

We are grateful to reviewer #1 for the critical comments and useful suggestions that have helped us to improve our paper. As indicated in the responses that follow, we have taken all these comments and suggestions into account in the revised version of our paper.

01. page 4851 line 1: For precise expression, the title should be “Distribution of the Fukushima-derived radionuclides in seawater in the Pacific off the coast of Miyagi, Fukushima, and Ibaraki Prefectures, Japan from March 2011 to February 2012”

The title suggested by the reviewer#1 seems to be too long. We do not change the title.

02. page 4853 lines 5 - 19: Progression of the NDNPP accident is described in detail. But there are no references in the text. Please add necessary quotations.

Page 4853, line 5

We will revise “... **the Great East Japan Earthquake (M9.0) and ... of Japan (Japan Meteorological Agency, JMA, 2011a; JMA, 2011b).**”

We will add two references in the References.

JMA (Japan Meteorological Agency): "Report (No.14) of the 2011 off the Pacific coast of Tohoku Earthquake, 13th March 2011", 2011a. (in Japanese)

JMA (Japan Meteorological Agency): "Report (No.15) of the 2011 off the Pacific coast of Tohoku Earthquake, 13th March 2011", 2011b. (in Japanese)

Line 12

“... **waves (Nuclear Emergency Response Headquarters, Government of Japan, NERH, 2011).**”

Line 23

“... **April 2011 (NERH, 2011).**”

03. page 4853 lines 19 - 20: The explosions are not only the causes of the radionuclides discharge to the atmosphere. The radionuclide continued to leak to the atmosphere after the explosions.

Page 4853 line 19-20

We will revise the sentence, “As a result of these explosions, a large amount of radioactive material was discharged into the environment.” to “**The reactor accident caused discharge of a large amount of radioactive materials into the environment.**”

04. Page 4853 line 27 - page 4854 line 4: Please add necessary quotations.

Page 4854 line 4

We will add references (Kawamura et al., 2011; Tsumune et al., 2012) after the phrase “...conduits into the ocean.”

05. page 4854 lines 14 - 17: It is useful for the readers to mention major revisions of the radionuclide data in this paper. At least the data available not in the MEXT web page but in

this paper should be noticed.

Page 4854 line 17

We will add a sentence **“Especially revision was made for the radioactivity data that have lacked information on error on the MEXT website”**

06. page 4855 lines 6 - 8: Please add necessary quotations.

Page 4855 line 8

We will add a reference, **“(Oikawa et al., 2011)”** after “...the Chernobyl accident.”

07. page 4855 line 14: Why did the authors conclude that direct discharge of waste water is a main source of radiocesium in the coastal seawaters?. Please show quantitative evaluation or add necessary quotations

page 4855 lines 14-15: We will revise a phrase, “In case of the FDNPP accident, however, direct discharge of wastewater is a main source for the coastal seawaters” to **“In the case of the FDNPP accident, however, not only the atmospheric deposition, but also direct discharge of wastewater were important sources in seawaters off the coast of Miyagi, Fukushima, and Ibaraki Prefectures, where physical processes, such as coastal currents and vertical mixing driven by diurnal tidal currents, are also thought to transport radiocesium downward.”**

08. page 4855 lines 22 - 25: If this sentence explains about oceanographic condition in the coastal region off Miyagi, Fukushima, and Ibaraki Prefs., please add necessary quotations.

Although this sentence do not indicate the coastal region off those three prefectures, the winter mixing occurs in oyashio region near the coastal area.

page 4855 lines 25.

We will added a reference **“(Yoshie et al., 2003))”** to the end of the sentence.

Reference

Yoshie, N., Yamanaka, Y., Kishi, J. M., and Saito, H.: One dimensional ecosystem model simulation of the effects of vertical dilution by the winter mixing on the spring diatom bloom. *J. Oceanogr.*, 59, 563-571, 2003.

09. page 4855 line 28: What are “these two processes” ? diapycnal and isopycnal mixings?

These processes indicate (1) transportation by sinking particles and (2) vertical turbulent mixing.

page 4855 lines 28

We will revise to **“...these two processes (i.e. transportation by sinking particles and vertical turbulent mixing)...”**

10. page 4856 line 21: During the Phase 1, 500-ml or 20-l of seawater samples were collected (not “a few liters”).

We will revise to **“During the Phase 1, 500-ml or 2-l of seawater samples were.”**

11. page 4857 line 5: I believe that Niskin type samplers were used on board of the JAMSTEC cruises during Phase 2 as same as those during the Phase 1.

