Biogeosciences
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Interactive comment on "Ocean dynamic processes causing spatially heterogeneous distribution of sedimentary caesium-137 massively released from the Fukushima Dai-ichi Nuclear Power Plant" *by* H. Higashi et al.

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Received and published: 5 October 2015

We are grateful to the referee for her/his comments that help us to improve our manuscript. Our responses are described below.

Referee #3 comment:

The authors employed complex model for Cs-137 concentration in seawater and sediment to represent the spatially heterogeneous distribution in sediment. I don't agree

with their results of total amount of Cs-137 in sediment which is 10 times larger than previous estimated value based on observation. Main problem is that they only considered sedimentation and resuspension process in a similar manner to Europe. In Fukushima case, very high concentration of Cs-137 passed through on the sediment in the earlier period. Therefore, absorption and desorption process on sediment is dominant (Otosaka and Kobayashi, 2013). Sediment properties are a major factor of Cs-137 on absorption and desorption process. They did not considered these dominant processes. They simulated the sedimentation rates of Cs-137. The sedimentation rates were observed by sediment trap (Honda et al., Biogeosciences, 2013; Buesseler et al., ES & T, 2015). They should validate the sedimentation rates in comparison with observed data if they believe that sedimentation process is dominant. I think their simulated sedimentation rates are overestimated to observed value. If they simulate more than 2 or 3 years, difference between observation and their simulation is getting larger. Because their model focus on sedimentation process which is not dominant in Fukushima case.

Response:

Our model definitely includes the adsorption process of dissolved ^{137}Cs in bottom seawater to surface sediment, although this description was omitted in our original manuscript. In our model, exchange of dissolved ^{137}Cs between bottom seawater (Eq. (1)) and surface seabed (Eq. (5)) was calculated from diffusion equation. Diffusion coefficient was derived from bottom seawater turbulent and sedimentary bioturbation. Adsorption of dissolved ^{137}Cs in bottom seawater to surface sediment could occur through the diffusion process in our model. When high concentration of the dissolved ^{137}Cs passed through just above the seabed, the dissolved ^{137}Cs infiltrated into sediment porewater through the diffusion process, and was adsorbed to sediment particles afterward. We will add these explanations to Sects. 2.3 and 2.4 as a below section of Revise #3-1.

We consider that "absorption and desorption process on sediment is dominant (Oto-

saka and Kobayashi, 2013)" in the referee comment should be limited within shallow regions. Indeed, Otosaka and Kobayashi (2013) described "Higher levels of sedimentary radiocesium in the shallow regions are attributable to a higher contact probability of dissolved radiocesium with sediment as well as the efficient vertical transport of radiocesium to the deeper layer of sediment via bioturbation" as their second conclusion. They estimated amunts of dissolved- and particulate-^{137}Cs southwardly-transported to their study field (the coastal area of north Ibaraki) using available surface seawater data (the 4–5th paragraphs in "Discussion" of Otosaka and Kobayashi, 2013). Their estimated results indicated that lateral flux of the dissolved ^{137}Cs in the seawater was dominant rather than that of the particulate phase. Hence, they would reach the aforementioned conclusion. However, they did not discuss about sedimentation process of the horizontally-transported dissolved-^{137}Cs from the surface seawater to the seabed. We consider that the ^{137}Cs downward transport probably included the adsorption to particulate matter, which was suspended from seabed in their study field, and subsequent sinking. The direct adsorption of dissolved ^{137}Cs to sediment as described by the referee also occurred in their field because surface and bottom seawater can be easily mixed by oceanic turbulence in shallow regions, as shown in Fig. 12b of our manuscript. However, in hotspot swath of our target area located on offshore of shelf break (50–100 m depths), our simulation indicated that the dissolved-^{137}Cs horizontally-transported from the shallow region did not reach the bottom by vertical mixing as shown in Fig. 12g. There, horizontal transport of particulate ^{137}Cs that was suspended from the shallow seabed is probably dominant for ^{137}Cs sedimentation as described in Sect. 4.3 of our manuscript (we will add the similar sentence to our manuscript as a below section Revise #3-2). Indeed, Otosaka and Kobayashi (2013) also reached the conclusion (their 4th conclusion) of "resuspension and lateral transport of the fine-grained sediments plays an important role to redistribute the sedimentary radiocesium" through their observations.

