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Sedimentology models from activity concentration measurements: application to the "Bay of Cadiz" Natural Park (SW Spain)

R.A. Ligero*, J. Vidal, M.J. Meléndez, M. Hamani, M. Casas-Ruiz

Departamento de Física Aplicada. Universidad de Cádiz, 11510 Puerto Real, Cádiz, Spain

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ABSTRACT

A previous study on seabed sediments of the Bay of Cadiz (SW of Spain) enabled us to identify several relations between sedimentological variables and activity concentrations of environmental radionuclides such as ^{137}Cs, ^{226}Ra, ^{232}Th and ^{40}K. In this paper the study has been extended to a large neighbouring inter-tidal area in order to establish if the above mentioned models can be generalized. As a result we have determined that the measured activity concentrations are closely to the values predicted by the theoretical models (correlation coefficient range = 0.85–0.93).
Furthermore, the proposal model for granulometric facies as a function of activity concentrations of the abovementioned radionuclides provides for the sediments distribution a representation which agrees with the values of the tidal energy distribution obtained using numeric models calibrated with experimental data from current meters and water level recorders.

1. Introduction

The Natural Park of the Bay of Cadiz is located in the southwest of the Spain. This geographic area is important in terms of ecology, economics, tourism and general social and cultural factors. Its configuration is the result of a natural evolution determined by geological agents, mainly the littoral dynamics, the mouth of the Guadalete river and the aeolian regime characteristic of the zone, and by biological factors such as the vegetation and fauna. One significant further factor is the profound alteration of the environment caused by human activities such as the exploitation of salt lagoons and fish farming. The Bay can be divided into two distinctive parts: the Outer Bay (located to the north of the Carranza Bridge, where the hydrodynamic conditions due to the strong tidal currents and the mouth of the river are important) and the Inner Bay to the south of this bridge, which is less exposed to the action of erosive agents. It is drained by the main channel that runs from the Puntales strait, continues along the creek of La Carraca, and flows into the open sea through the tidal channel of Sancti Petri (Fig. 1).

The Sancti Petri tidal channel is an inflow–outflow channel which extends from the Inner zone of the Bay of Cadiz to the outlet in the Atlantic Ocean. With a length of 17 km, this channel connects with a number of secondary channels which in turn, supply a vast area of tidal flats. The central course of this channel is deeper than the much shallower margins to each side. The depth varies between approximately 9 and 3 m and the cross-sectional area of the channel at the mouth decreases towards the more inland part of the channel.

From the oceanographic perspective, the Sancti Petri Channel links two bodies of water with different hydrodynamic characteristics, where the tide wave enters the two ends with different amplitudes and phases. A hydrodynamically critical point appears at Vicario Island (approximately 11 km from the southern end) where a maximum occurs in the phases of the tide constituents (Vidal and Tejedor, 2005). The tide in the Sancti Petri Channel is semidiurnal, with an amplitude of about 1.70 m in spring tides, and 0.65 in neap tides, and where the component of astronomical origin M2 is the most important, accounting for more than 85% of the total tidal energy (Vidal et al., 2002). The flow is mainly tidally driven with average values of 0.50 m/s. The velocities are greater at the ends than in the intermediate points of the channel. The lowest velocities occur in places where the maximum is located for the elevation phases.

Previous research has shown the dependence of the radionuclide concentration on grain size, magnetic susceptibility, composition or organic content (De Meijer et al., 1985, 1990; Elejalde et al., 1996). The concentration is also influenced by other factors dependent on the medium, like pH or redox potential (Fauré et al., 1996) and the physical or chemical state of the radionuclides (Cundy and Croudace, 1995). Previous studies show:

- Radionuclide concentrations increase when grain size of the materials decreases (He and Walling, 1996).

* Corresponding author. Tel.: +34(9)56016068.
E-mail address: rufino.ligero@uca.es (R.A. Ligero).

