

# Soil to Plant transfer of $^{226}\text{Ra}$ in a Marsh Area: Modelling Application

A. Martínez-Aguirre & R. Perriñez

Dept. Física Aplicada, EUITA, Universidad de Sevilla, Carretera de Utrera, km 1, 41013  
Sevilla, Spain

## ABSTRACT

*The Odiel river forms an estuarine system which surrounds a large marsh area. A phosphate fertilizer processing complex releases its wastes into the estuary. The presence of  $^{226}\text{Ra}$  in soils and plants (*Spartina Densiflora*) from the marsh has been investigated. Concentrations up to 700 and 15 mBq  $\text{g}^{-1}$  have been detected in soil and plant samples, respectively. Soil to plant concentration ratios have been calculated and some activity ratios have also been investigated. A model which is able to simulate the dispersion of radionuclides in the marsh has been applied. The model includes the ex-change of radionuclides between water and the solid phase (suspended matter and bottom sediments) and the transfer of radionuclides to the plants. Model results are, in general, in good agreement with measurements. © 1998 Elsevier Science Ltd. All rights reserved.*

## INTRODUCTION

The Odiel marsh is located close to the town of Huelva, in the southwest of Spain. The Odiel river forms, at its lowest reaches, a tidal estuary which discharges into the Atlantic Ocean. This estuary surrounds a large marsh area, the Odiel marsh. A phosphate fertilizer processing complex releases part of its wastes directly into the Odiel river; thus, enhanced concentrations of natural radionuclides (U, Th, Po) have been detected in plant (*Spartina Densiflora*) and soil samples collected from the marsh (Martínez-Aguirre and García-León, 1997; Martínez-Aguirre *et al.*, 1997). The fertilizer complex seems to be responsible for the enhanced levels of radioactivity measured in the marsh. Concentration ratios,  $C_R$ , which are defined as

the ratio between the concentration of an element in the plant and that in its substrate were also calculated for these radionuclides.

A model which describes the dispersion of radionuclides in the marsh has been developed (Periáñez and Martínez-Aguirre, in press). This model includes the exchange of radionuclides between water and the solid phase (suspended matter and bottom sediments). It also includes the transfer of radionuclides to soils, which are covered by water during part of the tidal cycle (while bottom sediments are always under water). Finally, the transfer of radionuclides from soil to plant was implemented in the model. This model was successfully applied to simulate the contamination of soils and plants by  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{210}\text{Po}$  (Periáñez and Martínez-Aguirre, in press).

Due to the radiological relevance of  $^{226}\text{Ra}$ , the previous work has been extended to this radionuclide.  $^{226}\text{Ra}$  concentrations are measured in the same soil and plant samples as in those references mentioned, using alpha and gamma spectrometry. Concentration ratios for  $^{226}\text{Ra}$  are also calculated. Finally, the same model is applied to this radionuclide. Thus, an extra validation of the model is provided, i.e. the model is validated for elements with different geochemical behaviours: U, Th, Po and Ra.

In the next section the experimental work is presented. In Section 3 the model is described briefly and, finally, experimental and model results are presented and discussed.

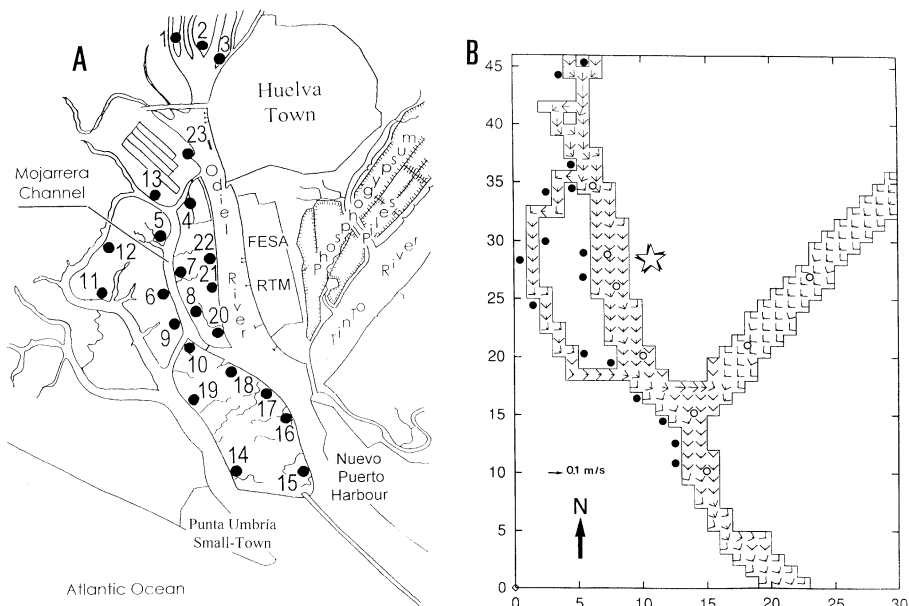
## SAMPLES AND EXPERIMENTAL METHODS

Several stations were chosen across the Odiel Marsh area of Huelva at the southwest of Spain. At each station, surface soils and *Spartina Densiflora* samples were collected at the same time, at the border of the channels (see Fig. 1). All samples are affected by the concentration of radionuclides in the water, since they are covered by water during part of the tidal cycle.

$^{226}\text{Ra}$  activity concentrations were determined in all samples. The enhanced activity concentrations in soil samples allow their  $^{226}\text{Ra}$  analysis by  $\gamma$ -spectrometry using a Reverse Coated Germanium detector. In case of plant samples, with lower activity concentration, a proportional counter (LB770) was used.

