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Modelling the Dispersion of Non-Conservative Radionuclides in Tidal Waters — Part 2: Application to ²²⁶Ra Dispersion in an Estuarine System

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ABSTRACT

The Odiel river is an estuary system located in the south-west of Spain and affected by tidal dynamics. A phosphate fertilizer processing plant releases part of its wastes directly into the Odiel river. The model described in a previous paper (J. Environ. Radioactivity, **31**(2), 127–141) has been applied to study ²²⁶Ra (discharged from the fertilizer plant) dispersion in the Odiel river. The model adequately reproduces the observed behaviour of ²²⁶Ra in suspended matter and the dissolved phase along the Odiel river, as well as the general behaviour of the measured distribution coefficients. Some predictive studies as well as the corresponding sensitivity studies are also included in this paper. Copyright © 1996 Elsevier Science Limited

1 INTRODUCTION

In the first part of this work, which was presented in a separate paper (Periáñez *et al.*, 1996), we described a mathematical model to study non-conservative radionuclide dispersion in a tidal system in conditions of disequilibrium for the ionic transfers.

Within each grid cell, four phases or sub-compartments are distinguished: water, suspended matter and two fractions of sediments (a smalland a large-grain fraction). Non-conservative radionuclides are exchanged among these four phases. The processes involved and the equations for the time evolution of the activity in each phase were presented in Periáñez *et al.* (1996).

In this paper we present the validation of the model. It has been applied to study ²²⁶Ra dispersion in the Odiel river, a tidal estuary located in the south-west of Spain. A phosphate fertilizer plant releases part of its wastes directly into the Odiel river, thus anomalously high ²²⁶Ra concentrations were measured in water (Periáñez & García-León, 1993) and suspended matter (Periáñez *et al.*, 1994*a*) samples collected from the river. As can be seen in the above-mentioned references two sampling campaigns were performed, one during July 1990 (dry season) and another during March 1991 (wet season). For each campaign, samples were collected for both high and low water to study the influence of tidal oscillations on activity concentrations. Distributed coefficients k_d were also measured for each sample.

We will reproduce the experimental results by applying the model described in Part 1 of this paper. As will be seen, an acceptable agreement between both sets of data is achieved, which shows the appropriateness of the description of the ionic exchanges. Moreover, the model is used to study the process of sediment contamination. Results obtained are in reasonable agreement with experimental measurements of ²²⁶Ra in sediments.

In the next section, values for the different parameters involved in the model are given. These parameters are summarized in a table in Part 1 of this paper. Results are presented in Section 3. Finally, some sensitivity tests have been performed to study the model response to variations in parameters involved in the model. They are presented in Section 4.

2 PARAMETERS FOR THE MODEL

The part of the river which is studied is divided into 456 grid cells. The spatial resolution of the model is dx = dy = 100 m. The time step is fixed as dt = 6 s. Resolution of the model was selected so as to satisfy a stability condition (the CFL criterion) and to minimize numerical dispersion (Periáñez *et al.*, 1994*b*). The calibration and validation of the hydrodynamic model, as well as the dispersion equation are also presented in Periáñez *et al.* (1994*b*). There the bed friction coefficient, diffusion coefficients, and boundary conditions for hydrodynamics and dispersion are defined.

As an example, water elevations and velocity fields computed by the model when the water level is increasing and decreasing in a situation of medium tides are shown in Figs 1(a) and (b), respectively. The grid used in the model can also be seen in that figure, as well as the location of the estuary.

The parameters for the description of suspended matter dynamics are: the deposition velocity, the resuspension velocity, critical resuspension and deposition velocities and the dry weight fraction, f, of small particles (diameter $\phi < 62.5 \,\mu$ m) in sediments. The last parameter has been obtained from field measurements (Universidad de Sevilla, 1991) and the others are defined in Periáñez *et al.* (1996), where suspended matter distribution and sedimentology of the Odiel river are studied. In a later figure the sedimentation rates (averaged over some tidal cycles) can be seen along the river.

The bulk density of the sediment has also been obtained from field measurements (Universidad de Sevilla, 1991). The density of particles in suspension is taken as $\rho = 2.6 \,\mathrm{g \, cm^{-3}}$, which is the established value for the density of soil particles (Baver *et al.*, 1972) and this value should not change from one soil to another.

