ANTHROPOGENIC EMISSIONS OF ^{210}Po, ^{210}Pb AND ^{226}Ra IN AN ESTUARINE ENVIRONMENT

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An extensive study on the distribution of natural radionuclides in an estuarine ecosystem located in Southwestern Spain is presented. This environment is highly affected by the wastes released by a phosphoric acid industry which uses phosphate rocks as raw material for fertilizer production. This rock has generally high concentrations of U and its daughters. The estuary is formed by two rivers, Odiel and Tinto, which have a common mouth into the Atlanic Ocean and a salt marsh (Odiel marsh) affected by the income of Odiel riverwaters. This river receives directly the liquid and part of the solid (gypsum) wastes released from the industries. Besides that, most of the phosphogypsum wastes are stored in uncovered piles at the right margin of the Tinto river. The study has concluded that the wastes from such industries are the cause of the enhanced concentrations found at the bed of both river channels as well as the enhancement found in surface soils in certain zones of the Odiel wet marshland. Indeed, the Northern marsh and the Mojarrera channel at the Odiel marsh seem to be the main sinks of the contaminant released by the phosphoric acid industry.

On a historical time scale, sediments may be regarded as, at least, a temporary sink for much of the material which passes through the various chemical and biological cycles operating on the earth's surface. At present, the material which is released and takes part in these cycles can have its composition markedly affected by anthropogenic emissions. Sediments became an environmental host for many of the waste products discharged by society. The effects of these man-made emissions, in some situations, can be sufficiently strong to affect highly the composition of the deposited sediment.

In the marine environment, near-shore sediments retain the strongest memory of the impact of the released material on the earth's surface. This is because these sediments accumulate at rates which can be as much as several orders of magitude higher than those in deep sea and because they are being formed in areas in which many anthropogenic substances initially reach the sea. Most industrial complexes are located in estuarine areas close to the sea. Thus, they behave as sinks of much of the anthropogenic emissions, which with time will probably be transported to their close sea environment. Thus, the study of natural environmental radioactivity in estuarine systems is of great importance and interest in health physics and many other purposes.