As pointed out by the referee, we also consider that total amount of sedimentary ^{137}Cs should be quantitatively validated more. However, we could not do adequately at the

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present, although we attempted to validate that by comparing between simulated and observed vertical sedimentary ^{137}Cs profiles as mentioned in Sect. 3.3. One of the reasons was a lack of observations, necessary for the validation, after the accident in the deeper sediment, especially near 1FNPP where massive sedimentary ^{137}Cs remains even today. Meanwhile, about the sediment trap measurements by Honda et al. (2013) and Buesseler et al. (2015) as described by the referee, their sites were located on far offshore out of our target area, unfortunately. Another reason why we could not validate was uncertainty related to our simulation conditions. As mentioned in Sect. 2.4, because we used the ideal sediment assumption, our simulation would overestimate sedimentary suspension unless actual seabed mainly consisted of fine particles. In addition, our results included uncertainty of ^{137}Cs inflow conditions, especially atmospheric deposition– which will be described in Sect. 2.2 of our manuscript (please refer to our response to RC C4982 of Reviewer #2 comment). It goes without saying that the total amount of sedimentary ^{137}Cs directly depends on that of ^{137}Cs inflow.

We consider that the aforementioned quantitative uncertainty included in our results should be described in the manuscript briefly and more clearly. Hence, we will revise our manuscript (abstract, discussion, and conclusions, etc.) as a below section Revise #3-3. Especially, because the total amount of sedimentary ^{137}Cs would include relatively large uncertainty, its description in the abstract will be deleted. We will largely revise Sect. 4.1 to describe that our result of the total sedimentary ^{137}Cs amount included uncertainty.

Although our simulation includes the aforementioned quantitative uncertainties, we consider that sequential ocean dynamic processes of ^{137}Cs accumulation in the hotspot swath (as described in the first paragraph in Sect. 4.4) would be qualitatively reasonable at least. This is because ^{137}Cs accumulation in the hotspot swath is governed mainly by ocean dynamics: spatiotemporal variation of bottom shear stress. That is, the quantitative uncertainty in our simulation conditions would affect amounts of suspension and subsequent horizontal transport of sedimentary ^{137}Cs on shallow shelf,

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but not ^{137}Cs accumulating location in the offshore. Besides, our simulation reproduced several sedimentary ^{137}Cs characteristics found in the observations, and indicated new reasonable findings of the causing ocean dynamics, i.e., the spring-neap tidal variation would cause decrease trend in the sediment-surface ^{137}Cs near shore region (as described in Sect. 4.2 of our manuscript). In discussion and conclusions of the revised manuscript, those will be also described as the below section Revise #3-3.

Revise #3-1: (Bold type shows revised sentences.)

(P. 12725, L1 – L5, Sect. 2.3)

At the bottom boundaries in the seawater ^{137}Cs simulation, **diffusion flux of the dissolved ^{137}Cs and** sedimentation/suspension fluxes of the particulate ^{137}Cs and suspended particulate matter, which were evaluated by the sediment ^{137}Cs model described in Sect. 2.4, were specified. At the lateral boundaries, the three variables in the Region-1 simulation were set to zero. We used the hourly Region-1 results in the Region-2 domain.

(P. 12728, L4 – L13, Sect. 2.4)

Net flux of suspended particulate matter/particulate ^{137}Cs evaluated from Eqs. (9)/(12) and (10)/(13) was given as surface seabed condition of Eq. (7) (but unnecessary)/(6) and bottom boundary of Eq. (3)/(2). Exchange of dissolved ^{137}Cs between bottom seawater (Eq. (1)) and surface seabed (Eq. (5)) was calculated from diffusion equation. Diffusion coefficient was derived from bottom seawater turbulent and sedimentary bioturbation. Adsorption of dissolved ^{137}Cs in bottom seawater to surface sediment could occur through the diffusion process in our model. At the bottom boundaries of Eqs. (5) and (6), we specified flux conditions as advection-outflow at the sedimentation, or zero-inflow at the suspension if deeper

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^{137}Cs activity was assumed zero. These fluxes were consistent with mass balance of the particulate matter expressed by the identical Eq. (7). However, there is an issue in the method, in that Eq. (7) cannot be restricted to the suspended amount of sediment matter, i.e., it is possible that the fine particulate matter is infinitely and endlessly supplied from deeper levels. Therefore, the present procedure using the relative vertical-axis z' and uniform sediment assumption, which facilitates simulation of the vertical profile of sedimentary ^{137}Cs, probably overestimates the suspension in regions whose seabed does not actually have sufficient suspendable particulate matter.

