Geochemical Journal, Vol. 46, pp. 311 to 320, 2012

Low levels of ^{134}Cs and ^{137}Cs in surface seawaters around the Japanese Archipelago after the Fukushima Dai-ichi Nuclear Power Plant accident in 2011

MUTSUO INOUE,[1]* HISAKI KOFUJI,[2] SEIYA NAGAO,[1] MASAYOSHI YAMAMOTO,[1] YASUNORI HAMAJIMA,[1] KEN FUJIMOTO,[3] KEISUKE YOSHIDA,[1] ATSUO SUZUKI,[4] HIDEAKI TAKASHIRO,[5] KAZUICHI HAYAKAWA,[6] KAZUHITO HAMATAKA,[1] SHOTA YOSHIDA,[6] MASAYUKI KUNUGI[7] and MASAYUKI MINAKAWA[3]

[1]Low Level Radioactivity Laboratory, Institute of Nature and Environmental Technology, Kanazawa University, Nomi, Ishikawa 923-1224, Japan
[2]Japan Marine Science Foundation, Minato, Mutsu, Aomori 035-0064, Japan
[3]Fisheries Research Agency, National Research Institute of Fisheries Science, Fukuura, Kanazawa, Yokohama 236-8648, Japan
[4]Shizuoka Prefectural Environmental Radiation Monitoring Center, Omaezaki, Shizuoka 437-1612, Japan
[5]College of Bioresource Science, Nihon University, Shimoda, Shizuoka 415-0014, Japan
[6]Institute of Medical, Pharmaceutical and Health Sciences, Kanazawa University, Kakuma, Kanazawa 920-1192, Japan
[7]Environmental Safety Center, Tokyo University of Science, Noda, Chiba 278-8510, Japan

(*Received March 2, 2012; Accepted June 18, 2012*)

A total of 150 surface seawater samples were collected from around the Japanese Archipelago between July 2009 and February 2012, with sample collection peaking in July and October 2011. Low-background measurements revealed that, except for coastal and offshore areas around western Japan, ^{134}Cs and ^{137}Cs concentrations in surface waters were significantly higher than those recorded before the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, although their concentration levels were generally very low (<~0.1 to 1 mBq/L for ^{134}Cs). In June 2011, concentration peaks of 1 mBq/L for ^{134}Cs and 2.5 mBq/L for ^{137}Cs were recorded in the Tsugaru Strait and off western Hokkaido, toward the Japan Sea approximately 150–300 km north of the high-deposition area of Akita in northern Honshu. In October 2011, ^{134}Cs was detected only in the surface waters off western Hokkaido at a concentration of 0.2 mBq/L, which is markedly lower than the values observed in June 2011. These findings indicate that the observed radionuclide distributions could primarily be attributed to the atmospheric deposition of radionuclides emitted from the FDNPP and transported by ocean currents.

Keywords: radiocesium, surface seawater, water circulation, deposition, Fukushima Dai-ichi Nuclear Power Plant accident

INTRODUCTION

The massive East Japan Earthquake and tsunami that hit the Pacific Ocean side of northeastern Japan on March 11, 2011 caused extensive damage to the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). The FDNPP accident resulted in the widespread release of large amounts of ^{134}Cs (half-life: 2.06 y) and ^{137}Cs (half-life: 30.2 y) into the atmosphere, both of which were deposited over extensive ocean and land areas in eastern Japan (e.g., Hirose, 2012; Momoshima *et al*., 2012; Oura and Ebihara, 2012; Sakaguchi *et al*., 2012). Analysis of immediate and continuous records of these radionuclides in the marine environment following the FDNPP accident are particularly important for obtaining knowledge of initial input of ^{134}Cs and ^{137}Cs because their concentrations in seawater are strongly affected by physical processes such as advection and mixing. While the ^{134}Cs and ^{137}Cs concentrations of highly contaminated surface seawater samples around the FDNPP have been studied extensively (Tsumune *et al*., 2012; Honda *et al*., 2012), relatively little information is available on radionuclide concentrations in seawater for other sea areas around the Japanese Archipelago. We previously reported low levels of ^{134}Cs and ^{137}Cs concentrations in surface water samples from the Pacific Ocean side of northeastern Japan, the Japan Sea, and the southwestern Okhotsk Sea before and after the FDNPP accident; however, the scope of these studies was somewhat cursory (Inoue *et al*., 2012a, b). Nonetheless, these studies illustrated the need for more extensive temporal and spatial investigations to assess the oceanic behaviors and fate of Fukushima-derived radionuclides around the Japanese Islands.

We therefore used low-background γ-spectrometry to characterize the lateral variations of ^{134}Cs and ^{137}Cs concentrations in surface waters around the Japanese Archipelago in June and October 2011. These data were combined with ^{134}Cs deposition data (Ministry of Education,

*Corresponding author (e-mail: mutsuo@llrl.ku-unet.ocn.ne.jp)

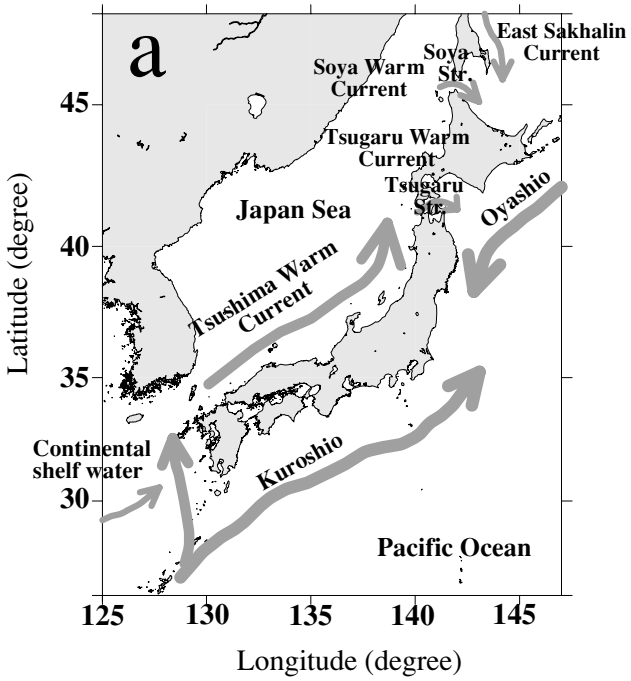
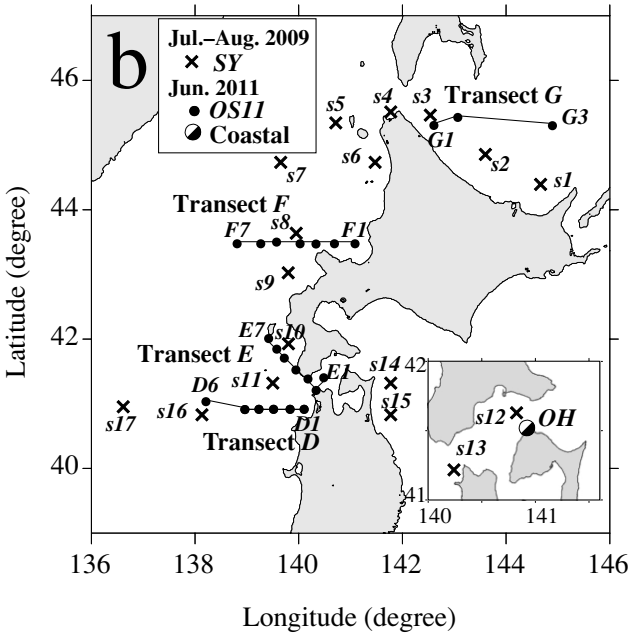


Fig. 1a). Map showing main ocean current systems around the Japanese Archipelago.



