Response to Reviewer #1

The authors are most grateful to the reviewer for thorough analysis of manuscript and for his constructive criticism and suggestions. We have taken his remarks into account, and the paper has been revised in many places accordingly.

Scientific significance: This is an inspiring paper which includes a dynamic pelagic food chain as well as benthic food chain. This is seldom seen in radioecology and hopefully opens up a world of new ideas in radioecology. At the same time it needs to maintain its connections to marine ecology, where the benthic and pelagic food webs have been studied for a long time. Thus it needs to be understandable both for radioecologist and marine ecologist, which can be difficult to achieve. Below are some comments how this can be improved.

The presentation quality of paper is good, well written and structured, even if the connection between the two sites seem to be only the model and Cs. I don't see any discussion or comparison what the difference is between the sites, just examples.

The scientific quality has a good appearance, but when looking closer to the supporting model and references the results are weak. There is simple to little data to support the modelling results (exemplified below) . Moreover the scientific nomenclature is not consistent with e.g. marine ecological nomenclature and exact description of e.g. species.

Answer. The model results were compared with observations in two very different marine environments: in the North Western Pacific and in the Baltic Sea before and after Fukushima and Chernobyl accidents, respectively. The added observations for 2015-2016 in Figs. 3 and 4 support the generic model predictions. These figures and updated Supplement are given after text of response. The detailed answers on the rest of reviewer comments are given below.

1. *Although it is a step forward to include the foodchains, both the bentic and pelagic foodweb presented here miss the important microbial loops and even meiofauna. The microbial loop has been discussed the last three decades in oceanography and limnology (see review in Fenchel 2008 Journal of Experimental Marine Biology and Ecology 366 (2008) 99–103 . The depicted foodchain in figure 1 doesn't include and nothing is mentioned in the text. I can understand that there reasons not to include them in the model, but there a no reasons to omit them without explanation. This will certainly cause doubts of sound science of this paper by marine ecologist on this paper.*

Answer: We agree that a full model of pelagic and benthic food webs should include a variety of transfer processes in water and in the sediment. However, we consider here the more limited task of biota model development and its implementation into the compartment model which is in turn a component of the decision-support system RODOS for nuclear emergency. Therefore, a number of simplifications have been made in order that the model is robust and generic, requiring a minimum number of parameters. It is assumed that the radioactivity concentrations in organic and mineral fractions of bottom deposit are in mutual equilibrium, and the radioactivity concentrations in microbial biota and non-living organic matters also are in equilibrium, and only organic matter in the bottom deposit is bioavailable. The text was changed accordingly (see answer on comment #2). To explain model assumptions and limitations we reworked text in lines 94-99 as

“To describe transfer pathways of ^{137}Cs from bottom sediments to marine organisms the dynamic model BURN was extended. The model was developed to assess doses from marine products in the decision-support system RODOS for off-site nuclear emergencies (Lepicard et al., 2004). For such aim it was necessary to use a robust and generic model requiring a minimal number of parameters. Therefore, in the model the marine organisms are grouped into a few classes based on trophic levels and types of species. The radionuclides are also grouped in several classes in terms of tissues in which a specific radionuclide accumulates preferentially. These simplifications allow for a limited number of standard input parameters.”

2. *There are problems with the classifications in the paper of the different trophic groups discussed in the paper and shown in figure 1. They are not consistent and classification with the same variables. E.g what is a costal predator and what difference compared to piscivoures fish? The example given is cod. In the Baltic Sea it certainly would be regarded a Piscivorus fish you can everywhere not only coast. Algae (fig 1) and phytoplankton are the same, you maybe mean benthic algae or macroalgae. In table 1 you call it macroalgae Demersal fish and Benthic predator what is the difference? Example is given with European flounder which certainly is a benthic predator and demersal fish at the same time. In Fukushima we can read about Rockfish , what is that? There are least a dozen fish genera which can be called rockfish, they have different position in the food chain. This needs better description or at least the scientific (latin) name.*

Answer: We agree with referee’s comment #30 “*that it can never become clearcut where different species belong.”* A good example is the omnivorous predator Atlantic cod (Gadus morhua) in the Baltic Sea. Diet of cod in deep Central Baltic can be dominated by herring and sprat. However in shallow Western Baltic (box 45 in Fig. 5, depth 31.4 m) diet is diverse, including herring, sprat, Gobiidae, the molluscs, various Polychaeta and crustaceans (Sparholt, 1994). Therefore for this basin the cod is considered as “coastal predator” feeding by both pelagic and benthic preys. Following reviewer’s comment, “Algae” in Fig.1 renamed to “Macroalgae”. According Gibson and al. (2015) European flounder (Platichthys flesus) belongs to group of “Polychaete and small crustacean feeders”. See more details in answer on comment #37. Rockfish is “Japanese rockfish” (Sebastes Cheni). The Latin names are given for species when observations and simulation results are compared (see answers on comments #28 and #36).

The text and caption to Fig. 1 were changed to extend description of food web and explain classification approaches used in the paper.

Lines 99-114 “The transfer scheme of radionuclides through the marine food web is shown in Fig. 1 where transfer of radionuclides through the food web is shown by arrows whereas the direct transfer from water is depicted by the shadowed rectangle surrounding 11 biota compartments (i=1,…,11). The different food-chains exist in both pelagic and benthic zones. Pelagic organisms are divided into primary producer, phytoplankton (i=1), and consumers which consist of zooplankton (i=2), forage (non-piscivorous) fish (i=3), and piscivorous fish (i=4). The benthic food web includes three primary pathways for radionuclides: (i) transfer from water to macroalgae (i=5), then to grazing invertebrates (i=6,..,8); (ii) through the vertical detritus flux and zooplankton faeces (Fowler et al., 1987) to detritus-feeding invertebrates, and (iii) through contaminated bottom sediments to deposit feeding invertebrates. Concentrations of radionuclides in water and in the upper layer of bottom sediment are calculated using the box model POSEIDON-R described below. The output from this model is shown by external boxes in Fig. 1. The radionuclides adsorbed on the organic matter in the sediments are bioavailable for benthic organisms but the mineral component of sediments is not (Ueda et al., 1977; Ueda et al., 1978) although Koyanagi et al. (1978) found relatively rapid and more intensive transfer of several sediment adsorbed radionuclides (^{54}Mn, ^{60}Co, ^{65}Zn) to particular organs of the demersal fishes in contrast to flesh. It is assumed that (i) radioactivity concentrations in organic and mineral fractions of bottom deposit are in mutual equilibrium, (ii) that radioactivity concentrations in microbial biota and non-living organic matter also are in equilibrium and (iii) that only organic matter in the bottom deposit is bioavailable. The benthic invertebrate group (surrounded in Fig. 1 by dashed rectangle) includes molluscs (e.g. filter—feeders) (i=7), crustaceans (e.g. detritus-feeders) (i=6) and subsurface and surface deposit feeders (e.g. annelid). It is assumed that radioactivity is transferred from invertebrates to benthic invertebrate feeding demersal fishes (i=9), and on to omnivorous bottom predators (i=10) (Fig. 1). The marine food web also includes "coastal predators" (i=11) feeding in the whole water column in shallow waters."

Line 443. "Calculated and observed ^{137}Cs concentrations in the coastal predator (cod) also agree well with the measurements (Fig. 7d). The diet of Atlantic cod in shallow Western Baltic is diverse, including herring, sprat, Gobiidae, molluscs, various Polychaeta and crustaceans (Sparholt, 1994). Therefore for this basin the cod is considered as "coastal predator" feeding by both pelagic and benthic preys. The geometric mean of the simulated-to-observed ratios is 0.91 with a geometric standard deviation of 1.37 for a total number of observations N=95 in the whole Baltic Sea."

Sparholt, H.: Fish species interactions in the Baltic Sea. Dana, 10, 131-162, 1994.

Caption to Fig. 1
"Figure 1. Scheme of radionuclide transfer to marine organisms. A transfer of radionuclides through food web is shown by arrows whereas direct transfer from water is depicted by shadowed rectangle surrounding biota compartments. The output from the compartment POSEIDON-R model is shown by external boxes."

3. *There are other filterfeeders than mollusc and mollusc can be grazers and deposit feeders. Benthic algae are consumed by grazer also. Why the difference between deposit feeding invertebrates and crustacean invertebrates Crustaceans are certainly many of the Zooplankton.*

Answer: As shown in Fig. 1 and in Table 2 benthic algae are part of diet of crustaceans, molluscs and deposit-feeding invertebrates (e.g. echinoidea). Deposit feeders include subsurface deposit feeders (e.g. worms). We consider "crustaceans" as a part of benthic food web.

4. *No explantion in figure 1 what are the arrows, boxes, dotted lines, where are the explantions of the categories. What are the numbers?The dotted box with a waterbox outside? Water deposit what is that and why is that box outside ?*

Answer: See answer on Comment #2.

5. *Figure 2. What are deep water boxes ? What are coastal box? Describe or give criteria or point to text where that is described*

Answer: The text and figure caption have been changed accordingly.

Line 228 “The model was customized for the Northwestern Pacific Ocean, the East China and Yellow Seas and the East/Japan Sea. A total of 176 boxes cover this entire region (Fig. S1). In the deep-sea regions a three-layer box system was built to describe the vertical structure of the radioactivity transport in the upper layer (0-200 m), intermediate layer (200-1000 m) and lower layer (>1000m). The compartments around the FDNPP are shown in Fig. 2. The “coastal” box 15x30 km is nested into large “regional” box 90 to provide more detailed description in the area around the FDNPP. It covers observation data within a circular-shaped surface area of a radius 15 km with a center at the FDNPP. This box has one vertical layer for the water column and three bottom sediment layers. The depth of coastal box is less than that in the one layer outer box 90. The water exchange fluxes with the outer box are equal in both directions. The parameters of the coastal box are given in Table S1. The averaged advective and diffusive water fluxes between regional compartments were calculated for a ten-year period (2000-2009) using the Regional Ocean Modeling System (ROMS). Details of customization are given by Maderich et al. (2014a,b). The values for parameters ϕ_{org}=0.01and $T_{migr;i}$=0.7 y for i=3,4,9,10,11 were used.”

Caption to Figure 2. “The box system for the area close to Fukushima NPP. The shaded boxes represent the deep-sea water boxes divided on three vertical layers. The NPPs are shown by filled circles. Coastal box around the FDNPP (marked by “F” is inside of box 90. Thick line limits the area of the Fukushima accident fallout .”
Caption to Figure 2S “The compartment system for the Northwestern Pacific. The shaded boxes represent the deep-sea water boxes divided on three vertical layers...”

6. *Figure 3 Explain what the legend means e.g correction of wha?t . kg of what drymatter ?? Something strange that the estimated KD for the sediment is different before and after Fukushima, especially the before values the ratio seem low if you expect a KD of 1000 l/kg*

Answer: The “dry” (weight) was added in axis title in Figs. 3b and 6b. The K_d in the simulation is constant in time. The value of K_d is given in Table S1. The caption was changed accordingly.

“Figure 3. Comparison between calculated and observed ^{137}Cs concentrations in seawater (a) and in bottom sediment (b) in the coastal box around the Fukushima Dai-ichi NPP. The dashed line in (b) shows results of simulations using standard POSEIDON-R model, whereas solid line presents simulation with correction term in equation (S3).”

7. *The paper identifies that an important process is missing resuspension, and it is compensation with some unclear equations. In the Baltic Sea this certainly is a much more important process than e.g. diffusion. I can imagine that it could be important outside Fukushima especially since the organic content is so low (<25% line 50). Thus it would certainly lift this to a through discussion and conclusion, not just an equation fix.*

Answer: We used “standard” parameterization of transfer between water and bottom sediments following approach developed in series of EC MARINA projects. In this approach resuspension was not included, however, “standard” parameterization was successfully used e.g. for the Baltic Sea (MARINA-BALT). The Chernobyl case simulation confirms that the standard parameterization describes well exchange processes for the Baltic Sea (see answers on Comment #40). In the Fukushima case study we identified that ^{137}Cs decreases in upper layer of sediments faster than model predicts using standard parameterization. A several possible mechanisms were mentioned in Lines 286-291 but there have been no study confirming dominance of one of these mechanisms. Therefore, the very simple parameterization was used in the model because the main aim of our study is transfer radiocaesium through the benthic food chain. The text was changed accordingly.

Line 468 "It was found that ^{137}Cs decreases in upper layer of sediments in the Fukushima case study faster than POSEIDON-R predicts using the standard for marine compartment model parameterization of exchange between water and sediment by diffusion mechanism. A simple parameterization calibrated on measurements was therefore used to correct this exchange. However, the further studies of exchange mechanisms are necessary."

8. *Line 26 : What does the biomagnification effect mean here? See e.g. Gray 2002 Marine Pollution Bulletin 45 (2002) 46–52 Biomagnification in marine systems: the perspective of an ecologist*

Answer: See answer on Comment 27

9. *Line 50 says that it is bound to organic matter. It is unclear if that means dead matter och e.g microbes or both.*

Answer: There is no explicit discussion on the origin of organic matter in these papers. However, it can be concluded from the description of methods in (Ono et al., 2014) that the total organic matter passed through the 2 mm mesh sieve was tested. See also answer on comments #1 and 2.

10. *Line 66: What is an underground leakage in this context?*

Answer: The routes of radioactive water from the FDNPP were not exactly identified yet (Kanda, 2013). It can be assumed that possible pathway is transport by ground water leaked from damaged facilities. Text was changed accordingly

Line 66 "In that study the flux of radionuclides due to the ground water leakage of contaminated waters from FDNPP (Kanda, 2013) was taken into account."

11. *Line 109: I don't understand "rapid and more intensive transfer of several sediment adsorbed radionuclides to particular organs of the demersal fishes" in contrast invertebrates? Or as another source of contaminants?*

Answer: The text was changed:

Line 109 "although Koyanagi et al. (1978) found relatively rapid and more intensive transfer of several sediment adsorbed radionuclides (^{54}Mn, ^{60}Co, ^{65}Zn) to particular organs of the demersal fishes in contrast to flesh."

12. *Line 111: In this context I don't understand the role of macroalgae (and why not benthic microalgae) I would also assume that crustaceans and molluscs are able to graze the algae not only deposit feed. Moreover there no data about the macralgae and for me if the depth of the coastal box is 60m there must be large areas outside the photic zone. How is that estimated?*

Answer: The macroalgae were considered in the food chain because they are a component in the diet of the molluscs, crustaceans and invertebrates with dominant deposit feeding (Table 2). They also are part of human diet and are important for dose estimates. We used a simple approach where the benthic component with macroalgae was included in the shallow one-layer compartments adjacent to the shore that guaranteed range of depth for macroalgae photic zone. The text was changed accordingly:

Line 209 “The model for the pelagic food web component was implemented for the upper water layer of all compartments, whereas the benthic component was included in the shallow one-layer compartments adjacent to the shore”.

13. *Line 130: Why not call the food abstraction coefficient assimilation efficiency which is the normal biological word*

Answer: The extraction coefficient was changed on “assimilation efficiency” in Line 130

14. *Lines around 155: Since the classification not very systematic the relationships between these fish types are unclear also.*

Answer: See answers on comment #2.

15. *Line 191 and forward: The description of the POSIEIDN model should be helped with a figure showing the compartments and where the additional food web interact with POSEIDON. Moreover the parameter value for the two sites should be tabulated somewhere, without this information it is not possible to reproduce the results.*

Answer: The POSEIDON model equations and figure with compartment structure (Fig. S1) are given in Supplement. The parameters of two boxes from the Pacific Ocean and the Baltic Sea are given in Table S1. Text was changed accordingly:

Line 193 “ The compartments describing the water column containing suspended matter are subdivided into a number of vertical layers as shown in Fig. S1.”
Line 208 “The model equations are given in Supplement”.
Line 234 “The parameters of the coastal box are given in Table S1”.

