# Redissolution and long-term transport of radionuclides released from a contaminated sediment: a numerical modelling study

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## Abstract

A numerical model based upon residual circulation has been developed to simulate the long-term (years) transport of non conservative radionuclides in the English Channel. The transfer of radionuclides between water, suspended matter and bottom sediments has been described in terms of kinetic transfer coefficients, so that the model can be applied in situations out of equilibrium. The model has been used to study and compare the effects of direct discharges of radionuclides, the case of a continuous release and the redissolution of radionuclides from a contaminated sediment. These numerical experiments have been carried out for two radionuclides with a different geochemical behaviour: $^{137}$Cs and $^{239,240}$Pu. It has been found that natural equilibrium in the partition of radionuclides between the liquid and solid phases is reached in the case of instantaneous releases and in the case of redissolution from a contaminated sediment. However, in the case of a continuous release such partition remains out of natural equilibrium. Also, the behaviour of $^{137}$Cs and $^{239,240}$Pu is similar, but the time scales of the processes are clearly different due to the different affinities of both radionuclides to remain fixed to the solid phases.

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*Keywords:* English Channel; Advection–diffusion; Distribution coefficient; Sediment; Suspended matter; Cs; Pu

## 1. Introduction

Numerical models to simulate the dispersion of radionuclides in the marine environment have been developed in the last years. They have been applied to simulate the dispersion of radionuclides released from nuclear fuel reprocessing plants like Sellafield, in the Irish Sea (Aldridge, 1998; Periáñez, 1999), and Cap de la Hague, in the English Channel (Breton & Salomon, 1995; Periáñez, 2000), and also from dumped nuclear materials in the Artic Ocean (Harms, 1997). Once that the models have been adequately validated, through comparison of the model output with measured distributions of radionuclides, they can be used as predictive tools that can be applied in the assessment of contamination following accidental or deliberate releases of radionuclides. Also, models can be applied to assess the radiological consequences of the releases through the calculation of doses (Baxter, Harms, Osvath, Povinec, & Scott, 1998).

However, discharges from such nuclear fuel reprocessing plants have been reduced in the last years. The reduction of the discharges has not been reflected in a potential reduction of activity levels in waters. This has revealed the importance of bed sediments as a source of radionuclides, due to redissolution, to the water column. Indeed, there is evidence that suggests that sediments cannot be considered as a final repository for artificial radionuclides (Cook, Mackenzie, Mcdonald, & Jones, 1997). Numerical models have not been used to study these redissolution and transport processes of radionuclides. In part, this is due to the difficulty of including in models all the processes that are involved: absorption–desorption reactions between water, suspended matter and bottom sediments, deposition and erosion of the sediment, and advection and diffusion of radionuclides in water and suspended matter.

The objective of this work consists of studying the redissolution of radionuclides from a contaminated sediment in the English Channel and the long-term

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transport of such radionuclides after redissolution. Results from this study will be compared with the effect of an instantaneous release and a continuous release, since it is expected that translocation of remobilized radionuclides differs from that of direct discharges. Two radionuclides with a different geochemical behaviour will be used for the study: $^{137}$Cs and $^{239,240}$Pu. A long-term dispersion model for non-conservative radionuclides, including all the processes cited above, has been developed. The model is based upon the kinetic transfer coefficient formulation used for previous models developed to simulate the tidal dispersion of radionuclides. Although long-term dispersion models for the English Channel have already been developed (Breton & Salomon, 1995; Janin, 1996), they can only be applied to conservative radionuclides. Also, a model to simulate the tidal dispersion of non conservative radionuclides in the Channel has been previously developed by the author (Periáñez, 2000). However, such model is not adequate to carry out simulations over long temporal scales (years) since it solves simultaneously the hydrodynamic equations, the suspended matter equation and the radionuclide dispersion equations and, as a consequence, the model is not suitable for the studies we are now interested in (it was applied to study the near-field tidal dispersion of radionuclides). A long-term dispersion model based upon residual circulation is required to carry out simulations over long temporal scales. Such new long-term dispersion model for non-conservative radionuclides is presented in this paper.

The English Channel has been selected for the study since it is a well-known environment from the point of view of oceanography, and there is also a source (La Hague nuclear fuel reprocessing plant) of radionuclides to the sea.

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

## 2. The model

The model is based upon residual (averaged) circulation. Thus, simulations over long temporal scales (years) can be carried out. The residual circulation in the English Channel has been obtained from the hydrodynamic model previously developed to simulate the tidal dispersion of conservative radionuclides released from La Hague (Periáñez & Reguera, 1999), which includes the two main tidal constituents $M_2$ and $S_2$. The model is depth-averaged; the hydrodynamic equations (Pugh, 1987) include the non-linear terms, Coriolis term (earth rotation) and a quadratic law for bottom friction in which the bed friction coefficient increases from 0.0015 in the west Channel to 0.0875 in the Dover Strait to obtain a better agreement between observed and computed currents. Indeed, Prandle (1975) found that friction increases in the region around Dover. A wide comparison between observed and computed currents is presented in Periáñez and Reguera (1999) and will not be repeated here. Average wind speed and direction (southwest, $6\,\mathrm{m\,s^{-1}}$) has also been considered to obtain the average circulation in the Channel from the tidal model under mean climate conditions. Spatially varying wind fields are not considered in the model. This approximation, consisting of considering a temporal averaged wind that is uniform over the modelled area, has been used before (Prandle, 1984) even over a much larger region (the European Continental Shelf).

