Three-dimensional modelling of the tidal dispersion of non-conservative radionuclides in the marine environment. Application to ^{239,240}Pu dispersion in the eastern Irish Sea

R. Periáñez *

Dpto. Física Aplicada, E.U. Ingeniería Técnica Agrícola, Universidad de Sevilla, Ctra. Utrera km 1, 41013-Sevilla, Spain

Abstract

A three-dimensional model to simulate the tidal dispersion of non-conservative radionuclides in a marine environment has been developed. The model solves the three-dimensional hydrodynamic equations using normalized σ coordinates in the vertical and a flow dependent formulation for the eddy viscosity. Simultaneously, the suspended matter equation is also solved in σ coordinates, which includes advective–diffusive transport of particles plus settling. Erosion and deposition are incorporated into the sea bed boundary condition of the equation. It is considered that radionuclides can be present in three phases: solution, suspended matter particles and bottom sediments. Three equations have been formulated, whose solutions give the time evolution of specific activity into each phase. The exchanges of radionuclides between the liquid and solid phase (suspended matter and sediments) have been described in terms of kinetic transfer coefficients, so that the model can be applied under non-equilibrium conditions. The model has been applied to simulate the dispersion of ^{239,240}Pu in the eastern Irish Sea, which is released from Sellafield nuclear fuel reprocessing plant. Computed and measured suspended matter concentrations have been compared for several points in the sea to test the suspended matter sub-model. Also, computed and observed ^{239,240}Pu distributions in water, suspended matter and sediments have been compared for several years. k_d distribution coefficients have been computed as well. Model results are, in general, in good agreement with observations. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Over the past years, there has been an increasing interest in developing mathematical models to simulate the dispersion of radionuclides in aquatic environments since these models can be used as predictive instruments that can be applied in the assessment of radioactive contamination following an accidental release of radionuclides. On the other hand, useful oceanographic information (for instance, flushing and transit times) can be obtained by means of the application of such models (Prandle, 1984; Salomon et al., 1995).

* Tel.: +34-95423-3669; Fax: +34-95423-2644; E-mail: perianez@cica.es

Radionuclides are said to behave conservatively in water if they remain in solution, that is, no fraction is removed from the water column due to geochemical processes. In this case, radionuclide transport is governed by an advection diffusion equation. When radionuclides have a certain affinity to be fixed to the solid phase (suspended matter particles and bottom sediments), they are said to be non-conservative. Thus, adsorption–desorption reactions must be included in models developed to simulate the dispersion of non-conservative radionuclides. On the other hand, deposition of suspended particles and erosion of the sediment will produce an exchange of radionuclides between suspended matter and the sediment. Thus, the suspended matter dynamic must be solved also. A detailed discussion of these processes can be found in Periáñez et al. (1996a).

The first models developed to simulate the dispersion of non-conservative radionuclides in the sea were long-term dispersion models (Gurbutt et al., 1987; Abril and García-León, 1993a) in which the time steps used allowed to describe the transfers of radionuclides between the liquid and solid phases in terms of equilibrium distribution coefficients, k_d. The same approach was used by Ng et al. (1996) to simulate the dispersion of Cd and Zn in an English estuary. Periáñez et al. (1996a) developed a mathematical model to simulate the dispersion of non-conservative radionuclides in tidal waters based upon kinetic transfer coefficients, thus the model could be used under non-equilibrium conditions, and applied this model to study the dispersion of ^{226}Ra, ^{238}U and ^{232}Th in a Spanish estuary (Periáñez and Martínez-Aguirre, 1997a; Periáñez et al., 1996b).

All these models are two-dimensional (integrated in the vertical). Prandle et al. (1993) have pointed out that vertical structure can be of primary concern, even in shallow water (depth ~ 50 m), when typical simulated times are in the order of one month or less if tidal mixing keeps the vertical diffusion coefficient smaller than 10^{-3} m^2 s^{-1} (this value can be considered representative of a strong tidal action). However, few three-dimensional dispersion models have been developed. Nies et al. (1997) and Baxter et al. (1998) studied the three-dimensional transport of radionuclides in the Arctic Ocean, but the authors could not compare the model results with observations. Periáñez (1998a) developed a three-dimen-

sional model of the Irish Sea to study the three-dimensional dispersion of dissolved ^{137}Cs, released from Sellafield nuclear fuel reprocessing plant. Some interesting qualitative results were obtained from this work: for instance, it was found an alternating, with the same frequency as tidal oscillations, of well-mixed and vertically structured states. This model was improved using normalized σ-coordinates in the vertical and a flow dependent eddy viscosity (Periáñez, in press). Observed and computed ^{137}Cs distributions in the eastern Irish Sea were compared for a number of years. Model results were, in general, in good agreement with observations. These models can be used for conservative radionuclides.

The objective of this paper is to present a three-dimensional model for non-conservative radionuclides dispersion in tidal waters. The model solves the three-dimensional hydrodynamic equations in σ-coordinates and with a flow dependent eddy viscosity. Simultaneously, it solves the suspended matter dynamic equation, which is also written in σ-coordinates and includes the deposition and erosion terms in the sea bed boundary condition. Finally, the equations for radionuclides are solved. The model includes three phases: radionuclides in solution, in suspended matter and in the active fraction of the sediment (as denoted by Benes et al., 1994), that is, in muds (particles with a diameter < 62.5 μm that can be eroded and incorporated into the water column as suspended matter). Thus, three equations more are solved. These equations include advective and diffusive transport (in the case of radionuclides in solution and in suspended matter) plus the transfer of radionuclides between phases, which is formulated using kinetic transfer coefficients. The model has been applied to simulate the dispersion of ^{239,240}Pu in the eastern Irish Sea, which is released from Sellafield nuclear fuel reprocessing plant. Observed and computed plutonium distributions in water, suspended matter and sediments have been compared. Also, k_d values over the sea and k_d profiles in the water column have been obtained with the model.

The model equations are presented in the Section 2, where the methods used to solve them are also briefly described. After, the values for all the parameters involved in the model are given and finally model results are presented and discussed.

