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Levels of ^{137}Cs in muddy sediments on the seabed in the Bay of Cádiz (Spain). Part II. Model of vertical migration of ^{137}Cs

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Abstract

This second part of the study reports the development of a model to describe the vertical migration of the artificial radioisotope ^{137}Cs in the sediment column on the seabed of the Bay of Cádiz. The application of the model provides an overall picture of the process of sedimentation in the Inner Bay of Cádiz. The spatial distribution of the rate of sedimentation enables us to study the sources of sediments and the means by which the sediments have been transported. A method has been derived from the rate of sedimentation to perform the dating of the layers of sediment. The model describes the behaviour of ^{137}Cs in the area under study, taking into account the time of residence in the zones that are the source of accumulation, the origin of the sedimentary material, together with the diffusion of the radionuclide in the sediment of the seabed.

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Keywords: Seabed sediment; Migration model; Radionuclide; Dating; Sedimentation rate; ^{137}Cs

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1. Introduction

As a consequence of thermonuclear weapons testing, ^{137}Cs began to be detectable on a global scale from 1954 onward. The intensification of nuclear tests in 1958–59 produced an initial peak of activity, and in 1963 there was a second peak that was responsible for the greater part of the activity of ^{137}Cs found in the natural environment. The accident at the Chernobyl nuclear power station in 1986 released into the atmosphere a large quantity of fission products, and this release has been employed as a radioactive marker in the northern hemisphere (Callaway et al., 1996). The dating of sediments by means of ^{137}Cs can be performed by identifying peaks in the profile of activity and then assigning the corresponding historical dates to the peaks. However, the shape of the profiles of activity against depth that were determined for the Bay of Cádiz in the first part of this study has shown that it is not possible to apply this technique directly. The behaviour of ^{137}Cs in this system does not allow clearly defined peaks for these activity profiles to be identified. On the one hand, the tidal dynamic leads to a relatively dispersed deposition of the radionuclide in these sediments, in comparison with the accumulation that takes place in lakes or glaciers. On the other hand, from such dispersed profiles, it can be stated that this radionuclide presents a vertical mobility within the body of the sediment column. The presence of ^{137}Cs in deep sediments is indicative of the diffusion of the radionuclide from the superficial layers, of greater activity, towards the deeper layers. The diffusive flux of ^{137}Cs in sediments is a phenomenon produced in consequence of the combined action of various processes of transport (Robbins and Edgington, 1975; Robbins, 1990); notable among these processes is the molecular diffusion of the radionuclide in the interstitial water of the sediment, while another is the physical movement of particles of smaller size (< 1 μm) that have already incorporated ^{137}Cs in earlier times.

The most important factor determining the shape of the ^{137}Cs profile is the temporal evolution of the activity flux at the water-sediment interface. This flux, in general, does not follow precisely the temporal pattern associated with changes in the concentration of ^{137}Cs in the atmosphere. The high solubility of caesium in the aquatic medium leads to the transfer of the radionuclide to the sediment being delayed with respect to the evolution of the radioactive fallout from the atmosphere. In the profiles of ^{137}Cs obtained in the Bay of Cádiz, this time-lag effect translates into a zone of approximately constant activity being present; this zone extends from the surface down to the depth at which the activity of the radionuclide begins to decrease very sharply. The interpretation of the profiles of ^{137}Cs therefore requires the development of a mathematical model that represents the physical phenomena participating in the process of incorporation and subsequent mobility of the radionuclide in the sediment.

The model that is proposed attributes the mobility of ^{137}Cs in the sediment to the combined effect of diffusion and advective flow (vertical transport generated by the sedimentation on the interface). Hence, the evolution of the activity in this type of system is determined by an equation of diffusion–advection for a substance subject to radioactive decay. The choice of the model employed is conditioned by the available experimental information. Since the profiles of activity have been measured at only one point in time, subsequent to the deposition of activity, the model only permits the average rate of sedimentation and the average coefficient of diffusion to be determined; for this reason both magnitudes are considered to be independent of depth, to simplify the fitting parameters, and to avoid the problems of over-parameterisation presented by other more complex models (Beck and Arnold, 1977).

2. Development of the model

To describe the profile of activity of ^{137}Cs, let us consider, for a generic sediment column, a volume V representative of the column, limited by two horizontal surfaces, at depths a and b. The activity flux takes place across both surfaces and is attributable to two components:

Advective flow J_A. The transport of material in the direction z produces an activity flux $J_A = v\tilde{a}$, where $\tilde{a}$ is the activity by unit of volume and v is the rate of sedimentation (L T^{-1}). In general, the rate of sedimentation v will depend on the depth due to compaction. However, as the presence of ^{137}Cs is limited to the more superficial strata, the effect of compaction can be discounted. The rate of sedimentation can also vary with time, if the rate of supply of material to the interface changes, but considering an interval of time longer than the time scale associated with the phenomenon that produces the sedimentation, as occurs when the supply of material is related to the tides, this dependence can be ignored. In consequence, the rate of sedimentation can be taken as constant, representing the mean value of the rate for the depths of column considered and in the period of time studied.

