# Modelling the tidal dispersion of  $137Cs$  and 239,240Pu in the English Channel

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#### Abstract

A numerical model to simulate the tidal dispersion of non-conservative radionuclides in the English Channel has been developed. The model solves the shallow water hydrodynamic equations and, simultaneously, the suspended matter equation which includes deposition and resuspension terms. The model considers that radionuclides can be present in three phases: water, suspended matter and bottom sediments. Thus the equations whose solutions give the temporal evolution of activities in the three phases must be solved too. The transfer of radionuclides between the liquid and solid phases has been described in terms of kinetic transfer coefficients. The model has been applied to simulate the dispersion of  $137Cs$  and <sup>239,240</sup>Pu released from a nuclear fuel reprocessing plant at La Hague (France). The model gives a realistic estimation of the activity levels detected in the Channel. The transit time of radionuclides from La Hague to the Dover Strait has also been calculated, using the crosscorrelation function method, under different weather conditions.

*Keywords:* Model; Tidal; Dispersion; Radionuclides; English Channel; Cap de la Hague

#### 1. Introduction

A nuclear fuel reprocessing plant releases radionuclides  $(^{125}Sb, ^{99}Tc, ^{137}Cs,$ 239,240Pu) to the sea at Cap de La Hague, located on the French coast of the English Channel (Fig. 1). Some models have been developed to simulate the dispersion of conservative radionuclides (that remain in solution) in part of the Channel (Salomon, Guegueniat, Orbi & Baron, 1988) and in the whole channel plus the southern North Sea (Breton & Salomon, 1995; Salomon, Breton & Guegueniat, 1995; Janin, 1996).

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Fig. 1. Computational domain. The star is La Hague nuclear fuel reprocessing plant. The line is the section along which radionuclide concentrations are obtained (see Section 3.2). Each unit in the *x*- and *y*-axis is 5000 m.

However, these models work with residual circulation (averaged currents) and, as a consequence, they can only be used to obtain the long-term dispersion of radionuclides (temporal scale in the order of years).

Recently, Periánez and Reguera (1999) developed a numerical model to simulate the tidal dispersion of dissolved radionuclides in the English Channel. The model solves the shallow water hydrodynamic equations to obtain the instantaneous water velocities over the Channel and, simultaneously, the advection diffusion dispersion equation is also solved. Model predictions were in good agreement with observations in the case of  $125Sb$  and  $99Tc$ . The model underestimated the concentrations in the case of 137Cs. This can be due to the fact that this radionuclide is less conservative than <sup>125</sup>Sb and <sup>99</sup>Tc (IAEA, 1985) and thus part of the released  $137Cs$  is fixed to the bottom sediment. Bottom sediments may be a source of radionuclides to the water column when concentrations in water are low enough to allow the desorption reaction to dominate absorption. During the simulated period the input of  $137\overline{Cs}$  was smaller than some months before and thus the  $137Cs$  contribution from the sediment could be higher than the input from the source. This effect was not considered in the model and could explain the underestimation of the 137Cs concentrations.

The objective of this work consists of extending the previous model to nonconservative radionuclides. Thus, the shallow water hydrodynamic equations are solved and, simultaneously, the suspended matter equation, which includes advection and diffusion of suspended particles plus the deposition and resuspension processes, and the equations that govern the dispersion of radionuclides in the three phases considered (water, suspended matter and bottom sediments) are solved too. The transfers of radionuclides between the liquid and solid phases are described in terms of kinetic transfer coefficients. The model has been applied to simulate the dispersion of  $137$ Cs: the objective is to improve the results of Periáñez and Reguera (1999). It has also been applied to studying the dispersion of  $239,240$ Pu. The dispersion of this radionuclide, which has a high affinity for the solid phases, in the English Channel has not been studied before with numerical models. Also, the transit times of radionuclides from Cap de La Hague to the Strait of Dover (see Fig. 1) have been calculated.

The model is presented in the next section. Thereafter, the results are presented and discussed.