The mixing depth L has been taken as L = 0.1 m, following previous modelling work (Abril & García-León, 1993).

As commented in the first part of this work, the small grain-size fraction of sediments is composed of particles with $\phi < 62.5 \,\mu\text{m}$ and a step function was adopted, as a first approach, for the size distribution of such particles. Thus, the mean radius, *R*, of suspended matter particles and small grain-size particles in the sediment is taken as $15 \,\mu\text{m}$ as a first approach, since grain-size distributions are not known. The mean radius of large grain particles was obtained from field measurements (Universidad de Sevilla, 1991). Sediment samples were passed through three different sieves of well-known pore size. By knowing the mass fraction retained by each sieve, the mean radius of sediment particles can be estimated. This value ranges from $R_1 = 350 \,\mu\text{m}$ to $R_1 = 450 \,\mu\text{m}$ depending on the sampling point. Consequently, we have used the mean value, which is $R_1 = 440 \,\mu\text{m}$.

The coefficients χ_1 and k_2 , which govern transfers among the dissolved and solid phases, were obtained from laboratory experiments. They consist of tracing, at the laboratory, unfiltered Odiel river water with ¹³³Ba; a γ -emitter whose chemical behaviour is very similar to that of Ra. The conditions in the laboratory reproduced natural conditions (temperature, salinity, pH, movement of water). The time increase of the ¹³³Ba activity in suspended matter was to be measured to obtain the above-mentioned coefficients. Details can be seen in Periáñez (1995). The obtained values are $\chi_1 = 0.55 \times 10^{-7} \,\mathrm{m \, s^{-1}}$ and $k_2 = 8.17 \times 10^{-6} \,\mathrm{s^{-1}}$. Nevertheless, as these values may change slightly from one season to



Fig. 1. Grid used in the model and map showing the location of the estuary. The dimensions of compartments are $\Delta x = \Delta y = 100$ m. (a) Water elevation and velocity map when water level is increasing in a medium tide situation. Step between lines is 3 mm. (b) When water level is decreasing. Step between lines is now 5 mm.

another because of variations in water salinity and temperature, some sensitivity tests are presented in Section 4 to study the model response to changes in these parameters.

The geometrical accessibility factors for sediments have been obtained from a calibration exercise. The adopted values are $\psi = 0.01$ and $\psi' = 0.005$ since good results are obtained with them. Nevertheless, a sensitivity test is presented in Section 4 to study the model response to changes in these parameters.

Boundary conditions for activities in water and suspended matter are the same as those used when the advective-diffusive dispersion equation was calibrated (see Periáñez *et al.*, 1994*b*). In both fractions of sediments, activities in the last row of the grid (in both the northern and southern borders) were made equal to the activity in the previous row.

As the rate of change of activity in sediments is slow, there are no significant changes during periods of some days (typical simulated times). Consequently, we use experimental values of 226 Ra activity concentrations in sediments as initial conditions, instead of considering zero activities at t = 0. The experimental 226 Ra values are taken from Martínez-Aguirre *et al.* (1994).

3 MODEL RESULTS

3.1 ²²⁶Ra dispersion

The source terms of dissolved and particulate 226 Ra to the river were unknown. Thus, the input rates were changed, by trial and error, until the model reproduced the experimental data. The source is located in compartment (7, 23), since high activity peaks were measured at this point (Periáñez & García-León, 1993; Periáñez *et al.*, 1994*a*).

The results of the model for the 1990 sampling campaign are shown in Fig. 2, both for high and low water samples. Lines are the computed activity concentrations in dissolved phase $(mBq l^{-1})$ and suspended matter $(mBq g^{-1})$ and points are the corresponding experimental measurements. The x-axis is the location of the sampling station along the river (compartment number).

High and low water samples were collected with a time difference of 18 h between them. Generally speaking, activity peaks should correspond to discharges of 226 Ra which occurred shortly before sampling. When a flat distribution is measured, it should correspond to the activity which remained in the river after the homogenization of discharges performed earlier than sampling.



