The reducing rate of radiocesium ^{137}Cs in the North Pacific surface water after the TEPCO Fukushima Dai-ichi Nuclear Power Plant accident

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Abstract

We started monitoring the radiocesium released from Fukushima in the North Pacific surface water from May 2011 after the accident soon to June 2016, using the cruise of the *Oshoro-maru* of Hokkaido University. We found that the reducing rate from the ocean surface of ^{137}Cs regardless of the distance from Fukushima was almost constant at 0.0033 ± 0.0005 day^{-1}. This finding indicated that 96 % of ^{137}Cs in the surface water over the North Pacific were removed by 1000 days after the accident from the surface water to the ocean interior excluding the decrease by radioactive disintegration.

Keywords

Fukushima Dai-ichi nuclear power plant accident, ^{137}Cs, North Pacific Ocean, Reducing rate

Introduction

The Great East Japan Earthquake that happened on March 11th, 2011 was one of the most powerful in world history, and the subsequent massive tsunami caused extensive damage to the coastal areas of the Tohoku region of Japan. In the Fukushima Dai-ichi Nuclear Power Plant (FNPP1), a nuclear accident occurred as the result. It is then estimated that 15.2–18.3 PBq (P; Peta = 10^{15}) of radiocesium (^{137}Cs) were released to the North Pacific Ocean through atmospheric deposition and the direct discharge of contaminated water by the collapse of a nuclear reactor [1]. Most of the contaminated water was discharged into the ocean via the harbor facility for FNPP1 during the first two months of the accident [2].

It was important to estimate the source term of the radioactive materials in the ocean, the atmosphere, and the land area in those days. Many research institutions carried out intensive observations to obtain the surface and vertical distributions of radiocesium in the North Pacific in the years following the accident [1–11]. It became clear that there are two routes for the inflow of radioactive cesium into the ocean. Immediately after the accident: first, the radioactive cesium released into the atmosphere by the explosion of the nuclear reactor fell into the ocean, and second, the contaminated water was discharged directly into the ocean. As a result, the total amount of ^{137}Cs released to the ocean by the FNPP1 accident was estimated at 15.2–18.3 PBq by using compiled data and model simulations [1]. This accident is characterized by the widespread dispersion of radioactive materials via the oceans, compared to other nuclear accidents that have occurred around the world [12–14]. The Fukushima-derived radiocesium discharged in the North Pacific Ocean had been transported primarily eastward by the surface currents of the Kuroshio Current and Kuroshio Extension Current. The behavior of the Fukushima-derived radiocesium simulated numerically by a regional ocean model also reproduced being transported from the coastal area of FNPP1 to the eastern region of the North Pacific Ocean [13, 14]. Furthermore, in the observations, the main body of Fukushima-derived radiocesium had spread to around 170°W by the summer of 2012 [9], and the subsurface maximum of radiocesium was found at 200–600 m depth at the same time [10]. It arrived further eastward to the Canadian continental shelf by June 2013 [7].

The present monitoring of the radioactive materials in the ocean continues only in the vicinity of FNPP1, with little ongoing research being conducted in the North Pacific. The *Oshoro-maru*, the research and training vessel of Hokkaido University Japan carried out about 10 cruises for the fieldwork of the students every year during the period from 2011 to 2016. This vessel has no regular routes every year but covered the wide areas of the North Pacific including the Bering Sea. We tried to continue to measure ^{137}Cs concentration in the surface water collected along the route of almost all the cruises of the *Oshoro-maru* beginning approximately two months after the accident at FNPP1. Therefore, the *Oshoro-maru* cruises were possible to monitor continuously the radioactive material over the North Pacific.

In this article, using the results obtained by the sampling of the Oshoro-maru Cruise during the period from approximately two months after the accident of FNPP1 to December 2016, we describe the behavior of concentration and spatiotemporal distribution of ^{137}Cs released from Fukushima in the surface waters of the North Pacific. We also estimate the reducing rate of ^{137}Cs in the surface water of the ocean from the behavior of ^{137}Cs activity concentrations and discuss the relationship between the distance from FNPP1 to the observed sea area and each reducing rate in that sea area.

Experimental

Sampling and analytical methods

Seawater samples for radiocesium measurement were collected on the surface layer in the wide region of the North Pacific Ocean including from the Japan Sea to the Bering Sea, by using the *Oshoro-maru* cruises performed from May 2011 to June 2016 (Fig. 1). The number of water samples obtained in this study was 501 samples, which were collected in 10 L plastic bags by pumping up under the water surface to about 5m when the ship overpassed these stations. These seawater samples were preserved at normal temperatures without filtering. In our laboratory, the activities of ^{137}Cs in the collected seawater samples were measured in the following analytical procedure according to Levy et al. [15]. After acidifying by the addition of concentrated nitric

(16 mL; FUJIFILM Wako Pure Chemical Co., Ltd., Guaranteed Reagent) acid in the seawater sample (8 L), cesium chloride (0.10 g; FUJIFILM Wako Pure Chemical Co., Ltd., 99.9 %) was added. Then, 1.60 g of ammonium phosphomolybdate (AMP; KANSO Co., Ltd., High Purity) was added. AMP/Cs compounds that absorbed cesium in a seawater sample were formed by one-hour bubbling. Thereafter, AMP/Cs compounds were collected by filtering with a membrane filter (pore size 0.45 μm) and were dried by using the desiccator. The activities of ^{137}Cs in these compounds were measured for 48 hours by using the well–type germanium semiconductor detector. The detection limit of ^{134}Cs and ^{137}Cs was 0.002 mBq L^{-1}. We corrected the measured ^{137}Cs to the value at the sampling time and defined it as $^{137}Cs_{obs}$. In addition, we corrected the measured ^{137}Cs to the value at the FNPP1 accident (March 11th, 2011(acc)) and defined it as $^{137}Cs_{acc}$.