Diffusive flux J_D. The presence of a fraction of activity dissolved in the interstitial water permits the molecular diffusion of the radionuclide in any direction in the body of the sedimentary medium. In addition, the brownian movement of solid particles of microscopic size, also constitutes a means of transport for the material, and therefore for the activity, which is similar in effect to molecular diffusion. In general, diffusive transport, encompassing all the phenomena that give rise to it, produces a net flux in the direction of the gradient of activity, the direction of which is from zones of greater activity to those of less activity. Therefore, in our case, this gives $J_\mathrm{D} = -D(\delta a/\delta z)$, where D ($\mathrm{L}^2\ \mathrm{T}^{-1}$) is the constant of diffusion that encompasses all the processes capable of producing diffusive transport. The biological activity generated by benthic organisms (which, in the case we are concerned with, consist of species of annelids, crustaceans and molluscs), together with the deposition of marine vegetation at the water-sediment interface, produces an alteration of the sediment stratification that occurs preferentially in the more superficial layers. The effect that this surface bioturbation produces is known traditionally as layer mixing, and consists of the total or partial homogenisation of the activity of the radionuclides in a layer of determined thickness, and a rapid reduction of this homogenisation at greater depth. The effect of layer mixing has been modelled by several authors (Robbins et al., 1977), assuming that in this layer the value of the coefficient of diffusion D is constant. In addition, the coefficient of diffusion D is independent of the time when the exchange of the radionuclide between the phases present in the sedimentary medium, regulated by the coefficient of distribution k_D, reaches thermodynamic equilibrium (Kirchner, 1998). ^{137}Cs undergoes a slow but progressive "fixation" by clay minerals in soils and sediments (Chamard et al., 1993), and for this reason the diffusion of this element may diminish with time in clay-type sediments. Hence, in the case where the fraction of the element that undergoes the settling is limited, or in the case where the time of relaxation of this process is sufficiently long, in comparison with the time during which the element remains in the sediment, it is also possible to consider a constant coefficient of diffusion, D.

Taking into account both the advective and diffusive flows, and the decay of the radionuclide, the evolution of the activity, within the control volume considered, will be given by the equation

$$\frac{\partial \tilde{a}}{\partial t}=\frac{\partial}{\partial z}\left(D\frac{\partial \tilde{a}}{\partial z}\right)-\frac{\partial}{\partial z}(v\tilde{a})-\lambda\tilde{a} \tag{1}$$

that, with the hypotheses of constancy for v and D, previously explained, can be simplified to

$$\frac{\partial \tilde{a}}{\partial t}=D\frac{\partial^2 \tilde{a}}{\partial z^2}-v\frac{\partial \tilde{a}}{\partial z}-\lambda\tilde{a} \tag{2}$$

This equation describing the evolution of the activity in a sediment column, in absence of another condition lacks analytical solutions; therefore we propose its resolution by using a method of finite differences.

The unknown function $\tilde{a}(z,t)$ is defined in a spatial and temporal domain $z\geq 0, t\geq 0$. Generating a network of discrete values for $\tilde{a}$ in this domain, defining $z_p=p\delta z, t_q=q\delta t$, where δz and δt represent, respectively, the spatial and temporal interval of the network, and $p,q\geq 0$ are integers that provide the position in this network. Utilising the simplified notation $\tilde{a}(z_p t_q)=\tilde{a}_p^q$, and writing the partial derivatives in discretised form (Haberman, 1987), gives

$$\tilde{a}_p^{q+1}=\tilde{a}_p^q(1-\lambda\delta t)+D\frac{\delta^2 t}{\delta z^2}(\tilde{a}_{p+1}^q-2\tilde{a}_p^q+\tilde{a}_{p-1}^q)-v\frac{\delta t}{\delta z}(\tilde{a}_p^q-\tilde{a}_{p-1}^q) \tag{3}$$

This expression describes the temporal evolution of the activity, providing the value of $\tilde{a}$ at an instant of time t_{q+1} in function of the value of $\tilde{a}$ in a previous time t_q. The activity at the depth z_p at the instant of time t_{q+1} is determined in function of the activity in the instant of previous time t_q, at the depths z_{p+1}, z_p, z_{p-1}.

Since boundary conditions are established as the activity in $z=0$, $\tilde{a}_{p=0}^q=c(t)$, which represents the phenomenon of interaction of the interface with the aquatic medium, and the disappearance of the activity from a sufficiently great depth, $\tilde{a}(z\to\infty, t)=0$. At the same time, solution of eq. (2) requires knowledge of the initial profile of activity, $\tilde{a}(z_p, t=0)$. Considering the initial point in time $t=0$ as the moment when the activity of ^{137}Cs begins to reach the sediment, this assumes, given the artificial character of this radionuclide, the previous absence of this radionuclide, and therefore the initial condition $\tilde{a}_p^{q=0}=0$.

In this way, the determination of the profile of activity at a point in time t is obtained by applying the expression in an iterative process (3). From the initial profile, $t=0$, the profile at a subsequent point in time t_1 is deduced, and thence successively up to $t_N=t$ (N=number of iterations).

The first boundary condition, $c(t)$, represents the phenomenon of the incorporation of the radionuclide from the aquatic medium to the sediment column. Given that the activity of ^{137}Cs measured in the sediments of the Bay of Cádiz is very small, it is possible to accept that its presence originated in the peak activity of the 1960s, and the year 1963 will be accepted as the initial date ($t=0$), which implies that the date when the samples were extracted (1998) corresponds to $t=35$ years.

A first hypothesis would consist of assuming that the arrival of the activity at the interface takes place a short time after its fall-out over the hydrosphere. In a relatively short period of time, the ^{137}Cs would be absorbed by the matter in suspension present in the water column, and would subsequently be deposited on the water-sediment interface, or else the dissolved radionuclide would pass directly to this interface. The subsequent sedimentation of inactive material would result in a profile with zero or very low values of ^{137}Cs at the interface and a maximum value at a particular depth. However, as has been seen in the first part of this study, the profiles found in the Bay of Cádiz do not present this form, therefore the model of rapid incorporation “post-fallout“ is not applicable. The experimental profiles obtained show relatively high activity values from the surface down to a certain depth, which suggests that the supply of sediment originates from zones, that we shall designate source zones F, where ^{137}Cs has previously accumulated. The erosion of these zones, transport and subsequent deposition on the seabed explain how the activity of the ^{137}Cs can be incorporated in the sediment column continuously over a relatively long period of time. We are therefore dealing with a mechanism of “slow” incorporation, in which the evolution undergone by the activity at the interface, $c(t)$, is the consequence of the variation of the activity source, A^F, itself. This activity decreases with time, not only due to the radioactive decay of the radionuclide ($T_{1/2}$) but also to the loss of activity in the sediment in consequence of its residence time in the source zone ($T_{1/2}^S$). The global evolution of the activity is, therefore, an exponential decay with a time characteristic $T_{1/2}^T$, that is produced as the combination of both effects, complying with

