Marine radioecology after the Fukushima Dai-ichi nuclear accident: Are we better positioned to understand the impact of radionuclides in marine ecosystems?

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HIGHLIGHTS

- Marine radioecology studies at the FDNPP coast: process-based modelling and field investigations
- Dynamic modelling of transfer between seawater, sediments and the biological compartments
- New data on submarine groundwater discharges and ocean circulation of radionuclides
- We formulate a strategy for marine radioecology based on processes-based research.
- We highlight the need for more ecology knowledge in marine radioecology.

GRAPHICAL ABSTRACT
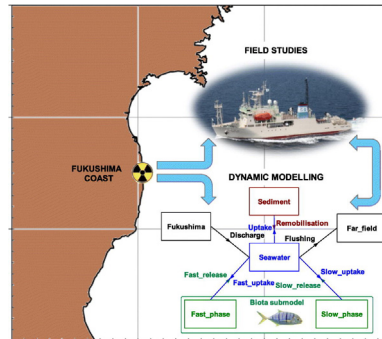
ABSTRACT

This paper focuses on how a community of researchers under the COMET (CO-ordination and iMplementation of a pan European projecT for radioecology) project has improved the capacity of marine radioecology to understand at the process level the behaviour of radionuclides in the marine environment, uptake by organisms and the resulting doses after the Fukushima Dai-ichi nuclear accident occurred in 2011. We present new radioecological understanding of the processes involved, such as the interaction of waterborne radionuclides with suspended particles and sediments or the biological uptake and turnover of radionuclides, which have been better quantified and mathematically described.

We demonstrate that biokinetic models can better represent radionuclide transfer to biota in non-equilibrium situations, bringing more realism to predictions, especially when combining physical, chemical and biological

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interactions that occur in such an open and dynamic environment as the ocean. As a result, we are readier now than we were before the FDNPP accident in terms of having models that can be applied to dynamic situations. The paper concludes with our vision for marine radioecology as a fundamental research discipline and we present a strategy for our discipline at the European and international levels. The lessons learned are presented along with their possible applicability to assess/reduce the environmental consequences of future accidents to the marine environment and guidance for future research, as well as to assure the sustainability of marine radioecology. This guidance necessarily reflects on why and where further research funding is needed, signalling the way for future investigations.

1. Introduction

Marine radioecology is a research discipline investigating and quantifying the biogeochemical and ecological processes that control the fate and transfer of radionuclides in the marine environment. A context for improving knowledge and consideration of these processes is, most unfortunately, provided by the Fukushima Dai-ichi nuclear power plant (FDNPP) accident in 2011, which led to the most important accidental release of artificial radionuclides to the oceans ever. As a result, the FDNPP accident was rapidly identified as the subject for marine radioecology investigations.

Many questions about the behaviour and fate of FDNPP-derived radionuclides appeared unsolved at the time of the accident. There was an ongoing need to obtain reliable data to assess and follow the radiological state of the local environment. Measurements in marine biota were not available in the acute accident phase (i.e. the first 30-days), when activity concentrations in the Fukushima coast were highest. Therefore, dynamic modelling tools for radionuclide dispersion and biokinetic transfer were urgently required to complete the timeline of contamination in the various marine compartments, including marine biota species, both locally and in the wider marine environment.

In 2012, a strategic research agenda (SRA) for radioecology was published (Hinton et al., 2012), as the outcome of work within a series of European Commission (EC)-supported initiatives. This SRA identified three challenges: (1) predict human and wildlife exposure more robustly by quantifying the key processes that most influence radionuclide transfers; (2) determine ecological consequences under realistic exposure conditions and (3) improve human and environmental protection by integrating radioecology (Hinton et al., 2012).

The main lines of research from Challenge One of the SRA are directly relevant to the study of the Fukushima accident. One such line involves identifying and mathematically representing key processes that make significant contributions to the environmental transfers of radionuclides and resultant exposures of humans and wildlife. Another research line calls for acquiring the data necessary for parameterisation of the key processes controlling the transfer of radionuclides. A third line focusses on developing transfer and exposure models that incorporate physical, chemical and biological interactions, enabling predictions of radionuclide concentrations in biota to be made, both spatially and temporally. These research lines provided the reference point for the present project.

This paper focuses on the development within the EC-funded project COMET (Vandenhove et al., 2016) of modelling tools adapted to the highly dynamic FDNPP post-accident situation, improving the existing models by making them more dynamic and process-based, and trying to make more realistic estimates of marine dispersion and transfer to biota with these models in order to assess radiological consequences, using improved data for parameterisation, calibration and validation. We also report new research on marine processes undertaken to investigate in detail the fate of the FDNPP-derived radionuclides in the ocean, providing an integrated radioecological picture of what releases subsist and the environmental concentrations arising thereof. We then use this experience to propose a strategy for the future of marine radioecology.

1.1. State of knowledge on the impact of the FDNPP accident on the marine environment

The FDNPP accident that occurred on 11 March 2011 was a highly significant event for marine radioecology, since about 80% of the radioactive fallout occurred over the Pacific Ocean (IAEA, 2015; Sanial et al., 2017; Stohl et al., 2011; UNSCEAR, 2014), especially in coastal waters near the FDNPP. Direct discharges of radionuclides to the ocean continued during the first months after the accident, as a consequence of emergency cooling efforts at the FDNPP (Buesseler et al., 2011), whereas atmospheric releases ceased in a few days. This chain of events constituted the largest 'single' accidental release of manmade radioactivity to the marine environment from civilian nuclear plants (Beresford and Vives Batlle, 2012). A full description of the accident is given elsewhere (IAEA, 2011; IAEA, 2015; IRSN, 2012; PMJHC, 2011; Povinec et al., 2013; UNSCEAR, 2014), so only a brief summary is presented here.

Most efforts on the investigation of the contamination in the ocean have centred on the radiocaesium isotopes ^{134}Cs (T$_{1/2}$ = 2.06 y) and ^{137}Cs (T$_{1/2}$ = 30.2 y), of which between 5 and 11.7 PBq would have been deposited onto the regional ocean (Estournel et al., 2012; Kawamura et al., 2011; Kobayashi et al., 2013). The direct liquid discharges to sea were estimated to be about 2.8–5.9 PBq (Miyazawa et al., 2013; Tsumune et al., 2012; Tsumune et al., 2013), although other authors produced larger figures, ranging from 13.5 to 27 PBq (Bailly du Bois et al., 2012; Charette et al., 2013; Rypina et al., 2013). The combined inputs of fallout and direct discharges would amount to about 15–18 PBq (Aoyama et al., 2015; Inomata et al., 2016; Tsubono et al., 2016), which represent an increase by 22–27% of the existing ^{137}Cs in the North Pacific due to global fallout (Aoyama et al., 2015; Buesseler et al., 2017).

The concentrations of radiocaesium in the coastal ocean of Japan decreased with time after the accident, but remained consistently above background values over the following years, with some episodic, significant increases. Concentrations of ^{137}Cs in surface seawater from the vicinity of the FDNPP increased from 1 to 2 Bq m^{-3} before the accident to ~10^7 Bq m^{-3} near the plant in early April 2011 (Buesseler et al., 2011), then decreasing to ca. 1000 Bq m^{-3} during 2013–2015. By late 2011, inputs of radiocaesium were already ~4–5 orders of magnitude lower than those estimated at the time of the accident (Bailly du Bois et al., 2012; Charette et al., 2013; Estournel et al., 2012; Kanda, 2013; Rypina et al., 2013; Tsumune et al., 2012).

