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Report of Working Group 7

Modelling and Data for Radiological Impact
Assessments (MODARIA II) Programme

ENHANCEMENT OF MODELLING
APPROACHES FOR THE ASSESSMENT
OF RADIONUCLIDE TRANSFER
IN THE MARINE ENVIRONMENT

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ENHANCEMENT OF MODELLING
APPROACHES FOR THE ASSESSMENT
OF RADIONUCLIDE TRANSFER
IN THE MARINE ENVIRONMENT

REPORT OF WORKING GROUP 7

MODELLING AND DATA FOR RADIOLOGICAL IMPACT
ASSESSMENTS (MODARIA II) PROGRAMME

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2024

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FOREWORD

The IAEA has been organizing international model testing programmes for the transfer of radionuclides in the environment and the estimation of radiation exposures since the 1980s. These programmes have contributed to a general improvement in such models, both in the transfer of data and in the capabilities of modellers in Member States. IAEA publications on this subject over the past three decades demonstrate the comprehensive nature of the programmes and record the associated advances which have been made.

In 2012 the IAEA organized a programme entitled Modelling and Data for Radiological Impact Assessments (MODARIA). The first phase of the programme ran until 2015 (MODARIA I). From 2016 to 2019 the IAEA organized the second phase of the programme, MODARIA II, where seven working groups continued much of the work of MODARIA I. This publication describes the work on the assessment of fate and transport of radionuclides released in the marine environment carried out in Working Group 7 that started during MODARIA I.

Eight different institutions and companies participated in the work described in this publication: ABmerit (Slovakia), Defence Academy of the United Kingdom (United Kingdom), Institute of Mathematical Machines and Systems Problems (Ukraine), Japan Atomic Energy Agency (Japan), Korea Atomic Energy Research Institute (Republic of Korea), Korea Institute of Ocean Science and Technology (Republic of Korea), Norwegian Radiation and Nuclear Safety Authority (Norway) and University of Seville (Spain).

The IAEA wishes to express its gratitude to all those who participated in Working Group 7 of the MODARIA II programme and gratefully acknowledges the valuable contribution of R. Perriñez (Spain) as leader of Working Group 7. The IAEA officer responsible for this publication was P. McGinnity of the Marine Environment Laboratories.

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SUMMARY

In recent years there has been a development in the complexity of models and approaches to effectively assess the transfer of radionuclides in marine environments. Through the Environmental Modelling for Radiation Safety (EMRAS and EMRAS II) and Modelling and Data for Radiological Impact Assessments (MODARIA I and MODARIA II) programmes, the IAEA has facilitated knowledge sharing on this topic, including through intercomparison, testing and development of models. This publication describes the work undertaken by Working Group 7, Assessment of Fate and Transport of Radionuclides Released in the Marine Environment of the IAEA's Modelling and Data for Radiological Impact Assessments (MODARIA II) programme (2016–2019). The work carried out was a natural continuation of that completed during the first phase of the IAEA's MODARIA programme (2012–2015) by Working Group 10, Modelling of Marine Dispersion and Transfer of Radionuclides Accidentally Released from Land Based Facilities. In MODARIA II, the degree of complexity of the models used was increased in order to effectively consider additional processes, specifically uptake by biota, and the spatiotemporal scales of the simulations were also expanded.

Firstly, general information on the simulation of radionuclides in the marine environment using dispersion and transport models was compiled. This is intended for scientists in Member States considering development or adoption of models for radiological assessment following incidents or emergencies involving releases of radionuclides into the marine environment. In particular, methods of simulating water–sediment interactions were analysed in detail and some information is provided on the experimental determination of distribution coefficients. Furthermore, complementary information that also needs to be collected to ensure that these distribution coefficients are useful for modelling purposes is provided.

A Fukushima Daiichi Nuclear Power Station accident scenario which had already been studied during MODARIA I was expanded from a regional to an oceanic scale (including almost the whole of the North Pacific Ocean). An assessment of biological uptake was included, and simulations were performed over a period of two years. Other simulation exercises were undertaken for areas with significantly different characteristics, namely the North Atlantic Ocean and the European Shelf Seas. These exercises consisted of simulating historical releases of ^{137}Cs from the nuclear fuel reprocessing facilities of Sellafield (UK) and La Hague (France) and thus simulations were performed over some 50 years.

A theoretical study concerning the interpolation schemes which are included in Lagrangian transport models was carried out to investigate whether the different schemes could lead to significantly different results and thus constitute a source of model uncertainty. A series of transport problems of increasing complexity was designed for this purpose.

Finally, an exploratory model intercomparison exercise was also carried out. This was a blind test of three models and involved computation of the radionuclide activity concentrations resulting from the routine discharges from a hypothetical nuclear power plant located on the US Atlantic coast. This work was carried out in collaboration with MODARIA II Working Group 3, Assessments and Control of Exposures to the Public and Biota for Planned Releases to the Environment.

1. INTRODUCTION

1.1. BACKGROUND

The IAEA organized a programme from 2016 to 2019, entitled Modelling and Data for Radiological Impact Assessments (MODARIA II), which had the general aim of enhancing the capabilities of Member States to simulate radionuclide transfer in the environment and, thereby, to assess exposure levels of the public and in the environment in order to ensure an appropriate level of protection from the effects of ionizing radiation, associated with radionuclide releases and from existing radionuclides in the environment.

The following topics were addressed in seven working groups:

- Working Group 1: Assessment and Decision Making of Existing Exposure Situations for NORM and Nuclear Legacy Sites;
- Working Group 2: Assessment of Exposures and Countermeasures in Urban Environments;
- Working Group 3: Assessments and Control of Exposures to the Public and Biota for Planned Releases to the Environment;
- Working Group 4: Transfer Processes and Data for Radiological Impact Assessment;
- Working Group 5: Exposure and Effects to Biota;
- Working Group 6: Biosphere Modelling for Long Term Safety Assessments of High Level Waste Disposal Facilities;
- Working Group 7: Assessment of Fate and Transport of Radionuclides Released in the Marine Environment.

The activities and results achieved by the Working Groups are described in individual IAEA TECDOCs. This publication describes the work of Working Group 7.

The work of the MODARIA II Programme provides Member States with technical information that can be used by their competent authorities to develop and improve their modelling capability for radiological environmental impact assessment (REIA) to support them in meeting the requirements of IAEA Safety Standards Series No. GSR Part 3, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards [1], with respect to safety assessment and REIA for facilities and activities. The results of the MODARIA II programme also support Member States in the implementation of IAEA Safety Standards Series No. GSG-10 [2]. In particular, the IAEA TECDOCs written from the work undertaken in the MODARIA II programme provide information on the improvement of environmental transfer models for reducing uncertainties associated with the results of REIA. The work of MODARIA II also include the development of new assessment approaches and methodologies to strengthen the evaluation of the radiological impact of radionuclides in the environment on people, as well as to flora and fauna.

The work described in this publication is a natural continuation of that completed during the MODARIA I programme (2012–2015) by Working Group 10 (Modelling of Marine Dispersion and Transfer of Radionuclides Accidentally Released from Land Based Facilities) and reported in the respective MODARIA I Working Group 10 report [3]. Eight different institutes and companies actively participated in the work described in this publication: ABmerit (Slovakia), Defence Academy of the United Kingdom (AoD), Institute of Mathematical Machines and Systems Problems (IMMSP, Ukraine), Japan Atomic Energy Agency (JAEA, Japan),

Korea Atomic Energy Research Institute (KAERI, Republic of Korea), Korean Institute of Ocean Science and Technology (KIOST, Republic of Korea), Norwegian Radiation and Nuclear Safety Authority (DSA, Norway) and University of Seville (USEV, Spain). The following models were used: I/K THREETOX (IMMSP/KIOST), I/K Lagrangian (IMMSP/KIOST), ESTE (ABmerit), ESTE AI (ABmerit), USEV (USEV), SEA-GEARN (JAEA), LORAS (KAERI), POSEIDON R (IMMSP) and NRPA (DSA)

The aims of the MODARIA I programme were to make progress in relation to the assessment of radionuclides in the environment and their impact to humans and biota. MODARIA Working Group 10 specifically dealt with the modelling of marine dispersion and transfer of radionuclides accidentally released from land based facilities. Different models under development in Member States were applied in order to estimate radionuclide activity concentrations in the marine environment in two different scenarios: (i) the accidental releases from the Fukushima Daiichi Nuclear Power Station (FDNPS) accident into the Pacific Ocean; and (ii) the deposition onto the Baltic Sea following atmospheric releases from the Chernobyl accident in 1986. Models with different characteristics and levels of complexity were used, ranging from box models, to those making use of the shallow water and advection–diffusion equations [3]. For the scenario of the accidental release from the FDNPS, it was found that the different hydrodynamic circulation data used to drive each model (the numerical description of water currents in the model domain, which is a fundamental input of any marine transport model) had a significant impact on model results in the highly dynamic and energetic waters off the coast of the FDNPS. These waters are characterized by the convergence of warm and cold currents and the consequent generation of unsteady eddies. Consequently, there was high variability between the results of the different transport models for this scenario, with similar hydrodynamic models predicting differing current fields which, in turn, led to very different radionuclide dispersion patterns. Small differences in the circulation inputs produced different dispersion patterns and these differences tended to be amplified with time. Thus, it was concluded that water circulation is a very significant source of model uncertainty in highly dynamic waters. In contrast, the results of the transport models for the Baltic Sea scenario were in good agreement despite the different hydrodynamic circulation modelling approaches.

In addition to the work described in this publication, a number of peer reviewed scientific papers have been published by members of Working Group 7 during the MODARIA II programme:

- The FDNPP accident simulations were reported in detail in Ref. [4];
- A review of current marine transport modelling techniques, uncertainties and practical items to consider for radiological assessments following incidents or emergencies involving releases of radionuclides into the marine environment was published in Ref. [5];
- The analysis of marine distribution coefficients, k_d , was described in Ref. [6].

Summaries of the work reported in these papers are given in the relevant sections of this publication.

1.2. OBJECTIVE

The work carried out under MODARIA II was a natural continuation of that completed during the first phase of the IAEA's MODARIA I programme (2012–2015). A key objective was to assess recent significant developments of the models used to simulate the dispersion and transport of radionuclides in the marine environment in several areas including more effective consideration of uptake of radionuclides by biota, and expansion of the spatiotemporal scales

of the simulations compared to those considered in MODARIA I. A biological uptake model was interfaced with the marine transport models: it is understood that this has not been done previously on these spatiotemporal scales. Three different types of biological uptake model, equilibrium, dynamic and allometric models, were tested. Model predictions for ^{137}Cs activity concentrations in seawater (at three different depth layers), bed sediment and biota (for four classes of organisms) were compared with measurements over a model domain covering the entire North Pacific Ocean.

A significant conclusion of the work undertaken in the MODARIA I programme was that the assumptions on water circulation used as input to the models had a major influence on model results in the highly dynamic and energetic waters in the western North Pacific Ocean off the coast of Fukushima. Therefore, this was further tested in MODARIA II with the same water circulation data being used by all of the marine transport models used.

A further objective was to provide information on the simulation of radionuclides in the marine environment using dispersion and transport models for scientists in Member States considering development or adoption of models for radiological assessment following incidents or emergencies involving releases of radionuclides into the marine environment. In particular, details of methods of simulating water–sediment interactions are described in detail and some information is provided on the experimental determination of distribution coefficients. Several dynamic models for describing water–sediment interactions were investigated and compared (e.g. one step versus two step models; single layer versus layered sediment models). The relative advantages of each approach are described and some information on their use is given. The review also includes sources of uncertainties in models and practical items for consideration, with particular focus placed on water–sediment interactions and uptake of radionuclides by biota.

1.3. SCOPE

The Working Group compiled information on the simulation of radionuclides in the marine environment using dispersion and transport models and, in particular, on methods of simulating water–sediment interactions, the experimental determination of distribution coefficients and information that also needs to be collected to ensure that these distribution coefficients are useful for modelling purposes. Some general information on the use of models for such assessments are provided and the relative advantages and disadvantages for each transport model type (box, Eulerian, Lagrangian) are described.

A theoretical study was carried out to investigate whether the different interpolation schemes included in Lagrangian transport models could lead to significantly different results giving rise to a source of model uncertainty. A series of transport problems of increasing complexity was designed for this purpose.

Several model intercomparison exercises were carried out for releases to the marine environment, covering a reconstructed accidental release scenario to the North Pacific Ocean from the Fukushima Daiichi Nuclear Power Station; a simulation of historic releases from nuclear fuel reprocessing facilities to the North Atlantic Ocean and the European Shelf Seas; and a routine discharge from a hypothetical nuclear power plant on the US Atlantic Coast.

The simulations of historical releases of ^{137}Cs from the European nuclear fuel reprocessing facilities are presented in Section 4 involved the consideration of a large model area (almost the whole North Atlantic Ocean including the European Shelf Seas) and an even longer duration

(50 years). Biological uptake models were also coupled with the transport models. The transport models participating in the exercise were driven using the results of ocean global circulation models (OGCMs) – several OGCMs were tested – throughout the model domain. Model results – activity concentrations in seawater, sediment and biota – were compared with measurements obtained from the IAEA’s Marine Radioactivity Information System, MARIS . The limitations for this type of application of OGCMs used to calculate water circulation are presented.

As a collaboration with MODARIA II Working Group 3, an exploratory model intercomparison for a hypothetical nuclear power plant (NPP) release scenario into a coastal area was carried out. Some customizations of existing models were needed and a box model was specifically designed for the scenario. This was a completely blind model intercomparison for which the only data provided to modellers were radionuclide releases from the hypothetical site.

Descriptions of the models used have, in general, not been included in this publication as most were previously presented in detail in the MODARIA I Working Group 10 report [3]. Only new information on biological uptake models and two models not described in Ref. [3] are included in the Appendix.

1.4. STRUCTURE

Section 1 provides a brief description of the background of the MODARIA programme and the work of the Working Group covering Assessment of Fate and Transport of Radionuclides Released in the Marine Environment. Section 2 summarizes a detailed review of current marine transport modelling techniques for radiological assessments following incidents or emergencies involving releases of radionuclides into the marine environment that is intended for scientists in Member States considering development or adoption of marine dispersion and transport models for such purposes.

Sections 3 and 4 describe comparisons of the results of the analyses of two contrasting marine modelling scenarios that were undertaken as part of MODARIA II: (i) the dispersion of radionuclides released into the Pacific Ocean following the FDNPS accident; and (ii) the dispersion of historical releases of ^{137}Cs from the European nuclear fuel reprocessing facilities of Sellafield (UK) and La Hague (France) in the Atlantic Ocean.

An analysis of model interpolation schemes used in Lagrangian models for both temporal and spatial interpolations is described in Section 5. A series of exercises of increasing levels of complexity was specifically designed for this purpose. Section 6 gives the conclusions of the work undertaken by the Working Group.

In the Appendix, the results of an exploratory model intercomparison for a hypothetical nuclear power plant (NPP) release scenario into a coastal area are presented. Some customizations of existing models were needed and a box model was specifically designed for the scenario: these are also described in the Appendix.

2. USE OF MARINE MODELS FOR TECHNICAL ASSESSMENTS FOLLOWING NUCLEAR OR RADIOLOGICAL INCIDENTS OR EMERGENCIES

A summary of the main conclusions of a review of current marine transport modelling techniques, sources of uncertainties in models, and practical items to consider when models are applied for radiological assessments following incidents or emergencies involving releases of radionuclides into the marine environment, is presented in this section. The main approaches are summarized in Section 2.1. Some general information on the use of models for such assessments is provided in Section 2.2, including advantages and disadvantages for each transport model type (i.e. box, Eulerian, Lagrangian). The main sources of uncertainty involved when performing such modelling are presented in Section 2.3 and, finally, an analysis of best practice for modelling water–sediment interactions is presented in Section 2.4.