Page 4857, line5

We will revise “During the phase” to **“During the phase 2, we used a Van Dorn type or Niskin type water sampler with a CTD system (SBE 19, Sea-Bird Electronics, Inc., Bellevue, WA, USA) to collect water samples.”**

12. page 4857 lines 7 - 8: “Although – standpoint, “ is not necessary.

We will delete “Although it would be difficult to define “bottom water” precisely from a physical oceanographic standpoint,”

13. page 4857 lines 20 - 22: This sentence is awkward. What is “detection limit” in this sentence mean? If the detection limit varied widely by analyses, please consider to add each detection limit for activity below the detection on the Tables (for example, <5.0E+00 instead of “-”). I believe this could upgrade value of the dataset in this paper.

page 4857 lines 18 - 22:

From further consideration, we will revise the sentences describing detection limit during phase 1. For the nature of emergency, radio activities of radiocesium in water samples were calculated by using counts which were obtained by deducting row counts from background data, so we did not consider detection limits (e.g., less than 3σ of counting errors).

page 4857 lines 18 - 22:

We will revise the sentence to **“Considering the serious impact of the accident to the environment, activity of the samples was calculated for all the samples as long as total counts were greater than the background without setting any detection limit shown below.”**

14. page 4857 line 23: How about YK11-E01 and NT11-E02? In addition, what does “R” for YK11-E05R and KR11-E04R indicate?

We will add those two cruise and we removed “R” because we put it into the cruises mistakenly.

page 4857 line 23

We will revise **“(i.e. NT11-E01, NT11-E02, MR11-E02, YK11-E02, YK11-E05, and KR11-E04)”**

15. page 4858 lines 3 - 4: How was the recovery of Cs with AMP (=100%) estimated?

page 4858 line 4

We will revise to **“Cs with AMP turned out to be about 100% gravimetrically”**

16. page 4859 lines 18 - 25: The authors explained the temporal changes in I-131 activity and I-131/Cs-137 ratio by the direct discharge of polluted waters from the FDNPP. However there were not discussion about their differences between the direct

discharge and atmospheric fallout. Please add more quantitative discussion about the differences.

page 4859 lines 19 – 24

We will revise the sentences as follow.

“Activities of ^{131}I in surface water were highly variable with time. They decreased rapidly from a geometric mean of 39.2 Bq/L on 23 March 2011 to 7.0 Bq/L on 1 April 2011, and then increased again to 34.1-161 Bq/L in the middle of April. After that, it decreased again until the end of April 2011 (Fig. 2A). The $^{131}\text{I}/^{137}\text{Cs}$ activity ratios were scattered in the period from 23 March to 3 April 2011. After 9 April 2011, the ratio seems to be converged to a value that followed its decay line. The variability of ^{131}I activity and its ratio to ^{137}Cs in surface water reflect variable contributions from the direct discharge of polluted waters and airborne dust (Chino et al., 2011) from the FDNPP, both of which have different activity ratio from each other (see Fig. 11 in a comparison paper Kusakabe et al. (2013)). Tsumune et al. (2012) inferred that although both nuclides in the area were derived mainly from the atmosphere until 9 April 2011, they were dominated by direct discharge after that date.”

17. page 4860 lines 14 - 15: This sentence is inconsistent with the previous one, “the ratio fluctuated until the beginning of the April and then decreased (page 4859 lines 22 – 23)”.

page 4860 lines 14 - 15:

We will delete the sentence because this sentence is not necessary in this section.

18. page 4860 line 17: Was the Cs-134/Cs-137 ratio (0.93) calculated using the values decay-collected to the sampling date, or the date of the FDNPP accident? In the former case the ratio is underestimated compared to those obtained in previous works (e.g. about 0.97 in Buesseler et al., 2012, PNAS).

Although the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio (0.93) calculated using the values was decay-collected to the sampling date, the data were derived from only Table 2 for comparison of $^{131}\text{I}/^{137}\text{Cs}$. We re-calculated the ratio to be 1.03 by using all the data during the phase 1.