After radionuclides entered the water by wet/dry atmospheric deposition and direct discharges, numerous physical and biogeochemical processes occurred. These included (a) transport and dilution of radionuclides by oceanographic advection and dispersion, (b) exchange processes between seawater and suspended particulate matter (sorption and ion exchange), (c) accumulation of radionuclides in sediments (sorption, particle scavenging and deposition), (d) biological uptake and turnover of radionuclides by marine biota (seawater uptake, food/sediment ingestion, depuration and transfer across the foodweb) and (e) remobilisation of sediment-borne radionuclides by desorption, sediment resuspension or biological activity. A conceptual representation of these key processes is given in Fig. 1. In this study, we focussed
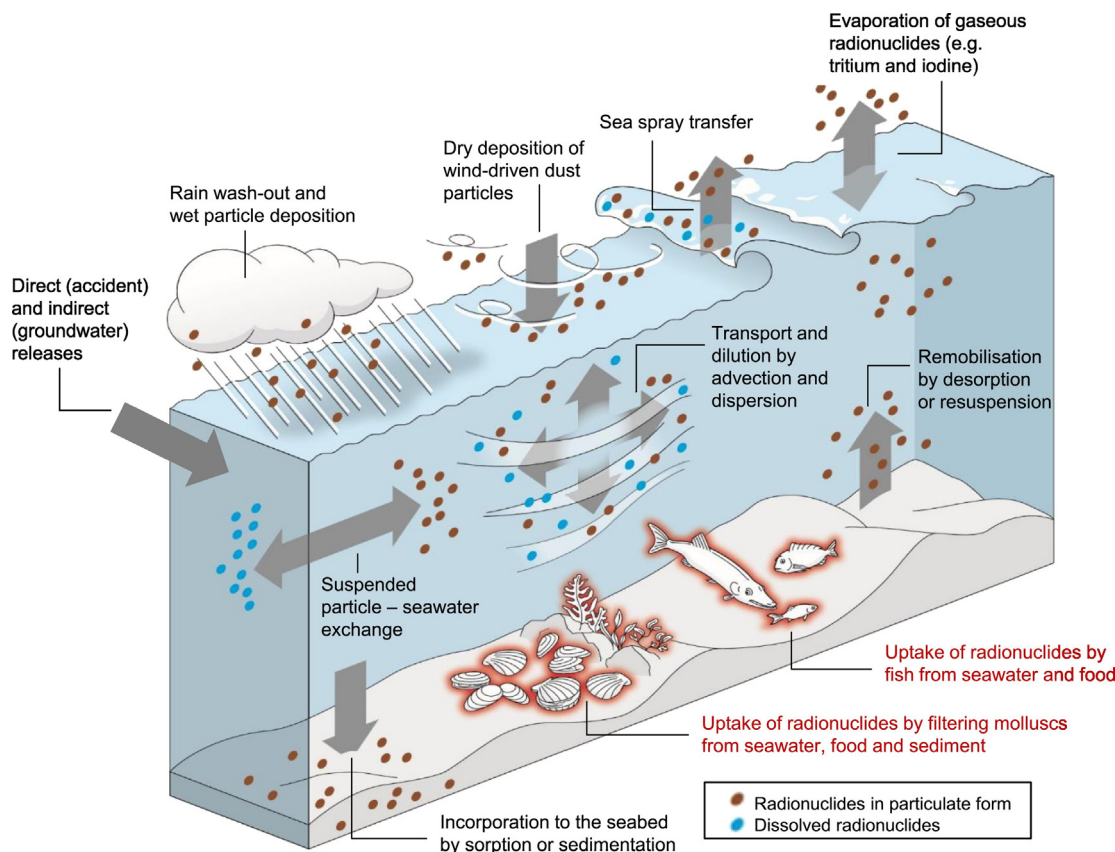
Fig. 1. Fate of radionuclides in the marine environment including the principal processes of dry and wet deposition, advection and dispersion, particulate transfer, sedimentation, seabed remobilisation and biological uptake.
(adapted from Hervé Bouilly, IRSN)

principally on processes such as oceanographic dispersion, infiltration of groundwater to sea, uptake and turnover by sediments and bioaccumulation, capitalising on knowledge of the local ecosystem.

Radiocaesium was transported by ocean currents, particularly the southward flowing Oyashio Current and the northward and eastward flowing Kuroshio Current in the area off the FDNPP. The transport eastwards was driven by the North Pacific Current. Aoyama et al. (2016) estimated an average propagation velocity of about 7 km day^{-1} until March 2012, and 3 km day^{-1} from March 2012 through August 2014, consistent with drifter-based estimates (Rypina et al., 2014). FDNPP-derived Cs isotopes were detected on the Canadian continental shelf by June 2013 and in the coastal waters of North America in February 2015, with 5.6 Bq m^{-3} and 1.4 Bq m^{-3} for ^{137}Cs and ^{134}Cs, respectively (http://OurRadioactiveOcean.org/results; http://fukushimainform.ca/). Based on ocean circulation models, it is expected that the ^{137}Cs activities in coastal waters of North America will decrease to pre-accident levels of 1–2 Bq m^{-3} by 2020 (Behrens et al., 2012; Rossi et al., 2014).

In addition to radiocaesium, other long-lived radionuclides were released to the marine environment. Due to its low volatility, the releases of ^{90}Sr to the atmosphere were several orders of magnitude lower than those of ^{137}Cs (Povinec et al., 2012; Steinhauser et al., 2014; Tanaka et al., 2014). Most of the ^{90}Sr released from the FDNPP was directly discharged to the North Pacific Ocean, with estimates of total inventories ranging from 0.04–1.0 PBq (Casacuberta et al., 2013; Povinec et al., 2012). Approximately 1 kg of the long-lived ^{129}I was also released from the FDNPP, mainly through direct discharges into the ocean (Guilderson et al., 2014), slightly increasing the concentrations of ^{129}I in seawater near the FDNPP due to global fallout. The potential release of plutonium has also been reviewed by Zheng et al. (2013), who estimated that at most about 1.0 to 2.4 × 10^9 Bq ^{239,240}Pu would have been released as a consequence of the accident, which is a relatively low amount due to its low volatility and the limited explosive releases from the FDNPP.

Only a small fraction of radiocaesium is scavenged by particles that eventually accumulate on the sea floor, whether they are biogenic or lithogenic and resuspended from the shelf. Various studies of samples collected using sediment traps detected FDNPP-derived Cs after the accident (Buesseler et al., 2015; Honda and Kawakami, 2014; Honda et al., 2013; Otosaka and Kato, 2014), allowing the calculation of Cs removal rates of <2% y^{-1} from the water column (Honda et al., 2013).

The radioactive contamination in seabed sediments from sites off the east coast of Japan is dominated by ^{134}Cs and ^{137}Cs (Ambe et al., 2014; Black and Buesseler, 2014; Kusakabe et al., 2013; Nagaoka et al., 2015; Otosaka and Kato, 2014; Otosaka and Kobayashi, 2013; Sohtome et al., 2014; Thornton et al., 2013). The inventories of ^{137}Cs in the sea floor from 35.5 to 39° N off Japan out to 4000-m depth range from <50 Bq m^{-2} (comparable to the inventories due to global fallout) to some 10^5 Bq m^{-2} in sediments closest to the FDNPP, totalling <1% of the estimated total of 15–30 PBq of radiocaesium released during the FDNPP disaster (Black and Buesseler, 2014; Kusakabe et al., 2013), most of which ended in the ocean and originated from atmospheric deposition. Albeit with a large spatial variability, ^{137}Cs inventories tend to decrease with increasing distance from the FDNPP and increasing water depth, and are higher in shelf sediments to the south of the FDNPP relative to the north (Ambe et al., 2014; Black and Buesseler, 2014; Kim et al., 2016; Ono et al., 2015; Otosaka and Kobayashi, 2013).

Since radionuclides from the FDNPP have entered the marine environment, they have also been taken-up by the local biota. Measurements carried out by the site operator and the Japanese authorities give the contamination evolution in the various environmental compartments (including biota) over the years since the accident. Several databases have become available online, such as the Japanese Nuclear

Regulatory Authority (NEA - http://radioactivity.nsr.go.jp/en/list/205/list-1.html), the Japanese Atomic Energy Agency (JAEA - http://emdb.jaea.go.jp/emdb/en/) and the Tokyo Electric Power Company (TEPCO - http://www.tepco.co.jp/en/nu/fukushima-np/f1/smp/index-e.html). These databases can be complemented with published data (Buesseler and Aoyama, 2012; Buesseler et al., 2012; Honda et al., 2012). When combining these sources, a quasi-continuous record of concentrations emerges. Despite significant data scatter, it shows that benthic fish, which live and feed in close proximity to bottom sediments, have higher Cs activity concentrations than pelagic fish (Wada et al., 2013). It is also clear that biota landed in the Fukushima Prefecture have levels higher than elsewhere - see Fig. 5 of Buesseler et al. (2017) and Sohtome et al. (2014).

