# INHALATION RISKS TO PEOPLE LIVING NEAR A CONTAMINATED AREA.-

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### 1. INTRODUCTION

As the result of an air accident, the nuclear fuel of two thermonuclear bombs was burned out on 17th January 1966. This resulted in the production of an aerosol which contaminated in varying degress a total area of about 226 hectares. Contaminated vegetation and the surface layer of soil (about 5 cms. thick) in areas with alpha-emitting radionuclide contamination levels above 3.2 x 10<sup>-2</sup> microCi/100 cm<sup>2</sup> were taken up and eliminated as radioactive wastes. The rest of the area was plowed over down to a depth of about 25 cms. so as to eliminate the radionuclides contaminating the surface soil and dilute the concentration of - radioactive elements to what was considered to be a safe level, at which there would be no risks of inhalation capable of causing relatively serious internal contamination to the people living in - the area in subsequent years.

In order to establish practically the correctness of the -theoretical reasoning behind the preceding actions, and to observe
the effects of weather conditions and farming on the contaminating
elements in the soil, and therefore on the amount of environmental
radioactivity in the area, a research program was drawn up, designed to measure the environmental alpha activity in the area
for a time long enough to enable researchers to determins how great the risk would be to people living there during all or the -greater part of their lives, including infancy and childhood, under
any circumstances whatsoever.

The weather conditions in the area (mean annual rainfall - of 200 l/m<sup>2</sup>; mean annual temperature of 18.20C; wind velocities - reaching 70 Km/hr fairly frequently) are such as to make it very-likely that the atmosphere will pick up a sizable load of particles from the soil.

## 2. SAMPLING STATIONS

The best places for setting up sampling stations had to be - determined on the basis of the size of the contaminated area, its - topographical features, the sparse population distribution (the area is rural and not a compact urban center), and the location of the most highly contaminated areas remaining after the elimination of the ones with the highest original contamination as described above. The area was thus divided up into three zones, circumscribing each of the two points where the thermonuclear bombs hit the ground, and the town containing 90% of the area's population. Four sampling stations were then set up (see fig. 1 for locations): two in zona 2, which is the largest one, one in the town, and one in zone 3.

- a) Station 2-1 This station is located in the hills near point of impact no 2. The soil in this zone is rocky and sparsely covered with wild scrub. There are some parts where plowing and adequate dilution of the radioactive particle concentration proved impossible. The station stands in the center of an area where contamination levels were between 3.2 x 10<sup>-1</sup> and 3.2 x 10<sup>-2</sup> microCi/100 cm<sup>2</sup>.
- b) Station 2-2 This station is located on farmland in zone 2, near a small group of inhabited houses.

As a result of the topographical features of this land, the aerosol stream contaminated it at levels lying between 3.2 x 10<sup>-2</sup> and -3.2 x 10<sup>-3</sup> microCi/100 cm<sup>2</sup>; however, it is surrounded by other farmland which suffered even higher levels of contamination.

- situated so as to be able to take a representative sampling of the air being breathed by the inhabitants, thereof, since it openly faces the directions of the prevailing winds in the area, which blow from the various contaminated ed sectors. The contamination of the area near this station was invariably lower than -3.2 x 10<sup>-2</sup> microCi/100 cm<sup>2</sup>.
- d) Station 3-1. This station is located in the center of the -strip with the highest remaining contamination
  in the area surrounding the point of impact of thermonuclear bomb no. 3. Since this entire zone is a plain lying about 4 meters below the level at which impact took place, it seemed unnecessary to set up more than
  a single sampling station there. The surrounding area was contaminated at levels
  lying between 3.2 x 10<sup>-1</sup> and 3.2 x 10<sup>-3</sup> microCi/100 cm<sup>2</sup>.

As figure 2 shows, each of these sampling stations is comprised of a filter support, a 1425 r.p.m., 1/3 HP air pump, a gas meter to measure the volume of air sampled, and a voltage regulator. These devices are installed inside a wooden shed which protects them from the elements.

