



Surface pathway of radioactive plume of TEPCO Fukushima NPP1 released ^{134}Cs and ^{137}Cs

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Abstract. ^{134}Cs and ^{137}Cs were released to the North Pacific Ocean by two major likely pathways, direct discharge from the Fukushima NPP1 accident site and atmospheric deposition off Honshu Islands of Japan, east and northeast of the site. High density observations of ^{134}Cs and ^{137}Cs in the surface water were carried out by 17 cruises of cargo ships and several research vessel cruises from March 2011 till March 2012. The main body of radioactive surface plume of which activity exceeded 10 Bq m^{-3} travelled along 40°N and reached the International Date Line on March 2012, one year after the accident. A distinct feature of the radioactive plume was that it stayed confined along 40°N when the plume reached the International Date Line. A zonal speed of the radioactive plume was estimated to be about 8 cm s^{-1} which was consistent with zonal speeds derived by Argo floats at the region.

Plant (hereafter FNPP1). The station blackout developed into a disaster that left three of the six FNPP1 reactors heavily damaged and caused radionuclides to be discharged into the air and ocean (Chino et al., 2011; Morino et al., 2011; Stohl et al., 2012; Tsumune et al., 2012; Kawamura et al., 2011; Estournel et al., 2012).

^{134}Cs and ^{137}Cs were released into the North Pacific Ocean by two major likely pathways, direct discharge from the FNPP1 accident site and atmospheric deposition off the Honshu Islands of Japan, east and northeast of the site. ^{134}Cs and ^{137}Cs activities in the surface water in the North Pacific Ocean were already reported (Aoyama et al., 2012a; Honda et al., 2012) and those ranged from a few to 1000 Bq m^{-3} in April–May 2011. Distributions of ^{134}Cs and ^{137}Cs activities in the surface water off Honshu and coastal stations around Japan during the period from April 2011 to November 2011 (Inoue et al., 2012a, b; Aoyama et al., 2012a; Buesseler et al., 2011, 2012) were also reported and discussed.

During the first month of the release period, ^{134}Cs and ^{137}Cs activities ratios were very close to one (0.99 ± 0.03 for FNPP1 north and south discharge channels) and extremely uniform (Buesseler et al., 2011). The presence of ^{134}Cs is a unique isotopic signature for tracking these waters and calculating mixing ratios. In the oceans, the behaviour of caesium is thought to be conservative, i.e. it is soluble ($< 1\%$ attached to marine particles) and is carried primarily with ocean waters, as such has been used as a tracer of water mass mixing and transport (Buesseler et al., 2011).

Results of observations of ^{134}Cs and ^{137}Cs activities in surface water at Hasaki, a coastal station 180 km south of

1 Introduction

On 11 March 2011, an extraordinary earthquake of magnitude 9.0 centred approximately 130 km off the Pacific coast of Japan's main island, at 38.3°N , 142.4°E , was followed by a huge tsunami with waves reaching up to 40 m height in Iwate region and about 10 m in the Fukushima region (The 2011 Tohoku Earthquake Tsunami Joint Survey Group, 2011; Mori et al., 2011). These missing and extensive damage. One of the consequences was a station blackout (total loss of AC electric power) at the Tokyo Electric Power Company (hereafter TEPCO), Fukushima Dai-ichi Nuclear Power

the FNPP1 accident site, April 2011 to December 2011, was presented and the maximum in radiocaesium activity around 2000 Bq m^{-3} at Hasaki was observed in June 2011, representing a delay of two months from the corresponding maximum in April 2011 at FNPP1. Directly discharged ^{134}Cs and ^{137}Cs were transported dominantly southward along the coastline of north-eastern Honshu. The reasons for the two-month delay at Hasaki are not yet clear; however, clockwise current associated with a warm water eddy, whose center is located at 36.5° N , 141.4° E off Iwaki between Onahama and Hasaki, in mid-May 2011 might have prevented southward transport of ^{134}Cs and ^{137}Cs released from FNPP1 to Hasaki until the end of May 2011 (Aoyama et al., 2012b).

Fukushima-derived ^{134}Cs and ^{137}Cs were detected throughout waters 30–600 km offshore, with the highest activities associated with near-shore eddies and the Kuroshio Current acting as a southern boundary for transport in June 2011. A total inventory of 1.9–2.1 PBq ^{137}Cs in an ocean area of $150\,000\text{ km}^2$ has been calculated (Buesseler et al., 2012).

However, there is no report of ^{134}Cs and ^{137}Cs activities in the surface water in the North Pacific Ocean after June 2011. High density sampling of surface seawater to measure ^{134}Cs and ^{137}Cs activities were carried out by 17 cargo ships as Voluntary Observing Ship (here after VOS) cruises and several research vessel cruises since March 2011 till March 2012 in the North Pacific Ocean.

It is also important to consider the pre-existing ^{137}Cs before the FNPP1 accident. ^{137}Cs was already present as it originated from the nuclear weapon tests conducted in the late 1950s and in the early 1960s (Aoyama et al., 2011; Aoyama and Hirose, 2008). In the western North Pacific Ocean, ^{137}Cs activity in surface water was $10\text{--}100\text{ Bq m}^{-3}$ in the late 1950s and in the early 1960s, then it decreased gradually and the ^{137}Cs activity in surface water decreased to around a few Bq m^{-3} (Aoyama et al., 2011, 2006). In the 1986 Chernobyl accident, however, a contribution of atmospheric deposition of Chernobyl derived ^{137}Cs in the North Pacific Ocean was around 3 % of total deposition derived from global fallout before 1986 (Aoyama et al., 1986); therefore, the effect of Chernobyl accident in the North Pacific Ocean is negligible. Before the FNPP1 accident, distribution and inventory of ^{137}Cs which originated from atmospheric weapons tests had been studied in the Pacific Ocean since the late 1950s and the ^{137}Cs inventory in the North Pacific Ocean was $290 \pm 30\text{ PBq}$ in January 1970 based on $10^\circ \times 10^\circ$ mesh data of the ^{137}Cs deposition (Aoyama et al., 2006). In 2003, the ^{137}Cs inventory in the North Pacific Ocean was 86 PBq by the model study (Tsumune et al., 2011) and 85 PBq by the observation (Aoyama et al., 2012a), then it decreased to 69 PBq in 2011 due to decay (Aoyama et al., 2012a). In the 2000s just before the FNPP1 accident, the ^{137}Cs activity in surface water was a few Bq m^{-3} and showed less change compared with the decreasing trend of ^{137}Cs activity we observed before 2000. A horizontal distribution of ^{137}Cs in the 2000s in the surface water showed a very homoge-

neous distribution, but relatively high ^{137}Cs activity regions in surface water were observed in the western part of the subtropical gyre in both the North Pacific Ocean and the South Pacific Ocean where ^{137}Cs activity exceeded 2 Bq m^{-3} and 1.5 Bq m^{-3} , respectively (Aoyama et al., 2012a).

In this paper we present the results of our measurements of ^{134}Cs and ^{137}Cs activities in the surface water in sea area both close to the site and the North Pacific Ocean based on the monitoring data and on our observation, respectively. We also discuss the behaviours of the radioactive plume in the North Pacific Ocean through March 2012.