### 2. The model

#### *2.1. Hydrodynamics*

The two-dimensional shallow water hydrodynamic equations are presented in Periáñez and Reguera (1999) and will not be repeated here. They include the nonlinear terms, a Coriolis term and the effect of wind. The use of a two-dimensional model is justified due to the dominance of barotropic over baroclinic mechanisms in the shallow and well-mixed waters of the Channel (Breton & Salomon, 1995).

Instead of using a constant value for the bed friction coefficient, as is usual in most models, its value has been increased from 0.0015 in the west Channel to 0.0875 in the Dover Strait to obtain a better agreement between the observed and computed currents. Indeed, Prandle (1975) has found in previous modelling work that friction increases in the region around Dover and has noted that the value of the friction coefficient must be selected with care in this area so as to account for such an increase in the bottom friction.

#### *2.2. Suspended matter dynamics*

As usual in suspended matter studies, we will consider that only particles with a diameter  $< 62.5 \text{ µm}$  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, Kershaw & Durance, 1987; Clarke, 1995; Periáñez, Abril & García-León, 1996a). Indeed, Eisma (1981) has pointed out that for all practical purposes muds can be regarded as synonymous with suspended matter. The suspended matter equation is

$$
\frac{\partial (Hm)}{\partial t} + \frac{\partial (uHm)}{\partial x} + \frac{\partial (vHm)}{\partial y} = \frac{\partial}{\partial x} \left( HK_x \frac{\partial m}{\partial x} \right) + \frac{\partial}{\partial y} \left( HK_y \frac{\partial m}{\partial y} \right)
$$

$$
- w_s m \left( 1 - \frac{q}{q_{cd}} \right) + Efq^M,
$$
(1)

where *m* is the suspended matter concentration in  $g/m<sup>3</sup>$ , *u* and *v* are the components of the water velocity along the *x* and *y* directions, *H* is the instantaneous water depth and  $K_x$  and  $K_y$  are the diffusion coefficients along the *x* and *y* directions. The last two terms represent deposition and erosion of the sediment. The deposition term has been based on the concept given by Teisson (1991), also used by Clarke (1995):  $w_s$  is the settling velocity of suspended particles,  $q = \sqrt{u^2 + v^2}$  is the current magnitude and  $q_{cd}$  is a critical deposition velocity. Thus, deposition occurs only if  $q < q_{cd}$ ; otherwise this term is set to zero. *E* is the erosion constant (Nicholson & O'Connor, 1986), *M* is some power of the water velocity, typically in the range  $2-5$  (Prandle, 1997) and  $f$  gives the fraction of small particles (diameter  $<$  62.5 µm) in the sediment. This way, the formula derived by Fukuda and Lick (1980) and Lavelle, Mojfeld and Baker (1984) has been used for the erosion rate, although the factor *f* has been included to take into account that only particles with diameter  $< 62.5 \,\mu m$  can be incorporated as suspended matter into the water column. Erosion occurs only if the water velocity is larger than a critical erosion velocity,  $q_{ce}$ ; otherwise the erosion term is set to zero.

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

$$
w_s = a_1 m^{a_2},\tag{2}
$$

where  $a_1$  and  $a_2$  have to be obtained from measurements or from model calibration.

Finally, a source term must be included in the suspended matter equation along the coastline. This term represents the input of particles from runoff of continental waters (Periáñez, 1999).

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.

### *2.3. Radionuclide dispersion*

As commented above, 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). It is considered that the transfer of radionuclides from water to the solid phase is governed by a coefficient  $k_1$  and the inverse process by a coefficient  $k_2$ , which are denoted kinetic transfer coefficients. The adsorption of radionuclides  $\alpha_2$ , which are denoted kinetic transfer coefficients. The adsorption of radiomicines<br>will depend on the surface of particles per water volume unit. This quantity has been denoted as the exchange surface (Periáñez, Abril & García-León, 1996b; Periáñez & Martínez-Aguirre, 1997a; Periáñez, 1999). Thus

