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# Direct observation of $^{134}$Cs and $^{137}$Cs in surface seawater in the western and central North Pacific after the Fukushima Dai-ichi nuclear power plant accident

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**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 a broad area in the western and central North Pacific in July 2011, 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. High $^{134}$Cs and $^{137}$Cs concentrations more than 10 m Bq kg$^{-1}$ were observed in the area of the northern part of Kuroshio Extension at 144° E and 155° E in July 2011, in the area 147–175° E around 40° N in October 2011, and the northern part of Kuroshio Extension at 155° E and 175°30′ E in July 2012. Combining the result of direct observations and particle tracking experiment, the radioactive Cs derived from the 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 the FNPP during study period is suggested from temporal change in the activity ratio of $^{134}$Cs / $^{137}$Cs, which was decay-corrected on 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 of the reactors and hydrogen explosions. Radioactive materials were released into the ocean not only as atmospheric fallout but also as the direct release and leaking of the heavily contaminated coolant water (cf. Buesseler et al., 2011). Radioactive cesium (Cs), with a relatively long half-life (2.07 yr for $^{134}$Cs and 30.07 yr for $^{137}$Cs), is a serious concern as it contaminates oceanic environments and marine biota, especially for fishery 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 of the FNPP (MEXT, 2012), and those in fishery products in the broad area around Japan (Fisheries Agency, 2012). However, the information on radioactive contamination covering the broad area in the North Pacific is still quite limited (Aoyama et al., 2012, 2013). In some model experiments, the dispersion of radioactive Cs from the FNPP is discussed (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 discuss 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 a broad area of western and central North Pacific. In June–July 2011, three north–south transects were set along 144° E, 155° E and 175°30′ E, and seawater samples were collected at 32 stations (Fig. 1a). A total of 34 samples were collected in the area within 30–41° N and 140–176° E during October and November 2011 (Fig. 1b), and 57 samples were collected in the three north–south transects along 144° E, 155° E and 175°30′ E during July and August 2012 (Fig. 1c). The seawater sample was unfiltered, transferred into a 20 L plastic bag and acidified to a pH of 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 elsewhere (cf. Aoyama et al., 2000; Aoyama and Hirose, 2008; Aoyama et al., 2012). The seawater sample was transferred to the container and weighed 18 kg. As a carrier, 0.52 g of CsCl was added to 18 kg of sample and stirred for at least 1 h. Then 8.0 g of AMP were added to sample and stirred for at least 1 h. 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 a 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, the dried AMP/Cs compound was weighed to determine the weight yield of the AMP/Cs compound. The weight yield of AMP/Cs compound 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 and 796 keV for $^{134}$Cs and 662 keV for $^{137}$Cs obtained by more than 7200 s of 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 m Bq kg$^{-1}$. When the $^{137}$Cs was not detected with the 7200 s measurement, longer time measurements (> 40000 s) were carried out. The detection limits of $^{137}$Cs by the more than 40000 s measurements ($\leq$ 1.4 m Bq kg$^{-1}$) had almost the same level of the concentration of $^{137}$Cs in the sea surface water in the North Pacific before the accident of the FNPP (2.4 m Bq kg$^{-1}$; cf. Povinec et al., 2004).

