Marine radionuclide transport modelling: Recent developments, problems and challenges

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A B S T R A C T

A brief overview of the current status on the subject of numerical modelling of radionuclide transport in the marine environment is given: main transport processes occurring in the sea, basic approaches to solve such processes numerically, up-to-date trends to deal with water/sediment interactions (in the case of non-conservative radionuclides), and topics in which work is currently in progress (like the integration of biological uptake models within marine transport models). A brief review of models applied to simulate Fukushima Daiichi nuclear power plant releases in the Pacific Ocean after the 2011 accident is also included, since the most recent modelling efforts have been focused in this problem. A discussion of the main sources of uncertainty in models is given, as well as the problems these uncertainties pose in relation to emergency modelling, which is one of the most relevant applications of dispersion models.

Keywords:
Hydrodynamics
Dispersion
Marine environment
Numerical model
Sediment

1. Introduction

Assessments of radioactivity doses to humans and non-human biota rely on measurement data and model outputs. Models should describe the main radionuclide transport processes within the environment and modelling tools depend on the assessment character, which may be "predictive", when expected doses are estimated for future release scenarios, or "retrospective", when doses are estimated for past time sources (Hunt, 2004). The modelling approach can also differ for cases of regular or accidental releases.

In the case of the marine environment, radionuclide pathways depend on sources (wet and dry atmospheric deposition, direct releases due to regular or accidental discharges from nuclear installations, rivers, etc.), dispersion by currents and uptake by sediments and biota. The main pathways for human exposure are given in Fig. 1, although only processes within the dashed square are considered in this review. Modelling radionuclide transport in the sea is an interdisciplinary science which requires basic knowledge of different topics, namely ocean dynamics, numerical methods, sedimentology and bio-geochemistry. Consequently, it is useful to have a review of the basic required principles.

Numerical models which simulate the transport of radionuclides in the marine environment have been continuously improved since the pioneering work of Prandle (1984), who simulated the transport of dissolved ^{137}Cs in the European Shelf Seas. This radionuclide is released from Sellafield (UK) nuclear fuel reprocessing plant into the Irish Sea. Later, models were developed to simulate the fate of releases from other

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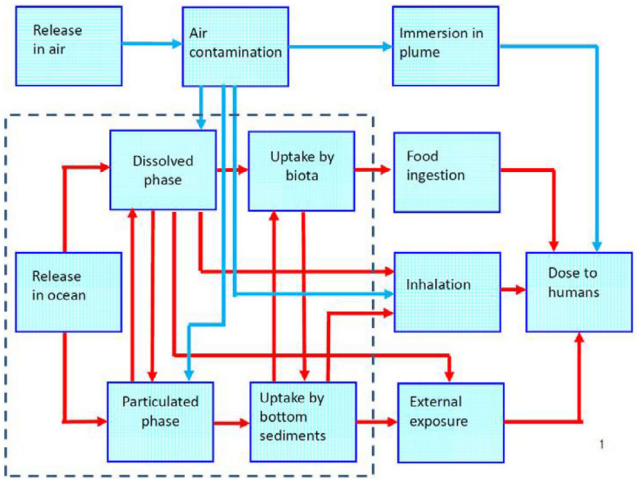

Fig. 1. Pathways for human exposure from releases of radioactivity into the ocean environment (based on Hunt, 2004). The dotted rectangle identifies processes under consideration in this review.

nuclear facilities, such as La Hague nuclear fuel reprocessing plant in the English Channel (Breton and Salomon, 1995) or from nuclear waste dumped in the sea (Harms, 1997; Cetina et al., 2000). Marine transport models attracted more attention after the accident in Fukushima Daiichi Nuclear Power Plant (FDNPP) resulting from the March 2011 earthquake and tsunami (e.g. Kawamura et al., 2011; Behrens et al., 2012; Tsumune et al., 2012; Dvorzhak et al., 2012; Masumoto et al., 2012; SCJ, 2014, etc.). However, it should be commented that last reviews of marine radionuclide transport models were published more than a decade ago (Harms et al., 2003; Periáñez, 2005a).

The International Atomic Energy Agency (IAEA) has organized activities on marine radioactivity transport model testing since the VAMP (Validation of Model Predictions) program in 1988 (see IAEA, 2000, for the aquatic group work). The most recent programmes are the MODARIA[2] (Modelling and Data for Radiological Impact Assessments) project, launched in 2012, and MODARIA-II,[3] launched in 2016. The marine working groups in these programs were motivated by the recent developments in marine science and marine modelling, as well as the radioactive pollution due to the FDNPP disaster. In general, IAEA model intercomparisons are also motivated by the need to link modelling and data with radiological impact assessments, to enhance the capabilities of member states to simulate radionuclide transport and also to understand their effects in the environment. State-of-the-art models were applied by different teams to several radionuclide transport problems and results were compared , with special emphasis on the sources of discrepancies between models. In addition to what was said above, it may be useful to have a summary of the basic principles of such models, which include all processes within the dashed square in Fig. 1 and are the most advanced radionuclide transport models at the present time.

The purpose of this paper, in view of our previous comments, consists of addressing three main points. These points correspond to the main sections of the paper:

1. To provide an updated review of marine radionuclide transport modelling techniques, including a brief description of model types (box models, Eulerian and Lagrangian models). Different model structures, techniques presently used to obtain water circulation and to describe other processes like water/sediment interactions and biological uptake are described in Section 3. First, the main radionuclide transport processes in the sea are discussed in Section 2.
2. To briefly review modelling works which were carried out to simulate FDNPP releases in the Pacific Ocean since, as mentioned above, the most recent modelling efforts were devoted to this task. This review in presented in Section 4.
3. To discuss the main difficulties and problems in marine radionuclide transport models found during the MODARIA and MODARIA-II programs, with special emphasis on sources of model uncertainties. Challenges in developing models for emergency situations (as FDNPP accident for instance) are also discussed. All this is presented in Section 5.

2. Radionuclide transport processes in the marine environment

Radionuclides are considered to be conservative or non-conservative according to their geochemical behaviour. Some radionuclides remain dissolved in the water column since adsorption by the solid phases (suspended and bed sediment particles) is negligible. These are the so-called conservative radionuclides. Radionuclides which are significantly adsorbed by sediment particles (suspended in the water column and on the seabed) are denoted as non-conservative.

In general, the sea is a stratified environment where the pycnocline usually acts as a barrier for vertical mixing. Radionuclides may be deposited on the sea surface due to atmospheric fallout (dry and/or wet deposition) or directly introduced into the sea from a point source due to releases from a given industrial facility (chronic or accidental), an accident in a nuclear vessel etc. Other sources of radionuclides in the sea are river runoff, distributed influx due to the coastal wash-off and undersea groundwater discharges (Sanial et al., 2017). Once in the water column, radionuclides are transported by the currents (advection) and also mixed by turbulent diffusion. Diffusion produces a transport from high to low concentration regions, which makes peak concentrations decrease as the radionuclide patch size increases. Radionuclides may be fixed to suspended matter particles present in the water column, which sink by gravity (settling) if their densities are higher than water density and reach the deep layer. Advection and diffusion processes also occur in the deep layer. Suspended particles transporting radionuclides may be deposited on the seabed. Also, direct adsorption/desorption reactions between the seabed and water may occur. Radionuclides introduced into the seabed sediment are eventually buried by the further deposition of suspended particles or resuspended by near bed turbulence. Eventually, there will be a transfer of radionuclides to biota. Other processes, as sea spray transfer from sea to land and evaporation of gaseous radionuclides may occur as well (Vives i Batlle et al., 2018).

Plutonium and thorium isotopes are examples of non-conservative radionuclides. Radionuclides which are considered to be conservative are, for instance, ^{90}Sr, ^{125}Sb, ^{99}Tc and ^{129}I. It is quite usual to find modelling studies in which ^{137}Cs is treated as a conservative radionuclide (Prandle, 1984; Estournel et al., 2012; Behrens et al., 2012; Tsumune et al., 2012, among many others). Actually ^{137}Cs is measured in seabed sediments, thus this is just an approximation in which adsorption–desorption processes are neglected. These processes were considered for ^{137}Cs in Abril and García-León (1993), Margvelashvily et al. (1997), Aldridge (1998), Aldridge et al. (2003) and Goshawk et al. (2003) among others.

All these processes, which are represented in Fig. 2, can be described by means of appropriate differential equations. A computer code can then be written to numerically solve the equations, which constitutes the transport model. The following ingredients are required to simulate the transport of non-conservative radionuclides in the marine environment:

[2] http://www-ns.iaea.org/projects/modaria/default.asp?l=116.
[3] http://www-ns.iaea.org/projects/modaria/modaria2.asp?s=8&l=129.

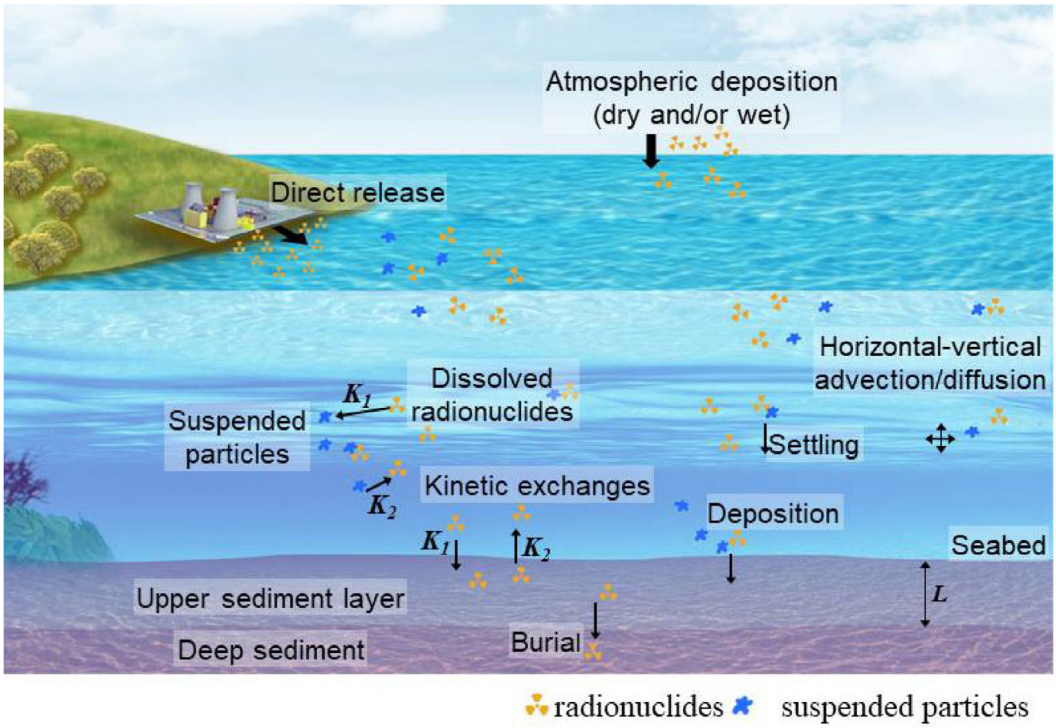

Fig. 2. Processes affecting the dispersion of non conservative radionuclides in a marine system. Kinetic rates k_1 and k_2 describe uptake and release of radionuclide by solid particles. L is thickness of the upper sediment contaminated layer.

1. Hydrodynamic model: it provides the water currents, which determine advective transport. Also, water currents and density stratification may be used to derive eddy diffusivities, which are used to evaluate turbulent mixing (see Section 3.3 for more details).
2. Sediment transport model: it provides suspended matter concentrations and erosion and deposition fluxes over the model domain. Details on the mathematical formulation of the physical processes are described for instance in Eisma (1993), van Rijn (1993), Winterwerp and van Kester (2004) and Lick (2008), but are not included in the present review.
3. Radionuclide transport model: it includes the description of advection/diffusion processes and the description of adsorption/desorption reactions between the dissolved and solid phases (Periáñez, 2005a). Other relevant processes could be included as well, as for instance migration of radionuclides in the bottom sediments due to molecular diffusion and reworking of sediments by animals -bioturbation- (Maderich et al., 2017). Also redox reactions may be included in the case of plutonium (Periáñez, 2003b), which presents a complex speciation and oxidized and reduced species coexist in solution.

The different ways in which equations describing these transport processes are written and numerically solved lead to different models. These are briefly described in the following section.

3. Radionuclide transport models

3.1. Box, Eulerian and Lagrangian models

Three types of transport models are used: Eulerian, Lagrangian and box-models. They are very briefly described below.

3.1.1. Box models

In these models the marine area under consideration is divided into a number of large boxes or compartments. These boxes are interconnected according to water circulation and it is assumed that the transport of dissolved radionuclides between boxes is proportional to the difference in radionuclide concentration between them. It is assumed that mixing of radionuclides within each box is uniform and instantaneous. Other processes such as transfers of dissolved radionuclides to suspended matter, deposition of suspended matter and adsorption or radionuclides in bed sediments can be described. This is usually done using an equilibrium distribution coefficient k_d (see Eq. (15) below for its definition). Box models are well suited for assessments of radionuclide dispersion involving large spatial and temporal scales (thousand kilometres; years to decades). Some recent examples of box models are POSEIDON-R (Lepicard et al., 2004; Maderich et al., 2014a,b; Bezhenar et al., 2016), models developed in NRPA (Iosjpe et al., 2002, 2009) and others (Sánchez-Cabeza et al., 2002; Håkanson, 2005; Smith and Simmonds, 2009; for instance).

The basic equation for box models provides the temporal evolution of radionuclide activity in the water column in box i, A_i (Bq). The system of differential equations is (Nielsen, 1995):

$$\frac{\partial A_i}{\partial t} = \sum_{j=1}^{n} q_{ji} A_j - \sum_{j=1}^{n} q_{ij} A_i - q_i A_i + S_i \quad (1)$$

where n is the number of boxes in the model, q_{ij} (expressed in s^{-1}) is the transfer rate from box i to box j, q_i (s^{-1}) is the rate of radionuclide loss from box i without transfer to another box (due to radioactive decay, sedimentation, etc.) and S_i is the external source of radionuclides to box i (i.e., Bq per unit time which are released into the sea at that box due to atmospheric fallout and/or point sources). Transfer rates q_{ij} are deduced from known oceanographic features of the region under study and they parameterize the advection and diffusion processes described in the previous section. They can also be obtained from the currents calculated by hydrodynamic models.

It is assumed that, at any time, activity in the water column is partitioned between the dissolved phase and suspended matter particles. This partition depends on the suspended matter concentration in box i, SM_i, and the radionuclide k_d (equilibrium is assumed). If sedimentation rate of suspended particles is SR_i, then the activity transfer rate q_i from water to bed sediment due to particle sedimentation is

$$q_i = \frac{k_d SR_i}{d_i(1 + k_d SM_i)}, \quad (2)$$

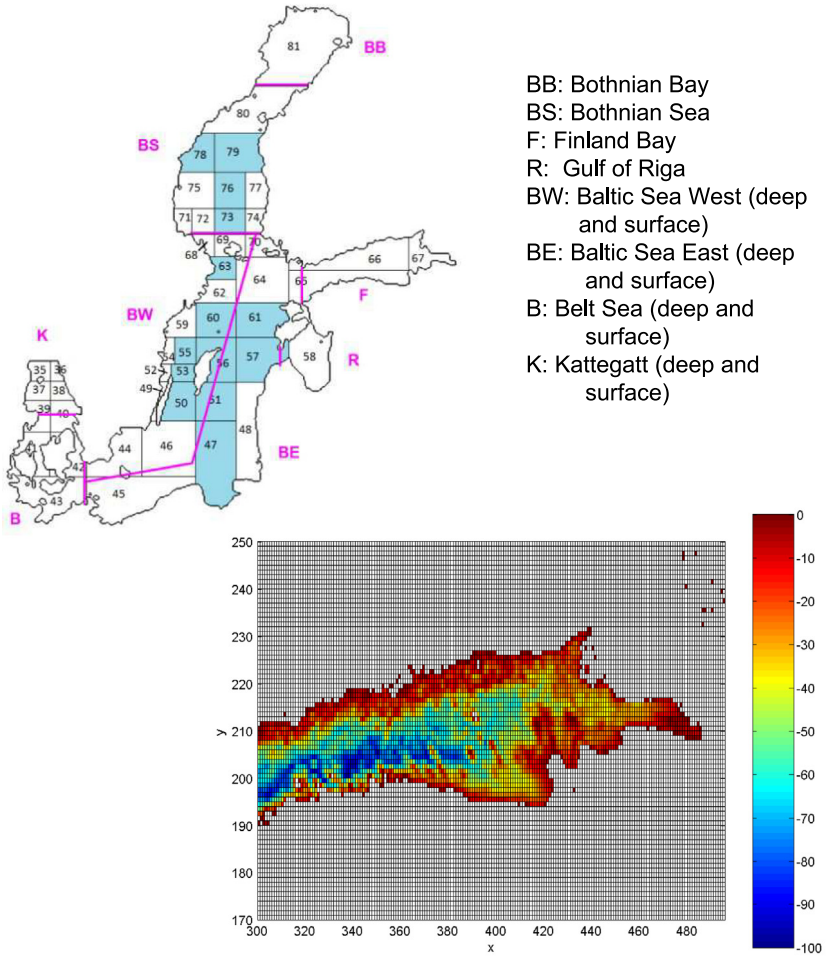

Fig. 3. POSEIDON (Lepicard et al., 2004) and NRPA (Iosjpe et al., 2002) model box structures for the Baltic Sea. Pink lines define NRPA model boxes and numbered boxes correspond to POSEIDON. Blue boxes are those divided into two water layers (surface and deep). A grid suitable for an Eulerian model covering the Gulf of Finland is also shown. It was used in the model described in Periáñez et al. (2015b). The colorbar indicates water depths in m, which are specified for each grid cell. A similar grid is required in Lagrangian models to derive radionuclide concentrations from the number of particles per volume.

where d_i is mean water depth in box i. If radioactive decay is also considered, then λ would be added to the q_i above, which is the radioactive decay constant of the considered radionuclide. Similarly, activity in the bottom deposit is partitioned between pore water and sediment particles using the distribution coefficient. The equation for the temporal evolution of combined aqueous and adsorbed activity in the bottom deposit is derived taking into account molecular diffusion, bioturbation and burial processes.

