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Rapid recirculation of FNPP1 derived radiocaesium suggesting new pathway of subtropical mode water in the western North Pacific to the Sea of Japan

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Abstract.

The rapid recirculation of ^{137}Cs derived from the Fukushima Nuclear Power Plant Accident (FNPP1-^{137}Cs) occurred in the Sea of Japan (SOJ) in several year timescale after released to the environment in March 2011. The recirculation of FNPP1-^{137}Cs had started in 2012 and reached to the maximum in 2015/2016 in the East China Sea (ECS) and the western North Pacific Ocean. The recirculation of FNPP1-^{137}Cs has been still continued in the coastal site of Sea of Japan in the year of 2016. The re-circulated FNPP1-^{137}Cs activity concentrations showed subsurface peak in the seawater of which density correspond to the Subtropical Mode Water (STMW). These suggests that FNPP1-^{137}Cs injected into the western North Pacific Ocean at south of Kuroshio were subducted into ocean interior just after the accident, then transported southward/southwestward. A part of FNPP1-^{137}Cs in STMW entered into the ECS between Kyushu Island and Okinawa Islands. Then it obducted in the region west of Kyushu Island, north of the ECS, following then entered into the SOJ associated with northward transport with Tsushima Warm Current in several year time scale. This rapid pathway might be new finding of transport process from the western North Pacific Ocean to the SOJ. Almost same value of the ^{134}Cs/^{137}Cs activity ratio in the coastal region of the Japanese islands (ECS, SOJ, and Ogasawara) also support this re-circulation route. The integrated amount of FNPP1- ^{137}Cs entered in the SOJ until 2016 was estimated to be 0.21±0.03 PBq, which corresponds to 5.1 (3.4-8.0) % of the total amount of FNPP1-^{137}Cs in the STMW. The integrated amount of FNPP1-^{137}Cs back to the North Pacific Ocean through the Tsugaru Straight in the surface layer was 0.1±0.02 Bq, which corresponds to 0.6 (0.4-1.0) % of the total amount of FNPP1-^{137}Cs in the STMW.

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1 Introduction

Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident in March 2011 released radiocaesium (^{137}Cs ($T_{1/2}$ 30.07 yr) and ^{134}Cs ($T_{1/2}$ 2.06 yr)) by atmospheric deposition and direct discharge of liquid contaminated water, mostly in March and April 2011 (Aoyama et al., 2016a; Buesseler et al., 2016; Hirose, 2016; Tsumune et al., 2013). The ^{137}Cs activity concentration in the surface seawater in the North Pacific Ocean after FNPP1 accident ranged from a few Bq m^{-3} to ca, 1 kBq m^{-3} (e.g., Aoyama et al., 2012, 2013; Honda et al., 2012; Kaeriyama et al., 2013, 2014). Basin scale measurement indicated that the FNPP1 derived radiocaesium located in a region from 25°N to 50°N and from 135°E to 135°W in April and May of 2011 (e.g., Aoyama et al., 2013; 2016a; Inomata et al., 2016; Tsubono et al., 2016). The radiocaesium were moved to eastward around 40°N with the North Pacific current system such as Kuroshio and Kuroshio extention with approximately 8 cm sec^{-1} and reached to 165°E during July-September 2011 and 180 meridian during January-March 2012 (Aoyama et al., 2012, 2013). After then, the speed become to slow (3.5 cm s^{-1}) from March 2012 to August 2014 (Aoyama et al., 2016a). FNPP1 derived radiocaesium were also found in southward in the North Pacific Ocean with a subsurface maximum at a depth of about 300 m (Kaeriyama et al., 2013, 2014, 2016; Kumamoto et al., 2013, 2017). Because the subsurface maxim of ^{137}Cs were found in the Subtropical Model Water (potential density (σ_θ) =25.0-25.6 kgm^{-3}) and Central Mode Water regions (σ_θ=26.0-26.5 kgm^{-3}) (Aoyama et al., 2016a, Kumamoto et al., 2014, 2015), these were concluded that atmospheric-deposited ^{137}Cs in south of the Kuroshio extension had been transported to eastward along with the surface ocean current as well as southward due to formation/subduction of mode water within 10 months after the FNPP1 accident.

Before the FNPP1 accident, ^{137}Cs have already existed in the North Pacific Ocean and its marginal sea. These were injected into the environment by the large scale atmospheric nuclear weapons testing occurred in the late 1950s and early 1960s and the Chernobyl accident in 1986 (Aoyama et al., 2006; Inomata et al., 2009). Furthermore, dumping of radioactive wastes by Russia and former USSR occurred in the north part of the Sea of Japan, although there is no significant increase of the activity concentration (Miyao et al., 1998). The higher global fallout occurred in the Kuroshio and Kuroshio extension areas at 20-40°N in the 1950-1960s (Aoyama et al., 2006). The global-fallout ^{137}Cs were eastward transport with Kuroshio and Kuroshio extension within several years and reached to the eastern North Pacific Ocean. Subsequently, ^{137}Cs activity concentrations in the eastern North Pacific Ocean were decreased associated with subduction to the subsurface layer (Inomata et al., 2012). In 2000s, just before the FNPP1 accident, the ^{137}Cs activity concentrations were ranged from 1 to 2.5 Bq m^{-3} with homogenous distribution, although a relatively high activity in the western part of the subtropical gyre in the North Pacific Ocean (exceed 2 Bq m^{-3}) and South Pacific Ocean (exceed 1.5 Bq m^{-3}) (Aoyama et al., 2012). At the FNPP1 accident, mainly March-April 2011, FNPP1-derived ^{137}Cs and ^{134}Cs (FNPP1-^{137}Cs, FNPP1-^{134}Cs) were released in the North Pacific Ocean. Therefore, there are two source of ^{137}Cs, global fallout ^{137}Cs (Global fallout-^{137}Cs) and FNPP1-^{137}Cs in the North Pacific Ocean and its marginal sea. In contrast, ^{134}Cs derived from global fallout had already decayed due to the shorter lifetime. Actually, ^{134}Cs derived from the Chernobyl accident had already less than the detection limit in 1993 (Miyao et al., 1998). The ^{134}Cs, therefore, is used as a chemical tracer of radiocaesium derived from the FNPP1 accident.

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After the several years later at the FNPP1 accident, it appeared the increase of ^{137}Cs activity concentration in the Sea of Japan (SOJ). As preliminary results, Aoyama et al. (2017) had reported that the increased radiocaesium in the coastal region of SOJ from November 2015 to March 2016 was resulted by re-circulation of FNPP1 derived radiocaesium. The SOJ locate among the region between Eurasia Continent and the Japanese archipelago. The area is 1008000 km^2 and a mean depth of 1667 m (Menard and Smith, 1966). The SOJ is divided into two regions around the 40°N, located the Polar Front. In the south of Polar Front, the seawater is characterized as higher temperature and higher salinity due to the Tsushima Warm Current (TWC). The increased FNPP1-^{137}Cs radioactivity in the SOJ suggests the possibility of shorter transport route from the North Pacific Ocean to the SOJ through the ECS rather than previously considered that the main part of subducted Global fallout-^{137}Cs will be expected return to the coastal region of Japan along sub-tropical gyre in a few decade time scale (Aoyama et al., 2008a). Although behaviors of radiocaesium in shallower layers have not still understood well, combining the temporal and spatial variation of FNPP1 derived ^{137}Cs and fallout ^{137}Cs, it is deduced that the ocean transport processes may have the two different time scales in the subtropical gyre. We discussed about the FNPP1-^{137}Cs activity concentrations in the SOJ and these recirculation pathway from the western North Pacific Ocean to the SOJ in time scale of several years. We estimated total amount of FNPP1-^{137}Cs which were transported to the SOJ from the subtropical gyre in the western North Pacific Ocean. We also discussed about the long-term trend of ^{137}Cs in the SOJ.