16. *Line 204: An important transfer from sediment to water column is resuspension.*

Answer: See answer on Comment #7 .

17. *Line 210: shallow one layer compartment ? another sedimentlayer or a description of the sectors in POSEIDON? If the later wasn’t that the same compartment as the pelagic food web?*

Answer: We described a structure of compartments in the Supplement and in the text. See answers on Comment #5. The shallow one water column layer and three sediment layer compartments include both pelagic and benthic food webs. The text was refined as:

Line 209 “The model for the pelagic food web component was implemented for the upper water layer of all compartments, whereas the benthic component was included in the shallow one-layer compartments adjacent to the shore”.

18. *Line 235: Somewhere I am missing a table giving the parameters of the model. Also a description of the average depth of the site and bottom substrate is missing*

Answer: We added Table S1 where these parameters were given.

19. *Line 275: Do you mean the geometric mean of the ratio? Between measured to observed values?*

Answer: It is geometric mean of ratio between simulated by model and observed in ocean values. The text was changed in several places as

Lines 298, 312, 327, 420, 433, 441,444 “…geometric mean of the simulated-to-observed ratios..”

20. *Line 293: .... "cannot account"... you mean maybe " not included in the model", it is not clear.*

Answer: The text was changed accordingly.

Line 291 "Only several of these mechanisms are included in the POSEIDON-R model."

21. *Line 294: it is not easy to understand where these terms are added into which equation. This addition seem crucial to the model and needs to be presented clearer and completely to be transparent. Moreover I get the impression that this is some sort of calibration to make the model fit for the measurements. Or how is it obtained?*

Answer: The text was changed accordingly.

Line 293 "Therefore, to take into account the vertical transfer of ^{137}Cs we added the exchange terms $(C_{s,1}-C_{s,2})\lambda_s$ and $-(C_{s,1}-C_{s,2})\lambda_s$ to the right hand side of the equations (S3) and (S4) for the concentration of radioactivity in upper ($C_{s,1}$) and medium ($C_{s,2}$) layers of sediment in the coastal box, respectively. Here λ_s is an empirical parameter. The value of λ_s=0.4 y^{-1} was obtained to fit observation data for $C_{s,1}$. As seen in Fig. 3b the corrected by additional exchange term concentration of ^{137}Cs is described well in period 2008-2015."

22. *Line 304: You mention sea urchin here, is that detritus feeding or a grazer in real life and what group is it represented in the model, invertebrate?*

Answer: According to Lawrence (2007) the principal foods of sea urchin (*Strongylocentrotus nudus*) include large and small algae, detritus, sand, shells, sessile animals and fish. The model diet for deposit feeding invertebrates includes both macroalgae and organic matter in the bottom deposit that grossly represent transfer of ^{137}Cs through food to S. nudus. Notice that among of benthic invertebrates only data on the sea urchin were available for the 15 km area around the FDNPP. The text was added:

Line 305 "This is consistent with model diet that includes macroalgae and deposit organic matter grossly representing diet of S. nudus (Lawrence (2007). The macroalgae contribution in food contamination first prevails, then after 2012 the bottom contamination dominates."

Lawrence J. M. (ed): Edible sea urchins: Biology and ecology. Developments in Aquaculture and Fisheries Science, 37, Elsevier, Amsterdam, Netherlands, 529 pp., 2007.

23. *Line 306: Here you mention depuration constant for the first time. I am unsure if it can be called depuration constant at least from the ecotoxicological viewpoint, moreover this constant could be mentioned the first time it occurs and explained what it is.*

Answer: We defined depuration constant as a decrease constant in the fitted exponential function of concentration (see Line 305). The depuration constant is equal to $(\ln 2 T_{e1/2})^{-1}$, where $T_{e1/2}$ is ecological half-life. The term "depuration rate constant" is used in marine radioecology (see e.g. Sohtome et al., 2014; Tateda et al., 2013;2015).

24. *Line 309: What do you mean with transfer coefficient here ? Concentration ratio?*

Answer: See answer on Comment #25

25. *Line 311: What kind of polychaete, deposit feeding or filterfeeding, to unspecific without species name*

Answer: The text was changed accordingly comments #24-25:

Line 311. "The field studies of several species of polychaeta (deposit or filter feeders: *Flabelligeridae*, *Terebellidae* and *Opheliidae*; herbivore or carnivore feeders: *Glyceridae, Eunicidae,* and *Polynoidae*) off the coast of Fukushima and rearing experiment for *Perinereis aibuhitensis* demonstrated that ^{137}Cs concentration in all specimens was much lower than that in the sediment (Shigenobu et al., 2015). Results of rearing experiment using contaminated sediments from near the FDNPP showed that transfer coefficient (concentration ratio between *P. aibuhitensis* (Bq kg^{-1}-wet) and contaminated sediment (Bq kg^{-1}-wet)) was less than 0.1. "

26. *Line 318: Reference needed for the experimental value or/and description of the experiment. Crucial is how they are fed and how the radionuclide is added*

Answer: The data are the field data from TEPCO (2015). The text was corrected accordingly.

Line 317 "The simulated values of the depuration constant is 0.46 y^{-1} whereas estimated from the field data for 2012-2015 in Fig.4b is 0.48 y^{-1}"

27. *Line 322: What do you mean with biomagification effect (see earlier comment) and how should that affect the CR in demersal fish mechanistically?*

Answer: The text was changed accordingly comments #8 and #27.

Line 24 "The estimated from model transfer coefficient from bulk sediment to demersal fish in the model for 2012-2020 (0.13) is larger than that to the deposit feeding invertebrates (0.07)."
Line 320 "Notice that the predicted transfer coefficient from bulk sediment to demersal fish for the period of 2012-2020 is approximately 0.13. This value is larger than that for deposit feeding invertebrates. The observed in this area BCF for demersal fish (flounders) in 2013-2015 is 0.9 m^{3}kg^{-1}, whereas the standard value of BCF for fish is 0.1 m^{3}kg^{-1}(IAEA, 2004) that confirms the importance of transfer of radiocaesium to demersal fish from the sediments."

28. *Line 323: Again inexact species and categorisation, there are several genus called rockfish, what is the scientific name? How does it fit into the classification? Coastal predator?*

Answer: We added in text scientific names for all organisms presented in Fig. 4.

Line 302 "The symbols in Fig. 4 are observation data for sea urchin (*Strongylocentrotus nudus*) (a), flounders (*Microstomus achne, Kareius bicoloratus*, *Pleuronectes yokohamae*) (b) and Japanese rockfish (*Sebastes cheni*) (c). The open and filled symbols in Fig. 4d are data for seabass (*Lateolabrax japonicas*) and fat greenling (*Hexagrammos otakii*), respectively."
Line 323 "Comparison of simulations with observations for a bottom predator (Japanese rockfish) in Fig. 4c shows a good agreement."

29. *Line 326: The legend to the figure could be put into figure text*

Answer: See answer on comment 28.

30. *Line 327-333: This is interesting results and probably support your approach, but it is messed up with inconsistent classifications. My suggestion that you first of all make a consistent classification, a clear description what that means and finally give examples of species in the area for each group. This should be done in methods, the you adhere*

to the classification when you mention different species with common and scientific names (latin) . I know that it can never become clearcut where different species belong, but you tell at least the reader where they are in the model.

Answer: See reworked text with description of each group of organisms in comment #2. The scientific species names are given in answers on comments#28 and #36.

31. *Line 334: ".. which are known with high uncertainity." Maybe better ... known to have a high...*

Answer: Done.

32. *Line 352: probably figure S3b not 3b*

Answer: The figure number was changed to Fig. S4b.

33. *Line 355: why this different numbers ?*

Answer: The text was changed accordingly:
"The maximum ^{137}Cs concentration for zooplankton using the maximal value of $T_{0.5,i}$ was increased by a factor 2.7 compared with a case when the minimum value of $T_{0.5,i}$ was used. This factor for pelagic fish and coastal predator was in the range 2.4-1.7 whereas for the rest organisms it was smaller."

34. *Line 363: also probable wrong figure number*

Answer: The figure number was changed to Fig. S4d.

35. *Line 363-384: I would suggest to omit this part, there are assumptions and limits with different relevance in different parts of the world. From my horizon (responsible for dose assessments) I cannot see the point of this section. Omit it the also from conclusion*

Answer: Done.

36. *Line 385-410: If the modelling of the Baltic sea should be useful, this section should at least tabulate the drivers (fluxes over borders) and parameter values for the modelled box for the result. It is not reproducible with the current information. I am also missing general data on the bathymetry and which species are considered in the model foodweb.*

Answer: We added Table S2 with river runoff into the Baltic Sea and Table S1 with parameters for box 45 in the Baltic Sea.The scientific names for all organisms presented in Fig. 7 were also added.

Line 425 "The symbols in Fig. 7 are observation data for echinoderms (*Echinodermata*) (a), sprat (*Sprattus sprattus*) (b), European flounder (*Platichthys flesus*) (c) and Atlantic cod (*Gadus morhua*) (d)."

37. *Line 440: Polychaete feeding is that valid for the Baltic Sea*

Answer: We used information on diet of the European flounder (Platichthys flesus) from Gibson, R. N., Nash, R. D. M. Geffen, A. J., and Van der Veer, H. W. (eds.): Flatfishes: biology and exploitation. - Second edition Wiley-Blackwell, Chichester, UK, 2015. According Gibson and al. (2015) these fishes belong to group of "Polychaete and small crustacean feeders" (Table 11.1). This table provides more detailed information the European flounder in the Baltic Sea: it feeds by oligochaetes, amphipods, chironomids and smaller sizes harpacticoids. Text was changed accordingly.

Line 440 “Notice that European flounder diet in the Baltic Sea includes oligochaetes, amphipods, chironomids and smaller sizes harpacticoids. (Gibson et al., 2015).

38. *Line 459: As commented earlier the classification system needs to be reworked*

Answer: See answers on Comment #2.

39. *Line 464: Suggest to omit “strongly”, it is not relevant for the Baltic Sea and I don’t think I adds something more for Fukushima area.*

Answer: The text was changed accordingly.

Line 464: “The compartment model was applied to two regions (north western Pacific (NWP)) and the Baltic Sea) which were contaminated due to accidents on the Fukushima Dai-ichi and Chernobyl NPPs.”

40. *Conclusion or discussion I am missing a more rigorous comparision between the Baltic Sea and Fukushima, otherwise I don’t see the point include both in this paper.*

Answer: The text was added accordingly.

Line 387 “The Baltic Sea is an important case because of its transfer of ^{137}Cs originating from the Chernobyl fall-out. It was chosen to verify the ability of the model with generic parameters to describe transfer processes in a semi-enclosed sea with very different oceanography.”

Line 451 “The observed BCFs in this area for sprat, European flounder and Atlantic cod in 1990-2010 are 0.11, 0.14 and 0.15 m^3kg^{-1}, respectively. This is close to the standard value of BCF for fish 0.1 m^3kg^{-1}(IAEA, 2004) taking in account that waters in the Baltic Sea are brackish that affects the uptake rate of radiocaesium. These results essentially differ from the Fukushima case where BCF for demersal fish was an order greater confirming importance of transfer of radiocaesium from the sediments to demersal fish for that case.”

Line 494-500 “The results of the application of POSEIDON-R with an extended dynamic model to the Baltic Sea which is semi-enclosed and filled by brackish waters are in good agreement with available measurements in the Baltic Sea. Unlike the highly dynamical off coast processes caused by eddy dominated currents in the Pacific Ocean where the FDNPP is located, weak water exchange with the North Sea and regular circulation in the Baltic Sea results in a slow quasi-equilibrium evolution of water-sediment-biota system. The Chernobyl case confirms that the standard parameterization of water-sediment exchange used in POSEIDON-R describes well the exchange processes for the Baltic Sea whereas in the Fukushima study the observed value of ^{137}Cs decreases faster in the upper layer of the sediments than that the model predicts using the standard parameterization. In the Fukushima accident case the concentration of ^{137}Cs in piscivorous fish decreases faster than in the coastal predators whereas in the Chernobyl case these concentrations decrease simultaneously. The obtained results demonstrate the importance of the benthic food chain in the long-term transfer of ^{137}Cs from contaminated bottom sediments to marine organisms and the potential of a generic model for use in different regions of the World Ocean.”

41. *No explanation in figure 1 what are the arrows, boxes, dotted lines, where are the explanations of the categories. What are the numbers? The dotted box with a waterbox outside? Water deposit what is that and why is that box outside ?*

Answer: The text and capture for Fig. 1 were changed accordingly. See answer on Comment #2.

42. *Figure 2. What are deep water boxes ? What are coastal box? Describe or give criteria or point to text where that is described*

Answer: See answer on Comment # 5.

43. *Figure 3 Explain what the legend means e.g correction of wha?t . kg of what drymatter ??*

Answer: See answer on Comment #21

44. *Figure 5 explain color-coding*

Answer: The text was added accordingly

"Figure 5. Compartment system of POSEIDON-R model for the North-Eastern part of the Atlantic Ocean, the North Sea and the Baltic Sea. The shaded boxes represent boxes divided on two vertical layers."

45. *Figure 6 is the concentration in bottom sediment for the bul sediment or organic fraction (the same question applies for Fukushima)*

Answer: The text was changed accordingly.

"Figure 3. Comparison between calculated and observed ^{137}Cs concentration in seawater (a) and in bulk bottom sediment (b) in the coastal box around the Fukushima Dai-ichi NPP."
"Figure 6. Comparison between calculated and observed ^{137}Cs concentrations in seawater (a) and in bulk bottom sediment (b) for box 45."

46. *FigS3 There no figtext for d)*

Answer: The text in the figure caption was corrected inserting (d).

Response to Reviewer #2

The authors are most grateful to the reviewer for his constructive criticism and suggestions. We have taken his remarks into account, and the paper has been revised accordingly.

The paper is substantially corrected which makes reader more being easier to understand the result of the paper. Following points are recommended to reconsider or revise before publishing.

1) Citation of Matsumoto et al., 2015 should not be cited. The methodology in this paper (gross counts of Cs-137 energy band in whole body fish minus background counts without fish) contains bottom-up effect on net count by neglecting Compton effect from K-40 in fish flesh, which makes the turnover rate being overestimated.

Answer: Text was changed accordingly. The reference on Matsumoto et al. (2015) was excluded from text.

Line 153 “The biological half-life data for fish flesh (Baptist and Price, 1962; Coughtrey and Thorne, 1983; Tateda, 1994,1997; Zhang et al., 2001) show variability in a large range (35-180 days) due to the differences between species and due to the differences in the experiment methodology.“

2) Rational for radiocesium turnover in bone etc. other than flesh should be shown by citation or theoretical assumption. It will help following further research by similar approach.

According to Yankovich et al. (2010) the concentration of radiocaesium in muscle is 1.65 times higher than in the bone for marine fish. In combination with ratio between weight fractions of muscle and bone (0.845 and 0.135, respectively) the total amount of caesium in fish bone can be estimated only as 9% compared with muscle (90%) and organs (1%). Therefore, in a first approximation radiocaesium turnover in bones and organs was not considered. Text was added accordingly.

Line 187 “According to data from Yankovich et al. (2010) amounts of radiocaesium in flesh, bone and organs are 90%, 9% and 1%, respectively.”

Line 16 “released”

Answer: Text was changed accordingly.

