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Iodine-129 concentration in seawater near Fukushima before and after the accident at the Fukushima Daiichi Nuclear Power Plant

T. Suzuki[1], **S. Otosaka**[1], **J. Kuwabara**[2], **H. Kawamura**[1], **and T. Kobayashi**[1]

[1]Japan Atomic Energy Agency, Ibaraki, Japan
[2]Japan Atomic Energy Agency, Aomori, Japan

Correspondence to: T. Suzuki (suzuki.takashi58@jaea.go.jp)

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Abstract. Anthropogenic radionuclides were released into the environment in large quantities by the Fukushima Daiichi Nuclear Power Plant (1FNPP) accident. To evaluate accident-derived ^{129}I, the ^{129}I concentrations in seawater before and after the accident were compared.

Before the accident (2008–2009), the ^{129}I concentrations in the western margin of the North Pacific between 32° N and 44° N showed a latitudinal gradient that was expressed as a linear function of latitude. The highest and average ^{129}I concentrations after the accident were 73 times and approximately 8 times, respectively, higher than those before the accident in this study area. Considering the distribution of ^{129}I in surface seawater, the accident-derived ^{129}I in the southern and northern stations of the 1FNPP was predominantly supplied by seawater advection and atmospheric deposition (including microbial volatilization), respectively.

As of October 2011, depth profiles of ^{129}I revealed that ^{129}I originating from the 1FNPP existed mainly in the upper 100 m depth. From the depth profiles, the cumulative inventories of accident-derived ^{129}I were estimated to be $(1.6–9.6) \times 10^{12}$ atoms m^{-2} in this study area.

On the basis of the ^{129}I data in the seawater near Fukushima, the effective dose of ^{129}I from seafood ingestion was much smaller than the annual dose limit.

1 Introduction

Significant fission products such as ^{134}Cs (half-life: 2.06 yr), ^{137}Cs (half-life: 30.2 yr), and ^{131}I (half-life: 8.02 days) were released into the environment by the Fukushima Daiichi Nuclear Power Plant (1FNPP) accident caused by the Great East Japan earthquake and tsunami on 11 March 2011 (Honda et al., 2012; Momoshima et al., 2012). From the viewpoint of environmental safety with respect to nuclear accidents, ^{134}Cs, ^{137}Cs, and ^{131}I are regarded as important radionuclides because of their high fission yield and high γ-ray energy.

Radioiodine can enter the human body via food and drinking water and then selectively accumulate in the thyroid, creating an irradiation risk. A significantly higher incidence of thyroid cancer in children living around the Chernobyl accident site has been reported (Baverstock et al., 1992). Although thyroid cancer related to ^{131}I exposure as a result of nuclear accidents has been of considerable concern, less attention has been paid to the increasing inventory of ^{129}I (half-life: 1.57×10^7 yr) from the 1FNPP accident. Because ^{129}I has a long half-life, the amount and behavior of accident-derived ^{129}I in the environment should be investigated to address concerns about the radiological impacts to future generations. Measuring ^{129}I in environmental samples also offers an opportunity to retrospectively analyze the migration of accident-derived ^{131}I, which has decayed and become undetectable from the passage of post-accident time.

Anthropogenic ^{129}I is released into the environment from nuclear fuel reprocessing plants and is then transported mainly to the northern hemisphere (Suzuki et al., 2010; Snyder et al., 2010; Toyama et al., 2012). Thus, the ^{129}I level in the environment is already increased from its natural level. Soil and seawater play a role as a reservoir of ^{129}I (Muramatsu et al., 2004). The impact of ^{129}I on land from the 1FNPP accident has been reported (Miyake et al., 2012), but not the impact on the ocean. This study focuses on the impact of ^{129}I on seawater from the 1FNPP accident. To precisely