The average suspended matter distribution and average deposition (or erosion) have also been obtained from the suspended matter model developed to simulate the tidal dispersion of non conservative radionuclides in the Channel (Periáñez, 2000). This suspended matter model is depth-averaged and includes advection–diffusion of suspended particles, a standard formula to represent flocculation (Clarke, unpub. data; Clarke & Elliott, 1998; Mehta, 1989; Pejrup, 1988), and erosion and deposition terms described using threshold erosion and deposition velocities. The deposition term is based on the concept given by Teisson (1991) and also used by Clarke (unpub. data), Clarke and Elliott (1998) and Prandle et al. (2000). The erosion term formulation is based on the erosion constant concept (Nicholson & O'Connor, 1986; Prandle, 1997). Details are given in Periáñez (2000). The hydrodynamic and suspended matter models have been validated directly (by comparison of computed currents, water and suspended matter fluxes through Dover Strait and suspended matter concentrations with observations) and indirectly, through comparison of computed and measured radionuclide distributions (Periáñez, 2000; Periáñez & Reguera, 1999).

The model considers that radionuclides can be present in three phases: solution, suspended matter and active bottom sediments (particles with a diameter $< 62.5\,\mu\mathrm{m}$), as denoted by Benes, Cernik, and Slavik (1994). It is considered that the exchange of radionuclides between the liquid and solid phases is governed by a single reversible reaction (Nyffeler, Li, & Santschi, 1984). Thus, 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 [dimensions $(T)^{-1}$]. Radionuclide dispersion models based upon kinetic transfer coefficients have also been used by other authors (Aldridge, 1998; Margvelashvily, Maderich, & Zheleznyak, 1997; Piasecki, 1998). The absorption of radionuclides depends on the surface of particles per water volume unit, denoted as the exchange surface (Periáñez, 1999; Periáñez, 2000; Periáñez, Abril, & Garcıa-León, 1996a; Periáñez & Martinez-Aguirre, 1997):

$$k_1 = \chi_1 (S_m + S_s) \qquad (1)$$

where $S_m$ and $S_s$ are the exchange surfaces, per unit water volume [dimensions $(L)^{-1}$], for suspended matter and bottom sediments respectively, and $\chi_1$ is a parameter with the dimensions of a velocity denoted as the exchange velocity (Periáñez et al., 1996a). Assuming spherical particles and a step function for the size distribution of suspended particles, it can be easily obtained (Periáñez et al., 1996a) that:

$$S_m = \frac{3m}{\rho R} \tag{2}$$

$$S_s = \frac{3Lf\phi}{RH} \tag{3}$$

where $m$ is the suspended matter concentration, $\rho$ is the suspended particle density, $R$ is the mean radius of suspended matter and active sediment particles, $L$ is the average mixing depth (the distance to which the dissolved phase penetrates the sediment), $f$ gives the fraction of active sediment, $H$ is the water depth and $\phi$ is a correction factor that takes into account that not all the surface of the sediment particles is in contact with water since it will be partially hidden by other particles. This formulation has been successfully applied in the simulation of the tidal dispersion of radionuclides in estuarine (Periáñez, Abril, & Garcıa-León, 1996b; Periáñez & Martınez-Aguirre, 1997) and marine environments (Periáñez, 1999, 2000).

The equation that gives the time evolution of specific activity in water, $C_d$ (Bq m$^{-3}$), is:

$$\frac{\partial C_d}{\partial t} = (\text{adv} + \text{dif}) - k_1 C_d + k_2 C_s m + k_2 \frac{A_s L \rho_s f \phi}{H} \tag{4}$$

where $C_s$ and $A_s$ are, respectively, specific activity (Bq kg$^{-1}$) in suspended matter and the active bottom sediment. Since the model is two-dimensional, it is considered that $C_d$ and $C_s$ have an uniform distribution over the water column. The sediment bulk density, $\rho_s$, is expressed in kg m$^{-3}$ and (adv + dif) means advective plus diffusive transport of radionuclides. Kinetic coefficient $k_1$ is obtained from Eqs. (1)–(3); thus it can be seen that, although the exchange velocity $\chi_1$ is constant, $k_1$ depends on the suspended matter concentration and the distribution of active sediments over the model domain. On the other hand, $k_2$ is considered constant since the desorption process does not depend upon the available particle surface, as is the case with absorption.

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

$$\frac{\partial (mC_s)}{\partial t} = (\text{adv} + \text{dif}) + k_1 C_d + k_2 C_s m + \text{ED}_m \tag{5}$$

where $k_1$ is now given by the first term of Eq. (1). Symbol ED$_m$ is the erosion-deposition term. If there is a net deposition, such term is:

$$\text{ED}_m = -\frac{WC_s}{H} \tag{6}$$

and if there is a net erosion:

$$\text{ED}_m = \frac{WA_s}{H} \tag{7}$$

where $W$ is the net erosion or deposition rate in kg m$^{-2}$s$^{-1}$.

