### **Response to Reviewer #3**

The authors appreciate the reviewer's valuable comments. They are very helpful for improving our paper. We have now modified the manuscript accordingly, and the changes are noted point by point. Please, find our responses below.

The authors present a multi-compartment kinetic-allometric model for radionuclide bioaccumulation in fish. First, the authors present the development of the model. They tested their model on data from laboratory experiments and several radionuclides. Finally, they used their model to simulate real-case scenarios. I believe that the development of such model could have strong contribution for the risk assessment of radionuclides. However, the manuscript would need some clarifications before considering it for publication.

## **General comments**

The authors should give more information on the MCKA model structure: I would suggest adding a schematic representation of the model (In part 2. Model) as it would be of a great help to visualize the structure of the model and better understand the relationships between the compartments. The authors should also better present the parameters of the model by given a table with the definition of all parameters, their values and units. Especially, the values parameter related to the radionuclides should be clearly presented. Table 1 only presents the parameters in allometric relations. The last part (Part 4. Model applications) is really interesting as it presents model applications for simulating radionuclide bioaccumulation in real contexts. This part may even represent another manuscript. I understand why the authors wanted to keep this part here, but more explanations should be added. The relationships between the fish and their prey should be better explained. How do you handle the preference type (Pj) in the MCKA models? Could you give the parameter values? Also, you discussed that there was no major impact of temperature on uptake and elimination in laboratory experiments, but what about in real-case scenarios (l. 110)? Similarly, what about salinity? Are the abiotic parameters taken into account in the POSEIDONR model? Without more information, the coupling between the POSEIDON-R model and the MCKA model is still unclear, and the simulation results could be questioned. They also compare the results to a one-compartment model that was not presented in the simulation cases before, hence it is hard to conclude on the comparison with this model. Have you compared the simulations of the one-compartment model and your MCKA model for laboratory experiments? It would help to better explain the importance of adding complexity in the MCKA model compared to a single-compartment model. With more clarifications on those different points, I believe the manuscript would be worth for publication.

Answer. Thank you for the discussion and the important suggestions.

(1) We added a schematic description of the model in Fig. 2 and included the following text: 1.74 "A schematic representation of the model is shown in Fig. 2."

(2) Following a suggestion by Reviewer#1 we updated the tables with model parameters. The model parameters are presented in Tables 1 and 2, and in several tables in the Supplementary Material:

**Table S1** Parameters of the MCKA model for fish used to simulate the experiments (Mathews and Fisher, 2008; Mathews et al., 2008) on pulse-like feeding.

**Table S2**. Transfer rate  $k_{2,3}$  (d<sup>-1</sup>) for radionuclides used to simulate the experiments (Mathews and Fisher, 2008; Mathews et al., 2008) on pulse-like feeding.

**Table S3**. Transfer rate  $k_{2,4}$  (d<sup>-1</sup>) for radionuclides used to simulate the experiments (Mathews and Fisher, 2008; Mathews et al., 2008) on pulse-like feeding.

**Table S4**. Transfer rate  $k_{2,5}$  (d<sup>-1</sup>) for radionuclides used to simulate the experiments (Mathews and Fisher, 2008; Mathews et al., 2008) on pulse-like feeding.

**Table S5**. Parameters of MCKA model for fish used to simulate the experiments (Mathews and Fisher, 2008; Jeffree et al., 2006) on uptake of activity from sea water.

**Table S6** Transfer rate  $k_{1,3}$  (d<sup>-1</sup>) for radionuclides used to simulate the experiments (Mathews and Fisher, 2008; Jeffree et al., 2006) on uptake of activity from sea water.

**Table S7** Transfer rate  $k_{1,4}$  (d<sup>-1</sup>) for radionuclides used to simulate the experiments (Mathews and Fisher, 2008; Jeffree et al., 2006) on uptake of activity from sea water.

**Table S8** Transfer rate  $k_{1,5}$  (d<sup>-1</sup>) for radionuclides used to simulate the experiments (Mathews and Fisher, 2008; Jeffree et al., 2006) on uptake of activity from sea water.

**Table S9**. Food preference  $P_{ij}$  for predator of type *i*, and prey of type *j*.

**Table S11** Parameters of the MCKA model for prey fish (*Clupea harengus membras*) and predator fish (*Esox lucius, Hexagrammos otakii, Triakis scyllium, Squatina japonica, Sebastes cheni, Lateolabrax japonicus*) in POSEIDON-R model applications.

(3) We added a table (Table S9) for food preference in the Supplementary Material together with an extra line of text. Food preferences  $P_{ij}$  for predator of type i, and prey of type j are obtained from Bezhenar et al. (2016).

1. 284 "Table S9 in the Supplementary Material contains food preferences for organisms in the food web used in the Poseidon-R model."

(4) The modification of the dynamic radionuclide uptake model for strontium and caesium by salinity driven transfer parameters for the marine food web and its integration in POSEIDON-R was given in a paper by Heling and Bezhenar (2009). More data is necessary to verify temperature dependence of the MCKA parameters in further studies.

(5) A detailed comparison with analysis of the one-compartment model and MCKA model for FDNPP case study was added in the paper following suggestions by Reviewer #2. See answers on these comments. We did not compare MCKA and the one-compartment model with the single-feeding experiment because a one-compartment model cannot describe the fast transfer processes governed by eqn. (1).