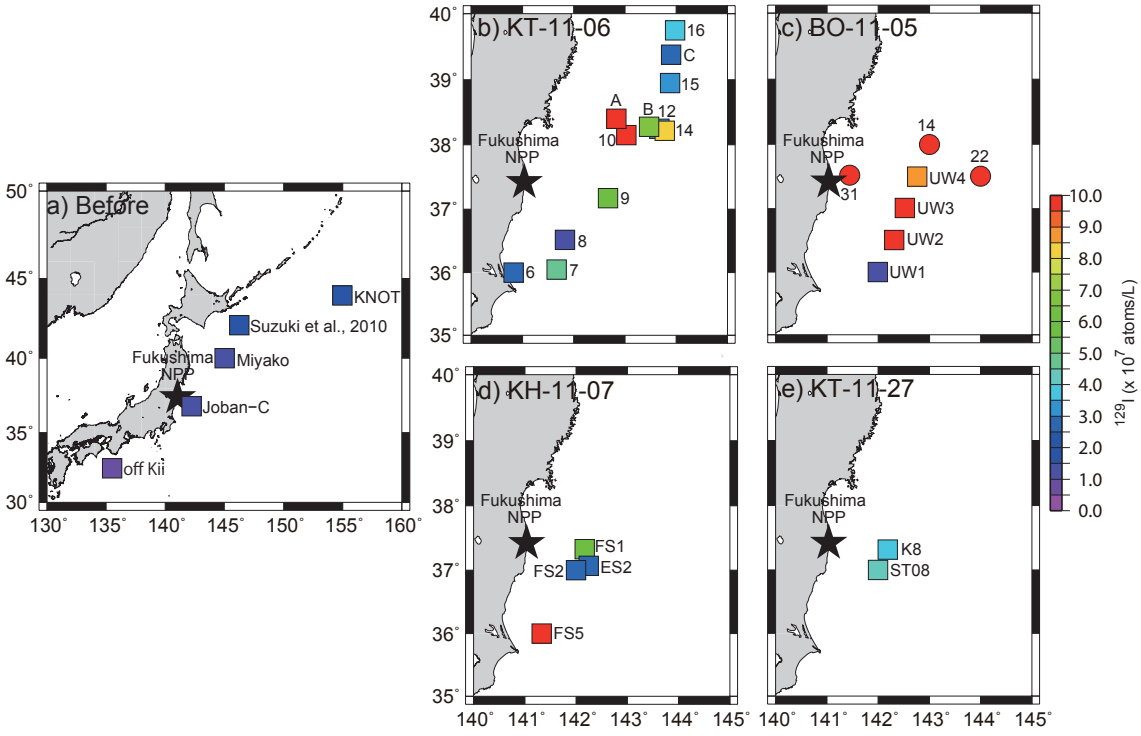

Fig. 1. Map of sampling locations and the result of surface ^{129}I concentrations before the 1FNPP accident **(a)** and afterwards **(b–e)**. After the accident, seawater sampling was undertaken during four cruises: **(b)** KT-11-06, **(c)** BO-11-05, **(d)** KH-11-07, and **(e)** KT-11-27. In **(c)**, circle symbols were cited from Hou et al. (2013). Red at the sampling locations indicates that the ^{129}I concentration is higher than 10×10^7 atoms L^{-1}.

evaluate the increase of ^{129}I caused by the accident, information about the ^{129}I before the accident in the western North Pacific Ocean is crucial. Many studies about ^{129}I in seawater have been conducted around nuclear fuel reprocessing plants (Keogh et al., 2007; Raisbeck and Yiou, 1999; Alfimov et al., 2004; Michel et al., 2012; Hou et al., 2001), but only a few studies have been undertaken in the Pacific Ocean (Suzuki et al., 2010; Povinec et al., 2010). Therefore, the background level of ^{129}I in the western North Pacific Ocean had not previously been determined.

Prior to the 1FNPP accident, a new nuclear fuel reprocessing plant was tested for routine operation at Rokkasho, Japan. Anticipating the release of ^{129}I from the operation of the reprocessing plant, seawater samples in the western North Pacific Ocean were collected to clearly define the background level before plant operations commenced. After the 1FNPP accident, seawater samples collected before the accident proved useful for evaluating the background level of ^{129}I in the western North Pacific Ocean. In this paper, we report the distributions of ^{129}I in the western North Pacific before and after the 1FNPP accident and evaluate the accident-derived ^{129}I in seawater. In addition, we infer the migration of ^{129}I off Fukushima after the accident and assess the internal dose of accident-related ^{129}I as a result of seafood consumption.

2 Experimental

2.1 Seawater sampling

Seawater sampling before the 1FNPP accident were conducted at four stations: KNOT (154°58′ E, 43°58′ N) on 10 May 2008 by the T/S *Oshoro Maru*; off Kii (135°31′ E, 32°28′ N) on 5 August 2008 by the R/V *Soyo Maru*; Joban C (142°13′ E, 36°48′ N) on 18 July 2009 by the R/V *Soyo Maru;* and Miyako (145°00′ E, 40°00′ N) on 20 July 2009 by the R/V *Soyo Maru*, as shown in Fig. 1a. Figure 1a includes another station that can provide a background level for 1FNPP in the western North Pacific Ocean (Suzuki et al., 2010).

Seawater sampling after the 1FNPP accident were undertaken by four expeditions: from 27 April to 1 May 2011 by the R/V *Tansei Maru*, cruise KT-11-06 (Fig. 1b); from 12 to 13 June 2011 by the R/V *Bosei-Maru*, cruise BO-11-05 (Fig. 1c); from 2 to 3 August 2011 by the R/V *Hakuho Maru*, cruise KH-11-07 (Fig. 1d); and on 29 October 2011 by the R/V *Tansei Maru*, cruise KT-11-27 (Fig. 1e). Seawater samples at eight stations were collected from surface to deep layers at stations A, B, and C by cruise KT-11-06; at stations FS1, ES2, and FS2 by cruise KH-11-07; and at stations ST08 and K8 by cruise KT-11-27. Fourteen surface seawater samples were collected at other stations.

2.2 Analytical procedure

Iodine was extracted from seawater samples by the solvent extraction technique (Suzuki et al., 2008). Ascorbic acid with hydrochloric acid was added to a seawater sample to reduce iodate to iodide after passing through a 0.45 µm filter. Iodide was oxidized to molecular iodine by the addition of sodium nitrite for the extraction to chloroform. Back extraction was carried out from chloroform using sodium sulfite by reducing molecular iodine to iodide. The extracted iodide was precipitated as silver iodide by silver nitrate. The silver iodide sample was washed by nitric acid and pure water and then loaded to a copper target holder.

Iodine isotopic ratios were measured by accelerator mass spectrometry at the Aomori Research and Development Center of the Japan Atomic Energy Agency. All measured data was normalized to the standard reference material having $^{129}I/^{127}I = (9.85 \pm 0.12) \times 10^{-13}$, which was obtained from the National Institute of Standards and Technology (NIST SRM 3230) (Suzuki et al., 2006).

Total iodine (iodate + iodide) concentrations in seawater samples were measured by cathodic stripping square wave voltammetry or quadrupole inductively coupled plasma mass spectrometry (Campos, 1997).

