Identification of suitable areas for high-resolution sea-level studies in SW Europe using commonly applied ^{210}Pb models

Identificación de aéreas validas para la realización de estudios de alta resolución sobre variaciones del nivel marino en el SO de Europa mediante modelos comunes de ^{210}Pb

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RESUMEN

Se ha analizado la distribución vertical de ^{210}Pb y ^{137}Cs en siete nuevos sondeos cortos (50 cm) de cinco estuarios diferentes de la costa Atlántica de la Península Ibérica con el fin de identificar posibles áreas donde desarrollar estudios de alta resolución sobre cambios en el nivel marino. De los siete sondeos, dos presentaron excelentes perfiles sugiriendo que esas zonas podrían ser adecuadas para este tipo de estudios (Urdaibai y Miño). Por otro lado, dos sondeos parecen reflejar las actividades humanas en las áreas colindantes, con elevadas tasas de sedimentación (Urdaibai y Mira). Un sondeo presenta una interrupción en la sedimentación (Mira), invalidando su uso, mientras que los dos restantes (Plentzia y Sado) presentan cronologías que dependen del modelo aplicado y requieren, por tanto, de indicadores adicionales antes de poder ser utilizados en este tipo de estudios.

***Key words:** Sea-level; marsh environment; ^{210}Pb; ^{137}Cs; SW Europe.*

Introduction

Global estimates of sea-level rise for the 20th and 21st centuries are *ca.* 1.8 mm yr^{-1} (Church and White, 2006; IPCC, 2007). These values represent an acceleration in comparison to previous centuries. However, these estimates are based on geographically limited instrumental data that in most cases span short periods of time (Douglas, 2008). Furthermore, they do not capture local and regional sea-level variability (Katsman *et al.*, 2008). Geological data can usefully complement instrumental data, e.g., in northern Spain sea-level observations have been extended back to the late 17th century. (Leorri *et al.*, 2008; Garcia-Artola *et al.*, in press). These studies depend strongly on a precise chronology (Leorri *et al.*, 2008). Typically, to cover the last 100-150 years chronologies are based on ^{210}Pb and supported by ^{137}Cs and other chrono-horizons (Leorri *et al.*, 2008). We analyze here the vertical distribution of ^{210}Pb and ^{137}Cs in seven new short (50 cm) cores in order to identify possible areas suitable for sea-level studies.

Study area

We collected seven 50-cm sediment cores for geochemical analyses in five different estuarine areas along the northern and western Iberian coast: two cores (Kanala and Mape) in the Urdaibai estuary (northern Spain), one core in the Plentzia estuary (north of Spain), one core in the Minho estuary (northern Portugal), one core in the Sado estuary (southern Portugal) and two final cores (Xisto and Vilanova de Milfontes) in the Mira estuary (southern Portugal) (Fig. 1). Sampling sites were chosen in the high marsh environment (above the mean high water level). The size of the estuaries ranges from < 200 ha (Plentzia) to more than 23,500 ha (Sado). Tidal ranges are mesotidal and the mean tidal range varies from 1.8-2.0 m in Minho and Sado to 2-2.5 m in Plentzia, Urdaibai and Mira (Fig. 1).

Materials and Methods

Dating recent marsh sediments usually relies on the determination of the vertical distribution of unsupported ^{210}Pb (half life 22.3 years), a naturally occurring fallout radionuclide, which then allows ages to be ascribed to sedimentary layers based on the known decay rate of ^{210}Pb (Appleby and Oldfield, 1992). On the other hand, ^{137}Cs (half life 30 years) is an artificially produced radionuclide present in the environment due to atmospheric fallout from nuclear weapons testing, reactor accidents, and discharges from nuclear facilities. Global dispersion and fallout of ^{137}Cs occurred from 1954 onwards following the detonation of high-yield thermonuclear weapons. In the northern hemisphere, distinct maxima in fallout