The comprehensive UNSCEAR study (2014) included an evaluation of the biological uptake of FDNPP radionuclides and the resulting doses to marine biota have been published (Strand et al., 2014; Vives i Batlle et al., 2014). Based on an extensive record of measured data from biological samples collected in the early post-acute period (April 2011–August 2012) activity concentrations of ^{137}Cs in marine biota ranged from a few Bq kg^{-1} up to above 10^3 Bq kg^{-1}. The highest dose rates to marine biota calculated directly from these measured concentrations were approximately in the range 0.17–0.25 µGy h^{-1} (ascidians, macroalgae, sea urchins and holothurians) and 0.10 to 0.17 µGy h^{-1} (benthic fish, crustaceans and molluscs). An absolute maximum total dose rate of 4.4 µGy h^{-1} was calculated for fat greenling (*Hexagrammos otakii*) in August 2012.

As the earliest information became available (MEXT, 2011; MEXT, 2013; TEPCO, 2011), it was possible to conduct an early dynamic modelling based assessment of doses to non-human biota in the FDNPP environment in the first 3 months after the accident. A dynamic transfer model (Vives Batlle et al., 2008) was used to reconstruct biota concentrations 1 month after the accident, when seawater concentrations were peaking but biota monitoring had not been established (Vives i Batlle and Vandenhove, 2014). This allowed the refinement of early estimates that had identified the potential for ecological consequences (Garnier-Laplace et al., 2011) but, since equilibrium of the biota with maximum ambient concentrations had been assumed, this led to higher exposures than subsequently calculated (Buesseler et al., 2011; Kryshev et al., 2012; Vives i Batlle, 2011).

These early dynamic modelling efforts were refined in the UNSCEAR study, allowing maximum dose rates (^{131}I and ^{134,137}Cs combined) for fish of approximately 140 µGy h^{-1} to be retrospectively calculated for the closest vicinity of the FDNPP (North drainage channel). This implies an accumulated dose to fish over 1 year of about 0.32 Gy. Maximum calculated exposures for macroalgae (exceeding 20 mGy h^{-1}) were predicted to have occurred at day 23 after the accident, decreasing to below 10 mGy h^{-1} by the 32nd day due to the decreasing contribution of ^{131}I, and giving an accumulated dose of 6.8 Gy over the first 90 days. Further away (Iwasawa shore), maximum dose rates for all biota were <3 µGy h^{-1}, except that a maximum ^{131}I dose rate of 633 µGy h^{-1} for seaweed was recorded 30 days post-accident, decreasing 2 orders of magnitude after one month (Vives i Batlle et al., 2014).

The conclusion of UNSCEAR, based on both monitoring and modelling, was that exposures of marine non-human biota were generally too low for acute effects to be observed, and that effects (if any) would have been transient. This transience was due to the quick radioactive decay of the then dominant isotope (^{131}I, T$_{1/2}$ = 8 days). Any effects would have been confined to some species (seaweeds) and areas close to the release point, with exposures generally falling below thresholds for likely population effects in a few months after the accident, so that observable effects on populations were not to be expected (UNSCEAR, 2014). The more recent IAEA study provides an evaluation consistent with these findings (IAEA, 2015).

Other dynamic modelling methodologies have been used to study the transfer of FDNPP radionuclides to marine biota (Heling et al., 2002; Keum et al., 2015; Kryshev et al., 2012; Maderich et al., 2014;

Psaltaki et al., 2013). Additional data and model simulations have extended the analysis to subsequent years (Vives i Batlle, 2016; Vives i Batlle et al., 2016). Sediment transfer and source-term estimation are now factorised in dynamic transfer modelling (Vives i Batlle, 2015; Vives i Batlle and Vandenhove, 2014), as will be shown in Results below.

Observations after 2011 showed that radionuclide levels in fish off Fukushima are highly variable but remain elevated. By 2012, ^{137}Cs levels of several thousand Bq kg^{-1} in benthic fish were being reported, exceeding the Japanese radioactivity in food limit of 100 Bq kg^{-1} (Buesseler, 2012; Buesseler and Aoyama, 2012; Wada et al., 2013). However, the activity concentrations in marine products in Fukushima Prefecture (even within the 20-km radius area) decreased drastically during the five years after the FDNPP accident. Nevertheless, by 2015, ^{137}Cs concentrations of >10 kBq kg^{-1} were still detectable in some sedentary rockfishes (*Sebastes cheni*, *S. oblongus*, and *S. pachycephalus*) from the interior of the FDNPP port (Wada et al., 2016).

We consider that this did not indicate significant risks at the level of populations. For 2012–2014, only the most exposed fish nearest to the FDNPP somewhat exceeded the ERICA 'screening' benchmark of 10 µGy h^{-1} with a median dose rate of 45 µGy h^{-1} (Johansen et al., 2015), but this would be for the most exposed individuals and not for fish populations as a whole. Recent work shows a tendency towards even lower exposures. Regarding dose to human fish consumers, by 2013 Fukushima-derived doses were already 3–4 orders of magnitude below naturally occurring ^{210}Po-doses.

Dynamic transfer modelling continues to be applied. Prior to this study, this could simulate successfully the acute phase of the accident, but not the sustained concentrations in fish observed over a longer period of several years (Johansen et al., 2015). Our work has helped to reconcile this difference by refining the biokinetic model with a two-component biological half-life[1] (T$_{B1/2}$) so it can now reproduce the long-term ecological half-lives observed in the FDNPP environment (Iwata et al., 2013; Tagami and Uchida, 2013; Vives i Batlle, 2016), as shown below.

2. Methodology

The objectives of the present study, in line with the ALLIANCE SRA, were to (a) identify and mathematically represent key processes governing the environmental transfer of radionuclides resulting from the Fukushima accident and resultant exposures to biota in the marine environment, (b) acquire the data necessary for parameterisation of the key processes controlling the transfer of radionuclides and (c) develop transfer and exposure models that incorporate physical, chemical and biological interactions, enabling time-dependent predictions of radionuclide concentration to biota to be made. To achieve the objectives, we undertook research to improve and validate radioecological models for the improved understanding of impact to humans and the environment in dynamic (non-steady state) exposure situations. We also performed field investigations on the impact of direct and indirect releases from the FDNPP, which included research cruise work.

For the modelling effort, we aimed to go beyond the simple models included in decision support systems for emergency situations by selecting process-based models used to represent more accurately the Fukushima post accidental situation, whence we linked some of these models with the ERICA dosimetric approach (Brown et al., 2008), bridging the gap between research and assessment modelling. The main focus was to use time-dependent (dynamic) models as a basis for the work with three goals in mind:

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[1] The biological half-life of a radionuclide for a particular organism is the time required by the organism to eliminate half the activity taken-up through biological processes (that is, excluding radioactive decay).

a) Implementation and use of classical radioecological models based on dynamic transfer (biokinetic) equations to evaluate concentrations in marine organisms (algae, fish, molluscs, crustaceans), combined with improvement of radioecological parameters (concentration factors and single or multicomponent biological half-lives) for ^{137}Cs, ^{134}Cs and ^{90}Sr. This task included comparisons with observed data series from Fukushima.

b) Inclusion of the processes governing the interaction of radionuclides with seabed sediments in dynamic transfer modelling, in order to better understand the benthic transfer pathway. This work followed our ongoing studies adapting existing dynamic models and testing them with the available data to see if they were capable of reproducing radionuclide concentrations in sediment to give a more realistic calculation of concentrations in biota, improving the dose models for humans and biota based on K_d values.

c) Inclusion of ecological and environmental processes in process-oriented modelling for mid- and long-term predictions: modelling trophic transfer to pelagic fishes, including foodweb transfers and establishing whether there is real potential for biomagnification in the Fukushima marine food chain.