$$
k_1 = \chi_1(S_m + S_s),\tag{3}
$$

where  $S_m$  and  $S_s$  are the exchange surfaces for suspended matter and bottom sediments, respectively, and  $\chi_1$  is a parameter with the dimensions of a velocity. It is denoted as the exchange velocity (Periáñez et al., 1996b). As a first approach, assuming spherical particles and a step function for the grain size distribution of particles, it can be easily obtained (Periancez et al., 1996b) that

$$
S_m = \frac{3m}{\rho R},\tag{4}
$$
\n
$$
S_c = \frac{3Lf\phi}{\rho R},\tag{5}
$$

$$
S_s = \frac{S_{\text{avg}}}{RH},\tag{5}
$$

where  $\rho$  is the suspended particle density,  $R$  is the mean radius of suspended matter and active sediment particles,  $L$  is the averaged mixing depth (the distance to which the dissolved phase penetrates the sediment) and  $\phi$  is a correction factor that takes into account that not all the mass of the sediment is in contact with water.

The transfer coefficient  $k_2$  is considered constant. This description of the transfer of radionuclides between the dissolved and the solid phases has been used successfully in previous modelling studies (Periánez, Abril  $\&$  García-León, 1996c; Periánez  $\&$  Martínez-Aguirre, 1997aPeriáñez & Martínez-Aguirre, 1997b; Periáñez, 1999).

The equation that gives the temporal evolution of radionuclide concentration in the dissolved phase,  $C_d$  (Bq/m<sup>3</sup>), is

$$
\frac{\partial (C_d H)}{\partial t} = (adv + dif) - k_1 C_d H + k_2 C_s mH + k_2 A_s L \rho_s f \phi \times 10^3 - \lambda C_d H, \quad (6)
$$

where ( $adv + dif$ ) means advective plus diffusive transport of dissolved radionuclides, *C*s (Bq/g) and *A*s (Bq/g) are radionuclide concentrations in suspended matter and active sediment particles, respectively, the sediment bulk density  $\rho_s$  is expressed in kg/m<sup>3</sup> and  $\lambda$  is the radioactive decay constant. The external source of radionuclides should be added to this equation at the points where it exists. The transfer coefficient  $k_1$  is given by

$$
k_1 = \chi_1 \left(\frac{3m}{\rho R} + \frac{3Lf\phi}{RH}\right).
$$
\n(7)

The equation for the temporal evolution of specific activity in suspended matter particles is

$$
\frac{\partial (mC_s H)}{\partial t} = (adv + dif) + k_1 C_d H + k_2 mC_s H + (eros - dep) - \lambda mC_s H, \qquad (8)
$$

where  $k_1$  is given by the first term of Eq. (7) and the erosion and deposition terms are given by

$$
eros = Efq^M A_s,\tag{9}
$$

$$
\text{dep} = w_s m C_s \bigg( 1 - \frac{q}{q_{cd}} \bigg). \tag{10}
$$

Again, a source term should be included in Eq. (8) if there exists an external input of radionuclides fixed to solid particles.

The equation for the temporal evolution of specific activity in the active fraction of the sediment is

$$
\frac{\partial A_s}{\partial t} = k_1 \frac{C_d H}{L \rho_s f} 10^{-3} - k_2 A_s \phi + (\text{dep} - \text{eros}),\tag{11}
$$

where  $k_1$  is given by the second term of Eq. (7) and the deposition and erosion terms are given by

$$
\text{dep} = \frac{w_s m C_s}{L \rho_s f} \bigg( 1 - \frac{q}{q_{cd}} \bigg) 10^{-3},\tag{12}
$$

$$
eros = \frac{Eq^M A_s}{L\rho_s} 10^{-3}.
$$
\n<sup>(13)</sup>

#### 2.4. Model configuration

To solve the equations, a spatial and temporal discretization is carried out: the Channel was divided into 3750 grid cells (forming a matrix  $75 \times 50$ ). The grid extends from  $48.3^{\circ}$ N to  $51.0^{\circ}$ N and from  $4.0^{\circ}$ W to  $1.5^{\circ}$ E; the grid cell size is  $\Delta x = \Delta y = 5000$  m and the time-step is fixed as  $\Delta t = 60$  s.