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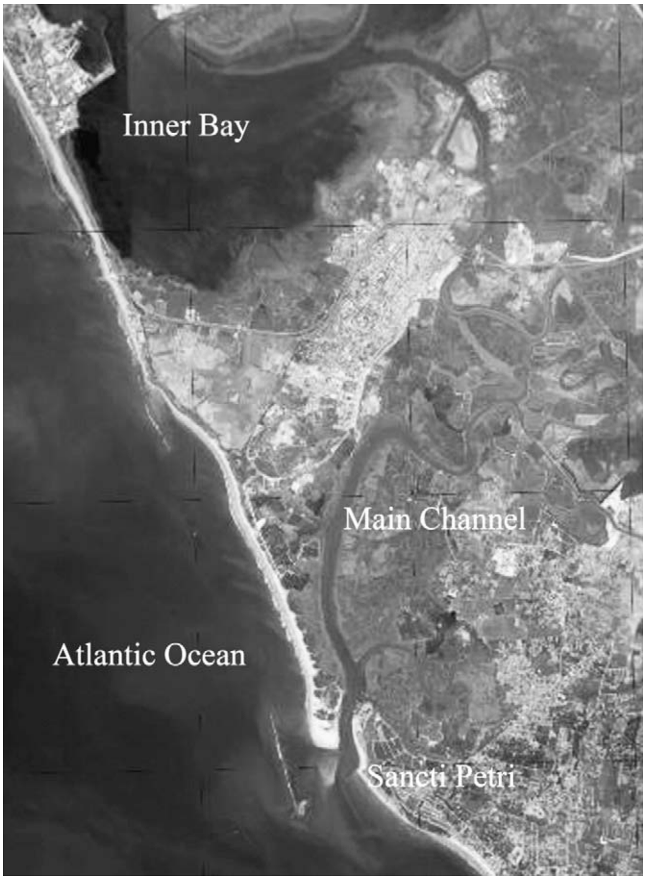

Fig. 1. "Bay of Cadiz" Natural Park.

Table 1
Geographic location of sampling points.

Simple number	Latitude (36N) min	Longitude (6W) min
1	29.938	11.085
2	29.808	10.177
3	29.689	8.975
4	28.928	10.932
5	26.406	11.965
6	29.317	10.837
7	29.251	9.926
8	26.027	12.468
9	25.538	12.709
10	25.015	12.802
11	23.946	12.539
12	23.376	12.679
13	22.877	12.518
14	22.966	13.046
15	22.921	12.761
16	27.500	10.650
17	23.833	11.184
18	24.345	11.916
19	24.988	11.969
20	24.563	12.733
21	23.648	12.104
22	26.905	12.135
23	25.983	11.796
24	26.034	11.123
25	26.542	11.046
26	26.037	9.988
27	27.993	11.230
28	28.410	10.830
29	28.131	8.935
30	27.437	10.092
31	27.577	10.724
32	27.005	10.510
33	27.431	11.095
34	27.262	9.771
35	26.544	10.088
36	27.954	9.914

- Higher activity concentrations are found in high density materials (Schuiling et al., 1985).
- Concentrations also depend on the composition of the sediments (Cho et al., 1996). Thus, U and Th are associated mainly with heavy minerals, while their concentrations in the light fraction of the materials are very low. This light fraction is generally quartz and feldspar and can contain high concentrations of K associated with the feldspar.

Multivariate analysis techniques were used to study the relationship between the activities of the different radionuclides and the measured sedimentological parameters (organic content, apparent density and sand/silt/clay content (Ligero et al., 2001)). The multivariate analysis known as analysis of variance (ANOVA) is a statistical technique used to explore the relationship between qualitative (non metric) variables called "factors" and quantitative (metric) variables.

The application of ANOVA to the seabed sediments of the Bay of Cadiz can be summarised as follows (being a, b and c, parameters derived from general correlation matrix):

(1) There is a high correlation between the activity concentrations of ^{226}Ra and ^{232}Th and there are no significant correlations between the activity of these radionuclides and other variables. Such a correlation in the entire Bay, including zones with different hydrodynamic conditions or geomorphological features, indicates that the concentrations of ^{226}Ra and ^{232}Th are representative of a common geological origin where these radionuclides are associated with a specific mineralogical component of the sediments,

$$A_{\mathrm{Ra}} = a + bA_{\mathrm{Th}} \tag{1}$$

(2) The apparent density is correlated only with ^{137}Cs activity concentration and organic content. Therefore these variables allow granulometric components within the mud (clay and silt, for example) to be distinguished,

$$A_{\mathrm{Cs}} = a + b\mathrm{OC} + c\rho \tag{2}$$

(3) The radionuclide ^{232}Th is related not only to ^{226}Ra, but also to ^{40}K and organic content, which confirms that there must be a mineralogical component associated with ^{40}K.

