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## ***Interactive comment on “Distribution of the Fukushima-derived radionuclides in seawater in the Pacific off the coast of Miyagi, Fukushima, and Ibaraki Prefectures, Japan” by S. Oikawa et al.***

**Anonymous Referee #1**

Received and published: 22 March 2013

### General comments

This manuscript entitled "Distribution of the Fukushima-derived radionuclides in seawater in the Pacific off the coast of Miyagi, Fukushima, and Ibaraki Prefectures, Japan" provides a significant dataset of the Fukushima-derived radionuclides in the northwest-ern North Pacific from March 2011 to February 2012. This dataset must contribute to elucidate the total amount and behavior of the Fukushima-derived radionuclides in the Pacific Ocean. However, discussion on the mechanisms that control distribution of the radionuclides in the water column are very poor. In addition, the metadata is still insuffi-cient although the authors revised the first manuscript. I recommend modifications

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listed below before the publication in Biogeosciences.

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Specific comments

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”
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.
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.
04. Page 4853 line 27 - page 4854 line 4: Please add necessary quotations.
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.
06. page 4855 lines 6 - 8: Please add necessary quotations.
07. page 4855 line 14: Why did the authors conclude that direct discharge of waste water is a main source of radio cesium in the coastal seawaters?. Please show quantitative evaluation or add necessary quotations
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.
09. page 4855 line 28: What are “these two processes” ? diapycnal and isopycnal mixings?
10. page 4856 line 21: During the Phase 1, 500-ml or 20-l of seawater samples were

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collected (not “a few liters”).

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.

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

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.

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

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

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.

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)”.

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).

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

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

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.

22. page 4861 line 3: “0.93”, same as 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/m<sup>3</sup>). 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.

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).

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

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?

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

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28. page 4863 lines 1 - 4: This sentence is speculative. The inhibition by the high-salinity 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.

29. page 4863 line 13: What is “dynamic 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.

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

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.)

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

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

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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.

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?

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

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.

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

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

#### Technical comments

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

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

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

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

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