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Opening Fukushima floodgates: Modelling ^{137}Cs impact in marine biota

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ABSTRACT

A numerical model was applied to simulate the transport of ^{137}Cs released with the waters which were used to cool Fukushima reactors. These stored waters will be released to the Pacific Ocean according to Japanese government plans. The radionuclide transport model is Lagrangian and includes radionuclide interactions with sediments and an integrated dynamic foodweb model for biota uptake. Calculations made from a conservative approach indicate that expected concentrations in sediments and marine fish would be orders of magnitude below those detected after Fukushima accident and also lower than those resulting from global fallout background.

1. Introduction

Water has been pumped by the operators of Tokyo Electric Power Company (TEPCO) into the damaged Fukushima Dai-ichi NPP (FDNPP) to cool the nuclear reactors during the past 10 years. Consequently, huge volumes of radioactive water have been stored. Underground water and rain water seepage into the reactors have contributed to the accumulation of radioactive water (Buesseler, 2020) as well. According to TEPCO operators, about 1000 tanks used to store the contaminated water will run out of space by 2022, when they will contain nearly 1.3 million tons of radioactive water (TEPCO, 2020). The Japanese government has decided to dump this contaminated radioactive water into the ocean in 2022. This decision is being fiercely opposed by international communities, local fishermen, and residents. Although most radionuclides in the water have been treated by an Advanced Liquid Processing System (ALPS), the water still contains radionuclides whose half-lives are comparatively long, such as ^{3}H, ^{14}C, ^{125}Sb, ^{137}Cs, ^{90}Sr etc. (Buesseler, 2020). These radionuclides may potentially contaminate marine ecosystems and threaten human health (Buesseler, 2020; Shozugawa et al., 2020).

The consequences of the releases have been recently investigated using numerical modelling in the case of ^{3}H by Zhao et al. (2021). Concentrations of ^{3}H in Pacific Ocean waters arising from different release scenarios were calculated. It was found that that the majority of tritium would be mixed and diluted rapidly in the coastal waters off Japan, and then transported eastward along the Kuroshio Current extension. The radionuclide patch would arrive to the north-American Pacific coast in about 4.5 years. In addition, different discharge durations would lead to different dispersion patterns in the ocean. If the discharge duration is short (one month to one year) the polluted water would shift with the currents and finally arrive to the coastal area of the US and Canada involving larger polluted areas. For long release durations, the core of polluted water would be more constrained around the discharge area.

This paper describes a further step in assessing the effects of these planned releases. Thus, a numerical model is applied to simulate ^{137}Cs transport in the Pacific Ocean from one of the same release scenarios as in Zhao et al. (2021). Moreover, interactions of radionuclides between water and sediments are incorporated in the model. Also, a dynamic foodweb biota uptake model is integrated within the marine transport model to evaluate ^{137}Cs concentrations in biota resulting from the releases. Actually, ^{137}Cs was measured in fish after FDNPP accident [Wada et al., 2016; Men et al., 2017; Johansen et al., 2014] and these new planned releases are a matter of concern for local fisheries which are not recovering (Buesseler, 2020).

The methodology is described in Section 2. First, the numerical

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Table 1
Generic parameters used in the dynamic biological uptake model (from Maderich et al., 2014a) for ^{137}Cs. The concentration factor for phytoplankton is CR_{phyto}=20 l/kg.

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

model is briefly presented in Section 2.1; later (Section 2.2) the model setup is described. Results are presented and discussed in Section 3.

2. Methods

2.1. The model

The model is a Lagrangian transport model which includes three dimensional advection and diffusion by ocean currents, interactions of radionuclides with sediments and a dynamic foodweb biota uptake model integrated within the transport model. This model was used in the assessment of Fukushima accident, simulating ^{137}Cs transport during the first two years after March 2011 and participated in a model intercomparison exercise organized by the International Atomic Energy Agency (IAEA) in the frame of MODARIA-II program (IAEA, 2019; Periáñez et al., 2019a). A brief summary of the model is provided here.