2 Sampling and measurements

After the FNPP1 accident, there are many measurements of radiocaesium in the SOJ and western North Pacific Ocean (Fig. 1). In order to elucidate the temporal and spatial distributions of the radiocaesium activity concentrations, it is necessary to use as many data as possible. We, therefore, compiled available all data from literature and report researches. Most of data before the FNPP1 accident was included into the database, “Historical Artificial Radionuclides in the Pacific Ocean and its Marginal Seas (HAM database)” (Aoyama and Hirose, 2003 and their updated version). The part of data after the FNPP1 accident were shown in Aoyama et al. (2016a).

We also focus on the Japanese government 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 conducted once a year (from middle of May to early June). Near the Aomori sites, off shore the Rokkasho Reprocessing Plant (39.5-41.4°N, 141.5-142.3°E), monitoring was also conducted twice a year (May and October). Monitoring data (304-01(33.0°N, 127.7°E), 105-11(37.3°N, 131.3°E)) by Korean government was also used to this analysis (Korea Institute of Nuclear Safety, 2011, 2012, 2013, 2014, 2015, 2016) (Fig. 1). In these monitoring sites, the measurements were conducted four times (February, April, August, October) in a year.

Because most of the monitoring data did not detect ^{134}Cs due to lower capability of measurements, we also collected seawater samples to investigate the ^{134}Cs activity concentrations. The volume of samples ranged from a few liters to ten liters.

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Radiocaesium in the sample seawater was extracted by ammonium phosphomolybdate (AMP)/Cs compound method by improved by Aoyama and Hirose (2008b). The AMP/Cs compound was measured using ultra-low-background gamma-ray detectors in Low Level Radioactivity concentration Laboratory, Kanazawa University (Hamajima and Komura, 2004) to measure ^{134}Cs as well as ^{137}Cs. Total amount of ^{134}Cs and ^{137}Cs in the sample was expected to be only a few mBq and around a few to 20 mBq, respectively. The detail of measurement has been described in Aoyama and Hirose (2008b) and Aoyama et al. (2016a). All radioactivity concentration shown in this research were decay corrected at the time of the samples collected. In addition to this, we also showed the activity concentration ratio of radiocaesium at the time to the FNPP1 accident (11 March 2011).

3 Results

3.1 Increased ^{137}Cs activity concentrations in the surface seawater

Fig. 2 shows the time variation of ^{137}Cs activity concentrations in the surface seawater in the SOJ after the 1960's. The circle and error bars mean the measurement value and these standard deviations. The half-year average value was also plotted. Location of the measurement data observed before (1740 records) and after (473 recrods) March 2011 also show in Fig. A1. The activity concentrations are exponentially decreased from the years of 1950-1960. The higher activity concentrations in the 1950 and 1960 was derived from the atmospheric nuclear weapons testing. Small increase at 1986 were derived from the Chernobyl accident (Miyao et al., 1998). In the 2000s, the ^{137}Cs activity concentrations in the surface seawater become to be small (1.9-2.2 Bq m^{-3} in the year of 2005) and the apparent half-residence time during the period from 1970 to 1990, which are mainly due to controlled by advection and diffusion, was estimated as 23 years (Inomata et al., 2009). After the FNPP1 accident, it appeared that the ^{137}Cs activity concentrations are gradually increased to about ~3.4 Bq m^{-3}. These are the same level in the year 1990s-2000s.

Fig. 3 shows the FNPP1 derived ^{137}Cs activity concentrations (FNPP1–^{137}Cs) and ^{134}Cs/^{137}Cs ratio at several monitoring sites ((a) Tomari, Aomori area, (b) Niigata, Fukui, Ishikawa, Shimane, (c) Saga, Kagoshima) measured by Japanese government around the Japanese islands. These monitoring sites locate along a branch of Kuroshio in the ECS and TWC. In these, the data measured by other researches were also added. FNPP1-^{137}Cs was estimated by the difference between the measurement value and the extrapolation value of the half-year average value in the SOJ during the period from 2000 to 2010 (Fig. A2). The long term variation of ^{137}Cs activity concentrations and FNPP1-^{137}Cs at each site are also shown in Fig. A3-10. FNPP1-^{137}Cs in 2011, particularly Aomori area (Fig. A9), were influenced the contribution from the atmospheric deposition. FNPP1-^{137}Cs were gradually increased between 2012 and 2014 or 2015 in all sites. FNPP1-^{137}Cs at 2016 were almost same level to those in 2015 or slight decrease were found in the Saga and Kagoshima sites. The largest value of FNPP1-^{137}Cs were at Kagoshima and Saga, on the other hand the smallest one was found at Tomari and Aomori area (Fig. 3a-c). The lower activity at Niigata (Fig. 3b) were collected at different monitoring site, near Sado islands (Niigata off). The ^{134}Cs/^{137}Cs activity concentration ratios in 2015/2016 were ranged from 0.36 to 0.51 (Fig. 3d-f).

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Fig. 4 shows the temporal variation of ^{137}Cs activity concentrations and FNPP1-^{137}Cs at two Korean monitoring sites (western site of the SOJ). Although the data number was small, ^{137}Cs activity concentrations were seasonal variation with higher in spring and the largest activity concentration (about 3 Bq m^{-3}) was observed in spring, 2015. (Fig. 4a). Except for the data measured in 2011, which was contributed from atmospheric deposition, FNPP1-^{137}Cs were gradually increased until 2015 and tended to decrease in 2016 (Fig. 4b).

3.2 Vertical distributions of ^{137}Cs activity concentrations

Fig. 5 shows the vertical distribution of ^{137}Cs activity concentration against depth and potential density (σ_{θ}, kg m^{-3}), and these horizontal distributions from 2011 to 2016 in the SJPN. The ^{137}Cs activity concentrations show subsurface maximum around 300 m depth and the activity concentrations were maximum in 2012-2013. After 2014, ^{137}Cs activity concentrations were gradually decreased (Fig. 5a). The vertical distributions of ^{137}Cs activity concentrations against the σ_{θ} were gradually increased with increasing σ_{θ} with a peak around 25.5 σ_{θ}, mainly existed in the STMW (25.0-25.6 kg m^{-3}) (Fig. 5b). Higher activity concentrations were found along 132°E (the region between Kagoshima and Amami islands) and along 142°E in the SJPN (Fig. 5c).

In the ECS, ^{137}Cs activity concentrations also show subsurface peak with about 100 m and increased until 2015 and tend to decrease in 2016 (Fig. 6a). The higher activity concentration of ^{137}Cs in 2014-2015 were found at the density range from 24.5 to 25.5 σ_{θ}, corresponds to the STMW (Fig. 6b). Horizontal distribution of ^{137}Cs indicated that the ^{137}Cs activity concentrations were relatively large at north of 30°N (Fig. 6c).