Line 23 "evaluated as caused"

Answer: Text was changed accordingly.

Line 26 Is it sure to say? Better to refer only in main text as suggestion.

Answer. "due to the biomagnification effect. " was excluded from text

Line 33 "...suggest the substantial contribution…"

Answer: Text changed accordingly.

Table 3 Is there citation for the BHL of radiocesium in fish bone? Or theoretical assumption?

Answer: Text was added

Line 161"The biological half-life for bone was estimated using data for ^{90}Sr, which is mainly accumulated in bone. This value was calculated for non-piscivorous and piscivorous fish using equation (1) in equilibrium approximation to satisfy BCF values from IAEA (2004)."

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~~TRANSFER OF RADIOCAESIUM FROM CONTAMINATED BOTTOM SEDIMENTS TO MARINE ORGANISMS THROUGH BENTHIC FOOD CHAIN IN POST-FUKUSHIMA AND POST-CHERNOBYL PERIODS~~Transfer of radiocaesium from contaminated bottom sediments to marine organisms through benthic food chain in post-Fukushima and post-Chernobyl periods

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Abstract. After the earthquake and tsunami on 11 March, 2011 damaged the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), an accidental release of a large amount of radioactive isotopes into both the air and the ocean occurred. Measurements provided by the Japanese agencies over the past ~~four~~ five years show that elevated concentrations of ^{137}Cs still remain in sediments, benthic organisms and demersal fishes in the coastal zone around the FDNPP. These observations indicate that there are ^{137}Cs transfer pathways from bottom sediments to the marine organisms. To describe the transfer quantitatively, the dynamic food chain model BURN has been extended to include benthic marine organisms. The extended model takes into account both pelagic and benthic marine organisms grouped into several classes based on their trophic level and type of species: phytoplankton, zooplankton, and fishes (two types: piscivorous and non-piscivorous) for the pelagic food chain; deposit feeding invertebrates, demersal fishes feeding by benthic invertebrates and bottom omnivorous predators for the benthic food chain; crustaceans, molluscs and coastal predators feeding on both pelagic and benthic organisms. Bottom invertebrates ingest organic parts of bottom sediments with adsorbed radionuclides which then migrate up through the food chain. All organisms take radionuclides directly from water as well as food. The model was implemented into the compartment model POSEIDON-R and applied to the Northwestern Pacific for the period of 1945-2010 and then for the period of 2011-2020 to assess the radiological consequences of ~~releases of~~ ^{137}Cs released due to FDNPP accident. The model simulations for activity concentrations of ^{137}Cs in both pelagic and benthic organisms in the coastal area around the FDNPP agree well with measurements for the period of 2011-2015. The decrease constant in the fitted exponential function of simulated concentration for the deposit ~~ingesting~~ feeding invertebrates (0.45 y^{-1}) is close to the decrease constant for the sediment observations (0.44 y^{-1}), indicating that the gradual decrease of activity in the demersal fish (decrease constant is 0.46 y^{-1}) ~~was~~ evaluated as caused by the transfer of activity from organic matter deposited in bottom sediment through the deposit feeding invertebrates. The estimated from model transfer coefficient from bulk sediment to demersal fish in the model for 2012-2020 (0.13) is larger than that to the deposit feeding invertebrates (0.07)~~due to the biomagnification effect~~. In addition, the transfer of ^{137}Cs through food webs for the period of 1945-2020 has been modelled for the Baltic Sea that was essentially contaminated due to global fallout and the Chernobyl accident. The model simulation results obtained with generic parameters are also in good agreement with available measurements in the Baltic Sea. Due to weak water exchange with the North Sea of the semi-enclosed Baltic Sea the chain of water-sediments-biota slowly evolves into a quasi-equilibrium state unlike the processes off the open Pacific Ocean coast where the FDNPP is located. Obtained results ~~demonstrate the importance~~ suggest the substantial contribution of the benthic food chain in the long-term transfer of ^{137}Cs from contaminated bottom sediments to marine organisms and the potential of a generic model for the use in different regions of the World Ocean.

1 Introduction

A catastrophic earthquake and tsunami that occurred on 11 March, 2011 severely damaged the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). The loss of power and the subsequent overheating, meltdowns, and hydrogen explosions at the FDNPP site resulted in the uncontrolled release of radioactivity into the air and ocean (Povinec et al., 2013). The atmospheric fallout over the land and the ocean peaked in mid-March whereas the direct release to the ocean from FDNPP peaked in the beginning of April. Approximately 80% of the radioactivity released due to the accident in March-April 2011 was either directly discharged into the ocean or deposited onto the ocean surface from the atmosphere. The concentration of ^{137}Cs in the ocean reached a maximum in mid-April of 2011 and has thereafter declined (by a factor of 10^5), except for the area around the FDNPP, where continuous leaks of contaminated water have been reported (Kanda, 2013). However, the concentration of ^{137}Cs in the bottom sediment that was contaminated by water with high concentrations in April-May 2011 remains quite high and is showing signs of very slow decrease with time (Otosaka and Kobayashi, 2013; Kusakabe et al. 2013; Ambe et al 2014; Black and Buesseler, 2014). The concentration of organically bound ^{137}Cs in coastal areas is several times higher than that of the bulk sediment (Otosaka and Kobayashi, 2013; Ono et al., 2015) due to ^{137}Cs adsorption on organic matter. It is worth noting that organic content in the shelf of Fukushima and Ibaraki Prefectures varies in the range of 0.1-25% (Otosaka and Kobayashi, 2013; Ambe et al., 2014; Ono et al., 2015). The preferential adsorption of ^{137}Cs on organic matter can be explained by the partial coverage of fine mineral sediment by organic substances and subsequent blocking of sorption (Kim et al., 2006; Ono et al., 2015). Comparison of the concentration of ^{137}Cs in the sediment and benthic invertebrates (Sohtome et al., 2014) and in the demersal fishes (Buesseler et al., 2012; Wada et al., 2013; Tateda et al., 2013) suggests that the continual ingestion of organic matter from sediments can be an important contamination pathway for all components of the benthic food web. However, in most of the benthic food web models applied to the FDNPP accident, the deposit ingestion is not included as a transfer process in the food-chain (Tateda et al., 2013; Keum et al., 2015; Vives i Batlle 2015; Tateda et al., 2015a,b; Vives i Batlle et al., 2015a,b).

Several models were used to perform long term assessments of the radiological impact in the marine environment due to the FDNPP accident (Nakano and Povinec, 2012; Maderich et al., 2014a,b). In particular, the compartment model POSEIDON-R (Maderich et al., 2014a,b) correctly predicted the concentration of ^{137}Cs and ^{90}Sr in water and sediments in the coastal box (30x15 km) around the FDNPP for 2011-2013. In ~~that study~~ these studies the flux of radionuclides due to the ~~underground~~ ground water leakage of contaminated waters from FDNPP (Kanda, 2013) was taken into account. However, the version of the dynamic food-chain model BURN (Biological Uptake model of Radionuclides) used in the POSEIDON-R model (Heling et al., 2002; Lepicard et al., 2004, Maderich et al., 2014a,b) did not take into account the benthic food web. Nevertheless the results of simulations still agreed well with observations for the first months and years when transfer from water dominated (Maderich et al., 2014a,b). Measurements following the Fukushima Dai-ichi accident suggest that transfer of radioactivity from bottom deposits through the benthic food web over a prolonged period of time can be an increasingly important factor in the radiological assessment of released radioactivity.

Another relevant case is the significant contamination of the Baltic Sea in 1986 by the deposition of activity originating from the Chernobyl accident. Unlike the coastal sea region near FDNPP, the Baltic Sea is a semi-enclosed relatively shallow sea filled by brackish waters and connected with the ocean by the narrow and shallow Danish Straits (Leppäranta and Myrberg, 2009). Within HELCOM (Helsinki Convention on the Protection of the Marine Environment of the Baltic Sea Area, www.helcom.fi) the group MORS (Monitoring of Radioactive Substances) established an internationally agreed monitoring network in 1986 and collected all the data in a common data base (MORS, 2015). Therefore, this event also represents a good test case to validate models (Periañez et al., 2015).

In this study, an extended food web model is presented that considers both pelagic and benthic foodchains. This dynamic model was implemented into the compartment model POSEIDON-R and applied to the northwestern Pacific for the period of 1945-2020 to assess the radiological consequences from ^{137}Cs released as a result of global fallout and the Fukushima Dai-ichi accident. The model was also applied to the Baltic Sea for the period 1945-2020 to show the versatile applicability of this model. The paper is organized as follows. Descriptions of the compartment model and of the extended dynamic food web model are given in Section 2. Section 3 presents the model application for the Fukushima Dai-ichi accident. The results of the model application to the Baltic Sea are given in Section 4. Section 5 summarizes the findings.

2 Model description

To describe transfer pathways of ^{137}Cs from bottom sediments to marine organisms the dynamic model BURN was extended. The model ~~is based on the approach devised by Heling~~ was developed to assess doses from marine products in the decision-support system RODOS for off-site nuclear emergencies (Lepicard et al.~~(2002)in which the~~ , 2004). For such aim it was necessary to use a robust and generic model requiring a minimal number of parameters. Therefore, in the model the marine organisms are grouped into a few classes based on trophic ~~level~~ levels and types of species. The radionuclides are also grouped in several classes in terms of tissues in which a specific radionuclide accumulates preferentially. These simplifications allow for a limited number of standard input parameters. The ~~scheme of transfer of~~ transfer scheme of radionuclides through the marine food web is shown in Fig. ~~1.~~ 1 where transfer of radionuclides through the food web is shown by arrows whereas the direct transfer from water is depicted by the shadowed rectangle surrounding 11 biota compartments (i=1,...,11). The different food-chains exist in both pelagic and benthic zones. Pelagic organisms are divided into primary producer~~(phytoplankton~~ , phytoplankton (i=1), and consumers which consist of zooplankton (i=2)~~and consumers: zooplankton~~ , forage (non-piscivorous) fish (i=3), and piscivorous fish ~~. This food web has been implemented in the compartmental POSEIDON-R model (Lepicard et al., 2004; Maderich et al., 2014 a,b).~~

(i=4). The benthic food web includes three primary pathways for radionuclides: (~~i) through water contamination in a manner similar to the BURN model, (ii)~~I) transfer from water to macroalgae (i=5), then to grazing invertebrates (i=6,..,8); (II) through the vertical detritus flux and zooplankton faeces (Fowler et al., 1987) to detritus-feeding invertebrates (i=8), and (~~iii~~III) through contaminated bottom sediments ~~.~~ to deposit feeding invertebrates (i=6). Concentrations of radionuclides in water and in the upper layer of bottom sediment are calculated using the box model POSEIDON-R described below. The output from this model is shown by external boxes in Fig. 1. The radionuclides adsorbed on the organic matter in the sediments ~~is~~ are bioavailable for benthic organisms but the mineral component of sediments is not (Ueda et al., 1977; Ueda et al., 1978) although Koyanagi et al. (1978) found ~~a~~ relatively rapid and more intensive transfer of several sediment adsorbed radionuclides (^{54}Mn, ^{60}Co, ^{65}Zn) to particular organs of the demersal fishes in contrast to flesh. It is assumed that ~~radioactivity is transferred from organic bottom deposits to deposit feeding invertebrates, then~~ (i) radioactivity concentrations in organic and mineral fractions of bottom deposit are in mutual equilibrium, (ii) radioactivity concentrations in microbial biota and non-living organic matter also are in equilibrium and (iii) only organic matter in the bottom deposit is bioavailable. The benthic invertebrate group (surrounded in Fig. 1 by dashed rectangle) includes molluscs (e.g. filter-feeders) (i=7), crustaceans (e.g. detritus-feeders) (i=8) and subsurface and surface deposit feeders (e.g. annelid)(i=6) .

In the model radioactivity is transferred from invertebrates to benthic invertebrate feeding demersal fishes (i=9), and on to omnivorous bottom predators (i=10) (Fig. 1). The ~~components of this system also include crustaceans (e.g detritus-feeders),molluscs (filter-feeders), and coastal predators~~marine food web also includes 'coastal predators' (i=11) feeding in the whole water column in shallow waters.

In the extended model utilised in this study, the concentration of radioactivity in phytoplankton C_1 is calculated using the Biological Concentration Factor (BCF) approach due to the rapid uptake from water and the short retention time of radioactivity, namely,

$$C_1 = CF_{ph} C_w, \tag{1}$$

where C_w is concentration of radioactivity in water and CF_{ph} the BCF for phytoplankton. For the macroalgae, a dynamic model is used to describe radionuclide concentrations due to the longer retention times

$$\frac{dC_5}{dt} = (CF_{ma} C_w - C_5) \ln 2 T_{0.5,5}^{-1}, \tag{2}$$

where C_5 is the concentration of radioactivity in the ~~macro-algae~~macroalgae, CF_{ma} is corresponding BCF, $T_{0.5,5}$ is the biological half-life of the radionuclide in the plant and t is the time. The concentration of a given radionuclide in the zooplankton (i=2), invertebrates (i=6,7,8) and fish (i=3,4,9,10,11; see Table 1 for a description of the different fish groups in the model) is described by the following differential equation:

$$\frac{dC_i}{dt} = a_i K_{f,i} C_{f,i} + b_i K_{w,i} C_w - \ln 2 T_{0.5,i}^{-1} C_i, \tag{3}$$

where C_i and $C_{f,i}$ are the concentrations of radioactivity in the marine organism and food, respectively, a_i is the ~~food extraction coefficient~~assimilation efficiency, b_i is the water extraction coefficient, $K_{f,i}$ is the food uptake rate, $K_{w,i}$ is the water uptake rate and $T_{0.5,i}$ is the biological half-life of the radionuclide in the organism.

The activity concentration in the food of a predator $C_{f,i}$ is expressed by the following equation, summing up for the total of n prey types,

$$C_{f,i} = \sum_{j=0}^{n} C_{prey,j} P_{i,j} \frac{drw_{pred,i}}{drw_{prey,j}}, \tag{4}$$

where $C_{prey,j}$ is the activity concentration in prey of type j, $P_{i,j}$ is preference for prey of type j, $drw_{pred,i}$ is the dry weight fraction of predator of type of i, and $drw_{prey,j}$ is the dry weight fraction of prey of type j. The index ~~“~~‘0~~”~~’ corresponds to the bottom deposit. ~~It is assumed that (i) radioactivity concentration in organic and mineral fractions of bottom deposit are in mutual equilibrium and (ii) that only organic matter in the bottom deposit is bioavailable. Therefore, the concentration of~~ The concentration of assimilated radioactivity from the organic fraction of bottom sediment ~~can be~~ is related with the radioactivity concentration of the upper layer of bulk sediment as $C_{prey,0} = \phi_{org} \cdot C_s$. Here ϕ_{org} is an empirical parameter $\phi_{org} = (1-p) f_{org} C_{org} C_s^{-1}$ where p is porosity, f_{org} is the organic matter fraction, $C_{org}C_s^{-1}$ is the ratio of concentration C_{org} (Bq kg^{-1}-dry) in the organic matter to in the bulk sediment concentration C_s (Bq kg^{-1}-dry). The value of ϕ_{org} is in the range 0.1-0.01 (Ono et al. 2015).

