We thank reviewers for their comments. We address each individual comment in detail below. Please note that all changes made in this revised version are written in bold font.

This revised version takes into account all reviewers remarks and comments. Moreover, more recent data on plankton contamination reported especially by Kaeriyama et al. (2014) and Baumann et al. (2015) were included in this paper and are taken into account in the model validation (see section 3.4) and in the different discussion parts.

Response to reviewer # 1

Question 1:

I fail to see the point of this paper. The authors' goal of the paper was stated as:

"We presented a modelling approach based on an ecosystem model to estimate the 137Cs activity in marine plankton populations following the Fukushima nuclear power plant (FNPP) accident" I am not going to quibble too much about the strengths of the model and its details, which seem to be mostly sensible, but why do this in the first place? Why not simply measure Cs activities in the plankton, or rely on other people's measurements?

Answer 1:

The aim of this paper was to present an approach for modelling the transfer of cesium in planktonic populations as a first step before modelling its transfer to fish in an accidental situation. Of course it is worth having field data but as it appears in the case of Fukushima, there are not so many data on plankton and in case of an accident the only way to get clues about the impact on the marine food chains is to have tools: modelling is one of them.

The radioecological models once validated using the field observations, enable one to predict the evolution of radionuclide concentrations in the biological compartments in both spatial and temporal scales, compensating therefore the lack of observation data, especially in the case of planktonic populations for which the data are generally scarce, as is the case for the post Fukushima accident period. For example, the study carried out and described in this paper, covers a large spatial scale compared to what has been taken into account by the monitoring plans. Consequently, it enables the prediction of contamination levels even for the planktonic populations located in the areas that are not taken into account by the monitoring plans.

In addition, modelling is a primordial tool to understand and explain some processes that are generally difficult to determine by direct measurements. In our study for example, we have estimated some parameters (e.g. the importance of different contamination routes of radiocesium accumulated by different zooplankton size-classes, the trophic transfer factors, the apparent concentration ratio, etc), we have studied the influence of the accidental conditions on their basic values (in normal conditions), and we have studied their relations with some environmental conditions in the area (food availability for example). This is generally more difficult to do using only the field measurements.

Finally, it should be important to note that, in this study, we have proposed an innovative approach that enables the modelling of radionuclides bioaccumulation by planktonic populations in parallel with their ecological functioning (growth, mortality, grazing, etc) and the dynamics of their surrounding environment (ocean circulation, temperature, light, etc), which is more logical and closer to reality.

All these arguments are well explained in the introduction of this paper.

Question 2:

Betinetti & Manca - there are much better references for this statement

Answer 2:

The reference "Bettinetti and Manca, 2013" is replaced by : Fowler and Fisher (2004) ,

(Fowler,S.W., Fisher,N.S., 2004. Radionuclides in the biosphere. In: Hugh,D.L. (Ed.), Radioactivity in the Environment. Elsevier, pp. 167–203 (Chapter6))

Question 3:

"Plankton populations were largely affects by this contamination …" Really? How? I doubt it.

Answer 3:

It is perhaps a problem of language. In English contaminated is likely the proper word. The term "largely" has been deleted.

« Plankton populations were contaminated by these releases»

Question 4:

Statement that reads “Consequently, the effective consideration of all these factors implies that the modelling approach of radionuclide transfer to marine biota should be driven by an ecosystem model describing different ecological processes and transfers between organisms in the food web” is not at all convincing. Again, why develop a model to estimate Cs in plankton when there are direct measurements of this?

Answer 4:

Please see answer 1.

Question 5:

For the statement “The simple linear method based on the bioconcentration factor, defined as the ratio of the amount of radionuclide in the organism divided by the concentration in the water, is the most commonly used to assess the radionuclide concentration in marine biota (Buesseler, 2014)” Buesseler 2014 is the wrong reference for this statement.

Answer 5:

The reference “Buesseler 2014” is replaced by “IAEA (2004)”

Question 6:

Statement that photosynthesis plays a prominent role in regulation of radionuclide concentrations in primary producer populations - this is normally not true.

Answer 6:

This statement has been deleted.