The explicit finite difference scheme described in Flather and Heaps (1975) was used to solve the hydrodynamic equations. Second-order accuracy finite difference schemes (Kowalick  $\&$  Murty, 1993) have been applied to solve the advection and diffusion terms in the suspended matter and radionuclide dispersion equations. In the case of advection, it consists of a second order approximation to the upwind scheme. All the stability conditions (Kowalick & Murty, 1993) are satisfied by the spatial and temporal resolutions of the model.

The computational domain is presented in Fig. 1. Along the open western boundary, water elevations were specified for each time-step from observations (Howarth & Pugh, 1983). A radiation condition (Kowalick & Murty, 1993) is applied along the northeastern open boundary (Dover Strait). Along the coast, the current component that is normal to the boundary is set to zero. Only the two main tidal components  $(M_2 + S_2)$  are considered. Good agreement with observations is obtained although only two components are used, as will be seen.

In the case of the dispersion equations, there is no flux of radionuclides or suspended matter through a closed boundary. Along open boundaries, the following boundary condition is applied:

$$
C_i = \psi C_{i-1},\tag{14}
$$

where  $C_i$  represents the concentration of radionuclides or suspended matter at the open boundary and  $C_{i-1}$  represents the concentration just inside the computational domain. This condition has been previously applied in Perianez (1998a,b,1999) and Periáñez and Reguera (1999).

A FORTRAN code was developed to solve the equations involved in the model and was implemented on a HP SPP 2000 X-Class computer.

#### 3. Results and discussion

#### *3.1. Hydrodynamics and suspended matter dynamics*

The magnitude and direction of the computed and observed currents have been compared for 12 points in the channel for spring, medium and neap tides. Observed and computed  $\overrightarrow{M_2}$  corange charts have also been compared. Results are, in general, in good agreement with observations. Details can be seen in Periancez and Reguera (1999) and will not be repeated here.

The following parameters are used to solve the suspended matter equation: diffusion coefficients are fixed as  $K_x = K_y = 51 \text{ m}^2/\text{s}$  and it is assumed that  $\psi = 0.9$  in Eq.  $(14)$  (see Periáñez & Reguera, 1999, for details).

One of the main difficulties in sediment transport modelling is the method of obtaining the erosion and deposition thresholds and the erosion constant. These parameters are specific for each site and they are often selected after a model calibration process. Thus they are selected in such a way that they produce the best fit of the model results with the observations. Of course, these parameters must be physically realistic.

After some model runs, the erosion constant has been fixed as  $E = 2.2 \times 10^{-5}$  g m<sup>-11/2</sup>s<sup>5/2</sup>. Unconventional units are due to the fact that *E* also acts as a scaling factor: the erosion rate must be expressed in  $g m^{-2} s^{-1}$  and it has been assumed that  $M = 3.5$  (as commented above,  $\overline{M}$  is typically in the range 2–5).

It has been selected, also after a model calibration, that  $a_1 = 5.7 \times 10^{-6}$  and 1. has been selected, also alter a model canolation, that  $u_1 = 3.7 \times 10^{-4}$  and  $u_2 = 1.6$  in Eq. (2) is  $w_s$  is expressed in m/s and *m* in g/m<sup>3</sup>. Since suspended matter  $a_2 = 1.6$  in Eq. (2) is  $w_s$  is expressed in m/s and *m* in g/m . Since suspended matter concentrations in most of the Channel are of the order of 1 g/m<sup>3</sup>, settling velocities of the order of  $10^{-6}$  m/s are obtained, which is the order of magnitude that can be found in the literature (Prandle, Jago, Jones, Purdie & Tappin, 1993; Prandle, 1997) for the settling velocity of cohesive sediments. Indeed, it can be found in the literature that  $a_2$  ranges between 0.5 and 2 (Pejrup, 1988; Eisma, 1993). A value of 1.6 was obtained by Mehta (1989) and was also used in previous modelling work (Periánez et al., 1996a). It can also be seen that  $a_1$  ranges from  $10^{-3}$  (Clarke, 1995) to  $10^{-9}$  (Mehta, 1989).