2. The model

2.1. Hydrodynamics

The three-dimensional hydrodynamic equations are written using normalized σ coordinates in the vertical. This way a constant number of grid boxes is used in the vertical at each horizontal grid point and vertical resolution is not lost in the shallower areas. The transformation to σ coordinates is (see Davies, 1985a, for instance):

$$\sigma = \frac{z + \zeta}{h + \zeta} \tag{1}$$

where h is the undisturbed (mean) depth of water, ζ is sea surface displacement from the mean level due to tidal oscillations and z coordinate is measured from the mean sea level to the bottom. Thus, the hydrodynamic equations are transformed from the interval $-\zeta \le z \le h$ into the constant interval $0 \le \sigma \le 1$. The form of the equations, for a homogeneous sea, in normalized coordinates has been given previously (Davies, 1985a; Davies and Stephens, 1983) and will not be repeated here. The boundary conditions applied at the sea bottom and sea surface can also be seen in Periáñez (in press). Wind effects are not considered and a linear law for bottom friction has been used. Although a quadratic law for bottom friction is now more extended than a linear formulation, the linear law has been adopted since a faster convergence of the equations is obtained. Also, there are no appreciable differences in dispersion patterns (for a dissolved radionuclide) obtained when the hydrodynamic model is calibrated using a linear and a quadratic law for bottom friction (Periáñez, in press).

A flow-dependent eddy viscosity has been used in the model. This formulation has been used previously and has given good results for tidal flow studies (Davies and Lawrence, 1994; Jones and Davies, 1996; Davies et al., 1997a):

$$N = C_N \sqrt{\bar{u}^2 + \bar{v}^2} \, h \tag{2}$$

where $C_N = 0.0025$ is a dimensionless experimentally measured coefficient and $\bar{u}$ and $\bar{v}$ are depth mean currents along the x and y axis, respectively. Eddy viscosity has been taken constant in the verti-

cal. This approximation has also been used by Davies and Lawrence (1994) and Davies et al. (1997b).

2.2. Suspended matter dynamics

As usual in suspended matter studies, we will consider that only particles with a diameter < 62.5 μm can remain in the water column as suspended matter, since larger particles will sink rapidly to the bottom and, as a consequence, their horizontal movement is negligible (Belderson, 1964; Gurbutt et al., 1987; Abril and García-León, 1994; Clarke, 1995; Periáñez et al., 1996c). Indeed, Eisma (1981) has pointed out that for all practical purposes muds can be regarded as synonymous of suspended matter. Although bed load transport of coarse particles may exist, it is not included in the model since, as will be shown below, specific activity in large sediment particles can be neglected when compared to specific activity in the active sediment.

The three-dimensional suspended matter equation has been obtained by transforming the equation proposed by Nicholson and O'Connor (1986) into σ coordinates. Such equation includes advective–diffusive transport plus vertical fall (settling) of suspended particles:

$$\frac{\partial m}{\partial t} + u \frac{\partial m}{\partial x} + v \frac{\partial m}{\partial y} + w * \frac{\partial m}{\partial \sigma}$$

$$= \frac{\partial}{\partial x} \left(K_h \frac{\partial m}{\partial x} \right) + \frac{\partial}{\partial y} \left(K_h \frac{\partial m}{\partial y} \right)$$

$$+ \frac{1}{(h + \zeta)^2} \frac{\partial}{\partial \sigma} \left(K_v \frac{\partial m}{\partial \sigma} \right)$$

$$- \frac{1}{h + \zeta} \frac{\partial (w_s m)}{\partial \sigma} \tag{3}$$

where m is the suspended matter concentration in ppm (parts per million), $w *$ is the vertical water velocity along the σ axis, K_h and K_v are the horizontal and vertical diffusion coefficients respectively and w_s is the settling velocity.

The vertical diffusion coefficient can be written as a function of the eddy viscosity (Launder and Spalding, 1972):

$$K_v = \epsilon N \tag{4}$$

where the non-dimensional number ε ranges from 0.1 to 0.5.

It is assumed that there is no flux of particles through the sea surface. Deposition and erosion of the sediment are incorporated into the sea bed boundary condition (Nicholson and O'Connor, 1986):

$$\frac{1}{h+\zeta}\left(K_{\mathrm{v}}\frac{\partial m}{\partial \sigma}\right)_{\sigma=1} = Efq^M - w_{\mathrm{s}}(b)m(b)$$

$$\times\left(1-\frac{q}{q_{\mathrm{cd}}}\right) \quad (5)$$

The first term of the right member of the equation represents erosion: E is the erosion constant (Nicholson and O'Connor, 1986) that also acts as a scaling factor, M is some power of the velocity typically in the range 2–5 (Prandle, 1997) and f gives the fraction of active sediments. It is considered that erosion occurs only when the near-bed current magnitude, $q=\sqrt{u^2(b)+v^2(b)}$ is larger than a critical erosion velocity, q_{ce}. Otherwise this term is set to zero. The second term represents deposition of particles on the sea bed: $w_{\mathrm{s}}(b)$ is the near-bed settling velocity and q_{cd} is a critical deposition velocity. Thus, deposition occurs only if $q<q_{\mathrm{cd}}$; otherwise this term is set to zero.

Note that (b) means that the corresponding magnitude is evaluated in the water layer that is in contact with the sediment.

The settling velocity of suspended particles, at low concentration (smaller than 1000 ppm), increases as the suspended matter concentration increases. A standard formula used to represent this process is (Pejrup, 1988; Mehta, 1989; Nicholson and O'Connor, 1986; Clarke, 1995):

$$w_{\mathrm{s}} = a_1 m^{a_2} \quad (6)$$

It has been taken $a_1 = 1.7 \times 10^{-6}$ and $a_2 = 1.6$ if w_{s} is measured in m s^{-1} and m in ppm. Since suspended matter concentrations in the Irish Sea are of the order of 1 ppm, settling velocities of the order of 10^{-6} m s^{-1} are obtained, which is the order of magnitude that can be found in literature (Prandle et al., 1993; Prandle, 1997) for the settling velocity of mud particles. Although the accepted interval for a_2 is 0.5 – 1.2 (Pejrup, 1988), a value of 1.6 was

obtained by Mehta (1989) and was also used in previous modelling work (Periáñez et al., 1996c). It can be found in literature that a_1 ranges from $\sim 10^{-3}$ (Clarke, 1995) to $\sim 10^{-9}$ (Mehta, 1989).

A value of 0.18 m s^{-1} has been used for the critical deposition velocity (Eisma, 1993; Periáñez et al., 1996c). A typical value of 0.21 m s^{-1} was taken for the critical erosion velocity (Heathershaw, 1981; Pugh, 1987).