$$T_{1/2}^T=\frac{T_{1/2}T_{1/2}^S}{T_{1/2}+T_{1/2}^S} \tag{4}$$

If $T_{1/2}^S \gg T_{1/2}$, the activity of the source evolves with the half-life, $T_{1/2}^T=T_{1/2}$, since a long residence time causes a loss of activity by elimination that is negligible against the radioactive decay. On the contrary, if $T_{1/2}^S \ll T_{1/2}$, the total evolution of the activity will be determined by this time of residence. $T_{1/2}^T=T_{1/2}^S \ll T_{1/2}$.

Given that the activity $c(t)$ at the interface of the sediment follows an evolution similar to that undergone by the activity of the source zone, A^F, we propose

$$c(t) = c_o e^{-\frac{\ln 2}{T^T_{1/2}} t} \tag{5}$$

It should be observed that the deposition of ^{137}Cs in consequence of the Chernobyl accident has not been taken into account; this is because, according to experimental measurements, the activity corresponding to the year 1986 is insignificant in comparison with the fallout deposited in the 1960s.

Equation (2) considers the activity by unit of volume, $\tilde{a}$, instead of the activity by unit of mass, a, since the diffusive flux of activity is produced by the spatial gradient of $\tilde{a}$. However, the activity of radionuclides is generally expressed by unit of mass, a (Bq/kg). If the volume V contains a mass of sediment m, $A = \tilde{a}V = am$, from which $\tilde{a} = \rho a$, with ρ being the bulk density of sediment in the layer. It is only possible to express eq. (2) in terms of activity by unit of mass a in the special case where the density of sediment is constant.

In the columns of sediment studied, the variations of ρ with depth are moderate, as can be seen in Fig. 1. The introduction of $\rho(z)$ transforms the profile of activity by unit of mass into the corresponding profile of activity by unit of volume, $\tilde{a}(z) = \rho(z)a(z)$; this produces a distortion of the profile of activity by unit of mass, which is seen to be relatively deformed in the layers of higher density (Fig. 2). For this reason, eq. (2) is solved in terms of activity by unit of volume. The direct application of this equation in terms of activity by unit of mass would lead to erroneous values for the parameters of the model.

The determination of the parameters $v, D, c_o, T^T_{1/2}$ associated with each measured profile of ^{137}Cs has been carried out by minimising the quadratic difference, χ^2, between the theoretical and experimental functions, as shown in schematic form in the diagram of Fig. 3.

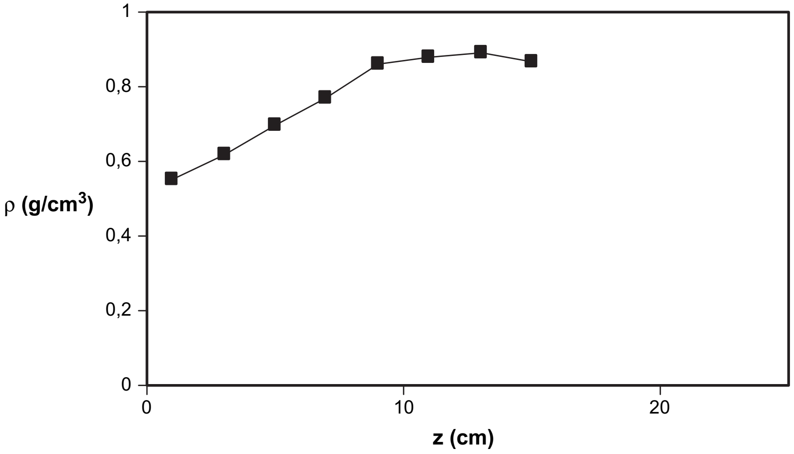

Fig. 1. Density against depth for the sediments.

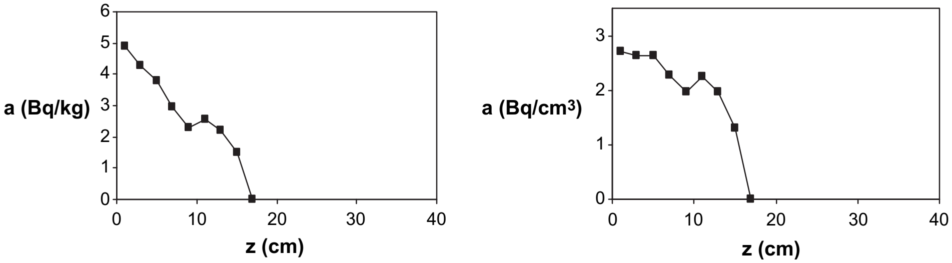

Fig. 2. Profiles of activity by unit of mass, and of activity by unit of volume, against depth.

3. Application of the model

Table 1 shows the parameters $v, D, c_o, T^T_{1/2}$ obtained at each experimental sampling station of the Inner Bay of Cádiz by applying the migration model proposed. Fig. 4 presents, by way of example, the fits between the theoretical function provided by the model and the experimental data for some of the sediment columns measured, where a good correlation between theoretical and experimental activities by unit of volume is shown.