Finally, the equation for the temporal evolution of specific activity in the active bottom sediment is:

$$\frac{\partial A_s}{\partial t} = k_1 \frac{C_d H}{L \rho_s f} - k_2 A_s \phi + \text{ED}_s \tag{8}$$

where $k_1$ is given by the second term of Eq. (1). If there is a net deposition:

$$\text{ED}_s = \frac{WC_s}{L \rho_s f} \tag{9}$$

and for the case of a net erosion:

$$\text{ED}_s = -\frac{WA_s}{L \rho_s f} \tag{10}$$

To solve the equations, a spatial and temporal discretisation is carried out: the Channel was divided into 3750 grid cells (forming a matrix 75 × 50). The grid extends from 4.0°W to 1.5°E and from 48.3° to 51.0°N. The grid cell size is $\Delta x = \Delta y = 5000$ m ($x$ and $y$ measured eastward and northward respectively) and time step is $\Delta t = 3600$ s. The hydrodynamic equations in the tidal model used to obtain the residual water circulation were solved using the standard explicit finite difference scheme described by Flather and Heaps (1975). Second order accuracy explicit schemes were used to solve advection and diffusion in the suspended matter model. In particular, a three-point formula was used for advection (Kowalick & Murty, 1993).

In the long-term dispersion model, the MSOU (Monotonic Second Order Upstream) explicit finite difference scheme, as described in Vested, Baretta, Ekebjæerg, and Labrosse (1996), was applied to solve the advection terms and a second order accuracy scheme (Kowalick & Murty, 1993) was also applied to the diffusion ones. The boundary conditions described in Periáñez (2000) and Periáñez and Reguera (1999) have been used along the open borders of the grid.

## 3. Results

The average circulation in the Channel, obtained with the mean wind speed and direction, is presented in Fig. 1. The parameters used in the model are the following: $L = 0.1$ m following previous modelling work (Periáñez et al., 1996b; Periáñez & Martínez-Aguirre, 1997), the density of suspended matter particles is taken as

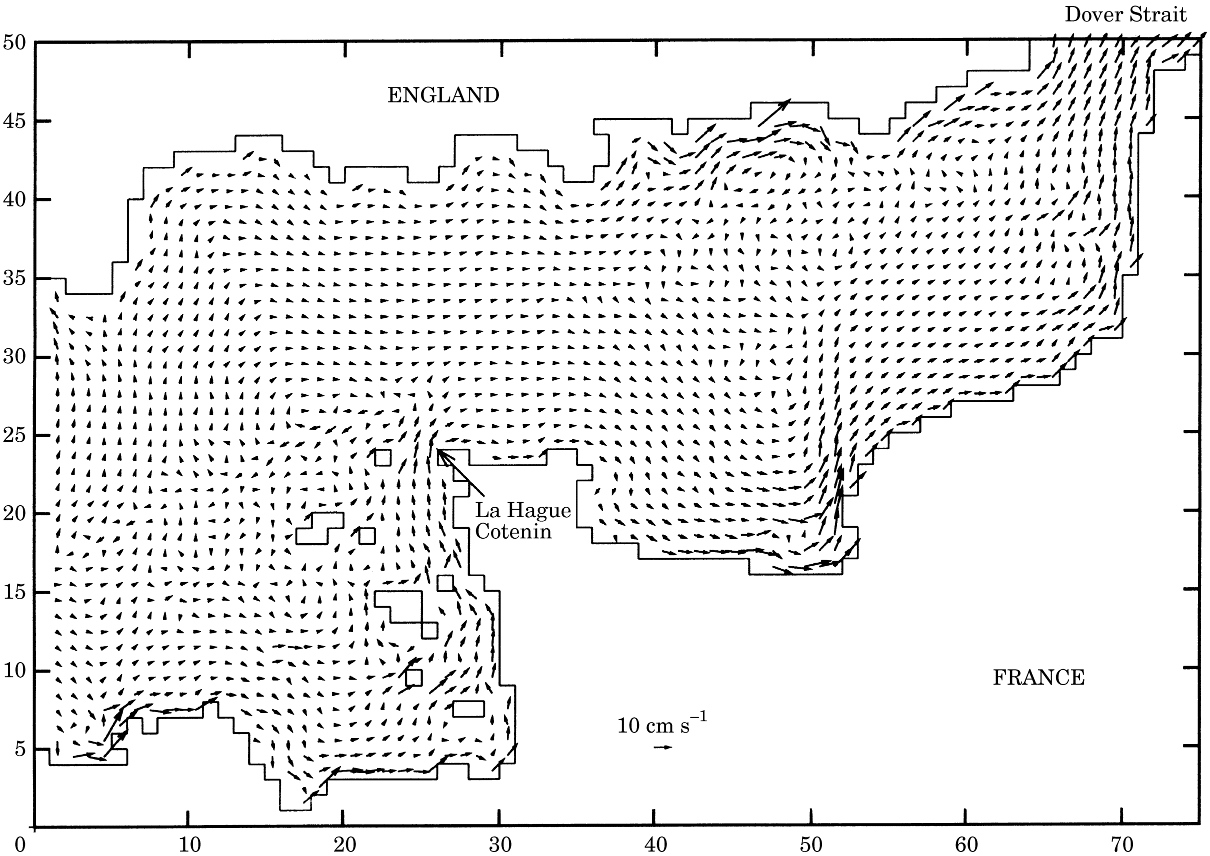

Fig. 1. Residual currents in the Channel for average wind conditions. Each unit in the $x$ and $y$ axis is 5000 m.