Fig. 2. Model results for the high (A) and low (B) water samples corresponding to the 1990 campaign. Radium-226 concentrations in water are expressed in mBq l⁻¹ and in suspended matter they are expressed in mBq g⁻¹. Points are the measured concentrations and lines are the model results. The x-axis is the position in the grid.

Thus, to reproduce the sampling campaign conditions a first activity input was introduced in tidal cycle 1 and lasted 3.3 h. High water concentrations were obtained two tidal cycles later (cycle 3). A second activity input started in cycle 4 and lasted 9 h, low water concentrations were obtained from this tidal cycle. In this way, 18 h elapsed between the timepoints in which concentrations for high and low water are obtained. A more detailed discussion about the reasons for selecting this time sequence for the input rates can be seen in Periáñez *et al.* (1994*b*).

The input rates for the first activity inputs were 9.0×10^7 and 9.9×10^6 Bq per time step for the dissolved phase and the suspended matter, respectively. During the second activity input they were, for dissolved phase and suspended matter, respectively, 2.0×10^4 and 2.5×10^3 Bq per time step. The ratio between the activity discharged in dissolved and suspended forms is obtained from a trial and error exercise. The first activity input is larger than the second one since it is used to create the ²²⁶Ra background which is present in the river, although there are no discharges from the fertiliser plant.

As can be seen in Fig. 2, there is very good agreement between computed and measured activities for both phases and for both high and low water conditions. It is necessary to point out that meteorological conditions (wind speed and direction, atmospheric pressure), which affect the water dynamics, corresponding to the sampling dates were also introduced in the model (Periáñez *et al.*, 1994*b*).

Sediments are not included in this discussion because there are no apparent changes in their activity concentrations during a time period of several tidal cycles. Thus, their concentrations correspond to the initial conditions. Nevertheless, some studies involving sediments will be presented later (Section 3.2).

Measured and computed values of the distribution coefficients can be seen in Fig. 3. The general behaviour of k_d all along the river is reproduced by the model. To compute k_d values, it must be considered that the model is taking into account only the ²²⁶Ra activity which is present in the surface layer of the suspended matter particles, since only this activity will participate in the ionic exchanges. As ²²⁶Ra is a naturally occurring radionuclide it will be present in the inner part of the particles. Thus, the specific activity inside the particles must be added to the specific activity computed by the model (see Section 4.4, in Periáñez *et al.*, 1996). We have adopted a value of 45 mBq g⁻¹ for the specific activity inside particles, since this is the measured activity concentration in suspended matter far upstream of the fertilizer complex (Periáñez *et al.*, 1994*a*). Therefore, such a value can be considered as the ²²⁶Ra natural background in suspended matter. This correction has been taken into account in Figs 2 and 3.

During the 1991 sampling campaign, low water samples were collected a week later than the high water samples. Thus, there are no tidal correlations among the results. For high water samples, the activity input began at the same tidal cycle during which activity concentrations were going to be obtained. This input lasted 3h. The input rates were 1.2×10^4 and



Fig. 3. Computed (line) and measured (points) distribution coefficients (lg⁻¹) for the 1990 sampling campaign for the high (A) and low (B) water samples.

 1.5×10^3 Bq per time step for the dissolved phase and suspended matter, respectively. In Periáñez *et al.* (1994b) it was shown that the method of creating the background is not decisive in the model results. The background was created, to save CPU time, by taking initial ²²⁶Ra concentrations all over the grid. These concentrations were $10 \text{ mBq } 1^{-1}$ and 220 mBq g^{-1} for water and suspended matter, respectively. Several tidal cycles were elapsed so as to avoid correlations between high and low water concentrations. In the case of low water samples, the activity input began at the middle of the tidal cycle in which activities were going to be

obtained and lasted 3 h. Input rates were 1.2×10^3 and 4.0×10^2 Bq per time step for water and suspended matter, respectively. The specific activity in the inner part of particles for this sampling campaign was again 45 mBq g^{-1} .

The results for the 1991 sampling campaign are shown in Figs 4 (activity concentrations in water and suspended matter) and 5 (distribution coefficients). The general behaviour of activity concentrations and distribution coefficients along the river is again reproduced by the model. The decrease in suspended matter activity concentration around compartment



Fig. 4. As Fig. 2 but for the 1991 sampling campaign.