$^{226}\text{Ra}$  in soil samples was determined by its emission at 182.6 keV, after the contribution of the  $\gamma$ -emission of  $^{235}\text{U}$  to this peak was subtracted. The efficiency curve was determined by adding known activities of  $^{152}\text{Eu}$  and  $^{133}\text{Ba}$  to a soil sample with similar density to that of the soil samples.

For  $^{226}\text{Ra}$  analysis in *Spartina Densiflora* samples, a LB770  $\alpha$  counter with ten different proportional counters was used. The samples were first



**Fig. 1.** (a) Map of the Odiel marsh showing the sampling stations. (b) Residual water circulation in the modelled area. Each unit in the x- and y-axis is equal to 250 m. This map also shows the fertilizer complex (star), the points where bottom sediments were collected (white circles) and the points in the marsh where soil and plants samples were collected (black circles).

acid-digested with  $\text{HNO}_3$  and aqua regia and finally redissolved with 10 to 15 ml of concentrated  $\text{HNO}_3$ . The final solution was taken to 0.5 l with distilled water. Five milligrams of Ba was added to the samples and precipitation of Ra– $\text{BaSO}_4$  was then carried out. The precipitate was collected by filtration through  $0.45 \mu\text{m}$  pore size Millipore filters. After 20 days (to allow secular equilibrium of  $^{226}\text{Ra}$  daughters) the ingrown alpha activity of the samples was counted. To determine the  $^{226}\text{Ra}$  activity of the samples, calibration of the detectors was necessary. All samples had the same geometry but different thickness. Since  $^{226}\text{Ra}$  is an  $\alpha$ -emitter, the self-absorption of the samples must be taken into account. For such purpose, several calibration samples with known  $^{226}\text{Ra}$  activities and different Ba quantities, to allow different thickness, were prepared. Their total recovery was then calculated as

$$R_T = \frac{N}{4TA}, \quad (1)$$

where  $N$  is the ingrown alpha counts,  $A$  is the activity (Bq) added to the calibration samples,  $T$  is the counting time (s) and 4 appears due to the fact

that three  $\alpha$ -emitters ( $^{226}\text{Ra}$  daughters) are also counted together with  $^{226}\text{Ra}$ .

The calibration curve was similar for all counters,

$$R_T = 23.34 - 1.732 * e, \quad (2)$$

$e$  being the thickness of the samples in  $\text{mg cm}^{-2}$ .

Thus, for a sample with a known thickness, the above curve allows determination of its total recovery and, thus, its  $^{226}\text{Ra}$  activity in  $\text{Bq g}^{-1}$  is given by

$$A_{226} = \frac{N}{4TR_T M}, \quad (3)$$

were  $M$  is the mass in g of the sample.

Some of the soil samples gave quite low  $^{226}\text{Ra}$  activity concentration; thus their  $\gamma$ -analysis gives rise to quite high uncertainties. For this reason, they were also analysed using the same method described above for plant samples. This allowed a considerable reduction in the uncertainties in their activity concentrations.

## MODEL DESCRIPTION

The model is described in detail in Periañez and Martínez-Aguirre (in press). Therefore, only a brief description of the processes included will be given here.

Radionuclides are released to the river in dissolved and particulate form and are transported along the river by advection and diffusive processes. Thus, water circulation must be known to solve the advective–diffusive equation. Ionic exchanges between water and the solid phase (suspended matter and bottom sediments) also take place. The model includes two kinds of suspended matter particles: contaminated particles released from the fertilizer complex and natural suspended matter particles in the river. Deposition and resuspension of both types of suspended particles occurs, which lead to an exchange of radionuclides between suspended matter and bottom sediments. Radionuclides can be incorporated into soils (during the part of the tidal cycle when they are covered by water) by the same processes by which they are incorporated into bottom sediments: deposition of suspended particles and exchanges with water. Finally, radionuclides can be transferred from soil to plant.

The model must run for long time scales since the contamination of soils and plants has been taking place over the operational period of the fertilizer complex (30 years). Thus, the model works with residual (averaged)

circulation, which permits the use of larger time steps. The residual circulation in the estuary has been obtained from field measurements (Periáñez and Martínez-Aguirre, in press) and can be seen in Fig. 1.

The averaged suspended matter concentration and sedimentation rates over the whole study area must be known to compute ionic exchanges of radionuclides between water and suspended matter and the exchanges of radionuclides between suspended matter and bottom sediments due to deposition and resuspension processes. A detailed formulation of the processes can be found in Periáñez *et al.* (1996a).

Ionic exchanges of radionuclides are described in terms of kinetic transfer coefficients. Thus, the transfer of radionuclides from water to the solid phase is governed by a coefficient  $k_1$  and the inverse process by a coefficient  $k_2$ . The concept of exchange surface (surface of particles per water volume unit in the grid cell) is used to obtain an analytical expression for  $k_1$  (Periáñez *et al.*, 1996b; Periáñez and Martínez-Aguirre, in press).

Radionuclides can also be incorporated in soils, during the part of the tidal cycle that they are covered by water, by the same processes as they are incorporated into bottom sediments. Then they can be absorbed by plants. The transfer of radionuclides from soil to plant is described by the concentration ratio  $C_R$ . The specific activity ( $\text{mBq g}^{-1}$ ) in the plant and in the soil,  $C_p$  and  $C_s$  respectively, can be written as (Periáñez and Martínez-Aguirre, in press)

$$C_p^* = C_p + \frac{C_R C_s - C_p}{1 + C_R M_p / M_s}, \quad (4)$$

$$C_s^* = C_s - \frac{C_R C_s - C_p}{C_R + M_s / M_p}, \quad (5)$$

where \* denotes that the quantity is evaluated in the new time step and  $M_s$  and  $M_p$  are the total masses of soils and plants (g), respectively, in the compartment.

