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STUDIES ON THE DISPERSION OF RADIONUCLIDES IN AQUATIC SYSTEMS: MODELLING AND LABORATORY EXPERIMENTS

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ABSTRACT

In this paper a mathematical model which describes the dissemination of non-conservative radionuclides in an aquatic tidal system is presented. Among other things, it is useful to estimate the radioactive environmental impact of two phosphoric acid factories located in an estuary at the Southwest of Spain. The ability of the model as a predictive tool is analysed. Further extensions and applications of it are also commented in the paper.

INTRODUCTION

The huge amount of experimental observations on the presence of radionuclides in different environmental systems has to be interpreted. And this because it is apparent that the understanding of the information which resides in this ample amount of data can provide with a lot of knowledge on the behaviour of radionuclides in the environment and on the behaviour of the environment itself. This task not only requires statistical works but also mathematical modelling. With a model the Nature is described in a more or less complicated way through some mathematical equations. They try to reproduce the processes which take place in the environment. The agreement between the model outputs and the observational information supports the way under which the Nature has been described and, consequently, the assumed behaviour of, for instance, a given radionuclide in it.

The first and simpler approaches used averaged box model of long tradition in Radiopharmacology works. Their viewpoint consists of the partition of the studied environment in boxes or cells which exchange radionuclides between them at a rate given by the so-called transfer coefficients. Earlier applications of this philosophy to the marine environment considered such a coefficients as fitting parameters to merely reproduce the experimental data. Under this approach the transfer coefficient has not any physical meaning and cannot be used to describe any other set of experimental data *(Hallstadius et al. 1987)*. A further approach was able to relate the transfer coefficients to oceanographical information and the geometry of the partition of the studied environment *(Abril and García-León 1991)*. This way, the coefficients acquired a clear physical meaning as well as some kind of universality. From such very moment it was not necessary any previous radiological history of the studied site to reproduce how radionuclides could disseminate in it. Within this framework models were developed to successfully describe the behaviour of conservative and non-conservative radionuclides in the Irish Sea *(Abril and García-León 1992, 1993a and 1993b)*. They were annually averaged box models. Thus the ionic exchange mathematical formalism was based on distribution coefficients since the situation of equilibrium exchange was accomplished each time step.

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After, the problem of the behaviour of natural radioactivity in estuarine areas was afforded. This gave us the opportunity of studying the environmental impact of two phosphoric acid factories in the Southwest of Spain, whose description appears elsewhere in these Proceedings *(García-León et al. 1997)*. The time and spatial scales involved in these environments requires a different mathematical approach. Now, hydrodynamical models must be used and the description of the ionic exchanges for non-conservative radionuclides has to be done in terms of kinetic transfer coefficients. The reason behind this need resides in the fact that the equilibrium exchange cannot be achieved for the short time steps used during the calculations.

In this paper we describe this approach which has permitted the description of the behaviour and dissemination of several natural radionuclides in the estuary formed by the Odiel and Tinto rivers. It is also presented the ability of the developed model as a tool for predictive studies useful for rehabilitation and planning works in the studied environment.

CONCEPTUAL MODEL

The studied environment is divided into a number of grid cells. Four phases are present inside each grid cell: water, suspended matter and two grain size fractions of sediments. In Fig. 1 a grid cell is presented. Inside it, radionuclides can be associated either to the dissolved or suspended phase. As tides produce a continuous movement of water, radionuclides in both phases will be transported from one cell to another by advection and diffusion. As is usual *(Gurbutt et al. 1987)*, we will consider that only particles with a diameter ϕ<62.5 μm will be present in the water column as suspended matter. Larger particles will quickly sink to the bottom. Therefore, it makes sense to consider two grain size fractions in the sediment: particles with ϕ<62.5 mm, the small grain fraction, and particles with ϕ>62.5 μm, the large grain size fraction. Only the small grain size fraction can be resuspended into the water column and participate in the movement of the suspended phase. Similarly , the suspended matter deposited onto the sediment bed will be incorporated in the small grain size fraction. Thus, deposition and resuspension processes produce an exchange of radionuclides between suspended matter and the small grain size fraction of the sediment.
The dissolved phase is in contact with the other three phases. Consequently, ionic exchanges take place among them.

