Response to Reviewer #2

The authors are most grateful to the reviewer for thorough analysis of the manuscript and the constructive criticism and suggestions. We have followed your suggestions and revised the manuscript accordingly. Please, find our responses below.

General comments

The authors present a multi-compartment model for radionuclide bioaccumulation in fish. The compartments for this model are muscle, bones, and organs. Uptake can be by direct absorption through gills, or from food. Transfer is also allowed between compartments. The model was tested on a set of radionuclides, with good agreement with lab experiments. The model was implemented into the POSEIDON-R, and applied to several real-world scenarios. This seems to work better than the previous single compartment model that was previously used. Overall, the paper is detailed and well-written. The authors present a novel method, integrated into a current software with applications to real-world problems.

My major concern about the work here is in the comparison between the MCKA model and one-compartment or equilibrium models. The Forsmark results seem to show improvements in estimates from the MCKA model as opposed to the one-compartment model and Equilibrium models. However, I'm not convinced that it's not just because of poor-quality estimates of parameters for the one-compartment model and equilibrium. The equilibrium model consistently underpredicts by a factor of _10 for 54Mn over a period of decades. If you want a fair comparison for the underlying model, then you need to make sure they all the parameters are consistent. Are the parameters consistent between models? That is, you could use the MCKA model to estimate equivalent one-compartment parameters and BAF parameters such that the equilibrium concentrations are all identical. In that case, are the results significantly different? If the results are still different, then you have shown that your additional model complexity is needed for higher accuracy in these dynamic problems. If they aren't, then it just shows that your method can be used to estimate these factors for a given ecosystem model. This would still be an excellent finding, as it will help with model building, but it wouldn't be necessary to explicitly track all the concentrations inside the model. Judging from the results in figure 2, it looks like the inter-compartment equilibrium is reached quite quickly (<2 days?) in this case, the system should behave identically to a single-compartment system, should it not? If this issue is resolved then I would highly recommend publication.

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

(i) For the Forsmark simulation we used generic parameter values for all models including the MCKA. The aim was to demonstrate the ability of using the MCKA model without *a posteriori* information.

(ii) As follows from a comparison of the equations (1)-(3) and (7) of the MCKA model and the equation (11) of the standard whole-body model, the main difference between them is the description of the whole-body elimination rate λ_{wb}. Therefore, we considered in more detail the timely variations of the calculated λ_{wb} in the MCKA model. The results are presented in a new Fig. 11.

(iii) Following your suggestion we compared simulation results on the FDNPP accident obtained with the MCKA model and the one-compartment model. In both models identical AE values are used. The λ_{wb} computed using the MCKA model was variable, while the λ_{wb} in the one-compartment model was set to a fix value of 0.0027 d^{-1} for piscivorous fish. This value was

obtained from the MCKA model representing the conditions prior to 2011 (Figure 11 (c)). The results from this comparison were added in Figs. 10 and 11, which were merged as Fig. 10.
(iv) We fixed some errors in the calculations presented in Fig. 2. Therefore, input of the different tissues is clearly visible now.

The text and figures were reworked as follows:

l. 364 "Comparison of the equations (1)-(3) and (7) of the MCKA model and the equation (11) of the standard whole-body model demonstrates that the main difference is found in the description of the whole-body elimination rate λ_{wb}. Whereas in the whole-body model the value of λ_{wb} is constant, in the MCKA model it is the ratio of activity weighted tissue elimination rates to whole-body activity

$$\lambda_{wb} = \frac{\sum_{i=3}^{5} \mu_i \lambda_i C_i}{C_{wb}}. \quad (34)$$

Therefore, in the MCKA model, the value of λ_{wb} can vary over time, depending on the uptake of radionuclide and the tissue elimination rates. The time variation of λ_{wb} computed from (34) is shown in Fig. 11 for three different cases: (a) pulse-like feeding experiment (Matthews, Fisher (2008), (b) release of ^{60}Co during normal operation of the Forsmark NPP and (c) accumulation of ^{90}Sr in the fish due to the FDNPP accident. As seen in the plots, λ_{wb} varies considerably when there is non-equilibrium, such as in the case of a pulse-like feeding or an accident. Even in case of a routine release, λ_{wb} follows any changes in the release rate (Fig. 11b). In case of the FDNPP accident, the calculated λ_{wb} shows some tendency towards an equilibrium value, but after a pulse-like release of ^{90}Sr in 2011 λ_{wb} doubled following the release of activity and then slowly converged to the quasi-equilibrium state governed by the global deposition. Notice that in this case we extended simulation period to 2040 extrapolating deposition data and FDNPP release data in Fig.9b. Therefore, whole-body model with a constant λ_{wb}, that is calibrated using observational data, cannot to correctly describe such transient processes in the organism. This is confirmed in the Fig. 10 by comparing the results from the MCKA model and the one-compartment model. Here the AE value in both two models are the same, whereas the equilibrium value λ_{wb} is calculated in the MCKA model using value before 2011 for piscivorous fish (λ_{wb} =0.0027 d^{-1}). As seen in Figs. 10c and 10f, the target tissue (TT) model (Maderich et al., 2015) underestimates the concentration in fish comparatively with MCKA model and observations. The one-compartment model simulation results using parameters from MCKA model are close to the MCKA model results at initial stage of accidental release. However, over time, the concentration ^{90}Sr in fish tends to equilibrium faster than the MCKA model predicts, which is explained by the time-dependent behavior of λ_{wb} in the MCKA model. The calculation results confirm that generic parameters of the MCKA model make it possible to correctly estimate λ_{wb} without preliminary calibration on the local measurement data, which may be impossible in an accident. "