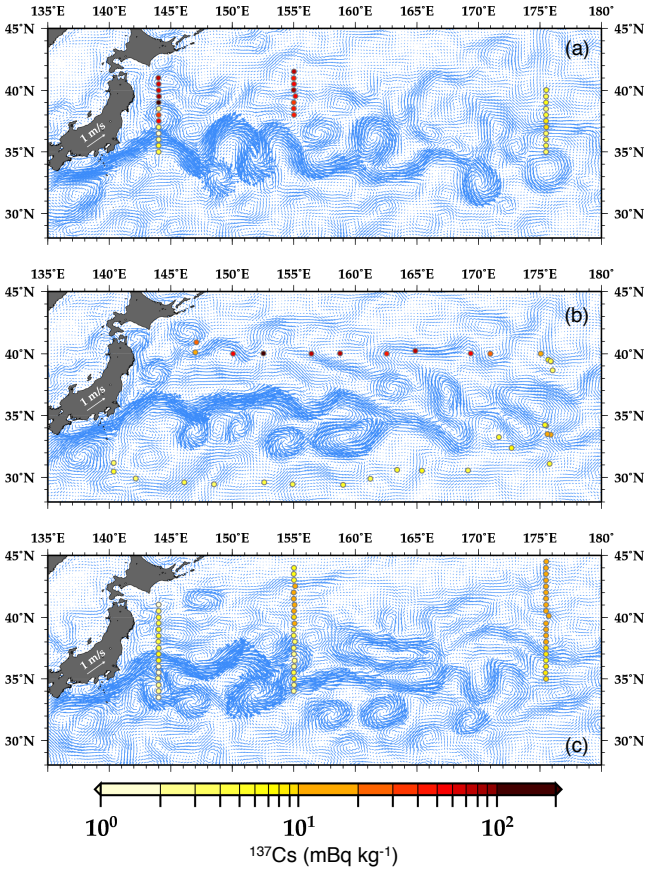


**Fig. 1.** Sampling locations for surface seawater in the western and central North Pacific. Closed circles indicate the sampling stations. Color of the closed circles indicates concentration of $^{137}$Cs in the surface seawater. Blue arrows indicate the estimated temporal mean velocity vectors for 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.

### 2.3 Particle tracking experiment

A numerical particle tracking experiment was carried out to investigate the sea surface horizontal distribution pattern

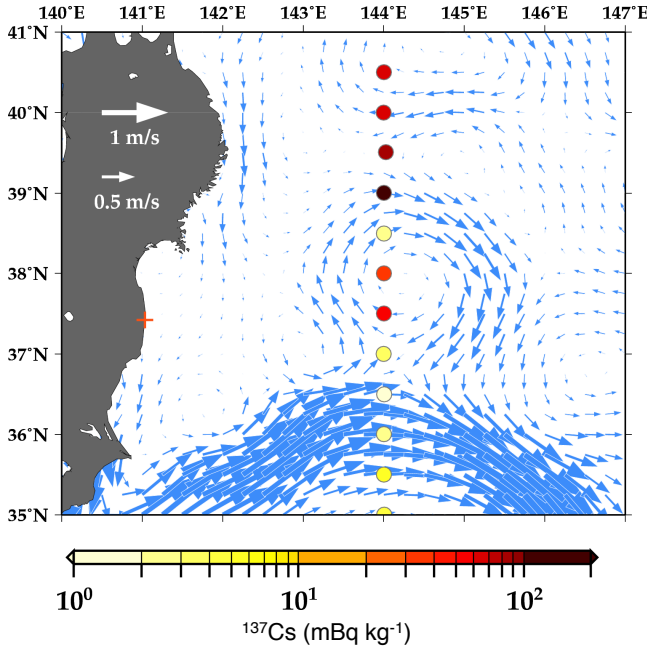


**Fig. 2.** Sampling locations for surface seawater around the anticyclonic eddy observed in July 2011. Color of the closed circles indicates concentration of $^{137}$Cs in the surface seawater. Blue arrows indicate the estimated temporal mean velocity vectors for the period between 30 June and 29 July 2011.