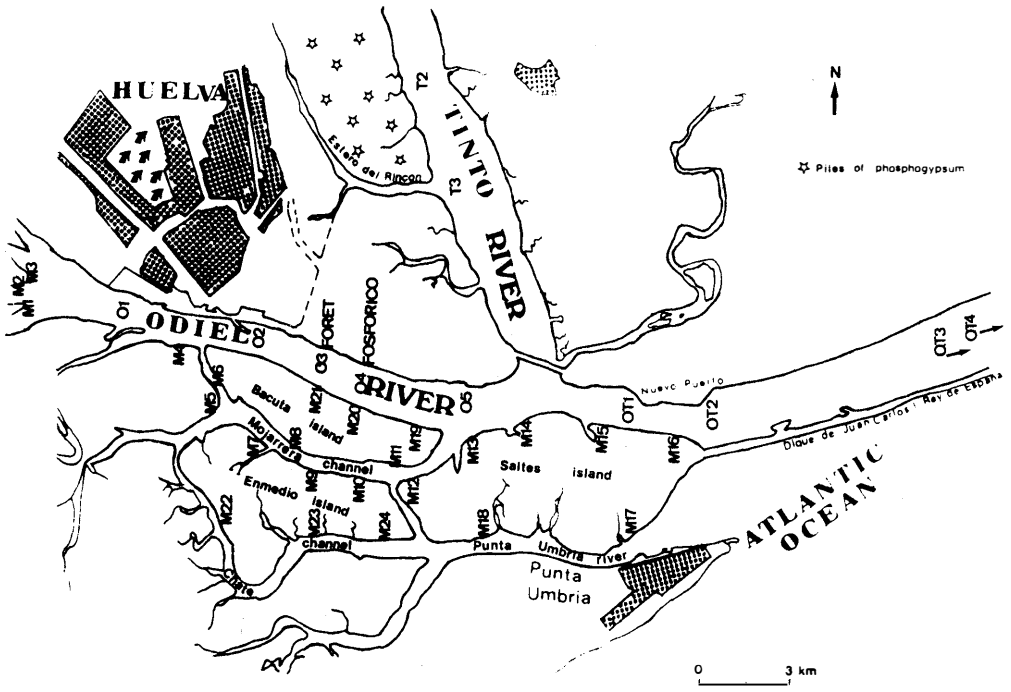

Fig. 1. Map of the Estuarine area of Huelva. Bottom sediments from the Odiel and Tinto rivers are denoted by "O" and "T", respectively, followed by a number which indicates the location of the sample. Soil samples from the Odiel marsh are denoted by "M" followed by the number of the station

In Spain a large industrial complex is located near the city of Huelva, along the estuary formed by the Odiel and Tinto rivers. A phosphoric acid industry for fertilizer production is located at the left margin of the Odiel river (see Fig. 1).

Phosphate ores used for phosphoric acid production contain important amounts of natural radioactive elements, specially ^{238}U and daughters, in concentrations that depend on their geographical and geological origin, but which can reach up to 300 ppm for U. The radioactive impact of the industrial complex over the Odiel and Tinto rivers has been widely studied for the case of U and Th isotopes.[1,2] A dose estimation due to the ^{222}Rn inhalation emitted by the phosphogypsum piles has been also published.[3] The movements of these waters due to the flow of the water during change in tide, produce the logical distribution of the contaminant upwards and downwards of the point of release.

Next to the rivers, there is a large saline wet marshland. This area is a natural reservation with high biological activity. It is located at the right side of the Odiel river

in front of the fertilizer industry and is highly affected by the income of the Odiel river waters. The radiological significance of ^{210}Po, ^{210}Pb and ^{226}Ra in the future dose received by the population motivated the extention of previous works. The present paper discusses the results obtained in the study in the presence of these radionuclides in bottom sediments from the Odiel and Tinto rivers as well as in surface soil samples from the wet Marshland. The general aim was to study the reach and influence that the fertilizer production has over all the estuarine area.

Experimental

Two sampling campaigns were performed in February and November 1993, respectively. In each campaign bottom sediments from the Odiel and Tinto rivers and surface soil sampes from the Odiel Marsh were collected. Bottom sediments were collected at the center of the rivers, whereas soil samples were collected during the low tide in areas covered with water during the high tide.

The samples were dried, powdered and homogenized before the analysis. Some 1 g of sample was spiked with well known ^{208}Po activities. After digestion of the sample with HNO_3 and aqua regia, Po is extracted by solvent extraction and the final solution is self-deposited onto silver planchets.[4] ^{210}Po activity is determined by α-spectrometry with surface barrier or ion-implanted detectors. ^{210}Pb and ^{226}Ra are measured by γ-spectrometry with Reverse Coated Germanium detectors. ^{210}Pb duplicate analysis were performed in sediment aliquots,[5,6] being significantly concordant with those obtained by γ-spectrometry.

Results and discussion

To simplify the discussion, the data obtained on bottom sediments from the Odiel and Tinto rivers for both sampling campaigns will be first discussed. For the sake of clarity, we will denote by "O" those samples collected at the Odiel river and by "T" those from the Tinto river. Samples collected in the confluence of both river will be denoted by "OT". The number following these data will denote the location of the sampling station (see Fig. 1). Latelly, data obtained for the same sampling campaigns in surface soils from the Odiel wet marshland will be presented and discussed. The location of the soils collected at this area is shown in the same Figure.

Bottom sediments

In Tables 1 and 2 the ^{210}Po, ^{210}Pb and ^{226}Ra activity concentrations in mBq/g together with some activity ratios, for February and November campaigns are presented, respectively.