Revise #3-2: (Bold type shows revised sentences.)

(P. 12737, L16 – L28, Sect. 4.3)

In contrast to the shallow region, in the offshore region along the shelf break (50–200 m depths), impacts of the tide and strong wind on the bottom disturbance were much weaker (< 50 m depths). Even the extratropical cyclone that caused the strong bottom disturbance in the shallow region at the end of May could not increase bottom friction beyond the critical shear stress (Figs. 10c and 12f), so little sediment was suspended (Figs. 11c and 12h). **Although strong vertical mixing then occurred in seawater, dissolved ^{137}Cs activity in bottom seawater did not rise (Fig. 12g) in contrast to the shallow result (Fig. 12b).** Nevertheless, sedimentary ^{137}Cs activities in the offshore region began to increase significantly just after that strong wind event (MEXT stations C3, E1, D1, G0, G1, I0, I1, J1 in Fig. 6, and E1 in Fig. 12j). This increase in sedimentary ^{137}Cs resulted from sedimentation of particulate ^{137}Cs (Fig. 12i), which was suspended and horizontally transported into and from the adjacent shallow shelf by the wind event. This horizontal transport is supported by the fact that both concentrations of suspended matter and particulate ^{137}Cs suddenly increased in the upper seawater, without bottom suspension or upward diffusion (Fig. 12h and i).

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Revise #3-3: (Bold type shows revised sentences.)

(P. 12715, L3 – L9, Abstract)

(The 3rd paragraph in the abstract was deleted.)

(P. 12718, L17 – P. 12719, L10, Sect. 1)

Our previous study (Higashi et al., 2014) developed a comprehensive model for simulating oceanic ^{137}Cs behaviour in both seawater and seabed, with consideration of vertical ^{137}Cs transport in the sediment. We then roughly assumed that sediment matter in the entire simulation domain had ideal properties such as identical bulk density, uniform porosity, and particle aggregates of a single grain diameter. **The reason why we used this assumption was not only that spatiotemporal variation of the sediment properties just after the tsunami disturbance was unknown but also that the assumption enabled direct simulation of vertical ^{137}Cs behaviour in the sediment.** This type of assumption has also been used in other models (Kobayashi et al., 2007; Choi et al., 2012), except for the ^{137}Cs behaviour in sediment. Our earlier simulations using the developed model agreed reasonably well with the sampling of ^{137}Cs activity in both seawater and sediment off east Japan in the Pacific during March and December 2011. However, we could not effectively simulate the heterogeneous sedimentary ^{137}Cs distribution, mainly because of a lack of spatial resolution.

We performed a downscaling simulation of oceanic ^{137}Cs behaviour using the usual one-way nesting method to resolve the heterogeneous sedimentary ^{137}Cs distribution, especially in the hotspot swath. **The present simulation also employed the aforementioned assumption of the ideal sediment properties in entire domain.** The model and the numerical procedure are described in Sect. 2. Simulated results of the spatiotemporal ^{137}Cs distributions in seawater and sediment within the nested region are shown in Sect. 3 as compared with observations, to evaluate our model

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performance. In Sect. 4, we discuss ocean dynamic processes causing the spatially heterogeneous distribution, especially in the hotspot swath, **including our model uncertainties**.

(P. 12733, L10 – P. 12734, L28, Sect. 4.1)

4.1 **Total amount of sedimentary ^{137}Cs and its uncertainty**

The total ^{137}Cs amount rapidly increased at the beginning of April in our simulation because of atmospheric deposition and direct discharge from 1FNPP (Fig. 3). After that, it stabilized at $\sim$ 12 PBq by the end of May, and strongly declined to 4.3 PBq at the end of 2011. The latter decrease was caused by the seawater ^{137}Cs dispersed from Region-2 to the open ocean. Sedimentary ^{137}Cs also increased steadily until onset of the significant seawater dispersion, but suddenly declined at the end of May. This rapid decrease resulted from short but strong suspension induced by an extratropical cyclone that originated as typhoon 201102 (SONGDA) and passed over the southern part of Region-2. Afterward, the sedimentary ^{137}Cs rapidly recovered, indicating that the suspended ^{137}Cs returned to the sediment. **Such behaviours of sedimentary ^{137}Cs before and after the cyclone were also simulated by Choi et al. (2013).**