As an example, two different box structures adopted for simulating the transport of radionuclides in the Baltic Sea (Periáñez et al., 2015b) are presented in Fig. 3.

Although in box models water/sediment interactions are described by equilibrium distribution coefficients, as commented above, the present trend is to apply kinetic rates. Thus, water/sediment interactions are described dynamically. This dynamic description is presented in Section 3.4.

3.1.2. EulerIan models

In Eulerian models the differential equations giving temporal and spatial evolution of the radionuclide concentrations in different states (e.g. dissolved in water column and pore water in sediments, fixed on the suspended and bottom sediment etc.) are solved. The general compact form of these equations for concentration of radioactivity C_α in state α per unit of volume (Bq m^{-3}) or per unit of mass (Bq kg^{-1}) are written in Cartesian coordinates as:

$$\frac{\partial C_\alpha}{\partial t} + \frac{\partial (u_\alpha C_\alpha)}{\partial x} + \frac{\partial (v_\alpha C_\alpha)}{\partial y} + \frac{\partial (w_\alpha C_\alpha)}{\partial z} = \frac{\partial}{\partial x}\left(K_h \frac{\partial C_\alpha}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_h \frac{\partial C_\alpha}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_v \frac{\partial C_\alpha}{\partial z}\right) + \sum_{\beta=1}^{n} k_{\beta\alpha} C_\beta + S_\alpha - \lambda C_\alpha \quad (3)$$

where (x, y, z) are Cartesian coordinates, u_α, v_α and w_α are components of flow field for the radionuclide in the state α. In general, velocity can differ for different states (e.g. due the presence of settling velocity for suspended sediment or it may be zero in the bottom deposit). K_h and K_v are turbulent or molecular diffusivities in the horizontal and vertical directions respectively, and/or biodiffusivity in the bottom deposit, which are variable in time and space. The term $\sum_{\beta=1}^{n} k_{\beta\alpha} C_\beta$ describes first order reactions between the radionuclides in different states, where $k_{\beta\alpha}$ are kinetic transfer coefficients (they are defined in Section 3.4) and $\sum_{\beta=1}^{n} k_{\beta\alpha} = 0$ for each α; S_α is the source term (defined as in the box models) and λ is the radionuclide decay constant. Equations for the water column and bottom sediment layer are linked by fluxes of activity. A numerical solution of these equations is required; usually applying finite differences (Periáñez, 2005a). An example of a computational grid which may be used to apply a finite difference technique

can be seen in Fig. 3. The area under study is divided into a number of small grid cells (in this example 1884 × 3712 m^2), which allows a larger spatial resolution than in box models.

Some examples of Eulerian radionuclide transport models are Breton and Salomon (1995), Harms (1997), Koziy et al. (1998), Preller and Cheng (1999), Cetina et al. (2000), Bailly du Bois and Dumas (2005), Estournel et al. (2012), Periáñez et al. (2012), Maderich et al. (2016, 2017), among many others.

3.1.3. Lagrangian Models

In Lagrangian models the released activity is represented by a number of particles, each one equivalent to a given amount of activity (Bq). Many examples of Lagrangian models applied to radionuclide transport are found in literature (Schonfeld, 1995; Harms et al., 2000; Periáñez and Elliot, 2002; Toscano-Jiménez and García-Tenorio, 2004; Periáñez, 2005b; Nakano et al., 2010; Kawamura et al., 2011; Kobayashi et al., 2007; Min et al., 2013; Periáñez et al., 2016a, among many others). The path followed by each particle is calculated and radionuclide concentrations are obtained from the number of particles per volume or mass unit. The equations describing variations of particle (in state α) position over each time increment dt are given by the Itô (Protter, 2004) stochastic differential equations:

$$dx = u_\alpha dt + \frac{\partial K_h}{\partial x}dt + \sqrt{2K_h}dW_x, \tag{4}$$

$$dy = v_\alpha dt + \frac{\partial K_h}{\partial y}dt + \sqrt{2K_h}dW_y, \tag{5}$$

$$dz = w_\alpha dt + \frac{\partial K_v}{\partial z}dt + \sqrt{2K_v}dW_z, \tag{6}$$

where u_α, v_α and w_α are velocity components on coordinate axis (x, y, z) for state α; W_x, W_y, W_z are independent components of the stochastic motion, which have zero mean and variance dt ($\overline{dW_x^2} = \overline{dW_y^2} = \overline{dW_z^2} = dt$), and for a finite time step Δt they can be simulated as $\Delta W_x = \sqrt{\Delta t}R_x$, $\Delta W_y = \sqrt{\Delta t}R_y$, $\Delta W_z = \sqrt{\Delta t}R_z$, where (R_x, R_y, R_z) are normally distributed random variables having zero mean and standard deviation one. Derivatives of the diffusion coefficients above prevent the artificial accumulation of particles in regions of low diffusivity (Proehl et al., 2005; Lynch et al., 2015).

A method based on the solution of the Kolmogorov equation for the transfer probability is used in the stochastic approach for simulating transfers between different states of the radionuclide. If a particle at a point in time is in state α, then the probability $p_{\alpha\beta}$ of transfer to state β during time dt is found from the solution of the Kolmogorov equation with initial conditions $p_{\alpha\beta}(0) = \delta_{\alpha\beta}$, where $\delta_{\alpha\beta}$ is the Kronecker delta.[4] This equation, known as the Kolmogorov forward equation (Parzen, 1962), is written as:

$$\frac{dp_{\alpha\beta}}{dt} = \sum_{\gamma=1}^{n} k_{\gamma\beta}p_{\alpha\gamma}. \tag{7}$$

For a two-state transfer between dissolved (state 1) and adsorbed radionuclides (state 2), considering a one-step reaction and one sediment size (see Section 3.4), the equations for the transfer probabilities are:

$$\frac{dp_{12}}{dt} = k_1 p_{11} - k_2 p_{12}, \tag{8}$$

$$\frac{dp_{21}}{dt} = -k_1 p_{21} + k_2 p_{22}, \tag{9}$$

$$\frac{dp_{11}}{dt} = -k_1 p_{11} + k_2 p_{12}, \tag{10}$$

$$\frac{dp_{22}}{dt} = k_1 p_{21} - k_2 p_{22}, \tag{11}$$

[4] This function is defined as:

$$\delta_{\alpha,\beta} = \begin{cases} 1 & \alpha = \beta \\ 0 & \alpha \neq \beta \end{cases}$$

where we have the following adsorption and desorption coefficients: $k_1 = -k_{11} = k_{12}$ and $k_2 = -k_{22} = k_{21}$. From the solution of the system of Eqs. (8)–(9), the probabilities to change state during each time step will be equal to:

$$p_{12} = \frac{k_1}{k_1 + k_2}(1 - \exp(-(k_1 + k_2)\Delta t)), \tag{12}$$

$$p_{21} = \frac{k_2}{k_1 + k_2}(1 - \exp(-(k_1 + k_2)\Delta t)), \tag{13}$$

whereas $p_{11} = 1 - p_{12}$ and $p_{22} = 1 - p_{21}$ are the probabilities to remain in the previous state. Similarly, "death" of particles due to the radioactive decay can be described. If more than two possible states (adsorption on multifractional sediments and/or two step kinetic reaction etc.) are considered, then the system of Eqs. (7) should be solved numerically. For small $k_{\alpha\beta}\Delta t$, we can approximate equation (7) by the first order numerical scheme that yields

$$p_{\alpha\beta}(\Delta t) - p_{\alpha\beta}(0) \approx \sum_{\gamma=1}^{n} k_{\gamma\beta}p_{\alpha\gamma}(0)\Delta t = \sum_{\gamma=1}^{n} k_{\gamma\beta}\delta_{\alpha\gamma}\Delta t = k_{\alpha\beta}\Delta t. \tag{14}$$

As seen from (12)–(13) and (14), for small $k_{\alpha\beta}\Delta t$ solutions for $p_{\alpha\beta}(\Delta t)$ coincide, but (14) is applicable for any number of states.

A comparison of the main advantages and disadvantages of each model type is included in Section 5.5, where the particular situations in which each model type is advantageous are discussed.

3.2. Model spatial and temporal resolution

Models may adopt different structures depending on the physical characteristics of the area to be modelled. A one-dimensional model may be enough to simulate dispersion in an essentially one-dimensional structure, like a channel, where mixing in depth and in the transverse direction is fast. Such a model was applied to the Suez Canal (Abril et al., 2000). A simple model in which the conservation of mass, heat, salt and tracer are considered only in the vertical direction and inflows and outflows through straits act as forcing terms for the vertical circulation and feedback for the Mediterranean sub-basins was developed by Maderich (1998) to reconstruct ^{137}Cs contamination in the period 1960–2010 for the Mediterranean Seas chain.

Two-dimensional depth-averaged models may be applied to estuaries, lakes, bays and coastal areas. They assume that the water column is homogeneous, i.e., no vertical structure exists either in water circulation or in radionuclide concentrations (Periáñez et al., 2013c). In general, these conditions are more easily satisfied in shallow waters and in winter: wind-induced mixing is more intense during this season; although some buoyancy driven mixing due to surface cooling may also occur. Calm conditions in summer may lead to vertical stratification and, thus, the validity of a depth-averaged model would be seasonally dependent. An example where this happens is the North Sea (van Leeuwen et al., 2015). Winter cooling in the tropics is not enough to destroy stratification, which is permanent. In contrast, stratification is virtually non-existent in polar regions.

A full three-dimensional model is required if there is vertical structure in the water column. These models present the highest level of complexity (Harms et al., 2000; Hazell and England, 2003; Gao et al., 2004; Orre et al., 2007; Kobayashi et al., 2007; Maderich et al., 2017; Min et al., 2013, etc.). In special situations other intermediate approaches are valid. For instance, two-layer models consist of two depth-averaged models, one over the other. These models can be applied if the marine area may be treated as two well-mixed water layers which move in different directions. This is the case in the Alborán Sea, western Mediterranean, (Periáñez, 2008). A vertical two-dimensional model can also be applied in fjords, for instance, where transverse mixing is fast but a significant vertical structure exists due to stratification.

Temporal resolution of the model (time step used for integration of the corresponding differential equations) is another key factor to

be defined. If we are interested in solving transport induced by tides, temporal resolution must be much higher than the tidal period. Average circulation fields are used for longer range calculations. These fields can be daily averages, monthly averages, annual averages etc, again depending on the problem to be solved. Time step is also limited by numerical stability conditions (Kowalik and Murty, 1993).

3.3. Water circulation

The description of water circulation is a key factor in radionuclide transport modelling since, as commented before, water currents are the main vector in radionuclide transport in the sea. Even in box models, exchange rates between boxes are derived from water circulation patterns, as already mentioned. In addition, water currents and density stratification may also be used to derive the diffusion coefficients describing turbulent mixing; for instance using the Richardson number based schemes (Pacanowski and Philander, 1981), or the generic length-scale turbulence model (Umlauf and Burchard, 2003) for vertical diffusivity and the Smagorinsky's scheme (Cushman-Roisin and Beckers, 2011) for horizontal diffusivity. Other methods exist to derive diffusivities (Kowalik and Murty, 1993) since turbulence is still an open problem in fluid dynamics and can only be parameterized. Most of the circulation models are written in Boussinesq and hydrostatic approximations. Besides physical parameterizations (turbulent mixing, exchange with atmosphere by heat and mass, ice cover), they differ in the vertical discretization (e.g. z-, σ and isopycnal coordinate systems) and horizontal discretization (structured and unstructured meshes). Reviews on physics and numerics of models and formulation of diffusivities are given in James (2002) and Fox-Kemper et al. (2019). A description of equations, approaches and numerical methods used in hydrodynamic models may be seen in the books by Kowalik and Murty (1993), Müller and von Storch (2004), Chassignet and Verron (2006), Miller (2007), Kampf (2009, 2010), Cushman-Roisin and Beckers (2011) and Glover et al. (2011), among many others. Herzfeld et al. (2011) present an excellent review on the specification of open boundary conditions in ocean models.

Generally speaking, there are two ways of obtaining water circulation for a radionuclide transport model: to use pre-computed fields from a global ocean forecasting system and to solve the fluid dynamic equations in regional applications. These methods are briefly described in the following subsections.

3.3.1. Global models

One option is to use pre-computed circulation by an operational global ocean forecasting system or reanalysis, in which past observation data are assimilated by the circulation model. These circulation data may be downloaded from each model web page in binary format. For instance, a number of different ocean forecasting models were applied to simulate the dispersion of FDNPP releases in the Pacific Ocean (Periáñez et al., 2015a). Some examples of global models are:

MOM, Modular Ocean Model. The model was developed and supported by researchers at NOAA Geophysical Fluid Dynamics Laboratory (GFDL)[5]. Initially it was developed by K. Bryan and M.Cox in 1960–1980s as a first global circulation model being permanently improved and extended. The model uses generalized orthogonal horizontal coordinates and variable vertical coordinates.

HYCOM, Hybrid Coordinate Ocean Model. HYCOM[6] consortium is a multi-institutional effort sponsored by the National Ocean Partnership (NOPP) as a part of U.S. Global Ocean Data Assimilation Experiment (GODAE) (Bleck, 2001). HYCOM is a primitive equation general circulation model with vertical coordinates that remain isopycnic in the open, stratified ocean.

NEMO, Nucleus for European Modelling of the Ocean. NEMO[7] is a framework of ocean related models for ocean dynamics and thermodynamics, for sea–ice dynamics and thermodynamics, and for tracer transport. The model uses a curvilinear orthogonal grid in the horizontal direction, and in the vertical direction it uses z−coordinates, terrain-following coordinates, or a mixture of the two. The embedded zooms are created using the two-way nesting package AGRIF.[8] This is a package for the integration of adaptive mesh refinement features within a multidimensional model. The nesting capability allows resolution to be focused over a region of interest by introducing an additional grid.

OFES, Ocean global circulation model For the Earth Simulator. OFES[9] was developed by the Japan Agency for Marine-Earth Science and Technology (JAMSTEC). Horizontal resolution is 0.1°. and there are 54 vertical levels, with increasing thickness from the surface towards the sea bottom. A comparison of model performance with data in several regions of the global ocean may be seen in Masumoto et al. (2004). This model was used by Periáñez et al. (2016a) to simulate historical releases from European nuclear fuel reprocessing plants in the North Atlantic.

The reanalysis data can be downloaded from the Copernicus Marine Environment Monitoring Service (CMEMS)[10], from the HYCOM web site or from several other databases.

As an example, the surface circulation in the Pacific Ocean (averaged value for March 2011), as applied to simulate the transport of Fukushima releases in the whole North Pacific (Periáñez et al., 2019), is presented in Fig. 4. This water circulation was calculated in JAMSTEC (Japan Agency of Marine-Earth Science and Technology) with FORA[11] (Four-dimensional Variational Ocean ReAnalysis) model (Usui et al., 2016). FORA is the first-ever dataset covering the western North Pacific over the last three decades (1982–2014) at eddy-resolving resolution. This is a cooperative work of JAMSTEC and the Meteorological Research Institute, Japan Meteorological Agency (JMA/MRI) using the Earth Simulator. The domain extends 117° E-160° W, 15° N-65° N with horizontal resolution 1/10° in both longitude and latitude and 54 vertical levels (0-6300 m). The general large scale circulation in the western Pacific Ocean is dominated by the interaction between the Kuroshio and Oyashio currents. The Kuroshio Current is the western boundary current in the north Pacific, which flows along the coast of Japan towards the north and curves to the central Pacific Ocean, then forming the so-called Kuroshio Extension. The Oyashio Current is a cold current which flows from the north. These two current systems converge in the coastal waters off Fukushima coast, which leads to the generation of unsteady eddies in the area. These features may be seen in Fig. 4. Transport calculations were carried out in the frame of the IAEA MODARIA-II program (Periáñez et al., 2019) with these hydrodynamic fields (monthly means from 2011 to 2014).