Page 4860 lines 15-18:

We will revise to **“In contrast, the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in the surface waters after the direct discharge was apparently constant at 1.03 (Table 1 in Supplement), a value higher than the corresponding ratio of 0.5 in the Chernobyl fallout (UNSCEAR, 2000). This ratio is consistent with that of 0.99 ± 0.03 in the water collected at the north and south discharge channels of the FDNPP in March and April 2011 (Buesseler et al., 2011) and that of nearly 1 in the western North Pacific in April and May 2011 (Honda et al., 2012).”**

Reference

Buesseler, K., Aoyama, M., and Fukasawa, M.: Impacts of the Fukushima nuclear power plants on marine radioactivity. *Environ. Sci. Technol.* 45, 9931, doi:10.1021/es202816c, 2011.

19. page 4860 line 19: In Table 3 there are data from bottom (or intermediate) water of KH11-E01 too.

This is our mistake. In Table 3 we will change MR11-E01 to KH11-E01.

20. page 4860 lines 26 - 27: There is no I-131/Cs-134 ratio in Table 2.

page 4860 line 26

This is our mistake. We will delete I-131/Cs-134 ratio because it is good enough to discuss the ratio I-131/Cs-137.

21. page 4860 lines 27 - 29: What is “complexity”? I believe that authors should discuss the large variation in the ratios using the different ratio between the direct discharge and atmospheric fallout. In addition, there is no I-131/Cs-134 ratio in Figure 2b.

page 4860 lines 27 – 29

We will revise to

“This scatter may also reflect variability in sources of atmospheric dust and direct discharge as shown previous paragraph.

page 4860 line 29

We will delete I-131/Cs-134 ratio

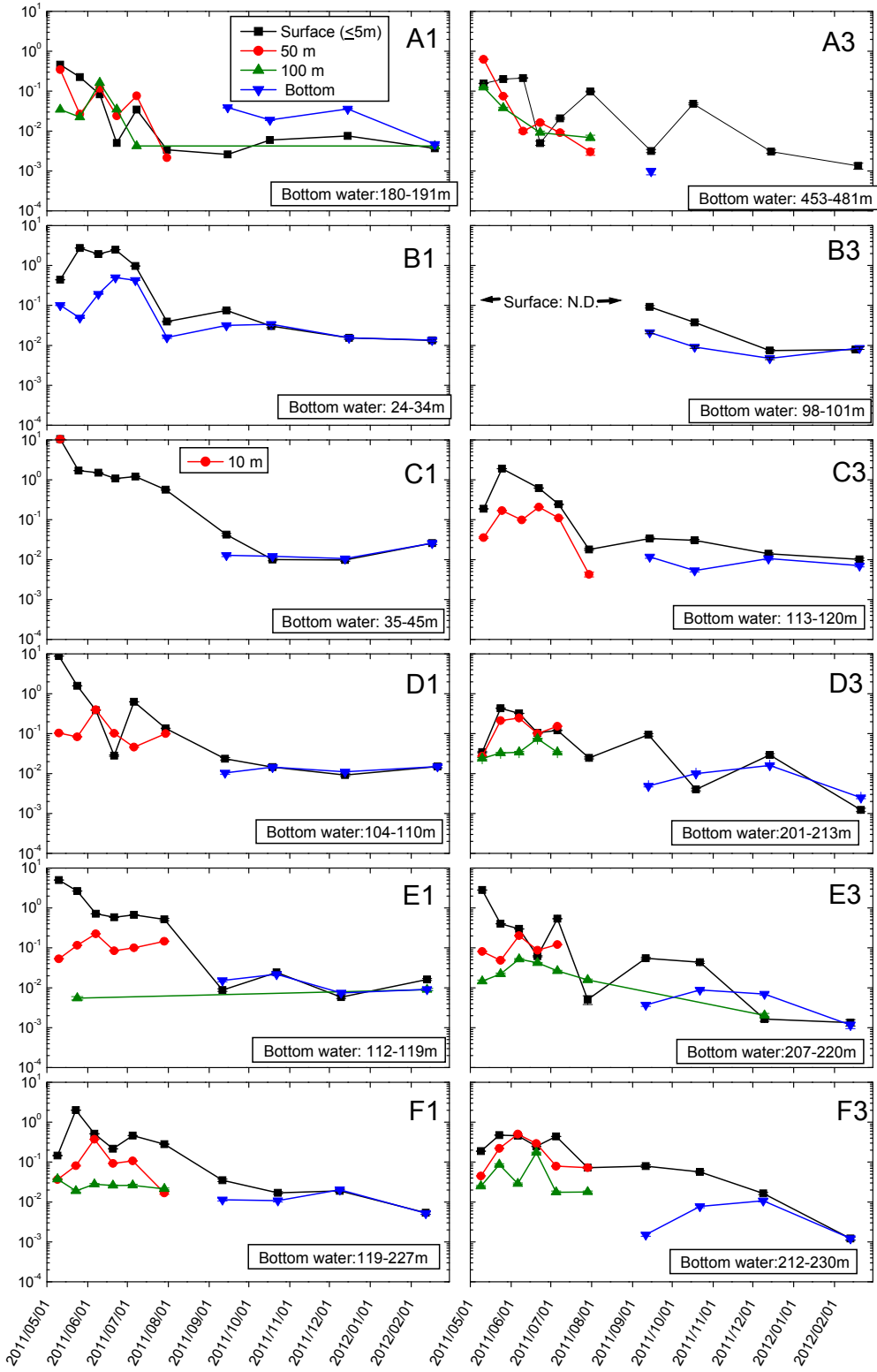
22. page 4861 line 3: “0.93”, same as comment 18.