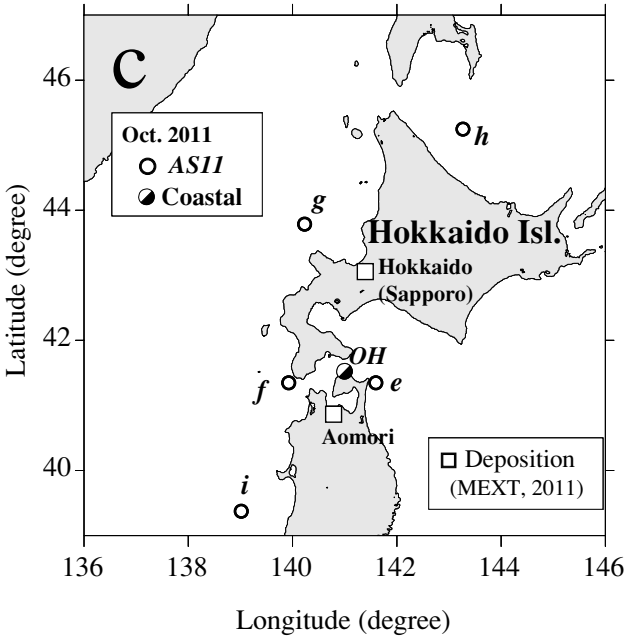


Fig. 1b). Sample locations for seawater collection near Hokkaido Island in b) July–August 2009 (SY) and June 2011 (OS11) and c) in October 2011 (AS11).

Culture, Sports, Science and Technology in Japan (MEXT), 2011) to more accurately clarify the delivery and transport patterns of radiocesium around Japan.

Samples and Experimental Procedures

The locations of the seawater sample sites used in the present study are shown in Fig. 1. Surface water samples were collected near Japan on three expeditions; the *Oshoro Maru* in June 2011, and the *Asuka-II* and *Mizuho Maru* in October 2011. Forty-four samples were collected onboard the *Oshoro Maru* (*OS11* samples), and 22 each were obtained from the *Asuka-II* (*AS11* samples) and *Mizuho Maru* (*MZ11* samples) voyages. To compare these data with pre-Fukushima accident values, we used 33 surface water samples (*SY* samples) collected near Japan during the *Soyo Maru* expeditions (July–August 2009 and 2010). We examined the fine temporal variations in ^{134}Cs and ^{137}Cs concentrations through a comparison of 29 coastal water samples collected from the Pacific Ocean side of Japan (*OM*, *IZ*, *MY*, and *TN* samples) and from the Tsugaru Strait (*OH* samples) between May 2011 and February 2012. ^{134}Cs and ^{137}Cs concentrations in the *OS11*, *OH*, *SY* (*s1–s24*), and *TN* (*1106-2*) and *MY* (*1106-2*) samples were reported in previous studies (Inoue *et al.*, 2012a, b). All water samples were unfiltered.

Detailed explanations of the γ-spectrometry procedures are described elsewhere (Inoue *et al.*, 2012b). Briefly, ^{134}Cs and ^{137}Cs were quantitatively separated by coprecipitation with ammonium phosphomolybdate (AMP)/Cs, by adding 0.52 g of CsCl and 8.0 g of AMP to an 18–21 L aliquot of water. Low-background γ-spectrometry was performed on the AMP sample using Ge-detectors at the Ogoya Underground Laboratory in

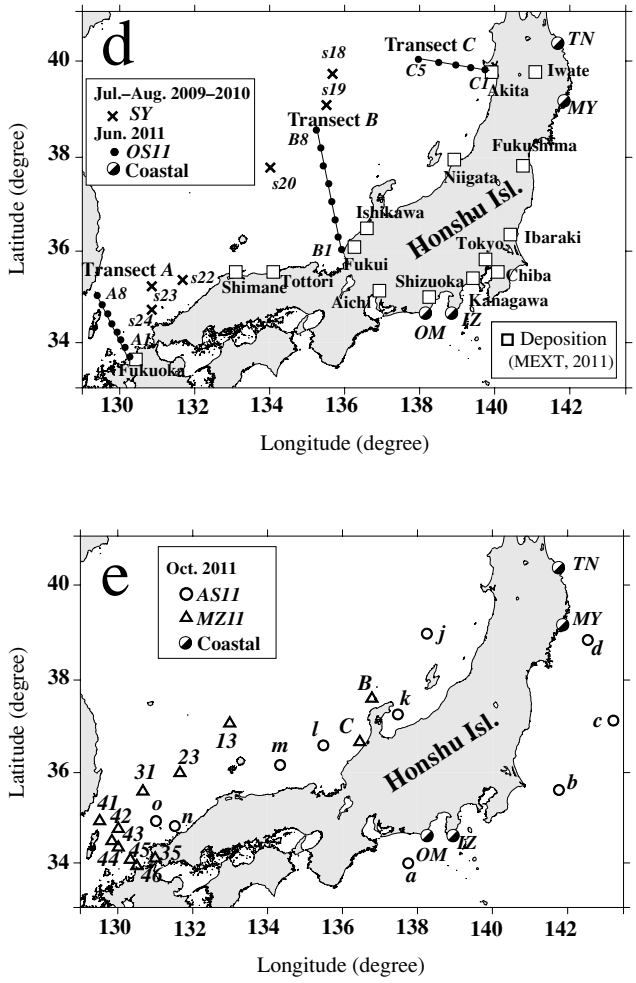


Fig. 1d). Collection points within the Japan Sea in July–August 2009–2010 and June 2011 (OS11), e) in October 2011 (AS11 and MZ11).