2.1. OVERVIEW OF THE MAIN APPROACHES FOR MODELLING RADIONUCLIDES IN THE MARINE ENVIRONMENT

Three types of models are generally used to simulate the transport of radionuclides in the marine environment, namely box models, Eulerian models and Lagrangian models. The main equations and assumptions involved in each approach are summarized in Ref. [5] and are not repeated here. Essentially, box models divide the model domain – the spatial extent of the marine environment considered – into boxes or compartments in which material is exchanged according to the fluxes of water between the boxes. These fluxes are derived from oceanographic observations (measurements), from numerical ocean circulation models or from a combination of the two (reanalysis of forecast data assimilating observations). Eulerian models are based on an advection–diffusion differential equation for the radionuclide concentration. In a Lagrangian model a radionuclide release is simulated by a number of particles (each one being representative of a certain activity in becquerels (Bq)); the path followed by each particle is calculated and the final activity concentrations are calculated from the number of particles in each volume unit of water. Eulerian and Lagrangian models use detailed water current fields derived from ocean circulation models (forecast or reanalysis datasets). Methods for considering water–sediment interactions and other biogeochemical processes are also described in detail in Ref. [5].

The general modelling approach is shown in Fig. 1, with the different types of model which may be applied to physical transport (advection and diffusion) and biogeochemical processes, as well as with the potential sources of the necessary inputs. The outputs of circulation models provide detailed water current fields which are used by Eulerian and Lagrangian models to simulate radionuclide physical transport. Box models use water fluxes, which are obtained from observations and/or calculated from the water currents provided by circulation models.

Biogeochemical processes (water–sediment interactions and biota uptake of radionuclides) are described by equilibrium models or dynamic models. The equilibrium approach is generally used in box models, since long temporal scales are simulated with these models. Dynamic models are, in contrast, more frequently used in Eulerian and Lagrangian models. Suspended matter concentrations and sedimentation rates also have to be known if adsorption–desorption reactions of radionuclides with suspended particulate matter are included in the model. Mean values deduced from observations for these parameters are generally used in box models.

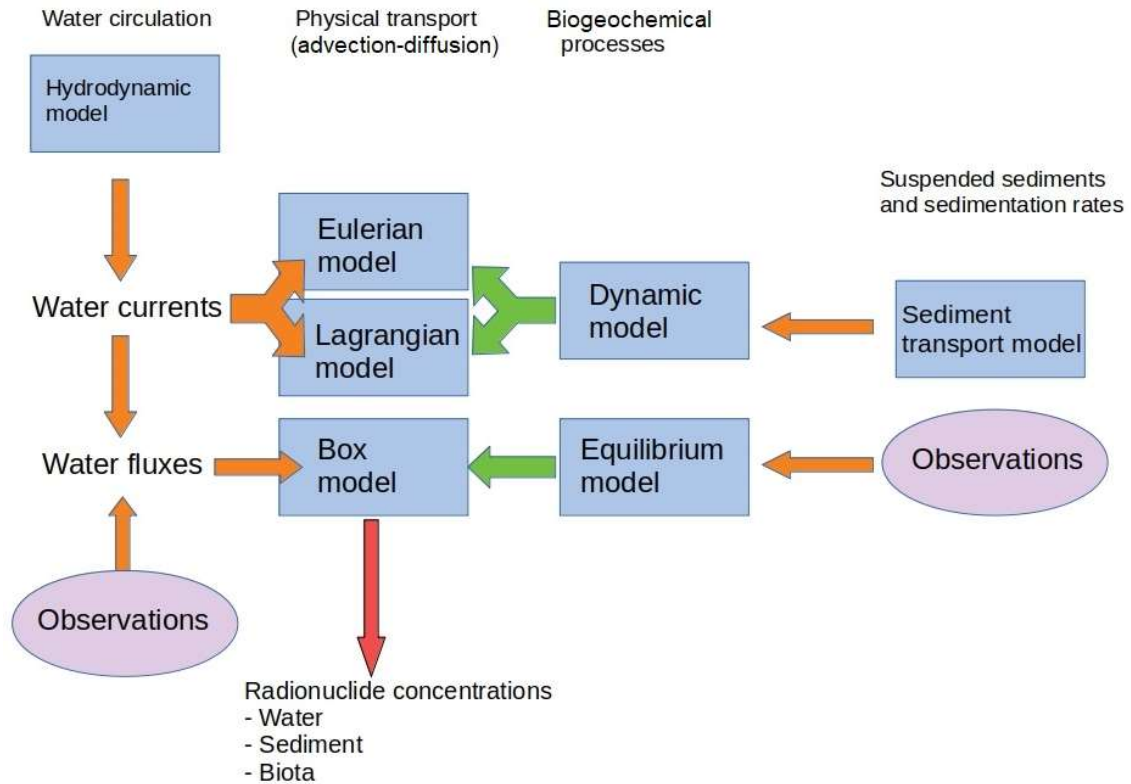


FIG. 1. The general modelling approach for the marine environment [5].

A sediment transport model is more often used in Eulerian and Lagrangian models to derive such parameter values. The physical transport model, together with the added biogeochemical model, provides radionuclide activity concentrations in seawater, sediment and the biota classes considered. Dynamic biological uptake models often include consideration of phytoplankton, zooplankton, non-piscivorous and piscivorous fish [5].

A review of the numerous transport models for the marine environment which have been applied in the Pacific Ocean after the FDNPP accident is also presented in Ref. [5]. This review covers the first modelling studies published soon after the accident took place up to the most recent model-model and model-measurement comparison exercises for seawater, sediment and biota, including those carried out within the frame of MODARIA I [3, 7] and MODARIA II [4].

2.2. GENERAL INFORMATION ON THE USE OF MODELS IN RADIOLOGICAL ASSESSMENTS

Models of varying degrees of complexity may be used to calculate radionuclide activity concentrations in the marine environment following an incident or emergency involving the release of radionuclides into the marine environment. A general principle is that a simple model is always better than a more complex one, if it provides comparable results for a particular application as the results will be easier to interpret and it will be easier to apply in different scenarios. However, as presented in Ref. [5], a simple model might not be universally applicable for application to the diverse range of potential modelling scenarios, marine systems and environmental circumstances that other more intricate and wide-ranging models are capable of simulating. For example, peak ^{137}Cs activity concentrations are reached concurrently in

seawater and fish if an equilibrium model is used. In contrast, if dynamic models are applied to the same scenario, there is a delay before peak activity concentrations in fish are reached which is more realistic. As a result, the simple (equilibrium) model is not suitable for assessing the initial phase following non-routine releases and a more complex model, capable of simulating non-equilibrium conditions, needs to be used. The same point applies to the situation of water–sediment interactions in dynamic conditions.

The method used to compare models in model–model and model–measurement comparisons needs to be carefully considered. Comparisons of Lagrangian model outputs at specific points are difficult to make because a number of discrete particles are released, and so scattered concentration maps are generally produced, especially in the case of large spatial model domains (i.e. thousands of km). Consequently, it may be more convenient to divide the oceanic space into a number of boxes and to obtain average activity concentrations over such boxes. Construction of the box structure needs to carefully consider water currents in the area of interest and also support greater detail nearer to the release location (i.e. normally smaller boxes are defined for areas close to the release and gradually increase in area with distance). Thus, as mentioned above, generic box models with fixed structures might not be suitable for every situation. As a simple example, a release of radionuclides following an incident or emergency in a nuclear powered ship or submarine could, in theory, occur anywhere.

Currents derived from OGCMs are often used for predicting transport in coastal waters. However, since these models might not adequately describe currents in coastal regions, use of carefully tested local forecasts of currents need to be considered and ideally be available for all waters which could potentially be affected by an incident or emergency involving the release of radionuclides.

The different formulations and numerical treatments of processes involved in the transport of radionuclides in the marine environment lead to different modelling approaches. Advection and mixing may be solved using a box, a Eulerian or Lagrangian model, as mentioned previously. Water–sediment interaction and biota uptake may be described, essentially, using an equilibrium or a dynamic model. However, there is no single model available which can be applied to all situations (i.e. to all spatiotemporal scales), because of practical computational limitations. This has been analysed in detail in a recently published review paper [5].

The basic assumptions of box models, namely uniform and instantaneous mixing of radionuclides within each box [3], render these models well suited to long term assessments over large spatial scales. Therefore, they are useful tools in the longer term after an incident or emergency has occurred, as well as for the environmental assessment of ongoing routine discharges from nuclear facilities. In addition, the mathematics involved are relatively simple and these models are easy to develop and to adapt to specific cases. It has to be pointed out that the NRPA (Norwegian Radiation and Nuclear Safety Authority) box model which has been used in the work described in this publication, and also described and applied in Ref. [3], assumes instantaneous mixing in the release box only, not in the whole oceanic space, in order to make it more compatible with hydrodynamic models. Specific mathematical details may be found in Ref. [8]. Finally, for box models, detailed water circulation patterns are not needed. The only parameterization needed is the fluxes of water between boxes.

Eulerian and Lagrangian models make use of detailed water current fields [3], changing in time and space. Thus, these models are appropriate for the acute phases (up to a few months) of the assessment of an incident or emergency. The mathematical formulations used in these models are generally more complex than in box models and they are therefore more difficult to develop

and customize. Moreover, these models need the previously mentioned water current fields as input data; information which is not always easy to obtain and sufficiently accurate, as analysed in detail during the MODARIA I programme [3].

Lagrangian models are especially well suited to the acute phase of an incident or emergency, since they do not introduce numerical diffusion [5] and can therefore handle the very high concentration gradients between contaminated and clean seawater which would be expected after an acute radionuclide release into the marine environment. In addition, computation can be significantly faster than for Eulerian models when the contaminated area is initially a small part of the computational domain and the number of particles in the simulation can be kept relatively low (typically a few tens of thousands of particles). This is another advantage to be considered for incident or emergency modelling: normally a rapid response is needed for the provision of information to decision makers. Finally, a real point source of radionuclides may be defined in Lagrangian models; in Eulerian models, in contrast, the initial plume size is defined by spatial resolution of the calculation grid (where the minimum size is obviously equal to one grid cell). Consequently, lower peak activity concentrations are to be expected from Eulerian models.

However, Eulerian models have a significant advantage over Lagrangian, in that additional processes are easier to include since only the addition of new terms to the transport differential equations is needed. For example, the inclusion of multistage water–sediment interactions and/or redox reactions can be implemented in a simpler way in Eulerian models [9]. In contrast, the stochastic treatment of these processes needed by the Lagrangian approach [3, 5] makes it more difficult to implement. Moreover, the number of particles needed in a Lagrangian simulation increases as the number of sub-compartments (e.g. different oxidation states in seawater, different speciation states in sediments) increases. Consequently, a 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 suitable for simulating spatially extended radionuclide sources (e.g. due to atmospheric fallout) over large areas since many particles would be needed in a Lagrangian simulation, which would be computationally much more expensive.

Water–sediment interactions and biota uptake, can be described using equilibrium or dynamic models, as mentioned previously. Again, each approach has advantages and disadvantages. The obvious advantage of equilibrium models is their simplicity and the fact that few parameters are needed, that is only water–sediment distribution coefficients and biota concentration ratios. The equilibrium assumption means that these models are ideal for application in long term assessments over wide spatial scales and are well suited for inclusion within box transport models. In contrast, these models are not appropriate for emergency purposes and for assessments of ongoing routine discharges near the release point, since the steady state or equilibrium assumption is not valid in these cases [6]. Instead, dynamic models have to be used in such cases. However, difficulties can arise as these models are necessarily more complicated and a larger number of input parameters is needed. These parameters can be radionuclide and location specific and information about them is generally scarce and incomplete, especially in the acute phases of assessments of unplanned releases. Tentative values have to be used in many cases. Water–sediment interactions are presented in more detail in Section 2.4.

To summarize, each type of model can be a useful tool for marine radionuclide transport assessments, provided that it is applicable to the specific modelling challenge presented. Specifically, because of practical computational limitations, currently no model exists which is applicable to all spatiotemporal scales.

2.3. SOURCES OF UNCERTAINTY

Poor knowledge of model input parameters is a major source of uncertainty. The numerical description of turbulence is still a challenge in computational fluid dynamics. Diffusivities have to be parameterized and different schemes and approaches may be applied [5] and these different approaches can lead to different model results. In the case of fully conservative radionuclides (those that remain in solution, without any interaction with sediments), the only relevant parameters are the horizontal and vertical diffusion coefficients. For non-conservative radionuclides the situation is much more complex and a larger number of parameters are needed, such as kinetic rates, particle sizes, and density and thickness of the sediment. These parameters are site specific and information about them is generally scarce and so, in many cases only estimated values can be used.

A numerical solution of the equations is needed in the three types of model (i.e. box, Eulerian and Lagrangian). The most common method is the use of finite differences, by which differential equations are converted into algebraic equations. A discretization in time and space is needed to apply finite difference techniques, thus the different time steps and different grid sizes, in addition to the particular finite difference scheme selected, will contribute to model uncertainty. Spatial discretization is needed even in Lagrangian models to evaluate activity concentrations from the number of particles per water volume unit, which results in averaging of quantities. Since averaging leads to errors, a numerical solution, irrespective of whether a box, Eulerian or Lagrangian model is applied, is only an approximation to the exact solution (due to errors such as rounding errors or truncation errors, which necessarily appear).

Another issue is the so called ‘numerical diffusion’. A mathematical treatment can be seen in Ref. [10] which consists of the artificial smoothing of the radionuclide concentration gradients which appears in box and Eulerian models due to the numerical schemes. This problem is minimized in the case of Lagrangian models. High order accuracy numerical schemes are needed to decrease numerical diffusion in Eulerian models [10], but such schemes are difficult to handle and are computationally expensive.

Finally, the radionuclide release area has to be considered, since the initial radionuclide spatial extent may be quite different in the three types of model. A real point source may be defined in Lagrangian models since particles can be released at a given position. However, the radionuclide input is instantaneously mixed within a grid cell in a Eulerian model, and within the box where the source is located in the case of a box model. As a consequence, the initial concentration peak and spatial extent may be significantly different between the three models for the same release. Obviously, this is a source of uncertainty and discrepancy between models as time elapses. In the particular case of a box model, making the size of boxes in the release area smaller avoids the initial diffusion to the extent possible. As a consequence, the same box structure is not valid for all potential emergencies (i.e. all potential location of sources) which can occur in the sea.

2.4. MODELLING WATER–SEDIMENT INTERACTIONS

The simplest way of estimating the partitioning of a radionuclide released into the marine environment between the seawater and the sediment consists of the use of a distribution coefficient, k_d , which is defined as the ratio between the activity concentration of the radionuclide in the sediment C_s and the activity concentration in the seawater C_w , once equilibrium conditions have been achieved:

$$k_d = \frac{C_s}{C_w} \quad (1)$$

The basic assumption in the k_d model is that the partition of radionuclides between seawater and sediment is in equilibrium. However, there are cases where the distribution coefficient is used, but this basic assumption is not satisfied. Essentially, following a release of radionuclides into the marine environment, higher activity concentrations will initially be measured in seawater relative to activity concentrations in the sediment. Some time is needed for activity concentrations to build up in the sediment. After the release, radionuclides in seawater will be flushed off (more or less quickly, depending on the intensity of currents), while the sediment acts as a buffer or sink of radionuclides and thus behaves as a long term delayed source. When the partition of radionuclides is not in equilibrium, the ratio of C_s and C_w is denoted as an ‘apparent’ k_d [6]. If an equilibrium distribution coefficient is used to model these processes, there will be an overestimation of radionuclides in the sediment in the initial phase and an overestimation of radionuclide concentration in seawater in the late phase (or underestimation in the sediment, depending on the model numerical scheme).

A number of numerical experiments were carried out to study water–sediment interactions and to evaluate the performances of equilibrium and kinetic models, to compare one step and two step kinetic models (i.e. those consisting of, respectively, a single reversible reaction and two consecutive reversible reactions) and to compare single layer and layered sediment models. This work is described in detail in Ref. [6] and only the main conclusions are given here. Some indications about how best to provide experimental k_d data (i.e. in such a way that they are useful for modelling purposes) are also given, as well as indications about how changing environmental conditions (pH and salinity) can be treated in numerical models for radionuclide transport. These changing conditions may appear in estuaries, where salinity increases from freshwater to marine values, and in coastal areas close to industrial facilities, where changes in pH could be caused by waste releases. The main conclusions of the study are as follows:

- (1) The marine environment is an open system affected by external perturbations. As a consequence, steady state (understood as a radionuclide concentration which remains constant in time) does not necessarily mean equilibrium in the partition of tracers between the liquid and solid phases. This is especially relevant close to a radionuclide point source, such as a release point from a nuclear facility. In contrast, true equilibrium in radionuclide partition between seawater and sediment is found away from the source (i.e. 10 km to 100 km, depending on the physical characteristics of the area). Thus, measurements of k_d values in the sea, far away from radionuclide releases are more likely to be representative of an equilibrium situation. However, in practice the term ‘apparent’ k_d is used for the ratio between measured radionuclide activity concentrations in sediment and seawater. Only if equilibrium in the partition really exists, then this ratio is a ‘true’ distribution coefficient k_d . In other words, the apparent k_d turns to be a true k_d . Immediately after an accident, there is a decrease in the apparent k_d due to the faster contamination of seawater than sediments. Subsequently the apparent k_d increases as the sediment is contaminated, until reaching a maximum value. Afterwards there is a slow decrease in the apparent k_d towards the equilibrium value as radionuclides are slowly redissolved from the sediment. This behaviour was predicted by the calculations described in Ref. [6], and are supported by measurements in Fukushima seawaters and sediments [11, 12]. This temporal trend is presented in a very clear form in Ref. [12], where k_d measurements off the coast of Fukushima are presented over the period 1985–2018. It has also been demonstrated that the decrease in the apparent k_d offshore Fukushima towards equilibrium may take several decades [6].