Fig. 5 shows the spatial distribution of the rates of sedimentation, derived from the model, in the Inner Bay of Cádiz. It can be observed that most sedimentation takes place in the eastern zone, close to the outlet of the Sancti Petri Channel, where rates of more than 1 cm/year are reached. From the rate of sedimentation v, it is possible to determine the *rate of mass sedimentation* v_{m}, defined as the quantity of sediment accumulated per unit of surface and time, $v_m = \rho v$, with ρ being the density of the sediment. The spatial distribution of sedimentation in the Inner Bay determined using the model is in agreement with the distribution of the inventory of ^{137}Cs obtained in the first part of this study, which reveals the correlation existing between inventory and sedimentation: the global accumulation of the radio element ^{137}Cs occurs as a consequence of the sedimentation of material that has previously incorporated radiocaesium.

Further, the spatial distribution of the rate of sedimentation identifies the possible sources of supply of sedimentary material. The sedimentation in the eastern zone of the Bay is constituted by a supply of suspended matter originating in the zone of salt marshes that is therefore undergoing a net process of erosion. In conditions of ebb tide, the sediment is transported by the network of channels running through the marsh, removed in suspension along the Sancti Petri channel, and later deposited in the proximities of the channel's outlet. The strong tidal currents in the navigation channel transport the sediment in a westerly direction; as can be appreciated in Fig. 5, this produces a "tongue" of sedimentation that extends towards the west.

The sedimentation in the western part of the Bay must be associated with the transport of matter in suspension caused by the entry of the masses of water existing in this zone. This takes place through the Puntales strait in the flow tide, while in the ebb tide, the transport is by the River Arillo that constitutes the artery into which

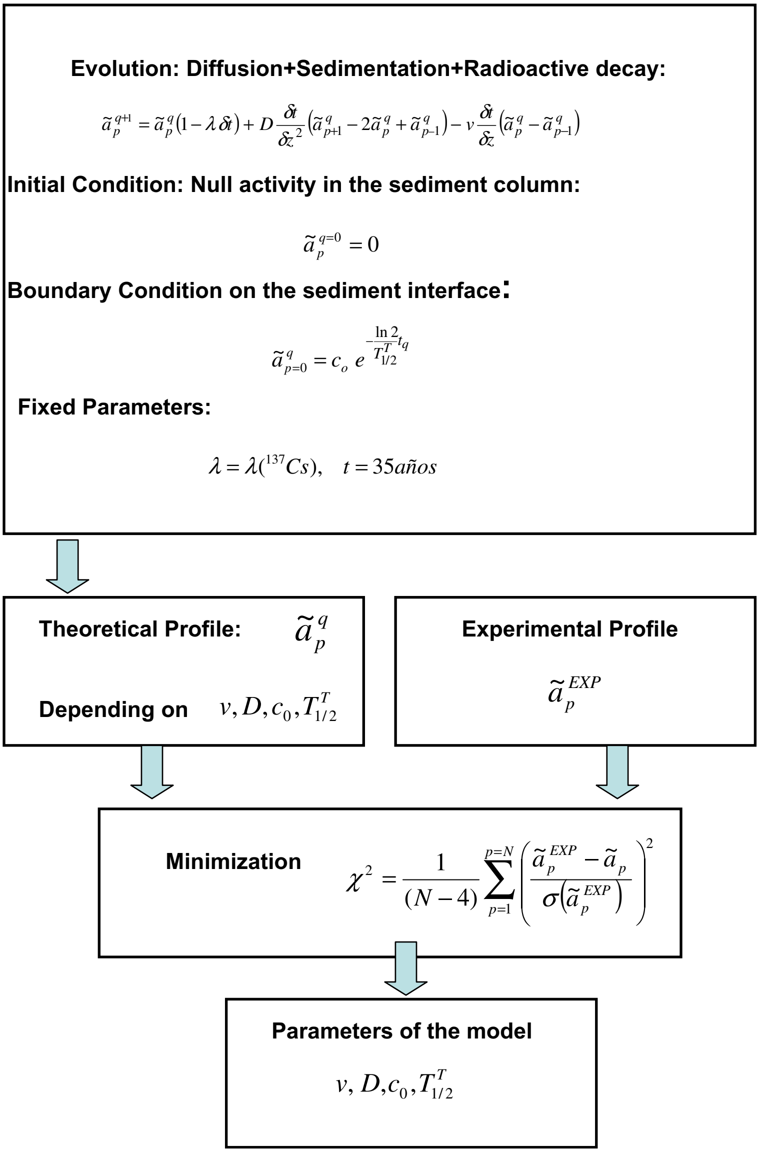

Fig. 3. Diagram of the algorithm for determining the parameters of the model.

flow the interlinking channels of the marshes extending to the south of the Bay. The erosion of the marshes and the subsequent transport of the sediment towards the north, generated by the tidal currents in the ebb tide, seem to be the factors responsible for the sedimentation in the south of the Bay. The peak of sedimentation appearing in this central-western zone coincides with the clockwise vortex of currents

Table 1
Parameters of the model for all sampling stations

Station	v (cm/year)	D (cm^2/year)	c_o (Bq/cm^3)	$T_{1/2}^{T}$ (years)
R7	0.32	0.73	5.0	30.2
S8	0.23	0.11	4.9	22.3
T7	0.15	0.27	6.3	30.2
T9	0.49	0.18	5.9	21.8
U2	0.26	0.25	5.4	30.2
U4	0.28	0.12	5.1	30.2
U6	0.46	0.23	6.1	27.5
U8	1.58	0.27	11.9	15.4
V1	0.45	0.11	5.4	30.2
V3	0.85	0.43	10.7	15.0
V5	0.24	0.16	4.9	30.2
V7	0.38	0.05	10.2	20.3
W2	0.63	0.40	4.9	30.2
W4	0.45	0.19	7.3	30.2
W6	0.23	0.23	5.4	30.2
X3	0.51	0.12	4.6	30.2
X5	0.39	0.09	5.7	30.2
Y4	0.51	0.28	2.7	30.2
Range	0.15–1.58	0.05–0.73	2.7–11.9	15.0–30.5
Average	0.47	0.24	6.3	
σ	0.32	0.16	2.4	