3.3.2. Regional models

Another option is to solve the fluid dynamic Navier–Stokes equations to calculate water circulation (Kowalik and Murty, 1993) in the area of interest. This can be done either in on-line (water circulation and radionuclide transport calculated at the same time) or off-line (circulation is calculated in advance and stored) modes (Periáñez, 2005a). A numerical method is required and, as in the case with the transport model, the model structure must be selected according to the characteristics of the area (2D, 3D model etc.). Three well known regional models used in radionuclide transport studies are:

[5] https://www.gfdl.noaa.gov/mom-ocean-model/.
[6] https://hycom.org/.
[7] http://www.nemo-ocean.eu/.
[8] http://forge.ipsl.jussieu.fr/nemo/wiki/Users/SetupNewConfiguration/AGRIF-nesting-tool.
[9] http://www.jamstec.go.jp/esc/research/AtmOcn/product/ofes.html#cite_note-1.
[10] http://marine.copernicus.eu/.
[11] http://synthesis.jamstec.go.jp/FORA/e/index.html.

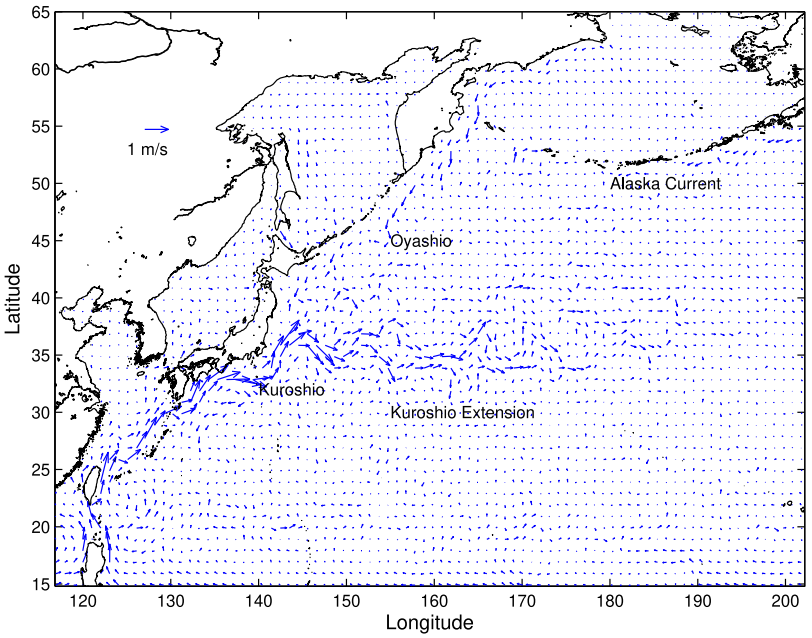

Fig. 4. Example of water circulation obtained from an ocean model. It corresponds to the average surface circulation in March 2011. The model is FORA (Four-dimensional Variational Ocean ReAnalysis model; (Usui et al., 2016). This type of velocity vectors are used in Eulerian and Lagrangian dispersion models to evaluate advective transport of radionuclides in the water column, and often also diffusion coefficients.

POM, Priceton Ocean Model. POM[12] was first described by Blumberg and Mellor (1987). It is a sigma coordinate (terrain-following), free surface ocean model with embedded turbulence and wave sub-models, as well as wet–dry capability. This model has been used since the 1980s, and continues with innovative new developments until today.

ROMS.[13] The 3D baroclinic free-surface, terrain-following, primitive equations (Haidvogel et al., 2000; Shchepetkin and McWilliams, 2009) are solved. The model, widely used for different applications, includes ice and sediment transport modules, and the two-way nesting package AGRIF. Model equations are evaluated using orthogonal curvilinear coordinates in the horizontal and terrain-following coordinates in the vertical.

SELFE/SCHISM. The 3D primitive equation model SELFE (Zhang and Battista, 2008; Roland et al., 2012) renamed now as SCHISM[14] is an open-source community supported modelling system with an embedded wave model and a sediment transport model as well. This model is based on unstructured grids and localized sigma coordinates with shaved cell (LSC^2), designed for seamless simulation of 3D baroclinic circulation across the scales.

These models were used in many radionuclide transport calculations (e.g. Kobayashi et al., 2007; Miyazawa et al., 2012; Tsumune et al., 2012, 2013; Periáñez et al., 2016b; Maderich et al., 2017).

3.4. Water/sediment interactions and other processes

Radionuclide exchange between the dissolved and solid phases is a significant process in the transport of non-conservative radionuclides in the marine environment. A good general review is given in the book by Duursma and Carroll (1996). Water–sediment interactions are schematically shown in Fig. 5 (Maderich et al., 2017). In the water column, radionuclides in the dissolved and particulate phases are transported by currents (advection processes) and turbulent diffusion. Radionuclides in the dissolved phase interact with the particulate phase in suspended sediments and bottom deposits. Exchange of activity between the dissolved and particulate phases is described by adsorption/desorption processes. Settling of contaminated suspended sediments and bottom erosion result in radionuclide exchanges between the bottom and suspended sediment. The transfer of activity between the water column and the pore water in the bottom sediment is governed by bottom boundary layer turbulence regulated diffusional processes (Maderich et al., 2017). The migration of activity in the sediments is due to molecular diffusion, diffusion driven by bioturbation, bioirrigation and also due to advection driven by surface waves and by subsurface groundwater flow (Maderich et al., 2017). A basic microscopy theory of radionuclide water/suspended sediment interactions was given by Abril (1998).

Radionuclide transfers between water and sediments were initially described in models in terms of the equilibrium distribution coefficient, k_d, of the considered radionuclide [as for instance in the models by Abril and García-León (1993)]. The marine distribution coefficient for a given radionuclide, k_d, is defined (Carroll et al., 1999; Johansson et al., 2001; IAEA, 2004; Takata et al., 2016; Periáñez et al., 2018) as the ratio between the radionuclide concentration in the solid phase (suspended matter or bed sediment) and the concentration in water (dissolved phase):

$$k_d = \frac{C_s}{C_w}, \tag{15}$$

where C_s and C_w are, respectively, radionuclide concentrations in the solid (Bq kg^{-1}) and dissolved (Bq m^{-3}) phases. Such concentrations have to be at equilibrium, i.e., after the partition of the radionuclide between phases has reached equilibrium. This k_d is measured in SI units of m^{3}kg^{-1}. Table 1 summarizes recommended k_d values (IAEA, 2004) for ocean margin and open ocean and for a number of elements.

Thus, in the first models it was assumed that partition of the radionuclide between the liquid and solid (suspended matter and/or bed sediments) achieves instantaneous equilibrium. From the radionuclide concentration in water, knowing the radionuclide k_d, concentration in

[12] http://www.ccpo.odu.edu/POMWEB/.
[13] https://www.myroms.org/
[14] http://ccrm.vims.edu/schism/.

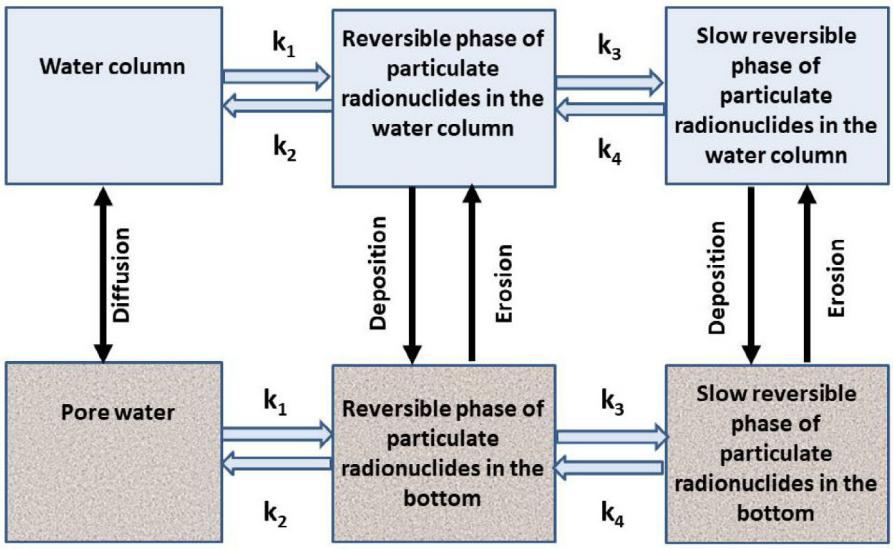

Fig. 5. Scheme representing exchanges of radionuclides between dissolved and solid (suspended matter and bed sediments) phases (from Maderich et al., 2017). Kinetic rates k_1 and k_2 describe uptake and release, respectively, for the first fast reaction. Rates k_3 and k_4 describe the second, slower, reaction. Only k_1 and k_2 are considered in the case of 1-step models; the full scheme is applied in 2-step kinetic models.

Table 1
IAEA (2004) recommended k_d values (m^3kg^{-1}) for open ocean and ocean margins for a number of elements.

Element	Open ocean	Ocean margin
H	1×10^{-3}	1×10^{-3}
Cr	4×10^{2}	5×10^{1}
Mn	2×10^{5}	2×10^{3}
Co	5×10^{4}	3×10^{2}
Ni	3×10^{2}	2×10^{1}
Sr	2×10^{-1}	8×10^{-3}
Tc	1×10^{-1}	1×10^{-1}
Cd	3×10^{0}	3×10^{1}
Sb	4×10^{0}	2×10^{0}
Cs	2×10^{0}	4×10^{0}
Ba	5×10^{0}	2×10^{0}
Pb	1×10^{4}	1×10^{2}
Po	2×10^{4}	2×10^{4}
Ra	4×10^{0}	2×10^{0}
Th	5×10^{3}	3×10^{3}
U	5×10^{-1}	1×10^{0}
Pu	1×10^{2}	1×10^{2}
Am	2×10^{3}	2×10^{3}

the sediment is calculated through equation (15). This can be an adequate simplification in the long-term assessments usually done with box models; however it is clear now for the marine modelling community that such is not the case in the near field, in both cases of accidental and chronic releases. Carroll et al. (1997) noted that in dynamic coastal environments the distribution of radionuclides between water and sediment may not reach equilibrium. For instance, in the Amazon Shelf the rates between measured concentrations of ^{234}Th in sediment and water varied over two orders of magnitude during a single tidal cycle.

In the case of an accident, radionuclide concentration in the sediment would be overestimated in the initial phase and underestimated later if equilibrium is assumed. Moreover, as demonstrated in Periáñez (2003a), Periáñez et al. (2018), equilibrium is never reached in the near field in the case of chronic releases, even if they are constant in time.

As a consequence, the state-of-the-art approach involves the use of kinetic models, by which adsorption/desorption reactions are described in a dynamic way and non-equilibrium situations can be treated in a more valid way (Nyffeler et al., 1984; Laissaoui et al., 1998; Periáñez et al., 2018). Uptake/release of radionuclides may be considered to be described by a single reversible reaction (1-step model, Fig. 5). This reaction is described by kinetic rates k_1 and k_2 for adsorption and release respectively, which are reaction rates measured in s^{-1}.

The differential equations whose solution gives the time evolution of activity in water and sediment, A_w and A_s, are:

$$\begin{aligned} \frac{\partial A_w}{\partial t} &= -k_1 A_w + k_2 A_s \\ \frac{\partial A_s}{\partial t} &= k_1 A_w - k_2 A_s \end{aligned} \tag{16}$$

Note that kinetic rates k_1 and k_2 operationally include all mechanisms for adsorption, like electrostatic attraction, ion exchange etc, and desorption. Also, in steady-state conditions (time derivatives are equal to zero) it is found from Eqs. (16) (Periáñez et al., 2018) that:

$$k_d = \frac{1}{m}\frac{k_1}{k_2}, \tag{17}$$

where m is the concentration of sediment (mass of sediment per water volume unit). Thus, kinetic rates are related with the equilibrium k_d.

Nevertheless, there has been evidence to suggest that uptake takes place in two stages: fast surface adsorption followed by slow migration of ions to pores and interlattice spacings (Nyffeler et al., 1984; Turner et al., 1992; Turner and Millward, 1994; Oughton et al., 1997; Ciffroy et al., 2001; El-Mrabet et al., 2001). Thus, a 2-step model (Fig. 5) has also been included in some marine radionuclide transport models. A 2-step model considers that exchanges are governed by two consecutive reversible reactions: surface adsorption is followed by another process that may be a slow diffusion of ions into pores and interlattice spacings, inner complex formation or a transformation such as an oxidation. The forward and backward rates for this second reaction are k_3 and k_4 respectively. Thus, radionuclides adsorbed by sediments are divided into two phases: a reversible and a slowly reversible fraction. It was shown that the 2-step model reproduces both the adsorption and release kinetics of ^{137}Cs in the Irish Sea, where it is released from Sellafield nuclear fuel reprocessing plant (Periáñez, 2003b). It was also recently included in THREETOX, which is the marine radionuclide transport model implemented within the JRODOS decision support system (Maderich et al., 2008, 2016).

Although a kinetic model provides a more realistic description of adsorption/desorption processes than a k_d model, it requires the specification of kinetic rates. Obtaining values of kinetic rates is not easy since requires laboratory adsorption experiments with marine water

Table 2
Kinetic rates (s^{-1}) for several radionuclides, as derived from adsorption experiments: Børretzen and Salbu (2000) for ^{109}Cd and ^{60}Co, Børretzen and Salbu (2002) for ^{134}Cs and Nyffeler et al. (1984) for the rest of radionuclides. A zero rate means that such reaction is not relevant in the temporal frame of the experiment.

	k_1	k_2	k_3	k_4
^{134}Cs	3.4×10^{-4}	1.2×10^{-5}	1.0×10^{-6}	2.0×10^{-7}
^{109}Cd	5.9×10^{-4}	1.2×10^{-5}	1.0×10^{-6}	1.0×10^{-7}
^{60}Co	2.2×10^{-2}	1.2×10^{-5}	1.0×10^{-6}	4.0×10^{-8}
^{7}Be	4.6×10^{-2}	1.2×10^{-6}	6.9×10^{-8}	0
^{54}Mn	1.9×10^{-2}	4.1×10^{-7}	2.1×10^{-7}	0
^{59}Fe	1.7×10^{-1}	2.7×10^{-7}	6.9×10^{-8}	0
^{133}Ba	2.9×10^{-4}	1.2×10^{-5}	0	0
^{125}Sb	1.7×10^{-3}	5.8×10^{-6}	0	0
^{65}Zn	4.5×10^{-3}	3.7×10^{-6}	0	0
^{75}Se	2.1×10^{-3}	1.2×10^{-5}	0	0
^{203}Hg	4.1×10^{-1}	2.3×10^{-6}	0	0

and sediments, as those described in Børretzen and Salbu (2000, 2002) and Nyffeler et al. (1984). As an example, kinetic rates derived from these experiments are shown in Table 2. It should be taken into account that these values must be considered as tentative since kinetic rates are site specific, depending on the water and sediment properties. Overall, information about kinetic rates is generally scarce in current literature.

More complex water/sediment interaction models which involve parallel and consecutive reactions have been formulated as well (Barros and Abril, 2004; Benkdad et al., 2008), but they have not been yet implemented in a marine radionuclide transport model.

From our experience, it is also relevant to develop models of radionuclide migration in the seabed for heterogeneous environments where the sediment characteristics (sediment fractions, densities and porosity) vary in space (Aldridge et al., 2003; Higashi et al., 2015; Maderich et al., 2017): a few years after the Fukushima accident, bottom sediments on the Japan shelf have become the main source of radionuclide remobilization to the water column due to the rapid dilution of radionuclides by intensive currents and eddies (Buesseler et al., 2017). Similarly, desorption from bottom sediments is the main source of dissolved ^{137}Cs in the Irish Sea following the reduction of routine releases from Sellafield reprocessing plant (Mitchell et al., 1999). A detailed discussion on the water/sediment interaction problem can be found in Periáñez et al. (2018).