- (2) Distribution coefficients for suspended and for seabed sediments may vary in orders of magnitude even if all the environmental conditions and respective particle properties are identical. As a consequence, it is essential to know, when k_d data are provided, whether they correspond to suspended matter–seawater or bed sediment–seawater. This is due to a purely geometry effect, that is part of the seabed sediment particle surface may be partially hidden by other particles and, thus, it is not available for interactions with seawater. Additionally, it is also essential to know the details of how measurements of radionuclides in sediments used to determine a measured k_d have been carried out. For example, if only the readily exchangeable phase has been extracted (which would provide what is denoted as the ‘fast’ k_d [6]) or, alternatively, whether a more aggressive treatment has been applied to the sediment to additionally extract more strongly fixed radionuclides from the sediment (which would provide the ‘total’ k_d [6]). Significant errors in predictions can be expected if the wrong k_d (fast versus total) is used in a model when it is utilized for an assessment of an incident or emergency.
- (3) During the initial phase of an accidental radionuclide release, one step and two step models produce essentially the same results. Thus, for rapid radiological assessments after an incident or emergency, a one step model may be adequate and it also needs fewer parameters. However, a two step model is expected to give better results when used for longer term calculations. In the case of the FDNPS accident, equilibrium in the partition of radionuclides between sediments and seawater is not rapidly achieved. If a two step model dominates radionuclide kinetics, as generally seems to be the case, decades are needed in order to achieve equilibrium, time during which the apparent k_d will slowly decrease from its maximum. This maximum k_d is also delayed with respect to the maximum concentration in seawater for a period of almost one year.
- (4) Single sediment layer and layered models have also previously been described and compared [6]. Model–model comparisons were carried out, as well as model comparisons with laboratory adsorption experiments. It was found that the use of each model is determined by the objectives of the particular modelling study. Moreover, a multilayer model has to be used to evaluate sediment inventories of radionuclides; but a single layer model, adequately tuned, may produce satisfactory results for radionuclide activity concentrations in the surface sediment. This is generally the case for radiological assessments performed following accidents.
- (5) As a general conclusion for the assessment of radiological incidents or emergencies, which imply short temporal scales, it is important to ensure the availability of site-specific values of kinetic rates which could be used in radionuclide transport models for areas where accidents might occur. That is to say, as is the case with water currents, careful selection of the ocean model is needed and after a detailed comparison with local measurements of currents in each area potentially exposed to an accident [3, 7]. Moreover, kinetic rates have to be characterized as best as possible.
- (6) A semi-empirical equation has been proposed to modulate the freshwater adsorption kinetic rate, k_1 , due to changes in salinity and pH. The mathematical details and a graphical representation are presented in Ref. [6]. Essentially k_1 is modulated by a function of pH and salinity $F(S, pH)$, $k_1 = k_{10} F$, where k_{10} is the freshwater k_d for $pH=7$. An increase in salinity reduces the adsorption rate because of competition of radionuclides with ions dissolved in water. Moreover, a reduction in pH increases dissolution and, hence, also reduces adsorption.

3. FUKUSHIMA DAIICHI NUCLEAR POWER STATION ACCIDENT SCENARIO: LARGE SCALE SIMULATIONS AND BIOTA TRANSFER OF CAESIUM-137

The transport of radionuclides released after the FDNPS accident was studied in detail during the MODARIA I programme. Moreover, results were described in the Working Group 10 report [3] and in a scientific paper jointly prepared by various participants in the programme [7]. Several Eulerian and Lagrangian transport models, which included water–sediment reactions, were applied and differences between them analysed. Two sources of radionuclides were considered: (1) direct releases into the Pacific Ocean; and (2) deposition on the sea surface of radionuclides released into the atmosphere. Direct releases were reconstructed by the Japan Atomic Energy Agency (JAEA) and deposition of atmospheric releases was estimated using two atmospheric dispersion models; details of these models are summarized in Refs [3] and [4]. There were five Lagrangian transport models used in the exercise by participants of the Working Group: ESTE (Slovakia), SEA-GEARN (Japan), LORAS (Republic of Korea), I/K Lagrangian (Ukraine and Republic of Korea) and USEV (Spain); and one Eulerian model: I/K THREETOX (Ukraine and Republic of Korea).

It was found that the main source of discrepancies in results was due to the hydrodynamic circulation data (a numerical description of water currents in the model domain) selected to drive each transport model. In highly dynamic environments, like the waters off the coast of Fukushima which are characterized by the convergence of currents and the generation of unsteady eddies, the transport model is extremely sensitive to this numerical description of water circulation. Small differences in magnitude and direction of currents predicted by the ocean circulation models result in differences in the dispersion patterns produced by the transport models, and these discrepancies tend to be amplified over time. In this work, the same water circulation data were used in the applied transport models with the objective of significantly improving agreement, both model–model and model–measurement.

In MODARIA II, simulations of the transport of the FDNPS accident releases were expanded by extending the spatiotemporal scale of the simulations. The simulation time was extended up to two years, from March 11, 2011 (it was only three months in MODARIA I) and the spatial domain of models covered most of the North Pacific Ocean.

Biological uptake models were also coupled with the transport models in order to predict the transfer of ^{137}Cs activity concentrations to biota.

Model results were compared with radionuclide measurements over the North Pacific Ocean for seawater (three depth layers), sediment and biota (four classes were included: phytoplankton, zooplankton, non-piscivorous and piscivorous pelagic fish). Model results were also compared with measured ^{137}Cs activity concentrations in seawater and sediment.

All of the model calculations and results are described in detail in Ref. [4] and, thus, only a summary is presented in this section.

For the simulations performed during the MODARIA I programme, two sources of radionuclides were considered, direct releases to the Pacific Ocean and radionuclides depositing on the sea surface from releases to atmosphere. In MODARIA II, data describing direct releases into the ocean (again reconstructed by the JAEA) were provided as input for the simulation models from 25 March to 31 December 2011. Data describing deposition of atmospheric releases onto the ocean surface were provided on a daily basis for the whole computational

domain for the period from 12 March to 1 June 2011 from the average results obtained by two atmospheric dispersion models run by JAEA and the Korea Atomic Energy Research Institute (KAERI).

The model domain for simulations extended longitudinally and latitudinally, 117°E–160°W and 15°N–65°N, respectively. Water circulation data calculated using the FORA (Four-dimensional Variational Ocean Reanalysis for the Western North Pacific) model for 1982-2014 [13] was used as input for the simulations in all the participating models as well as monthly climatological data from 2011 to 2014. The horizontal resolution of FORA is 0.1° with 54 vertical levels.

The six marine transport models listed above were used to conduct simulations. Eulerian and Lagrangian models were applied with different formulations for horizontal and vertical mixing. A dynamic approach was used by all models to describe water–sediment interactions, based on a desorption kinetic coefficient and the radionuclide equilibrium distribution coefficient, which is used to derive the adsorption kinetic coefficient. A summary of the characteristics of each model is given in Ref. [4] and detailed descriptions are included in the MODARIA Working Group 10 report [3].

Three different types of biological uptake models were applied, that is an equilibrium model based on a concentration ratio between biota and seawater, a dynamic model, and an allometric approach based on empirical relations between biota characteristics (such as daily ingestion rates, seawater intake rates and biological half-lives for different radionuclides) and the mass of the organism. The classes of biota included in the biological uptake models were phytoplankton, zooplankton, non-piscivorous and piscivorous pelagic fish. Details of the different biological uptake models are provided in Ref. [4]. Additional details on the dynamic biological uptake model used are provided in the Appendix (POSEIDON-R model, case of uniform radionuclide distribution in biota).

A compilation was made of measurements of ^{137}Cs activity concentrations in seawater (from surface, intermediate and deep levels), sediment and biota samples from the North Pacific Ocean during the period over which simulations were undertaken. The North Pacific Ocean was divided into a number of boxes or compartments according to the long term average currents in different regions of the ocean and the locations of the releases (both direct releases and deposition of airborne releases). Model results were averaged for each box and these averaged values were compared with measurements. It is worth mentioning that a model–measurement comparison for specific points in a large domain such as the North Pacific Ocean is not feasible with Lagrangian models which assume the release of individual particles and, consequently, produce scattered concentration patterns at long distances from the radionuclide source. Therefore, it is better to use averages over given model boxes. The box structure does not only depend on the water circulation, but also on the release locations. This implies that different box structures may be needed in order to simulate the consequences of different nuclear or radiological incidents or emergencies. For this reason, generic box models with fixed structures might not always be suitable for addressing all modelling challenges, although it has been demonstrated that these can effectively deal with large temporal and spatial scales (see Ref. [14]). As implemented previously in Ref. [15], a box model needs to be linked to a high-resolution hydrodynamic model which allows water fluxes for a flexible system of model boxes to be derived which may be adapted to any emergency conditions.

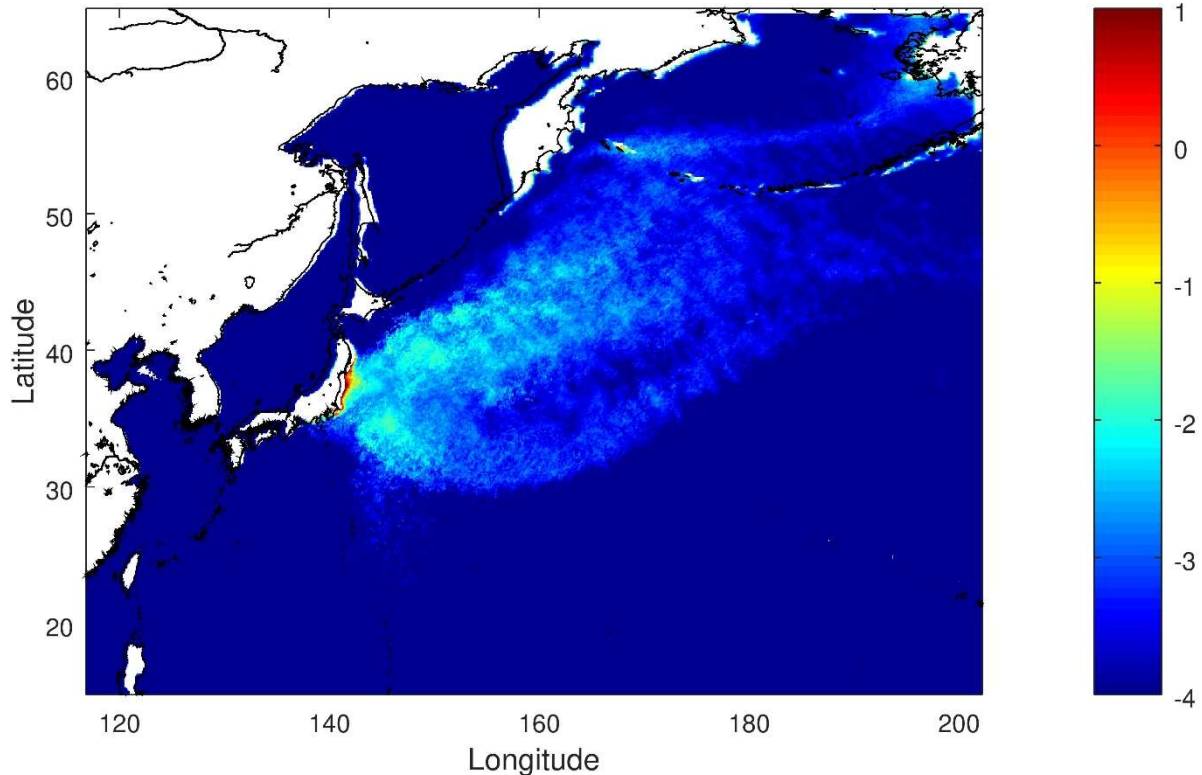


FIG. 2. Example biological uptake model output from the POSEIDON-R model showing ^{137}Cs activity concentrations in non-piscivorous fish using a biological uptake model (Bq/kg fresh weight) in April 2011. The scale in the colour bar is logarithmic.

An example of a biological uptake model output from the POSEIDON-R model is provided in Fig. 2, in which calculated ^{137}Cs activity concentrations over the North Pacific Ocean in non-piscivorous fish in April 2011 are presented. These activity concentrations are spatially averaged over the defined model boxes [4] to enable comparison of model results with measurements.

Detailed model–model and model–measurement comparisons are presented in Ref. [4], and only a brief summary is provided here. In some model compartments located far from the FDNPS, the results indicated increases in ^{137}Cs concentration in surface seawater (approximately one order of magnitude above background), occurring soon after the accident. These can be attributed to the deposition of atmospheric releases. In other compartments also located far from the FDNPP, there are no such increases. High activity concentrations were estimated for some compartments closer to the FDNPS, mainly due to direct releases to the ocean but also because of atmospheric deposition. Models and measurements both demonstrate a trend towards background activity concentrations after approximately one year. Models underestimate activity concentrations in the compartment into which direct releases occurred during the acute phase of the accident, since the average concentration over the whole box is calculated (and, additionally for box models, it is implicitly assumed that an instantaneous mixing occurs when obtaining model results). After the initial phase of the accident passed, model results agreed within an order of magnitude with the measurements.

In general, the models predicted which model compartments for surface, intermediate and deep waters were affected by the FDNPS accident (either by direct releases, deposition of airborne releases or both) and which model compartments were not affected.

Similarly, the results from each model for ^{137}Cs activity concentrations in sediment showed agreement in predicting which areas were affected by the accident and which were not. It is interesting to note that a decreasing trend in ^{137}Cs activity concentrations in sediments, some located more than 100 km away from Fukushima, is not observed (neither in measurements nor models). This indicates that sediments far from the FDPP could be acting as sinks of radionuclides and could potentially become a delayed source. In contrast, a decreasing trend in ^{137}Cs activity concentrations was observed in sediments near (less than 100 km) the FDNPP [16]. These sediments were heavily contaminated during the acute phase of the accident, after which, as the activity concentration in seawater decreased (due to both the reduction of releases and the presence of water currents), a more efficient redissolution of radionuclides occurred (where the desorption rate was proportional to the activity concentration in the sediment).

Biological uptake model predictions for the activity concentrations in zooplankton from the three types of models (equilibrium, dynamic and allometric) were found to be of the same order of magnitude, although there was an underestimation with respect to measured activity concentrations. Since radionuclides transfer from seawater to zooplankton (in contrast to phytoplankton where activity concentrations are in equilibrium with those in seawater), significant differences between equilibrium and dynamic models were not observed. However, the situation is different for both classes of fish (non-piscivorous and piscivorous). In the case of equilibrium models, peak activity concentrations in seawater and fish are simultaneous. However, there is a delay in the time of peak fish activity concentrations with respect to peak activity concentrations in seawater in the case of dynamic models. This is described in more detail in Ref. [17].

The results show, in general, a good agreement between models and between models and measurements. The approach used in the study to compare models is helpful, that is dividing the oceanic space into several boxes and obtaining average activity concentrations over such boxes. This is because comparisons of Lagrangian model outputs at specific points are difficult in such a large domain since several discrete particles are released, and consequently scattered concentration maps are produced.

