

Abstract

The horizontal distribution of radioactive cesium (Cs) derived from the Fukushima Dai-ichi Nuclear Power Plant (FNPP) in the North Pacific is still unclear due to the limitation of direct measurement of the seawater in the open ocean. We present the result of direct observation of radioactive Cs in surface seawater collected from broad area in the western and central North Pacific in July, October 2011 and July 2012. We also conducted a simple particle tracking experiment to estimate the qualitative spatial distribution of radioactive Cs in the North Pacific. ^{134}Cs was detected at 94 stations out of 123 stations and ^{137}Cs was detected at all stations. The high ^{134}Cs and ^{137}Cs concentrations more than 10 mBq kg^{-1} were observed in the area where the northern part of Kuroshio extension at 144° E and 155° E in July 2011, in the area 147° E – 175° E around 40° N in October 2011, and the northern part of Kuroshio extension at 155° E and $175^\circ 30'\text{ E}$ in July 2012. Combining the result of direct observations and particle tracking experiment, the radioactive Cs derived from FNPP had been dispersed eastward to the central North Pacific during 2011. It was considered from the horizontal distribution that radioactive Cs was dispersed not only eastward but also north- and southward in the central North Pacific. Pronounced dilution process of radioactive Cs from FNPP during study period is suggested from temporal change in the activity ratio of $^{134}\text{Cs}/^{137}\text{Cs}$ which was decay corrected at 6 April 2011, and relationships between radioactive Cs and temperature.

1 Introduction

After the Tohoku earthquake of magnitude 9.0 and tsunami on 11 March 2011, a loss of electric power at the Fukushima Dai-ichi Nuclear Power Plant (hereafter FNPP) resulted in overheating the reactors and hydrogen explosions. Radioactive materials were released into the ocean not only atmospheric fallout but also the direct release and leaking of the heavily contaminated coolant water (cf. Buesseler et al., 2011).

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Radioactive cesium (Cs) with relatively long half-life time (2.07 yr for ^{134}Cs and 30.07 yr for ^{137}Cs) is a serious concern as contaminants in oceanic environment and marine biota, especially for fisheries products as a food (cf. Buesseler et al., 2012; Honda et al., 2012; Madigan et al., 2012).

The Japanese government conducted intensive monitoring on ^{131}I , ^{134}Cs and ^{137}Cs in seawater near offshore FNPP (MEXT, 2012), and those in fisheries products in the broad area around Japan (Fisheries Agency, 2012). While the information of radioactive contamination covering the broad area in the North Pacific is still quite limited (Aoyama et al., 2012, 2013). Some model experiments are discussed the dispersion of radioactive Cs from FNPP (cf. Kawamura et al., 2011; Bailly du Bois et al., 2012; Dietze and Kriest, 2012; Tsumune et al., 2012; Miyazawa et al., 2012) and estimated amounts of ^{137}Cs discharged directly into the ocean were ranged from 2.3 to 14.8 PBq, with considerable uncertainties (Masumoto et al., 2012).

In this study, we focus on the horizontal distribution of ^{134}Cs and ^{137}Cs in the western and central North Pacific based on the direct observation of surface seawater collected during 29 June 2011 and 1 August 2012 and discussed the horizontal distribution patterns of radioactive Cs in the North Pacific with a simple particle tracking experiment based on the sea surface velocity field.

2 Materials and methods

2.1 Sample collection

Surface seawater samples were collected on board with a bucket from broad area of western and central North Pacific. In June–July 2011, three north-south transects were set along with 144°E , 155°E and $175^\circ 30'\text{E}$ and seawater samples were collected at 32 stations (Fig. 1a). The total of 34 samples were collected in the area within 30°N – 41°N and 140°E – 176°E during October and November 2011 (Fig. 1b), and 57 samples were collected in the three north-south transects along with 144°E , 155°E

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and 175° 30' E during July and August 2012 (Fig. 1c). The seawater sample was not filtered, transferred into 20 L plastic bag and acidified to pH = ca 1.6 by adding 40 mL of concentrated nitric acid. The differences in the concentrations of radioactive Cs after the FNPP accident between filtered seawater and unfiltered seawater were negligible (Honda et al., 2012; Buesseler et al., 2012). Thus, the results of this study based on unfiltered seawater could be comparable with previous studies.

2.2 Analysis of ^{134}Cs and ^{137}Cs in seawater

The ^{134}Cs and ^{137}Cs in seawater samples were concentrated by adsorption onto ammonium phosphomolybdate (AMP) using a modified method described in elsewhere (cf. Aoyama et al., 2000, 2012; Aoyama and Hirose, 2008). The seawater sample was transferred to the container and weighed 18 kg. As carrier, 0.52 g of CsCl was added to 18 kg of sample and stirred at least one hour. Then 8.0 g of AMP were added to sample and stirred at least one hour. After settling the AMP (almost 14–16 h, but no longer than 24 h), decanted supernatant and AMP/Cs compound with a small amount of supernatant (up to 2 L) was transferred into a 5 L beaker. The AMP/Cs compound was collected onto glass fiber filter (GA-100, ADVANTEC co ltd.) by filtration and washed with nitric acid. The AMP/Cs compound was dried at 60–70°C for more than 48 h. Then, dried AMP/Cs compound was weighed to determine the weight yield of AMP/Cs compound. The weight yield of AMP/Cs compound were exceeded 95 %.

