Characteristic times in the English Channel from numerical modelling: supporting decision-making

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Abstract

A numerical model that simulates the dispersion of radionuclides in the English Channel has been applied to study the dispersion of conservative and non-conservative radionuclides released from the La Hague nuclear fuel reprocessing plant. The model is based upon previous work and now is able to simulate dispersion over long timescales (decades), explicitly including transport by instantaneous tidal currents and variable wind conditions. Wind conditions are obtained from meteorological statistics using a stochastic method. Outputs from the model are treated using time-series analysis techniques. These techniques allow the determination of characteristic times of the system, transport velocities and dispersion factors. This information may be very useful to support the decision-making process after an emergency situation. Thus, we are proposing that time-series analysis can be integrated with numerical modelling for helping decision-making in response to an accident. The model is first validated through its application to actual releases of ^{99}Tc and ^{125}Sb, comparing measured and computed concentrations, and characteristic times for three radionuclides are given next: a perfectly conservative one, a very reactive one (^{239,240}Pu) and ^{137}Cs, which has an intermediate behaviour. Characteristic transport velocities and dispersion factors have been calculated as well. Model results are supported by experimental evidence.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Numerical models that simulate the dispersion of radionuclides in the sea have been widely developed (Prandle 1984, Breton and Salomon 1995, Thiessen *et al* 1999, Aldridge *et al* 2003,

Periáñez 2005a, Monte *et al* 2006 among many others). These models can be applied in the assessment of contamination following accidental or deliberate releases of radionuclides into the marine environment and of their radiological consequences through the calculations of doses. It is also useful to determine characteristic timescales of a given system for contamination assessments. Many published works concern the determination of timescales for different systems such as the atmosphere, ground waters, rivers, estuaries and the sea (Zimmerman 1988, Prandle 1984, Salomon *et al* 1995, Waugh and Hall 2002, Holzer and Hall 2000, Braunschweig *et al* 2003, Delhez and Deleersnijder 2002, Shen and Haas 2004, Beckers *et al* 2007, Mercier and Delhez 2007) since the earlier papers by Bolin and Rodhe (1973) and Takeoda (1984). In the marine environment, the concept of age (as the transit time is often denoted) has been used to infer the horizontal circulation of shelf seas (Prandle 1984, Salomon *et al* 1995, Delhez and Deleersnijder 2002) and has also been applied in estuaries (Shen and Haas 2004). In this sense, the age of a tracer particle was defined as the time elapsed since it was released into the sea from a point source. Solid theoretical studies on the concept of age have also been published in recent years (Delhez *et al* 1999, Deleersnijder *et al* 2001, Delhez and Deleersnijder 2002).

On the other hand, time-series analysis is a very interesting mathematical technique that can be used in the study of the spatial and temporal evolution of a magnitude when environmental data analysis is required (Hewit 1992). Usually, it is directly applied to experimental data (Baeza *et al* 2001) and has been applied, for instance, to calculate transit times, mean speed and dispersion factors of tritium released from a nuclear power plant in the Tagus River (Baeza *et al* 2001, 2005).

In this paper both techniques, numerical modelling and time-series analysis, are combined. Time-series analysis is applied to study the output from a numerical model that simulates the dispersion of radionuclides in the marine environment. In particular, the model simulates the dispersion of radionuclides in the English Channel. Radionuclides are released from Cap de La Hague nuclear fuel reprocessing plant in France (see figure 1). Using the time-series analysis, some characteristic timescales of the system and dispersion factors for any point of interest may be calculated. Having an estimation of these magnitudes may be very useful in the assessment of contamination after an accident: even without running a dispersion model, the time required for a contamination patch to travel from the source to a given point (for instance a coastal town), the time during which such a point is being affected by contamination or the speed of the contamination patch may be known, as well as the expected dilution of the patch.

Thus, we are proposing that time-series analysis can be integrated with modelling to support the decision-making process after an emergency situation. A first approach may be obtained from the timescales and dispersion factors given by the time-series analysis and, if more detailed results are required, the dispersion model is run. Nevertheless, the information provided by the time-series analysis has oceanographic interest in itself.

Transit times of radioactive tracers have already been determined in the English Channel from experimental data (Guegueniat *et al* 1993, Bailly du Bois *et al* 1995, Guegueniat *et al* 1994, Povinec *et al* 2003), as well as from numerical models (Salomon *et al* 1995, Deleersnijder *et al* 2001, Delhez and Deleersnijder 2002). However, a new aspect of this work is that, to the authors' knowledge, characteristic times for non-conservative radionuclides have not been calculated before using numerical modelling. This is due to the difficulties in modelling the dispersion of these radionuclides, since interactions with suspended matter and bed sediments must be described.

The model used to simulate the dispersion of non-conservative radionuclides in the English Channel is based upon some previous work (Periáñez and Reguera 1999, Periáñez 2000, 2003). The first two papers describe a model that simulates tide-induced dispersion of conservative

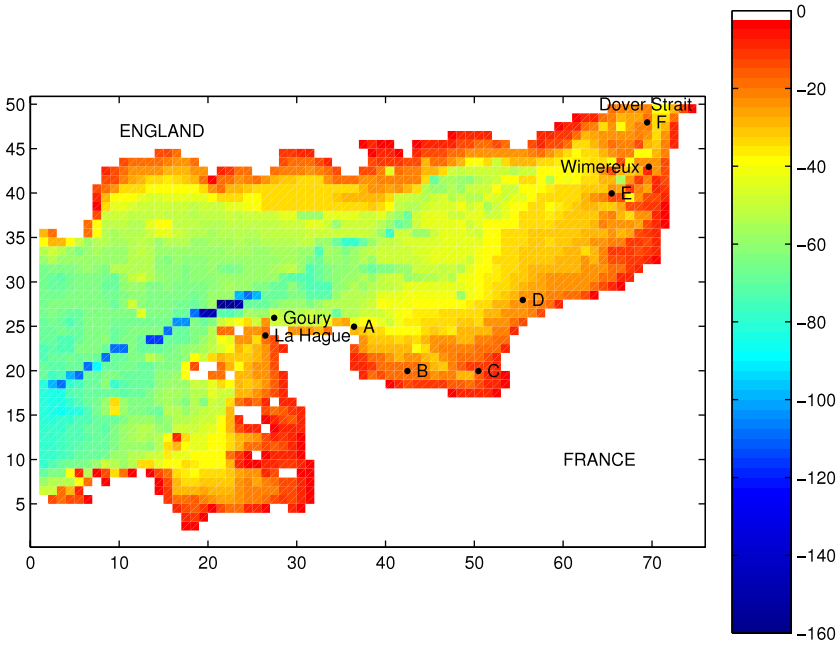

Figure 1. Topography of the English Channel showing the locations of all sites mentioned in the text. Water depths are given in metres. Each number in the axis gives the grid cell number and thus corresponds to 5000 m.