Fig. 7 display the vertical distribution of ^{137}Cs activity concentrations against σ_{θ} in the SON. In the monitoring sites (Kagoshima, Saga, 314-01) in the ECS, western Kyusyu, the ^{137}Cs was found in the seawater which corresponds to the potential density at 22-26 σ_{θ}. The largest activity concentration was found in 2015 and gradually degreased in 2016. On the other hand, ^{137}Cs in the monitoring sites in the eastern coast site of SOJ (Shimane, Fukui, Niigata, Aomori, Tomari) along the eastern TWC were found in more denser seawater (24-28 σ_{θ}). The σ_{θ} in ^{137}Cs existed seawater become to be large associated with northward transport. The ^{137}Cs activity concentrations in 2015/2016 were almost similar. In the case of trend at station 105-11, which are enroute along the western TWC, the ^{137}Cs existed in the seawater corresponding to the 25.5-27.5 σ_{θ} indicating cooling at surface layer after transported from the Tsushima Straight, then more dense seawater rather than STMW. Decrease of the ^{137}Cs activity concentrations started in 2016 at station 105-11.

4 Discussion

4.1 Pathway from subsurface layer in the western North Pacific Ocean to ECS and SOJ

Before the FNPP1 accident, ^{137}Cs activity concentration showed maxima in the surface layer and gradually decreased with increasing depth in the SOJ (Miyao et al., 1998). However, ^{137}Cs activity concentrations after the FNPP1 accident showed

subsurface peak and these were existed in the layer of which density corresponds to that of STMW, as shown in the vertical distributions in the monitoring sites in the coastal region in Japan (Fig. 7).

There are three pathways of entering FNPP1 derived radiocaesium in the North Pacific Ocean. The dominant pathway was the injection at north of Kuroshio front by atmospheric deposition. Deposited radiocaesium north of Kuroshio was transported eastward by the Kuroshio and Kuroshio extention at mid-latitude in the North Pacific Ocean (Aoyama et al., 2016a). Another two pathways were subduction in the western North Pacific Ocean associated with the formation of CMW and STMW from surface layer (Aoyama et al., 2016a; Kumamoto et al., 2014; Kaeriyama et al., 2014, 2016). We observed that the re-circulated FNPP1-^{137}Cs activity concentrations showed subsurface peak in the seawater of which density correspond to that of STMW. This suggests that FNPP1-^{137}Cs injected into the western North Pacific Ocean (south of Kuroshio) were subducted into ocean interior just after the accident, then transported southward/southwestward. A part of FNPP1-^{137}Cs in STMW entered into the East China Sea (ECS) between Kyushu Island and Okinawa Islands. The vertical distribution of ^{137}Cs activity concentrations against the potential density (Fig. 6b) suggest the signature of the obduction of intruded STMW in the northern ECS. The subsurface peak of ^{137}Cs activity concentrations near Kagoshima (T3-T5) and almost same activity concentrations at vertical distribution near TWC (T2) support the obduction of FNPP1-^{137}Cs in this region (Fig. A11b-e). Following then FNPP1-^{137}Cs were entered into the SOJ associated with northward transport with Tsushima Warm Current in several year time scale.

This pathway might be new funding for ocean circulation in the western North Pacific Ocean. This recirculation route has several year time scale from the North Pacific Ocean to the SOJ. In the SJPN, ^{137}Cs activity concentration was maximum in 2012 and 2013, whereas the maximum activity concentrations were found in 2014/2015 in the ECS (Kagoshima, Saga) and the western SOJ (314-01, 105-10) (Fig. 7). In these sites, decrease of ^{137}Cs activity concentrations have already started in 2016. In the eastern SOJ (Shimane, Fukui, Ishikawa, Niigata, Aomori, and Tomari), the ^{137}Cs activity concentrations were almost same in 2015/2016. The time lag of peak activity concentration of ^{137}Cs suggests that the center of FNPP1 derived radioactive labelled seawater had already passed though in this transport route (Fig. 7). Although the increase of ^{137}Cs activity concentrations have still continued in the ECS and the western SOJ at the time of 2016, the ^{137}Cs activity concentrations will be decrease within 1-2 years.

Fig. 8 shows the latitudinal distribution of ^{134}Cs/^{137}Cs activity concentration ratios which are decay corrected to 11 March 2011. The ^{134}Cs/^{137}Cs activity concentration ratio ranged from 0.1 to 0.72 and the ratio at Ogasawara (OGA, 27.09°N, 142.19°E) and the ECS are almost same to those in the SOJ (Fig. 8a). Considering that the released ^{134}Cs/^{137}Cs at the FNPP1 accident were almost 1, FNPP1-labelled seawater was mixed with the Global-fallout labelled seawater (no contaminated seawater by the FNPP1-^{137}Cs) during transport, resulting with the decay corrected ^{134}Cs/^{137}Cs activity concentration ratio should be changed by the seawater mixing processes. The highest activity concentration ratio (0.72), which was found near Kagoshima, might be signal of the larger contribution of the FNPP1-^{137}Cs and ^{134}Cs associated with the obduction processes. In contrast, relatively lower ^{134}Cs/^{137}Cs activity concentration ratio in the region between 30 and 34°N suggests that contribution from global fallout ^{137}Cs were larger than those in the re-circulated seawater. In north of 34°N at the coastal site

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of SOJ, the $^{134}Cs/^{137}Cs$ activity concentration ratios were almost same as those in south 30°N, suggesting that the recirculated FNPP1 derived radiocaesium is entered along with TWC.

The scattering plot between ^{137}Cs activity concentration and $^{134}Cs/^{137}Cs$ activity concentration ratio is shown in Fig. 9. The $^{134}Cs/^{137}Cs$ activity concentration ratio are increased with increasing ^{137}Cs activity concentration (> about 1.5 Bq m^{-3}). In particular, the highest activity concentration ratio (0.72) was found in relatively high ^{137}Cs activity concentration (2 Bq m^{-3}). The ratio in the ECS, SOJ, and OGA are almost same. This is also a signature of re-circulation of FNPP1-^{137}Cs transport. The lower ^{137}Cs activity concentration (<1.5 Bqm^{-3}) would be less contaminated by FNPP1 and have larger contribution from the global-fallouted ^{137}Cs. In other words, these reflect the different large scale seawater circulation.

4.2 Integrated recirculated FNPP1-^{137}Cs amount budget in the Sea of Japan

Several researches reported that the atmospheric deposited FNPP1-^{137}Cs in the North Pacific Ocean were 11.7-14.8 PBq (Aoyama et al., 2016b; Tsubono et al., 2016). The direct discharged FNPP1- ^{137}Cs amount were 3.5±0.7 PBq (Tsumune et al., 2012, 2013). Considering the atmospheric fallout and direct release from the FNPP1, the FNPP1-^{137}Cs inventory in the North Pacific Ocean was estimated to 15.2-18.3 PBq in April-May 2011 by Aoyama et al. (2016b). The inventory estimated by different methods (optimal interpolation analysis 15.3±2.6 PBq, Inomata et al., 2016; Ocean circulation model 16.1±1.4 PBq, Tsubono et al., 2016) were almost same value. In these, 4.2±1.1 PBq of ^{134}Cs (FNPP1-^{137}Cs) decay corrected on 11 March 2011 were intruded into the subsurface water in the western North Pacific Ocean associated with the formation of STMW (Kaeriyama et al., 2016).