To computing the advection-diffusion processes, as well as the resuspension and deposition of suspended matter, the water and suspended matter dynamics must be known for the system under study. The detailed description of the model can be seen in *Periáñez et al. 1996a*.

Hydrodynamics

The water circulation is obtained from the hydrodynamic equations, whose solutions gives the instantaneous water state (water displacement from the mean level due to tides and water velocity) for each grid cell and each time step. These equations include the Coriolis term, bed friction, response to wind stress and response to changes in atmospheric pressure. Details can be seen in *Periáñez et al. 1994*.

When long time scales are involved in the studied problem a residual circulation approach can be adopted. In this case the residual circulation is obtained at each point by averaging the water velocity over a complete tidal cycle. For it field data on water velocities are used. A full account of the procedure is given in *Periáñez and Martínez-Aguirre, 1997b.*

Suspended matter dynamics

An advective-diffusive dispersion equation governs the horizontal movement of the suspended matter. The vertical movement is governed by the deposition and resuspension terms. They have been formulated using critical deposition and resuspension velocities in such a way that there is deposition only if the water velocity is smaller than the critical deposition velocity. For larger values of the water velocity the deposition is hindered by water turbulence. On the other hand, there is resuspension only if the water velocity is larger than the critical resuspension velocity, otherwise there will not be enough energy to lift up particles from the sediment. The mathematical formulation of these terms and the application of these equations to study the sedimentology of an estuary can be found in *Periáñez et al. 1996c*.

