A particle-tracking method for simulating the dispersion of non-conservative radionuclides in coastal waters

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Abstract

A particle-tracking method has been used to simulate the dispersion of non-conservative radionuclides in the sea. Three dimensional turbulent diffusion and the interactions between water, suspended matter and bottom sediments are simulated using a stochastic method. Kinetic transfer coefficients, as in finite difference models, are used to describe the transfers between the liquid and solid phases. Deposition of suspended matter and erosion of sediment are also included in the model. The method has been applied to simulate the dispersion of ^{137}Cs and ^{239,240}Pu in the English Channel and the results have been compared with those of a finite difference model. The results from both techniques are, in general, in good agreement.

Keywords: Model; Dispersion; Particle-tracking; Finite difference; Radionuclides; English Channel

1. Introduction

The state-of-the-art models used to simulate the dispersion of radioactivity in the sea consist of finite elements or, mainly, finite difference models that solve the hydrodynamic equations together with the advection–diffusion dispersion equation, in either a two dimensional or a three dimensional form (Harms, 1997; Periáñez &

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Reguera, 1999). If the model is applied to simulate the dispersion of non-conservative radionuclides, then the interactions with the solid phases (suspended matter and bottom sediments) must also be considered. This implies that the suspended matter equation, including the settling of particles, deposition and erosion of the sediment, must be solved too (Aldridge, 1998; Margvelashvily, Maderich, & Zheleznyak, 1997; Piasecki, 1998; Periáñez, 1999, 2000). Absorption/desorption reactions are usually described using kinetic transfer coefficients instead of the less appropriate equilibrium distribution coefficients, k_d. These models are computationally expensive due to the fact that a large number of equations must be solved. Also, since finite difference models work with radionuclide concentrations, the whole computational grid must be swept each time step. Finally, the CFL condition (Kowalick & Murty, 1993) limits the size of the time step that can be used if an explicit scheme is used to solve the hydrodynamic equations. In consequence, large CPU times may be required even using recent supercomputers (Periáñez, 1999).

An alternative approach is to make use of particle-tracking techniques. In this kind of model, the impact of the CFL criterion can be reduced by making the hydrodynamic computations off-line. Also, advection can be simulated to a high degree of accuracy since numerical dispersion (which appears when the advection equation is solved with finite differences) is not introduced. Particle-tracking models have already been used to simulate the dispersion of conservative tracers and oils spills (Hunter, 1987; Elliott, Dale, & Proctor, 1992; Proctor, Elliott, & Flather, 1994a, b). In such applications, a release of radioactivity to the sea is modelled as a number of discrete particles, each particle being equivalent to a number of units (Bq, moles, atoms, etc.). Then the path followed by each individual particle is computed, turbulent diffusion being modelled as a three dimensional random walk (Monte Carlo) process. The density of particles is calculated to obtain the radioactivity concentrations at the end of the simulation. The main difficulty that appears in the simulation of the dispersion of non-conservative radionuclides is the treatment of absorption and desorption: how to decide if each particle is fixed to suspended matter or bottom sediments (if initially dissolved) or if it is redissolved (if initially present in the suspended matter or the bottom sediment). The main contribution of this paper consists of a new method developed to solve this problem: a formulation (suitable for a particle-tracking model) to describe the transfers of radionuclides between water, suspended matter and bottom sediments, based upon kinetic transfer coefficients and a stochastic method, is presented. It is important to point out that exactly the same physical parameters as in the equivalent finite difference models are used.

The particle-tracking modelling technique is well suited to problems in which high contaminant gradients are involved, since numerical diffusion is not introduced. This, together with the fact that it can give very fast answers, even in a PC, allows the technique to be considered as a very useful predictive tool in the assessment of contamination following accidental or deliberate releases of radionuclides.

The particle-tracking model is presented in the next section, then an application to the English Channel is shown. The model has been used to simulate the dispersion of an instantaneous hypothetical release of radionuclides from La Hague nuclear fuel

reprocessing plant, located on the French shore of the English Channel. Simulations have been carried out for two radionuclides with very different geochemical behaviours: the relatively conservative ^{137}Cs and the high reactive ^{239,240}Pu. A finite difference model of the Channel has been previously developed and validated through the study of the dispersion of these radionuclides, comparing observed and computed concentrations in water, suspended matter and bottom sediments (Periáñez and Reguera 1999; Periáñez, 2000). Thus results of the particle-tracking model will be compared with the output of the finite difference model in equivalent simulations and the relative advantages of each technique will be assessed.

2. The particle-tracking model

2.1. Advective transport

The position vector of a given particle, $\mathbf{r}(t+\Delta t)$, at time $t+\Delta t$ is computed from

$$\frac{\mathbf{r}(t+\Delta t)-\mathbf{r}(t)}{\Delta t}=\mathbf{q}(t), \tag{1}$$

where Δt is the time step used in the model and $\mathbf{q}$ the current vector of components u and v along the x and y axes, respectively. Currents are obtained by running a hydrodynamic model in advance. Standard tidal analysis is used to determine the tidal constants (tide amplitude and phase) for each grid cell of the hydrodynamic model. These constants are evaluated for both components of the flow and can be derived for as many tidal constituents as desired. In this work, only the two main semidiurnal tides, M_2 and S_2, will be considered. Once the tidal constants are known, computation of the flow vector, $\mathbf{q}$, just involves the calculation and addition of a few cosine terms. As a consequence, the evaluation of the tidal advective transport of particles is very fast and is not limited by the CFL criterion.

For real applications, first order accuracy in the particle-tracking scheme is adequate. In simulations of the movement of drogues in an estuarine environment, Elliott and Clarke (1998) found no improvement in the results when a second order accuracy scheme was used to simulate the movement of surface drifters by the particle-tracking technique. Moreover, in ocean dispersion problems, the effects of turbulence will mask any small errors in the advection scheme.

The net residual current in the modelled area must be added to $\mathbf{q}$ since a residual transport cannot be generated with the pure harmonic tidal currents that are used in particle-tracking calculations. The residual flow vectors can also be obtained from the (previously run) hydrodynamic model. Wind-induced transport can be included in the model by assuming that the surface wind-induced current is a percentage of the wind speed, generally 2–3% (Proctor et al., 1994b). This current decreases logarithmically below a depth z_1 (the thickness of the wind-driven surface layer) to zero at a depth z_2. Eckman theory (Pugh, 1987) predicts that the surface wind-induced current due to a steady wind blowing over deep water should be deflected to the right of the wind direction (in the northern hemisphere). However, observational

evidence suggests that this deflection angle can be neglected (Proctor et al., 1994b) in shallow coastal waters.

