

On the sensitivity of a marine dispersion model to parameters describing the transfers of radionuclides between the liquid and solid phases

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Abstract

The sensitivity of a marine dispersion model for non-conservative radionuclides, previously developed and validated for the English Channel, to parameters describing the exchanges between the liquid and solid phases (suspended matter and bottom sediments) has been studied using a Monte Carlo method. A probability distribution is assigned to each parameter. They are sampled to obtain a set of model parameters and a model run is carried out. This process is repeated to obtain a distribution of model outputs. Partial correlation coefficients are calculated to assess the relative influence of each parameter on model output. Errors are also assigned to model results. Three situations are studied: an instantaneous release of radionuclides, a continuous release and the case of a contaminated sediment behaving as a long-term source of radionuclides. Calculations have also been carried out for two radionuclides with different geochemical behaviour: ^{137}Cs and $^{239,240}\text{Pu}$. The results indicate that all parameters are relevant, depending on the phase we are interested in obtaining the result and on the source term (instantaneous, continuous or due to sediments). However, parameters that are, in general, more influential are kinetic rates, mixing depth in the sediment and mean radius of suspended and sediment particles. This suggests that including several particle sizes in future radionuclide dispersion models could lead to an improvement in model results. Differences have also been found with respect to the relevance of some parameters depending on the geochemical behaviour of the radionuclide.

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1. Introduction

Numerical models have been widely used to assess the effect of radioactivity releases to the environment. Model predictions may be used as the basis for important decisions on issues such as emergency response and countermeasures, waste management, and environmental remediation and restoration. The consequences of such decisions may be significant in terms of human, ecological or economic costs (Thiessen et al., 1999). Thus, it is essential to evaluate the reliability of the model predictions.

Model uncertainties have been typically investigated through simple sensitivity studies in which each of the parameters in the model was varied in turn, leaving the others fixed at their nominal values (Periáñez et al., 1996; Tappin et al., 1997; Margvelashvily et al., 1999). Indeed, sensitivity of a model to a parameter that has a high natural variability or about which little is known can increase the uncertainty associated with model predictions. Now, the Monte Carlo method has proven to be generally a more suitable approach in carrying out a quantitative analysis of sensitivity and propagation of uncertainties in radionuclide transport codes (Whicker et al., 1999). It is also possible to calculate partial correlation coefficients (r_{part}) for relationships between model outputs and values of the parameters selected for analysis. A partial correlation estimates the linear correlation between an output variable and a parameter after removing the effects of the other parameters. A rank of model parameters can be obtained according to their partial correlation coefficients, which gives an indication of their relative influence on model output (Whicker et al., 1999). The effort for obtaining site-specific values for model parameters, for a given model application, should be focused on the most influential ones. A parameter with a high r_{part} may deserve more study to improve confidence in it and perhaps to reduce its uncertainty. In contrast, research on a parameter with a small r_{part} may not be useful because of the low relationship between its value and the output.

The objective of this work consists of studying the sensitivity of a marine dispersion model for non-conservative radionuclides previously developed and validated for the English Channel (where radionuclides are released from Cap de La Hague nuclear fuel reprocessing plant). In particular, only the model sensitivity to parameters governing the exchanges of radionuclides between the liquid and solid phases (suspended matter and bottom sediments) will be studied. Model sensitivity to parameters related to tidal propagation and sediment dynamics is not included in this work. Model sensitivity will be studied in three typical situations, which correspond to the different source terms that can generally be found: (1) the case of a hypothetical instantaneous release of radionuclides from the reprocessing plant, (2) the case of a continuous release and (3) the case in which there is no external input of radionuclides but the bottom sediments are initially contaminated, so that they behave as a long-term source of previously released waste radionuclides. Errors

will be assigned to model predictions and r_{part} s will be calculated for the three situations studied. This will allow establishing a ranking of partial correlation coefficients to analyze the relative influence of each parameter. Calculations are made for two radionuclides with different geochemical behaviour: ^{137}Cs and $^{239,240}\text{Pu}$.