## RESULTS AND DISCUSSION

### Experimental results

In Tables 1 and 2,  $^{226}\text{Ra}$  activity concentrations in soil and *Spartina Densiflora* samples are given, respectively. Also, the  $^{226}\text{Ra}/^{238}\text{U}$  and  $^{210}\text{Po}/^{226}\text{Ra}$  activity ratios ( $^{210}\text{Po}$  and  $^{238}\text{U}$  activities were taken from Martínez-Aguirre and García-León, 1997) in each sample are given.

**TABLE 1**

Data obtained in soil samples collected at the Odiel marsh in 1993.  $^{226}\text{Ra}$  concentrations are given in  $\text{mBq g}^{-1}$ . Errors are  $1\sigma$

<i>Code</i>	$^{236}\text{Ra}$	$^{226}\text{Ra}/^{238}\text{U}$	$^{210}\text{Po}/^{226}\text{Ra}$
M1	166 ± 25	0.917 ± 0.144	1.20 ± 0.19
M2	711 ± 45	1.02 ± 0.08	1.09 ± 0.09
M3	681 ± 36	0.935 ± 0.064	0.944 ± 0.065
M4	598 ± 25	1.11 ± 0.06	0.970 ± 0.059
M6	149 ± 20	1.014 ± 0.142	0.946 ± 0.135
M7	130 ± 21	0.823 ± 0.137	1.00 ± 0.17
M8	33.0 ± 1.7	0.509 ± 0.036	1.96 ± 0.16
M9	149 ± 22	1.17 ± 0.18	1.26 ± 0.20
M10	21.1 ± 1.1	0.199 ± 0.013	1.32 ± 0.12
M11	52.9 ± 2.6	0.702 ± 0.042	3.67 ± 0.25
M12	14.6 ± 0.7	0.326 ± 0.019	3.03 ± 0.22
M13	161 ± 24	0.821 ± 0.126	1.38 ± 0.22
M14	11.3 ± 0.6	0.299 ± 0.020	4.81 ± 0.42
M15	134 ± 16	1.42 ± 0.18	0.978 ± 0.128
M16	484 ± 32	0.947 ± 0.071	0.911 ± 0.073
M17	2.29 ± 0.16	0.130 ± 0.011	17.9 ± 1.9
M18	673 ± 39	1.120 ± 0.076	0.840 ± 0.059
M19	15.8 ± 0.8	0.525 ± 0.034	3.44 ± 0.29
M20	245 ± 26	0.950 ± 0.106	0.951 ± 0.110
M21	3.57 ± 0.18	0.199 ± 0.013	4.54 ± 0.74
M22	25.1 ± 1.3	0.775 ± 0.051	1.482 ± 0.126
M23	242 ± 23	0.988 ± 0.099	0.868 ± 0.094

From  $^{226}\text{Ra}$  data in Table 1, the same conclusions previously obtained through the analysis of other natural radionuclides are obtained (Martínez-Aguirre and García-León, 1997). The activity concentration ranges from 2 to 711  $\text{mBq g}^{-1}$ . Thus, the wide range of activity concentrations found in the Odiel Marsh is a clear effect of the inflow of contaminated Odiel riverwaters into the marsh. Again, only the areas further away from the fertilizer industries show activity concentrations typical of undisturbed soils. The data obtained in the analysis of both activity ratios in Table 1 is more interesting. In the case of the  $^{226}\text{Ra}/^{238}\text{U}$  activity ratios, quite different data are obtained, ranging from  $0.199 \pm 0.013$  to  $1.17 \pm 0.18$ , with the exception of sample M15 with  $1.41 \pm 0.18$ . The values seems to be dependent on the activity concentration in the soils. This fact can be clearly observed in Fig. 2a, where this activity ratio vs. the concentration of  $^{226}\text{Ra}$  has been plotted. As can be seen, the ratio increases as the  $^{226}\text{Ra}$  concentration increases in the soil. Thus, in undisturbed soils, this ratio is below the