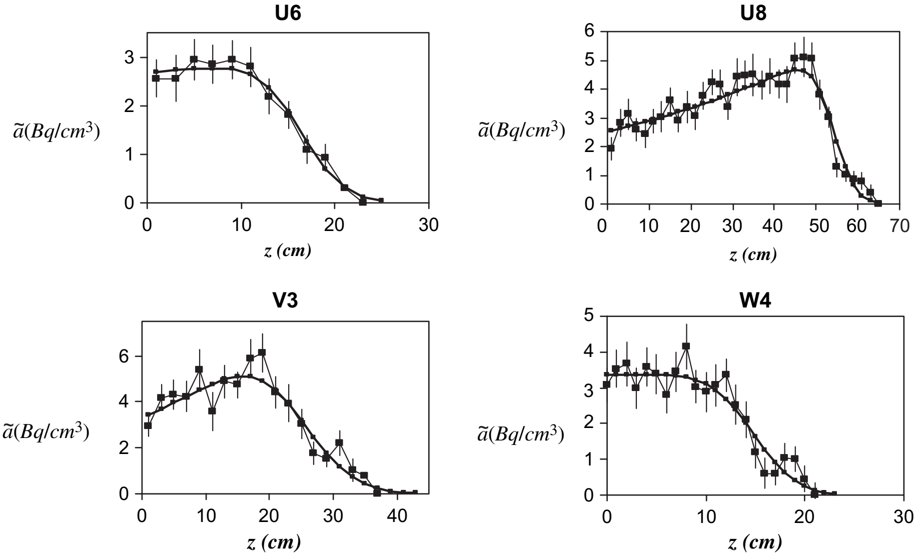

Fig. 4. Fit between the profiles of experimental activity by unit of volume and those given by the model, for some of the sampling stations.

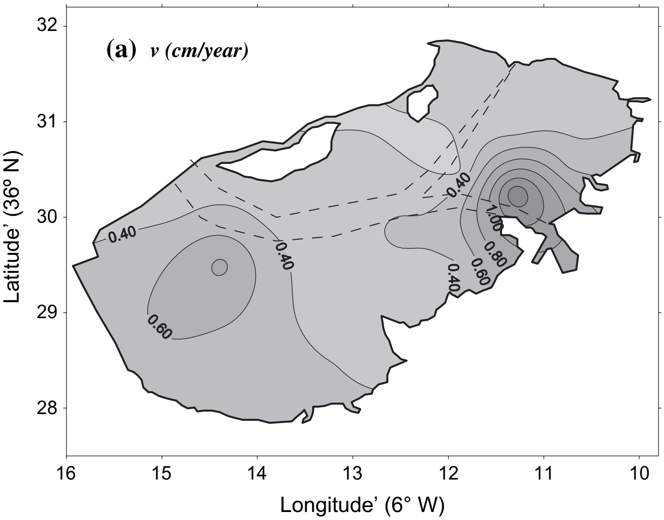

Fig. 5. Distribution of the rate of linear sedimentation derived from the model for the Inner Bay.

produced by the flow tide (Álvarez, 2000). It is probable that part of the sediment leaving the Inner Bay through the Puntales strait in the flow tide is returned again by the ebb tide currents. This sediment would be trapped by this vortex and deposited later, giving rise to this zone of peak sedimentation previously mentioned. The scheme of erosion, transport and subsequent sedimentation presented here coincides notably with the transport paths determined in a previous study of the zone (Gutiérrez-Mas et al., 1999; Achab, 2000). This work cited was conducted utilising the proportions found between the various minerals of the sedimentary clay, together with satellite images and other data. According to this study, the Inner Bay is a source that supplies sediment to the Outer Bay, and to the adjacent continental shelf.

When the rates of sedimentation are known, and under the hypothesis that these rate are constant at each station, it is possible to deduce the date of formation of the layers of sediment. A layer of accumulated mass thickness δm (quantity of matter per unit of surface) will have been deposited during a period of time $\Delta t = \delta m / v_m$, and therefore the surface layer is formed in the interval of time $(t = -\delta m_1 / v_\mathrm{m}, 0)$. In general, a layer N is deposited in the interval of time $(t = -\delta m_N / v_m, -\delta m_{N-1} / v_\mathrm{m})$, where δm_N is the accumulated mass thickness, from the base of the layer N up to the interface. Taking into account the date of sampling, $t_\mathrm{o} = 1998$, the date of formation of the layer is determined by means of date $= t_\mathrm{o} + t$. Although this dating is only possible for the layers in which ^{137}Cs is detected, and that therefore are less than 35 years old, it is possible to extrapolate the dating to deeper layers, accepting the hypothesis $v_\mathrm{m} = cte$. Following this method, sedimentary layers to a maximum age about 200 years have been dated, as shown in Table 2.