This type of sensitivity analysis has been carried out in box models. For instance, [Breshears et al. \(1992\)](#) present such a study for an agroecosystem model that represents processes affecting milk contamination after a fallout deposition of radionuclides. [Carroll and Harms \(1999\)](#) studied uncertainty in a model that simulates the dispersion of radionuclides in a shallow Arctic Bay. The model uses partition coefficients to describe the distribution of radionuclides between water and suspended matter particles. Probability distributions were assigned to suspended matter concentration and distribution coefficients k_{d} s (parameters needed to calculate the partition coefficient). These distributions were sampled and probability distributions were then obtained for the partition coefficient. The dispersion of radionuclides was then computed for the partition coefficient probability distribution mode and for the lowest and the highest values. Thus, in this work, probability distributions are used for the partition coefficients, although the sensitivity analysis is carried out in the simple way mentioned above. This is the first time, to the author's knowledge, that a detailed Monte Carlo-based sensitivity study is carried out on a marine dispersion model for non-conservative radionuclides.

The model and method used for the study are described in the next section. Following this, results are presented and discussed.

2. Materials and methods

2.1. The model

The model used for the study has been described and tested before ([Periáñez, 2003](#)), so few details are given here. It is a long-term dispersion model for non-conservative radionuclides. The model is based upon residual (averaged) circulation obtained from a previously validated tidal model of the Channel ([Periáñez and Reguera, 1999](#)). The averaged suspended matter concentrations over the Channel have also been obtained from a sediment model which includes advection/diffusion of particles, settling, deposition and erosion of the sediment. Details can be seen in [Periáñez \(2000\)](#). The model domain, showing the residual circulation for average wind speed and direction (southwest 6 m/s), is presented in [Fig. 1](#). The position of Cap de La Hague nuclear fuel reprocessing plant is also indicated in the map. The measured suspended matter concentrations in the Channel ([Eisma and Kalf, 1987](#)) range from some 3 ppm along the French shore to 5 ppm along the British shore, although concentrations up to 10 ppm can be found in some locations. The model results are in agreement with these values ([Periáñez, 2000](#)). Since the Channel is a highly dynamic environment in terms of tides, the sediments are essentially composed of coarse material ([Boust, 1999](#)). Mud deposits can only be found along

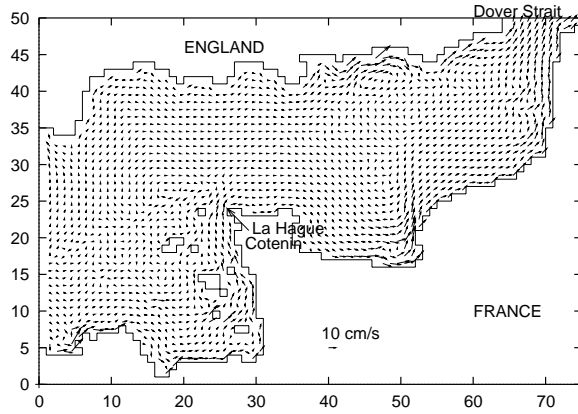


Fig. 1. Residual currents in the Channel for average wind conditions. Each unit in the x and y axes is 5000 m.

both shores in the areas of weaker tidal currents (amplitudes of the order of 0.5 m/s or less).

The model considers that radionuclides can be present in three phases: solution, suspended matter and active bottom sediments (particles with a diameter $< 62.5 \mu\text{m}$). It is considered that the exchange of radionuclides between the liquid and solid phases is governed by a single reversible reaction. 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 $[T]^{-1}$). The adsorption of radionuclides depends on the surface of particles per water volume unit, denoted as the exchange surface (Perianez, 1999, 2000, 2002a):

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

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 χ_1 is a parameter with the dimensions of a velocity denoted as the exchange velocity. Assuming spherical particles and a step function for the size distribution of suspended particles, it can be easily obtained (Perianez, 1999, 2000, 2002a) that:

$$S_m = \frac{3m}{\rho R} \quad (2)$$

$$S_s = \frac{3Lf\phi}{RH} \quad (3)$$

where m is the suspended matter concentration, ρ 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 ϕ 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 dispersion of non-conservative radionuclides in estuarine (Periáñez et al., 1996, 2002a) and marine environments (Periáñez, 1999, 2000).

Three partial differential equations are then obtained (Periáñez, 2003), whose solutions give the time evolution of specific activities in water, suspended matter and active sediment. In the case of water, C_d (Bq/m³), is:

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

where C_s and A_s are specific activity (Bq/kg) in suspended matter and the active bottom sediment, respectively. Since the model is two-dimensional, it is considered that C_d and C_s have a uniform distribution over the water column. The sediment bulk density, ρ_s , is expressed in kg/m³ 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 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 adsorption.