$$A_{\mathrm{K}} = a + b\mathrm{OC} + cA_{\mathrm{Th}} \tag{3}$$

(4) By applying a Discriminant Analysis, we found a function which allows us to determine the percentage of mud in the sediment using the activities of the radionuclides as experimental variables:

$$F = -0.048A_{\mathrm{Ra}} + 0.24A_{\mathrm{Th}} + 0.65A_{\mathrm{Cs}} - 0.0020A_{\mathrm{K}} - 3.6 \tag{4}$$

where A_{Ra}, A_{Th}, A_{K} and A_{Cs} are the ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs activities in Bq/kg, respectively. The function F classifies the sediment in one of these three groups: a) sand and muddy sand, b) sandy mud, c) mud. The positive values of F designate

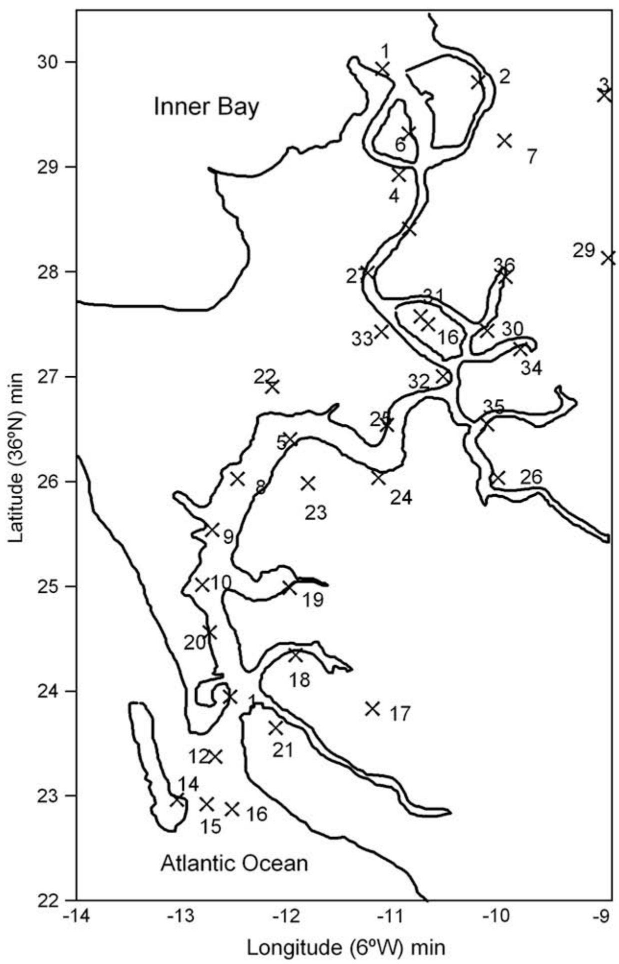

Fig. 2. Sampling stations.

sediments where the mud content is higher than 80% (muddy sediments), the values of F below -1.5 indicate that the sand fraction in the sediment is above 50% (sand and muddy sand sediments) and the values of F in the range $[-1.5, 0]$ designate sediments with a mud content between 50 and 80% (sandy mud sediments).

These results will be applied in this work to sediments of the samples collected from the Sancti Petri zone to determine if the same conclusions can be reached. If so, the consequences derived from this radiometric model will be compared with those deduced from the previously mentioned oceanographic study. They should be compatible if both models describe the same environmental space.

2. Experimental

To study the zone adequately, 36 sampling stations were distributed over the area covering the main channel, secondary channels, upper-tidal and inter-tidal areas, from the mouth in the Atlantic Ocean to the Inner Bay. However, if this distribution had been followed strictly, some stations would have been located in inaccessible points. In these cases, the positioning of the station in question was moved to the nearest accessible point. In Table 1 the geographical coordinates of the stations are given, and in Fig. 2 the location of each is indicated on a map of the zone, marked by a cross.

The sediment samples were dried in the laboratory at 60 °C, and milled using an agate mortar to a grain size of less than 0.25 mm, to ensure their homogeneity for the spectrometry measurements.