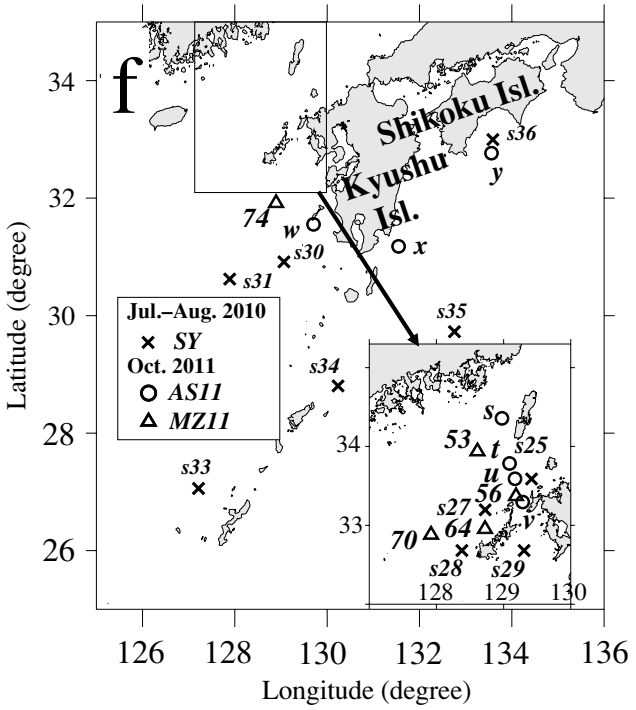


Fig. 1f). Collection points around western Japan in July–August 2010 and October 2011 (AS11 and MZ11).

Japan (Hamajima and Komura, 2004). The precision of the analyses, based on a standard deviation of counting statistics, was 5–20% for ^{134}Cs and 3–10% for ^{137}Cs. In addition, the ^{134}Cs concentration was corrected for cascade summing (21–27%).

Results and Discussion

Eastern East China Sea, Japan Sea, Southwestern Okhotsk Sea

γ-spectrometry results for the seawater samples are shown in Table 1. The lateral profiles of ^{134}Cs deposition on Honshu Island along the Japan Sea side, in addition to ^{134}Cs and ^{137}Cs concentrations of the surface waters of the Japan Sea, are presented in Fig. 2. The deposition characteristics of ^{134}Cs and ^{137}Cs over Japan revealed constant ^{134}Cs/^{137}Cs ratios (approximately 1) in radioactive aerosols emitted from the FDNPP (MEXT, 2011). In addition, the spatial distribution of the cumulative deposition of ^{134}Cs during March–May 2011 indicates a marked increase of ^{134}Cs deposition from Fukuoka (1 MBq/km^2) to Akita (174 MBq/km^2), and a decrease from Akita to Hokkaido (Sapporo) (8 MBq/km^2) (Fig. 2a). The highest deposition rates were observed in April 2011.

The mean ^{134}Cs and ^{137}Cs concentrations in water samples collected along transects *A–F* (*OS11* samples) on the Japan Sea side showed a gradual increase from transects *A–D* (<~0.1 to 1 mBq/L for ^{134}Cs and 1.5–2.5 mBq/L for ^{137}Cs) before plateauing from transects *D–F* (Inoue *et al.*, 2012a). The highest observed ^{137}Cs deposition area (500–1000 MBq/km^2) expanded further westward (~250 km) from Akita on the Japan Sea side (March–April 2011) (Yasunari *et al.*, 2011). In the Japan Basin (site *SY07*; N41°02′, E138°10′) in July 2007, the concentration of ^{7}Be, a short-lived nuclide (half-life: 53.3 d) deposited to the sea surface from the atmosphere, exhibited a steep decrease from 20 m to 50 m depths (8.5 mBq/L at 5 m depth, 9.3 mBq/L at 10 m depth, and 0.7 mBq/L at 50 m depth for dissolved fraction; *n.d.* for particle fraction (>0.5 μm)) (unpublished data of our lab). Assuming that the distribution of ^{134}Cs from the ocean surface to a depth of 50 m was vertically homogenous as a maximum depth and that the deposition value on transects *D–F* was approximately equal to that for Akita in April 2011 (130 MBq/km^2), the ^{134}Cs concentration in water shallower

Table 1. ^{134}Cs *and* ^{137}Cs *concentrations in surface seawater samples around Japan Islands*

Sampling site		Location		Depth (m)	Sampling date	Salinity	Concentration (mBq/L)	
		N	E				^{134}Cs	^{137}Cs
*SY09**	*s1*	44°33′	144°41′	5	Aug. 2, 2009	32.12	n.d.	0.93 ± 0.11
	s2	44°53′	143°22′	5	Aug. 1, 2009	32.00	n.d.	0.96 ± 0.12
	s3	45°24′	142°29′	5	Aug. 3, 2009	33.90	n.d.	1.58 ± 0.15
	s4	45°35′	141°56′	5	Aug. 1, 2009	33.43	n.d.	1.65 ± 0.11
	s5	45°17′	140°47′	5	Aug. 4, 2009	33.66	n.d.	1.47 ± 0.17
	s6	44°50′	141°21′	5	Jul. 31, 2009	33.35	n.d.	1.54 ± 0.14
	s7	44°42′	139°48′	5	Aug. 5, 2009	33.85	n.d.	1.61 ± 0.13
	s8	43°42′	139°45′	5	Aug. 5, 2009	33.80	n.d.	1.49 ± 0.10
	s9	42°58′	139°38′	5	Jul. 27, 2009	33.72	n.d.	1.50 ± 0.10
	s10	41°58′	139°46′	5	Aug. 6, 2009	33.18	n.d.	1.49 ± 0.11
	s11	41°18′	139°47′	5	Jul. 21, 2009	33.90	n.d.	1.83 ± 0.15
	s12	41°35′	140°52′	5	Jul. 20, 2009	33.71	n.d.	1.59 ± 0.13
	s13	41°20′	140°19′	5	Jul. 21, 2009	33.51	n.d.	1.69 ± 0.14
	s14	41°21′	141°49′	5	Jul. 20, 2009	33.85	n.d.	1.82 ± 0.11
	s15	40°53′	141°49′	5	Aug. 6, 2009	32.79	n.d.	1.43 ± 0.10
	s16	41°00′	138°00′	5	Jul. 21, 2009	—	n.d.	1.69 ± 0.15
	s17	40°56′	136°38′	5	Jul. 25, 2009	33.50	n.d.	1.49 ± 0.12
	s18	39°50′	135°53′	5	Jul. 23, 2009	—	n.d.	1.64 ± 0.09
	s19	39°05′	135°32′	5	Jul. 24, 2009	—	n.d.	1.51 ± 0.10
*SY10**	*s20*	37°41′	134°03′	5	Jul. 21, 2010	33.99	n.d.	1.61 ± 0.08
	s22	35°21′	131°35′	5	Jul. 21, 2010	33.09	n.d.	1.31 ± 0.08
	s23	34°35′	130°54′	5	Jul. 22, 2010	32.19	n.d.	1.46 ± 0.09
	s24	34°24′	130°44′	5	Jul. 22, 2010	32.25	n.d.	1.33 ± 0.13
	s25	33°33′	129°20′	5	Jul. 25, 2010	32.68	n.d.	1.45 ± 0.12
	s27	33°04′	128°20′	5	Jul. 25, 2010	31.77	n.d.	1.37 ± 0.09
	s28	32°19′	128°15′	5	Jul. 26, 2010	31.90	n.d.	1.52 ± 0.13
	s29	32°13′	129°39′	5	Jul. 26, 2010	33.08	n.d.	1.51 ± 0.11
	s30	31°49′	129°00′	5	Jul. 27, 2010	33.32	n.d.	1.56 ± 0.11
	s31	30°30′	128°06′	5	Jul. 28, 2010	33.25	n.d.	1.57 ± 0.11
	s33	26°59′	127°18′	5	Jul. 30, 2010	34.22	n.d.	1.43 ± 0.11
	s34	28°44′	130°17′	5	Aug. 4, 2010	34.39	n.d.	1.54 ± 0.09
	s35	29°48′	133°05′	5	Aug. 5, 2010	34.39	n.d.	1.34 ± 0.13
	s36	33°03′	133°49′	5	Aug. 7, 2010	33.32	n.d.	1.58 ± 0.09
*OS11**	*A1*	33°38′	130°13′	0	Jun. 25, 2011	31.90	n.d.	1.45 ± 0.07
	A2	33°50′	130°08′	0	Jun. 24, 2011	34.09	n.d.	1.60 ± 0.07
	A3	34°00′	130°02′	0	Jun. 24, 2011	33.44	n.d.	1.53 ± 0.07
	A4	34°10′	129°56′	0	Jun. 24, 2011	33.30	n.d.	1.56 ± 0.06
	A5	34°20′	129°49′	0	Jun. 24, 2011	33.11	n.d.	1.44 ± 0.09
	A6	34°30′	129°43′	0	Jun. 24, 2011	34.00	n.d.	1.68 ± 0.10
	A7	34°40′	129°36′	0	Jun. 24, 2011	33.25	n.d.	1.68 ± 0.12
	A8	34°50′	129°30′	0	Jun. 24, 2011	33.14	n.d.	1.58 ± 0.12
	mean of Transect A (A1–A8)						n.d.	1.56 ± 0.09

