

Relationships between sea-bed radionuclide activities and some sedimentological variables

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Abstract

Natural radionuclides (^{232}Th , ^{226}Ra , ^{40}K) and ^{137}Cs , coming from atmospheric radioactive fallout, have been measured in sea-bed sediments of the Bay of Cádiz (South Western Spain). In this report, multivariate analysis methods have been employed to study the relationships between the activities of the radionuclides and some sedimentological variables like granulometric facies, organic content and apparent density. The correlation functions found show that it is possible to determine, with a satisfactory degree of approximation, the granulometric facies of the sediments using only radiometric information. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Some investigations have shown the dependence of radionuclide concentrations on grain size, magnetic susceptibility, composition or organic content (Elejalde, Herranz, Romero, & Legarda, 1996; de Meijer et al., 1985; De Meijer, Lesscher, Schuiling, & Eldburg, 1990). Furthermore, the concentration is influenced by some other factors dependent on the medium, like pH or redox potential (Fauré, Sardin, & Vitorge, 1996) and the physical or chemical state of the radionuclides (Cundy & Croudace, 1995).

Previous studies show the following:

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

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- Higher activities appear in high-density materials (Schuiling, De Meijer, Riezebos, & Scholten, 1985).
- Concentrations also depend on the composition (Young-Hwan Cho, Chan-Ho Jeong, & Pil-Soo Hahn, 1996). So, the radionuclides 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.

In the process of mineral formation, the radionuclides are incorporated as trace elements in the crystal lattice. The concentrations of these elements depend on the type of mineral and the abundance in the parent magma. So, the activity depends on the mineral and the provenance. Later on and through erosive processes, these minerals are transported and can reach the coast or the marine environment becoming part of the sediments. Besides, there are some radionuclides incorporated in sediments through the aquatic medium experiencing anthropogenic influence, such as from phosphated fertilizers or other industrial wastes.

The radionuclides and trace elements which reach the marine environment can be retained by the sediments through the following processes:

1. Fixation on suspended matter and sedimentation.
2. Direct precipitation of colloidal forms.
3. Direct fixation by adsorption.
4. Deposition of organic waste which previously had incorporated the radionuclides.

From the above, it can be deduced that the interactions of the radionuclides with the biota or the suspended matter, which also depend on the chemical state of the element, play an important role in the fixation to sediments. Besides, in coastal sediments, where the organic content is higher, the organic fraction is very important for the binding and fixation of these elements to the sediment. The organic matter is important in the process of sea-bed-water-column transfer, due to the high number of functional groups of the organic molecules associated with the sediment (Alonso Santos & Díaz del Río, 1994).

In this paper, the relationships between the activities and sedimentological variables which could explain the concentrations of radionuclides in sediments are studied. The main objective is to derive sedimentological features from the values of radiometry, for this specific environment. Although in this study an HPGe detector has been used to obtain the most precise functions, the advantage of the method consists in utilising other types of detectors such as a portable gamma detector once the relationships have been established. So, instead of measuring many samples in the laboratory, radiometric maps can be obtained *in situ* and only a few samples are required for calibration (De Meijer, Tánzos, & Stapel, 1996). This paper is a new approach to a problem that has been addressed recently, namely the search for an empirical relationship to determine the granulometric facies of the sediments as a function of the radiometric variables (Venema, De Meijer, Van Os, & Gieske, 1999).

2. Zone of study and experimental methods

The Bay of Cádiz is located in the southwest of the Iberian Peninsula, and unfolds to the Gulf of Cádiz. It can be divided into two different parts (see Fig. 1): The Exterior Bay, located to the north of José León de Carranza Bridge, where there are important hydrodynamic conditions due to the strong tidal currents and the mouth of two rivers. The Interior Bay, to the south of this Bridge, is less exposed to the action of erosive agents. The sea-bed of the former is composed of sand and mud sediments, with an important Pliocene rock substratum and the sea-bed of the latter is composed of fine sediments, silt and clay, which allow the development of an exceptional biotope with an important ecological value (the zone was declared the Natural Park of Cádiz Bay from 1989).