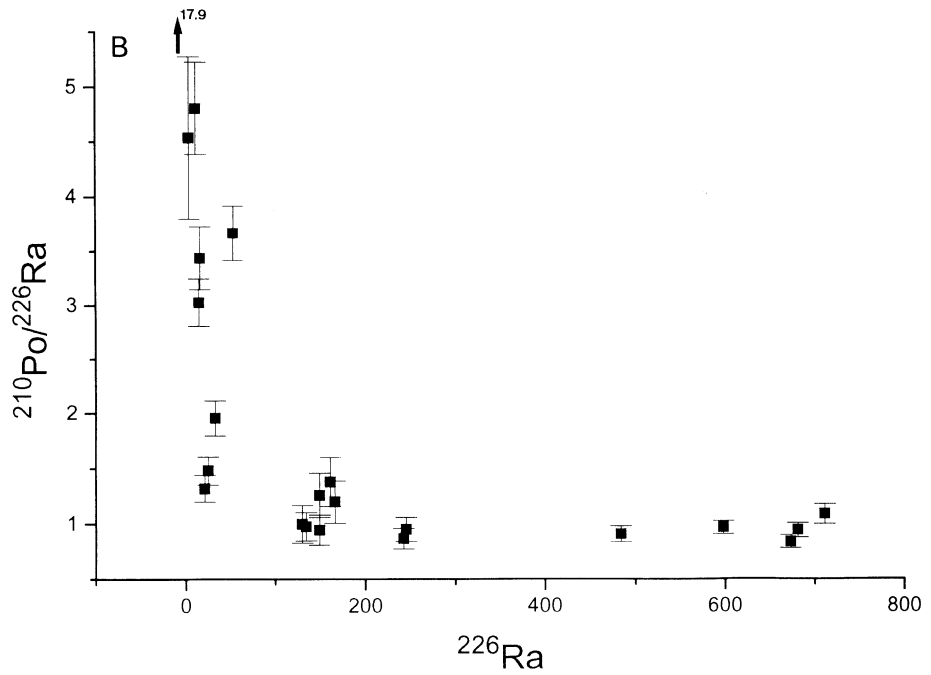
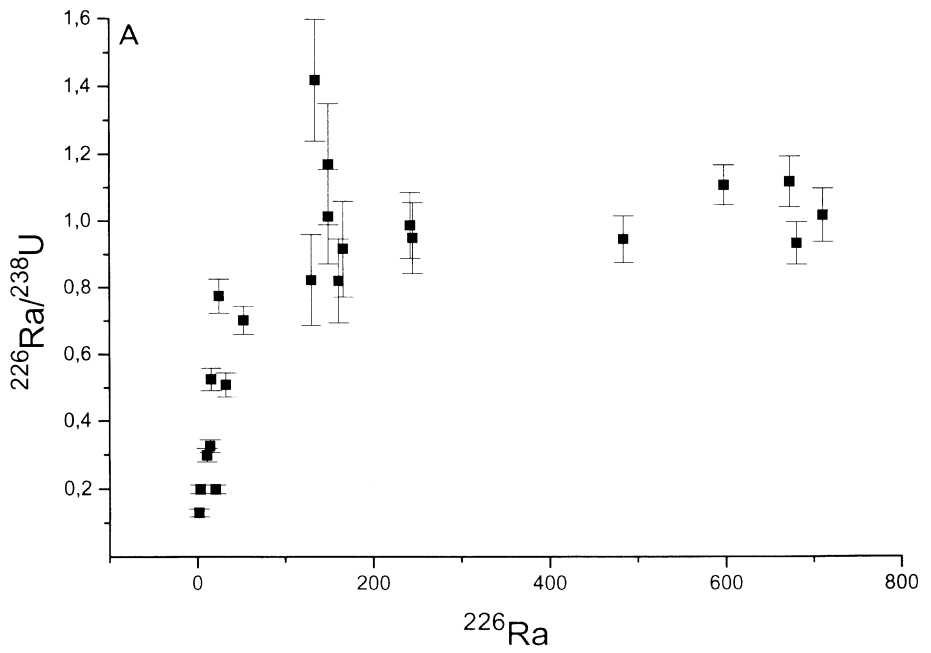
TABLE 2

Data obtained in *Spartina Densiflora* samples collected at the Odiel marsh in 1993, including the  $C_R$ .  $^{226}\text{Ra}$  concentrations are given in  $\text{mBq g}^{-1}$ . Errors are  $1\sigma$

Code	$^{226}\text{Ra}$	$^{226}\text{Ra}/^{238}\text{U}$	$^{210}\text{Po}/^{226}\text{Ra}$	$C_R$
M1	5.36 ± 0.21	0.323 ± 0.025	4.40 ± 0.34	0.032 ± 0.005
M2	6.36 ± 0.25	0.421 ± 0.035	3.33 ± 0.22	0.089 ± 0.007
M3	12.2 ± 0.5	0.466 ± 0.039	2.05 ± 0.13	0.018 ± 0.001
M4	15.2 ± 0.6	0.357 ± 0.027	2.97 ± 0.32	0.025 ± 0.001
M6	1.76 ± 0.07	0.432 ± 0.049	3.70 ± 0.28	0.012 ± 0.002
M7	5.25 ± 0.21	0.473 ± 0.039	1.70 ± 0.16	0.040 ± 0.007
M8	3.21 ± 0.13	0.287 ± 0.024	2.96 ± 0.27	0.097 ± 0.006
M9	4.90 ± 0.20	0.715 ± 0.065	1.83 ± 0.14	0.033 ± 0.005
M10	5.96 ± 0.24	0.518 ± 0.038	2.15 ± 0.16	0.28 ± 0.02
M11	1.44 ± 0.06	0.466 ± 0.044	4.74 ± 0.41	0.027 ± 0.002
M12	5.57 ± 0.22	0.561 ± 0.042	2.33 ± 0.16	0.38 ± 0.02
M13	2.46 ± 0.10	0.370 ± 0.030	3.50 ± 0.27	0.015 ± 0.002
M14	3.06 ± 0.12	1.42 ± 0.16	5.16 ± 0.44	0.27 ± 0.02
M15	4.28 ± 0.17	0.515 ± 0.046	2.32 ± 0.17	0.032 ± 0.004
M16	13.0 ± 0.5	0.726 ± 0.046	1.15 ± 0.07	0.027 ± 0.002
M17	2.60 ± 0.10	0.549 ± 0.063	3.31 ± 0.26	1.14 ± 0.09
M18	13.8 ± 0.6	0.852 ± 0.073		0.021 ± 0.002
M19	2.80 ± 0.11	1.18 ± 0.11	2.99 ± 0.27	0.18 ± 0.01
M20	8.87 ± 0.35	0.464 ± 0.032	3.45 ± 0.25	0.036 ± 0.004
M21	4.34 ± 0.17	1.48 ± 0.19	1.32 ± 0.12	1.22 ± 0.08
M22	2.87 ± 0.11	0.683 ± 0.066	3.41 ± 0.27	0.114 ± 0.007
M23	8.42 ± 0.34	0.663 ± 0.063	2.05 ± 0.23	0.035 ± 0.004