Other processes may be included in marine transport models, if required. For instance, the behaviour of plutonium in aquatic systems is of considerable complexity due to the fact that it can exist in different oxidation states simultaneously and these change in time. Thus, Pu (III) and Pu (IV) predominate as the reduced forms and Pu (V) and Pu (VI) as the oxidized forms (Mitchell et al., 1995). The reduced Pu is highly reactive with particles and possesses a distribution coefficient that is two orders of magnitude higher than that of the more soluble oxidized Pu (Mitchell et al., 1995). Hence the k_d values observed in field measurements represent the properties of the mixture of oxidation states that is present in the particular sample. Also, the oxidation state of Pu may change with time. For instance, Pu is released from Sellafield reprocessing plant in a reduced form, but after some days an equilibrium in the partition of Pu between the reduced and oxidized species is achieved. Redox reactions may be included in a model using kinetic rates, similarly to uptake/release reactions between water and sediments. Details may be seen in Perianez (2003b). A model for plutonium behaviour in the marine environment representing the oxidation state distribution and partitioning of plutonium between the soluble, colloidal, suspended particulate and seabed sediment fractions in the vicinity of the Sellafield plant was presented in Vives i Batlle et al. (2007). However, this model consisted of a single box in the Sellafield area and, in consequence, spatial distributions could not be obtained from it.

Table 3
Generic parameters used in the dynamic biological uptake model (from Maderich et al., 2014a) for ^{137}Cs. The concentration factor for phytoplankton is $CR_{phyto} = 20$ l/kg.

	Zooplankton	Non-pisc. fish	Pisc. fish
$T_{0.5}$ (day)	5	75	200
a	0.2	0.5	0.7
b	0.001	0.001	0.001
K_1 (day^{-1})	1.0	0.035	0.0055
K_w (m^3/kg day)	1.5	0.1	0.075

3.5. Biological uptake models

A further step is to integrate dynamic biota uptake models and turnover models within marine dispersion models. This was done in most box models using an equilibrium approximation based on a concentration factor, CR, between water and biota. This CR, in analogy with the k_d, is defined as the ratio between radionuclide concentration in a given species of biota and concentration in water:

$$CR = \frac{C_{bio}}{C_w}. \tag{18}$$

Thus, concentration in biota, C_{bio}, can be calculated from the CR and the calculated concentration in water, assuming equilibrium (Carvalho, 2018).

In a recent model intercomparison carried out within the IAEA MODARIA programme (Vives i Batlle et al., 2016) it was shown that dynamic biota models, which handle situations out from equilibrium, perform better than equilibrium models. It was also demonstrated that a more correct description of radionuclide concentration in biota, by means of kinetic modelling, has a significant influence for radioecological dose assessment and, therefore, for decision-making.

A basic dynamic model consists of four species (Heling et al., 2002; Maderich et al., 2014a,b) : phytoplankton, zooplankton, non-piscivorous and piscivorous fish (Fig. 6, from (Maderich et al., 2014a). The basic equation connecting concentration of activity in predator C_{pred} (Bq kg^{-1} wet weight) with activity concentration in food C_f (Bq kg^{-1} wet weight) is:

$$\frac{\partial C_{pred}}{\partial t} = aK_1C_f + bK_wC_w - K_{0.5}C_{pred}, \tag{19}$$

where K_1 (s^{-1}) is food uptake rate, a is the transfer coefficient through food, K_w is water uptake rate (s^{-1}), b is the transfer coefficient from water and C_w is activity concentration in water (Bq m^{-3}). $K_{0.5}$ is the radionuclide elimination rate from the body of the organism given by $K_{0.5} = \ln 2 T_{0.5}^{-1}$, where $T_{0.5}$ is the biological half-life of the radionuclide(s). Thus, all organisms take radionuclides from water, phytoplankton is the food for zooplankton, zooplankton is the food for non-piscivorous fish and this is the food for piscivorous fish (as summarized in Fig. 6). Phytoplankton exchanges radionuclides only with the water via adsorption and desorption processes. Due to the rapid uptake and short retention time of radioactivity, the concentration of radionuclides in phytoplankton is calculated using the equilibrium approach:

$$C_{phyto} = CR_{phyto}C_w, \tag{20}$$

where CR_{phyto} (m^3kg^{-1}, wet weight) is the concentration ratio for phytoplankton (see Eq. (18)). Standard literature values for all these parameters for the four considered species may be seen in Table 1 in Maderich et al. (2014a), which is reproduced in Table 3. Further improvement of biota models includes the description of radionuclide transfer using metabolic rates of the marine organism, model which was developed by Konovalenko et al. (2014). The dynamic approach given by (19), combined with a model of spatial and temporal biomass dynamics, was applied by Walters and Christensen (2018).

An organism is described as a single box in most dynamic biota models. In D-DAT model (Vives i Batlle et al., 2008) the organism

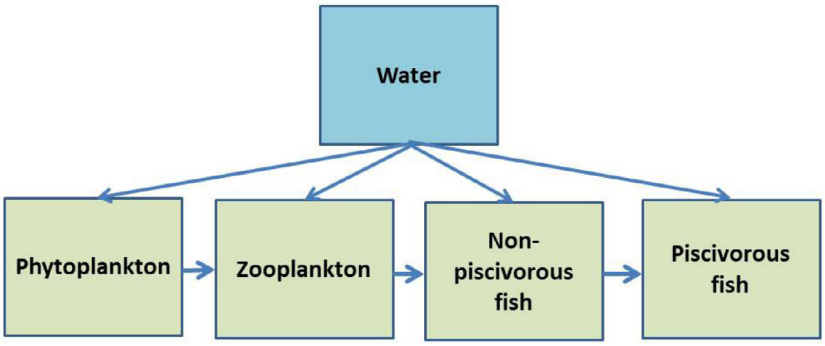

Fig. 6. Scheme of radionuclide transfer (arrows) in a dynamic food chain model (from Maderich et al., 2014a).

comprises two compartments describing fast and slow uptake and depuration rates, which allows a better description of the temporal variations of radionuclide concentrations after an accidental release. Physiologically-based pharmacokinetic multi-compartmental models (Thomann et al., 1997) have an interesting potential. In these models well-mixed compartments correspond to the fish organs/tissues or groups of organs/tissues according to specific metabolic functions. The kinetics of exchange is first order, and transport between compartments is governed by blood flux. However, these models require many parameters, which should be calibrated from controlled experiments. Therefore, this approach was rarely used in radioecology (Garnier-Laplace et al., 2000).

Dynamic food web models can be included within a marine dispersion model in order to simulate radionuclide concentrations in biota at oceanic scales. Thus, previous equations must be included in the marine transport model. They are solved for each grid cell in the domain. From the calculated concentration in water, Eqs. (19) and (20) provide radionuclide concentrations in each grid cell, time and species.

This work was recently carried out in MODARIA-II program (Periáñez et al., 2019): ^{137}Cs concentrations in phytoplankton, zooplankton, non-piscivorous and piscivorous fish were calculated over the whole north Pacific Ocean up to two years after the Fukushima accident. Several transport models with different models for biota uptake were applied and results compared with measurements in water, sediment and biota (zooplankton and fish). Three types of biological uptake models were tested: an equilibrium model based on a concentration ratio, the dynamic model described here and an allometry method. Dynamic models provided the known patterns of delayed rise of activity concentration in biota. Details may be seen in the cited reference.

A more complex model of ^{14}C transfer in the marine trophic web was recently described by Tierney et al. (2018). However, advection was treated in a limited way and measurement data could not be reproduced in some areas.

As a resume, a compilation of the basic characteristics of some radionuclide marine transport models are listed in Tables 4–6 for, respectively, transport models linked with hydrodynamic models, standalone radionuclide transport models which import water circulation, and box models; including all the previously cited characteristics.

In general, equilibrium or dynamic models may be used to describe water/sediment interactions and biota uptake. The main advantages and disadvantages of each approach are discussed in Section 5.5.

Fig. 7 presents a scheme of the modelling procedures. The hydrodynamic model provides water currents which are required by Eulerian and Lagrangian models for simulating physical transport (advection and diffusion). Box models need water fluxes which are deduced from observations and/or calculated from water currents. Bio-geochemical processes (water/sediment interactions and uptake of radionuclides by biota) may be described by equilibrium or dynamic models. In general, the equilibrium approach is used in box models, since long temporal scales are simulated with them. In contrast, dynamic models are more frequently used in Eulerian and Lagrangian models. Suspended matter concentrations and sedimentation rates may also be required if radionuclide interactions with suspended particulate matter are included. Mean values of these magnitudes, deduced from observations, are generally used in box models. Values calculated with a sediment transport model (which can be Eulerian or Lagrangian) are more often used in Eulerian and Lagrangian radionuclide transport models. The physical transport model, with the added bio-geochemical model, provides radionuclide concentrations in water, sediment and biota species.

4. Models applied to simulate and estimate Fukushima releases in the Pacific Ocean

Significant amounts of radioactive material were released to the environment from FDNPP as a consequence of the March 2011 earthquake and tsunami. Radionuclides released to the atmosphere were transported eastwards by a jet stream, and they reached the coast of North America in four days (Takemura et al., 2011). Some of these radionuclides were deposited on the Pacific Ocean surface by wet and dry processes. In addition, water used to cool a damaged nuclear reactor directly leaked into the ocean (Kobayashi et al., 2013). Since most of the recent efforts in modelling marine radionuclide dispersion were focused in Fukushima releases in the Pacific Ocean, a brief review of some of the models applied to this problem is included. A review on dispersion patterns of^{137}Cs released from Fukushima in the Pacific derived from field measurements (not models) may be seen in Kaeriyama (2017).

The first modelling studies into the dispersion of Fukushima releases in the Pacific Ocean were published soon after the accident. Thus, the spreading of ^{131}I and ^{137}Cs using the Lagrangian model SEA-GEARN, developed at JAEA, was simulated by Kawamura et al. (2011). A Lagrangian code was applied to simulate the dispersion of ^{137}Cs and ^{134}Cs in the world ocean up to 30 years after the accident (Nakano and Povinec, 2012). Annually averaged water circulation was used for this purpose. The dispersion of ^{137}Cs, using a high resolution (1 km) regional Eulerian model, during the first three months after the accident was simulated by Tsumune et al. (2012). Later, simulations were extended to one year in Tsumune et al. (2013).

Ten year long simulations of ^{137}Cs dispersion in the Pacific Ocean were also performed by Behrens et al. (2012). Water circulation of the past 10 years was used for this purpose. They found that the initial current field was relevant for ^{137}Cs spreading in the first months after the accident, but this relevance fades in the long-term. Also, these authors found that concentrations would be nearly homogeneous over the whole Pacific after some 10 years. Simulations finally indicated a fast mixing over the upper 500 m of the water column. Similar results were found by other authors (Kawamura et al., 2014): radioactive caesium concentration was efficiently diluted in the North Pacific 2.5 years after the accident. The meso-scale eddies in the Kuroshio Extension (see Fig. 4) played an important role in diluting the radioactive

Table 4
Transport models linked with hydrodynamic models. Transport of radionuclides only in solute is denoted by S, in suspended sediments by SS and in bottom sediments by BS.

	Dimensions	Density stratification	Sediment transport	Ice transport	Radionuclide phases	Radionuclide transport mode
Prandle (1984)	2D	No	No	No	S	Eulerian
Onishi and Trent (1992)	3D	Yes	Yes	No	S-SS-BS	Eulerian
Abril and García-León (1993)	2D	No	Yes	No	S-SS-BS	Eulerian
Harms (1997)	3D	Yes	No	Yes	S	Eulerian
Margvelashvily et al. (1997)	3D	Yes	Yes	No	S-SS-BS	Eulerian
Zheleznyak et al. (1992)	2D	No	Yes	No	S-SS-BS	Eulerian
Koziy et al. (1998)	3D	Yes	Yes	Yes	S-SS-BS	Eulerian
Aldridge (1998)	2D	No	Yes	No	S-SS-BS	Eulerian
Periáñez (1999)	3D	No	Yes	No	S-SS-BS	Eulerian
Kobayashi et al. (2007)	3D	Yes	Yes	No	S-SS-BS	Lagrangian
Choi et al. (2013)	3D	Yes	Yes	No	S-SS-BS	Lagrangian
Misumi et al. (2014)	3D	Yes	Yes	No	S-BS	Eulerian
Higashi et al. (2015)	3D	Yes	Yes	No	S-SS-BS	Eulerian
Maderich et al. (2017)	3D	Yes	Yes	No	S-SS-BS	Eulerian

Table 5
Stand-alone transport models. Transport of radionuclides only in solute is denoted by S, in suspended sediments by SS and in bottom sediments by BS.

	Dimension	Hydrodynamics	Time averaging	Sediment transport	Radionuclide transport	Transfer to biota
Breton and Salomon (1995)	2D	Tidal model	Tidal	No	S	No
Lyons et al. (1998)/CUMBRIA	2D	Tidal model	1 year	No	S-SS-BS	No
Goshawk et al. (2003)/MEAD	2D	Mike21	1 year	Yes	S-SS-BS	No
Gleizon and McDonald (2010)/MARISA	2D	Mike21	1 year	Yes	S-SS-BS	Yes
Nakano et al. (2010)	3D	MRI.COM	1 month	Yes	S-SS	No
Nakano and Povinec (2012)/LAMER	3D	Diagnostic	1 year	Yes	S-SS	Yes

Table 6
Box models. CR is a concentration factor.

	Number of boxes	Daughter products	Transfer to biota	Dose assessment
MARINA I CEC (1990)	44	No	CR	sea food ingestion sea sprays, sediments
MARINA II EC (2002)	72	No	CR	sea food ingestion sea sprays, sediments
NRPA Iosjpe et al. (2002)	129	No	CR	sea food ingestion sea sprays, sediments
PC-CREAM-08 Smith and Simmonds (2009)	55	No	CR	sea food ingestion inhalation of sea sprays (beach)sediments
MOIRA Monte (2011)	55	No	CR	sea food ingestion sea sprays, sediments
POSEIDON Lepicard et al. (1998)	44	Yes	CR	sea food ingestion
POSEIDON-R Lepicard et al. (2004)	Variable	Yes	CR/dynamic	sea food ingestion

patch. The ^{137}Cs concentrations in the surface, intermediate, and deep layers reduced to the pre-Fukushima values over the North Pacific some 2.5 years after the Fukushima accident. Similar conclusions were also obtained by Rossi et al. (2013, 2014): the Fukushima plume was rapidly diluted within the Kuroshio system over a time-scale of a few months. Over the subsequent decades a significant amount of Fukushima-derived radionuclides will spread across the North Pacific basin. The model estimated that a component of Fukushima ^{137}Cs was injected into the interior ocean via subduction, before eventually returning to the surface by coastal upwelling along the west coast of North America. The comparison of ^{137}Cs measurements along line P, placed about 1500 km west British Columbia, Canada (Smith et al., 2017), with modelling (Rossi et al., 2013, 2014; Tsubono et al., 2016) showed good agreement. Some discrepancies in the calculated time evolution of concentrations could be explained because of the assumption that releases occurred solely by direct discharges from FDNPP (Rossi et al., 2013, 2014). Simulations with both sources (atmospheric deposition and direct release) matched ocean measurements well (Tsubono et al., 2016). The relevance of atmospheric deposition was also studied by other authors (Honda et al., 2012), finding that the high^{137}Cs concentrations detected in surface waters north of 40° N one month after the accident should be attributed to atmospheric deposition.

The residence time of ^{137}Cs in the shelf was estimated, using a model, as 43±16 days (Dietze and Kriest, 2012). The effective horizontal diffusivities on the shelf and effective diffusivity for cross-shelf transport were evaluated as well. As discussed in Section 5.2, these authors highlighted the effects of numerical (artificial) diffusion which appears in Eulerian transport models.

It was found, using a Lagrangian model, that the Kuroshio current acts as a barrier (Rypina et al., 2013), as previously described by Jayne et al. (2009), which prevents the migration of radionuclides released from Fukushima towards the south (they would not travel south beyond the latitude of Tokyo). Instead, they are transported towards the central Pacific.