Cellulose filters with a diameter of 47 mm. and a pore size of 1.2 microns are used for sampling. Sampling goes on 24 hours a day, every day, throughout the year. The daily samples are placed one by one on stainless steel plates of equal size, which are covered with mylar paper with a thickness of 12.7 microns and sent to Madrid for analysis and subsequent chemical treatment at the laboratories of the Spanish JEN's Division of Medicine and Protection.

Samples are taken at a height of 1.70 m. above the ground.

## 3. ANALYTICAL TECHNIQUES

The daily aerosol samples from each station that arrive at the laboratories of the Division of Medicine and Protection are there — prepared for measurement of their gross alpha activity due to long lived radioactive elements, and for quantitative analysis of their — plutonium—239, uranium—235, and uranium—234 contents, by means of the techniques described in summary fashion below.

# 3.1. Measurement of gross alpha activity

Alpha activity is measured directly from each filter - received by means of an "Eberline" methane-flow proportional counter with a rate of methano flow of 80 ml/min. Counting - efficiency with this device is 35.65%.

## 3.2. Plutonium-239 measurement

Plutonium-239 content is measured by a combined —technique involving ion exchange separation, electrodeposition, and alpha spectrometry. For reasons of the complexity of —the plutonium-239 and uranium-235 quantitative analysis — procedure, compound samples prepared from the individual —samples from each of the sampling stations for every ten-day period in a month are used for these analysis. This means

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that each ultimate alpha spectrometry reading obtained - corresponds to a volume of air on the order of 100 m<sup>3</sup>, as explained below.

The membrane-type filters carrying the aerosol samples from each station for every ten-day period undergo wet - digestion by concentrated nitric acid in a glass tube at - temperatures of 250-300°C over an aluminium block, until the residue takes on a white color. Before digestion, 1 ml of a plutonium-236 solution containing 3 d.p.m. of this radionuclide is added to each tube so that the porcentage of recovery in - each analysis can be calculated subsequently.

The residue is dissolved in 50 ml of HNO<sub>3</sub> (8 M), and the solution is passed through an ion-exchange anionic resin, Dowex AG 1X-2 (50-80 meshes) in chloride form, at a speed of flow of less than 1 ml/min. Under the analysis -- conditions, plutonium and thorium are retained by the resin.

To eliminate whatever traces of uranium may have been picked up, the resin column is washed with several batches of 8 M HNO<sub>3</sub> to a total volume of 150 ml. The effluent from these washing operations is collected for subsequent uranium analysis.

To eliminate the thorium retained in the resin along with plutonium, the column is washed once again with concentrated HCl, and the effluent from this washing operation is discarded.

The plutonium fixed in the resin is eluted with three 5 ml batches of 0,5 N HCl.

The effluent from this operation is readied for plutonium - electrodeposition through evaporation to dryness at a - temperature no lower than 75°C, redisolution of the residue in

concentrated HCl and a new desiccation.

A normal HCl solution of this residue, with ammonium oxalate 4% added, is submitted to electrolysis in the apparatus shown in figure 3. Plutonium is electrodeposited on stainless steel plates with a diameter of 14 mm., at a voltage of 20 V. and a current of 200 mA., for three hours.

The samples obtained by this procedure are submitted to alpha spectrometry using "Ortec" silicon-barrier detectors with a surface area of 300 mm<sup>2</sup>, and a "Packard", 400-channel pulse-height analyzer. Figure 4 shows the kind of alpha energy spectrum obtained.

The counting efficiency of this equipment is 31%.

# 3.3. Uranium-235 and uranium-234 analysis

The 8 M nitric acid solution used to wash the resin - column fixing plutonium and thorium is employed for uranium-235 and uranium-234 analysis.