A source term must be included to the suspended matter equation in those grid cells located along the coastline. This term represents the input of particles from runoff of continental waters (Abril and García-León, 1994) and can be written as:

$$\alpha I_{i,j} \quad (7)$$

where α is a constant to be fixed that represents the mass of suspended load per continental water volume unit and $I_{i,j}$ is the total volume of continental water that enters compartment i,j. $I_{i,j}$ will be different from zero when the compartment is situated along the coastline.

The solution of the suspended matter equation provides the suspended matter concentration at each position in the model domain and at each instant of time, but also gives information about the sedimentation processes that take place in the area under study. Thus, the sedimentation rate is obtained as the net balance between the deposition and erosion terms.

It has been shown (Davies and Lawrence, 1995) that wave-current interaction must be considered when studying sediment transport, and this is particularly important during major wind events (when wave heights are large and swell is significant). In this situation suspended matter concentration will increase and the near bed current (magnitude influenced by wave-current interaction) will be the primary mechanism moving the suspended sediment. However, at this stage of model development, wave-current interaction has not been considered. Nevertheless, the model reproduces historically deduced net sedimentation rates, and realistic plutonium concentrations in water, suspended matter and sediments are given by the model, as will be shown. It is our objective to study, in the next future, the influence of wind in the dispersion patterns of ra-

dionuclides. Then wave-current interaction will have to be considered.

2.3. Radionuclides dispersion

As was commented in Section 1, the model considers that radionuclides can be present in three phases: solution, suspended matter and active bottom sediments (particles with a diameter < 62.5 μm). In some preliminary calculations, the larger grain size sediment (particles with diameter > 62.5 μm) was also considered, as in Periáñez et al. (1996a). However, it was found that specific activity in the large grain size fraction of the sediment could be neglected when compared to specific activity in the active sediment. Thus, only the active sediment was included in the final model. This is not in agreement with the calculations of Abril and García-León (1993b), that found that specific activities in both sediment fractions are comparable. Nevertheless, our model confirms previous experimental work: Aston et al. (1985) found that virtually all the activity is associated with the active sediment, and McKay and Walker (1990) found that Pu specific activity in the active sediment is of the order of 20 times greater than that of the sand.

We will consider that the transfer of radionuclides from water to the solid phase (suspended matter and bottom sediments) is governed by a coefficient k_1 and the inverse process by a coefficient k_2, which are denoted kinetic transfer coefficients. It is known that actinide adsorption tends to be a surface phenomenon (Ramsay and Raw, 1987) and will depend on the surface of particles per water volume unit into the grid cell. This quantity has been denoted as the exchange surface (Periáñez and Martínez-Aguirre, 1997a; Periáñez et al., 1996a). Thus (Periáñez et al., 1996a):

$$k_1 = \chi_1 (S_m + S_s) \qquad (8)$$

where S_m and S_s are the exchange surfaces for suspended matter and bottom sediments, respectively, and χ_1 is a parameter with the dimensions of a velocity. It is denoted as the exchange velocity (Periáñez et al., 1996a). As a first approach, assuming spherical particles and a step function for the grain size distribution of particles, it can be easily obtained (Periáñez et al., 1996a) that:

$$S_m = \frac{3m}{\rho R} \qquad (9)$$

$$S_s = \frac{3Lf\phi}{R\Delta\sigma H} \qquad (10)$$

where ρ is the suspended particles density, R is the mean radius of suspended matter and active sediment particles, $H = h + \zeta$ is total water depth, L is the average mixing depth (the distance to which the dissolved phase penetrates the sediment), $\Delta\sigma$ is spacing in the vertical direction and ϕ is a correction factor that takes into account that not all the mass of the sediment is in contact with water. Then the kinetic transfer coefficient k_1 is written as:

$$k_1 = \chi_1 \frac{3m}{\rho R} + \chi_1 \frac{3Lf\phi}{R\Delta\sigma H} = k_{11} + k_{12} \qquad (11)$$

The transfer coefficient k_2 is considered constant. This description of the transfer of radionuclides between the dissolved and solid phases has been used successfully in previous modelling studies (Periáñez, 1998b; Periáñez and Martínez-Aguirre, 1997a,b; Periáñez et al., 1996b; Martínez-Aguirre and Periáñez, 1998).

Thus, the equation which gives the time evolution of radionuclide concentration in the dissolved phase (C_d, in Bq m^{-3}) in each grid cell is:

$$\frac{\partial C_d}{\partial t} = (\text{adv} + \text{dif})_{3D} - (k_{11} + \pi k_{12}) C_d$$
$$+ k_2 \left(mC_s + \pi \frac{A_s L \rho_m f\phi}{\Delta\sigma H} \times 10^3 \right) - \lambda C_d. \qquad (12)$$

The time derivative is interpreted as in a forward finite differences scheme. $(\text{adv} + \text{dif})_{3D}$ represents the three-dimensional advective plus diffusive transport of dissolved radionuclides from one grid cell to another. $\pi = 0$ unless we are solving the equation for a grid cell that is in contact with the sediment; in this case $\pi = 1$ to allow the transfer of radionuclides between water and the bottom sediment. C_s is the concentration of radionuclides in the suspended matter particles (Bq g^{-1}), A_s is the concentration of radionuclides in the active sediment (Bq g^{-1}) and the sediment bulk density, ρ_m, is expressed in kg

m^{-3}. Finally, λ is the radioactive decay constant. The external source of radionuclides should be added to this equation if it exists.

The equation for the time evolution of specific activity in suspended matter (Bq g^{-1}) is:

$$\frac{\partial C_s}{\partial t} = (\text{adv} + \text{dif})_{3D} - \text{sett} + k_{11}\frac{C_d}{m*}$$

$$- k_2 C_s \frac{m}{m*} + \pi(\text{res} - \text{dep}) - \lambda C_s \frac{m}{m*}$$

$$\tag{13}$$

where sett represents settling (see Eq. 3) and the $*$ means that the corresponding suspended matter concentration must be evaluated in the new time step. The deposition term (Bq g^{-1} s^{-1}) is written as:

$$\text{dep} = \frac{w_s(b)C_s(b)m(b)}{H\Delta\sigma m*(b)}\left(1 - \frac{q}{q_{cd}}\right). \tag{14}$$

Similarly, the resuspension term (Bq g^{-1} s^{-1}) is written as:

$$\text{res} = \frac{fEq^M A_s}{\Delta\sigma H m*(b)}. \tag{15}$$

Details about how the expressions for these terms can be deduced (in the case of a two-dimensional model) can be seen in Periáñez et al. (1996a).