For the field-based work we focused on sampling seawater, sediments and biota using purposely-designed research cruises providing controlled sampling in terms of spatial and temporal location. An important objective of this cruise work was to quantify the fluxes of chemical elements associated with the offshore transport via submarine groundwater discharge. We participated in the 2014 KS-14-20 and 2015 KS-15-13 Shinsei Maru cruises organised by Japan off the Fukushima coast, where samples of water were collected and analysed for ^{134}Cs, ^{137}Cs, ^{90}Sr, ^{129}I, ^{236}U and Pu-isotopes. Biota samples were collected and analysed for ^{134}Cs and ^{137}Cs. This fieldwork aimed to address the following four questions:

a) What is the fate of the contamination from the FDNPP accident in the ocean and what fraction of the total releases is stored in marine sediments?

b) How much radioactivity is still leaking from the FDNPP site and which are the mechanisms that govern the releases to the ocean, such as the role of submarine groundwater discharge?

c) To what extent have the concentrations of contaminants in the ocean changed since 2011?

d) What are the current activity levels and doses to marine biota?

3. Results

3.1. Biological transfer modelling

The main results of the model development effort have been published elsewhere (Belharet et al., 2016; Duffa et al., 2015a; Vives i Batlle, 2016) and are included in reports (Iosjpe et al., 2016a) and conference presentations (Duffa et al., 2015b; Iosjpe et al., 2016b; Vives i Batlle, 2014), Thus they will only be summarised here.

For the goal of implementing and using models based on dynamic transfer (biokinetic) equations, two kinetic radioecological models were perfected. The first one was the model STERNE, which is now optimised to obtain fast results for the first analysis of any radiological situation. STERNE calculates on a tri-dimensional grid both radionuclide transport using advection and diffusion equations offline from hydrodynamic calculations, and radioecological transfers to biota with a simple model based on first order kinetic transfer equations (Fievet and Plet, 2003). The required radioecological parameters (concentration factors and $T_{B½}$s) were compiled for some relevant radionuclides and for generic marine species including macroalgae, fish, molluscs and crustaceans (Duffa et al., 2015a).

The second model was D-DAT (Dynamic Dose Assessment and Transfer), which includes the dynamics of radionuclide uptake and turnover in biota, as determined by a balance between the residence time of radionuclides in seawater and the organism's $T_{B½}$ of elimination. The model calculates activity concentrations of ^{131}I, ^{134}Cs, ^{137}Cs and ^{90}Sr in fish, crustaceans, molluscs and macroalgae starting from measured activity concentrations in seawater. Although the model does not explicitly include the incorporation of radionuclides via food intake, this process is captured indirectly by imposing the condition that the concentration ratio at equilibrium equals the concentration factor (CF), which incorporates all processes. D-DAT uses the radionuclide concentrations in biota to calculate absorbed dose by using dose rate per unit concentrations (DPUC, in μGy h^{-1} per Bq kg^{-1}) derived from template ERICA Tool simulations for marine biota, using default parameters from the latest version of the Tool and other sources (IAEA, 2014).

For the goal of modelling radionuclide interaction with sediments, D-DAT was expanded to include a three-layer sediment sub-model derived from the marine model POSEIDON (Lepicard et al., 2004; Lepicard et al., 1998) in which the processes of particle scavenging, molecular diffusion, pore water mixing and bioturbation are considered, with parameters from Simmonds et al. (2004). Some of these parameters were recalibrated for the coastal area closest to the FDNPP (Vives i Batlle, 2016). The new version of D-DAT also implements a dual $T_{B½}$ approach, as many organisms depurate radionuclides from their bodies via a fast, short-term process followed by a longer-term process (Vives i Batlle et al., 2005; Wilson et al., 2005). This is essential if the model is to reproduce the biological transfer over both short and long timescales. D-DAT was further adapted to back-calculate the amount of radionuclide discharged in the initial accident, based on deducing the radionuclide flux into the seawater compartment by a mass balance method and integrating it over time.

An additional Multi-Analysis RadiologIcal aSSessment cAlculator (MARISSA) tool was developed to calculate dose rates to marine biota sampled during the field study, obviating the need for repeated runs using the ERICA Tool. This tool can categorise some 200 + different species of marine biota within nine ERICA reference organism categories (benthic & pelagic fish, Benthic mollusc, crustacean, macroalgae, phytoplankton & zooplankton, polychaete worm and vascular plant), plus six newly defined reference organisms (small crustacean, octopus, squid, sea urchin, ascidian and holothurian). Internal and external dose rates for ^{129}I, ^{131}I, ^{134}Cs, ^{137}Cs and ^{236}U can be calculated using dose per unit concentration values extracted from template runs with the ERICA Tool. An averaging algorithm pairs biota concentration at a particular location with water and sediment concentrations from nearby times and locations to match external and internal dose rates.

For the goal of modelling trophic transfer and possible biomagnification, we improved modelling tools for simulation of transfer to biota were improved, such as a previous dynamic approach (Iosjpe et al., 2016a; Psaltaki et al., 2013) based on the Thomann (1981) model. This uses a set of first-order differential equations to represent transfer of radionuclides within a foodchain containing phytoplankton, zooplankton, small fish (preying on the former) and large fish (preying on zooplankton and small fish) (Iosjpe et al., 2016b). Zooplankton and fish can also take up radionuclides directly from seawater. The model assumes that excretion/elimination rates are independent of the uptake route, assimilation efficiency is independent of food type, and that predators do not assimilate the activity concentration in gut content of their prey and zooplankton are a homogeneous group. The model is also linked to exposures to humans consuming the biota.

A further model of radiocaesium transfer to marine biota was also developed to take into account large scale organism displacements in the FDNPP area (Belharet et al., 2016). This is a trophic level ecosystem model coupled with ocean circulation. The different radioecological parameters of the model were estimated by calibration using published data, and a sensitivity analysis of parameter uncertainties showing a

high sensitivity to the ^{137}Cs concentration in seawater, the rates of accumulation from water and the radionuclide assimilation efficiency.

3.2. Field investigations

The main results of the field investigations have been published elsewhere (Aoyama et al., 2016; Buesseler et al., 2017; Casacuberta et al., 2017; Castrillejo et al., 2016; Vives Batlle, 2016), as well as in various conference proceedings (Buesseler, 2015a; Buesseler, 2015b; Buesseler, 2015c; Masqué, 2015). The main aspects are summarised below.

The principal focus of the research was on ^{137}Cs, ^{134}Cs and ^{90}Sr, although longer-lived radionuclides such as ^{129}I, ^{236}U and Pu-isotopes were also investigated. The 2014 and 2015 Shinsei Maru cruises resulted in a suite of seawater samples, sediment cores and biota samples collected at specific sampling stations (see Fig. 2). These stations covered the most heavily affected area off the coast of Fukushima and overlapped with previous expeditions, allowing a time-series of the changing contamination levels. In addition to the artificial radionuclides, natural radium isotopes were analysed to quantify the fluxes of chemical elements associated with the offshore transport. Groundwater sampling along the shoreline was conducted to determine the rate of submarine groundwater discharge (SGD) to constrain the radiocaesium flux to the ocean.

Surface ocean concentrations of ^{137}Cs, ^{90}Sr and ^{129}I measured during the field investigation were found to be higher near the FDNPP and they were above the global fallout levels, suggesting that there are still ongoing releases from the Nuclear Power Plant site. Particularly interesting was the evolution of atomic and/or activity ratios of ^{137}Cs/^{90}Sr, ^{129}I/^{137}Cs, ^{129}I/^{90}Sr with time, which helped to corroborate the ongoing releases of radionuclides to the coast off Japan (Casacuberta et al., 2017; Castrillejo et al., 2016). Overall, the results strongly suggested that a continuous surveillance of inputs of artificial radionuclides to the Pacific Ocean is still required.