This elution is evaporated to dryness and redissolved - in 10 N HCl in order to make it acid enough to fix uranium when passed through a cationic resin, Dowex AG-1 X-10 (100-200 meshes), in chloride form. After the resin column has been washed with 10 N HCl, all the uranium previously fixed therein is eluted with 0.5 N HCl.

The uranium in a normal HCl solution of the elution - obtained from the resin column is electrodeposited under - conditions similar to the ones described above with reference to plutonium.

Quantitative analysis of uranium-235 and uranium-234 by alpha spectrometry is done with the use of silicon-barrier -- detectors with a surface area of 300 mm<sup>2</sup>.

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The total recovery with this procedure is on the order of 75%.

The alpha energy spectra for the electrodeposited samples invariably reveal the presence of uranium-235 and uranium-234 (4.72 and 4.77 MeV). Since both isotopes are significant from the standpoint of the risk involved, the results of these analysis are expressed as the sum of the values -- for both radionuclides.

Nevertheless, more precise spectrometric analyses have also been conducted to allow determination of the porcentage - of the total concentration accounted for by each isotope. -- Better resolution was obtained with a silicon-barrier detector with a surface area of 7 mm<sup>2</sup>, and as a result, the alpha -- energy spectrum could be obtained from a soil sample from the most heavily contaminated area. Analysis of this - spectrum has made it possible to calculate that the ratio of uranium-234 to uranium-235 activity is 43.16, and that the ratio of uranium-235 to uranium-234 atoms is thus 98.68.

## 4. RESULTS

#### 4.1. Concentration of total alpha activity

Table I contains the monthly mean values for total alpha activity at each of the sampling stations in the Palomares
area.

It also contains the maximum and minimum values for - alpha activity during each of the months listed. These values are for the second half of 1966. Table II contains the - values for 1967.

Comparing the values contained in the above-mentioned

tables with the maximum permissible concentration of any indiscriminated mixture of alpha-beta and gamma-emitting - radionuclides for the general public, 2 x 10<sup>-14</sup> microCi/cm<sup>3</sup> (ENEA, Radiation Protection Norms, Revised edition 1968), one finds that:

- The mean monthly concentrations of environmental gross alpha activity in the Palomares area remained below the maximum permissible concentrations during the entire period mentioned. In August of 1966, the mean values approached the M.P.C. levels at the stations in the town (1.7 x 10<sup>-14</sup> microCi/cm<sup>3</sup>) and in zone 2-2 (1.4 x 10<sup>-14</sup> microCi/cm<sup>3</sup>).
- The maximum values for gross alpha activity exceeded the M.P.C. en 14 occasions in all, during the said time. Four of these occasions were recorded at station 2-1, seven at station 2-2, one at station P, and two at station 3. The M.P.C., was exceeded by a factor in excess of 10 on only 3 days.
- The maximum level was recorded at station 2-2 on 4th April 1967. The wind on that day reached a maximum velocity of 17 km/hr in North-South and East-West directions.
- The days on which the maximum values for total alpha activity were recorded, were characterized by wind velocities exceeding 17 km/hr and ranging as high as 64 km/hr.

#### 4.2. Plutonium-239 concentration

The values for plutonium-239 concentracion in the aerosol samples taken in the Palomares area are shown in

tables ITI and IV. Table III is for the second half of 1966, and table IV is for 1967. The values listed normally represent the mean concentrations for ten-day periods. The arrows marked in the tables indicate that, for any of a variety of reasons, a small number of samples was collected during the period in question, and that the samples taken were therefore included with the ones for the period immediately proceding or following, for the purposes of analysis.

According to the "Radiation Protection Norms" of the European Nuclear Energy Agency (revised edition, 1968), the maximum permissible concentrations of insoluble,—compounds of plutonium—239 in the air for the general public is  $10^{-12}$  microCi/cm<sup>3</sup>, and the M.P.C. of soluble plutonium—239 compounds in the air is  $6 \times 10^{-14}$  microCi/cm<sup>3</sup>. The plutonium—239 contaminating the Palomares area is found as plutonium dioxide, which is a highly insoluble compound. To provide a guarantee of maximum safety, however, the—latter value may be taken as a basis for comparison with—the data obtained on plutonium—239 concentration.