than 50 m could be estimated as 2.6 mBq/L; this value is sufficient to explain the ^{134}Cs concentrations in the surface water samples collected along transects *D–F*. Since the Japan Sea is isolated from the sea area in front of the FDNPP by the Japanese Islands, the ^{134}Cs and ^{137}Cs nuclides in the Japan Sea are likely to have been derived from atmospheric deposition of radioactive aerosols from the FDNPP.

The peaks in concentrations of ^{134}Cs and ^{137}Cs in the surface water samples collected in June 2011 shifted ap-

Table 1. (continued)

Sampling site		Location		Depth (m)	Sampling date	Salinity	Concentration (mBq/L)	
		N	E				^{134}Cs	^{137}Cs
*OS11**	*B1*	36°03′	135°48′	0	Jun. 22, 2011	33.83	n.d.	1.43 ± 0.09
	B2	36°25′	135°46′	0	Jun. 22, 2011	34.27	n.d.	1.71 ± 0.07
	B3	36°46′	135°43′	0	Jun. 22, 2011	34.37	n.d.	1.52 ± 0.13
	B4	37°06′	135°41′	0	Jun. 22, 2011	34.32	0.18 ± 0.02	1.72 ± 0.07
	B5	37°29′	135°39′	0	Jun. 22, 2011	34.07	0.27 ± 0.04	1.88 ± 0.09
	B6	37°49′	135°36′	0	Jun. 22, 2011	33.95	0.26 ± 0.03	1.83 ± 0.05
	B7	38°10′	135°34′	0	Jun. 21, 2011	34.02	0.35 ± 0.03	1.84 ± 0.07
	B8	38°28′	135°30′	0	Jun. 21, 2011	34.39	0.16 ± 0.04	1.72 ± 0.10
	mean of Transect B (B1–B8)						0.15 ± 0.14	1.70 ± 0.15
	C1	39°42′	139°54′	0	Jun. 19, 2011	30.61	0.86 ± 0.06	2.12 ± 0.11
	C2	39°45′	139°34′	0	Jun. 19, 2011	34.15	0.43 ± 0.04	2.20 ± 0.09
	C3	39°49′	139°12′	0	Jun. 19, 2011	34.25	0.41 ± 0.05	1.82 ± 0.11
	C4	39°51′	139°47′	0	Jun. 19, 2011	34.04	0.33 ± 0.05	2.04 ± 0.10
	C5	39°55′	138°22′	0	Jun. 20, 2011	33.97	0.57 ± 0.05	2.10 ± 0.09
	mean of Transect C (C1–C5)						0.52 ± 0.21	2.05 ± 0.14
	D1	41°00′	140°09′	0	Jun. 16, 2011	32.65	1.20 ± 0.06	2.41 ± 0.11
	D2	41°00′	139°54′	0	Jun. 16, 2011	32.15	1.07 ± 0.07	2.61 ± 0.12
	D3	41°00′	139°37′	0	Jun. 16, 2011	32.66	0.90 ± 0.06	2.40 ± 0.10
	D4	41°00′	139°19′	0	Jun. 16, 2011	33.49	0.68 ± 0.06	2.13 ± 0.11
	D5	41°00′	138°59′	0	Jun. 15, 2011	33.96	0.99 ± 0.05	2.59 ± 0.09
	D6	41°12′	137°41′	0	Jun. 15, 2011	33.91	0.93 ± 0.06	2.50 ± 0.11
	mean of Transect D (D1–D6)						0.96 ± 0.18	2.44 ± 0.18
	E1	41°19′	140°25′	0	Jun. 8, 2011	33.28	0.98 ± 0.07	2.78 ± 0.13
	E2	41°09′	140°11′	0	Jun. 8, 2011	32.82	1.02 ± 0.06	2.59 ± 0.11
	E3	41°22′	140°09′	0	Jun. 8, 2011	33.36	1.21 ± 0.09	2.48 ± 0.14
	E4	41°30′	139°55′	0	Jun. 8, 2011	33.44	1.09 ± 0.05	2.79 ± 0.08
	E5	41°40′	139°41′	0	Jun. 8, 2011	33.29	1.00 ± 0.08	2.63 ± 0.14
	E6	41°51′	139°29′	0	Jun. 9, 2011	33.95	0.57 ± 0.06	2.31 ± 0.13
	E7	41°59′	139°16′	0	Jun. 9, 2011	34.11	0.70 ± 0.05	2.19 ± 0.10
	mean of Transect E (E1–E7)						0.94 ± 0.22	2.54 ± 0.23
	F1	43°39′	141°14′	0	Jun. 13, 2011	33.07	0.86 ± 0.05	2.15 ± 0.09
	F2	43°38′	140°50′	0	Jun. 13, 2011	33.76	1.12 ± 0.06	2.64 ± 0.11
	F3	43°38′	140°24′	0	Jun. 13, 2011	33.19	0.99 ± 0.08	2.43 ± 0.14
	F4	43°38′	139°55′	0	Jun. 13, 2011	33.83	0.76 ± 0.05	2.17 ± 0.10
	F5	43°38′	139°29′	0	Jun. 13, 2011	33.89	1.31 ± 0.10	2.59 ± 0.16
	F6	43°38′	138°57′	0	Jun. 14, 2011	33.90	1.49 ± 0.08	2.86 ± 0.13
	F7	43°38′	138°28′	0	Jun. 14, 2011	33.90	0.82 ± 0.06	2.51 ± 0.10
	mean of Transect F (F1–F7)						1.05 ± 0.27	2.48 ± 0.25
	G1	45°19′	142°29′	0	Jun. 10, 2011	33.80	0.44 ± 0.04	2.08 ± 0.09
	G2	45°28′	142°48′	0	Jun. 10, 2011	32.30	0.73 ± 0.05	1.51 ± 0.08
	G3	45°23′	145°02′	0	Jun. 11, 2011	32.58	0.57 ± 0.04	1.58 ± 0.08
	mean of Transect G (G1–G3)						0.58 ± 0.14	1.72 ± 0.31

