

## Reply to reviewer 2

Thank you very much for valuable your comments and questions. According to your comments, the manuscript was revised. The answer to your comments are described as below.

### **Major points:**

- **Although English is not my mother tongue, I found that grammar and typing mistakes are everywhere in the manuscript; furthermore, it contains many awkward sentences that confuse the reader and make the scientific message difficult to understand. I suggest the authors to consider English professional editing for the revised version.**
- A. The author ask to the English editing company to correct English.
- **Analyses: to properly analyze the circulation and to estimate transport (by the way, how this is done in this manuscript?), especially through the straights or across frontal structure, I suggest the authors to present cross-sections (longitude or latitude versus depth) of radiocaesium.**
- A. In order to clearly show the propagation of FNPP1-<sup>137</sup>Cs, latitude-time cross sections at the potential density surface were depicted in Figure 7. We selected the  $25.2\pm 0.5 \text{ kg m}^{-3}$  surface data because the maximum <sup>137</sup>Cs activity concentration was observed at this layer. In other words, we think that the center of FNPP1-<sup>137</sup>Cs transported in the North Pacific Ocean was located in this layer. In this latitude-time cross sections, the <sup>137</sup>Cs activity concentrations in the ECS were maximum in 2014/2015, and then tended to decrease in 2016. In southwest SOJ, <sup>137</sup>Cs activity concentrations were gradually increased until 2016. In the northeastern Japanese monitoring stations, the <sup>137</sup>Cs activity concentrations were gradually increased. These suggest the propagation of the <sup>137</sup>Cs occurred from southwestern SOJ and northeastern SOJ.

(Page 5, Line 30 ; Page 6, Line 5)

Fig. 7 displays the latitude-time cross sections of the <sup>137</sup>Cs activity concentrations at potential densities of  $25.2\pm 0.5 \text{ kg m}^{-3}$  along the western TWC,  $25.7\pm 0.5 \text{ kg m}^{-3}$  along the eastern TWC, and  $26.7\pm 0.5 \text{ kg m}^{-3}$  along the eastern TWC. These potential density surfaces were selected to show the maximum <sup>137</sup>Cs activity concentrations observed in the ECS. The vertical distributions of the <sup>137</sup>Cs activity concentrations with depth and potential density at each monitoring station are displayed in Fig. SI11. Note that in the SOJ, the vertical distributions of the <sup>137</sup>Cs activity concentrations below 250 m were almost constant, and the subsurface peak of <sup>137</sup>Cs was not found at the monitoring stations along the eastern TWC (Fig. SI11). It is noted that the <sup>137</sup>Cs activity concentrations before the FNPP1 accident were approximately  $1.5 \text{ Bq m}^{-3}$ . As shown in Fig. 7a, in the ECS, the <sup>137</sup>Cs activity concentrations gradually increased and attained the maximum in 2014/2015. The <sup>137</sup>Cs activity concentrations in the ECS tended to decrease in 2016 in a layer with a density of  $25.2\pm 0.5 \text{ kg m}^{-3}$ . In the southwestern part of the SOJ (Shimane, Fukui, Ishikawa, Niigata), the <sup>137</sup>Cs activity concentrations also gradually increased beginning in 2012, and the activity concentrations attained a maximum of  $2.5 \text{ Bq m}^{-3}$  in 2015/2016; these trends were almost the same as those at the monitoring stations in the ECS. In the northwestern SOJ (Aomori and Tomari), the <sup>137</sup>Cs activity concentrations were slightly increased and higher activity concentrations above  $2 \text{ Bq m}^{-3}$  were observed in 2016. These results revealed that the propagation of FNPP1-<sup>137</sup>Cs occurred within several years from the ECS to the SOJ along the TWC.

On the other hand, the latitude-time cross section along the western TWC indicated that the higher activity concentrations were observed in the layers with potential densities of  $25.7\pm 0.5$  and  $26.7\pm 0.5 \text{ kg m}^{-3}$  than other layers (Fig. 7b,c). Higher <sup>137</sup>Cs activity concentrations were observed in the higher potential density layer in comparison with those in the seawater along with eastern TWC. In the 105-11 station, the

decrease of  $^{137}\text{Cs}$  activity concentrations were started from 2015/2016.