The equation for the time evolution of specific activity in the active fraction of the sediment, A_s (Bq g^{-1}) is:

$$\frac{\partial A_s}{\partial t} = k_{12}\frac{C_d(b)H\Delta\sigma}{L\rho_m f} \times 10^{-3} - k_2 A_s \phi$$

$$+ (\text{dep} - \text{res}) - \lambda A_s. \tag{16}$$

The deposition and erosion terms are now written as follows:

$$\text{dep} = \frac{w_s(b)m(b)C_s(b)}{L\rho_m f}\left(1 - \frac{q}{q_{cd}}\right) \times 10^{-3} \tag{17}$$

$$\text{res} = \frac{Eq^M A_s}{L\rho_m} \times 10^{-3}. \tag{18}$$

Again, details about how these terms are obtained, in the case of a two-dimensional model, can be seen in Periáñez et al. (1996a).

2.4. Numerical solution

The hydrodynamic equations are solved using a explicit finite difference scheme, although the Saul'ev (1957) implicit method is applied to the vertical diffusion term to retain stability (Davies, 1985b). Some boundary conditions must also be given. At land boundaries, the normal component of the current and the flux of suspended particles and radionuclides are zero. Along open boundaries, amplitude and phase of the surface elevation at the M_2 frequency are specified from observations and the surface current component that is normal to the boundary is obtained from a radiation condition (Kowalick and Murty, 1993; Glorioso and Davies, 1995). Of course, better results would be obtained if other tidal components (at least S_2 and N_2) are included, since they induce currents that may affect the advection of radionuclides. However, we are mainly interested in tuning and testing the three-dimensional formulation of the dispersion of non conservative radionuclides: as will be shown below, realistic results are obtained although only the M_2 tide is used. This is also the reason why surface wind stress is not considered in the model. On the other hand, inclusion of the S_2 and N_2 tides would influence bed stress, and thus, sediment resuspension. However, as will be shown, slight variations in computed suspended matter concentrations are obtained when E and M are changed. This suggests that suspended matter concentrations in the eastern Irish Sea are not dominated by erosion events. Thus, it is not expected that the increase in bed stress produced by these other components produce a significant change in erosion and in suspended matter concentrations.

The open boundary condition described in Periáñez (1998a, in press) is applied to the dispersion equation of suspended matter and radionuclides:

$$C_i = \beta C_{i-1} \tag{19}$$

where C_i is the concentration in the open boundary and C_{i-1} represents the concentration just inside the computational domain. The non-dimensional number β is obtained from a calibration exercise.

The advection and horizontal diffusion terms are treated using explicit second order accuracy finite

difference schemes (Kowalick and Murty, 1993) and vertical diffusion is again solved using the Saul'ev method.

A FORTRAN code was developed to solve the equations involved in the model. The code was implemented on a HP SPP 2000 X-Class computer. It has 24 PA-8000 processors at 180 MHz rate. Memory is 3 GB. The equations are solved, for each time step, in the following sequence.

(1) Hydrodynamic equations are solved to obtain the water elevation and components of the water velocity for each grid cell. Eddy viscosity is also computed.

(2) Suspended matter equation is solved to obtain the suspended matter concentration into each grid cell. Deposition and erosion are also evaluated.

(3) Radionuclide dispersion: Equations for dissolved radionuclides, radionuclides in suspended matter and radionuclides in the active sediments are solved in this sequence.

(4) Sources: External sources of suspended matter, dissolved radionuclides and radionuclides in particulate form are introduced into the grid cells in which these sources exist.

One month of simulation takes about 2.5 h of CPU time. Model results will be compared with observations.

3. Application of the model

The model has been applied to study the dispersion of ^{239,240}Pu in the eastern Irish Sea. These radionuclides are discharged from a nuclear fuel reprocessing plant at Sellafield. The model has a horizontal resolution $\Delta x = \Delta y = 5000$ m. Ten layers are used in the vertical direction, thus, $\Delta\sigma = 0.1$. Time step is fixed as $\Delta t = 60$ s. Stability conditions (Kowalick and Murty, 1993) are satisfied with this selection. The computational domain is presented in Fig. 1, where the location of Sellafield plant is also shown. Water depths are introduced from bathymetric maps and range from 55 m in the west of the computational domain to a shallower area around the British coast. The fraction of active sediments (muds) has been obtained from Pentreath (1985) and Abril and García-León (1994).
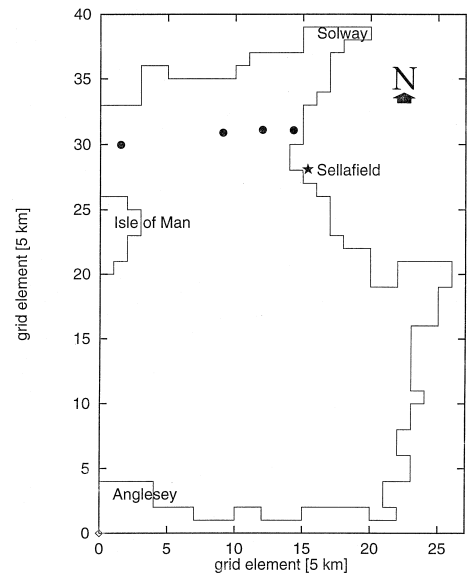

Fig. 1. Map of the computational domain. The star is Sellafield nuclear fuel reprocessing plant and circles are the points where suspended matter concentrations were measured (see Section 4). Each unit in the x and y axis is 5000 m (grid element number).

Surface elevations and phases along the open boundary have been taken from the observations of Howarth (1990).

The mean value 0.3 has been taken for ε in Eq. 4. The horizontal diffusion coefficient has been fixed as $K_h = 500$ m^2 s^{-1} since results in good agreement with observations are obtained with this value. Indeed, Bowden (1950) suggested that for the Irish Sea $500 < K_h < 900$ m^2 s^{-1} and, on the other hand, this value has been successfully used in previous modelling work (Periáñez, in press).