Please see response to comment 18.

23. page 4861 lines 4 - 7: Before the FDNPP accident Cs-137 derived from the nuclear weapon tests and Chernobyl accident was measured in the surface layers of the western North Pacific (about 1 - 2 Bq/m³). Thus the observed Cs-137 after the FDNPP accident is a mixture of those from the global fallouts and FDNPP accident. In contrast, Cs-134 is an ideal tracer for the FDNPP because there was no background concentration before the accident because of its short half-life. Indeed the concentration of Cs-134 was lower than that of Cs-137 but could be measured significantly in mBq/L range. In addition, the authors discussed the background-level concentration of radiocesium in this paper. Therefore Cs-134 should be used instead of Cs-137 for discussion on the Fukushima-derived radiocesium.

As suggested by the reviewer, discussion on Cs-134 data for the better understanding distribution of FDNPP derived radiocesium, but for long term monitoring research in future using Cs-137 data is important because of its relatively long half-life even though there were background concentrations, so we used the Cs-137 data. However, in order to elucidate whether the subsurface water was contaminated by the FDNPP derived radiocesium, in Figure 5, we will add Cs-134 data.

Decay-corrected ^{134}Cs (Bq/L)



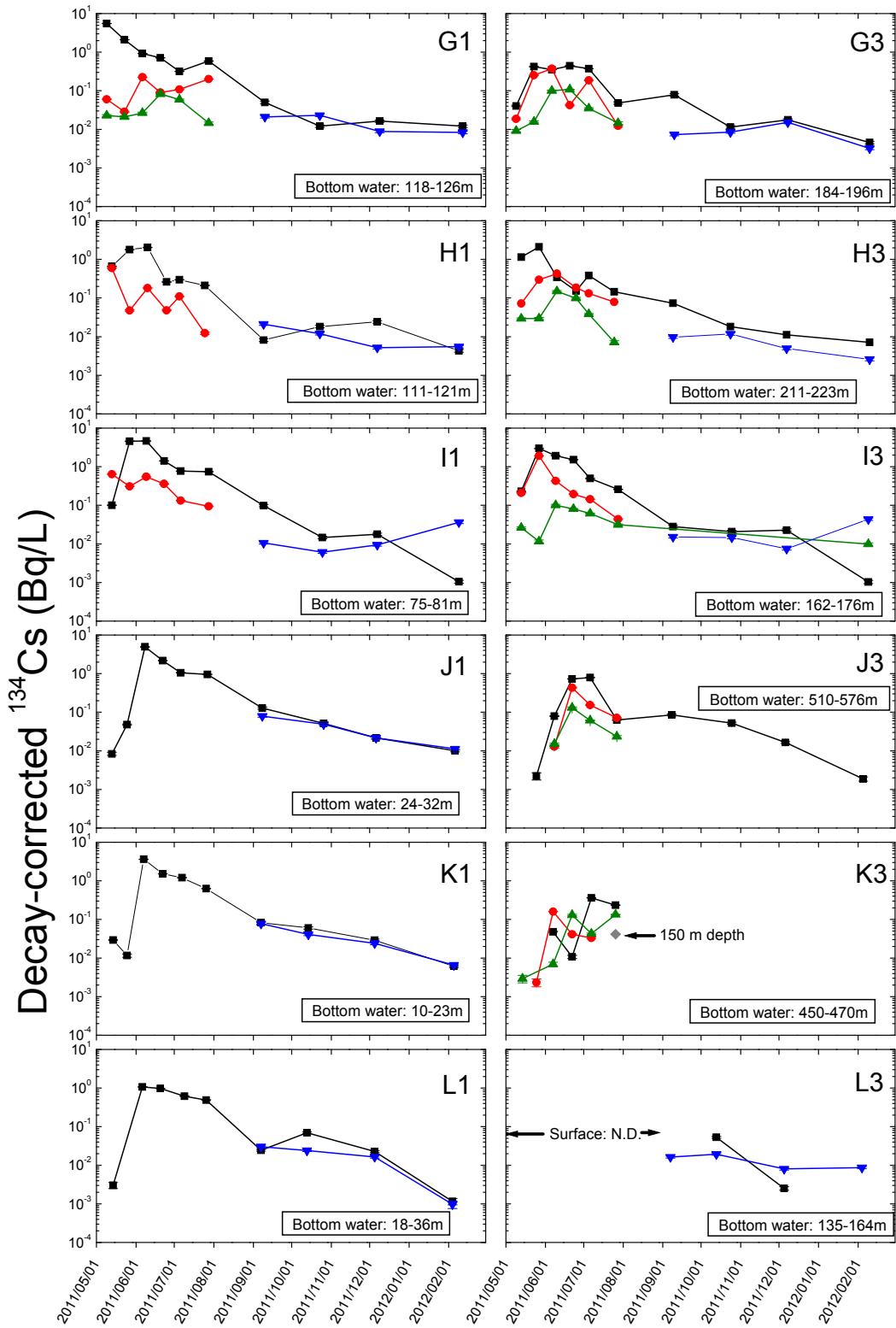


Figure caption

Fig. 5 Temporal changes of ^{134}Cs and ^{137}Cs activities in seawater at each sampling depth. Error bars are less than or equal to the size of the symbols. Activities of ^{134}Cs were decay-corrected to 26 March 2011. Activities of both radiocesium in bottom waters at all the stations from 9–14 May to 26–31 July were below detection limits.

24. page 4861 lines 13 - 18: I cannot see the meaning of this sentence because high concentration of Cs-137 was measured in the subsurface waters before 26 April 2011 (Table 3).

This is our mistake.

page 4861 line 14: We will change “26 April 2011” to “**26 March 2011**”.

25. Page 4861 lines 26 - page 4862 line 2: Why are not data from KY11-E03 and KR11-E07 discussed?

In order to focus on time dependent variation on Cs-137 concentrations in surface and subsurface seawaters, we used 11WM01 (9–14 May 2011) to 11WM14 (4–21 February 2012)) because we can evaluate monthly variation on radiocesium in coastal seawaters.