From the results for all radionuclides in both sampling campaigns,[7] it can be concluded that all bottom sediments from the Odiel river present activity concentrations

Table 1
Radioactive concentrations in mBq/g in bottom sediments from the Odiel and Tinto rivers collected in Februay 1993

Code	^{210}Po	^{210}Pb	^{226}Ra	$^{210}Po/^{210}Pb$	$^{210}Pb/^{226}Ra$
O1	518 ± 26	506 ± 31	406 ± 96	1.02 ± 0.08	1.24 ± 0.30
O2	726 ± 21	922 ± 38	584 ± 88	0.79 ± 0.04	1.57 ± 0.25
O3	4919 ± 126	2743 ± 64	3395 ± 198	1.79 ± 0.06	0.81 ± 0.05
O4	1300 ± 100	1176 ± 41	1140 ± 129	–	1.03 ± 0.12
O5	884 ± 33	763 ± 35	723 ± 65	1.16 ± 0.07	1.05 ± 0.11
T1	76 ± 10	31 ± 16	70 ± 20	2.45 ± 1.30	0.44 ± 0.26
T2	221 ± 16	82 ± 20	≤ 203	2.70 ± 0.69	–
T3	1498 ± 75	914 ± 40	618 ± 110	1.64 ± 0.11	1.47 ± 0.27
OT1	474 ± 13	445 ± 27	211 ± 68	1.07 ± 0.07	2.11 ± 0.69
OT2	1062 ± 47	629 ± 31	364 ± 77	1.69 ± 0.11	1.73 ± 0.37
OT3	56 ± 5	20 ± 15	≤ 67	2.80 ± 2.11	–
OT4	54 ± 8	< 26	< 62	–	–

Table 2
Radioactive concentrations in mBq/g in bottom sediments from the Odiel and Tinto rivers collected in November 1993

Code	^{210}Po	^{210}Pb	^{226}Ra	$^{210}Po/^{210}Pb$	$^{210}Pb/^{226}Ra$
O1	567 ± 38	511 ± 86	405 ± 55	1.11 ± 0.20	1.26 ± 0.27
O2	500 ± 30	419 ± 66	363 ± 75	1.19 ± 0.20	1.15 ± 0.30
O3	2080 ± 140	925 ± 134	972 ± 109	2.25 ± 0.36	0.95 ± 0.17
O4	1330 ± 80	1160 ± 189	831 ± 83	1.15 ± 0.20	1.40 ± 0.27
O5	1660 ± 140	1450 ± 213	1273 ± 112	1.15 ± 0.19	1.14 ± 0.19
T1	–	98 ± 22	94 ± 22	–	1.04 ± 0.34
T2	233 ± 16	73 ± 14	87 ± 20	3.19 ± 0.65	0.84 ± 0.25
T3	788 ± 50	733 ± 116	718 ± 62	1.08 ± 0.18	1.02 ± 0.18
OT1	600 ± 24	408 ± 70	407 ± 45	1.47 ± 0.26	1.00 ± 0.21
OT2	618 ± 39	529 ± 81	416 ± 29	1.17 ± 0.19	1.27 ± 0.21
OT3	21 ± 2	≤ 35	≤ 32	–	–
OT4	19 ± 2	15 ± 10	19 ± 10	1.27 ± 0.86	0.79 ± 0.67

above background levels.[8] Thus, radioactive concentrations above 470, 445 and 211 mBq/g for ^{210}Po, ^{210}Pb and ^{226}Ra, respectively, are found along the estuary of the Odiel river in February. Radioactive concentrations in November are similar to those found in February. The highest activity concentration is found, as it was expected, at the point of release of the phosphoric acid industry (O3), being the radioactive concentration in February much higher than that in November for all radionuclides. This activity concentration decreases downstream the river channel but still high radioactive concentrations were found at the common confluence with the Tinto river (OT1 and OT2). Pattern distributions of ^{210}Po and ^{210}Pb along the Odiel river for samples collected in November are shown in Fig. 2. These pattern distributions are similar to that for ^{226}Ra, and reflects all said above. Samples OT3 and OT4 were collected at the Atlantic Ocean coast close to the mouth of the river. From the data in Tables 1 and 2 for stations OT3 and OT4 is easy to conclude that the contamination in the Odiel river does not yet reach the coast. In fact, radioactive concentrations are similar to typical background levels (~ 25 mBq/g).[9]

In the case of the Tinto river channel, three samples were collected. The radioactive concentration in both sampling campaigns are similar and excepting station T3, they are