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

$$\frac{\partial(mC_s)}{\partial t} = (\text{adv} + \text{dif}) + k_1 C_d - k_2 C_s m + ED_m \quad (5)$$

where k_1 is now given by the first term of Eq. (1). Symbol ED_m is the erosion–deposition term. Finally, the equation for the temporal evolution of specific activity in the active bottom sediment is:

$$\frac{\partial A_s}{\partial t} = k_1 \frac{C_d H}{L \rho_s f} - k_2 A_s \phi + ED_s \quad (6)$$

where k_1 is given by the second term of Eq. (1). The form of the erosion–deposition terms for suspended matter and bottom sediments, ED_m and ED_s , can be seen in Periáñez (2003).

For the case study (3), redissolution from a contaminated sediment, the kinetic model is replaced by a two-step model consisting of two consecutive reversible reactions. The first, governed by the same coefficients k_1 and k_2 , is a reversible adsorption or isotopic exchange with non-specific sites on particle surfaces. The second and slower reaction, governed by rates k_3 and k_4 (forward and backward, respectively), is a reversible transfer of radionuclides to more specific sites in the particles. The second process may be a slow diffusion of ions into pore and inter-lattice spacings, inner complex formation or a transformation, such an oxidation. Thus, in the two-step model, the solid phase is divided into two phases, which are denoted as non-specific and specific. This has been done since very recent experiments (see for instance Ciffroy et al., 2001) have shown that a two-step model is

more adequate than a one-step model to simulate both the uptake and release kinetics. On the other hand, such a two-step model has been implemented in a plutonium dispersion model of the Irish Sea (Periáñez, 2002b). Results indicate that there are no appreciable differences between both models when simulating the dispersion of radionuclides introduced from an external source of radionuclides over relatively short time scales (several months), but a two-step model should be used if the source is due to redissolution from contaminated sediments since in this case a one-step model produces a too fast decrease in sediment activity if compared with experimental evidences (see Periáñez, 2002b). Thus, for cases (1) and (2), a one-step model is used since it is computationally cheaper, and the two-step model is used for case (3).

To solve the equations, a spatial and temporal discretization 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 (see Fig. 1). 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 equations are solved explicitly using second order accuracy finite difference schemes.

2.2. Method for the sensitivity analysis

The sensitivity analysis has been limited to parameters governing the exchanges of radionuclides between the liquid and solid phases: χ_1 , k_2 , ρ , ρ_s , R , L , ϕ and f , as well as k_3 and k_4 in the case of redissolution. Other parameters concerning tidal propagation or sediment dynamics have not been included in this study since we are mainly interested in the description of the transfers between phases as commented above.

A Gauss probability distribution is assigned to each parameter, which is defined by its average value and standard deviation. A random sampling of each distribution is carried out using a Monte Carlo method, so that a set of model parameters is obtained, and the model is run. This process is repeated 200 times, so that a distribution of model results is obtained. The set of model results allows assigning uncertainties to model output and calculating partial correlation coefficients.

Partial correlation coefficients are calculated in the following way: if a set of variables (x_1, x_2, \dots, x_p) is given, the r_{part} between variables 1 and 2, for instance, is calculated obtaining the correlation coefficient in the regression:

$$e_{1,3,4,\dots,p} = A \cdot e_{2,3,4,\dots,p} + B \quad (7)$$

where $e_{1,3,4,\dots,p}$ and $e_{2,3,4,\dots,p}$ are the residues of the multiple regression of variables 1 and 2, respectively, with respect to the rest of variables $3, \dots, p$. The correlation coefficient for regression Eq. (7) is the partial correlation coefficient between variables 1 and 2.

In the three cases analyzed, the model can produce an extensive amount of information: distribution maps of radionuclide concentrations for each phase and at any desired time, and/or temporal evolution of concentration in each phase and for each desired point of the model domain. Considering that 200 simulations are

carried out for each case, the volume of data obtained would be rather difficult to handle. Thus, to simplify the model output, we have fixed our attention in a given *target grid box* and at a defined instant of time after the beginning of the simulation ($t = 0$). This target means a sensitive point in the domain, where a local population (human or not) could receive an external dose due to contamination of water and/or sediments or an internal dose due to ingestion of contaminated material, for instance. The instant of time at which results are obtained is the time required for the peak activity to travel from La Hague to the target grid box. Thus, the model would provide information on maximum activity levels reached in that point (in water, suspended matter and bottom sediments) so as to determine, for instance, if any kind of remedial action should be taken. For simulations carried out with ^{137}Cs , we have defined grid box (50,20) as the target and the results are obtained 40 days after the beginning of the release from La Hague or the redissolution from the sediment. In the case of $^{239,240}\text{Pu}$, target grid box is (30,26). This grid box is closer to La Hague since due to the high reactivity of plutonium, this radionuclide moves slower than Cs.