We will discuss both KY11-E03 and KR11-E07 at second paragraph in section 3.2.

Page 4861 21-22.

We will revise the sentence to “**At Stns 2-26 located 100-300 km away from FDNPP, ^{134}Cs and ^{137}Cs ranged between 0.2 mBq/L and 60 mBq/L and between 1.4 mBq/L and 72 mBq/L in subsurface (100-200 m depth) in August 2011 (cruise: KY11-E03, Table 1 in Supplement). Similar result was observed in November (cruise: KR11-E07). These results indicate that the FDNPP derived radiocesium were spread widely and vertically within 2-5 months.**”

26. page 4862 line 10: What is “complexity”? Do the data shown in Fig.4 support the discussion of Tsumune et al. (2012) or not?

This word indicates that the surface distribution is dependent on coastal currents.

Our field observation might also support simulation in May 2011 by Tsumune et al. (2012).

page 4862 lines 10-14

We will revise

“The relatively high activities in the northeastern area can be dependent on pattern of coastal currents. It is thought that a water mass with high ^{137}Cs was transported northward. Tsumune et al. (2012) have also argued from model analysis that a water mass with high ^{137}Cs activity had been transported southward along the coast by the coastal current until April, and that the water mass was then advected northward during May, the result being a northward dispersion of ^{137}Cs following the initial southward dispersion.”

27. page 4862 line 19: Aoyama et al. (2012, Geochemical Journal, 46, 321-325) also discussed influence of the eddy off the Ibaraki Pref. using the observed radiocesium

activity in the coastal region. Please compare the data presented in this paper with those of Aoyama et al. (2012), which will provide information about spatial scale of the eddy

We will add the report: Aoyama et al. (2012, *Geochemical Journal*, 46, 321-325)

28. page 4863 lines 1 - 4: This sentence is speculative. The inhibition by the highsalinity water mass should be confirmed by spatial distribution of Cs-137 (or salinity) in the north of Stations A1-3. If the Cs-137 activity there is higher than those at Stations A1-3, the blocking by the high-salinity water is improbable.