2 Sampling and measurements

We collected 2 L surface seawater samples at more than 300 stations as shown in Fig. S1. The samples were treated by an improved ammonium phosphomolybdate, AMP, procedure developed by one of the authors (Hirose et al., 2005; Aoyama and Hirose, 2008). The improvement of the AMP procedure realised that the weight yield of an AMP/Cs compound basically exceeds 99 % for 2 L samples as well as radiochemical yield of radiocaesium; furthermore, the activities of AMP/Cs compounds were measured at the Ogoya Underground Facility of the Low Level Radioactivity Laboratory of Kanazawa University using high-efficiency, well-type ultra low background Ge-detectors (Hamajima and Komura, 2004). One example of the best performance at this underground facility was reported to have a detection limit of ^{137}Cs is 0.18 mBq for a counting time of 10 000 min (Hirose et al., 2005). Therefore, this development permits us to use the residue of nuclear weapon tests as useful tracers in oceanography (Aoyama et al., 2011; Povinec et al., 2011; Sanchez-Cabeza et al., 2011) and also to measure released ^{134}Cs and ^{137}Cs from the FNPP1 in 2 L samples of which activity was less than 1 Bq m^{-3} .

Because reagents can add trace levels of radioactivity, skewing small volume measurements, it is important to know the specific activity of analytes such as ^{137}Cs in the reagents. The ^{137}Cs activity in CsCl was measured to be 0.03 mBq g^{-1} by using extremely low background γ -spectrometry and we neglect this amount of ^{137}Cs because we use only 0.26 g as carrier. The ^{137}Cs activity in AMP we used was 0.024 mBq g^{-1} and we subtract corresponding amount of ^{137}Cs in the AMP used to extract radiocaesium from the samples because we use 4–6 g for extraction. There is no serious contamination of ^{137}Cs from other reagents. For ^{134}Cs contaminations, we did not observe any ^{134}Cs contaminations from the reagents.

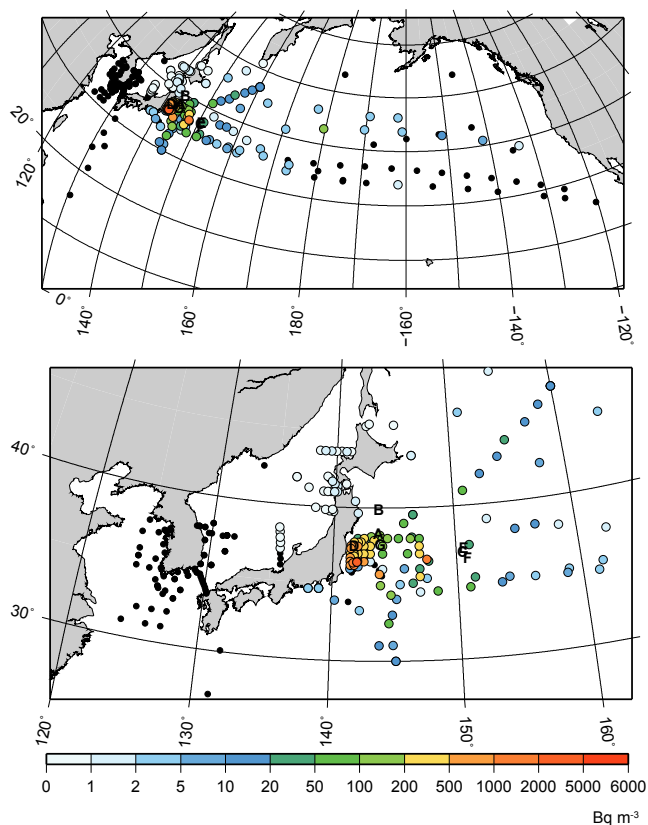


Fig. 1. ^{134}Cs activity in the surface water during the period from April 2011 to June 2011 for the North Pacific Ocean (upper) and close to Japan (lower). Positions of Argo floats on 15 May 2011 are marked “A–G”.

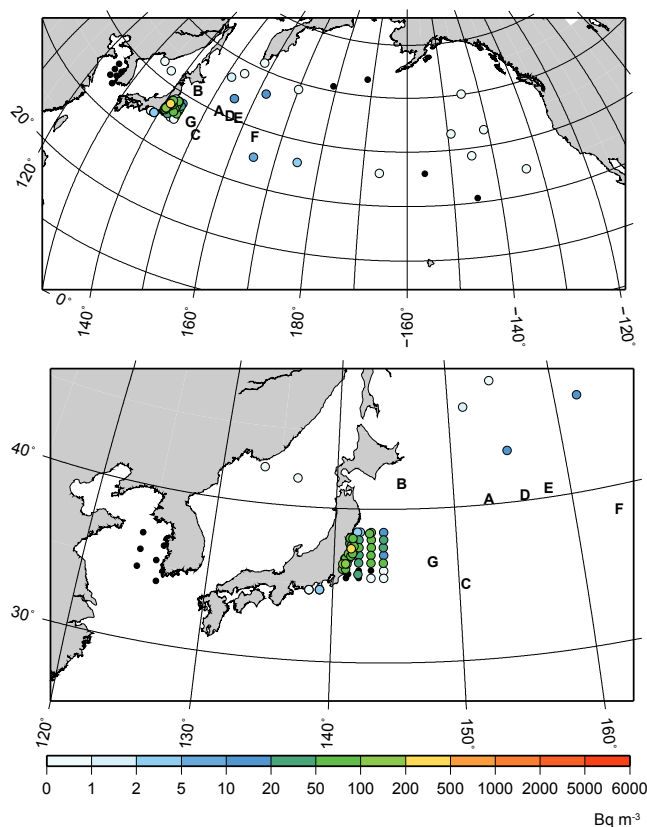


Fig. 2. ^{134}Cs activity in the surface water during the period from July 2011 to September 2011 for the North Pacific Ocean (upper) and close to Japan (lower). Positions of Argo floats on 15 August 2011 are marked “A–G”.

3 Results

3.1 Trend of ^{134}Cs and ^{137}Cs close to the accident site

In addition to our own data, we compiled monitoring data from the Ministry of Education, Culture, Sports, Science & Technology (hereafter MEXT) and TEPCO to discuss the trend of source term at the accident site. The measured ^{137}Cs concentration in a seawater sample near the FNPP1 site reached 68 MBq m^{-3} on 7 April (Buesseler et al., 2011). An analysis of ^{137}Cs concentrations and $^{131}\text{I}/^{137}\text{Cs}$ activity ratios suggest that the major direct release of ^{137}Cs from the FNPP1 reactors occurred for 12 days, from 26 March to 6 April 2011 (Tsumune et al., 2012) then decreased but still continued until July 2011 (Buesseler et al., 2011) and thereafter. During the period from August 2011 to July 2012, the activities of ^{134}Cs and ^{137}Cs near the FNPP1 site were kept around $1000\text{--}10\,000 \text{ Bq m}^{-3}$, which means that direct discharge becomes very small but still continues until July 2012 as shown in Fig. S2.