Water circulation provided by FORA ocean model was used for transport calculations. This model, Four-dimensional Variational Ocean ReAnalysis for the Western North Pacific (FORA-WNP30), is the first-ever dataset covering the western North Pacific over the last three decades (1982–2014) at eddy-resolving resolution. It is a cooperative work of the Japan Agency for Marine-Earth Science and Technology (JAMSTEC) and the Meteorological Research Institute, Japan Meteorological Agency (JMA/MRI) using the Earth Simulator (Usui et al., 2017; Tsujino et al., 2010). The domain used in the present calculations extends 117° E-160° W and 15° N-65° N in longitude and latitude, respectively. Horizontal resolution is 0.1° and there are 54 vertical levels (0–6300 m) with increasing thickness from the surface to the sea bottom. This ocean model is the same which was used in the previous applications in the frame of the IAEA exercises (IAEA, 2019; Periáñez et al., 2019a). Monthly data from years 2011 and 2012 were used. This two year sequence, selected as typical one, was repeated five times to carry out simulations over 10 years.

Advection is obtained from the following equation for each released particle:

$$L_x = u\Delta t + \frac{\partial K_h}{\partial x}\Delta t \quad (1)$$

$$L_y = v\Delta t + \frac{\partial K_h}{\partial y}\Delta t \quad (2)$$

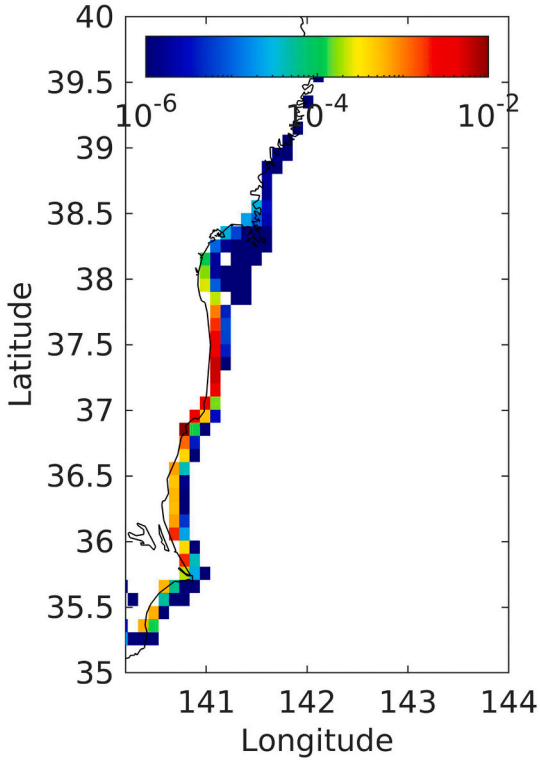

Fig. 2. Concentrations of ^{137}Cs in bed sediments (Bq/kg) one year after starting the one month release.

