80.43	93.43	84.92	49.54	54.81	
63	62.22	100.00	58.09	39.00	100.00	
40	84.78	97.18	55.43	65.33	25.90	
20	58.70	59.62	21.51	61.30	7.53	
10	58.70	72.24	11.97	76.78	10.54	
5	67.39	46.48	6.65	100.00	5.42	

TABLE XI. MINERALOGICAL DISTRIBUTION OF SIZE FRACTIONS

		Parcel 2-2				
I Size μm	Chlorite	Moscovite- Illite	Quartz	Calcite	Dolomite	
	$d(\text{\AA})-14$	$d(\text{\AA})-10$	$d(\text{\AA})-4.24$	$d(\text{\AA})-3.03$	$d(\text{\AA})-2.83$	
	23.73	24.40	71.52	53.63	86.36	
1000	33.90	32.37	100.00	38.64	50.91	
250	44.07	35.75	97.43	14.52	100.00	
63	38.98	92.75	57.60	43.56	51.14	
40	100.00	100.00	59.10	63.70	68.18	
20	61.02	93.72	20.77	61.59	57.95	
10	27.12	33.09	13.06	79.16	39.77	
5	37.29	31.64	5.78	100.00	--	

I = reflexion intensity in arbitrary units, taking 100 as the maximum value of I of the mineral in the X-Ray diffraction diagrams.

TABLE XII. MINERALOGICAL DISTRIBUTION OF SIZE FRACTIONS

		Parcel 3-1				
I Size μm	Chlorite	Moscovite- Illite	Quartz	Calcite	Dolomite	
	$d(\text{\AA})-14$	$d(\text{\AA})-10$	$d(\text{\AA})-4.24$	$d(\text{\AA})-3.03$	$d(\text{\AA})-2.88$	
1000	3.89	22.55	85.96	58.37	55.23	
250	5.63	29.30	100.00	53.47	45.35	
63	100.00	100.00	67.98	66.12	41.86	
40	6.36	55.32	64.45	58.18	100.00	
20	5.65	90.21	35.09	58.78	29.65	
10	9.89	78.30	18.20	71.43	19.19	
5	13.78	85.74	11.84	95.51	27.33	
	14.19	64.26	8.11	100.00	11.63	

TABLE XIII. MINERALOGICAL COMPOSITION OF SIZE FRACTIONS

		Parcel 3-2				
I Size μm	Chlorite	Moscovite- Illite	Quartz	Calcite	Dolomite	
	$d(\text{\AA})-14$	$d(\text{\AA})-10$	$d(\text{\AA})-4.24$	$d(\text{\AA})-3.03$	$d(\text{\AA})-2.88$	
1000	27.59	58.26	99.77	62.00	16.15	
250	31.03	39.99	79.25	57.20	85.84	
63	81.03	99.08	75.76	82.00	50.66	
40	24.14	42.20	100.00	56.80	100.00	
20	93.10	100.00	56.88	75.20	40.27	
10	34.48	66.51	30.30	53.60	10.62	
5	100.00	91.74	14.92	85.20	7.96	
	58.62	64.68	8.39	100.00	4.42	

TABLE XIV. MINERALOGICAL COMPOSITION OF SIZE FRACTIONS

Parcel 5-3B					
I Size μm	Chlorite	Moscovite- Illite	Quartz	Calcite	Dolomite
	$d(\text{\AA})-14$	$d(\text{\AA})-10$	$d(\text{\AA})-4.24$	$d(\text{\AA})-3.03$	$d(\text{\AA})-2.88$
1000	53.49	40.51	62.27	100.00	30.00
250	51.16	58.56	89.64	65.24	92.50
63	88.37	83.33	97.16	35.24	46.67
40	27.91	46.30	100.00	57.14	90.00
20	100.00	99.07	77.29	47.38	71.67
10	90.70	100.00	26.84	55.24	26.67
5	79.07	81.48	15.03	62.38	21.67
	69.77	74.07	21.74	71.90	10.00