The critical deposition and erosion velocities have been taken from literature data. A value of 0.18 m/s has been used for the critical deposition velocity (Eisma, 1993; Periancez et al., 1996a) and a typical value of  $0.21$  m/s was assumed for the critical erosion velocity (Heathershaw, 1981; Pugh, 1987). Indeed, it has already been pointed out (Clarke, 1995) that due to the small size of cohesive sediments they tend to be slow falling and thus there is some lag between the end of deposition and erosion beginning. During the intermediate time interval, the sediment suspension is maintained by water turbulence with no apparent effect on erosion or deposition. Thus, usually,  $q_{cd} < q_{ce}$  (Clarke, 1995).

The suspended matter source-term from continental runoff was selected after a calibration process. Along the French coast, it was considered that a mean supply of  $0.027 \times 10^9$  kg/year occurs. Along the British coast it was fixed as  $0.35 \times 10^9$  kg/year. Of course, runoff will not be constant through the year, but these values may be considered as annual averages that give a mean suspended matter distribution over the Channel that is, in general, in agreement with the observed features of suspended matter concentrations. For comparison, suspended matter supply to the North Sea from different rivers ranges from  $10^7$  (Ems river) to  $10^9$  kg/year (Thames plus Humber, Rhine plus Meuse rivers), and the supply from coastal erosion (East Anglia plus Holderness coasts) is  $0.7 \times 10^9$  kg/year (Eisma, 1981).

The mean suspended matter concentrations computed by the model are presented in Fig. 2a and are, in general, in agreement with those given by Eisma and Kalf (1987), shown in Fig. 2b, obtained from observations. Suspended matter concentrations are somewhat higher along the English coast (some 5 ppm) than along the French coast (some 3 ppm). Also, suspended matter concentrations of 5 ppm are obtained to the north of La Hague. Unfortunately, there are no measurements in the western part of the Channel (from La Hague to the west). Again, the use of a two-dimensional model is justified since Eisma and Kalf (1987) have found that suspended matter concentrations in the bottom water are rather similar to those in the surface water. Measured (Brylinski, Dupont & Bentley, 1984) and computed suspended matter concentrations at Dover Strait have also been compared: measured concentrations 10 and 15 km from Cap Gris-Nez are 7 and 5 ppm, respectively; the corresponding mean computed values are 5.5 and 5.2 ppm.

The suspended matter flux through the Dover Strait into the North Sea has been computed as  $1.3 \times 10^{13}$  g/year. Eisma (1981) estimated a flux of  $1.0 \times 10^{13}$  g/year, although the value obtained by Van Alphen (1990),  $1.7\times10^{13}$  g/year, is considered the most accurate estimate (Eisma, 1990). Thus the computed #ux is in rather good agreement with previous estimates.

#### *3.2. Radionuclide dispersion*

The computed distribution of a conservative radionuclide  $(^{99}Tc)$  in the Channel is presented in Fig. 3 as an example (from Periáñez & Reguera, 1999). In the case of a conservative radionuclide,  $\chi_1 = k_2 = 0$ . It can be seen that concentrations are a conservative radionuclide,  $\chi_1 = \kappa_2 = 0$ . It can be seen that concentrations are higher on the French side of the Channel than on the English side. The banded structure, showing decreasing activities off the French coast, has been observed by Guegueniat, Herrmann, Kershaw, Bailly du Bois and Baron (1996) and is also apparent here. Waters of the Norman Breton Gulf are affected by the discharges from La Hague due to the existence of gyres around the Channel Islands (Orbi & Salomon, 1988). The Dover Strait acts as a bottleneck and thus gradients are enhanced in this area. This effect was also found by Breton and Salomon (1995).