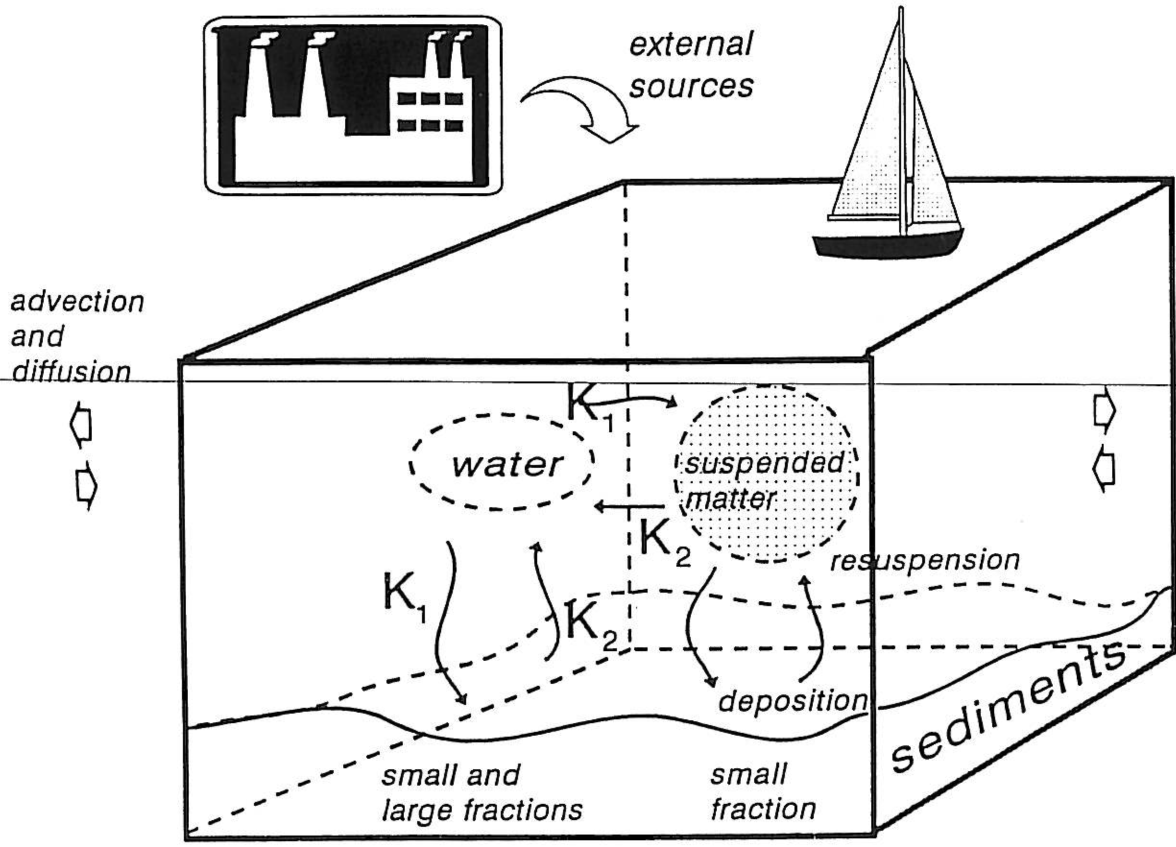


Figure 1 - Grid cell in which the radionuclide transfer processes among the four phases are presented.

Radionuclide dispersion

To complete the description of the dissemination of radionuclides along the system the processes of ionic exchanges which take place inside each grid cell must be included. This is done with four differential equations, one for each phase.

The formulation of ionic exchanges between the dissolved and the solid phases (suspended matter and both grain size sediment fractions) is done in terms of kinetic transfer coefficients instead of distribution coefficients. This is due to the fact that the short time scales involved during the simulations make unrealistic to using distribution coefficients typical of equilibrium situations.

A full account of the ionic exchange mathematical description is given in *Periáñez et al. 1996a and Abril and Fraga 1997*. Briefly a coefficient k_1 governs the transfer of radionuclides from water to the solid phase and a coefficient k_2 the transfer from the solid phase to the dissolved phase. Both coefficients can be found from laboratory experiments where the exchange between the dissolved phase and the other three solid phases are simulated. The dependence of such a coefficients on several relevant parameters, as supended matter load and water conductivity has been also done *(Laissaoui et al. 1997)*.

Computational scheme