Fig. 5. As Fig. 3 but for the 1991 sampling campaign.

9 is due to the fact that a factory releases important amounts of nonradioactive material to the river at this point. This effect has been included in the suspended matter dynamics model (Periáñez *et al.*, in press). These wastes have large contents of Fe, Zn, and Pb which produce a decrease in specific content for other elements (Respaldiza *et al.*, 1993).

As seen in Figs 3 and 5, the model produces systematically lower k_{ds} than those observed in the low water samples of both campaigns at the south part of the grid (samples of compartment number 2). This effect could be corrected by taking into account the probable existence of two populations of particles: natural particles and 'hot' particles released from

the source. Hot particles contain a larger activity in the inner part than natural particles. Thus, when the border of the hot-particle cloud released from the source reaches cleaner water, a mixture of natural and hot particles is taken when collecting a sample, and then, k_d must increase. In this way, when water level is decreasing, hot particles will reach the south of the grid and k_d will increase. Predictions of the model could be improved by considering these two kinds of particles.

In conclusion, the model seems to reproduce the experimental results for two sampling campaigns performed in the Odiel river. Run time for a simulation over six tidal cycles is about 2 h on a VAX computer.

3.2 Predictive studies

With the help of the previously described model some prediction studies were carried out. Firstly, we investigated the process of radioactivity cleaning of the river. For that, a simulation over 10 tidal cycles was performed, during which there were no activity discharges into the river. Concentrations in sediments were those measured by Martínez-Aguirre *et al.* (1994) and arbitrary, although realistic, concentrations were assumed as initial concentrations for water and suspended matter. These should simulate the ²²⁶Ra background remaining in the river after the activity discharges stop. In a first trial they were 50 mBq l^{-1} in water and 25 mBq g^{-1} in the surface layer of suspended matter particles. For the central part of the particles, the above-mentioned value of 45 mBq g^{-1} was taken.

After the simulation, it was verified that no apparent changes in sediment activity concentrations appeared for such a time period. This revealed that the study of sediment cleaning has to be performed by using residual (averaged) water circulation, which allows a larger time step. In this way, simulations over several years may be carried out.

The results for time evolution of activity concentrations in water and suspended matter and distribution coefficients can be seen in Figs 6(a) and (b), respectively, for a compartment in the central part of the grid. The concentrations in both phases follow tidal oscillations. Distribution coefficients also oscillate following tides, showing that they depend on the tidal state.

It is interesting to note that activity concentrations increase, both for water and suspended matter, although in this case there were no external sources of ²²⁶Ra. These activities must arise from sediments, which are cleaned very slowly. Activity in water and suspended matter will increase until extreme conditions such as spring tides, heavy rains or strong winds clean the river. Afterwards, more ²²⁶Ra is extracted from the sediments



Fig. 6. (a) Time evolution of ²²⁶Ra concentrations in water, in mBq 1^{-1} (continuous line), and the surface layer of suspended matter, in mBq g^{-1} (dotted line) for a compartment in the middle of the grid. (b) Time evolution of the distribution coefficient $(1g^{-1})$ in the same compartment.

until the next extreme conditions clean the river again and so on. Thus, it seems that to study the sediment cleaning process, extreme conditions should be included in the model.

These results have been obtained with arbitrary initial ²²⁶Ra concen-



Fig. 7. Same as Fig. 6 but starting from different initial concentrations for water and the surface layer of suspended matter.

trations in water and suspended matter. In Fig. 7 we present the same simulation but with other initial concentrations: $50 \text{ mBq } 1^{-1}$ and 100 mBq g^{-1} for water and suspended matter, respectively. As happened before, the concentrations in both phases oscillate following tides and concentration in water increases again. However, it is interesting to note that, at the end of the simulation, k_{ds} have reached the same values as in

the previous case. Thus, it seems that the distribution coefficients tend to an equilibrium value. The relevant question now, is what is such a k_d equilibrium value?