The ^{134}Cs and ^{137}Cs radioactivities in the AMP/Cs compounds were measured by Ge semiconductor detectors. The ^{134}Cs and ^{137}Cs radioactivities were determined by comparing the photopeak area corresponding to the energy of 605 keV, 796 keV for ^{134}Cs and 662 keV for ^{137}Cs obtained by more than 7200 s counting of the sample with that of the standard sample which had the same geometry and was tagged with known radioactivity. The detection limit of ^{137}Cs by the 7200 s counting was nearly 5.0 mBq kg⁻¹. When the ^{137}Cs had not been detected with 7200 s measurement, longer time measurements (> 40 000 s) had been carried out. The detection limits of ^{137}Cs by the more than 40 000 s measurements (≤ 1.4 mBq kg⁻¹) were almost the same level

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of the concentration of ^{137}Cs in the sea surface water in the North Pacific before the accident of FNPP (2.4 mBqkg^{-1} ; cf. Povinec et al., 2004).

2.3 Particle tracking experiment

A numerical particle tracking experiment was carried out to investigate the sea surface horizontal distribution pattern of radioactive Cs released from FNPP. The time series of sea surface velocity field we used was estimated every 7 days and 0.25 degree for large and meso scale motions by combining satellite altimeter data and sea surface drifter data (Ambe et al., 2010). The estimation method was based on that of Uchida and Imawaki (2003), estimation processes for the two types of data were improved by matching those two representative spatiotemporal velocity scales to reduce the data missing regions, a geostrophic velocity components and noise. Horizontal advective equation was solved by 4th-order Runge-Kutta method. Horizontal diffusion effect is also adopted by the random-walk method based on a Smagolinsky-type parameterization. 2000 of particles were released at a near point from the FNPP ($37^{\circ} 25.2' \text{ N}$ and $141^{\circ} 4.8' \text{ E}$) everyday for the period from 26 March and 6 April 2011, when the most of the discharge from FNPP to the ocean was estimated (Tsumune et al., 2012). Because this experiment does not consider the atmospheric deposition, the degree of vertical mixing, adsorption onto particles and absorption due to biological processes, it cannot provide the quantitative concentration field of the radioactive Cs. But it can be allowed to reveal a certain qualitative tendency of horizontal dispersion of radioactive Cs directly discharged to the ocean by reason of using the realistic velocity data. For example, because the spatial motion scale of meteorological phenomenon is generally much larger than that of oceanographic phenomenon, this experiment provides the tendencies of horizontal dispersion by the oceanic meso-scale motions on an instantaneous field. The estimated distributions were compared with the direct observations in July, October 2011 and July 2012.

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3 Results

3.1 Concentrations of ^{134}Cs and ^{137}Cs in surface seawater

In July 2011, the concentrations of ^{134}Cs and ^{137}Cs in the surface seawater were highly elevated more than 10 mBq kg^{-1} , up to 140 mBq kg^{-1} and 153 mBq kg^{-1} at stations C43–C50, located in the northern part of Kuroshio Extension (KE) along with 144° E , estimated from satellite altimeter data, and all stations at 155° E (Fig. 1a and Table 1). At station C48 ($38^\circ 30'\text{ N}$, $144^\circ 00'\text{ E}$), ^{134}Cs was not detected ($< 1.4\text{ mBq kg}^{-1}$) and ^{137}Cs was lower than those of adjacent stations, although station C48 located in the northern part of KE (Fig. 1a). The concentrations of ^{137}Cs in central North Pacific ($175^\circ 30'\text{ E}$ transect) ranged from 3.2 to 9.3 mBq kg^{-1} and were lower than those in the western part of studied area (144° E and 155° E transects), still higher than those of background level (2.4 mBq kg^{-1} ; cf. Povinec et al., 2004). Furthermore, ^{134}Cs was detected at three of 11 stations clearly indicating the existence of radioactive Cs derived from FNPP accident at these three stations in the central North Pacific.

The northern part of KE, east-west gradient of ^{134}Cs and ^{137}Cs in the surface water was observed at the stations around 40° N in October 2011 (Fig. 1b and Table 2). More than 10 mBq kg^{-1} of ^{134}Cs and ^{137}Cs were observed between 147° E and $175^\circ 05'\text{ E}$, and the highest concentrations observed at $152^\circ 31'\text{ E}$ (Table 2). On the other hands, southern part of KE, the concentrations of ^{137}Cs were relatively lower than those in the northern part of KE. Slightly increase of ^{137}Cs was observed at the eastern stations (stations W16–W20). ^{134}Cs had not been detected at almost stations located at the southern part of KE mainly due to the short time measurement; the detection limits of ^{134}Cs were mostly $3\text{--}4\text{ mBq kg}^{-1}$ by 7200 s counting.

After one year from the observation in July 2011, ^{134}Cs and ^{137}Cs were decreased drastically at 144° E and 155° E transects (Fig. 1c and Table 3), the concentrations of ^{134}Cs and ^{137}Cs at $175^\circ 30'\text{ E}$ transect were almost the same level between two years or slightly increased in July 2012 compared with those in July 2011 (Tables 1 and 3).