Values of the model parameters are given in Table 1. The parameters for pelagic and benthic food webs were compiled from published data (Baptist and Price, 1962; Cammen, 1980; De Vries and De Vries, 1988; Coughtrey and Thorne, 1983; Tateda, 1994,1997; Vives i Batlle et al., 2007; Tateda et al., 2013; Iwata et al., 2013; Sohtome et al., 2014). The biological half-life data for fish flesh (Baptist and Price, 1962; Coughtrey and Thorne, 1983; Tateda, 1994,1997; Zhang et al., 2001~~; Matsumoto et al., 2015) show variations~~) show variability in a large range (~~35-270~~ 35-180 days) due to the differences between species and due to the differences in the experiment methodology~~(Matsumoto et al. , 2015).~~ . In this generic model, values of $T_{0.5,i}$ were divided into two groups: $T_{0.5,i} = 75$ d for non-piscivorous fishes and those demersal fishes feeding on invertebrates ($i = 3, 9$) and $T_{0.5} = 150$ d for predatory fishes ($i = 4, 10, 11$). This is based on the assumptions that (a) larger fishes have longer $T_{0.5,i}$ due to the slower metabolic rate~~(Matsumoto et al., 2015),~~ , and (b) predatory fishes are generally larger than prey fishes. The results of sensitivity study for $T_{0.5,i}$ are given in next section to assess robustness of this simplification. Additional restriction on the values of the model parameters is the condition that at equilibrium state BCF of the components of the food chain should be relevant to the values from IAEA(2004). The ~~values~~ biological half-life for bone was estimated using data for ^{90}Sr, which is mainly accumulated in bone. This value was calculated for non-piscivorous and piscivorous fish using equation (3) in equilibrium approximation to satisfy BCF values from IAEA (2004). The values of prey preference are given in Table 2. They are compiled from data on food habits of organisms (Fujita et al., 1995; Kasamatsu and Ishikawa, 1997; Iwata et al., 2013; Sohtome et al., 2014).

It is well known that the uptake of caesium decreases with increasing salinity due to the increase of competing ions from potassium with higher concentration. This was taken into account when introducing the salinity-dependent correction factor F_K for phytoplankton and macro-algae since caesium enters the foodweb mainly via the lowest trophic level whereas the uptake from water contributes in a relatively minor way (Heling and Bezhenar, 2009). Based on laboratory experiments with marine plants for caesium, the correction factor was verified against field measurements in the Dnieper-Boog Estuary (Heling and Bezhenar, 2011). It is expressed as

$$F_K = \frac{0.05}{\exp(0.73 \ln(K^+/39.1) - 1.22 \cdot 10^3 \Theta^{-1})}, \tag{5}$$

where K^{+} is the potassium concentration (mg L^{-1}) and Θ is temperature (^{o}K). For water with a K^{+} concentration of above 1.5 mg L^{-1}, the potassium concentration could be linked to the salinity using the following linear relationship (Heling and Bezhenar, 2009):

$$K^{+} = 11.6S - 4.28, \tag{6}$$

where S is the salinity in g L^{-1}. Then the BCF for phytoplankton and macro-algae can be expressed by:

$$CF_{ph} = F_K CF^{*}_{ph}, \qquad CF_{ma} = F_K CF^{*}_{ma}, \tag{7}$$

where CF^{*}_{ph}=20Lkg^{-1} and CF^{*}_{ma}=50 Lkg^{-1} are standard BCFs for marine environments (IAEA, 2004).

According to a review of radiological data (Coughtrey and Thorne, 1983; Yankovich et al., 2010), every radionuclide in fish accumulates mostly in a specific (target) tissue. According to data from Yankovich et al. (2010) amounts of radiocaesium in flesh, bone and organs are 90%, 9% and 1%, respectively allowing do not consider caesium turnover in bone and organs. It is assumed that the target tissue (bone, flesh, organs and stomach) controls the overall elimination rate of the nuclide ($T_{0.5,i}$) in the organism. The radioactivity in the food for the predator is therefore the activity concentration in the target tissue diluted by the remaining body mass of the prey fish, calculated by multiplying the predicted level in the target tissue by its weight fraction. For radiocaesium the target tissue is flesh. To calculate the concentration in the edible part of fish from the calculated levels in the target tissues, a target tissue modifier (TTM) is introduced. This is based on tissue distribution information (Coughtrey and Thorne, 1983; Yankovich et al., 2010). Values of the described parameters for the fish in a dynamic food chain model are given in Table 3.

The dynamic food-chain model is part of the POSEIDON-R (Lepicard et al., 2004; Maderich et al., 2014a,b) compartment model where the marine environment is modelled as a system of compartments representing the water column, bottom sediment and biota. The compartments describing the water column containing suspended matter are subdivided into a number of vertical layers as shown in Fig. S1. The model assumes partition of the radionuclides between the dissolved and particulate fractions in the water column, described by a distribution coefficient. The radionuclide concentration for each compartment is governed by a set of differential equations including the temporal variations of concentration, the exchange with adjacent compartments and with the suspended and bottom sediments, radioactive sources and decay. The exchange between the water column boxes is described by fluxes of radionuclides due to advection, sediment settling and turbulent diffusion processes. The activity loss on suspended sediments occurs through settling in underlying compartments and, finally, to the bottom. A three-layer model describes the transfer of radionuclides in the bottom sediment. The transfer of radioactivity from the upper sediment layer to the water column is described by diffusion in the interstitial water and by bioturbation. Radioactivity in the upper sediment layer migrates downwards by diffusion and by burial at a rate assumed to be the same at which particles settle from the overlying water. The upwards transfer of radioactivity from the middle sediment layer to the top sediment layer occurs only by diffusion. Burial causes an effective loss of radioactivity from the middle to the deep sediment layer, from which no upward migration occurs. The model equations are given in Supplement. The model for the pelagic food web component was implemented for the upper ~~layer of the compartment~~water layer of all compartments, whereas the benthic component was included in the shallow one-layer compartments ~~.~~ adjacent to the shore

The POSEIDON-R model can handle different types of radioactive releases: atmospheric fallout, runoff from land deposited radionuclide by river systems, point sources associated with routine releases from nuclear facilities located either directly on the coast or inland at river systems, and point sources associated with accidental releases (Lepicard et al., 2004). For coastal discharges occurring in the large (‘regional’) boxes, ‘coastal’ release boxes are nested into the regional box system. Advection and diffusion of zooplankton are not taken into account due to the short biological half-life (five days), except in the coastal box, where diffusion was considered. It was assumed that crustaceans, molluscs, and fish are not transported by ocean flows. When calculating the radionuclide concentration in fish in small coastal boxes, random fish migration is taken into account as in Maderich et al. (2014a,b). For this purpose, the right hand side of equation (3) for radionuclide concentration in fish, both in the inner ($C_{in,i}$) and outer ($C_{out,i}$) compartments, is extended by the term $-(C_{in,i} - C_{out,i})/T_{migr,i}$ for the coastal compartment and by the term $(C_{in,i} - C_{out,i})/(\delta T_{migr,i})$ for the outer compartment. Here $T_{migr,i}$ is the characteristic time of fish migration from a coastal compartment, depending on compartment scale and fish species, and δ is the ratio between the volumes of the outer and the coastal compartments.

3 Application to the Fukushima Dai-ichi accident

3.1 Model setup

The model was customized for the ~~northwestern~~ Northwestern Pacific Ocean, the East China and Yellow Seas and the East/Japan Sea. A total of 176 boxes cover this entire region (Fig. ~~S1).~~ S2). In the deep-sea regions a three-layer box system was built to describe the vertical structure of the radioactivity transport in the upper layer (0-200 m), intermediate layer (200-1000 m) and lower layer (>1000m). The compartments around the FDNPP are shown in Fig. 2. The ~~coastal box (~~‘coastal’ box 15x30 km ~~), which is placed around FDNPP,is located inside box 90. It was chosen to cover~~ is nested into large ‘regional’ box 90 to provide more detailed description in the area around the FDNPP. It covers observation data within a circular-shaped surface area ~~with~~ of a radius 15 km ~~centred around~~ with a center at the FDNPP. ~~The~~ This box has one vertical layer for the water column and three bottom sediment layers. The depth of coastal box is less than that in the one layer outer box 90. The water exchange fluxes with the outer box are equal in both directions. The averaged advective and diffusive water fluxes between ~~compartments were based on~~ regional compartments were calculated for a ten-year ~~average over the period~~ period (2000-2009) using the Regional Ocean Modeling System (ROMS). ~~Details of the~~ The parameters of the coastal box are given in Table S1. Details of customization are given by Maderich et al. (2014a,b). ~~In the simulations the~~ The values for parameters ϕ_{org}=0.01 and $T_{migr,i}$=0.7 y for i=3,4,9,10,11 were used.

The simulation of dispersion and fate of ^{137}Cs was carried for the period 1945-2010 to provide background concentrations of radiocaesium for the radiological assessment of the FDNPP accident for the period 2011-2020 and to verify the model with available data. The main source of ^{137}Cs in the northwestern Pacific in the period 1945-2010 was from fallout due to atmospheric nuclear weapon tests. The fallout includes a global component, caused by the transport of radioactivity due to the general atmospheric circulation and subsequent deposition on the surface of the sea and a regional component, caused by fallout from weapon tests carried out in the Marshall Islands, resulting in the contamination of the surface layer of the ocean. The annual deposition of ^{137}Cs on the ocean for the period 1945-2005, compiled from Nakano (2006) and Hirose et al. (2008), is shown in Fig. ~~S2a~~S3a. The concentrations of ^{137}Cs at the eastern and southern boundaries (Fig. ~~S2b~~S3b) of the computational domain (Fig. ~~S1~~S2) were estimated by using both observations from the MARIS (Marine Information System) database (MARIS, 2015), and observations from Kang et al. (1997) and Nakano and Povinec (2003). These values represent both the effect from global deposition of ^{137}Cs on the northeastern Pacific and the regional effect of weapon tests carried out in the Marshall Islands. For the prediction of the concentration of ^{137}Cs for the period 2005-2020, five-years-averaged deposition and the boundary concentrations during the period of 2000-2004 were extrapolated and corrected for radioactive decay. The simulation for the period 1945-2010 was continued for the period of 2011-2020 with a source term estimated from the Fukushima accident. It was assumed that the release of activity directly to the ocean took place over the period 1-10 April 2011. Amounts of 5 PBq of ^{134}Cs, and 4 PBq of ^{137}Cs were transferred directly into the coastal box. These quantities are in accordance with widely accepted source terms for the Fukushima accident simulations (see Povinec et al., 2013). The atmospheric deposition data was obtained from simulations with the MATCH model (Robertson et al., 1999) where the dispersion of ^{137}Cs for the period 12 March-5 April was computed (Maderich et al., 2014a). The ECMWF meteorological data with a source term reported by Stohl et al. (2012) was used in the simulation. The amount of deposited ^{137}Cs in the computational domain was 8.5 PBq. The deposition of ^{134}Cs was estimated at 10.2 PBq using an activity ratio ^{134}Cs/^{137}Cs=1.2 (NISA, 2011). The atmospheric deposition was distributed between compartments as shown in Fig.2. The continuous leakage into the coastal box from the middle of 2011 with a release rate of 3.6 TBq y^{-1} (Kanda, 2013) was taken into account.

3.2 Results

The results from the modelling of the ^{137}Cs concentration in the water and in the upper layer of sediments of the coastal box are shown in Fig. 3. Model results for the water demonstrate good agreement both with yearly averaged observations by MEXT (the Japanese Ministry of Education, Culture, Sports, Science and Technology) for the period 1950-2010 (MEXT, 2010) and with observation by TEPCO (Tokyo Electric Power Company) for the period of ~~2011-2014~~ 2011-2016 (TEPCO, ~~2014~~2016). Comparison of Fig. 3a with Fig. 9a from Maderich et al. (2014a) confirms that the model correctly simulated the almost constant concentration of ^{137}Cs in the water in the FDNPP vicinity due to the continued leak of radioactivity from FDNPP (Kanda, 2013). The geometric mean of the simulated-to-observed ~~values is 1.0~~ ratios is 1.03 for the period ~~1984-2014~~ 1984-2016 when data were available, with a geometric standard deviation of ~~1.9~~ 1.89 for a total number of observations N=~~48.~~ 51.

The model also predicts well the concentration of ^{137}Cs in the bottom sediment before the accident and the sudden increase in concentration by more than ~~five~~ three orders magnitude as a result of the accident. However, as seen in Fig. 3b, the observed concentration from 2013 decreases faster than the model prediction without including the correction of vertical transfer. The details of this correction are described below. The estimated decrease constant of the fitted exponential function of the observed sediment concentration for ~~2012-2014~~ 2012-2015 is λ_s = 0.44 y^{-1}. The observed concentrations of ^{137}Cs in the bottom sediment of the coastal areas (B,C,D) with a depths less than 50 m in the Fukushima Prefecture (Sohtome et al., 2014) show a similar decrease. The decrease constant for area B located north of FDNPP is 0.44 y^{-1} whereas for the smaller areas C and D located south of the FDNPP it is 0.63 y^{-1} and 0.7 y^{-1}, respectively. For the deeper offshore area F adjacent to the areas C and D the value of the decrease constant is much less (0.24 y^{-1}). Several possible mechanisms could be responsible for the observed time-spatial redistribution of radioactivity in the surface layer of sediment. According to Ambe et al. (2014) the vertical transfer of ^{137}Cs by resuspension and redeposition by the ocean currents and waves, desorption to the pore water and bioturbation can result in a decrease of ^{137}Cs concentration in the upper layer of sediments. Resuspension and lateral transport of the fine-grained sediments also can redistribute radiocaesium in the coastal sediment (Otosaka and Kobayashi, 2013). Only several of these mechanisms are included in the POSEIDON-R model. The simplified representation of the exchange processes in the upper layer of the sediment and the lack of re-suspension cannot account for the mechanisms described above. ~~Instead~~Therefore, to take into account the vertical transfer of ^{137}Cs we added the exchange terms $(C_{s,1} - C_{s,2})\lambda_s$ and $-(C_{s,1} - C_{s,2})\lambda_s$ to the right hand side of the equations (S3) and (S4) for the concentration of radioactivity in upper ($C_{s,1}$) and medium ($C_{s,2}$) layers of sediment in the coastal box, respectively. ~~It can be seen that corrected prediction~~ Here λ_s is an empirical parameter. The value of λ_s =0.4 y^{-1} was obtained to fit observation data for $C_{s,1}$. As seen in Fig. 3b ~~is in good agreement with the observations for 2012-2014~~the corrected by additional exchange term concentration of ^{137}Cs is described well in period 2008-2015. The geometric mean of the simulated-to-observed ~~values is 0.93~~ratios is 0.97, with a geometric standard deviation of 1.26 for a total number of observations N=~~42~~ 46 for the period ~~1984-2014~~1984-2015.