At equilibrium conditions, the radionuclide transfer from water to suspended matter must be equal to transfer from suspended matter to water. So, following our formulation it is:

$$k_1 A_d = k_2 A_s \tag{1}$$

where A_d and A_s are total activities in water and suspended matter, respectively. If these activities are converted into activity concentrations (C_d, C_s) we have:

$$k_{\rm d} = \frac{C_{\rm s}}{C_{\rm d}} = \frac{1}{m} \frac{k_1}{k_2}$$
(2)

where *m* is the suspended matter concentration. If we consider that the value for k_1 is:

$$k_1 = \chi_1 \frac{3m}{\rho R} \tag{3}$$

(eqn (15) in Periáñez et al., 1996) then we have that:

$$k_{\rm d} = \frac{\chi_1}{k_2} \frac{3}{\rho R} \tag{4}$$

By substituting the different values, we have $k_d = 0.52 \ \text{lg}^{-1}$. Nevertheless, it must be taken into account that when laboratory experiments for determining the transfer coefficients were performed, all the tracer activity added at the beginning could be exchanged, since it was a manmade radionuclide (not present in the central part of particles). The $k_{\rm d}$ calculated by the model includes the contribution of that central activity, since this is the k_d which would be obtained from experimental measurements. The specific activity in the central part of the particles is $45 \,\mathrm{mBg \, g^{-1}}$ and the activity concentration in water at the end of the simulation is 80 mBq l^{-1} (see Fig. 7(a)), then the ratio among these two values (which is $0.56 lg^{-1}$) must be subtracted to obtain the computed k_d : $k_{\rm d} = 1.1 - 0.56 = 0.54$ lg⁻¹. This value is in very good agreement with that obtained from the former equation. It can be concluded that after the simulation time, ionic exchanges among water and suspended matter have reached equilibrium (with slight deviations produced by tidal oscillations). Moreover, the equilibrium vales of k_d agrees, within an order of magnitude, with that defined in current literature. Indeed, in IAEA (1985) it can be seen that ²²⁶Ra k_d in coastal water ranges from 0.5 to 50 lg⁻¹, with a mean value of $5 \lg^{-1}$, which is very close to $1 \cdot 1 \lg^{-1}$.

The contamination process of the sediment has also been studied. The model was started from idealistic conditions in which water, suspended matter and sediments were free from ²²⁶Ra contamination. A continuous activity input to water and suspended matter was then performed at a rate equal to the mean input rates used to reproduce experimental results of the 1990 and 1991 sampling campaigns $(4.2 \times 10^3 \text{ and } 9.9 \times 10^2 \text{ Bq per time step for water and suspended matter, respectively).}$

The activity concentration evolution in sediments of the compartment into which the discharges are performed can be seen in Fig. 8. Concentration increases almost linearly. Indeed, a slope of $1.25 \times 10^{-4} \,\mathrm{mBq \, g^{-1}}$ min⁻¹ can be obtained with a correlation factor r = 0.9985. The activity concentration measured at this point is about 1400 mBq g⁻¹ (Martínez-Aguirre *et al.*, 1994), then a time of about 21 years would be necessary to reach that concentration (if the input rate persists over time). Industrial activity started in the middle of the sixties, thus, it seems that the model produces a contamination rate which is faster than the real one. Nevertheless it must be taken into account that extreme conditions (such as spring tides, strong winds, heavy rains or even dredgings) have not been included in the model, and these conditions can produce eventual cleaning of sediments. Moreover, the rates of discharge will not have been constant over all this time.

A ²²⁶Ra concentration map in sediments after a simulation over eight



Fig. 8. Time evolution of 226 Ra concentrations (mBq g⁻¹) in the sediment of the compartment where discharges are performed.

tidal cycles can be seen in Fig. 9(B). This concentration pattern has been compared with that measured by Martínez-Aguirre *et al.* (1994). Of course, we must compare the relative variations from one point to another, and not the absolute values. Results from this comparison can be seen in Table 1. Computed relative variations of concentrations to the point of discharges show very good agreement with measured variations. Thus, the model clearly reproduces the process of contamination of the sediments.