3. Results and discussion

The values given to the parameters are presented in Table 1 for the ^{137}Cs application. Each value, which is the nominal value previously used in this model (Periáñez, 2003), is taken as the average value for each Gauss distribution. The errors given in Table 1 (arbitrarily taken as 20% of the nominal values) correspond to the standard deviation of each probability distribution. The probability distribution of each parameter is sampled using a Monte Carlo method over a 3σ distance around the average value. Details on the selection of the nominal values for the parameters can be seen in Periáñez (2003) and will not be repeated here.

Initial conditions for the simulations are the following. In cases 1 and 2, the model is started from zero concentrations in water, suspended matter and bottom sediments over all the domain. In the case of an instantaneous release, an input of

Table 1
Nominal value and error assigned to each parameter in the model for the application to ^{137}Cs

Symbol	Description	Value
ρ	Particle density	$2600 \pm 520 \text{ kg/m}^3$
ρ_s	Sediment bulk density	$900 \pm 180 \text{ kg/m}^3$
χ_1	Exchange velocity	$(2.1 \pm 0.4) \times 10^{-8} \text{ m/s}$
k_2	Kinetic coefficient	$(1.16 \pm 0.23) \times 10^{-5} \text{ s}^{-1}$
L	Sediment mixing depth	$0.10 \pm 0.02 \text{ m}$
ϕ	Correction factor	0.10 ± 0.02
R	Particle mean radius	$15 \pm 3 \text{ }\mu\text{m}$
f	Active sediment fraction	0.10 ± 0.02
k_3	Kinetic coefficient	$(1.16 \pm 0.23) \times 10^{-7} \text{ s}^{-1}$
k_4	Kinetic coefficient	$(1.16 \pm 0.23) \times 10^{-8} \text{ s}^{-1}$

5.0×10^{12} Bq of dissolved ^{137}Cs is assumed at $t = 0$ and at grid box (26,24), where wastes from La Hague are released. In the case of a continuous release, 5.0×10^{10} Bq are introduced each time step. In study 3, (redissolution) an initial specific activity of 1.0×10^5 Bq/kg was considered in the active fraction of the bottom sediment in grid box (26,24) (in the specific-site sediment phase), which means an inventory of 2.25×10^{13} Bq in the entire compartment. Zero concentrations are assumed for water, suspended matter and sediment (unless in the mentioned point) over all the domain.

As an example, the distribution of dissolved ^{137}Cs over the model domain 40 days after the instantaneous release from La Hague is presented in Fig. 2. The position of the target grid box is also shown in the map. It can be seen that the patch of radionuclides essentially moves along the French shore and that the target grid box is effectively in the area of the activity peak at this instant of time. In the case of a continuous release, a banded structure showing higher concentrations along the French shore is now extending from La Hague to Dover (see Periañez, 2003). In the case of redissolution, concentrations at La Hague in water and suspended matter initially increase due to the input from the sediment and after decrease due to advective/diffusive processes. A patch extending along the French shore is then observed. The sediments are contaminated as the patch travels above them.

The frequency histograms of results have been obtained for C_d , C_s and A_s in the ^{137}Cs instantaneous release experiment as an example. Such histograms have been fitted to Gauss distributions. The results are presented in Table 2, where the average value and width of the distributions are given together with the correlation coefficient of the numerical fitting. It can be seen that, in general, the fitting of the histograms to the distributions is good: Gauss distributions are obtained for the model results from the probability distributions of parameters. Uncertainty can be assigned to model results now. Thus, it can be concluded that for the target grid

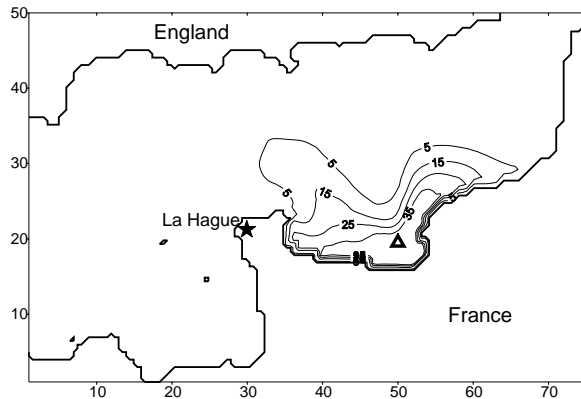


Fig. 2. Distribution of dissolved ^{137}Cs (Bq/m^3) 40 days after an instantaneous release at La Hague. The position of the *target grid box* is indicated by the triangle.