A key finding of the cruise work was that groundwater transported FDNPP radionuclides have entered the ocean, both locally and further along the coasts. Relatively constant ^{137}Cs fluxes of up to 10^{11} Bq d^{-1} were estimated between May 2013 and October 2014 from the FDNPP site, suggesting ongoing steady state inputs, in addition to episodic inputs of Cs and other radionuclides. By 2016, the ongoing releases were still in the order of several GBq per day (Aoyama, M., pers. Comm.). At distances of 30–100 km from FDNPP, elevated activities of ^{137}Cs $> 10^4$ Bq m^{-3} were measured in groundwater below several beaches, with the highest levels associated with brackish groundwater underneath beaches along 180 km of coastline from about 36°50′N to 38°20′N, having salinities from 5 to 25‰ (Sanial et al., 2016; Sanial et al., 2017). These submarine groundwater beach samples were most likely enriched due to ongoing desorption of radiocaesium from subsurface sands enriched from contaminated seawater that took-up this radionuclide shortly after the accident, given exchanges of ocean and groundwater through tidal pumping and other processes in the subterranean estuary. Ongoing releases of radiocaesium to the ocean via the beach sands may be of similar magnitude as current inputs from the nuclear power plant and the dissolved component that desorbs from river born sediments entering the ocean primarily with heavy rains and storm events. It is highly likely that the main source of these ongoing releases is highly contaminated water within the FDNPP site. Discharges of only a few m^3 per day, whose ^{137}Cs activity concentration is as high as 10^{10} Bq m^{-3}, would be sufficient to account for the estimated ongoing releases.

In addition to radiocaesium, the field study also investigated the occurrence of other long-lived radionuclides, namely ^{90}Sr, ^{239,240}Pu, ^{236}U and ^{129}I. It was estimated that the discharge rates of ^{129}I from 2011 to 2015 varied from 30 to 500 kBq d^{-1}, some 5–10% of the ^{129}I released during March–April 2011 (Casacuberta et al., 2017). We also investigated the occurrence of Pu isotopes and ^{236}U in ocean waters in 2013 and 2014, but could not detect any clear signal attributable to emissions from FDNPP the (Casacuberta et al., 2017). It was also estimated that FDNPP leaked ^{90}Sr at a rate of 2.3–8.5 GBq d^{-1} into the North Pacific Ocean in September 2013 (Castrillejo et al., 2016). Our results were used to calculate that some 0.1 kg of ^{129}I have been released during the years following the accident up until 2015 (Casacuberta et al., 2017).

For non-human biota, the main scientific results (Vives i Batlle, 2014; Vives i Batlle, 2016) can be summarised as follows. A generally
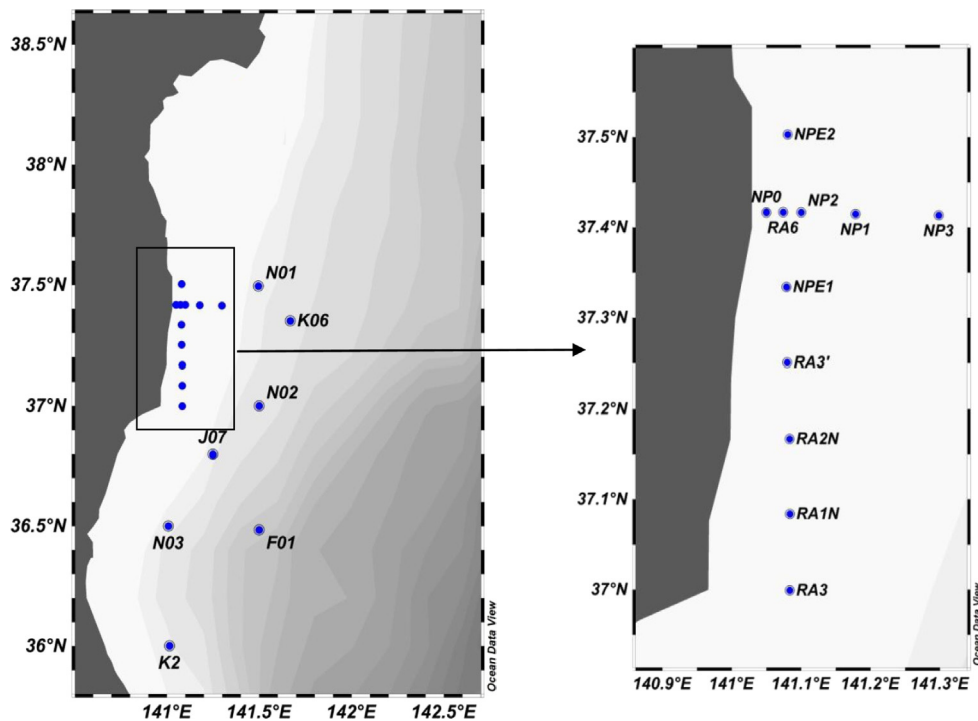

Fig. 2. Stations sampled for radionuclide analysis during the Shinsei Maru cruises.

exponential decrease of activity concentrations with time is observed for all the main categories of marine biota, as would be expected, since contamination clears the local environment by dispersion and dilution. The fact that activity concentrations for marine biota decrease much more slowly over time than would be expected given their $T_{B\frac{1}{2}}$s can also be explained through the modelling work with D-DAT (Vives i Batlle, 2016). We suggest that this is related to hold-up processes by bottom sediments, meaning that the ecological half-life reflects not only the biological turnover rates but also the desorption rate of radionuclides from sediments. In effect, sedimentary deposits offshore from the FDNPP have become increasingly a source for radionuclides in the local environment, due to their ability to retain radiocaesium.

From our own analysis of the published data (Vives i Batlle, 2016) and new field measurements, different rates of decrease exist for different organism groups, being slowest (consequently with a higher residence half-time) for benthic biota and fastest for pelagic biota. This is compatible with differences in $T_{B\frac{1}{2}}$s for these radionuclides and in agreement with other studies (Sohtome et al., 2014; Wada et al., 2016; Wada et al., 2013) as well as the aforesaid hold-up processes by sediments. There was a high variability in the ^{137}Cs activity concentrations, which could be due to sampling, measurement or geographical variability but may be partially explained also (particularly in the latter years) by the presence of secondary emissions (such as groundwater releases).

The complex time evolution of ^{137}Cs in marine biota from 2011 to present follows a multicomponent exponential decrease influenced by a bi-phasic depuration mechanism and the radionuclide hold-up processes by seabed sediments. Our latest results show that this is better reproduced by D-DAT when using dual $T_{B\frac{1}{2}}$ release with a short component of 4 days, a long component of 54 days and a phase transition time of 80 d. The default CR of 0.1 m^3 kg^{-1} was optimised to 0.061 m^3 kg^{-1} using pelagic fish (mackerel) concentrations from the RV Shinsei Maru KS-14-20 Research cruise (17–26 October 2014) and dividing them by a mean water concentration of 17.1 ± 0.9 Bq m^{-3}. In this way, the D-DAT prediction for benthic fish at the time of the cruise (1320 days after the accident) is 30 Bq kg^{-1}, close to the mean of 43 Bq kg^{-1} measured during the cruise.

We performed a sensitivity analysis of the D-DAT sediment submodel which identified the sediment reworking rate (R_W) as the most sensitive parameter for the turnover of radionuclides by the upper sediment layer. For a Station "T-1" contiguous to the FDNPP port in shallow waters (37.43°N, 141.03°W, 5 m depth) where sediment data have been continuously monitored (http://radioactivity.nsr.go.jp/en/), we calibrated R_W at 10^{-2} m d^{-1} leading to a good match with the observed sediment concentrations between 2011 and 2016 (Vives i Batlle, 2017). This high value is likely due to the shallow depth of the station and the proximity to the port area, leading to higher sediment reworking.

The influx of radioactivity to sea necessary to sustain the activity concentrations observed in the FDNPP vicinity was predicted by D-DAT. This requires knowing the half-time of water from the coastal region into the open ocean, which was deduced from the ^{137}Cs monitoring data in the first 50 days post-accident. The data was fitted to a double exponential curve and a flushing time of 2.2 ± 0.3 days was obtained by a standard method (Choi and Lee, 2004; Periáñez, 2012). With this information, the model gives integrated releases of 103, 30 and 3 PBq for ^{131}I, ^{137}Cs and ^{90}Sr, in line with the reported UNSCEAR (2014) ranges.