3.2 In the North Pacific Ocean

After the FNPP1 accident, both ^{134}Cs and ^{137}Cs are observed in a wide area in the North Pacific Ocean as shown in Table 1 and Figs. 1–4. The differences between ^{134}Cs and ^{137}Cs activities observed after the FNPP1 accident were $1\text{--}2 \text{ Bq m}^{-3}$ which are consistent with pre-existing ^{137}Cs originated from the nuclear weapons tests as described in Sect. 1. When we take into account the pre-existing ^{137}Cs , it is also clear that ^{134}Cs and ^{137}Cs activities ratios were close to 1 which is also consistent with observed ^{134}Cs and ^{137}Cs activities ratio of 0.99 ± 0.03 very close to the source region of the FNPP1 (Buesseler et al., 2011). This is clear evidence that observed ^{134}Cs and excess ^{137}Cs originated from the FNPP1 accident as shown in Table 1. The horizontal distribution of FNPP1-origin ^{134}Cs in the western North Pacific Ocean, except for just in front of the FNPP1 site, showed that the high concentration area located close to the FNPP1 accident site might have received both atmospheric deposition – showing good consistency with previous atmospheric transport model study (Honda et al., 2012) – and direct discharge (Tsumune et al., 2012) from the FNPP1 site. We see

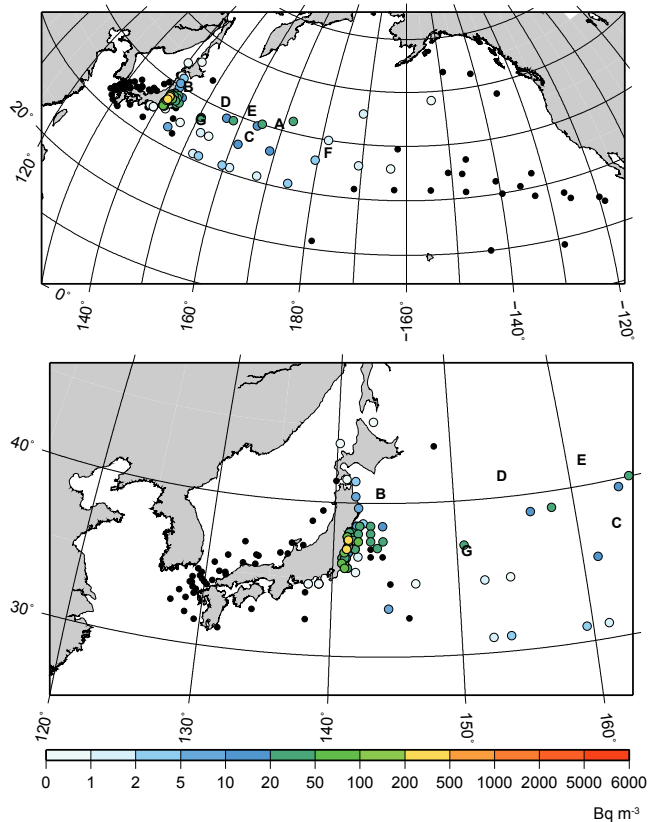


Fig. 3. ^{134}Cs activity in the surface water during the period from October 2011 to December 2011 for the North Pacific Ocean (upper) and close to Japan (lower). Positions of Argo floats on 15 November 2011 are marked “A–G”.

another high concentration area near the International Date Line in April–June 2011 as shown in Fig. 1 (upper panel). This high concentration region may be more likely explained by atmospheric deposition because of the transport distance compared to surface current. At the sea area east of the International Date Line north of 40°N in the Pacific Ocean in April 2011, ^{134}Cs activity in the surface water was less than 12 Bq m^{-3} .

In July–September 2011, a relatively high concentration area for which ^{134}Cs activity exceed 10 Bq m^{-3} moved eastward and arrived at 165°E as shown in Fig. 2. In October–December 2011, relatively high concentration area for which ^{134}Cs activity exceed 10 Bq m^{-3} moved more east and arrived at 172°E along 40°N as shown in Fig. 3. In January–March 2012, it arrived at the International Date Line as shown in Fig. 4.

4 Discussions

The atmospheric deposition occurred mainly in March 2011 (Chino et al., 2011); therefore, ^{134}Cs and ^{137}Cs activity in surface water derived by atmospheric deposition, except for

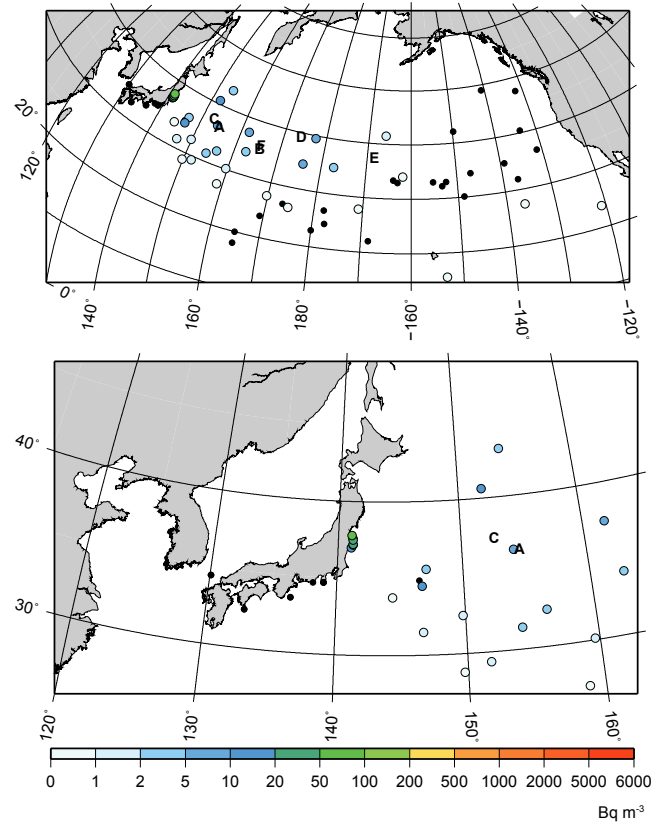


Fig. 4. ^{134}Cs activity in the surface water during the period from January 2012 to March 2012 for the North Pacific Ocean (upper) and close to Japan (lower). Positions of Argo floats on 15 February 2012 are marked “A–G”.

close to the area of the FNPP1 site, should rapidly decrease by dispersion, while eastward movement of the radioactive plume with relatively higher activity exceeding 10 Bq m^{-3} was observed as shown in Figs. 1–4. The radioactive plume were formed by the atmospheric deposition close to the FNPP1 site and direct discharge. It is interesting to estimate a zonal speed of radioactive plume based on our observations. A distinct feature of the radioactive plume was that it stayed confined along 40°N when the plume reached the International Date Line, as stated in Sect. 3.2. The radioactive plume travelled 1800 km (from 160°E to 178°E) for 270 days (9 months) (Fig. 5); therefore, an average zonal speed (u) of the surface radioactive plume was calculated to be about 8 cm s^{-1} which was consistent with the speed of the reported surface current of $4\text{--}16\text{ cm s}^{-1}$ in the region (Maximenko et al., 2009).

Eleven Argo floats were deployed off Fukushima on 31 March–13 April at $37.001\text{--}37.709^\circ\text{N}$, $141.250\text{--}141.399^\circ\text{E}$ after the accident (Argo Information Center, 2012). Nine of 11 floats were still operational until around January–March 2012; therefore, we can compare our observations and trajectories of 9 Argo floats. In Figs. 1–4, midway into each of

Table 1. ^{134}Cs and ^{137}Cs activity in the surface water in the North Pacific Ocean until March 2012.