and non-conservative radionuclides, respectively, in the English Channel. Thus, they could only simulate relatively short timescales (a few months). The last work describes a long-term dispersion model for non-conservative radionuclides that simulates timescales of the order of decades. This model uses a residual (mean) circulation in the English Channel, as well as the annual mean wind speed and direction. These models have now been modified to be able to simulate long-term dispersion (years to decades) of non-conservative radionuclides explicitly including transport by instantaneous tidal currents and variable wind conditions deduced from meteorological statistics. Consequently, the new developments described in this paper may be summarised in the following:

- Development of a long-term (decades) dispersion model for non-conservative radionuclides that includes mixing by instantaneous tidal currents and variable wind conditions. The long-term capability is required for highly reactive radionuclides, which have little mobility. Thus, long simulations are required to evaluate timescales along the English Channel.
- The application of time-series analysis techniques to the evaluation of characteristic times of the system for both conservative and non-conservative radionuclides.
- The demonstration that the determination of characteristic times may be combined with numerical modelling for a more efficient decision-making process after an accident.

The model is described in section 2, together with the time-series analysis that has been carried out. Results are presented in section 3. First, the new features of the model are validated through its application to a real case: dispersion of actual ^{99}Tc and ^{125}Sb discharges from La Hague. Then some numerical experiments are carried out to determine characteristic times in three cases: for a conservative radionuclide, for ^{137}Cs and for ^{239,240}Pu.

2. Model description

2.1. Water circulation

A 2D depth-averaged barotropic model has been used. This is justified by the dominance of barotropic over baroclinic mechanisms in the shallow and well-mixed waters of the English Channel (Breton and Salomon 1995).

First, the 2D hydrodynamic equations (Periáñez 2005a) are solved over the domain for the two main tidal constituents, M_2 and S_2 (Moon and Sun main tides respectively; more details on these concepts may be seen in Pugh (1987) and Periáñez (2005a)). The output of the tidal model has been validated through the comparisons of measured and computed tidal currents and surface elevations at several points over the English Channel. Details may be seen in Periáñez and Reguera (1999) and are not repeated here. Nevertheless, tidal analysis (Pugh 1987) has now been carried out and tidal constants (amplitudes and phases; each tide constituent may be described as a pure harmonic function) for each constituent are stored in files that are later read by the dispersion model for a fast computation of tidal currents. This is a common procedure in rapid response models (for instance Periáñez 2005a; 2005b; 2006).

The residual circulation (strictly speaking Eulerian residual transport velocities as described in (Delhez 1996)) corresponding to the annual mean winds, southwest 6 m s^{-1} (Breton and Salomon 1995), is described in Periáñez (2003). These mean currents are directed to the east. This residual flow will be used in some simulations. Now, a residual wind-induced current database has been created. It contains mean currents produced by winds blowing from each sector at their corresponding mean speeds, which are given by Bailly du Bois and Dumas (2005). A stochastic wind may be generated from the wind rose in the English Channel, which gives the wind direction frequency in percentage of time, and a Monte Carlo procedure (Elliott 2004). This stochastic wind reproduces the mean annual wind in the English Channel. A stochastic wind is generated for each simulated day and, following Bailly du Bois and Dumas (2005), wind on day i, W_i, is affected by winds on the 3 days before:

$$W_i = 0.65W_i + 0.28W_{i-1} + 0.05W_{i-2} + 0.02W_{i-3}. \quad (1)$$

These authors have found that this represents better the time required for the model to reach equilibrium after a shift in the wind direction. The wind-induced current is obtained from the database of residuals for each day using the weighting function given above.

Stochastic winds are required, since long-term simulations must be carried out (100 years in the case of plutonium). Thus, actual meteorological data or forecasts cannot be used. It must be commented that stochastically generated winds have already been successfully used in oil spill simulations in the Irish Sea (Elliott 2004).

2.2. Radionuclide dispersion model

The dispersion model is based upon an advection/diffusion dispersion equation to which the terms describing the interactions between the dissolved radionuclides and the sediments are added.

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 English Channel (Periáñez 2000). This suspended matter model is depth-averaged and includes advection–diffusion of suspended particles, a standard formula to represent flocculation (Pejrup 1988, Mehta 1989, Clarke and Elliott 1998), 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

and Elliott (1998) and Prandle *et al* (2000). The erosion term formulation is based on the erosion constant concept (Nicholson and O'Connor 1986, Prandle 1997). Details can be seen in Periáñez (2000).

The dispersion model considers that radionuclides can be present in three phases: solution, suspended matter and active bottom sediments (particles with a diameter <62.5 μm), as denoted by Benes *et al* (1994). It is considered that the exchange of radionuclides between the liquid and solid phases is governed by a single reversible reaction (Nyffeler *et al* 1984). Thus, 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 (dimensions $[T]^{-1}$). Bed sediments which are able to interact with water are considered to be in a well-mixed surface sediment layer with thickness $L = 10$ cm. A parameter, f, describes the fraction of active sediment. Full equations have been presented before and are not repeated here (see for instance Periáñez 2000, 2003, 2005a).

The diffusion coefficient has been selected according to a standard formulation that relates it to the grid spacing. Indeed, following Breton and Salomon (1995), it was taken as $K = 51$ m^2 s^{-1}.

To solve the equations, spatial and temporal discretisation is carried out: the English Channel was divided into 3750 grid cells (forming a 75×50 matrix). 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). The hydrodynamic equations were solved using the standard explicit finite difference scheme described by Flather and Heaps (1975). In the dispersion model, the MSOU (monotonic second order upstream) explicit finite difference scheme, as described in Vested *et al* (1996), was applied to solve the advection terms and a second-order accuracy scheme (Kowalick and Murty 1993) was also applied to the diffusion ones.