4. ATLANTIC OCEAN SCENARIO: HISTORICAL RELEASES FROM EUROPEAN NUCLEAR FUEL REPROCESSING FACILITIES

The objective of the Atlantic Ocean model exercise was to attempt to simulate in a comprehensive manner the transport of historical ^{137}Cs discharges from the European nuclear fuel reprocessing facilities located in Sellafield (UK) and La Hague (France). This exercise included transport models driven by an ocean general circulation model (OGCM), that is a single, all-encompassing description of currents. Discharges from these European nuclear fuel reprocessing facilities were simulated through all affected marine areas of the European Shelf Seas and the North Atlantic Ocean, and for the entire period from the start of discharges to 2006. To attempt to achieve this objective, both large spatial and temporal scales were considered. In addition, biological uptake models were also coupled with the transport models. Where possible, modelled activity concentrations in seawater, sediment and biota were compared with measurement results obtained from the MARIS database¹.

Spatially, a model domain was defined in the North Atlantic Ocean extending longitudinally from 50°W to 25°E and latitudinally from 45.1°N to 75.1°N. Annual releases of ^{137}Cs from both facilities were compiled from 1952 to 2006; details are available via the IAEA's Database on Discharges of Radionuclides to the Atmosphere and the Aquatic Environment (DIRATA)² and OSPAR's Data and Information Management System³, as well as Ref. [18]. The transport of ^{137}Cs was simulated over the same timeframe. Additionally, ^{137}Cs fluxes from the Baltic Sea (relevant after the Chernobyl accident in 1986) and deposition from global fallout (uniform values for latitude bands) were considered.

Monthly averages of currents were calculated using the JAMSTEC (Japan Agency for Marine-Earth Science and Technology) OGCM for the Earth Simulator (OFES) model⁴. This model produces data on currents for 54 vertical levels of increasing depth intervals from the surface to the sea bottom with horizontal resolution of 0.1°. The performance of this circulation model has been assessed through comparison with ocean observations from several regions of the global ocean (including the North Atlantic Ocean) and found to be acceptable; the results are summarized in Ref. [19].

The OFES circulation data were used as input for running each transport model included in the exercise (i.e. the Lagrangian ESTE (Slovakia), LORAS (Republic of Korea), and USEV (Spain) models (all described in Ref. [3]), and the POSEIDON R (Ukraine) box model [20]). The NRPA (Norway) box model [8] was also included but its own estimations of currents were used. Time series of radionuclide activity concentrations in seawater, sediment and four classes of biota (i.e. phytoplankton, zooplankton, non-piscivorous and piscivorous pelagic fish) were calculated using each transport model for a number of defined areas in the European Shelf Seas and the North Atlantic Ocean. These defined areas included those regions where most of the radionuclides released from Sellafield and La Hague are known to have been transported, including the Celtic Sea, English Channel, North Sea (divided into 3 regions) and Norwegian coastal waters (divided into two regions). As an example, the temporal evolution of ^{137}Cs activity concentrations in biota in the southern Norwegian coastal area are illustrated in Fig. 3.

¹ Available online at <https://maris.iaea.org/>

² Available online at <https://dirata.iaea.org>

³ Available online at <https://odims.ospar.org/>

⁴ Available online at <http://www.jamstec.go.jp/ofes/>

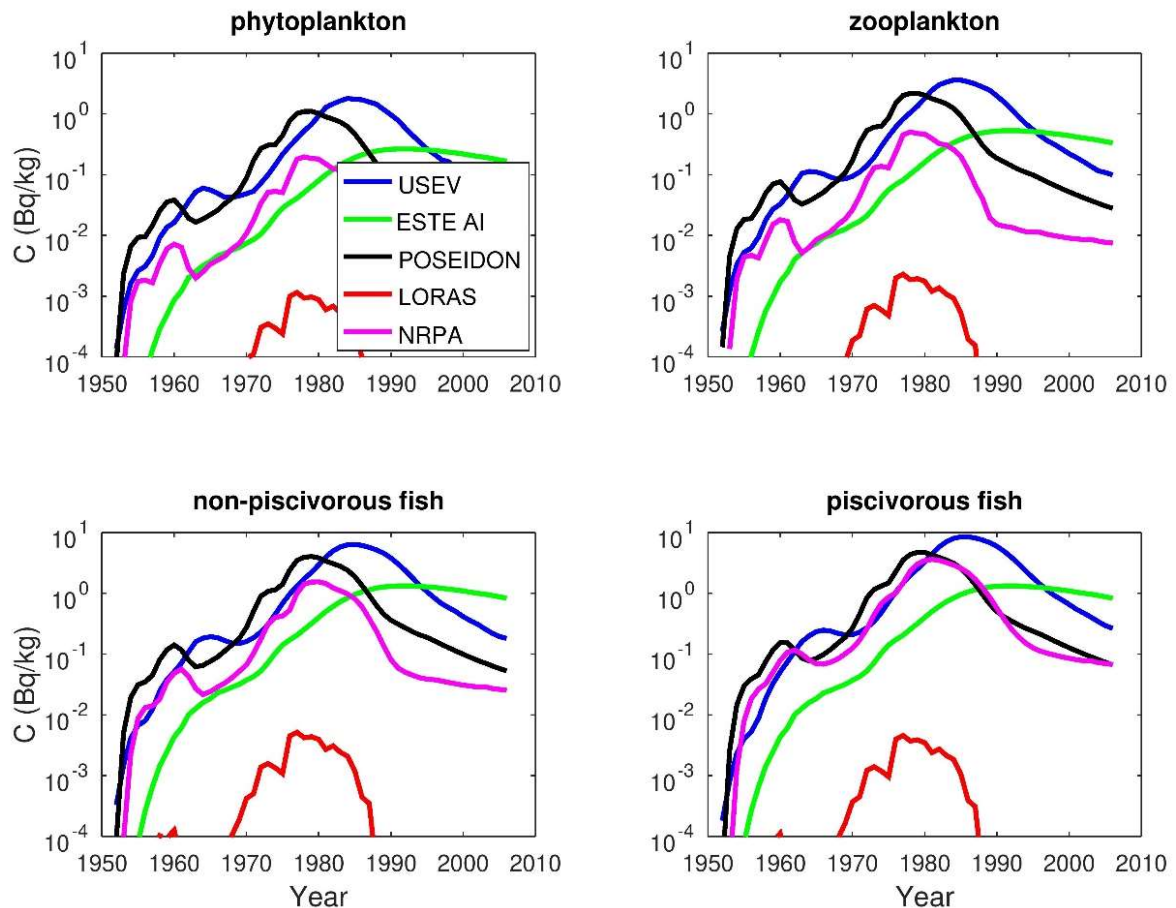


FIG. 3. Temporal evolution of ^{137}Cs calculated activity concentrations (Bq/kg fresh weight) in the biotic components of the model in the southern Norwegian coastal current using JAMSTEC circulation data.

Figure 3 shows that, apart from the LORAS model, the models produced a very consistent picture, with peak activity concentrations arising in the 1980s. The results of the LORAS model are not unexpected owing to the coarse resolution scale of the model being used for small, enclosed sea areas.

It is well known that the majority of the discharges from the Sellafield facility into the Irish Sea are advected towards the north, leaving the Irish Sea through the North Channel; they are subsequently advected around Scotland and enter the North Sea along the east coast of Britain. However, the results of all transport models included in this exercise indicated that most of the discharges were advected towards the south and onwards to the Celtic Sea, with only a minor flux towards the north. In other words, no transport model was able to simulate the well-known currents and radionuclide transport patterns in the Irish Sea. Thus, it was concluded that the circulation data from the OFES model were not sufficiently detailed to describe water currents within the Irish Sea.

Some alternative OGCMs were subsequently tested, including the HYCOM, FIO and NEMO models. HYCOM (HYbrid Coordinate Ocean Model), is a multi-institutional effort – by the HYCOM Consortium – sponsored by the National Ocean Partnership Program (NOPP), as a part of the U.S. Global Ocean Data Assimilation Experiment (GODAE) [21]. The FIO model

was developed by the First Institute of Oceanography (China) [22], and lastly, NEMO (Nucleus for European Modelling of the Ocean), is a state-of-the-art modelling framework for research activities and forecasting services in ocean and climate sciences, developed in a sustainable way by a European consortium⁵. An example of the application of NEMO is summarized in Ref. [23].

However, the application of alternative OGCMs did not significantly improve the results of the transport models. Most of the radionuclides released from the Sellafield facility continued to be wrongly simulated as travelling southwards. None of the models used was of sufficient spatial resolution to effectively describe water currents within the Irish Sea and water exchanges with the Atlantic Ocean through the North Channel. In some cases, further deviations from known transport patterns resulted. For example, HYCOM described the water flux through the Dover Strait (which connects the English Channel with the North Sea) less effectively than the JAMSTEC model. Most of the flux predicted using HYCOM was directed to the west, that is from the North Sea through the Dover Strait to the English Channel, while it is known that it ought to be directed to the east. This means that the simulated ¹³⁷Cs activity concentrations in the North Sea were underestimated, since most releases from the La Hague facility were simulated as not entering the North Sea but instead moving eastwards to the Celtic Sea. As an example of the results of simulations driven by HYCOM circulation data, Fig. 4 shows a comparison of calculated and measured (retrieved from the MARIS database) activity concentrations in the southern North Sea. (Note that measurement data for surface and deep seawaters are available from MARIS for this area, but not for bed sediments.) Despite conflicting with observations, it is interesting to note that there is relatively good agreement between the different transport models, both box and Lagrangian.

These conflicting results can be attributed to the relatively coarse spatial resolution of the circulation data provided by the OGCMs, approximately 0.1° in both longitude and latitude, which is about 10 km in the area simulated. A relatively coarse resolution is needed for OGCMs in order to cover the world ocean in a computationally efficient way, but it is clearly not sufficient to numerically describe currents in narrow sea areas and straits. It has to be noted that the North Channel in the Irish Sea is only 21 km wide at its narrowest section and is described by only two grid cells in each OGCM. Similarly, the narrowest section of the Dover Strait in the English Channel is 33 km.

An additional aspect so far not considered is the potential influence of tidal residual currents on transport within the Irish Sea. Tides produce an oscillating movement of water. If this movement is integrated along a tidal cycle there is a net transport in a given direction (which depends on the bathymetry, shape of the coastline and characteristics of the tide). This net transport is the so-called tidal residual. Tidal residual currents are very weak, typically only a few centimetres per second (cm/s), but may have a significant long term influence in areas with relatively large tides, such as the Irish Sea. A tidal model was applied by the University of Seville (the same as that applied in Ref. [24]) and the tidal residual was calculated for the main constituent in the Irish Sea, M₂, related to the direct gravitational effect of the Moon.

⁵ Available online at <https://www.nemo-ocean.eu/>

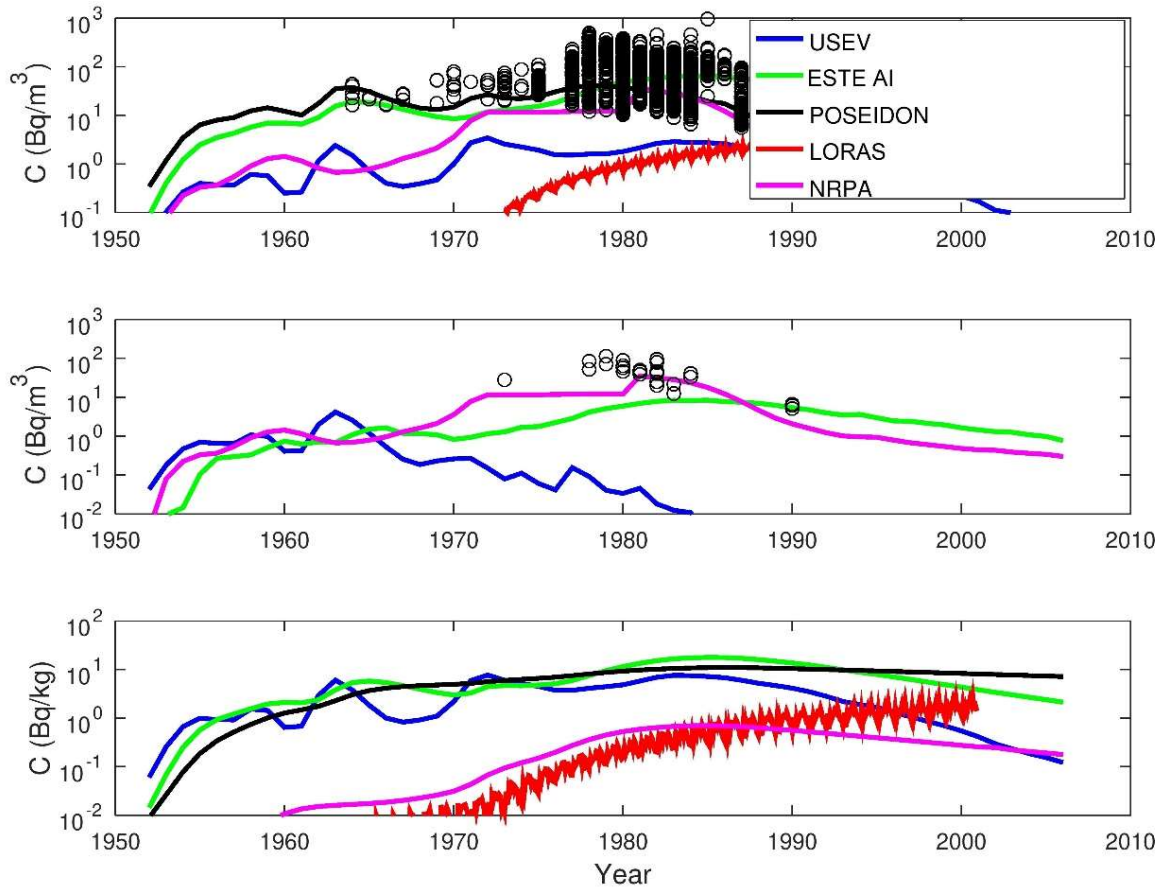


FIG. 4. Measured and calculated ^{137}Cs activity concentrations in the south North Sea in surface seawater (top), deep seawater (middle) and bed sediments (bottom). Measurements (points) were obtained from MARIS database.

Figure 5 illustrates the south–north component of the residual current in the Irish Sea. Only the positive values (thus indicating the current directed to the north) are depicted. Effectively, the tidal residual is mostly directed to the north, but currents are weak (i.e. a few cm/s, reaching the order of 10 cm/s in the area of the North Channel). The net flow through the North Channel calculated from these residuals is $7.5 \times 10^4 \text{ m}^3/\text{s}$. For comparison, measurements of high frequency radar complemented with ADCP (Acoustic Doppler Current Profiler) records indicate a flux of $8.0 \times 10^4 \text{ m}^3/\text{s}$ [25], which is relatively consistent with the model calculation.

Further research is necessary to investigate whether the tidal residuals related to the direct gravitational effect of the Sun (S_2) can dominate the fluxes provided by OGCMs and produce consistent fluxes through the straits. In a recent modelling study of ^{99}Tc [26], the importance of tidal advection was investigated by comparing transport when the model was run either with tides included or with tides filtered out. The results pointed to a systematic Lagrangian tidal drift in the Irish Sea and the North Sea that eventually impacted the ^{99}Tc activity concentration levels far downstream.