Station	Latitude	Longitude	Date	^{134}Cs Bq m^{-3}	\pm	error	^{137}Cs Bq m^{-3}	\pm	error	Ref*
VOS11-001	35.68° N	143.77° E	20110331	507	\pm	33	546	\pm	28	a
VOS11-043	34.95° N	143.86° E	20110331	132	\pm	9	146	\pm	8	a
VOS11-003	36.60° N	147.60° E	20110401	1000	\pm	70	1080	\pm	60	a
VOS11-005	37.42° N	151.08° E	20110401	32.4	\pm	2.6	34.7	\pm	2.0	b
VOS11-044	35.07° N	146.44° E	20110401	34.0	\pm	2.6	36.6	\pm	2.1	a
VOS11-045	35.29° N	151.41° E	20110401	33.4	\pm	2.7	40.3	\pm	2.3	b
VOS11-007	38.18° N	154.97° E	20110402	17.7	\pm	1.6	21.3	\pm	1.3	b
VOS11-009	38.08° N	158.58° E	20110402	1.9	\pm	0.5	3.5	\pm	0.4	b
VOS11-046	35.12° N	154.14° E	20110402	9.2	\pm	1.0	11.1	\pm	0.8	a
VOS11-047	34.89° N	158.76° E	20110402	6.6	\pm	0.8	6.5	\pm	0.5	a
VOS11-048	34.76° N	161.27° E	20110402	2.6	\pm	0.5	3.4	\pm	0.4	a
VOS11-085	33.85° N	141.31° E	20110402	BD	\pm	NA	1.6	\pm	0.2	a
VOS11-011	37.38° N	162.40° E	20110403	2.0	\pm	0.4	3.4	\pm	0.3	b
VOS11-013	36.67° N	166.15° E	20110403	2.2	\pm	0.4	3.9	\pm	0.3	b
VOS11-049	34.09° N	173.28° E	20110403	3.4	\pm	0.6	6.1	\pm	0.5	a
VOS11-086	34.33° N	144.68° E	20110403	158	\pm	11	181	\pm	9	a
VOS11-087	34.51° N	148.40° E	20110403	98.4	\pm	6.7	117	\pm	6	a
VOS11-015	35.28° N	173.53° E	20110404	2.1	\pm	0.4	3.8	\pm	0.3	b
VOS11-050	35.09° N	173.38° E	20110404	BD	\pm	NA	2.4	\pm	0.3	b
VOS11-088	34.66° N	150.97° E	20110404	73.3	\pm	5.6	86.8	\pm	4.9	b
VOS11-089	34.81° N	153.51° E	20110404	10.2	\pm	1.1	12.5	\pm	0.8	a
VOS11-090	34.98° N	156.16° E	20110404	2.7	\pm	0.7	6.0	\pm	0.6	a
VOS11-017	35.13° N	179.67° W	20110405	BD	\pm	NA	2.1	\pm	0.2	b
VOS11-051	33.85° N	179.69° E	20110405	BD	\pm	NA	2.0	\pm	0.2	b
VOS11-091	35.15° N	158.93° E	20110405	3.3	\pm	0.6	7.3	\pm	0.6	a
VOS11-092	35.32° N	161.70° E	20110405	3.4	\pm	0.6	5.1	\pm	0.4	a
VOS11-019	34.08° N	173.87° W	20110406	BD	\pm	NA	1.8	\pm	0.2	b
VOS11-093	35.66° N	167.30° E	20110406	3.4	\pm	0.6	4.7	\pm	0.4	b
VOS11-094	35.99° N	173.05° E	20110406	BD	\pm	NA	2.2	\pm	0.2	b
VOS11-095	36.35° N	178.99° E	20110406	BD	\pm	NA	2.2	\pm	0.4	a
VOS11-021	41.12° N	167.75° W	20110407	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-052	33.91° N	173.89° W	20110407	BD	\pm	NA	1.8	\pm	0.2	b
VOS11-053	33.96° N	168.73° W	20110407	BD	\pm	NA	1.5	\pm	0.2	b
VOS11-096	36.70° N	174.98° W	20110407	BD	\pm	NA	1.8	\pm	0.2	b
VOS11-023	42.33° N	159.88° W	20110408	BD	\pm	NA	1.8	\pm	0.2	b
VOS11-054	33.86° N	161.73° W	20110408	1.0	\pm	0.3	2.3	\pm	0.2	b
VOS11-097	36.99° N	169.34° W	20110408	BD	\pm	NA	1.6	\pm	0.2	b
VOS11-025	43.00° N	151.95° W	20110409	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-055	33.46° N	154.15° W	20110409	BD	\pm	NA	1.4	\pm	0.2	b
VOS11-098	36.84° N	163.23° W	20110409	BD	\pm	NA	1.9	\pm	0.2	b
VOS11-027	43.62° N	143.57° W	20110410	BD	\pm	NA	2.2	\pm	0.2	b
VOS11-056	32.52° N	146.59° W	20110410	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-099	36.50° N	157.55° W	20110410	BD	\pm	NA	2.1	\pm	0.2	b
VOS11-100	35.88° N	151.92° W	20110410	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-125	33.29° N	142.20° E	20110410	3.1	\pm	0.6	3.5	\pm	0.4	b
VOS11-029	38.18° N	134.97° W	20110411	BD	\pm	NA	1.4	\pm	0.2	b
VOS11-057	31.32° N	140.20° W	20110411	BD	\pm	NA	1.8	\pm	0.2	b
VOS11-101	34.97° N	146.43° W	20110411	BD	\pm	NA	1.9	\pm	0.2	b
VOS11-127	35.36° N	147.57° E	20110411	2.2	\pm	0.6	3.3	\pm	0.3	b
VOS11-102	33.92° N	141.12° W	20110412	BD	\pm	NA	1.9	\pm	0.2	b
VOS11-103	32.50° N	135.86° W	20110412	BD	\pm	NA	1.6	\pm	0.2	b
VOS11-129	39.01° N	152.70° E	20110412	1.8	\pm	0.4	3.5	\pm	0.3	b
VOS11-058	28.94° N	130.73° W	20110413	BD	\pm	NA	1.4	\pm	0.2	b
VOS11-059	27.66° N	126.44° W	20110413	BD	\pm	NA	1.9	\pm	0.2	b

Table 1. Continued.