A higher spatial resolution would be required to solve small-scale features of water circulation in the English Channel. However, the model includes the transport of radionuclides fixed to suspended sediments and radionuclide exchanges between water, suspended sediments and bed sediments. Because of these processes, a new stability condition for the time step arises (Periáñez 2005a). This condition is more restrictive as the radionuclide reactivity increases. In the case of plutonium, for instance, the time step had to be reduced to 6 min. Moreover, simulations for up to 100 years are required, as commented on below. Thus, we had to find a compromise between minimum acceptable spatial resolution and running times.

2.3. Characteristic times

Since studies on oceanic timescales started in the 1980s several concepts have been applied, such as transit and residence times (Takeoda 1984, Prandle 1984). More recently, the general theory of 'age' has been described (Delhez *et al* 1999, Deleersnijder *et al* 2001, Hall and Haine 2004) and more sophisticated concepts, such as age distributions, introduced. Each defined timescale supplies information about a given aspect of the behaviour of the studied system.

The following characteristic times have been considered in our system for the dissolved phase, given the objectives of the work:

- $T_{\mathrm{L}} - T_{\mathrm{d}}$: give the time interval during which a given point is affected by contamination. T_{L} is the time required for the arrival of the leading edge of the contamination patch to the point and T_{d} is the time required by the patch to pass the point.
- T_{p}: the period of the oscillations in radionuclide concentration at a given point.
- T_{t}: the transit time of the radionuclide patch from the source to a given point.

The first two characteristic times, T_L and T_d are directly obtained from the time evolution of the radionuclide concentrations at the selected point. Their determination has been recommended by the USEPA (2002) and, indeed, they are calculated in the program QTRACER2 (USEPA 2002).

For the evaluation of T_p it has been considered that the temporal evolution of the radionuclide concentration at a given point may be described as a Fourier series (Miró and Periáñez 2006). Then the concept of a periodogram is applied. It is obtained by transforming the time-series from a time domain to a frequency domain through a fast Fourier transform. The periodogram provides the most relevant frequencies w_k, which correspond to the peaks in such periodograms. The periodogram intensity $I(w_k)$ is defined as (full details may be seen in Hewit (1992)):

$$I(w_k) = \frac{2}{N}\left\{\left[\sum_{t=1}^{t=N} C(t)\cos(2\pi w_k t)\right]^2 + \left[\sum_{t=1}^{t=N} C(t)\sin(2\pi w_k t)\right]^2\right\} \tag{2}$$

where $k = 1, 2, \ldots, q$; w_k is the frequency, $C(t)$ is the radionuclide concentration at time t, and $q = (N-1)/2$ for odd N and $q = N/2$ for even N. The periodogram is then the plot of $I(w_k)$ against w_k up to the Nyquist frequency of 0.5 cycles per sampling interval, $w_k = 0.5$ (which corresponds to the smallest identifiable wavelength of two samples) (Hewit 1992). Then it is finally obtained that

$$T_p = 1/w_k. \tag{3}$$

The transit time from the source to a given point has been experimentally obtained in the English Channel and the North Sea by measuring the time elapsing between a given concentration maximum at La Hague and the (same) maximum at the considered point. The works of Kautsky (1973), Mauchline (1980), Guegueniat *et al* (1993, 1994) Dahlgaard *et al* (1995) and Povinec *et al* (2003) are relevant in this sense. Although very interesting results are provided, it cannot be guaranteed that the measured pulses at both points are the same, since a patch rapidly becomes distorted by turbulent mixing. Moreover, the obtained transit times depend on the particular environmental conditions (for instance meteorology). Consequently, results which are difficult to interpret are sometimes obtained. As commented before, transit times may also be evaluated from numerical models (Prandle 1984, Salomon *et al* 1995, Delhez and Deleersnijder 2002 among others).

In this work, the transit time is defined as the elapsed time which gives the best similarity between signals (radionuclide concentrations versus time) in the two locations. First, time-series of radionuclide concentrations at the source and all the points of interest are obtained from the numerical simulation. Next, the transit time is obtained from the maximum of the cross-correlation function between the source and each point (Salomon *et al* 1995). This function estimates the correlation $R(k)$ between two time-series, which are obtained from the results of the simulations. One, $C_s(t)$, corresponds to the time-series of the radionuclide concentration at the source. The other, $C_j(t)$, corresponds to radionuclide concentrations at a given sampling point, j, whose values are shifted in time with respect to the first series as a function of a lag k. In our case, the values of the lag were increased in 1-day steps. Thus, the similarity between both signals is quantified by the cross-correlation function:

$$R(k) = \frac{\langle C_s(t)C_j(t+k)\rangle}{\sqrt{\langle C_s(t)^2\rangle}\sqrt{\langle C_j(t+k)^2\rangle}} \tag{4}$$

where R is the cross-correlation function and $\langle\,\rangle$ means time averaging. The maximum of the cross-correlation function occurs for a given value of k, which is a statistical evaluation of the transit time between the two points:

$$T_t = k. \tag{5}$$

The uncertainty in T_t has been obtained in the following way: the uncertainty ϵ in the maximum of R has been obtained from its standard error (SE) as $\epsilon(R_{max}) = 1.96SE$ (Box and Jenkins 1976, Zar 1984). Thus, a confidence interval for R_{max} is obtained, $R_{max} \pm \epsilon(R_{max})$, from which a confidence interval for the transit time, $T_t \pm \epsilon(T_t)$ can be deduced.

It is necessary to discuss the definition which has been adopted for the transit time. Since dissolved material moves in fluids by both advection and diffusion, a patch becomes rapidly distorted and the transit time is not a unique and simple concept to define. Experimentally it is even more complicated since any specific pattern, like a sharp peak for instance, easily identifiable in one place does not necessarily produce a similar pattern at some distance. Thus, the definition given by equation (5), used by Salomon *et al* (1995) and Baeza *et al* (2001, 2005), corresponds to a statistical evaluation of the mean transit time between two points.

One limitation of this approach (Delhez and Deleersnijder 2002) is that the maximum of the cross-correlation function is generally poorly defined because of the distortion of the signal at the observation point. This is caused by the spreading/diffusion of the patch on its way from the source. This is especially true when this definition is applied to experimental data or if the model is run for real discharges from the source, since variations on the discharge rates may not be as intense as they can be in the case of a numerical experiment. Indeed, the numerical experiments in this paper have been designed, with the appropriate definition of the source term, to avoid this problem and to have significant correlations, as will be shown below.