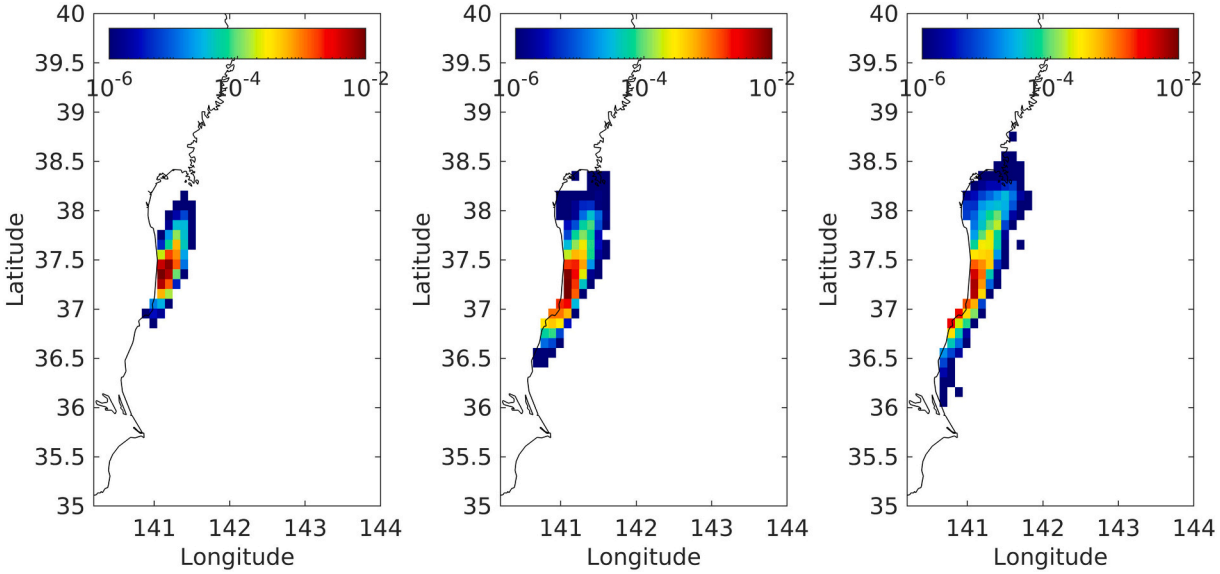

Fig. 1. Concentrations of ^{137}Cs in bed sediments (Bq/kg) 30, 60 and 120 days (left to right) after starting the one month release.

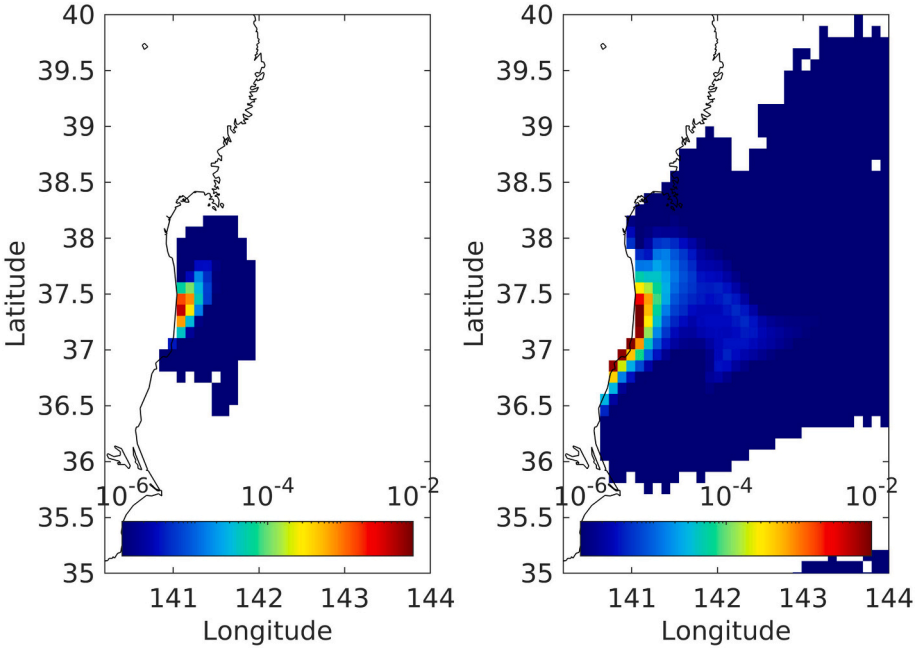

Fig. 3. Concentrations of ^{137}Cs in non-piscivorous fish (Bq/kg wet weight) 30 (left) and 120 (right) days after starting the one month release.

where L_x and L_y are the changes in particle position; u and v are water velocity components at the particle position and depth and for the corresponding calculation time step, since currents change in time. Derivatives of the horizontal diffusion coefficient (K_h) prevent the artificial accumulation of particles in regions were diffusion coefficients are smaller (Proehl et al., 2005).