Station	Latitude	Longitude	Date	^{134}Cs Bq m^{-3}	\pm	error	^{137}Cs Bq m^{-3}	\pm	error	Ref*
VOS11-104	31.02° N	130.83° W	20110413	BD	\pm	NA	2.2	\pm	0.2	b
VOS11-131	42.20° N	157.80° E	20110413	8.9	\pm	1.4	16.7	\pm	1.2	b
MR1103-02	36.16° N	142.03° E	20110414	6.0	\pm	1.0	9.0	\pm	1.0	c
MR1103-03	37.09° N	142.72° E	20110414	109	\pm	3	117	\pm	4	c
MR1103-04	37.86° N	143.31° E	20110414	273	\pm	4	284	\pm	5	c
VOS11-060	25.42° N	120.36° W	20110414	BD	\pm	NA	1.5	\pm	0.2	b
VOS11-105	29.24° N	126.05° W	20110414	BD	\pm	NA	1.8	\pm	0.2	b
VOS11-133	44.80° N	163.82° E	20110414	2.8	\pm	0.7	3.1	\pm	0.4	b
MR1103-05	38.21° N	143.79° E	20110415	145	\pm	4	148	\pm	5	c
MR1103-06	38.11° N	143.08° E	20110416	172	\pm	4	174	\pm	5	c
MR1103-07	38.99° N	145.78° E	20110416	53	\pm	2	61	\pm	3	c
VOS11-137	46.25° N	177.01° E	20110416	2.3	\pm	0.6	2.7	\pm	0.4	b
VOS11-141	46.23° N	169.31° W	20110416	2.4	\pm	0.5	3.4	\pm	0.3	b
MR1103-09	40.96° N	150.87° E	20110417	67	\pm	2	72	\pm	3	c
MR1103-10	41.96° N	152.46° E	20110417	17	\pm	2	19	\pm	2	c
MR1103-11	42.97° N	154.14° E	20110417	13	\pm	2	14	\pm	1	c
MR1103-12	43.99° N	154.99° E	20110417	41	\pm	2	43	\pm	3	c
MR1103-13	44.96° N	157.05° E	20110418	18	\pm	1	21	\pm	2	c
MR1103-14	45.97° N	158.50° E	20110418	14	\pm	2	16	\pm	2	c
VOS11-143	46.23° N	162.05° W	20110418	2.7	\pm	0.6	3.0	\pm	0.4	b
MR1103-15-01	47.00° N	160.00° E	20110421	8	\pm	1	9	\pm	1	c
MR1103-15-02	47.00° N	160.00° E	20110421	10	\pm	1	7	\pm	1	c
MR1103-15-11	47.00° N	160.00° E	20110421	8	\pm	1	7	\pm	2	c
MR1103-15-12	47.00° N	160.00° E	20110421	5	\pm	1	10	\pm	1	c
VOS11-165	30.07° N	131.81° E	20110421	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-166	27.16° N	131.24° E	20110422	BD	\pm	NA	1.5	\pm	0.2	b
VOS11-167	24.23° N	130.48° E	20110422	BD	\pm	NA	1.6	\pm	0.3	b
MR1103-16	38.08° N	146.42° E	20110426	48	\pm	2	53	\pm	2	c
MR1103-17	37.00° N	146.05° E	20110426	67	\pm	2	67	\pm	2	c
MR1103-18	36.02° N	145.77° E	20110426	5	\pm	1	9	\pm	1	c
MR1103-19	35.00° N	145.40° E	20110426	4	\pm	1	5	\pm	1	c
MR1103-20	34.04° N	145.12° E	20110427	9	\pm	1	9	\pm	1	c
MR1103-21	33.02° N	144.78° E	20110427	13	\pm	1	17	\pm	1	c
MR1103-22	32.47° N	144.50° E	20110427	56	\pm	2	52	\pm	2	c
MR1103-23	31.03° N	144.82° E	20110428	6	\pm	1	7	\pm	1	c
MR1103-25-1	30.00° N	145.00° E	20110428	14	\pm	1	18	\pm	1	c
MR1103-25-2	30.00° N	145.00° E	20110428	3	\pm	1	5	\pm	1	c
MR1103-24	31.01° N	143.74° E	20110503	11	\pm	1	11	\pm	1	c
MR1103-28	33.95° N	140.25° E	20110503	6	\pm	1	6	\pm	1	c
VOS11-147	53.81° N	146.08° W	20110508	BD	\pm	NA	1.6	\pm	0.2	b
VOS11-301	36.49° N	147.73° E	20110509	30.0	\pm	2.6	38.1	\pm	2.3	b
VOS11-310	38.31° N	156.66° E	20110510	12.7	\pm	1.5	14.6	\pm	1.1	b
VOS11-151	53.41° N	178.21° W	20110511	BD	\pm	NA	1.2	\pm	0.2	b
VOS11-307	41.31° N	170.45° E	20110511	3.2	\pm	0.6	5.1	\pm	0.5	b
VOS11-313	43.66° N	170.30° W	20110511	3.3	\pm	0.6	6.4	\pm	0.5	b
VOS11-314	43.61° N	160.96° W	20110512	3.8	\pm	0.7	6.8	\pm	0.5	b
VOS11-317	42.86° N	179.62° E	20110512	164	\pm	11	196	\pm	10	b
VOS11-319	42.72° N	151.12° W	20110513	13.5	\pm	1.5	13.6	\pm	1.1	b
VOS11-321	40.97° N	141.46° W	20110514	7.9	\pm	0.9	10.9	\pm	0.7	b
VOS11-175	15.70° N	130.45° E	20110515	BD	\pm	NA	1.1	\pm	0.2	b
VOS11-322	38.76° N	133.33° W	20110515	1.5	\pm	0.4	3.6	\pm	0.4	b
VOS11-176	21.02° N	131.77° E	20110516	BD	\pm	NA	1.7	\pm	0.3	b
VOS11-177	25.50° N	132.92° E	20110517	BD	\pm	NA	1.2	\pm	0.2	b
KH1108-260	43.37° N	155.00° E	20110815	11.2	\pm	0.8	14.7	\pm	0.8	b

Table 1. Continued.