$\rho = 2600\,\mathrm{kg\,m^{-3}}$, which is the established value for soil particle density (Baver, Gardner, & Gardner, 1972), $R = 15\,\mu\mathrm{m}$, since only particles with diameter $< 62.5\,\mu\mathrm{m}$ can remain in the water column as suspended matter (Clarke, unpubl. data; Gurbutt, Kershaw, & Durance, 1987) and $\rho_s = 900\,\mathrm{kg\,m^{-3}}$ is a typical value for sediment bulk density (Kershaw & Young, 1988). According to Okubo (1971), a value that depends on a horizontal length scale (set as the grid spacing) is chosen for the diffusion coefficient (Breton & Salomon, 1995; Dick & Schönfeld, 1996; Schönfeld, 1995). Indeed, following Breton and Salomon (1995) such coefficient was taken as $51\,\mathrm{m^2\,s^{-1}}$. After a calibration exercise, the geometry correction factor was taken as $\phi = 0.1$, a value that has also been used in previous modelling work (Periáñez, 1999). The selection of these parameters is widely justified in Periáñez (2000) and will not be repeated here. As commented above, the model will be applied to $^{137}$Cs and $^{239,240}$Pu. The different geochemical behaviour of these radionuclides is described by the different values of the exchange velocity $\chi_1$ and the kinetic transfer coefficient $k_2$. In the case of Cs $\chi_1 = 2.10 \times 10^{-8}\,\mathrm{m\,s^{-1}}$, and for Pu $\chi_1 = 1.51 \times 10^{-5}\,\mathrm{m\,s^{-1}}$. The exchange velocity of Pu is three orders of magnitude larger than that of Cs, thus Pu has a higher affinity to be fixed to the solid phases than Cs. The kinetic transfer coefficient $k_2$ is $1.16 \times 10^{-5}\,\mathrm{s^{-1}}$ and $0.93 \times 10^{-5}\,\mathrm{s^{-1}}$ for Cs and Pu, respectively. These parameters have been deduced from the laboratory experiments carried out by Nyffeler et al. (1984) and the method given by Periáñez and Martínez-Aguirre (1997), and their values have already been used in Periáñez (2000). The sensitivity of the formulation of the transfer of radionuclides between the liquid and solid phase has been studied in Periáñez et al. (1996b), through the study of the model response to changes in $\chi_1$, $k_2$ and $\phi$, and will not be repeated here. However, it has been observed that if $\chi_1$ is increased in a factor 2 and $k_2$ is decreased in the same factor so as to enhance the transfers to the solid phases, concentrations in these phases are also increased in a factor 2 approximately. A reduction in concentrations in the solid phases of the order of 2 is obtained if $\chi_1$ and $k_2$ are now decreased and increased, respectively, in a factor 2. On the other hand, an increase in $\phi$ implies a higher water–sediment interaction, thus the redissolution process is enhanced and higher concentrations are obtained in water. In particular, dissolved concentrations are increased in a factor 2 when $\phi$ is increased in a factor 10.

In all simulations, background radionuclide concentrations in water, suspended matter and active bottom sediments are considered over the Channel (unless in the indicated points) to start calculations.

The transport of a conservative radionuclide has initially been studied. In this case $\chi_1 = k_2 = 0$. The patch moves along the French shore of the Channel. A banded structure, showing decreasing activities off the French shore is apparent, which was also observed by Gueguenia‍t, Herrmann, Kershaw, Bailly du Bois, and Baron (1996). This probably corresponds to a plume affected by Coriolis acceleration to stay on the right side of the Channel. After 84 days the patch is arriving in the Strait of Dover. This is in agreement with the 3 months transit time between La Hague and Dover calculated with the tidal model and the cross-correlation function method (Periáñez, 2000; Salomon, Breton, & Gueguenia‍t, 1995).

A map showing the computed distribution of $^{137}$Cs corresponding to February 1991 is presented in Fig. 2. The model was started from a uniform $^{137}$Cs background (initial condition for concentrations) corresponding to the average concentration measured in the Channel in September 1990 (Herrmann, Kershaw, Bailly du Bois, & Gueguenia‍t, 1995). Average discharge from La Hague corresponding to that period of time was used (see for instance Gueguenia‍t et al., 1996). Measured $^{137}$Cs concentrations in February 1991 from La Hague to Dover range from 3.5 to 13.8 Bq m$^{-3}$ (Herrmann et al., 1995) with a mean value of $10 \pm 3$ Bq m$^{-3}$ (14 samples). Computed concentrations are in agreement with these values. As in the case of a conservative radionuclide, concentrations are, in general, still higher along the French coast than along the British coast. There is, however, an activity maximum situated to the west of La Hague. Bailly du Bois, Rozet, Thoral, and Salomon (1997) estimated the average impact of $^{137}$Cs in the Channel waters by normalizing the measured activities to the averaged discharges from La Hague. The distribution map obtained by these authors is very similar to that presented in Fig. 2, also showing a clear maximum at the west from La Hague. The magnitude of the computed maximum (relative to concentrations in the rest of the Channel) is also in agreement with the estimation of Bailly du Bois et al. (1997).