We have developed a code to solve the equations involved in our mathematical model. The code was implemented on a VAX-VMS computer. The equations are computed according to the following sequence for each time step.

1 The hydrodynamic equations are solved so as to obtain the water elevation and velocity for each grid cell.
2 The suspended matter advective-diffusive equation is evaluated, as well as the resuspension and deposition terms. The suspended matter concentrations and the sedimentation rate are then obtained for each grid cell. To solve these equations, the instantaneous water state must be known.
3 The four equations which govern the time evolution of the radionuclide concentration in each phase are now solved for each grid cell.
4 The external sources of suspended matter, dissolved radionuclides and radionuclides in particulate form are introduced into the grid cells in which these sources exist.

The output provides detailed information: time evolution of radionuclide concentrations in each of the four phases in desired positions into the grid, activity concentration maps at desired times, etc.

RESULTS

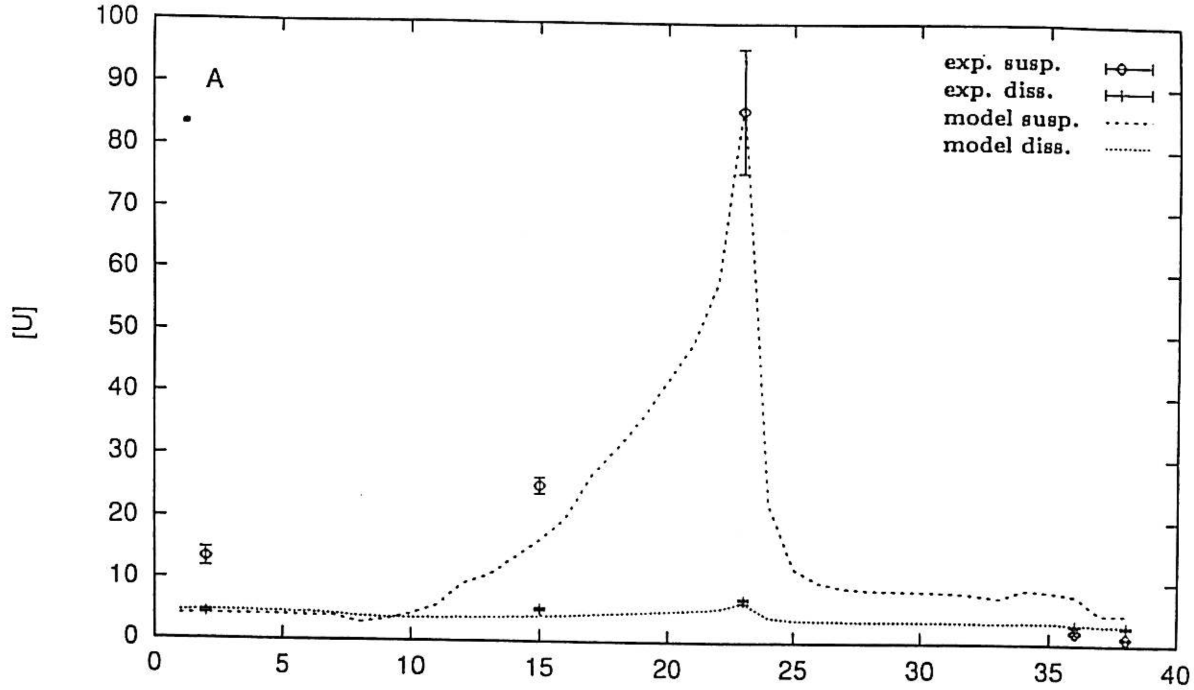
The model has been applied to study the dispersion and behaviour of some natural radionuclides in the estuarine area formed by the Odiel and Tinto river at Huelva, Southwest Spain. As can be seen in (the Proceedings) there are two phosphoric acid factories in this area which release natural radioactivity into the estuary through different ways.