Response to reviewer # 2

General comments:

Question 1:

The output from the simulated result is regulated by the contamination source input to the model. Thus it is necessary to show the information of what kind/amount of the radiocesium source (e.g. atmospheric: 2.0?? PBq within radius of ??km, initial liquid release 3.5?? PBq) was introduced into the model as the source input for this study analysis. At the same time, it should be stated what other possible sources (e.g. redistributed by river discharge, late continuous release etc.) were not considered in this study. Otherwise, the reader cannot identify limits of the authors result, the applicability of which is regulated by the source used in this modeling study.

Answer 1:

The following text has been added at the end of paragraph 2.4 (page 7):

"…the amount of atmospheric deposition included was 0.26 PBq within a radius of 80 km. The direct leakage was about 5.5 PBq released between 12 March and 30 June 2011. The simulation was extended until 31 December 2012, The inverse method described in Estournel et al., (2012) and used to calculate the source term in the first three months after the accident was applied to the whole period. After June 2011, the concentrations at the two outlets of the nuclear power plant were simplified to a linear decrease from 40 and 20 Bq/L on 1st July 2011 to 8 Bq/L for both outlets at the end of 2011 and then remained constant at this value for 2012. However, no other additional source (e.g. terrestrial runoff, rivers flow, etc) has been considered in this simulation."

Question 2:

Bio-kinetic parameters in the model are calibrated by the measured concentrations in zooplankton collected from Sendai Bay (MEXT?). As Kaeriyama et al., (2015) discussed about the biota data from these coastal waters, the analyzed values of zooplankton samples collected by Bongo/sledge nets were higher variable, probably because they contained suspended particles with or within the zooplankton. A similar tendency for higher variability in concentrations in collected plankton samples has been pointed out in the conference presentations by Aono (NIRS) and Ishimaru (Tokyo Univ MST)(unpublished data). The effect of this kind of variability in the data on model calibration is more or less significant for the final calculated output. One may use a bias-based calibrated parameter as an apparent (calibrated) value, especially in the model simulation carried in an area of similar conditions such as the coastal waters around Fukushima. However, in contrast, the application of bias-based calibrated parameters will generate overestimations in the case when applied to the North Pacific Ocean where the contribution of contaminated suspended particles is negligible. Thus, in this paper the authors should mention that their result of the Ocean simulation may be overestimated. Or if possible, they should consider deriving a correction factor (see. Tateda et al., 2015). If this cannot be done, mention of the extent of the assumed error would be helpful to avoid the reader's misunderstanding about the result being a maximum value or bias-based estimation.

Answer 2:

For the parameters calibration we used zooplankton data from coastal areas presented in (Kaeriyama et al., 2014). According to these authors, zooplankton gut content in these areas may contain particles with high ^{137}Cs levels (this is just an hypothesis among others that was not confirmed by the authors). Consequently, it is very difficult (if not impossible) to quantify the biases that could be generated in the calibrated parameters (if this hypothesis is true), since the samples composition has not been reported. For this reason, we have simply added the following discussion in the paragraph 3.2 :

"However, for the calibration we used zooplankton data from coastal areas presented in (Kaeriyama et al., 2014). According to these authors, zooplankton gut content in these areas may contain particles with high ^{137}Cs levels, which could affect the calibrated values. Consequently, over-estimations in ^{137}Cs concentrations in these populations could be generated especially in the open ocean where the particles contribution is generally negligible."

Question 3:

The significant findings in this paper are that the time-dependent radiocesium concentration in zooplankton is theoretically explainable by temporal changes of plankton biomass and the food ingestion/composition rates reconstructed from the ecosystem model. In addition, it is worth reporting that limited oceanic winter food condition reduces the radiocesium concentration in zooplankton, though it should be evaluated by multi-year simulation by validation with filed observations (Kitamura, Nishikawa unpublished data).

Answer 3:

We agree with the reviewer though the required data are not available, so the following paragraph is added in the conclusion:

"It should be important to note that although the model results indicate that the spatio-temporal dynamics of ^{137}Cs concentrations in zooplankton populations in non-accidental conditions are mainly depending on the food availability (i.e. phytoplankton biomasses in the area), with an apparent decrease of cesium concentrations in these populations during the limited-food conditions (e.g. winter), this finding has to be verified and validated by multi-years field observations once these data are available."