A very important parameter in environmental radioactivity studies is the $k_d$ coefficient. It gives the partition of a tracer between the solid and dissolved phases. Distribution coefficients are defined and experimentally obtained as the ratio between specific activities in the solid and liquid phases, and are given in m$^3$ kg$^{-1}$. If a system is not externally perturbed, then the distribution coefficient gives the partition of the tracer between the liquid and solid phases that is characteristic of the natural conditions of the system. Such coefficient will be denoted as the ‘natural equilibrium‘ distribution coefficient, $k_d^0$. In previous work (Periáñez et al., 1996b), it was shown that the following relation between $k_d^0$, $\chi_1$ and $k_2$ holds:

$$k_d^0 = \frac{\chi_1}{k_2} \frac{3}{\rho R} \tag{11}$$

If the system is perturbed (for instance through an external input of radionuclides), the reversible reaction that governs the exchanges of radionuclides may, of course, reach an equilibrium (or steady) state, but the distribution coefficient will now be, in general, different to $k_d^0$. Thus, the model can compute the $k_d$ that would be obtained after a field measurement as the ratio between computed specific activities in the solid and dissolved phases. That computed $k_d$ can be compared with the corresponding natural equilibrium value given by Eq.

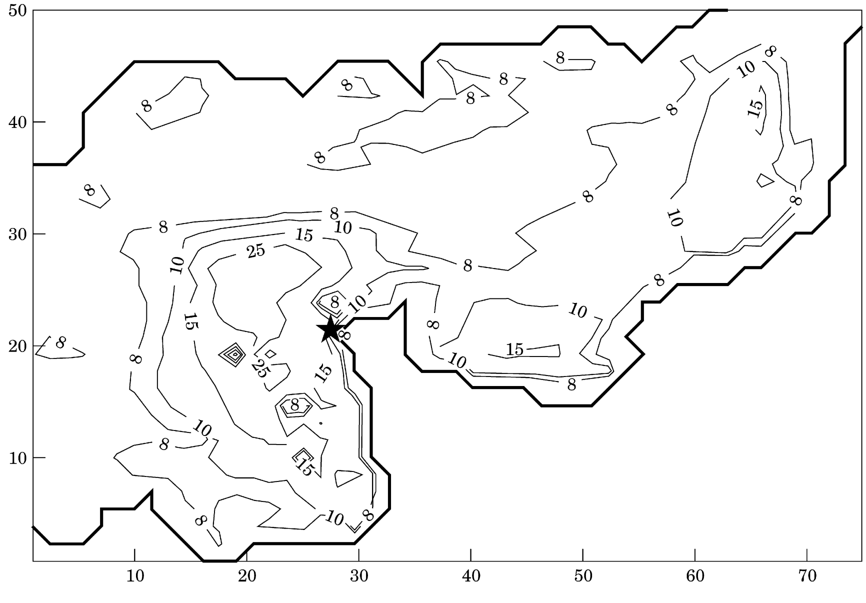

Fig. 2. Computed distribution of $^{137}$Cs in water (Bq m$^{-3}$) for February 1991. The star is La Hague nuclear fuel reprocessing plant.

(11). If the same value is obtained, it means that the partition is in natural equilibrium.

A first experiment was carried out with $^{137}$Cs. An initial specific activity of $10^5$ Bq kg$^{-1}$ was considered in the active fraction of the bottom sediment in compartment (26, 24), where discharges from La Hague are carried out. Such content implies a total amount of $2.25 \times 10^{13}$ Bq in the entire compartment. The time evolution of specific activities in water, suspended matter and active bottom sediments, as well as the suspended matter-water $k_d$ distribution coefficient, in this compartment are presented in Fig. 3.

It can be seen in Fig. 3a that concentrations in the three phases decrease during approximately the first 100 days. Thus, radionuclides are redissolved from the sediment and incorporated into the water column. Then they are removed from the area by advection processes: a patch that moves along the French coast, as in the case of an instantaneous release of a conservative radionuclide, is observed. After the initial 100 days, concentrations have almost stationary values in the three phases. Also, partition between the solid phase and water has reached natural equilibrium. This can be seen with the help of Fig. 3b: with the parameters considered in the model, $R = 15\,\mu$m and $\rho = 2600$ kg m$^{-3}$, and the corresponding exchange velocity and $k_2$ values for Cs (see above), it is obtained [from Eq. (11)] that the natural equilibrium distribution coefficient is $k_d^0 = 139 \times 10^{-3}$ m$^3$ kg$^{-1}$. From Fig. 3b, the distribution coefficient is initially around $50 \times 10^{-3}$ m$^3$ kg$^{-1}$, below the natural equilibrium value. Then it increases and tends to the natural equilibrium value. Thus, redissolution does not take place under equilibrium (this implies that the process should not be studied with a model based upon $k_d^0$ distribution coefficients). Only after a characteristic time of the order of 100 days (in the case of Cs), when the redissolution process is completed, natural equilibrium is reached.

