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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 since March 2011 till March 2012. Main body of radioactive surface plume of which activity was exceed 10 Bq m^{-3} had been travelling along 40°N , and reached International Date Line on March 2012 one year after the accident. A feature was that the radioactive plume was confined along 40°N when the plume reached 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 and satellite observations at the region.

1 Introduction

On 11 March 2011, an extraordinary earthquake of magnitude 9.0 centred about 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 Fukushima region (The-2011-Tohoku-Earthquake-Tsunami-Joint-Survey-Group, 2011; Mori et al., 2011). These events caused the loss of about 16 000 lives, missing of about 4000 lives 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 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).

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¹³⁴Cs and ¹³⁷Cs were released to the North Pacific Ocean by two major likely pathways, direct discharge from the FNPP1 accident site and atmospheric deposition off Honshu Islands of Japan, east and northeast of the site. ¹³⁴Cs and ¹³⁷Cs activities in the surface water in the North Pacific Ocean were already reported (Aoyama et al., 2012a; Honda et al., 2012) and those ranged a few to 1000 Bq m⁻³ in April–May 2011. Distributions of ¹³⁴Cs and ¹³⁷Cs 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 release period, ¹³⁴Cs and ¹³⁷Cs 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 ¹³⁴Cs 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 and as such has been used as a tracer of water mass mixing and transport (Buesseler et al., 2011).

Results of observations of ¹³⁴Cs and ¹³⁷Cs activities in surface water at Hasaki, a coastal station 180 km south of 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 ¹³⁴Cs and ¹³⁷Cs 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 of which center located at 36.5° N, 141.4° E off Iwaki between Onahama and Hasaki in mid of May 2011 might prevent southward transport of ¹³⁴Cs and ¹³⁷Cs released from FNPP1 to Hasaki until the end of May 2011 (Aoyama et al., 2012b).

Fukushima-derived ¹³⁴Cs and ¹³⁷Cs were detected throughout waters 30–600 km offshore, with the highest activities associated with near-shore eddies and the Kuroshio

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Current acting as a southern boundary for transport in June 2011. They calculate a total inventory of 1.9–2.1 PBq ¹³⁷Cs in an ocean area of 150 000 km² (Buesseler et al., 2012).

However, there is no report of ¹³⁴Cs and ¹³⁷Cs activities in the surface water in the North Pacific Ocean after June 2011. High density sampling of surface seawater to measure ¹³⁴Cs and ¹³⁷Cs 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.

In this paper we present the results of our measurements of ¹³⁴Cs and ¹³⁷Cs 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 these 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). This improvement of AMP procedure realized that the weight yield of AMP/Cs compound basically exceed 99 % for 2 L samples as well as radiochemical yield of radiocaesium. And their activities of AMP/Cs compound 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 (Hama-jima and Komura, 2004). One example of the best performance at this underground facility was reported that a detection limit of ¹³⁷Cs 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 to measure released

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^{134}Cs and ^{137}Cs from the FNPP1 in 2L samples of which activity was less than 1 Bq m^{-3} , too.

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.

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 of the Ministry of Education, Culture, Sports, Science and Technology (hereafter MEXT) and TEPCO to discuss about 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 (Buessler et al., 2011). An analysis of ^{137}Cs concentrations and $^{131}\text{I}/^{137}\text{Cs}$ activity ratios suggest that 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 it decreased much but it was still continuing until July 2011 (Buessler et al., 2011) and thereafter. During the period from August 2011 to July 2012, the activities of ^{134}Cs and ^{137}Cs at near 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.

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3.2 In the North Pacific Ocean

Before the FNPP1 accident, ^{137}Cs was already exist which was 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., 2006, 2011). In 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° by 10° mesh data of the ^{137}Cs deposition (Aoyama et al., 2006). In 2003, ^{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 because due to decay (Aoyama et al., 2012a). In 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 homogeneous 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).

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. S3–S6. The differences between ^{134}Cs and ^{137}Cs activities observed after the FNPP1 accident were consistent with pre-existing ^{137}Cs originated from the nuclear weapons tests as described above.