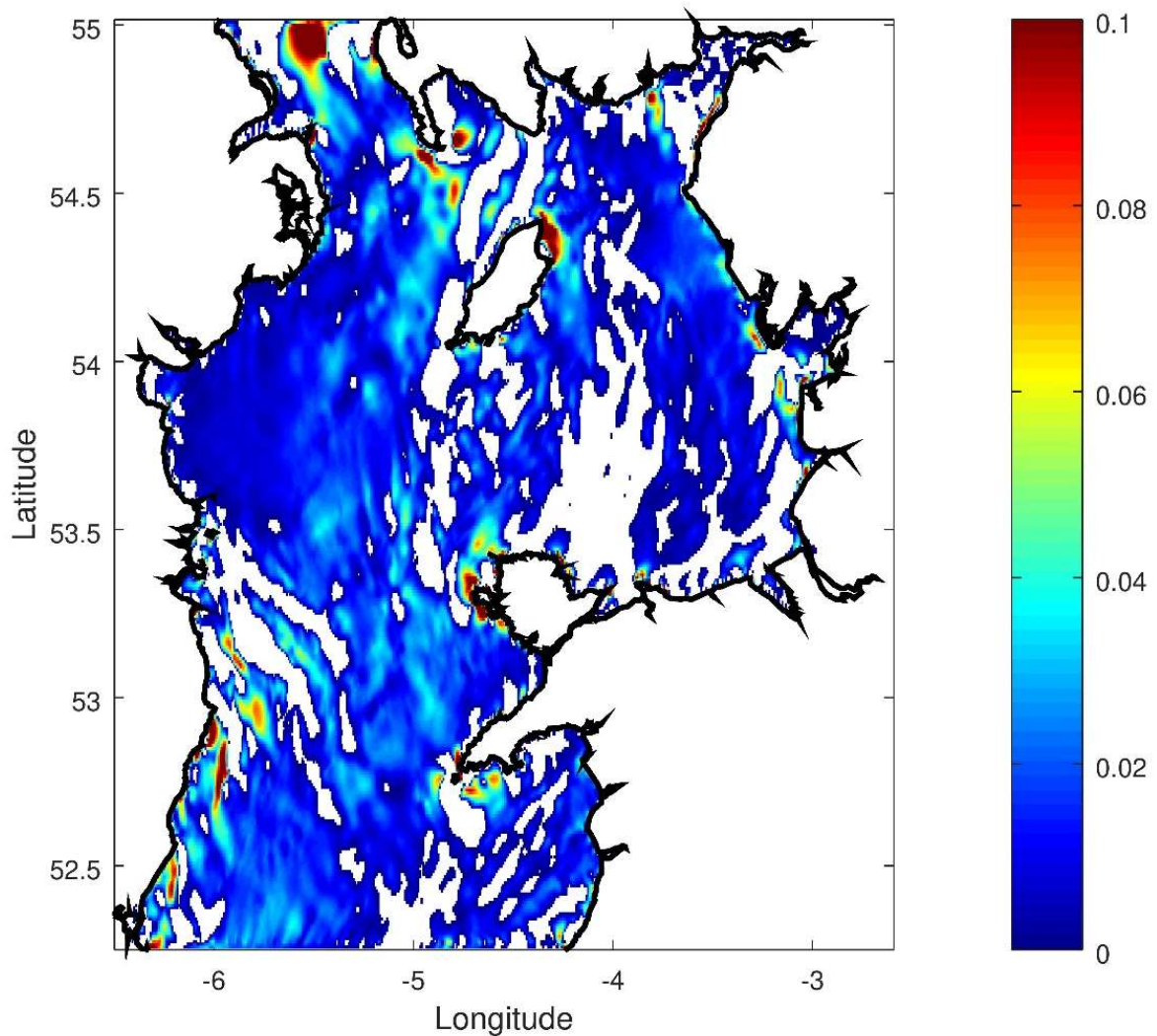


FIG. 5. South to north component (m/s) of the M_2 tidal residual current in the Irish Sea calculated with a tidal model. Only positive values are drawn.

A conclusion of this exercise is that care needs to be taken if currents derived from OGCMs are used for predicting transport in coastal waters, since the models might not adequately represent the complex behaviours in these regions. Instead, validated local forecasts of currents have to be used. Ideally, these local forecasts need to be available for all waters which could potentially be affected by a nuclear accident. This reinforces one of the main conclusions presented in Ref. [27].

These observations and conclusions have been well-known to the many marine modelling groups who have investigated radionuclide transport in seawater and transfer to different compartments of the marine environment in specific areas within the model domain for many years.

As far back as 2003, the benefits of increasing computer power for marine transport models were commented upon, and it was generally concluded that better spatial resolution improves the results in nearly all cases [28]. Discharges from Sellafield have been simulated in the Irish Sea [29–33] and discharges from both Sellafield and La Hague have been simulated in the

English Channel [34, 35] for many years using models carefully validated with measurements at different temporal and spatial scales, often nested within OGCMs for computational efficiency and for calculation of boundary conditions. For the Irish Sea in particular, M_2 and S_2 tidal residual currents, the seasonal stratification effect of the summer gyre in the Western Irish Sea [36] and influence of radionuclides remobilized from sediment [37] from are also routinely considered.

Nevertheless, it may be useful to continue to conduct research into a standard approach for the comprehensive simulation of the transport of radionuclides released from the nuclear fuel reprocessing facilities of Sellafield and La Hague into the Atlantic Ocean and European Shelf Seas. One possibility may be to use 'real' hydrodynamic circulation data (i.e. to compile monthly or weekly averages for a given year and to use these as representative of all years in a multiannual simulation) instead of the climatological data (i.e. averages from many years) such as that derived from the OFES, HYCOM, FIO and NEMO models which were applied in the simulations described here.

5. TESTING NUMERICAL INTERPOLATION SCHEMES IN LAGRANGIAN MODELS: PRELIMINARY RESULTS

Some preliminary exercises were carried out to analyse the differences between numerical schemes applied in Lagrangian transport models. The motivation for this work was that these Lagrangian techniques are the most commonly applied in transport models developed for radiological assessments of incidents or emergencies.

A radionuclide release is simulated in a Lagrangian model by means of a number of particles. Each particle represents a number of units (for instance Bq); the trajectory followed by each particle is calculated, and from the position of particles at a given time, the corresponding radionuclide activity concentration is obtained [3]. Diffusion is calculated in Lagrangian models using a stochastic method and advection (transport due to the water current) is calculated from a simple equation, which is actually the definition of velocity:

$$\vec{r}(t + \Delta t) = \vec{r}(t) + \vec{v} \cdot \Delta t \quad (2)$$

where r is the position vector of the particle and v is water velocity vector at the particle position. This vector equation is written as three scalar equations (also adding diffusion terms) in the following form:

$$\begin{aligned} x(t + \Delta t) &= x(t) + u\Delta t + \frac{\partial K_h}{\partial x} \Delta t + \sqrt{2K_h} dW_x \\ y(t + \Delta t) &= y(t) + v\Delta t + \frac{\partial K_h}{\partial y} \Delta t + \sqrt{2K_h} dW_y \\ z(t + \Delta t) &= z(t) + w\Delta t + \frac{\partial K_v}{\partial z} \Delta t + \sqrt{2K_v} dW_z \end{aligned} \quad (3)$$

where (u, v, w) are the three components of the water velocity vector in the directions (x, y, z) , and K_h and K_v are, respectively, the horizontal and vertical diffusion coefficients. Usually, x and y are defined in the east-west and south-north directions respectively, and z is measured downwards from the sea surface.

Horizontal and vertical diffusion coefficients are different. Actually, in the marine environment K_h is much greater than K_v owing to the following two reasons:

- (1) The sea presents stable stratification, thus the vertical movements of water are much more difficult (energy demanding) than horizontal movements;
- (2) The ocean extends much further horizontally (thousands of km) compared to vertically (a few km). Thus much larger eddies may form horizontally which enhance horizontal mixing.

The derivatives of the diffusion coefficients are normally introduced to avoid the artificial accumulation of particles in regions of low diffusivity. However, zero or constant diffusion coefficients were used in the exercises reported here and thus these terms are not relevant.

The last variable in Eq. (3) describe mixing. W_x , W_y and W_z are independent components of the stochastic motion, which have zero mean and variance dt :

$$\overline{dW_x^2} = \overline{dW_y^2} = \overline{dW_z^2} = dt \quad (4)$$

and for a finite time step Δt they can be simulated as $\Delta W_x = R_x (\Delta t)^{1/2}$, $\Delta W_y = R_y (\Delta t)^{1/2}$ and $\Delta W_z = R_z (\Delta t)^{1/2}$, where (R_x, R_y, R_z) are normally distributed random variables having zero mean and a standard deviation of one.

Thus, the position of a particle at the new time step is calculated from its position in the previous time step, its velocity at the initial particle position plus the corresponding diffusion terms.

Water velocities are discrete values provided by hydrodynamic models that are only defined over the nodes of the computational grid used in the ocean circulation model. Since the particles are not necessarily located in nodes, but anywhere, some interpolation is needed to evaluate water velocity at the particle position from velocities at the nearest points. Similarly, interpolation in time may be needed since water velocities are provided only at given moments (e.g. monthly values, while the time step in the model may be one hour). Several approaches are available to carry out interpolations, from the simplest, consisting of directly using velocity at the nearest point (no interpolation), to more complex interpolation schemes (linear or quadratic).

In addition, a grid is not needed to evaluate diffusion and advection in a Lagrangian model, but it is needed to calculate the radionuclide activity concentrations from the position of particles, that is grid cells are defined, then the number of particles inside each cell is counted and the resulting concentration C is the total activity within the cell divided by the cell volume V :

$$C = \frac{1}{V} \sum_{i=1}^N R_i \quad (5)$$

where in the case of a regular grid and the simpler case of a 2-D depth averaged model, the cell volume is $V=dx \times dy \times H$. Here dx and dy are cell dimensions in the west–east and south–north directions and H is the mean water depth of the cell. N is the number of particles within the cell and R_i is the activity in Bq corresponding to particle i . It was also investigated, during this exercise, if the cell dimensions selected (dx and dy) have a significant effect in the resulting activity concentrations.

The purpose of the exercises reported here was to compare interpolation schemes applied by the different modelling teams using Lagrangian models by means of a series of simple numerical exercises in order to evaluate if the choice of scheme could have a significant effect on the model results.

A simple 1-D case was initially considered that consists of a uniform channel (constant geometry) in which a tide propagates. Thus, water velocities change along the channel and with time. The tidal channel is 100 km long, the period of the tide is 12 hours and its amplitude is 0.25 m. The channel depth is constant and equal to 1 m and its width is also constant, but this number is not relevant since the exercise is 1-D and thus results are averaged over the channel width. From the analytical solution of the 1-D wave equation, water velocities and surface elevations along the channel can be calculated at any time and position. These data are provided in 12 files (one per hour) to be read by the transport codes. The channel is divided into 1000 cells (segments in this case) with $dx=100$ m. Each file contains the water velocity and elevation in the centre of each cell. In addition to the tide, a net residual current (net transport) to the right, equal to 0.1 m/s, is included. The 12 files contain velocity and elevation profiles along the channel at $t=1$ h, $t=2$ h, etc. The sequence is repeated to simulate as many days as needed. For example, two of the velocity profiles along the channel, as provided to the modellers, are shown in Fig. 6.

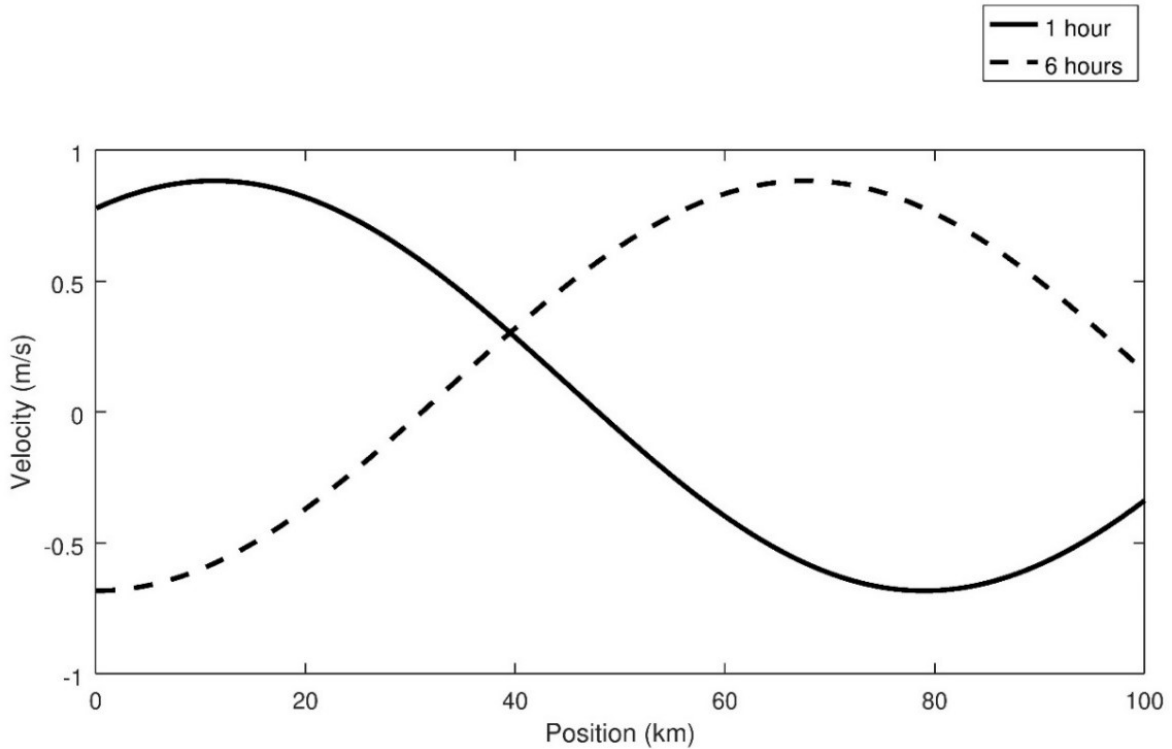


FIG. 6. Velocity along the channel at $t=1$ h and $t=6$ h.

A constant horizontal diffusion coefficient equal to $1 \text{ m}^2/\text{s}$ is considered and no vertical diffusion is added. An initial deposition of $100 \text{ Bq}/\text{m}^2$ was assumed to occur on the first 500 m of the channel over its whole depth (note that depth is not 1 m, since there will be a surface elevation due to tides). The endpoint of the exercise is to provide a profile of radionuclide concentration along the channel length after 6 tidal cycles (i.e. 72 hours).

Initially, the dependence of results on the channel segment length used to evaluate radionuclide activity concentrations was studied with the Lagrangian model developed by USEV (see Ref. [38]). For computational economy in doing this, it was considered acceptable to use the same grid applied to generate water circulation for evaluating radionuclide activity concentrations. Thus, in this case, a value for dx of 100 m was assumed. Other values for dx were tested and the results are presented in Fig. 7 which also shows the final radionuclide activity concentrations along the channel after 72 hours for a number of different values of dx . The graphic has been split into two separate panels to improve clarity. It can be seen that for dx values smaller than 10 m there is a high degree of variability in the calculated activity concentrations. A large value for dx (500 m) resulted in a significant artificial diffusion (lower peak activity concentrations and a wider distribution) as well as the centre of the distribution being displaced downstream. In this particular case, reasonable results (low variability and relatively small artificial diffusion) were obtained for $dx=100$ m, but it seems that, in general, the optimum grid size used to evaluate radionuclide activity concentrations from the distribution of particles in a Lagrangian model has to be tested.

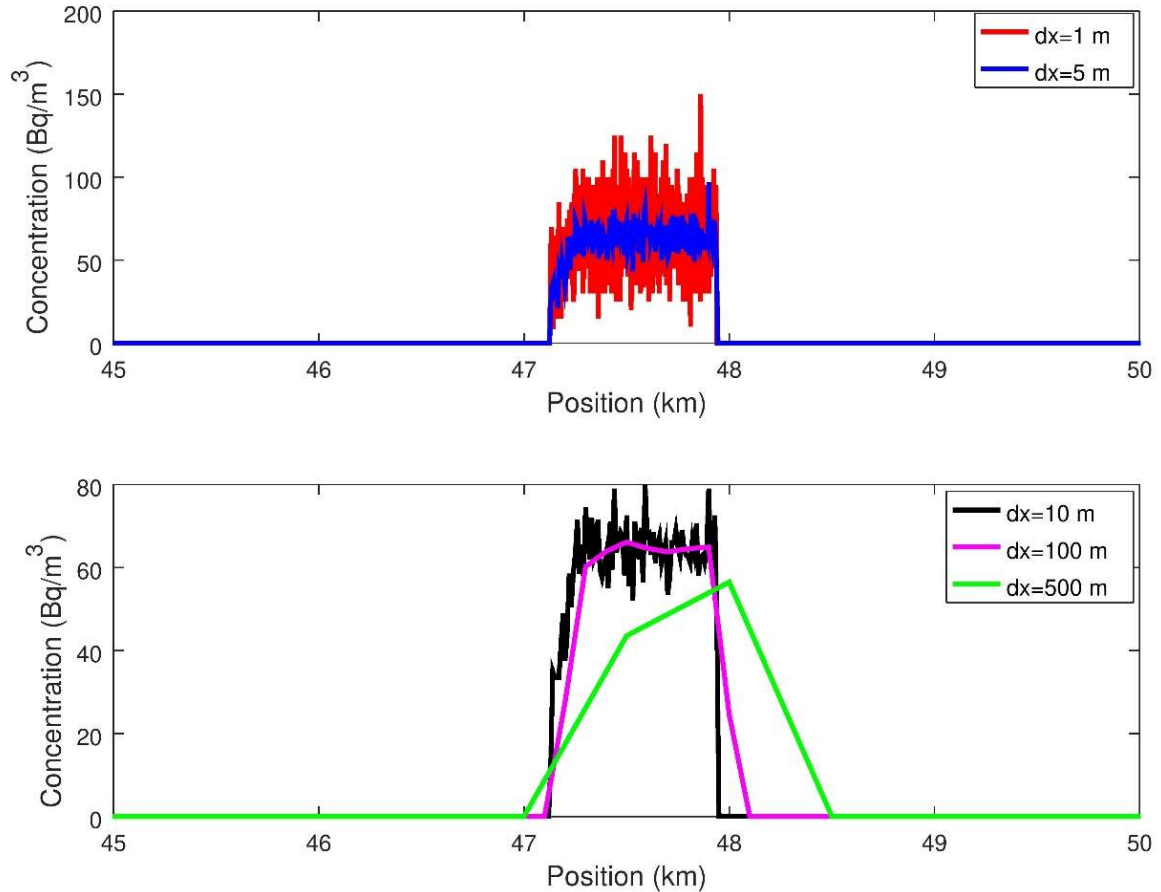


FIG. 7. Radionuclide activity concentrations along the channel calculated for different values of dx .