Other modelling studies, using an Eulerian model (Estournel et al., 2012), indicated that radionuclides stay close to the coastline for relatively long times and suggested the role of freshwater discharges

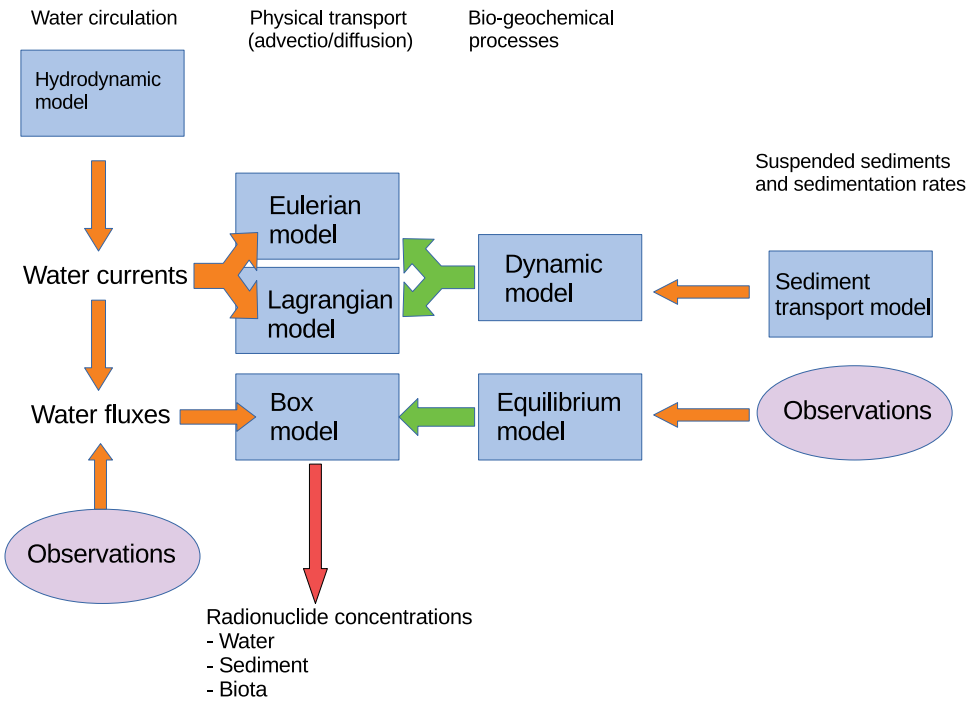

Fig. 7. Scheme showing the models required to simulate the dispersion of radionuclides in the marine environment.

from land in offshore dispersion events. The relevant role of winds in the shelf region was highlighted by means of sensitivity analysis, using as well an Eulerian dispersion model for ^{137}Cs, carried out by Miyazawa et al. (2012). In this sense, sensitivity analysis showed that a tuning of the wind drag coefficient was required for a better reproduction of ^{137}Cs measurements (Bailly du Bois et al., 2014).

In addition to pure advection–diffusion simulations, as those cited above, the transport of Fukushima radionuclides through drifter data and statistical methods was evaluated as well (Rypina et al., 2014).

One of the key problems in the marine dispersion modelling is the determination of radionuclide sources. There were several major sources of radionuclide contamination to the marine environment due to the FDNPP accident: (i) atmospheric deposition of radionuclides onto the sea surface, (ii) direct release of radionuclides into the ocean; (iii) releases from land via river and coastal runoff; (iv) groundwater release. The first two sources dominated during the first year after the accident. However, later ongoing groundwater and river releases were locally important. A feasible method for determining the source term is to combine radionuclide measurement data and advection–diffusion models ("inverse modelling"). A number of atmospheric transport models using different tracer inversion algorithms were used to estimate deposition onto the ocean surface (see review in SCJ (2014)). Scenarios of direct release in the ocean were constructed using monitoring data in the vicinity of FDNPP to scale computations (Kawamura et al., 2011; Tsumune et al., 2012, 2013). The estimated total direct releases of ^{137}Cs were 4 PBq (Kawamura et al., 2011) and 3.5 PBq (Tsumune et al., 2012, 2013). These scenarios were used in several subsequent studies (e.g. Tsumune et al., 2013; Kawamura et al., 2014; Tsubono et al., 2016; Maderich et al., 2014a,b; Bezhenar et al., 2016). A total direct release of 5.1–5.5 PBq, using a four-step inverse approach based on the measured ^{137}Cs activity south and north outlet channels of FDNPP, was also estimated (Estournel et al., 2012). The 27 PBq direct release estimate by Bailly du Bois et al. (2012) was based on interpolated monitoring data in a 50-km area around FDNPP and the environmental half-time for it, which was deduced from observations. However, this source term was considered to be significantly overestimated by Dietze and Kriest (2012). Inverse estimation of direct releases based on the Green function approach (Enting, 2002) was also carried out by Miyazawa et al. (2013). An inversion method based on minimizing the differences between model and cruise data was applied by Rypina et al. (2013) to estimate releases. Corresponding total direct release was 16.2 PBq. The total release of ^{137}Cs from FDNPP harbour was estimated by Kanda (2013) as 2.25 PBq. This value was comparable with estimates of Kawamura et al. (2014) and Tsumune et al. (2012). A 3.6 TBq y^{-1} continuous underground leak of contaminated water from FDNPP was also suggested by Kanda (2013). This value was confirmed by comparison of modelling results and measurements within an area with 15 km radius around FDNPP in the period 2012–2015 (Maderich et al., 2014a,b; Bezhenar et al., 2016). According to Kanda (2013), total river flux of ^{137}Cs in Fukushima, Ibaraki and Miyagi prefectures in 2012 was 1.56 TBq y^{-1}.

All modelling studies mentioned above (which does not try to be an exhaustive list) had the common feature that ^{137}Cs was treated as a conservative radionuclide which did not interact with sediments. The first models including ^{137}Cs contamination of bed sediments were described by Periáñez et al. (2012) and Min et al. (2013). In the first case a local study was carried out, covering only the coastal region of Japan. A larger domain was considered in the second paper. In both cases, calculated and measured ^{137}Cs concentrations in bed sediments were compared. Also, water–sediment interactions were described in a dynamic way in both studies. Adsorption by bottom sediments was considered by other authors as well (Choi et al., 2013; Misumi et al., 2014; Higashi et al., 2015). All these papers agree on the fact that significant adsorption occurs in the first months after the accident, most of radionuclides staying on the sea bed once they were adsorbed, which may be indicative of a two-step kinetics. Later, a box model (POSEIDON-R) was used to perform a radiological assessment of the accident in the period 2011–2040 (Maderich et al., 2014a). This box model included not only adsorption to sediments, but also the transfer of radionuclides through the marine food web and subsequent doses to humans. The benthic food chain was included in this model (Bezhenar et al., 2016). The simulation results indicated a substantial contribution of the benthic food chain in the long-term transfer of ^{137}Cs from contaminated bottom sediments to marine organisms. ^{137}Cs levels in coastal biota in the area near FDNPP were reconstructed by Tateda et al. (2013) using a circulation model (Tsumune et al., 2012) to

calculate concentration in water and a dynamic 12 component biota model.

A radioecological model to estimate ^{137}Cs concentrations in phytoplankton and zooplankton populations, representing the lower levels of the pelagic trophic chain, was developed by Belharet et al. (2016). This model was coupled to a lower trophic level ecosystem model and an ocean circulation model to take into account the site-specific environmental conditions in the area. Results showed that the maximum concentrations in plankton after the accident were about 2 to 4 orders of magnitude higher than those observed before the accident, depending on the distance to Fukushima power plant.

Some models have finally been applied to other radionuclides, as ^{90}Sr (Periáñez et al., 2013a; Maderich et al., 2014b). The same coastal model commented above for ^{137}Cs was used in the first case. In the case of Maderich's work, POSEIDON-R model was again applied. Some preliminary simulations for plutonium were presented by Periáñez et al. (2013b).

Some exercises comparing model performances when applied to simulate the transport of Fukushima releases in the Pacific Ocean have been carried out, as for instance in Masumoto et al. (2012). These authors concluded that most of the discrepancies between the five participating models were due to the different calculated current fields in the coastal waters of Japan, off Fukushima, which led to different radionuclide distributions. Differences in current fields were caused by the different ocean models and dispersion model settings used by the research groups. However, a systematic assessment aimed at investigating the reasons of differences was not carried out.

The Science Council of Japan (SCJ, 2014) carried out a similar intercomparison study, with eleven models involved. Again, significant differences between models were found. Models were different in concept (Eulerian vs. Lagrangian), with different setting and even different source terms. Thus, it was concluded that a simple comparison is not straightforward and that detailed systematic comparison studies, such as ones that use the same radionuclide forcing with different models and/or the same model with different forcing scenarios, were required. This intercomparison exercise was carried out in the frame of IAEA MODARIA program (Periáñez et al., 2015a). Different dispersion models were compared, using both different and identical circulation fields. Simulations with identical parameters (like diffusion coefficients for instance) were also carried out. It was found that the main source of discrepancy between different dispersion models was due to the different circulation fields. Model/model and model/measurement comparisons for both the dissolved phase and bed sediments were carried out in this study. Alternatively, the same dispersion model forced with different circulation fields has been tested as well, although water/sediment interactions were not included in this study (Kawamura et al., 2017).

The most recent model intercomparison exercise was made in the frame of IAEA MODARIA-II program (Periáñez et al., 2019). This recent program is highlighting the relevance for the international scientific community of the numerical modelling of radionuclide transport in the marine environment topic. Simulations extended over larger spatio-temporal scales (two years after March 2011 and the whole North Pacific Ocean) and models included water/sediment interactions and biota uptake models integrated within the marine transport models. In general, models agreed in predicting areas in the ocean which were affected by FDNPP releases (direct and/or atmospheric deposition) and regions which were not. Also, predicted concentrations in the different compartments (three water layers, bed sediment and pelagic biota species) were within the same order of magnitude in most cases.

5. Problems and challenges

Sources of uncertainty in marine dispersion models are discussed in this section, together with the implications of these uncertainties when models are used for emergency response. Finally, advantages and disadvantages of each modelling approach and situations for which each model type is best suited are discussed.

5.1. Uncertainties in model parameters

If the model is applied to a perfectly conservative radionuclide (remaining dissolved, without any interactions with sediments) the only involved parameters are the horizontal and vertical diffusion coefficients. Since turbulence is still an open problem in physics, different schemes and approaches are used to evaluate the diffusion coefficients, as already mentioned in Section 2. These different approaches may lead to different model results.

Nevertheless, the situation is even more complex for non-conservative radionuclides. A number of parameters are required in this case, such as kinetic rates, particle sizes, density and thickness of the sediment, etc. These parameters are site-specific and information about them is generally scarce. Thus, estimated values have to be used in many cases. Kinetic modelling requires more parameters in comparison with simpler approaches based on sediment distribution coefficients (k_d) and concentration ratios (CR) for biota (food or wildlife). At the same time, these models are more flexible and less site-dependent, as demonstrated by the application of a dynamic food web model to the North Western Pacific and the Baltic Sea (Bezhenar et al., 2016). However, most dynamic biota models were developed and calibrated for several radionuclides only, whereas model parameters (e.g. transfer coefficients through food and water, and biological half-lifes) should be determined for each radionuclide and organism from experiment data, which often are unavailable. An unsolved issue in these models is also the representation of biota migration through areas with heterogeneous contamination levels. Examples are fish migration along the Japan shelf and trans-Pacific transport of Fukushima-derived radionuclide by bluefin tuna (Madigan et al., 2012). Several random movement models were developed in the frame of the diffusion approach (Monte, 2002; Maderich et al., 2014a).

Due to uncertainties in model parameters, sensitivity analysis should be an obligatory component of dispersion modelling. Essentially (Uusitalo et al., 2015), sensitivity analysis consists of evaluating how the model output would behave if some input data were changed within their reasonable range or within an assigned a probability distribution. The simplest way is to alter input values and/or parameters of the model and study the subsequent changes in model output (e.g. Bezhenar et al., 2016a). Monte Carlo methods may also be used: model input parameters are drawn randomly from their probability distribution and the resulting set of model outputs can be seen as a random sample of the distribution of the output of interest (see examples in e.g. Periáñez, 2004).

Probability density functions are now being provided for some parameters, like the k_d, in order to be able to take into account parameter variability in uncertainty analysis of transport and risk assessment models (Ciffroy et al., 2009; Boyer et al., 2018). This also allows carrying out Monte Carlo sensitivity analysis as mentioned above.

5.2. Uncertainties in model numerics

A second source of uncertainty is due to the numerical solution of the equations. Different procedures may be used, but finite differences (Periáñez, 2005a) is the most commonly applied method. A discretization is required to transform a derivative to an algebraic expression. Discretization in time is always required (in box, Eulerian and Lagrangian models). With respect to spatial discretization, even in Lagrangian models a discretization is required when concentrations are derived from the number of particles per water volume unit, resulting in averaging of quantities. A discretization always implies averaging magnitudes. Since averaging leads to errors, a numerical solution is only an approximation to the exact solution (rounding errors, truncation errors etc appear).

Numerical diffusion (see for instance Kowalik and Murty, 1993, for mathematical definition) is an artificial smoothing of concentration gradients produced by the numerical scheme in Eulerian models (one

of the advantages of Lagrangian models is that they do not introduce this numerical diffusion). In particular, some numerical experiments with a Fukushima Eulerian advection–diffusion model were performed and effective diffusivities along time were evaluated (Dietze and Kriest, 2012). High initial values were attributed to numerical diffusion in the initial release phase, due to the high concentrations gradients. After, effective diffusivities decreased and finally increased again once radionuclides entered the high-eddying Kuroshio system.

Effective diffusivity increases with the model cell size, since a larger cell size implies that larger eddies are not solved, and thus have to be accounted for as sub-grid mixing. Eddy and non-resolving eddy models were compared in Behrens et al. (2012) for instance. They found that the non-eddy model underestimated lateral dispersion of the Fukushima plume in the Pacific. Eddy and non-resolving eddy models were also compared in Simonsen et al. (2017) for the North Atlantic Ocean, where transport of ^{99}Tc released from Sellafield reprocessing plant was simulated.

Constructing an accurate Lagrangian numerical scheme for simultaneous simulation of advection and diffusion is also a challenging problem. In the general case of diffusion in a complex flow field, the centre of mass of the particle distribution may not follow the streamlines of the flow field, whereas the random distribution of particle locations due to diffusion can depend on the flow field.

Finally, the radionuclide release area size has to be considered. In Eulerian models, radionuclides are homogeneously distributed into the release cell where the accident occurs; this would be the initial patch minimum size. Thus, the initial patch size depends on the model spatial resolution. In contrast, a real point source can be used in a Lagrangian model.

5.3. Uncertainties in water circulation models

Existing state-of-the-art marine dispersion models are robust tools in our opinion, providing consistent results (Periáñez et al., 2016b). However there are problems with the predictions of hydrodynamic models in energetic regions characterized by strong current variability, like Fukushima waters and the North Western Pacific region, characterized by the very strong and fluctuating Kuroshio current and its extension (Masumoto et al., 2012) -a map of currents may be seen in Fig. 4. In the frame of IAEA MODARIA program it was shown that the energetics of the considered system (magnitude and variability of currents) control the agreement between different dispersion models (Periáñez et al., 2016b). Good agreement could be achieved between models of very different type in environments characterized by weak currents. However, even similar models led to significantly different results in highly dynamic systems characterized by strong and variable currents. Two marine environments were studied: a highly dynamic system (Fukushima coastal waters) and a semi-enclosed basin (Baltic Sea). The models applied to the Baltic Sea included two box models, a full three dimensional model including water and ice thermodynamics and a depth-averaged two dimensional model forced with mean annual winds. Thus, very different approaches were used (details may be seen in Periáñez et al., 2015b). In the case of the Baltic Sea results of models were in good agreement despite of the different approaches and simplifications applied by models. On the contrary, in the case of FDNPP accident, even similar hydrodynamic models led to different current fields which, in turn, led to very different radionuclide dispersion patterns. Given the intensity and variability of currents in this area, as well as the presence of unsteady eddies due to the instability of currents, small differences in the hydrodynamics may produce different dispersion patterns. These differences tend to be amplified with time. For highly dynamic environments, the dispersion model output is extremely sensitive to the ocean model which is used to obtain circulation. Simulation results from a single oceanic advection–diffusion model and multiple oceanic general circulation models were compared in Kawamura et al. (2017), arriving at similar conclusions. Thus, we may state that the ocean model should be selected with great care and after a detailed comparison with local measurements of currents.

In this sense, the spatio/temporal scale of interest is also relevant. When simulating the transport of Fukushima releases in the Pacific Ocean, it was found that the initial current field is relevant for ^{137}Cs spreading in the first months after the accident. However, this relevance fades in the long-term (Behrens et al., 2012).

Additionally, it can be pointed out that, if measurements of radioactivity concentration are available, then assimilation of observations is a powerful tool to improve the predictive capabilities of radionuclide transport models (Yuschenko et al., 2005).

5.4. Emergency modelling

One of the main applications of marine dispersion models is their use as predictive tools to assess radionuclide concentrations after an accident in order to support decision making. Three stages after a nuclear accident in a coastal facility were defined by our group (Periáñez et al., 2016b): emergency phase, post-emergency phase and long term phase. They are characterized by increasing spatio-temporal scales, and each one requires a specific kind of model to give response to decision makers. It must be noted that an ideal model which could be applied for all spatio-temporal scales does not exist. Of course physical–chemical processes are the same, but depending on the scales in which we are interested the numerical realization and involved simplifications are different. This leads to the different modelling approaches defined in Section 3.1.