Table 2
Dating of the sediment strata for all the sampling stations

z (cm)	R7	S8	T7	T9	U2	U4	U6	U8	V1	V3	V5	V7	W2	W4	W6	X3	X5	Y4
0–2	1995(3)	1994(4)	1992(6)	1997(2)	1993(5)	1995(3)	1996(2)	1997(1)	1997(1)	1997(1)	1995(3)	1995(3)	1996(2)	1996(2)	1994(4)	1996(2)	1995(3)	1996(2)
2–4	**1990(3)**	1986(5)	1979(8)	1994(2)	1985(4)	**1989(4)**	1993(2)	1996(1)	1994(2)	1995(2)	**1988(4)**	1991(2)	1993(2)	1992(2)	1986(4)	1993(1)	1990(3)	1993(2)
4–6	1985(3)	1977(5)	1965(7)	1990(2)	1977(5)	**1981(4)**	1989(2)	1995(1)	1989(3)	1993(1)	1981(4)	1986(3)	1990(2)	1989(2)	1978(5)	**1990(2)**	**1986(2)**	1990(2)
6–8	1979(4)	1968(5)	1952(7)	1987(2)	1969(3)	1974(4)	1985(2)	1994(1)	1984(3)	1990(2)	1974(4)	1981(3)	1987(2)	1984(3)	1968(6)	**1987(2)**	1982(2)	**1986(2)**
8–10	1972(4)	1960(5)	1939(6)	1983(3)	1962(4)	1966(4)	1980(3)	1993(1)	1978(4)	**1988(1)**	1964(6)	1976(3)	1984(2)	1979(3)	1958(4)	1984(2)	1977(3)	1982(2)
10–12	1965(4)	1951(5)	1927(7)	1978(3)	1957(3)	1959(4)	1976(2)	1991(1)	1972(3)	1985(2)	1953(5)	1970(4)	1980(2)	1974(3)	1949(5)	1980(2)	1972(3)	1978(2)
12–14	1958(4)	1942(5)	1914(7)	1973(3)	1951(3)	1951(4)	1972(2)	1990(1)	1967(3)	1982(2)	1943(5)	1964(3)	1976(2)	1969(3)	1940(5)	1975(3)	1966(3)	1974(2)
14–16	1951(4)	1933(5)	1902(6)	1968(2)	1945(4)	1944(4)	1967(3)	1989(1)	1963(2)	1979(2)	1933(5)	1958(3)	1971(3)	1964(3)	1928(7)	1970(3)	1959(4)	1969(3)
16–18	1944(3)	1922(6)	1890(6)	1964(3)	1938(3)	1937(3)	1962(3)	1988(1)	1959(2)	1975(2)	1923(5)	1953(3)	1966(3)	1958(3)	1915(7)	1965(3)	1952(4)	1965(2)
18–20	1938(4)	1909(7)	1878(6)	1959(3)	1932(4)	1931(4)	1956(3)	1986(1)	1956(2)	1972(2)	1912(6)	1947(3)	1962(2)	1953(3)	1901(7)	1960(3)	1944(5)	1960(3)
20–22	1931(4)	1895(7)	1866(7)	1954(3)	1925(3)	1924(3)	1950(4)	1985(1)	1953(2)	1969(2)	1900(6)	1942(3)	1957(3)	1948(3)	1888(7)	1954(3)	1936(4)	1956(2)
22–24	1924(4)	1882(8)	1853(6)	1949(3)	1918(4)	1917(4)	1944(3)	1984(1)	1949(2)	1966(2)	1890(5)	1936(3)	1952(3)	1942(3)	1874(7)	1949(3)	1929(4)	1952(2)
24–26	1916(4)	1867(8)	1841(6)	1944(3)	1911(3)	1909(4)	1939(3)	1982(1)	1945(2)	1962(2)	1879(6)	1930(3)	1948(2)	1937(3)	1861(7)	1944(3)	1922(4)	1947(3)
26–28	1909(3)	1852(7)	1828(8)	1939(2)	1904(4)	1900(5)	1933(3)	1981(1)	1942(2)	1958(2)	1868(6)	1925(3)	1943(3)	1932(3)	1849(6)	1938(3)	1914(4)	1943(2)
28–30	1903(4)	1838(7)	1813(8)	1935(3)	1896(4)	1890(5)	1928(3)	1980(1)	1938(2)	1955(2)	1856(6)	1920(3)	1939(2)	1927(3)	1836(7)	1932(4)	1907(4)	1938(3)
30–32	1987(3)	1824(7)		1930(3)	1889(4)	1880(6)	1923(3)	1979(1)	1934(2)	1951(2)	1845(6)	1916(2)	1934(3)	1921(3)	1823(7)	1924(4)	1899(4)	1933(3)
32–34	1890(4)	1810(7)		1925(3)	1881(4)	1869(5)	1917(3)	1978(1)	1930(2)	1947(2)	1834(6)	1911(3)	1929(3)	1914(4)	1810(7)	1916(4)	1891(4)	1928(3)
34–36	1883(3)			1920(3)	1873(5)	1859(5)	1912(2)	1976(1)	1926(2)	1943(2)	1821(7)	1906(2)	1924(3)	1907(4)		1910(3)	1883(5)	1923(3)
36–38	1877(4)			1915(3)	1865(4)	1850(4)	1907(3)	1975(1)	1923(2)	1939(2)	1808(7)	1901(3)	1918(3)	1899(4)		1903(4)	1875(4)	1919(2)
38–40	1869(4)			1910(3)	1857(4)	1841(6)	1902(2)	1973(1)	1919(2)	1935(2)		1897(2)	1912(3)	1890(5)		1896(4)	1867(4)	1915(2)
40–42	1862(3)			1905(2)	1849(5)	1831(5)	1897(3)	1972(1)	1915(2)	1930(3)		1892(3)	1905(4)	1882(4)		1890(3)	1859(4)	1911(2)
42–44	1856(4)			1901(2)	1840(4)	1822(5)	1891(3)	1971(1)	1911(2)	1924(3)		1887(3)	1899(3)	1874(5)		1884(3)	1850(5)	1907(2)
44–46	1848(4)			1897(3)	1832(5)	1813(5)	1885(3)	1970(1)	1907(2)	1920(2)		1882(3)	1893(3)	1866(4)		1877(4)	1841(4)	1902(3)

Peaks of activity attributed to the Chernobyl accident are given in bold.