Results and discussion

The horizontal distributions of ^{137}Cs in the surface water of the North Pacific

The radioactive cesium data in this study were continuously obtained in the surface water on the course line of the "Oshoro-maru" cruise. To investigate the time-series variations in the horizontal distributions of ^{137}Cs in the surface water, the radioactive cesium data obtained from the samples collected at regular intervals after the FNPP1 accident were compiled and plotted on the map (Fig. 2). Therefore, these data were grouped into the nine periods from May 2011 to June 2016 as follows, the period I (from May 2011 to October 2011), the period II (from December 2011 to June 2012), the period III (from July 2012 to December 2012), the period IV (from May 2013 to September 2013), the period V (from October 2013 to April 2014), the period VI (from May 2014 to November 2014), the period VII (from December 2014 to May 2015), the period VIII (from June 2015 to December 2015), the period IX (from March 2016 to June 2016). The data sets for the periods I through IX are represented in Figs. 2 (a) – (i), respectively. During the period I, a high concentration of ^{137}Cs was existed in the off region from Fukushima Prefecture to eastern Hokkaido. It was confirmed that this high

concentration area was distributed along with the Kuroshio Extension Current toward the east side of the North Pacific (Fig. 2 (a)). Although the concentration level decreased half a year after the accident, the same distribution tendency as in the period I was observed in the period II, the first year after the accident (Fig. 2 (b)). It makes us imagine that even one year after the accident, ^{137}Cs was being transported to the Northeastern Pacific Ocean by the effect of the Kuroshio Extension Current. The ^{137}Cs existing in the ocean during our observation period was mostly due to the accident at the FNPP1 [3]. The horizontal distribution of ^{137}Cs on the ocean surface immediately after the FNPP1 accident simulated by Tsumune et al. [13, 14] and the spatial distribution during the period I are very similar. It was also found that the radioactive cesium released from the FNPP1 was mixed with the Kuroshio Extension Current and transported to the eastern North Pacific Ocean. After the period III, the concentration levels have decreased further, making it In the surface water at all the regions, the radiocesium concentration had decreased to the background levels in the ocean until the summer of 2016. Reports of radioactive cesium detected in seawater from the middle revel depths of the North Pacific Ocean [10], the sediment particles collected from time-series sediment traps in the western North Pacific Ocean for one year immediately after the accident [16], and the zooplankton community collected during a survey carried out 10 months after the accident in the western North Pacific [17, 18] have been published, respectively. Although the radioactive cesium released by the FNPP1 accident spread in the whole North Pacific Ocean, it was shown the result which their activities in surface sea water were removed by the physical movement and the biochemical substance circulation mechanism. We were able to demonstrate the spatiotemporal attenuation of ^{137}Cs in the surface water for approximately five years after the FNPP1 accident.

Time-series of ^{137}Cs in the surface water

Activity concentrations of ^{137}Cs in all the observation sites from May 2011 to July 2016 are shown on the elapsed time after the FNPP1 accident (Fig. 3 (a)). Though we already reported in the previous paper [11] about the ^{137}Cs data until October 2014, the concentration of ^{137}Cs was a maximum of 0.425 Bq L^{-1} in the observation from May to July 2011. We have not detected ^{134}Cs whose half–life is about two years by the

observation in 2013 and afterward. However, even during the period when ^{134}Cs were detected, the radioactivity ratio of ^{134}Cs / ^{137}Cs was less than 1, as reported by Aoyama et al. [19]. By half a year after the FNPP1 accident, the concentrations of ^{137}Cs showed a decreasing trend, moving to an equilibrium state of 0.001 Bq L^{-1} grade in almost all areas [20]. This temporal variation was the same as the decreasing trend of ^{137}Cs concentration in the private port belonging to FNPP1 reported by Kanda [12]. The large–scale and direct discharge to the ocean of radioactive material contaminated water were stopped by the beginning of April 2011. It is thought that it is the result of removing radioactive cesium from the ocean surface water with the progress of time. It is almost measured at a low level in the observation after October 2014 in the whole area. In our previous reports [11], we noted that several observations in 2012, 2013, and 2014 from October to December showed relatively high values of ^{137}Cs. These observation sites were found to be in shallow coastal waters. Therefore, it was assumed that the high values were due to the supply of ^{137}Cs to the surface layer caused by the sudden inflow of radioactive materials from the river [21, 22] and the resuspension of coastal marine sediments due to strong winds. Uchiyama [23] proved these phenomena using the numeric model. From 2015 downward, the high value of the ^{137}Cs did not have been measured, including the near coast sites. We have suggested the possibility that the inflow from a land area is decreasing.