2.2. Turbulent diffusion

Three dimensional diffusion is simulated using a random walk method. It has been shown (Proctor et al., 1994b; Hunter, 1987) that it is a simulator of Fickian diffusion provided that the maximum size of the horizontal step given by the particle, D_h, is

$$D_h = \sqrt{12 K_h \Delta t} \tag{2}$$

in the direction 2πRAN, where RAN is a random number between 0 and 1. This equation gives the maximum size of the step. In practice, it is multiplied by RAN to obtain the real size at a given time and for a given particle. Similarly, the size of the vertical step is

$$D_v = \sqrt{2 K_v \Delta t} \tag{3}$$

given either towards the sea surface or the sea bottom. K_h and K_v are the horizontal and vertical diffusion coefficients, respectively.

2.3. Radioactive decay

Consider the radioactive decay equation:

$$\frac{\partial C}{\partial t} = -\lambda C, \tag{4}$$

where λ is the radioactive decay constant. This equation can be treated using a stochastic method if it is assumed that the probability p of removal of a particle at each time step is (Hunter, 1987; Proctor et al., 1994b)

$$p = 1 - e^{-\lambda \Delta t}. \tag{5}$$

In practice, a random number is generated for each particle on each time step. If RAN $\leqslant p$ then the particle is removed from the computation.

2.4. Transfers between water, suspended matter and bottom sediments

Consider a two phase system. If the transfers of radionuclides between the two phases are described through the kinetic transfer coefficients k_1 and k_2, the equations that give the time evolution of activity in the two phases are

$$\frac{\partial A_1}{\partial t} = -k_1 A_1 + k_2 A_2, \tag{6}$$

$$\frac{\partial A_2}{\partial t} = k_1 A_1 - k_2 A_2.$$

These equations are easily solved using finite differences. In particle tracking, a label is given to each particle to differentiate if it is in phase l or 2. If the particle is in phase 1, the probability p_1 that the particle goes to phase 2 in each time step is

$$p_1 = 1 - e^{-k_1 \Delta t}. \tag{7}$$

Similarly, if the particle is in phase 2, the probability p_2 that it goes to phase 1 each time step is

$$p_2 = 1 - e^{-k_2 \Delta t}. \tag{8}$$

Thus, in particle tracking, the exchanges between two phases can be modelled as two decay processes with probabilities p_1 and p_2. These processes are treated as the radioactive decay process described above. If a given particle goes from one phase to the other, its label is changed and the new corresponding decay process is considered at the next time step.

This method has been compared with the finite difference solution of the system of Eq. (6). It has been considered that all radionuclides are, at $t = 0$, in phase 1. Thus, the solution given by each method refers to the percentages of radionuclides that are in phase 1 at each following time step. Results obtained by both methods are presented in Fig. 1, using 200 and 10,000 particles in the stochastic simulation. High fluctuations occur with 200 particles, but the finite difference solution is well modelled if 10,000 particles are used in the particle-tracking calculation. The mean value and standard deviation of the difference between finite difference and stochastic solutions are presented in Table 1 for different numbers of particles in

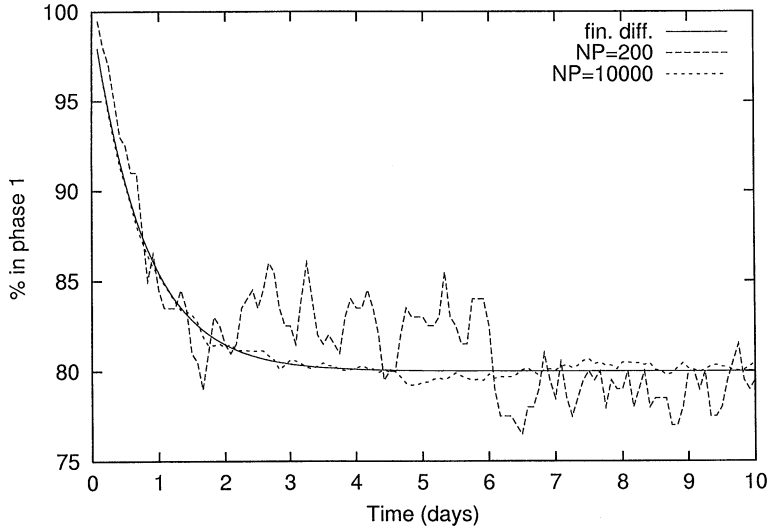

Fig. 1. Time evolution of the percentage of radionuclides in phase 1 given by the solution of the differential equations using finite differences and the particle-tracking method, with 200 and 10,000 particles.

Table 1
Mean value and standard deviation of the difference between the percentage of radionuclides in phase 1 given by finite differences and the stochastic method[a]

NP	$\langle \Delta \rangle$	σ
100	−0.654	2.861
200	−0.637	2.220
500	0.296	1.557
1000	0.335	1.021
10,000	1.56×10^{-2}	0.309
100,000	6.17×10^{-2}	0.114

[a] NP is the number of particles used in the simulation.

the stochastic method. It can be seen that both the mean value and standard deviation of the difference decrease as the number of particles considered in the stochastic simulation increases. Acceptable results are obtained for numbers of particles of the order of 10,000.

The stochastic method can be extended to the case in which there are three different phases: water, suspended matter and active bottom sediments (particles with a diameter $<62.4\,\mu m$). It is considered that the transfer of radionuclides from the solid phases to water is governed by a kinetic transfer coefficient k_2, the transfer from water to suspended matter by k_{1m} and the transfer from water to the sediment by k_{1s}. The absorption of radionuclides depends on the surface of particles per water volume unit. Thus the exchange surface and exchange velocity concepts have been used (Periáñez, Abril, & García-León, 1996; Periáñez & Martínez-Aguirre, 1997; Periáñez, 1999, 2000). Following these papers:

$$k_{1m} = \chi_1 \frac{3m}{\rho R}, \tag{9}$$

$$k_{1s} = \chi_1 \frac{3Lf\phi}{RH}, \tag{10}$$

where χ_1 is the exchange velocity, m is the suspended matter concentration, ρ and R are the density and mean radius of suspended matter particles, L is the average mixing depth (the distance to which the dissolved phase penetrates the sediment), ϕ is a correction factor that takes into account that not all the mass of the sediment is in contact with water and H gives the thickness of the water layer above the sea bottom that interacts with the sediment. In a two dimensional depth-averaged model, H is equal to the water depth. Since particle tracking is three dimensional, H is left as a free parameter to be calibrated.