TABLE XV. LINEAR COEFFICIENT OF CORRELATION BETWEEN THE ACTIVITY CONCENTRATION OF $\text{Pu}^{239+240}$ AND THE MINERALS

Parcel	Correlated Parameters				
	Activity Concentration of $^{239}\text{Pu} + ^{240}\text{Pu}$ Versus Mineral				
	Chlorite	Moscovite-Illite	Quartz	Calcite	Dolomite
2-0	0.39	0.73	0.33	-0.78	0.84
2-2	-0.04	0.03	0.51	-0.72	0.78
3-1	0.66	0.75	0.06	-0.25	0.02
3-2	0.50	0.66	0.02	0.37	-0.06
5-3B	0.15	0.16	-0.64	0.55	0.31

TABLE XVI. CHEMICAL COMPOSITION PER CENT OF MACROCONSTITUENTS IN
SAMPLES OF SOILS

Oxides \ Parcels	TOTAL SAMPLE				
	2-0	2-2	3-1	3-2	5-3B
*SiO ₂	53.00	58.00	53.00	55.00	56.00
Al ₂ O ₃	12.10	8.20	13.30	12.20	9.00
CaO	10.60	11.80	9.40	9.00	10.80
MgO	2.10	1.60	2.20	2.20	2.00
MnO	0.05	0.06	0.09	0.07	0.06
Fe ₂ O ₃	4.70	3.60	5.50	5.10	4.30
Na ₂ O	0.86	0.53	0.96	0.85	0.63
K ₂ O	2.60	1.50	2.70	2.60	1.90
TiO ₂	0.60	0.47	0.56	0.55	0.51
P ₂ O ₅	0.16	0.09	0.19	0.19	0.12
**P.C.	13.00	14.00	12.00	12.00	15.00

*). SiO₂ calculated by difference to 100.

**). P.C.: Calcination losses at 900°C during 2 hrs.

TABLE XVII. ADDITIONAL SIGNIFICANT COMPONENTS FROM SAMPLES OF SOILS
TOTAL SAMPLE

Components \ Parcel	TOTAL SAMPLE				
	2-0	2-2	3-1	3-2	5-3B
% Total C	3.030	3.730	2.740	2.680	3.560
% Org. C	0.389	0.321	0.226	0.192	0.210
% Total S	0.011	0.022	0.279	0.170	0.247
U ₃ O ₈ ppm	4.000	4.000	3.000	4.000	4.000
Humic acids ppm	1710	1590	1166	917	1545

TABLE XVIII. ACTIVITY ($\text{mol.l}^{-1} \cdot 10^{-3}$) AND IONIC STRENGTH CALCULATED IN MACERATION-ELUTRIATION WATERS FROM SAMPLES OF SOILS

Ions	Parcels				
	2-0	2-2	3-1	3-2	5-3B
HCO_3^-	2.54	4.56	1.43	1.53	1.04
Cl^-	0.48	10.71	15.67	9.49	30.24
NO_3^-	0.07	0.06	3.54	1.01	2.68
NO_2^-	0.03	0.01	0.41	0.34	0.12
PO_4^{3-}	-	0.01	-	-	-
SO_4^{2-}	0.59	0.72	6.74	2.47	8.77
Na^+	2.25	10.94	10.67	7.44	10.44
K^+	1.71	1.61	1.93	1.09	2.03
Ca^{++}	0.64	0.50	3.07	5.49	4.94
Mg^{2+}	0.25	0.79	3.88	2.30	2.04
Ionic Strength	8.37	24.18	89.99	57.16	118.81

TABLE XIX. PHYSICAL-CHEMICAL PARAMETERS OF THE SYSTEM WATER/SOIL FROM THE SAMPLES OF SOIL

Parcels	Parameters		
	Eh (mV)	pH	Conductivity μS
2-0	340.4	7.70	484
2-2	312.0	8.20	1247
3-1	344.4	7.60	2500
3-2	328.4	8.43	2330
5-3B	321.4	8.36	2410

TABLE XX. PLUTONIUM CONCENTRATION IN MACERATION-ELUTRIATION WATERS

<u>Parcels</u>	<u>PuO₂, (Mol.l⁻¹ · 10⁻¹⁵)</u>	<u>% leached</u>
2-0	250	12 x 10 ⁻⁴
2-2	10	2 x 10 ⁻⁴
3-1	10	21 x 10 ⁻⁴
3-2	5	5 x 10 ⁻⁴
5-3B	1	5 x 10 ⁻⁴