proximately 150–300 km northward from the high deposition area of Akita (transect *C*) to Aomori and Sapporo (transects *D–F*), although these nuclide concentrations on Honshu Island along the Japan Sea side may not directly reflect the amounts of radiocesium deposited over the adjacent offshore and coastal areas (Yasunari *et al.*, 2011; Inoue *et al.*, 2012a). In October 2011, the ^{134}Cs and ^{137}Cs concentrations in water samples collected along Honshu Island (*AS11-h* to *-o*) were similar to pre-Fukushima accident levels. On the Honshu side of the Japan Sea, the

Table 1. (continued)

Sampling site		Location		Depth (m)	Sampling date	Salinity	Concentration (mBq/L)	
		N	E				^{134}Cs	^{137}Cs
AS11	*a*	34°04′	137°55′	5	Oct. 11, 2011	34.04	n.d.	1.57 ± 0.09
	b	35°29′	141°49′	5	Oct. 13, 2011	34.47	0.24 ± 0.03	1.64 ± 0.08
	c	37°04′	143°33′	5	Oct. 13, 2011	33.88	31.17 ± 0.88	52.53 ± 0.77
	d	38°40′	142°22′	5	Oct. 14, 2011	33.68	2.52 ± 0.11	3.91 ± 0.16
	e	41°24′	141°40′	5	Oct. 15, 2011	33.64	2.75 ± 0.11	4.38 ± 0.16
	f	41°24′	140°01′	5	Oct. 15, 2011	33.45	n.d.	1.62 ± 0.09
	g	43°51′	140°11′	5	Oct. 15, 2011	33.83	0.22 ± 0.03	1.59 ± 0.06
	h	45°16′	143°08′	5	Oct. 17, 2011	31.33	0.28 ± 0.04	1.02 ± 0.07
	i	39°27′	139°01′	5	Oct. 18, 2011	—	n.d.	1.77 ± 0.08
	j	38°45′	138°12′	5	Oct. 19, 2011	—	n.d.	1.59 ± 0.07
	k	37°08′	137°32′	5	Oct. 19, 2011	—	n.d.	1.54 ± 0.09
	l	36°32′	135°45′	5	Oct. 20, 2011	33.17	n.d.	1.62 ± 0.10
	m	36°12′	134°00′	5	Oct. 20, 2011	33.37	n.d.	1.65 ± 0.08
	n	34°55′	131°32′	5	Oct. 21, 2011	33.92	n.d.	1.55 ± 0.08
	o	34°56′	131°02′	5	Oct. 21, 2011	33.91	n.d.	1.46 ± 0.08
	s	34°20′	129°01′	5	Oct. 23, 2011	34.12	n.d.	1.69 ± 0.11
	t	33°58′	129°01′	5	Oct. 23, 2011	34.12	n.d.	1.54 ± 0.11
	u	33°41′	129°07′	5	Oct. 23, 2011	34.13	n.d.	1.67 ± 0.06
	v	33°18′	129°16′	5	Oct. 23, 2011	34.03	n.d.	1.51 ± 0.07
	w	31°37′	129°34′	5	Oct. 24, 2011	33.99	n.d.	1.53 ± 0.07
	x	31°17′	131°26′	5	Oct. 24, 2011	34.13	n.d.	1.52 ± 0.08
	y	32°43′	133°53′	5	Oct. 24, 2011	34.06	n.d.	1.40 ± 0.08
MZ11	*B*	37°14′	136°37′	0	Oct. 15, 2011	—	n.d.	1.51 ± 0.10
	C	36°30′	136°20′	0	Oct. 17, 2011	—	n.d.	1.61 ± 0.11
	4	36°15′	133°50′	0	Oct. 18, 2011	—	n.d.	1.58 ± 0.06
	7	35°40′	133°50′	0	Oct. 18, 2011	—	n.d.	1.54 ± 0.08
	13	37°02′	132°50′	0	Oct. 19, 2011	—	n.d.	1.52 ± 0.06
	19	35°40′	132°50′	0	Oct. 20, 2011	—	n.d.	1.67 ± 0.12
	23	36°02′	131°35′	0	Oct. 20, 2011	—	n.d.	1.49 ± 0.13
	26	34°45′	131°35′	0	Oct. 21, 2011	—	n.d.	1.67 ± 0.07
	31	35°29′	130°35′	0	Oct. 26, 2011	—	n.d.	1.52 ± 0.07
	35	34°16′	130°35′	0	Oct. 21, 2011	—	n.d.	1.68 ± 0.11
	41	34°53′	129°21′	0	Oct. 27, 2011	—	n.d.	1.55 ± 0.05
	42	34°42′	129°40′	0	Oct. 27, 2011	—	n.d.	1.51 ± 0.09
	43	34°28′	129°34′	0	Oct. 27, 2011	—	n.d.	1.62 ± 0.07
	44	34°17′	129°46′	0	Oct. 27, 2011	—	n.d.	1.50 ± 0.09
	45	34°07′	129°58′	0	Oct. 25, 2011	—	n.d.	1.65 ± 0.08
	46	33°57′	130°11′	0	Oct. 25, 2011	—	n.d.	1.57 ± 0.12
	53	33°58′	128°51′	0	Oct. 28, 2011	—	n.d.	1.54 ± 0.06
	56	33°33′	129°24′	0	Oct. 28, 2011	—	n.d.	1.55 ± 0.08
	61	33°23′	128°08′	0	Oct. 28, 2011	—	n.d.	1.47 ± 0.13
	64	32°56′	128°42′	0	Oct. 29, 2011	—	n.d.	1.37 ± 0.10
	70	32°40′	127°34′	0	Oct. 29, 2011	—	n.d.	1.70 ± 0.06
	74	32°02′	128°45′	0	Oct. 30, 2011	—	n.d.	1.62 ± 0.09
*OH**	*1106-4*	41°32′	140°54′	0	Jun. 25, 2011	33.33	0.51 ± 0.07	1.96 ± 0.13
	1110-1			0	Oct. 8, 2011	32.74	0.12 ± 0.04	1.51 ± 0.07
*TN**	*1106-2*	40°25′	141°43′	0	Jun. 19, 2011	33.22	1.07 ± 0.13	2.84 ± 0.16
	1110-1			0	Oct. 1, 2011	32.59	5.05 ± 0.29	6.70 ± 0.19