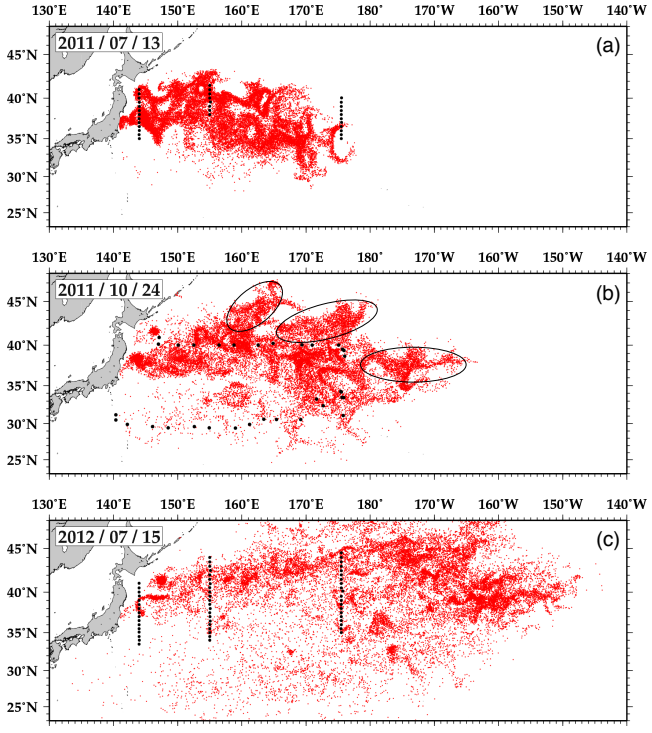


**Fig. 3.** The result of particle tracking experiment at the median date of each sampling period. Red circles indicate pseudo-particle, and black circles indicate sampling locations: **(a)** 13 July 2012, **(b)** 24 October 2011, and **(c)** 15 July 2012.

of radioactive Cs released from the FNPP. The time series of sea surface velocity field we used was estimated every 7 days and 0.25 degree for large- and mesoscale 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, geostrophic velocity components and noise. A horizontal advective equation was solved by the fourth-order Runge–Kutta method. A horizontal diffusion effect is also adopted by the random-walk method based on a Smagorinsky-type parameterization. A total of 2000 particles were released at a point near the FNPP (37°25.2′ N and 141°4.8′ E) everyday for the period from 26 March to 6 April 2011, when most of the discharge from the FNPP into 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 reveal a certain qualitative tendency of horizontal dispersion of radioactive Cs directly discharged into the ocean by using the realistic velocity data. For example, because the spatial motion scale of meteorological phenomena is generally much larger than that of oceanographic phenomena, this experiment provides the tendencies of horizontal dispersion by the oceanic mesoscale motions on an instantaneous field. The estimated distributions were compared with the direct observations in July 2011, October 2011 and July 2012.

## 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 by more than 10 m Bq kg$^{-1}$, up to 140 m Bq kg$^{-1}$ and 153 m Bq kg$^{-1}$ at stations C43–C50, located in the northern part of Kuroshio Extension (KE) along 144° E, estimated from satellite altimeter data, and all stations at 155° E (Fig. 1a and Table 1). At station C48 (38°30′ N, 144°00′ E), $^{134}$Cs was not detected ($< 1.4$ m Bq kg$^{-1}$), and $^{137}$Cs was lower than those of adjacent stations, although station C48 was located in the northern part of KE (Fig. 2). The concentrations of $^{137}$Cs in central North Pacific (175°30′ E transect) ranged from 3.2 to 9.3 m Bq kg$^{-1}$ and were lower than those in the western part of the studied area (144° E and 155° E transects), still higher than those of the background level (2.4 m Bq kg$^{-1}$; cf. Povinec et al., 2004). Furthermore, $^{134}$Cs was detected at 3 of 11 stations, clearly indicating the existence of radioactive

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

Cs derived from the FNPP accident at these three stations in the central North Pacific.

At the northern part of KE, an 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 m Bq kg$^{-1}$ of $^{134}$Cs and $^{137}$Cs were observed between 147° E and 175°05′ E, and the highest concentrations were observed at 152°31′ E (Table 2). On the other hand, at the southern part of KE, the concentrations of $^{137}$Cs were relatively lower than those in the northern part of KE. A slight increase of $^{137}$Cs was observed at the eastern stations (stations W16–W20). $^{134}$Cs had not been detected at most stations located at the southern part of KE mainly due to the short time measurement; the detection limits of $^{134}$Cs were mostly 3–4 m Bq kg$^{-1}$ by 7200 s counting.

One year after the observation in July 2011, $^{134}$Cs and $^{137}$Cs decreased drastically at 144° E and 155° E transects (Fig. 1c and Table 3); the concentrations of $^{134}$Cs and $^{137}$Cs at 175° E transect were almost the same level between the two years or slightly increased in July 2012 compared to those in July 2011 (Tables 1 and 3).