As described by the reviewer, data on salinity or Cs-137 in the northern area of Stn A1-3 are necessary to elucidate the blocking by high-salinity water. We will delete phrase describing the blocking by the high-salinity.

Page 4862 line 28 to page 4863 line 4

We will revise as follow.

“The water mass at around Stns. A1, A2 and A3 was different from that at around Stns. B1, B2, B3, and B4 with respect to salinity (Fig. 4); they are >34 and >33, respectively. It may imply intrusion of a water mass with low ¹³⁷Cs. ”

29. page 4863 line 13: What is “dynamic mixing”?

We will revise to “water mixing”

30. page 4864 lines 4 - 5: Here I recommend again (see comment 23) that Cs-134 should be used in the Figure 5 instead of Cs-137 in order to trace the Fukushima-derived radiocesium. In addition, I do not doubt that the Fukushima-derived radiocesium penetrated into 200-m depth within six months after the accident. Thus I feel uneasy again that all the radiocesium activity in bottom layers between May and August were “not detected” (or less than 0.1 – 1.0 mBq/L, page 4858 line 15) in Figure 5 and Supplementary Table 1. Please confirm again whether those were measured using the AMP method or the direct analysis. If the latter, the detection limit should be about 10 Bq/L, not 1.0 mBq/L.

We will add Cs-134 data into Fig.5.

page 4864 lines 4 – 5

We will add a sentence **“Similar observation from ¹³⁴Cs can support the vertical transport of FDNPP radiocesium into 200 m depth.”**

The data during May-August periods were measured using the AMP method, so the detection limit was expected to be about 1.0 mBq/L.

31. page 4866 lines 8 - 10: Please add necessary quotations.

We will add a reference “(Matsuura et al., 2007)”

Matsuura, H., Isoda, Y., Kuroda, H., Kuma, K., Saitoh, Y., Kobayashi, N., Aiki, T., Wagawa, T., Yabe, I., and Hoshiba, Y.: Water mass modification process of the passage-flow waters through the Tsugaru Strait. *Umi to Sora*, 83,21–35 (in Japanese with English abstract), 2007.

32. page 4866 lines 14 - 16: Please add the data from the Phase 1 in Figure 6. I think the time series data at D1, E1, and F1 during the Phase 2 can be connected with those at 1-1, 1-3, 2-1 during the Phase 1, respectively. In addition, Cs-137 activities at D1, E1, and F1 can be also connected with those from Phase 1 in Figure 5. These are of help to understand the temporal changes in oceanographic condition and Cs-137 activity near the FDNPP. (In Figure 3 the temporal change of Cs-137 at each station is not clear because lots of data are overlapped in this figure.)

As suggested by the reviewer, connection data during the Phase 1 with that during the Phase 2 is helpful to understand the temporal changes in oceanographic condition.

We will add new density data on Stn. 1-1, 1-3, 2-1 during Phase 1 into Figure 6.

This new graph will show that the σ_t was nearly constant throughout the surface-bottom water column during the Phase 1.