Data from the KS-14-20 2014 cruise show activity concentrations in fish ranging from 0.8 to 500 Bq kg^{-1}. These concentrations rank as ctenophore > benthic fish (rockfish > greenling and flounder) > pelagic fish (mackerel) > plankton. Except for benthic fish, dose rates to marine biota are predominantly dominated by internal exposure, then external exposure to sediment and then external exposure to water. However, the total dose rates to benthic fish are dominated by external exposure to sediment, showing that sediments are now an important source of

radiation exposure to the benthic biota. From these data, it seems that, as of 2016, total ^{137}Cs dose rates to pelagic fish are generally down to about 2 × 10^{-3} μGy h^{-1} and the highest total doses for benthic fish (rockfish) are of the order of 0.15 μGy h^{-1}. These doses are below the ERICA no-effects dose screening level of 10 μGy h^{-1} (Andersson et al., 2009; Beresford et al., 2007; Brown et al., 2008), confirming that the fish populations near the FDNPP are not at risk.

The above results help to explain mathematically why biota and sediment activity levels remain somewhat elevated, as well as the long ecological half-lives for biota observed in the field. The dual component biokinetic model setup improves predictions of biota concentrations over the single component model. The recalibration of the sediment model likewise improves the simulation of long-term activity trends in sediments. However, the scattering in biota sampling locations does not permit to corroborate the short-term variations predicted by the model.

4. Discussion on the impact of our results and the future of radioecology

4.1. How have the challenges of marine radioecology been addressed?

The FDNPP accident highlights the importance of potential accidental inputs to the marine environment, their radioecological consequences and the need to "predict human and wildlife exposure more robustly by quantifying key processes that influence radionuclide transfers, and incorporate the knowledge into new dynamic models" (Hinton et al., 2012), as identified by Challenge 1 in the SRA. We have progressed in this direction through marine biota transfer modelling work and collection of new field data from the vicinity of the FDNPP. Important marine processes, such as the interaction of seaborne radionuclides with suspended particles and sediments, the role of submarine discharges or the biokinetic processes of uptake and turnover, have been better quantified and mathematically described. Models have incorporated physical, chemical and biological interactions. In some cases, large spatial scales incorporating ocean circulation and land-sea interactions have been used. Whether or not this work has substantially decreased uncertainties is debatable, though it certainly has contributed significantly to quantify them.

In some respects, Challenge 2 of the SRA on the determination of ecological consequences remains the "weakest link" for marine radioecology. There is a need to develop and improve models (Wilson et al., 2010) and experimental approaches to determine the impact of radionuclides at the level of marine populations and ecosystems under the realistic conditions that these organisms are exposed.

Regarding the SRA Challenge 3 "to improve human and environmental protection by integrating radioecology" (Hinton et al., 2012), we have improved knowledge and tools for risk assessment, but only the first steps in developing radiation protection and decision support systems. The model STERNE has been developed to be included in a decision support system and the model D-DAT is set-up for dose assessment and has been tested in the context of the UNSCEAR Fukushima accident assessment (Vives i Batlle et al., 2014).

Ultimately, through our work, we have demonstrated that biokinetic models can better represent radionuclide transfer to biota in non-equilibrium situations, bringing more realism to predictions, especially when combining processes occurring in such an open and dynamic environment as the ocean. For example, such models can quantify how much and for how long activity released is retained (by biota, sediments or local waters) or is removed to the far field. Only by combining biogeochemical processes in a model is it possible to find out how quickly the radionuclides are cycled and cleared within the marine environment. This research is now ready to be integrated into the more general debate about what information is needed in case of accidental marine contamination, developing an assessment approach linked to the

wider scientific aspects (including interactions between radiological and non-radiological pollution).

4.2. What scientific questions have been answered and what questions remain?

The main achievement during this project has been to directly address temporal dynamics in the assessment process. There has also been an attempt to involve more biological factors in the modelling process, by including (for example) $T_{B\frac{1}{2}}$s, bioturbation, fish migration, trophic transfer and uptake/elimination. A consensus has gradually formed around the modelling approach for non-equilibrium situations. However, trophic transfer, particularly in the benthic foodweb, remains a challenge. Much work remains to be done regarding biokinetic transfer of radionuclides in marine biota, such as adopting a more refined parameterisation of transfer models using locally derived $T_{B\frac{1}{2}}$ data and the derivation of generic parameters for use in other environments.

In particular, the complex time evolution of ^{137}Cs from 2011 to present in marine biota from Japan requires further examination. Radioactivity levels in fish and other species have remained higher than initially anticipated, and are decreasing more slowly than concentrations in seawater, more akin to the gradual decline of radionuclides in sediments. Local radioactive hotspots for biota very close to the FDNPP could potentially persist. The necessary research to understand what this means has not been completed and should not stop with the conclusion of the present project, even if radioactivity levels in marine biota are below current knowledge-based thresholds for measurable effects on populations.

The questions that remain are: (a) what is the eventual fate of the radioactive contamination leaving the FDNPP zone including releases from delayed sources (b) what are the inputs by rivers whose catchment basins are still contaminated, (c) what are the long-term effects on marine biota species and ecosystems, taking into account the spatial heterogeneity of the contamination and the presence of some hotspots and (d) what is the likely time before the radionuclides released to the marine environment reach a steady state.

From our field research, it is clear that specific studies on the sediments should be addressed, particularly regarding the processes of remobilisation to the water column. Noteworthy here are the field investigations on ^{129}I for circulation studies, or using ^{137}Cs and the transuranic elements Pu and Am for sedimentation and dating studies, to mention the most obvious examples.

Ultimately, the contamination of the marine environment near the FDNPP is significant, even if risk estimates (in terms of dose) are low. We are convinced that the FDNPP accident remains a challenge for Marine Radioecology, requiring an ongoing study of the marine environment for years to come, in the same way that the Irish Sea remains a marine radioecology challenge to this day.

4.3. International dimension of the research performed

A particular emphasis in the present project was the collaboration with Japanese scientists, especially during the field research near the FDNPP aboard R/V Shinsei Maru and subsequent analytical determinations, as well as observations of radiocaesium in seawater from coastal sites around the FDNPP and the provision of data from the Japanese authorities for use in modelling.

At the broader international level, marine radioecology connects to the expertise of IAEA in the scope of marine studies and associated technical programmes aiming at developing marine models for radiation safety such as with the ongoing programme MODARIA II's two working groups dedicated to model exposure and effects to biota and radionuclide transport in the marine environment, respectively (http://www-ns.iaea.org/projects/modaria/modaria2.asp?s=8&l=129). Work presented here, particularly the dynamic transfer modelling knowledge and expertise developed, was actually utilised in a wider international context provided by the MODARIA I model intercomparisons (Vives i Batlle et al., 2016).

Marine radioecology has at its core an international dimension and this will influence science and risk assessment in the future. In this context, it is important to look at the results of activities of radioecologists outside the present network of collaborators (Baumann et al., 2015; Fukuda et al., 2017; Tateda et al., 2015; Tateda et al., 2017; Tateda et al., 2013; Tateda et al., 2016; Wang et al., 2016) in order to arrive at a true synthesis of marine radioecology research.

There have been radioactive releases to European marine waters before, both accidental (Chernobyl, Palomares, Thule) and operational (Sellafield, La Hague, Marcoule, etc.), but it is not clear how much of that knowledge/competence was applied to the FDNPP accident. There is an impression that some expertise from those situations (admittedly very different from the FDNPP accident) has not really been passed on to the current generation of radioecologists and marine radiochemists. Indeed, there seems to be less research activity in marine radiochemistry and radioecology than in the 1980′s. A number of research institutes and monitoring networks have been shut down, some networks for radionuclide sampling and radio-analytical laboratories are no longer available and fewer programmes are left in place for researching marine radioactivity and radioecology and to ensure the training of the next generation.

However, the increasing international level of attention on protection of the environment has permeated to marine radioecology, as exemplified by the first internationally-coordinated effort to assess the consequences of the FDNPP accident to the marine environment (UNSCEAR, 2014). This effort showed that, even if the immediate stakes in term of accidental exposure were probably not the environmental but the human ones, the situation characterisation makes scientific knowledge of the marine processes involved a necessity.

4.4. Are we well prepared in case of an accidental release in European marine waters?

After the present study, we are now better prepared than before the FDNPP accident in terms of having models that can be applied to dynamic situations. We have certainly contributed to identifying and expressing mathematically (implementing into radioecological models) what processes are most relevant to explain the marine distribution of radionuclides and their uptake and turnover by organisms.