On the other hand, each average value of the nine periods shown in the previous chapter is shown in Fig. 3 (b). The average value of the ^{137}Cs in the period I immediately after the accident was 0.051 Bq L^{-1}. From period I to period II, the average value decreased sharply to 0.010 Bq L^{-1}, and thereafter became almost constant in the low range of 0.0043–0.0063 Bq L^{-1}. We have not measured ^{134}Cs and ^{137}Cs prior to the start of our measurements. Therefore, our data alone cannot be used to estimate the background value of Cs prior to the Fukushima nuclear accident. Inomata et al. [24] reported ^{137}Cs values of 0.0011–0.0032 Bq L^{-1} (average 0.0022 ± 0.0003 Bq L^{-1}) in the ocean surface water prior to the Fukushima nuclear accident in the western North Pacific including the area off Fukushima for the period 1990–2005. Consequently, we estimated the contribution ratio of the background value to the ^{137}Cs from the Fukushima nuclear accident using the average value of 0.0022 ± 0.0003 Bq L^{-1} as the background value of

^{137}Cs before the Fukushima nuclear accident. As a result, for the concentration of ^{137}Cs in ocean surface water, the background accounted for only 4 % of the average concentration of 0.0515 ± 0.0105 Bq L^{-1} in the ocean surface water in the period I of 2011 (May 2011 to October 2011), immediately after the Fukushima nuclear accident (Fig. 3 (b)). In contrast, in the period IX of 2016 (March 2016 to June 2016), the background value accounted for 38 % of the average concentration of 0.0057 ± 0.0004 Bq L^{-1} in ocean surface water, suggesting that there are still significant residual effects from the Fukushima nuclear accident (Fig. 3 (b)).

The figure inserted in Fig. 3 (b) shows the time-series changes in the natural logarithm of the average value until period IV when an almost constant value of ^{137}Cs. The temporal change in the average concentration of ^{137}Cs could be expressed almost linearly, it found that this slope had a reducing rate of 0.0036 day^{-1} until the end of 2013 when it passed after the accident for about 1000 days. It is presumed that it was removed from the surface of the ocean at a constant rate.

The reducing rate of ^{137}Cs in the surface water

It could be shown, as in many reports, that the concentration of cesium on the sea surface we measured decreased after the accident. Here, we attempted to study the locational reducing rate of the cesium concentration in the surface water of the western North Pacific near Japan. Therefore, the relationship between the distance from FNPP1 to the observed sea area and the reducing rate of ^{137}Cs in that sea area was analyzed. First, the distances from the position of FNPP1 to each sampling point were calculated respectively. The starting point was the position of the dedicated pier for FNPP1 (37° 25.28' N, 141° 2.2' E). Between FNPP1 and the sampling point so that it is not blocked by land areas, we have picked up data from observation points on the Pacific Ocean, excluding the Japan Sea, the Okhotsk Sea, and the Bering Sea in our survey area. Then, groupings were performed according to the distance from FNPP1. It was divided into the following 6 sea areas, within 100 miles from FNPP1, between 100 and 200 miles, between 200 and 300 miles, between 300 and 400 miles, between 400 and 600 miles, and between 600 and 800 miles. In addition, it was analyzed using data within 800 miles, because the number of observation sites in this study was small at locations more than

800 miles away. Fig. 4 shows the relation between the time-series of $^{137}Cs_{obs}$ in the surface water and elapsed time for each distance from FNPP1. The concentration of $^{137}Cs_{obs}$ (the value corrected at the sampling time) in the period I was represented by "1". The elapsed time was set to the median of each time compartment (as before chapter, periods I–IX). The ratio of concentration per elapsed time did not exceed 1 in all periods when the concentration in period I of each region was set to 1 (Fig. 4 (a)). In other words, it shows the trend that the concentration of $^{137}Cs_{obs}$ was decreasing in time series in all regions. Therefore, if the concentration ratio of each region shown in Fig. 4 (a) is replaced with the natural logarithm, a linear decreasing tendency can be seen, and a constant reduction can be obtained (Fig. 4 (b)). Although the characteristics of the reducing rate of ^{137}Cs were shown for each region, the ^{137}Cs concentration clearly decreased regardless of the distance from FNPP1.

Table 1 summarizes the reducing rates (RR) of $^{137}Cs_{obs}$ (the value corrected for radioactive decay to sampling time) and $^{137}Cs_{acc}$ (The value corrected for radioactive decay to the FNPP1 accident (March 11th, 2011(acc))) in each region. The RR calculated from $^{137}Cs_{obs}$ and $^{137}Cs_{acc}$ ranged from $-0.00166\ day^{-1}$ to $-0.00402\ day^{-1}$ ($-0.00337 \pm 0.0005\ day^{-1}$ on average) and $-0.00159\ day^{-1}$ to $-0.00477\ day^{-1}$ ($-0.00331 \pm 0.0005\ day^{-1}$ on average) in all regions, respectively. The straight lines showing the RR showed highly correlated results. The RR calculated from $^{137}Cs_{obs}$ reflects both the radioactive decay of ^{137}Cs and the process of removing ^{137}Cs from the surface water into the ocean. The RR calculated from $^{137}Cs_{acc}$ has been corrected for radioactive decay to the acc, so it reflects only the process of removing ^{137}Cs from the surface water into the ocean. Using the RR estimated from $^{137}Cs_{acc}$, we estimated the percentages of ^{137}Cs concentration removed from the surface water by 1000 days from the acc (Fig. 3 (b)) to be 96.3%, indicating that most of the ^{137}Cs from the FNPP1 accident dispersed to the ocean surface removed into the ocean interior by 1000 days after the accident.