A second limitation found by Delhez and Deleersnijder (2002) is that this method gives an average advection timescale and slightly underestimates the transit times. From a management perspective, this is not an objection since conservative estimations would be provided.

Finally, the temporal variations in transit times (caused by changes in hydrodynamic conditions and/or by different discharge scenarios, which lead to different statistics) cannot be assessed with this approach. Nevertheless, the objective of the work consists of giving estimations of the average values of the characteristic times of the system. From a management perspective, it is very different to have a transit time to a given sensible point of the order of several hours than of the order of several days. A calculation of the actual transit time for an actual release would require to run the hydrodynamic model using meteorological forecasts, to calculate the expected actual currents, and then to run the dispersion model using such computed currents. Consequently the approach completely loses its utility to help the decision-making process after an emergency. Moreover, in previous calculations made with conservative radionuclides, Miró and Periáñez (2006) found that the characteristic times in the English Channel do not vary significantly with the discharge scenario. On the other hand, hydrodynamic conditions are changing in the present simulations as are meteorological conditions. Thus the characteristic times which we are providing may indeed be considered as mean values.

Characteristic times here defined tend to respond to the following questions: after an accident, when will the patch begin to arrive at my location? How long will such a location be affected by contamination? What is the mean speed of the patch? How much will the contaminant be diluted? Fast answers are required by decision-making managers. This way of asking questions in based upon the USEPA QTRACER2 program (USEPA 2002). Thus, although other definitions of the transit time are more theoretically founded, as the general theory of 'age' developed by Delhez *et al* (1999) and Deleersnijder *et al* (2001), the method used here, keeping in mind its limitations, is sufficient for the purposes of the work. We must add the fact that numerical models have never been used before to calculate transit times of non-conservative radionuclides.

Some characteristic times have also been defined for the bed sediments. The sediment half-life has been defined before (Periáñez 2003) as the time in which radionuclide concentration in the sediment decreases by a factor of 2. If the decrease in radionuclide concentration at a

given point in the sediment (after the contamination patch has passed) begins at time t_s, the time-series giving the evolution of this concentration is fitted, from t_s on, to an exponential decay curve

$$C(t) = C(t_s)\exp\left(-\frac{\ln 2}{T_h}t\right) \tag{6}$$

where T_h is the sediment half-life. T_h and t_s, as well as their uncertainties, are obtained from numerical fitting to a exponential curve. It must provide a regression coefficient larger than 0.996 (Zar 1984).

Finally, the delay between the maximum concentrations, for a given point, in the dissolved phase and the bed sediment, T_{lag}, has been determined from the maximum of the cross-correlation function between both time-series.

3. Results and discussion

3.1. Model validation

Computed tides have been compared with observations in the English Channel (Periáñez and Reguera 1999) and results will not be repeated here. The suspended matter distribution is described in Periáñez (2000), where all required input parameters are also described. In particular, since the English Channel is a highly dynamic environment in terms of tides, sediments are essentially composed of coarse material (Boust 1999). Mud deposits can only be found along both shores in the areas of weaker tidal currents (amplitudes of the order of 0.5 m s^{-1} or less). Thus, as an approximation, it was considered that the parameter f, describing the fraction of active sediments (section 2.2), is constant over all the domain and with a value $f = 0.1$. The water–sediment interaction module has previously been tested in the English Channel (Periáñez 2000, 2003).

The new process of wind generation by the stochastic method has been tested by applying the model to simulate the dispersion of ^{99}Tc and ^{125}Sb, using real discharges from La Hague over the years 1991–1992 (monthly values). A comparison between the measured (Masson *et al* 1995) and computed time-series of concentrations of these radionuclides at two points (figure 1) in the English Channel can be seen in figure 2. Generally speaking, the model produces activity concentration levels which are similar to measurements for both points and radionuclides. It has to be noticed that the model is forced with stochastically produced wind conditions, not with actual meteorological data for the period of interest (the objective of this exercise is to observe if the stochastic wind generation process produces realistic results). Consequently, a perfect fitting to measurements should never be expected. Instead, it should be observed if activity levels and the general trends are reproduced. The spatial distribution shows a banded structure with decreasing concentrations away from the French shore, as has been observed (Guegueniat *et al* 1996) in the English Channel. As an example, the distributions of ^{125}Sb in March and December 1992 are shown in figure 3.

It is known that some of the Sellafield releases in the Irish Sea may travel to the south and enter the English Channel. However, the influence of Sellafield on ^{99}Tc activities in the English Channel plume is expressed as a mean background level of 0.3 Bq m^{-3} (Bailly du Bois *et al* 1995) and, consequently, has been neglected here.

3.2. Characteristic times, velocities and dispersion factors

Some numerical experiments have been carried out to evaluate the characteristic times. A discharge of an arbitrary magnitude (10^{10} Bq s^{-1}) is carried out from La Hague during 1 month