### 3.2 Particle tracking experiments

The pseudo-particles as tracers of radioactive Cs from the FNPP were distributed in a broad area in the western North Pacific in July 2011 (Fig. 3a). The particles were mainly distributed in the northern area of the KE (35–43° N and 143–170° E). In October 2011, the particles were distributed more broadly than those in July 2011, and three dense areas were observed within 41–47° N, 158–166° E; 40–44° N, 167° E–180° W; and 36–39° N, 180–165° W (Fig. 3b). Sixteen months after the FNPP accident, the dense area moved eastward, and was observed between 37–42° N and 155–175° W. The relatively dense area was also observed between 40–45° N and 145° E–160° W (Fig. 3c).

**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 (m Bq kg$^{-1}$) | $^{134}$Cs (m Bq 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 |

## 4 Discussions

The FNPP is located in 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 the FNPP is close to the Kuroshio Extension (KE), which is the strongest jet off the east coast of Japan (cf. Mizuno and White, 1983), KE could play an important role in the dispersion of the radioactive Cs derived from the 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 ($< 3.6$ m Bq kg$^{-1}$ for $^{134}$Cs and $< 5.0$ m Bq kg$^{-1}$ for $^{137}$Cs) than those at the northern part of the same section (Fig. 1a and Table 1). Such a result suggests that the radioactive Cs from the FNPP was transported eastward by KE before dispersing south of KE (as shown in Fig. 3a). 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. 3b). The lower concentration of radioactive Cs south of KE was also reported based on direct observations of seawater (Buesseler et al., 2011; Buesseler et al., 2012; Aoyama et al., 2012) or based on simulation models (Masumoto et al., 2012). Thus the majority of radioactive Cs directly released into the ocean from the FNPP would not be dispersed south of KE near the east coast of Japan until October 2011. On the contrary, the detection of $^{134}$Cs at three stations along th 175°30′ E transect and stations located south of KE (such as station C55 in July 2011 and stations around 30° N in October 2011) may indicate the effect of atmospheric deposition, although no or fewer particles were observed around these stations by the particle tracking experiment (Fig. 3a and b), which was taken into account for only the direct discharge from the FNPP. The atmospheric deposition occurred mostly