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3.2 Particle tracking experiments

The pseudo particles as tracers of radioactive Cs from the FNPP were distributed broad area in the western North Pacific in July 2011 (Fig. 2a). The particles were mainly distributed in the northern area from the KE (35° N–43° N and 143° E–170° E). In October 2011, the particles were distributed broader than those in July 2011 and two dense areas were observed within 40° N–43° N, 155° E–175° W and 35° N–40° N, 170° E–165° W (Fig. 2b). After 16 month from the FNPP accident, the dense area was moved to eastward, and observed between 37° N–42° N and 155° W–175° W. The relatively dense area was also observed between 40° N–45° N and 145° E–160° W (Fig. 2c).

4 Discussions

FNPP was located at the southern part in the Kuroshio–Oyashio transition area, which represents the area between the extensions of the subtropical Kuroshio and the subarctic Oyashio. Since FNPP was close to the Kuroshio Extension (KE) that is the strongest jet off the east coast of Japan (cf. Mizuno and White, 1983), KE could play an important role of dispersing the radioactive Cs derived from FNPP far eastward in the North Pacific. In July 2011, the northern edge of the KE estimated from sea surface height was distributed near 37° N at 144° E, where the radioactive Cs was lower ($< 3.6 \text{ mBqkg}^{-1}$ for ^{134}Cs and $< 5.0 \text{ mBqkg}^{-1}$ for ^{137}Cs) than those at the northern part of the same section (Fig. 1a and Table 1). Such result suggests that the radioactive Cs from FNPP was transported eastward by KE before dispersing south of KE as shown in Fig. 2a. Similarly, the concentrations of ^{134}Cs and ^{137}Cs at stations W21–W34 south of KE were lower than those at stations W01–W10 north of KE in October 2011 (Fig. 1b, Table 2 and Fig. 2b). The less concentration of radioactive Cs in south of KE was also reported based on direct observations of seawater (Buesseler et al., 2011, 2012; Aoyama et al., 2012) or based on simulation models (Masumoto et al., 2012). Thus the radioactive Cs from FNPP would not be dispersed south of KE near the east coast of Japan until

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October 2011. The detection of ^{134}Cs at three stations along with $175^{\circ}30'$ E transect in July 2011 may indicate the effect of atmospheric deposition, although no particle was observed around these stations by the particle tracking experiment (Fig. 2a) which was taken into account for only the direct discharge from FNPP. The atmospheric deposition occurred mostly in March 2011 (Chino et al., 2011), and most of the direct discharge occurred during late March and early April 2011 (cf. Tsumune et al., 2012). Aoyama et al. (2013) reported the high radioactive Cs concentration area around the International Date Line in April–July 2011. They estimated an eastward speed of the radioactive plume to be 8 cm s^{-1} from direct observation data, trajectories of Argo floats and satellite observations. Thus, high radioactive Cs area observed in the central North Pacific in July 2011 may be derived from atmospheric deposition.

In the observation of July 2011, the local minima of concentration of ^{137}Cs and ^{134}Cs were seen at station C48 whereas the adjacent stations had high concentrations. Judging from the sea surface velocity field, station C48 was located at the edge of an anti-cyclonic eddy (Fig. 1a). Since surface water of anti-cyclonic eddies is originated from KE (Itoh and Yasuda, 2010a; Yasuda et al., 1992), the water at C48 would not contain much water derived from FNPP. As there are a lot of meso-scale eddies originated from both the KE and Oyashio in the western Kuroshio–Oyashio transition area (cf. Itoh and Yasuda, 2010b), the concentration of radioactive Cs would be patchy corresponding to such eddies there.

An area with high concentration over 50 mBq kg^{-1} of ^{137}Cs was distributed around 40° N between 150° E and 170° E in October 2011 (Table 2). Since Isoguchi et al. (2006) showed the existence of two quasi-stationary jets which flow northeastward from KE to the subarctic front between 150° E and 170° E , the radioactive Cs from FNPP might be dispersed along these jets at about this observation period also as shown in Fig. 2b. Judging from Fig. 2c, dispersion of radioactive Cs from FNPP along with these jets seemed to be continued until July 2012.

The concentrations of radioactive Cs at 144° E and 155° E transects in July 2012 were much less than those in the previous year (July 2011). These differences strongly

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suggest that the water with high concentration of radioactive Cs were transported eastward after 16 month from FNPP accident, and the concentrations of radioactive Cs in the western North Pacific were almost decreased to $\leq 10 \text{ mBqkg}^{-1}$. On the contrary, the concentrations of radioactive Cs at $175^\circ 30' \text{ E}$ transect were almost comparable between the two years. Since the eastward current forms circum-seamount circulations northward and southward around the Emperor Sea Mounts (located between 170° E and 175° E) (Wagawa et al., 2012), the high radioactive Cs waters would be dispersed not only eastward but also northward and southward as shown in Fig. 2b, c. Then the water will be transported further northward and southwestward by the recirculations of subarctic and subtropical gyres, respectively.