The decay equations that are equivalent to the differential equations that describe transfers between the three phases, presented for instance in Periáñez (2000), are

$$\frac{\partial C_d}{\partial t} = -k_{1m} C_d - k_{1s} C_d, \tag{11}$$

$$\frac{\partial C_s}{\partial t} = -k_2 C_s, \tag{12}$$

$$\frac{\partial A_s}{\partial t} = -k_2 \phi A_s. \tag{13}$$

where C_d, C_s and A_s are radionuclide concentrations in water, suspended matter and active bottom sediments, respectively. A label is given to each particle to classify in which phase it is present. Depending on the label of the particle, the corresponding decay equation is treated. If the particle is in suspended matter, the probability that it goes to the dissolved phase, in each time step, is

$$p = 1 - e^{-k_2 \Delta t}. \tag{14}$$

Similarly, if the particle is in the bottom sediment, the probability that it is redissolved is

$$p = 1 - e^{-k_2 \phi \Delta t}. \tag{15}$$

If the particle is initially dissolved and its distance to the sea bottom is smaller than H, it can go to any of the two solid phases with a probability

$$p = 1 - e^{-(k_{1m} + k_{1s})\Delta t}. \tag{16}$$

A random number is generated to decide if the particle is effectively removed from solution. If it is, the normalized probability that the particle goes to the sediment is calculated as

$$p = \frac{p_s}{p_m + p_s}, \tag{17}$$

where

$$p_m = 1 - e^{-k_{1m} \Delta t}, \tag{18}$$

$$p_s = 1 - e^{-k_{1s} \Delta t}. \tag{19}$$

A second random number is then generated. If RAN $< p$, then the particle goes to the sediment. If RAN $> p$, then it goes to the suspended matter. Of course, if the distance of the particle to the sea bottom is larger than H, only the decay to suspended matter is considered since such particles cannot interact with the sediment.

A numerical experiment has been carried out to test the method in which a volume of water with a given suspended matter concentration and active sediment on the bottom is considered. A dissolved radioactive tracer is added and the equations that give the time evolution of activities in the three phases are solved using finite differences and the stochastic method. The following realistic parameters have been used: $\chi_1 = 2.1 \times 10^{-8}$ m/s, $k_2 = 1.2 \times 10^{-5}\,s^{-1}$, $m = 10$ ppm, $R = 15\,\mu m$, $\rho = 2600\,kg/m^3$, $L = 0.01$ m, $f = 1$, $\phi = 0.1$, $H = 0.2$ m with 10,000 particles being used in the particle-tracking simulation. The comparison between both methods is presented in Fig. 2, where the time evolution of the fraction of tracer that is dissolved, in suspended matter and in the sediment is presented. The simulation

shows that the stochastic method solution is in very good agreement with the finite difference solution for the three phases. Indeed, solutions corresponding to both methods cannot be distinguished in the case of water and sediment.

2.5. *Suspended matter deposition and sediment erosion*

The suspended matter concentrations over the model domain can be obtained by running in advance a finite difference suspended matter model. Results are then analyzed in a similar way to currents, so that the suspended matter concentration at each point and for any time can be obtained as the simple calculation of cosine functions.

Suspended matter falls to the sea bottom with a settling velocity w_s. If a particle (in the particle-tracking sense, not a suspended matter particle) is fixed to suspended matter, its position above the bottom, z, at time $t + \Delta t$ is obtained from

$$\frac{z(t+\Delta t) - z(t)}{\Delta t} = -w_s. \tag{20}$$

If $z(t + \Delta t) \leqslant 0$, then the particle is considered to fix to the sediment and its label is appropriately changed.

A standard formula for flocculation has been used to represent the increase in the settling velocity as the suspended matter concentration increases (Clarke & Elliott, 1998; Eisma, 1993; Periáñez, 2000)

$$w_s = a_1 m^{a_2}, \tag{21}$$

where a_1 and a_2 are obtained from measurements or from model calibration. The particle-tracking model is three dimensional. If the finite difference suspended matter model is depth-averaged, its output is the depth-averaged suspended matter concentration. A Rouse profile is then used to resolve the vertical structure of suspended matter. This allows the calculation of the suspended matter concentration at height z above the bottom, m_z, from the depth-averaged suspended matter concentration m (Clarke & Elliott, 1998):

$$m_z = m\left(1 - w_s/\beta k u_*\right)\left(\frac{h}{z}\right)^{w_s/\beta k u_*}, \tag{22}$$

where h is water depth, k is the von Karman constant (0.4), β is an arbitrary constant usually taken as 1 (Eisma, 1993; Clarke & Elliott, 1998) and u_* is the scalar friction velocity

$$u_* = \frac{k|\mathbf{q}|}{\ln\left(h/z_0\right) - 1}, \tag{23}$$

where z_0 is the bottom roughness. The corresponding value of m_z is used to calculate $k_{1\mathrm{m}}$ at the position of each particle from Eq. (9).

Erosion of the sediment has been described in terms of the erosion constant concept (Nicholson & O'Connor, 1986). Thus, the probability that a particle is removed from the sediment and incorporated to the water column as suspended

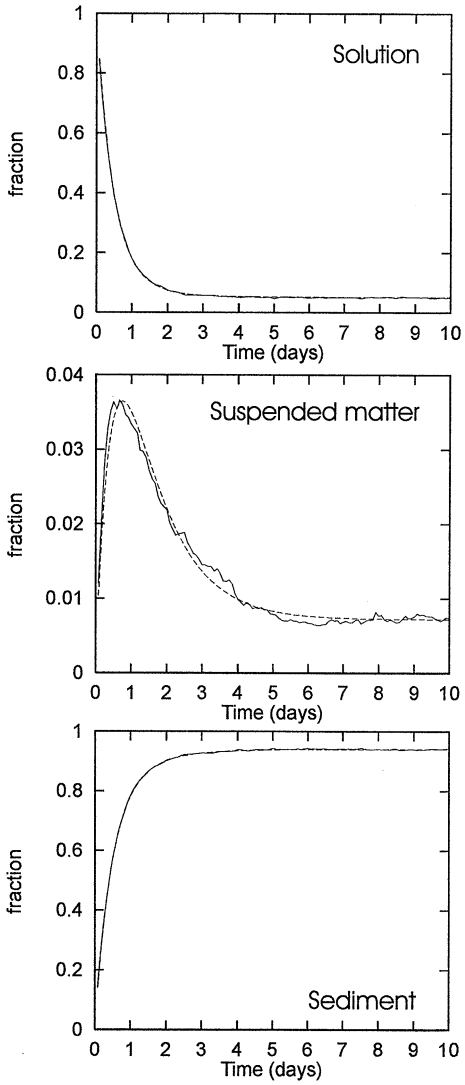

Fig. 2. Time evolution of the fraction of radionuclides in water, suspended matter and bottom sediments given by finite differences and by the particle-tracking method with 10,000 particles. Solid lines correspond to the stochastic solution and dashed lines to the finite difference solution.

matter is

$$p = 1 - \mathrm{e}^{-Ef|\mathbf{q}|^M \Delta t}, \tag{24}$$

where E is the erosion constant and M is some power of the water velocity typically in the range 2–5 (Prandle, 1997). Thus, the erosion description used in previous finite difference models (Periáñez, 1999, 2000) has been converted into a stochastic form. It is considered that erosion can only take place if the water velocity is larger than a

critical erosion velocity, q_{ce}. If this is the case, a random number is generated to decide if the particle is effectively eroded or not.