(Page 7, Line 13-21)

In this study, we revealed that FNPP1- $^{137}\text{Cs}$  entered the SOJ via the ECS; then, FNPP1- $^{137}\text{Cs}$  was transported northward with the TWC. In the SOJ, a time lag of the propagation of FNPP1-radiocaesium of approximately one year was observed (Fig. 7). Based on measurements of phosphate, one of the dominant seawater nutrients, Kodama et al. (2016) revealed that the phosphate concentrations in surface seawater during winter were significantly positively correlated with the concentrations in the saline ECS seawater in the preceding summer, and the surface water of the southern SOJ was almost entirely replaced by the ECS seawater during May–October. Kodama et al. (2016) suggested that the transport of water-soluble constituents from the ECS to the SOJ takes at least approximately 0.5 years. The propagation of FNPP1-radiocaesium in the SOJ was consistent with the propagation time scale of nutrients concentration change from the ECS to the SOJ (Kodama et al., 2016).

(Figure 7)

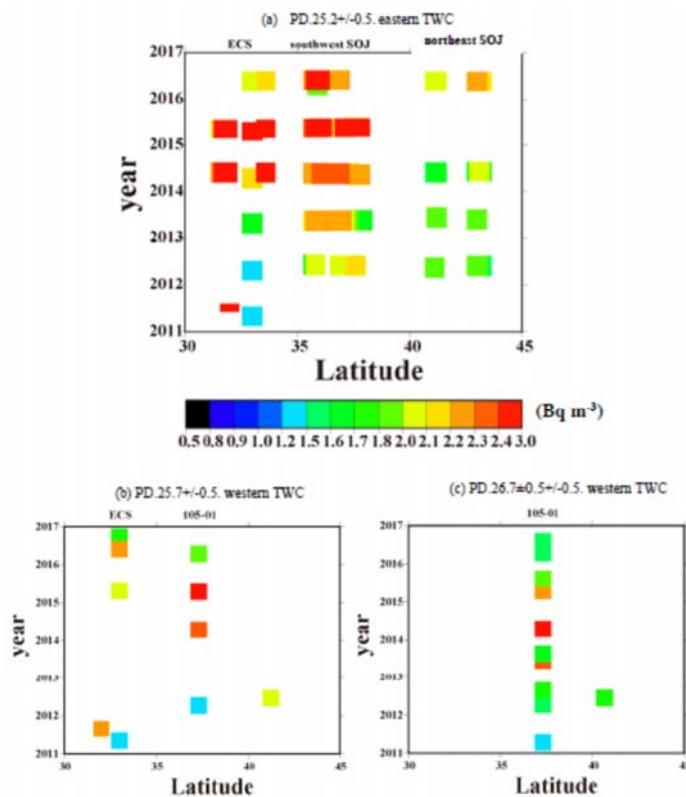


Figure 7: Latitude-time cross sections of the  $^{137}\text{Cs}$  activity concentrations at a potential density. (a) along with an eastern TWC at a potential density of  $25.2 \pm 0.5 \text{ kg m}^{-3}$ , (b) along with a western TWC at a potential density of  $25.7 \pm 0.5 \text{ kg m}^{-3}$ , (c) along with a western TWC at a potential density of  $26.7 \pm 0.5 \text{ kg m}^{-3}$ . The ECS stations described on the x-axis are Kagoshima and Saga stations. Southwestern SOJ includes the monitoring stations Shimane, Fukui, Ishikawa, and Niigata. The northwestern SOJ includes the monitoring stations Aomori and Tomari. Colour indicates the  $^{137}\text{Cs}$  activity concentrations ( $\text{Bq m}^{-3}$ ).

- Looking at Fig. 1, it seems that the authors have access to at least a few almost-synoptic measurements along “sections” (including at the straights), so this seems feasible.