Conclusions

To monitor the radiocesium released by the accident of FNPP1 on 11 March 2011, we measured concentrations of ^{137}Cs at about five hundred sites in the surface water over the North Pacific Ocean using the cruise of the *Oshoro-maru* from May 2011 to June 2016. During our one-year observation period after the accident of FNPP1, we were able to similarly assess the eastern transfer of the FNPP1-derived radiocesium along the Kuroshio Extension Current on the surface of the ocean, as previously reported. And, in all the survey areas, it became difficult to detect cesium one year after the accident. The ^{137}Cs concentrations in surface water were shown to have decreased to natural levels regardless of the distance from FNPP1, even though the accident had not been treated. The ^{137}Cs concentrations in surface water were shown to have decreased to natural levels regardless of the distance from FNPP1, even though the accident had not been treated.

Results of analyzing these data, we obtained the following three new findings;

(i) The average ^{137}Cs in the North Pacific had a reducing rate of 0.0036 day^{-1} until the end of 2013 when it passed after the accident for 1000 days.

(ii) The reducing rate of ^{137}Cs in the surface water regardless of the distance was almost constant at 0.0033 ± 0.0005 day^{-1} until 1000 days after the accident.

(iii) ^{137}Cs in the surface water over the North Pacific were removed by 96 % until 1000 days from the surface water to the ocean interior excluding the decrease by radioactivity decay.

Most of the radiocesium derived from Fukushima directly or via the atmosphere to the surface water is rapidly transferred to deeper layers of the ocean, suggesting that it remains in the ocean until radioactive decay due to its half-life. By the latest report, it was indicated that FNPP1-derived radiocesium was again detected in the coastal water around the island of Japan [25, 26]. Although most of the FNPP1-derived radiocesium has once disappeared from the surface side, continued monitoring at the surface is needed in the future as part of our role to monitor recirculation from the deep to the surface layer by oceanic physical and biochemical dynamics.

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Funding and/or Conflicts of interests/Competing interests

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Figure captions

Fig. 1 Sampling stations of surface water for radiocesium measurement from May 2011 to June 2016. We here used the Ocean Data View [27] to draw these figures [http://odv.aw.de].

Fig. 2 The horizontal distributions of ^{137}Cs in the sea surface water of the North Pacific during the period from May 2011 to June 2016, (a) period I (from May 2011 to October 2011), (b) period II (from December 2011 to June 2012), (c) period III (from July 2012 to December 2012), (d) period IV (from May 2013 to September 2013), (e) period V (from October 2013 to April 2014), (f) period VI (from May 2014 to November 2014), (g) period VII (from December 2014 to May 2015), (h) period VIII (from June 2015 to December 2015), (i) period IX (from March 2016 to June 2016). Dots are the sampling site. The range of ^{137}Cs radioactivity for creating these figures is indicated using three scales, following as (a) is from 0 to 0.2 Bq L^{-1}, (b) to (e) are from 0 to 0.05 Bq L^{-1} and (f) to (i) are from 0 to 0.025 Bq L^{-1}. We here used the Ocean Data View [27] to draw these figures [http://odv.aw.de].

Fig. 3 Time-series of ^{137}Cs in the surface water over the North Pacific from May 2011 to June 2016. (a) All the concentrations of ^{137}Cs in this study. (b) The averaged value of ^{137}Cs radioactivity as each during the 9 periods (I: May 2011 to October 2011, II: December 2011 to June 2012, III: July 2012 to December 2012, IV: May 2013 to September 2013, V: October 2013 to April 2014, VI: May 2014 to November 2014, VII: December 2014 to May 2015, VIII: June 2015 to December 2015, IX: March 2016 to June 2016). The inserted figure in (b) is the Natural logarithm of $^{137}Cs_{obs}$ content in the surface water between the period I and IV. Errors show the standard errors.