What matters now is how effectively such models can be applied to another environment. European oceanographers, meteorologists and biologists work constantly to have models for these seas, but the transferability of existing models has yet to be demonstrated. Many of the oceanographic processes are the same for some European waters, at least for open oceans like the North Sea/Atlantic and the Arctic Ocean, but there are differences for seas with more restricted water movement such as the Baltic or the Mediterranean Seas or for a semi-enclosed shelf sea like the Irish Sea. We refer here not only to circulation patterns but also, for example, other differences such as salinity, suspended or sinking particulates and colloidal matter concentration or sediment biogeochemistry which can have an effect on the speciation and mobility of radionuclides (Mitchell et al., 2001).

One lesson learned from the FDNPP accident is how important is it to have properly georeferenced data, including data of different environmental compartments (e.g. biota/water/sediment) from the same location and time, along with key characteristics of the waters being sampled (temperature, salinity, sample depth, seafloor depth, etc.). This was often lacking and made for difficulties in trying to implement models that calculate transfer between different compartments (e.g. seawater and biota), requiring for input data and for validation concurrent reference points in these compartments.

In terms of actually dealing with an accidental release such as that from the FDNPP, it is necessary to prepare guidance and operational tools for different marine environments. In the first instance, it is

necessary to optimise tools for dispersion modelling (Min et al., 2013) and perform intercomparisons between different models. For the FDNPP accident, this has successfully been done in the context of the IAEA MODARIA I and II inter-comparisons of radionuclide transfer models for seawater and sediments (Periáñez et al., 2016; Periáñez et al., 2015; Periañez et al., 2015).

The next step is to integrate these models in decision support systems, as has been done in the JRODOS platform (Heling et al., 1997; Lepicard et al., 2004; Raskob et al., 2011). It is necessary to think about a good indicator for environmental impact that should be used for recommendations. For example, in assessing the radiation risks to non-human biota, it is necessary to have benchmarks for acute exposures as well as for low-level routine exposures, a point considered in the UNSCEAR study (Vives i Batlle et al., 2014).

Lastly, an accidental release from a nuclear installation (or from other industrial installations linked to river or marine waters) is not only a scientific issue, but also a concern for society. Radioecological research could contribute to an inter-disciplinary approach to develop tools addressing scenarios of general societal concern.

4.5. Advice on research prioritisation

Prioritisation of marine radioecology research should be strongly grounded on basic science criteria, selecting projects with potential to lead to a knowledge breakthrough. Societal relevance is a complementary indicator to help to identify research topics, but like any scientific discipline, radioecology needs to be independent. By this we mean that research prioritisation should never be based on public opinion or political decisions, as this has proven in the past to harm the scientific process (Liu et al., 2009).

Basic science in the context of this advice means developing models and measurements that raise the level of the science by bringing forth new mechanistic understandings at the level of processes (e.g. foodweb modelling, eventual fate in marine environment, etc.). The scope of marine radioecology should be broadened to include the whole marine environment and its interacting parts, inducing a conceptual change in the way we interpret the marine environment by linking to the big themes of other sciences. We must also strive to bring a new understanding of the effects of radionuclides on humans and the environment both by the radionuclides themselves and by their interaction with other environmental stressors.

The potential to improve risk assessment by developing practical tools for decision-makers is a natural consequence of working through the above priorities, rather than being an *a priori* research objective. For example, modelling the migration of FDNPP radionuclides to faraway continents is not crucial from a risk assessment perspective due to the low levels involved, but it will improve our understanding on global circulation using marine tracers and thus lead to better predictive models.

Scientists can benefit from listening to societal concerns on issues such as releases from nuclear installations, becoming involved in two-way communication with the public. This should not be limited to when there is an accident, because an uninformed public may raise unnecessary concerns and this fosters the distrust of agencies providing information. The public stands to benefit greatly from the dissemination marine radioecology in a balanced and impartial way.

4.6. Developing a strategy for marine radioecology

Marine radioecology connects to larger scientific issues such as the use of radionuclides as tracers for marine processes involving the dispersion of contaminants that in the broader sense can be both radioactive and non-radioactive. Consequently, there is a need to integrate the approaches of marine radioecology not only with those of oceanography and radiochemistry but also ecology, ecotoxicology and climate science, thereby bringing more ecological thinking into marine

radioecology. This is a logical consequence of the way radionuclides are investigated as tracers for marine biogeochemical processes. Yet, despite decades of research, the role of biological and ecological processes (e.g. speciation, radionuclide incorporation in biomass, the biological processes promoting desorption of radionuclides from sediments) in radionuclide dispersion is not sufficiently understood. This is true in particular for radioelements other than Cs and Sr. The mechanisms by which uptake of one element/radionuclide enhances or decreases uptake of another are also not fully understood from a basic science perspective. There is a need for radioecologists to have common projects with the wider research community in order to gain a better level of understanding on these issues.

A future strategy for marine radioecology should therefore include gaining increased process understanding of biogeochemical interactions and their influence on radionuclide dispersion in marine environments. This should include physico-chemical processes at the land/marine interfaces (estuaries, submarine groundwater discharges), considering the mid- and long-term behaviour of radionuclides in sediments and the dynamic modelling of interactions with particulate matter, both suspended particles and colloidal matter, particularly important for understanding the behavior of transuranic radionuclides in the water column (Vives i Batlle et al., 2008).

Although the FDNPP accident will continue to be a significant topic for marine radioecology in the future, this does not mean neglecting other problems. At the European level, there is a need to consider three research topics: the radioactive releases from sunken nuclear submarines to Northern European seas, the NORM radionuclides released from offshore oil platforms and the continuing releases of European nuclear power plants and reprocessing industries under either their active or decommissioning phases (see section on Future directions in marine radioecology below). Moreover, there could be potential accidental discharges from nuclear reactors or modern operative nuclear submarines in European waters, acting as an additional incentive to improve dispersion and biological transfer models. Assessing such events would require better kinetic bioaccumulation parameters for a wide set of radionuclides (Iosjpe et al., 2016b; Iosjpe et al., 2011), validated hydrodynamic models with good resolution available over all European seas and knowledge of the possible source terms.

Another element of a strategy for improving marine radioecology is that the effects of radiation on marine organisms and hence foodchains need to be studied. Although initial work in this direction has been performed (Vives i Batlle, 2012; Vives i Batlle et al., 2009; Wilson et al., 2010), practical models and vital data for use in making realistic predictions are limited.

4.7. Need for further radioecology funding

Marine radioecology is not only of purely scientific but also of social relevance. It seems legitimate to propose increased resources (not only funding but also infrastructure, education and training) for marine radioecology, particularly at the European level where funding is sporadic yet there is an urgent need to concentrate efforts and maintain research capacity.

Funding for marine radioecology peaked after the period of maximum actinide discharges and subsequent technetium discharges from Sellafield, decreasing gradually in the last decades. The focus changed to the terrestrial/freshwater environments after Chernobyl. By that time, few laboratories were prepared to work in the ocean, and the state of modelling was less advanced than it should have been. Although there has been some EC funding within the COMET project, the present series of EC CONCERT calls have so far overlooked marine radioecology. It is now imperative to consolidate the pool of marine radioecologists and fund new projects. We cannot afford a weak position to face the challenges of the future, including those posed by future accidental situations.

The first step would be to pursue the strategy proposed herein, whilst envisaging possible scenarios with which to interest individual countries having installations that pose risks towards marine ecosystems and their neighbouring countries. Since this is an international issue, it is legitimate to seek international funding – for example, in Europe, support at the EURATOM level. Individual research institutions have a very limited capacity to sponsor research in marine radioecology, so it is wholly legitimate to instate European funding, thereby ensuring that a cross-border harmonised approach is adopted.

Recently, a first proposal for a 5-year roadmap of the radioecology SRA was developed within the COMET project in collaboration with the European Radioecology Alliance, which was established in 2009 to strengthen coordination and integration of research in radioecology (http://www.er-alliance.org/media-centre/publications/radioecology-roadmap/). This roadmap is a basis of an implementation plan for the priority research activities that have been identified in the SRA, with emphasis on integrating research methods and approaches in marine radioecology to harmonise readiness for accidental marine releases at the European level. Thus, the roadmap acts as possible vehicle for implementation of a marine radioecology strategy, though our view is this strategy should not be limited to accidents because marine radioecology concerns itself with issues much broader than emergency preparedness.