The Maritime Bay, Exterior and Interior, was reticulated in a grid oriented according to parallels and meridians, with half a nautical mile long sides. There is a sampling station at every intersection point where sea-bed sediments were collected with a Van Veen grab. However, based on the granulometric facies map previously

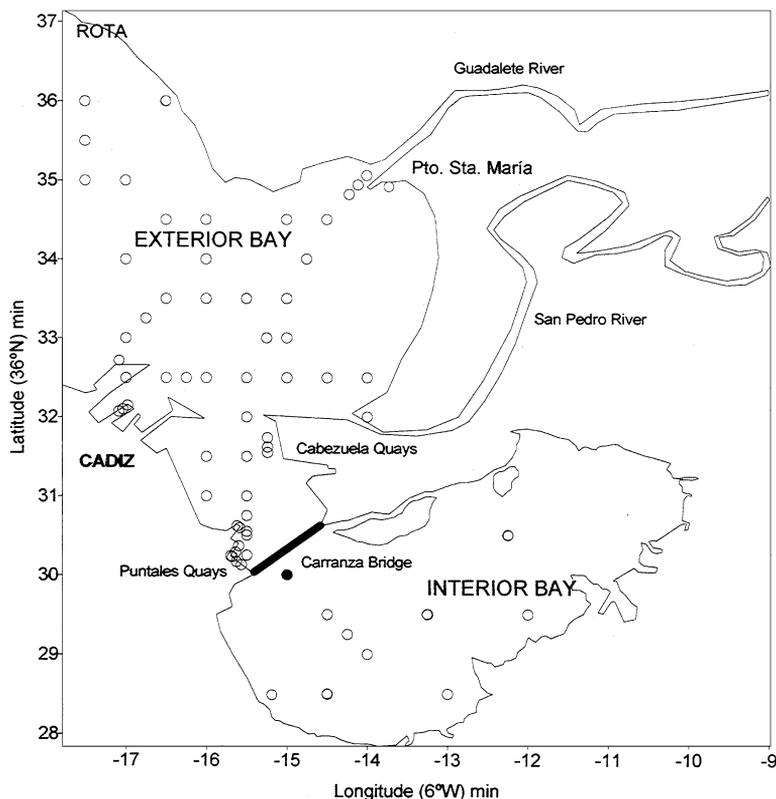


Fig. 1. Bay of Cádiz: Situation map and sampling stations.

determined (Parrado Román, 1997), the density of the grid was increased in zones where strong changes in granulometry could be found over shorter distances.

So, 70 samples representative of zones with different hydrodynamic and granulometric features, were collected through 1996 and 1997 (Fig. 1). From every sample, three aliquots were taken to measure the organic content by chemical procedures (Gaudette, Flight, Toner, & Folger, 1974; El-Rayis, 1985), the gamma-emitting radionuclides and the percentage of sand/mud.

The sediments collected from the sea-bed were dried at 95°C. Their water content and apparent density were determined by gravimetric and pycnometric methods, respectively. They were powdered and sieved to a grain size smaller than 0.5 mm to guarantee the homogeneity of the samples which were kept closed in cylindrical containers of 64 mm inner diameter filled to 55 mm height (Barrera, Ramos-Lerate, Ligeró, & Casas-Ruiz, 1999). The containers were kept sealed for a month before being subjected to gamma-spectrometry, in order to ensure the secular equilibrium between ^{222}Rn and ^{226}Ra . The activities of the radionuclides ^{226}Ra , ^{232}Th , ^{137}Cs and ^{40}K were measured.

The gamma radioactivity measurements were performed with a CANBERRA-supplied p-type coaxial HPGe detector system (Ramos-Lerate, Barrera, Ligeró, & Casas-Ruiz, 1997).

The activities of the sediments were measured in cylindrical geometry. The sample containers were placed at a distance of 3 mm from the detector window. The efficiency, with summing corrections, was determined (Ramos-Lerate et al., 1997; Ramos-Lerate, Barrera, Ligeró, & Casas-Ruiz, 1998), obtaining a function depending on the apparent density of the sediment and the energy of the gamma ray to be measured, with an uncertainty of 10% for a confidence level of 95%. In order to determine the background distribution, due to the existence of natural radionuclides in the environment, an empty polyethylene container, with the same geometry and the same measuring conditions as for the rest of the samples, was used.

3. Multivariate analysis

Multivariate analysis techniques have been used to study the relationships between the activities of the different radionuclides and the measured sedimentological parameters (organic content, apparent density and sand/silt/clay content).

The multivariate analysis known as analysis of variance (ANOVA) is a statistical technique used to explore the relationships between qualitative (non-metric) variables called 'factors' and quantitative (metric) variables.