l. 396 "The main difference between MCKA and whole-body models was found in the description of the whole-body elimination rate λ_{wb}. Whereas in the whole-body model the value of λ_{wb} is constant, in the MCKA model it is the ratio of activity weighted tissue elimination rates to whole-body activity as described (34). The elimination rate λ_{wb} varies considerably in non-equilibrium state of fish, such as in the case of a pulse-like feeding or an accident."

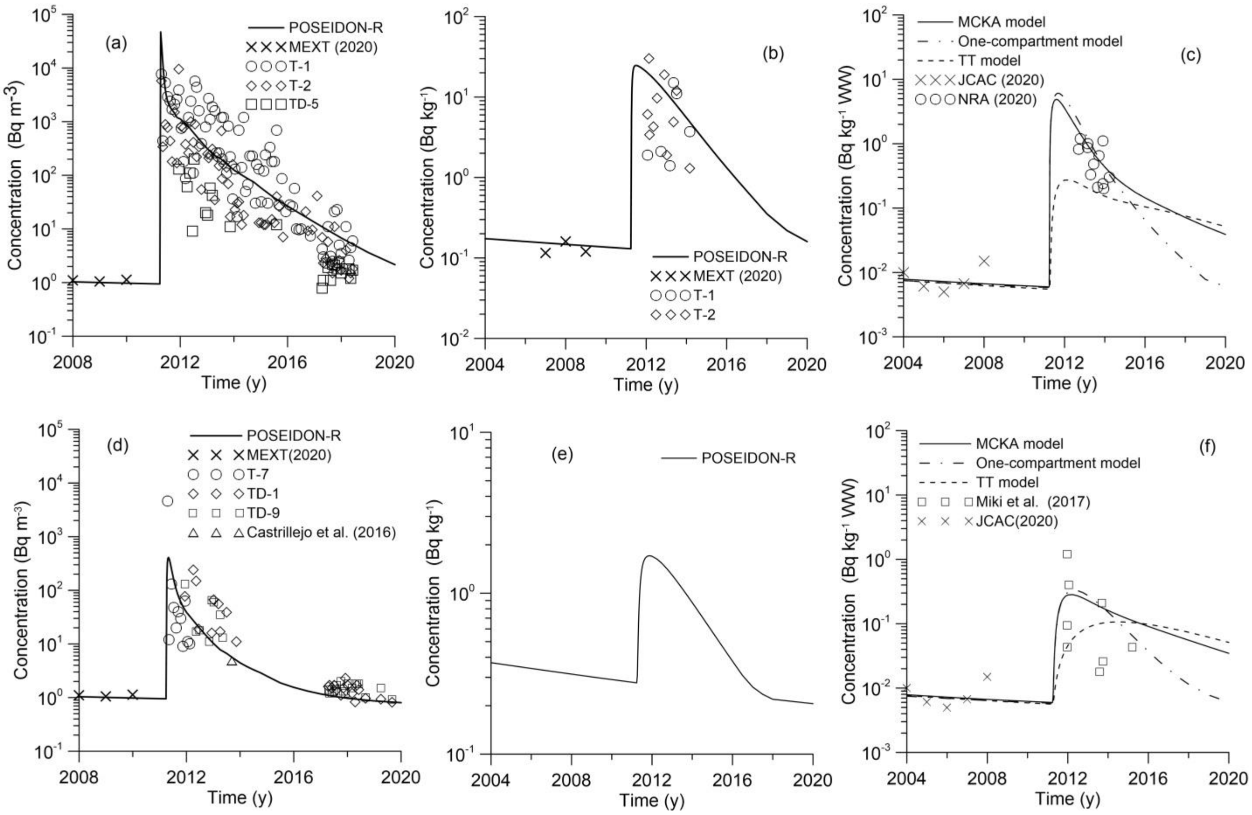

Figure 10. Comparison between calculated and measured ^{90}Sr concentrations in water (a), bottom sediment (b), piscivorous fish (c) for the coastal box and in water (d), bottom sediment (e), and piscivorous fish (f) for box no. 173.