Some parameters must be known to simulate the dispersion of non-conservative radionuclides. The mixing depth L was taken as 0.1 m, following previous modelling works (Periáñez et al., 1996c; Periáñez & Martínez-Aguirre, 1997a; Periáñez & Martínez-Aguirre, 1997b; Periáñez, 1999). The density of suspended matter particles is taken as  $2600 \text{ kg/m}^3$ , which is the established value for soil particle density (Baver, Gardner & Gardner, 1972). The mean radius of suspended matter and active sediment particles has been selected as  $R = 15 \mu m$ , since only particles with a diameter  $\lt 62.5 \text{ µm}$  can be present in the water column as suspended matter, as





Fig. 2. (a) Average suspended matter concentrations (ppm) calculated by the model. (b) Suspended matter concentrations (ppm) from observations of Eisma and Kalf (1987).



Fig. 3. Computed  $99$ Tc (mBq/L) distribution map.

discussed above. Measurements of sediment bulk density in the Channel have not been found in the literature and thus a typical value of 900 kg/m<sup>3</sup> (Kershaw & Young, 1988) has been used over the whole Channel. After a calibration exercise, the geometry correction factor was taken as  $\phi = 0.1$ , a value that has been used in previous modelling work (Periáñez, 1999).

In the case of  $137Cs$ , the values for the exchange velocity and the kinetic transfer coefficient  $k_2$  have been obtained from the laboratory experiments carried out by Nyffeler, Li and Santschi (1984):  $\chi_1 = 2.10 \times 10^{-8}$  m/s and  $k_2 = 1.16 \times 10^{-5}$  s<sup>-1</sup>. These values have been successfully used in previous modelling work (Periánez, 1998c). In the case of  $239,240$ Pu, the situation is more complicated since, in solution, plutonium 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 that is approximately two orders of magnitude higher than that of the more soluble oxidized Pu (McKay & Pattenden, 1993). Hence, the  $k_d$  values observed in field measurements represent the properties of the mixture of oxidation states that is present in the particular sample. To overcome this problem, mean kinetic transfer coefficients were estimated by Periáñez (1999) to simulate the dispersion of Pu in the eastern Irish Sea:  $\chi_1 = 1.51 \times 10^{-5}$  m/s and  $k_2 = 1.16 \times 10^{-5}$  s<sup>-1</sup>. However, the eastern film sea.  $\chi_1 = 1.51 \times 10^{10}$  m/s and  $\kappa_2 = 1.10 \times 10^{10}$  s. However,<br>kinetic transfer coefficients depend on the physico-chemical environmental conditions (water temperature, salinity and pH). Thus, although the values given above gave good results in the Irish Sea, in the English Channel it has been assumed that  $k_2 = 0.93 \times 10^{-5}$  s<sup>-1</sup> since a better agreement with measurements is obtained.

The inputs of 137Cs and 239,240Pu from Cap de La Hague into the Channel are presented, for instance, in Guegueniat et al. (1996). In the case of <sup>137</sup>Cs, observed and computed concentrations in water in 1991 have been compared. Indeed, the simulation presented in Periánez and Reguera (1999) has been repeated, but including the



Fig. 4. Computed and measured  $137Cs$  (mBq/L) profiles along the Channel. The *x*-axis corresponds to the grid element number in Fig. 1, from the western boundary to Dover thus, each unit is 5000 m.

transfers between the liquid and solid phases. A transect of  $137Cs$  activity along the Channel is presented in Fig. 4. The line represents the computed concentration, from the open western boundary to Dover Strait, approximately along the central Channel (see Fig. 1). Measured concentrations have been taken from Herrmann, Kershaw, Baily du Bois and Guegueniat (1995). The model gives a realistic estimation of the activity levels measured in the Channel. Results obtained previously (Periance & Reguera, 1999), when  $137Cs$  was considered as a conservative radionuclide, have been improved. Now it seems that the model is not underestimating the activity levels. However, it seems that the model produces a peak that does not appear in observations. This is due to the fact that model results were also extracted from points located very close to the source (following the line in Fig. 1) and there are no measurements in this area to compare with.