A. The data used in this study was collected by literature reserach except for our own data to measure the  $^{134}\text{Cs}$  activity concentrations. The most of sections data did not measure repeatedly. Instead of this, we investigated the time variation at the monitoring sites by the Japanese and Korean government.

(Page 2, Line 41- Page 3, Line 16)

## 2.1. Data sources

After the FNPP1 accident, many radiocaesium measurements were taken in the SOJ and western North Pacific Ocean (Fig. 1). To elucidate the temporal and spatial distributions of the radiocaesium activity concentrations, we use as many data points as possible. We, therefore, compiled all available data from the literature and reported studies. Most of the data before the FNPP1 accident was included in the database, “Historical Artificial Radionuclides in the Pacific Ocean and its Marginal Seas (HAM database)” (Aoyama and Hirose, 2003 and their updated version). The data observed after the FNPP1 accident were shown in Aoyama et al. (2016a). The term “surface seawater” used in this study defines a sample collected at less than 10 m depth.

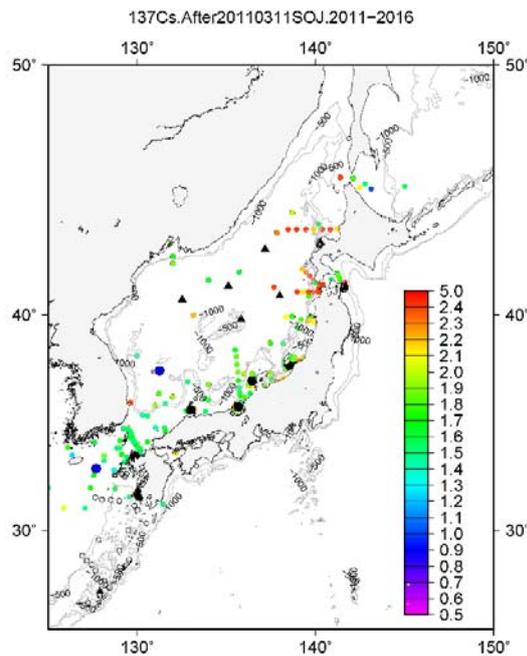
We also focused on the Japanese government’s monitoring data at Tomari (42.98-43.17°N, 140.21-140.30°E), Aomori (41.13-41.22°N, 141.50-141.67°E), Niigata (37.62-38.10°N, 138.38-138.84°E), Ishikawa (36.87-37.29°N, 136.43-136.47°E), Fukui (35.75-36.09°N, 135.50-135.83°E), Shimane (35.67-35.80°N, 132.87-133.2°E), Saga (33.57-33.62°N, 129.73-129.98°E), and Kagoshima (31.58-31.93°N, 130.02-130.15°E) (Marine Ecology Research Institute, 2011, 2012, 2013, 2014, 2015, 2016) (Fig. 1). These measurements were taken once a year (from the middle of May to early June). Near the Aomori sites, offshore monitoring was also conducted twice a year (May and October) at the Rokkasho Reprocessing Plant (39.5-41.4°N, 141.5-142.3°E). Seawater was sampled from 0-664 m with different depths at each monitoring site. Monitoring data (304-01(33.0°N, 127.7°E), 105-11(37.3°N, 131.3°E)) from the Korean government was also used in this analysis (Korea Institute of Nuclear Safety, 2011, 2012, 2013, 2014, 2015, 2016) (Fig. 1). At these monitoring sites, the surface seawater (0 m) measurements were taken four times (February, April, August, October) a year.