As an example we give in Fig. 2 results on the concentration of ^{226}Ra and ^{238}U in water and suspended matter during high tides compared to experimental data measured by our group. The x-axis is the location of the sampling station along the river. The agreement between experimental data and computed results is good. Also by using the average residual circulation approach we habe been able to reproduce the experimental results in river and marsh sediments, as well as in marsh plants. The details can be seen in *Periáñez et al. 1996b and 1997 and Periáñez and Martínez-Aguirre 1997a*.

All this confirming that the description of the transfer processes is essentially correct.

Consequently, the model becomes a very powerful tool to performing predictive studies.

Thus, the process of radioactivity cleaning of the Odiel river was analysed. And, for instance, a simulation over 10 tidal cycles was performed, during which there was not activity discharges into the river. In Fig. 3 we present the time evolution of the ^{226}Ra activity concentration in the dissolved and suspended matter phases *(Periáñez et al. 1996a)*. They follow the tidal oscillations and seem to increase during the simulation time period. In the absence of source inputs this increase should be attributed to a very slow cleaning of the sediments. However, it was verified that no apparent changes



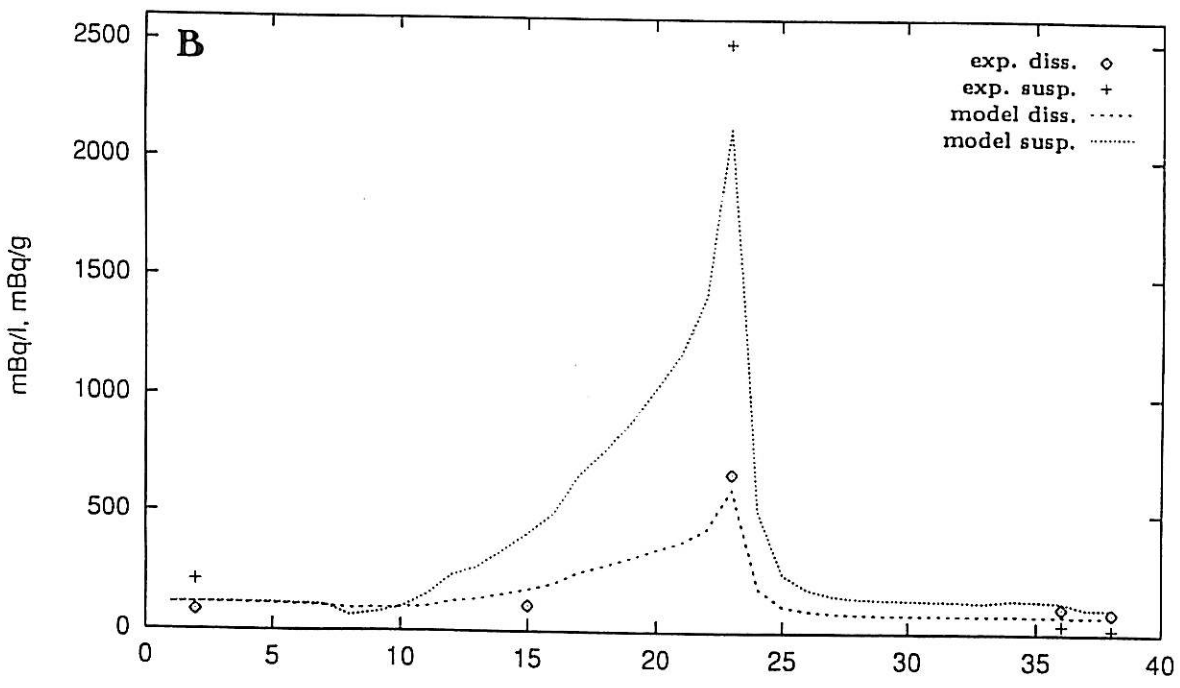


Figure 2 - Computed (lines) and measured (points) ^{238}U (A) and ^{226}Ra (B) concentrations in water and suspended matter during high water and low water conditions respectively. The x-axis is the position in the grid, being each x-unit 100 m. Water concentrations are given in mBq/L for Ra and μg/l for U. Similarly suspended matter concentrations are given in mBq/g for Ra and μg/g for U.

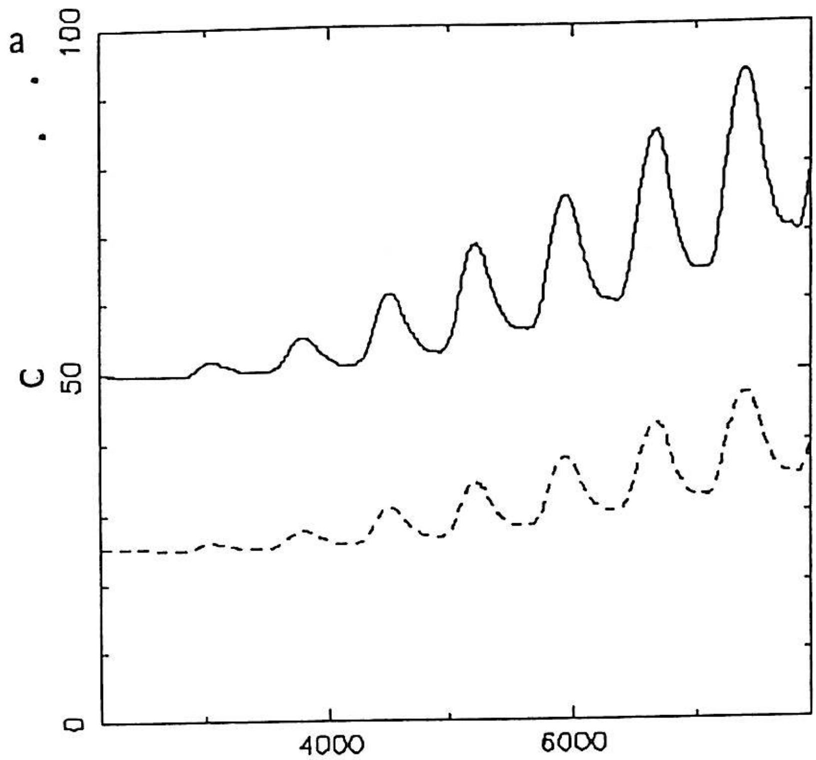


Figure 3.- Time evolution of ^{226}Ra concentrations in water, in mBq/L (continuous line), and suspended matter, in mBq/g (dotted line), for a compartment in the middle of the grid.