The simulated ^{137}Cs concentrations in deposit feeding invertebrates, demersal fishes, bottom predators and coastal predators in the coastal box are shown in Fig. 4 along with observed concentrations by the Japan Fisheries Research Agency (JFRA, 2015). The symbols in Fig. 4 are observation data for sea urchin (*Strongylocentrotus nudus*) (a), flounders (*Microstomus achne, Kareius bicoloratus, Pleuronectes yokohamae*) (b) and Japanese rockfish (*Sebastes cheni*) (c). The open and filled symbols in Fig. 4d are data for seabass (*Lateolabrax japonicas*) and fat greenling (*Hexagrammos otakii*), respectively. As seen in Fig. 4a, just after the accident the simulated ^{137}Cs concentration in the deposit feeding invertebrates and the observed concentration in the sea urchin increase due to the high concentration of ^{137}Cs in the water (Fig. 4a). After that the concentration trend becomes equal to the bottom sediment trend. ~~The~~ This is consistent with model diet that includes macroalgae and deposit organic matter grossly representing diet of *S. nudus* (Lawrence, 2007). The macroalgae contribution in food contamination first prevails, then after 2012 the bottom contamination dominates. The decrease constant of the fitted exponential function of simulated concentration (depuration constant) is 0.45 y^{-1}, which is close to the decrease constant for the sediment observations (0.44 y^{-1}). It agrees with the conclusion by Sohtome et al. (2014) that both observed decrease rates of concentration in sediment and in deposit-feeding benthic invertebrates are almost identical. The predicted transfer coefficient from bulk sediment to deposit feeding benthic invertebrates for the period of 2012-2020 is approximately 0.07. ~~Results of observations and~~ The field studies of several species of polychaeta (deposit or filter feeders: *Flabelligeridae*, *Terebellidae* and *Opheliidae*; herbivore or carnivore feeders: *Glyceridae*, *Eunicidae*, and *Polynoidae*) off the coast of Fukushima and rearing experiment for ~~benthic polychaete~~ *Perinereis aibuhitensis* demonstrated that ^{137}Cs concentration in all specimens was much lower than that in the sediment (Shigenobu et al., 2015)~~showed that this coefficient~~. Results of rearing experiment using contaminated sediments from near the FDNPP showed that transfer coefficient (concentration ratio) between *P. aibuhitensis* (Bq kg^{-1}-wet) and contaminated sediment (Bq kg^{-1}-wet) was less than 0.1. The geometric mean of the simulated-to-observed ~~values is 0.95~~ratios is 0.98, with a geometric standard deviation of ~~1.43~~ 1.41 for a total number of observations N=~~20.~~ 21.

The results of simulation of the ^{137}Cs concentration in the demersal fishes (Fig. 4b) agree well with observations documented for several species of flounders. The geometric mean of the simulated-to-observed ~~values is 1.18~~ratios is 1.16, with a geometric standard deviation of ~~1.30~~ 1.31 for a total number of observations N=~~47. The simulated depuration rate~~ 49. The simulated value of the depuration constant is 0.46 y^{-1} whereas ~~the experimental value for 2012-2014~~ value estimated from the field data for 2012-2015 in Fig.4b is 0.48 y^{-1}. The gradual decrease of activity in demersal fish caused by the transfer of activity from organic matter deposited in the bottom sediment is similar to observations by Wada et al. (2013). Notice that the predicted transfer coefficient from bulk sediment to demersal fish for the period of 2012-2020 is approximately 0.13. This value is larger than that for deposit feeding invertebrates~~due to the biomagnification effect~~. The observed in this area BCF for demersal fish (flounders) in 2013-2015 is 0.9 m^{3}kg^{-1}, whereas the standard value of BCF for fish is 0.1 m^{3}kg^{-1}(IAEA, 2004) that confirms the importance of transfer of radiocaesium to demersal fish from the sediments. Comparison of simulations with observations for a bottom predator (Japanese rockfish) in Fig. 4c shows a good agreement. The geometric mean of the simulated-to-observed ~~values is 0.8~~ratios is 0.84, with a geometric standard deviation of ~~1.75~~ 1.73 for a total number of observations N=~~46.~~ 48. The comparison of simulated and observed concentrations of ^{137}Cs in coastal predators is given in Fig. 4d. The open and filled symbols are data for seabass and fat greenling, respectively. The geometric mean of the simulated-to-observed ~~values~~ ratios for coastal predators is ~~1.19~~1.16, with a geometric standard deviation of ~~1.85~~ 1.89 for a total number of observations N=~~66~~ 69 for the period of ~~1984-2014~~1984-2015. As seen in Fig. 4d, the simulated concentration of ^{137}Cs in coastal predators feeding on both pelagic and benthic organisms is similar to the simulated concentration in pelagic piscivorous fish in the period of 2011-2013, whereas after 2013 the concentration in coastal predators decreases more slowly than in piscivorous fish due to the omnivorous predation diet of coastal predators that includes benthic organisms.

The model output can be sensitive to the model parameters which are known ~~with~~ to have a high uncertainty. Therefore, a sensitivity study was carried out for the major benthic food web parameters including the water uptake rate $K_{w,i}$, the food uptake rate $K_{f,i}$, the biological half-life of ^{137}Cs in the organism ~~$T_{0.5}$~~ $T_{0.5,i}$ and for the concentration ratio of assimilated radioactivity from the organic fraction in bottom sediment to the radioactivity in bulk bottom sediment ϕ_{org}. The effects from variations in these parameters were estimated for the following model output: maximum ^{137}Cs concentration in the ~~i~~i=2,..11 types of organisms in the coastal box after the FDNNP accident. The range for $K_{w,i}$, $K_{f,i}$, $T_{0.5,i}$, and ϕ_{org} is defined following Keum et al. (2015) as follows: minimum value is half the reference value and maximum value is twice the reference value. The reference values for $K_{w,i}$, $K_{f,i}$, and $T_{0.5,i}$ are given in Tables 1 and 2 whereas $\phi_{org} = 0.01$. The model output sensitivity was estimated using sensitivity index (SI). It was calculated following Hamby (1994) as

$$SI = \frac{D_{max} - D_{min}}{D_{max}}, \tag{8}$$

where D_{max} and D_{min} are output values for maximal and minimal values in the parameter range, respectively.

Figure ~~S3a~~ S4a shows that all organisms (except the primary producers) are most sensitive to the variation of $K_{f,i}$. Effect of the food uptake rate for zooplankton $K_{f,2}$ slightly decreases up the pelagic food web ($i = 2,3$), whereas it is much less for the benthic food web ($i = 7-11$) because of its diverse diet. The biological half-life for zooplankton $T_{0.5,i}$ was also one of most sensitive parameters both for pelagic and benthic food webs (Fig. ~~3b~~S4b). The maximum ^{137}Cs ~~concentrations for zooplankton , non-piscivorous and piscivorous fishes, algae, deposit-feeding invertebrates, molluscs,~~

~~crustaceans, demersal fish, bottom predators and coastal predators using~~ concentration for zooplankton using the maximal value of $T_{0.5,i}$ ~~were~~ was increased by a factor 2.7 ~~, 2.4, 2, 1.3, 1, 1.3, 1.9,1,1.3, and 1.7, respectively,~~ compared with a case when ~~a minimum values~~ the minimum value of $T_{0.5,i}$ was used. This factor for pelagic fish and coastal predator was in the range 2.4-1.7 whereas for the rest organisms it was smaller. The biological half-life $T_{0.5,6}$ of deposit feeding invertebrates essentially influences ^{137}Cs concentration in demersal fish ($i = 9$). Figure ~~S3c~~ S4c shows that the effect of variations in the water uptake rate of zooplankton $K_{w,2}$ decreased for organisms of higher trophic levels, showing good agreement with results by Keum et al. (2015). The concentrations of ^{137}Cs in ~~algae~~ macroalgae and deposit-feeding invertebrates are found to be three times more sensitive to the variations in water uptake than in the rest of organisms. The benthic organisms were less sensitive to the parameter ϕ_{org} (Fig. ~~3d~~S4d).

~~To estimate the contribution of benthic organisms in the individual ingestion dose rate due to the consumption of contaminated marine products, a hypothetical reference group is considered. It is assumed that this reference group is located in the Fukushima region and consumes only marine products from the coastal compartment near the Fukushima NPP. According to data given by Povinec et al. (2013), the annual consumption of marine products in Japan is 23.4 kg of fish, 2 kg of crustaceans, 1.3 kg of molluscs, and 3.7 kg of macro-algae. We compare two cases assuming that the consumed fish are pelagial or benthic species. In the first case consumption of piscivorous and non piscivorous species are equal. In the second case consumption of each of the three species of fish (demersal fish, bottom predator and coastal predator) is 1/3. In both cases we consider the period of 2014-2020 because the simulated and observed radiocaesium concentration in fish for 2011-2013 exceeds the Japanese regulatory level of seafood safety (100 Bq kg^{-1}) for Fukushima offshore waters (Fig.4). In the first case the fish consumption is equally divided between piscivorous and non piscivorous species. The dose contributions of the two caesium-isotopes ^{134}Cs and ^{137}Cs for the period of 2014-2020 are 1.4 μSv and 6.3 μSv, respectively. In the second case the consumption of each of the three species (demersal fish, bottom predator and coastal predator) is 1/3 from 23.4 kg. The corresponding dose contributions are 29μSv and 56 μSv,respectively. The total dose for consumed marine products including pelagic fish (7.7 μSv) is one order smaller than for marine products including benthic fish (85 μSv) but both of them are much less than the maximum annual effective dose commitment for the public, equal to 1000 μSv according to IAEA regulations (IAEA, 2014). Notice that we considered a conservative scenario with a continuous leak of radiocaesium from FDNPP in the period of 2012-2020, whereas ending of this leak results in a return of ^{137}Cs concentration to background value within one year (Maderich et al. , 2014a).~~

4 Modelling the effects from the Chernobyl accident on marine organisms in the Baltic Sea

4.1 Model setup

The Baltic Sea is an important case because of transfer of ^{137}Cs originating from the Chernobyl fall-out through its water-sediment-biota system. It was chosen to verify the ability of the model with generic parameters to describe transfer processes in a semi-enclosed sea with very different oceanography. The model was customized for the Baltic Sea, the North Sea, and the North Atlantic Ocean. The box system contains a total of 81 regional boxes~~followed by an additional 16 boxes for the inflow from rivers into the Baltic Sea~~. A plot of the box system is shown in Fig. 5. The volume and average depth for the 47 boxes describing the Baltic Sea are derived from bathymetric data. A water column with a depth of more than 60 m is divided into two layers (surface and bottom) to allow for activity stratification in the water column. These ~~multi-layered~~ boxes are marked blue in Fig.5. The exchange of water between the boxes in the Baltic Sea is based on a ten year average (1991-2000) of three-dimensional currents from reanalysis based on the Swedish Meteorological and Hydrological Institute (SMHI) model (SMHI, 2013). The exchange rates for the remainder of the boxes were adopted from the standard POSEIDON configuration (Lepicard et al., 2004). To consider the water balance of the Baltic Sea and the inflow of radioactivity from river runoff, an additional 16 boxes were defined to represent main rivers in the basin (Table S2). The inflow of river water for each box is based on information reported by Leppäranta and Myrberg (2009). The total inflow of water into the rivers accumulates to 484 km^{3}y^{-1}. A concentration of suspended sediments (different for each box) was calculated by a 3D hydrodynamic THREETOX model (Margvelashvily et al., 1997; Maderich et al., 2008). The bottom sediment classes for simulation were determined using data from Winterhalter et al, (1981). The simulation of transport and fate of ^{137}Cs in the Baltic Sea was carried out for the period 1945-2020. The main sources of ^{137}Cs as included in this model are: global deposition from weapon testing and from the Chernobyl accident (HELCOM,1995), release from the Sellafield and La Hague reprocessing plants (HELCOM, 2009), regional deposition from the Chernobyl accident in May 1986 (HELCOM, 1995), and river runoff. Details of these main sources are shown in Fig. ~~S4a~~ S5a (global deposition), and in Fig. ~~S4b~~ S5b (Sellafield and La Hague releases), as well as in Table ~~S1~~ S3 (Chernobyl accident). The river runoff from corresponding catchments was calculated using a generic model by Smith et al. (2004). The value for parameter ϕ_{org} is 0.02.

4.2 Results

The simulation results for the period of 1945-2020 are shown in Fig. 6-7 for box 45 where data for concentration in the water, in the sediment and in the biota are most detailed (MARIS, 2015; MORS, 2015). Time variations of ^{137}Cs concentration in the water and sediments in Fig. 6 show two maxima related with weapon testing and the Chernobyl accident and then with a decreasing tendency due to outflow to the North Sea and radioactive decay. The decrease constants of the fitted exponential function of the simulated concentration in the water (0.081 y^{-1}) and sediments (0.070 y^{-}1) are close unlike the Fukushima accident where the plume of contaminated water quickly dissolves in the open ocean. The simulation results are in good agreement with the measurements. The geometric mean ~~ratios between the predicted and observed values~~ of the simulated-to-observed ratios for concentration in the water and sediment are 0.89 and 0.86, respectively. The geometric standard deviation for concentration in the water is of 1.42 for a total number of observations N=378, whereas corresponding value for concentration in the sediment is of 2.17 for a total number of observations N=163 in the whole Baltic Sea.

Figure 7 shows a comparison between the calculated and observed ^{137}Cs concentration in marine organisms for box 45. The symbols in Fig. 7 are observation data for echinoderms (*Echinodermata*) (a), sprat (*Sprattus sprattus*) (b), European flounder (*Platichthys flesus*) (c) and Atlantic cod (*Gadus morhua*) (d). Comparison of the calculated concentrations of ^{137}Cs in the deposit-feeding invertebrates with the measurements (Fig. 7a) shows that the model correctly predicts the time-varying concentration in these organisms. The assessment of the model accuracy in this case is, however, hardly possible because of the small number of measurements. Calculated and observed concentrations of ^{137}Cs in pelagic non-piscivorous fish (sprat) demonstrate a good agreement with the measurements (Fig. 7b). The geometric mean ~~ratio~~ for the simulated-to-observed ~~values~~ ratios is 0.91 with a geometric standard deviation of 1.32 for a total number of observations N = 24 in the whole Baltic Sea. Using the standard model with a constant value of CF_{ph} (IAEA, 2004) for brackish waters leads to an essential underestimation of the concentration in fish: the geometric mean value is 0.68 with a geometric standard deviation of 1.33. Comparison of calculated and observed concentrations of ^{137}Cs in demersal fish (European flounder) is shown in Fig. 7c. It can be seen that the concentration of ^{137}Cs in demersal fish reveals a pattern with significantly more delay in time compared with that in the non-piscivorous fish (Fig. 7b) due to the difference in the food chain between these species (Fig. 1). The benthic food web depends on the ^{137}Cs concentration in the bottom sediment (Fig. 6b), which follows the ^{137}Cs concentration in water with some delay (Fig. 6a). Notice that European flounder ~~belongs to the polychaete and small crustacean feeding group~~ diet in the Baltic Sea includes oligochaetes, amphipods, chironomids and smaller sizes harpacticoids (Gibson et al., 2015). The geometric mean ~~ratio~~ for the simulated-to-observed ~~values~~ ratios is 0.92 with a geometric standard deviation of 1.67 for a total number of observations N=70 in the whole Baltic Sea. Calculated ~~and observed~~ ^{137}Cs concentration in the coastal predator (cod) also agree well with the measurements (Fig. 7d)~~: the geometric mean ratio for~~. The diet of Atlantic cod in shallow Western Baltic is diverse, including herring, sprat, *Gobiidae*, molluscs, various Polychaeta and crustaceans (Sparholt, 1994). Therefore for this basin the cod is considered as 'coastal predator' feeding by both pelagic and benthic preys. The geometric mean of the simulated-to-observed ~~values~~ ratios is 0.91 with a geometric standard deviation of 1.37 for a total number of observations N=95 in the whole Baltic Sea. The concentration of ^{137}Cs in the coastal predators is greater than in piscivorous fish due to the additional benthic food chain included in the web (Fig. 7d).