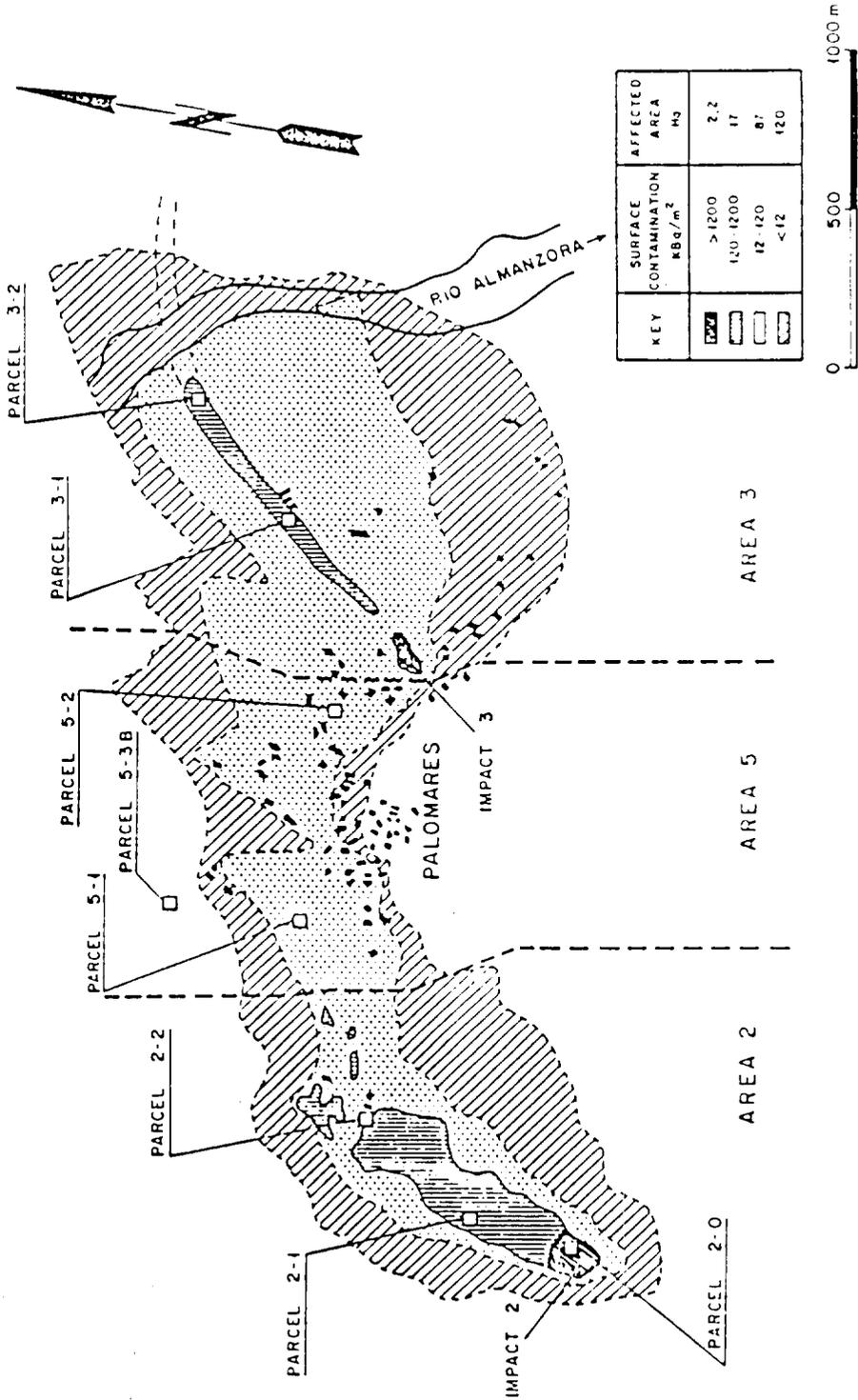
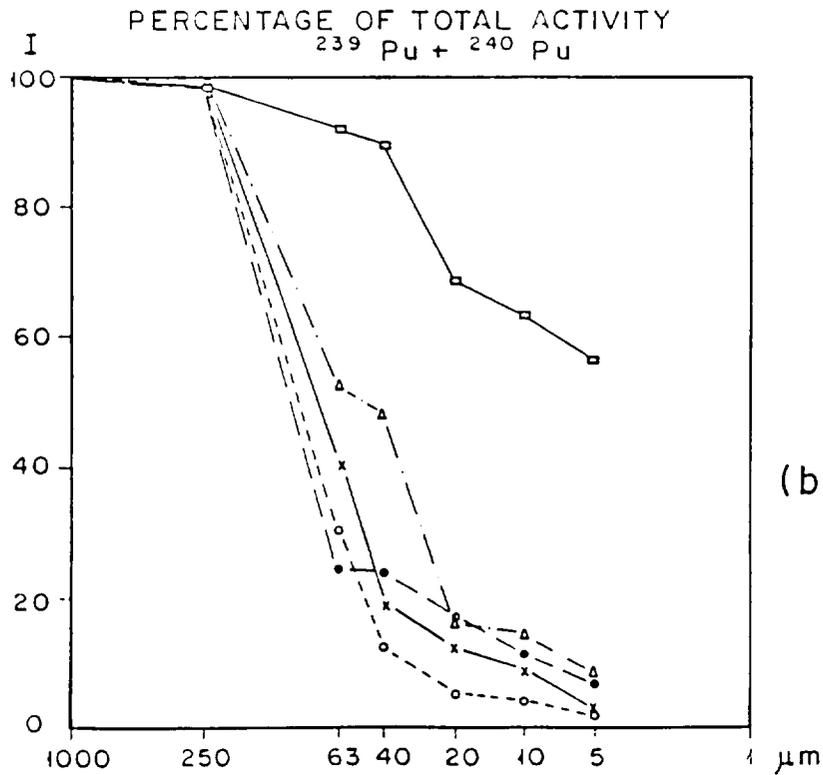