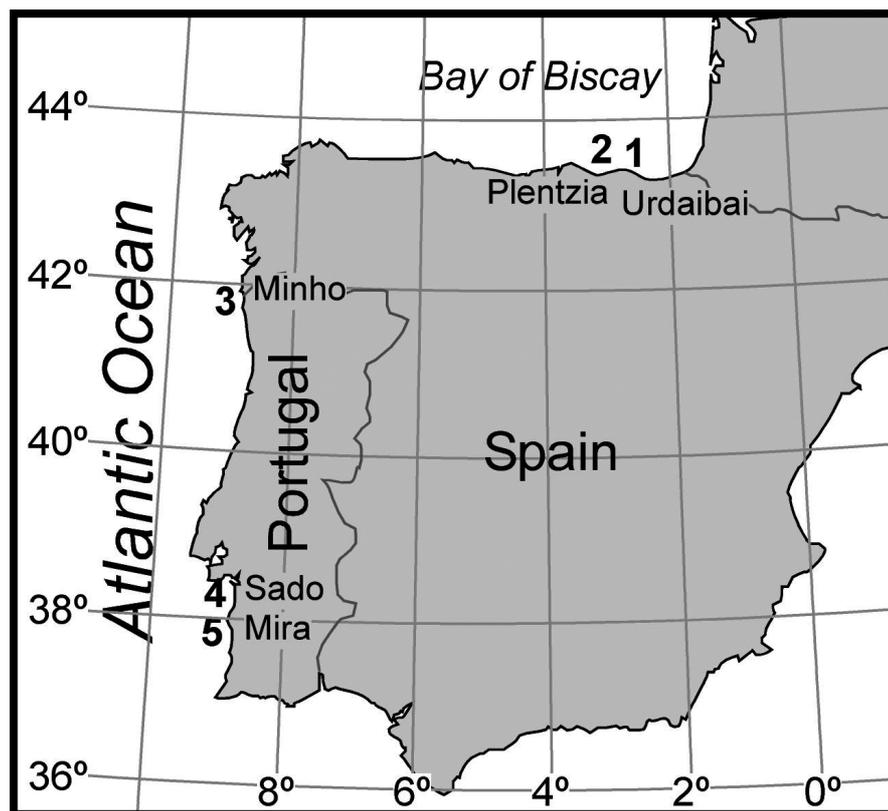**Fig. 1.- Location map. Key: 1-Urdaibai; 2-Plentzia; 3-Minho; 4-Sado; 5-Mira.**

Fig. 1.- Mapa de localización. Clave: 1-Urdaibai; 2-Plentzia; 3-Miño; 4-Sado, 5-Mira.

occurred in 1958 and 1963 (from nuclear weapons testing) and in 1986 (from the Chernobyl incident). In favorable conditions, these periods of peak fallout provide subsurface activity maxima in accumulating sediments, which can be used to derive rates of sediment accretion and support ^{210}Pb derived chronologies. The remote position of the study areas from major nuclear facility discharges (e.g. Sellafield, La Hague) and the Chernobyl plume means that ^{137}Cs in these marshes is likely to be dominantly derived from nuclear weapons testing, with peak fallout in 1963 (Cearreta *et al.*, 2002).

Samples from the cores recovered in Urdaibai, Plentzia and Sado were analyzed following the methodology described by Appleby (2001). All samples were packed and sealed in gas tight containers and stored for 21 days to allow equilibration between ^{214}Pb and its parent radioisotope ^{226}Ra prior to measurement by gamma spectrometry. Activities of the target radionuclides were measured using an EG&G Ortec planar (GEM-FX8530-S N-type) HPGe gamma spectrometry system built to ultra-low background specification for ^{210}Pb detection. Total ^{210}Pb was measured by its gamma emissions at 46.5 KeV and its

unsupported component (^{210}Pb$_{Excess}$) calculated by subtraction of ^{226}Ra activity, which in turn was measured by the gamma emission of ^{214}Pb at 295 and 352 KeV.

In the Minho and Mira estuaries, total ^{210}Pb was measured by alpha spectroscopy following the methodology of Nittrouer *et al.* (1979). Approximately 1.5 g of sediment was spiked with ^{209}Po, as a yield determinant, and then was partially digested with 8 N nitric acid (HNO$_3$) by microwave heating. Polonium-209 and ^{210}Po in solution was then electroplated onto nickel planchets in a dilute acid solution (modified from Flynn, 1968). ^{210}Pb$_{Excess}$ was determined by subtracting the ^{210}Pb activity supported by ^{226}Ra from the total ^{210}Pb activity, where the supported ^{210}Pb activity for a given core was assumed to be equal to the uniform background activity found at depth (Nittrouer *et al.*, 1979).