The $^{134}\text{Cs}/^{137}\text{Cs}$ ratio was calculated using the data those were decay corrected at 6 April 2011, when the maximum direct radioactivity discharged in to the ocean (Buesseler et al., 2011). The ratios were calculated only for the samples detected both of ^{134}Cs and ^{137}Cs , and the measurement errors were smaller than 10%. The ratios were varied from 0.55 ± 0.17 to 1.14 ± 0.15 throughout the studied periods (Fig. 3). The mean values of $^{134}\text{Cs}/^{137}\text{Cs}$ ratio were gradually decreased from 0.96 ± 0.10 ($n = 15$) in July 2011, 0.90 ± 0.07 ($n = 8$) in October 2011 and 0.85 ± 0.12 ($n = 39$) in July 2012. Such decrease trend of the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio may indicate the dilution process of radioactive Cs derived from FNPP with the background ^{137}Cs mostly originated from the nuclear weapons tests conducted in the late 1950s and the 1960s (Hirose and Aoyama, 2003).

At each station where seawater sample was collected, a CTD observation was conducted in July 2011 and July 2012. The relationship between ^{137}Cs concentration and temperature may also be indicating the dilution process of the FNPP derived radioactive Cs within the studied area (Fig. 4a). The relationship between ^{137}Cs concentration and latitude was also shown as Fig. 4b, since temperature in the studied area mainly depends on the latitude, high temperature in low latitude (subtropical region) and low temperature in high latitude (subarctic region). Although both of the concentration of ^{137}Cs and temperature were almost same ranges between two years at $175^\circ 30' \text{ E}$

transect, the ^{137}Cs concentrations at the low temperature (15–25°C) and high latitude (37° 30′–41° 30′ N) area were apparently higher than those at high temperature (> 25°C) and low latitude (35°–37° N) area in July 2011 at two western transects. On the contrary, a gently sloping decrease of ^{137}Cs concentration with increase of temperature was observed between 15°C and 27°C (Fig. 4a), and from 42° 00′ N to 33° 30′ N in July 2012 (Fig. 4b). It's note worthy that the ^{137}Cs concentrations around 25°C were almost comparable between these two years. The difference of relationship between ^{137}Cs and temperature among two years with convergence at a few Bq/kg around 25°C suggests that the high radioactive Cs derived from FNPP observed around 15°C and 25°C in July 2011 had been diluted in the Kuroshio–Oyashio transition area, and considerable amounts of them had been transported away from the studied area during 16 month from the FNPP accident. Taking into account of the result of particle tracking experiment and the circum-seamount circulations around the Emperor Sea Mounts in the central North Pacific, the most of the radioactive Cs from FNPP would be transported eastward within one year, and then it would be diluted broadly not only eastward but also north- and southward with decreasing its concentration.

5 Conclusions

Radioactive Cs derived from FNPP was distributed broadly in western and central North Pacific, especially in the northern part of Kuroshio extension during July 2011 and July 2012. The dense radioactive Cs water masses had been moved away from the western North Pacific until October 2011, and the concentration of radioactive Cs at 144° E transect has been decreased almost one order of magnitude after 16 month from the FNPP accident. A particle tracking experiment, $^{134}\text{Cs}/^{137}\text{Cs}$ ratio and relationship between temperature and concentration of ^{137}Cs suggest that a considerable amount of radioactive Cs from FNPP has been dispersed eastward from the western North Pacific to the central North Pacific during a first year from FNPP accident, and then dispersed not only eastward but also north- and southward in the central

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and western North Pacific. We focus on the radioactive Cs in the surface seawater, thus significant uncertainties remain to understand the comprehensive distribution and dispersion pattern of radioactive Cs derived from FNPP in the North Pacific. Future studies must include vertical distribution patterns of the radioactive Cs, estimation of atmospheric fallout to the North Pacific, terrestrial input of radioactive Cs such as river runoff.

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Table 1. Concentrations of ^{134}Cs and ^{137}Cs in the surface seawater collected during 30 June and 29 July 2011.

