Anonymous reviewer 3

3.1) Page 1524, line 24-28 (the end of section 2.3): To agree with the authors' comments on sensitivity of the results to the number of released particles, it is good to add one figure or table showing this tendency.

We have added the following quantitative assessment of the sensitivity to the revised paper (see Appendix 2):
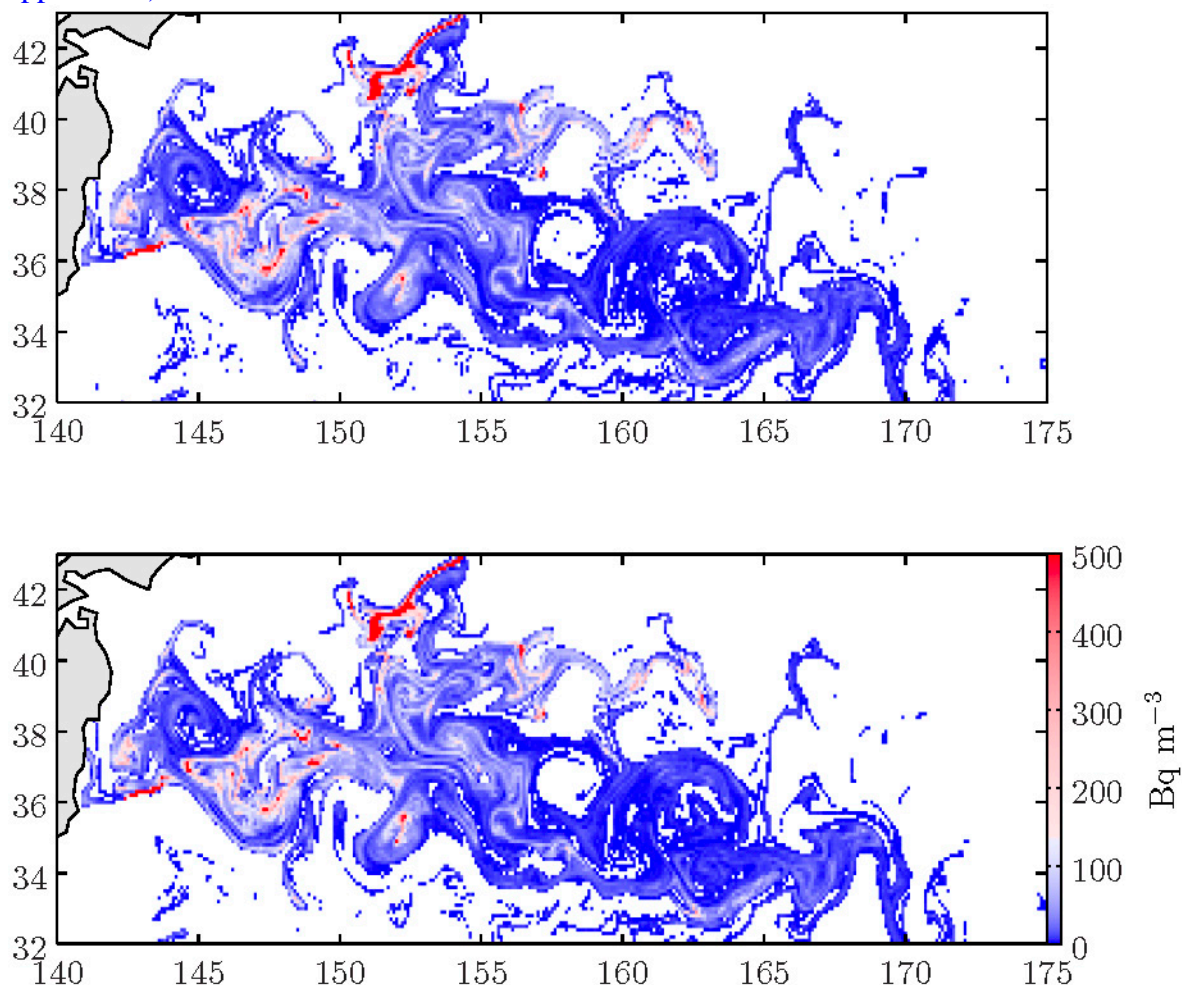

Fig. 12. Modeled ^{137}Cs concentration within the mixed layer on June 20 computed using 2*10^7 particles (top) and 4*10^7 particles (bottom). Colors, source domain and source strength as in Fig. 5. ^{137}Cs concentration values were cut off at the detection limit (1.5 Bq m^{-3}).