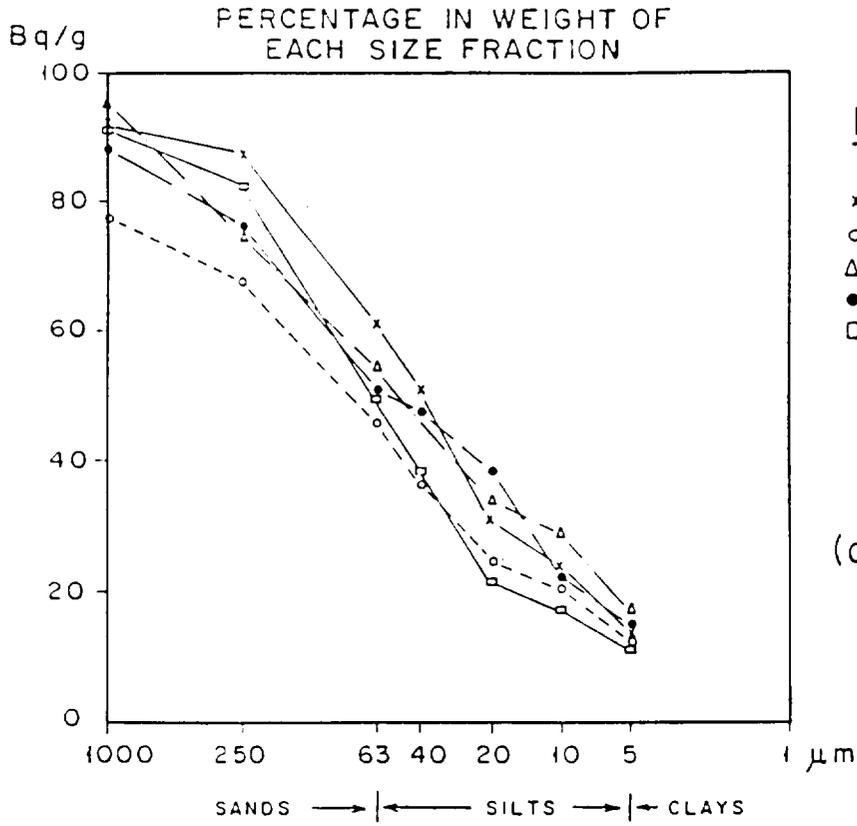