The prediction of radioactivity dispersion in the emergency phase (days–weeks) should be carried out using robust models and numerical tools. Two approaches can be used. The first consists of the use of local forecasts of marine circulation linked to the transport and biota models (e.g. Duffa et al., 2016) when this local model is operational. However, such local forecasts are often unavailable or result in large computational burdens. Therefore, in several decision-support systems (DSS) another approach is used: the forecast of marine circulation from operational ocean models. In the JRODOS DSS (Maderich et al., 2016) the dispersion of radioactivity was calculated using velocity fields from operational ocean models (Copernicus Marine Environment Monitoring Service[15]). It covers the European seas with 3–6 km resolution and the global ocean with resolution about 10 km. A similar approach was used by Kobayashi et al. (2017): the Short-Term Emergency Assessment system of Marine Environmental Radioactivity (STEAMER) was developed to predict radionuclide migration for a nuclear accident in the ocean around Japan at 8 and 30 days using operational ocean models.

An interesting modelling study was carried out by Kauler et al. (2016). The model was used to determine the areas where a nuclear accident (involving a nuclear powered vessel) would affect a sensitive point (in this case a fishery zone). Thus, the model was not used to deal with an emergency; instead it was used as a prevention tool since traffic of nuclear vessels could be banned across given areas.

Although global ocean models produce realistic pictures of the general circulation in the ocean, their outputs differ in the local scale in dynamic environments. This may be, at least in part, attributed to their relatively coarse spatial resolution. The problem is then to assess the best way to develop a reliable model to support decision-making after an emergency. A multi-model approach, as described by Monte et al. (2008), may be of interest when environmental processes are complex. Through this approach, the conclusions that obtain the greatest degree of consensus among modellers are made evident and the aspects that are subject to dispute and which should therefore be handled carefully also become clear. Nevertheless, a multi model application is not the perfect choice when an emergency is involved and a rapid response

[15] http://marine.copernicus.eu/web/69-interactive-catalogue.php.

from the model is required. In any case, it may help to select the most adequate characterization of water circulation to be used in the operational dispersion model in the development stage. However, there may be cases when an "outlier model" is closer to observations than the "consensus". An example is provided in IAEA (1995) -pages 26–28. Care should be taken with these cases. In our opinion, site specific tools should be carefully developed, tested and then made available for any marine area potentially exposed to a radionuclide release. In other words, we cannot be a priori confident in generic models which import ocean forecasts of currents if a highly dynamic environment is involved.

The source term information is an important but not solved issue in emergency modelling. Usually, it cannot be directly obtained and it is necessary to solve the inverse problem of source determination using marine monitoring data, as already commented. If these data are not available in the emergency phase, then an automatic prediction mode can be used assuming a unit release rate directly to the ocean and/or in the atmosphere (Kobayashi et al., 2017). These predictions can be useful for prohibiting fishing and sailing over given sea areas and setting up an emergency ocean monitoring corresponding to a realistic marine pollution area. At the post-emergency phase an inverse modelling, as described in Section 4, could be used.

5.5. Selecting a radionuclide transport model

The most significant processes governing the transport of radionuclides in the marine environment are advection by currents, turbulent mixing, water/sediment interactions and biota uptake. The different formulations and numerical treatments of these processes lead to the different modelling approaches which have been discussed in the paper. Thus, advection and mixing may be solved using a box, an Eulerian or a Lagrangian model. Water/sediment interaction and biota uptake may be described, essentially, using an equilibrium or a dynamic model. But, as commented before there is not a model which can be applied to all situations, i.e., to all spatio-temporal scales.

The basic assumptions in box models (uniform and instantaneous mixing of radionuclides within each box) make these models well suited to long-term assessments over large spatial scales. Thus, they are useful tools in the long-term phase of an emergency (Section 5.4), as well as for the environmental assessment of chronic releases from nuclear facilities. In addition, involved mathematics are relatively simple and these models are easy to program or to adapt to specific cases. Finally, detailed water circulation patterns are not required since the only needed parameterization is water fluxes between boxes.

Eulerian and Lagrangian models make use of detailed water circulation fields, changing in time and space. Thus, these models provide distributions of radionuclides in space and time, which make them appropriate for the emergency and post-emergency phases of an accident (Section 5.4). The mathematical formulation and solution on these models are more complex than in box models and therefore they are more difficult to program or to customize. Moreover, these models require the mentioned water circulation fields as input data; information which is not always easy to obtain and accurate enough, as already discussed (Section 5.3).

Lagrangian models are specially well suited to the emergency phase of an accident, since they do not introduce numerical diffusion (Section 5.2) and thus can handle the very high concentration gradients between contaminated and clean water which would be expected after an acute radionuclide release into the sea. In addition, computation can be significantly faster than in Eulerian models when the contaminated area initially is a small part of the whole computational domain and if the number of particles in the simulation is reasonable (typically a few tens of thousands). This is another advantage to be considered in emergency modelling, when a fast response must be forwarded to decision-makers. Finally, a real point source may be defined in Lagrangian models (Section 5.2); in Eulerian models the initial patch size is defined by the grid spatial resolution. Consequently, lower peak concentrations are expected from Eulerian models.

Eulerian models present the advantage, over Lagrangian ones, that the inclusion of additional processes is simpler since only the addition of new terms to the transport differential equations is required. For instance, including multi-stage water/sediment interactions and/or redox reactions can be done in an easier way in Eulerian models. Also, the number of particles required in a Lagrangian simulation increases as the number of sub-compartments (i.e., different oxidation states in water, different speciation states in sediments) increases. Consequently, an Eulerian model may be more efficient than a Lagrangian one if these processes are the main focus of the simulations. Eulerian models are also more appropriate for simulating spatially extended radionuclide sources (for instance due to atmospheric fallout) over large areas since many particles would be required in a Lagrangian simulation, which would be computationally more expensive.

Regarding water/sediment interactions and biota uptake, they can be described using equilibrium or dynamic models, as commented. Again, each approach has advantages and disadvantages. The obvious advantage of equilibrium models is their simplicity and the fact that few parameters are required; only water/sediment distribution coefficients and biota concentration ratios. The equilibrium assumption implies that these models may be applied in long-term assessments over wide spatial scales; thus they are well suited to be included within box transport models. In contrast, these models should not be used for emergency purposes and for assessments of chronic releases near the radionuclide source, since equilibrium is not achieved. Dynamic models should be used in these cases; but the main difficulty with these models, in addition to their more complicated formulation, is that a significant number of parameters are required. These parameters are radionuclide and site specific; information about them is generally scarce and only tentative values can be used in many cases.

Thus, all model types are useful tools for assessments of marine radionuclide transport, provided that each model is applied to suitable spatio-temporal scales. A model which can be applied in all situations does not exist because of practical computational limitations.

6. Conclusions

Significant advances in techniques for simulating the transport of radionuclides in the marine environment have taken place in the last years. Currently, most models do not only solve the transport in the dissolved phase (advection and turbulent mixing), but also include interactions with sediments and biota. There is a general trend consisting of describing sediment processes in a dynamic way, by means of kinetic transfer coefficients; instead of using an equilibrium approach based upon distribution coefficients, k_ds, since an equilibrium approach leads to significant errors in the near field, both in the cases of chronic and accidental releases. More detailed processes, such as redox reactions (which are relevant in the case of plutonium) may be also described in dynamic models. The most recent efforts are directed to include biological uptake models within the marine dispersion model. It has been found that biological uptake is also better described using dynamic than equilibrium models.

Generally speaking, three kinds of models exist: box models, Eulerian and Lagrangian models. Models also differ in structure (from one-dimensional to full three-dimensional models) and resolution, i.e., the same physico-chemical processes are described in different ways. A universal model which is able to describe all the spatial and temporal scales in a marine dispersion problem does not exist because of practical computational limitations. Thus, it is essential to have the different implementations mentioned in this paper.

Recently, it was found that the main source of uncertainty in marine transport models is due to water circulation in highly dynamic environments characterized by strong and variable currents. Thus, marine transport models are robust tools, providing consistent results,

but in energetic regions characterized by strong current variability, like Fukushima waters and north Pacific, differences between model outputs appear. Although several hydrodynamic models may be providing a coherent general picture of water circulation in the area of interest, small differences in current magnitude and/or direction in the area of release result in different initial transport pathways. Even small differences are then amplified in time. For this reason a careful selection of the ocean model is needed and should be done after a detailed comparison with local measurements of currents. In this sense, local forecasts of marine circulation should be used for emergency modelling if they are available.

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References

Abril, J.M., 1998. Basic microscopic theory of the distribution, transfer and uptake kinetics of dissolved radionuclides by suspended particulate matter. Part 1: Theory development. J. Environ. Radioact. 41, 307–324.

Abril, J.M., Abdel-Aal, M.M., Al-Gamal, S.A., Abdel-Hay, F.A., Zahar, H.M., 2000. Marine radioactivity studies in the Suez Canal, Part 2: Field experiments and a modelling study of dispersion. Estuar. Coast. Shelf Sci. 50, 503–514.

Abril, J.M., García-León, M., 1993. A 2d 4-phases marine dispersion model for nonconservative radionuclides 1. Conceptual and computational model. J. Environ. Radioact. 20 (2), 71–88.

Aldridge, J.N., 1998. CSERAM: A model for prediction of marine radionuclide transport in both particulate and dissolved phases. Radiat. Prot. Dosim. 75, 99–103.

Aldridge, J.N., Kershaw, P., Brown, J., McCubbin, D., Leonard, K.S., Young, E.F., 2003. Transport of plutonium (Pu-239/240) and caesium (Cs-137) in the Irish Sea: comparison between observations and results from sediment and contaminant transport modelling. Cont. Shelf Res. 23, 869–899.

Barros, H., Abril, J.M., 2004. Experimental and modelling study on the uptake and desorption kinetics of ^{133}Ba by suspended estuarine sediments from southern Spain. Water Res. 38, 749–755.

Behrens, E., Schwarzkopf, F.U., Lubbecke, J., Boning, C.W., 2012. Model simulations on the long-term dispersal of ^{137}Cs released into the Pacific Ocean off Fukushima. Environ. Res. Lett. 7, 034000, 10 pp..

Belharet, M., Estournel, C., Charmasson, S., 2016. Ecosystem model-based approach for modeling the dynamics of ^{137}Cs transfer to marine plankton populations: application to the western North Pacific Ocean after the Fukushima nuclear power plant accident. Biogeosciences 13, 499–516.

Benkdad, A., Laissaoui, A., El Bari, H., Benmansour, M., Ibn-Majah, M., 2008. Partitioning of radiostrontium in marine aqueous suspensions: laboratory experiments and modeling studies. J. Environ. Radioact. 99, 748–756.

Bezhenar, R., Jung, K.T., Maderich, V., de With, G., Willemsen, S., Qiao, F., 2016. Transfer of radiocaesium from contaminated bottom sediments to marine organisms through benthic food chain in post-Fukushima and post-Chernobyl periods. Biogeosciences 13, 3021–3034.

Bleck, R., 2001. An oceanic general circulation model framed in hybrid isopycnic-Cartesian coordinates. Ocean Model. 4, 55–88.

Blumberg, A.F., Mellor, G.L., 1987. A description of a three-dimensional coastal ocean circulation model. In: Heaps, N. (Ed.), Three-Dimensional Coastal Ocean Models. American Geophysical Union, p. 208.

Bailly du Bois, P., Dumas, P., 2005. Fast hydrodynamic model for medium and long term dispersion in seawater in the English Channel and southern North Sea, qualitative and quantitative validation by radionuclide tracers. Ocean Model. 9, 169–210.

Bailly du Bois, P., Garreau, P., Laguionie, P., Korsakissok, I., 2014. Comparison between modelling and measurement of marine dispersion, environmental half-time and 137-Cs inventories after the Fukushima Daiichi accident. Ocean Dyn. 64, 361–383.

Bailly du Bois, P., Laguionie, P., Boust, D., Korsakissok, I., Didier, D., Fiéve, B., 2012. Estimation of marine source-term following Fukushima Dai-ichi accident. J. Environ. Radioact. 114, 2–9.

Børretzen, P., Salbu, B., 2000. Estimation of apparent rate coefficients for radionuclides interacting with marine sediments from Novaya Zemlya. Sci. Total Environ. 262, 91–102.

Børretzen, P., Salbu, B., 2002. Fixation of Cs to marine sediments estimated by a stochastic modelling approach. J. Environ. Radioact. 61, 1–20.

Boyer, P., Wells, C., Howard, B., 2018. Extended k_d distributions for freshwater environment. J. Environ. Radioact. 192, 128–142.

Breton, M., Salomon, J.C., 1995. A 2d long-term advection–dispersion model for the Channel and southern North-Sea. A: Validation through comparison with artificial radionuclides. J. Mar. Syst. 6, 495–513.

Buesseler, K., Dai, M., Aoyama, M., Benitez-Nelson, C., Charmasson, S., Higley, K., Maderich, V., Masque, P., Morris, P.J., Oughton, D., Smith, J.N., 2017. Fukushima Daiichi-derived radionuclides in the Ocean: transport, fate, and impacts. Annu. Rev. Mar. Sci. 9, 173–203.

Carroll, J., Boisson, F., Fowler, S.W., Teyssie, J.L., 1997. Radionuclide adsorption to sediments from nuclear waste dumping sites in the Kara Sea. Mar. Pollut. Bull. 35, 296–304.

Carroll, J., Boisson, F., Teyssie, J.L., King, S.E., Krosshavn, M., Carroll, M.L., Fowler, S.W., Povinec, P.P., Baxter, M.S., 1999. Distribution coefficients (k_ds) for use in risk assessment models of the Kara Sea. Appl. Radiat. Isot. 51, 121–129.

Carvalho, F.P., 2018. Radionuclide concentration processes in marine organisms: A comprehensive review. J. Environ. Radioact. 186, 124–130.

CEC, 1990. The radiological exposure of the population of the European Community from radioactivity in North European marine waters. In: Project MARINA, Commission of the European Communities. EUR, Bruxelles, p. 12483.

Cetina, M., Rajar, R., Povinec, P., 2000. Modelling of circulation and dispersion of radioactive pollutants in the Japan Sea. Oceanol. Acta 23, 819–836.

Chassignet, E.P., Verron, J., 2006. Ocean Weather Forecasting an Integrated View of Oceanography. Springer, The Netherlands.

Choi, Y., Kida, S., Takahashi, K., 2013. The impact of oceanic circulation and phase transfer on the dispersion of radionuclides released from the Fukushima Dai-ichi Nuclear Power Plant. Biogeosciences 10, 4911–4925.

Ciffroy, P., Durrieu, G., Garnier, J.M., 2009. Probabilistic distribution coefficients (Kds) in freshwater for radioisotopes of Ag, Am, Ba, Be, Ce, Co, Cs, I, Mn, Pu, Ra, Ru, Sb, Sr and Th - Implications for uncertainty analysis of models simulating the transport of radionuclides in rivers. J. Environ. Radioact. 100, 785–794.

Ciffroy, P., Garnier, J.M., Pham, M.K., 2001. Kinetics of the adsorption and desorption of radionuclides of Co, Mn, Cs, Fe, Ag and Cd in freshwater systems: experimental and modelling approaches. J. Environ. Radioact. 55, 71–91.

Cushman-Roisin, B., Beckers, J.M., 2011. Introduction To Geophysical Fluid Dynamics. Elsevier.

Dietze, H., Kriest, I., 2012.^{137}Cs off Fukushima Dai-ichi, Japan. Model based estimates of dilution and fate. Ocean Science 8, 319–332.

Duffa, C., Bailly du Bois, P., Caillaud, M., Charmasson, S., Couvez, C., Didier, D., Dumas, F., Fievet, B., Morillon, M., Renaud, P., Thebault, H., 2016. Development of emergency response tools for accidental radiological contamination of French coastal areas. J. Environ. Radioact. 151, 487–494.

Duursma, E.K., Carroll, J., 1996. Environmental Compartments. Springer, Germany.

Dvorzhak, A., Puras, C., Montero, M., Mora, J.C., 2012. Spanish experience on modeling of environmental radioactive contamination due to Fukushima Daiichi NPP accident using JRODOS. Environ. Sci. Technol. 46, 11887–11895.

EC, 2002. MARINA II. Update of the MARINA project on the radiological exposure of the European Community from radioactivity in north European marine waters. In: Radiation Protection, vol. 132. European Commission, Luxembourg.

Eisma, D., 1993. Suspended Matter in the Aquatic Environment. Springer-Verlag, Berlin.

El-Mrabet, R., Abril, J.M., Manjón, G., García-Tenorio, R., 2001. Experimental and modelling study of plutonium uptake by suspended matter in aquatic environments from southern Spain. Water Res. 35, 4184–4190.

Enting, I., 2002. Inverse Problems in Atmospheric Constituent Transport. In: Cambridge Atmospheric and Space Science Series, Cambridge University Press, Cambridge, UK.