**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 (m Bq kg$^{-1}$) | $^{134}$Cs (m Bq kg$^{-1}$) |
|---|---|---|---|---|---|
| A23 | 35°00.0′ N | 175°29.6′ E | 2 Jul 2012 | 9.6 ± 0.5 | 5.4 ± 0.4 |
| A22 | 35°29.5′ N | 175°26.9′ E | 2 Jul 2012 | 8.6 ± 0.6 | 5.6 ± 0.4 |
| A21 | 36°00.1′ N | 175°30.3′ E | 3 Jul 2012 | 8.1 ± 0.5 | 5.1 ± 0.4 |
| A20 | 36°30.1′ N | 175°30.0′ E | 3 Jul 2012 | 6.4 ± 0.5 | 4.0 ± 0.4 |
| A19 | 37°00.0′ N | 175°30.4′ E | 3 Jul 2012 | 8.5 ± 0.5 | 4.3 ± 0.4 |
| A18 | 37°30.0′ N | 175°30.0′ E | 4 Jul 2012 | 9.4 ± 0.7 | 5.8 ± 0.5 |
| A17 | 38°00.1′ N | 175°30.0′ E | 4 Jul 2012 | 12 ± 0.7 | 7.7 ± 0.5 |
| A16 | 38°30.4′ N | 175°30.3′ E | 4 Jul 2012 | 12 ± 0.7 | 7.3 ± 0.5 |
| A15 | 39°00.1′ N | 175°30.0′ E | 5 Jul 2012 | 15 ± 0.4 | 7.4 ± 0.4 |
| A14 | 39°30.1′ N | 175°29.9′ E | 5 Jul 2012 | 16 ± 0.8 | 9.5 ± 0.5 |
| A13 | 40°06.5′ N | 175°43.3′ E | 5 Jul 2012 | 14 ± 0.7 | 8.3 ± 0.5 |
| A12 | 40°30.0′ N | 175°29.9′ E | 6 Jul 2012 | 13 ± 0.6 | 7.8 ± 0.4 |
| A11 | 41°00.0′ N | 175°30.0′ E | 6 Jul 2012 | 18 ± 0.7 | 11 ± 0.5 |
| A10 | 41°30.7′ N | 175°29.6′ E | 6 Jul 2012 | 19 ± 0.8 | 12 ± 0.5 |
| A09 | 42°00.1′ N | 175°30.0′ E | 7 Jul 2012 | 19 ± 0.8 | 11 ± 0.5 |
| A08 | 42°30.1′ N | 175°29.9′ E | 7 Jul 2012 | 18 ± 0.7 | 12 ± 0.5 |
| A07 | 43°00.2′ N | 175°30.2′ E | 7 Jul 2012 | 19 ± 0.8 | 12 ± 0.5 |
| A06 | 43°30.0′ N | 175°30.1′ E | 8 Jul 2012 | 16 ± 0.7 | 10 ± 0.4 |
| A05 | 44°00.0′ N | 175°30.0′ E | 8 Jul 2012 | 17 ± 0.7 | 11 ± 0.5 |
| A04 | 44°30.6′ N | 175°30.6′ E | 8 Jul 2012 | 15 ± 0.7 | 9.3 ± 0.5 |
| B42 | 44°00.0′ N | 155°00.4′ E | 12 Jul 2012 | 6.0 ± 0.5 | 3.8 ± 0.4 |
| B41 | 43°30.0′ N | 154°59.9′ E | 13 Jul 2012 | 6.9 ± 0.5 | 3.6 ± 0.3 |
| B40 | 43°00.7′ N | 155°00.3′ E | 13 Jul 2012 | 5.6 ± 0.5 | 2.7 ± 0.4 |
| B39 | 42°31.8′ N | 155°06.0′ E | 13 Jul 2012 | 13 ± 0.7 | 6.6 ± 0.5 |
| B38 | 42°00.3′ N | 154°59.8′ E | 14 Jul 2012 | 18 ± 0.7 | 11 ± 0.5 |
| B37 | 41°30.1′ N | 154°59.7′ E | 14 Jul 2012 | 17 ± 0.7 | 11 ± 0.5 |
| B36 | 40°59.8′ N | 155°03.8′ E | 14 Jul 2012 | 18 ± 0.7 | 9.4 ± 0.4 |
| B35 | 40°30.7′ N | 155°00.4′ E | 15 Jul 2012 | 13 ± 0.7 | 8.1 ± 0.5 |
| B34 | 40°00.1′ N | 155°00.3′ E | 15 Jul 2012 | 9.2 ± 0.5 | 5.0 ± 0.4 |
| B33 | 39°30.3′ N | 155°02.9′ E | 15 Jul 2012 | 11 ± 0.6 | 6.8 ± 0.4 |
| B32 | 39°00.0′ N | 154°59.7′ E | 16 Jul 2012 | 4.3 ± 0.6 | < 1.2 |
| B31 | 38°30.0′ N | 154°59.3′ E | 16 Jul 2012 | 4.0 ± 0.5 | 1.7 ± 0.4 |
| B30 | 38°00.6′ N | 155°04.9′ E | 16 Jul 2012 | 3.6 ± 0.5 | 2.0 ± 0.38 |
| B29 | 37°30.0′ N | 155°00.1′ E | 17 Jul 2012 | 3.4 ± 0.4 | < 0.98 |
| B28 | 37°00.6′ N | 155°00.8′ E | 17 Jul 2012 | 2.2 ± 0.4 | < 0.97 |
| B27 | 36°27.7′ N | 155°03.5′ E | 17 Jul 2012 | 2.2 ± 0.4 | < 0.95 |
| B26 | 36°00.3′ N | 155°00.5′ E | 18 Jul 2012 | 2.6 ± 0.4 | < 0.90 |
| B25 | 35°29.9′ N | 154°59.9′ E | 18 Jul 2012 | 4.2 ± 0.5 | 2.2 ± 0.30 |
| B24 | 35°00.1′ N | 155°00.4′ E | 18 Jul 2012 | 7.2 ± 0.6 | 4.2 ± 0.39 |
| B24X | 34°29.9′ N | 155°00.3′ E | 19 Jul 2012 | 2.3 ± 0.4 | < 0.86 |
| B24Y | 33°59.9′ N | 155°00.0′ E | 19 Jul 2012 | 2.1 ± 0.4 | < 0.81 |
| C58 | 33°30.0′ N | 144°00.0′ E | 27 Jul 2012 | 1.7 ± 0.3 | < 0.71 |
| C57 | 34°00.1′ N | 144°00.4′ E | 28 Jul 2012 | 2.5 ± 0.3 | < 0.77 |
| C56 | 34°30.1′ N | 144°00.0′ E | 28 Jul 2012 | 2.4 ± 0.4 | < 0.87 |
| C55 | 35°02.3′ N | 143°58.5′ E | 28 Jul 2012 | 2.1 ± 0.4 | < 0.69 |
| C54 | 35°30.0′ N | 144°00.0′ E | 29 Jul 2012 | 1.2 ± 0.3 | < 0.79 |
| C53 | 35°59.9′ N | 144°00.1′ E | 29 Jul 2012 | 1.8 ± 0.4 | < 0.90 |
| C52 | 36°30.3′ N | 143°59.9′ E | 29 Jul 2012 | 6.5 ± 0.5 | 3.3 ± 0.31 |
| C51 | 37°00.1′ N | 144°00.0′ E | 30 Jul 2012 | 6.3 ± 0.5 | 2.9 ± 0.33 |
| C50 | 37°29.9′ N | 143°59.9′ E | 30 Jul 2012 | 4.1 ± 0.4 | 1.5 ± 0.31 |
| C49 | 38°00.5′ N | 143°59.5′ E | 30 Jul 2012 | 4.7 ± 0.4 | 1.7 ± 0.34 |
| C48 | 38°29.9′ N | 144°00.0′ E | 31 Jul 2012 | 5.2 ± 0.4 | 2.1 ± 0.30 |
| C47 | 39°00.0′ N | 144°00.0′ E | 31 Jul 2012 | 5.3 ± 0.5 | 2.5 ± 0.32 |
| C46 | 39°30.6′ N | 144°00.9′ E | 31 Jul 2012 | 4.1 ± 0.5 | 2.7 ± 0.33 |
| C45 | 39°59.9′ N | 144°00.0′ E | 1 Aug 2012 | 5.6 ± 0.5 | 3.2 ± 0.35 |
| C44 | 40°29.9′ N | 144°00.0′ E | 1 Aug 2012 | 3.3 ± 0.4 | 1.5 ± 0.30 |
| C43 | 41°01.7′ N | 144°00.1′ E | 1 Aug 2012 | 1.7 ± 0.4 | < 0.84 |