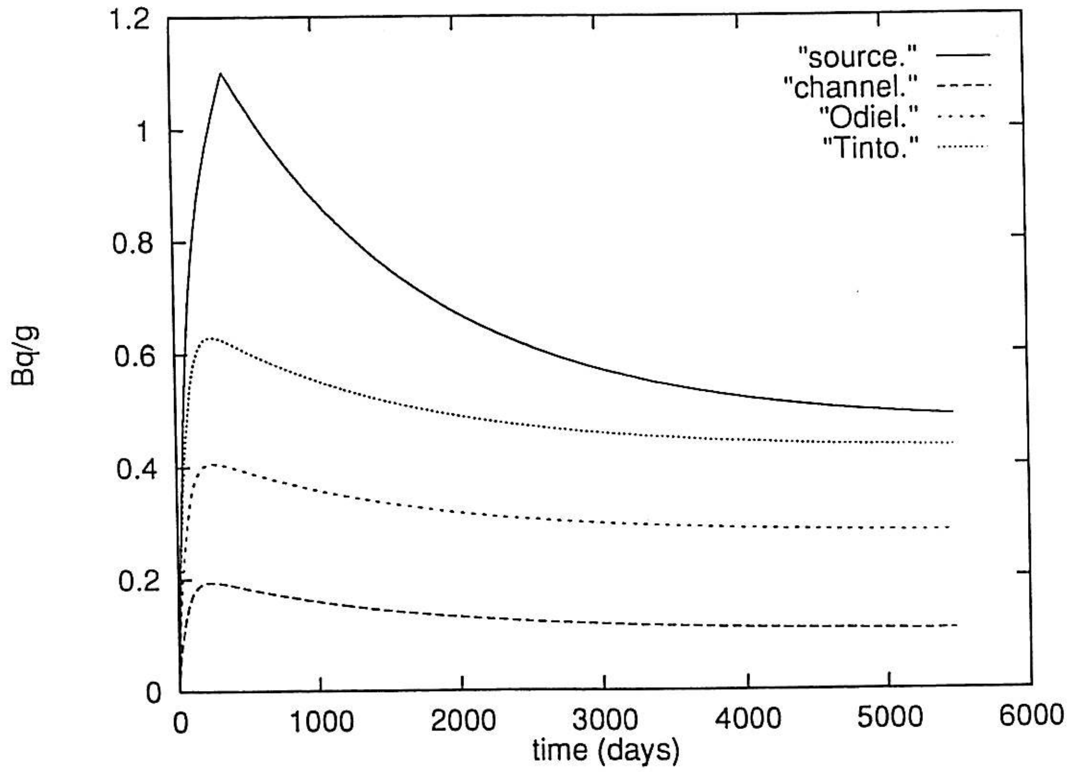


Figure 4.- Time evolution of ^{210}Po concentration in sediments in four sites of the modelled area. See text for an easier comprehension.

occurred in the sediment activity concentration during the simulation period. This revealing that the study of sediment cleaning has to be performed by using average water circulation. In this way simulations over several years can be carried out. And, indeed, this was done. In Fig. 4 a numerical experiment on the time evolution of the ^{210}Po activity concentration in sediments from 4 interesting sampling points in the estuarine area under study is depicted *(Periáñez and Martínez-Aguirre 1997b)*. Fourteen years after the ceasing of source discharges, the sediment activity concentration upstream the source in the Odiel river and in the Tinto river decreased by a 30%, a 42% in the sediment at the Odiel marsh area and by a 47% in the source itself which is located at the Odiel river *(García-León et al. 1997)*.

Many other applications have been done or are in progress. Some of them have already been referred to in the Introduction of this paper. Others have not. Among them, the description of the dispersion of conservative and non-conservative radionuclides in the English and the Suez Channels, the vertical migration of non-conservative radionuclides along deep ocean water columns, etc. With them it has been possible to study, for instance, the effect of hypothetical radioactivity releases on the ocean surface and on the sea sediment, from radioactive dumped material *(Periáñez 1997)*.

CONCLUSIONS AND FUTURE DEVELOPMENTS

A versatile mathematical model have been developed to describing the dispersion of non-conservative radionuclides in aquatic systems. Among its different applications, that of the case of the estuary formed by the Odiel and Tinto rivers at the Southwest of Spain can be underlined. There, two phosphoric acid factories are present which release natural radionuclides into the estuarine environment. The agreement between the experimental observations and model results confirm the goodness of the description of the transfer processes. This convert the model in a very interesting tool to studying the migration of radionuclides, as well as in a very powerful predictive tool for short, medium and long term problems. It seems clear that the model can help very much in optimising the wasting practices from these factories by estimating the fate, residence times, etc. of the different radionuclides in the studied system.