The results for this comparison, based on tables III and IV, are:

- Particles of plutonium dioxide mixed with farmed soil are resuspended in the air, as the presence of this compound has been recorded at all four sampling stations.
- The plutonium-239 concentration in the air is normally less than one tenth the M.P.C. for the general -- public.

Concentrations exceeding one tenth the maximum --

permissible concentration were recorded on only 7 occasions, 2 of which exceeded the M.P.C. (both at station 2-2). Both of the latter values (2.5 x 10<sup>-13</sup> and 0.86 x 10<sup>-13</sup> microCi/cm<sup>3</sup>) - did not exceed one—tenth the M.P.C. for insoluble plutonium-239 compounds.

- The plutonium-239 concentration was null in - 29.61% of the analyses of samples from all - stations.

The highest incidence of the presence of plutonium-239 in the air was recorded at station 2-2, where null values for this element were obtained in only 13.2% of the ten-day sample analyses. This is also the station that recorded the highest also the station that recorded the highest acconcentrations. In 1966, it recorded an overall mean value of 1.2 x 10<sup>-15</sup> microCi/cm<sup>3</sup>, which aircrease to 11.9 x 10<sup>-15</sup> in 1967.

- In the zone containing the town, the plutonium-239 concentrations were null in 30.6% of the tenday periods. The station in this zone normally recorded were less than one thousandth the M. P.C., 42.86% were less than one hundredth the M.P.C., and 6.12% were less than one tenth the M.P.C., and 6.12% were less than one tenth the M.P.C.. The mean value for 1966 was 0.38 x 10<sup>-15</sup> microCi/cm<sup>3</sup>. This value dropped considerably in 1967 to a mean of o.09 x 10<sup>-15</sup> microCi/cm<sup>3</sup>.
- At station 2-1, the mean values for 1966 and 1967 were 1.17 x 10<sup>-15</sup> microCi/cm<sup>3</sup> and 0.38 x 10<sup>-15</sup> microCi/cm<sup>3</sup>, respectively.

This mean values for station 3 were  $0.73 \times 10^{-15}$ 

and  $0.47 \times 10^{-15}$  microCi/cm<sup>3</sup>, microCi/cm<sup>3</sup>, - respectively.

- The incidence of the highest plutonium-239 - concentration values coincides with periods in which wind velocities reach levels higher than 35 km/hr.

### 4.3. Uranium-234 and uranium-235 concentration

Table V contains the concentration of the activity due to the two isotopes, uranium-234 and uranium-235, in the ten-day sets of air samples from each of the sampling stations
during 1967. The figures for 1966 are not shown, because
separate analyses were not scheduled at first for these -radionuclides.

The maximum permissible concentration of uranium-234 or uranium-235 in the air for the general public, according to ENEA norms (1968), is the same for both: 4 x 10<sup>-12</sup> microCi/cm<sup>3</sup> for insoluble compounds, which are the most dangerous ones. Moreover, the isotopes scattered as a result of the air accident are actually found in the form of insoluble compounds in this area.

The results implicit in the data contained in table V - are these:

- Some particles of the uranium-234 and uranium-235 oxides contained in farmed soil are placed in suspension in the air and scattered over the entire area by the wind, for the presence of these compounds has been recorded at all four sampling stations.
- The concentration of activity due to the two isotopes, uranium-234 and uranium-235, is lower in all cases

than the maximum permissible concentration of these radionuclides in the air.

The maximum mean concentration value for all -tendayy periods was less than one tenth the above-mentioned M.P.C..