The organic carbon content, OC, was determined by applying a modification of the technique of Gaudette et al. (1974), developed by El-Rayis (1985); this involves the oxidation of the sediment sample with potassium dichromate in a strongly acid medium at a temperature of 135 °C. The OC content is obtained by measuring the surplus dichromate remaining after the oxidation, with ferrous ammonium sulphate. The percentage of OC detected by this method depends on the type of sediment, but is between 80 and 95% of the true total. In this technique, the elemental carbon remains unaltered and the carbonates do not present interference problems.

Table 2
Experimental data (activity concentration in Bq/kg, no data given for quantities below Minimum Detectable Activity).

Sample number	A_{Ra}	Δ_{Ra}	A_{Cs}	Δ_{Cs}	A_K	Δ_K	A_{Th}	Δ_{Th}	OC (%)	ρ (g/cm^3)
1	16.45	1.75	2.13	0.45	496.09	35.79	19.92	2.54	6.17	1.21
2	15.57	1.82	5.56	0.73	777.17	53.06	26.37	3.21	2.20	0.95
3	16.00	1.76	4.55	0.45	737.31	49.86	30.95	3.31	2.17	1.18
4	18.02	2.09	6.15	0.82	781.45	54.43	27.17	3.38	1.72	0.95
5	15.79	1.71	3.73	0.57	628.32	43.54	22.34	2.73	1.33	1.19
6	14.65	1.66	2.71	0.50	563.61	39.87	24.56	2.86	2.72	1.19
7	14.18	1.70	3.62	0.59	744.48	50.64	24.26	2.99	2.06	1.01
8	13.65	1.63	3.90	0.58	590.49	41.56	21.39	2.68	1.33	1.04
9	13.00	1.54	6.37	1.01	585.70	40.76	21.40	2.63	3.61	1.11
10	15.67	1.83	4.30	0.66	666.06	46.70	23.41	2.96	1.27	0.95
11	3.80	0.72	–	–	223.74	17.85	4.72	1.12	0.26	1.61
12	3.25	0.65	–	–	161.30	13.98	3.47	1.00	0.15	1.68
13	4.72	0.77	–	–	119.94	11.72	4.19	1.13	0.17	1.48
14	1.96	0.62	–	–	91.34	10.10	–	–	0.16	1.38
15	3.54	0.65	–	–	69.13	8.07	3.05	0.90	0.18	1.78
16	14.25	1.68	6.06	0.74	698.65	47.95	30.43	3.28	2.37	1.02
17	4.50	1.00			189.00	17.81	8.36	1.70	0.35	1.43
18	12.98	1.55	3.75	0.57	521.37	37.46	18.96	2.55	1.27	1.04
19	6.38	0.97	1.41	0.33	320.12	24.12	9.65	1.57	0.32	1.32
20	9.54	1.16	1.03	0.32	341.07	25.53	13.61	1.88	0.59	1.43
21	12.86	1.52	3.29	0.53	541.43	38.25	18.75	2.41	1.34	1.10
22	10.69	1.37	3.60	0.54	541.97	38.10	17.61	2.32	1.62	1.13
23	14.33	1.63	4.96	0.65	740.38	49.76	24.99	2.91	1.68	1.01
24	14.85	1.68	3.83	0.57	676.85	46.31	25.81	2.97	1.40	1.09
25	14.93	1.62	3.55	0.53	527.51	37.37	19.57	2.46	1.37	1.24
26	4.89	0.77	–	–	69.33	8.43	5.55	1.12	0.26	1.49
27	14.39	1.77	5.38	0.72	759.40	52.07	30.07	3.40	1.65	0.95
28	14.13	1.65	5.07	0.66	671.59	46.17	23.25	2.80	2.03	1.06
29	13.12	1.65	4.43	0.65	722.28	49.42	25.40	3.02	1.33	1.01
30	15.45	1.84	5.01	0.71	740.41	50.95	29.13	3.33	1.48	0.97
31	12.85	1.62	5.26	0.69	748.22	50.86	25.65	3.04	2.00	1.01
32	17.04	1.83	4.74	0.65	713.74	48.73	24.23	2.91	1.83	1.15
33	15.47	1.80	7.03	0.83	726.28	50.01	26.42	3.14	1.92	0.96
34	14.24	1.71	5.34	0.70	739.26	50.43	26.55	3.11	1.71	1.04
35	14.96	1.68	5.75	0.69	692.64	47.05	26.90	3.03	1.70	1.09
36	12.37	1.55	3.07	0.54	564.66	40.08	21.62	2.71	1.23	1.04