The maximum size of the horizontal step given by a particle due to turbulent mixing, D_h, is (Periáñez and Elliott, 2002):

$$D_h = \sqrt{12K_h\Delta t} \tag{3}$$

in the direction θ=2π*RAN*, where *RAN* is a uniform random number between 0 and 1 and Δt is time step in the Lagrangian model. This equation gives the maximum step size. The real size at a given time and for a given particle is obtained by multiplying the equation by another independent random number. In this way it is ensured that a Fickian diffusion process is simulated. The time step used to integrate the Lagrangian model was fixed at Δt=1800 s.

The size of the vertical step given by the particle is written as (Periáñez and Elliott, 2002; Periáñez, 2020):

$$D_v = \sqrt{2K_v\Delta t} \tag{4}$$

given either upward or downward. In this equation K_v is the vertical diffusion coefficient. The Smagorinsky scheme (Cushman-Roisin and Beckers, 2011) was used to obtain K_h and a constant and uniform K_v was set (Periáñez et al., 2019a): K_v=1.0 × 10^{-5} m^2/s.

Radioactive decay is solved using a stochastic method (Periáñez and Elliott, 2002; Periáñez, 2020). The probability of radioactive decay is defined as:

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

where λ is the radioactive decay constant. A new random number is generated. If $RAN \leq p_d$ the particle is considered to decay and it is removed from the computation. Radioactive decay is considered for dissolved radionuclides and also for those fixed to the sediments.

The interactions of radionuclides between water and the seabed sediments are also solved using a stochastic method, and they are described in terms of a kinetic adsorption rate k_1 and a desorption rate k_2. The probability that a dissolved particle is adsorbed by the sediment is:

$$p_a = 1 - e^{-k_1\Delta t} \tag{6}$$

If a new generated independent random number is $RAN \leq p_a$, then the particle is fixed to the sediment. The probability that a particle which is attached to the sediment is released to water is:

$$p_r = 1 - e^{-k_2\phi\Delta t} \tag{7}$$

and the same procedure is used. ϕ is a correction factor that takes into account that part of the surface the sediment particle is hidden by the surrounding sediments. Thus, this hidden part is not interacting with the water. More details may be seen in Periáñez and Elliott (2002) and Periáñez (2020), for instance.

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

$$\frac{\partial C_{pred}}{\partial t} = aK_1C_f + bK_wC_w - K_{0.5}C_{pred}, \tag{8}$$

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

$$C_{phyto} = CR_{phyto}C_w, \tag{9}$$

where CR_{phyto} (m^{3}kg^{-1}, wet weight) is the concentration ratio for phytoplankton. Standard literature values for all these parameters for the four considered species may be seen in Table 1 in Maderich et al.