In this study, we confirmed the ^{137}Cs recirculation route from the SJPN to the SOJ via the ECS. The FNPP1-^{137}Cs in the western North Pacific Ocean are subducted to 200-300m depth (σ_θ 25-26.3) associated with the formation of the STMW. These are westward transported in the southern region of the Japanese islands, the western North Pacific Ocean, and obducted to the surface layer around the west of Kyushu islands, the northern part of ECS (>30°N). In the northern ECS, FNPP1-^{137}Cs is transported northward and enter the SOJ through the Tsushima Straits by TWC. The TWC flowed into the Japan Sea through two channels, the channel east and channel west. Previous physical oceanographic research revealed that the annual average volume transport of the TWC into the Sea of Japan for the 10 years average (1997-2007) was 2.65±0.41 × 10^6 m^3 s^{-1}, and these through east channel and west channel of the Tsushima islands were 1.20±0.20 × 10^6 m^3 s^{-1} and 1.45±0.34 × 10^6 m^3 s^{-1}, respectively (Fukudome et al., 2010). The volume transport along Tsushima Straight though the east channel and west channel showed seasonal variation as minimum in January and maximum in March at east channel and minimum in January and maximum in October at east channel. The seasonal variation of seawater volume in west channel is consistent with the seasonal variation of FNPP1-^{137}Cs activity concentration in the station 105-11 at east coast of Korean Peninsula (37.3°N, 131.3°E, Fig. 1). However, in east channel, the monitoring data in Japan (except Aomori area, two times per year) was only once a time per year. It is, therefore, difficult to discuss the seasonal variation based on the monitoring data by Japanese Government, although the variation of ^{137}Cs activity concentrations at Aomori were similar to the seawater

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volume variation (higher activity concentrations in October is consistent with those in higher sweater volume transport) (Fig. A9b). The annual flux of FNPP1-^{137}Cs at the entrance of the SOJ, western TWC, eastern TWC were estimated as the product of annual average FNPP1-^{137}Cs activity concentrations (Saga and 304-01 (using average value), Shimane, and 105-11) and annual volume transport at Tsushima straight for the period from 2012 to 2016 and integrated for whole period to estimate total amount of FNPP1-^{137}Cs pass through Tsushima Straight. For the total amount of outgoing flux though Tsugaru Straight, the annual flux of FNPP1-^{137}Cs were estimated as the product of annual average FNPP1-^{137}Cs activity concentration at stations off Aomori and volume transport at Tsugaru straight ($1.30\pm0.3 \times 10^6$ m^3 s^{-1}) for the period from 2012 to 2016 and integrated for whole period to estimate total amount of FNPP1-^{137}Cs pass through Tsugaru Straight.

Table 1 shows the estimated flux (inflow) of recirculated FNPP1-^{137}Cs. In the ECS (Saga and 304-11 monitoring sites's average), just the entrance region, the accumulated FNPP1-^{137}Cs amount from 2012 to 2016 is estimated to be 0.21±0.03 PBq, which correspond to 5.1 (3.4-8.0) % of the amount FNPP1-^{137}Cs in STMW. Connected with the separation of TWC, the flux at east channel was estimated to be 0.12±0.02 PBq, which correspond to 2.8 (1.9-4.4) % of the FNPP1-^{137}Cs injected into the STMW. On the other hand, the flux at west channel by using the 105-11 Korean monitoring site was estimated to be 0.08±0.02 PBq, which correspond to 2.0 (1.2-3.3) % of the FNPP1-^{137}Cs injected into the STMW. The integrated outflow at the Aomori off where the exit region of seawater from the SOJ in the North Pacific Ocean was estimated to be 0.09±0.02 PBq, which correspond to 1.6-3.6 % of the total amount in the North Pacific Ocean derived from the FNPP1 accident. Remaining 0.03±0.01 PBq (0.4-1.0 %) would transport to the northern part of SOJ (west of Hokkaido) or part of this might be transported to deep region associated with deep convection and surface mixing.

5 Conclusion

A part of FNPP1 derived radiocaesium in the North Pacific Ocean already re-circulate through in sub-surface layer in several year time scale. These are a signature of existing the new pathway of ocean current from the North Pacific Ocean to the SOJ, which are shorter time scale in comparison with the accepted view: The FNPP1-^{137}Cs subducted into the STMW in the western North Pacific Ocean were westward transport in the subsurface layer and obducted in the west of Kyushu islands (northern ECS), following then enter the SOJ associated with northward transported TWC (Fig. 10). The re-circulation had already started from 2012 and the increase of ^{137}Cs activity concentrations have still continued in the SOJ at the time of 2016. The slight decrease of ^{137}Cs activity concentrations were observed in the upstream region (SJPN, ECS). The ^{137}Cs activity concentration will be decrease within 1-2 years. Almost similar ^{134}Cs/^{137}Cs activity concentration ratio in the surface layer in the ECS, SOJ, and OGA (located in SJPN) also support this new transport route.

The integrated amount of recirculated FNPP1-^{137}Cs for the period from 2012 to 2016 was estimated to be 0.21±0.03 PBq, which corresponds to 5.1 (3.4-8.0) % of the amount of FNPP1- ^{137}Cs in the STMW. A small part of recirculated FNPP1-^{137}Cs (0.09±0.02 PBq; 1.3-3.6 %) passed through Tsugaru Straight and already back to the North Pacific Ocean.

Author contribution

Y. Inomata (corresponding author) conducted data analysis and preparing the manuscript with contributions from all co-authors. Prof. M. Aoyama developed the database of radioactivity and pretreatment of seawater samples for measurement of radiocaesium. Prof. Hamajima measured the radiocaesium activity concentrations. Prof. Yamada organized the seawater sampling for radiocaesium measurement.

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Figure 1: Location of the sampling points (open circles) after the FNPP1 accident. Red large circles are monitoring sites by the Japanese government. Blue circles are monitoring sites by the Korean government. Around Japan was divided into 3 regions; SOJ, ECS, and SJPN (<141.5◦E).

Figure 2: Temporal variations of ^{137}Cs activity concentrations in the surface seawater in the Sea of Japan during the period from1960 to 2016. Circles are measurement value, and red circles are data after the accident (2011March11). Black circles are the 0.5 years averaged value until 2010.

Figure 3: Temporal variations of FNPP1-^{137}Cs activity concentration (left panel) and ^{134}Cs/^{137}Cs activity concentration ratio (right panel) in the surface seawater in the monitoring sites in the Sea of Japan after 2011. FNPP1-^{137}Cs: (a) Tomari, Aomori, (b) Niigata, Ishikawa, Fukui, Shimane, (c) Saga, Kagoshima. ^{134}Cs/^{137}Cs activity ratio: (d) Tomari, Aomori, (e) Niigata, Ishikawa, Fukui, Shimane, (h) Saga, Kagoshima.

Figure 4: Temporal variations of (a) ^{137}Cs activity concentrations and (b) FNPP1-^{137}Cs activity concentrations in the surface seawater in the Korean monitoring sites after 2011.

Figure 5: Vertical distributions and horizontal distributions of ^{137}Cs activity concentrations in the SJPN. (a) Vertical distribution against depth, (b) vertical distribution against σ_θ, (c) horizontal distribution. For (a) and (b), color indicates the collected time (year). In the case of (c), color indicates the ^{137}Cs activity concentrations (Bq m^{-3}). The data in the surface seawater were shown in circle, and the data obtaining vertical profile (including surface seawater) were shown in square.

Figure 6: Vertical and horizontal distributions of ^{137}Cs activity concentrations in the ECS. (a) Vertical distribution against depth, (b) vertical distribution against sigma, (c) horizontal distribution. For (a) and (b), color indicates the collected time (year). In the case of (c), color indicates the ^{137}Cs activity concentrations (Bq m^{-3}). The data in the surface seawater were shown in circle, and the data obtaining vertical profile (including surface seawater) were shown in square. The red circles with arrows (T1-T5) show the data having the temporal variation (Fig. A.11).

Figure 7: Vertical distributions ^{137}Cs activity concentrations against s_q in the monitoring sites along the Tsushima Warm Current. (a) Kagoshima, (b) Saga, (c) Shimane, (d) Fukui, (e) Ishikawa, (f) Niigata, (g) Aomori, (h) Tomari, (i) 314-01, (j) 105-11. Color indicates the collected time.