2.6. Computation of activity concentrations

The output of a particle-tracking model is the position of each particle. Concentrations can be obtained by counting the density of particles (number of particles per surface unit) in each phase. Thus, concentrations in water, suspended matter and bottom sediments are, respectively

$$C_d = N_d R/h, \tag{25}$$

$$C_s = N_s R/mh, \tag{26}$$

$$A_s = N_{sed} R/Lf\rho_s, \tag{27}$$

where N_d, N_s and N_{sed} are the densities of particles in each phase, ρ_s is the bulk density of the sediment and R is the number of units (for instance Bq) that is equivalent to each particle

$$R = \frac{I}{NP}, \tag{28}$$

where I is the radioactivity input and NP is the number of particles used in the simulation.

2.7. Computational scheme

The computational scheme is summarised in Fig. 3. As commented above, a hydrodynamic and a suspended matter model must be run off-line in advance. The results are then analyzed and the tidal constants, for each current component and each constituent, are stored in files that will be used by the particle-tracking program. A similar analysis is carried out with the output of the suspended matter model: in this case, mean suspended matter concentrations, amplitudes and phases must be stored.

Some boundary conditions must be specified when computing the advection and diffusion of particles. In the case of an open boundary, particles that cross it are removed from the computation. In addition, as with a finite difference model, fluxes of particles through the land and sea bed boundaries are not permitted.

The particle-tracking program makes the following operations:

1. Read water depths, input the tidal constants and mean flow database created by the hydrodynamic and suspended matter models.
2. Calculate advective transport, three dimensional diffusion, settling of particles in suspended matter, erode particles in the sediment and compute interactions between particles in solution and particles in the solid phases.
3. Remove from computation particles that have decayed (radioactive decay) and particles that have left the model domain.

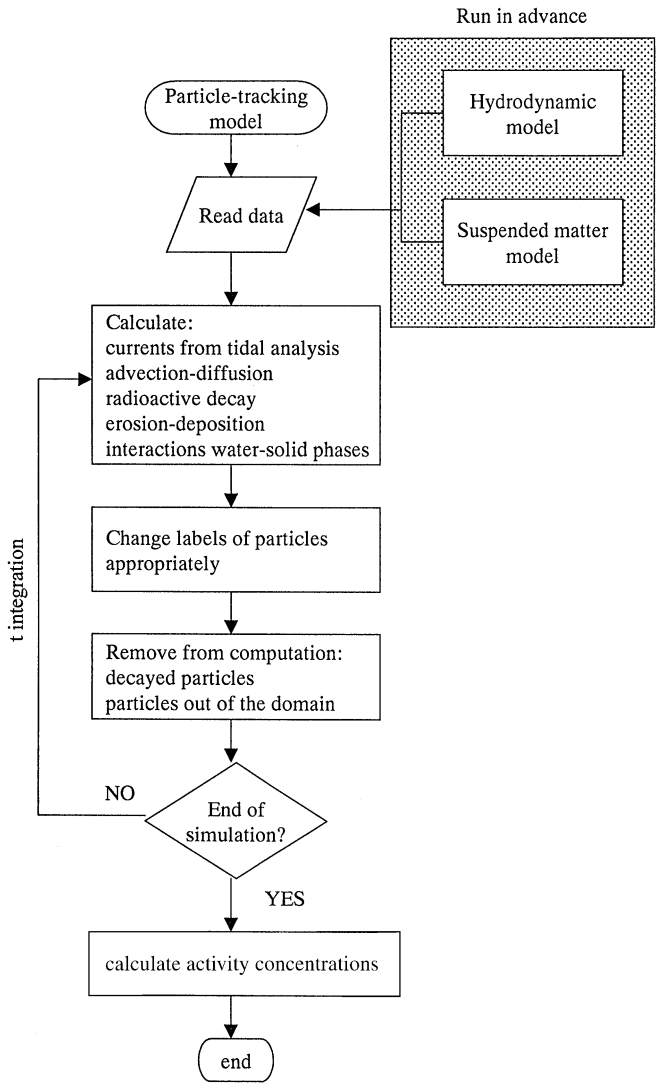

Fig. 3. Flow chart showing the computational scheme.

4. Calculate the activity concentrations at the desired times.

When a continuous release is simulated, the number of particles that are being tracked increases linearly during the computation until the time at which the release ceased is reached. At this time (and for longer times if the computation extends beyond the end time of the release), there are N particles in the simulation (some of which may have decayed or moved out of the region of interest) and therefore the

required accuracy is achieved at the end of the simulation. At intermediate times, for example at 10% of the way through the computation, there will only be $N/10$ particles in the simulation and therefore the accuracy of the intermediate results will be lower than those obtained at the end of the simulation. In such circumstances, the model user must decide on the simulation time at which the highest accuracy results are required. If the interest is in short time-scales after the release, the model should not be run to simulate a long continuous release. However, there is no computational reason why the technique cannot be applied to releases that continue over decades and algorithms have been devised that can mimic both sporadic and long-term spill scenarios. The results of such techniques applied to oil spill simulations are given in Proctor et al. (1994a, b).

3. Application to the English Channel: comparison with a finite difference model

3.1. Results

The modelling technique described above has been used to simulate the dispersion of radionuclides in the English Channel, where they are released from La Hague nuclear fuel reprocessing plant. Simulations have been carried out for two radionuclides with different geochemical behaviours, the relatively soluble ^{137}Cs and the reactive ^{239,240}Pu, so as to test the model response. A finite difference model to simulate the dispersion of these radionuclides in the Channel has already been developed and validated (Periáñez, 2000). The output from both models will be compared.