When the concentrations (in Bq/kg) were measured, it could be appreciated that the stations situated in the Interior Bay showed higher activities, so that the factor "Type of Bay", Exterior (E) and Interior (I), was introduced in order to apply ANOVA.

The results of the analysis show that all the measured variables are clearly separated by the factor "Type of Bay" (Interior or Exterior), as observed in Table 1, in which the average values of every variable in each group are shown.

On the other hand, in order to study the influence of the granulometry on every measured variable (activity of the radionuclides, organic content and apparent density), the percentage of sand and mud (granulometric facies) was considered as a non-metric variable (groups: sand, S; muddy sand, MS; sandy mud, SM; and mud, M). The application of ANOVA to this new factor leads to the results shown in Table 2 and the average values shown in Table 3. The following can be deduced:

1. There are only significant differences in the ^{226}Ra and ^{232}Th contents between the groups sand and mud. The stations muddy sand and sandy mud can be included in any of the previous groups.
2. With respect to ^{137}Cs , ^{40}K and organic content, their concentration values in mud are significantly different from their values in the rest of the groups. Although in the muddy sand and sandy mud stations the average values are higher than in sand, the statistical uncertainty does not allow us to state that these groups are different from sand.
3. If we consider the values of apparent density, its differences are statistically significant for the four granulometric groups.

An analysis of Tables 2 and 3 shows that the observed differences between the Exterior and Interior Bays agree basically with a difference in the apparent density, or as is the same, with a different grain size, i.e. mud in the Interior Bay and muddy sand in the Exterior Bay.

The fact that all the measured variables can be classified according to the granulometric factor justifies the application of a statistical study to every homogeneous subgroup into which the stations can be divided.

Table 1
Average values calculated with the factor “Type of Bay”; 95% confidence level (E — Exterior Bay, I — Interior Bay, OC — organic content)

	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{137}Cs (Bq/kg)	^{40}K (Bq/kg)	OC (%)	Apparent density (g/cm ³)
E	9 ± 1	10.8 ± 1.4	0.9 ± 0.5	220 ± 30	0.8 ± 0.4	1.36 ± 0.10
I	14 ± 2	16 ± 2	4.6 ± 0.7	460 ± 50	3.0 ± 0.5	1.11 ± 0.06

Table 2
ANOVA with the factor “granulometry” (OC — organic content)

Group	^{226}Ra		^{232}Th		^{137}Cs		^{40}K		OC		Apparent density			
	1	2	1	2	1	2	1	2	1	2	1	2	3	4
Sand	X		X		X		X		X		X			
Muddy sand	X	X	X	X	X		X		X			X		
Sandy mud	X	X	X	X	X		X		X				X	
Mud		X		X		X		X		X				X

Table 3

Average values calculated with the factor “granulometry”; 95% confidence level (OC — organic content)

	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{137}Cs (Bq/kg)	^{40}K (Bq/kg)	OC (%)	Apparent density (g/cm ³)
Sand	6.8 ± 2.5	7.3 ± 2.7	0.5 ± 0.5	160 ± 60	0.2 ± 0.5	1.50 ± 0.10
Muddy sand	10.2 ± 3.0	12.2 ± 2.7	0.5 ± 1.0	200 ± 70	0.5 ± 0.7	1.36 ± 0.05
Sandy mud	10.6 ± 4.5	13.0 ± 5.0	0.7 ± 1.5	240 ± 100	0.7 ± 1.0	1.24 ± 0.06
Mud	13.4 ± 2.0	15.5 ± 2.0	4.0 ± 1.5	440 ± 40	2.9 ± 0.5	1.13 ± 0.05

With the objective of obtaining the relationships between the measured variables, a correlation analysis was carried out. In Tables 4–6, the correlation matrices for all the samples and the subgroups sand and mud, are shown, respectively.

In the subgroup sand, the ^{137}Cs concentrations are below the minimum detectable activity (0.5 Bq/kg) and, as shown in Table 5, there is only a significant correlation between ^{226}Ra and ^{232}Th activities ($r=0.9959$).

Later on, a factorial analysis was done with the aim of establishing the features of the sediments (called factors) as a function of the minimum number of measured variables.

Tables 7 and 8 show the weight of all the measured variables in every factor found in the factorial analysis for all of the samples and for the subgroup mud, respectively. The subgroup sand was not considered as caesium had an activity lower than the minimum detectable.