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It is also clear that ^{134}Cs and ^{137}Cs activities ratios when we take into account the pre-existing ^{137}Cs were close to 1 which is also consistent with observed ^{134}Cs and ^{137}Cs activities ratio of 0.99 ± 0.03 at very close to the source region of the FNPP1 (Buesseler et al., 2011). These are clear evidences 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 just in front of the FNPP1 site showed that the high concentration area located close to the FNPP1 accident site which 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 another high concentration area near the International Date Line in April–June 2011 as shown in Fig. S3 (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 less than 12Bq m^{-3} .

In July–September 2011, relatively high concentration area for which ^{134}Cs activity exceed 10Bq m^{-3} moved eastward and arrived at 165°E as shown in Fig. S4. In October–December 2011, relatively high concentration area for which ^{134}Cs activity exceed 10Bq m^{-3} moved more east and arrived at 172°E along 40°N as shown in Fig. S5. In January–March 2012, it arrived International Date Line as shown in Fig. S6.

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 close area of the FNPP1 site should decrease by dispersion with time rapidly, while eastward movement of radioactive plume relatively higher activity exceed 10Bq m^{-3} was observed as shown in Figs. 1 and S3–S6. The radioactive plume were formed by the atmospheric deposition close to the FNPP1 site and direct discharge. It is

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interesting to estimate a zonal speed of radioactive plume based on our observations. A feature was that the radioactive plume was confined along 40°N when the plume reached 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. 1), therefore an average zonal speed (u) of the surface radioactive plume was calculated to be about 8cm s^{-1} which was consistent with a speed of reported surface current of $4\text{--}16\text{cm s}^{-1}$ at the region (Maximenko et al., 2009).

Eleven Argo floats were deployed off Fukushima on 31 March–13 April at 37.001°N – 37.709°N , 141.250°E – 141.399°E after the accident (Argo-Information-Center). 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. S3–S6, positions of Argo floats at mid time of each three months periods (Table S1) were plotted marked “A”. 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 more 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 more east up to 172°E as shown in Fig. S5. Almost one year after the accident, again we observed that both 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 0.1cm s^{-1} to 15.6cm s^{-1} with an average of 7.8cm 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 -1.9cm s^{-1} to 20.1cm s^{-1} with an average of 7.7cm s^{-1} (Table S2). It between November 2011 to February 2012 ranged from -1.7cm s^{-1} to 16.7cm s^{-1} with an average of 8.9cm s^{-1} (Table S2). These zonal speeds by Argo floats showed excellent agreement with zonal speed of Fukushima radioactive plume, about 8cm s^{-1} , derived by our observations.

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

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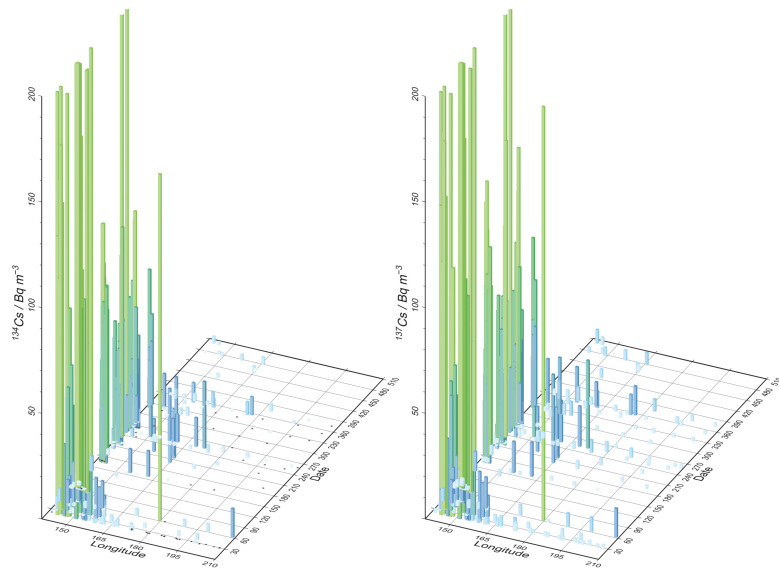


Fig. 1. ^{134}Cs (left) and ^{137}Cs (right) activity in the surface water during the period from 11 March 2011 (day 0) to 31 July 2012 (day 510).