In contrast to the open Pacific Ocean coast where the FDNPP is located, concentrations in demersal fish, pelagic and coastal predators after the Chernobyl accident decrease with almost the same rate (about 0.075 y^{-1}). The variation in decrease rate is approximately 10% with a decrease rate 0.081 y^{-1} for water and 0.07 y^{-1} for sediment. The observed BCFs in this area for sprat, European flounder and Atlantic cod in 1990-2010 are 0.11, 0.14 and 0.15 m^3kg^{-1}, respectively. This is close to the standard value of BCF for fish 0.1 m^3kg^{-1} (IAEA, 2004) taking in account that waters in the Baltic Sea are brackish that affects the uptake rate of radiocaesium. These results essentially differ from the Fukushima case where BCF for demersal fish was an order greater confirming importance of transfer of radiocaesium from the sediments to demersal fish for that case. The weak water exchange with the North Sea of the semi-enclosed Baltic Sea results in a slow evolution of water-sediments-biota system in quasi-equilibrium state. Notice that the food-web model parameters, except for the correction for brackish waters, are the same as for the FDNPP case study demonstrating generic character of the model.

5 Conclusions

A generic dynamic food web model was extended to include the benthic food chain. In the model pelagic organisms are grouped into phytoplankton, zooplankton, non-piscivorous fish and piscivorous fish (Heling et al., 2002). The benthic organisms are grouped into deposit feeding invertebrate, demersal fish, and bottom predators. The components of this system also include crustaceans, molluscs and coastal predators. The model takes into account the salinity effect on the intake of radiocaesium. The foodweb model is embedded into the POSEIDON-R compartment model (Lepicard et al., 2004; Maderich et al., 2014a,b) where the marine environment is modelled as a system of compartments comprising the water column, bottom sediment and biota. The compartment model was applied to two regions (~~north western Pacific~~ North Western Pacific (NWP) and the Baltic Sea) ~~that were strongly~~ which were contaminated due to accidents on the Fukushima Dai-ichi and Chernobyl NPPs. Results of simulations were compared with available data for the period of 1945-2015. The modeling confirmed the presence of a continuous leakage of ^{137}Cs from Fukushima Dai-ichi NPP with a rate of 3.6 TBq y^{-1} ~~from 2012~~ resulting in an almost constant concentration of ^{137}Cs in an area of 15x30 km around the NPP. It was found that ^{137}Cs decreased in upper layer of sediments in the Fukushima case study faster than POSEIDON-R predicted using the standard for marine compartment model parameterization of exchange between water and sediment by diffusion mechanism. A simple parameterization calibrated on measurements was therefore used to correct this exchange. However, the further studies of exchange mechanisms are necessary. The decrease rate for the simulated concentration in the deposit ~~ingesting~~ feeding invertebrates (0.45 y^{-1}) is close to the decrease rate for the sediment concentration (0.44 y^{-1}) found experimentally. This is due to a diverse diet of invertebrates, and this is conformed with the conclusions by Sohtome et al. (2014) that the decrease of observed concentration in sediment and deposit-feeding benthic invertebrates is almost identical. The predicted-by-model low (0.07) transfer coefficient of radiocaesium from bulk sediment to deposit-feeding benthic invertebrates in the area around the FDNPP for the period of 2012-2020 is consistent with observations and rearing experiments (Shigenobu et al., 2015). The findings are comparable with observations by Wada et al. (2013) showing a gradual decrease of activity in the demersal fish (decrease constant is 0.46 y^{-1}) caused by transfer of activity from organic matter deposited in bottom sediment through the deposit feeding invertebrates. The estimated model transfer coefficient from bulk sediment to demersal fish for the period of 2012-2020 (0.13) is larger than that for deposit feeding invertebrates~~due to the biomagnification effect~~. This value can be used for mapping of demersal fish contamination from the bottom sediments. The concentration in coastal predators that feed on both pelagic and benthic organisms is similar to the concentration in pelagic piscivorous fish for the period of 2011-2013 when effects of water contamination were dominant. After 2013 the concentration in coastal predators decreases slower than in piscivorous fish due to the omnivorous predation diet of coastal predator that includes benthic organisms.

The ~~total individual dose of a reference group for consumed marine products including only pelagic fish contaminated by two caesium-isotopes ^{134}Cs and ^{137}Cs from a coastal compartment in the period 2014-2020 is 1.4 μSv and 6.3 μSv, respectively. The total dose contribution for marine products including pelagic and benthic fish of the two caesium-isotopes ^{134}Cs and ^{137}Cs for the same period are 29 μSv and 56 μSv, respectively. The total dose for consumed marine products from pelagic fish (7.7 μSv) is one order smaller than when including pelagic and benthic fishes (85 μSv) but both of them are much less than the maximum annual effective dose commitment for the public, equal to 1000 μSv according to IAEA regulations (IAEA, 2014).~~

~~The~~ results of the application of POSEIDON-R with an extended dynamic model to the Baltic Sea which is semi-enclosed and filled by brackish waters are in good agreement with available measurements~~in the Baltic Sea~~. Unlike the highly dynamical off coast processes caused by eddy dominated currents in the Pacific Ocean where the FDNPP is located, weak water exchange with the North Sea ~~results in~~ and regular circulation in the Baltic Sea results in a slow quasi-equilibrium evolution of ~~water-sediments-biota~~ water-sediment-biota system. The Chernobyl case confirms that the standard parameterization of water-sediment exchange used in POSEIDON-R describes well the exchange processes for the Baltic Sea whereas in the Fukushima study the observed value of ^{137}Cs decreases faster in the upper layer of the sediments than that the model predicts using the standard parameterization. In the Fukushima accident case the concentration of ^{137}Cs in piscivorous fish decreases faster than in the coastal predators whereas in the Chernobyl case these concentrations decrease simultaneously. In general, the obtained results demonstrate the importance of the benthic food chain in the long-term transfer of ^{137}Cs from contaminated bottom sediments to marine organisms and the potential of a generic model for use in different regions of the World Ocean.

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Table 1. Parameters of dynamical food chain model.

i	Organism	drw	$K_{f,i}$ d^{-1}	a_i	$K_{w,i}$ m^{3}kg^{-1}d^{-1}	b_i	$T_{0.5,i}$ d
1	Phytoplankton	0.1					
2	Zooplankton	0.1	1.0	0.2	1.5	0.001	5
3	Non-piscivorous fish	0.25	0.03	0.5	0.1	0.001	Table 3
4	Piscivorous fish	0.3	0.007	0.7	0.075	0.001	Table 3
5	Macroalgae	0.1			0.6	0.001	60
6	Deposit feeding invertebrate	0.1	0.02	0.3	0.1	0.001	15
7	Mollusc	0.1	0.06	0.5	0.15	0.001	50
8	Crustacean	0.1	0.015	0.5	0.1	0.001	100
9	Demersal fish	0.25	0.007	0.5	0.05	0.001	Table 3
10	Bottom predator	0.3	0.007	0.7	0.05	0.001	Table 3
11	Coastal predator	0.3	0.007	0.7	0.075	0.001	Table 3

Table 2. Preference of predator of type i for prey of type j.

Predator Prey	2	3	4	6	7	8	9	10	11
0				0.5			0.1		
1	1.0				0.6	0.1			
2		1.0			0.2	0.8			
3			1.0						0.2
5				0.5	0.2	0.1			
6							0.7	0.3	0.25
7							0.1	0.2	0.1
8							0.1	0.2	0.2
9								0.3	0.25

Table 3. Parameters for the fish in dynamical food chain model.

Target tissue	Bone	Flesh	Organs	Stomach
Weght fraction	0.12	0.80	0.05	0.03
Target tissue modifier	0.5	1.0	0.5	0.5
Biological half-life of non-piscivorous fish (d)	500	75	20	3
Biological half-life of piscivorous fish (d)	1000	150	40	5
Biological half-life of demersal fish (d)	500	75	20	3
Biological half-life of bottom predator fish (d)	1000	150	40	5
Biological half-life of coastal predator fish (d) fish (d)	1000	150	40	5

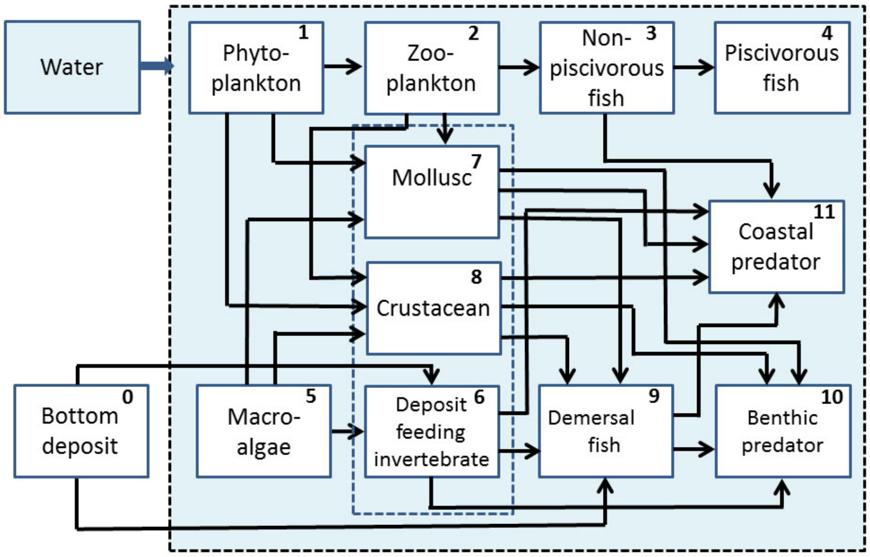

Figure 1. Scheme of radionuclide transfer to marine organisms. A transfer of radionuclides through food web is shown by arrows whereas direct transfer from water is depicted by shadowed rectangle surrounding biota compartments. The output from the compartment POSEIDON-R model is shown by external boxes.

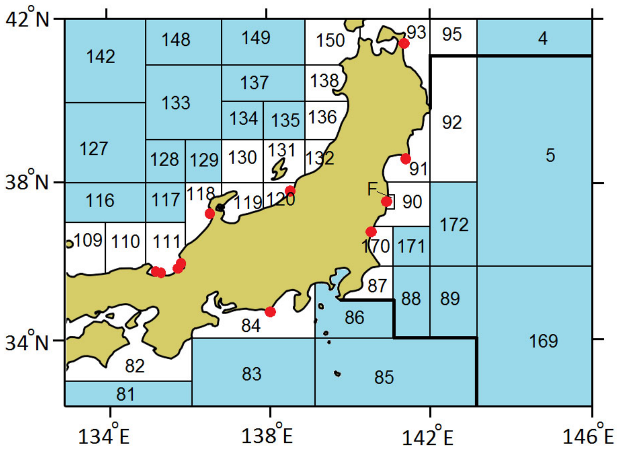

Figure 2. The box system for the area close to Fukushima NPP. The shaded boxes represent the ~~deep~~ deep-sea water boxes divided on three vertical layers. The NPPs are shown by filled circles. Coastal box around the FDNPP (marked by “F” is inside of box 90. Thick line limits the area of the Fukushima accident fallout.

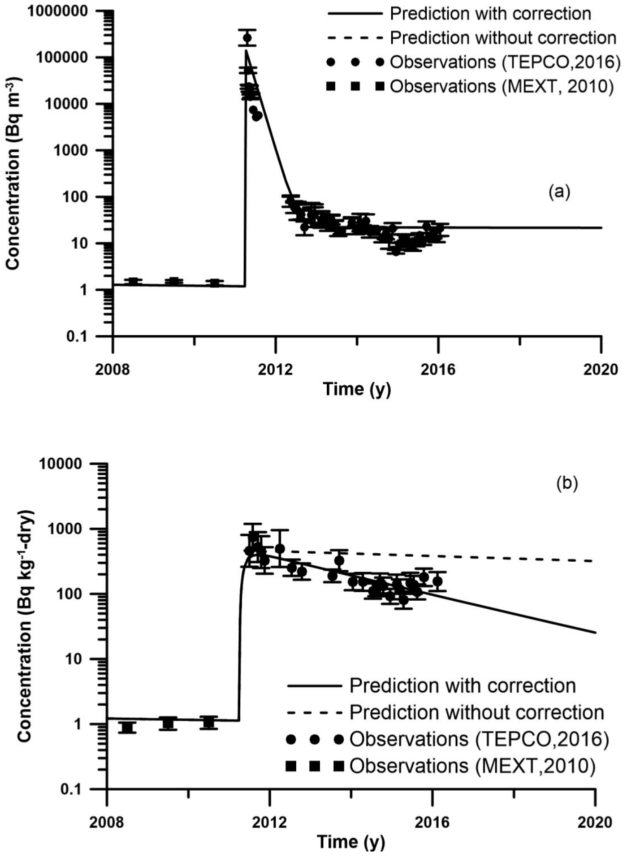

Figure 3. Comparison between calculated and observed ^{137}Cs concentration in seawater (a) and in bulk bottom sediment (b) in the coastal box around the Fukushima Dai-ichi NPP ~~.~~The dashed line in (b) shows results of simulations using standard POSEIDON-R model, whereas solid line presents simulation with correction term in equation (S3)

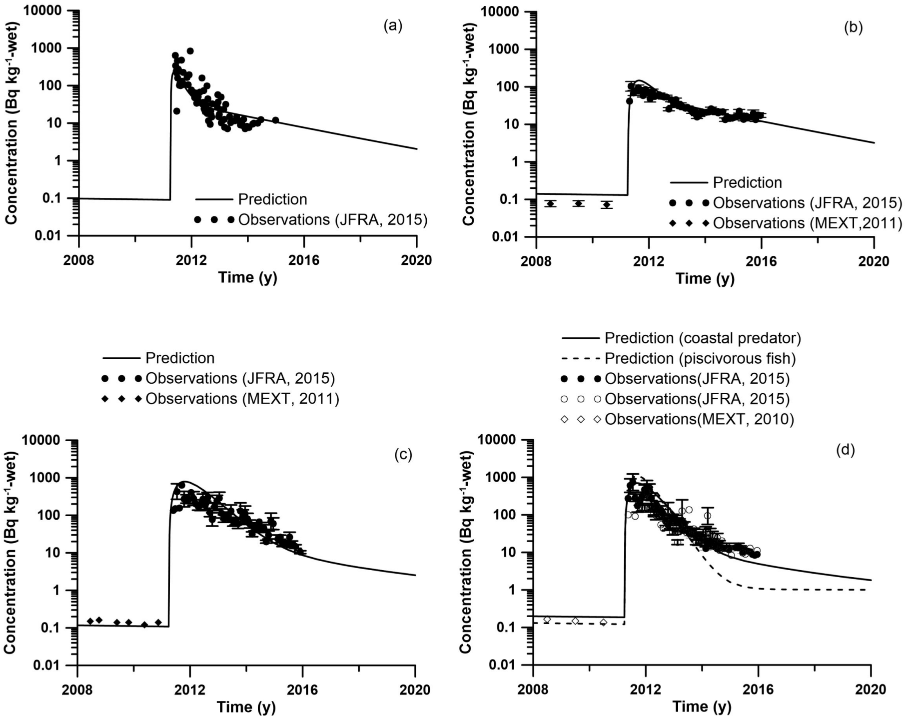

Figure 4. Comparison between calculated and observed ^{137}Cs concentration in deposit feeding invertebrate (a), demersal fish (b), bottom predator (c) and coastal predator (d) around the FDNPP.