The parameters used in the particle-tracking model are the same as those used in the finite difference model. They were obtained from the literature or from model calibration (Periáñez, 2000): $E = 2.2 \times 10^{-5}$, $M = 3.5$, $a_l = 5.7 \times 10^{-6}$, $a_2 = 1.6$, $q_{ce} = 0.21\,\text{m/s}$, $L = 0.1\,\text{m}$, $\phi = 0.1$, $R = 15\,\mu\text{m}$, $\rho = 2600\,\text{kg/m}^3$, $\rho_s = 900\,\text{kg/m}^3$, $K_h = 51\,\text{m}^2/\text{s}$. In the case of ^{137}Cs, $\chi_1 = 2.10 \times 10^{-8}\,\text{m/s}$ and $k_2 = 1.16 \times 10^{-5}\,\text{s}^{-1}$; for ^{239,240}Pu, $\chi_1 = 1.51 \times 10^{-5}\,\text{m/s}$ and $k_2 = 0.93 \times 10^{-5}\,\text{s}^{-1}$. The selection of these parameters has been justified in Periáñez (2000) and will not be repeated here. Simulations have been carried out with NP = 50,000 particles and the time-step is fixed as $\Delta t = 3600\,\text{s}$. While there is no stability criterion equivalent to the CFL condition in the particle-tracking calculations, it is wise to ensure that each particle does not move through a distance that exceeds the grid spacing during each time-step. This was satisfied by using a time-step of 3600 s. The vertical diffusion coefficient has been taken as $K_v = 0.01\,\text{m}^2/\text{s}$, the bottom roughness of muddy sediments as $z_0 = 4.0 \times 10^{-4}\,\text{m}$ and good results are obtained, as will be seen, if the thickness of the bottom layer is specified using $H = 3\,\text{m}$.

The model domain is presented in Fig. 4. It extends from 48.3°N to 51.0°N and from 4.0°W to 1.5°E. The particle-tracking model runs in only 6.5% of the time required by the finite difference model (for the same simulation and computer). Particle-tracking simulations can be even faster if the number of particles is reduced.

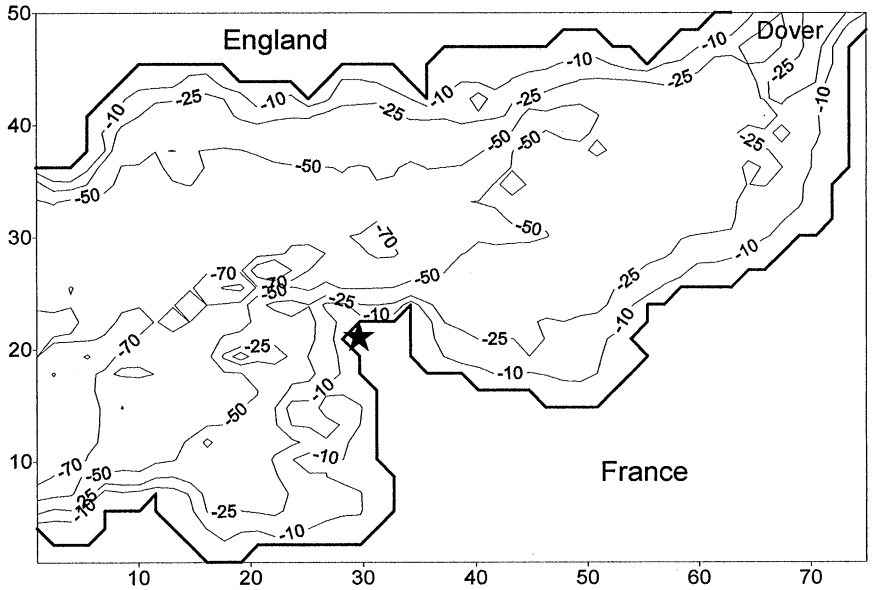

Fig. 4. Model domain. Depths are given in m and the star is La Hague nuclear fuel reprocessing plant. Each unit in the x and y axis is 5000 m.

A hypothetical instantaneous release from La Hague of 5×10^{12} Bq of ^{137}Cs has been considered. The position of dissolved particles at several times after the release is shown in Fig. 5 (snap-shots are presented), which shows that particles move towards Dover Strait due to the fact that a residual current in this direction exists in the Channel. Also, particles are concentrated towards the French shore, as has been deduced from observations (Guegueniat, Herrmann, Kershaw, Bailly du Bois, & Baron, 1996).

Mass conservation in the model has been tested. Thus, total activities present in water, suspended matter and bottom sediments over the model domain have been added from the computed activity concentrations. The magnitude of the initial input is obtained with 100% accuracy.

A comparison between particle-tracking and finite difference results is presented in Fig. 6, for the dissolved phase (depth-averaged) and bottom sediments. Activity maps have been obtained 40 days after the release from La Hague. An excellent agreement between the results of both models is obtained for the dissolved phase. Results for the bottom sediments are also in good agreement. However, results for suspended matter cannot be obtained with the particle-tracking model due to the low affinity of ^{137}Cs for the solid phases. Indeed, 10.260% and 0.048% of the total activity is present in the bottom sediments and in suspended matter, respectively. This implies that, if 50,000 particles are used in the simulation, 5130 and 24 particles are, respectively, in the bottom sediment and in suspended matter. The number of particles in the sediment is enough to calculate the activity concentrations over the