secular equilibrium, showing the higher solubility of  $^{226}\text{Ra}$  in waters compared to  $^{238}\text{U}$ , whereas in samples with higher concentration both radionuclides are close to secular equilibrium, showing the artificial release of these radionuclides. Similar results are obtained from the study of the  $^{210}\text{Po}/^{226}\text{Ra}$  activity ratio in Fig. 2b. Thus, this ratio decreases when the  $^{226}\text{Ra}$  concentration increases in the soil, which means that in undisturbed soils,  $^{210}\text{Po}$  is found in excess (mainly through atmospheric deposition of  $^{210}\text{Pb}$ ), whereas in contaminated soils both radionuclides are again close to secular equilibrium.

In the case of *Spartina Densiflora* samples,  $^{226}\text{Ra}$  activity concentrations range from 1.44 to 15.2  $\text{mBq g}^{-1}$ , with a distribution pattern similar to that found in the case of other natural radionuclides (Martínez-Aguirre *et al.*, 1997).  $^{226}\text{Ra}/^{238}\text{U}$  activity ratios below unity are found, exception being samples M14, M19 and M21, with values generally below those in soils, with a clear excess of  $^{238}\text{U}$  in all samples.  $^{210}\text{Po}$  is clearly in excess in all the plants, with  $^{210}\text{Po}/^{226}\text{Ra}$  well above unity in most cases.



**Fig. 2.** (a)  $^{226}\text{Ra}/^{238}\text{U}$  and (b)  $^{210}\text{Po}/^{226}\text{Ra}$  ratios vs.  $^{226}\text{Ra}$  concentrations (mBq g<sup>-1</sup>) in soil samples.



The model requires a plant/substrate transfer coefficient, referred to as a concentration ratio ( $C_R$ ), which describes the amounts of nuclide expected to enter a plant from its substrate and is defined as the concentration of a nuclide in the plant divided by the concentration of the same nuclide in the substrate soil,  $C_R = C_p/C_s$ .

The individual  $C_R$  data range from 0.01 to 1.2, with an average plant to soil ratio of 0.1835. Whitehead *et al.* (1971) report an average plant to soil ratio of 0.815 and Ng (1982) report values ranging from  $7 \times 10^{-5}$  to 0.75. Ashkinazi (1980) found  $C_R$  ranging from  $10^{-3}$  to 0.4 according to the type of soil, the highest being associated with sandy soils. In our case and with the exception of samples M17 and M21 which have  $C_R$  values of 1.14 and 1.22, respectively,  $C_R$  values below 0.38 have been found. In fact, according to what was found by Ashkinazi, those two samples have a larger content of sandy particles, which is reflected in their higher density and lower organic content. Thus, with exception of these two soil samples, an average  $C_R$  value of 0.084 (range 0.009–0.38) is found. In general, our  $C_R$  values agree with the ranges of  $C_R$  values given in the literature.

The  $^{226}\text{Ra}$  activity concentration in the plant is linearly related to the  $^{226}\text{Ra}$  in the substrate soil. This line can be modelled by the equation

$$C_p = A + BC_s. \quad (6)$$

By fitting the data,  $A = 2.84 \pm 0.53$  and  $B = 0.017 \pm 0.002$  (errors  $1\sigma$ ) with a correlation coefficient of 0.9042. However, since the  $y$  intercept is clearly different from zero, the  $C_R$  for  $^{226}\text{Ra}$  is clearly dependent on the soil concentration. Moreover, for a zero concentration in the substrate soil, the concentration in the plant would be  $A$ . Due to this fact, the linear relationship must be rejected. Another approach is the hyperbolic dependence of the concentration in the plant with the concentration in the soil. This means fitting a function of the form

$$C_p = aC_s^b \quad (7)$$

in our case giving  $a = 1.95 \pm 0.30$  and  $b = 0.25 \pm 0.05$  (errors  $1\sigma$ ) with a correlation coefficient of 0.8007.

By using the above equation, the  $C_R$  can be modelled by the function

$$C_R = \frac{C_p}{C_s} = 1.95C_s^{-0.75}. \quad (8)$$

Thus, for a given concentration in the substrate soil, the transfer of  $^{226}\text{Ra}$  to the plant can be estimated by the above equation (see Fig. 3).