Question 4:

The order of appearance of some tables and figures do not correspond to the order they are discussed in the text. Since tables and Fig. 2 are not necessary to shown in the main text, it is suggested to put them in an Appendix at the end of the paper, or in Supplementary Material.

Answer 4:

The order of appearance of different tables and figures has been reviewed and corrected in accordance with their appearance in the text. Figure 2 has been deleted.

Specific comments:

Question:

"assess the radionuclide concentration in marine biota" -> "assess the radionuclide distribution between marine biota and the environment"? or "reconstruct the radionuclide concentration in marine biota"?

Answer:

The two propositions are correct but in this paragraph I mean " the most commonly used to reconstruct the radionuclide concentration in marine biota".

Question:

Missing (ZP) in "predatory zooplankton (ZP) such as krill and/or jellyfish"

Answer:

(ZP) is added to the "predatory zooplankton".

Question:

The source information used for the simulation has to be shown, such as atmospheric, initial effluent and continuous release (if included in this paper).

Answer:

Information related to the source term and atmospheric deposits used in the simulations are added at the end of the section 2.4 as follow:

« The amount of atmospheric deposition included was 0.26 PBq within a radius of 80 km. The direct leakage was about 5.5 PBq released between 12 March and 30 June 2011. The simulation was extended until 31 December 2012, The inverse method described in Estournel et al., (2012) and used to calculate the source term in the first three months after the accident was applied to the whole period. After June 2011, the concentrations at the two outlets of the nuclear power plant were simplified to a linear decrease from 40 and 20 Bq/L on 1st July 2011 to 8 Bq/L for both outlets at the end of 2011 and then remained constant at this value for 2012. However, no other additional source (e.g. terrestrial runoff, rivers flow, etc) has been considered in this simulation. »

9505 13:

Question:

If the radionuclide contribution from terrestrial runoff is not estimated and included in this simulation, this should be mentioned in the text. (See Nagao, S., Kanamori, M., Ochiai, S., Tomihara, S., Fukushi, K., Yamamoto, M., 2013. Export of 134Cs and 137Cs in the Fukushima river systems at heavy rains by Typhoon Roke in September 2011. Biogeosciences 10, 2767-2790.; Tateda, Y., Tsumune, D., Tsubono, T., Aono, T., Kanda, J., Ishimaru, T., 2015. Radiocesium biokinetics in olive flounder inhabiting the Fukushima accidentaffected Pacific coastal waters of eastern Japan. J. Environ. Rad. 147, 130-141)

Answer:

Terrestrial runoff contribution is not included in this simulation, I added this information at the end of the section 2.4 as follows:

"However, no other additional source (e.g. terrestrial runoff, rivers flow, etc) has been considered in this simulation"

9505 17:

Question:

Instead MEXT (2014), Kaeriyama et al., (2015) should be cited. See "Kaeriyama, H., Fujimoto, K., Ambe, D., Shigenobu, Y., Ono, T., Tadokoro, K., Okazaki, Y., Kakehi, S., Ito, S., Narimatsu Y., Nakata K., Morita, T., Watanabe T., Fukushima-derived radionuclides ^{134}Cs and ^{137}Cs in zooplankton and seawater samples collected off the Joban-Sanriku coast, in Sendai Bay, and in the Oyashio region. Fish Sci (2015) 81, 139–153"

Answer:

MEXT (2014) reference is replaced by Kaeriyama et al. (2015) in all the text and in the bibliography list.