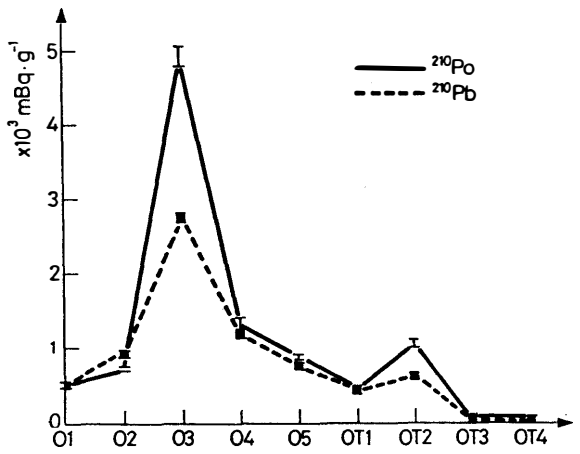

Fig. 2. Comparison of ^{210}Po and ^{210}Pb activity concentration in mBq/g in bottom sediments from the Odiel river collected in February 1993

lower than those found in the Odiel river channel. Therefore, the exception of station T3, has an activity concentration even higher than in most location of the Odiel river. This station is located at the confluence of a small natural rivulet (Estero del Rincón) with the Tinto river. This rivulet crosses the phosphogypsum storage area and probably,

it can redissolve part of the radioactivity concentration contained in these wastes. Station T2 also seems to be contaminated. This sample was collected close to an artifical stream which crosses the phosphogypsum storage area. Upstream this stations, the activity concentrations are similar to background.

From the results it is clear that the wastes from the phosphoric acid production enhance the radioactive concentrations in bottom sediments from the Odiel and Tinto rivers. However, this contamination seems to be restricted to the estuarine area of the rivers, thus being the radioactive concentrations at the coastal area similar to background levels.

It is also interesting to discusse the $^{210}Po/^{210}Pb$ and $^{210}Pb/^{226}Ra$ activity ratios which are also given in Tables 1 and 2 for February and November campaigns, respectively.

The $^{210}Po/^{210}Pb$ activity ratio reveals the existence of secular equilibrium in those stations less contaminated whereas those highly enhanced show an excess of ^{210}Po compared to ^{210}Pb. The excess is confirmed by the values of this activity ratio in stations O3, T2 and T3. This excess should reflect either a higher presence of ^{210}Po in the wastes from the industries or a different environmental behaviour after the release. The excess disappears downstream the river channel. The excess of ^{210}Po found at station ST1, located upstream from the phosphogypsum piles in the Tinto river, which seems to be unenhanced by the industries, must be related with the higher ^{210}Po association with solid particles.

In the case of the $^{210}Pb/^{226}Ra$ activity ratio, most stations present values which indicate the existence of secular equilibrium between both radionuclides or a little excess of ^{210}Pb compared to ^{226}Ra. However, station O3 from the Odiel river and T1 from the Tinto river, present a clear excess of ^{226}Ra related to ^{210}Pb (mainly in samples collected in February) given $^{210}Pb/^{226}Ra$ activity ratios clearly below unity. This situation should be explained in different ways for each sample. Sample O3, was collected at the point of discharge from the phosphoric acid industry into the Odiel river. Thus, the excess of ^{226}Ra should indicate the existence of a similar excess in the wastes. As it is well know, phosphogypsum (mainly $CaSO_4$) are enriched in ^{226}Ra comparing with other radionuclides from the ^{238}U decay series.[10] The equilibrium is restored downstream the Odiel channel due to a higher association of Pb[11] to solid phases as well as to the redissolution of ^{226}Ra in saline waters.[8,12]

The case of station T1 located upstream from the industries must be related to a higher presence of ^{226}Ra in solution. Thus, due to the solubility of this radionuclide in natural waters, releases of wastes with high concentration of ^{226}Ra would produce a general increase of ^{226}Ra in solution in compare with uncontamined rivers. This excess of Ra would produce a higher contain of ^{226}Ra in bottom sediments in locations upwards, compared to ^{210}Po which has been already incorporated to the sediments.