Table 1. (continued)

Sampling site		Location		Depth (m)	Sampling date	Salinity	Concentration (mBq/L)	
		N	E				^{134}Cs	^{137}Cs
*MY-a**	*1106-2*	39°39′	141°58′	0	Jun. 19, 2011	33.18	1.51 ± 0.14	2.99 ± 0.12
MY-b	*1110-1*	39°40′	141°59′	0	Oct. 1, 2011	33.29	9.43 ± 0.33	11.48 ± 0.22
IZ	*1106-1*	34°41′	138°57′	0	Jun. 9, 2011	33.98	1.17 ± 0.10	2.89 ± 0.18
	1106-2			0	Jun. 16, 2011	—	2.01 ± 0.10	3.53 ± 0.15
	1106-3			0	Jun. 23, 2011	34.20	0.96 ± 0.09	2.59 ± 0.18
	1106-4			0	Jun. 30, 2011	34.12	1.07 ± 0.07	2.51 ± 0.13
	1107			0	Jul. 7, 2011	34.07	1.09 ± 0.06	2.63 ± 0.12
	1108			0	Aug. 24, 2011	33.69	3.23 ± 0.14	5.46 ± 0.21
	1109			0	Sep. 15, 2011	33.50	1.35 ± 0.11	2.94 ± 0.20
	1110			0	Oct. 18, 2011	34.06	0.79 ± 0.05	2.37 ± 0.10
	1111			0	Nov. 16, 2011	33.88	0.74 ± 0.05	2.12 ± 0.10
	1112			0	Dec. 14, 2011	34.46	0.19 ± 0.04	1.93 ± 0.11
	1201			0	Jan. 13, 2012	34.61	n.d.	1.63 ± 0.10
	1202			0	Feb. 14, 2012	34.57	n.d.	1.63 ± 0.10
	1203			0	Mar. 14, 2012	34.54	n.d.	1.45 ± 0.08
OM	*1105*	34°38′	138°07′	0	May 25, 2011	—	2.50 ± 0.24	4.58 ± 0.35
	1106-1			0	Jun. 4, 2011	31.88	2.18 ± 0.10	3.62 ± 0.15
	1106-2			0	Jun. 18, 2011	33.72	0.70 ± 0.05	2.13 ± 0.10
	1107			0	Jul. 9, 2011	32.81	0.84 ± 0.06	2.36 ± 0.11
	1108			0	Aug. 12, 2011	33.42	0.63 ± 0.06	2.12 ± 0.11
	1109			0	Sep. 18, 2011	32.69	0.54 ± 0.08	2.16 ± 0.16
	1110			0	Oct. 29, 2011	33.93	0.41 ± 0.04	1.90 ± 0.10
	1111			0	Nov. 19, 2011	33.82	0.13 ± 0.03	1.78 ± 0.08
	1112			0	Dec. 24, 2011	34.48	n.d.	1.58 ± 0.10
	1201			0	Jan. 28, 2012	32.45	n.d.	1.44 ± 0.08

The analytical error is based on 1 sigma of counting statistics.
"n.d." ("not detected") denotes "no peak" or "concentration <3σ of error" (<~0.1 mBq/L under present analytical conditions).
Concentrations are decay-corrected based on sampling date.
**^{134}Cs and ^{137}Cs concentrations of OS11, SY (s1–s24), TN (1106-2), MY-a and OH samples are from Inoue et al. (2012a, b).*

northeastward flowing Tsushima Warm Current (TWC) exhibits major features of the flow patterns of surface currents after passing through the Tsushima Strait, particularly in spring–summer (Hase *et al.*, 1999). The shift in the peak area of ^{134}Cs and ^{137}Cs concentrations in the surface waters appears to be related to the advective transport of contaminated water and exchange with non-contaminated TWC water flowing through the Tsushima Strait. Interestingly, a small amount of ^{134}Cs (0.2 mBq/L) was detected in the *AS11-g* surface water sample collected near western of Hokkaido (Fig. 2b).

The surface waters of the southwestern Okhotsk Sea generally exhibit lower ^{137}Cs concentrations than those of the Japan Sea, which reflects considerable mixing between the ^{137}Cs-poor East Sakhalin Current water, that flows along the east coast of Sakhalin Island and the ^{137}Cs-rich Soya Warm Current water from the Japan Sea (Takizawa, 1982; Inoue *et al.*, 2012c). The mean ^{137}Cs concentration in the water samples along line *G* was markedly higher than that observed in *SY09* water samples collected in August 2009; ^{134}Cs was also detected (~0.6 mBq/L) (Figs. 2b and c). While the ^{134}Cs and ^{137}Cs concentrations in the *AS11-h* sample collected from the Okhotsk Sea side of Japan in October 2011 were higher than those from the Japan Sea side (*AS11-g*), those of the *AS11-h* sample showed lower concentrations than those obtained in July 2011 (*OS11-G* samples). The decrease in ^{134}Cs and ^{137}Cs concentrations in the southwestern Okhotsk Sea is attributed to a lower contribution of the Soya Warm Current water and a gradual decrease of the ^{134}Cs and ^{137}Cs concentrations in the current water.

Conversely, 0.1–1.6 MBq/km^2 of atmospheric ^{134}Cs was deposited at the East China Sea (ECS) side of Kyushu Island during March–May 2011 (MEXT, 2011). Such a negligible amount suggests that only small amounts of ^{134}Cs and ^{137}Cs have been deposited onto the surface of the ECS, compared with those deposited in eastern Japan. The surface waters of the ECS pass through the Tsushima Strait and flow into the Japan Sea. ^{134}Cs and ^{137}Cs concentrations in the surface waters of the Tsushima Strait (samples *OS11-A* and *MZ11-41–46*) were comparable to pre-Fukushima accident levels in 2006 (sites *TE* and *TW*; Inoue *et al.*, 2007), suggesting a negligible contribution of Fukushima-derived ^{134}Cs and ^{137}Cs near the