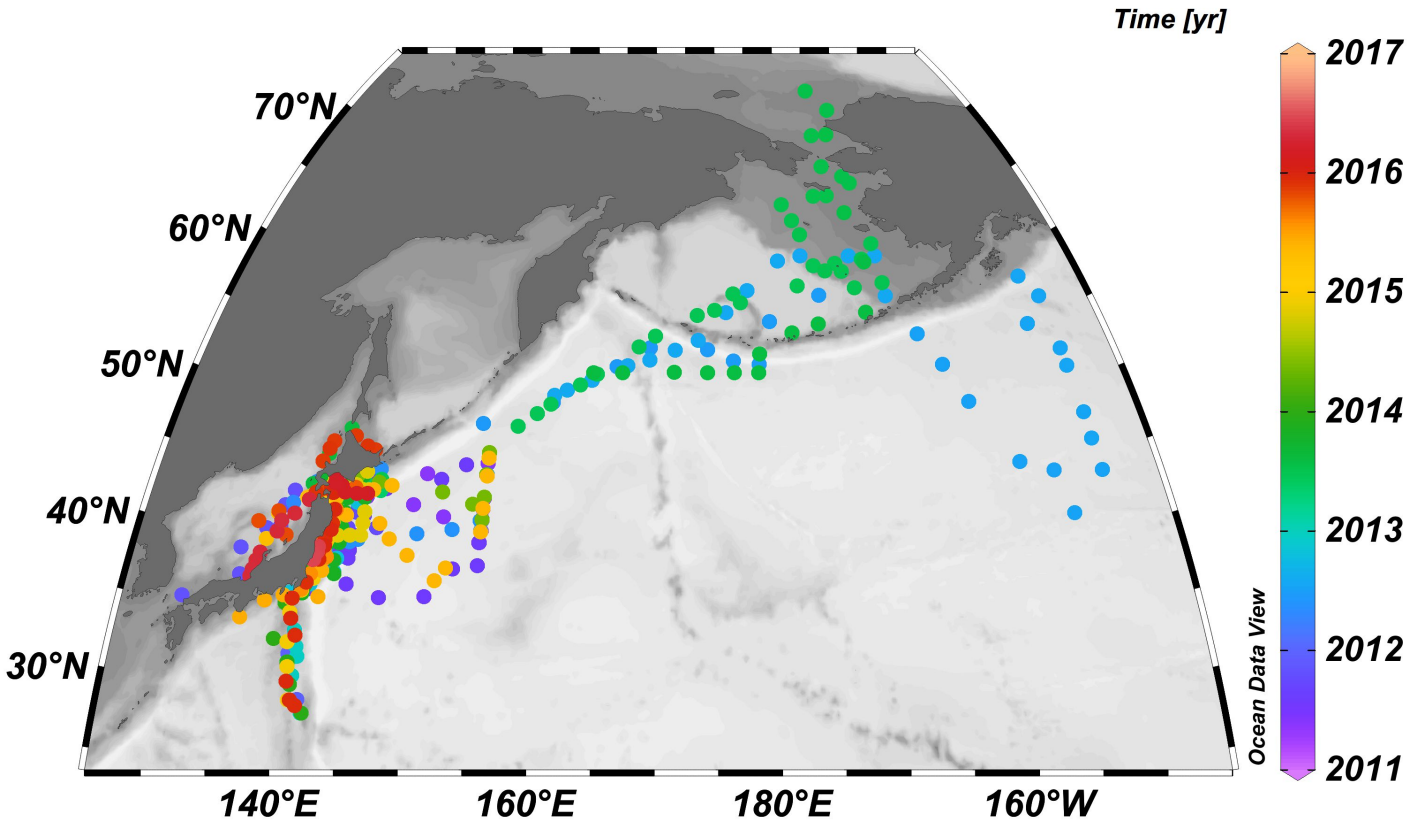
380 **Fig. 4** The relation between the time-series of $^{137}Cs_{obs}$ in the surface water and elapsed
381 time for each distance from FNPP1. (a) Time-series of the ratio of $^{137}Cs_{obs}$ to that in the
382 period I. (b) Same as (a) but the Natural logarithm of the ratio of $^{137}Cs_{obs}$ to that in the
383 period I for the vertical axis. Each distance was divided as follows; the regions within
384 100 miles (open circle), between 100 miles and 200 miles (closed circle), between 200
385 miles and 300 miles (open square), between 300 miles and 400 miles (closed square),
386 between 400 miles and 600 miles (open triangle) and between 600 miles and 800 miles
387 (closed triangle) from the position of FNPP1. This mile is a nautical mile, 1.8 km.
388

Tables

Table 1 Reducing rates (RR) of ^{137}Cs content in the surface water for each distance from FNPP1 (day^{-1}). (i) RR based on the time series of $^{137}Cs_{obs}$; (ii) RR based on the time series of $^{137}Cs_{acc}$.

			The distance from FNPP1 (miles)							
			0-100	100-200	200-300	300-400	400-600	600-800	Average	SE
(i)	$^{137}Cs_{obs}$	RR (day^{-1})	−0.00378	−0.00342	−0.00484	−0.00166	−0.00253	−0.00402	−0.00337	0.00046
		(r)	(0.89)	(0.84)	(0.84)	(0.95)	(0.78)	(0.92)		
(ii)	$^{137}Cs_{acc}$	RR (day^{-1})	−0.00371	−0.00335	−0.00477	−0.00159	−0.00247	−0.00395	−0.00331	0.00046
		(r)	(0.89)	(0.84)	(0.83)	(0.95)	(0.77)	(0.92)		

$^{137}Cs_{obs}$ is the value corrected for radioactive decay to sampling time. $^{137}Cs_{acc}$ is the value corrected radioactive decay to the FNPP1 accident (March 11th, 2011). RR of $^{137}Cs_{obs}$ reflects both the radioactive decay of ^{137}Cs and the processes of removing ^{137}Cs from the surface water to the ocean interior. RR of $^{137}Cs_{acc}$ reflects only the processes of removing ^{137}Cs from the surface water to the ocean interior due to correcting the measured value to that at the FNPP1 accident. The value of r in parentheses is the correlation coefficient. Errors show the standard errors.


Time [yr]
2017
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Ocean Data View
70°N
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50°N
40°N
30°N
140°E
160°E
180°E
160°W