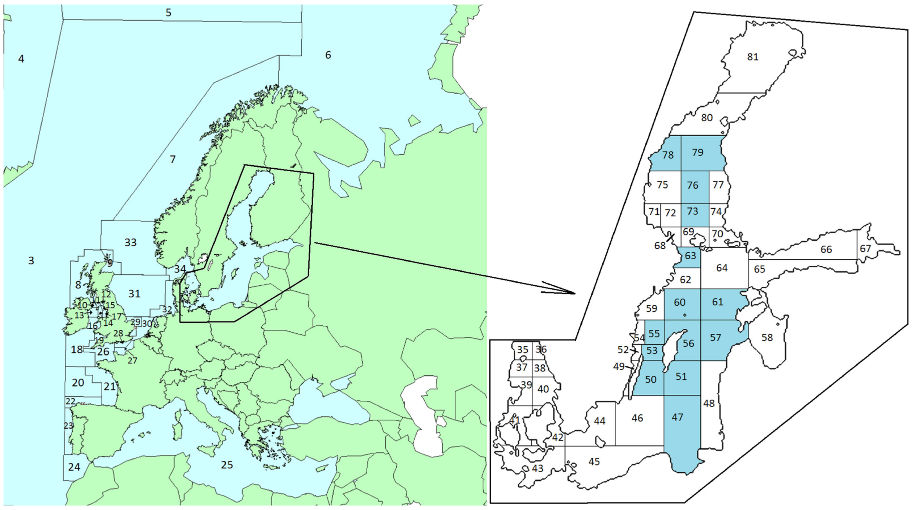

Figure 5. Compartment system of POSEIDON-R model for ~~north-eastern~~ the North-Eastern part of the Atlantic Ocean, the North Sea and the Baltic Sea. The shaded boxes represent boxes divided on two vertical layers.

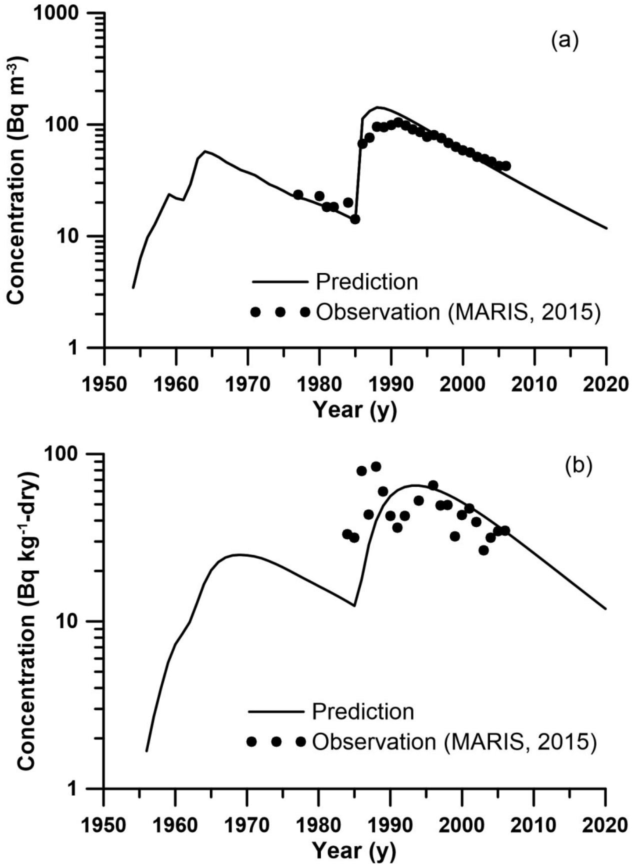

Figure 6. Comparison between calculated and observed ^{137}Cs concentrations in seawater (a) and in bulk bottom sediment (b) for box 45.

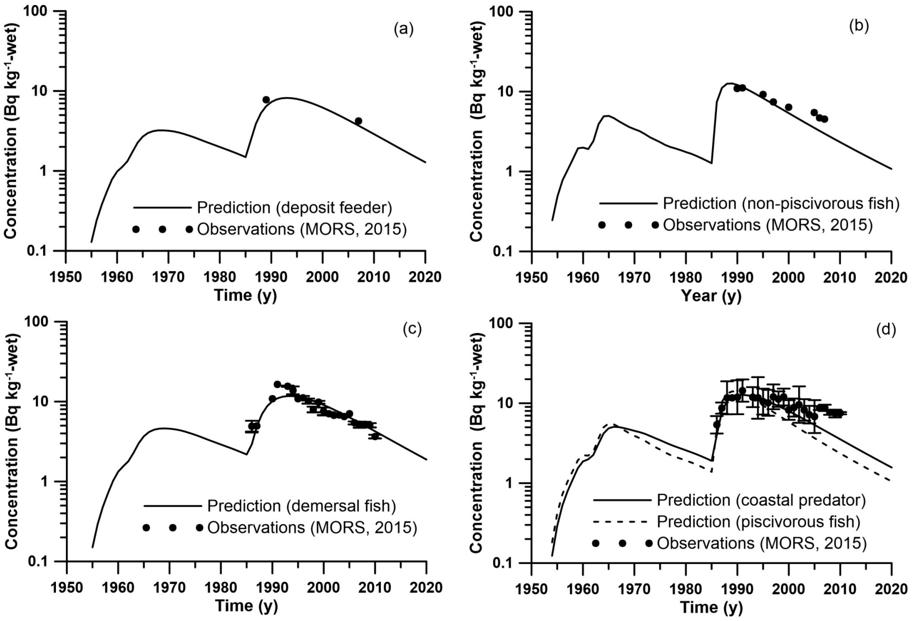

Figure 7. Comparison between calculated and observed ^{137}Cs concentrations in deposit-feeding invertebrate (a), non-piscivorous fish (b), demersal fish (c) and coastal predator (d) for box 45.

Supplementary materials to the paper “Transfer of radiocaesium from contaminated bottom sediments to marine organisms through benthic food chain”

Poseidon-R model

The mechanisms of radionuclide transfer in the POSEIDON-R model (Lepicard et al., 2004) are as follows. Activity entering the water column is transported by currents and turbulent diffusion and lost to bottom sediments through sorption on suspended particles which then settle out. The exchange of activity between the upper layer of the sediment and the water column is described as diffusion and bioturbation (modelled as a diffusion process). Activity in the upper sediment layer may diffuse downward but there is also an effective downward transfer via the continued sedimentation at the top of the sediment layers. Return of activity from the middle sediment to the top sediment occurs only through diffusion. The effective loss of activity from middle sediment to deep sediment arises from the continued deposition of sediment. A more detailed composition of the water column and its sediment layers, as well as its interaction with neighbouring volumes is shown in Fig. S1.

The POSEIDON-R equations are obtained by averaging the three dimensional transport equations for the dissolved radionuclide concentration C_w (Bq·m^{-3}) and the concentration in the three layers of the bottom sediment. It is assumed that the activity in the water column is partitioned between the water phase and the suspended sediment material, resulting in the following relation:

$$C_{ss} = K_d C_w . \tag{S1}$$

where C_{ss} (Bq·kg^{-1}) is the concentration of radioactivity sorbed by suspended sediment, K_d is the radionuclide distribution coefficient (m^3·kg^{-1}). The equations read as follows.

For the water column layers:

$$\frac{\partial C_{w,i}}{\partial t} = \sum_j \left[\frac{F_{ji}}{V_{w,i}} C_{w,j} - \frac{F_{ij}}{V_{w,j}} C_{w,i} \right] + \gamma_{0i} C_{w,(i,j,k-1)} - (\gamma_{1i} + \lambda) C_{w,i} + \frac{L_{t,i}}{h_i} \gamma_2 C_{s,1} + Q_{si}; \tag{S2}$$

for the upper sediment layer:

$$\frac{\partial C_{s,1}}{\partial t} = -(\gamma_2 + \gamma_3 + \lambda) C_{s,1} + \frac{h_i}{L_{t,i}} \gamma_{1,i} C_{w,i} + \frac{L_{m,i}}{L_{t,i}} \gamma_4 C_2 , ; \tag{S3}$$

for the middle sediment layer:

$$\frac{\partial C_{s,2}}{\partial t} = -(\gamma_4 + \gamma_5 + \lambda) C_{s,2} + \frac{L_{t,i}}{L_{m,i}} \gamma_3 C_{s,1} . \tag{S4}$$

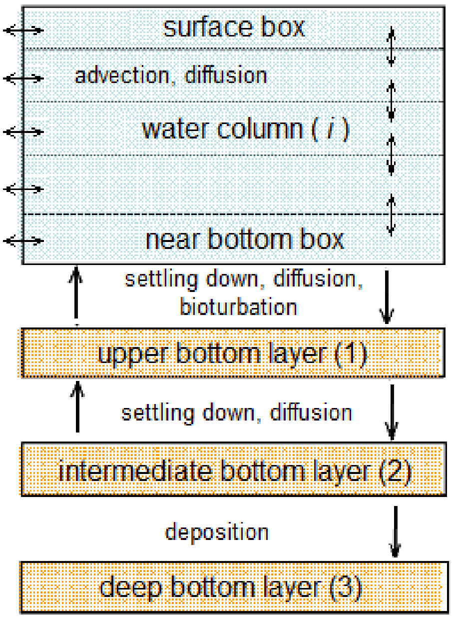

Fig.S1 Vertical structure and radionuclide transfer processes in the compartment of POSEIDON-R model. Arrows show exchange between boxes and layers.

Here subscript (0) denotes the water column, subscripts (1) and (2) denote the upper and middle sediment layer, respectively; $C_{w,i}$ is the box averaged concentration of radionuclide C_w in the water column layer i; $C_{s,1}$ is the averaged concentration of radionuclide in the upper sediment layer; $C_{s,2}$ is the averaged concentration in the middle sediment layer; λ (y^{-1}) is the radionuclide decay constant; F_{ij} is the water flux ($t \cdot y^{-1}$) from box i to box j; $V_{w,i}$ is the box volume (m^3); h_i is the depth of the water box layer (m); L_t, L_m are the depth (m) of top and middle bottom sediment layers respectively; Q_{si} is the point source of the activity in box i (Bq y^{-1}); $\gamma_0 \ldots \gamma_5$ are the coefficients, their values depend on the characteristics of the radionuclide and sediments.

For the surface water layer, the coefficients are as follows:

$$\gamma_{0i} = 0,$$
$$\gamma_{1i} = \frac{K_d SSW}{h_i(1+K_d SS)}, \tag{S5}$$
$$\gamma_2 = 0.$$

and for the layers in the water column below the surface water layer, the coefficients are defined as follows:

$$\gamma_{oi} = \frac{K_d SSW}{h_i(1+K_d SS)},$$
$$\gamma_{1i} = \frac{K_d SS}{h_i(1+K_d SS)}, \tag{S6}$$
$$\gamma_2 = 0.$$

In the near bottom layer located at the bottom of the water column just above the bottom sediment, the coefficients are defined as:

$$\gamma_{oi} = \frac{K_d SSW}{h_i(1+K_d SS)},$$

$$\gamma_{1i} = \frac{K_d SSW}{h_i(1+K_d SS)} + \frac{1}{(1+K_d SS)} \frac{1}{L_b \min(L_b, L_t)} + \frac{K_d SSW}{(1+K_d SS)} \frac{B}{L_b \min(L_b, L_t)},$$

$$\gamma_2 = \frac{1}{R} \frac{D}{L_t \min(L_b, L_t)} + \frac{(R-1)}{R} \frac{B}{L_t \min(L_b, L_t)},$$

$$\gamma_3 = \frac{R-1}{R} \frac{SSW}{L_t(1-\varepsilon)\rho} + \frac{1}{R} \frac{D}{L_t \min(L_t, L_m)},$$

(S7)

$$\gamma_4 = \frac{1}{R} \frac{D}{L_m \min(L_t, L_m)},$$

$$\gamma_5 = \frac{(R-1)}{R} \frac{SSW}{L_m(1-\varepsilon)\rho},$$

where the coefficient R is defined as:

$$R = 1 + \frac{\rho(1-\varepsilon)}{\varepsilon} K_d \tag{S8}$$

Here L_b (m) is the length scale of the bottom boundary layer, SS is the different for each box concentration of suspended sediments (t·m^{-3}), obtained from observations or model simulation, W_g is the settling velocity calculated as a function of suspended particles size; $SSW=SS \cdot W_g$ is the fixed sediment flux (t·m^{-2}·yr^{1}); D is the coefficient of vertical diffusion in the bottom; B is the coefficient of bioturbation in the top bottom; ε is the porosity of the bottom sediment; ρ is the sediment density.

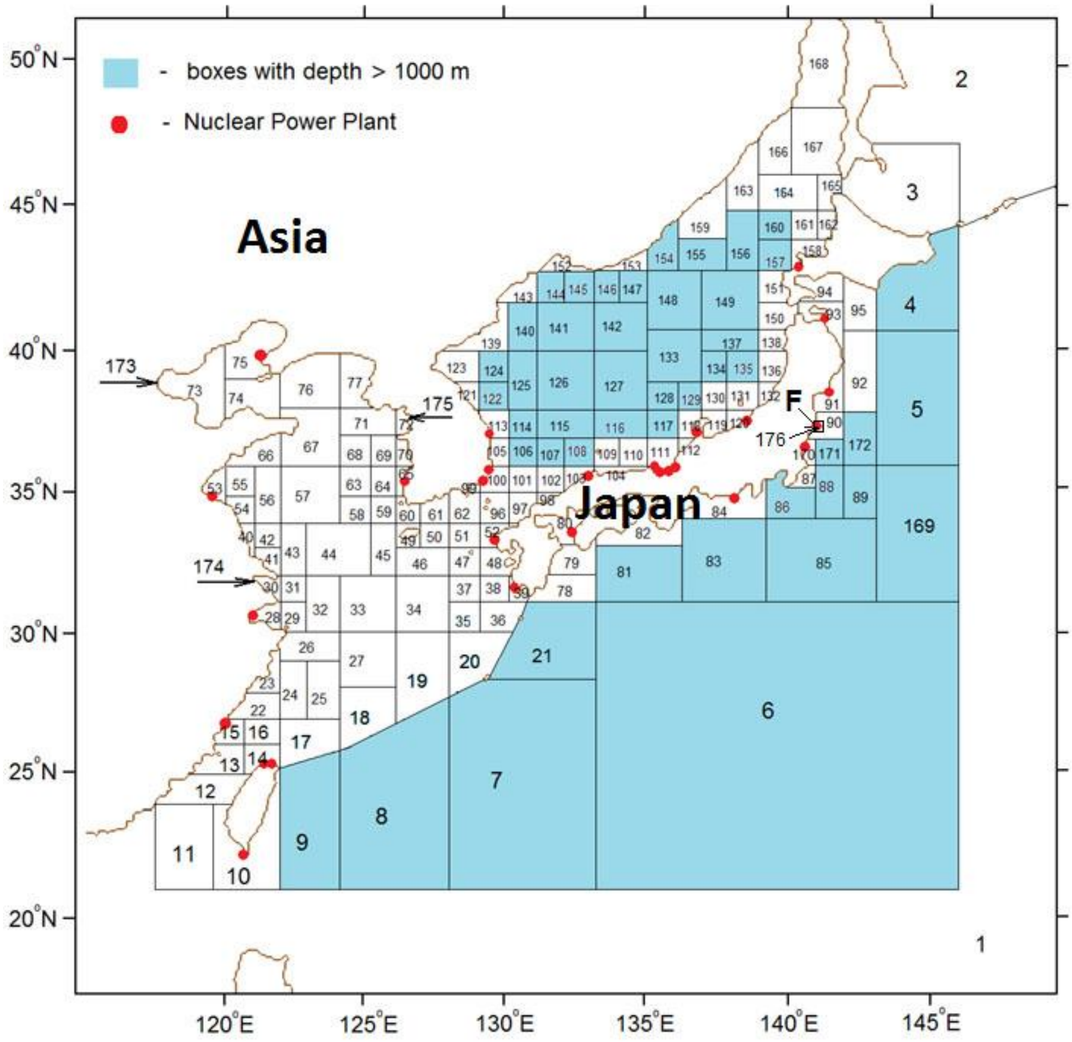

Fig. S2. The compartment system for the Northwestern Pacific. The shaded boxes represent the deep water boxes. The arrows with numbers show the compartments representing estuaries of large rivers (174 – the Yangtze River, 173 – the Huanghe River and 175 – the Han River. The NPPs are shown by filled circles. Letter "F" represent the Fukushima Dai-ichi NPP.