- The uranium-235 and uranium-234 content was null in only 2.98% of the samples analyzed.
- In the zone containing the town, much lower concentrations were detected than in the other zones, as the mean concentration values for each of the stations in 1967 show. These mean values are:
  - Station 2-1: 9.57 x 10<sup>-15</sup> microCi/cm<sup>3</sup>
  - Station 2-2:  $9.60 \times 10^{-15}$
  - Station P: 0.42 x 10<sup>-15</sup>
  - Station 3:  $11.81 \times 10^{-15}$

These values mean that the annual mean concentrations at station 2-1, 2-2 and 3 were on the order of  $5 \times 10^{-2}$  times the M.P.C., and on the order of  $10^{-4}$  times the M.P.C. in the town.

## 5. CONCLUSIONS

The following conclusions may be drawn from an analysis of the results set forth in the preceding section:

The elimination of the radionuclides contaminating the surface layer of soil, and their dilution in the subsoil
to a depth of 25 cm., has proven effective in reducing the mean values for radionuclide concentration
in the air to levels consistently below the permissible
maxima.

On very few occasions was the daily concentration of radionuclides in the air higher than the maximum - permissible value, and on all of those occasions, the wind velocity fluctuated between 17 and 64 km/hr.

The combined action of the weather conditions and farming of the soil have resulted in the resuspension of particles of plutonium and uranium oxides in the air, although the concentration of these radionuclides in the air has consistently remained well below the -maximum permissible concentrations for these elements,
in the insoluble form in which they are found in the -area of the accident.

The incidence of the presence of uranium in the air - is higher than that of plutonium, for 30% of the samples analyzed showed no trace of plutonium, as compared - with 3% which contained no uranium.

In the zone containing the town, the airborne—concentrations of plutonium and uranium have remained far below the ones in other areas where larger.—concentrations of these elements were mixed into the soil to a depth of 25 cm. In the latter areas, the—annual mean concentration values were practically the same for each of the years studied. The values for—1967 were lower than the ones for 1966, with the—exception of the concentration of plutonium—239 in area 2-2, which increased significantly in 1967.

TABLE I. AIRBORNE GROSS ALPHA ACTIVITY DURING 1966

				SAMPLING	1	STATION. microcuries	Ì	x 10 <sup>-15</sup> /cm.				
Month	7	Statión 2-	-1	5	Statión 2	- 2	Statión	Station P.( in the town)	town)	<i>S</i>	Statión 2	en 1
	Max	Min.	Mean	Max.	Min.	Mean	Max.	Min.	Mean	Max.	Min.	Mean
January								•				
February												
March		•										
April												
Мау						,						
June	3.0	< 0.7	1.4	7.3	< 0, 7	1. 69	8.9	<0.7	1.9	1.69	<0.7	1.2
July	33, 8	< 0.7	4.6	13.2	< 0.7	6, 1	16.6	<0.7	4.4	16.9	< 0.7	4.1
August	12, 2	<0.7	4.6	236.6	2 '0>	14 .5	422.5	<0,7	17.1	13. 9	<0.7	4
September	15.0	< 0.7	4.9	74.4	< 0.7	6.6	16.1	<0.7	4.4	72.7	<0.7	7.9
October	22.0	< 0.7	4.2	120.0	<0.7	7, 3	9.3	<0.7	3.5	6.3	<0.7	2.4
November	6.6	<0°.	5,5	14.9	2 .0>	3.7	10.3	<0.7	3,5	8.3	<0.7	2.4
December	16. 1	<0.7	3, 4	23, 7	<0.7	3.9	7.3	<0.7	2.7	16.4	<0.7	3.4