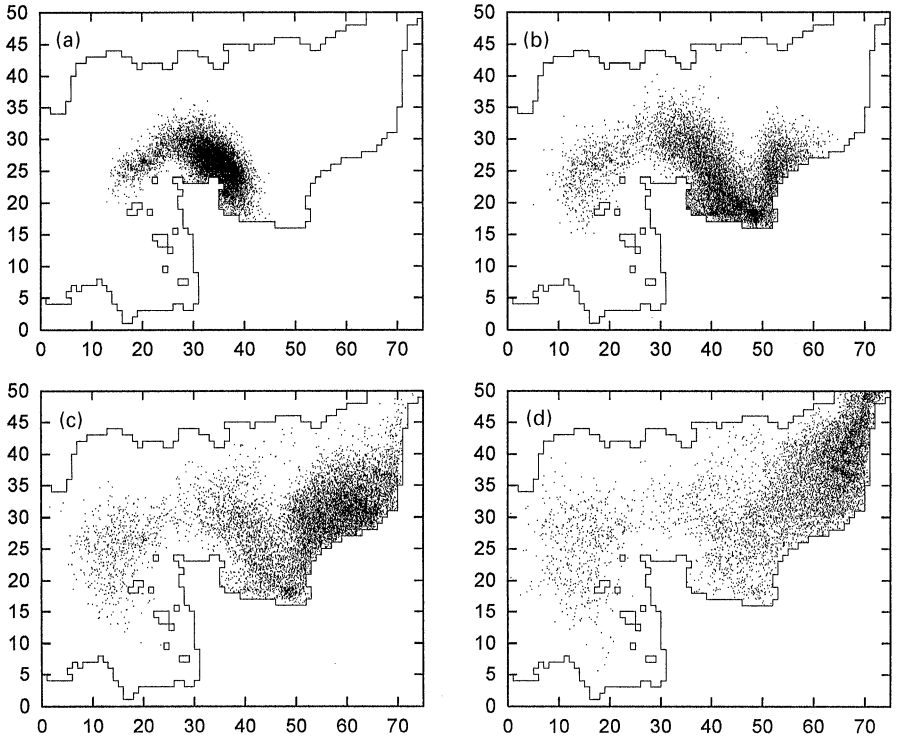

Fig. 5. Dispersion of an instantaneous release of ^{137}Cs from La Hague. The movement of particles in solution 20 (a), 40 (b), 60 (c) and 80 (d) days after the discharge is shown.

Channel from the density of particles but this is clearly not the case for the suspended matter.

A similar simulation has been carried out for a release of $^{239,240}Pu$. Results are presented in Fig. 7 for water, suspended matter and bottom sediments. It shows that, due to the high reactivity of Pu, it remains essentially close to the source, as simulated by the finite difference and particle-tracking models. Activity levels given by the two models are, in general, in good agreement for the three phases. However, it seems that slightly higher concentrations are produced by the particle-tracking model. This discrepancy may be caused by the finite difference model. Effectively, concentration gradients in the case of Pu are larger than in the case of Cs, since all the released Pu remains close to La Hague. Thus, the finite difference model introduces a numerical diffusion (even although a second order accuracy advection scheme is being used) that is more apparent than in the case of Cs. In contrast, the particle-tracking method does not introduce numerical diffusion and, thus, higher concentrations are produced.

In the case of Pu, it is possible to calculate activity concentrations in the three phases since 6.44%, 90.08% and 3.48% of Pu are in water, sediments and suspended

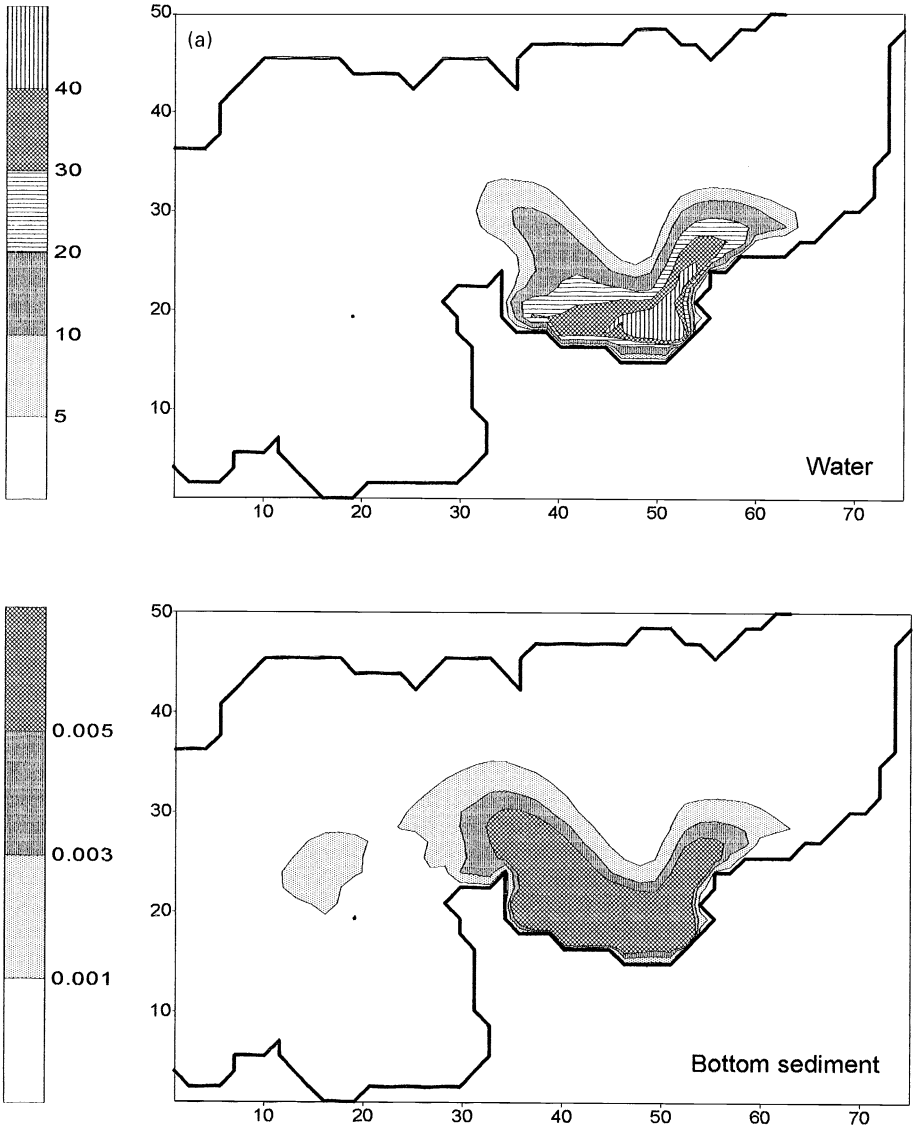

Fig. 6. ^{137}Cs activity concentrations in water (mBq/l) and the active fraction of the sediment (Bq/g) given by the finite difference (a) and the particle-tracking (b) models.

matter, respectively. This implies that, using 50,000 particles, 3220 are in solution, 45,040 in the sediment and 1740 in suspended matter. Thus there are enough particles in each phase to calculate the corresponding densities and activity concentrations.