In our simulation, total sedimentary ^{137}Cs amount was 0.10 PBq (0.66% of total ^{137}Cs inflow) in the upper 3 cm layer, 0.40 PBq (2.6% of total ^{137}Cs inflow) in the upper 10 cm layer, and 3.2 PBq (21% of total ^{137}Cs inflow) in the entire seabed over all of Region-2 (1.4 $\times$ 10^5 km^2) at the end of 2011. Kusakabe et al. (2013) estimated 0.042–0.052 PBq of total sedimentary ^{137}Cs between September and December 2011 in the upper 3 cm seabed off Miyagi, Fukushima, and Ibaraki prefectures (2.2 $\times$ 10^4 km^2 domain) on the basis of the MEXT (2011) observations. Otosaka and Kato (2014) estimated total sedimentary ^{134}Cs **regarded as almost equivalent to ^{137}Cs amount** at 0.20 $\pm$ 0.06 PBq (decay-corrected to 11 March 2011) within the region less than 200 m depth (1.5 $\times$ 10^4 km^2 domain) in October 2011 **using their sam-**

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pling data in upper 10 cm sediments and the MEXT observations. Accounting for the difference in study area, our results of the sedimentary ^{137}Cs amounts in the upper layers were almost comparable to the two studies. However, the simulated amounts in the upper 3 and 10 cm layers were only 3 and 13% of the total sedimentary ^{137}Cs, respectively, while the remaining was present in the deeper sediment.

We could not adequately validate the simulated result of a large amount of ^{137}Cs in the deeper layer. One of the reasons was a lack of observations, necessary for the validation, after the accident in the deeper sediment, especially near 1FNPP where massive sedimentary ^{137}Cs remains even today, as described in Sect. 3.3. Although we described in Sect. 3.2 that the simulated concentrations of sediment-surface ^{137}Cs was roughly comparable with observations except for overestimation in the region northeast of 1FNPP, only this agreement could not indicate validation of ^{137}Cs migration flux from seawater to sediment. The insufficient of the deeper data also became the reason why the earlier estimations of the total sedimentary ^{137}Cs might be underestimated as described by their authors (Kusakabe et al., 2013; Otosaka and Kato, 2014). Indeed, recent surveys have detected high activity $O(10^3–10^4)$ Bq kg^{-1} ($= O(10^6–10^7)$ Bq m^{-3}) through the present, in both surface and lower (> 30 cm) sediment at several sampling stations near 1FNPP (Thornton et al., 2013; NRA, 2014a). Our simulation also revealed 1.0 PBq **(31% of the total sedimentary ^{137}Cs amount)** in a large amount of sedimentary ^{137}Cs in the 30 × 30 km square domain (140.88–141.21E, 37.29–37.56N in Fig. 9, **except for the land**) around 1FNPP at the end of 2011.

Another reason why we could not validate the sedimentary ^{137}Cs amount in the deeper layer was uncertainty related to our simulation conditions. As mentioned in Sect. 2.4, because we used the ideal sediment assumption, our simulation would overestimate sedimentary suspension unless actual seabed mainly consisted of fine particles. In fact, our simulation overestimated some sediment-

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surface ^{137}Cs activities in the region northeast of 1FNPP and Sendai Bay whose seabed was dominated by coarse sand, as mentioned in Sect. 3.2. The simulated amount of sedimentary ^{137}Cs in the 30 × 45 km rectangular region in Sendai Bay (141.03–141.37E, 37.71–38.11N in Fig. 9) reached 0.52 PBq {bf (16% of total sedimentary ^{137}Cs amount). **Furthermore, our result included uncertainty of ^{137}Cs inflow conditions, especially atmospheric deposition, as mentioned in Sect. 2.2. It goes without saying that the total amount of sedimentary ^{137}Cs directly depends on that of ^{137}Cs inflow.**

(P. 12737, L1 – L14, Sect. 4.2)

The bottom disturbance caused by the tide or strong wind did not occur in every shallow region (< 50 m depths) because of the seabed topography and other factors. In the narrow nearshore region from south Fukushima to north Ibaraki, bottom friction did not increase even during extratropical cyclone passage (Fig. 10c). The Otosaka and Kobayashi (2013) station O-S4, where the apparent downward movement of **sedimentary** ^{137}Cs was found in both observation and simulation (Fig. 8i), was just located in that region. This area was where the bottom disturbance rarely occurred; if anything, the sedimentation slightly dominated the suspension over a long period (Fig. 11d). This indicates that the apparent vertical transport of **sedimentary** ^{137}Cs found at station O-S4 was caused by relatively fresh suspended particulate matter settling on earlier sediment containing substantial ^{137}Cs. It is inconceivable that the amount of sedimentation over only several months became so large under the stable seabed condition. **Although this is probably caused by the uncertainty related to the ideal sediment assumption as mentioned in Sect. 4.1,** this may have been possible in the unstable **seabed** state just after the extraordinary disturbance of the tsunami.