"To illustrate the sensitivity of our results to the number particles released, Figure 12 shows the comparison between the simulated mixed-layer ^{137}Cs concentrations from the direct discharge source on June 20 (the end of our numerical run) computed using different numbers of released water parcels. The upper panel with 2 x 10^7 particles is the baseline calculation as this number of released particles that was used for the results presented in the text. The lower panel has twice the number of particles, 4 x 10^7 particles. The distributions of ^{137}Cs are similar in both cases and

the concentration values (c_1 and c_2) differ, on average, by only 8.4%. The correlation, $\langle 2c_{1*}c_2/(c_1{}^2 + c_1{}^2)\rangle$, between the two cases is also high and, ignoring ^{137}Cs concentration values below the detection limit, is equal to 95.2%."

3.2) Page 1526, line 28 – Page 1527, line 4 (the second paragraph of section 3.2): This mixed-layer depth argument is problematic in the following two points. First, the authors mentioned that some part of Cs-137 is transported to the layer below the mixed-layer, which is no longer contributing to the surface (or mixed-layer average) concentration of Cs-137. However, the authors use a constant value for the mixed-layer depth during the whole period of calculation. In this case, the above detrainment effect is not included, resulting in increase of surface Cs-137 concentration. Second, as the authors also mentioned in the text, the mixed-layer depth gets shallower from March to June. If we use the constant mixed-layer depth of March or April throughout the calculation, this creates underestimate tendency of the surface Cs-137 concentration, simply because Zml is overestimated. Relatively large number of the source amplitude, especially for direct discharge component of 16.2 PBq, seems to be related to this uncertainty of the mixed-layer specification. Do the results differ with different values for the mixed-layer depth? How sensitive the results are to the Zml values?

We agree with the reviewer that converting 2d Cs concentration (Bq/m^2) resulting from the observation-based model to 3d mixed layer concentrations (Bq/m^3) by dividing by the April (May for the atmospheric source) mixed layer depths, z_{mld}, is an approximation. We also agree that the resulting surface concentrations, c_{surf}, depend on the choice of z_{mld}. The value of c_{surf} is inversely proportional to z_{ML} and thus when z_{ML} is half as deep, c_{surf} is doubled. We have clarified this dependency in the revised paper (see page 10).

That said, we think that because of the irreversible nature of the vertical mixing, Cs that is mixed down to the bottom of the April mixed layer, does not re-concentrate in the surface ocean even when the mixed layer shoals later in spring. Thus, in situations with temporally-changing mixed layer depths, such as the western Pacific waters in late spring, it is the deepest mixed layers that should be used for the conversion. Since the peaks of the atmospheric and oceanic sources occurred on March 18 and April 7, respectively, we use the March/April z_{ML} values for estimating surface concentrations of ^{137}Cs from the atmospheric/oceanic sources. We have extended the discussion of this issue in the revised manuscript (see pages 10-11).

3.3) Page 1529, line 22 – Page 1530, line 7 (the first paragraph of section 3.3): The source estimations are performed with only the KOK observations. There are some more Cs-137 concentration values observed by TEPCO and MEXT, Japan. Why these other data are not utilized for the error minimization procedure? If these data are used as well as the KOK observations, how do the results change? I think the near coastal values, with the direct discharge source magnitude of 16.2 PBq, may be overestimated. Comparisons with other observed data should be conducted.

The focus of our paper is the KOK cruise dataset, and the main goal is to provide the physical oceanographic context for the measured Cs distribution (Fig. 1(b)) and to investigate the processes that influence the spreading of Cs into the broader Pacific Ocean over a time interval

of several months. Comparison with, and interpretation of, other available measurements (including the earlier near-shore measurements mentioned by the reviewer) is interesting but lies beyond the scope of the current paper. This is stated in section 5 of the revised manuscript:

"In this research we have focused on … the assessment and interpretation of the KOK observations. … comparison and interpretation of other available measurements, are subjects of future study"

Note also that because of the coarse temporal and spatial resolution, our observation-based oceanographic model might not be well-suited for simulating the earlier small-scale near-shore distribution of Cs. For these reasons comparison with earlier near-shore Cs measurements may be misleading. The deficiencies of our modeling, including the resolution issue, are clearly stated in section 5 of the revised manuscript.