- **There are also many other ways to gain insights into transport processes in the ocean. For surface pathways, one could be looking at satellite data (SST patterns and SSH data to derive geostrophic currents). For subsurface pathways, one could examine the subsurface dispersal of ARGO floats in the region and exploit numerical models. The latter includes (1) exploring the outputs of Eulerian mesoscale models (looking at the list of references, some outputs may be sourced by the authors themselves; the Japanese modelling group at the Earth-Simulator possibly run routinely models in the region; there are also some outputs publicly available, for instance on <http://marine.copernicus.eu/services-portfolio/access-to-products/>) as well as (2) performing ad-hoc Lagrangian experiments to unveil pathways (see also some references provided below). Altogether, these additional analyses could really help to grasp the three-dimensional structure of the flow and could provide further evidences to support their conclusions.**

A. Data analysis by using satellite data, ARGO, model simulation, lagrangian experiments give us very useful information to investigate the transport of radiocaesium in the seawater. Authors recognized the importance of these data to interpret the transport as well as spatial and temporal distributions of FNPP1-radiocaesium. However, the evaluation of radiocaesium propagation from interior to the surface seawater did not obtain by using the satellite data. Although model simulation is very useful to interpret the transport and distribution of radiocasium, these also include the uncertainty. It is better to investigate the spatial and temporal variation of radiocaesium based on the measurement results at first stage. This matter is also suggested by the reviewer 1. The authors analyzed the radiocesium distributions by using only the observation data. The transport of radiocaesium by using the numerous data such as Japan Coastal Ocean Predictability Experiment (JCOPE) will be investigated as next step.

**Discussion: I found that the results could be discussed further. For instance, the authors assumed that atmospheric deposition occurred only over the Pacific but what if airborne radiocaesium also fell down over the Sea of Japan?**

- A. Just after the FNPP1 accident,  $^{137}\text{Cs}$  activity concentrations were increased in comparison with those before the FNPP1 accident (about  $1.5 \text{ Bq m}^{-3}$ ) in the northern part of the Sea of Japan (north of  $40^\circ\text{N}$ ). The higher  $^{137}\text{Cs}$  activity concentrations exceed the concentrations before the FNPP1 accident was observed in the region north of 40 degree. In Fig. 1,  $^{137}\text{Cs}$  activity concentrations measured in 2011 were shown in color circles.



(Page 2, Line 16-21)

In the northeastern part of the SOJ, atmospherically deposited radiocaesium were caused to approximately 1-2 times higher activity concentrations of radiocaesium in May 2011 (1.5-2.8 Bq m<sup>-3</sup>) in northeastern SOJ than those of before the FNPP1 accident (~1.5 Bq m<sup>-3</sup>) (Fig. 1). By the end of 2011, the <sup>137</sup>Cs activity concentrations in the northeastern part of the SOJ had rapidly decreased to almost the same levels as those before the FNPP1 accident (Inoue et al., 2012). In fact, the <sup>137</sup>Cs activity concentration in surface water at an SOJ coastal monitoring site after July 2011 was almost the same as the pre-FNPP1 accident level (<http://radioactivity.nsr.go.jp/ja/list/195/list-8.html>).

- **They conclude with a transport pathway crossing a well-established oceanic front : what kind of physical processes (front destabilization? Mesoscale processes? Etc...) could explain this route? What about discussing the effect of diffusive mixing?... A recent publication (Sania et al. PNAS 2017) suggests that continuous submarine groundwater discharge could also contribute to the radioactive elements measured in Japanese coastal waters; this process could also occur on the western shores...etc...**

A. In this study, we focus on the description of the spatial and temporal distributions of radiocaesium activity concentrations, FNPP1 derived radiocaesium activity concentrations, and <sup>134</sup>Cs/<sup>137</sup>Cs activity ratio. In the revised manuscript, the authors discuss about the advection and vertical mixing of FNPP1-<sup>137</sup>Cs in the SOJ. As for the Sania et al.(2017), the unexpected high activity concentrations of <sup>137</sup>Cs (up to 23000 Bq m<sup>-3</sup>) were observed in ground water. In the coastal site of SOJ, the <sup>137</sup>Cs associated with inflow from the ground water as well as river water might be negligible to the <sup>137</sup>Cs activity concentrations in the seawater. Therefore, the author think that the contribution from a ground water is minor.