5. Future directions in marine radioecology

The future directions of marine radioecology are open-ended. Some of these directions may depend on unforeseen nuclear events, but in this paper we are making a much broader case for marine radioecology as a fundamental discipline, working alongside radiochemistry and the ocean sciences to provide quantitative understanding about the processes that happen when radionuclides enter the marine environment.

Despite an abundance of field research, marine radioecology is singularly lacking in underlying theories and principles that are experimentally testable. Some of the work has an inherent weakness in that it tries to combine data from various sources to derive results, often providing qualitative explanations without a clear underlying theoretical basis. The SRA has already acknowledged the need to rectify this situation, studying the behaviour of radioactive contaminants in the seas and their consequences for man and environment from a 'first-principles', classical science basis.

There is also a need to reinforce the experimental base by carrying out laboratory and field studies, such as on radionuclide transfer in marine biota, in order to better parameterise, calibrate and validate dispersion and dynamic transfer models. The usability of models as assessment tools is a consequence of research steps addressing (and answering) fundamental science questions.

One example of much needed research is the inclusion of speciation in the models to represent the dynamic repartitioning between particulates, colloidal and dissolved phases. Such process studies were initiated in the late 1980's, but they are rarely used in assessment models (Vives i Batlle et al., 2008). Much scope is left for such studies in complex and highly dynamic brackish environments such as fjords and estuaries, especially near inputs.

Besides Fukushima, marine radioecology should reopen research on other seas. An important topic is discharges of produced water from oil and gas installations. Discharges of produced water from the petroleum industry are a multi-contaminant situation involving metals, radionuclides and organic compounds (Hosseini et al., 2012). As part of this, it is important to address the bioaccumulation and combined toxicity of the different components in produced water, both in the water column and in sediments, for various marine species. This calls for expertise in marine dispersion and bioaccumulation modelling that goes beyond radioecology and intersects with the field of ecotoxicology, requiring a multidisciplinary approach. In Europe, the Norwegian shelf discharging NORM radionuclides in North-East Atlantic waters is a logical place to begin this research.

Beyond Europe, Marine Radioecology needs also to focus on studying other environments such as (sub-) tropical environments, because tropical countries and small islands have ecosystems that are particularly sensitive to anthropogenic changes as they are among the most active in terms of urbanisation and industrialisation. Tracking the behaviour of heavy metals through their radiotracers offers a way to study threats to such environmentally fragile ecosystems (Aarkrog, 1994; IAEA, 2004). New nuclear power plants will be operated in other countries (Arabic countries, India, China and Southeast Asia) and some of these countries are also asking Europe for advice in marine dispersion and knowledge on the fate of radionuclides in marine waters.

More research is also needed on the role of sediments as sinks and sources for radionuclides. The long-term evolution of the radionuclide inventory in the oceans including the fine sedimentary deposits is also of interest. Much work remains to be done by providing models that include biogeochemical processes affecting the transfer and remobilisation of radionuclides in the seafloor. Such processes include radionuclide scavenging from the water column (sinking biogenic particles, interactions at the benthic nepheloid layers), mixing deeper into the sediment column (bioturbation) or the role of sediment as a new source term (diffusion, non-redox driven microbial processes, sediment resuspension during storms), with links to how this would affect the uptake by benthic and pelagic biota.

An important aspect for modelling is dynamic food webs and investigating the possibility of bioaccumulation of radionuclides as they ascend into the foodchain in various specific environments. Linked to the above is the need to calculate doses to populations of marine biota in response to chronic radiation-induced changes. Population models have been used (mainly under IAEA MODARIA I and II) to study how radiation effects could alter the natural balance of the ecosystem in the presence of ecological factors such as resource limitation, migration, predation and species competion (Kryshev et al., 2008; Vives i Batlle et al., 2009; Wilson et al., 2010; Woodhead, 2003) and the way is open to future investigations. We call also for studies of interspecies differences in uptake/turnover of radionuclides by biota, as well as variability within species with age, size and sex and the transgenerational effects of historical doses of radiation. This will lead to a more refined version of the biological uptake and population dose effects models for marine biota in order to better justify the radiation dose benchmarks used.

In order to parameterise models, it is important to address gaps in the biokinetic data used by transfer models (such as multicomponent $T_{B\frac{1}{2}}$s). One possible approach is to use extrapolation approaches such as allometric analysis, which allows to effect an interpolative scaling of the biokinetic parameters linked to metabolism by means of a power function of the mass (Vives i Batlle et al., 2007). A biokinetic database for marine biota developed as part of the IAEA MODARIA I exposures and effects in biota group (Beresford et al., 2015a; Beresford et al., 2015b) is being analysed in search of such allometric mathematical relationships. Time series of measurements from existing monitoring programmes offer the opportunity to determine new parameter values (e.g. CF and $T_{B\frac{1}{2}}$s) for different radionuclides.

It is vital that we continue to support marine radioecology for these findings to come to fruition. Only in this way we can enhance the status of our discipline as a fully-fledged fundamental scientific research discipline, dedicated to the betterment of the environment and the generations of humans and wildlife that will inhabit it (and live with any consequences) into the far future.

6. Conclusions

By the complexity of their physical and chemical properties, radionuclides have entered every component of the FDNPP marine ecosystem: soluble phase, suspended particulates, seabed sediments and the life forms inhabiting therein. The circulation and fate of these radionuclides is inextricably linked to dynamic processes occurring in a

complex environment. There is a need to understand water circulation at the local and global level, and marine biogeochemical processes generally, by means of new field data on the behaviour of radiotracers. There is also a need to use dynamic modelling to reach an adequate understanding of the consecquences of marine radioactivity for life and the environment, underpinned by such data. Our primary contribution in this study is the combination of both approaches to understand the radioecological impact of the FDNPP accident.

The present study has attended to specific aspects of the Fukushima situation that had not been well investigated. This included, for instance, the implementation of the modelling of uptake of radionuclides by biota and the assessment of the radiological impact, as well as the evaluation of the relevance of groundwater discharges providing a continuous input of radionuclides. Moreover, we assessed the presence of radionuclides not previously investigated, such as ^{236}U and Pu isotopes. The main achievements of the project are therefore the quantification of marine processes such as the interaction of waterborne radionuclides with suspended particles and sediments, the biokinetic uptake and turnover of radionuclides and radiation doses arising thereof. It is now demonstrated that biokinetic models can better represent radionuclide transfer to biota in non-equilibrium situations. As a result, we are readier now than we were before Fukushima in terms of having models that can be applied to dynamic situations.

The importance of adopting a dynamic representation of radionuclide transfer between seawater, sediment and the biological compartments has benefited from our effort to involve more biological factors in the modelling process, by including (for example) dual $T_{B\frac{1}{2}}$s to represent multicomponent depuration, bioturbation, fish migration, trophic transfer and uptake/elimination. The inclusion of sediment processes in the dynamic models has resulted in improved mid- and long-term modelling predictions, especially for the benthic ecosystem. The transfer models developed and tested in the present project contain a better process quantification and increased understanding of the uptake of radionuclides to wildlife, with the consequent increased capacity to address temporal dynamics in radiological assessment.

The research cruises undertaken in the FDNPP area and resulting data interpretation have allowed us to increase understanding on the fate and transport of ^{90}Sr, ^{129}I, radiocaesium, plutonium and ^{236}U from the FDNPP in the ocean, the inputs from submarine groundwater discharges and the uptake and turnover of radionuclides by marine biota. Ocean circulation and land-sea interactions have been considered in interpreting these results, showing that the radionuclides are far from being permanently immobilised, experiencing instead a dynamic exchange between sediments, the water column and the biomass in an ongoing biogeochemical cycle. Releases are still ongoing but activity concentrations in fish are below the allowed human consumption levels except for the most affected species (e.g. rockfish) from high exposure areas (e.g. the FDNPP harbour).

The present research has contributed to strengthen capacity, competence and skills in marine radioecology. *We make a loud call for support to continue marine research* in line with the priorities identified in the SRA and the advancement of the proposed strategy for the future of marine radioecology, signalling the direction for future investigations.

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