Table 2

Average value (\bar{x}), width (w) and correlation coefficient for the numerical fitting of model results to Gauss distributions for each phase in the ^{137}Cs instantaneous release experiment. The width of the distribution is $w = 2\sigma$

Phase	\bar{x}	w	r^2
C_d (Bq/m ³)	35.39	3.04	0.949
C_s (Bq/kg)	5.28	4.60	0.829
A_s (Bq/kg)	11.12	7.97	0.680

box 40 days after the discharge, ^{137}Cs concentrations are 35.4 ± 1.5 Bq/m³, 5.3 ± 2.3 Bq/kg and 11 ± 4 Bq/kg for water, suspended matter and active bottom sediment, respectively (errors are 1σ). The model results are more precise in the case of the dissolved phase (relative error 4%), while uncertainties are larger for the suspended matter and bottom sediments (relative errors 43% and 36%, respectively). This method for estimating model uncertainties could be applied to any other modelling application.

Absolute values of the partial correlation coefficients for the three cases studied and for each parameter are presented in Fig. 3 in the case of ^{137}Cs . The most influential parameters are arbitrarily defined as those having r_{part} above 0.5 (Breshears et al., 1992). Moderately influential parameters are defined as those having r_{part} between 0.3 and 0.5.

It can be seen that all parameters are relevant in the description of the transfers between the liquid and solid phases, depending on the phase in which we are interested and on the source term. For instance, if we are interested in obtaining specific activity in suspended matter particles (due to any reason), it is clear that particle density, ρ , must be precisely specified for the three situations investigated ($r_{\text{part}} > 0.77$). This parameter, however, is not relevant if our interest is focused in water or bottom sediments ($r_{\text{part}} < 0.16$ in all cases).

Sediment bulk density, ρ_s , is relevant for A_s in the three situations ($r_{\text{part}} > 0.79$). It is also relevant for the dissolved phase in the case of redissolution ($r_{\text{part}} = 0.84$), but is only moderately relevant ($r_{\text{part}} = 0.47$) for suspended matter in this case. It may be due to the fact that transfer to suspended matter from the sediment is indirect, the dissolved phase being an intermediate step. In cases of instantaneous or continuous releases, ρ_s is not relevant either for the suspended matter or for the dissolved phase ($r_{\text{part}} < 0.08$).

The exchange velocity χ_1 and the kinetic coefficient k_2 are always relevant parameters, for all situations and phases ($r_{\text{part}} > 0.59$). The only exception is χ_1 in the dissolved phase for the redissolution experiment, where the influence is moderate ($r_{\text{part}} = 0.42$). It seems that the activity content in water is essentially more dominated by the transfer from the sediment to water (desorption is dominating) at La Hague than by the subsequent transfer to suspended matter and bottom sediments that is produced as the patch of redissolved radionuclides travels along the Channel.

The mixing depth in the sediment, L , is relevant for the dissolved phase in the three cases ($r_{\text{part}} > 0.75$) since the kinetic coefficient k_1 depends on L (see Eq. (3))