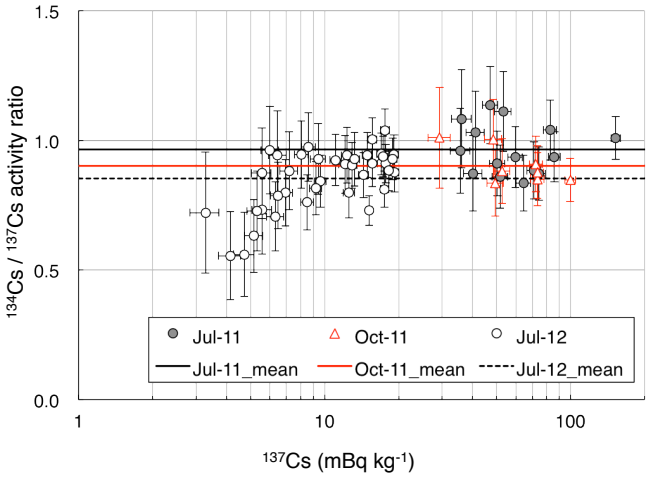


**Fig. 4.** Relationship between concentrations of $^{137}$Cs decay corrected on sampling date and $^{134}$Cs / $^{137}$Cs activity ratio decay corrected on 6 April 2011, when the maximum direct radioactivity discharged into the ocean (Buesseler et al., 2011).