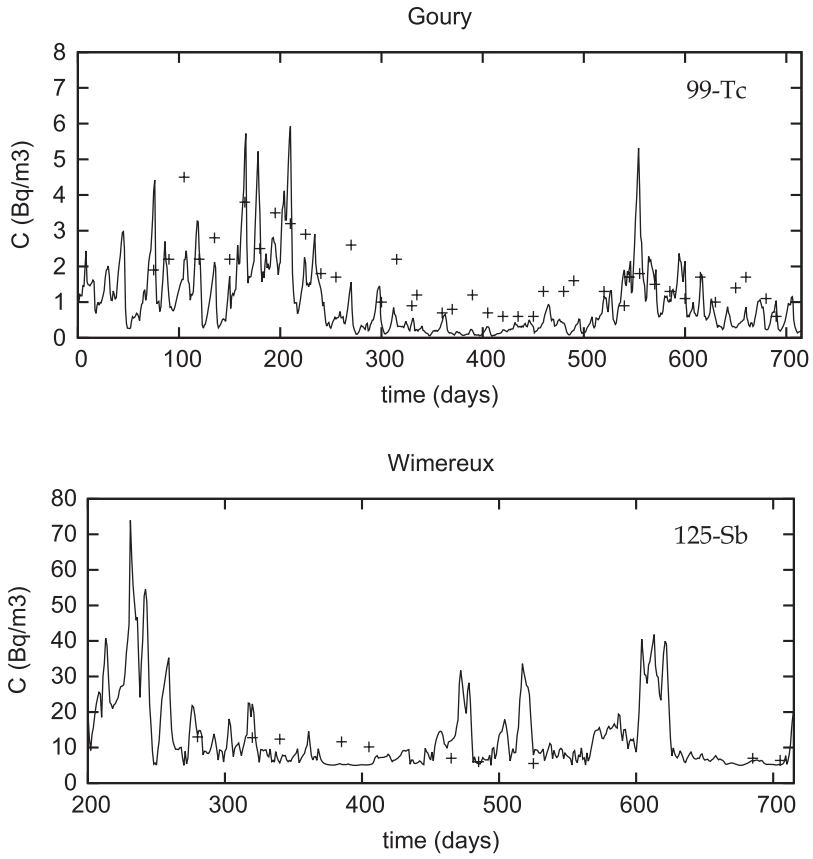

Figure 2. Computed (lines) and measured (points) ^{99}Tc and ^{125}Sb concentrations (Bq m^{-3}) at two points in the English Channel (Goury and Wimereux, see figure 1 for locations). Day 1 corresponds to 1 January 1991.

at a constant rate. Other authors have carried out radionuclide releases similar to those used in the present exercise to study dispersion in the aquatic environment. This is the case of tritium releases from the Ascó nuclear power plant to the Ebro River (Spain) during 1991 (Pujol and Sánchez-Cabeza 1999) or the releases of ^{99m}Tc into Manila Bay (Hughes *et al* 2004). The temporal evolution of activity concentrations at several locations in the English Channel are obtained and stored in files that are later used for the time-series analysis. The selected points are located approximately along the path followed by the maximum of the contamination patch as it travels along the English Channel. As has been shown in Periáñez (2003) and figure 3, the patch essentially moves along the French shore. Points are shown in figure 1 and their grid coordinates, distances from the source and water depths are detailed in table 1. These points are selected just as examples of possible sensitive areas where contamination assessment is required.

The following numerical experiments have been carried out:

(i) Conservative radionuclide. The residual circulation corresponding to the annual mean winds (southwest, 6 m s^{-1} (Breton and Salomon 1995)) described in Periáñez (2003) is used. Simulation time: 1 year.
(ii) Conservative radionuclide. Stochastic winds. Simulation time: 1 year

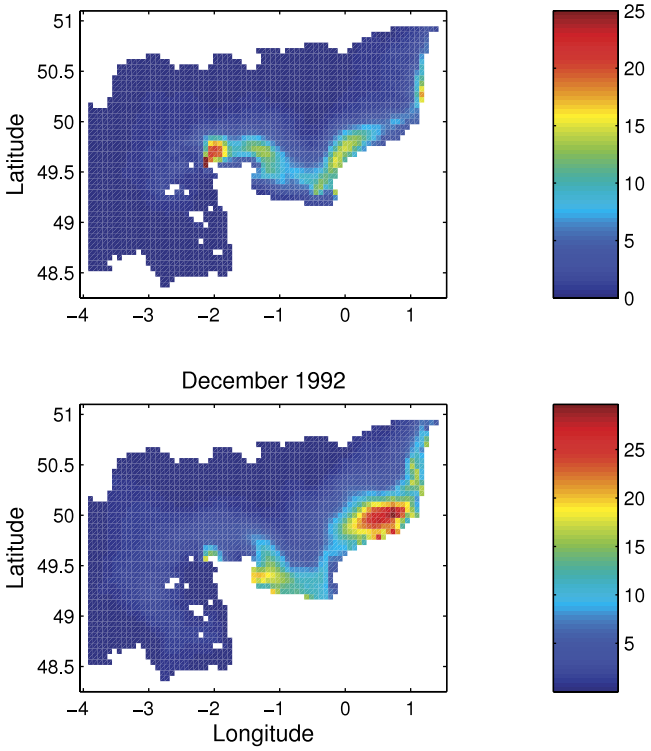

Figure 3. Computed distribution of dissolved ^{125}Sb (Bq m^{-3}) in March and December 1992.

Table 1. Points where model results are obtained. LH is La Hague plant. Distances from each point to La Hague are also given.

Point	Coordinates	Depth (m)	Distance (km)
LH	(26, 24)	20	0
A	(36, 25)	45	50.25
B	(42, 20)	20	82.46
C	(50, 20)	12	121.65
D	(55, 28)	25	146.37
E	(65, 40)	25	210.77
F	(69, 48)	20	246.22

(iii) ^{137}Cs. Residual circulation corresponding to the annual mean winds (southwest, 6 m s^{-1}). Simulation time: 10 years.

(iv) ^{239,240}Pu. The same residual circulation as in the ^{137}Cs experiment. Simulation time: 100 years.

In the case of a perfectly conservative radionuclide the simulations are carried out over 1 year, since this time is longer than the transit time from La Hague to Dover Strait, which has been estimated to be of the order of some months (2–8) from ^{125}Sb measurements (Guegueniat *et al* 1994). However, the simulation time in the case of ^{137}Cs was increased to 10 years since, because of interactions with the solid phase, reactive radionuclides are less mobile than conservative ones and transit times are significantly increased. In the case of ^{239,240}Pu

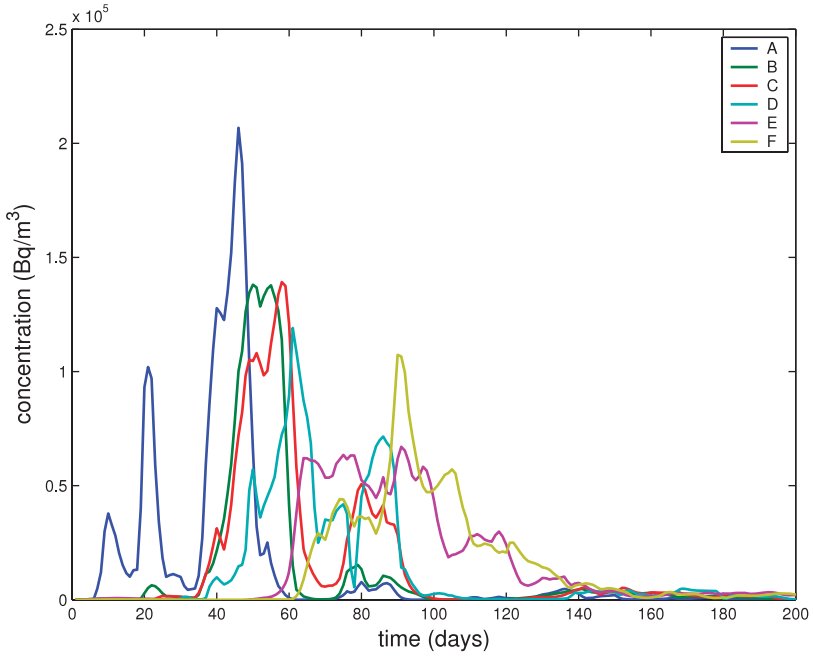

Figure 4. Computed temporal evolution of concentrations of a conservative radionuclide at points indicated in figure 1 and table 1 (experiment 2).