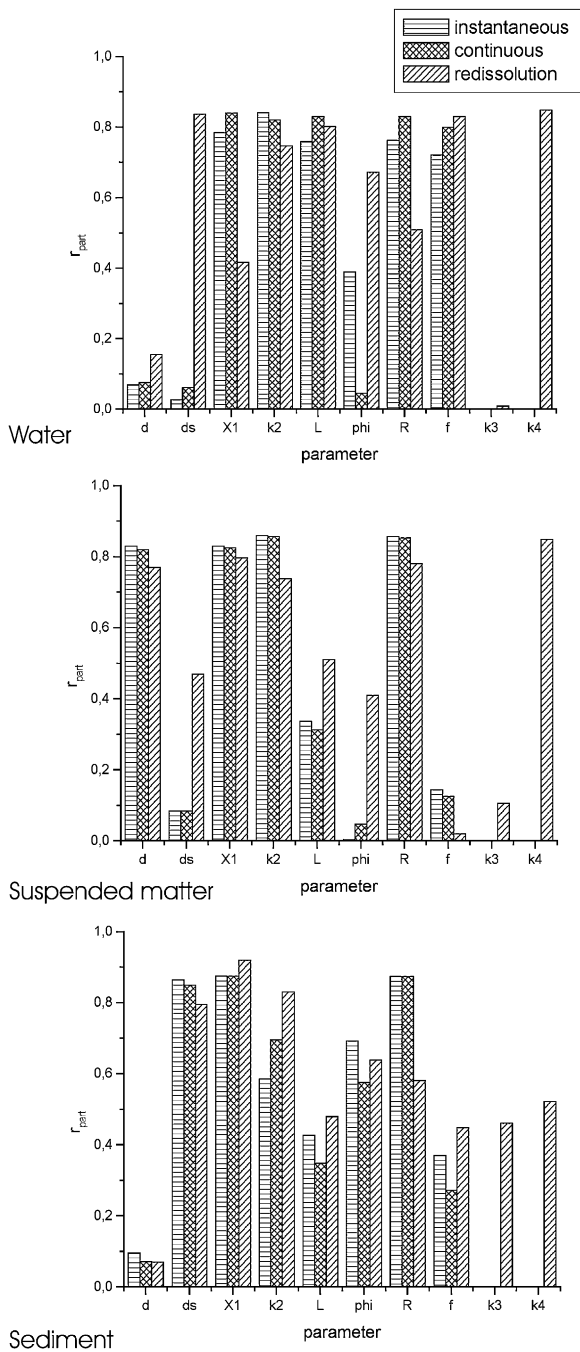


Fig. 3. Values of the partial correlation coefficient for the three phases and in the three situations studied ($d = \rho$, $ds = \rho_s$) in the case of ^{137}Cs .

and the transfer from the sediment to water that takes place at La Hague, which is the dominant process in the redissolution experiment as seen before, also depends on L (see last term in Eq. (4)). It is moderately influential for the rest of phases and all the experiments ($0.31 < r_{\text{part}} < 0.48$). In the case of suspended matter for the redissolution experiment, it is slightly more relevant ($r_{\text{part}} = 0.51$).

The correction factor ϕ is relevant for the bottom sediment in the three situations analyzed ($r_{\text{part}} > 0.57$). In the case of redissolution, it is relevant for the dissolved phase ($r_{\text{part}} = 0.67$), as also was L , and moderately influential for suspended matter. Again, this is because the transfer from sediment to suspended matter is indirect through the dissolved phase. Parameter ϕ does not seem to be relevant for water and suspended matter in the cases of instantaneous and continuous releases ($r_{\text{part}} < 0.05$) with the exception of the dissolved phase in an instantaneous release situation ($r_{\text{part}} = 0.39$).

The medium radius of suspended particles and bottom sediments, R , is relevant for all the phases and experiments ($r_{\text{part}} > 0.51$). Thus, it may be interesting, for future radionuclide dispersion models, to include several grain sizes of suspended particles and sediments since the results are rather sensitive to this parameter. However, it must be taken into account that an increase in model complexity must always be accompanied with appropriate field data to provide values for new parameters, that appear when adding features to a model, and to carry out model validation and testing.

The relevance of the active sediment fraction, f , is similar to that of L : it is relevant for the dissolved phase in all cases ($r_{\text{part}} > 0.72$), not relevant for the suspended matter and moderately influential for the sediment.

As commented above, a two-step kinetic model was used in the redissolution experiments. Thus, r_{part} s have also been calculated for k_3 and k_4 (rates for the second reaction) in the redissolution experiment. They are presented in Fig. 3 as well. It can be seen that k_4 is relevant for the three phases ($r_{\text{part}} > 0.52$), since all activity is considered to be initially in the specific sites of the sediment and k_4 governs the transfers from the specific sites to the non-specific sites, where radionuclides can be redissolved. k_3 is not relevant for water and suspended matter ($r_{\text{part}} < 0.11$). However, it is moderately influential for the sediment phase.