Lastly, we would like to stress that the source amplitude estimation (section 3.3) is not the only result of our paper. Even without the quantitative assessment of the source strengths, the importance of the Kuroshio Current and near-shore eddies in the dispersal of Fukushima radionuclides, as well as the relatively minor role played by the vertical velocity and vertical shear are, in our mind, just as interesting as the source strength estimation. Another important finding is the general utility of the observation-based model based on AVISO and NCEP in realistically simulating the dispersal of passive tracers in the ocean over a time interval of several months.

3.4) Fig.8: I do not understand the meaning of line-connected figure. Instead, I suggest to make a scatter diagram of the observed vs simulated values for this kind of point-to-point comparison.

The intention of Fig 8 is to show that the two curves are in qualitative agreement with each other, showing minima/maxima at same stations (i.e., at same geographical locations). We have added this sentence to the discussion of Fig. 8 in the revised paper (see page 16). We agree with the reviewer that a scatter diagram would be an alternative way to illustrate the same point, but another reviewer has specifically asked us to include a station-by-station comparison between the measured and simulated Cs concentrations, therefore Fig. 8 will be included in the revised paper.

Comments by G. Hong

Rypina et al.'s manuscript addresses the dispersal of the introduced dissolved substances based on the field data collected from the scientifically well designed field sampling campaign and state of the art numerical modeling of the water movement in the ocean and its conservative analysis of the model outcomes. This manuscript demonstrates that the basic oceanographic research is very useful to understand the scene of the aftermath of the nuclear power plants located in the coastline in the world, if the accident occurs.

Number of important findings, such as dissolved substance is not readily entrained into the core of the eddy and strong ocean current can form an effective barrier for the dispersal of dissolved substance, should be recognized and will lead to further scientific inquiry.

Operators of the seashore based nuclear power plants or other hazardous installations should invest to the probable dispersal pattern if accidental discharge of dissolved substances from the plants using this manuscript as a model. Therefore, this manuscript deserves a close scrutiny for the practical point of view as well as scientific point of view.

A few points may be offered to the authors to providing balanced view on their data.

H1) The distribution of 137Cs in the particulate phase should be mentioned although it is very small and may not affect the model results.

We agree with the reviewer and added the following phrase to clarify this issue: *"Our model does not take into account the small fraction of Cs in the particulate phase."*

H2) Page 1530 Line 5. The manuscript says "As a large part of the initial fallout distribution of 137Cs lies south of the Kuroshio," but a number of literatures said a large part of the atmospheric emission directed to the northeast, that is, north of the Kuroshio (e.g., Lozano et al., 2011. Environmental International; Stohl et al., 2012; and others). If it is true, then, the authors should provide the reference to it.

We see now that the phrase "a large part" may be confusing. We thank the reviewer for pointing this out. In the revised manuscript, the sentence in question has been clarified and now reads *"Unlike the oceanic source that lies entirely to the north of Kuroshio, the southern edge of the initial fallout distribution of ^{137}Cs lies to the south of the Kuroshio (Fig. 2(b))."*

To further illustrate this point, we have added the mean Kuroshio position to Fig. 2(b).

H3) Page 1532 Line 1. If the authors could elaborate the mechanism of why dissolved substances are not mixed into the interior of the eddy for about 3 months time periods. That would be very useful for further studies.

This is an excellent question and a topic of an ongoing research. We have added the following discussion of this issue to the paper:

"Fluid exchange processes are different at the perimeter of an eddy compared to its core. At eddy perimeters the so-called "chaotic advection" mechanism is at play (Mancho et al., 2006; Rypina et al., 2010), which leads to the sequential stretching and folding of fluid elements and facilitates the exchange with the outer waters. Within the Lagrangian core of an eddy, on the other hand, a more "regular" state of motion exists where the behavior of fluid parcels is similar to that found in steady flows. Similar to the steady case, fluid parcels near the core follow paths around the center of the eddy without much exchange with outside fluid. This is why eddies are generally able to transport the water in their cores over long distances without "spilling" it along the way (Chelton et al., 2007; Early et al., 2011; Beron-Vera et al. 2013). The ability of oceanic eddies to trap fluid in their Lagrangian cores has been a subject of an ongoing research."