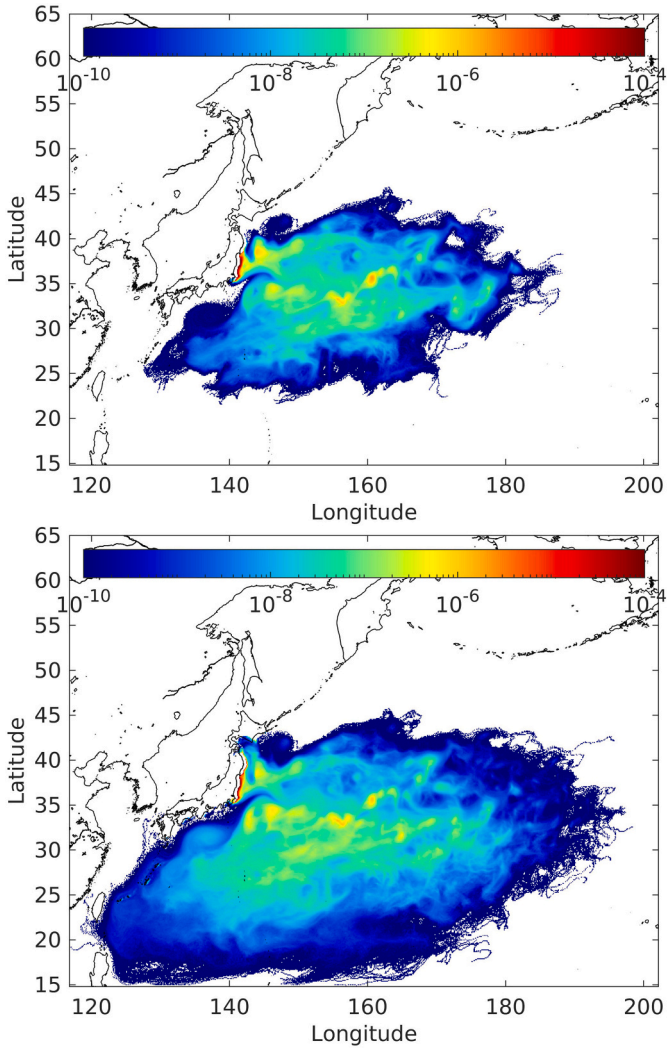

Fig. 4. Concentrations of ^{137}Cs in non-piscivorous fish (Bq/kg wet weight) one (top) and two (bottom) years after the one month release.

(2014a), which is reproduced in Table 1 in the present paper. The movement of fish in the ocean is not considered in the present model.

This foodweb model was integrated within the marine transport model to obtain the spatio/temporal variations of ^{137}Cs concentrations in biota. Only pelagic biota was considered, in a 200 m thick surface layer. The number of Lagrangian particles in the simulation was fixed as NP=5 × 10^6. Note that, although the transport model is Lagrangian, the biota model solves a set of differential equations (Eq. (8)). Thus it is Eulerian in concept.

The coupling between both models is carried out in the following way. First radionuclide concentrations in surface water, C_w, and sediments, C_{sed} (although sediment concentration is not required by the biota model), are calculated in each grid cell of the 0.1° × 0.1° resolution hydrodynamic model from the following equations:

$$C_w\left(i,j\right) = \frac{RN_{surf}\left(i,j\right)}{\Delta x \Delta y d_{pic}} \tag{10}$$

where $\Delta x\Delta y$ is the cell surface, $N_{surf}(i, j)$ is the number of particles within the surface layer in cell (i, j) and d_{pic} is the surface layer thickness (thus we count only particles at depth less than d_{pic}=200 m, as mentioned before). Finally R is the number of Bq corresponding to each particle, which is deduced from the number of particles used in the simulation and the magnitude of the release M (total released activity):

$$R = \frac{M}{NP}. \tag{11}$$

Radionuclide concentration (Bq/kg) in seabed sediment of cell (i, j) is:

$$C_{sed}\left(i,j\right) = \frac{RN_{sed}(i,j)}{\Delta x \Delta y L \rho_s} \tag{12}$$

where $N_{sed}(i, j)$ is the number of particles in the bed sediment of cell (i, j), L is sediment thickness (set as 0.05 m) and ρ_s is sediment bulk density. This is defined as:

$$\rho_s = \rho_m(1 - por) \tag{13}$$

where ρ_m=2600 kg/m^3 is mineral particle density and por=0.6 is sediment porosity, for which standard values were used. A label is assigned to each particle to identify if it is in water or sediment phase, which is appropriately changed along the simulation.

Once that radionuclide concentration in the surface water for each grid cell and time step is known, Eq. (8) are solved for each cell using such calculated C_w. The model implicitly assumes that adsorption of radionuclides by biota does not reduce the concentration in water. This does not lead to a mass conservation problem due to the very low amount of radionuclides which are adsorbed by biota in comparison with those in dissolved form.

2.2. Model setup and numerical experiment

The amount of ^{3}H contained in the tanks was reported to be about 1 PBq (Buesseler, 2020), and it was estimated to be about 1.22 PBq in 2022, when the Japanese government plans to start the discharges (Zhao et al., 2021). The concentration of ^{137}Cs in the water of the tanks is a factor 10^6 lower than that of ^{3}H (Buesseler, 2020). Consequently, the total amount of ^{137}Cs contained in the tanks in 2022 was fixed as 1.2 × 10^9 Bq.