Figure 8: Latitudinal and horizontal distributions of ^{134}Cs/^{137}Cs activity concentrations ratio measured in the coastal site of SOJ and ECS in 2015-2016. The value was radioactive decay corrected to 11th March 2011. The data measured in OGA (red circles in (a)) were also added. (a) Latitudinal distribution, (b) horizontal distribution.

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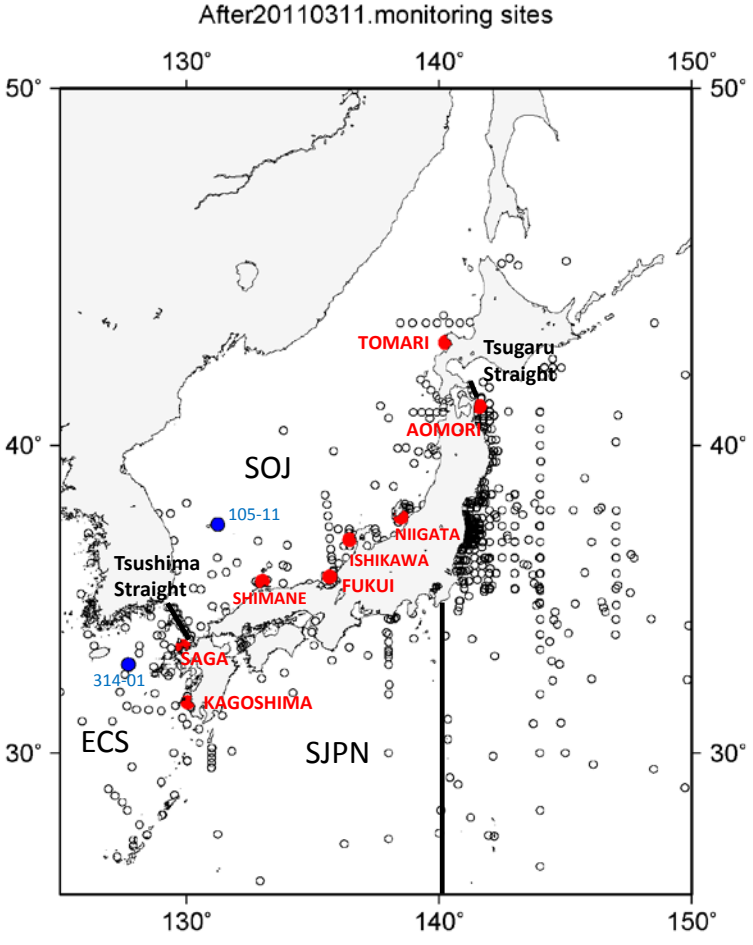

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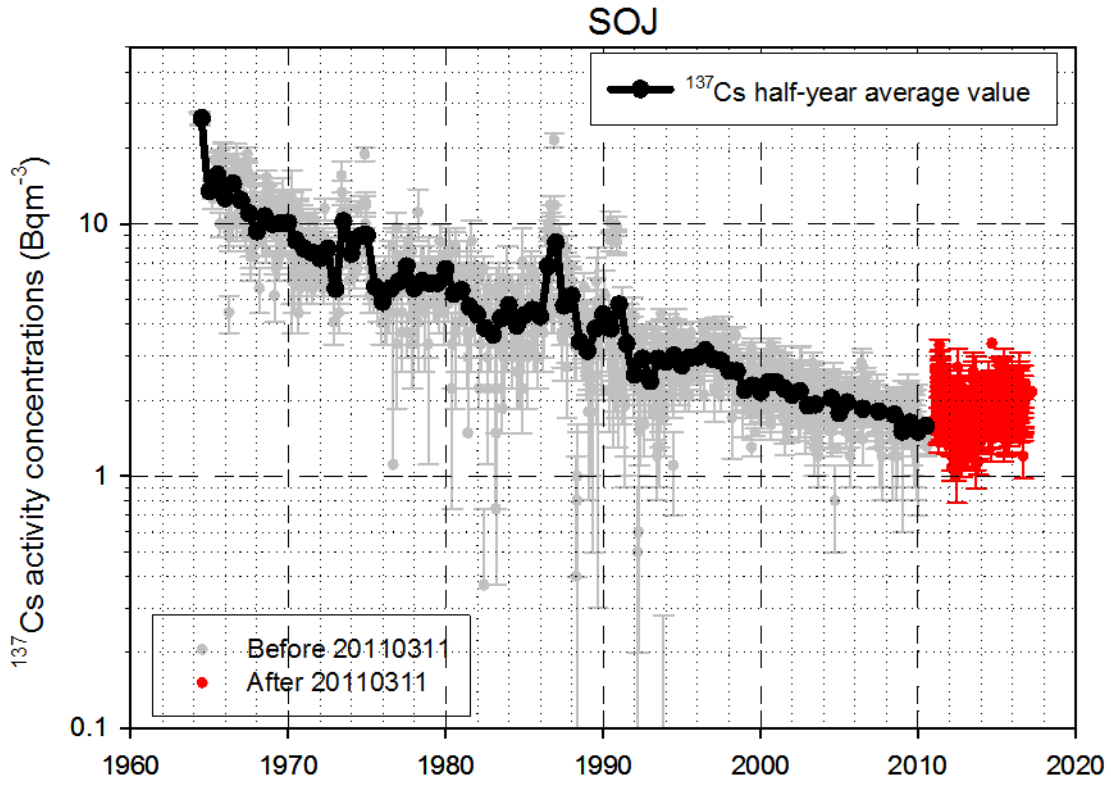

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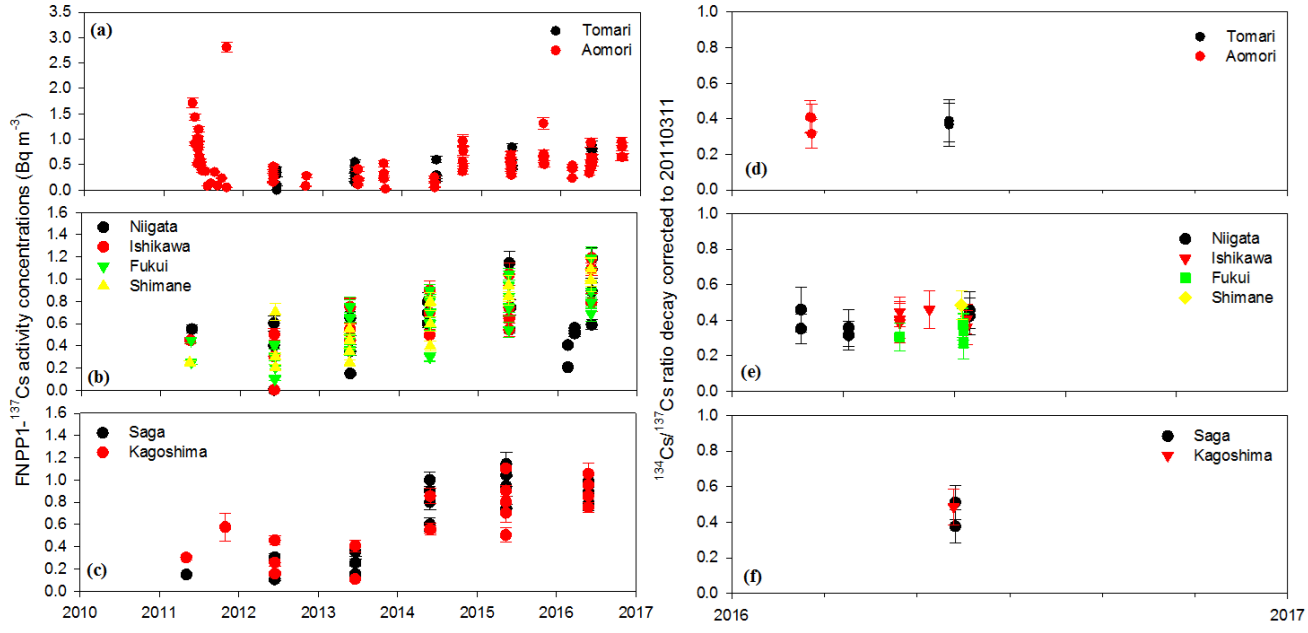