Computed  $137Cs$  distribution maps in solution and suspended matter are presented in Figs. 5a and b, respectively. It can be seen that there are differences in the distribution of  $137Cs$  with respect to the distribution of  $99Tc$ . Although, in general, activity levels are still higher along the French coast than along the English coast, there is an activity maximum situated to the west of La Hague. In this area, there are also higher suspended matter concentrations (see Fig. 2). Thus, the suspended matter distribution is now affecting the dispersion pattern of  $137\text{Cs}$ , which obviously does not occur in the case of a perfectly conservative radionuclide. Bailly du Bois, Rozet, Thoral and Salomon (1997) estimated the average impact of  $137Cs$  in the Channel waters by normalizing the measured activities to the averaged discharges from La Hague. The distribution map obtained by these authors is surprisingly similar to that presented in Fig. 5a, showing a clear impact to the west of La Hague. Since 137Cs was considered by these authors as a conservative radionuclide, the maximum was attributed to the existence of another source of  $137Cs$  that labels the waters of the





Fig. 5. Computed <sup>137</sup>Cs distribution maps in water, mBq/L, (a) and suspended matter, mBq/g, (b).



Fig. 6. Measured and computed  $2^{39,240}$ Pu specific activities in water (a), suspended matter (b) and bottom sediments (c). The *x*-axis corresponds to the grid element number in Fig. 1, from the western boundary to Dover; thus, each unit is 5000 m.

Channel. However, now it seems that the maximum may be due to the influence of suspended matter, since Cs is not a totally conservative element.

As expected, the  $137Cs$  distribution map in suspended matter (Fig. 5b) can be clearly correlated with the suspended matter distribution map (Fig. 2).

Boust, Mitchell, Garcia, Condren, Leon-Vintro and Leclerc (1996) measured  $239,240$ Pu specific activity in surface water, suspended matter and bottom sediments at several distances east and west of La Hague. Experimental and model results are presented in Figs. 6a (water), b (suspended matter) and c (bottom sediments). Lines represent the computed concentration activity transects following the directions of the sampling by Boust et al. (1996). In general, the model gives a realistic representation of the activity levels detected along the Channel in the three phases.

A map showing the distribution of dissolved  $^{239,240}$ Pu over the channel is presented in Fig. 7a. It can be seen that, again, it is different to that of Fig. 3 (obtained for a conservative radionuclide), since Pu is essentially attached to solid particles. Thus, its distribution must be governed by suspended matter dynamics. The plutonium distribution in suspended matter is presented in Fig. 7b. Due to the high reactivity of this element, the impact of Pu is essentially restricted to the area around La Hague: after release, it is quickly fixed to suspended matter and bottom sediments.



Fig. 7. (a) Computed 239,240Pu distribution in water (mBq/L). (b) Computed distribution in suspended matter (mBq/g). (c) Computed  $k_d$  ( $\times 10^6$  L/kg) map over the Channel.



Fig. 7. (*Continued*)

Distribution coefficients  $k_d$  have also been calculated with the model. A map showing the computed  $k_d$  values over the Channel is given in Fig. 7c. Computed  $k_d$  values are of the order of 10<sup>4</sup>-10<sup>5</sup> L/kg. IAEA (1985) recommended  $k_d$  values for Pu range  $10^4$ – $10^6$  L/kg, with an average of  $1.0 \times 10^5$  L/kg. The model results are consistent with these.

#### *3.3. Transit times*

Having developed and validated a model to simulate dispersion over the Channel, the model can be used to determine transit times, a time constant associated with water quality studies.

Consider a bounded sea region in which material (in this case radionuclides) is introduced at some location  $(x_0, y_0)$ . The transit time at any position  $(x, y)$  within the region is defined as the time to travel from  $(x_0, y_0)$  to  $(x, y)$ . Since dissolved material moves both by advection and diffusion, a patch becomes rapidly distorted and the transit time is not simple to obtain, either experimentally or numerically (Salomon et al., 1995). In this work, the transit time concept of Salomon et al. (1995) is applied to estimate the transit time from La Hague to Dover Strait. As a definition of the transit time from one point to another, we will use the lapse of time which gives the best similarity between signals (concentrations versus time) at the two locations. This similarity will be quantified by the cross-correlation function between the two

local series, that is defined as

$$
\theta(\tau) = \frac{\langle C_1(t)C_2(t+\tau)\rangle}{\sqrt{\langle C_1(t)^2 \rangle} \sqrt{\langle C_2(t)^2 \rangle}},\tag{15}
$$

where  $\theta$  is the cross-correlation function,  $C_1$  and  $C_2$  are concentrations at La Hague and the central Dover Strait, respectively, and  $\langle \rangle$  means time-averaging. The maximum of the cross-correlation function occurs for a given value of  $\tau$ , which is a statistical evaluation of the transit time between the two points.