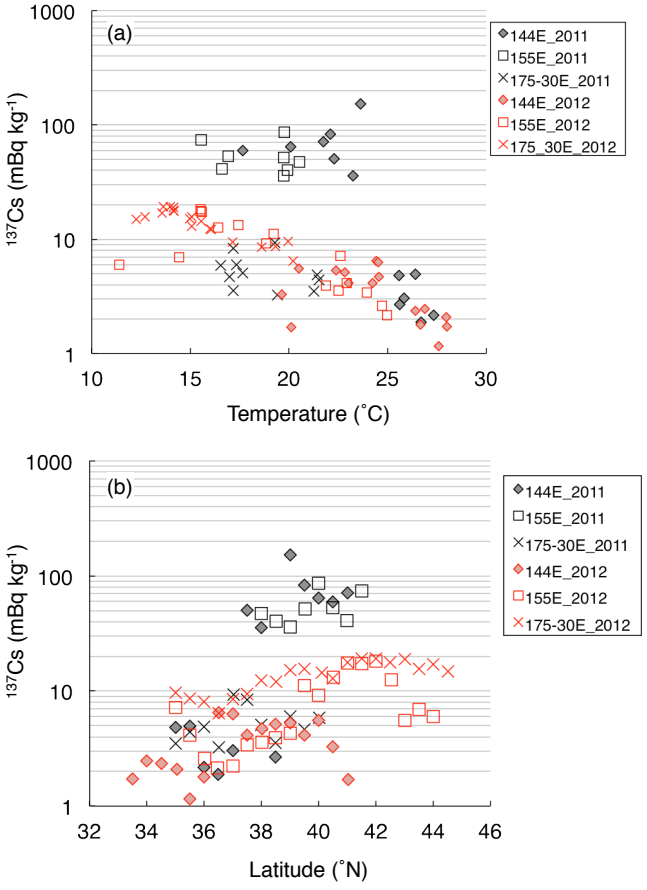


**Fig. 5. (a)** Relationship between concentrations of $^{137}$Cs and seawater temperature at 5 m depth during July 2011 and July 2012. **(b)** Relationship between concentrations of $^{137}$Cs and latitude.

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 the eastward speed of the radioactive plume to be 8 cm s$^{-1}$ from direct observation data, trajectories of Argo floats and satellite observations. Moreover, atmospheric deposition of radioactive Cs and iodine-131 south of KE near the east coast of Japan were strongly suggested by the numerical simulations (Kawamura et al., 2011; Kobayashi et al., 2013). Thus, the high radioactive Cs area observed in the central North Pacific in July 2011 and south of KE near the east coast of Japan 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 (Fig. 2). Judging from the sea surface velocity field, station C48 was located at the edge of an anti-cyclonic eddy (Fig. 2). Since surface water of anti-cyclonic eddies originated from KE (Itoh and Yasuda, 2010a; Yasuda et al., 1992), the water at C48 would not contain much water derived from the FNPP. As there are a lot of mesoscale eddies that 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 m Bq 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 that flow northeastward from KE to the subarctic front between 150° E and 170° E, the radioactive Cs from the FNPP might be dispersed along these jets in about this observation period also (as shown in Fig. 3b). Judging from Fig. 3c, dispersion of radioactive Cs from the FNPP along with these jets seemed to continue until July 2012.