(Page 7, Line 12-35)

#### 4.1. Advection and vertical mixing of FNPP1-<sup>137</sup>Cs in the SOJ

In this study, we revealed that FNPP1-<sup>137</sup>Cs entered the SOJ via the ECS; then, FNPP1-<sup>137</sup>Cs was transported northward with the TWC. In the SOJ, a time lag of the propagation of FNPP1-radiocaesium of approximately one year was observed (Fig. 7). Based on measurements of phosphate, one of the dominant seawater nutrients, Kodama et al. (2016) revealed that the phosphate concentrations in surface seawater during winter were significantly positively correlated with the concentrations in the saline ECS seawater in

the preceding summer, and the surface water of the southern SOJ was almost entirely replaced by the ECS seawater during May–October. Kodama et al. (2016) suggested that the transport of water-soluble constituents from the ECS to the SOJ takes at least approximately 0.5 years. The propagation of FNPP1-radiocaesium in the SOJ was consistent with the propagation time scale of nutrients concentration change from the ECS to the SOJ (Kodama et al., 2016). As shown in Fig. 6e, the  $^{137}\text{Cs}$  activity concentrations at station 105-11, located along the western TWC in the SOJ, were maximum at the surface and gradually decreased with increasing depth. This vertical distribution is different from those in the SJPN (Fig. 6a) and ECS (Fig. 6c). Particularly, the subsurface peak observed in the SJPN and ECS did not appear at station 105-11. At station 105-11, most of the  $^{137}\text{Cs}$  existed in seawater with a potential density of 25.7–27.3 kg m<sup>-3</sup> (Fig. 6f), which was located in a higher potential density layer than that in the SJPN and ECS. A similar vertical distribution was also observed at the western coast of the Japanese Islands along the eastern TWC (Fig. SI11). These distributions were due to cooling in the surface layer after water was transported from the Tsushima Strait. Physical processes such as the convergence and subduction of surface water inside the eddies are important mechanisms of downward transport of radiocaesium (Miyao et al., 1998; Budyansky et al., 2015). Based on the lagrangian analysis of the vertical structure of the eddies in the SOJ, the eddy in summer was characterized as unstable layer results in thinner mixed layer depth and weaker seasonal pycnocline in upper layer. During winter season, the eddy became to be very stable in the upper layers, which leads to increase mixed layer depth and become to weak seasonal pycnocline (Prants et al., 2015). There is a possibility of the seasonality of downward transport of radiocaesium. As expected, the past global fallout- $^{137}\text{Cs}$  had already penetrated and accumulated in the deeper layers of the SOJ.

- **Bibliography: this contribution contains a large number of auto-citations and crucially suffers from a lack of key references. A large body of bibliography is not cited nor discussed properly. Bibliographic items which have been totally omitted in the present manuscript, but which MUST be cited and properly discussed in a revised, include:**

**Behrens, E.; Schwarzkopf, F. U.; Lubbecke, J. F.; Boning, C. W. Model simulations on the long-term dispersal of  $^{137}\text{Cs}$  released into the Pacific Ocean off Fukushima. *Environ. Res. Lett.* 2012, 7, 034004.**

A. (Page 1, Line 34-36)

It was revealed that dilution due to horizontal and vertical dispersion in the vicinity of Kuroshio led to a rapid decrease of the radiocaesium activity concentrations (Behrens et al., 2012).

**Budyansky, M.V., V.A. Goryachev, D.D. Kaplunenko, V.B. Lobanov, S.V. Prants, A.F. Sergeev, N.V. Shlyk, M.Yu. Uleysky, Role of mesoscale eddies in transport of Fukushima-derived cesium isotopes in the ocean, *Deep Sea Research Part I: Oceanographic Research Papers*, Volume 96, 2015, Pages 15-27, <https://doi.org/10.1016/j.dsr.2014.09.007>.**

A. (Page 7, Line 28-30)

Physical processes such as the convergence and subduction of surface water inside the eddies are important mechanisms of downward transport of radiocaesium (Miyao et al., 1998; Budyansky et al., 2015).