If wind is not included in the model, the computed transit time due to tides is 4.1 months. However, if the average wind, southwest 6 m/s (Breton & Salomon, 1995), is included, the transit time is reduced to 3.0 months. This value is in close agreement with the previous calculation of Salomon et al. (1995) with a long-term model. This decrease is due to the fact that average wind conditions produce an increase in the residual drift to the North Sea relative to the tidal drift. This computed value is also in agreement with the previous experimental estimations of Guegueniat, Bailly du Bois, Gandon, Salomon, Baron and Leon (1994), who found a transit time between 2 and 8 months. A transit time of 3 months implies an average velocity of radionuclides discharged at La Hague of 83 km/month. Of course, the transit time depends upon weather conditions. Thus, in the case of a constant easterly wind (speed  $6 \text{ m/s}$ ), the computed transit time is increased to 7.1 months.

 $Of$  course, transit times are affected by the real changing and non-homogeneous wind fields. However, our calculations allow reasonable estimations of the order of magnitude of the transit times. Indeed, 3 and 7.1 months have been obtained for average wind and easterly wind conditions, respectively. The estimate by Guegueniat et al.  $(1994)$ , 2-8 months, represents the real mixture of different wind conditions since it has been deduced from observations. The calculations from this modelling study are in the range of variation given by Guegueniat et al. (1994) and thus they can be considered as realistic values.

These transit times have been computed for dissolved radionuclides, considering that  $\chi_1 = k_2 = 0$ . However, it has been observed that if radionuclides have a high that  $\chi_1 = \kappa_2 = 0$ . However, it has been observed that if radiofidently and affinity for the solid phase the transit time increases significantly. Thus, in the case of  $239,240$ Pu, the transit time to Dover is 7.0 months (in average wind conditions) a value that is higher than the transit time of a conservative radionuclide by a factor 2.3. The transit time of plutonium had not been calculated previously.

### 4. Conclusions

A model to simulate the dispersion of non-conservative radionuclides in the English Channel has been developed. The model solves the shallow water hydrodynamic equations, the suspended matter equation and the equations for the temporal evolution of activity concentrations in the three phases: water, suspended matter and active bottom sediments. The transfer of radionuclides between water and the solid phases has been described in terms of kinetic transfer coefficients.

The suspended matter sub-model has been tested by comparing observed and computed suspended matter distributions. Also, the suspended matter flux to the North Sea through the Dover Strait has been calculated. The computed flux is in agreement with previous estimates.

The model has been applied to simulate the dispersion of  $137Cs$  and  $239,240Pu$ released from the nuclear fuel reprocessing plant at La Hague. In the case of  $137Cs$ , the results obtained with an earlier model, in which this radionuclide was considered to remain in solution, have been improved. This suggests the importance of contaminated sediments as a source of radionuclides to the dissolved phase. The model gives a realistic estimation of 239,240Pu activity levels measured along the Channel in water, suspended matter and bottom sediments. Also, different distribution maps have been obtained for conservative and non-conservative radionuclides, revealing the influence of suspended matter on Cs and Pu dispersion. Computed  $k_d$  values for plutonium are consistent with those recommended by IAEA (1985).

Transit times of radionuclides from La Hague to the central Dover Strait have also been calculated using the cross-correlation function method. A transit time of 3 months has been calculated for dissolved radionuclides with average wind conditions, in close agreement with previous calculations. The transit time increases to 4 months if only tides are considered. On the other hand, a value of 7 months is obtained for easterly winds. In the case of a radionuclide with a high affinity for the solid phases, like  $2^{39}$ ,  $2^{40}$ Pu, a transit time significantly longer than that for a dissolved radionuclide has been obtained.

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