3 Results and discussion

3.1 Distribution of ^{129}I in surface seawater

3.1.1 Before the 1FNPP accident

With regard to the global distribution of ^{129}I in surface reservoirs, the latitudinal distribution provides us with information useful for understanding the fate of ^{129}I (Snyder et al., 2010). Thus, the ^{129}I concentrations in surface seawater before and after the 1FNPP accident with another published data (Hou et al., 2013) are plotted in Fig. 2 against the latitude of the sampling location. The surface ^{129}I concentrations are also plotted at the sampling locations in Fig. 1.

The concentration of ^{129}I before the 1FNPP accident was in the 0.94 (off Kii) – 1.83 (offshore of Kushiro: Suzuki et al., 2010) $\times 10^7$ atoms L^{-1} range (Table 1). The ^{129}I in surface seawater between 32° N and 44° N in the western North Pacific Ocean before the 1FNPP accident was high in the north and decreased with decreasing latitude. These concentrations correspond to the locations of nuclear fuel reprocessing plants, which is a primary source of ^{129}I in the environment. Since major reprocessing plants such as Sellafield (54°00′ N) in the United Kingdom, La Hague (49°30′ N) in France, and Hanford (46°37′ N) in the United States are located in the middle- to high-latitude regions of the Northern Hemisphere, ^{129}I originating from nuclear fuel reprocessing plants was mainly distributed over the Northern Hemisphere (Snyder et al., 2010; Suzuki et al., 2010; Moran et al., 1999). The latitudinal distribution of ^{129}I in surface seawater before the 1FNPP accident is attributed to the atmospheric deposition of ^{129}I originating from the major reprocessing plants and can be expressed as a linear function of latitude (Fig. 2).

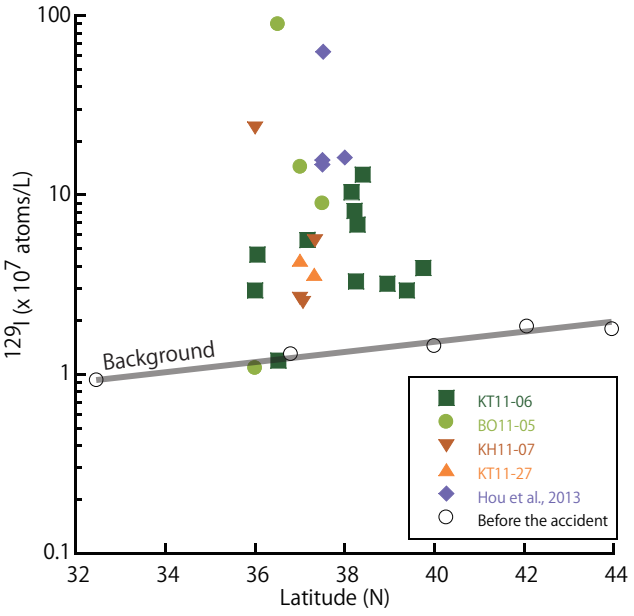

Fig. 2. ^{129}I concentrations in surface seawater before and after the 1FNPP accident as a function of latitude. The dark green, light green, dark orange, and light orange symbols indicate cruises KT-11-06, BO-11-05, KH11-07, and KT-11-27, respectively, after the 1FNPP accident. The purple symbols were cited from Hou et al. (2013). The white symbols indicate a cruise before the Fukushima NPP accident.

$$^{129}C_{BG} = 0.080 \times L - 1.7, (32 < L < 44), \quad (1)$$

where $^{129}C_{BG}$ (10^7 atoms L^{-1}) is the ^{129}I concentration in surface seawater before the 1FNPP accident and L (° N) is the latitude.

3.1.2 After the 1FNPP accident

In this study area, the ^{129}I concentrations in the surface seawater after the 1FNPP accident were in the $1.08–89.8 \times 10^7$ atoms L^{-1} (corresponding to $^{129}I/^{127}I = 4.47–362 \times 10^{-11}$) range (Figs. 1 and 2). The highest ^{129}I concentration ($^{129}I = 89.8 \times 10^7$ atoms L^{-1}, $^{129}I/^{127}I = 362 \times 10^{-11}$) was observed at station UW2 on 12 June 2011 during cruise BO-11-05, this value was 73 times higher than that before the accident, and this value was higher than that of the previous study (Hou et al., 2013). The average ^{129}I concentration in surface seawater after the 1FNPP accident was 9.3×10^7 atoms L^{-1}, a value approximately 8 times higher than that before the accident. Since routine operation of the Tokai reprocessing plant and test operation of the Rokkasho reprocessing plant had both ceased in 2008, the elevated ^{129}I can be considered to originate from 1FNPP. Accident-derived ^{129}I was detected

Table 1. Concentrations of ^{129}I in seawater samples.