In all cores, ^{137}Cs was determined by its gamma emissions at 662 KeV (with corrections for ^{214}Bi emissions). Samples from each core were counted for the order of 24 hours to the depth of limit of fallout ^{210}Pb or ^{137}Cs, i.e., until activity concentrations of both radionuclides dropped below the minimum detectable activity. Sample resolution was 1 cm in all

cases and no granulometric changes were identify within each core for the analyzed sections.

The processes affecting sediment deposition can vary greatly from place to place and there is not a unique model to provide a chronology based on ^{210}Pb (Irabien *et al.*, 2008). Most models are based on a series of assumptions among which the most common are: 1- steady-state system, 2- closed system, 3- continuous ^{210}Pb supply; and 4- no postdepositional vertical redistribution of ^{210}Pb. From the possible models, we have used here the simple model (Robbins, 1978), constant initial concentration (CIC; Robbins and Edgington, 1975) and constant rate of supply (CRS; Appleby and Oldfield, 1978) and when possible compared them.

We additionally measured water content and bulk density to account for possible changes in the radiometric activity due to these factors, e.g., possible changes of density due to autocompaction downcore.

Results and Discussion

Figure 2 summarizes the vertical distribution of both ^{210}Pb$_{Excess}$ and ^{137}Cs in all seven cores. With the exception of one core (Xisto), the agreement between ^{210}Pb derived chronologies and ^{137}Cs could be classified as good or excellent.

All cores, except Mape, showed an exponential, although variable, decline in ^{210}Pb$_{Excess}$ with depth and plots of the natural logarithm of ^{210}Pb were fairly linear. This suggested relatively constant rates of deposition, allowing us to use the simple and CIC model. We also calculated sedimentation rates based on CRS for Kanala, Isuzkiza and Sado. However, irregularities in the plot of the natural logarithm of ^{210}Pb$_{Excess}$ at Mape suggested that there could have been a short-term fluctuation in net deposition and therefore we used only CRS since these changes prevent the use of the other two methods (Marshall *et al.*, 2007).

In northern Spain, Irabien *et al.* (2008) showed the strong variability of sedimentation rates in response to environmental changes, spatial distribution and human impact. In this same area, several low-resolution sea-level reconstructions have been based on chronologies derived from ^{210}Pb (Leorri *et al.*, 2008; García-Artola *et al.*, in press). It is therefore essential to understand these depositional environments before we undertake any high-resolution sea-level study.

At the Kanala site, ^{210}Pb activity showed a general decline with depth. This profile did not suggest any major mixing or disruption in the sedimentation. Estimates derived from the simple, CIC and CRS models are in agreement and indicate that the uppermost 10 cm have been deposited over 100 years. The main difference occurred between 8.5 and 10.5 cm depth, where CRS indicates an inflexion in the sedimentation rates, i.e., lower sedimentation rates at the bottom of the core. On the other hand, ^{137}Cs shows a clear subsurface maximum in activity at 3.5 cm depth, declining with depth to negligible values at 18 cm (Fig. 2). Ascribing this subsurface activity maximum to AD 1963 supports all three estimates; the disagreement with the simple and CIC model estimates is less than 2 years. While all models are in good agreement, Marshall *et al.* (2009) indicated that the CRS model tended to produce dates that were progressively too old when compared to ages derived from local metal mining histories in four salt marshes of SW England. While the inflexion indicated by the CRS model could be an artifact rather than a real change in the sedimentation rates, the activity at these levels is very low, the associated uncertainty very large and therefore the difference between estimates is not statistically significant. Based on ^{210}Pb and ^{137}Cs profiles and the lack of compaction in the core, Kanala is an excellent site for high-resolution sea-level studies.