Figure 1

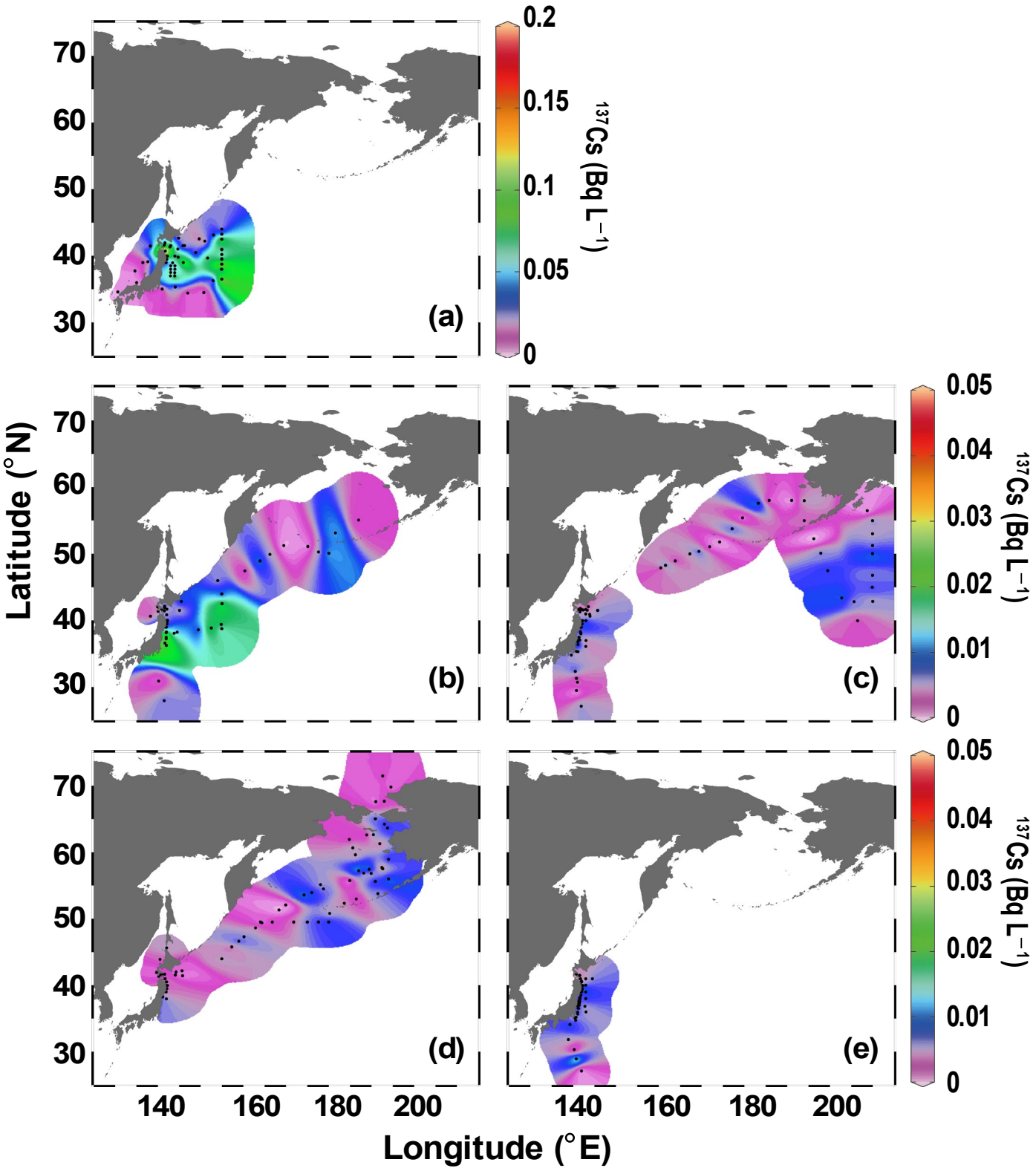
(a)
(b)
(c)
(d)
(e)
0.2
0.15
0.1
0.05
0
0.05
0.04
0.03
0.02
0.01
0
^{137}Cs (Bq L^{-1})
Latitude (°N)
Longitude (°E)
70
60
50
40
30
140
160
180
200