Cruise	Station	Date	E	N	Depth (m)	Salinity PSU	Potential Temp. °C	Density σ_θ	^{129}I/^{127}I $\times 10^{-11}$	^{129}I $\times 10^7$ atoms L^{-1}	^{129}I nBq L^{-1}
Before the 1FNPP accident											
OS08	KNOT	10 May 2008	154°58′	43°58′	4	33.178	5.477	26.176	5.85 ± 0.21	1.78 ± 0.07	24.9 ± 1.0
					10	33.178	5.474	26.176	6.22 ± 0.22	1.83 ± 0.08	25.6 ± 1.1
					20	33.217	5.339	26.223	6.25 ± 0.19	1.78 ± 0.06	24.9 ± 0.8
					30	33.289	5.084	26.309	5.53 ± 0.25	1.56 ± 0.08	21.8 ± 1.2
					39	33.400	4.717	26.438	6.38 ± 0.23	1.72 ± 0.07	24.1 ± 1.0
					48	33.428	4.661	26.467	5.70 ± 0.24	1.63 ± 0.09	22.7 ± 1.2
					74	33.572	5.079	26.534	4.79 ± 0.21	1.53 ± 0.08	21.4 ± 1.0
					99	33.563	4.544	26.586	5.70 ± 0.20	1.63 ± 0.07	22.8 ± 0.9
					122	33.531	3.930	26.625	5.31 ± 0.18	1.47 ± 0.05	20.5 ± 0.7
					149	33.512	3.116	26.687	4.42 ± 0.23	1.32 ± 0.08	18.4 ± 1.1
					197	33.637	2.859	26.810	3.98 ± 0.17	1.18 ± 0.06	16.5 ± 0.8
					299	33.869	3.191	26.965	2.65 ± 0.12	0.83 ± 0.04	11.7 ± 0.6
					396	33.991	3.181	27.063	2.06 ± 0.10	0.66 ± 0.04	9.2 ± 0.5
					495	34.118	3.279	27.155	1.45 ± 0.09	0.44 ± 0.03	6.2 ± 0.4
					743	34.309	2.842	27.348	0.56 ± 0.06	0.17 ± 0.02	2.3 ± 0.3
					990	34.414	2.528	27.459	0.22 ± 0.05	0.07 ± 0.01	1.0 ± 0.2
					1235	34.462	2.271	27.519	0.19 ± 0.04	0.06 ± 0.01	0.8 ± 0.2
					1483	34.510	2.079	27.573	0.07 ± 0.05	0.02 ± 0.02	0.3 ± 0.2
					1727	34.559	1.895	27.626	0.03 ± 0.05	0.01 ± 0.01	0.1 ± 0.2
					1974	34.592	1.722	27.666	0.03 ± 0.04	0.01 ± 0.01	0.1 ± 0.2
					2708	34.642	1.390	27.730	0.03 ± 0.04	0.01 ± 0.01	0.1 ± 0.2
					2955	34.651	1.320	27.743	0.01 ± 0.04	0.01 ± 0.01	0.1 ± 0.2
SY08	off Kii	5 Aug 2008	135°31′	32°28′	6	34.081	29.931	21.061	3.13 ± 0.12	0.94 ± 0.04	13.2 ± 0.6
					50	34.309	25.889	22.547	2.84 ± 0.11	0.86 ± 0.04	12.0 ± 0.5
					101	34.796	21.571	24.133	3.74 ± 0.15	1.12 ± 0.05	15.7 ± 0.8
					151	34.828	19.726	24.703	3.09 ± 0.12	0.92 ± 0.04	12.9 ± 0.6
					201	34.810	18.907	24.900	3.42 ± 0.11	1.08 ± 0.04	15.1 ± 0.6
					251	34.800	18.379	25.025	3.84 ± 0.14	1.17 ± 0.05	16.3 ± 0.7
					498	34.435	12.659	26.023	3.88 ± 0.12	1.23 ± 0.05	17.2 ± 0.7
					750	34.255	6.187	26.940	1.59 ± 0.13	0.52 ± 0.05	7.3 ± 0.7
					999	34.388	3.752	27.324	0.85 ± 0.07	0.27 ± 0.02	3.7 ± 0.3
					1248	34.460	2.931	27.461	0.60 ± 0.06	0.20 ± 0.02	2.8 ± 0.3
					1499	34.522	2.480	27.550	0.53 ± 0.05	0.17 ± 0.02	2.4 ± 0.2
					2002	34.599	1.883	27.660	0.84 ± 0.08	0.27 ± 0.03	3.7 ± 0.4
SY09	Joban C	18 Jul 2009	142°13′	36°48′	5	34.110	21.625	23.648	4.75 ± 0.28	1.29 ± 0.08	18.1 ± 1.1
					10	34.201	20.819	23.937	4.30 ± 0.25	1.24 ± 0.07	17.3 ± 1.1
					20	34.142	16.369	25.005	4.35 ± 0.25	1.19 ± 0.07	16.6 ± 1.0
					29	34.319	13.446	25.777	4.10 ± 0.26	1.20 ± 0.08	16.7 ± 1.1
					39	34.267	12.366	25.951	4.47 ± 0.26	1.29 ± 0.08	18.1 ± 1.1
					50	34.243	11.561	26.085	3.97 ± 0.25	1.13 ± 0.07	15.8 ± 1.0
					69	34.135	10.302	26.227	5.22 ± 0.29	1.47 ± 0.08	20.5 ± 1.1
					100	33.979	8.711	26.366	4.73 ± 0.28	1.42 ± 0.09	19.8 ± 1.2
					124	34.003	8.420	26.430	4.66 ± 0.27	1.32 ± 0.08	18.5 ± 1.1
					149	33.842	7.167	26.486	5.19 ± 0.24	1.48 ± 0.07	20.7 ± 1.0
					198	33.685	5.027	26.631	5.42 ± 0.28	1.49 ± 0.08	20.9 ± 1.1
					298	33.718	3.908	26.778	4.33 ± 0.25	1.31 ± 0.08	18.3 ± 1.1
					397	33.954	4.230	26.934	2.68 ± 0.20	0.83 ± 0.06	11.5 ± 0.9
					496	34.093	4.095	27.059	1.65 ± 0.16	0.51 ± 0.05	7.1 ± 0.7
					743	34.229	3.373	27.296	0.70 ± 0.14	0.21 ± 0.04	3.0 ± 0.6
					990	34.410	2.787	27.439	0.09 ± 0.11	0.03 ± 0.04	0.4 ± 0.5
					1236	34.476	2.397	27.527	0.04 ± 0.10	0.01 ± 0.03	0.2 ± 0.4
					1730	34.556	1.992	27.625	0.18 ± 0.11	0.06 ± 0.03	0.8 ± 0.5
					1976	34.586	1.814	27.664	0.03 ± 0.10	0.01 ± 0.03	0.1 ± 0.4
					2221	34.608	1.674	27.694	0.02 ± 0.10	0.01 ± 0.03	0.1 ± 0.5
	Miyako	20 Jul 2009	145°00′	40°00′	5	33.817	17.583	24.469	5.01 ± 0.25	1.43 ± 0.07	19.9 ± 1.0
					10	33.815	17.586	24.467	4.52 ± 0.24	1.28 ± 0.07	17.9 ± 1.0
					20	33.863	15.008	25.095	5.12 ± 0.28	1.36 ± 0.08	19.0 ± 1.1
					30	34.047	13.912	25.471	5.41 ± 0.27	1.53 ± 0.08	21.4 ± 1.1
					39	34.166	14.011	25.542	4.48 ± 0.24	1.25 ± 0.07	17.5 ± 1.0
					50	34.273	13.041	25.823	4.73 ± 0.26	1.34 ± 0.08	18.8 ± 1.1
					74	34.188	11.315	26.088	5.13 ± 0.28	1.46 ± 0.08	20.4 ± 1.1
					99	33.785	8.127	26.303	5.42 ± 0.30	1.46 ± 0.08	20.5 ± 1.1