After some model runs, a value of 2.7×10^{-4} gm$^{-11/2}$ s$^{5/2}$ was selected for the erosion constant and it was taken $M = 3.5$. On the other hand α (Eq. 7) was fixed as 25×10^6 kg km^{-3}. The input of continental waters along the coast was obtained from MAFF (1987), and is also presented in Abril and García-León (1994). Results in agreement with observations are obtained with these selections.

The mixing depth L was taken a 0.1 m, following previous modelling works (Abril and García-León, 1993b; Periáñez and Martínez-Aguirre, 1997b; Periáñez et al., 1996b). The density of suspended matter particles is taken as $\rho = 2600$ kg m^{-3}, which

is the established value for soil particles density (Baver et al., 1972). The mean radius of suspended matter and active sediment particles has been selected as $R = 15$ μm, since only particles with a diameter < 62.5 μm can be present in the water column as suspended matter (Gurbutt et al., 1987; Periáñez et al., 1996b). A mean value of 950 kg m^{-3} was selected for the sediment bulk density (Kershaw and Young, 1988; Abril and García-León, 1994). After a calibration exercise, the geometry correction factor was taken as $\phi = 0.1$ and a value of 0.9 was selected for β in equation 19 (Periáñez, 1998a, in press).

The source of virtually all plutonium to the Irish Sea is the discharge from Sellafield through a pipeline which extends 2.5 km beyond high water. The annual discharges are presented in Fig. 2, compiled from McKay and Pattenden (1993) and Hunt et al. (1997). It has also been reported that about 99% of the discharged Pu is associated with particulate matter (Pentreath, 1985).

Finally, to simulate the dispersion of ^{239,240}Pu the values of the exchange velocity, χ_1, and of the kinetic transfer coefficient, k_2, must be known for this radionuclide. These parameters can be obtained from the measurements carried out by Nyffeler et al. (1984) and the equilibrium distribution coefficient, k_d, following the method presented in detail in Periáñez and Martínez-Aguirre (1997a) and Periáñez (1998b). However, the behaviour of plutonium in aquatic systems is of considerable complexity due to the fact that, in solution, it can exist in different oxidation states simultaneously. Thus, Pu (III) and

Pu(IV) predominate as the reduced and Pu (V) and Pu (VI) as the oxidized forms. The reduced Pu is highly particle-reactive and has been shown to possess a distribution coefficient which is approximately two orders of magnitude higher than that of the more soluble oxidized Pu (McKay and Pattenden, 1993; Mitchell et al., 1995). Hence, the values observed in field measurements represent the properties of the mixture of oxidation states that is present in the particular sample. Pentreath et al. (1986) have found that oxidation of Pu occurs quickly following release from Sellafield to sea water, and Mitchell et al. (1995) report that the oxidation state of Pu is the same all over the Irish Sea, result that has also been confirmed by Boust et al. (1996). Thus, since there are no differences in the speciation of Pu over the sea, a constant value for the equilibrium distribution coefficient can be used. We have taken the recommended value for Pu in coastal waters, 1.0×10^5 L kg^{-1} (IAEA, 1985), since this value is in agreement with measurements in the Irish Sea (McKay and Walker, 1990; Garland et al., 1990; Mitchell et al., 1995). As commented above, from the value of the Pu distribution coefficient and the experiments of Nyffeler et al. (1984), it can be obtained (Periáñez, 1998b) that:

$$\chi_1 = 1.51 \times 10^{-5} \text{ m s}^{-1}$$

$$k_2 = 1.16 \times 10^{-5} \text{ s}^{-1}$$

This values have been successfully used in previous modelling work (Periáñez, 1998b).

4. Results and discussion

The hydrodynamic part of the model was tested by comparing observed and computed values of tidal elevation amplitudes and phases, current profiles at several locations and semi-major axis magnitude and orientation of the M_2 tidal current ellipse at several depths and locations. The difference between computed and observed tidal amplitudes is always $<$ 10%, only in one point is 17.4% (comparisons have been made for 12 points). In the case of tidal phases, absolute values of the differences between observed and computed phases range from 0° to 27°, being the mean value 10.5°. The difference between computed
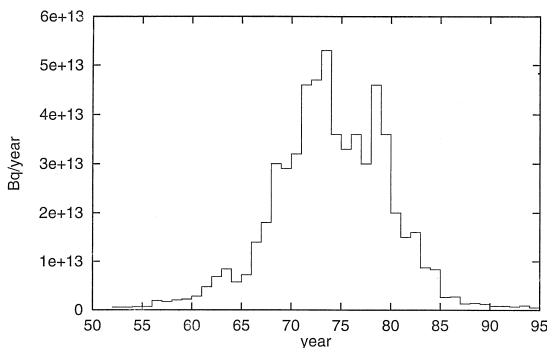

Fig. 2. Annual ^{239,240}Pu discharges (Bq year^{-1}) to the sea from Sellafield.

and observed semi-major axis magnitude of the tidal current ellipse ranges from 1.5% to 45%. Only in three points errors are above 25% (12 points used for comparison). The absolute value of the difference between observed and computed orientation of the axis ranges from 0.1° to 30.5°, being the mean value 7.6°. Details can be seen in Periáñez (in press). In that work, the advection–diffusion dispersion equation was calibrated by studying the dispersion of dissolved ^{137}Cs. The calibration consists of selecting the optimum boundary conditions and values of the diffusion coefficients, which were presented in Section 3.

Kershaw and Young (1988) measured the suspended matter concentration at several locations in the eastern Irish Sea and for both surface and sea bottom samples. Sampling points are shown in Fig. 1. Computed suspended matter profiles, along the west-to-east direction, following the points of Kershaw and Young (1988) for surface and bottom waters are presented in Fig. 3a,b, respectively, to-

Fig. 4. Computed suspended matter (ppm) distribution in surface waters. Each unit in the x and y axis is 5000 m (grid element number).

gether with the measured concentrations. As can be seen, the model gives a good estimation of the observed suspended matter concentrations for both set of samples. As expected, maximum concentrations are obtained close to the coast, and these concentrations decrease when entering into the open sea. A map showing the computed surface suspended matter concentration is presented in Fig. 4, which is in agreement with previous modelling work (Abril and García-León, 1994). Averaged sedimentation rates are small (range from -0.02 to 0.05 g cm^{-2} year^{-1}), thus the model does not violate historically deduced net sedimentation rates: Kirby et al. (1983) suggest that no major erosion or accretion of sediments is taking place in the Irish Sea at the present time, so that a state of equilibrium exists. Kershaw et al. (1988) have also found very low sedimentation rates (of the order of 10^{-2} g cm^{-2} year^{-1}) in the eastern Irish Sea.