Zhao et al. (2021) carried out 4 simulations (since the release procedure has not been announced yet) using different release durations, from one month to 10 years. Maximum peak concentrations are obtained in the shortest release, although more persistent concentrations are obtained in the release area in the case of long lasting discharges. Thus, we adopted a conservative methodology to predict which could be the highest concentrations and thus, a release lasting one month was simulated.

The radioactive decay constant of ^{137}Cs is 7.31 × 10^{-10} s^{-1}. The desorption rate k_2 was obtained from the experiments in Nyffeler et al. (1984), value which was used in many other modelling works (see for instance Periañez et al., 2013 and the review in Periáñez et al., 2019b): k_2=1.16 × 10^{-5} s^{-1}. The adsorption rate k_1 is deduced from the desorption rate k_2 and the radionuclide distribution (or partition) coefficient k_d for ocean waters, which can be obtained from IAEA (2004), following the procedure described in Periáñez (2008) for instance. The ^{137}Cs distribution coefficient k_d was taken as 4.0 m^3/kg, which is the recommended value for ocean waters by IAEA (2004) for this radionuclide. Parameters of the biota model are provided in Table 1, as already mentioned.

3. Results and discussion

The focus of this paper will be the resulting ^{137}Cs concentrations in sediments and biota (non-piscivorous and piscivorous fish) since the transport of radionuclides in the dissolved phase at oceanic scales was already discussed in detail in Zhao et al. (2021).

Calculated concentrations in bed sediments in the local area around the FDNPP 30, 60 and 120 days after the beginning of the 30 day long release are presented in Fig. 1. Contamination of the bed sediment is limited to this local area, and does not significantly extend offshore.

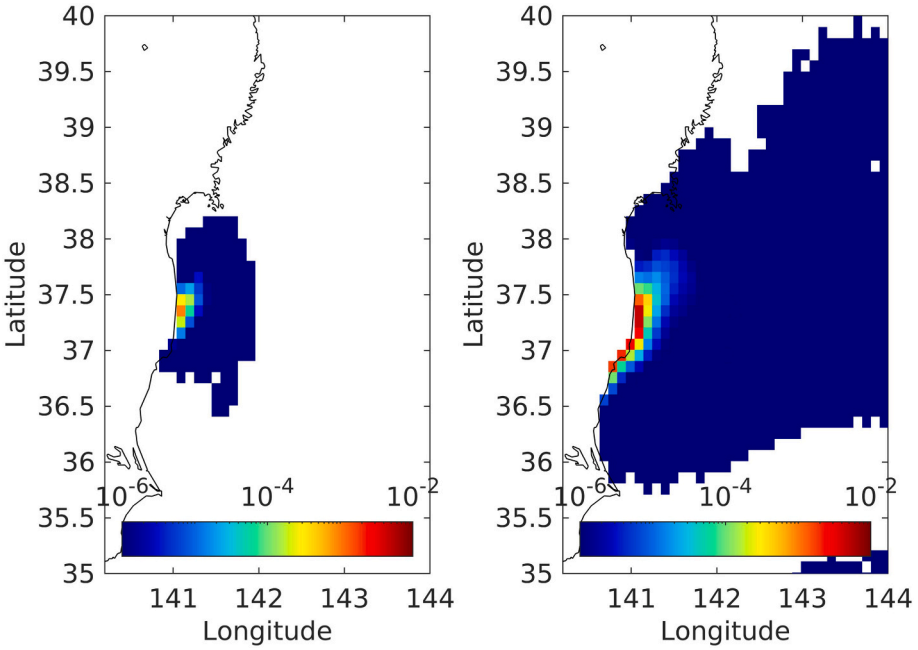

Fig. 5. Concentrations of ^{137}Cs in piscivorous fish (Bq/kg wet weight) 30 (left) and 120 (right) days after starting the one month release.