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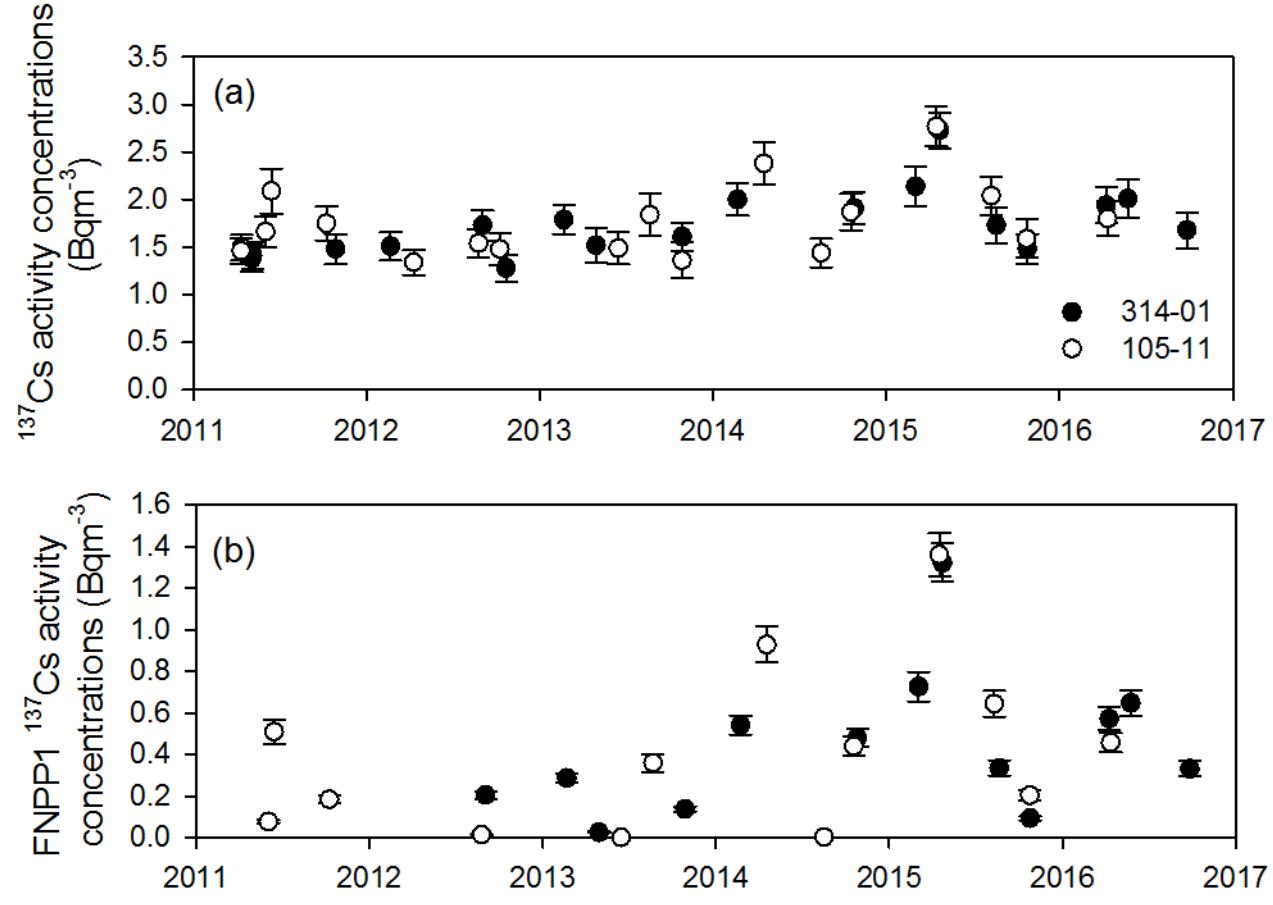

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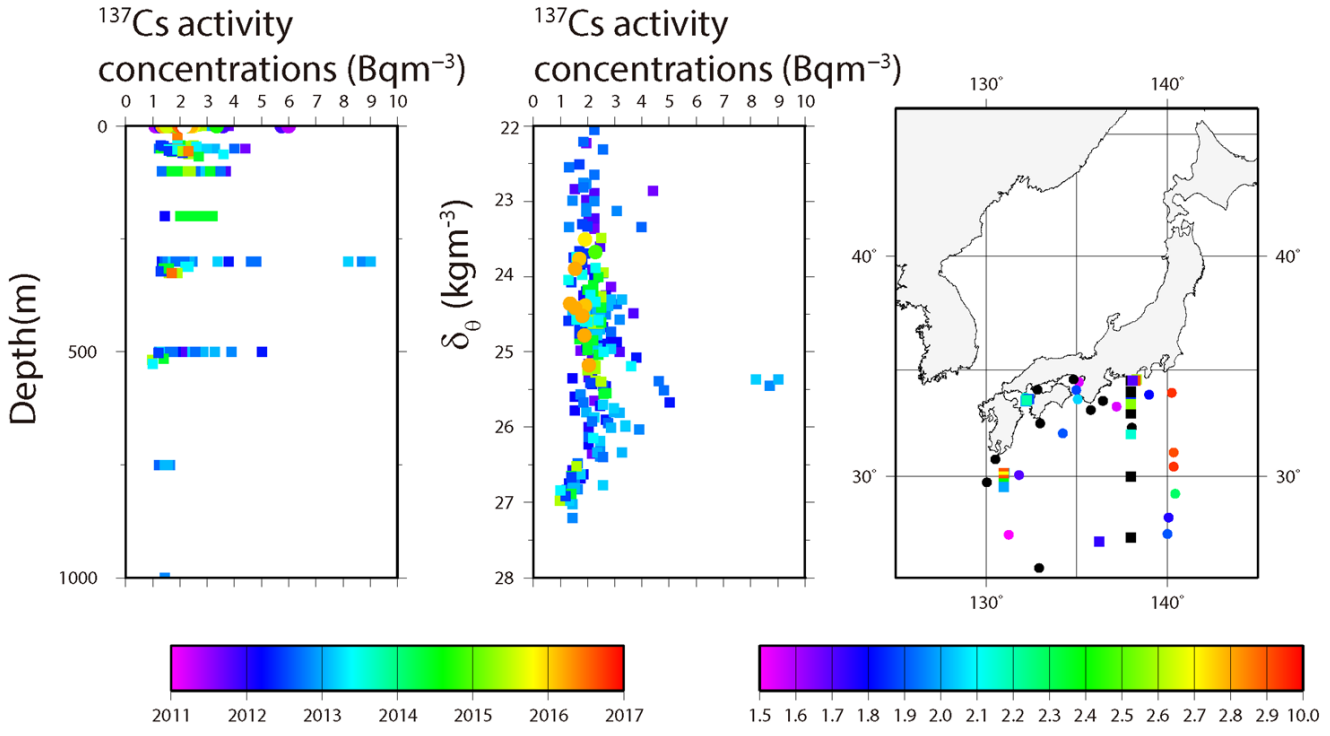