Estournel, C., Bosc, E., Bocquet, M., Ulses, C., Marselaix, P., Winiarek, V., Osvath, I., Nguyen, C., Duhaut, T., Lyard, F., Michaud, H., Auclair, F., 2012. Assessment of the amount of 137-Cs released into the Pacific Ocean after the Fukushima accident and analysis of its dispersion in Japanese coastal waters. J. Geophys. Res. 117 (C1014).

Fox-Kemper, B., Adcroft, A., Böning, C.W., Chassignet, E.P., Curchitser, E., Danabasoglu, G., Eden, C., England, M.H., Gerdes, R., Greatbatch, R.J., Griffies, S.M., Hallberg, R.W., Hanert, E., Heimbach, P., Hewitt, H.T., Hill, C.N., Komuro, Y., Legg, S., Le Sommer, J., Masina, S., Marsland, S.J., Penny, S.G., Qiao, F., Ringler, T.D., Treguier, A.M., Tsujino, H., Uotila, P., Yeager, S.G., 2019. Challenges and prospects in ocean circulation models. Front. Mar. Sci. 6 (65), http://dx.doi.org/10.3389/fmars.2019.00065.

Gao, Y., Drange, H., Bentsen, M., Johannessen, M., 2004. Simulating transport of non-Chernobyl ^{137}Cs and ^{90}Sr in the North Atlantic-Arctic region. J. Environ. Radioact. 71, 1–16.

Garnier-Laplace, J., Adam, C., Lathuilliere, T., Baudin, J., Clabaut, M., 2000. A simple fish physiological model for radioecologists exemplified for 54Mn direct transfer and rainbow trout (Oncorhynchus mykiss W). J. Environ. Radioact. 49, 35–53.

Gleizon, P., McDonald, P., 2010. Modelling radioactivity in the Irish Sea: from discharge to dose. J. Environ. Radioact. 101, 403–413.

Glover, D.M., Jenkins, W.J., Doney, S.C., 2011. Modelling Methods for Marine Science. Cambridge University Press, UK.

Goshawk, J.A., Clarke, S., Smith, C.N., McDonald, P., 2003. MEAD (Part I): A mathematical model of the long-term dispersion of radioactivity in shelf sea environments. J. Environ. Radioact. 68, 115–135.

Haidvogel, D.B., Arango, H.G., Hedstrom, K., Beckmann, A., Malanotte-Rizzoli, P., Shchepetkin, A.F., 2000. Model evaluation experiments in the North Atlantic Basin: Simulations in nonlinear terrain-following coordinates. Dyn. Atmos. Oceans 32, 239–281.

Harms, I.H., 1997. Modelling the dispersion of Cs-137 and Pu-239 released from dumped waste in the Kara Sea. J. Mar. Syst. 13, 1–19.

Harms, I.H., Karcher, M.J., Burchard, H., 2003. Modelling radioactivity in the marine environment: the application of hydrodynamic circulation models for simulating oceanic dispersion of radioactivity. In: Scott, E.M. (Ed.), Modelling Radioactivity in the Environment. Elsevier Science Ltd, Oxford, pp. 55–85.

Harms, I., Karcher, M.J., Dethleff, D., 2000. Modelling Siberian river runoff - implications for contaminant transport in the Arctic Ocean. J. Mar. Syst. 27, 95–115.

Hazell, D.R., England, M.H., 2003. Prediction of the fate of radioactive material in the South Pacific Ocean using a global high-resolution ocean model. J. Environ. Radioact. 65, 329–355.

Heling, R., Koziy, L., Bulgakov, V., 2002. On the dynamical uptake model developed for the uptake of radionuclides in marine organisms for the POSEIDON-R model system. Radioprotection 37 (C1), 833–838.

Herzfeld, M., Schmidt, M., Griffies, S.M., Liang, Z., 2011. Realistic test cases for limited area ocean modelling. Ocean Model. 37, 1–34.

Higashi, H., Morino, Y., Furuichi, N., Ohara, T., 2015. Ocean dynamic processes causing spatially heterogeneous distribution of sedimentary caesium-137 massively released from the Fukushima Daiichi Nuclear Power Plant. Biogeosciences 12, 7107–7128.

Honda, M., Aono, T., Aoyama, M., Hamajima, Y., Kawakami, H., Kitamura, M., Masumoto, Y., Miyazawa, Y., Takigawa, M., Saino, T., 2012. Dispersion of artificial caesium-134 and -137 in the western North Pacific one month after the Fukushima accident. Geochem. J. 46, 1–9.

Håkanson, L., 2005. A new general dynamic model predicting radionuclide concentrations and fluxes in coastal areas from readily accessible driving variables. J. Environ. Radioact. 78, 217–245.

Hunt, G.J., 2004. Radiological assessment of ocean radioactivity. In: Livingston, H. (Ed.), Marine Radioactivity. Elsevier, Amsterdam, pp. 205–236.

IAEA, 1995. Validation of models using Chernobyl fallout data from the Central Bohemia region of the Czech Republic. Scenario CB. First report of the VAMP Multiple Pathways Assessment Working Group. IAEA-TECDOC-795, Vienna.

IAEA, 2000. Modelling of the transfer of radiocaesium from deposition to lake ecosystems. Report of the VAMP Aquatic Working Group. IAEA-TECDOC-1143. Vienna.

IAEA, Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment. Technical Reports Series, vol. 422, Vienna.

Iosjpe, M., Brown, J., Strand, P., 2002. Modified approach for box modelling of radiological consequences from releases into marine environment. J. Environ. Radioact. 60 (1-2), 91–103.

Iosjpe, M., Karcher, M., Gwynn, J., Harms, I., Gerdes, R., Kauker, F., 2009. Improvement of the dose assessment tools on the basis of dispersion of the ^{99}Tc in the Nordic Seas and the Arctic Ocean. Radioprotection 44 (5), 531–536.

James, I.D., 2002. Modelling pollution dispersion, the ecosystem and water quality in coastal waters: a review. Environ. Model. Softw. 17, 363–385.

Jayne, S.R., Hogg, N.G., Waterman, S.N., Rainville, L., Donohue, K.A., Randolph Watts, D., Tracey, K.L., McClean, J.L., Maltrud, M.E., Qiu, B., Chen, S., 2009. The kuroshio extension and its recirculation gyres. Deep Sea Res. I 56 (12), 2088–2099.

Johansson, H., Lindström, M., Håkanson, L., 2001. On the modelling of the particulate and dissolved fractions of substances in aquatic ecosystems - sedimentological and ecological interactions. Ecol. Model. 137, 225–240.

Kaeriyama, H., 2017. Oceanic dispersion of Fukushima-derived radioactive cesium: a review. Fisheries Oceanography 26 (2), 99–113.

Kampf, J., 2009. Ocean Modelling for Beginners. Springer-Verlag, Heidelberg.

Kampf, J., 2010. Advanced Ocean Modelling. Springer-Verlag, Heidelberg.

Kanda, J., 2013. Continuing ^{137}Cs release to the sea from the Fukushima Dai-ichi Nuclear Power Plant through 2012. Biogeosciences 10, 6107–6113.

Kauler, F., Kaminski, T., Karcher, M., Dowdall, M., Brown, J., Hosseini, A., Strand, P., 2016. Model analysis of worst place scenarios for nuclear accidents in the northern marine environment. Environ. Model. Softw. 77, 13–18.

Kawamura, H., Furuno, A., Kobayashi, T., In, T., Nakayama, T., Ishikawa, Y., Miyazawa, Y., Usui, N., 2017. Oceanic dispersion of Fukushima-derived Cs-137 simulated by multiple oceanic general circulation models. J. Environ. Radioact. 180, 36–58.

Kawamura, H., Kobayashi, T., Furuno, A., In, T., Ishikawa, Y., Nakayama, T., Shima, S., Awaji, T., 2011. Preliminary numerical experiments on oceanic dispersion of ^{131}I and ^{137}Cs discharged into the ocean because of the Fukushima Daiichi nuclear power plant disaster. J. Nucl. Sci. Technol. 48, 1349–1356.

Kawamura, H., Kobayashi, T., Furuno, A., Usui, N., Kamachi, M., 2014. Numerical simulation on the long-term variation of radioactive cesium concentration in the North Pacific due to the Fukushima disaster. J. Environ. Radioact. 136, 64–75.

Kobayashi, T., Kawamura, H., Fujii, K., Kamidaira, Y., 2017. Development of a short-term emergency assessment system of the marine environmental radioactivity around Japan. J. Nucl. Sci. Technol. 54, 609–616.

Kobayashi, T., Nagai, H., Chino, M., Kawamura, H., 2013. Source term estimation of atmospheric release due to the Fukushima Dai-ichi Nuclear Power Plant accident by atmospheric and oceanic dispersion simulations. J. Nucl. Sci. Technol. 50, 255–264.

Kobayashi, T., Otosaka, S., Togawa, O., Hayashi, K., 2007. Development of a non-conservative radionuclides dispersion model in the ocean and its application to surface cesium-137 dispersion in the Irish Sea. J. Nucl. Sci. Technol. 44 (2), 238–247.

Konovalenko, L., Bradshaw, C., Kumblad, L., Kautsky, U., 2014. Radionuclide transfer in marine coastal ecosystems, a modelling study using metabolic processes and site data. J. Environ. Radioact. 133, 48–59.

Kowalik, Z., Murty, T.S., 1993. Numerical Modelling of Ocean Dynamics. World Scientific, Singapore.

Koziy, L., Maderich, V., Margvelashvili, N., Zheleznyak, M., 1998. Three-dimensional model of radionuclide dispersion in the estuaries and shelf seas. Environ. Model. Softw. 13, 413–420.

Laissaoui, A., Abril, J.M., Periáñez, R., García-León, M., García-Montaño, E., 1998. Kinetic transfer coefficients for radionuclides in estuarine waters: reference values for ^{133}Ba and effects of salinity and suspended load concentration. J. Radioanal. Nucl. Chem. 237, 55–61.

Lepicard, S., Heling, R., Maderich, V., 2004. POSEIDON/RODOS model for radiological assessment of marine environment after accidental releases: application to coastal areas of the Baltic, Black and North seas. J. Environ. Radioact. 72 (1–2), 153–161.

Lepicard, S., Raffestin, D., Rancillac, F., 1998. Poseidon: a dispersion computer code for assessing radiological i mpacts in a European sea water environment. Radiat. Prot. Dosim. 75, 79–83.

Lick, W., 2008. Sediment and Contaminant Transport in Surface Waters. CRC Press.

Lynch, D.R., Greenberg, D.A., Bilgili, A., Mcgillicuddy, D.J., Manning, J.P., Aretx-abaleta, A.L., 2015. Particles in the Coastal Ocean. Theory and Applications. Cambridge University Press, NY.

Lyons, M.G., Bradley, S.B., Parker, T.G., 1998. Developments in the CUMBRIA model and its application to radiological assessment in the Irish Sea. Radiat. Prot. Dosim. 75, 91–97.

Maderich, V., 1998. Modelling of the Mediterranean system: changes under climate variations and man-made impact. Environ. Model. Softw. 13 (5–6), 405–412.

Maderich, V., Bezhenar, R., Heling, R., de With, G., Jung, K.T., Myoung, J.G., Cho, Y.K., Qiao, F., Robertson, L., 2014a. Regional long-term model of radioactivity dispersion and fate in the northwestern Pacific and adjacent seas: application to the Fukushima Dai-ichi accident. J. Environ. Radioact. 131, 4–18.

Maderich, V., Brovchenko, I., Dvorzhak, A., Koshebutsky, V., Periáñez, R., 2016. Integration of 3D model THREETOX in JRODOS, implementation studies and modelling of Fukushima scenarios. Radioprotection 51, S133–S135.

Maderich, V., Heling, R., Bezhenar, R., Brovchenko, I., Jenner, H., Koshebutskyy, V., Kuschan, A., K., Terletska, 2008. Development and application of 3D numerical model THREETOX to the prediction of cooling water transport and mixing in the inland and coastal waters. Hydrol. Process. 22, 1000–1013.

Maderich, V., Jung, K.T., Bezhenar, R., de With, G., Qiao, F., Casacuberta, N., Masqué, P., Kim, Y.H., 2014b. Dispersion and fate of ^{90}Sr in the Northwestern Pacific and adjacent seas: Global fallout and the Fukushima Dai-ichi accident. Sci. Total Environ. 494–495, 261–271.

Maderich, V., Jung, K.T., Brovchenko, I., Kim, K.O., 2017. Migration of radioactivity in multi-fraction sediments. Environ. Fluid Mech. 17, 1207–1231.

Madigan, D.J., Baumann, Z., Fisher, N.S., 2012. Pacific bluefin tuna transport Fukushima-derived radionuclides from Japan to California. Proc. Natl. Acad. Sci. 109, 9483–9486.

Margvelashvily, N., Maderich, V., Zheleznyak, M., 1997. THREETOX - computer code to simulate three-dimensional dispersion of radionuclides in homogeneous and stratified water bodies. Radiat. Prot. Dosim. 73, 177–180.

Masumoto, Y., Miyazawa, Y., Tsumune, D., Kobayashi, T., Estournel, C., Marsaleix, P., Lanerolle, L., Mehra, A., Garraffo, Z.D., 2012. Oceanic dispersion simulation of Cesium-137 from Fukushima Dai-ichi nuclear power plant. Elements 8, 207–212.

Masumoto, Y., Sasaki, H., Kagimoto, T., Komori, N., Ishida, A., Sasai, Y., Miyama, T., Motoi, T., Mitsudera, H., Takahashi, K., Sakuma, H., Yagamata, T., 2004. A fifty-year eddy-resolving simulation of the world ocean - preliminary outcomes of OFES (OGCM for the earth simulator). J. Earth Simul. 1, 36–56.

Miller, R.N., 2007. Numerical Modelling of Ocean Circulation. Cambridge University Press, Uk.

Min, B.I., Periáñez, R., Kim, I.G., Suh, K.S., 2013. Marine dispersion assessment of ^{137}Cs released from the Fukushima nuclear accident. Mar. Pollut. Bull. 72, 22–33.

Misumi, K., Tsumune, D., Tsubono, T., Tateda, Y., Aoyama, M., Kobayashi, T., Hirose, K., 2014. Factors controlling the spatiotemporal variation of 137Cs in seabed sediment off the Fukushima coast: implications from numerical simulations. J. Environ. Radioact. 136, 218–228.

Mitchell, P.I., Vives i Batlle, J., Downes, A.B., Condren, O.M., León-Vintró, L., Sánchez-Cabeza, J.A., 1995. Recent observations on the physico chemical speciation of plutonium in the Irish Sea and the western Mediterranean. Appl. Radiat. Isot. 46, 1175–1190.

Mitchell, P.I., Condren, O.M., Vintro, L.L., McMahon, C.A., 1999. Trends in plutonium, americium and radiocaesium accumulation and long-term bioavailability in the western Irish Sea mud basin. J. Environ. Radioact. 44, 223–251.

Miyazawa, Y., Masumoto, Y., Varlamov, S.M., Miyama, T., 2012. Transport simulation of the radionuclide from the shelf to the open ocean around Fukushima. Cont. Shelf Res. 50–51, 16–29.

Miyazawa, Y., Masumoto, Y., Varlamov, S.M., Miyama, T., Takigawa, M., Honda, M., Saino, T., 2013. Inverse estimation of source parameters of oceanic radioactivity dispersion models associated with the Fukushima accident. Biogeosciences 10, 2349–2363.

Monte, L., 2002. A methodology for modelling the contamination of moving organisms in water bodies with spatial and time dependent pollution levels. Ecol. Model. 158, 21–33.

Monte, L., 2011. Customisation of the decision support system MOIRA-PLUS for applications to the marine environment. J. Environ. Radioact. 102, 1112–1116.

Monte, L., Boyer, P., Brittain, J., Goutal, N., Heling, R., Kryshev, A., Kryshev, I., Laptev, G., Luck, M., Periáñez, R., Siclet, F., Zheleznyak, M., 2008. Testing models for predicting the behaviour of radionuclides in aquatic systems. Appl. Radiat. Isot. 66, 1736–1740.

Müller, P., von Storch, H., 2004. Computer Modelling in Atmospheric and Oceanic Sciences. Springer, Germany.

Nakano, H., Motoi, T., Hirose, K., Aoyama, M., 2010. Analysis of ^{137}Cs concentration in the Pacific using a Lagrangian approach. J. Geophys. Res. 115, C06015.

Nakano, M., Povinec, P., 2012. Long-term simulations of the ^{137}Cs dispersion from the Fukushima accident in the world ocean. J. Environ. Radioact. 111, 109–115.

Nielsen, S.P., 1995. A box model for North-East Atlantic coastal waters compared with radioactive tracers. J. Mar. Syst. 6, 545–560.