The rest of the sediment was put into cylindrical plastic containers of 4.6 cm diameter for analysis by gamma spectrometry. The sample bulk density was determined, after drying, by gravimetric procedures, and was found to be in the range of 0.95–1.43 g/cm^3.

The gamma radionuclides were measured by gamma spectrometry, employing a coaxial HP Ge detector with an active volume of 90 cm^3, a relative efficiency of 20% (in comparison with a NaI (Tl) detector of 3 in × 3 in) and a resolution of 2 keV at 1332 keV. This detector is sensitive in the 50 keV–10 MeV energy range. The sample holder of the measuring equipment utilises lead shielding 10 cm thick, with internal sheets of Cu and Cd (1 mm thick) to reduce external radiation. ^{137}Cs was determined using the 662 keV gamma emission peak (85% intensity) emitted by its descendent, ^{137}Ba^m, with which it is in secular equilibrium.

The efficiency was obtained experimentally by measuring a sediment standard of variable height (H) with emissions in the energy range (121.78 keV $\leq E \leq$ 1408.02 keV) usually found in environmental measurements (Ramos-Lerate et al., 1998). Applying a correction factor that takes into account the density dependent self-absorption of the samples (Cutshall et al., 1983), the efficiency function $\varepsilon(E, H, \rho)$ was obtained (Barrera et al., 1999). This efficiency is applicable in the range of measurement of the sediment samples, $E = 100$–2000 keV, $H = 1$–5 cm, $\rho = 0.95$–1.43 g/cm^3, and its uncertainty is 5%.

3. Results and discussion

Table 2 shows all the experimental data from the samples analysed. Using the Surfer Program (Golden Software®), and by the Kriging linear interpolation method, we have constructed contours for activity concentrations of the four gamma-emitting