H4) Page 1532 Line 9: "homogenization". Homogenization may be an over simplification of the reality. The chemical composition of the seawater in the ocean is rarely homogeneous over this large area over several degrees in latitude and longitude. Therefore the sentence needs to be corrected.

We agree with the reviewer that the word "homogenization" is an oversimplification and can be misleading here. We have corrected the sentence as follows: *" … our modeling results suggest that the distribution of ^{137}Cs in mid-June was still filamentary and that the diffusion processes had not yet wiped away the sharp gradients in the ^{137}Cs field."*

H5) Page 1532 Line 10: "station by station comparison": The discrepancy between measured and simulated values becomes greater after station 28 to 52. A little bit of elaboration on this feature need to be addressed in the manuscript.

We have added the following discussion of this issue to the revised paper (on p. 16).

"The two curves in Figure 8 are in qualitative agreement with each other, showing minima/maxima at same stations (i.e., at same geographical locations). The quantitative agreement is better for stations 1-27 than for the near-shore stations 28-51. This is plausibly because the distribution is extremely filamentary close to shore, more so than for the off-shore stations. ^{137}Cs concentrations in the near-shore area span several orders of magnitude, with very sharp changes in concentration from one station location to the next (Figure 1b). Our observation-based model is not able to quantitatively match all of this variability due to its coarse spatial and temporal resolution, and the oversimplified representation of diffusion processes. Despite some quantitative mismatch, the model is still useful in explaining the qualitative features of the observed Cs distribution."

H6) Minor points Page 1524 Line 16: In the manuscript, half-life of 137Cs was given as 30.16 yr, however, other literature says 30.05 (e.g., Monographie BIPM-Comments Vol1-4,

2008) or 30.02 yr (Table of Radioactive Isotopes by E Browne and R Firestone, 1986, Wiley- Interscience). Although the difference may not change the outcome of the manuscript, it is still useful for some other purposes.

We thank the reviewer for this comment and have added a footnote about this in the revised paper: *"Slightly different half-life values of 30.05 and 30.02 years can be found in the Monographie BIPM-Comments, Vol1-4 (2008) and Browne et al. (1986), respectively, but these small differences are not relevant to the time-scales investigated here."*

H7) Page 1526 Line 28: "Note that once 137Cs is mixed down to a certain depth, it gets advected laterally by currents at depth and does not collect back to shallower depths as the mixed layer shoals." This sentence may need to be rephrased to assist the reader to understand the underlying physics. Such as "Once 137Cs is mixed down to a certain depth, and the 137Cs at that depth will remain if the mixed layer becomes shallower than that depth, due to heating at the sea surface (?). And the 137Cs at that depth gets advected by currents at that depth dispersed through isopycnal mixing in lateral direction as well as vertically."

We have clarified the text in question as follows (see p. 10-11):
"Due to the rapid warming and restratification of surface waters in late spring, the mixed layer depth over the western Pacific Ocean in April/June is roughly half/one-fifth the March value. … In situations with temporally-changing mixed layer depths, such as the western Pacific waters in late spring, it is the deepest mixed layers that should be used for the conversion, because once ^{137}Cs has mixed down, it cannot un-mix and re-concentrate in the surface ocean even when the mixed layer shoals later in spring. Additionally, this particular area is subject to little upwelling so fluid parcels generally do not rise up through the water column from depth. Rather, subsurface Cs left below the bottom of the receding mixed layer will be advected by subsurface currents at depth and will flow, to a first approximation, along subsurface isopycnal surfaces. Since the peaks of the atmospheric and oceanic sources occurred on March 18 and April 7, respectively, we use the March/April z_{ML} values in modeling the spreading of ^{137}Cs from the atmospheric/oceanic sources."

H8) Sampling stations are not found in the figures. Therefore, appropriate station map showing all the oceanographic sampling station numbers is needed.

We have added a figure with station numbers (Figure 1(d)).

H7) Figure 6: Location of the Kuroshio may be inserted for the readers to see the concentrations at certain locations in relation to the position of the Kuroshio.

We have added the location of the Kuroshio Current to fig. 6.

Comments by P.p.p Povinec (Editor)

E1) Background - As this is a modelling paper you should introduce, and critically discuss previous papers published on a similar topic – later you can explain why there are such big differences in the 137Cs marine source term - References on some previous papers are missing , eg. Kawamura et al., 2011, Journal of Nuclear…; Tsumune et al., 2013, Biogeosciences, the same issue...etc.