Surface soils

In Tables 3 and 4 radioactive concentrations for the same radionuclides and same activity ratios as those for bottom sediments are presented here but for surface soil samples from the Odiel wet marshland. In each Table results for February and November campaigns are presented, respectively.

The results reveal clearly that the Marsh is also enhanced by the phosphoric acid production.[13] The activity concentrations are, in general, below the maxima found in bottom sediments from the Odiel river. However, activity concentrations up to 370, 660 and 500 mBq/g for ^{210}Po, ^{210}Pb and ^{226}Ra, respectively, were found in February, being in November the levels up to 580, 670 and 600 mBq/l for ^{210}Po, ^{210}Pb and ^{226}Ra, respectively. Besides that, much higher radioactivity concentrations were found in samples collected in the Northern marsh (see Fig. 1) in the campaign of November.

In the Odiel river margin of the marsh, eight stations were sampled for soils. These located in front of the fertilizer industries, present radioactive concentrations in the campaign of February only slightly above background concentrations, being those upstream and downstream more contaminated. In November, the station in front of the industries presents the lowest radioactive concentrations, below the concentrations found in February. Thus, it seems that contamination in the transversal direction is not as high as in the longitudinal direction. Of course, this fact must be related with the flow velocity of the river water. The differences in stations M20 and M21 between both

Table 3
Radioactive concentrations in mB/g in surface soil samples collected at the Odiel wet marshland in February 1993

Code	^{210}Po	^{210}Pb	^{226}Ra	^{210}Po/^{210}Pb	^{210}Pb/^{226}Ra
M6	264 ± 412	454 ± 27	321 ± 40	0.581 ± 0.043	1.41 ± 0.20
M7	290 ± 31	424 ± 29	280 ± 43	0.684 ± 0.087	1.51 ± 0.25
M10	341 ± 19	514 ± 36	326 ± 56	0.663 ± 0.059	1.58 ± 0.29
M11	276 ± 14	435 ± 30	304 ± 39	0.634 ± 0.054	1.43 ± 0.21
M12	25.1 ± 1.7	37.4 ± 9.1	26.4 ± 14.8	0.671 ± 0.169	1.42 ± 0.87
M14	108 ± 6	160 ± 25	143 ± 35	0.675 ± 0.112	1.12 ± 0.32
M15	367 ± 34	658 ± 30	505 ± 44	0.558 ± 0.058	1.30 ± 0.13
M16	117 ± 7	137 ± 18	65.4 ± 25.4	0.854 ± 0.123	2.10 ± 0.86
M20	288 ± 25	125 ± 20	≤ 241	2.30 ± 0.42	–
M21	181 ± 14	125 ± 21	≤ 148	1.45 ± 0.27	–
M22	168 ± 9	236 ± 25	136 ± 36	0.716 ± 0.085	1.73 ± 0.50
M23	124 ± 8	205 ± 23	76.6 ± 35.9	0.605 ± 0.078	2.68 ± 1.29
M24	44.9 ± 2.9	34.8 ± 6.8	33.2 ± 11.4	1.29 ± 0.26	1.05 ± 0.41
M18	29.0 ± 1.8	34.0 ± 8.4	34.8 ± 13.7	0.853 ± 0.217	0.98 ± 0.45
M17	40.5 ± 2.3	53.2 ± 6.5	43.3 ± 10.5	0.761 ± 0.102	1.13 ± 0.33

Table 4
Radioactive concentrations in mBq/g in surface soil samples collected at the Odiel wet marshland in November 1993