The general correlation analysis (Table 4) leads to the following results:

1. There is a high correlation between ^{226}Ra and ^{232}Th in the sediments of the entire Bay and there are no significant correlations between the activities of these radionuclides and the 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.
2. The radionuclides ^{137}Cs , ^{40}K and organic content are correlated.
3. The apparent density is negatively correlated with ^{137}Cs , ^{40}K and O.C., so that the sediments with a low apparent density, or equivalently with a higher content in mud, show higher concentrations of ^{137}Cs , ^{40}K and O.C.

The factorial analysis confirms the previous results. Two factors are obtained (Table 7). Factor 1 is determined mainly by the variables ^{137}Cs , ^{40}K , organic content and apparent density and defines “the granulometry” of the sediment. In Factor 2, the most important weights correspond to ^{232}Th and ^{226}Ra , so that this factor represents the mineralogy of the sediment.

If the results in Table 6, corresponding to the correlation analysis for muddy stations, are examined, the following can be stated:

Table 4

General correlation matrix. The significance level is shown inside parentheses (OC — organic content, ρ — apparent density)

	^{226}Ra	^{232}Th	^{137}Cs	^{40}K	OC	ρ
^{226}Ra	1.0000 (0.0000)	0.8684 (0.0000)	0.2418 (0.0437)	0.3043 (0.0104)	0.4384 (0.0021)	-0.3088 (0.0093)
^{232}Th	0.8684 (0.0000)	1.0000 (0.0000)	0.3604 (0.0022)	0.4284 (0.0002)	0.5523 (0.0001)	-0.3654 (0.0019)
^{137}Cs	0.2418 (0.0437)	0.3604 (0.0022)	1.0000 (0.0000)	0.8845 (0.0000)	0.9006 (0.0000)	-0.7123 (0.0000)
^{40}K	0.3043 (0.0104)	0.4284 (0.0002)	0.8845 (0.0000)	1.0000 (0.0000)	0.8271 (0.0000)	-0.7300 (0.0000)
OC	0.4384 (0.0021)	0.5523 (0.0001)	0.9006 (0.0000)	0.8271 (0.0000)	1.0000 (0.0000)	-0.7110 (0.0000)
ρ	-0.3088 (0.0093)	-0.3654 (0.0019)	-0.7123 (0.0000)	-0.7300 (0.0000)	-0.7110 (0.0000)	1.0000 (0.0000)

Table 5

Correlation matrix for sandy stations. The significance level is shown inside parentheses. The ^{137}Cs activity has not been included as it is below the minimum detectable activity (OC — organic content, ρ — apparent density)

	^{226}Ra	^{232}Th	^{40}K	OC	ρ
^{226}Ra	1.0000 (0.0000)	0.9959 (0.0003)	-0.7361 (0.1562)	-0.6023 (0.2824)	0.3283 (0.5897)
^{232}Th	0.9959 (0.0003)	1.0000 (0.0000)	-0.6965 (0.1913)	-0.5896 (0.2954)	0.2460 (0.6900)
^{40}K	-0.7361 (0.1562)	-0.6965 (0.1913)	1.0000 (0.0000)	0.1498 (0.8100)	-0.4638 (0.4314)
OC	-0.6023 (0.2824)	-0.5896 (0.2954)	0.1498 (0.8100)	1.0000 (0.0000)	-0.5352 (0.3526)
ρ	0.3283 (0.5897)	0.2460 (0.6900)	-0.4638 (0.4314)	-0.5352 (0.3526)	1.0000 (0.0000)

1. There is a correlation between ^{226}Ra and ^{232}Th , although the coefficient is lower than that in the general matrix. This can be explained by a transfer of the ^{226}Ra , the more soluble, from the sediment to the aquatic medium, which occurs more easily in finer sediments (Jurado Vargas, Vera Tomé, & Martín Sánchez, 1995).
2. The radionuclide ^{232}Th is related not only to ^{226}Ra , but also to ^{40}K , which confirms that there must be a mineralogical component associated with ^{40}K .
3. The ^{137}Cs and organic content are correlated, indicating the affinity of both variables for incorporation into the sediment.
4. The apparent density only shows correlations with ^{137}Cs and organic content. So, these variables allow us to distinguish granulometric components within the mud (clay and silt, for example).