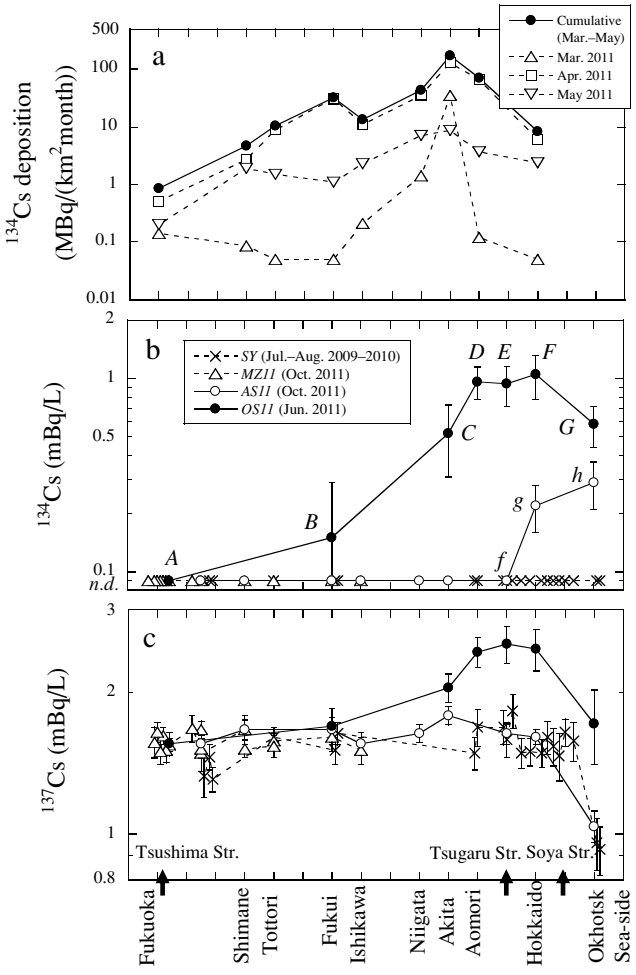


Fig. 2. Lateral profiles of a) cumulative ^{134}Cs deposition along the Japan Sea side of Honshu and Hokkaido islands (MEXT, 2011) and b) ^{134}Cs and c) ^{137}Cs concentrations in surface waters of the Japan Sea and the southwestern Okhotsk Sea. ^{134}Cs and ^{137}Cs concentrations in OS11 and SY water samples were previously reported (Inoue et al., 2012a, c). ^{134}Cs and ^{137}Cs concentrations in OS11 samples are mean values along transects A–F.

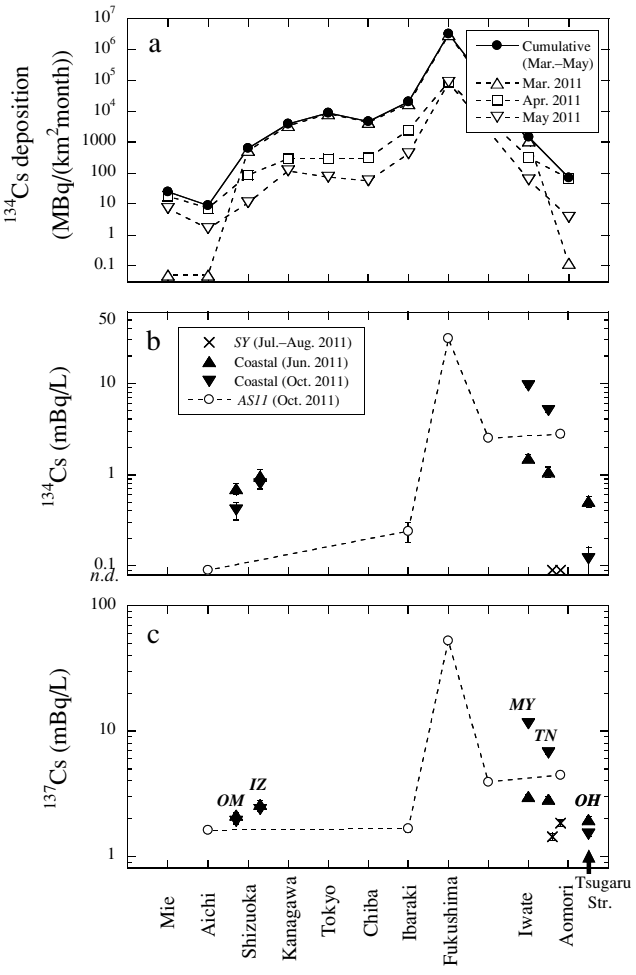


Fig. 3. Lateral profiles of a) cumulative ^{134}Cs deposition along the Pacific Ocean side of eastern and central Honshu Island (MEXT, 2011) and concentrations of b) ^{134}Cs and c) ^{137}Cs in surface waters of the Pacific Ocean.

ECS side of the Tsushima Strait in June and October 2011. ^{134}Cs and ^{137}Cs concentrations in the surface waters of the eastern ECS measured in October 2011 (*AS11-t–w* and *MZ11-53–74*) were also comparable to pre-Fukushima accident levels (*SY10s25–s33*).

Pacific Ocean side

The lateral profiles of ^{134}Cs deposited along the Pacific side of Honshu (MEXT, 2011) and the ^{134}Cs and ^{137}Cs concentrations in the surface waters of the Pacific Ocean are presented in Fig. 3. The cumulative deposition of ^{134}Cs along the Pacific side of Honshu showed a markedly wide variation below the detection limit with a total of 3.3×10^6 MBq/km^2 for March–May 2011; a significant peak occurred at Fukushima (Fig. 3a). ^{137}Cs concentrations in coastal waters near Fukushima were very high but decreased from 100 to 10 mBq/L through mixing as they were transported eastward and further offshore (Tsumune *et al.*, 2012; Honda *et al.*, 2012).

Temporal variations in ^{134}Cs and ^{137}Cs concentrations in the *OM* and *IZ* water samples are presented in Fig. 4. ^{137}Cs concentrations in the *OM* samples ranged from 1.5 to 4.6 mBq/L, which was 1–3 times higher than 1.3–1.5 mBq/L of the pre-Fukushima accident levels at the same sampling site when salinity >32 (Inoue *et al.*, 2007). In December 2011, however, the ^{134}Cs concentration at this site was below the detection limit at <~0.1 mBq/L under present analytical conditions. Kuroshio water moving northeastward along Honshu Island is dominant at site *OM*. Thus, the surface waters at this site may have gradu-

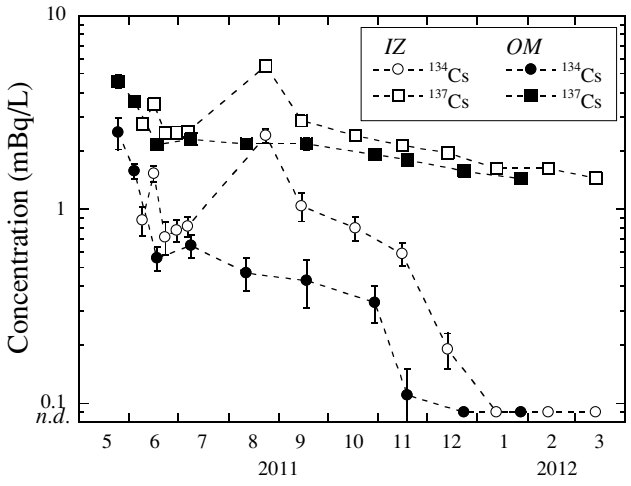


Fig. 4. Temporal variations in ^{137}Cs and ^{134}Cs concentrations in OM and IZ water samples collected along the coastline of the Pacific Ocean side of central Honshu.

ally mixed with Kuroshio water that contaminated little or no radiocesium after the deposition of radioactive aerosols on the ocean surface. Conversely, ^{134}Cs and ^{137}Cs concentrations in the *IZ* water samples collected in August 2011 exhibited high values, which is likely attributed to the local current or effluent of inland water.