9507 25:

Question:

Corresponding wet weight should be shown in the text, to compare with those of zooplankton (approximately, 200–600 ? mg w.w.m-3). In that case, please cite the reference for the Organic matter/Chl ratio used for the wet weight calculation

Answer:

The ecosystem model outputs are expressed in *mmol N m^{-3}*, the conversion to the *mg wet weight* unit is carried out using the ratios: C:N of 133/17, C:chl of 50 and the relationship between the nitrogen and the wet weight defined by Yamaguchi et al. (2005) for North western Pacific plankton.

$$log_{10}(X[mgNm^{-3}]) = -2.57 + 1.26log_{10}(M[mgwwm^{-3}])$$

Where ***M*** is the mass that we want to convert and ***X*** is the result of this conversion. By simplifying this equation we obtain:

$$X[mgwwm^{-3}] = 10^{\left(\frac{log_{10}(M[mgNm^{-3}])+2.57}{1.26}\right)}$$

So : 1 mg N m^{-3} → 110 mg ww m^{-3}

$\frac{12*133}{14*17}$ mg C m^{-3} → 110 mg ww m^{-3}

1 mg C m^{-3} → 16 mg ww m^{-3}

1/50 mg C m^{-3} → 16 mg ww m^{-3}

1 mg chl-a m^{-3} → 800 mg ww m^{-3}

So, to compare the chl-a concentrations reported in Fig.3 with the zooplankton biomasses expressed in *mg wet weight m^{-3}*, one has to multiply these values by a factor of 800.

The same reference has been used in this study to convert the mmol m^{-3} unit obtained from the model to mg wet weight in the case of zooplankton populations.

Reference: Yamaguchi, A., Watanabe, Y., Ishida, H., Harimoto, T., Maeda, M., Ishizaka, J., ... & Mac Takahashi, M. (2005). Biomass and chemical composition of net-plankton down to greater depths (0–5800m) in the western North Pacific ocean. *Deep Sea Research Part I: Oceanographic Research Papers*, *52*(2), 341-353.

9509 13 :

Question:

Calibrated elimination rate 0.03 – 0.11 d-1 for zooplankton is likely to be lower than the experimentally derived elimination rate 0.8 d-1 in zooplankton (the rotifer, Brachionus plicatilis) in Japan (Aomori Prefecture. 1990. Heisei-gannen Marine Environmental Radioactivity, General Review Report. Aomori Prefecture, Aomori, 91pp. (in Japanese)

The calibrated accumulation rate of 5 x 10-4 L g-1 d-1 (being approximately equivalent to 1.0 d-1 if assuming 500mg w.w.m-3) for zooplankton is also likely to be smaller than the experimentally derived elimination rate of 50 d-1 in zooplankton (Brachionus plicatilis) in Japan (Aomori Prefecture. 1990).

Answer:

We were not aware of this document published in japanese and we would like to know under what conditions this rate was determined since the rotifer *Brachionus plicatilis* is quite an euryhaline species.

9509 10 :

Question:

If the calibrated transfer rates in Table 1 are derived by fitting the simulated result to the observed result, they should be described as “apparent”.

Answer:

The term “apparent” is added in the text and in the table caption to the calibrated parameters

9509 26 :

Question:

Unify the term to “accumulation rate” instead of “uptake rate”.

Answer:

The term « uptake rate » in the text is replaced by « accumulation rate » .

9512 7, 10 :

Question:

“the simulated zooplankton” -> “simulated large ? zooplankton”. Are these discussion points for “large” or weight averaged ZS, ZL and ZP?

Answer:

The concentrations presented in Fig. 5 correspond to the weighted average of ^{137}Cs concentrations in the three size classes of zooplankton (ZS, ZL, and ZP). So. this discussion concerns these weighted average concentrations.

9512 22 :

Question:

Missing (R) in “calculated a ratio (R) of the 137Cs concentration

Answer:

(R) is added

9513 23 :

Question:

The vertical removal and transport of radionuclides to bottom layers is an important process in the open ocean as discussed in this paper. However, at present there is no quantitative proof for a significant contribution of this process around 1FNPP. Other processes are suggested as being critical in the Fukushima coastal waters, e.g. continuous releases, river discharged particles from highly contaminated land areas, etc.

Answer:

Even if there is no quantitative estimates related to the contribution of contaminated organic particles in contamination of sediment around 1FNPP, this phenomenon has already been observed following the Chernobyl accident as discussed in this paper. Consequently, we think that it is worth mentioning the existence of this process and its likely contribution in the contamination of sediment and benthic organisms especially in the closest vicinity of the 1FNPP. Though, of course, we are aware that continuous releases and run off processes are critical for the contamination of coastal areas close to 1-FNPP.