Average values of the absolute values of the partial correlation coefficients, for all phases and experiments, are presented in Table 3, together with their standard deviations. The average value gives an indication of the *overall influence* of each parameter, while its dispersion indicates the variability of the parameter relevance between experiments (source terms) and phases. It seems clear that, in general, the most influential parameters are χ_1 , k_2 and R , followed by k_4 and L . The large dispersion of the average value in the remaining parameters indicates that their relevance is limited to certain phases and source terms.

The same calculations have been repeated for $^{239,240}\text{Pu}$, a highly reactive radionuclide, to see if there are differences in model sensitivity depending on the radionuclide affinity to the solid phases. All parameters have the same values as in the application to ^{137}Cs (Table 1) except χ_1 and k_2 , that govern adsorption/release

Table 3

Average value of the partial correlation coefficients for all phases and experiments

Parameter	^{137}Cs	$^{239,240}\text{Pu}$
ρ	0.3 ± 0.4	0.3 ± 0.3
ρ_s	0.5 ± 0.4	0.23 ± 0.24
χ_1	0.80 ± 0.15	0.56 ± 0.08
k_2	0.78 ± 0.09	0.31 ± 0.25
L	0.53 ± 0.21	0.70 ± 0.11
ϕ	0.4 ± 0.3	0.29 ± 0.15
R	0.77 ± 0.13	0.51 ± 0.15
f	0.4 ± 0.3	0.62 ± 0.24
k_3	0.19 ± 0.24	0.18 ± 0.17
k_4	0.65 ± 0.17	0.65 ± 0.16

reactions. The following values are now used (Periáñez, 2003): $\chi_1 = (1.5 \pm 0.3) \times 10^{-5}$ m/s and $k_2 = (9.3 \pm 1.9) \times 10^{-6}$ s $^{-1}$. Again, uncertainties in these parameters have been fixed as 20% of their nominal values.

Absolute values of the partial correlation coefficients for the three phases and in the three experiments are shown in Fig. 4. It can be seen that, in general, χ_1 is less influential than in the case of ^{137}Cs (has smaller r_{part} values). k_2 is only clearly relevant in the case of the redissolution experiment and for water and sediments. This is related with the results of Carroll and Harms (1999) and Sholkovitz and Mann (1984), who found that uncertainties (thus sensitivities) decrease as the velocity of uptake increases. There are particular differences in parameter relevance with respect to ^{137}Cs (for instance $r_{\text{part}} > 0.5$ for suspended matter particle density in the sediment for the redissolution experiment or the behaviour of ϕ in the three phases). Also, in general, L and f are more relevant than in the case of ^{137}Cs . Thus, the parameters describing sediments (sediment composition f and mixing depth L) should be specified more carefully in the case of very reactive radionuclides. This is confirmed in Table 3, where the *overall influence* of each parameter is shown. χ_1 is still relevant for all phases and types of source term (although less than in the case of ^{137}Cs). This is not the case with k_2 , which is only moderately influential. The most influential parameters are now L , k_4 and f , followed by χ_1 and R . The remaining parameters show similar behaviour as in the case of ^{137}Cs .

Together with uncertainties in parameters describing the exchanges of radionuclides between the liquid and solid phases, there are other sources of error in a model. Although the use of residual circulation to compute advection can give results very similar to those obtained by explicitly calculating tidal currents and advection, if the residual circulation field is appropriately defined (Delhez, 1996), the use of an averaged wind speed and direction can be a source of uncertainty. Indeed, Carroll and Harms (1999) have found that changing wind patterns affects the dispersion of radionuclides with low k_d (Cs and Sr). However, they have little influence on particle-reactive radionuclides. Solving the advection–diffusion dispersion equation by finite differences also introduces errors in the model output since a