Observed and computed distributions of ^{239,240}Pu in water, suspended matter and bottom sediments have been compared for several years (1974, 1977, 1979 and 1989). The input from Sellafield (Fig. 2) was introduced in the model for each year. However, as can be seen in Fig. 2, the input has been taking place since the fifties. Thus, instead of starting the model from zero concentrations, we have assumed a uniform background in water, suspended matter and sediments that represents the effect of previous dis-
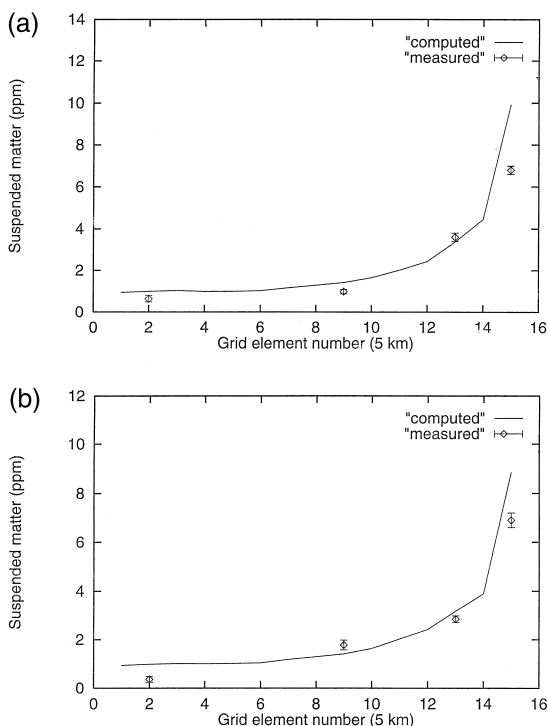

Fig. 3. Measured and computed suspended matter concentration (ppm) profiles following the points of Kershaw and Young (1988), shown in Fig. 1, in surface (a) and bottom (b) waters.
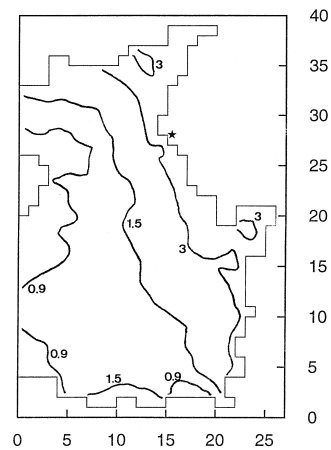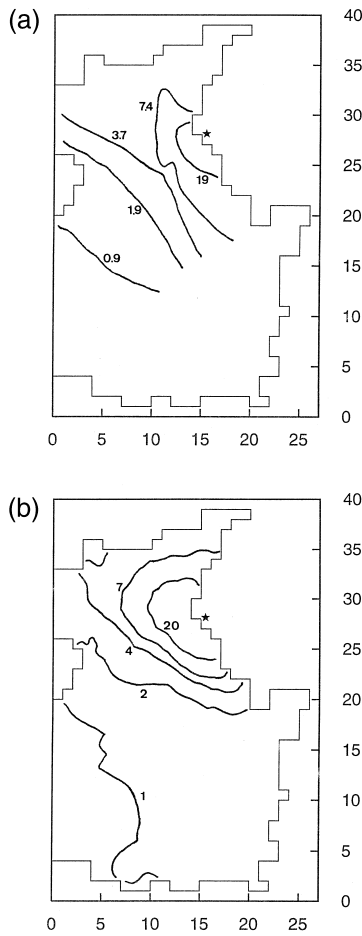
Fig. 5. Observed (a) and computed (b) ^{239,240}Pu distribution maps (Bq m^{-3}) in surface waters for year 1974. Each unit in the x and y axis is 5000 m (grid element number).

water. This technique of using an uniform background has been used in previous modelling works (Periáñez, 1998a, in press; Periáñez and Martínez-Aguirre, 1997a; Periáñez et al., 1996b).

Observed (from Hetherington, 1976) and computed ^{239,240}Pu specific activity in surface waters for year 1974 are presented in Fig. 5a,b, respectively. Fig. 6a,b are the same but for year 1979 (observed distribution is now taken from Pentreath, 1985). In general, the model gives a good estimation of the observed specific activities for both years, although it seems that it underestimates the values in the southeast area of the sea. However, it must be said that the observed distribution maps have been drawn by interpolation from several sampling points, and few samples were collected from this area.
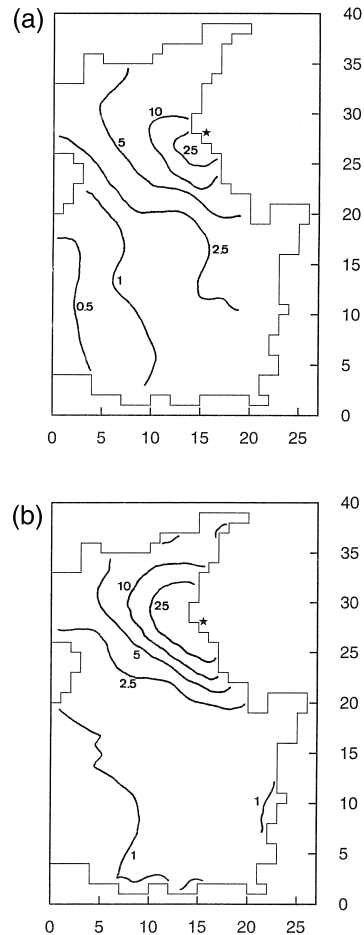

Fig. 6. Same as Fig. 5 but for year 1979.

charges. In Periáñez et al. (1994) it was shown that model results do not depend upon the way the background is created. Thus, the same results would be obtained if a large discharge is performed and some time is allowed to elapse so that the discharge is distributed over the sea. To save CPU time, the uniform background option was chosen. Thus, discharges from Sellafield are carried out over this uniform background and model results are obtained after a simulation period of two months. These results are compared with observations. The magnitude of the uniform background has been selected from a trial and error exercise: 0.1 Bq g^{-1} for suspended matter and bottom sediments and 0.05 Bq m^{-3} for

The computed distribution of ^{239,240}Pu in bottom waters, for year 1974, is shown in Fig. 7 as an example. The shape of the 1 and 2 Bq m^{-3} isolines is essentially the same in both surface (Fig. 5b) and bottom (Fig. 7) waters. However, close to Sellafield the shape of the isolines is different and specific activities are smaller than in surface waters. This must be due to the fact that close to Sellafield there is a mud bank that acts as a sink of plutonium, due to the high reactivity of this radionuclide (McKay and Pattenden, 1993; Boust et al., 1996). Thus, plutonium is removed from the deepest part of the water column and fixed to the bottom sediment. Indeed, the term $-\pi k_{12} C_d$ appears in Eq. 12, and k_{12} is proportional to f (Eq. 11).