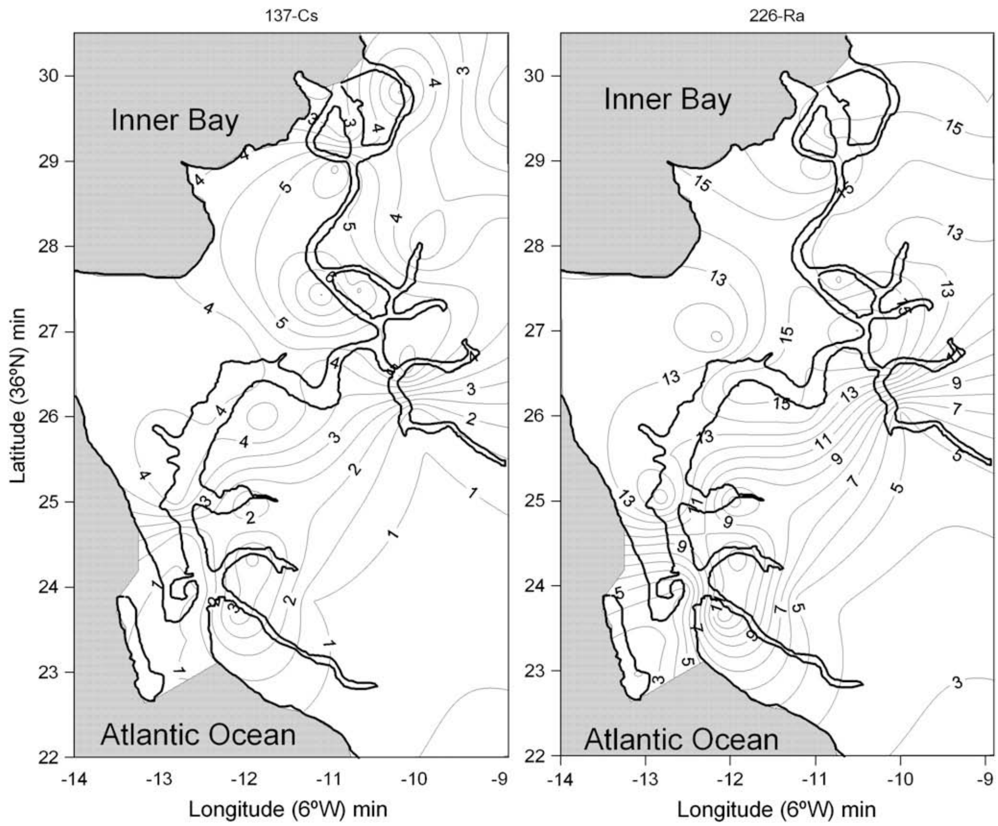

Fig. 3. Krigged contour lines for ^{226}Ra and ^{137}Cs.

radionuclides in sediments. The contour lines obtained for ^{226}Ra and ^{137}Cs are plotted in Fig. 3 as an example.

Comparing the values obtained for the activity concentrations of ^{226}Ra and ^{232}Th, a marked linear relationship (correlation coefficient of 0.93) can be seen between the two, in Fig. 4. This indicates that, in the zone of Sancti Petri, the mineralogical component is the same as that in the sediments of the bed of the Bay, which suggests that both possess a common origin and supports the result given by equation (1) for this zone.

Moreover the sediment of the zone of Sancti Petri is in general of the muddy type and, for this type of sediment, we find a relationship like that indicated by equation (2) where, for the submarine sediments of the Bay, the coefficients *a*, *b* and *c*, are found to be 3.22, 1.23 and −2.17, respectively. Since the supra-tidal zone of Sancti Petri is exposed to wind erosion, a factor that does not affect the sediments of the seabed, it is not expected that the same coefficients would be maintained in that zone. However, given the strong correlation existing between caesium, grain size (and therefore its apparent density) and the organic carbon content of the sediments, if our hypotheses are correct, the formal behaviour of these variables must be very similar to that established by equation (2). We have compared the values for the activity concentration of the ^{137}Cs measured experimentally with those calculated by equation (2), using the experimental values for OC

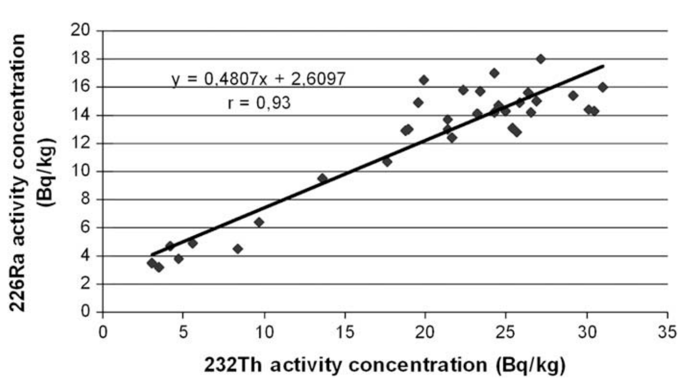

Fig. 4. Linear relationship between ^{226}Ra and ^{232}Th activity concentrations.

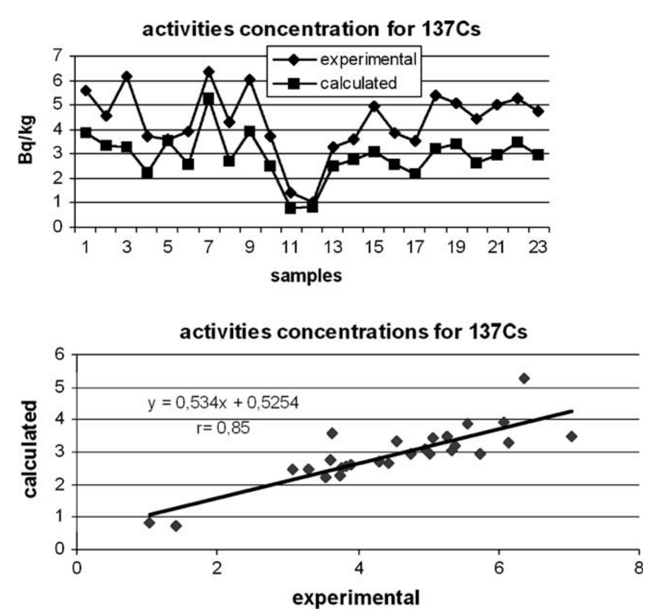

Fig. 5. Behaviour of the experimental and calculated activity concentrations of ^{137}Cs.