Table 1. Continued.

Cruise	Station	Date	E	N	Depth (m)	Salinity PSU	Potential Temp. °C	Density σ_θ	$^{129}I/^{127}I$ $\times 10^{-11}$	^{129}I $\times 10^{7}$ atoms L^{-1}	^{129}I nBq L^{-1}
					123	33.611	6.232	26.427	4.38 ± 0.24	1.27 ± 0.07	17.8 ± 1.0
					148	33.612	5.608	26.505	6.19 ± 0.30	1.72 ± 0.09	24.1 ± 1.2
					199	33.878	6.483	26.607	4.27 ± 0.24	1.21 ± 0.07	17.0 ± 1.0
					298	33.847	4.663	26.802	3.81 ± 0.20	1.09 ± 0.06	15.2 ± 0.8
					397	33.907	4.330	26.885	3.03 ± 0.21	0.08 ± 0.06	12.3 ± 0.9
					742	34.285	3.421	27.280	0.97 ± 0.14	0.30 ± 0.04	4.2 ± 0.6
					1482	34.518	2.177	27.580	0.15 ± 0.10	0.05 ± 0.03	0.7 ± 0.4
					1728	34.561	1.955	27.632	0.07 ± 0.10	0.02 ± 0.03	0.3 ± 0.4
After the 1FNPP accident											
KT11-06	6	27 Apr 2011	140°50′	36°00′	0				11.49 ± 0.45	2.93 ± 0.11	41.0 ± 1.6
	7	28 Apr 2011	141°40′	36°03′	0				18.88 ± 0.51	4.65 ± 0.13	65.1 ± 1.8
	8	28 Apr 2011	141°50′	36°31′	0				4.70 ± 0.29	1.19 ± 0.07	16.7 ± 1.0
	9	29 Apr 2011	142°40′	37°10′	0				22.70 ± 0.85	5.60 ± 0.21	78.3 ± 2.9
	10	29 Apr 2011	143°02′	38°09′	0				42.01 ± 0.92	10.37 ± 0.23	145.0 ± 3.2
	12	29 Apr 2011	143°40′	38°15′	0				13.14 ± 0.60	3.29 ± 0.15	46.1 ± 2.1
	14	30 Apr 2011	143°47′	38°13′	0				32.49 ± 0.81	8.15 ± 0.20	114.0 ± 2.8
	A	29 Apr 2011	142°50′	38°24′	9	33.988	11.262	25.941	50.78 ± 0.96	12.97 ± 0.25	181.4 ± 3.4
					29	33.936	10.511	26.034	70.37 ± 1.53	17.65 ± 0.38	246.9 ± 5.4
					49	34.252	10.403	26.229	30.92 ± 0.88	8.05 ± 0.23	112.6 ± 3.2
					98	33.741	6.928	26.437	12.59 ± 0.51	3.18 ± 0.13	44.4 ± 1.8
					198	33.701	5.484	26.589	5.89 ± 0.32	1.50 ± 0.08	21.0 ± 1.1
					594	34.192	4.269	27.116	1.29 ± 0.25	0.34 ± 0.07	4.8 ± 0.9
					1421	34.508	2.257	27.557	0.37 ± 0.15	0.10 ± 0.04	1.4 ± 0.6
	B	30 Apr 2011	143°28′	38°17′	9	34.273	12.759	25.657	27.73 ± 0.78	6.83 ± 0.20	95.5 ± 2.7
					50	34.274	11.513	25.894	11.64 ± 0.56	2.84 ± 0.14	39.7 ± 1.9
					99	34.212	10.396	26.094	8.82 ± 0.43	2.22 ± 0.11	31.1 ± 1.5
					198	33.987	8.294	26.435	5.66 ± 0.41	1.45 ± 0.10	20.2 ± 1.5
					497	34.070	4.073	27.039	2.53 ± 0.25	0.69 ± 0.07	9.6 ± 0.9
					989	34.375	2.932	27.392	0.91 ± 0.23	0.25 ± 0.06	3.5 ± 0.9
	C	1 May 2011	143°54′	39°23′	10	33.957	9.981	26.165	11.63 ± 0.69	2.93 ± 0.17	40.9 ± 2.4
					50	33.981	9.540	26.238	10.81 ± 0.50	2.73 ± 0.13	38.3 ± 1.8