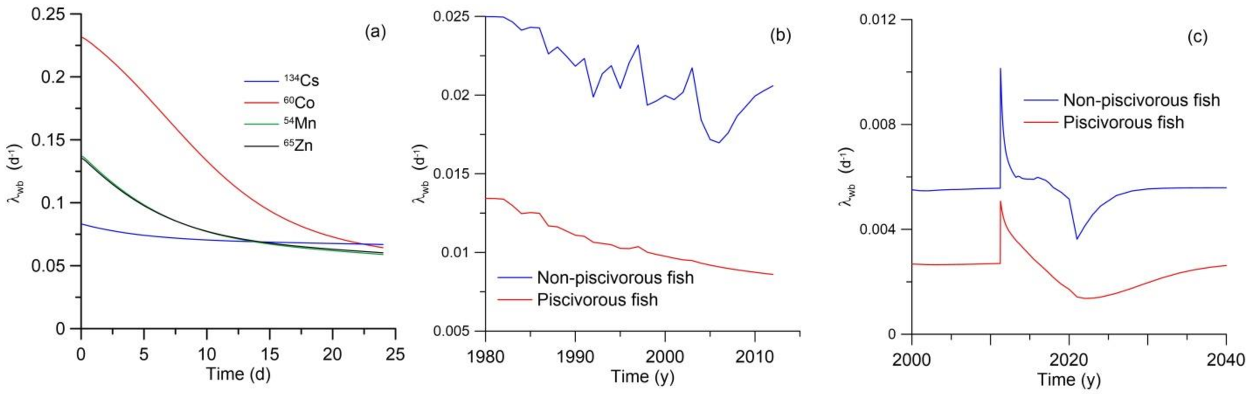

Figure 11. The calculated λ_{wb} for three scenarios: (a) the pulse-like feeding experiment (Matthews, Fisher, 2008), (b) the release of ^{60}Co during normal operation of the Forsmark NPP and (c) the accumulation of ^{90}Sr in the fish (coastal box) due to the FDNPP accident.