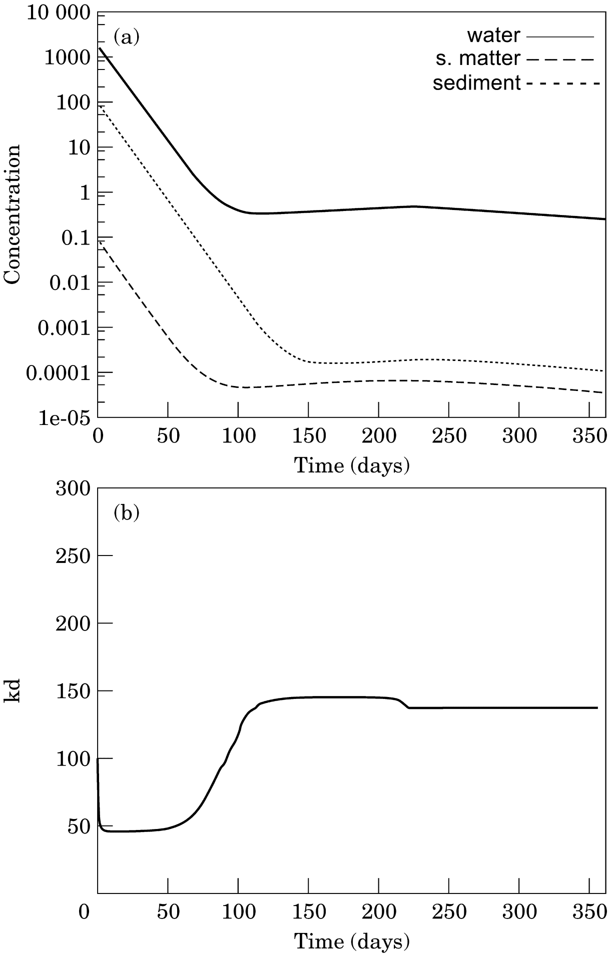

Fig. 3. Time evolution of $^{137}$Cs specific activities in water (Bq m$^{-3}$), suspended matter and active bottom sediment ($\times 10^3$ Bq kg$^{-1}$) (a), and $k_d$ distribution coefficient ($\times 10^{-3}$ m$^3$ kg$^{-1}$) (b) at La Hague in the redissolution experiment.

In analogy with the radioactive decay half-life, the sediment half-life has been defined as the time in which specific activity in the sediment is halved due to the redissolution process (it has been defined since the decrease in sediment concentration follows an exponential law). In the case of Cs, the resulting sediment half-life is 7.009 ± 0.016 days.

These results have been compared with the effect of a continuous release of dissolved $^{137}$Cs at La Hague. A release of $5.0 \times 10^{10}$ Bq per time step was introduced during a year. Concentrations increase quickly in the three phases and reach constant values: the removal rate becomes equal to the input rate. Although a constant value of the distribution coefficient is reached, the process is clearly out from natural equilibrium, since such steady $k_d$ is smaller than $k_d^0$. The situation some tens of km to the east from La Hague is different. Initially, concentrations correspond to background values in the three phases. Radionuclides released at La Hague begin to arrive, and constant values are quickly obtained. Here, the distribution of $^{137}$Cs between the liquid and solid phases is in natural equilibrium since the computed $k_d$ corresponds to $k_d^0$ during all the simulations. This is in contrast with the situation at La Hague, where the system remains out of natural equilibrium due to the existence of an external supply. A map showing the distribution of dissolved $^{137}$Cs 1 year after the beginning of the input is presented in Fig. 4. A banded structure showing higher concentrations along the French shore is now extending from La Hague to Dover.

Finally, the case of an instantaneous release of dissolved $^{137}$Cs has been studied. An input of $5.0 \times 10^{12}$ Bq was assumed at $t = 0$. It has been observed that the patch moves along the French coast as in the case of a conservative radionuclide. After a time of the order of 100 days, the distribution of $^{137}$Cs over the Channel is essentially the same as in the case of the redissolution experiment: natural equilibrium has been reached over

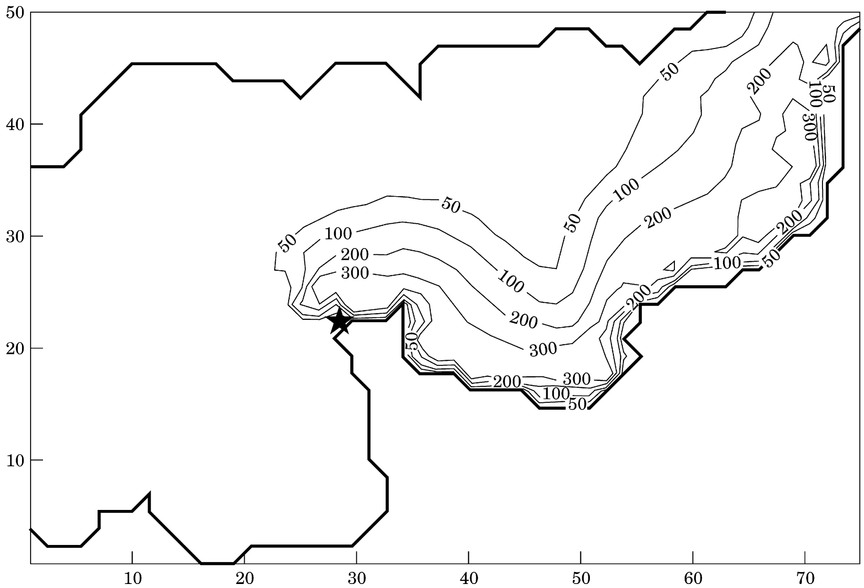

Fig. 4. Distribution map of dissolved $^{137}$Cs (Bq m$^{-3}$) 1 year after the beginning of a continuous release from La Hague. The star is La Hague nuclear fuel reprocessing plant.

the Channel in both cases. It is also observed that after a long time, still nonzero concentrations are found in the source region (although they go on decreasing slowly) since sediments, that initially absorb a significant amount of radionuclides, are now acting as a long term source to the water column.