9514 24 :

Question:

"poor" ->"oligotrophic"?

Answer:

« poor » is replaced by « less productive »

9515 8 :

Question:

"The time needed for …"-> "The time derived from the modeling analyses for these …

Answer:

The term « estimated » is added to the sentence → « the **estimated** time needed for …. »

9516 3.8 :

Question:

Is the term "TTF" is worth discussing? Discussed here is the apparent TTF (aTTF) under transition conditions. In addition, the TTF for Hg has completely different characteristics. The transfer time constant of Hg is extremely long or infinite. Thus TTF is appropriate concept for Hg transfer in the marine ecosystem, while the TTF concept is not useful for Cs.

Answer:

The trophic transfer factor (TTF) is an important parameter allowing us to compare the radionuclide concentrations in predator and its corresponding preys, and to see if it has any tendency for biomagnification along the trophic chain or not. For the radiocesium, this paper is not the first to use this concept, previous studies (Wang et al., 2000; Zhao et al., 2001; Mathews and Fisher, 2008, Harmelin-Vivien et al., 2012 ...) have already calculated this parameter (TTF) and used it to discuss the possibilities of biomagnification in different marine trophic chains.

Indeed, it is certainly not our intention to say that the biomagnification of ^{137}Cs is comparable to Hg that shows the greatest biomagnification factor.

9522 17:

Question:

Add JODC data archive location in the web.

Answer:

JODC data archive location in the web is added as follow:

The JODC (Japan Oceanographic Data Center) dataset is available at: http://www.jodc.go.jp

9523 23:

Question:

The MEXT reference should be replaced by Kaeriyama et al., 2014.

Answer:

MEXT(2014) is replaced by Kaeriyama et al., 2014 in the text, the figures captions and in the bibliography list.

Fig.3 :

Question:

Unify the case of letters in fig (A – F) and in figure caption (a - f). Add to the legend a mention of the three different taxonomic compositions (ZS,ZL, ZP?) in sub-fig B, D, F.

Answer:

the letters : "a,b,c,d,e,f " in the caption are replaced by "A,B,C,D,E,F" to unify them with the letters showed in the subfigures. A legend is added for the taxonomic composition.

Fig.4 :

Question:

Reference Kaeriyama et al., (2014) should be cited in the legend as a data source.

Answer:

Kaeriyama et al.(2014) is added at the end of the figure caption.

Fig.5 :

Question:

The parameters on the X-axes are small and are unreadable. Add plankton composition PS, PL, ZS, ZL, ZP to the legend.

Answer:

The X-axis font size is enlarged. Titles are added to the subfigures.

Fig6. :

Question:

Unify the case of letters in fig (A – D) and in figure caption (a - d). Show the unit (Bq kg d.w.-1) for the contour legend. For sub-figures B and C, are they not Buesseler et al. (2012) and C, Kitamura et al., (2013), respectively ?

Answer:

This figure has been replaced by another one containing 6 subfigures (A, B, C, D, E, F) and taking into account more field observations. The data are presented in Bq kg^{-1} ww as described in the figure caption.

Fig. 8 :

Question:

Missing (PS)(ZL)(ZP) in "… small phytoplankton (PS), large zooplankton (ZL) and predatory zooplankton (ZP) in the ….". The scale of the Y-axis in subfigure ZP is different from the others. Unify the Y-scales of all subfigures.

Answer:

(PS), (ZS) and (ZP) are added to their corresponding names in the caption text. "large zooplankton" is replaced by "small zooplankton" in the caption. The subfigures Y-scales are unified.

Fig. 11:

Question:

Y-scales in subfigure ZP are different. Unify the whole study area and 0-30km from FNPP.

Answer:

The Y-scales are unified for all sub-figures.

Fig.12:

Question:

What do the red bars and marks on the two figures represent?

Answer:

On each box, the central mark is the median, the edges of the box are the 25th and 75th percentiles, the whiskers extend to the most extreme data points not considered outliers, and outliers are plotted individually (the red marks).

This has been added in the figure caption