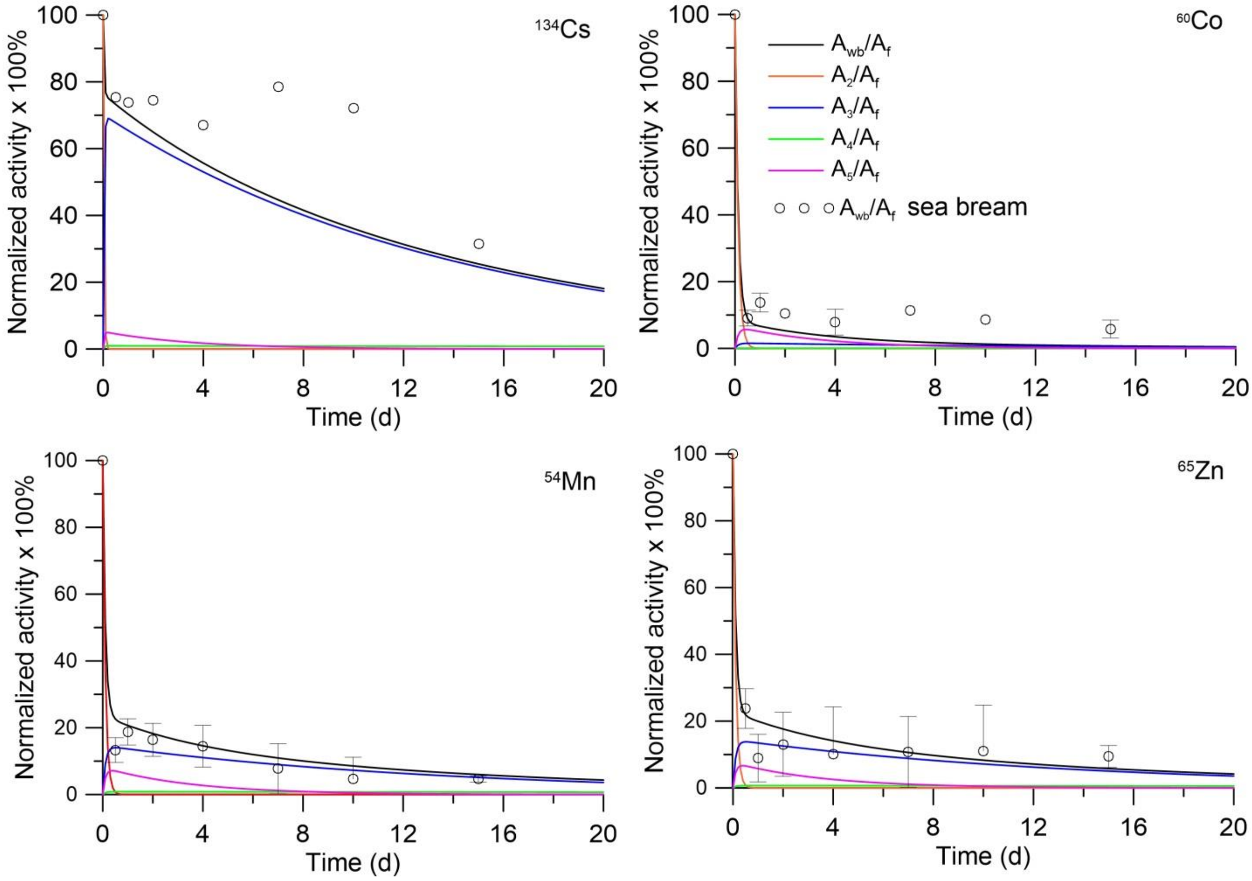

Figure 2. Retention of radionuclides in whole body and tissues of juvenile sea bream (*Sparus auratus*). The simulations are compared with whole body measurements by Mathews and Fisher (2008).

Specific comments:

l. 127 Not sure what is meant by "*The equations under (17) is used*"

Answer. The text was changed as

l. 131 "The equations (17) are used"

Should define BAF in equation somewhere. You may also note that IAEA uses concentration ratio CR or concentration factor CF to describe what you are using as bioaccumulation factor BAF, while you use CR for something different. I believe the IAEA document only has BAF_wb, not BAF_food, does it not?

Answer: We changed text accordingly.

l.139 "We define bioconcentration factor (*BCF*) as ratio of whole-body of fish to water concentrations with no dietary intake, bioaccumulation factor (*BAF*) as ratio of whole-body of fish to water concentrations with dietary intake, body-to-tissue concentration ratio (CR_i) as ratio of whole body to tissue concentrations, whereas ratio of food to water concentrations is indicated as BAF_{food}."

l.139 "Values of *BAF* for different radionuclides expressed as *CF* in (IAEA, 2004)."

l. 229 "*shown the importance of including in the model of the digestive tract compartment describing highly non-equilibrium transfer dynamics" this seems to show the importance of kinetics in the modeling, but not the digestive compartment per se, as opposed to just using a single compartment.*

Answer. Thank you for suggestion. The text in l. 229 was rewritten accordingly:

l. 239 "Comparison of the model against laboratory experiments on the retention of absorbed elements in fish after single feeding demonstrated the need to include the kinetic characteristics of the digestive tract in the model when highly non-equilibrium transfer dynamics are expected."

The half-life of 54Mn is only 312 days, so could be relevant compared to the biological half-lives. Was this accounted for in the modeling>?

Answer: The simplified equilibrium relations (17), which allow use of scaling (12) in (18) were obtained with the assumption that the elimination rates λ_i were much greater than the physical decay rates λ. Therefore, the physical decay of radionuclides was not taken into account in the transfer processes in fish. For ^{54}Mn, the decay constant λ=0.0022 was still less than the average value for fish λ_{wb}=0.009 in the Forsmark case study. Notice, that the physical decay was taken into account for processes of radionuclide transfer in the water and interaction with suspended and bottom sediments within the POSEIDON-R model used for long term simulation in the Forsmark and in the FDNPP case studies.

The text was added accordingly:

l. 131"This assumption also imposes requirement on the modelling of radionuclides with decay constant $\lambda<<\lambda_i$."

l. 321 *using compartments here as spatial regions may be confusing.*

Answer: Spatial "compartments" in text were replaced by "boxes"

Technical corrections:

Figure 1 should be regenerated in higher-quality.

Answer. Done

Regarding eq. 1-3, you describe all variables except C_i

Answer. Done.

l. 125 *should be lambda_g*

Answer. Done.

Figures 2-4 are low quality JPG. Avoid using lossy compression (jpg) on graphs – use lossless (e.g., png) or vector graphics (pdf/svg/wmf).

Answer. We used eps for all figures. These figures were reworked in higher quality.

l. 240 *space before 60Co*

Answer. Done.