Table 6

Correlation matrix for muddy stations. The significance level is shown inside parentheses (OC — organic content, ρ — apparent density)

	^{226}Ra	^{232}Th	^{137}Cs	^{40}K	OC	ρ
^{226}Ra	1.0000 (0.0000)	0.6955 (0.0120)	0.1661 (0.6059)	0.5115 (0.0892)	0.0280 (0.9312)	0.1116 (0.7300)
^{232}Th	0.6955 (0.0120)	1.0000 (0.0000)	0.1240 (0.7011)	0.8581 (0.0004)	-0.0258 (0.9366)	0.1328 (0.6808)
^{137}Cs	0.1661 (0.6059)	0.1240 (0.7011)	1.0000 (0.0000)	0.4229 (0.1708)	0.7289 (0.0072)	-0.5353 (0.0729)
^{40}K	0.5115 (0.0892)	0.8581 (0.0004)	0.4229 (0.1708)	1.0000 (0.0000)	0.4058 (0.1906)	-0.3383 (0.2821)
OC	0.0280 (0.9312)	-0.0258 (0.9366)	0.7289 (0.0072)	0.4058 (0.1906)	1.0000 (0.0000)	-0.6886 (0.0133)
ρ	0.1116 (0.7300)	0.1328 (0.6808)	-0.5353 (0.0729)	-0.3383 (0.2821)	-0.6886 (0.0133)	1.0000 (0.0000)

Table 7

Factorial analysis for all the stations: weights of every variable in the two factors

	Factor 1	Factor 2
^{226}Ra	0.23	0.93
^{232}Th	0.38	0.87
^{137}Cs	0.93	0.25
^{40}K	0.82	0.45
OC	0.93	0.22
ρ	-0.75	-0.42
Percent of variance	66	24

Table 8

Factorial Analysis for the muddy stations: weights of every variable in the two factors

	Factor 1	Factor 2
^{226}Ra	0.77	-0.23
^{232}Th	0.97	-0.12
^{137}Cs	0.21	0.87
^{40}K	0.89	0.21
OC	-0.10	0.87
ρ	0.37	-0.68
Percent of variance	47	34

The factorial analysis for the subgroup mud (Table 8) provides two factors slightly different from the general case. The first factor, dependent on the concentrations of the radionuclides ^{226}Ra , ^{232}Th and ^{40}K (not the apparent density), represents the “mineralogy” of the mud. The second factor, a function of the ^{137}Cs concentration, the organic content and apparent density, represents the “granulometry” of the sediment.

4. Discriminant functions analysis

The discriminant functions analysis allows us to show that using only the apparent density and organic content it is possible to distinguish between the clay and silt groups within the muddy sediments.

This multivariate technique provides lineal functions of experimental variables which allow classification of new samples into the appropriate groups.

We used, like a classificatory variable, the kind of mud (clay or silt) and, like experimental variables, the apparent density and the organic content. Based on previous studies in the Bay of Cádiz (Achab et al., 1999), a clay nature was assigned to the mud in the Interior Bay and a silt nature to the mud in the Exterior Bay, finding the following discriminant function:

$$H = -25 + 0.33 \times OC + 22\rho, \quad (1)$$

where OC is the organic content in per cent and ρ is the apparent density in g/cm^3 . The significance level of this function is 0.05 and the canonical correlation is 0.64.

The results shown allow us to state that, by only measuring the apparent density and the organic content of the sediments, and using Eq. (1), it is possible to distinguish between the clay or silt predominance inside the muddy sediments (clay if H is below 0.20 and silt if H is above) which represents an important advantage over the traditional granulometric methods.

Also, from the previous multivariate analysis, one can deduce a possible granulometric classification of the sediment as a function of its content of radionuclides. Table 9 shows the activities found in the sand, silt and clay fractions in the Bay of Cádiz, which are called “fingerprints” by some authors (De Meijer & Donahue, 1995).

Through a discriminant analysis, we found a function which allows the determination of the percentage of mud in the sediment using the activities of the radionuclides as experimental variables. Seven sandy stations and 12 muddy stations were used in the analysis, finding the function

$$F = -0.048A_{\text{Ra}} + 0.24A_{\text{Th}} + 0.65A_{\text{Cs}} - 0.0020A_{\text{K}} - 3.6, \quad (2)$$

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 significance level is 0.00012 and the canonical correlation is 0.89.