Maximum concentrations calculated by the model are of the order of 10^{-2} Bq/kg, which is two orders of magnitude below concentrations measured in sediments after FDNPP accident (Periáñez et al., 2012) and are also lower than background ^{137}Cs concentrations in sediments due to global fallout, which are in the order of 10^{0} Bq/kg. Sediments buffer radionuclides, which are slowly released back to the water column and thus the sediment acts as a long-term delayed radionuclide source. Actually, non zero (although extremely low) concentrations are still apparent after one year. This can be seen in Fig. 2: a band of sediments containing some ^{137}Cs can still be seen along the coast.

Due to the dynamic nature of the biota uptake model, radionuclide concentration build-up in fish is a slow process. Thus, peak concentrations are not found immediately after the release. This can be seen with the help of Figs. 3 and 4, where ^{137}Cs concentrations 30 and 120 days after the release (Fig. 2), as well as 1 and 2 years after if (Fig. 3) are presented for non-piscivorous fish as an example. Non-zero concentrations extend over a wider region as time elapses. Two years after the release the patch of fish containing ^{137}Cs has considerably increased its size.

Similar maps, but for the case of piscivorous fish, are presented in Figs. 5 and 6. The behavior is similar to that of non-piscivorous fish and the same comments are valid but, as this fish is the last step of the foodweb, radionuclide build-up is slightly slower than in the case of non-piscivorous fish.

However, it should be noted that peak concentrations are extremely low. In non-piscivorous they are of the order of 10^{-3} Bq/kg (wet weight) 30 days after the release start and increase to 10^{-2} Bq/kg (wet weight) after 120 days. After 1 year, peak concentration is again of the order of 10^{-3} Bq/kg (wet weight). Finally, it decreases a further single order of magnitude 10 years after the release. Peak concentrations in the case of piscivorous fish are one order of magnitude lower that those of non-piscivorous fish in the initial phase after the release, but after one year they remain of the same order of magnitude. These peak concentrations are much lower than those measured in fish immediately after FDNPP accident (Periáñez et al., 2019a) in the close area to FDNPP: 440 Bq/kg (wet weight) and 7300 Bq/kg (wet weight) respectively for non-piscivorous and piscivorous fish; as well as to ^{137}Cs concentrations due to fallout background, which are about 10^{-1} Bq/kg (wet weight).

As a final, example, calculated concentrations of ^{137}Cs in non-piscivorous fish 10 years after the release are presented in Fig. 7. Concentrations over the whole ocean are extremely low, although the patch of traced fish increases its size and expands into the Sea of Japan and Yellow Sea. Nevertheless, these concentrations would be well below detection limits.

There are other radionuclides contained in the tanks with similar (or even higher) concentrations to that of ^{137}Cs (Buesseler, 2020). That is the case of ^{90}Sr, whose geochemical behavior is similar to that of Ca. Thus, it is a significant radionuclide from a radiological point of view, since it is accumulated in bones. ^{60}Co, for instance, presents a strong affinity to be fixed to sediments. Consequently, high concentrations in the sediments of the release area might be produced. Therefore, additional studies for other radionuclides should be carried out due to their very different fates and toxicities in the marine environment.

4. Conclusions

The potential ^{137}Cs concentrations in sediments and biota resulting from the planned releases of FDNPP contaminated waters were evaluated using a Lagrangian numerical model which integrates a dynamic foodweb biological uptake model. The Lagrangian model includes physical transport (advection and diffusion), radioactive decay and interactions of radionuclides between water and sediments. A conservative approach was adopted to calculate the potential maximum concentrations, which implies assuming a short release (one month was used) scenario.

Contamination of the bed sediment is limited to a local area around FDNPP, and does not significantly extend offshore. Maximum concentrations calculated by the model are orders of magnitude below concentrations measured in sediments after FDNPP accident and also lower than background ^{137}Cs concentrations in sediments due to global fallout. Similarly, extremely low concentrations are calculated for non-piscivorous and piscivorous fish. Consequently, a significant effect of these releases in the environment should not be expected according to the present model.