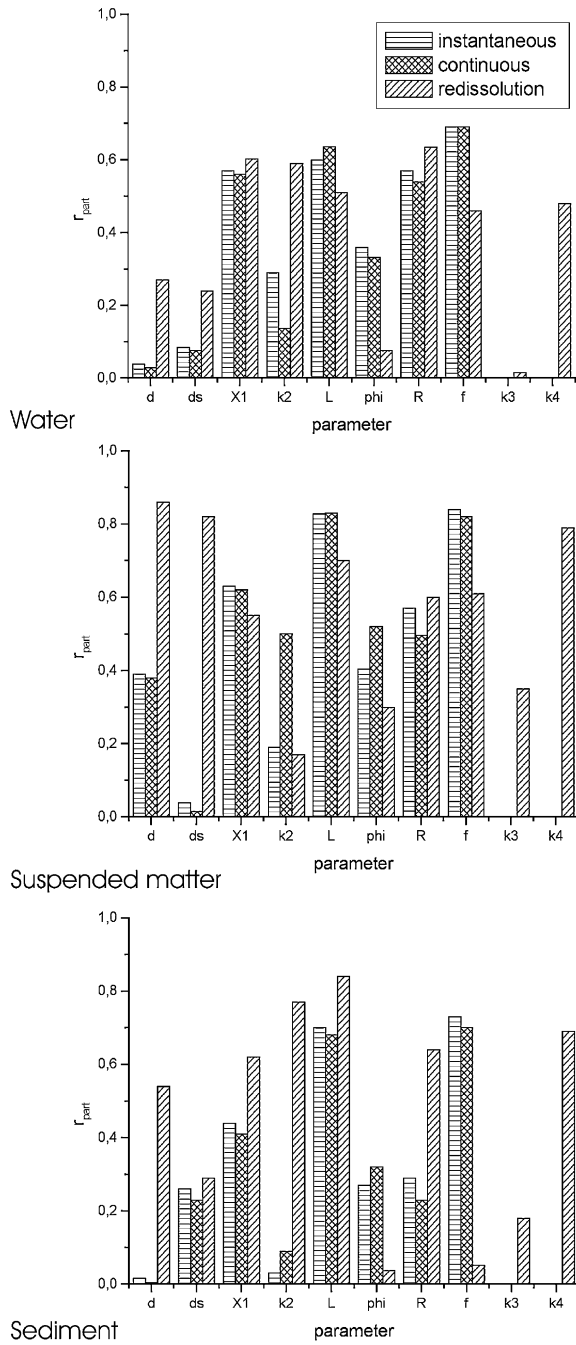


Fig. 4. Same as Fig. 3 but in the case of $^{239,240}\text{Pu}$.

finite difference scheme is always an approximation to the full differential equation. Open boundary conditions are often required (in our case along Dover Strait and the western boundary of the Channel). These conditions are mathematical artifacts that may introduce some errors. However, in real ocean dispersion problems, errors introduced by the finite difference scheme (if at least second order accuracy schemes are used) and boundary conditions are masked by turbulence. Turbulent mixing is rather complex [see for instance [Holt and Proctor \(2001\)](#) for a brief review], and is parameterized through a diffusion coefficient which is often fixed as a function of the model grid size. Indeed, ocean turbulence and diffusion constitute an active research field. The investigation of turbulence parameterization influence on model output is beyond the scope of this paper, although it constitutes a problem that should probably be addressed in the near future.

4. Conclusions

The sensitivity of a marine dispersion model for non-conservative radionuclides to parameters governing the transfers of radionuclides between the liquid and solid phases (suspended matter and bottom sediments) has been studied using a Monte Carlo method based upon random sampling of the probability distribution of each parameter and the calculation of partial correlation coefficients. A method for assigning errors to model results is also provided.

It has been found that all parameters in the description of the transfers are relevant, at least for certain phases and experiments (source terms). Thus, a parameter that is not relevant for the calculation of specific activity in water may be relevant if, for instance, interest lies on suspended matter. The most *overall* influential parameters in the case of ^{137}Cs seem to be χ_1 and k_2 , rates governing the adsorption/release kinetics, and R , the average radius of suspended matter and bottom sediment particles. In the case of $^{239,240}\text{Pu}$, the most relevant parameters are those describing sediments (L and f), followed by χ_1 and R . Efforts for obtaining site-specific parameters for a given model application should be focused on these parameters since they affect model results. Nevertheless, the particular situation that is going to be modelled, the geochemical behaviour of the radionuclide and the phases in which the interest lies, should be considered since the relevance of a given parameter may show a significant variability depending on the phase, radionuclide character and the source term.

Although the results of a model sensitivity study are specific for the particular model formulation, some general conclusions may be extracted. The geochemical behaviour of radionuclides affects model sensitivity to some parameters: it is necessary to specify more carefully parameters describing sediments (L and f) when the radionuclide presents a strong non-conservative character. In contrast, model output is less sensitive to kinetic rates than in the case of a more conservative radionuclide. Also, it seems that considering several particle sizes in future dispersion models could lead to an improvement in results.

Acknowledgements

This work was supported by ENRESA and the EU 5th Framework Programme (1998–2002) Nuclear Fission and Radiation Protection Contract FIGE-CT-2000-00085.

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