**Oka, E., Qiu, B., 2012. Progress of North Pacific mode water research in the past decade. *J. Oceanogr.* 68, 5–20, <http://dx.doi.org/10.1007/s10872-011-0032-5>.**

**Oka, E., Qiu, B., Kouketsu, S., Uehara, K., Suga, T., 2012. Decadal seesaw of the Central and Subtropical Mode Water formation associated with the Kuroshio Extension variability. *J. Oceanogr.* 68, 355–360.**

A. (Page 6, Line 32-33)

One of the region in which  $^{137}\text{Cs}$  was deposited in south of Kuroshio and Kuroshio Extension regions corresponded to the STMW formation region (Aoyama et al., 2016; Oka et al., 2012, 2013).

**Prants, S.V., M.V. Budyansky, V.I. Ponomarev, M.Yu. Uleysky, P.A. Fayman. Lagrangian analysis of the vertical structure of eddies simulated in the Japan Basin of the Japan/East Sea. Ocean Modelling. V.86 pp.128-140 (2015). <http://dx.doi.org/10.1016/j.ocemod.2014.12.010>.**

A. (Page 7, Line 30-33)

- Based on the lagrangian analysis of the vertical structure of the eddies in the SOJ, the eddy in summer was characterized as unstable layer results in thinner mixed layer depth and weaker seasonal pycnocline in upper layer. During winter season, the eddy became to be very stable in the upper layers, which leads to increase mixed layer depth and become to weak seasonal pycnocline (Prants et al., 2015).

**Prants, S.V., M.V. Budyansky, M.Yu. Uleysky. Statistical analysis of Lagrangian transport of subtropical waters in the Japan Sea based on AVISO altimetry data. Nonlin. Processes Geophys. V.24, p. 89-99, 2017 doi:10.5194/npg-24-1-2017.**

A. This paper described the northward transport of subtropical modewaters in the Sea of Japan. ; The subtropical water transported « gates » in specific places and time. In the frontal area, there are some « forbidden » zone that the northward transport did not observed. However, the radiocaesium data investigated in this manuscript was only measured at coastal site of SOJ. The focus of the Prants et al. (2017) is a little different from that of our manuscript. Therefore, the author used this paper as showing the feature of SOJ.

(Page 1, Line 26-34)

- The SOJ is located between the Eurasian continent and the Japanese archipelago. The area is 1008000 km<sup>2</sup>, and the mean depth is 1667 m (Menard and Smith, 1966). The SOJ is connected to the Pacific Ocean at its southwest through the Tsushima Straits and northeast through Tsugaru Straits. Warm and saline seawater passes through the Tsushima Strait as the Tsushima Warm Current (TWC), and this current splits into three paths. One is the nearshore current along the west coast of Honshu Island, Japan, and the seawater passes through the Tsugaru Straits and enters to the Pacific Ocean again. The seawater transported to the northward passes through the Soya Strait and connected to the Okhotsk Sea. Another current flows north of the Korean Peninsula. This current meets the North Korean Cold Current, which is the prolongation of the Liman Cold Current. Therefore, this northward warm subtropical waters and southward cold subarctic waters form the Polar Front meet at approximately 40°N, and the SOJ is largely divided into two regions (Prants et al., 2017).

**Rossi, V.; Van Sebille, E.; Sen Gupta, A.; Garçon, V.; England, M. H. Multi-decadal projections of surface and interior pathways of the Fukushima Cesium-137 radioactive plume. Deep Sea Res., Part I 2013, 80, 37–46.**

A. This paper was re-submitted as « Corridendum to « Multi-decadal projections of surface and interior pathways of the Fukushima cesium-137 radioactive plume » (Deep Sea Reserach 1, 93, (2014), 162-164). In order to describe the model simulation results, the author added Rossi et al. (2014) and Tsubono et al. (2016).