The same numerical experiments have been carried out for $^{239,240}$Pu. In the redissolution experiment, a specific activity of $10^5$ Bq kg$^{-1}$ is again considered in the active sediment of compartment (26, 24). The time evolution of specific activities in water, suspended matter and active sediment, as well as the time evolution of the $k_d$, at La Hague are presented in Fig. 5. As in the case of $^{137}$Cs, there is a decrease in specific activities in the three phases due to redissolution from the sediment and advective transport. However, the process is slower than in the case of $^{137}$Cs due to the larger affinity of Pu to remain fixed to the solid phases. Indeed, natural equilibrium is not reached until approximately 8 years, as can be deduced from the time evolution of the $k_d$ (from Eq. (11), the corresponding $k_d^0$ for Pu is 124 m$^3$ kg$^{-1}$). A map showing the distribution of $^{239,240}$Pu in solution and bottom sediments 7 years after the beginning of the redissolution process is presented in Fig. 6. An activity peak is observed to the east of Cotenin. This is in good agreement with the results of Boust, Colin, Leclerc, and Baron (1997), who also estimated a transit time of surficial sediments from La Hague to this area of 7 years.

The sediment half-life in the case of Pu is 73.4 ± 1.2 days, which is an order of magnitude larger than that of Cs.

In the case of a continuous release of plutonium ($5.0 \times 10^{10}$ Bq per time step), the results are similar to

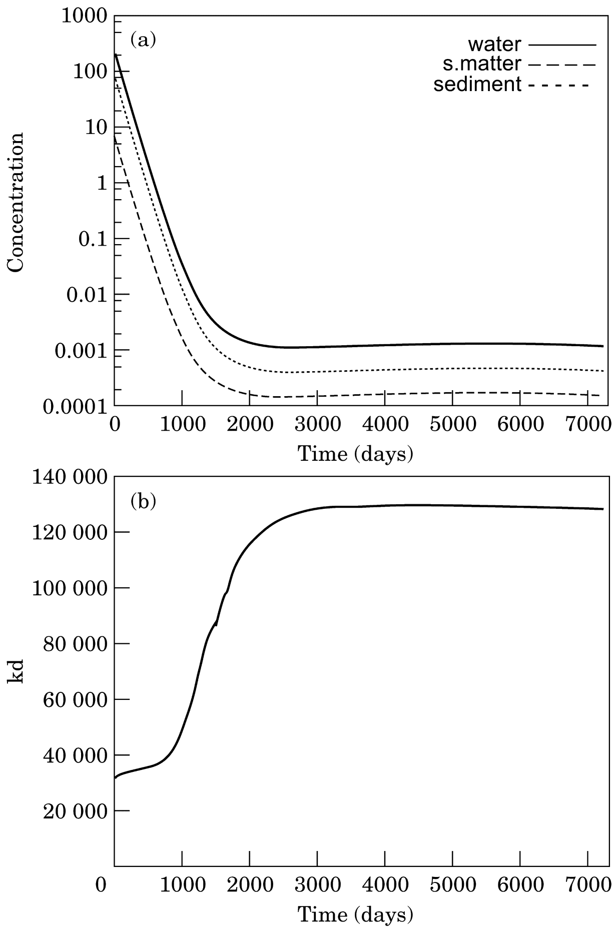

Fig. 5. Time evolution of $^{239,240}$Pu specific activities in water (Bq m$^{-3}$), suspended matter and active bottom sediment (×10$^3$ Bq kg$^{-1}$) (a), and $k_d$ distribution coefficient (×10$^{-3}$ m$^3$ kg$^{-1}$) (b) at La Hague in the redissolution experiment.