In the same estuary, the core recovered in Mape strongly differed from Kanala. In fact, the ^{210}Pb$_{Excess}$ profile showed greater values in the lower section of the core as it happens with the ^{137}Cs profile. In this case, we only attempted to provide a chronology based on the CRS model that indicates that the 50 cm were deposited in less than 100 years, but with variable rates up to 20 mm yr^{-1} in some sections. This is in general agreement with the presence on ^{137}Cs up to 50 cm depth that indicates that the core section has been deposited in *ca.* 50 years. These results are supported by historical aerial photography that show that the environmental change occurred in the area over the last 50 years in response to the deforestation of the nearby hills and increase of sediment load transported by one of the Oka river tributaries.

In the Plentzia estuary, the Isuskiza core showed some inflexions in the ^{210}Pb$_{Excess}$ profile which prevented the use of the CIC model. Also, the ^{137}Cs showed
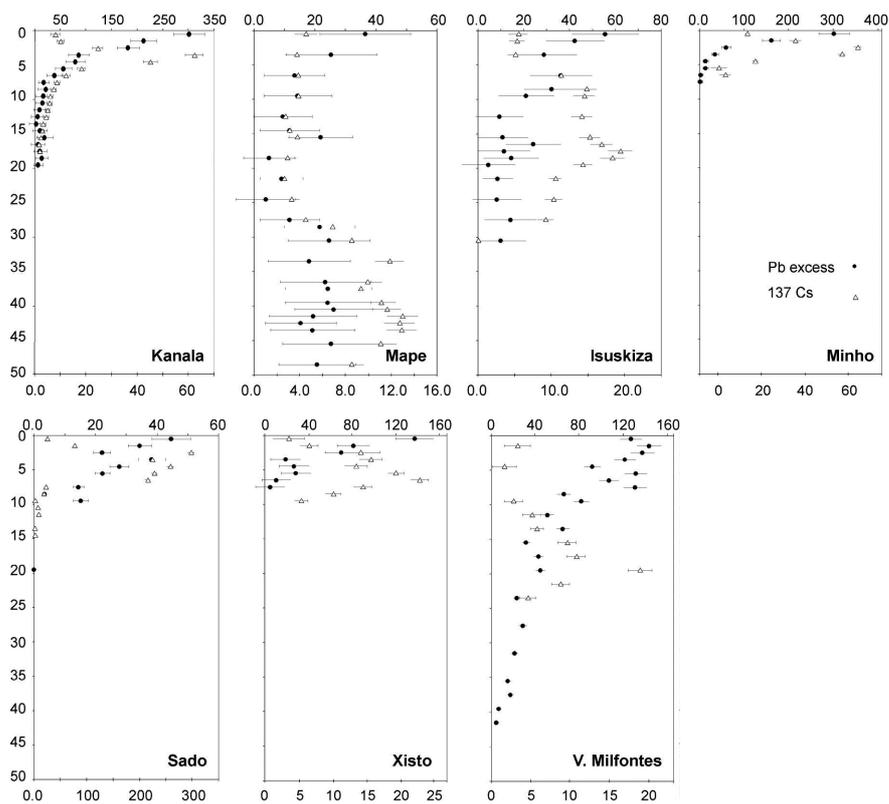

Fig. 2.- ^{210}Pb (upper axis) and ^{137}Cs (lower axis) in depth (cm) profiles for all cores analyzed.

Fig. 2.- Perfiles de ^{210}Pb (eje superior) y ^{137}Cs (eje inferior) según la profundidad (cm) en todos los sondeos analizados.

high mobility downcore that seems to be characteristic of most marsh cores. We tried to use additional chronological markers, such as ^{241}Am but this radioisotope did not present sufficient activity to permit measurement. Therefore, the chronology derived from this core should be considered carefully. As in Kanala, CRS-derived ages tend to show decrease in the sedimentation rates downcore, in this case, below 27 cm. This effect together with a better agreement between the simple model and ^{137}Cs derived chronologies (by 1 yr) suggests that the simple model is appropriate, providing a sedimentation rate of 4.2 mm year^{-1}.