(Page 1, Line 33-34)

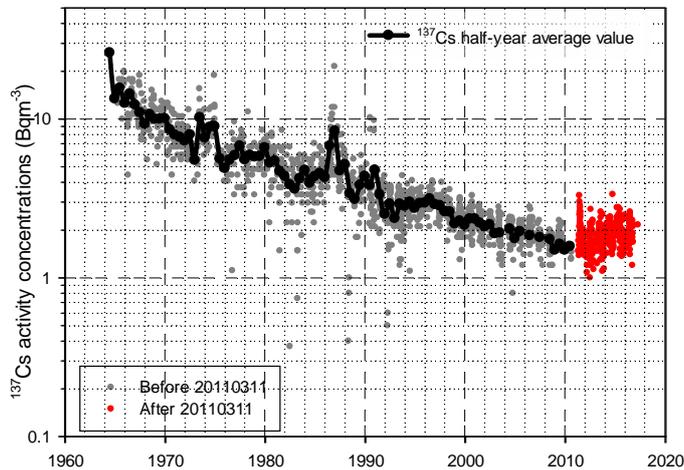
Ocean circulated models captured that the FNPP1-derived radiocaesium were transported in the North Pacific Ocean with advecting and diluting (Tsubono et al., 2016; Rossi et al., 2014).

- **Some figures are not necessarily well chosen and some are not very informative due to their poor content and/or low quality and low visual rendering. This is especially true for fig. 2**

(e.g. grey and red data points and error bars are not readable)

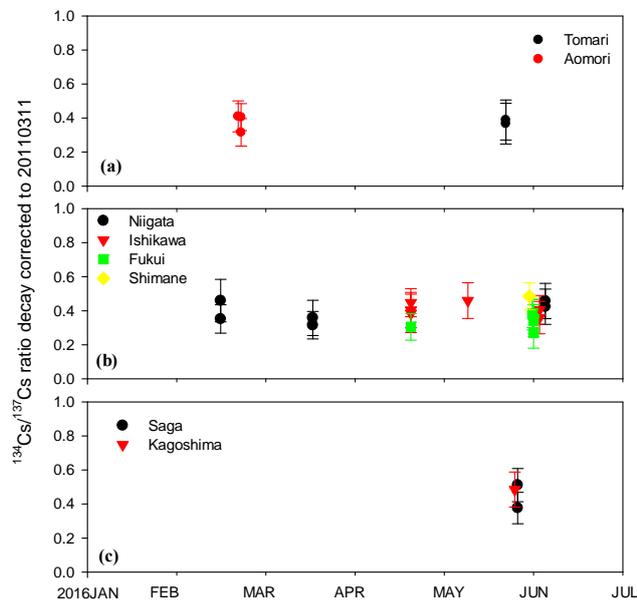
A. Almost all Figures were rewrite.

As for Fig.2, Authors think that the longtime variation of  $^{137}\text{Cs}$  activity concentrations in the SOJ was necessary to discuss the enhanced activity concentrations of  $^{137}\text{Cs}$  after the FNPP1 accident. In order to show clearly the activity concentrations of  $^{137}\text{Cs}$ , the error bars of each sample was removed in this Figure.



– **fig. 3 (panels d, e and f: cut the x-axis in mid-2016 since there is no data afterwards);**

A. The period in the x-axis range for Fig 3d, e, f were modified and these were mover to Figure 4.



– **fig. 5 (subplots are not numbered; what does black color means in panel 5c and 6c?);**

A. Fig. 5 was removed from the revised manuscript.

– **the scatters in fig. 9 are not clear to me, please clarify.**

- A. Before the FNPP1 accident,  $^{137}\text{Cs}$  were released from the large scale nuclear bomb experiment. After the FNPP1 accident,  $^{137}\text{Cs}$  were derived from two sources, global fallout and FNPP1 accident. Because of shorter lifetime,  $^{134}\text{Cs}$  ( $T_{1/2}=2.06$  year) was only derived from the FNPP1 accident. Therefore, the  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio is used as a useful tracer to investigate the transport of radiocaesium derived from the FNPP1. It tends to the positive relationship between the  $^{137}\text{Cs}$  activity concentrations above  $1.5 \text{ Bq m}^{-3}$ , which is the  $^{137}\text{Cs}$  activity concentrations before the FNPP1 accident, and the  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio. These relations in the SOJ suggests the contribution of  $^{137}\text{Cs}$  originated from the FNPP1 accident. However, this relationship might be unclear for the reviewers. Therefore, this Figure was removed in the revised manuscript.