In respect of the coefficient of diffusion D for ^{137}Cs provided by this model, its range of values is compatible with those determined for coastal sediments by other authors (Clifton et al., 1994). One of the mechanisms responsible for the migration of ^{137}Cs is molecular diffusion, which is produced as a consequence of the exchange of the radionuclide between the solid and liquid phases of the sedimentary medium. The affinity of the radionuclide for the solid phase, represented by the coefficient of distribution k_{D}, and the molecular diffusion, D^{molec}, are inversely related, i.e. the stronger the affinity of the radio element for the solid phase (k_{D}), the less the activity present in the interstitial water, and therefore the less the molecular diffusion. Taking into account the freedom of movement of the radioisotope in the interstitial water, some authors (Berner, 1980; Kirchner et al., 1993) approximate the coefficient of molecular diffusion by means of the expression

$$D^{\mathrm{molec}} \approx \frac{w^2 D_{\mathrm{o}}^{\mathrm{molec}}}{1+\frac{\rho}{w} k_D} \tag{6}$$

where w and ρ are the porosity and density of the sediment, respectively, and $D_{\mathrm{o}}^{\mathrm{molec}}$ is the coefficient of molecular diffusion of ^{137}Cs in water, the value of which is approximately 10^{-5} cm^2/s (Li and Gregory, 1974). Utilising this expression, and considering that the diffusion given by the model is molecular in character, the coefficient of distribution k_{D} has been calculated for the different stations (Table 3). A significant correlation, r, is found between this coefficient, the activity of ^{137}Cs at the interface ($r = 0.75$), and the content of organic carbon ($r = 0.70$). This demonstrates that the capacity of retention of ^{137}Cs in the top layer of sediment is linked to the organic matter and to the granulometry of the sediment. On the contrary, the coefficient of distribution does not present any correlation with the total inventories of ^{137}Cs, which demonstrates that the total content of ^{137}Cs at each station is not related to the capacity of absorption of the radionuclide. This is corroborated by the model proposed for the vertical migration of ^{137}Cs, which attributes the value of the inventories to the rate of sedimentation, rather than to this capacity of absorption. Table 4 show the values of sedimentation rate and total inventory of ^{137}Cs for all the stations, and Fig. 6 the fits between both quantities with a correlation coefficient equal to 0.94. The coefficient of distribution k_{D} in clay-type media, determined in various field and laboratory studies (Young-Hwan et al., 1996), has a value slightly higher than that given by this model, which indicates that the parameter D^{molec}, which we have employed to determine k_{D}, has been over-estimated. This means that the diffusion of ^{137}Cs is not only molecular in character, but also that part of the diffusion is due to the activity of benthic organisms on the biological mixture of the sedimentary material.

In previous studies conducted in the same zone, aimed at studying the evolution of heavy metal contamination (Gutiérrez, 1999; Ligero et al., 2002), the researchers utilised the method of dating by ^{210}Pb in the CRS (Constant Rate of Supply) model (Appleby and Oldfield, 1978). In order to test the validity of the model developed for the migration of ^{137}Cs, and the technique of dating the strata of sediment columns,

Table 3
Calculated distribution coefficient

Station	k_D (cm^3/g)
P1	120
R7	88
S8	560
T7	170
T9	307
U2	75
U4	246
U6	439
U8	262
V1	266
V3	94
V5	571
V7	1915
W2	364
W4	538
W6	600
X3	1451
X5	1878
Y4	99

derived from the model, Table 5 and Fig. 7 presents by way of example, the dating given by the two techniques at one of the stations studied. As can be observed, there is good agreement between the two procedures, which strengthens the validity of the model presented in this study.

Table 4
Sedimentation rate and total inventory of ^{137}Cs

Station	v (cm/year)	I (Bq/m^2)
P1	0.57	980
R7	0.32	319
S8	0.23	192
T7	0.15	166
T9	0.49	369
U2	0.26	195
U4	0.28	220
U6	0.46	481
U8	1.58	2141
V1	0.45	304
V3	0.85	816
V5	0.24	231
V7	0.38	560
W2	0.63	575
W4	0.45	567
W6	0.23	376
X3	0.51	507
X5	0.39	328
Y4	0.51	220

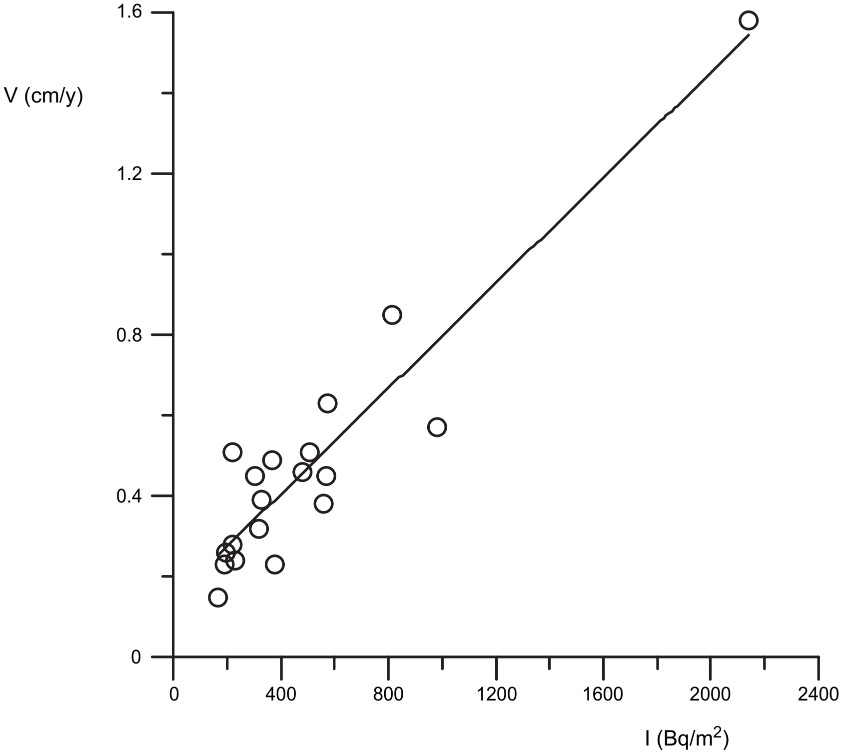

Fig. 6. Total inventory of ^{137}Cs versus sedimentation rate for all stations.