					99	33.985	9.199	26.294	11.70 ± 0.57	3.02 ± 0.15	42.2 ± 2.1
	15	1 May 2011	143°53′	38°57′	0				12.84 ± 0.61	3.19 ± 0.15	44.7 ± 2.1
	16	1 May 2011	143°59′	39°45′	0				15.84 ± 0.54	3.91 ± 0.13	54.7 ± 1.9
BO-11-05	UW1	12 Jun 2011	141°59′	36°00′	0	34.379	21.299	23.643	4.47 ± 0.27	1.08 ± 0.06	15.2 ± 0.9
	UW2	12 Jun 2011	142°19′	36°31′	0	33.426	17.677	24.576	362.25 ± 4.87	89.76 ± 1.21	1255 ± 16.9
	UW3	12 Jun 2011	142°31′	37°01′	0	34.011	18.224	24.443	58.99 ± 1.22	14.36 ± 0.30	200.8 ± 4.2
	UW4	13 Jun 2011	142°45’	37°30’	0	34.108	18.947	24.262	36.36 ± 0.75	8.98 ± 0.18	125.6 ± 2.6
KH-11-07	FS1	2 Aug 2011	142°10′	37°20′	0				23.29 ± 0.60	5.56 ± 0.14	77.8 ± 2.0
					101	33.772	8.277	26.437	7.33 ± 0.33	2.00 ± 0.09	28.0 ± 1.3
					198	33.573	4.948	26.878	6.76 ± 0.37	1.85 ± 0.10	25.9 ± 1.4
					297	33.658	3.589	27.022	5.43 ± 0.36	1.53 ± 0.10	21.3 ± 1.4
					496	34.011	3.703	27.011	2.32 ± 0.28	0.96 ± 0.08	13.4 ± 1.1
					692	34.247	3.506	27.030	2.61 ± 0.25	0.78 ± 0.08	10.9 ± 1.1
	ES2	2 Aug 2011	142°15′	37°04′	0				11.02 ± 0.48	2.52 ± 0.11	35.2 ± 1.5
					100	33.969	9.217	26.291	9.10 ± 0.41	2.44 ± 0.11	34.1 ± 1.5
					199	34.095	7.752	26.515	4.39 ± 0.24	1.21 ± 0.07	16.9 ± 0.9
					297	33.906	5.286	26.839	4.58 ± 0.29	1.28 ± 0.08	17.9 ± 1.1
					496	34.148	4.571	26.920	2.86 ± 0.19	0.83 ± 0.06	11.6 ± 0.8
					692	34.246	3.603	27.020	2.02 ± 0.27	0.59 ± 0.08	8.3 ± 1.1
					989	34.400	2.864	27.089	1.94 ± 0.26	0.57 ± 0.08	8.0 ± 1.1
					1978	34.591	1.862	27.171	0.48 ± 0.15	0.14 ± 0.05	2.0 ± 0.6
	FS2	2 Aug 2011	142°00′	37°00′	0				11.02 ± 0.48	2.65 ± 0.12	37.0 ± 1.6
					101	34.025	9.732	26.207	14.38 ± 0.52	2.91 ± 0.10	40.6 ± 1.5
					197	33.770	6.585	26.677	7.62 ± 0.45	2.03 ± 0.12	28.4 ± 1.7
					299	33.810	5.256	26.843	7.96 ± 0.38	2.18 ± 0.10	30.4 ± 1.4
					497	33.985	3.523	27.028	4.77 ± 0.22	1.39 ± 0.06	19.4 ± 0.9
					693	34.221	3.575	27.023	3.36 ± 0.38	0.98 ± 0.11	13.6 ± 1.5
					990	34.372	3.017	27.075	1.38 ± 0.18	0.41 ± 0.05	5.7 ± 0.7
					1307	34.480	2.420	27.127	1.09 ± 0.19	0.33 ± 0.06	4.6 ± 0.8
	FS5	3 Aug 2011	141°20′	36°00′	0				99.62 ± 2.12	23.66 ± 0.50	330.9 ± 7.0
KT-11-27	ST08	29 Oct 2011	142°00′	37°00′	0				15.84 ± 0.63	4.33 ± 0.17	60.6 ± 2.4
					49	33.881	17.928	24.515	11.62 ± 0.42	3.18 ± 0.12	44.5 ± 1.6

Table 1. Continued.