Station	Latitude	Longitude	Sampling date	^{137}Cs (mBq kg $^{-1}$)	^{134}Cs (mBq kg $^{-1}$)
A13	40° 00.8' N	175° 30.9' E	2 Jul 2011	5.9 ± 1.5	< 4.0
A14	39° 30.2' N	175° 30.3' E	2 Jul 2011	4.7 ± 0.7	4.2 ± 0.6
A15	39° 00.0' N	175° 30.1' E	2 Jul 2011	6.0 ± 1.7	< 3.9
A16	38° 29.6' N	175° 29.7' E	1 Jul 2011	3.5 ± 0.6	< 1.5
A17	38° 00.1' N	175° 30.1' E	1 Jul 2011	5.1 ± 0.7	< 1.6
A18	37° 30.2' N	175° 30.0' E	1 Jul 2011	8.4 ± 1.8	7.6 ± 1.5
A19	37° 01.2' N	175° 30.7' E	30 Jun 2011	9.3 ± 2.1	7.2 ± 1.8
A20	36° 30.1' N	175° 30.0' E	30 Jun 2011	3.2 ± 0.6	< 1.7
A21	36° 00.0' N	175° 30.1' E	30 Jun 2011	4.9 ± 1.4	< 3.7
A22	35° 30.2' N	175° 29.9' E	29 Jun 2011	4.4 ± 1.4	< 3.4
A23	34° 59.9' N	175° 29.9' E	29 Jun 2011	3.5 ± 0.6	< 1.6
B30	38° 00.3' N	155° 00.3' E	14 Jul 2011	47 ± 3.4	49 ± 2.9
B31	38° 30.2' N	155° 00.0' E	14 Jul 2011	40 ± 3.5	32 ± 2.6
B32	39° 00.4' N	154° 59.6' E	14 Jul 2011	36 ± 3.4	36 ± 2.9
B33	39° 31.4' N	155° 06.9' E	13 Jul 2011	52 ± 3.8	41 ± 2.8
B34	40° 00.1' E	155° 00.0' E	13 Jul 2011	86 ± 4.8	74 ± 3.6
B35	40° 29.9' N	155° 00.4' E	13 Jul 2011	53 ± 4.1	54 ± 3.4
B36	40° 59.0' N	154° 59.4' E	12 Jul 2011	41 ± 3.5	39 ± 2.7
B37	41° 30.2' N	155° 00.4' E	12 Jul 2011	75 ± 4.5	60 ± 3.5
C43	41° 00.3' N	143° 59.4' E	29 Jul 2011	71 ± 4.6	57 ± 3.5
C44	40° 30.0' N	144° 00.1' E	29 Jul 2011	60 ± 4.0	51 ± 3.0
C45	40° 00.0' N	143° 59.9' E	29 Jul 2011	65 ± 4.3	49 ± 3.1
C46	39° 30.4' N	144° 01.7' E	28 Jul 2011	83 ± 5.0	78 ± 3.9
C47	39° 00.1' N	144° 00.2' E	28 Jul 2011	153 ± 6.8	140 ± 5.2
C48	38° 29.9' N	144° 00.2' E	28 Jul 2011	2.7 ± 0.6	< 1.4
C49	38° 00.2' N	144° 00.2' E	27 Jul 2011	36 ± 3.3	31 ± 2.4
C50	37° 30.2' N	144° 00.1' E	27 Jul 2011	50 ± 3.6	42 ± 2.7
C51	37° 00.0' N	143° 59.9' E	27 Jul 2011	3.0 ± 0.6	3.6 ± 0.5
C52	36° 29.9' N	144° 00.3' E	26 Jul 2011	1.9 ± 0.4	< 1.2
C53	36° 00.2' N	144° 00.2' E	26 Jul 2011	2.2 ± 0.5	< 1.2
C54	35° 30.1' N	144° 00.1' E	26 Jul 2011	5.0 ± 1.4	< 3.6
C55	35° 00.2' N	144° 00.3' E	26 Jul 2011	4.8 ± 0.5	3.1 ± 0.5

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Table 2. Concentrations of ^{134}Cs and ^{137}Cs in the surface seawater collected during 14 October and 7 November 2011.

Station	Latitude	Longitude	Sampling date	^{137}Cs (mBq kg $^{-1}$)	^{134}Cs (mBq kg $^{-1}$)
W01	40° 05.9' N	147° 00.0' E	14 Oct 2011	15 ± 2.4	11 ± 1.8
W02	40° 54.7' N	147° 05.4' E	14 Oct 2011	24 ± 2.6	16 ± 1.9
W03	40° 00.0' N	150° 03.6' E	15 Oct 2011	53 ± 3.9	39 ± 2.7
W04	40° 00.1' N	152° 31.3' E	15 Oct 2011	100 ± 5.0	72 ± 3.5
W05	39° 59.9' N	156° 25.7' E	16 Oct 2011	74 ± 4.6	53 ± 3.1
W06	40° 00.1' N	158° 45.5' E	16 Oct 2011	72 ± 4.5	56 ± 3.1
W07	40° 00.0' N	162° 32.1' E	17 Oct 2011	49 ± 3.9	41 ± 3.0
W08	40° 12.2' N	164° 52.0' E	17 Oct 2011	73 ± 4.4	54 ± 3.1
W09	40° 01.2' N	169° 22.4' E	18 Oct 2011	50 ± 3.8	35 ± 2.7
W10	39° 59.9' N	170° 57.6' E	18 Oct 2011	29 ± 3.0	25 ± 2.3
W11	40° 00.0' N	175° 04.7' E	19 Oct 2011	12 ± 2.2	9.8 ± 1.7
W12	39° 30.0' N	175° 40.0' E	19 Oct 2011	7.4 ± 1.7	4.4 ± 1.2
W13	39° 21.4' N	175° 53.9' E	20 Oct 2011	3.7 ± 0.6	< 1.2
W14	39° 23.4' N	175° 52.8' E	21 Oct 2011	4.4 ± 0.6	2.6 ± 0.5
W15	38° 39.4' N	176° 01.5' E	22 Oct 2011	3.6 ± 0.5	< 1.2
W16	34° 10.7' N	175° 29.5' E	23 Oct 2011	5.6 ± 1.6	< 4.3
W17	34° 13.8' N	175° 24.0' E	24 Oct 2011	6.1 ± 1.7	5.8 ± 1.3
W18	33° 26.7' N	175° 52.9' E	25 Oct 2011	11 ± 1.9	5.8 ± 1.3
W19	33° 29.1' N	175° 36.8' E	26 Oct 2011	10 ± 2.0	5.9 ± 1.3
W20	31° 04.9' N	175° 47.8' E	27 Oct 2011	5.4 ± 1.5	< 3.1
W21	32° 22.8' N	172° 41.3' E	29 Oct 2011	7.0 ± 2.0	< 3.5
W22	33° 15.0' N	171° 39.7' E	30 Oct 2011	5.4 ± 1.7	7.1 ± 1.5
W23	30° 33.2' N	169° 09.2' E	1 Nov 2011	4.8 ± 1.4	< 3.3
W24	30° 32.0' N	165° 24.3' E	2 Nov 2011	6.8 ± 1.8	< 3.0
W25	30° 35.8' N	163° 24.4' E	2 Nov 2011	5.5 ± 1.6	< 3.2
W26	29° 52.4' N	161° 12.9' E	3 Nov 2011	4.5 ± 1.4	< 3.0
W27	29° 22.8' N	159° 00.3' E	3 Nov 2011	5.1 ± 1.5	< 3.1
W28	29° 26.2' N	154° 54.0' E	4 Nov 2011	4.2 ± 1.3	< 3.1
W29	29° 35.7' N	152° 35.5' E	4 Nov 2011	4.8 ± 1.6	< 3.5
W30	29° 26.2' N	148° 30.2' E	5 Nov 2011	3.7 ± 0.5	2.3 ± 0.4
W31	29° 35.7' N	146° 05.8' E	5 Nov 2011	4.5 ± 0.6	3.0 ± 0.5
W32	29° 53.8' N	142° 08.3' E	6 Nov 2011	3.9 ± 0.5	< 1.3
W33	30° 28.2' N	140° 21.0' E	6 Nov 2011	5.6 ± 1.4	< 3.7
W34	31° 09.6' N	140° 21.0' E	7 Nov 2011	3.5 ± 0.5	< 1.3