Subsequently, the approaches of the teams from ABmerit, Defence Academy of the United Kingdom (AoD), Institute of Mathematical Machines and Systems Problems (IMMSP), Japan Atomic Energy Agency (JAEA) and Korea Atomic Energy Research Institute (KAERI) participating in the Working Group were tested. A single particle test was initially carried out prior to performing the deposition exercise described above. Diffusion was not considered in order to avoid masking differences in interpolation schemes by turbulent mixing. Consequently, a single particle was assumed to be released on the surface (although this is not relevant) at position $x=10$ km from the left side of the channel at $t=0$. Its position was calculated during 6 tidal cycles (72 hours), as well as the water velocity at the point where the particle is located at each moment of time. All applied models used linear interpolation in both time and space.

Results of these calculations are illustrated in Fig. 8, where it is shown that there was very good agreement in the output of models, both in the cases of the evolution of the particle position (top panel) and in the water velocity at the particle position (bottom panel). The step or 'staircase curve' observed in velocity in the ABmerit model was due to the fact that only spatial interpolation was used. (In fact, this model applies linear interpolation in time and space, but only spatial interpolation is shown here as a comparison.) In any case, the staircase curve fitted closely to the curves produced when temporal interpolation was included. In this simple case all models produced essentially the same results, thus it seems that the mathematical description of the interpolation schemes, as applied by each team, do not affect the results.

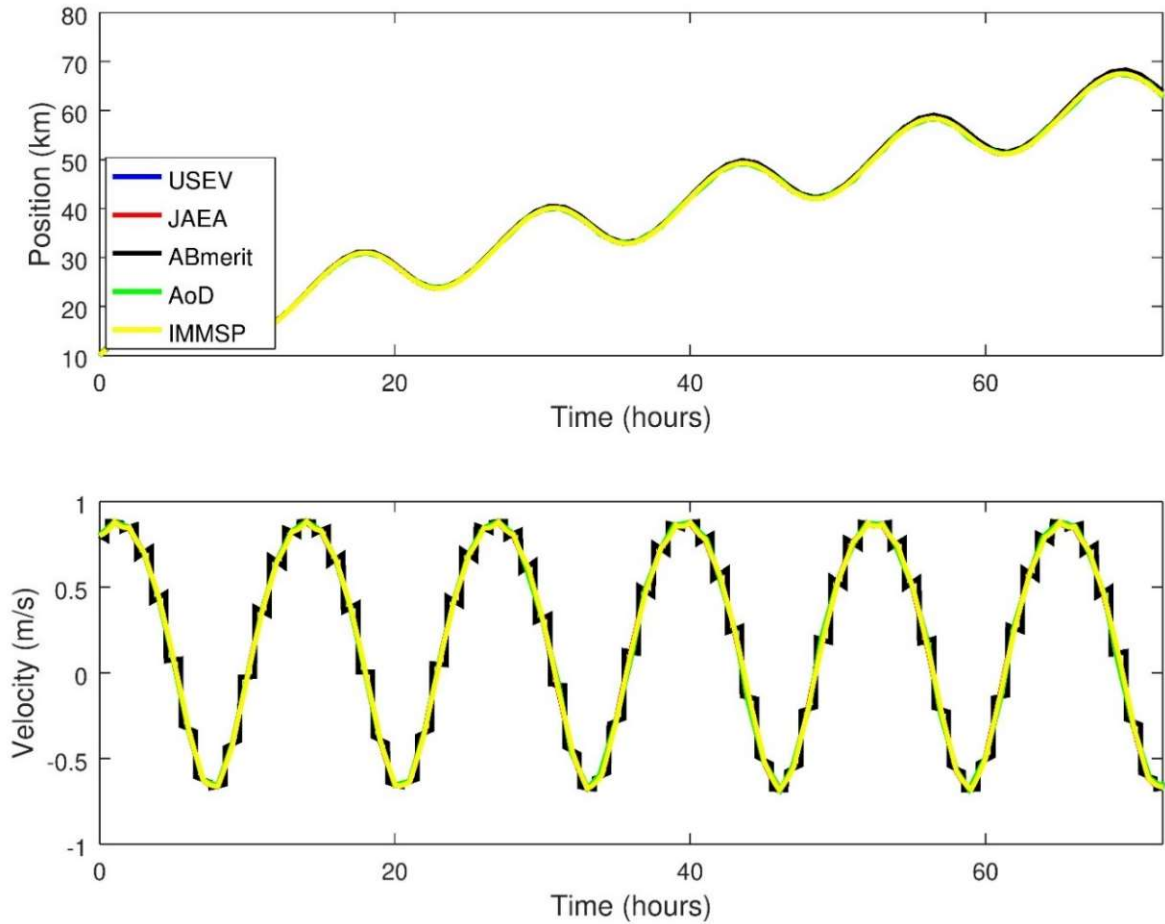


FIG. 8. Temporal evolution of the particle position (top) and velocity (bottom) as calculated by the different models. Position is measured from the left side of the channel.

To consider deposition, horizontal diffusion has to be added to the advection scheme. As mentioned previously and also described in detail in Refs [3, 5], diffusion is treated with a stochastic method in Lagrangian models.

The results of the deposition exercise are presented in Fig. 9, in which the initial radionuclide distribution is also shown. From this figure it can be seen that the models produced quite similar predictions of the amplitude of the peak. However, the predicted positions of the peak activity concentration and widths of the distribution were not the same.

Since there were no differences between models when considering only advection (Fig. 8), the slightly different results observed in Fig. 9 can probably be attributed to the particular numerical scheme used in each model to describe diffusion and it was concluded that this involves more detailed investigation. The differences were generally quite small, but in a much more complex case (e.g. 3-D movement in the ocean) it is possible that these could be significantly amplified.

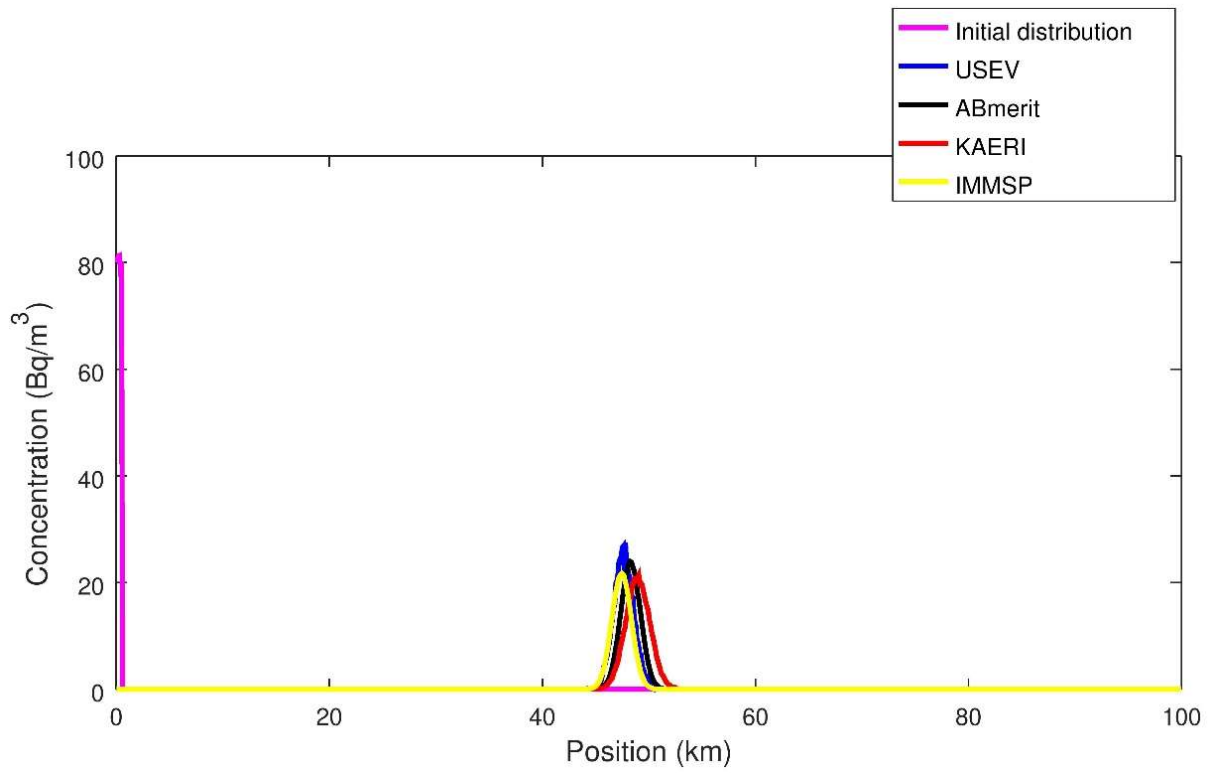


FIG. 9. Radionuclide distribution along the channel 72 hours after the initial deposition.

6. CONCLUSIONS

The work carried out by MODARIA II Working Group 7 on Assessment of Fate and Transport of Radionuclides Released in the Marine Environment was a continuation of that undertaken during the MODARIA I programme (2012–2015) by Working Group 10 on Modelling of Marine Dispersion and Transfer of Radionuclides Accidentally Released from Land Based Facilities [3].

In general, three types of transport models are available: box, Eulerian and Lagrangian. Water–sediment interactions and biota uptake of radionuclides can be treated using either an equilibrium approach or, alternatively, a dynamic model. The relative advantages of each methodology have been analysed and, for the case of radiological assessment following incidents or emergencies involving releases of radionuclides into the marine environment, information was provided to identify the most suitable approach for each particular situation and endpoint of the simulation.

One of the objectives of MODARIA II Working Group 7 was to improve radionuclide transport modelling for the marine environment through the addition, in manageable ways, of processes not previously implemented. This was done through long term simulations of releases from the FDNPS accident in the Pacific Ocean at oceanic scale and at a longer temporal scale than was carried out during the MODARIA I programme. Biological uptake models were coupled to the transport models. Model–model and model–measurement comparisons of ^{137}Cs activity concentrations in seawater, sediment and biota (zooplankton, phytoplankton, non-piscivorous and piscivorous fish) were undertaken and, in general, although there was good agreement, there is still room for improvement.

Application of the same types of model for simulation of the historical ^{137}Cs releases from European nuclear fuel reprocessing facilities (i.e. Sellafield and La Hague) in the north Atlantic Ocean and European Shelf Seas was not so successful. The transport models were driven by ocean general circulation models (OGCMs). In this case, the poor performance of the transport models in describing measured activity concentrations was attributed to the relatively coarse resolution (about 10 km) of the OGCMs from which water circulation data were obtained and to the use of climatological data. Although these models satisfactorily calculate currents in the open ocean, the spatial resolution is not high enough to describe narrow areas and straits which connect different areas of the European shelf (in particular the North Channel, connecting the northern Irish Sea with the Atlantic, and the Dover Strait, which connects the English Channel and the North Sea). Since fluxes through these narrow areas and straits are not correctly calculated by the circulation models, subsequent simulations of radionuclide dispersion are also not correct. This highlights the need for care to be taken when using currents derived from OGCMs to predict transport in coastal waters, since the resolution might not be adequate. Instead, carefully tested local forecasts of currents need to be used and these have to be available ideally for all coastal waters which could potentially be affected by a nuclear or radiological incident or emergency. In contrast, currents derived from OGCMs could be used to predict the fate of unplanned radionuclide releases in the open ocean.

A study of the description of water–sediment interactions was carried out using theoretical analysis and numerical experiments in order to compare the performances of several possible descriptions of these interactions. One step and two step kinetic models were compared, as well as descriptions of the bed sediment as a single layer or as multi-layered sediment. Since it is always preferable to use the simplest possible model, information is provided on when one step

and single layer models could be used, which depends on the temporal scale of the simulations and their particular objectives.

In addition, the k_d model was also assessed and some information on its use is given. The conclusion of this work is that the use of an equilibrium model in the area close to a radionuclide release (either unplanned or chronic) is not appropriate, since equilibrium conditions are not found in such areas due to the fact that the sea is an open environment affected by external perturbations. Some indications on good practice for measuring k_d in the field and on the necessary information for the use of distribution coefficients in modelling are given. For instance, it is necessary to know whether it is a suspended matter–seawater or a bed sediment–seawater k_d , since these may vary in orders of magnitude simply because of geometrical considerations.

A semi-empirical equation was proposed to handle changing environmental conditions (pH and salinity) in numerical models for radionuclide transport. These changing conditions may appear in estuaries, where salinity increases from freshwater to marine values, and in coastal areas close to industrial facilities, where changes in pH could occur as a result of waste releases.

A simple 1-D exercise was carried out in order to analyse differences in numerical schemes applied in Lagrangian models, since these models are commonly applied for emergency assessments. It was found that model results are sensitive to the grid spacing used to evaluate radionuclide concentration from the distribution of particles. Fine grids produce noisy results with high variability. In contrast, coarse grids lead to high artificial diffusion. It seems that the optimum grid spacing could be found for each particular situation, but this involves further investigation. The models compared produced the same results when only considering advection, but slight differences appeared when diffusion was added. These differences could be significantly larger in a true complex marine transport problem and might be due to the numerical scheme used in each model to simulate diffusion. However, it was concluded that this topic needs further research.

Finally, a completely exploratory model comparison exercise was carried out comparing the radionuclide activity concentrations in seawater and sediment resulting from routine discharges from a hypothetical (generic) NPP into the waters off the Atlantic coast of the USA. Three models were applied, that is a Eulerian model customized to the region (ESTE AI), the POSEIDON-R box model customized to the region and another box model specifically designed for the area. The only information provided to the modelling teams was radionuclide release data; all other relevant information (e.g. depths, water currents and fluxes, suspended matter concentrations, sedimentation rates) was obtained independently from a literature search or personal expertise by the modelling team. The three models have model compartments of differing spatial resolutions. However, in spite of this, the predicted activity concentrations in seawater, sediment and biota were within an order of magnitude for the radionuclides considered.

APPENDIX: EXPLORATORY INTERCOMPARISON OF MODELS APPLIED TO A HYPOTHETICAL GENERIC NUCLEAR POWER PLANT SCENARIO

The results of a collaboration with MODARIA II Working Group 3 on Assessments and Control of Exposures to the Public and Biota for Planned Releases to the Environment are reported in this Appendix. The objective of the collaboration was to explore the simulation of the transport of radionuclides released from a hypothetical (generic) NPP as routine discharges into coastal waters of the USA with three models that have model compartments of differing spatial resolutions.

The predicted results were average activity concentration of radionuclides in seawater, sediment and biota for defined coastal areas or boxes on several spatial scales ranging from small (a few km) to large (tens of km). Three models were used, that is the POSEIDON-R box model (customized for use on this problem by IMMSP, Ukraine), the USEV box model (which was specifically designed for the exercise), and a Eulerian model developed and customized to this region by ABmerit (the ESTE AI model). The three models are briefly described in Sections A.1–A.3 .