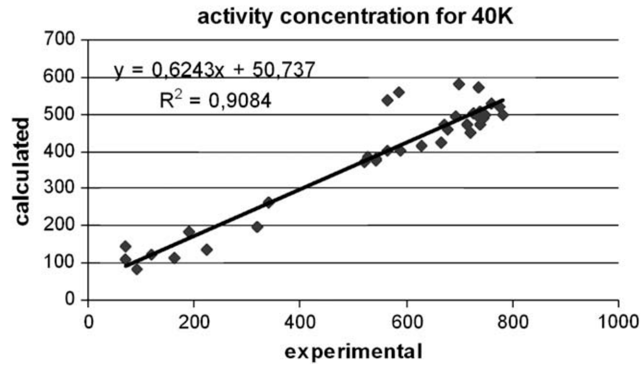

Fig. 6. Linear relationship between experimental activity concentrations of ^{40}K and activity concentrations calculated by equation (3).

and ρ: the results are shown in Fig. 5. It can be seen in Fig. 5 that both activities show the same qualitative behaviour, with a dual quadratic deviation of

$$\sigma = \sqrt{\frac{\sum\left(A_{\exp,i} - A_{\mathrm{cal},i}\right)^2}{n-1}} = 1.63 \quad (5)$$

and a linear correlation coefficient of 0.85; these values are sufficiently satisfactory to confirm our hypothesis in respect of the behaviour of these variables in the region of the Sancti Petri channel.

Further, the preliminary study had established that, although thorium and potassium could be considered elements linked to the mineralogy of the sediments, the existence in the Bay of Cádiz Natural Park of substantial salt extraction workings means that the presence of potassium may also be anthropogenic in origin, and is therefore connected to the organic matter present. In effect, it has been found that relationship (3) for the variables involved is verified also for the sediments of the zone of Sancti Petri, as can be observed in the correlation shown in Fig. 6.

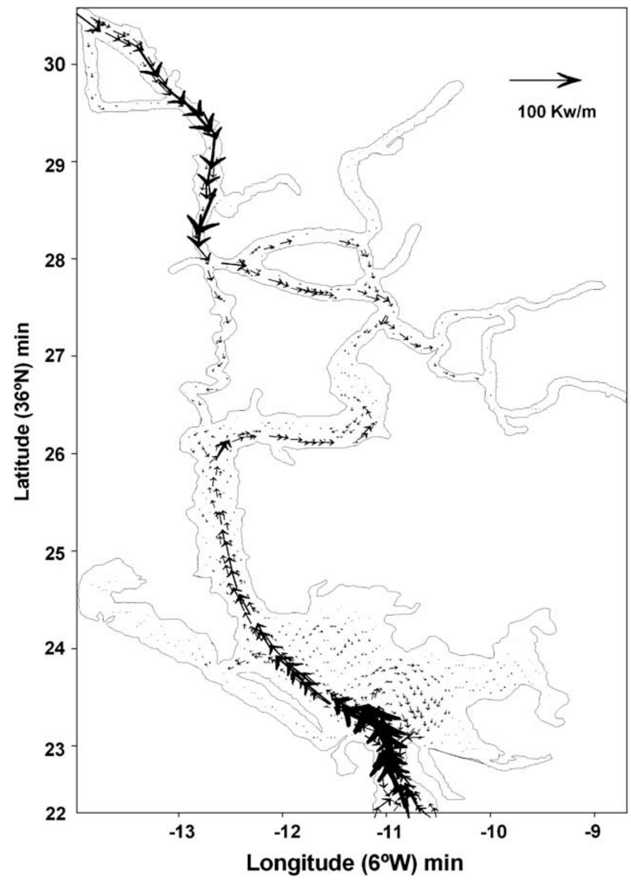

Fig. 8. Mean tidal energy flux per unit length (over a tidal cycle) for the main tidal constituent in the zone.