Code	^{210}Po	^{210}Pb	^{226}Ra	$^{210}Po/^{210}Pb$	$^{210}Pb/^{226}Ra$
M1	199 ± 10	236 ± 16	166 ± 25	0.843 ± 0.071	1.42 ± 0.24
M2	778 ± 41	820 ± 27	711 ± 45	0.949 ± 0.059	1.153 ± 0.082
M3	643 ± 28	795 ± 23	681 ± 36	0.811 ± 0.042	1.167 ± 0.070
M4	210 ± 11	233 ± 13	242 ± 23	0.901 ± 0.069	0.963 ± 0.106
M5	222 ± 11	156 ± 14	161 ± 24	1.42 ± 0.15	0.97 ± 0.17
M6	580 ± 26	673 ± 16	598 ± 25	0.862 ± 0.044	1.125 ± 0.054
M7	213 ± 10	287 ± 12	213 ± 21	0.742 ± 0.047	1.347 ± 0.144
M8	130 ± 7	212 ± 14	130 ± 21	0.613 ± 0.052	1.631 ± 0.285
M9	141 ± 7	161 ± 12	149 ± 20	0.876 ± 0.078	1.081 ± 0.166
M10	188 ± 10	161 ± 13	149 ± 22	1.168 ± 0.113	1.081 ± 0.182
M11	64.6 ± 4.0	76.2 ± 12.3	49.0 ± 18.6	0.848 ± 0.147	1.56 ± 0.64
M12	27.9 ± 2.0	24.8 ± 12.3	46.0 ± 22.3	1.13 ± 0.56	0.54 ± 0.37
M13	565 ± 23	681 ± 23	673 ± 39	0.830 ± 0.044	1.012 ± 0.068
M14	41.0 ± 3.2	18.4 ± 10.1	30.4 ± 16.5	2.23 ± 1.24	0.605 ± 0.467
M15	441 ± 20	589 ± 21	484 ± 32	0.749 ± 0.043	1.217 ± 0.091
M16	131 ± 7	162 ± 11	134 ± 16	0.809 ± 0.070	1.21 ± 0.17
M19	233 ± 11	268 ± 15	245 ± 26	0.869 ± 0.064	1.094 ± 0.131
M20	16.2 ± 2.5	15.4 ± 10.1	21.3 ± 16.0	1.05 ± 0.71	0.72 ± 0.72
M21	37.2 ± 2.5	22.9 ± 10.9	20.3 ± 17.7	1.62 ± 0.78	1.12 ± 1.12
M22	44.2 ± 2.5	26.6 ± 11.1	31.7 ± 17.2	1.66 ± 0.70	0.84 ± 0.57
M23	194 ± 9	121 ± 13	97.3 ± 20.7	1.61 ± 0.19	1.24 ± 0.30
M18	54.4 ± 3.7	51.9 ± 11.1	35.3 ± 17.6	0.830 ± 0.044	1.012 ± 0.068
M17	54.7 ± 3.7	51.9 ± 11.1	35.3 ± 17.6	0.68 ± 0.10	2.7 ± 1.7

campaigns must be related to the composition of the samples. Thus, samples collected in November had higher content of sand particles, which was reflected in the density (0.7 and 1.5 g/cm^3 dry weight in February and November, respectively), and in the organic content (10% and 1.5%, respectively) of the samples. Concentrations at the east side of the Saltés island, which clearly reveals a contamination, are similar to those found in bottom sediments at the confluence of the Odiel and Tinto rivers (OT1 and OT2). The differences found in stations at the east side of the Saltés island again must be partially related with the composition of the samples. Thus, values of density and organic content of stations M16 and M14 may reveal that they have a high amount of sandy particles, which produces a decrease in the specific activity of the samples.

In general, the differences in radioactive concentrations in stations along the Odiel river margin could be due to differences in organic content. If we normalize the radioactive concentrations to the organic content of the soil samples, the differences

between stations are less significative. This effect can be clearly observed in Fig. 3, where the ^{210}Po concentration in mBq/g are compared with the data normalized to the organic content for those soils along the Odiel river margin, which showed wider differences in composition. Only, station M14 does not seem to be contaminated in the same degree as those stations in the vicinity.