TABLE II. AIRBORNE GROSS ALPHA ACTIVITY DURING 1.967

				SAMPLING	1	STATION. microcuries	×	10 <sup>-15</sup> / cm.		·		
Month	S	Statión 2-	-	S	Statión 2 -	-2	Statión	Statión P.( in the town)	fown )	S	Statión 2-	. 3
	Max,	Min.	Mean	Max.	Min.	Mean	Max.	Min.	Mean	Max.	Min.	Mean
January	37,2	7,0>	3,9	87,0	<0,7	5,7	16,6	<0,7	2,5	15,5	<0,7	2,9
February	81,1	<0,7	7,4	41,7	7,0>	5,7	13,7	<0,7	3,7	118,0	<0,7	6,1
March	4,7	<0,7	2,0	18,6	7,0>	5,6	7,9	<0,7	2.0	9.9	202	2,0
April	11,5	<0,7	3,5	461,4	2,0>	4,6	2,5	<0,7	2,7	6,4	<0,7	2,4
Мау	8,1	<0,7	3,0	17,7	<0,7	4,7	11,2	<0,7	2,2	7,8	<0,7	2,5
June	12,3	<0,7	2,9	18,6	<0,7	3,2	17,6	<0,7	3,1	7,6	<0,7	3,2
July	11,7	7,0>	3,4	11,8	2'0>	3,8	7,9	<0,7	2,7	7,3	<0,7	2,5
August	3,9	<0,7	1,9	17,1	2,0>	3,5	6,9	40,7	1,7	3,9	<0,7	1,6
September	ω, π,	<0,7	3,2	10,1	3'0>	3,0	11,3	<0,7	2,6	5,6	<0,7	2.2
October	7,8	7,0>	1,9	១ ំន	<0,7	2,9	5,2	40,7	2,1	10,0	<0,7	2,5
November	7,8	<0,7	2,2	6,7	<0,7	2,5	5,0	<0,7	1,9	8,3	<0,7	2,5
December	3,9	<0,7	1,5	4,1	<0,7	1,9	3.2	<0,7	1,4	3,9	<0,7	1,9

TABLE III. AIRBORNE PLUTONIUM-239 CONCENTRATION DURING 1.966

								·				
Days				SAMPLING		STATION. microcuries	.s x 10 <sup>-15</sup> /cm <sup>3</sup>	, /cm <sup>3</sup>				
/		Statión 2-	-		Statión 2	-2.	Statión	Station P.(in the	e town)	<b>5</b> ,	Statión 2-3	. 3
Month	1st - 10	11 th 20 th	21 <sup>51</sup> 30 <sup>th</sup>	1st_10 th	11h; 20th	21 <sup>5</sup> t 30 <sup>th</sup>	1 <sup>st</sup> _10 <sup>th</sup>	11, ½20,11	zist 30th	1st 10th	11 <sup>th</sup> _20 <sup>th</sup>	21 <sup>51</sup> , 30 <sup>th</sup>
January												
February												
March			-									
April												
May												
June					0,67	0,34	1	0,47	0,83	l I	l l	0,67
July	00,00	0,13	0,99	0,29	0,14	0,40	ţ	1	i i	0,13	0,06	0,12
August	1,60	0,90	0,20	0,94	0,94	2,24		1	ł	0,77	0,10	0,73
September	0,37	0,16	0,89	0,44	5,10	2,71	0,00	00,0	0,00	5,12	0,50	1,60
October	2,59	0,74	10,33	0,49	0,10	0,00	0,30	2,90	0,29	0.05	0,09	00°¢.
November	0,08	00,0	1,57	1,57	0,13	0,22	0,00	0,00	0.08	0.23	0,00	0,00
December	0,05	00,00	0,47	0,62	4,57	2,17	0,00	0,08	0,41	0,05	0,44	0,22