Figure 2 Schematic of the multi-compartment kinetic-allometric model.

#### **Specific comments**

(1) In your introduction, you do not mention the POSEIDON-R model that you used to do the simulations of the accidental releases, you should at least present it. Without reading the

abstract, you do not expect to have a coupling of two models in the last part. More globally, I am not use to this type of presentation of an article, that is why I was a little bit confused reading the manuscript. I felt more like reading a report, even if I understand the importance of each part. **Answer.** We added text in the Introduction section:

1. 58 "The developed multi-compartment kinetic-allometric (MCKA) model was embedded into the box model POSEIDON-R (Lepicard et al., 2004; Maderich et al., 2014a,b; 2018b; Bezhenar et al., 2016), which describes transport of radionuclides in water, accumulation in the sediment, and transfer of radionuclides through the pelagic and benthic food webs."

(2) 1. 48: I am not a specialist of PBPK models but you wrote that fish PBPK models do not include scaling allometric relationship between metabolic rate and organism mass. However, the PBPK model of Grech et al. 2019 which takes into account the effect of growth on the cardiac output and oxygen consumption rate.

Answer. Thank you for suggestion. We changed text accordingly:

1. 51 "Note that, with the exception of model (Grech et al. 2019), PBPK fish models do not include scaling (allometric) relationships between metabolic rates and organism mass."

# (3) 1. 107: Could it have not be possible to adapt the dynamic budget theory (DEB) to model this? Could you explain better why this value of $\frac{3}{4}$ power? It is specific to fish?

**Answer.** We plan to consider possible use of DEB in future modeling. As mentioned in l. 113 "we employed quarter-power scaling for uptake, elimination and growth rates derived from general theory (West et al., 1997)..." applicable for whole specter of organisms. An important consequence of this theory for the considered model is the equation (18) in which dependence on the body mass excluded. The text was changed accordingly:

1. 113 "Here, we employed quarter-power scaling for uptake, elimination and growth rates derived from general theory (West et al., 1997). This theory predicts for all organisms a 3/4 power law for metabolic rates. It describes transport of essential materials through space-filling fractal networks of branching tubes assuming that the energy dissipation is minimized and that the terminal branch of the network is a size-invariant. The scaling relations are..."

(4) 1.115. So, if I understand well, the structure of the model if generic for different fish species but the parameters values are specific to the species depending on their weight? Maybe you should mention it for more clearly. For two different fish species of the same weight, could you not have an inter-species variability of the model parameters?

Answer. The text was modified as

1. 400 "The food and water uptake rates, elimination rate and growth rate depend on the metabolic rate, which is scaled by the fish' mass to the 3/4 power, but do not depend on the radionuclide. At the same time, the activity is distributed between the different tissues and organs according to the tissue assimilation efficiencies, which differ per radionuclide (Table 2), but that do not depend on fish mass. Therefore, the transfer rates can be associated with specific radionuclide and fish mass as shown e.g. in Tables S2-S4 and S6-S8. The position of the fish species in the trophic level also affects the concentration of activity in the organism."

(5) Fig 4. Why is there no curve for dogfish? If you could not simulate BCF for dogfish, maybe you should withdraw the concerning data points.

Answer. Calculated dogfish curve almost coincide with turbot curve. We restored it in figure.

(6) Mathews et al. 2008 (l. 190) should not be Mathews and Fisher 2008? (l. 208) Answer. Done

(7) 1. 242. A graphical representation of the results of the sensitivity analysis in SI rather than a table would be of a great help to clarify the results. Why did not you make a global sensitivity analysis to better understand interactions between parameters?

**Answer.** Thank you for suggestion. We replaced the table with a figure in Supplementary Material. In this paper, we limited ourselves to the One-At-a-Time method of relying on the results of previous studies (e.g. Bezhenar et al., 2016).

(8) 1. 275. A schematic representation of the one-compartment model would be required in SI as well as a table with the parameter values.

**Answer.** The two parameters of one-compartment model are given and discussed in the main text of paper (lines 289-293 and 379).

### (9) 1. 276. I do not understand the sentence.

Answer. The text was changed accordingly. See also answer on comment 10.

1. 295 "The nested boxes (`inner' and `coastal' boxes) inside the regional box no. 68 in the Baltic Sea box system were added to resolve the radionuclide concentration in the near field (Fig. 6)."

(10) 1. 277. What are the "inner box" and "coastal box" exactly? I do not really understand as I am not familiar with the POSEIDON-R model. It should be better clarified.

Answer. The inner and coastal boxes were explained as suggested:

1. 288 "The POSEIDON-R model can handle different types of radioactive releases: including atmospheric fallout and point sources associated with routine releases from nuclear facilities located directly on the coast or 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. The intermediary boxes between 'coastal' and 'regional' boxes are called 'inner' boxes."

### (11) Figures 7 and 8 could be coupled (6 panels).

Answer. Thank you for suggestion. We merged figures 7 and 8 and 9 and 10.

### **Technical corrections:**

- *Several spaces are missing:* 1. 107, 1. 241 **Answer.** Done

- *Error on the reference*: 1. 278 **Answer.** Done