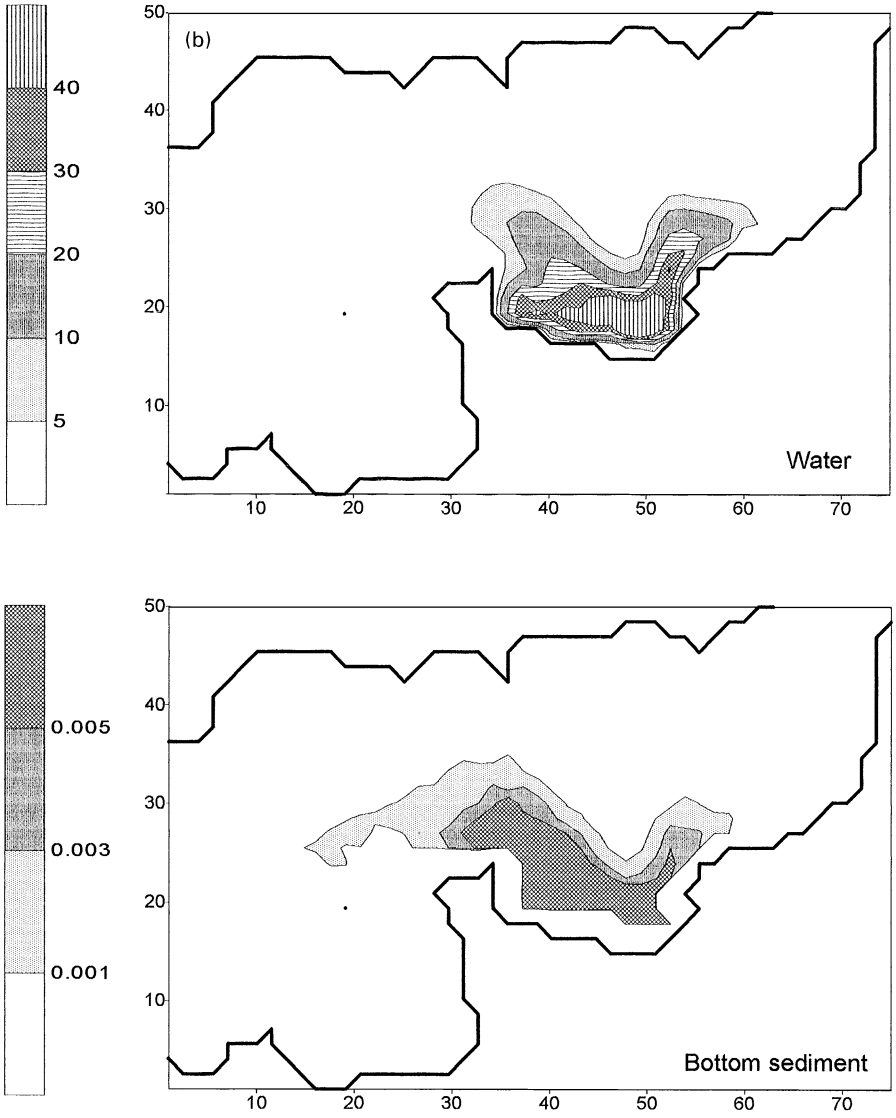

Fig. 6 (*continued*).

3.2. *Discussion*

Particle tracking is a powerful tool that can be applied in the assessment of radioactive contamination following an accidental release in aquatic environments in general. Also, the method can be applied to both conservative and non-conservative radionuclides, using the same conceptual approach for the interactions between

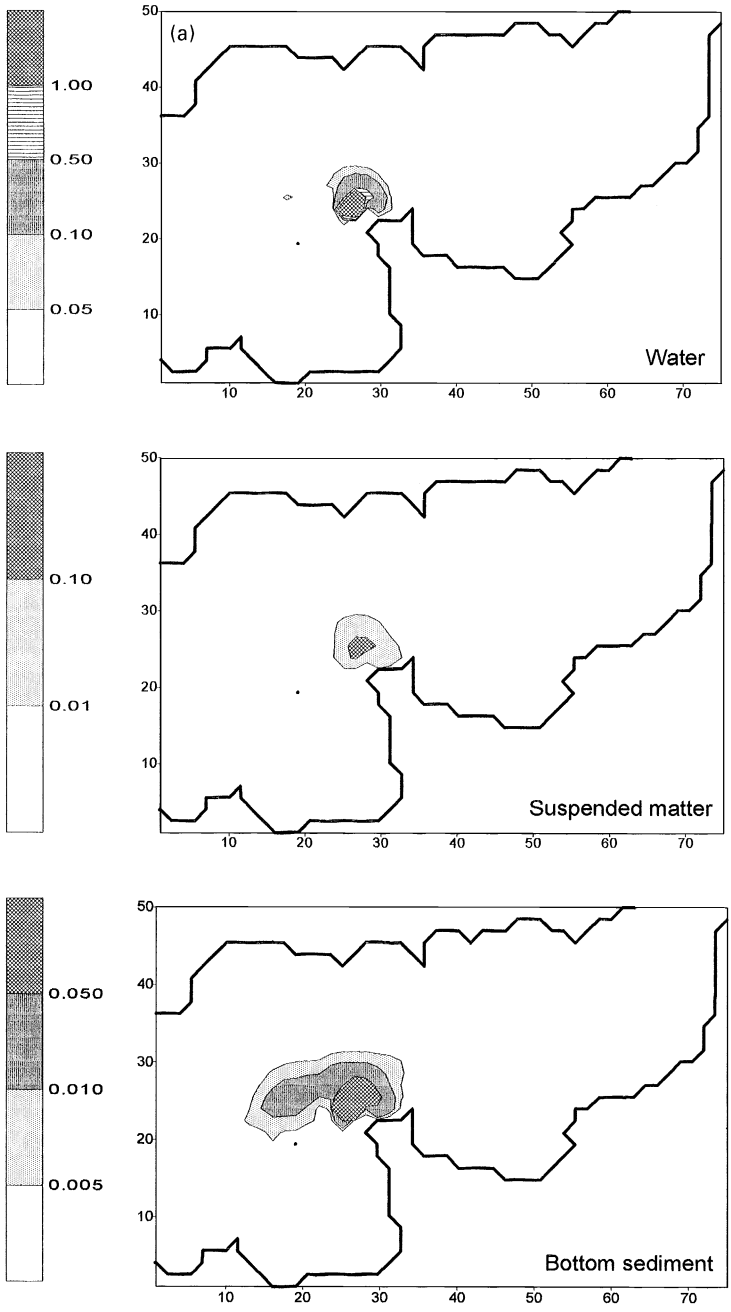

Fig. 7. ^{239,240}Pu activity concentrations in water (mBq/l), suspended matter (Bq/g) and active sediment (Bq/g) given by the finite difference (a) and the particle-tracking (b) models.