^{134}Cs and ^{137}Cs concentrations in surface waters collected at sites *TN* and *MY* 300–400 km north of the FDNPP and at sites *IZ* and *OM* approximately 300 km southwest of the FDNPP were similar to those recorded in the Tsugaru Strait (site *OH*) in June 2011. Although a negligible decrease in ^{134}Cs and ^{137}Cs concentrations in surface waters were observed at sites *IZ*, *OM* (Fig. 4), and *OH* in October 2011, these concentrations in *TN* and *MY* waters obtained in October were slightly and several times higher than the June 2011 values (Figs. 3b and c), respectively. This observation implies that radiocesium-contaminated water flowing past the FDNPP mixed with water from the *TN* and *MY* sites.

SUMMARY

A total of 150 surface seawater samples were collected near the Japanese Archipelago in June and October 2011 to determined ^{134}Cs and ^{137}Cs concentrations in surface waters. In June 2011 along the Japan Sea side, peak surface concentrations of ^{134}Cs (~1 mBq/L) and ^{137}Cs (~2.5 mBq/L) shifted ~150–300 northward from the high deposition area toward the Tsugaru Strait and the western side of Hokkaido. In October 2011, ^{134}Cs was detected only in the surface waters on the western side of Hokkaido. In addition, the concentrations of ^{134}Cs were less than those observed in June 2011. The variations in spatial distributions of the ^{134}Cs and ^{137}Cs concentrations in surface waters can be attributed to lateral transport by the TWC water following the deposition of radioactive aerosols. At sites ~300 km southwest of the FDNPP on the Pacific Ocean side of the Japanese Archipelago and the Tsugaru Strait, low concentrations of ^{134}Cs (~0.1–0.2 mBq/L) were detected in surface waters with concentrations reaching pre-Fukushima accident levels by the end of 2011.

Acknowledgments—We thank the researchers, captain, and crew of *Asuka-II* and R/Vs *Soyo Maru* and *Mizuho Maru* for their assistance during sampling. In addition, we thank two anonymous reviewers for constructive comments on this manuscript. This study was supported partly by a Grant-in-Aid for Scientific Research No. 21510011 (M. Inoue) from the Ministry of Education, Culture, Sports, Science and Technology.

REFERENCES

Hamajima, Y. and Komura, K. (2004) Background components of Ge detectors in Ogoya underground laboratory. *Appl. Radiat. Isot.* **61**, 179–183.

Hase, H., Yoon, J.-H. and Koterayama, W. (1999) The current structure of the Tsushima warm current along the Japanese coast. *J. Oceanogr.* **55**, 217–235.

Hirose, K. (2012) 2011 Fukushima Daiichi nuclear power plant accident: summary of regional radioactive deposition monitoring results. *J. Environ. Radioact.* **111**, 13–17.

Honda, M. C., Aono, T., Aoyama, M., Hamajima, Y., Kawakami, H., Kitamura, M., Masumoto, Y., Miyazawa, Y., Takigawa, M. and Saino, T. (2012) Dispersion of artificial caesium-134 and -137 in the western North Pacific one month after the Fukushima accident. *Geochem. J.* **46**, e1–e9.

Inoue, M., Tanaka, K., Kofuji, H., Nakano, Y. and Komura, K. (2007) Seasonal variation in the $^{228}Ra/^{226}Ra$ ratio of coastal water within the Sea of Japan: Implications for the origin and circulation patterns of the Tsushima Coastal Branch Current. *Mar. Chem.* **107**, 559–568.

Inoue, M., Kofuji, H., Nagao, S., Yamamoto, M., Hamajima, Y., Yoshida, K., Fujimoto, K., Takada, T. and Isoda, Y. (2012a) Lateral variation of ^{134}Cs and ^{137}Cs concentrations in surface seawater in and around the Japan Sea after the Fukushima Dai-ichi Nuclear Power Plant accident. *J. Environ. Radioact.* **109**, 45–51.

Inoue, M., Kofuji, H., Hamajima, Y., Nagao, S., Yoshida, K. and Yamamoto, M. (2012b) ^{134}Cs and ^{137}Cs activities in coastal seawater along Northern Sanriku and Tsugaru Strait, northeastern Japan, after Fukushima Nuclear Power Plant accident. *J. Environ. Radioact.* **111**, 116–119.

Inoue, M., Yoshida, K., Minakawa, M., Kofuji, H., Nagao, S., Hamajima, Y. and Yamamoto, M. (2012c) Spatial variations of ^{226}Ra, ^{228}Ra, ^{137}Cs, and ^{228}Th activities in the southwestern Okhotsk Sea. *J. Environ. Radioact.* **104**, 75–80.

Ministry of Education, Culture, Sports, Science and Technology in Japan (MEXT) (2011) Monitoring information of environmental radioactivity level; Reading of radioactivity level in fallout by prefecture. Available at http://radioactivity.mext.go.jp/old/en/monitoring_by_prefecture_

fallout/
Momoshima, N., Sugihara, S., Ichikawa, R. and Yokoyama, H. (2012) Atmospheric radionuclides transported to Fukuoka, Japan remote from the Fukushima Dai-ichi nuclear power complex following the nuclear accident. *J. Environ. Radioact.* **111**, 28–32.
Oura, Y. and Ebihara, M. (2012) Radioactivity concentrations of ^{131}I, ^{134}Cs and ^{137}Cs in river water in the Greater Tokyo Metropolitan area after the Fukushima Daiichi Nuclear Power Plant Accident. *Geochem. J.* **46**, this issue, 303–309.
Sakaguchi, A., Kadokura, A., Steier, P., Tanaka, K., Takahashi, Y., Chiga, H., Matsushima, A., Nakashima, S. and Onda, Y. (2012) Isotopic determination of U, Pu and Cs in environmental waters following the Fukushima Daiichi Nuclear Power Plant accident. *Geochem. J.* **46**, this issue, 355–360.
Takizawa, T. (1982) Characteristics of the Soya Warm Current in the Okhotsk Sea. *J. Oceanogr. Soc. Japan* **38**, 281–292.
Tsumune, D., Tsubono, T., Aoyama, M. and Hirose, K. (2012) Distribution of oceanic ^{137}Cs from the Fukushima Dai-ichi Nuclear Power Plant simulated numerically by a regional ocean model. *J. Environ. Radioact.* **111**, 100–108.
Yasunari, T. J., Stohl, A., Hayano, R. S., Burkhart, J. F., Eckhardt, S. and Yasunari, T. (2011) Cesium-137 deposition and contamination of Japanese soils due to the Fukushima nuclear accident. *PNAS*, doi:10.1073/pnas.1112058108.