The estimated ^{239,240}Pu distribution over the top 30 cm of the sediment for year 1977 is presented in Fig. 8a (Pentreath, 1985). We have computed the corresponding activity contained in the mixing layer, L, of the model, that is shown in Fig. 8b. Indeed, the mixing layer contains the greatest part of the total activity in the sediment (Abril and García-León, 1993b). The model again gives a realistic estimation of the activity levels, and the general features of isolines are reproduced by the model. As before, it seems that activity levels are underestimated in the southeast part of the sea.

Boust et al. (1996) measured ^{239,240}Pu surface specific activities in surface water and suspended

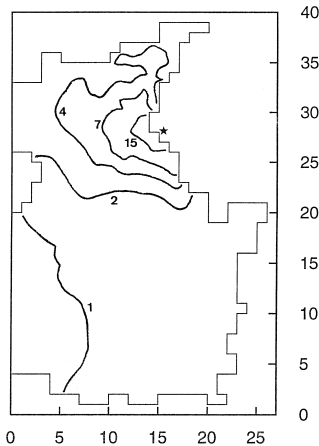Fig. 7. Computed ^{239,240}Pu distribution map in bottom waters (Bq m^{-3}) for year 1974. Each unit in the x and y axis is 5000 m (grid element number).
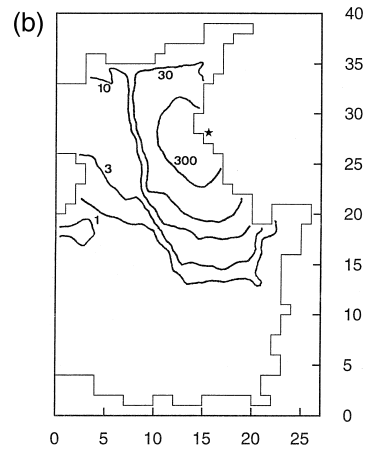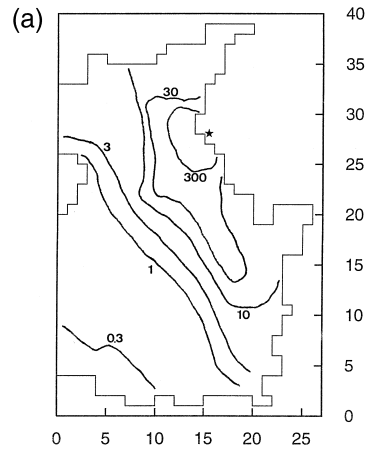

Fig. 8. Observed (a) and computed (b) ^{239,240}Pu distribution maps (kBq m^{-2}) in bottom sediments for year 1977. Each unit in the x and y axis is 5000 m (grid element number).

matter in 1989 at different distances southwest and northwest from Sellafield. Experimental and model results are compared in Fig. 9a (water samples) and Fig. 9b (suspended matter samples). Lines represent specific activity profiles following the directions of the sampling of Boust et al. (1996). It can be seen that there is a good agreement between measured and computed values for both water and suspended matter samples.

k_d distribution coefficients have also been calculated with the model. A map showing the computed k_d values in surface waters is presented in Fig. 10. It can be seen that k_ds diminish with increasing dis-
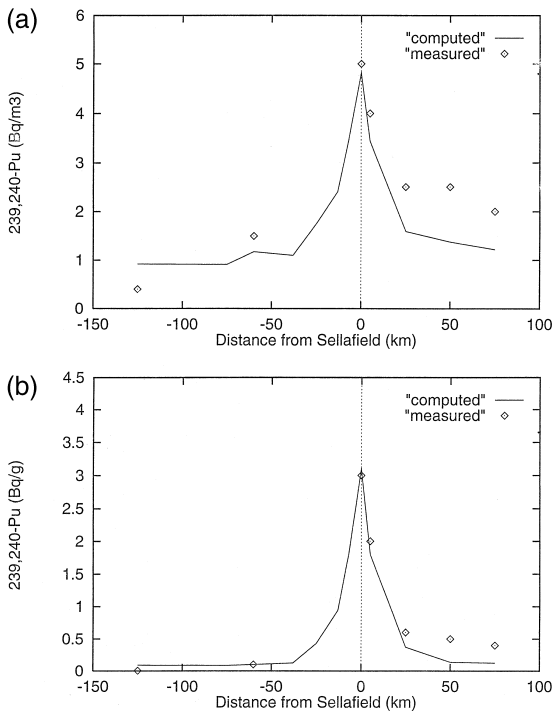
Fig. 9. Measured and computed ^{239,240}Pu specific activities in water (a) and suspended matter (b), in Bq m^{-3} and Bq g^{-1} respectively, for year 1989 at several points northwest (positive distances) and southwest (negative distances) from Sellafield.