Nyffeler, U.P., Li, Y.H., Santschi, P.H., 1984. A kinetic approach to describe trace element distribution between particles and solution in natural aquatic systems. Geochim. Cosmochim. Acta 48, 1513–1522.

Onishi, Y., Trent, D.S., 1992. Turbulence modeling for deep ocean radionuclide disposal. Internat. J. Numer. Methods Fluids 15, 1059–1071.

Orre, S., Gao, Y., Drange, H., Nielsen, J.E., 2007. A reassessment of the dispersion properties of ^{99}Tc in the North Sea and the Norwegian Sea. J. Mar. Syst. 68, 24–38.

Oughton, D.H., Børretzen, P., Salbu, B., Tronstad, E., 1997. Mobilisation of ^{137}Cs and ^{90}Sr from sediments: potential sources to Arctic water. Sci. Total Environ. 202, 155–165.

Pacanowski, R.C., Philander, S.G.H., 1981. Parameterization of vertical mixing in numerical models of tropical oceans. J. Phys. Oceanogr. 11, 1443–1451.

Parzen, E., 1962. Stochastic Processes. Holden-Day, San Francisco.

Periáñez, R., 1999. Three-dimensional modelling of the tidal dispersion of non-conservative radionuclides in the marine environment. Application to 239, 240-Pu dispersion in the eastern Irish Sea. J. Mar. Syst. 22 (1), 37–51.

Periáñez, R., 2003a. Redissolution and long-term transport of radionuclides released from a contaminated sediment: a numerical modelling study. Estuar. Coast. Shelf Sci. 56, 5–14.

Periáñez, R., 2003b. Kinetic modelling of the dispersion of plutonium in the eastern Irish Sea: two approaches. J. Mar. Syst. 38, 259–275.

Periáñez, R., 2004. On the sensitivity of a marine dispersion model to parameters describing the transfers of radionuclides between the liquid and solid phases. J. Environ. Radioact. 73, 101–115.

Periáñez, R., 2005a. Modelling the Dispersion of Radionuclides in the Marine Environment: An Introduction. Springer, Berlin.

Periáñez, R., 2005b. GISPART: a numerical model to simulate the dispersion of contaminants in the strait of gibraltar. Environ. Model. Softw. 20, 797–802.

Periáñez, R., 2008. A modelling study on ^{137}Cs and ^{239,240}Pu behaviour in the Alborán Sea, western Mediterranean. J. Environ. Radioact. 99, 694–715.

Periáñez, R., Bezhenar, R., Brovchenko, I., Duffa, C., Jung, K.T., Kobayashi, T., Lamego, F., Maderich, V., Min, B.I., Nies, H., Osvath, I., Outola, I., Psaltaki, M., Suh, K.S., de With, G., 2016b. Modelling of marine radionuclide dispersion in IAEA MODARIA program: lessons learnt from the Baltic Sea and Fukushima scenarios. Sci. Total Environ. 569/570, 594–602.

Periáñez, R., Bezhenar, R., Brovchenko, I., Jung, K.T., Kamidara, Y., Kim, K.O., Kobayashi, T., Liptak, L., Maderich, V., Min, B.I., Suh, K.S., 2019. Fukushima ^{137}Cs releases dispersion modelling over the Pacific Ocean. Comparisons of models with water, sediment and biota data. J. Environ. Radioact. 198, 50–63.

Periáñez, R., Bezhenar, R., Brovchenko, I., Jung, K.T., Kim, K.O., Maderich, V., 2018. The marine k_d and water/sediment interaction problem. J. Environ. Radioact. 192, 635–647.

Periáñez, R., Bezhenar, R., Iosjpe, M., Maderich, V., Nies, H., Osvath, I., Outola, I., de With, G., 2015b. A comparison of marine radionuclide dispersion models for the Baltic Sea in the frame of IAEA MODARIA program. J. Environ. Radioact. 139, 66–77.

Periáñez, R., Bezhenar, R., Min, Byung-Il, Duffa, C., Jung, K., Kobayashi, T., Suh, Kyung-Suk, Lamego, F., Maderich, V., Nies, H., Osvath, I., Psaltaki, M., 2015a. A new comparison of marine dispersion model performances for Fukushima releases in the frame of IAEA MODARIA program. J. Environ. Radioact. 150, 247–269.

Periáñez, R., Casas-Ruíz, M., Bolívar, J.P., 2013c. Tidal circulation, sediment and pollutant transport in Cádiz Bay (SW Spain): a modelling study. Ocean Eng. 69, 60–69.

Periáñez, R., Elliot, A.J., 2002. A particle-tracking method for simulating the dispersion of non-conservative radionuclides in coastal waters. J. Environ. Radioact. 58, 13–33.

Periáñez, R., Suh, Kyung-Suk, Min, Byung-Il, 2012. Local scale marine modelling of fukushima releases. Assessment of water and sediment contamination and sensitivity to water circulation description. Mar. Pollut. Bull. 64, 2333–2339.

Periáñez, R., Suh, Kyung-Suk, Min, Byung-Il, 2013b. Should we measure plutonium concentrations in marine sediments near Fukushima? J. Radioanal. Nucl. Chem. 298, 635–638.

Periáñez, R., Suh, Kyung-Suk, Min, Byung-Il, 2016a. The behaviour of ^{137}Cs in the North Atlantic Ocean assessed from numerical modelling: releases from nuclear fuel reprocessing factories, redissolution from contaminated sediments and leakage from dumped nuclear wastes. Mar. Pollut. Bull. 113, 343–361.

Periáñez, R., Suh, Kyung-Suk, Min, Byung-Il Min, Casacuberta, N., Masqué, P., 2013a. Numerical modelling of the releases of ^{90}Sr from Fukushima to the ocean: an evaluation of the source term. Environ. Sci. Technol. 47, 12305–12313.

Prandle, D., 1984. A modelling study of the mixing of ^{137}Cs in the seas of the European Continental Shelf. Philos. Trans. R. Soc. Lond. Ser. A 310, 407–436.

Preller, R., Cheng, A., 1999. Modeling the transport of radioactive contaminants in the Arctic. Mar. Pollut. Bull. 38, 71–91.

Proehl, J.A., Lynch, D.R., McGuillicuddy, D.J., Ledwell, J.R., 2005. Modeling turbulent dispersion on the North Flank of Georges Bank using Lagrangian particle methods. Cont. Shelf Res. 25, 875–900.

Protter, P.E., 2004. Stochastic Integration and Differential Equations, second ed. Springer, ISBN: 3-540-00313-4.

Roland, A., Zhang, Y.J., Wang, H.V., Meng, Y., Teng, Y.C., Maderich, V., Brovchenko, I., Dutour-Sikiric, M., Zanke, U., 2012. A fully coupled 3D wave–current interaction model on unstructured grids. J. Geophys. Res. 117, C00J33. http://dx.doi.org/10.1029/2012JC007952.

Rossi, V., Sebille, E., Gupta, A.E., Garçon, V., England, .M.H., 2013. Multi-decadal projections of surface and interior pathways of the Fukushima Cesium-137 radioactive plume. Deep Sea Res. I 80, 37–46.

Rossi, V., Sebille, E., Gupta, A.E., Garçon, V., England, M.H., 2014. Corrigendum to Multi-decadal projections of surface and interior pathways of the Fukushima Cesium-137 radioactive plume. Deep Sea Res. I 93, 162–164.

Rypina, I.I., Jayne, S.R., Yoshida, S., Macdonald, A.M., Buesseler, K., 2014. Drifter-based estimate of the 5 year dispersal of Fukushima-derived radionuclides. J. Geophys. Res. Oceans 119, 8177–8193.

Rypina, I.I., Jayne, S.R., Yoshida, S., Macdonald, A.M., Douglass, E., Buesseler, K., 2013. Short-term dispersal of Fukushima-derived radionuclides off Japan: modeling efforts and model-data intercomparison. Biogeosciences 10, 4973–4990.

Sánchez-Cabeza, J.A., Ortega, M., Merino, J., Masqué, P., 2002. Long-term box modelling of ^{137}Cs in the Mediterranean Sea. J. Mar. Syst. 33–34, 457–472.

Sanial, V., Buesseler, K.O., Charette, M.A., Nagao, S., 2017. Unexpected source of Fukushima-derived radiocesium to the coastal ocean of Japan. Proc. Natl. Acad. Sci. 201708659. http://dx.doi.org/10.1073/pnas.1708659114.

Schonfeld, W., 1995. Numerical simulation of the dispersion of artificial radionuclides in the English Channel and the North Sea. J. Mar. Syst. 6, 529–544.

SCJ, 2014. A Review of the Model Comparison of Transportation and Deposition of Radioactive Materials Released To the Environment As a Result of the Tokyo Electric Power Compny's Fukushima Daiichi Nuclear Power Plant Accident. Report of the Sectional Committee on Nuclear Accident Committee on Comprehensive Synthetic Engineering, Science Council of Japan.

Shchepetkin, A.F., McWilliams, J., 2009. Correction and commentary for ocean forecasting in terrain-following coordinates: formulation and skill assessment of the regional ocean modeling system by Haidvogel et al. J. Comput. Phys. 227, 3595–3624.

Simonsen, M., Saetra, Ø, Isachen, P.E., Christian Lind, O., Skjerdal, H.K., Salbu, B., Heldal, H.E., Gwynn, J.P., 2017. The impact of tidal and mesoscale eddy advection on the long term dispersion of ^{99}Tc from Sellafield. J. Environ. Radioact. 177, 100–112.

Smith, J.N., Rossi, V., Buesseler, K.O., Cullen, J.T., Cornett, J., Nelson, R., Macdonald, A.M., Robert, R., Kellogg, J., 2017. Recent transport history of Fukushima radioactivity in the northeast Pacific Ocean. Environ. Sci. Technol. 51, 10494–10502.

Smith, J.G., Simmonds, J.R. (Eds.), 2009. The Methodology for Assessing the Radiological Consequences of Routine Releases of Radionuclides To the Environment Used in PC-CREAM 08, HPA-RPD-058.

Takata, H., Aono, T., Tagami, K., Uchida, S., 2016. A new approach to evaluate factors controlling elemental sediment-seawater distribution coefficients (k_d) in coastal regions. Japan Sci. Total Environ. 543, 315–325.

Takemura, T., Nakamura, H., Takigawa, M., Kondo, H., Satomura, T., Miyasaka, T., Nakajima, T., 2011. A numerical simulation of global transport of atmospheric particles emitted from the Fukushima Daiichi Nuclear Power Plant. SOLA 7, 101–104.

Tateda, Y., Tsumune, D., Tsubono, T., 2013. Simulation of radioactive cesium transfer in the southern Fukushima coastal biota using a dynamic food chain transfer model. J. Environ. Radioact. 124, 1–12.

Thomann, R.V., Shkreli, F., Harrison, S., 1997. A pharmacokinetic model of cadmium in rainbow trout. Environ. Toxicol. Chem. 16, 2268–2274.

Tierney, K.M., Heymans, J.J., Muir, G.K.P., Cook, G.T., Buszowski, J., Steenbeek, J., Walters, W.J., Christensen, V., MacKinnon, G., Howe, J.A., Xu, S., 2018. Modelling marine trophic transfer of radiocarbon (^{14}c) from a nuclear facility. Environ. Model. Softw. 102, 138–154.

Toscano-Jiménez, M., García-Tenorio, R., 2004. A three-dimensional model for the dispersion of radioactive substances in marine ecosystems. Application to the Baltic Sea after the Chernobyl disaster. Ocean Eng. 31, 999–1018.

Tsubono, T., Misumi, K., Tsumune, D., Bryan, F.O., Hirose, K., Aoyama, M., 2016. Evaluation of radioactive cesium impact from atmospheric deposition and direct release fluxes into the North Pacific from the Fukushima Daiichi nuclear power plant. Deep Sea Research I 115, 10–21.

Tsumune, D., Tsubono, T., Aoyama, M., Hirose, K., 2012. Distribution of oceanic ^{137}Cs from the Fukushima Daiichi nuclear power plant simulated numerically by a regional ocean model. J. Environ. Radioact. 111, 100–108.

Tsumune, D., Tsubono, T., Aoyama, M., Uematsu, M., Misumi, K., Maeda, Y., Yoshida, Y., Hayami, H., 2013. One-year, regional-scale simulation of ^{137}Cs radioactivity in the ocean following the Fukushima Dai-ichi Nuclear Power Plant accident. Biogeosciences 10, 5601–5617.

Turner, A., Millward, G.E., 1994. Partitioning of trace metals in a macrotidal estuary. Implications for contaminant transport models. Estuar. Coast. Shelf Sci. 39, 45–58.

Turner, A., Millward, G.E., Balet, A.J., Morris, A.W., 1992. The solid-solution partitioning of trace metals in the southern North Sea-in situ radiochemical experiments. Cont. Shelf Res. 12, 1311–1329.

Umlauf, L., Burchard, H., 2003. A generic length-scale equation for geophysical turbulence models. J. Mar. Res. 6, 235–265.

Usui, N., Wakamatsu, T., Tanaka, Y., Hirose, N., Toyoda, T., Nishikawa, S., Fujii, Y., Takatsuki, Y., Igarashi, H., Ishikawa, Y., Kuragano, T., Kamach, M., 2016. Four-dimensional variational ocean reanalysis: A 30-year high-resolution dataset in the western North Pacific (FORA-WNP30). J. Oceanogr. 67, 449–479.

Uusitalo, L., Lehikoinen, A., Helle, I., Myrberg, K., 2015. An overview of methods to evaluate uncertainty of deterministic models in decision support. Environ. Model. Softw. 63, 24–31.

van Leeuwen, S., Tett, P., Mills, d., van der Molen, J., 2015. Stratified and nonstratified areas in the North Sea: Long-term variability and biological and policy implications. J. Geophys. Res. Oceans 120, 4670–4686.

van Rijn, L.C., 1993. Principles of Sediment Transport in Rivers, Estuaries, and Coastal Seas. Aqua Publications, Amsterdam, The Netherlands.

Vives i Batlle, J., Aoyama, M., Bradshaw, C., Brown, J., Buesseler, K.O., Casacuberta, N., Christl, M., Duffa, C., Impens, N.R.E.M., Iosjpe, M., Masqué, P., Nishikawa, J., 2018. Marine radioecology after the Fukushima Dai-ichi nuclear accident: Are we better positioned to understand the impact of radionuclides in marine ecosystems? Sci. Total Environ. 618, 80–92.

Vives i Batlle, J., Beresford, N., Beaugelin-Seiller, K., Bezhenar, R., Brown, J., Cheng, Jing-Jy, Cujic, M., Dragovic, S.S., Duffa, C., Fievet, B., Hosseini, A., Jung, K.T., Kamboj, S., Keum, Dong-Kwon, Kobayashi, T., Kryshev, A., Le Poire, D., Maderich, V., Min, Byung Il, Periáñez, R., Sazykina, T., Suh, K.S., Yu, C., Wang, C., Heling, R., 2016. Inter-comparison of dynamic models for radionuclide transfer to marine biota in a Fukushima accident scenario. J. Environ. Radioact. 153, 31–50.

Vives i Batlle, J., Bryan, S., McDonald, P., 2007. A process-based model for the partitioning of soluble, particulate and sediment fractions of plutonium and radiocaesium in the Eastern Irish Sea near Sellafield. J. Environ. Radioact. 99 (1), 62–80.

Vives i Batlle, J., Wilson, R.C., Watts, S.J., Jones, S.R., McDonald, P., Vives-Lynch, S., 2008. Dynamic model for the assessment of radiological exposure to marine biota. J. Environ. Radioact. 99, 1711–1730.

Walters, W.J., Christensen, V., 2018. Ecotracer: analyzing concentration of contaminants and radioisotopes in an aquatic spatial-dynamic food web model. J. Environ. Radioact. 181, 118–127.

Winterwerp, J.C., van Kester, W.G.M., 2004. Introduction To the Physics of Cohesive Sediment Dynamics in the Marine Environment, vol. 56. Elsevier.

Yuschenko, S., Kovalets, I., Maderich, V., Treebushny, D., Zheleznyak, M., 2005. Modelling the radionuclide contamination of the Black Sea in the result of Chernobyl accident using circulation model and data assimilation. Radioprotection 40, S685–S691.

Zhang, Y., Battista, A.M., 2008. SELFE: A semi-implicit Eulerian-Lagrangian finite-element model for cross-scale ocean circulation. Ocean Model. 21 (3–4), 71–96.

Zheleznyak, M.J., Demchenko, R.I., Khursin, S.L., Kuzmenko, Y.I., Tkalich, P.V., Vitiuk, N.Y., 1992. Mathematical modeling of radionuclide dispersion in the Pripyat-Dnieper aquatic system after the Chernobyl accident. Sci. Total Environ. 112 (1), 89–114.