Hypothetical annual discharges of several radionuclides for the year 2016 were provided, which were used as the source term for the models. These discharge data are provided in Section A.4. The most interesting aspect of this set of calculations is that the only information provided to the participants was the radionuclide releases. Thus, each modelling team had to investigate the parameters (e.g topography, hydrodynamics, sediment properties) to use in the area where according to the scenario, the hypothetical radionuclide releases occur. The area selected was the area of Long Island Sound (LIS) located off the Atlantic east coast of the USA. The customization of models to this region is described briefly below.

The configuration of the different models, as well as the parameters driving them, were selected by each modelling team from a literature review or from personal experience. The results for C_w and A_s , activity concentrations in seawater and sediments, respectively, are presented in Table 1. Box 1 results (small coastal box) are presented for the USEV model (see Section A.1). In the case of POSEIDON-R, only results for the large coastal box (see Section A.2) are presented, since, as the two boxes are similar in size and water exchanges, the predictions of this model are very similar for both boxes. The results provided by the Eulerian ESTE AI model (see Section A.3) are activity concentrations in close vicinity to the NPP, represented by the discharge cell (dimensions 2 km × 2 km).

It was demonstrated that there was relatively good agreement between the three models, as the predicted activity concentrations were generally within the same order of magnitude for both seawater and sediments.

Predictions for biota were reported only by the ESTE AI and POSEIDON-R models and these are presented in Table 2 for radionuclides which were calculated by both models. Predictions were, in general, in agreement within an order of magnitude.

TABLE 1. RADIONUCLIDE ACTIVITY CONCENTRATIONS IN SEAWATER AND SEDIMENT CALCULATED IN THE AREA OF THE NPP BY THE THREE APPLIED MODELS

Radionuclide	POSEIDON-R		USEV		ESTE AI (Eulerian)	
	C _w (Bq/m ³)	A _s (Bq/kg)	C _w (Bq/m ³)	A _s (Bq/kg)	C _w (Bq/m ³)	A _s (Bq/kg)
Cs-137	2.63 × 10 ⁻³	2.94 × 10 ⁻³	3.28 × 10 ⁻³	1.31 × 10 ⁻³	1.2 × 10 ⁻²	5.2 × 10 ⁻³
Cs-134	2.65 × 10 ⁻⁴	4.18 × 10 ⁻⁵	3.27 × 10 ⁻⁴	1.31 × 10 ⁻⁴	1.2 × 10 ⁻³	5.1 × 10 ⁻⁴
H-3	1.85 × 10 ²	3.4 × 10 ⁻¹	2.53 × 10 ²	2.53 × 10 ⁻²	9.18 × 10 ²	1.5 × 10 ⁻³
Mn-54	4.97 × 10 ⁻⁵	1.39 × 10 ⁻³	9.78 × 10 ⁻⁵	1.96 × 10 ⁻²	5.1 × 10 ⁻³	6.9 × 10 ⁻⁴
Co-60	7.04 × 10 ⁻⁴	1.66 × 10 ⁻²	1.90 × 10 ⁻³	5.70 × 10 ⁻²	1.3 × 10 ⁻²	3.9 × 10 ⁻³
Ni-63	1.13 × 10 ⁻³	7.20 × 10 ⁻³	1.47 × 10 ⁻³	2.94 × 10 ⁻³	NR	NR
Fe-55	8.90 × 10 ⁻⁶	1.1 × 10 ⁻¹	2.35 × 10 ⁻⁴	7.1 × 10 ⁻¹	NR	NR
Zr-95	7.84 × 10 ⁻⁷	4.55 × 10 ⁻⁶	6.16 × 10 ⁻⁷	1.23 × 10 ⁻⁴	7.1 × 10 ⁻⁵	1.7 × 10 ⁻⁵
Ag-110m	1.15 × 10 ⁻⁴	1.41 × 10 ⁻⁵	1.41 × 10 ⁻⁴	1.41 × 10 ⁻⁴	6.2 × 10 ⁻⁴	7.3 × 10 ⁻⁶
Sb-125	4.93 × 10 ⁻²	5.86 × 10 ⁻³	6.26 × 10 ⁻²	1.25 × 10 ⁻²	2.2 × 10 ⁻¹	9.1 × 10 ⁻²
I-131	2.42 × 10 ⁻⁴	7.91 × 10 ⁻⁸	2.84 × 10 ⁻⁴	1.99 × 10 ⁻⁶	1.2 × 10 ⁻³	7.6 × 10 ⁻⁹
Co-58	8.73 × 10 ⁻⁵	8.45 × 10 ⁻⁵	6.15 × 10 ⁻⁵	1.84 × 10 ⁻³	1.5 × 10 ⁻³	3.4 × 10 ⁻⁴

Note: NR = not reported.

TABLE 2. CALCULATED ACTIVITY CONCENTRATIONS IN BIOTA (Bq/kg WET WEIGHT) FOR SEVERAL RADIONUCLIDES BY POSEIDON-R AND ESTE AI MODELS

Radionuclide	Crustaceans		Molluscs		Algae	
	POSEIDON-R	ESTE AI	POSEIDON-R	ESTE AI	POSEIDON-R	ESTE AI
Ag-110m	3.06 × 10 ⁻²	1.2 × 10 ⁻¹	4.4 × 10 ⁻²	3.6 × 10 ⁻²	2.24 × 10 ⁻³	3.0 × 10 ⁻³
Co-58	1.18 × 10 ⁻³	6.4 × 10 ⁻³	2.26 × 10 ⁻³	1.8 × 10 ⁻²	2.1 × 10 ⁻³	5.5 × 10 ⁻³
Co-60	9.29 × 10 ⁻³	5.5 × 10 ⁻²	1.77 × 10 ⁻²	1.6 × 10 ⁻¹	1.65 × 10 ⁻²	4.7 × 10 ⁻²
Cs-134	5.76 × 10 ⁻⁵	5.8 × 10 ⁻⁵	7.73 × 10 ⁻⁵	6.9 × 10 ⁻⁵	5.11 × 10 ⁻⁵	5.8 × 10 ⁻⁵
Cs-137	5.71 × 10 ⁻⁴	5.8 × 10 ⁻⁴	7.66 × 10 ⁻⁴	7.0 × 10 ⁻⁴	5.07 × 10 ⁻⁴	5.8 × 10 ⁻⁴
I-131	2.42 × 10 ⁻³	3.6 × 10 ⁻⁶	6.5 × 10 ⁻³	1.2 × 10 ⁻⁵	1.11 × 10 ⁻²	1.2 × 10 ⁻²
Mn-54	1.34 × 10 ⁻²	5.1 × 10 ⁻³	1.93 × 10 ⁻²	5.1 × 10 ⁻²	1.17 × 10 ⁻³	6.1 × 10 ⁻³
Sb-125	0.26	6.3 × 10 ⁻²	0.37	6.3 × 10 ⁻²	3.81 × 10 ⁻³	4.2 × 10 ⁻³
Zr-95	2.59 × 10 ⁻⁴	2.8 × 10 ⁻⁶	3.69 × 10 ⁻⁴	7.0 × 10 ⁻⁵	9.49 × 10 ⁻⁶	4.2 × 10 ⁻⁵
H-3 (HTO)	0.64	0.89	0.64	0.89	0.64	0.89

A.1. USEV MODEL

Releases of radionuclides into coastal waters from the NPP are simulated by means of a simple two box model specifically designed for the particular site of interest. A small box (Box 1) covers the release area and a larger box (Box 2) covers the entire LIS. Moreover, the second box exports radionuclides to the open Atlantic Ocean.

The geography of the area and the structures of the boxes are shown in Fig. 10. Water depths were taken from GEBCO08 global bathymetry data⁶, with a spatial resolution of 30 seconds of arc in longitude and latitude. Bathymetry data allow the calculation of surface area, volume and mean water depth of each box. Releases occur inside Box 1 (see Fig. 10).

As shown in Fig. 10, Box 1 (where radionuclides are introduced) exchanges water with Box 2, which in turn exchanges water with the open Atlantic Ocean through the east and west open boundaries (F_e and F_w). There is an inflow of fresh water into LIS from river discharge, runoff and precipitation. In addition, there is outflow due to evaporation. However, these processes are not considered. It is assumed that the river inflow, R , exactly compensates the net outflow to the Atlantic Ocean ($R=F_e+F_w$). Thus, the volume of water in the boxes remain constant. Water fluxes between Box 1 and Box 2 are due to the residual (mean) circulation within LIS, which is denoted as Φ . All water fluxes used in this exercise are derived from selected published literature (see Refs. [39–41]).

The model is schematically represented in Fig. 11, where S is the source of radionuclides from the NPP. The deposition of atmospheric releases on the sea surface are not considered. Equations representing the rate of change of activity within each box, in both water and sediment, have a standard form in box models [3]. Since the present model was specifically designed for these calculations, the equations are summarized below (see Eqs 6–9). Furthermore, it has to be noted that transfer to biota is not included in this model.

In addition to water fluxes between the boxes, and the subsequent radionuclide transfers, interactions with suspended matter and bed sediments are also considered. Uptake–release reactions of radionuclides between seawater and suspended matter and sediments are also included. Furthermore, suspended matter particles are deposited on the seabed at a given sedimentation rate, where this process also produces an additional radionuclide flux to the bed sediments. Uptake and release reactions are described in a dynamic way, valid outside equilibrium conditions [3]. The desorption rate has been fixed as $k_2=1.16\times 10^{-5} \text{ s}^{-1}$ according to the experiments presented in Ref. [42], which is the value that has been used in other modelling investigations [3]. The k_d values, needed to evaluate the adsorption rate [6], are taken as the recommended values for ocean margins in Ref. [43]. Suspended sediment activity concentrations and sedimentation rates in LIS have also been derived from the literature [44, 45]. A summary of the model's box characteristics is given in Table 3 below.

⁶ Available online at https://www.gebco.net/data_and_products/gridded_bathymetry_data/

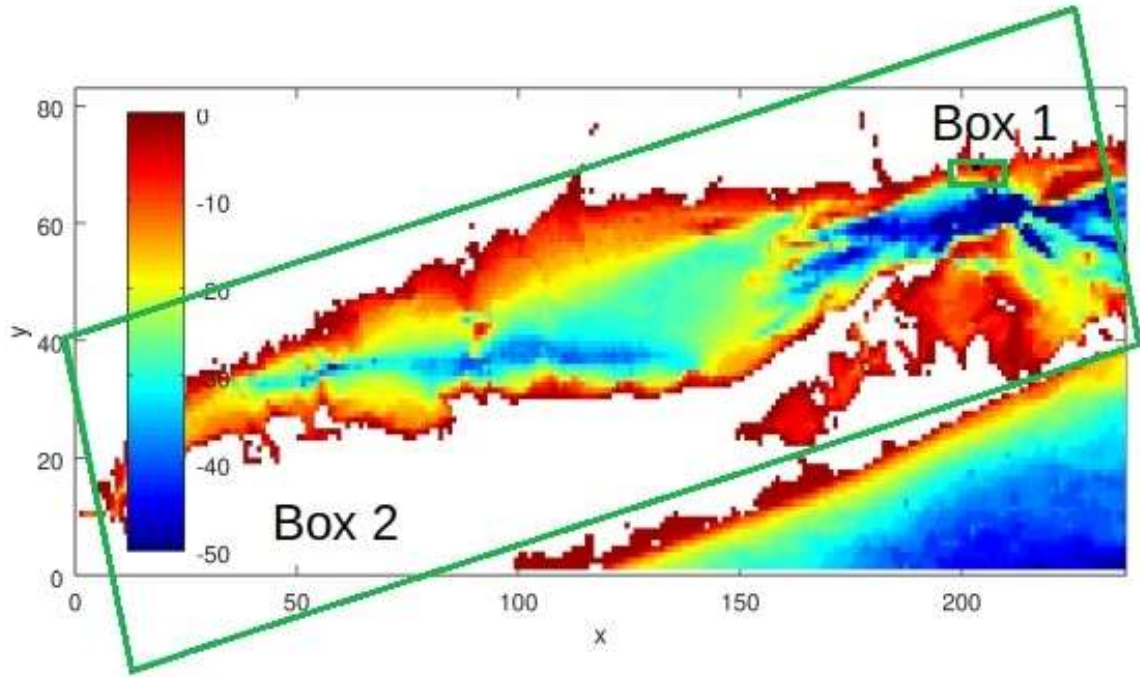


FIG. 10. The Long Island Sound area. Water depths in the colour bar are given in m. The two boxes are indicated by the green rectangles. Units correspond to 0.70 km and 0.93 km in the x (west–east) and y (south–north) directions respectively.

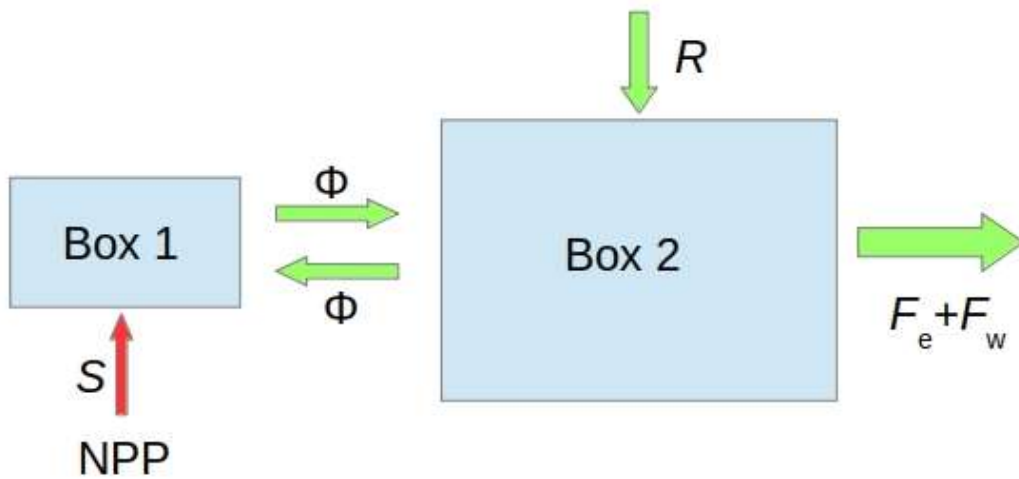


FIG. 11. Representation of the USEV box model.

TABLE 3. MAIN PARAMETERS IN THE USEV BOX MODEL

Parameter	Box 1	Box 2
Surface area (km ²)	18.8	4011
Volume (km ³)	0.138	75.1
Mean depth (m)	7.3	18.7
Flux F (m ³ /s)	0	835
Flux Φ (m ³ /s)	1016	1016
SR (g cm ² /y)	0.040	0.040
spm (mg/l)	4.5	4.5

The equation that gives the temporal evolution of radionuclide concentration in water (dissolved) in Box 1 due to water fluxes, interactions with the solid phases and radioactive decay is:

$$\frac{dC_i}{dt} = \frac{S_i}{V_i} + \Phi \frac{C_j - C_i}{V_i} - \frac{C_i}{V_i} F_i + I_i - \lambda C_i \quad (6)$$

where $i \neq j$, S_i is the radionuclide input (which occurs in Box 1, hence $S_2=0$) and fluxes F_i are: $F_1=0$; $F_2=F_e+F_w=835$ m³/s, as shown in Table 3. V is the water volume in the box and λ is the radioactive decay constant of the radionuclide. Finally, I_i is the term describing interactions with the solid phases, which is written as follows:

$$I_i = k_2 m_i (A_i - k_d \alpha C_i) + k_2 spm (P_i - k_d C_i) \quad (7)$$

where A_i and P_i are, respectively, radionuclide concentration in sediment and suspended matter particles, spm is suspended matter concentration (the same in both boxes) and α is a correction factor that takes into account that part of the sediment particle surface may be hidden by other sediment particles. Consequently, the surface available to adsorb radionuclides is smaller than if particles were suspended in the water column [6] and has been taken as $\alpha = 0.1$.

For radionuclide concentration in suspended matter, the equation is:

$$\frac{dP_i}{dt} = k_2 (k_d C_i - P_i) - \lambda P_i \quad (8)$$

and for the bed sediments:

$$\frac{dA_i}{dt} = k_2 (k_d \alpha C_i - P_i) + SR \frac{Surf}{m_i V_i} P_i - \lambda A_i \quad (9)$$

where $Surf$ is the area of the box, SR is the sedimentation rate, and m is the sediment concentration (total sediment mass divided by total water volume in the box).