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Table 3. Concentrations of ^{134}Cs and ^{137}Cs in the surface seawater collected during 2 July and 1 August 2012.

Station	Latitude	Longitude	Sampling date	^{137}Cs (mBqkg $^{-1}$)	^{134}Cs (mBqkg $^{-1}$)
A23	35°00.0'N	175°29.6'E	2 Jul 2011	9.6±0.5	5.4±0.4
A22	35°29.5'N	175°26.9'E	2 Jul 2011	8.6±0.6	5.6±0.4
A21	36°00.1'N	175°30.3'E	3 Jul 2011	8.1±0.5	5.1±0.4
A20	36°30.1'N	175°30.0'E	3 Jul 2011	6.4±0.5	4.0±0.4
A19	37°00.0'N	175°30.4'E	3 Jul 2011	8.5±0.5	4.3±0.4
A18	37°30.0'N	175°30.0'E	4 Jul 2011	9.4±0.7	5.8±0.5
A17	38°00.1'N	175°30.0'E	4 Jul 2011	12±0.7	7.7±0.5
A16	38°30.4'N	175°30.3'E	4 Jul 2011	12±0.7	7.3±0.5
A15	39°00.1'N	175°30.0'E	5 Jul 2011	15±0.4	7.4±0.4
A14	39°30.1'N	175°29.9'E	5 Jul 2011	16±0.8	9.5±0.5
A13	40°06.5'N	175°43.3'E	5 Jul 2011	14±0.7	8.3±0.5
A12	40°30.0'N	175°29.9'E	6 Jul 2011	13±0.6	7.8±0.4
A11	41°00.0'N	175°30.0'E	6 Jul 2011	18±0.7	11±0.5
A10	41°30.7'N	175°29.6'E	6 Jul 2011	19±0.8	12±0.5
A09	42°00.1'N	175°30.0'E	7 Jul 2011	19±0.8	11±0.5
A08	42°30.1'N	175°29.9'E	7 Jul 2011	18±0.7	12±0.5
A07	43°00.2'N	175°30.2'E	7 Jul 2011	19±0.8	12±0.5
A06	43°30.0'N	175°30.1'E	8 Jul 2011	16±0.7	10±0.4
A05	44°00.0'N	175°30.0'E	8 Jul 2011	17±0.7	11±0.5
A04	44°30.6'N	175°30.6'E	8 Jul 2011	15±0.7	9.3±0.5
B42	44°00.0'N	155°00.4'E	12 Jul 2011	6.0±0.5	3.8±0.4
B41	43°30.0'N	154°59.9'E	13 Jul 2011	6.9±0.5	3.6±0.3
B40	43°00.7'N	155°00.3'E	13 Jul 2011	5.6±0.5	2.7±0.4
B39	42°31.8'N	155°06.0'E	13 Jul 2011	13±0.7	6.6±0.5
B38	42°00.3'N	154°59.8'E	14 Jul 2011	18±0.7	11±0.5
B37	41°30.1'N	154°59.7'E	14 Jul 2011	17±0.7	11±0.5
B36	40°59.8'N	155°03.8'E	14 Jul 2011	18±0.7	9.4±0.4
B35	40°30.7'N	155°00.4'E	15 Jul 2011	13±0.7	8.1±0.5
B34	40°00.1'N	155°00.3'E	15 Jul 2011	9.2±0.5	5.0±0.4
B33	39°30.3'N	155°02.9'E	15 Jul 2011	11±0.6	6.8±0.4
B32	39°00.0'N	154°59.7'E	16 Jul 2011	4.3±0.6	<1.2
B31	38°30.0'N	154°59.3'E	16 Jul 2011	4.0±0.5	1.7±0.4
B30	38°00.6'N	155°04.9'E	16 Jul 2011	3.6±0.5	2.0±0.38
B29	37°30.0'N	155°00.1'E	17 Jul 2011	3.4±0.4	<0.98
B28	37°00.6'N	155°00.8'E	17 Jul 2011	2.2±0.4	<0.97
B27	36°27.7'N	155°03.5'E	17 Jul 2011	2.2±0.4	<0.95
B26	36°00.3'N	155°00.5'E	18 Jul 2011	2.6±0.4	<0.90
B25	35°29.9'N	154°59.9'E	18 Jul 2011	4.2±0.5	2.2±0.30
B24	35°00.1'N	155°00.4'E	18 Jul 2011	7.2±0.6	4.2±0.39
B24X	34°29.9'N	155°00.3'E	19 Jul 2011	2.3±0.4	<0.86
B24Y	33°59.9'N	155°00.0'E	19 Jul 2011	2.1±0.4	<0.81
C58	33°30.0'N	144°00.0'E	27 Jul 2011	1.7±0.3	<0.71
C57	34°00.1'N	144°00.4'E	28 Jul 2011	2.5±0.3	<0.77
C56	34°30.1'N	144°00.0'E	28 Jul 2011	2.4±0.4	<0.87
C55	35°02.3'N	143°58.5'E	28 Jul 2011	2.1±0.4	<0.69
C54	35°30.0'N	144°00.0'E	29 Jul 2011	1.2±0.3	<0.79
C53	35°59.9'N	144°00.1'E	29 Jul 2011	1.8±0.4	<0.90
C52	36°30.3'N	143°59.9'E	29 Jul 2011	6.5±0.5	3.3±0.31
C51	37°00.1'N	144°00.0'E	30 Jul 2011	6.3±0.5	2.9±0.33
C50	37°29.9'N	143°59.9'E	30 Jul 2011	4.1±0.4	1.5±0.31
C49	38°00.5'N	143°59.5'E	30 Jul 2011	4.7±0.4	1.7±0.34
C48	38°29.9'N	144°00.0'E	31 Jul 2011	5.2±0.4	2.1±0.30
C47	39°00.0'N	144°00.0'E	31 Jul 2011	5.3±0.5	2.5±0.32
C46	39°30.6'N	144°00.9'E	31 Jul 2011	4.1±0.5	2.7±0.33
C45	39°59.9'N	144°00.0'E	1 Aug 2011	5.6±0.5	3.2±0.35
C44	40°29.9'N	144°00.0'E	1 Aug 2011	3.3±0.4	1.5±0.30
C43	41°01.7'N	144°00.1'E	1 Aug 2011	1.7±0.4	<0.84