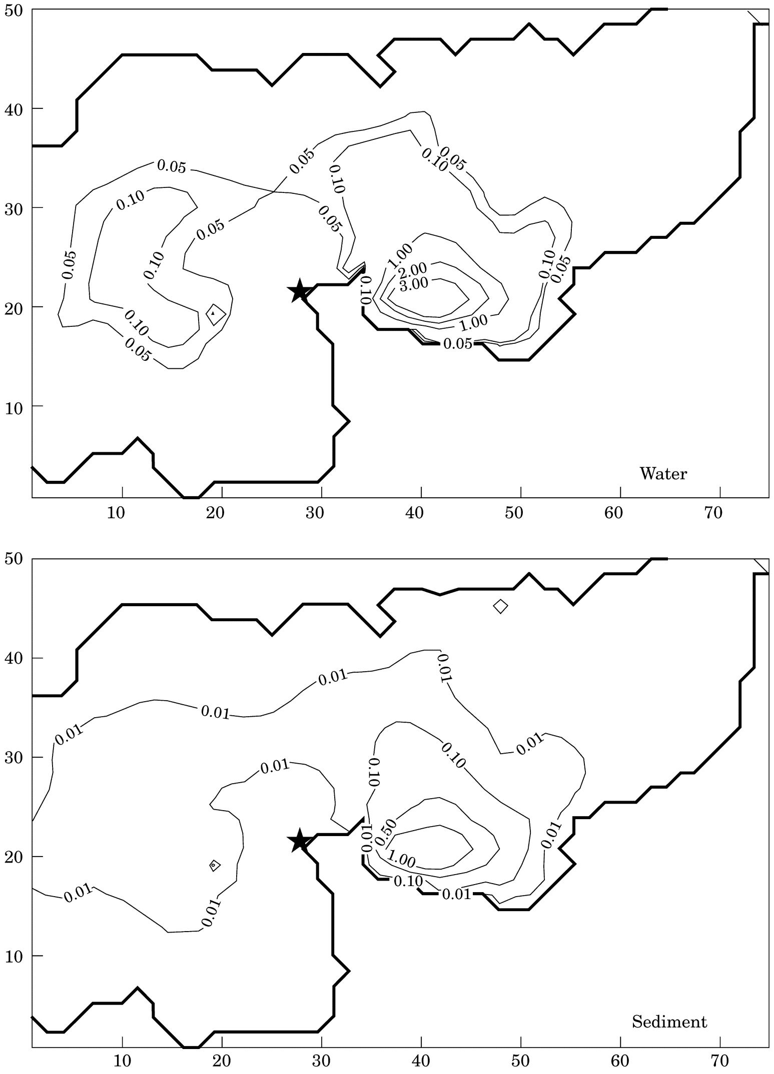

Fig. 6. Distribution of $^{239,240}$Pu in water (Bq m$^{-3}$) and the active bottom sediment (×10$^3$ Bq kg$^{-1}$) 7 years after the beginning of the redissolution process from La Hague.

those obtained for Cs. Concentrations in the source increase quickly and soon reach constant values, although they are not natural equilibrium values since the steady value of the $k_d$ is 27 m$^3$ kg$^{-1}$ (smaller than $k_d^0$. Again, a banded structure is observed in the Channel, although a longer time than in the case of Cs is required to develop it.

The effect of an instantaneous release of $^{239,240}$Pu has finally been studied. $5.0 \times 10^{12}$ Bq are introduced in solution in compartment (26, 24). Concentrations decrease in the three phases and after a time of the order of 4 years natural equilibrium is reached, that is one order of magnitude larger than that of $^{137}$Cs.

## 4. Conclusions

A numerical model based upon residual circulation has been developed to simulate the long-term transport of non-conservative radionuclides in the English Channel. The residual currents and average suspended matter distributions have been obtained from previously validated hydrodynamic and suspended sediment models. The exchanges of radionuclides between the liquid and solid phases have been described in terms of kinetic transfer coefficients instead of using equilibrium distribution coefficients. Thus, the model can be applied to situations out of equilibrium.

Three different situations (redissolution from a contaminated sediment, an instantaneous release and a continuous release) have been analysed. It has been observed that the redissolution from the sediment and an instantaneous release are similar processes in the following aspects:

1. After the initial specific activity increase, concentrations decrease quickly and tend to almost stationary values, now slowly decreasing.

2. These stationary values correspond to natural equilibrium values, as can be observed from the time evolution of the $k_d$ distribution coefficient.
3. In both cases, a patch moving along the French coast is obtained.

In the case of a continuous release, stationary concentrations are soon reached in the source area, although they remain out from natural equilibrium due to the presence of the external supply. A banded structure, showing decreasing concentrations out from the French shore, is observed from La Hague to Dover.

The behaviour of $^{137}$Cs and $^{239,240}$Pu is similar. However, the time scale of the processes are clearly different due to the different affinities of both radionuclides to remain fixed to the solid phases. Thus, the time required by Pu to reach equilibrium in the cases of redissolution and instantaneous release is more than one order of magnitude larger than the corresponding time for Cs. Also, the transit time of Pu from La Hague sediment particles to the east of Cotenin is 7 years, which is in agreement with previous estimations by other authors. This is in constrast with the 3 months transit time of Cs from La Hague to Dover, which is a characteristic value of a conservative radionuclide.

The sediment half-life concept has been introduced and computed for Cs and Pu. The corresponding value for Pu is an order of magnitude larger than the Cs value, which is again due to the larger affinity of Pu to remain fixed to the solid phases. However, this parameter has been calculated in an idealized situation in which contamination in the sediment is located in a reduced area ($5 \times 5\,\mathrm{km}^2$) and with background concentrations in water and suspended matter. In a real case, considerably higher values should be expected. Also, higher values would be obtained if a model consisting of two consecutive reversible reactions (Ciffroy, Garnier, & Pham, 2001) is applied to describe the exchanges of radionuclides between the liquid and solid phases. The first reaction describes an exchange process between dissolved ions and some non-specific sites present on the particles. The second reaction represents a reversible and slower sorption to more specific sites. Thus, the values given in this paper can simply be used to reflect the different kinetics of Cs and Pu.

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