We thank the editor for pointing out relevant references, all of which have been included and discussed in the revised paper. To put our work in the context with prior studies, we have added the following paragraph at the beginning of Sec. 2.3:

"Over the last two years there have been a number of numerical modeling studies aimed at assessing the aftermath of the Fukushima disaster (e.g., Tsumane et al., 2011, 2013; Kawamura et al., 2011; Estournel et al., 2012; Miyazawa et al, 2012, Masumoto et al., 2012; Bailly du Bois et al., 2012). Forward numerical models have allowed simulation of the spread of radioactive waters into the ocean. Inverse modeling efforts have been useful in constraining the source amplitudes of both the oceanic direct discharge and atmospheric fallout sources. The main goals of this investigation differ from this prior work in two respects. First, we focus on providing the physical oceanographic interpretation of the patterns observed by the KOK survey (Figure 1b). Second, we investigate the general utility of data-based velocity fields for realistically reproducing the dispersal of passive tracers in the ocean over a time scale of several months."

E2) 3.3 Source-term amplitude - eq. (1) – this is the crucial part of your paper – you should present results in the form of graphs (both for marine and atm. releases) - we need to see the minima which you describe in the text!

In the revised manuscript, we show the error function for 50 cases of the bootstrapping method that we have used to define the mean and STD of the source amplitude term. To make the text more streamlined, we decided to place this figure along with the corresponding discussion into the Appendix 1.
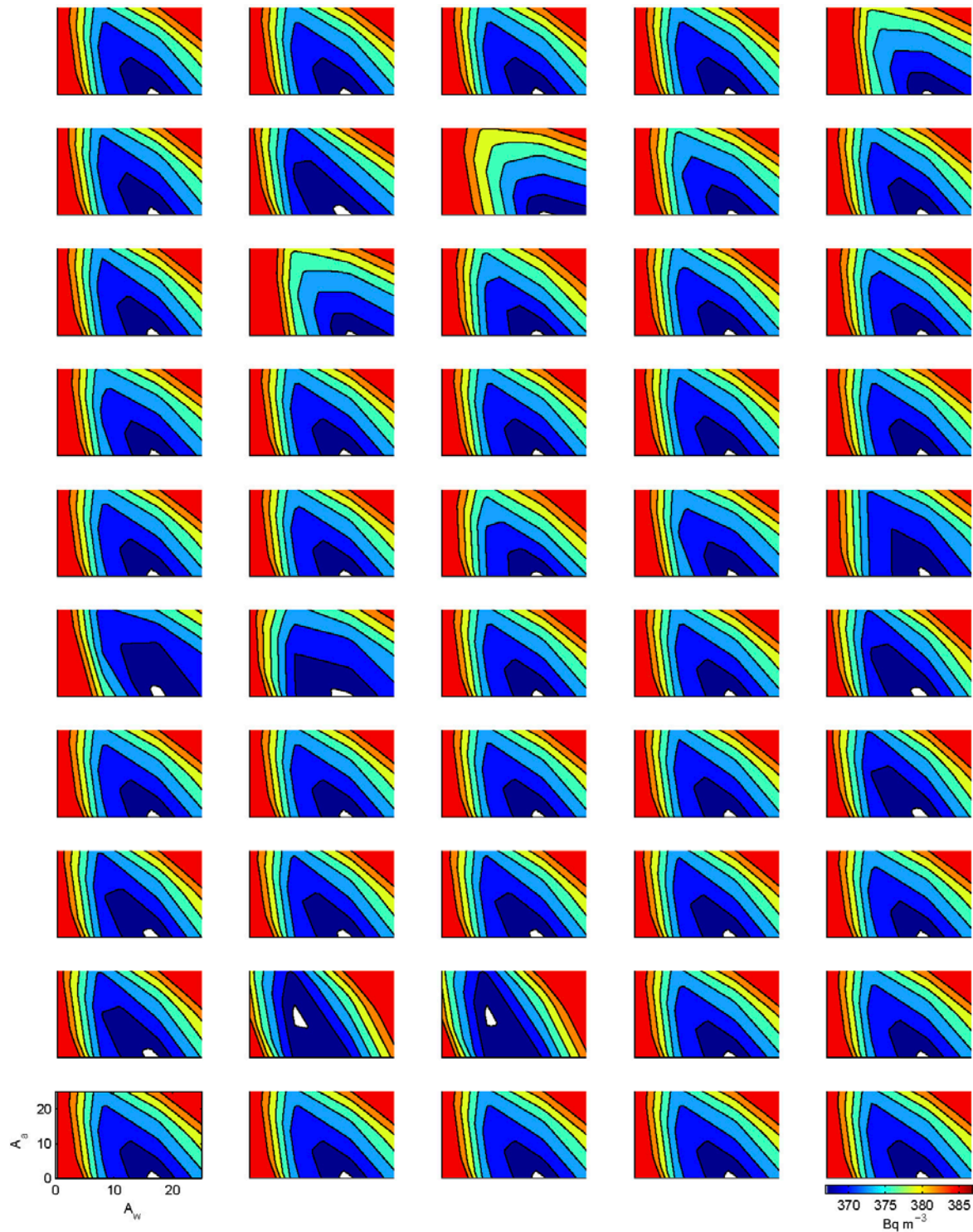
Fig. 11. Error function *Er* from Eq. (1) as a function of the oceanic and atmospheric source strengths, A_{atm} and A_{ocean}, for 50 cases resulting from the bootstrapping procedure of sequentially eliminating one station at a time. The position of the minimum is indicated as a small white region in each panel. Colorbars are slightly different for different subplots. To put the mean error values in perspective, the mean measured Cs concentration averaged over all stations is 415 Bq m^{-3}.