FIG. 1 - PALOMARES REGION
 LEVELS OF ORIGINAL CONTAMINATION AND LOCATION OF SAMPLE PARCELS



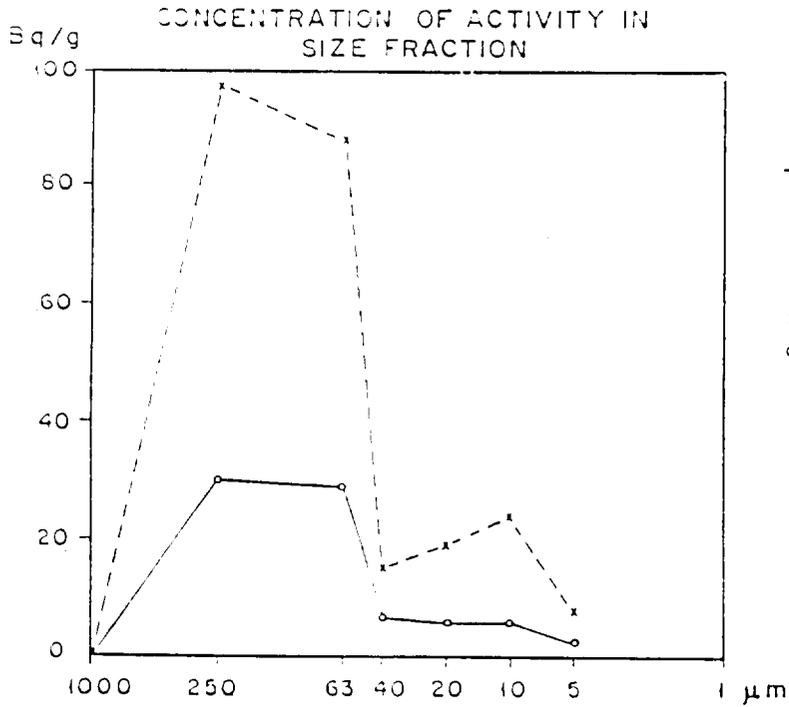
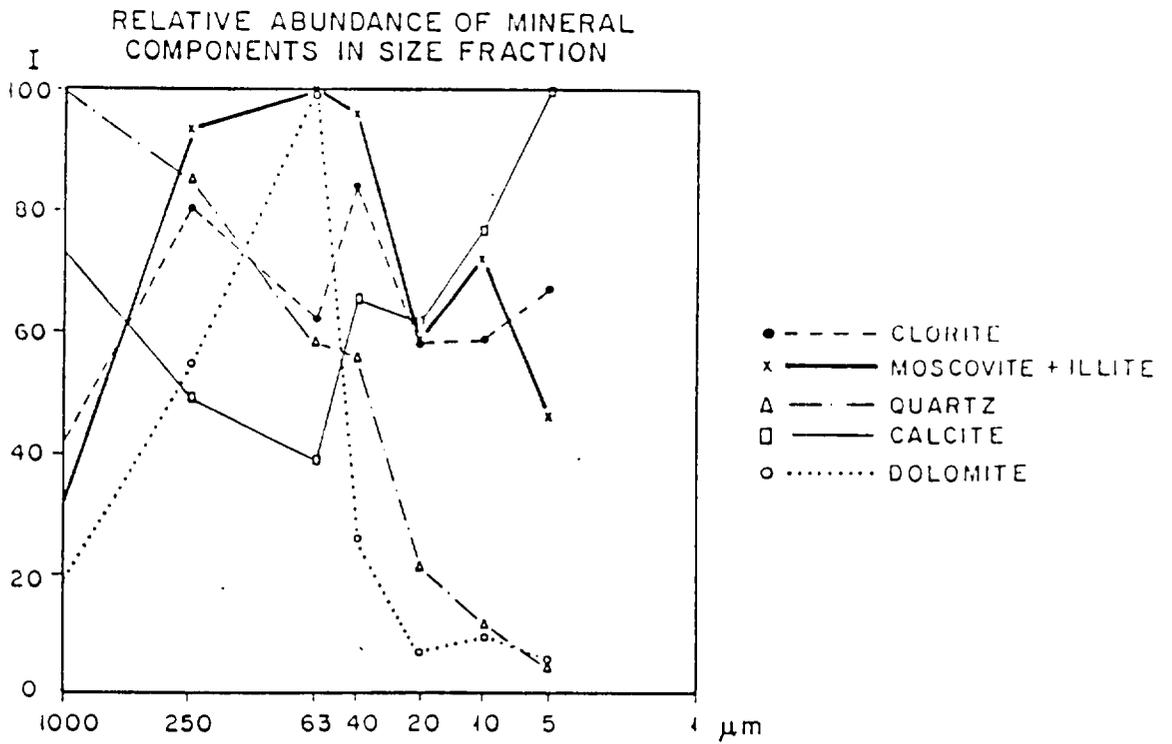