A.2. POSEIDON-R MODEL

The POSEIDON-R compartment model is described in detail in various published literature (see Refs. [3, 26]). It simulates the marine environment as a system of 3-D compartments (boxes) for the water column, bottom sediment and biota. The water column compartment is

vertically subdivided into layers (in the present study a single water layer in all boxes is considered). Radionuclides in the water column are assumed to be partitioned into dissolved and particulate fractions, which fall on the seabed, and this partitioning is described by a distribution coefficient. The radionuclide concentration in each water compartment is regulated by a set of differential equations which describe the exchange of radionuclides between adjacent compartments, the temporal variation in the respective concentration, and also between suspended radionuclides and those in the bottom sediment, as well as radioactivity sources and decay. Exchanges between the water column boxes are described by radionuclide fluxes due to turbulent diffusion, sediment settling and advection. The bottom sediment compartment is divided into three layers, and the transfer of radioactivity between the upper sediment layer and the water column resulting from resuspension, diffusion and bioturbation, and between the upper and middle sediment layers, resulting from diffusion only, are explained. Moreover, downward burial processes operate in all three sediment layers. More detailed information on the POSEIDON-R compartment model equations and parameters is provided in Ref. [20]. The POSEIDON-R model can deal with different types of radiological releases including river runoff of land deposited radionuclides, atmospheric fallout, point sources associated with both unplanned releases and routine discharges from nuclear facilities located inland (directly into rivers) and on the coast (directly into the marine environment) [26].

The POSEIDON-R compartment model [46] implements a dynamic biota uptake model which includes both benthic and pelagic food chains. In the biological uptake model, marine organisms are grouped into classes according to taxon and trophic level. Radionuclides are also classified according to the fish tissue type in which they are specifically accumulated. These simplifications allow for a limited number of standard input parameters. The food chains differ between the pelagic and the benthic zones where pelagic organisms comprise primary producers (phytoplankton) and consumers (zooplankton, non-piscivorous (forage) fish, and piscivorous fish). In the benthic food chain, transfer of radionuclides is from contaminated bottom sediments to deposit-feeding invertebrates, demersal fish, and in turn, benthic predators. Bottom sediments include both inorganic and organic components and radioactivity is assumed to be assimilated by benthic organisms from the organic component of the bottom deposits. Other food web components are molluscs (filter feeders), crustaceans (detritus feeders) and coastal predators, which feed throughout the water column in shallow coastal waters. Furthermore, all organisms take in radionuclides directly from the water and also through the food web.

There are some differences in the accumulation of radionuclides into marine organisms. The simplest case is when radionuclides are accumulated uniformly in the organism. An example of such radionuclides are isotopes of caesium, which are mostly accumulated in muscle, this being the dominant tissue in the fish organism. However, most radionuclides are accumulated non-uniformly in the organism. The ‘target’ tissue for these radionuclides is one or several organs such as the liver, kidney, stomach, gonads. Activity concentrations in each tissue type need to be summed with their weight fractions in order to estimate the concentration of the radionuclide in the organism. Another specific radionuclide is tritium, which forms organically bound compounds. To illustrate these processes, biological uptake models are briefly described below.

A.2.1. Uniform accumulation

Phytoplankton takes up radionuclides only from water and it is assumed that phytoplankton is in equilibrium with water due to its fast metabolism (i.e. rapid uptake rate of activity from water and a short retention time thereof). This means that radionuclide concentration in phytoplankton

is proportional to the concentration in water at any time. The coefficient of proportionality is called the biological concentration factor and it is already known for most radionuclides [43]. For other organisms, the dynamic approach is used for calculation of the activity concentration according to the following differential equation:

$$\frac{dC}{dt} = aK_f C_f + bK_w C_w - \frac{\ln(2)}{T_{0.5}} C \quad (10)$$

where C , C_f and C_w are the activity concentrations in each type of marine organism, their food and the surrounding water, respectively, a is the food extraction coefficient (assimilation efficiency), b is the water extraction coefficient (in gills), K_f is the food uptake rate, K_w is the water uptake rate, and $T_{0.5}$ is the biological half-life of the radionuclide in the body, which correlates with the time of the renewal of living cells. The diet of the marine organism may include one or several types of feedstuffs. To account for this, the activity concentration in the food, C_f is expressed as the total activity from all types of prey. Details and values of all parameters for this approach are described in Ref. [20].

A.2.2. Non-uniform accumulation

For radionuclides which mostly accumulate in tissues other than muscle, activity in muscle cannot be ignored because even low activity concentrations in muscle provide an essential contribution to the total concentration in the fish body due to large mass fraction. Therefore, in general, concentrations of radionuclides in all tissues need to be considered in order to obtain the total concentration in the fish body.

For this purpose, Eq. 10 for each type of fish is split into three equations (see Eq. (11)), which describe the accumulation of radionuclides in the main fish tissues (i.e. carcass, muscle, organ). Assimilation efficiency parameters for the different tissues (a_b for carcass, a_f for muscle and a_o for organs) were defined based on experimental ratios between the concentration of the radionuclide in the whole body of marine fish and concentration in the specific tissue [47]. Median values of assimilation efficiencies amongst a wide range of values for each radionuclide (e.g. isotopes of Co, Mn, Zn, Fe, Ni, Ag) were used in the study. The median values used were: $a_b = 0.005$; $a_f = 0.05$; $a_o = 0.5$.

$$\begin{aligned} \frac{dC_{bone}}{dt} &= a_b K_f C_f + b K_w - \frac{\ln 2}{T_{0.5b}} C_{bone} \\ \frac{dC_{fles}}{dt} &= a_f K_f C_f + b K_w - \frac{\ln 2}{T_{0.5f}} C_{fles} \\ \frac{dC_{organ}}{dt} &= a_o K_f C_f + b K_w - \frac{\ln 2}{T_{0.5o}} C_{organ} \end{aligned} \quad (11)$$

A.2.3. Tritium accumulation

Tritium enters the food chain in exchangeable form as tritiated water (HTO) and in the form of organically bound tritium (OBT). The HTO tritium in the body of the aquatic organism is considered as being in equilibrium with the HTO in the aquatic environment and is expressed by the following simple relationship [48]:

$$C_{HTO} = C_w (1 - drw) \times 0.001 \quad (12)$$

where C_{HTO} is the HTO concentration in an aquatic organism (Bq kg^{-1} fresh weight), C_w is the HTO concentration in water, 0.001 is the transformation coefficient m^3l^{-1} , and drw is the dry weight fraction of an aquatic organism.

OBT concentration in phytoplankton C_{OBT}^{phy} is assumed to be in equilibrium with water [49] as:

$$C_{OBT}^{phy} = 0.0004 \times drw^{phy} C_w \quad (13)$$

For other marine organisms in the food chain (only the pelagic food chain is considered for 3H), the same equation used for uniform accumulation is applied. The set of parameters which were used were taken from the literature review summarized in Ref. [48].

The system of boxes consists of three nested compartments (see Fig. 12) where the small coastal box (in red) describes the vicinity of the NPP (Jordan Cove) and the large coastal box (in green) corresponds to a larger area (Niantic Bay). The volume and average depth of these two boxes is taken from NOAA Chart 13211⁷. It is known that tidal currents are dominant in the area being considered. The exchange rates between boxes were estimated based on average currents for the Jordan Cove [50] and Niantic Bay [51], respectively. The average depth and exchange rate with the outer area for the regional box (in blue) were calculated from the Ocean Assimilation Model, with hydrodynamics of the NEMO system [52]. Default values for the suspended sediment activity concentrations and sedimentation rates for coastal areas [53] were used in the model. The parameters used are summarized in Table 4.



FIG. 12. Configuration of boxes in the area of interest in POSEIDON-R model.

⁷ Available for download at: <https://www.charts.noaa.gov/PDFs/13211.pdf>

TABLE 4. MAIN PARAMETERS IN POSEIDON-R BOX MODEL

Parameter	Small coastal box (Fig 12 red)	Large coastal box (Fig. 12 green)	Regional box (Fig. 12 blue)
Mean depth (m)	10	20	80
Volume (km ³)	1.0×10^{-2}	0.16	12
Exchange with outer box (m ³ /s)	363	1204	7312
spm (mg/l)	10	10	10
SR (g/cm ² y)	0.075	0.075	0.075

A.3. EULERIAN MODEL, ESTE AI

The programme ESTE AI (Annual Impacts) has implemented two dispersion models for marine environment, i.e. a Lagrangian particle tracking model and a Eulerian dispersion model. In the version ESTE AI used for the generic NPP Scenario, the Eulerian dispersion model was applied.

The model represents a Eulerian approach—solution to diffusion process. It is a 2-D depth averaged dispersion model, and the dispersion equation describes the time evolution of the radionuclides in the computational domain. The computational domain includes an impacted marine area of 200 km in size where the discharge point is situated in the centre of the computational domain. The particular domain is also defined by the shoreline. The dispersion equation is solved using the finite difference numerical method where the computational domain is discretized on small cells (squares of 2 km in size). Each square is defined by its depth in order to include real bathymetry data. The grid configuration is illustrated in Fig. 13.

The dispersion equation takes into account advection (transport of radionuclides due to the water currents), the diffusion process, radioactive decay, and interaction with sediments (water-suspended sediments, water—bottom sediment, suspended sediment—bottom sediment). The dispersion calculation is performed for each radionuclide separately and the outputs are the concentration in the water C (in the dissolved phase), the concentration in the suspended sediment C_{susp} and the concentration in the bottom sediment C_b .

The governing equation for activity concentration in water C includes advection, diffusion, desorption from suspended sediment, sediment settling, adsorption from bottom sediment, radioactive decay, and the source term (release into the sea):

$$\frac{\partial(hC)}{\partial t} + \frac{\partial(v_i h C)}{\partial x_i} = \frac{\partial}{\partial x_i} \left(h k \frac{\partial C}{\partial x_i} \right) - k_2 h C_{susp} - SR C_{susp} + f_2 C_b - \lambda h C + S \quad (14)$$

Where v_i is the depth averaged current field ($i=1, 2$), h is the depth, k is the diffusion coefficient, λ is the radioactive decay constant, SR is the sedimentation rate on the marine bottom, S is the source term, k_2 is the desorption kinetic coefficient (describing the desorption from the suspended sediment), and f_2 is a factor describing adsorption from the bottom sediment.

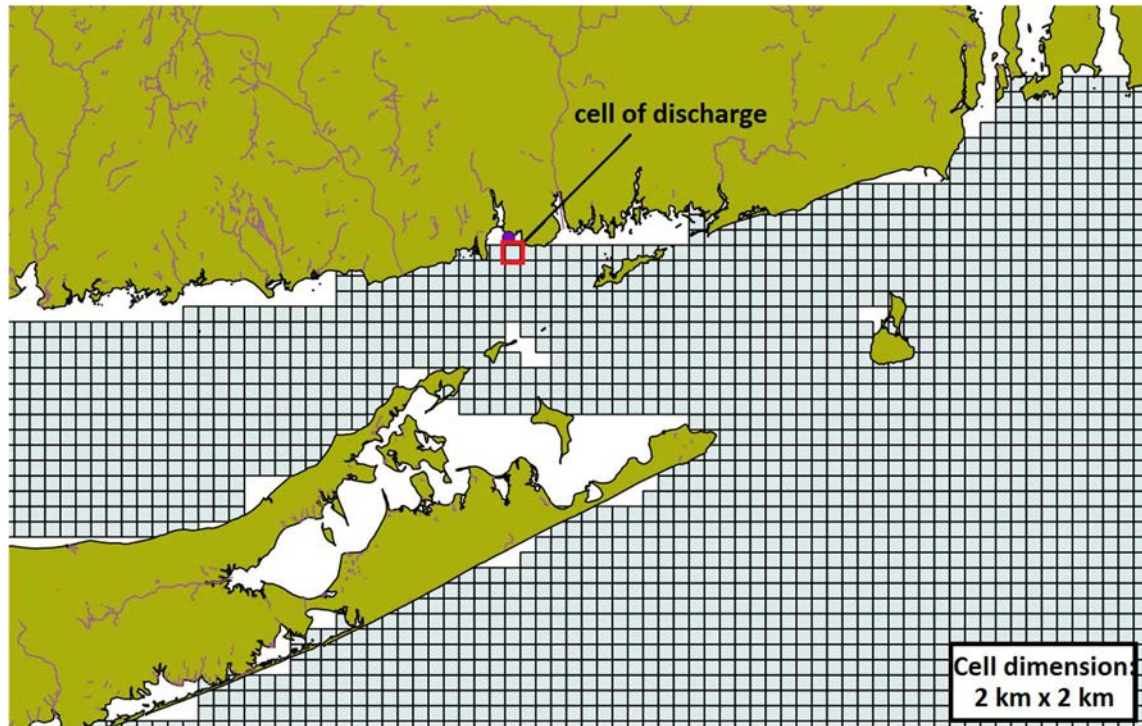


FIG. 13. Applied discretization of Long Island Sound area (area of interest in the generic NPP scenario) in the Eulerian model ESTE AI.

The governing equation for the bottom sediment C_b consists of desorption from bottom sediment, sediment settling, adsorption from water, radioactive decay, and the burial process of activity into deeper sediment layer.

As an input parameter for calculation, the model needs marine currents of the assumed marine area, covering the whole computational domain and the whole computational period. The currents are specified as monthly mean currents, and the points in which the current fields are defined are the same as the centres of the domain cells. The HYCOM database was used to generate the applied current fields.

Consequently, the programme calculates activity concentration in marine animals, including fish, shellfish (molluscs, crustaceans) and algae. The evaluation is based on the calculated activity concentration in water (dissolved phase) and the concentration factors given in Ref. [43]. Based on the activity in water, the programme also evaluates impacts on biota (represented by impacts on flatfish, crab and seaweed as reference animals and plants for the marine environment). Moreover, in biota calculation, the approach and factors defined in ICRP 108 [54, 55] are applied.

Exposure pathways for the representative person applied in ESTE AI for discharges into the marine environment are as follows:

- Marine fish consumption (internal exposure): consumption of fish caught locally;
- Swimming in the sea (external exposure during the time spent in the water);
- Fishing on the sea (external exposure during the time spent on the ship);

- Stay at shoreline (external exposure during shore activities);
- Drinking water (applied only in the case of the presence of desalination plants), it represents the pathway of direct ingestion of ‘seawater–desalination–drinking water–human’;
- Feeding water for animals (applied only in the case of the presence of desalination plants), the representative person consumes animal products fed by water from desalination plants.

A.4. RELEASE DATA

Total releases for the year 2016 are used as the source term for the simulations. These release data are given in Table 5 and were provided by MODARIA II Working Group 3.

TABLE 5. RADIONUCLIDE TOTAL RELEASES FOR YEAR 2016

Radionuclide	Total release S (Bq/year)
Cs-137	8.90×10^7
Cs-134	8.99×10^6
H-3	6.01×10^{12}
I-131	1.02×10^7
Xe-133	2.53×10^6
Mn-54	3.53×10^7
Fe-55	9.01×10^8
Co-58	1.21×10^7
Co-60	9.36×10^7
Ni-63	4.44×10^7
Zr-95	5.77×10^5
Nb-95	5.81×10^5
Nb-97	9.25×10^5
Ag-110m	4.18×10^6
Sb-125	1.64×10^9

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LIST OF ABBREVIATIONS

ADCP	Acoustic Doppler current profiler
FIO	First Institute of Oceanography
FDNPP	Fukushima Daiichi Nuclear Power Plant
GODAE	U.S. Global Ocean Data Assimilation Experiment
HTO	Tritiated water
HYCOM	HYbrid Coordinate Ocean Model
JAMSTEC	Japan Agency for Marine-Earth Science and Technology
k_d	distribution coefficient (marine)
LIS	Long Island Sound
MODARIA I	IAEA programme on Modelling and Data for Radiological Impact Assessments, 2012–2015
MODARIA II	continuation of MODARIA, 2016–2019
NEMO	Nucleus for European Modelling of the Ocean
NOPP	National Ocean Partnership Program
NPP	nuclear power plant
OBT	organically bound tritium
OGCM	ocean general circulation model
OFES	OGCM for the Earth Simulator
WG7	Working Group 7 of the MODARIA II programme

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