Table 5
Ages of the sediment levels in years, for one station, obtained from the model and by using ^{210}Pb dating technique

z (cm)	^{137}Cs	^{210}Pb
0–2	2 ± 2	2 ± 1
2–4	8 ± 3	6 ± 1
4–6	16 ± 4	12 ± 1
6–8	23 ± 4	19 ± 2
8–10	33 ± 6	28 ± 3
10–12	44 ± 5	38 ± 4
12–14	54 ± 5	45 ± 5
14–16	64 ± 5	52 ± 7
16–18	74 ± 5	61 ± 10
18–20	85 ± 6	72 ± 14
20–22	96 ± 5	87 ± 23
22–24	107 ± 5	111 ± 51

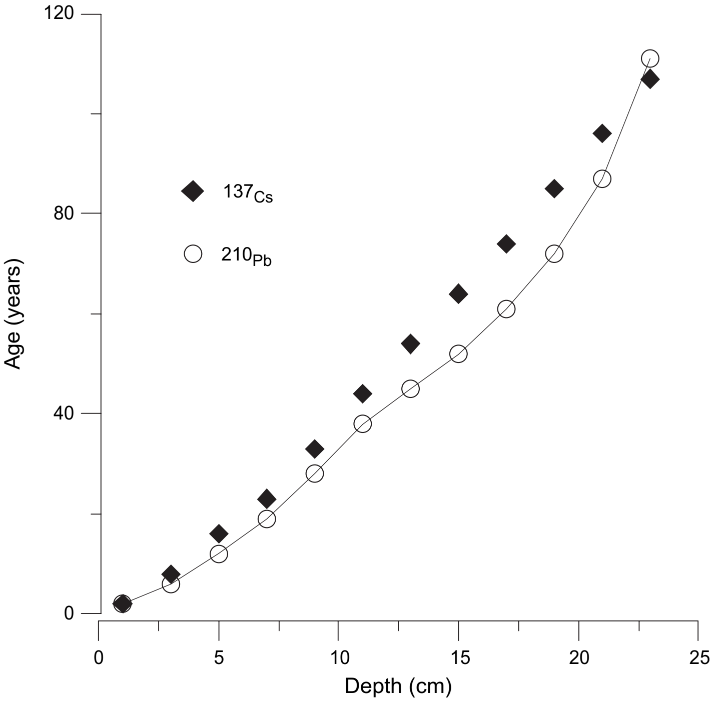

Fig. 7. Ages of the sediment levels for one station derived from the model (plots) and by ^{210}Pb dating technique (continuous line).

Further, the boundary condition utilised for the sedimentary interface, $c(t)$, only includes the fallout associated with the peak of 1963. This means that the radioactivity originating from the accident at Chernobyl present in the sediments of the Bay of Cádiz should be manifested as an increase of the experimental activity of ^{137}Cs, in comparison with that predicted theoretically. Because the time elapsed since the accident up to the date of extraction of samples is some twelve years, and considering the mean rate of sedimentation in the Inner Bay, 0.5 cm/year, the activity from Chernobyl should be located at an average depth of about 6 cm. In effect, although the exact depth in each profile depends on the rate of sedimentation associated with each station, in general, the columns of sediment that detect this activity peak do present it at approximately the expected depth.

4. Conclusions

The reduced value of the inventory of ^{137}Cs measured in the Inner Bay (460 Bq/m^2), in relation to the estimated deposition on the zone (2000 Bq/m^2) originating from the radioactive fall-out from nuclear tests, is because most of the activity that originally fell onto these waters has been removed from the Inner Bay by tidal currents before it could be deposited on and incorporated into the marine sediments. This rapid removal is the consequence of the high solubility of the radionuclide. Practically all the ^{137}Cs present in these sediments has its origin in the system of creeks and salt marshes of the zone to the south and southeast of the Inner Bay, where the radio element had previously been incorporated.

The migration model proposed, which takes into account the time of residence in the source zones, correctly interprets the experimental profiles of activity in respect of depth. Unlike what occurs at the interface, the global presence of ^{137}Cs in the sediments is not associated with characteristics of the sediment such as the granulometry, organic carbon content, and porosity, but rather reflects the differences in the accumulation of sediment taking place in each zone of the Bay.

In addition to the advective movement, in consequence of the incorporation of sediment on the interface, the ^{137}Cs enjoys an additional mobility within the body of the column that can be described in terms of a generic diffusion. On the one hand, the radionuclide dissolves in the interstitial water of the sediment, which gives rise to molecular diffusion, and on the other, the radionuclide is transported vertically in consequence of the mixing processes generated by the biological activity of benthic organisms.

The rate of sedimentation derived from the model provides an indirect method for dating the sediments which provides a similar age estimated as the method of ^{210}Pb dating. In addition, by means of extrapolation in the sediment column, the dating can be extended to an interval of time much longer than can be obtained using calculations of radioactive disintegration, as a result of the 30 years half-life of the ^{137}Cs.

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