In northern Portugal, the core recovered in the Minho estuary showed a very similar profile to Kanala, although with a significantly lower sedimentation rate. It presented, in fact, the lowest rate of all the study sites, recording the last 100 years in the top 4-6 cm. In this core, the limiting factor was the sampling resolution (1 cm samples). CIC estimates better agree with the ^{137}Cs (by 4 yrs) suggesting a sedimentation rate of 0.6 mm yr^{-1}. As in Kanala, this core did not present sedimentological changes and only below 33 cm there was a change in

the sample density that might be related to autocompaction. This suggests that the marsh has been accumulating very slowly at least over the last 100 years and suitable sea-level reconstructions might be possible for the last 500 years or more, albeit at low resolution.

In the south of Portugal, the core recovered in the Sado estuary shared some similarities with Isuskiza. In fact, both cores exhibit some inversion in the ^{210}Pb$_{Excess}$ profile. Also in this case, we should consider carefully the derived chronology. As in previous cases, CRS derived sedimentation changes indicate a decrease in the rates downcore. CRS and CIC provided rates between 1.2 and 1.0 mm yr^{-1}, while the simple model indicates much larger sedimentation rates (1.6 mm yr^{-1}) in better agreement with the maxima indicated by ^{137}Cs.

In Mira, further south, we found the same contrasting patterns between the two cores studied here as in Urdaibai. While Vilanova de Milfontes presented high sedimentation rates, Xisto had low rates. In the case of Vilanova de Milfontes, the ^{210}Pb$_{Excess}$ profile shows a top area of mixing (upper 7.5 cm) and both the simple model and CIC are in good agreement (4.4 and 4.0 mm yr^{-1},

respectively). Here, ^{137}Cs is in better agreement with the simple model (4.3 mm yr^{-1}). While in this case we do not have evidence of environmental change from historical data as happened in Mape, from the sedimentological evidence and the changes in density downcore it seems that the rapid sedimentation rates reflect the infilling of the marsh area, most probably in response to anthropogenic activities as occurred in Mape.

Finally, the core recovered in the Xisto marsh has one of the lowest sedimentation rates in all seven cores. Both the simple and CIC model estimates are in good agreement (0.7-0.8 mm yr^{-1}). However, they do not agree with the ^{137}Cs profile that indicates rates that are twice the ^{210}Pb estimates (1.5 mm yr^{-1}). The fact that the ^{137}Cs peak is concomitant with low ^{210}Pb$_{Excess}$ values suggests that the chronology from this core is unreliable. In fact, the ^{210}Pb$_{Excess}$ profile shows a significant jump between 2.5 and 3.5 cm depth. A similar effect was reported in Plentzia, where it was associated with a disruption in the sedimentation (Cearreta *et al.*, 2002), these events are fairly common in marsh environments.

The recent sediment accretion rates obtained from these estuarine areas are typical of many mesotidal marshes elsewhere in NW Europe (e.g. Cundy and Croudace, 1995). However, they are highly variable and strongly influenced by local factors. While Kanala and Minho apparently reflected naturally accreting marshes over the last 100 years, Mape and Vilanova de Milfontes seemed to reflect the human activities in these areas, although this has been proved only in Mape. Xisto on the other hand, showed a disruption in the sedimentation very recently. Isuskiza and Sado showed profiles that yield very different chronologies depending upon the different model used and therefore should be considered with caution.

Conclusions

The results derived from this study suggest that marshes from the northern and western coasts of the Iberian Peninsula can be used for high-resolution sea-level studies. Careful sedimentological and stratigraphic analysis should be performed prior to the selection of the different sites and cores should be selected from high marsh environments. During this study we found two sites that responded to other factors rather than natural accretion, thought to be of anthropogenic origin.

Furthermore, we were able to identify a discontinuity in the sedimentation in one of the cores. In two other cores, additional temporal markers should be required to support the chronology over the interval studied. In fact, the use of additional chronological marker should be strongly recommended in all cases.

While all three models used here are commonly used for dating recent salt marsh sediments, in general, CRS model estimates presented a greater offset in relation with the ^{137}Cs derived chronologies than the simple and CIC model estimates. Furthermore, CRS model presented slower sedimentation rates in the oldest (i.e., 100-120 years) layers in all four cores where we used this model. However, the discrepancies between models tend to be minimal if the associated errors are considered. This effect should be further investigated using additional chronological markers as it can have bearing on the interpretation of accelerations in sea-level rise from the 19th to the 20th century.

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