Figure 2

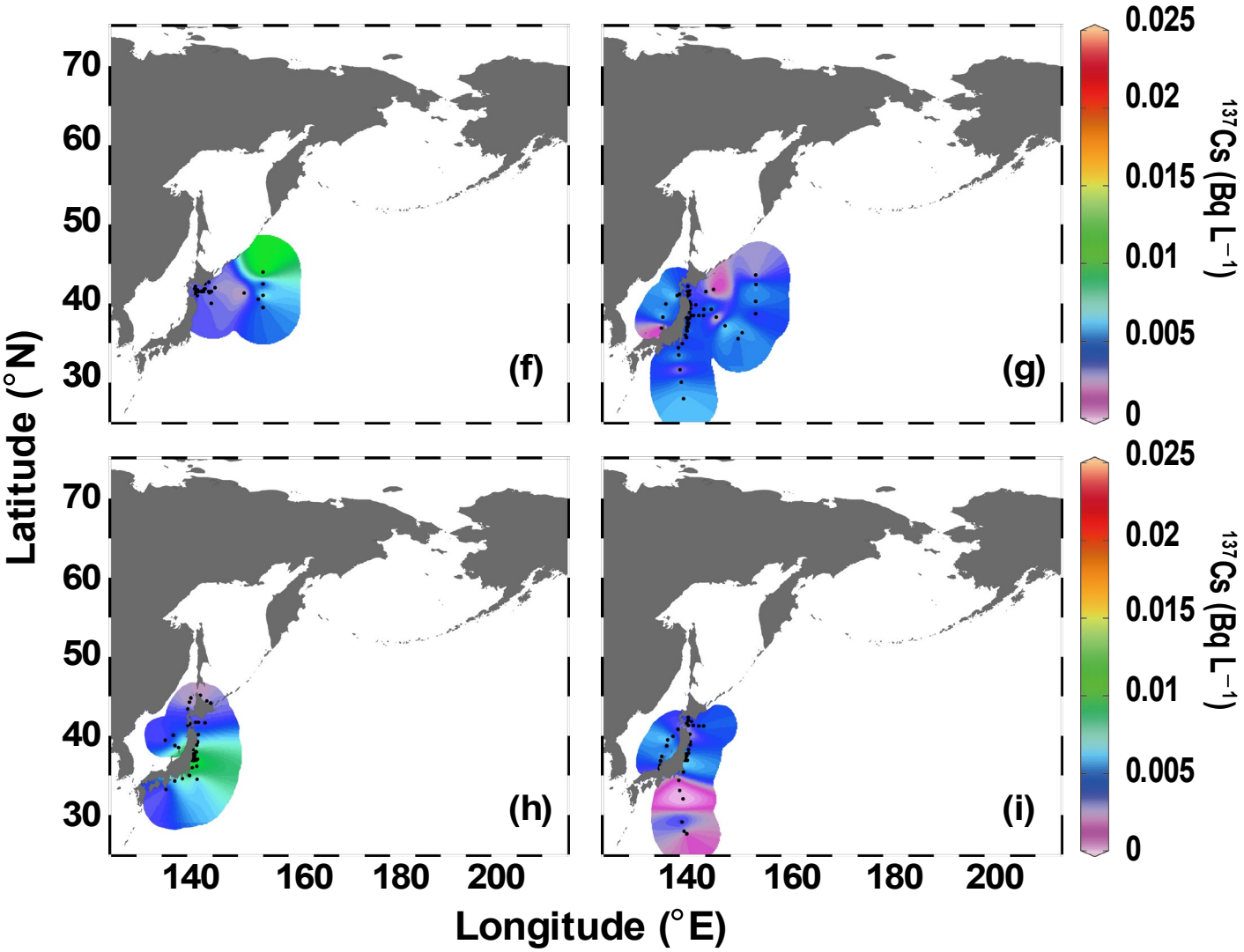

Figure 2 continued

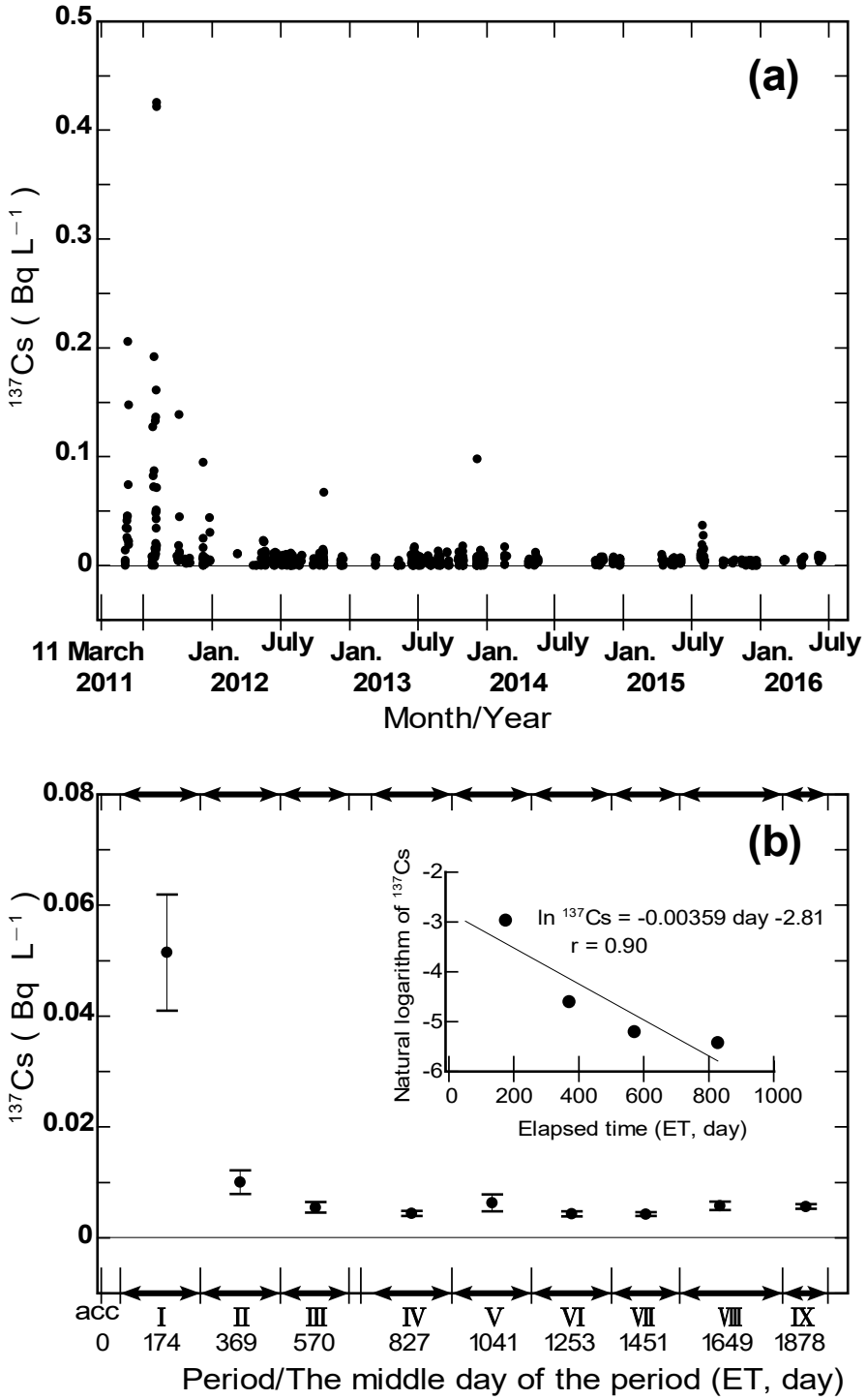
(a)
0.5
0.4
0.3
0.2
0.1
0
^{137}Cs (Bq L^{-1})
11 March 2011
Jan. 2012
July
Jan. 2013
July
Jan. 2014
July
Jan. 2015
July
Jan. 2016
July
Month/Year
(b)
0.08
0.06
0.04
0.02
0
^{137}Cs (Bq L^{-1})
ln ^{137}Cs = -0.00359 day -2.81
r = 0.90
Natural logarithm of ^{137}Cs
-2
-3
-4
-5
-6
0
200
400
600
800
1000
Elapsed time (ET, day)
acc
0
I
174
II
369
III
570
IV
827
V
1041
VI
1253
VII
1451
VIII
1649
IX
1878
Period/The middle day of the period (ET, day)