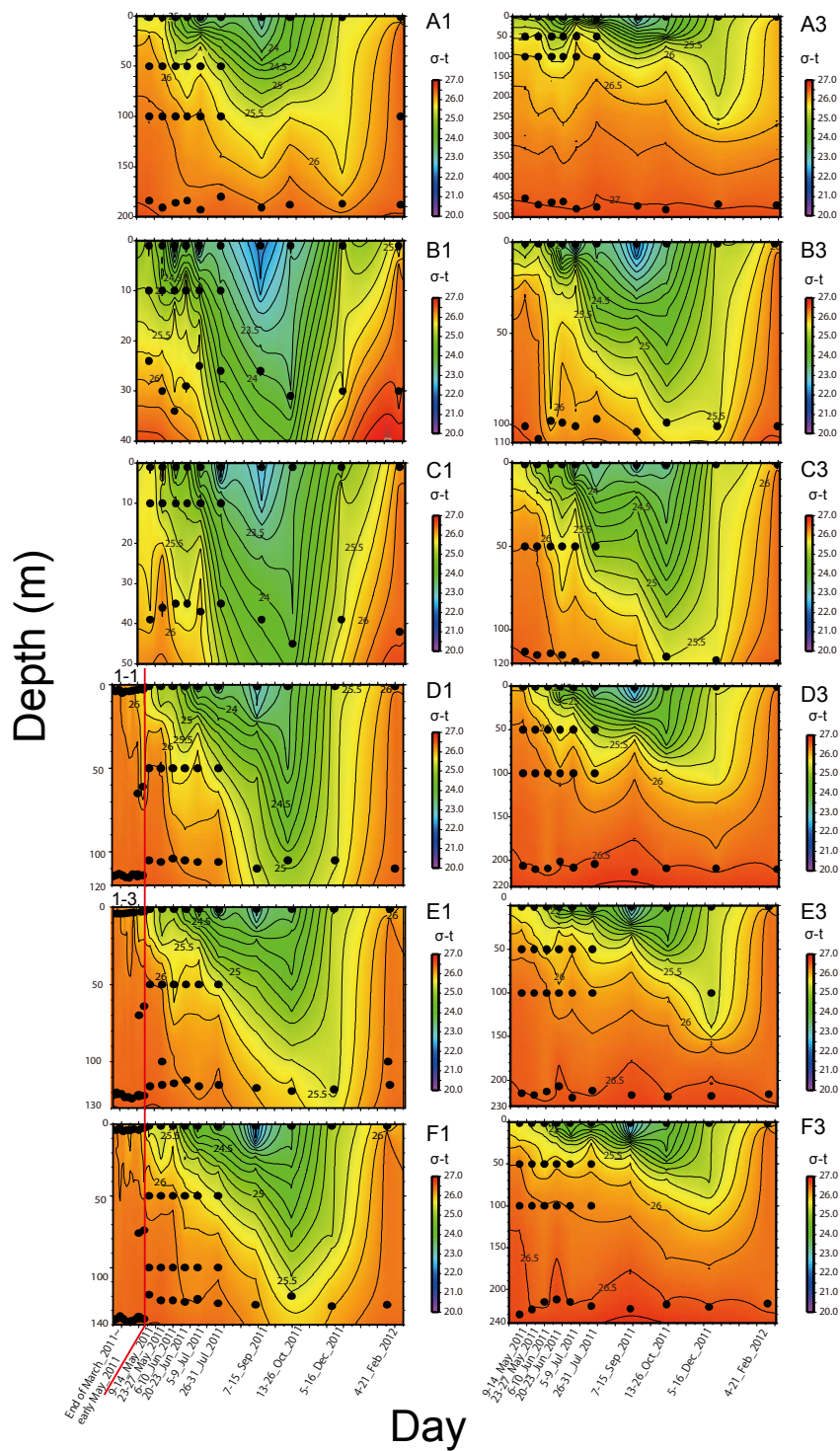


Figure caption

Fig. 6. Vertical distributions of σ_t and their temporal changes. Solid circles indicate

sampling depths. Data taken at Stns 1-1, 1-3 and 2-1 in the Phase 1, which had almost the same locations as those of Stns. D-1, E-1 and F-1, respectively, were also plotted on the left side of the red line in the correspondent figures.

We will revise the sentence in page 4866 lines 10-13:

“In this study, the formation of a vertically homogeneous water column during the Phase 1 could be due to vertical mixing during the winter (Fig. 6A). However, the homogeneous water column was not observed at each station from 9–14 May to 5–16 December 2011 (Fig. 6). Vertical gradients of σ_t at each depth indicate that little diapycnal mixing was occurring during the Phase 2.”

As for Cs-137 (or decay-corrected Cs-134) data for 1-1, 1-3, 2-1 during the Phase 1, it is not appropriate for connecting with those in Stn. D1, E1 and F1 in Figure 5 because most of data in seawaters were not detected. However, we will add following sentences in order to discuss whether relatively high radiocesium was vertically transported during vertical mixing during the Phase 1 before a sentence in page 4866 lines 2-4:

“Relatively high radiocesium (especially ^{134}Cs) activities were observed in subsurface waters during the Phase 1, at only Stns. 2-6 (49 m depth) and S4 (61 m depth) in 25 April and Stn. 1-B (30 m depth) in 5 May 2011. However, ^{137}Cs activities in subsurface waters, especially water depth of more than 100 m, were not detected. Thus, it is unlikely that the radiocesium derived from the FDNPP accident was transported to bottom depth during the Phase 1 period.”

33. page 4866 lines 16 - 17: Which isopycnal line reached bottom depth?