FIG. - 3
PARCEL 2 - 0



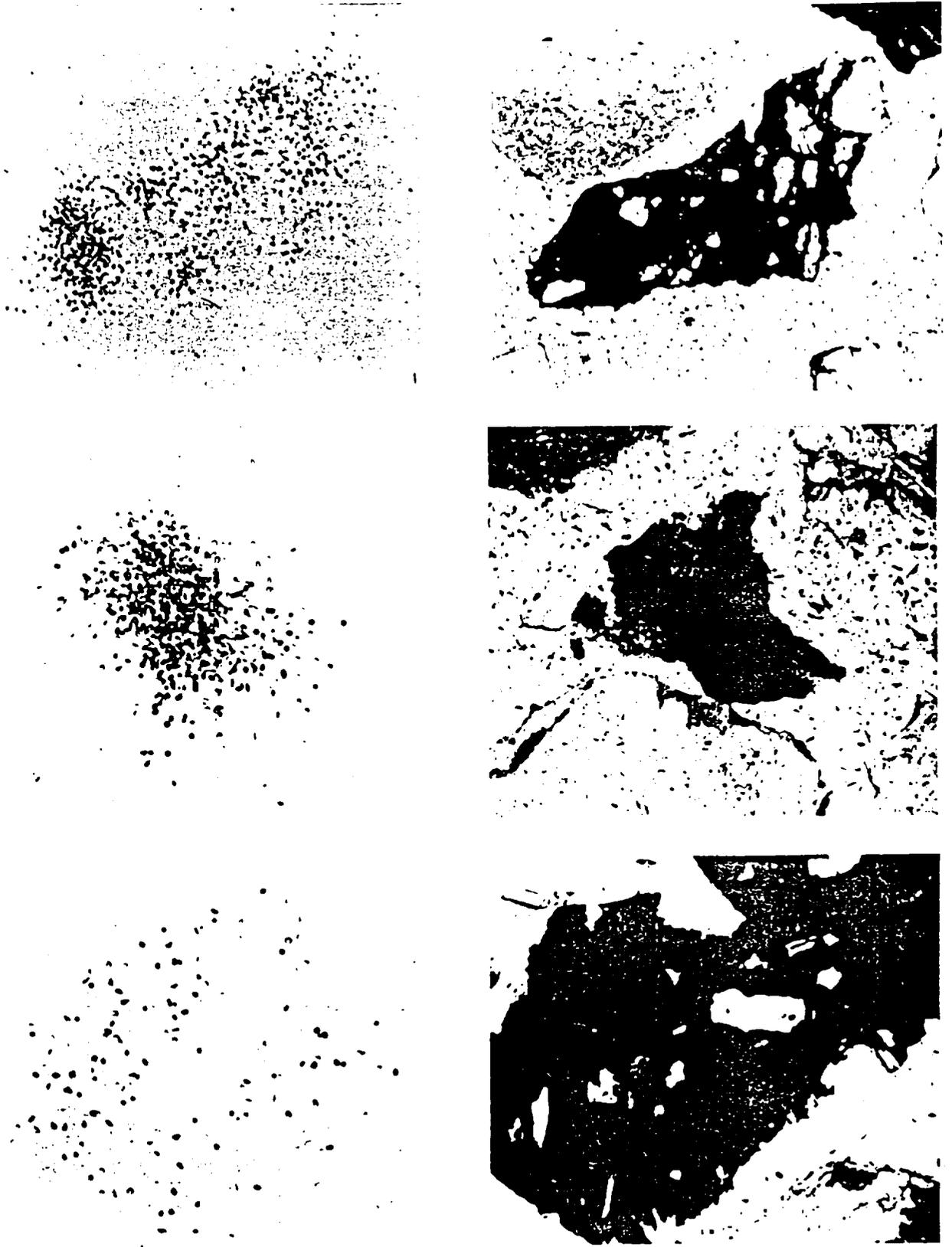


Fig.4. ∞ Autoradiography of iron oxy-hydroxides. L.N. x 75 and 150