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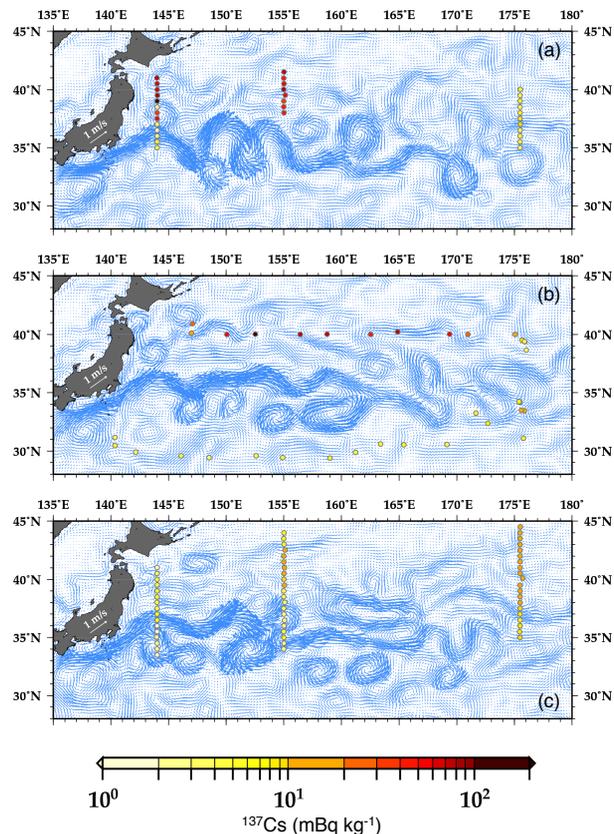


Fig. 1. Sampling locations for surface seawater in the western and central North Pacific. Closed circles indicate the sampling stations. Color of the closed circles indicates concentration of ^{137}Cs in the surface seawater. Blue arrows indicates the estimated temporal mean velocity vectors for the period between (a) 30 June and 29 July 2011, (b) 14 October and 7 November 2011, and (c) 2 July and 1 August 2012.