The lines indicate σ_t values of 25.5-26.5.

page 4866 lines 16 – 17:

We will revise the sentence to **“The isopycnal lines (σ_t : 25.5-26.5) reached bottom depths at Stns. A1–I1, B3, C3, and I3 or depths of about 200m at Stns. A3 and D3–H3 by 5–16 December 2011 (Fig. 6).”**

34. page 4867 lines 8 - 10: Negative. The deepening of the isopycnal from spring to autumn above 200-m depth does not mean downward move of seawater (isopycnal 25.5 - 26.5), i.e. Cs-137 activity. Primary cause of the deepening of isopycnal is probably the seasonal heating in surface layer one-dimensionally. Transport of Cs-137 on isopycnal layer should be discussed on the temporal changes both in horizontal and vertical oceanographic structure from March 2011.

As suggested by the reviewer, discussion on oceanographic structure from March 2011 is important to elucidate processes affecting radiocesium behavior. We will add the vertical water structure at Stns 1-1, 1-3, and 2-1 which are corresponding to Stns D1, E1, and F1, respectively.

We have discussed the processes using oceanographic data from March 2011 and will revise the sentence in page 4866 lines 10-13, and add sentences in order to discuss whether

relatively high radiocesium was vertically transported during vertical mixing during the Phase 1 before in page 4866 lines 2-4: Please see response to comment 32.

As for deepening of isopycnal layer:

We do not indicate the move of water mass. Activities in radiocesium could be diluted along the isopycnal lines. As suggested by the reviewer, the seasonal heating in surface layer one-dimensionally is one of factors the deepening. In our results, radiocesium activity decreased with isopycnal lines gradually. In addition, there were significant difference in the activities among isopycnal lines. For example, at Stns.D3 and E3, on isopycnal lines for 100 m depth (σ_t : 26-26.5), relatively high Cs-137 activities were observed, compared with those on other lines for 100 m depth (Fig. 6). This result suggests that activities in radiocesium were diluted along the isopycnal lines. Thus, it is indicated that isopycnal mixing is important. We will revise the sentence in page 4867 lines 8 – 10.

“Thus, ¹³⁷Cs in coastal waters was diluted along the isopycnal lines during the spring-to-autumn time interval, resulting in relatively rapid increase in activities of ¹³⁷Cs at bottom waters and/or 200 m depth within two months.”

35. page 4867 lines 24 - 25: What is “coastal current”? And isn't there any contribution of the atmospheric fallout to the Sr-90 activity in the surface water?

From our results, we cannot indicate, but coastal currents are thought to indicate oyashio, kuroshio, and/or branches of those two currents.

As for the atmospheric fallout, Povinec et al. (2012) have reported that three orders of magnitude lower amounts were released to the atmosphere than in the case of radio Cs in March 2011. Thus, we think that contribution of atmospheric radiostrontium input is relatively low to surface waters.

36. page 4867 line 27: What does “slowly” mean? Slower than the decrease of Cs-137 activity?

It is our mistake. We wanted to express gradual decrease in Cs-137.
We will change “slowly” to “gradually”.

37. page 4868 lines 19 - 22: Do authors consider that the high Sr-90/Cs-137 ratios in the early December are derived from the 2nd direct discharge of treated water on 4 December? If so, the discharged water from FDNPP reached to the station about 30 km away within one week. This time-scale should be compared with that estimated from the Cs-137 peak in the middle of April (Fig.3), which was derived from the first direct discharge of waste water from FDNPP in the early April.

We do not think that it is appropriate to comparison of the time-scale in dispersion process between the early April and December because oceanographic condition (current or structure of water mass) in the April was thought to be different from that in the December.

38. page 4868 lines 24 - page 4869 line 2: This paragraph is not necessary in “Conclusion”.
We will delete.

39. Supplementary Table 1: Why are sigma-t data of KH11-E01 missing while temperature and salinity are reported?

We will add the sigma-t data

Technical comments

40. page 4854 line 13: Abbreviation “MEXT” appears for the first time in the text. Please add a full-expression too.

We will add a full-expression.

41. page 4856 line 18: Should “1-A” and “1-B” be added too?

We will add both two stations.

42. page 4867 line 22: This paper is not in “References”.

It is our mistake

We will revise to “(Tsumune et al., 2012)”.

43. page 4867 lines 23 - 24: “0.22 - 043 mBq/L” should be replaced by “2.2 – 4.3 mBq/L”?

It is our mistake. We will replace the values.

44. page 4870 line 2: “crews” -> “crew”.

We will change it.