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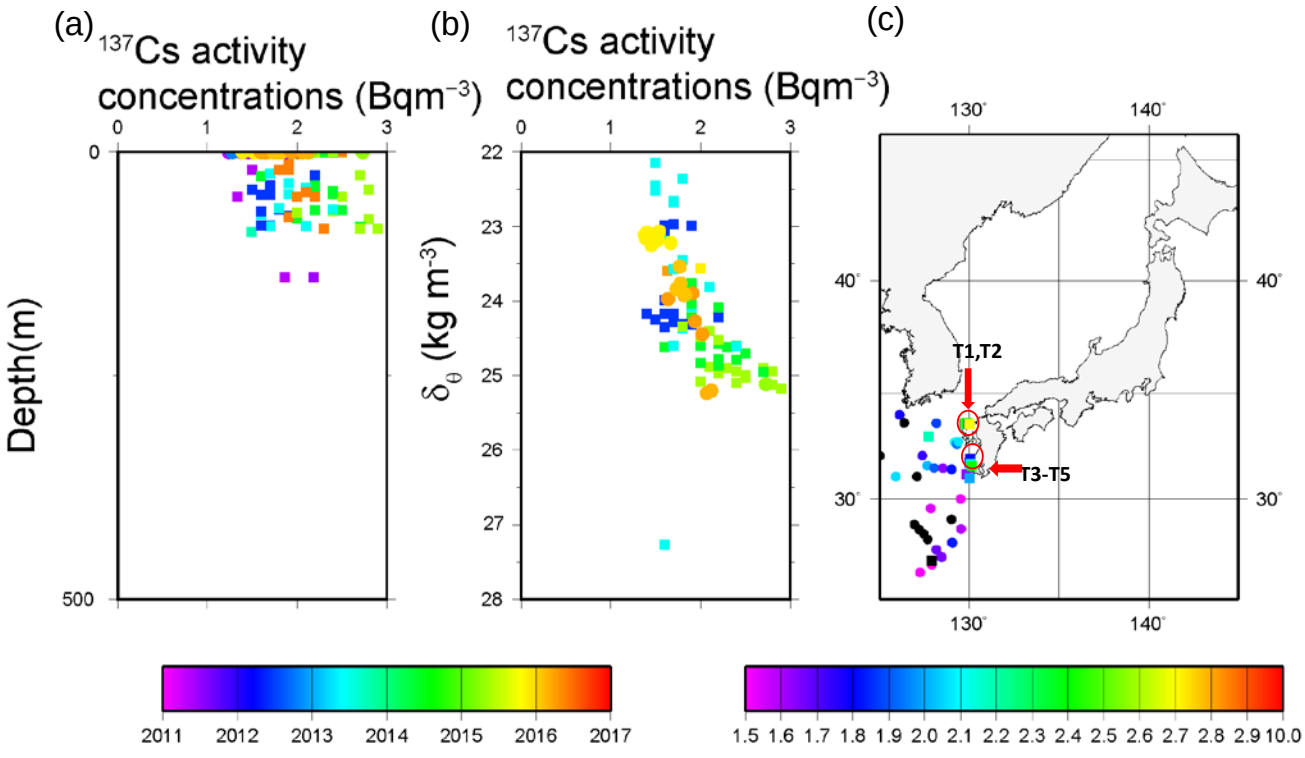

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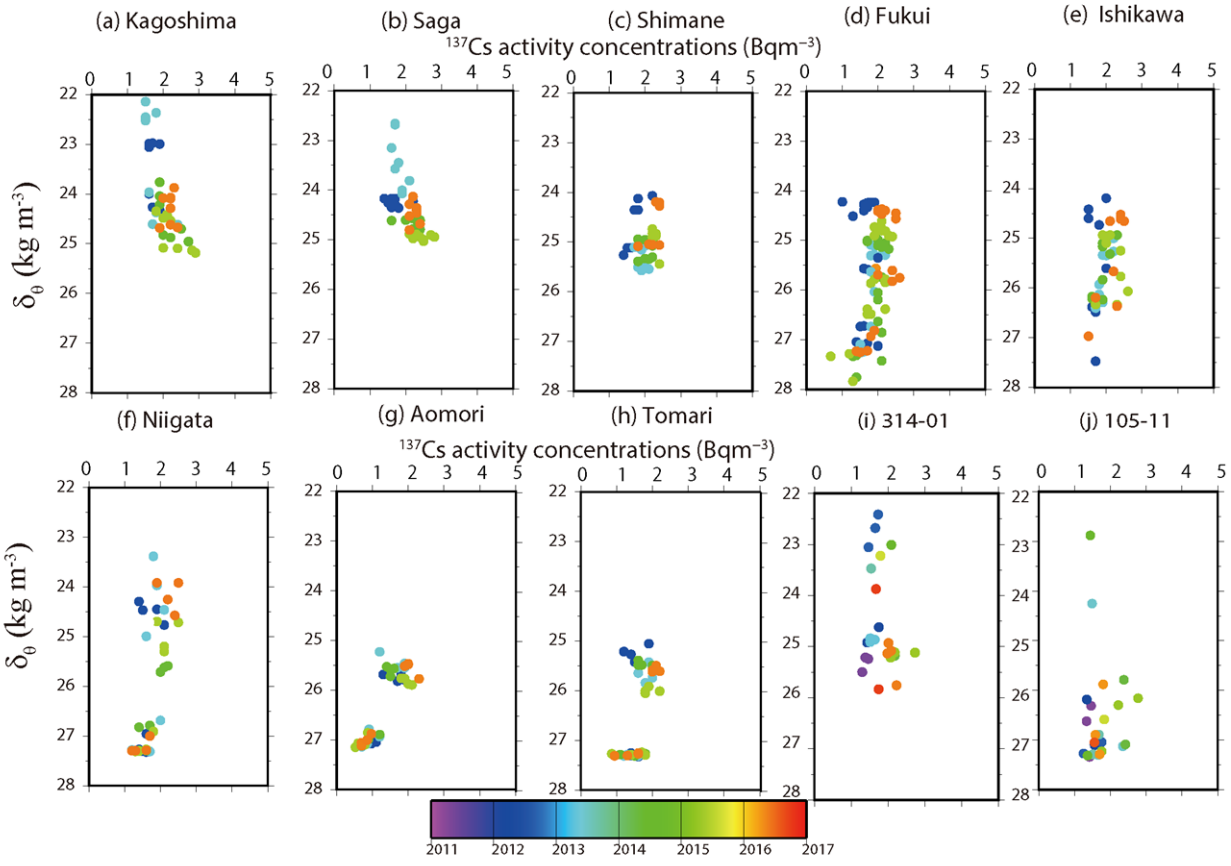