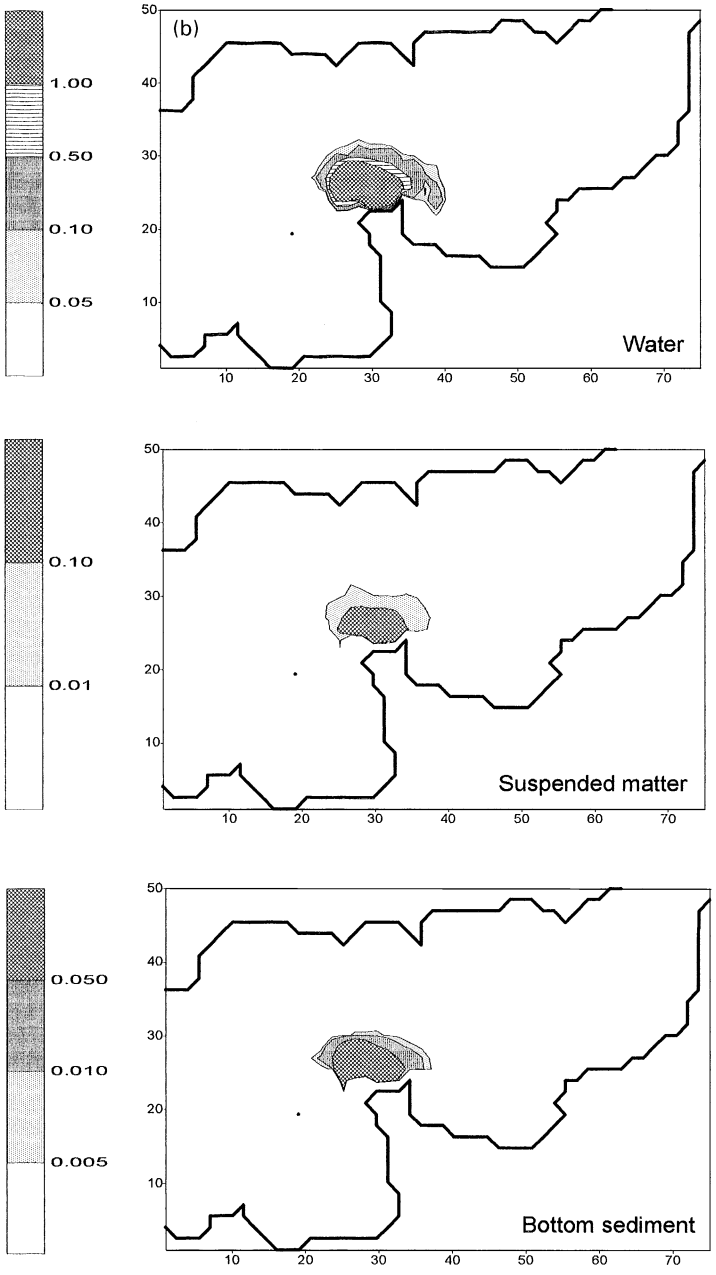

Fig. 7 (*continued*).

liquid and solid phases as used in finite difference models. The particle-tracking technique, combined with tidal analysis and mean flow databases for the hydrodynamics, presents two clear advantages over finite differences: speed of

computation and the fact that it does not introduce numerical diffusion. The number of particles used in simulations depends on the speed of computation and/or the accuracy required. For example, Proctor et al. (1994b) used 4000 particles to simulate an oil spill during real-time forecasting of an incident in the Arabian Gulf. However, it is possible that, due to the geochemical behaviour of certain radionuclides, the activity concentrations in some of the phases may not be resolved. This is the case for ^{137}Cs in suspended matter. The number of particles used in the simulation should be of the order of 10^6 to be able to calculate, with reasonable accuracy, activity concentrations in suspended matter but then the computing time would be similar to that of the finite difference model. However, if we are interested in the assessment of contamination following an accident, it is probably enough to calculate activities in water and bottom sediments, which are the phases where almost all the radioactivity is present. If the most important aspect is the speed of computation, the number of particles can be reduced. For instance, if ^{137}Cs dispersion is simulated using 20,000 particles, good results are still obtained in water and bottom sediments and the computation time is reduced to 2.6% of that required by the finite difference model. In the case of ^{239,240}Pu, the number of particles could be reduced more and activities in the three phases may still be calculated since Pu is more widely distributed between the three phases than Cs.

The particle-tracking method is fully 3-D and takes account of horizontal advection plus turbulent diffusion in the x–y–z directions. In the present application, the tidal and residual flows have been computed by a depth-averaged 2-D hydrodynamic model. A 2-D hydrodynamical model is adequate due to the very strong tidal currents and the vertically well-mixed character of the coastal waters. However, the particle-tracking method is suitable for use with a 3-D hydrodynamic model (e.g. Harms, Karcher, & Dethleff, 2000) and the manner in which vertical stability can inhibit vertical mixing can be parameterised in such applications by making the vertical step-size a function of the stability of the water column.

4. Conclusions

The purpose of this paper is to demonstrate the viability of new particle-tracking algorithms for the simulation of processes that involve chemical speciation. The particle-tracking method is commonly used for oil and chemical spill applications where there is a need for rapid response simulations. In such applications, a significant increase in computational speed is achieved by computing the hydrodynamics 'off-line' and then using tidal prediction and mean flow databases to reconstruct the water movement. This avoids the CFL criterion during the rapid response application, although the particle-tracking time-step must be kept reasonably short in order to maintain the accuracy of the first order advection scheme that is used to compute the particle movement. While it would also be feasible to solve finite difference speciation equations using a pre-computed flow field, the proposed technique has the advantage that the computational time will be very short if a relatively small number of particles is used in the simulation.

Moreover, the method will be more efficient if the contaminant patch covers only a small fraction of the grid domain (Hunter, 1987) and the accuracy of the results can be improved by releasing larger numbers of particles—although at the expense of an increased simulation time. One of the main advantages of the particle-tracking method is the ad-hoc manner in which particle characteristics can be defined. For example, non-Fickian dispersion can be readily simulated by attaching an age to each particle (equal to the time since the release of the particle into the computation) and then making the diffusive step of the particle a function of its diffusion time.

In the new model, the interactions between the liquid and solid (suspended matter and bottom sediment) phases have been described in terms of kinetic transfer coefficients, as in actual finite difference models. A stochastic method has been developed to simulate such interactions. Deposition of particles and erosion of the sediment are also included in the model. A Rouse profile is also used to estimate the vertical structure of suspended matter concentrations. As a demonstration of the method, the particle-tracking model has been used to simulate the dispersion of radionuclides in the English Channel. A finite difference model was previously developed and validated for the Channel. Thus the output from both models has been compared. Two radionuclides with different geochemical behaviours, ^{137}Cs and ^{239,240}Pu, have been used for the comparisons. The agreement between both models is, in general, rather good. However, it is possible that, due to the particular behaviours of certain radionuclides, activity concentrations in one of the phases cannot be calculated.

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