Station	Latitude	Longitude	Date	^{134}Cs Bq m^{-3}	\pm	error	^{137}Cs Bq m^{-3}	\pm	error	Ref*
KH1108-233	46.21° N	162.12° E	20110816	12.1	\pm	1.1	16.1	\pm	0.9	b
KH1108-234	48.83° N	169.97° E	20110817	0.7	\pm	0.2	2.0	\pm	0.1	b
KH1108-235	50.92° N	179.07° E	20110818	BD	\pm	NA	1.6	\pm	0.2	b
KH1108-236	53.08° N	171.65° W	20110819	BD	\pm	NA	0.9	\pm	0.1	b
KH1108-251	50.09° N	144.99° W	20110822	BD	\pm	NA	1.2	\pm	0.2	b
KH1108-251D	50.09° N	144.99° W	20110822	BD	\pm	NA	1.2	\pm	0.1	b
KH1108-273	38.62° N	144.99° W	20110829	BD	\pm	NA	1.7	\pm	0.2	b
KH1108-261	30.83° N	145.00° W	20110902	BD	\pm	NA	1.7	\pm	0.2	b
KH1108-263	34.54° N	133.34° W	20110903	BD	\pm	NA	1.8	\pm	0.2	b
KH1108-272	43.00° N	140.99° W	20110914	0.7	\pm	0.2	1.9	\pm	0.1	b
KH1108-226	42.98° N	149.20° W	20110916	BD	\pm	NA	1.5	\pm	0.1	b
KH1108-226D	42.98° N	149.20° W	20110916	0.5	\pm	0.2	1.6	\pm	0.1	b
KH1108-237	36.00° N	156.02° W	20110919	BD	\pm	NA	1.7	\pm	0.2	b
KH1108-240	35.99° N	166.23° W	20110922	0.4	\pm	0.2	2.0	\pm	0.2	b
KH1108-C065-bucket-383	36.00° N	175.00° E	20110927	2.0	\pm	0.3	3.9	\pm	0.3	b
KH1108-C070-bucket-424	35.01° N	165.00° E	20110929	9.5	\pm	0.8	13.3	\pm	0.8	b
KS1109-3101	40.03° N	165.01° E	20111012	23.3	\pm	1.6	30.2	\pm	1.6	b
VOS11-500	38.87° N	156.31° E	20111016	15.5	\pm	1.2	21.3	\pm	1.2	b
VOS11-504	39.50° N	163.97° E	20111017	17.5	\pm	1.3	22.8	\pm	1.3	b
VOS11-508	39.98° N	178.52° W	20111019	1.2	\pm	0.2	3.1	\pm	0.2	b
VOS11-512	39.49° N	162.07° W	20111021	BD	\pm	NA	1.8	\pm	0.1	b
VOS11-516	36.98° N	146.51° W	20111023	BD	\pm	NA	1.8	\pm	0.1	b
VOS11-520	32.53° N	132.63° W	20111025	BD	\pm	NA	1.6	\pm	0.1	b
VOS11-524	23.01° N	114.02° W	20111027	BD	\pm	NA	2.0	\pm	0.2	b
VOS11-589	33.12° N	144.46° E	20111108	6.3	\pm	0.7	9.6	\pm	0.6	b
VOS11-619	32.52° N	146.04° E	20111108	BD	\pm	NA	2.1	\pm	0.2	b
VOS11-587	31.00° N	152.40° E	20111109	1.7	\pm	0.3	3.9	\pm	0.3	b
VOS11-588	30.99° N	153.74° E	20111109	2.3	\pm	0.4	4.7	\pm	0.3	b
VOS11-585	31.00° N	159.51° E	20111110	2.1	\pm	0.4	3.9	\pm	0.3	b
VOS11-586	31.00° N	161.23° E	20111110	1.4	\pm	0.3	2.7	\pm	0.2	b
VOS11-584	31.00° N	167.52° E	20111111	1.6	\pm	0.3	3.3	\pm	0.3	b
VOS11-611	31.00° N	174.47° E	20111112	3.6	\pm	0.6	5.9	\pm	0.4	b
VOS11-612	31.75° N	171.22° W	20111112	BD	\pm	NA	1.6	\pm	0.2	b
VOS11-613	31.96° N	162.63° W	20111113	BD	\pm	NA	1.6	\pm	0.2	b
VOS11-608	31.78° N	154.78° W	20111114	BD	\pm	NA	1.3	\pm	0.2	b
VOS11-609	31.07° N	147.29° W	20111115	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-610	29.90° N	139.70° W	20111116	BD	\pm	NA	1.8	\pm	0.2	b
VOS11-620	28.47° N	132.92° W	20111117	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-621	26.59° N	125.68° W	20111118	BD	\pm	NA	1.5	\pm	0.2	b
VOS11-627	37.12° N	150.59° E	20111118	38.8	\pm	2.8	53.3	\pm	2.9	b
VOS11-668	34.74° N	144.59° E	20111118	BD	\pm	NA	2.3	\pm	0.2	b
VOS11-670	34.75° N	146.59° E	20111118	0.7	\pm	0.2	2.0	\pm	0.2	b
VOS11-672	34.75° N	152.04° E	20111118	1.3	\pm	0.2	3.1	\pm	0.2	b
VOS11-622	24.36° N	118.96° W	20111119	BD	\pm	NA	1.4	\pm	0.2	b
VOS11-629	38.94° N	158.10° E	20111119	24.5	\pm	2.0	31.3	\pm	1.8	b
VOS11-674	34.81° N	154.11° E	20111119	0.8	\pm	0.2	2.8	\pm	0.2	b
VOS11-631	42.07° N	172.01° E	20111120	31.5	\pm	2.5	41.5	\pm	2.3	b
VOS11-676	35.33° N	161.22° E	20111120	13.3	\pm	1.0	19.6	\pm	1.1	b
VOS11-678	35.88° N	168.70° E	20111120	13.5	\pm	1.1	19.0	\pm	1.1	b
VOS11-633	45.64° N	171.01° W	20111122	1.9	\pm	0.3	3.4	\pm	0.3	b
VOS11-680	36.03° N	179.37° E	20111122	2.3	\pm	0.3	4.6	\pm	0.3	b
VOS11-682	36.02° N	170.75° W	20111122	1.7	\pm	0.2	4.3	\pm	0.3	b
VOS11-684	35.78° N	163.53° W	20111123	0.5	\pm	0.2	2.2	\pm	0.2	b
VOS11-635	48.36° N	153.39° W	20111124	0.8	\pm	0.2	1.5	\pm	0.1	b

Table 1. Continued.