Fig. 9. (A) Averaged sedimentation rates $(g cm^{-2} year^{-1})$ over several tidal cycles all over the Odiel river. (B) Radium-226 concentration map in the sediment $(mBq g^{-1})$ obtained eight tidal cycles after the beginning of the discharges. Measured concentrations $(mBq g^{-1})$ and the sampling points can also be seen.

| TABLE 1 |
|---|
| Comparison Between Measured and Computed |
| Variations of ²²⁶ Ra Concentrations in the Sedi- |
| ment Relative to the Point where Discharges are |
| Performed |

| Measured | Computed |
|-----------------|------------------|
| 1400/40 = 35 | 0.5/0.015 = 33.5 |
| 1400/750 = 1.86 | 0.5/0.3 = 1.67 |
| 1400/225 = 6.2 | 0.5/0.1 = 5 |

4 SENSITIVITY TESTS

Some sensitivity tests have been performed to study the model response to changes in the parameters. As the model response to hydrodynamic parameters, diffusion coefficients, boundary conditions, suspended matter dynamic parameters, and source terms has already been studied (Periáñez *et al.*, 1994*b*; in press), only the sensitivity tests to new parameters (involved in the ionic transfers description) will be presented. These parameters are the transfer coefficients χ_1 and k_2 and the geometrical accessibility factors.

The model response to changes of the accessibility factors can be seen in Fig. 10. Results for low water of the 1991 sampling campaign are shown. As can be seen, there are no important changes in activity concentrations in suspended matter when such factors are decreased (1) or increased (2) by a factor 10. When they are increased, the larger variation corresponds to the dissolved phase since in this case there is a larger interaction among water and sediments. This variation suggests that k_d values increase far away from experimental values in the north part of the estuary (Fig. 10(B)).

The laboratory experiments to determine the ²²⁶Ra transfer coefficients were performed in controlled conditions which reproduce natural conditions (pH, salinity, etc). Such conditions may change slightly from one season to another. Therefore, a sensitivity test was carried out to study the model response when the transfer coefficients are slightly changed. Results are shown in Fig. 11. Firstly (1), the coefficient χ_1 has been increased by a factor 2 and k_2 has been decreased by the same factor so as to enhance the transfer from the water to the solid phases. In the second case (2), k_2 was increased and χ_1 decreased by a factor 2 so as to enhance the transfer towards water. In Fig. 11 it can be seen that in the first case there is an increase in suspended matter activity concentrations and in the second case there is a reduction in such concentrations. Of course, the larger activity concentration in water is obtained in case (2), and the smaller in case (1). The effect of these variations on the k_d can be seen in Fig. 11(B). In case (1), k_d are over the experimental values and in case (2) they are too small.

5 CONCLUSIONS

A 2D 4-phase model to study the dispersion of non-conservative radionuclides in an estuarine system has been developed. The model includes a wide set of complex processes. The transfers among water and solid phases have been formulated in terms of transfer coefficients, instead of



Fig. 10. Model response to changes in the geometrical accessibility factors. (A) Changes in activity concentrations in water and suspended matter. (B) Changes in k_d . See text for explanation of (1) and (2).

 $k_{\rm d}$. And this is because the small time step involved in calculations does not allow the transfers to reach equilibrium. With this new formulation processes at the scale of tidal flow can be simulated. Nevertheless, two transfer coefficients instead of one $k_{\rm d}$ must be known; but the possibility of predicting $k_{\rm d}$ values, we think, clearly compensates for this difficulty. The model has been applied to ²²⁶Ra dispersion in an estuarine system

The model has been applied to ²²⁰Ra dispersion in an estuarine system in the south-west of Spain, where a phosphate fertilizer processing plant



Fig. 11. Model response to changes in the transfer coefficients. (A) Changes in activity concentrations in water and suspended matter. (B) Changes in k_d . See text for explanation of (1) and (2).

releases its wastes. Experimental measurements of ²²⁶Ra concentrations in water, suspended matter, and distribution coefficients have been compared to model predictions, showing reasonable agreement between both sets of data for two sampling campaigns.

Some studies concerning scdiments have been carried out. Sediment decontamination has shown to be a very slow process and strongly influenced by extreme conditions, which should be included in the model in a further step. On the other hand, the model has been able to reproduce the sediment contamination process in very good agreement with available experimental observations.

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