an even longer time of 100 years was required. As an example, the time evolution of radionuclide concentrations at several points in the English Channel may be seen in figure 4 for a conservative radionuclide (experiment 2). Obviously, there is a delay in the arrival of the signal as going to the east in the English Channel and a decrease in concentrations. Similarly, the time evolution of ^{137}Cs concentrations in water and bed sediments, at the same locations, are presented in figure 5 (experiment 3). After the initial sediment contamination, there is a slow process of radionuclide redissolution, thus the sediment acts as a long-term delayed source of previously released radionuclides. Of course, this implies that the contamination also persists in the dissolved phase for a longer time.

Characteristic times for experiments 1 and 2 do not present any significant differences. In other words, essentially the same results are obtained with the mean annual wind and the stochastic winds (provided that these reproduce the annual mean value). This is not surprising and has already been observed by Sandery and Kampf (2005). Consequently, the residual circulation corresponding to the annual mean wind was used in the cases of non-conservative radionuclides since noise is reduced in the temporal sequences. Characteristic times for the dissolved phase and bed sediments may be seen in tables 2 and 3 respectively.

The periods of the radionuclide concentration oscillations, T_{p}, are not given in table 2. However, mean values over the English Channel are 16.5 ± 1.0, 14 ± 3 and 15.1 ± 1.0 days for experiments 1, 2 and 3 respectively. This corresponds to the spring–neap tidal cycle of 14.8 days. In the case of plutonium it could be calculated only at the point that is closest to La Hague, but the same value of 14.8 days was obtained. Thus, spring–neap tidal cycles are the most relevant processes producing oscillations in radionuclide concentrations (apart from the obvious semidiurnal tides with periods of about 12 h, which have been filtered out from the model output). As an example of these calculations, a periodogram is shown in

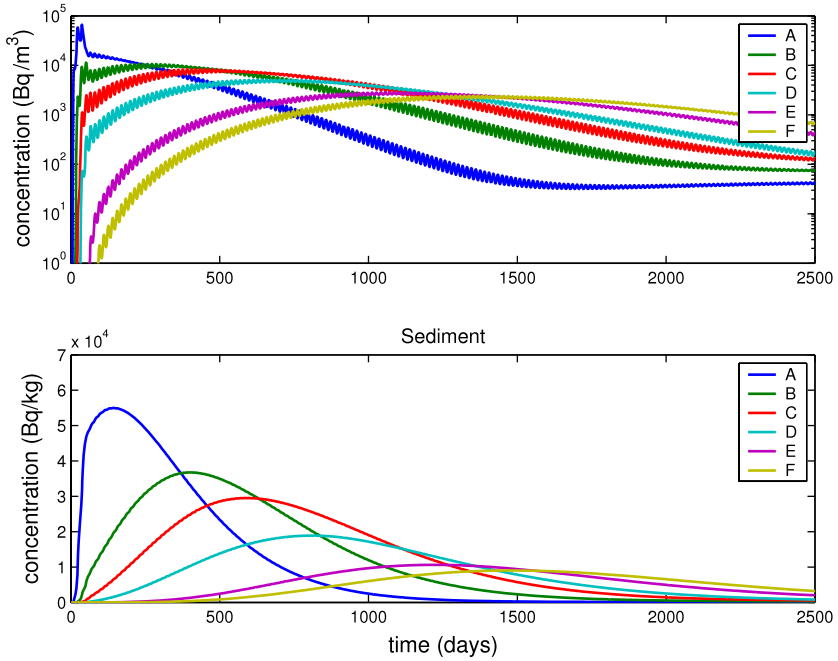

Figure 5. Computed temporal evolution of ^{137}Cs concentrations in water and bed sediments at points indicated in figure 1 and table 1 (experiment 3).

Table 2. Characteristic times (in days, d) for the dissolved phase. NC means that the time could not be calculated because the simulation was not long enough. Note the different units (years, y) for experiment 4.

	Experiment 1		Experiment 2		Experiment 3		Experiment 4	
Point	$T_L - T_d$ (d)	T_t (d)	$T_L - T_d$ (d)	T_t (d)	$T_L - T_d$ (d)	T_t (d)	$T_L - T_d$ (y)	T_t (y)
A	8–69	13 ± 4	6–134	18 ± 3	6–1700	12 ± 2	0–27	8.5 ± 0.3
B	19–93	28 ± 5	19–146	29 ± 3	15–2500	180 ± 30	3–63	18.6 ± 0.5
C	24–120	36 ± 5	35–135	31 ± 3	22–3000	340 ± 50	8–74	28.2 ± 1.6
D	32–135	40 ± 5	37–153	36 ± 4	NC	500 ± 60	12–90	36 ± 2
E	50–134	67 ± 6	54–172	64 ± 3	NC	850 ± 70	NC	54 ± 4
F	54–146	75 ± 7	62–176	64 ± 3	NC	1000 ± 80	NC	65 ± 4

figure 6. It corresponds to point A in experiment 1. The maximum intensity appears for $w_k = 0.060\,44$ day^{-1}, which corresponds to $T_p = 16.54$ days.

The transit time from La Hague to Dover is in the range 64–75 days for a conservative radionuclide. This value compares with previous calculations of about 3 months (Salomon *et al* 1995). Using the general theory of 'age', Delhez and Deleersnijder (2002) obtained a value of about 4 months. Gueguenia*t et al* (1994) estimated, from measurements, the transit time of ^{125}Sb from La Hague to Dover, obtaining values in the range 2–8 months. The transit time of non-conservative radionuclides increases since adsorption to sediments makes these radionuclides less mobile in the aquatic environment. Thus, from the present model, the transit time of ^{137}Cs to Dover is 2.7 ± 0.7 years and it is 65 ± 4 years in the case of ^{239,240}Pu.