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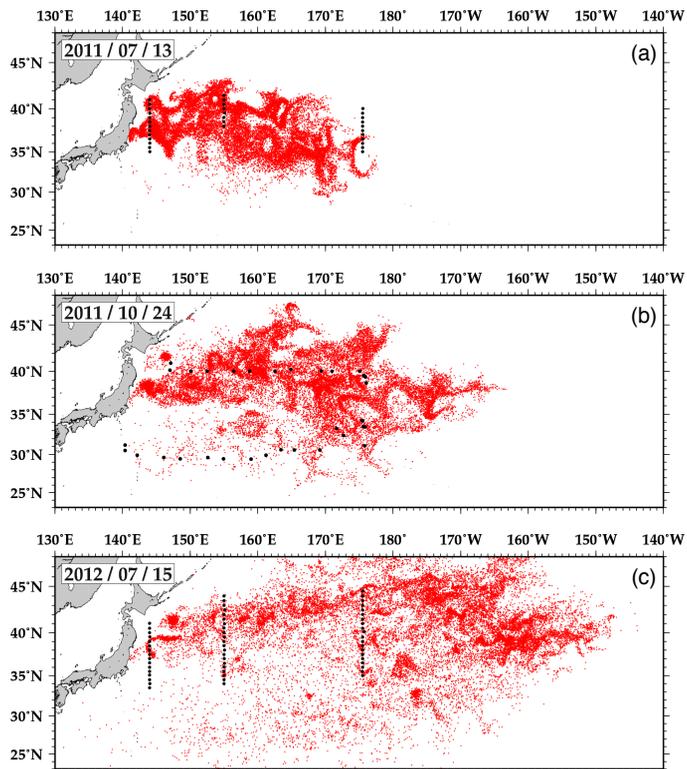


Fig. 2. The result of particle tracking experiment at the median date of each sampling period. Red circles indicate pseudo particle and black circles indicate sampling locations. **(a)** 13 July 2012. **(b)** 24 October 2011. **(c)** 15 July 2012.

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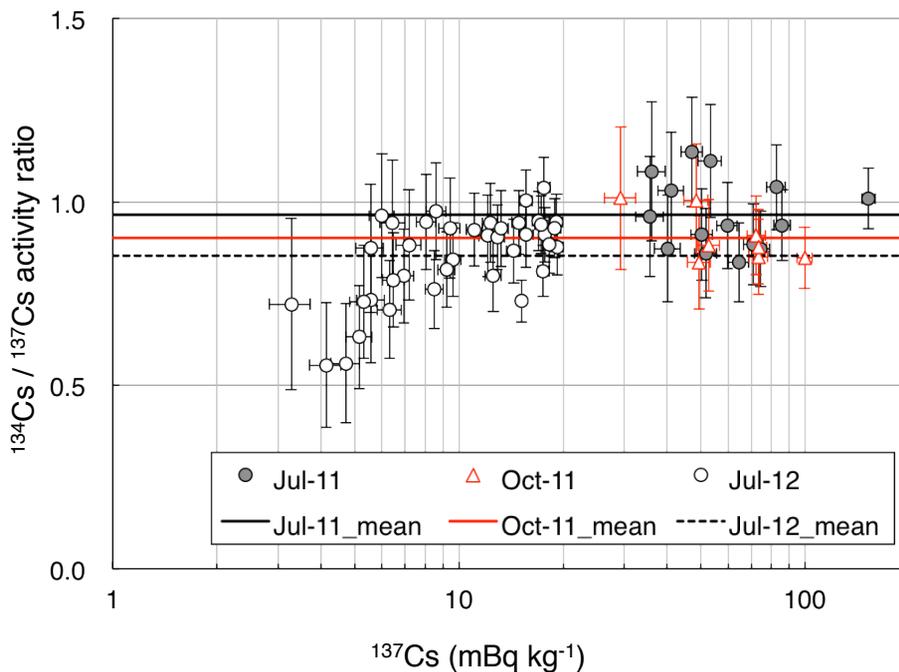


Fig. 3. Relationship between concentrations of ^{137}Cs decay corrected at sampling date and $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio decay corrected at 6 April 2011, when the maximum direct radioactivity discharged in to the ocean (Buessler et al., 2011).

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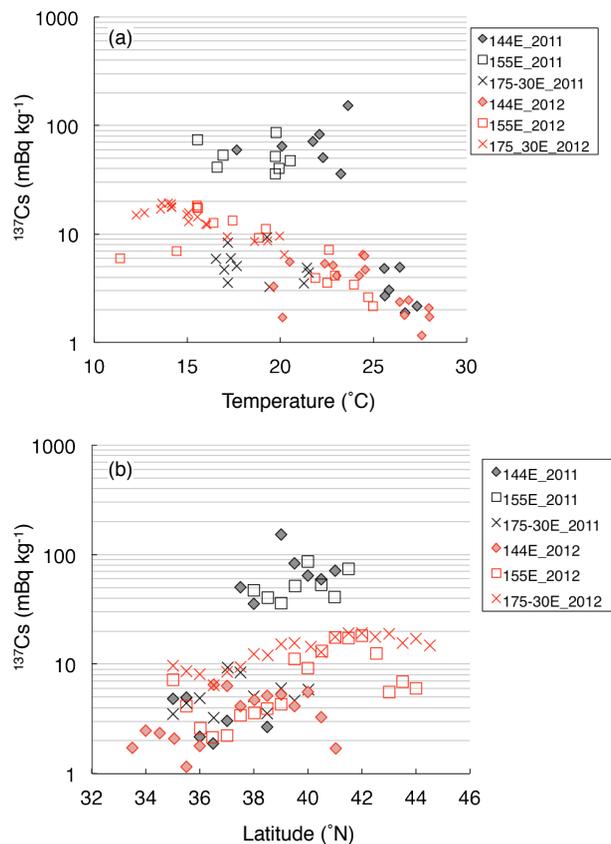


Fig. 4. (a) Relationship between concentrations of ^{137}Cs and seawater temperature at 5 m depth during July 2011 and July 2012. (b) Relationship between concentrations of ^{137}Cs and latitude.

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