Figure 3

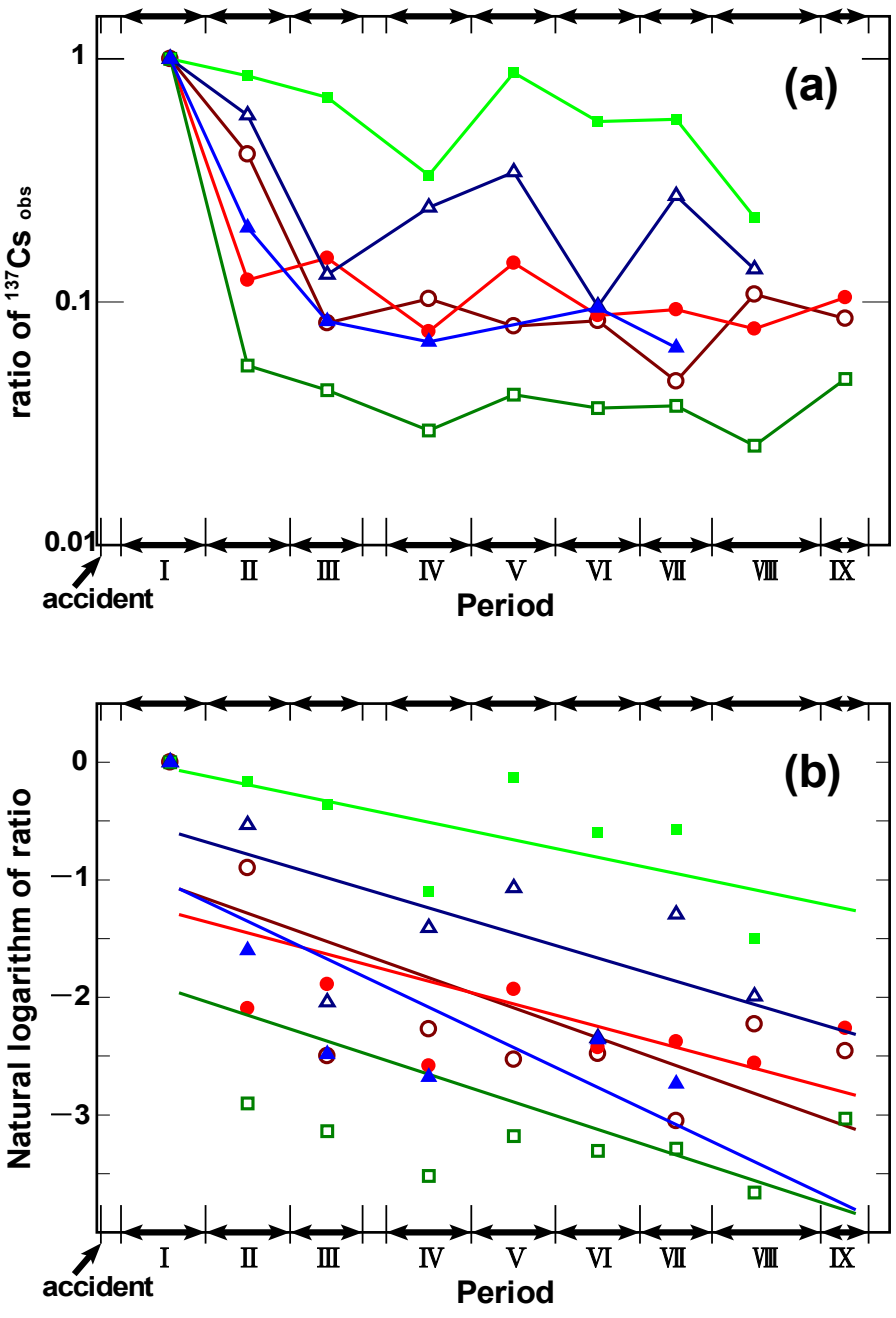
(a)
1
0.1
0.01
ratio of $^{137}Cs_{obs}$
accident
I
II
III
IV
V
VI
VII
VIII
IX
Period
(b)
0
−1
−2
−3
Natural logarithm of ratio
accident
I
II
III
IV
V
VI
VII
VIII
IX
Period

Figure 4