Table 9
Activity concentrations in the fractions sand, silt and clay

Fraction	Grain size	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	^{137}Cs (Bq/kg)
Sand	$63 \mu\text{m} < x < 2 \text{mm}$	6.8 ± 2.5	7.3 ± 2.7	160 ± 60	0.5 ± 0.5
Silt	$16 \mu\text{m} < x < 63 \mu\text{m}$	13.4 ± 2.0	15.5 ± 2.0	360 ± 80	3.3 ± 1.0
Clay	$x < 16 \mu\text{m}$	13.4 ± 2.0	15.5 ± 2.0	490 ± 120	4.5 ± 1.8

The function F was applied later to all the samples, classifying the sediment in one of these three groups: (a) sand and muddy sand, (b) sandy mud, and (c) mud. The positive values of F designate sediments where the mud content is higher than 80% (mud 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).

Using the activities measured in the Bay and Eq. (2), it is possible to evaluate the function F in the zone.

With the aim of obtaining a better resolution, the primitive experimental grid was extended using a well-known mathematical interpolation procedure whose efficiency has been proved in the literature (Burgess & Webster, 1980).

The distributions of the three kinds of sediments (sand, sandy mud and mud) that the function F allows us to discriminate are shown in Fig. 2.

Comparing these results with the granulometric facies studies performed in this area using traditional techniques (Achab et al., 1999), we find a good agreement between them. In particular:

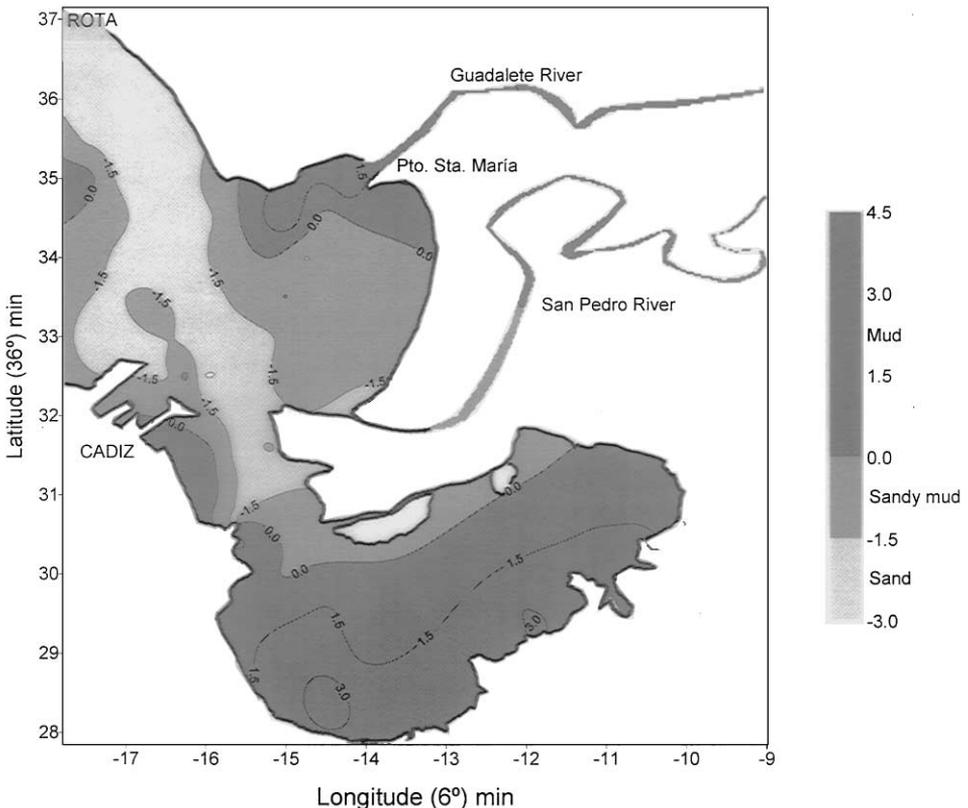


Fig. 2. Representation of the percentage of mud in the sediments obtained from the F function.