Cruise	Station	Date	E	N	Depth (m)	Salinity PSU	Potential Temp. °C	Density σ_θ	$^{129}I/^{127}I$ $\times 10^{-11}$	^{129}I $\times 10^{7}$ atoms L^{-1}	^{129}I nBq L^{-1}
					100	34.135	11.199	25.951	10.83 ± 0.50	2.94 ± 0.14	41.2 ± 1.9
					201	33.858	7.023	26.618	5.34 ± 0.35	1.50 ± 0.10	20.9 ± 1.4
					300	33.773	4.542	26.923	4.09 ± 0.27	1.15 ± 0.08	16.1 ± 1.1
					500	34.031	3.944	26.986	2.08 ± 0.17	0.60 ± 0.05	8.4 ± 0.7
					1000	34.365	2.901	27.086	0.62 ± 0.15	0.19 ± 0.05	2.6 ± 0.6
	K8	29 Oct 2011	142°11′	37°20′	0				13.32 ± 0.46	3.61 ± 0.12	50.5 ± 1.7
					49	34.194	17.051	24.726	9.38 ± 0.53	2.50 ± 0.14	35.0 ± 2.0
					99	34.082	10.493	26.077	9.99 ± 0.38	2.75 ± 0.11	38.4 ± 1.5
					201	33.706	5.621	26.799	4.41 ± 0.24	1.23 ± 0.07	17.3 ± 0.9
					299	33.927	5.463	26.818	3.25 ± 0.22	0.92 ± 0.06	12.8 ± 0.9
					500	34.093	4.278	26.952	1.71 ± 0.15	0.50 ± 0.04	7.0 ± 0.6
						34.231	3.469	27.033	0.92 ± 0.13	0.27 ± 0.04	3.8 ± 0.6
					1020	34.394	2.719	27.101	0.22 ± 0.15	0.07 ± 0.04	0.9 ± 0.6

at most stations after April 2011 except for two stations: station 8 during cruise KT-11-06 (28 April 2011) and station UW1 during cruise BO-11-05 (12 June 2011). Station 8 (141°50′ E, 36°31′ N) is located near station UW1 (141°53′ E, 36°00′ N). In the coastal regions, several water masses, such as the Kuroshio, meso-scale eddies associated with the Kuroshio, and fresh water from the land, coexist in a complex fashion. The satellite images (Ibaraki Prefectural Fisheries Experimental Station, 2012) of sea surface temperature indicate that the surface of station 8 at the time of observation was dominated by meso-scale eddies associated with the Kuroshio that were coming from low contamination regions (Buesseler et al., 2012). The lack of elevated ^{129}I concentrations at these two stations is considered to be caused by the complex seawater currents in the area near Fukushima.

The distribution patterns of surface ^{131}I and ^{137}Cs simulated numerically (Kawamura et al., 2011; Tsumune et al., 2012) showed that the accident-derived radionuclides supplied directly into the ocean were initially advected southward along the coast and then flowed eastward with the Kuroshio and its extension. The numerical simulation results from 25 March to 1 May 2011 also showed that the radionuclides were minimally advected northward near Fukushima (Kawamura et al., 2011). On the other hand, atmospheric dispersion simulations indicated that ^{131}I and ^{137}Cs were transported to the northeast and south of the 1FNPP and deposited on the surface of the western North Pacific (Kawamura et al., 2011; Terada et al., 2012). Thus, the observed accident-derived ^{129}I at northern stations can be inferred to be supplied to the ocean via atmospheric input. Another possibility mechanism to explain the high concentration at northern stations is the microbial volatilization at the southern stations. Released ^{129}I to the ocean transported to southward with seawater current and then volatilized by microbial activity and then moved to northward by wind and then deposited.

Considering the ^{129}I results, seawater current, and simulation results, we conclude that ^{129}I in the southern part from the 1FNPP was predominantly transported by seawater advection and that ^{129}I in the northern part of the 1FNPP was predominantly transported via the atmosphere including microbial volatilization.

3.2 Depth profile of ^{129}I

The ^{129}I concentrations in the seawater before and after the 1FNPP accident are plotted in Fig. 3 as a function of water depth. The all-depth profiles both before and after the 1FNPP accident show that the ^{129}I concentration had its maximum in the surface-mixed layer and decreased with depth below the layer. Because there is no ^{129}I source in the Pacific Ocean, the differences in the ^{129}I concentrations before and after the accident indicate the accident-derived ^{129}I in the water column. The ^{129}I concentrations after the accident at 1000 m depth were at levels similar to those before the accident. The results indicate that, as of October 2011, the accident-derived ^{129}I spread mainly in the upper 100 m depth of this study area and that only a small amount was transported to deep layers. Inventories of ^{129}I obtained in this study in the water column between the surface and 1000 m are summarized in Table 2.

Integrated depth profiles show the inventory of ^{129}I including the background and the accident-derived ^{129}I. The ocean inventories of ^{129}I depended on the distance from major reprocessing plants as well as water mass structures in the water column (Alfimov et al., 2004; Schink et al., 1995; Suzuki et al., 2010). Therefore, background inventories in the western margin of the North Pacific Ocean must be applied to evaluate the amount of ^{129}I derived from the 1FNPP accident. Since accident-derived ^{129}I was not detected below 1000 m depth, the inventories for all profiles up to 1000 m are tabulated in Table 2 to evaluate the influence of the 1FNPP accident. Before the 1FNPP accident, the inventories in the upper 1000 m were in the $6.3–8.4 \times 10^{12}$ atoms m^{-2} range and averaged an inventory of 7.3×10^{12} atoms m^{-2}.

Table 2. Inventory of ^{129}I before and after the 1FNPP accident and its influence.