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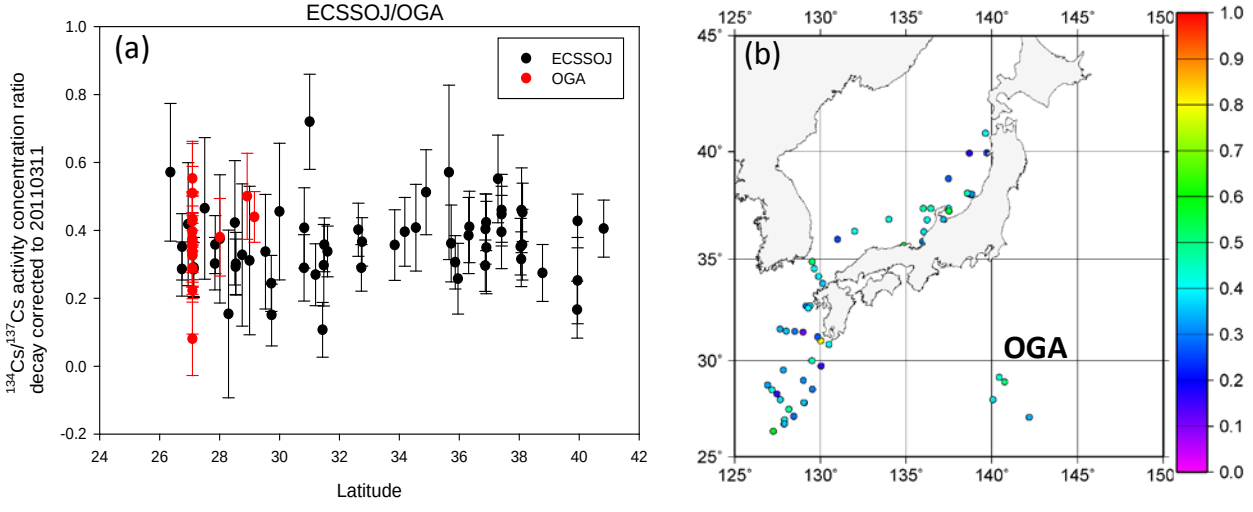

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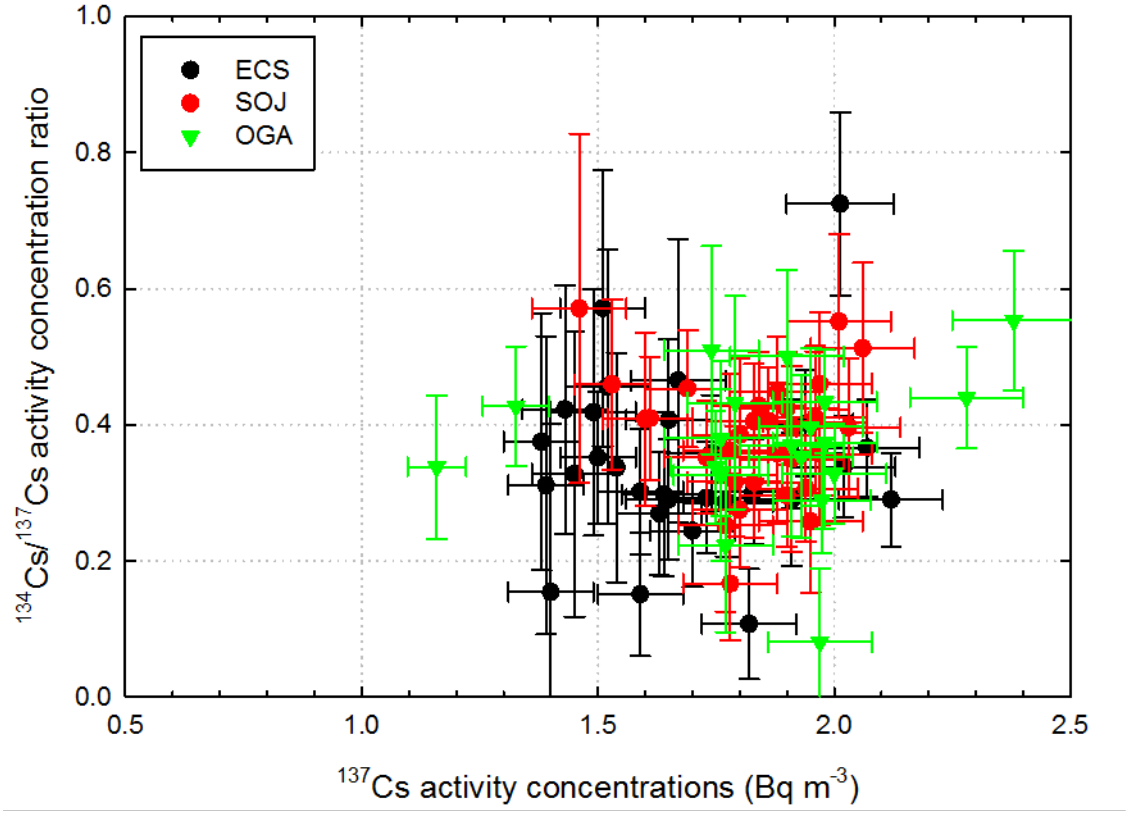

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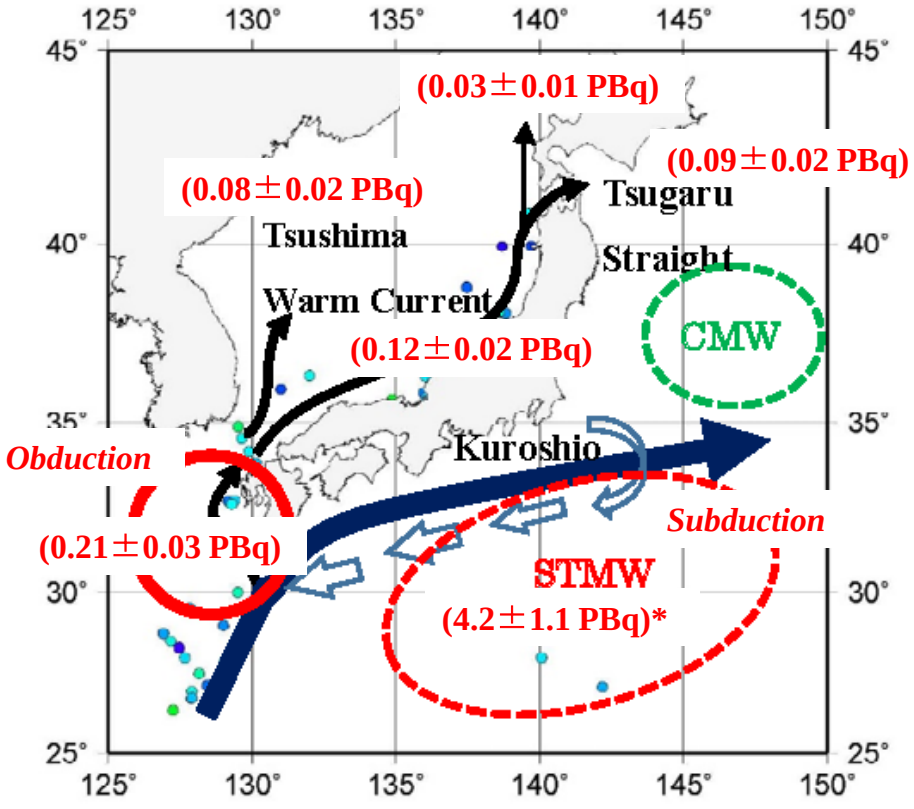

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	Station	Flux and Standard deviation (PBq*)
Inflow	ECS	0.21 ± 0.03
East flow with Tshushima Straight	Shimane	0.12 ± 0.02
West flow with Tsushima Straight	K105-11	0.08 ± 0.02
Out flow to the North Pacific Ocean	Aomori area	0.09 ± 0.02
North flow from the Tsugaru Straight	Tomari	0.03 ± 0.01

The value was decay corrected to 11th March 2011. ECS were estimated by using the average value of K314-01 and Saga monitoring sites. The Fluxes (*) were estimated by sum of the duration from 2012 to 2016.