Station	Latitude	Longitude	Date	^{134}Cs Bq m^{-3}	\pm	error	^{137}Cs Bq m^{-3}	\pm	error	Ref*
VOS11-686	34.83° N	150.78° W	20111124	BD	\pm	NA	1.8	\pm	0.1	b
VOS11-688	34.42° N	147.68° W	20111125	BD	\pm	NA	1.9	\pm	0.2	b
VOS11-637	47.91° N	135.59° W	20111126	BD	\pm	NA	1.3	\pm	0.2	b
VOS11-690	32.29° N	140.64° W	20111126	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-692	30.65° N	135.59° W	20111126	BD	\pm	NA	1.5	\pm	0.1	b
VOS11-528	18.82° N	128.98° W	20111128	BD	\pm	NA	1.6	\pm	0.1	b
VOS11-694	27.78° N	126.78° W	20111128	BD	\pm	NA	1.8	\pm	0.1	b
VOS11-696	25.27° N	119.89° W	20111129	BD	\pm	NA	1.7	\pm	0.1	b
VOS11-532	20.00° N	143.49° W	20111130	BD	\pm	NA	1.9	\pm	0.1	b
VOS11-698	22.58° N	113.60° W	20111130	BD	\pm	NA	1.2	\pm	0.1	b
VOS11-700	19.68° N	107.36° W	20111201	BD	\pm	NA	1.4	\pm	0.1	b
VOS11-638	52.44° N	141.29° W	20111204	BD	\pm	NA	1.3	\pm	0.1	b
VOS11-536	21.50° N	178.69° W	20111205	BD	\pm	NA	1.6	\pm	0.1	b
VOS11-639	53.50° N	149.15° W	20111205	BD	\pm	NA	1.2	\pm	0.1	b
VOS11-640	43.63° N	148.53° E	20111213	BD	\pm	NA	1.4	\pm	0.2	b
VOS11-646	32.33° N	138.03° E	20111217	BD	\pm	NA	1.4	\pm	0.2	b
VOS11-595	31.26° N	70.86° W	20111218	BD	\pm	NA	1.0	\pm	0.2	b
VOS11-602	31.62° N	62.81° W	20111219	BD	\pm	NA	1.0	\pm	0.1	b
VOS11-603	32.21° N	55.02° W	20111220	BD	\pm	NA	1.2	\pm	0.1	b
VOS11-604	32.85° N	46.70° W	20111221	BD	\pm	NA	1.3	\pm	0.1	b
VOS11-605	33.45° N	38.73° W	20111222	BD	\pm	NA	1.1	\pm	0.1	b
VOS11-606	34.09° N	30.34° W	20111223	BD	\pm	NA	1.1	\pm	0.1	b
VOS11-607	34.73° N	22.19° W	20111224	BD	\pm	NA	1.1	\pm	0.1	b
VOS11-596	35.43° N	12.96° W	20111226	BD	\pm	NA	1.2	\pm	0.1	b
VOS11-702	22.11° N	168.54° W	20120103	BD	\pm	NA	1.6	\pm	0.1	b
VOS11-704	22.97° N	179.98° E	20120105	BD	\pm	NA	1.6	\pm	0.2	b
VOS11-706	23.83° N	169.24° E	20120108	BD	\pm	NA	1.5	\pm	0.1	b
VOS11-708	28.79° N	149.73° E	20120112	0.9	\pm	0.2	2.6	\pm	0.2	b
VOS11-542	34.45° N	130.08° E	20120121	BD	\pm	NA	1.7	\pm	0.2	b
VOS11-543	34.45° N	130.08° E	20120121	BD	\pm	NA	1.4	\pm	0.1	b
VOS11-544	32.53° N	132.98° E	20120122	BD	\pm	NA	1.6	\pm	0.2	b
VOS11-545	32.53° N	132.98° E	20120122	BD	\pm	NA	1.3	\pm	0.1	b
VOS12-710	34.88° N	146.52° E	20120122	BD	\pm	NA	1.9	\pm	0.2	b
VOS11-546	33.57° N	136.44° E	20120123	BD	\pm	NA	1.3	\pm	0.1	b
VOS11-547	33.57° N	136.45° E	20120123	BD	\pm	NA	1.4	\pm	0.2	b
VOS12-716	40.61° N	151.98° E	20120124	17.4	\pm	1.3	26.5	\pm	1.4	b
VOS12-718	43.12° N	153.82° E	20120124	3.0	\pm	0.4	5.1	\pm	0.3	b
VOS12-756	32.41° N	156.27° E	20120126	3.7	\pm	0.5	6.6	\pm	0.5	b
VOS12-763	26.89° N	177.94° W	20120129	BD	\pm	NA	1.6	\pm	0.2	b
VOS12-759	27.84° N	170.90° W	20120130	0.8	\pm	0.2	2.1	\pm	0.2	b
VOS12-762	32.98° N	162.94° W	20120131	BD	\pm	NA	1.7	\pm	0.2	b
VOS12-761	33.05° N	155.28° W	20120201	BD	\pm	NA	1.6	\pm	0.2	b
VOS12-766	34.26° N	146.90° W	20120202	BD	\pm	NA	2.0	\pm	0.2	b
VOS12-768	35.16° N	139.09° W	20120203	BD	\pm	NA	2.2	\pm	0.2	b
VOS12-724	48.99° N	140.82° W	20120204	BD	\pm	NA	1.3	\pm	0.1	b
VOS12-769	36.36° N	131.17° W	20120204	BD	\pm	NA	1.7	\pm	0.2	b
VOS12-726	47.53° N	131.87° W	20120205	BD	\pm	NA	1.4	\pm	0.1	b
KS1202-3326	19.99° N	164.98° E	20120215	BD	\pm	NA	1.3	\pm	0.1	b
KS1202-3328	18.00° N	164.99° E	20120215	BD	\pm	NA	1.2	\pm	0.1	b
VOS12-773	26.82° N	173.34° E	20120217	BD	\pm	NA	2.4	\pm	0.3	b
VOS12-776	27.11° N	158.80° E	20120219	0.9	\pm	0.3	3.2	\pm	0.3	b
VOS12-778	29.35° N	151.75° E	20120220	1.4	\pm	0.3	3.7	\pm	0.3	b
VOS12-1028	32.29° N	153.33° W	20120224	BD	\pm	NA	1.6	\pm	0.2	b
VOS12-1044	34.07° N	162.63° E	20120227	4.2	\pm	0.4	7.9	\pm	0.4	b

Table 1. Continued.

Station	Latitude	Longitude	Date	^{134}Cs Bq m $^{-3}$	\pm	error	^{137}Cs Bq m $^{-3}$	\pm	error	Ref*
VOS12-1085	35.60° N	147.09° E	20120228	4.3	\pm	0.4	8.4	\pm	0.5	b
KH1201-EPO17	27.60° N	169.73° E	20120229	1.0	\pm	0.1	2.6	\pm	0.1	b
VOS12-1047	34.53° N	175.90° E	20120229	6.0	\pm	0.5	9.6	\pm	0.5	b
VOS12-1089	36.52° N	154.17° E	20120229	9.2	\pm	0.7	16.7	\pm	0.9	b
VOS12-1093	37.50° N	161.82° E	20120301	6.8	\pm	0.6	11.4	\pm	0.6	b
KH1201-EPO19	30.11° N	159.62° E	20120302	1.6	\pm	0.1	3.5	\pm	0.2	b
VOS12-1053	33.42° N	163.89° W	20120302	BD	\pm	NA	2.1	\pm	0.1	b
VOS12-1097	39.46° N	177.47° E	20120302	8.6	\pm	0.7	13.6	\pm	0.8	b
KH1201-EPO20	31.41° N	154.28° E	20120303	3.4	\pm	0.3	6.6	\pm	0.4	b
KH1201-EPO21	32.49° N	149.82° E	20120304	1.9	\pm	0.2	3.9	\pm	0.2	b
VOS12-1068	30.09° N	148.73° W	20120304	BD	\pm	NA	1.7	\pm	0.1	b
KH1201-EPO22	33.77° N	144.39° E	20120305	0.6	\pm	0.1	2.4	\pm	0.1	b
VOS12-1101	41.49° N	166.02° E	20120305	1.2	\pm	0.2	3.0	\pm	0.2	b
VOS12-1105	42.12° N	149.75° E	20120307	BD	\pm	NA	1.6	\pm	0.1	b
VOS12-1109	40.45° N	133.84° E	20120309	BD	\pm	NA	1.7	\pm	0.1	b
VOS12-1113	31.92° N	136.82° W	20120316	BD	\pm	NA	1.6	\pm	0.1	b
VOS12-1117	32.97° N	152.40° E	20120318	BD	\pm	NA	1.7	\pm	0.1	b
VOS12-1119	34.06° N	161.83° E	20120319	0.8	\pm	0.2	2.7	\pm	0.2	b
VOS12-1123	34.86° N	177.27° E	20120321	3.4	\pm	0.3	5.8	\pm	0.3	b

a: Aoyama et al. (2012a).

b: This study.

c: Honda et al. (2012).

BD: "below detection limit" and below ca. 0.4 Bq m $^{-3}$ of ^{134}Cs activity is stated as BD in this table.

NA: not available.

Radioactive decay was corrected at the time of collection.