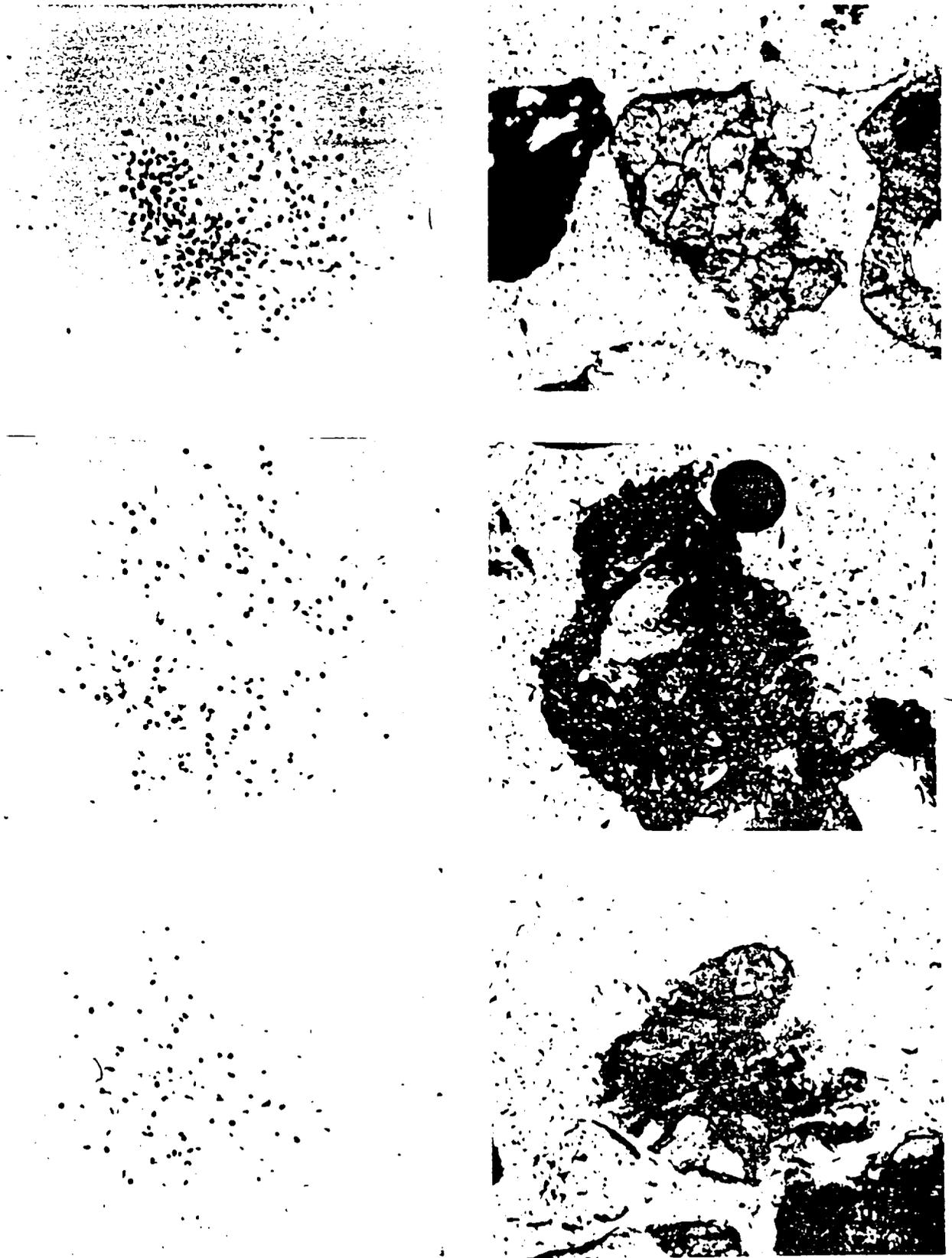
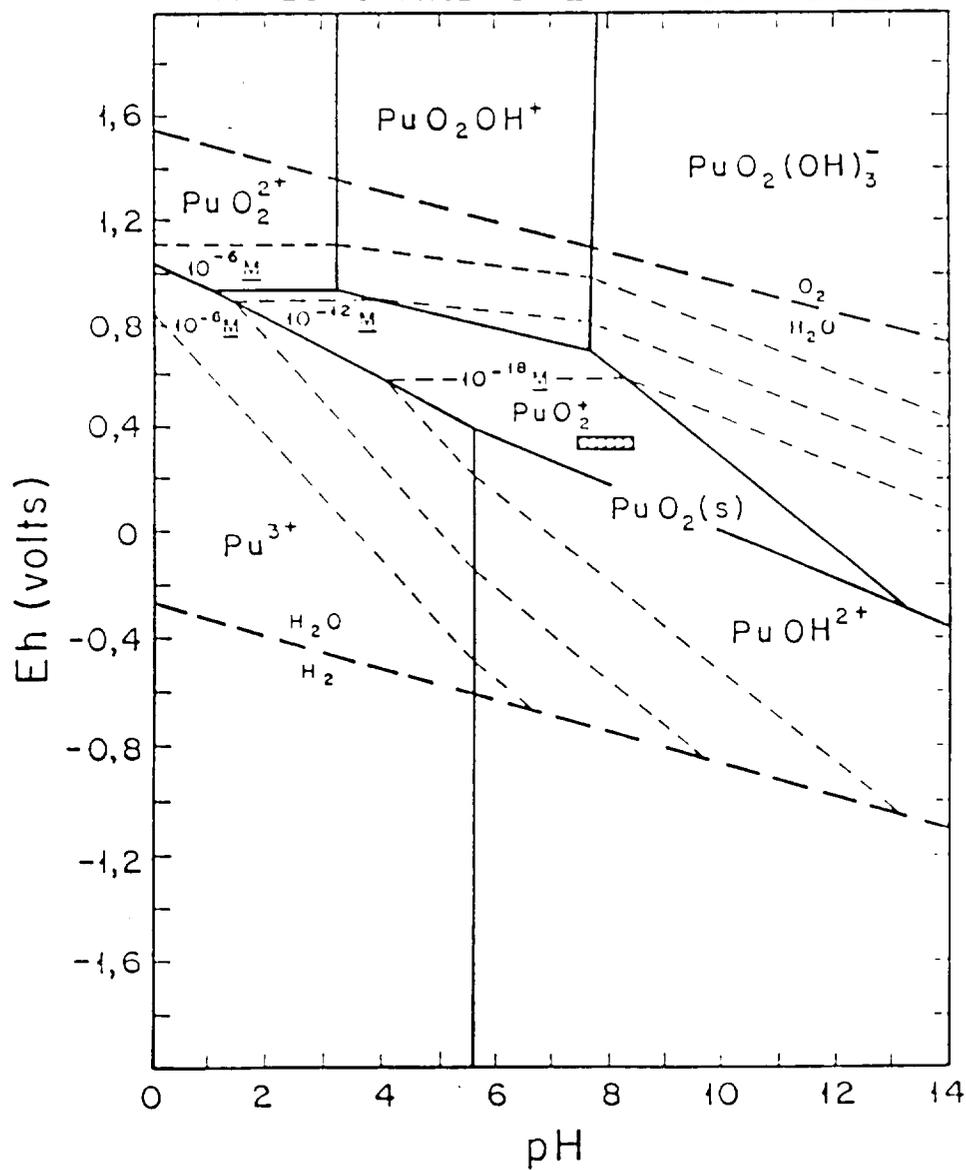


Fig.5. ∞ Autoradiography of carbonates and micaceous aggregates. L.N.x150

Eh-pH DIAGRAM FOR PLUTONIUM AT 25°C AND ONE ATMOSPHERE



 POSITION OF PALOMARES SOILS

FIG.- 6