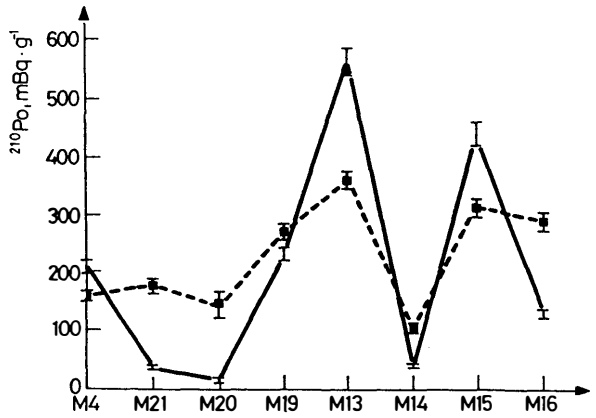

Fig. 3. Comparison of ^{210}Po activity concentration in mBq/g of dry soil sample (continuous line) with the ^{210}Po activity concentration normalized to the organic content of the soil samples (dashed line) collected along the Odiel river bank of the Odiel marsh in November 1993. Data of normalized concentrations (in mBq/g) have been divided by 10

Besides, that, a set of samples appears in which the influence of the wastes released from the industries is clearly reflected. Thus, soils along the Mojarrera channel have quite high radioactive concentrations. If we consider that the cause of the high activity concentrations found in sediments O1 and O2 was the movement of the contaminants upstream the Odiel river during high tide, this should be also the cause of the contamination along the Mojarrera channel. Thus, during the high tide the Odiel riverwater flows upstream. Part of this contaminated water will flow to the Northern marsh and the other part will flood the Odiel marsh through the north of the Bacuta island. The same could occur through the south of this island. Of this most of the contaminated water flows into the Mojarrera channel and a small part would flow into the Chate channel. The activity concentration at the Northwest of the Enmedio island decreases downstream the Chate channel and the Punta Umbría river, where background concentrations for ^{210}Po, ^{210}Pb and ^{226}Ra were found. Thus, the pattern distribution of radioactivity concentrations in soils across the marsh has clearly identified the way of

contamination into this area.The income of contaminated Odiel riverwaters through the north and probably also through the south of the Bacuta island is the cause of the levels found at the Odiel marsh. Besides that, the Mojarrera channel seems to be the main sink of the contamination in the Odiel marsh. However, the Northern marsh, with the highest concentrations, seems to be the main sink of all the estuary of Huelva.

Again, it is interesting to study the $^{210}Po/^{210}Pb$ and $^{210}Pb/^{226}Ra$ activity ratios. As in the case of bottom sediments, the $^{210}Pb/^{226}Ra$ activity ratio reflects, in general, the existence of a secular equilibrium or a slight excess of ^{210}Pb. This excess must be related to the higher Pb association with solid particles. However, redissolution of ^{226}Ra in areas of high salinity has been widely found for other estuaries,[8,12] which could also be the cause of a slight defect of ^{226}Ra. The case of the $^{210}Po/^{210}Pb$ activity ratio is different. Thus, in sediments from the Odiel river (excepting the point of release), the ratio reflects a secular equilibrium or a little excess of the daughter radionuclide (^{210}Po). However, soils from the marsh show, in general, a little defect of the same radionuclide, which means the concentration of ^{210}Po is below that of ^{210}Pb. If we consider that soil samples were collected in areas partially covered by water (during high tide), radioactive concentration in soils would be affected by the contamination of the waters. This contamination would present isotopic characteristics similar to those typical of dissolved phases, which in general present an excess of ^{210}Pb compared to ^{210}Po.

Conclusions

Two sampling campaigns have been performed in the estuarine area of Huelva in 1993. The data on bottom sediments from the Odiel and Tinto rivers, which directly receive the liquid and solid (gypsum) wastes from a phosphoric acid industry for fertilizer production, have unequivocally shown that this industry enhances all the estuarine area of both rivers. Surface soil samples from the wet marshland, located at the right margin of the Odiel river in front of the industries, have shown that the income of contaminated waters from the Odiel river produce the logical contamination of this area. However, the Northern marsh seems to be the main sink of the wastes released by the industries. Finally, data on sediments collected at the coastal zone (close to the confluence of the rivers with the Atlantic Ocean) have shown that the contamination is restricted to the estuary. In general, it seems that the bag of contaminant is moving to the Northern marsh and into the Mojarrera channel at the Odiel marsh due to water movement during tides.

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