Cruise	Station	Sampling date	Inventory ($\times 10^{12}$ atoms m^{-2})	Increase after the accident ($\times 10^{12}$ atoms m^{-2})	Reference
OS08	KNOT	10 May 2008	6.3		This study
SY08	off Kii	5 Aug 2008	8.4		This study
SY09	Joban C	18 Jul 2009	7.0		This study
	Miyako	20 Jul 2009	7.7		This study
SY07	Offshore of Kushiro	1 Aug 2007	7.2		Suzuki et al. (2010)
Average before the accident			7.3		
KT-11-06	A	29 Apr 2011	16.9	9.6	This study
	B	30 Apr 2011	11.2	3.9	This study
KH-11-07	FS1	2 Aug 2011	13.9	6.6	This study
	ES2	2 Aug 2011	10.8	3.5	This study
	FS2	2 Aug 2011	15.3	8.0	This study
KT-11-27	ST08	29 Oct 2011	10.7	3.4	This study
	K8	29 Oct 2011	8.9	1.6	This study

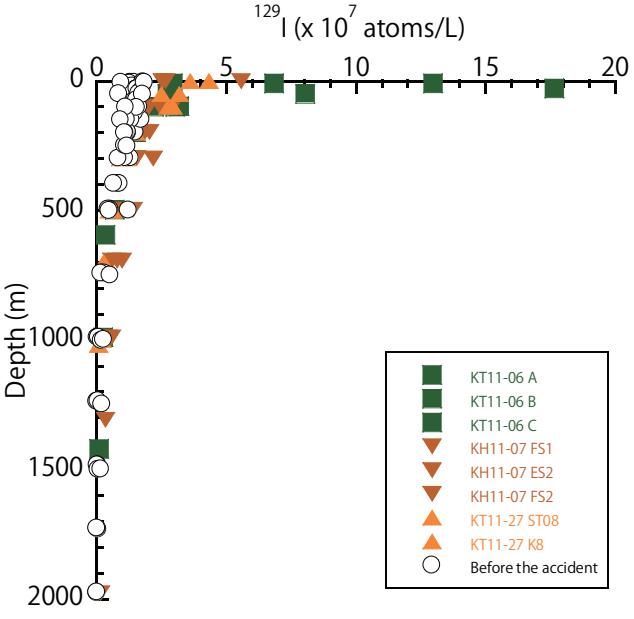

Fig. 3. Depth profiles of ^{129}I before and after the 1FNPP accident. The dark green, dark orange, and light orange symbols indicate cruises KT-11-06, KH11-07, and KT-11-27, respectively, after the 1FNPP accident. The white symbols indicate a cruise before the 1FNPP accident.

The inventories after the 1FNPP accident in the upper 1000 m were varied, (8.9–16.9) $\times 10^{12}$ atoms m^{-2}. Inventories after the 1FNPP accident increased at all stations observed in this study. As a result of the accident, the increase in ^{129}I across the study area ranged from 1.6–9.6 $\times 10^{12}$ atoms m^{-2}.

3.3 Dose estimation from ^{129}I

This study shows that the ^{129}I concentration has increased near Fukushima on account of the 1FNPP accident. Because iodine is a biophilic element, ^{129}I enters the food chain and bio-accumulates in seafood. Because of its long half-life, an internal dose may be a serious concern for many generations. To evaluate the internal dose, an effective dose was roughly estimated based on ingestion of contaminated marine food. The effective dose can be obtained from the ^{129}I concentration in seawater (Bq L^{-1}), the concentration factor for each marine food (L kg^{-1}), the total amount of marine food consumed in a year, and the effective dose coefficient (^{129}I: 1.1 $\times 10^{-7}$ Sv Bq^{-1} for adults) (ICRP, 1995).

Radioiodine is accumulated in marine foods such as fish, crustaceans, mollusks, and macroalgae at concentration factors of 9, 3, 10, and 10 000 L kg^{-1}, respectively (IAEA, 2004). The latest national survey revealed that the ordinary Japanese individual ingests 32.6 g fish, 4.4 g crustaceans, 2.9 g mollusks, and 11.0 g macroalgae in a day (MHLW, 2012). By assuming that the ^{129}I concentrations in surface seawater after the 1FNPP accident continue to have the value of 1.08–89.8 $\times 10^{7}$ atoms L^{-1} (corresponding to 15.2–1255 nBq L^{-1}) for a year, the effective dose was estimated to be 6.7–550 $\times 10^{-11}$ Sv yr^{-1}. It was quite lower than the annual dose limit of 1.0 $\times 10^{-3}$ Sv yr^{-1}. Since the ^{129}I concentration in seawater near Fukushima is expected to decrease by dilution in the ocean, the actual effective dose would be much lower than the estimated one. Therefore, we concluded that the internal dose from the ingestion of seafood is negligibly small.

4 Conclusions

This study focused on ^{129}I, a long-lived radionuclide, derived from the 1FNPP accident. ^{129}I concentrations in surface seawater before the 1FNPP accident, which can be used as the background level, showed a latitudinal gradient in horizontal distribution that could be expressed as a linear function of latitude between 32° N and 44° N. In the western margin of the North Pacific, the highest and average ^{129}I concentrations measured after the 1FNPP accident were 73 times and approximately 8 times, respectively, higher than those before the accident in this study area. The inventory of ^{129}I originating from the accident was 1.6–9.6 $\times$ 10^{12} atoms m^{-2} in this study area.

Based on the conservative estimation from measurement data of ^{129}I, the internal dose from the ingestion of seafood is negligibly small.

Because ^{129}I is one of the long-lived radionuclides, the data obtained in this study could be applied to the study of radionuclide migration from the 1FNPP accident. To investigate the impact of the 1FNPP accident more in detail, further investigation is required.

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