The concentrations of radioactive Cs at 144° E and 155° E transects in July 2012 were much lower than those in the previous year (July 2011). These differences strongly suggest that the water with a high concentration of radioactive Cs was transported eastward 16 months after the FNPP accident, and the concentrations of radioactive Cs in the western North Pacific almost decreased to $\leq$ 10 m Bq kg$^{-1}$. On the contrary, the concentrations of radioactive Cs at the 175°30′ E transect were almost comparable between the two years. In July 2011, radioactive Cs would have been derived as atmospheric deposition (see Fig. 5 of Kobayashi et al., 2013). In contrast, the concentration of radioactive Cs observed at the 175°30′ E transect in July 2012 would have been a result of a dilution process during 16 months after the FNPP accident in the western North Pacific. Since the KE jet is weakened eastward and its streamlines spread northward or southward by 175° E (see Fig. 1 of Qiu and Chen, 2011), the high radioactive Cs

waters would be stagnant around the central Pacific and dispersed not only eastward but also northward and southward slowly. Then the water would be transported further northward and southwestward by the recirculations of subarctic and subtropical gyres, respectively (Fig. 3c).

The $^{134}$Cs / $^{137}$Cs ratio was calculated using the data that were decay-corrected on 6 April 2011, when the maximum direct radioactivity discharged into the ocean (Buesseler et al., 2011). The ratios were calculated only for the samples for which both $^{134}$Cs and $^{137}$Cs were detected and had measurement errors smaller than 10 %. The ratios were varied from $0.55 \pm 0.17$ to $1.14 \pm 0.15$ throughout the studied periods (Fig. 4). The mean values of $^{134}$Cs / $^{137}$Cs ratio gradually decreased from $0.96 \pm 0.10$ ($n = 15$) in July 2011, $0.90 \pm 0.07$ ($n = 8$) in October 2011 to $0.85 \pm 0.12$ ($n = 39$) in July 2012. Such a decreasing trend of the $^{134}$Cs / $^{137}$Cs ratio may indicate the dilution process of radioactive Cs derived from the FNPP with the background $^{137}$Cs mostly originating from the nuclear weapons tests conducted in the late 1950s and the 1960s (Hirose and Aoyama, 2003).

At each station where a 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 indicate the dilution process of the FNPP-derived radioactive Cs within the studied area (Fig. 5a). The relationship between $^{137}$Cs concentration and latitude is also shown in Fig. 5b, since temperature in the studied area mainly depends on the latitude, high temperature in low latitude (KE and subtropical region) and low temperature in high latitude (subarctic region). Although both the concentration of $^{137}$Cs and temperature were almost in the same ranges between the two years at 175° 30′ 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 the 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. 5a), and from 42° 00′ N to 33° 30′ N in July 2012 (Fig. 5b). It is noteworthy that the $^{137}$Cs concentrations around 25 °C, located south of KE, were almost comparable between these two years. This may imply that the drastic changes in the concentration of radioactive Cs had been limited in the northern area of KE where temperature was lower than 25 °C. The difference in the relationship between $^{137}$Cs and temperature among the two years with convergence at a few m Bq kg$^{-1}$ around 25 °C suggests that the high radioactive Cs derived from the 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 the 16 months after the FNPP accident. Taking into account the result of particle tracking experiment and the weakness of the eastward KE jet around the central North Pacific, most of the radioactive Cs from the FNPP would be transported eastward within one year, and then it would be diluted broadly not only eastward but also north- and southward with a decrease in its concentration.

## 5 Conclusions

Radioactive Cs derived from the FNPP was distributed broadly in the western and central North Pacific, especially in the northern part of the Kuroshio Extension during July 2011 and July 2012. The dense radioactive Cs water masses moved away from the western North Pacific until October 2011, and the concentration of radioactive Cs at 144° E transect had decreased almost one order of magnitude 16 months after the FNPP accident. A particle tracking experiment, $^{134}$Cs / $^{137}$Cs ratio and relationship between temperature and concentration of $^{137}$Cs suggest that a considerable amount of radioactive Cs from the FNPP has been dispersed eastward from the western North Pacific to the central North Pacific during the first year after the FNPP accident, and then dispersed not only eastward but also north- and southward in the central and western North Pacific. We focus on the radioactive Cs in the surface seawater. Thus significant uncertainties remain in order to understand the comprehensive distribution and dispersion pattern of radioactive Cs derived from the FNPP in the North Pacific. Future studies must include vertical distribution patterns of the radioactive Cs, estimation of atmospheric fallout to the North Pacific, and terrestrial input of radioactive Cs such as river runoff.

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