The mobility of plutonium in the marine environment is much lower than that of conservative radionuclides and caesium. Indeed, Boust *et al* (1997) and Boust (1999) have

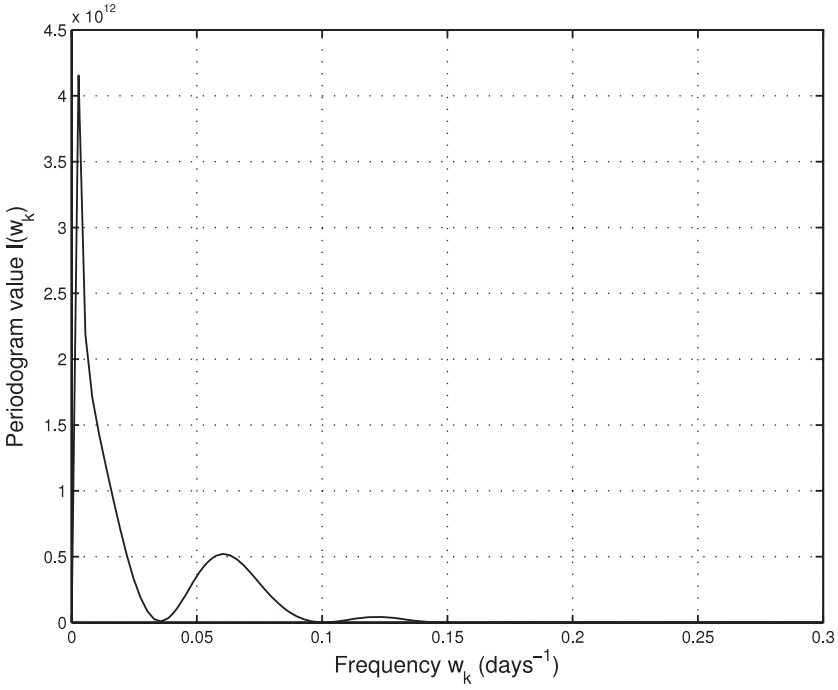

Figure 6. Periodogram obtained for experiment 1 at point A.

Table 3. Characteristic times for the sediments in the experiments for ^{137}Cs and ^{239,240}Pu. NC means that could not be calculated because the simulation was not long enough. Note the different units (days or years) for ^{137}Cs and ^{239,240}Pu.

	^{137}Cs		^{239,240}Pu	
Point	T_h (d)	T_{lag} (d)	T_h (y)	T_{lag} (y)
LH	79 ± 2	24 ± 7	0.6 ± 0.1	0.06 ± 0.01
A	157 ± 5	36 ± 17	2.3 ± 0.1	0.25 ± 0.48
B	238 ± 9	92 ± 61	4.4 ± 0.2	0 ± 1
C	277 ± 14	77 ± 67	7.2 ± 0.3	0 ± 2
D	330 ± 20	62 ± 69	7.7 ± 0.3	0 ± 3
E	400 ± 30	31 ± 40	8.3 ± 0.3	0 ± 4
F	425 ± 30	3 ± 25	NC	0 ± 4

estimated from measurements a transit time of plutonium in the eastern English Channel ranging from 10 to 50 years. Thus, the significant increase in the plutonium transit time that the model predicts is supported by experimental evidence.

In the case of ^{137}Cs, Kautsky (1973) has estimated, from measurements, transit times in the North Sea which are almost a factor 2 larger than those later deduced by Gueguenia*t et al* (1994) from ^{125}Sb measurements. These last authors attributed the discrepancy to a mistake in the experimental procedure of Kautsky (the measured pulses at both points were not the same). Nevertheless, it seems rather reasonable that the ^{137}Cs transit time is something intermediate between that of a conservative radionuclide and that of plutonium, as is the case for caesium mobility as well. Moreover, in a more recent work, Povinec *et al* (2003) have estimated a ^{137}Cs transit time between La Hague and the southern North Sea of about 1 year, about a factor

of 2 larger than the estimation of Salomon *et al* (1995) for a conservative radionuclide. Our results are consistent with the fact that the transit time of ^{137}Cs should be larger than that of a conservative radionuclide.

The confidence that we may have in the calculated transit times depends on the values of R_{max}. Thus, for experiment 1 R_{max} is in the range 0.958–0.860. For experiments 2 and 3 ranges are, respectively, 0.745–0.629 and 0.712–0.405. In the case of experiment 4 R_{max} is essentially constant, with a value of 0.303. In all cases these values are significantly greater than zero since the maximum error in R_{max} is 0.016. R_{max} decreases as the radionuclide reactivity increases since interactions with sediments make the pulse change its shape more rapidly. Also, R_{max} is larger for experiment 1 (annual mean wind) than for experiment 2 (stochastic wind). This is an expected result since variable winds enhance deformation of the patch, and this leads to a poorer correlation between signals at two different locations.

From the assessment of several models, Monte *et al* (2005) concluded that the main factor of model uncertainty is represented by the difficulties for predicting quantitatively the complex interactions of radionuclides with the sediments. Thus, not only the selected parameters for the present model, but also the formulation of the interaction processes itself, constitute a source of uncertainty which affects the computed characteristic times. A multi-model approach (Monte *et al* 2006) could be useful in this sense to point out if the presented results obtain or not consensus from modellers. An example of such a multi-model approach applied to simulate the behaviour of Chernobyl radionuclides in the Dnieper-Bug estuary (Ukraine) may be seen in the paper cited above.

The spreading of the radionuclide patch because of the wind shear and turbulent diffusion caused T_d to increase from points A to F. Thus, contamination persists for a longer time further from the source. This behaviour is the same for all radionuclides. The arrival of the patch, given by T_L, is similar for conservative radionuclides and for ^{137}Cs, although it is much longer in the case of the highly reactive ^{239,240}Pu. Nevertheless, although T_L may be similar in some cases, the transit time (in the statistical sense in which has been defined) is not.

The sediment half-life (table 3) increases from points A to F, although the kinetics of radionuclide redissolution is the same in the entire English Channel. This is again revealing that the sediment acts as a long-term delayed source and that points located to the east are affected by radionuclides released from sediments of the western areas for a longer time. Values of half-lives for ^{239,240}Pu are about one order of magnitude larger than for ^{137}Cs, which is again indicating that the former is less mobile than the latter. In the present work, a one-step model consisting of a single reversible reaction is used to describe adsorption/release kinetics, although the use of more complex models involving consecutive and/or parallel reactions will modify sediment half-lives (Periáñez 2004) and may affect transit times as well.