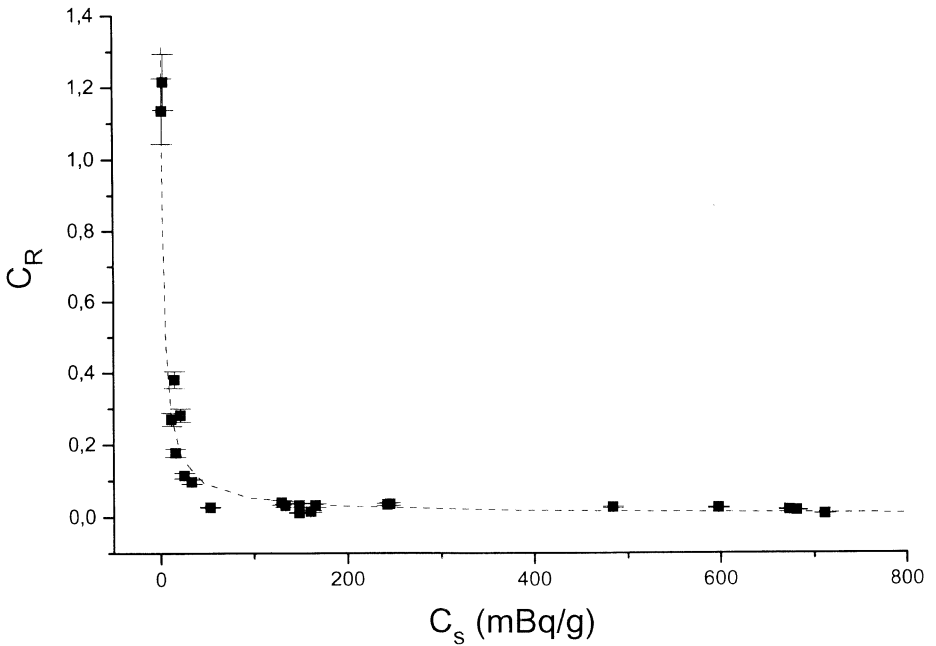


Fig. 3.  $C_R$  vs. concentration in the soil ( $\text{mBq g}^{-1}$ ).

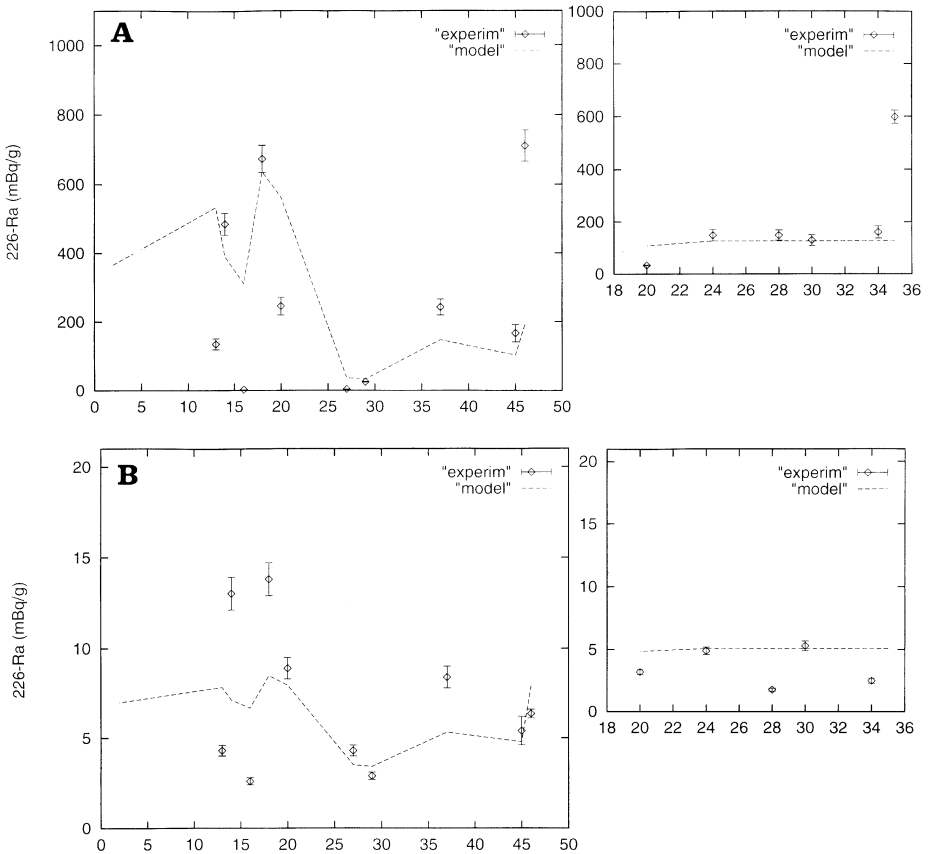
### Application of the model

The parameters for the model have been obtained from calibration exercises (diffusion coefficients), literature (mean density and radius of suspended particles, mixing depth in the sediments) or measurements in the Odiel estuary (bulk density of the sediment, dry weight fraction of small particles in the sediment). Details can be seen in Periañez and Martínez-Aguirre (in press). The transfer coefficients for  $^{226}\text{Ra}$  were obtained from laboratory experiments (Periañez, 1995) and the concentration ratio from this work. The parameters involved in the model, used to simulate the dispersion of  $^{226}\text{Ra}$  (with the exception of kinetic transfer coefficients and the source term of radionuclides from the fertilizer complex), are those that were used to simulate the U, Th and Po dispersion. Of course, the mean radius of suspended matter particles, for instance, does not depend upon whether we are dealing with U, Th, Po or Ra.

In Fig. 1, the points where sediment (Martínez-Aguirre *et al.*, 1996), soil and plant samples were collected are shown. The source of radionuclides to the river (the point where the fertilizer complex releases its wastes) is located in compartment (9, 29) (see Fig. 1). The magnitude of the source term is not

known. Thus, the source term magnitude was changed, by trial and error, until the model reproduced the experimental data. Experimental results represent the mean, averaged over 30 years, accumulation of radionuclides in soils and plants growing in such soils. Thus, we use a mean source term, which is constant in time. This, together with a mean suspended matter distribution and a residual water circulation, produces a mean distribution of radionuclides in soils and plants.