Errors stated in this table are extended uncertainty which include one sigma of counting error, uncertainty of sum effect correction factor, uncertainty of efficiency and uncertainty of assigned activity of standard material used for calibration of Ge-detectors.

the three month periods (Table S1) positions of Argo floats were plotted marked "A–G". In April–June 2011, a distribution of Fukushima radioactive plume and positions of Argo floats showed some discrepancy because the distribution of Fukushima radioactive plume was formed by both combination of atmospheric deposition and direct discharge as stated in Sect. 3.2. In July–September 2011 positions of Argo floats were moved further east up to 165° E, although observed results were so sparse radioactive plume also moved to east as well as Argo floats as shown in Fig. S4. Three months later, both Fukushima radioactive plume and Argo floats moved further eastward up to 172° E as shown in Fig. 3. Almost one year after the accident, again we observed that both the Fukushima radioactive plume and Argo floats moved more east up to 180° E as shown in Fig. S6. A zonal speed, u , based on trajectories of nine Argo floats between May 2011 to August 2011 ranged from 0.1 cm s $^{-1}$ to 15.6 cm s $^{-1}$ with an average of 7.8 cm s $^{-1}$ as shown in Table S2. A zonal speed, u , based on trajectories of nine Argo floats between August 2011 to November 2011 ranged from -1.9 cm s $^{-1}$ to 20.1 cm s $^{-1}$ with an average of 7.7 cm s $^{-1}$ (Table S2). It between November 2011 to February 2012 ranged from -1.7 cm s $^{-1}$ to 16.7 cm s $^{-1}$ with an average of 8.9 cm s $^{-1}$ (Table S2). These zonal speeds by Argo floats showed excellent agreement with zonal speed of the Fukushima ra-

dioactive plume, about 8 cm s $^{-1}$, derived by our observations. Therefore, we can say that deploying Argo floats just after nuclear reactor accidents near the coast line might be good to trace a radioactive plume which moves in the surface layer.

We can also assume that the Fukushima radioactive plume moved with surface water; therefore, it is also interested to compare between surface current speed by satellite observations and actual movement of Fukushima radiocaesium obtained by our observations as stated in Sect. 3.2. Furthermore, we look at surface current observation by satellite (Bonjean and Lagerloef, 2002) along 40° N (38–42° N) as shown in Tables S3-1 and S3-2. As shown in Tables S3-1 and S3-2, zonal speed during the period from April 2011 to March 2012 ranged from 1.5 cm s $^{-1}$ to 7.4 cm s $^{-1}$. The average speed estimated by advection of Fukushima radioactivity was 8 cm s $^{-1}$ on average and this zonal speed by observation showed slightly larger than satellite derived surface current zonal speed observed at 160–180° E. The average speed estimated by advection of Fukushima radioactivity was also larger than satellite derived surface current zonal speed observed at 140–160° E. The reasons for these differences are not clear at this moments, advanced research/discussion should be done in the near future.

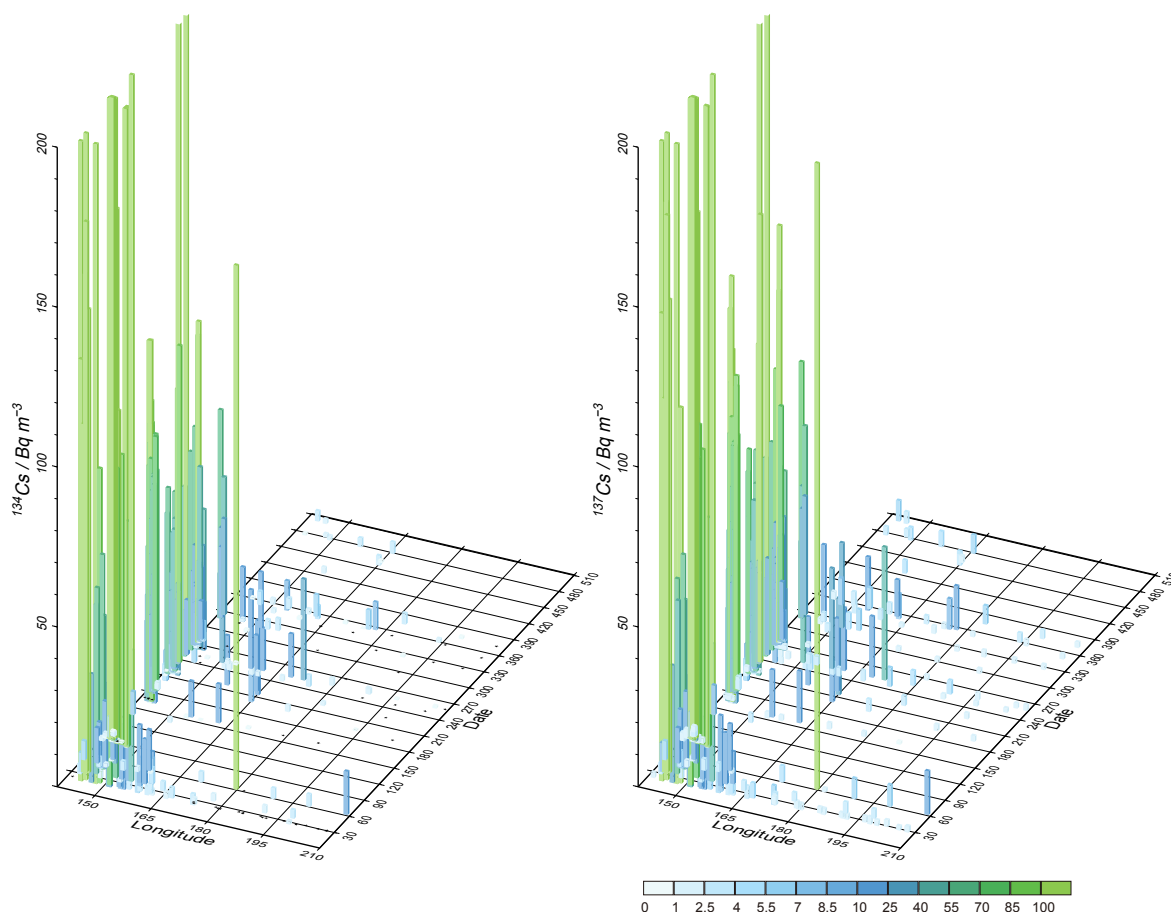


Fig. 5. ^{134}Cs (left) and ^{137}Cs (right) activity in the surface water during the period from 11 March 2011 (day 0) to 31 July 2011 (day 510) in the meridional zone of $38\text{--}42^\circ\text{N}$.

It should also be noted that there exist larger variability in the radioactive plume as shown in Fig. S3. Zonal speed, u , and meridional speed, v , showed temporal and spatial variation as shown in Fig. S7, the resulting positions of Argo floats also showed large variability which indicates that the movement of radioactive plume varied as well.

5 Conclusions

^{134}Cs and ^{137}Cs were released to the North Pacific Ocean by two pathways, direct discharge from the Fukushima NPP1 accident site and atmospheric deposition off Honshu Islands of Japan, east and northeast of the site. High density observations of ^{134}Cs and ^{137}Cs in the surface water were carried out by 17 VOS cruises and several research vessel cruises from March 2011 till March 2012. The main body of radioactive surface plume of which activity was larger than 10 Bq m^{-3} travelled along 40°N , and reached The International Date Line on March 2012, one year after the accident. A distinct feature of the radioactive plume was that it stayed confined along 40°N when the plume reached The International Date

Line. A zonal speed of the surface plume was estimated to be about 8 cm s^{-1} which was consistent with a zonal speed of surface current at the region observed by Argo floats.

Supplementary material related to this article is available online at: <http://www.biogeosciences.net/10/3067/2013/bg-10-3067-2013-supplement.pdf>.

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