(P. 12738, L11 – L24, Sect. 4.4)

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The hotspot swath in our simulation was just offshore of the shelf break (along the 50–100 m isobath) off southern Fukushima Prefecture through northern Sendai Bay at the end of 2011 (Fig. 5j). After the 1FNPP accident, the region of high sedimentary ^{137}Cs activity gradually expanded from south of 1FNPP to north in and around the shelf (< 100 m depths) by June (Fig. 5a–e). Afterward, in the shallow shelf (< 50 m depths), the sediment-surface ^{137}Cs significantly decreased because of the periodic tidal disturbance causing sediment suspension, horizontal transport in the seawater, and/or apparent downward movement in the seabed. Meanwhile, in the offshore region (50–100 m depths), the sedimentary ^{137}Cs that settled after being horizontally transported from the shallow region during the extratropical cyclone at the end of May remained largely stable, because of rare bottom disturbance. The present simulation suggests that these were the sequential processes causing the hotspot swath, and that its shape is closely related to spatiotemporal variation between bottom shear stress on the shallow shelf and that offshore of the shelf break. **Although our simulation includes quantitative uncertainty as mentioned in Sect. 4.1, these processes are qualitatively reasonable at least. This is because ^{137}Cs accumulation in the hotspot swath is governed mainly by ocean dynamics: spatiotemporal variation of bottom shear stress. That is, the quantitative uncertainty in our simulation conditions would affect amounts of suspension and subsequent horizontal transport of sedimentary ^{137}Cs on shallow shelf, but not ^{137}Cs accumulating location in the offshore.**

(P. 12739, L9 – L14, Sect. 5)

To clarify ocean dynamic processes causing the massive heterogeneous sedimentary ^{137}Cs distribution that persists in and around the shelf off Fukushima and adjacent prefectures, we numerically simulated oceanic ^{137}Cs behaviour for about 10 months after the 1FNPP accident. We succeeded in simulating such that distribution, especially the hotspot swath just offshore of the shelf break (along the 50–100 m isobath)

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shown by recent observations (Thornton et al., 2013; Ambe et al. 2014; NRA, 2014a), **although quantitative validations were not adequate. The result suggests that several spatiotemporal characteristics of the sedimentary ^{137}Cs would be produced by ocean dynamics.**

(P. 12740, L5 – L25, Sect. 5)

Our simulation also produced significant findings regarding sedimentary ^{137}Cs behaviour on the shallow shelf. There, the simulated bottom disturbance tended to occur frequently because of the periodic spring tide and occasional strong winds, steadily decreasing simulated sediment-surface ^{137}Cs per several observations. The simulation indicated that repeated bottom disturbances reducing sediment-surface ^{137}Cs over the long term caused sedimentary ^{137}Cs to not only be horizontally transported to the offshore region but also vertically toward deeper sediment. Consequently, in our simulation, relatively large amounts of ^{137}Cs in deeper sediment remained on the shallow shelf, especially near 1FNPP, even about 10 months after the 1FNPP accident. Hence, **total sedimentary ^{137}Cs at the end of 2011 reached 3.2 PBq, and 87% of that was present below 10 cm layers.** If our simulation **were** correct, ^{137}Cs in deeper sediment would be much greater than in upper sediment, and would remain stable over a long period. **However, the simulated sedimentary ^{137}Cs amount in the deeper layers would include relatively large uncertainty at the present. In future work, we will improve the model for quantitative simulation of the spatiotemporal variation of fine particulate matter in both seawater and sediment, and carry out long-term simulations including the tsunami disturbance to validate the model using recent observations of vertical sedimentary ^{137}Cs distribution.**

A manuscript that we will revise has been uploaded in PDF format as supplement file.

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Please also note the supplement to this comment:
http://www.biogeosciences-discuss.net/12/C6213/2015/bgd-12-C6213-2015-supplement.pdf

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