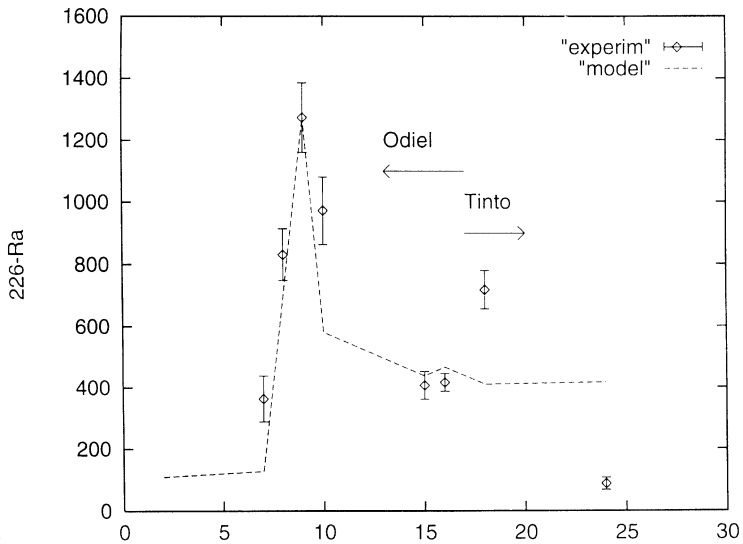
The magnitude of the source term was taken as  $5.7 \times 10^5$  and  $6.9 \times 10^5$  Bq per time step for dissolved and particulate radionuclides, respectively. This gives an annual input of  $6.6 \times 10^{12}$  Bq, which is of the same order of



**Fig. 4.** (a) Experimental (points) and model (line)  $^{226}\text{Ra}$  concentrations in soils in  $\text{mBq g}^{-1}$ . The x-axis represents the position in the grid. The smaller square shows samples collected along the Mojarrera Channel (see Fig. 1). (b) Same as (a) but for plant samples.

magnitude as the estimate of Van der Heijde *et al.* (1988), who calculated that  $1.6 \times 10^{12}$  Bq of  $^{226}\text{Ra}$  are annually discharged to an estuary in the Netherlands from phosphate rock processing plants.

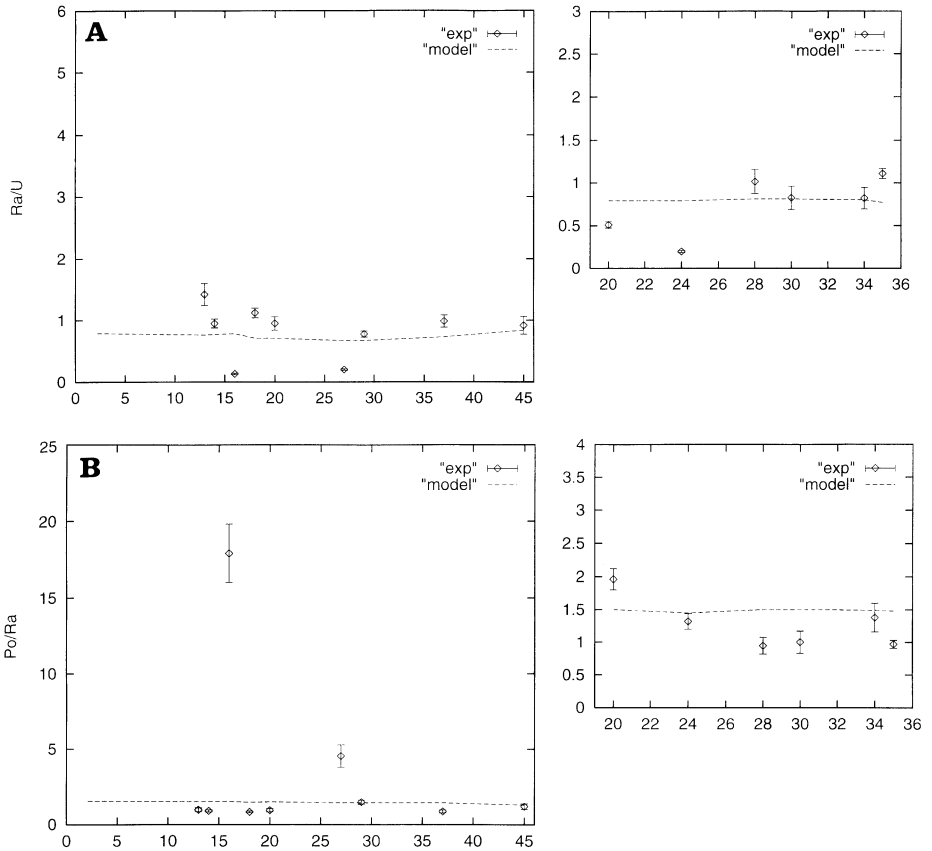
Experimental and computed  $^{226}\text{Ra}$  concentrations in soils and plants along the estuary are shown in Fig. 4(a) and (b), respectively, in which the  $x$ -axis is the position in the grid (compartment number) measured from south to north. Thus, the lines are specific activity profiles along the sections of the estuary following the points where samples were collected. As can be seen in both the figures, the general activity levels in soils and plants are reproduced by the model. Experimental and model results are, however, different for some samples. This may be an effect of taking mean parameters in the model: for some points, the bulk density of the sediment and the dry weight fraction of small particles in the sediment, for instance, may be different from the mean if there is a large sand content. This is the case with samples whose  $x$  coordinates are 13 and 16 in Fig. 4(a) and (b). On the other hand, it is assumed that the soils are flooded during half the tidal cycle (Periáñez and Martínez-Aguirre, in press): some soils will be under water almost all the tidal cycle while other soils will be flooded only during spring tide situations. Due to these considerations, we must compare the model and experimental results in terms of ‘general levels’ of activity.