Values for R_{max} in the determination of T_{lag} (see section 2.3) are again significantly greater than zero. It may be seen in table 3 that the delay, T_{lag}, between maximum concentrations in water and sediment decreases further from the source. As has been shown in Periáñez (2003), when radionuclides are introduced from a continuous source, water and sediment radionuclide partition is not at equilibrium in the source area. However, radionuclide partition between both phases is at equilibrium far from the source location. Consequently, the decrease in T_{lag} with increasing distance from the source is revealing such a trend towards equilibrium. Thus, it is confirmed that, from a management perspective, the equilibrium k_d is an adequate parameter to describe radionuclide speciation only far from the source location in the case of non-instantaneous releases. Nevertheless, if the radionuclide is highly reactive the adsorption kinetics will be faster and equilibrium will be reached sooner. In particular, water and sediment are at equilibrium as near as point B in the case of plutonium (table 3).

Table 4. Characteristic velocities. The mean values along the English Channel are given and errors are their corresponding standard deviations.

Experiment	v_L (km day^{-1})	v_t (km day^{-1})
1	4.8 ± 0.8	3.4 ± 0.3
2	4.7 ± 1.8	3.5 ± 0.6
3	5.0 ± 2.0	0.20 ± 0.04
4	0.037 ± 0.020	0.010 ± 0.003

Some velocities may also be calculated from the characteristic times. They are:

$$v_L = \frac{D}{T_{\mathrm{L}}} \tag{7}$$

and

$$v_t = \frac{D}{T_{\mathrm{t}}} \tag{8}$$

where D is the distance from the source to the corresponding point (table 1). The first velocity gives an indication of the speed of the edge of the contamination patch; thus it may be useful for management purposes to make conservative estimations. The second velocity is an indication of the speed of the patch as a whole. These velocities are summarised in table 4 for all the experiments. It may be seen that the same values are obtained if the residual circulation for the average wind conditions or the stochastic winds is used. v_L is some 5 km day^{-1}, slightly higher than v_t. This is due to the deformation of the patch by shear dispersion (Periáñez 2005a). For a conservative radionuclide, v_t is about 3.5 km day^{-1}, in agreement with the previous estimation in Periáñez (2000) with the short-term dispersion model, namely 2.8 km day^{-1}. The velocities are significantly reduced for non-conservative radionuclides since water–sediment interactions make them less mobile. In particular, the average velocity of plutonium is two orders of magnitude lower than the residual currents responsible of the transport of conservative elements. This is again in excellent agreement with the results, from measurements, of Boust *et al* (1997): these authors found average velocities ranging between some km per year to some tens of km per year (i.e. of the order of 10^{-3}–10^{-2} km day^{-1}).

Dispersion factors have been finally calculated along the English Channel. The dispersion factor (DF) is defined as:

$$\mathrm{DF} = \frac{A_t}{A_p} \tag{9}$$

where A_t is the total activity released by the source and A_p is the total accumulated activity in a given grid cell:

$$A_p = \sum_{t=T_{\mathrm{L}}}^{t=\infty} V(t)C(t). \tag{10}$$

In this equation $V(t)$ is the grid cell volume and $C(t)$ is the radionuclide concentration at time t.

DFs are given in table 5. Generally speaking, they increase from La Hague towards Dover, as should obviously be expected, where they are about 7 for all the simulations. It is worth observing that the DF remains essentially constant when the stochastic wind is used. This is due to the large dispersion, caused by changing wind episodes, which starts immediately after radionuclides are released.

Table 5. Dispersion factors along the English Channel for all considered points and experiments.

	Experiment			
Point	1	2	3	4
A	3.66	6.95	3.69	3.44
B	3.48	8.07	3.39	2.78
C	3.45	6.65	3.39	3.05
D	4.57	7.58	4.52	4.16
E	6.46	6.49	6.50	6.52
F	7.08	6.96	6.99	8.31

4. Summary and conclusions

A model to describe the dispersion of non-conservative radionuclides over long timescales (years to decades) in the English Channel has been developed from previous work. The new aspect of this model is that it explicitly includes tidal mixing (by instantaneous tidal currents) and variable wind conditions, which are obtained from meteorological statistics. This new model formulation was tested through its application to simulate the dispersion of actual ^{99}Tc and ^{125}Sb releases from the La Hague reprocessing plant. Computed concentrations of this radionuclide were, in general, in agreement with measurements.

Some numerical experiments were carried out once the model was validated. Model output from these experiments were processed using time-series analysis techniques to evaluate characteristic times for water, sediments, average transport velocities and dispersion factors. These magnitudes were calculated for three radionuclides with different geochemical behaviours: a perfectly conservative one, a very reactive one (^{239,240}Pu) and ^{137}Cs, with intermediate character. The computed transit time from La Hague to Dover in the case of a conservative radionuclide was a few months, and increased to about 65 years in the case of plutonium. Similarly, average velocity for this radionuclide was two orders of magnitude lower than the water residual velocity. These results are supported by experimental evidence, and it is worth commenting that characteristic times and velocities have not been calculated using numerical models before in the case of non-conservative radionuclides.

Essentially the same results were obtained if the annual mean wind or stochastic winds were used. This confirms results from previous modelling works. Also, it was confirmed that the equilibrium k_d should be used with care for contamination assessments: water–sediment radionuclide partition is not at equilibrium close to the source location. The meaning of *close* depends on the geochemical behaviour (reactivity) of the radionuclide.

Characteristic times, average velocities and dispersion factors for a given environment may be useful to support the decision-making process after an emergency situation. For instance, the time of arrival of a contamination patch to a given sensible point, the time during which the point is to be affected by contamination and the maximum concentration expected may be quickly estimated from them. These estimations may be carried out under more or less conservative approaches (for instance the use of v_L or v_t) and for radionuclides with different geochemical behaviours. If more details are required in a practical problem, a full model run may be carried out to complete the results. Of course, in the case of highly reactive elements (such as Pu) the calculation of transit times may not be relevant in this sense because of their extremely low mobility. However, valuable information about their environmental behaviour is obtained.

Although the model and the time-series analysis have been applied to the English Channel, these techniques may of course be applied to any other marine, estuarine or freshwater environment, as well as to other contaminants such as heavy metals.

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