Fig. 10. Computed k_d ($\times 10^5$ L kg^{-1}) map in surface waters. Each unit in the x and y axis is 5000 m (grid element number).

tance from the outfall. Mitchell et al. (1995) found this effect from k_d measurements. Since the oxidation state of Pu is identical in all the Irish Sea, this cannot account for the observed differences in k_d values. Thus, these authors concluded that such differences were due to the nature of discharges (almost all the plutonium in releases is associated with particulate matter). We have confirmed this hypothesis since we have not considered in our model spatial differences in the Pu oxidation state (see Section 3). On the other hand, computed distribution coefficients are in agreement with the measured values in the sea. Indeed, in nearshore waters along the English coast McKay and Walker (1990) obtained a k_d value of $(2.1 \pm 0.1) \times 10^5$ L kg^{-1}, and Mitchell et al. (1995) obtained a mean value of $(4 \pm 2) \times 10^5$ L kg^{-1} in the eastern Irish Sea. In the western Irish Sea k_d decreases to $(0.6 \pm 0.3) \times 10^5$ L kg^{-1} (Mitchell et al., 1995). Indeed, it can be seen in Fig. 10 that k_d

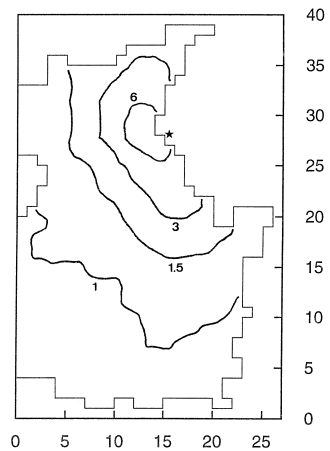decreases as we move towards the western sea (a value of 0.95×10^5 L kg^{-1} has been calculated between Anglesey and the Isle of Man).

k_d profiles in the water column for a point between Anglesey and the Isle of Man and for a point close to Sellafield outfall are presented in Fig. 11. It can be seen that there is no variation with depth for the point between Anglesey and the Isle of Man, confirming the findings of Mitchell et al. (1995). However, close to Sellafield, k_d values increase as we move towards the sea bottom. This is a logical result since wastes from Sellafield are discharged through a pipeline that is disposed on the sea bed. Pu
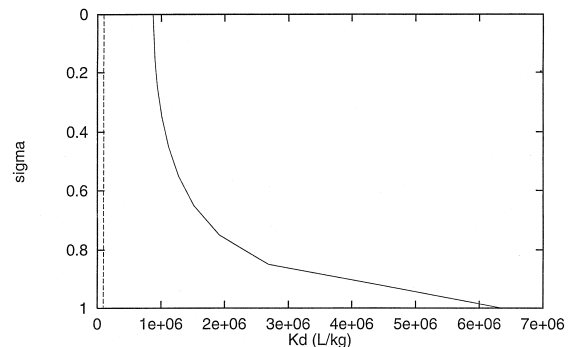

Fig. 11. Computed k_d (L kg^{-1}) profiles in the water column for a point between Anglesey and the Isle of Man, water depth 45 m, (dashed line) and for a point close to Sellafield outfall, water depth 20 m, (continuous line).

is discharged, as has been said before, in particulate form, thus higher k_d values must be found in bottom waters close to the release point. Nevertheless, computed k_ds are consistent with those recommended by IAEA (1985) and with measurements in the Irish Sea, as we have seen above.

The model sensitivity to the choice of boundary conditions for the dispersion equations (parameter β in Eq. 19) and to the diffusion coefficients has already been studied (Periáñez, in press). Suspended matter concentrations are slightly higher when E is increased by a factor 3, but they remain essentially unchanged if E is decreased by the same factor. This could be due to the fact that suspended matter concentrations in the eastern Irish Sea are not dominated by erosion events. Similar slight variations are obtained if it is taken $M = 2$ and $M = 5$. Too high suspended matter concentrations occur close to the coast if the source term for suspended matter (parameter α in Eq. 7) is increased by a factor 2. On the other hand, too low concentrations are obtained when it is decreased by the same factor. Suspended matter concentrations in agreement with observations are obtained when the value presented in Section 3 is used for α. Similarly, too high and too low concentrations are obtained when the settling velocity of suspended matter is decreased and increased, respectively, by a factor 3 (through parameter a_1 in Eq. 6). However, these variations affect mainly the regions close to the coast, where suspended matter concentrations are higher. Finally, it was found that essentially the same results are obtained if an erosion threshold velocity is not used in the model formulation. Indeed, it has been recently pointed out (Prandle, 1997) that the stipulation of a minimum threshold velocity below which no erosion occurs has little effect in tidal conditions where velocities exceed 0.5 m s^{-1}, as is the case (Periáñez, in press). On the other hand, very low suspended matter concentrations are obtained if a deposition threshold is not used. The conceptual approach to the transfer of radionuclides between the liquid and solid phase has already been used in a depth averaged model (Periáñez et al., 1996b). The sensitivity of the formulation to changes in the kinetic transfer coefficient k_2, the exchange velocity χ_1 and parameter ϕ has already been studied (Periáñez et al., 1996b) and will not be repeated here.

5. Conclusions

A numerical three-dimensional model to simulate the tidal dispersion of radionuclides in the sea has been developed. The model solves the hydrodynamic equations, using normalized σ coordinates in the vertical and a flow-dependent eddy viscosity formulation, and simultaneously, it solves the suspended matter equation and the equations that give the time evolution of specific activity in the liquid phase, suspended matter particles and active bottom sediments, which are also written in normalized coordinates. The transfer of radionuclides between the liquid and solid phases was described using kinetic transfer coefficients.

The model has been applied to simulate the dispersion of ^{239,240}Pu in the eastern Irish Sea. Plutonium is discharged to the sea, under authorization, from Sellafield nuclear fuel reprocessing plant. The hydrodynamic part of the model was tested in a previous paper (Periáñez, in press). Computed and measured suspended matter concentrations are in good agreement, for both surface and sea bottom samples. Also, the computed averaged sedimentation rates confirm the hypothesis that no erosion nor accretion of sediments is taking place in the eastern Irish Sea at present time.

Computed and measured plutonium distributions have been compared for the dissolved phase, suspended matter and bottom sediments for several years. Real inputs from Sellafield have been used in the calculations. It seems that the model underestimates the specific activities in the southeast part of the sea. We cannot be sure, at this moment, if this is due to the lack of measurements in the area or if it is due to the model itself. Nevertheless, computed and measured distributions are, in general, in good agreement, and the model gives a realistic representation of the activity levels that are measured in the sea.

Distribution coefficients, k_d, have also been computed with the model. Results are in agreement with literature values. Also, a decrease in k_d values with increasing distance from Sellafield has been obtained. This effect is due to the nature of the discharges, since 99% of Pu is released in particulate form. This result confirms previous hypothesis. On the other hand, no differences between surface and bottom samples have been obtained, unless close to

the release point. This effect is again due to the nature of the discharges.

Acknowledgements

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