TABLE IV. AIRBORNE PLUTONIUM - 239 CONCENTRATION DURING 1.967

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Days				SAMPLING	STATION.	microcuries		x 10 <sup>-15</sup> /cm.				
/		Statión 2	2-1	0,	Statión 2	-2	Statión	P.( in the	town)	J,	Statión 2-	- 3
Month	1st_10th	11 <sup>1</sup> h 20 <sup>1</sup> h	21 <sup>51</sup> -30 <sup>th</sup>	1st 10th	11th 20th	21 <sup>S1</sup> _30 <sup>th</sup>	ıst noth	11th_20th	21 <sup>51</sup> -30 <sup>th</sup>	151-10th	11 <sup>th</sup> _20 <sup>th</sup>	21 <sup>51</sup> , 30 <sup>th</sup>
January	00,00	1,89	00,0	0,15	0,32	6,17	00,00	0,00	0,08	9,04	0,04	0,04
February	08'0	00,0	00,00	85,90	0,88		0,12	00,00	0,05	0,09	0,13	0,05
March	2,16	. 0,30	0,23	1,15	00'0	0,29	0,06	0,04	0,04	0,26	0,17	0,07
April	00,00	0,50	00,00	24,89	†	0,18	0,11	0,28	0,05	0,05	0,07	0,06
May	00,00	00,00	0,49	6,27	1,88	0,35	0,18	0,05	0,09	96,0	0,10	0,33
June	00'0	00,00	0,00	00'0	0,49	00,0	00,00	0,07	0,08	50,0	60'0	00,00
July	00,00	00,00	0,93	0,13	00,00	0,51	00,00	0,16	0,04	0,10	00'0	00,00
August	4,36	1		250,84	1,05	4,71	90'0	0,08	0,07	00,00	00,00	00,00
September	1	00,00	00'0	0,32	2,70	00 0	0,00	00'0	0,00	00,00	00,00	0,00
October	0,19	00,00	00,00	0,71	0,28	98'0	80 0	0,04	0,09	00'0	00,00	00,00
November	0,19	0,17	00'0	0,87	0,35	0,00	0,58	60'0	0,05	00,0	90'0	0,29
December	0,07	0,07	P T	0,05	1,50		00,00	0,71	1	00,00	0,00	l I

TABLE V. AIRBORNE ACTIVITY CONCENTRATION FROM URANIUM -234 URANIUM -235 DURING 1.967

Days				SAMPLING	1	STATION. microcuries	ocuries x	10 <sup>-15</sup> /cm <sup>3</sup>				
/	<i>o</i>	Statión 2 -		<i>S</i>	Statión 2	- 2	Slatión	P.( in the	town)	ξ)	Station 2	- 3
Month	1st - 10th	11 <sup>th</sup> 20 <sup>th</sup>	21st 30th	181_10 <sup>th</sup>	11th. 20th	2131-30th	181-10 <sup>th</sup>	11th_20th	21 <sup>5</sup> t 30 <sup>th</sup>	1 <sup>st</sup> -10 <sup>th</sup>	11th 20th	21 <sup>51</sup> _ 30 <sup>th</sup>
January	4,59	0,68	2,33	83,48	33,72	37,03	0,12	0,06	0,28		0,44	0,57
February	208,77	3,80	8,20	5,58	1,69	1	0,11	0,76	0,16	3,15	0,79	0,50
March	4,51	1,04	3,17	1,19	57,99	2,69	0,14	0,12	0,07	3,31	0,17	0,18
April	4,16	2,78	2,96	3,31	†	1,53	0,20	0,16	0,21	0,06	0,19	0,23
Мау	1,66	0,97	09 0	0,49	3,74	2,00	00,0	0,15	0,18	20'0	0,08	12,84
June	0,78	0.35	2,03	13,55	10,44	11,67	0,22	0,14	0,10	7,01	10,25	11,50
July	7,42	11,24	15,98	5,07	17,87	0,38	0,06	0,11	0,04	327,43	0,38	0,66
August	0,18		-	4,43	3,22	00,00	0,57	0,23	0,31	14,44	2,03	1,46
September		2,34	-	1,62	0,17	4,32	1,14	0,34	0,47	5,22	1,19	4,16
October	0,32	0,21	0,67	0,20	0,64	2,53	0,17	0,24	0,34	89'0	0,17	0,22
November	2,32	1,45	1,42	1,25	0,07	0,50	1,84	0,67	2,29	80'0	0,26	0,84
December	0,03	00,00	ŀ	0,08	4,36	i i	2,78	00,00		2,17	0,14	!