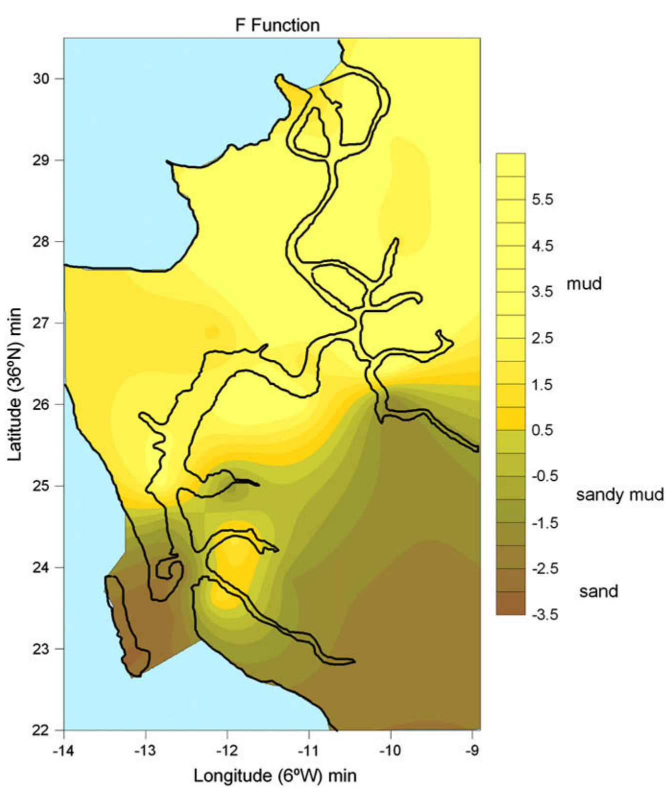

Fig. 7. Granulometric facies in the Natural Park, derived from discriminant function *F* defined by equation (4).

The granulometric facies obtained by applying the model described by equation (4) is represented in Fig. 7. The results show a granulometric variation along the length of the Sancti Petri channel, with three well-differentiated zones. In the mouth at the southern end are located sediments of the sandy and muddy sand types. In the central zone of the system of channels, adjacent to Isla Vicario, the sediments are of mud and sandy mud; and lastly, in the zone of the more northerly mouth, the sediments are sandy mud and muddy sands. This spatial distribution matches the results obtained by other authors (Vidal, 2002) based on the hydrological studies represented in Fig. 8, by means of the experimental tidal energy flux.

As already stated, there is a direct relationship between the morphology and the hydrodynamics of an estuary. One of the basic principles of marine geology is that the grain size of the seabed tends to decrease towards the areas of reduced physical energy. Since the estuary is formed by sedimentary material, whenever the water velocities are sufficient the material will be resuspended and transported in the same direction as the water flows. As the velocities decrease, the particles of larger grain size are deposited. Thus, the relationship between energy and fine sediment abundance can be observed by comparing Figs. 7 and 8. In the places where the energy flux is less, in the zone of Isla Vicario, the particle size of the sediment is smaller, giving sediments of the muddy type.

In contrast, in the zones near the channel mouths, where the energy flux is greater, larger grain sizes can be foreseen from these hydrological considerations. However, the zone of Sancti Petri (southern mouth) should be differentiated from the zone of La Carraca (northern mouth). In the southern mouth, the hydrodynamics of the channel produces higher water velocities (hence greater energy flux); these velocities, together with the presence of extensive sand beaches close by, conditions the presence of grain sizes typical of sandy sediment. This does not happen in the more northerly mouth, where the currents are less strong than at Sancti Petri, and hence the sediments of the Inner bay are of the sandy mud type.

A recent study (Tsabaris et al., 2007) has applied these models on the floor of the Butrint Lagoon and adjacent coast in the southwestern part of Albania. This approach predicts successfully the radionuclide content of the sediment, although the background radioactivity level is higher than that in the Bay of Cádiz.

4. Conclusions

The sedimentological models proposed, based on measurement of environmental radionuclides and developed in respect of more restricted areas, are valid for the whole of the Bay of Cádiz Natural Park, which suggests that it may be possible to extrapolate their results to other river and estuarine systems. In addition, the results provided by these models have been shown to be compatible with those derived from oceanographic models based on direct measurements, such as sea water levels and tidal currents rates.

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