CRediT authorship contribution statement

R. Periáñez: conceptualization, methodology, formal analysis,

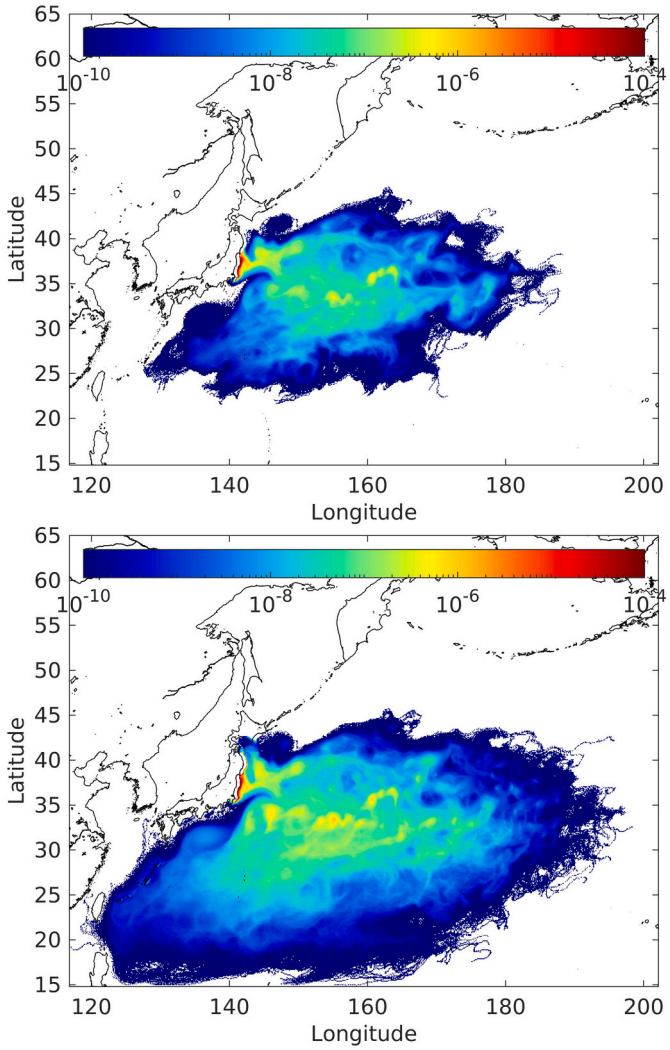

Fig. 6. Concentrations of ^{137}Cs in piscivorous fish (Bq/kg wet weight) one (top) and two (bottom) years after the one month release.

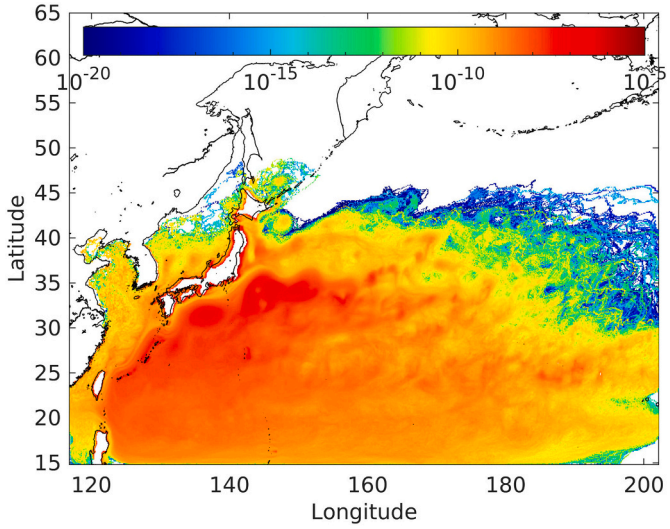

Fig. 7. Concentrations of ^{137}Cs in non-piscivorous fish (Bq/kg wet weight) ten years after the one month release.

writing (original draft).

All other authors: conceptualization, formal analysis, writing (review).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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