Acknowledgements
This work is a summary of our publications cited in the References. The finantial support of the Agencia de Medio Ambiente of the Junta de Andalucía, the European Union (Contract FI3P-CT92-0035) and ENRESA is deeply acknowledged.

REFERENCES

Abril J. M. and García-León M. (1991), *"A mathematical approach for modelling radionuclide dispersion along the marine environment"*, **J. Environ. Radioactivity** 13, 39-54.

Abril J. M. and García-León M. (1992), *"A marine dispersion for radionuclides and its calibration from non-radiological information"*, **J. Environ. Radioactivity** 16, 127-146.

Abril J. M. and García-León M. (1993a), *"A 2d 4-phases marine dispersion model for non-conservative radionuclides. Part 1: conceptual and computational model"*, **J. Environ. Radioactivity** 20, 71-88.

Abril J. M. and García-León M. (1993b), *"A 2d 4-phases marine dispersion model for non-conservative radionuclides. Part 2: two applications"*, **J. Environ. Radioactivity** 20, 89-115.

Abril J. M. and Fraga E. (1996), *"Some physical and chemical features of the variability of k_d distribution coefficients for radionuclides"*, **J. Environ. Radioactivity** 30, 253-270.

García-León M., Abril J. M., Bolívar J. P., García-Orellana I., García-Tenorio R., Martínez-Aguirre A., Periáñez R. (1997), *"Radioactive environmental impact during the phosphoric acid production"*, **these Proceedings.**

Gurbutt P. A., Kershaw P. J. and Durance J. A. (1987), *"Modelling the distribution of soluble and particle adsorbed radionuclides in the Irish Sea"*, In **Radionuclides. A tool for Oceanography**, ed. J. C. Guary, P. Gueguenia t and J. R. Pentreath, Elsevier, Oxford, UK, 395-490.

Hallstadius L., García-Montaño E. and Nilsson U. (1987), *"An improved and validated dispersion model for the North Sea and adjacent waters"*, **J. Environ. Radioactivity** 5, 261-274.

Laissaoui A., Periáñez R., Abril J. M. and García-León M., *"The effect of suspended matter concentration and water salinity in the kinetic transfer coefficients of radionuclides"*, in preparation.

Periáñez R., Abril J. M. and García-León M. (1994), *"A modelling study of ^{226}Ra dispersion in an estuarine system in South-West Spain"*, **J. Environ. Radioactivity** 24, 159-179.

Periáñez R., Abril J. M. and García-León M. (1996a), *"Modelling the dispersion of non-conservative radionuclides in tidal waters-Part 1: conceptual and mathematical model"*, **J. Environ. Radioactivity** 31, 127-141.

Periáñez R., Abril J. M. and García-León M. (1996b), *"Modelling the dispersion of non-conservative radionuclides in tidal waters-Part 2: application to ^{226}Ra dispersion in an estuarine system"*, **J. Environ. Radioactivity** 31, 253-272.

Periáñez R., Abril J. M. and García-León M. (1996c), *"Modelling the suspended matter distribution in an estuarine system. Application to the Odiel river in Southwest Spain"*, **Ecol. Modelling** 87, 169-179.

Periáñez R., Abril J. M. and García-León M. (1997), *"A four phases model to simulate the dispersion of ^{226}Ra, ^{238}U and ^{232}Th in an estuary affected by phosphate rock processing"*, **Radioprotection-Colloques** 32 C2, 79-84.

Periáñez R. and Martínez-Aguirre A. (1997a), *"U and Th concentrations in an estuary affected by phosphate fertilizer processing: experimental results and a modelling study"*, **J. Environ. Radioactivity** 35, 281-304.

Periáñez R. and Martínez-Aguirre A. (1997b), *"A 6-phases model to simulate the contamination by non-conservative radionuclides of sediments, soils and plants in a marsh area. Applications to the Odiel marsh in Southwest Spain"*, to be published in **J. Environ. Radioactivity**.

Periáñez R. (1997), *"Modelling the distribution of radionuclides in deep ocean water columns. Applications to ^{3}H, ^{137}Cs and $^{239+240}$Pu"*, to be published in J. **Environ. Radioactivity.**