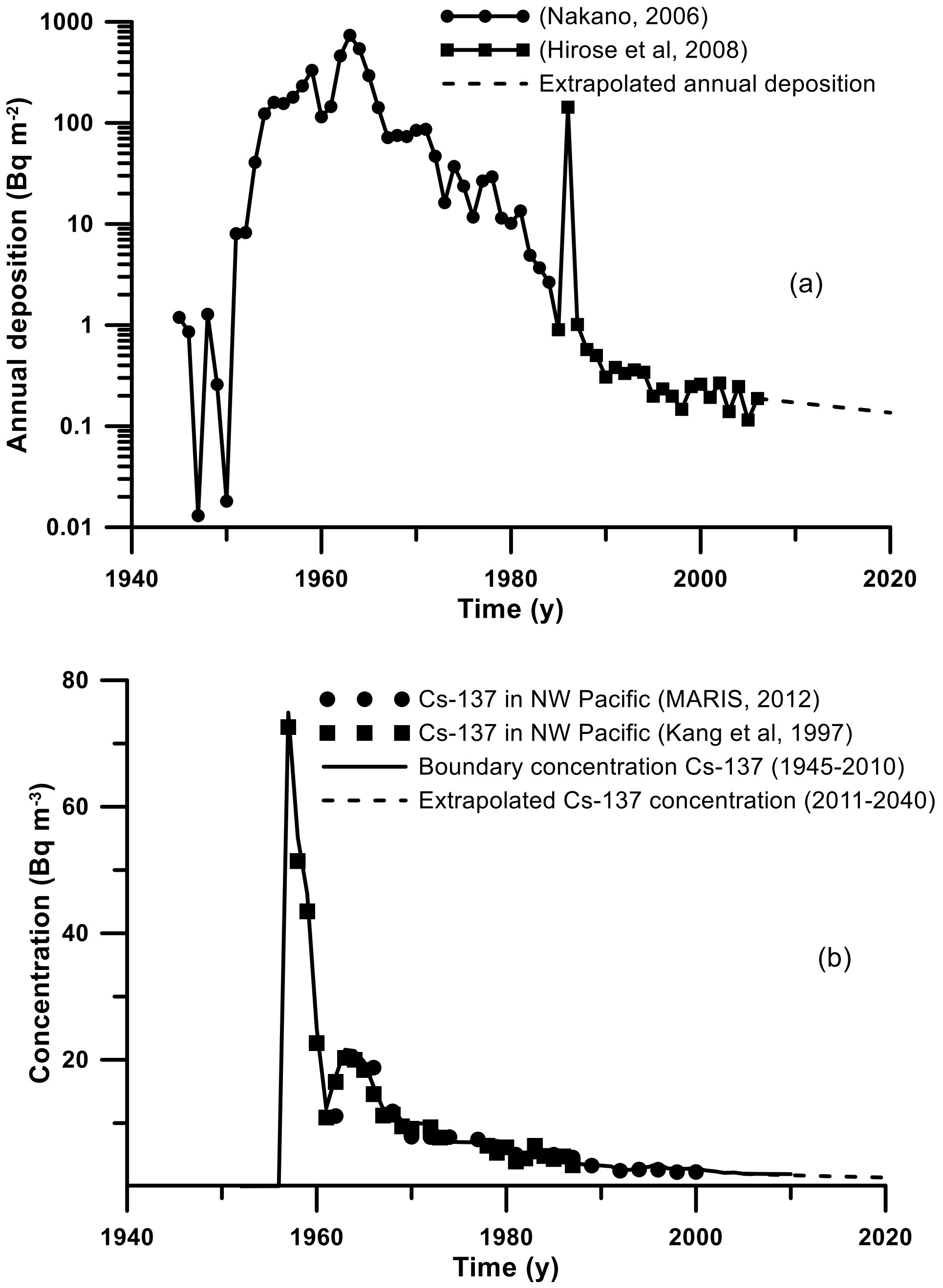

Fig. S3. Time variations of the annual deposition on the surface compiled from Nakano (2006) and Hirose et al (2008) (a) and the boundary values for the ^{137}Cs concentration in the NW Pacific compiled from MARIS (2012) database and Kang et al. (1997) (b).

Table S1 The model parameters for coastal box around the FDNPP and box 45 in the Baltic Sea.

Parameter	Coastal box (Fukushima case)	Box 45 (Baltic Sea case)
Volume, km^3	22.5	776.3
Average depth, m	50	31.4
Water exchange rate with adjacent compartments, km^{3}yr^{-1}	150	4430
Thickness of top sediment layer, m	0.1	0.05
Concentration of suspended sediments, kg/m^3	$8 \cdot 10^{-2}$	$1 \cdot 10^{-3}$
Sedimentation rate, kg(m^2 yr)$^{-1}$	$1 \cdot 10^{-2}$	$7.5 \cdot 10^{-2}$
Salinity, PSU	35	15
Sediment density, kg m^{-3}	2600	2600
Vertical diffusion coefficient in bottom sediments, m^{2}yr^{-1}	$3.15 \cdot 10^{-2}$	$3.15 \cdot 10^{-2}$
Bioturbation coefficient, m^{2}yr^{-1}	$3.6 \cdot 10^{-5}$	$3.6 \cdot 10^{-5}$
Porosity of bottom sediments	0.75	0.75
^{137}Cs distribution coefficient, K_d, m^3·kg^{-1}	2	2

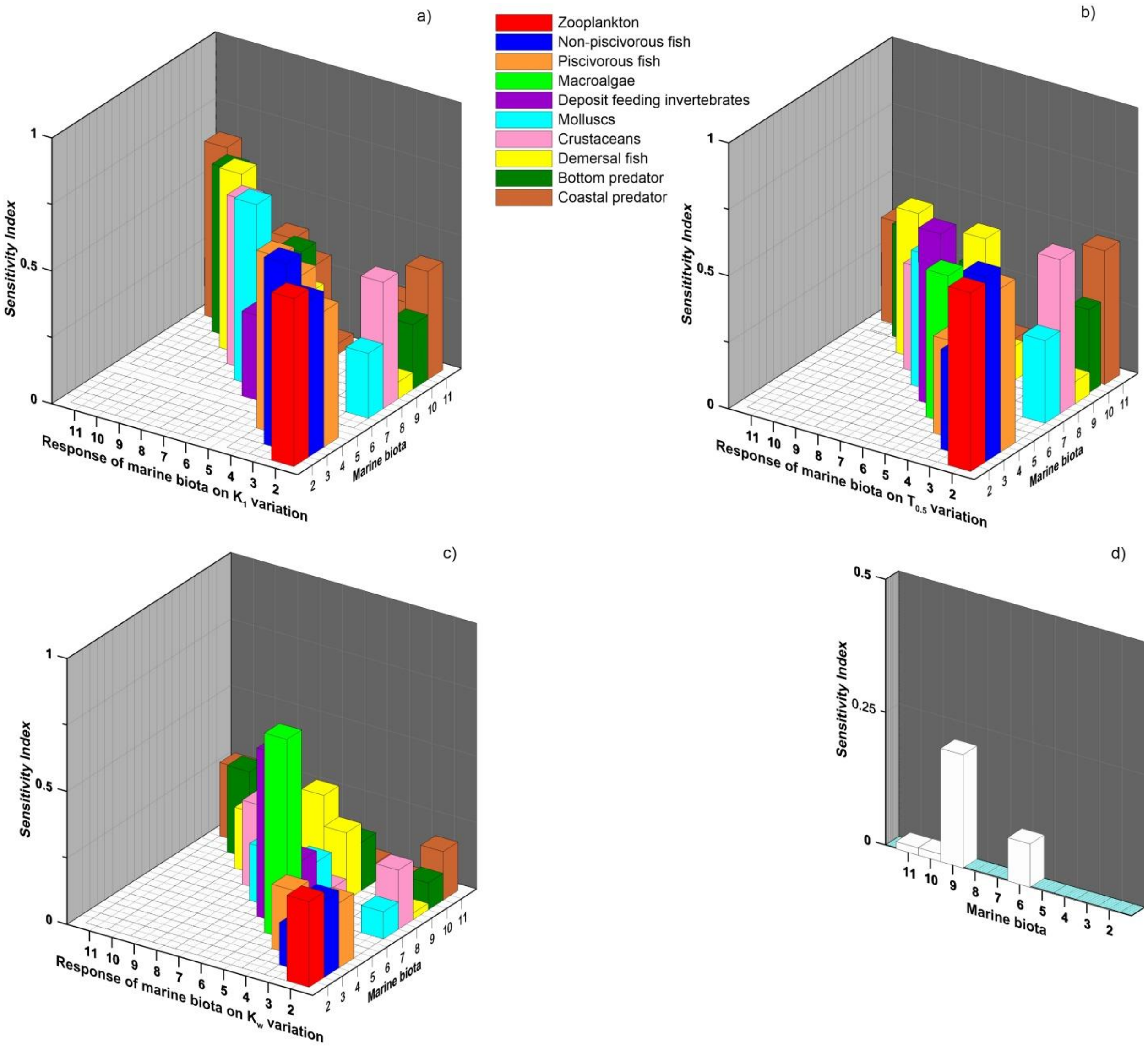

Fig. S4. Sensitivity indexes calculated for food uptake rate K_1 (a), biological half-life $T_{0.5}$ of ^{137}Cs in the organism (b), water uptake rate K_w (c) and for ratio of concentration of assimilated radioactivity from organic fraction of bottom sediment to the concentration of radioactivity of bulk bottom sediment ϕ_{org} (d).

Table S2 The river runoff into the Baltic Sea (Lepparanta and Myrberg, 2009.

River box → Baltic Sea box	Rivers	Inflow ($km^3 \cdot yr^{-1}$)
82 → 36	Gota-alv + small rivers	23
83 → 39	All Danish rivers	5
84 → 43	Small German rivers	9
85 → 45	Oder + small rivers	25
86 → 47	Wisla + small rivers	50
87 → 48	Neman + small rivers	30
88 → 59	Motala strem + Swedish small rivers	9
89 → 58	Daugava + small rivers	31
90 → 65	Narva + small rivers	20
91 → 66	Kymijoki + small rivers	13
92 → 67	Neva	79
93 → 71	Dalalven + small rivers	18
94 → 77	Kokemenjoki + other Finnish small rivers	25
95 → 78	Angerman-alv + Indals-alv + smaller rivers	47
96 → 80	Ume-alv + smaller rivers	22
97 → 81	Kemijoki + Oulujoki + Lijoki + Torne-alv + Kalix-alv + Lule-alv + smaller rivers	78

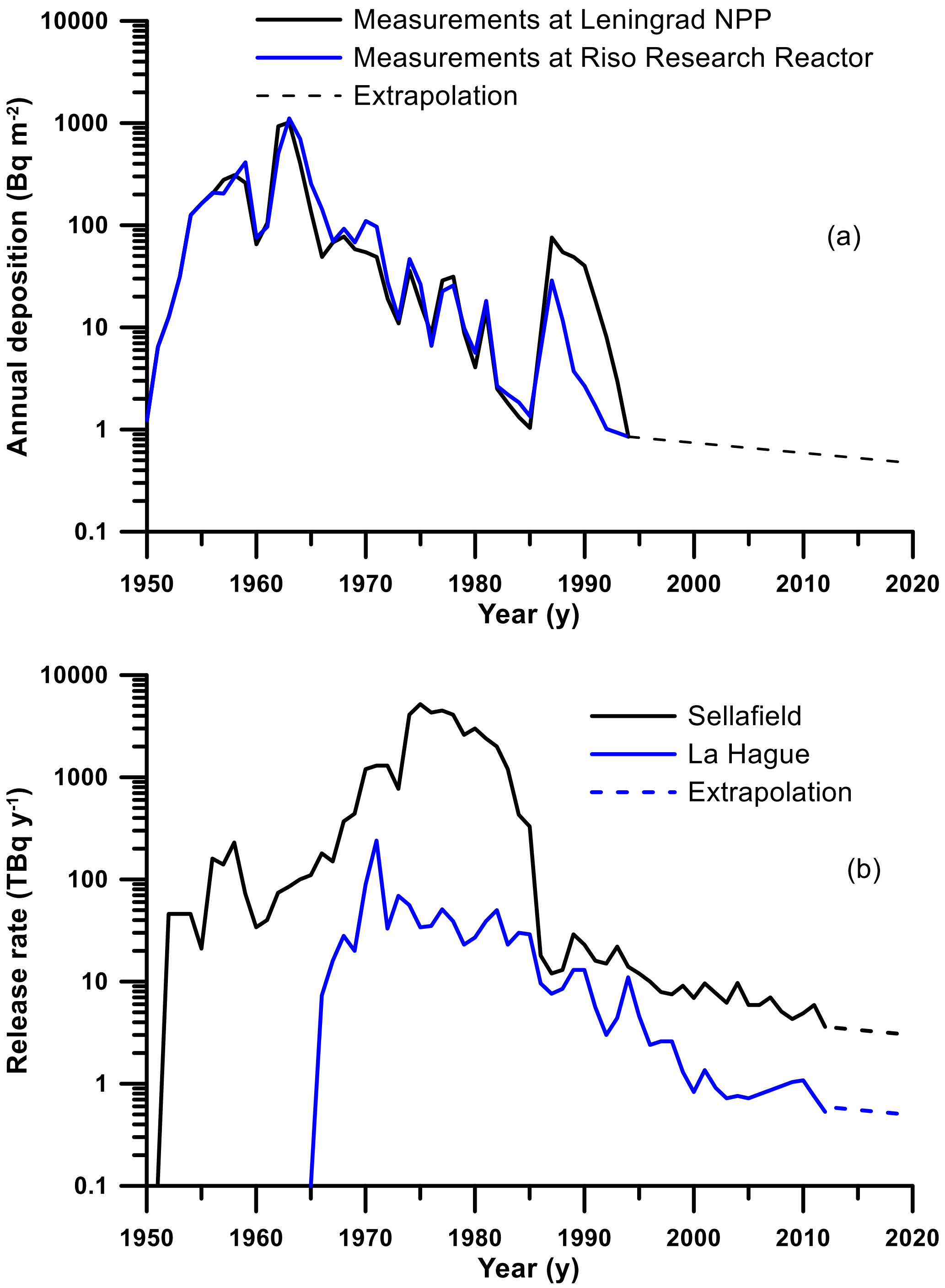

Fig. S5. Global atmosphere deposition rate of ^{137}Cs on the Baltic Sea (HELCOM, 1995) (a) and release of ^{137}Cs from Sellafield and La Hague reprocessing plants (HELCOM, 2009).

Table S3. Atmosphere deposition density of ^{137}Cs in 1986 due to the Chernobyl accident (HELCOM, 1995)

Basin	Deposition density, Bq m^{-2}	Inventory, PBq	Boxes
North-Atlantic	1000	35.4	3-34
Kattegat	1700	0.04	35-40
Belt Sea	1800	0.05	41-43
Baltic Proper	4500	0.82	44-57, 59-61
Gulf of Riga	5000	0.08	58
Gulf of Finland	15000	0.83	62-67
Aland Sea	72500	0.55	68-69, 71-72
Archipelago Sea	17300	0.04	70
Bothnian Sea	35000	1.94	73-79
Bothnian Bay	6900	0.31	80-81