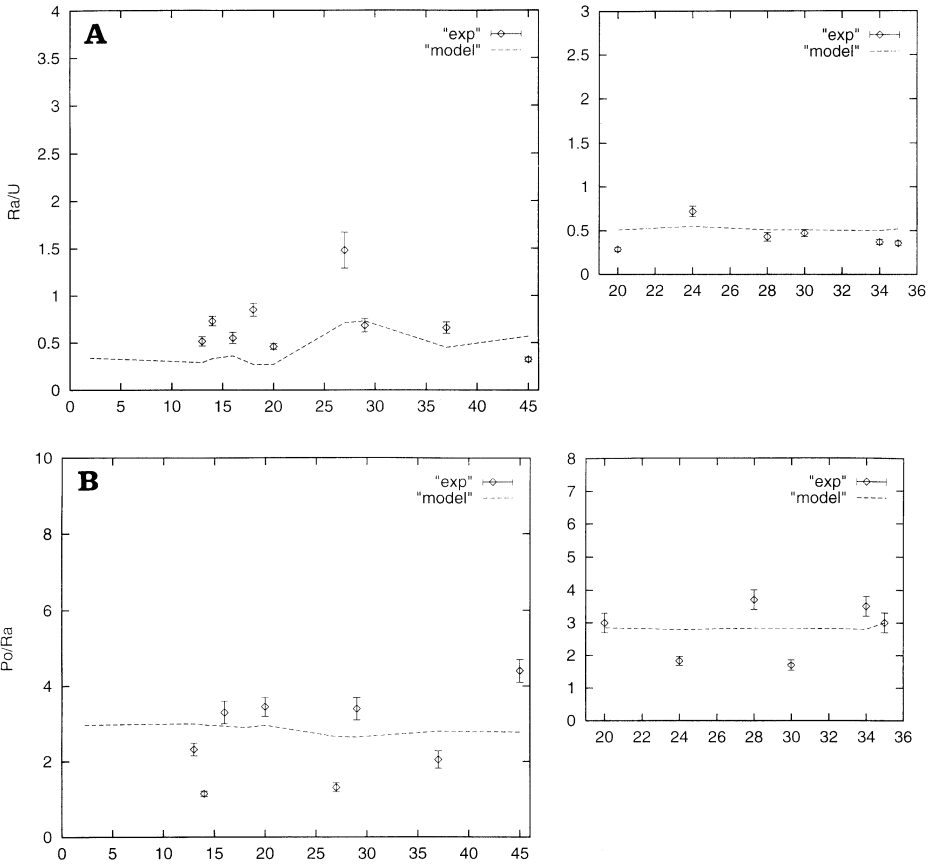
**Fig. 5.** Experimental (points) and computed (line)  $^{226}\text{Ra}$  concentration ( $\text{mBq g}^{-1}$ ) in bottom sediments collected from the Odiel and Tinto rivers. The  $x$  axis is the position in the grid.

Experimental and model results for bottom sediments are shown in Fig. 5. Experimental results were taken from Martínez-Aguirre *et al.*, (1996). In Fig. 5 the x-axis gives the position in the grid from west to east and the line is an activity profile following the points where sediments were collected. As can be seen, model results are in good agreement with experimental results.

Some activity ratios have also been investigated with our model. In Fig. 6(a) and (b) the experimental and computed  $^{226}\text{Ra}/^{238}\text{U}$  and  $^{210}\text{Po}/^{226}\text{Ra}$  ratios, respectively, in soils are given. Model results for  $^{238}\text{U}$  and  $^{210}\text{Po}$  have been taken from Periañez and Martínez-Aguirre (in press). As can be seen, the model gives a good approximation to the mean levels of such ratios along the river. Fig. 7(a) and (b) are similar to Fig. 6(a) and (b) but for plant samples. Again, the model reproduces the general behaviour of activity ratios along the estuary.



**Fig. 6.** Measured and computed (a)  $^{226}\text{Ra}/^{238}\text{U}$  and (b)  $^{210}\text{Po}/^{226}\text{Ra}$  ratios in soil samples.



**Fig. 7.** Same as Fig. 6 but for plant samples.

## CONCLUSIONS

The presence of  $^{226}\text{Ra}$  in soil and plant samples collected from a marsh area affected by phosphate fertilizer processing has been investigated. It seems that the operation of the complex, clearly, enhances the activity levels in the marsh. Concentration ratios between soil and plant have been calculated and some interesting activity ratios have also been investigated. Concentration ratios are, in general, in agreement with values found in current literature. It has been observed that the concentration ratios depend on the specific activity in the soil through an hyperbolic relation. The  $^{226}\text{Ra}/^{238}\text{U}$  and  $^{210}\text{Po}/^{226}\text{Ra}$  activity ratios are also dependent on the concentration in the soil: secular equilibrium exists in both cases in contaminated soils.

A numerical model has been applied to the Odiel marsh. The model includes the processes of contamination of bottom sediments, soils and plants. The model reproduces the general distribution pattern of  $^{226}\text{Ra}$  in soils and plants from the marsh. In the case of bottom sediments, results in good agreement with observations are obtained. The activity ratios have also been investigated with the model, giving results, in general, in agreement with measurements.

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