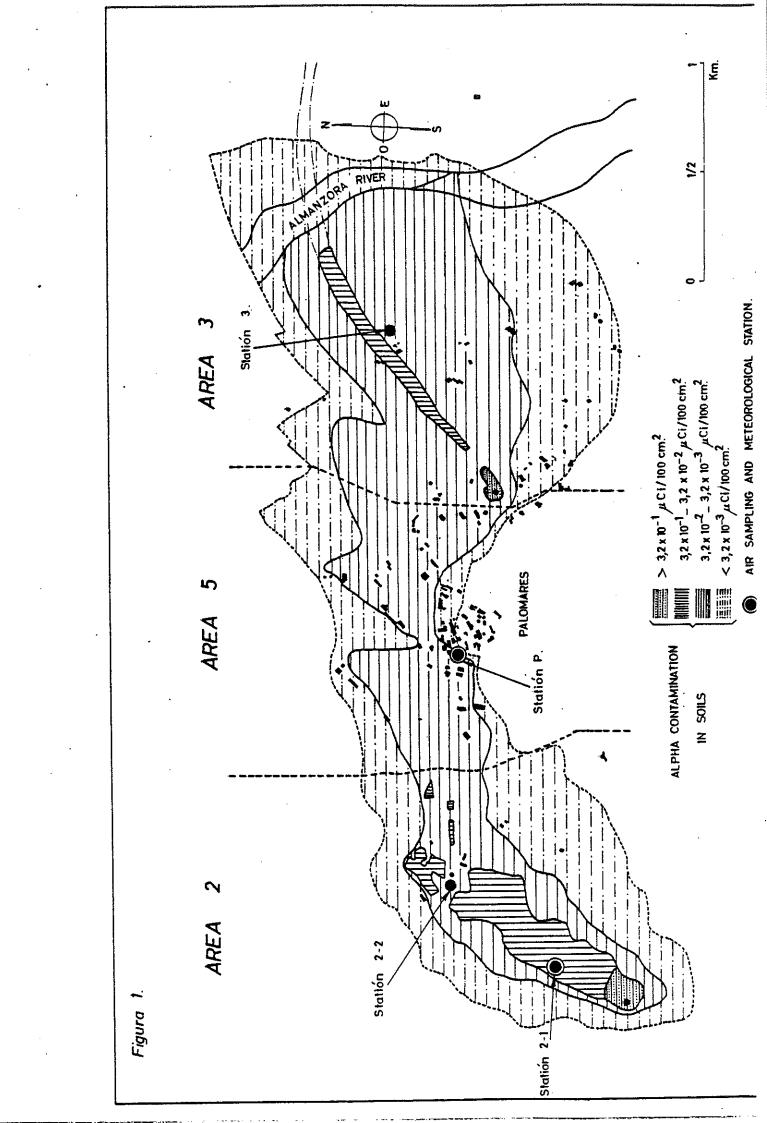




Fig. 2. - Sampling Station

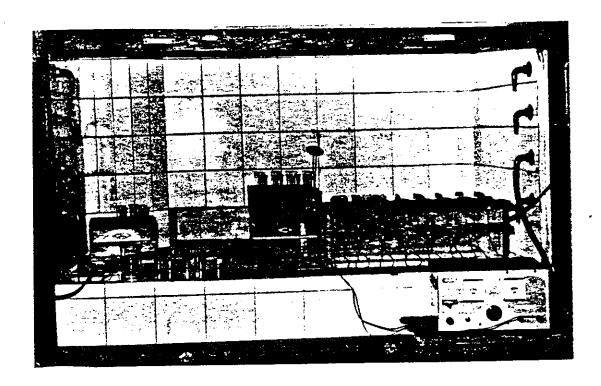


Fig. 3. - Device for Electrodeposition