E3) Fig. 9: - Why there is very good agreement between the theoretical and experimental data at the beginning, but later your model is predicting deep minima?

The detection limit for ^{137}Cs is 1.5 Bq m^{-3} (Buesseler et al., 2012) so in the revised paper we have cut off our simulated ^{137}Cs concentrations at the same level for a fair comparison. Note also that unlike our simulated concentrations, which correspond to Fukushima-derived ^{137}Cs, the measured values include both Fukushima-derived and pre-existing ^{137}Cs (the latter is at levels of 1-2 Bq m^{-3} (Buesseler et al., 2011; 2012)). This explanation has been added to the revised paper on p. 16.

Stations 28-52 correspond to the more densely-sampled near-shore area. The distribution of Cs here is extremely filamentary here, even more so than in the far field. Cs concentrations in the near-shore area span several orders of magnitude, with very sharp changes in concentration from one station location to the next (Fig 1(b)). Our observation-based model is not able to quantitatively match all of this variability due to its coarse spatial and temporal resolution, and the oversimplified representation of the diffusion processes. Despite this quantitative mismatch, the model is still useful in explaining the qualitative features of the observed Cs distribution such as the maximum associated with the perimeter of the eddy and intermediate concentrations at stations 33-37.

We have added the following discussion of this issue to the revised paper.

"The station-by-station comparison between the measured and simulated ^{137}Cs concentrations at the KOK stations is shown in Figure 8. Here, the detection limit for ^{137}Cs is 1.5 Bq m^{-3} (Buesseler et al., 2012) so we have cut off our simulated ^{137}Cs concentrations at the same level for a fair comparison. Note also that unlike our simulated concentrations, which correspond to Fukushima-derived ^{137}Cs, the measured values include both Fukushima-derived and pre-existing ^{137}Cs (the latter is at levels of 1-2 Bq m^{-3} (Buesseler et al., 2011; 2012)). The two curves in Figure 8 are in qualitative agreement with each other, showing minima/maxima at same stations (i.e., at same geographical locations). The quantitative agreement is better for stations 1-27 than for the near-shore stations 28-51. This is plausibly because the distribution is extremely filamentary close to shore, more so than for the off-shore stations. ^{137}Cs concentrations in the near-shore area span several orders of magnitude, with very sharp changes in concentration from one station location to the next (Figure 1b). Our observation-based model is not able to quantitatively match all of this variability due to its coarse spatial and temporal resolution, and the oversimplified representation of diffusion processes. Despite some quantitative mismatch, the model is still useful in explaining the qualitative features of the observed Cs distribution."

E4) (pls. change KBq/m3 to kBq/m3)

We thank the editor for this comment and have made the change in the revised manuscript.

E5) Pls. prepare a finale version of the manuscript taking into account all comments posted on your paper.

We have revised our paper to address all comments.