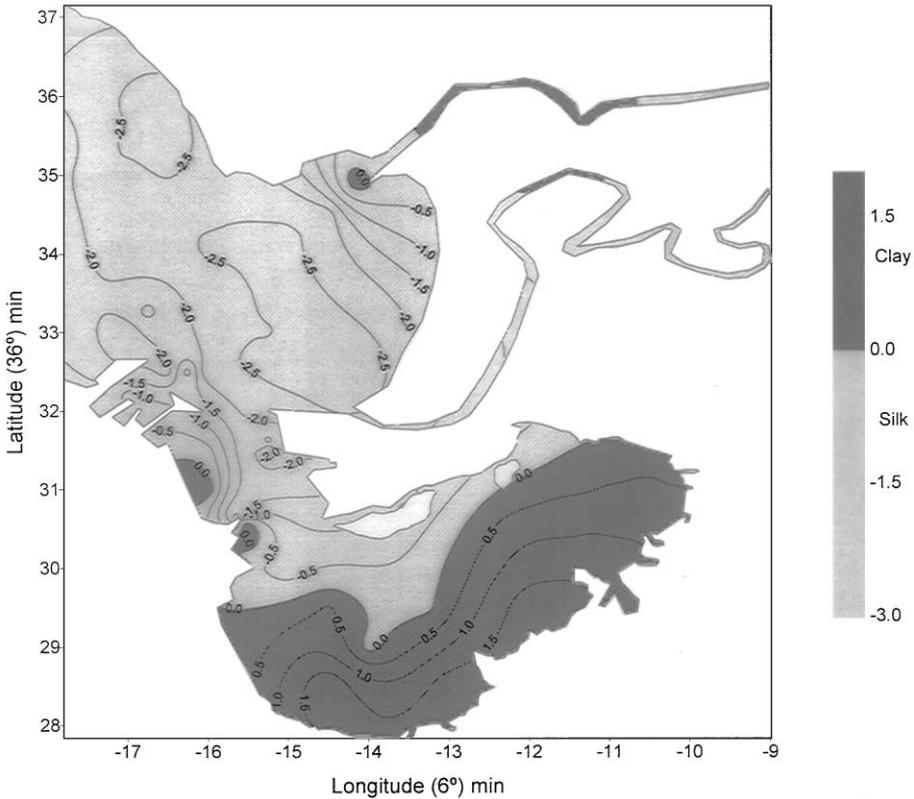


Fig. 3. Representation of the clay and silt predominance in the mud obtained from the G function.

- A muddy nature is confirmed for the whole Interior Bay (whose grain size increases in the north), for the Guadalete river mouth and for the west part of the Exterior Bay to the harbour docks of the city of Cádiz (a consequence of the dispersion to the west produced by the intertidal current prevalent in the Strait of Puntales which separates both Bays, flowing parallel to it). The radiological method also detects the muddy shoal situated between Cádiz and Rota.
- In the Exterior Bay there is a sandy central zone from Cabezuela to Rota. This sandy zone is connected with the mud through the lateral regions of sandy mud.

As the dependence of the ^{40}K and ^{137}Cs activities on the clay and silt contents has been shown, a discriminant analysis was used to find a function which determines the clay or silt predominance in the mud, obtaining

$$G = -3.5 + 0.0054A_K + 0.32A_{Cs} \quad (3)$$

with a significance level of 0.04 and a canonical correlation of 0.77, and where values $G > 0$ correspond to clay sediments and $G < 0$ to silt sediments.

Although the function G must be applied in a strict sense to muddy zones to discriminate clay or silt, it is interesting to evaluate it in the studied zone with the data from the entire Bay. Fig. 3 shows the results and it can be stated that

- (a) Most of the mud in the Interior Bay is clay.
- (b) Most of the mud in the Exterior Bay is silt, including the muddy zone between Cádiz and Rota.
- (c) The marginal fraction of the mud in the Exterior Bay, including the part contained in the sandy sediments, is also silt.

5. Conclusions

The radiometric method is an efficient tool for geological studies in a region, with the aim of determining granulometric facies in sediments.

The present possibility to monitor the sea-bed sediments with high-efficiency gamma detectors allows us to convert in real time the radiological map into a granulometric facies map, with an important saving of resources and time compared to the traditional techniques.

The functions F and G determine, with a good degree of approximation, the percentage of mud and the clay/silt characteristics of the mud, respectively, in the Bay of Cádiz. Although it is not possible to state that the same correlation functions can be applied to other geographic zones, it is reasonable that there is an analogous behaviour in other estuarine coastal zones with a common geological origin. The application of the radiometric techniques in such places requires a previous calibration using representative stations in these new zones.

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