EXPRESS LETTER

Dispersion of artificial caesium-134 and -137 in the western North Pacific one month after the Fukushima accident

MAKIO C. HONDA,^{1*} TATSUO AONO,² MICHIO AOYAMA,³ YASUNORI HAMAJIMA,⁴ HAJIME KAWAKAMI,⁵ MINORU KITAMURA,⁶ YUKIO MASUMOTO,⁷ YASUMASA MIYAZAWA,⁷ MASAYUKI TAKIGAWA⁷ and TOSHIRO SAINO¹

¹Research Institute for Global Change, Japan Agency for Marine-Earth Science and Technology,
2-15 Natsushima, Yokosuka 237-0061, Japan

²Research Center for Radiation Protection, National Institute of Radiological Sciences,
4-9-1 Anagawa, Inage-ku, Chiba 263-8555, Japan

³Geochemical Research Department, Meteorological Research Institute, Nagamine 1-1, Ibaraki 305-0052, Japan

⁴Low Level Radioactivity Laboratory, Kanazawa University, Wake, Nomi, Ishikawa 923-1224, Japan

⁵Mutsu Institute for Oceanography, Japan Agency for Marine-Earth Science and Technology,
690 Sekinehama, Sekine, Mutsu 035-0022, Japan

⁶Institute of Biogeoscience, Japan Agency for Marine-Earth Science and Technology,
2-15 Natsushima, Yokosuka 237-0061, Japan

⁷Research Institute of Global Change, Japan Agency for Marine-Earth Science and Technology,
3173-25 Showa-machi, Kanazawa, Yokohama 236-0001, Japan

(Received October 15, 2011; Accepted November 17, 2011; Online published January 16, 2012)

In March 2011, an accident at the Fukushima Daiichi nuclear power plant (FNPP-AC) was caused by the Tohoku earthquake and tsunami. Here we show the distribution of artificial caesium-134 and -137 (¹³⁴Cs and ¹³⁷Cs) in the western North Pacific one month after the FNPP-AC. In surface seawater, ¹³⁷Cs concentrations were from several times to two orders of magnitude higher than before the FNPP-AC. ¹³⁴Cs was also detected, and in many seawater samples the ¹³⁴Cs/¹³⁷Cs ratio was about 1. These findings indicate that radionuclides from the FNPP dispersed quickly in the western North Pacific. ¹³⁴Cs and ¹³⁷Cs concentrations in suspended solids and zooplankton at stations K2 and S1 were also one to two orders higher than before the accident. Numerical simulation results show that the higher caesium observed in the western North Pacific one month after the FNPP-AC was transported not only by diffusion and advection of seawater but also via the atmosphere as an aerosol.

Keywords: Fukushima, caesium, western North Pacific, zooplankton, suspended solids

INTRODUCTION

On 11 March 2011, a great earthquake occurred off Miyagi, Japan. This earthquake and the generated tsunami inflicted heavy damage on the FNPP. The supply of electricity to FNPP was stopped, which caused the reactor cooling system to malfunction. As a result, the fuel rods were exposed and a meltdown occurred. By venting and hydrogen explosions, the estimated amount of caesium-137 (¹³⁷Cs) emitted was 15 PBq (15×10^{15} becquerels) (NERH, 2011), about one-sixth the amount emitted as a result of the Chernobyl nuclear power plant accident

(about 85 PBq) (WHO, 1989). Large amounts of water to cool the fuel rods was supplied and this procedure increased the amount of water contaminated with radionuclides, and the contaminated water leaked onto the land and into the ocean. By early April (end of May), the estimated total amount of accidentally and deliberately emitted ¹³⁷Cs was 2.6 (3.5 ± 0.7) PBq (Tsumune *et al.*, 2011). The maximum ¹³⁷Cs concentrations at the north drain outlet of the plant and at 30 km off Fukushima were 68,000 Bq L⁻¹ (on 30 March: TEPCO, 2011a) and 186 Bq L⁻¹ (on 15 April: MEXT, 2011), respectively. Thereafter, these concentrations have been decreasing, but the level of radioactivity in seawater is still higher than the background level due to the atmospheric nuclear weapons test before 11 March (MOE, 2009). Moreover, about 0.11 million tons of radioactivity-contaminated water (total radioactivity, 720 PBq, including ¹³⁷Cs, 140 PBq) had

*Corresponding author (e-mail: hondam@jamstec.go.jp)

Copyright © 2012 by The Geochemical Society of Japan.

Fig. 1. Horizontal distribution of ^{137}Cs in surface seawater of the western North Pacific.

accumulated in buildings, trenches and pits at the FNPP by the end of May (TEPCO, 2011b), and it is feared that this contaminated water may leak into the ocean. Thus, different from the Chernobyl accident, an important long-term concern arising from the FNPP-AC is radioactivity-contaminated water and its effects on the marine environment, marine resources and consequently human health. To investigate the transport of radionuclides to the ocean and their dispersion, we conducted a cruise with the R/V *Mirai* in the western North Pacific about one month after the FNPP-AC and measured caesium (^{134}Cs and ^{137}Cs) in seawater, suspended solids (SS) and zooplankton.

METHODS

During the cruise (14 April to 5 May 2011), surface seawaters were collected along the cruise track with an underway surface pump (Fig. 1) and analysed by the ammonium phosphomolybdate/Cs compound method (Hirose *et al.*, 2008). Zooplankton was collected from the surface mixed layer and subsurface layers at two time-series stations (K2, 47°N, 160°E; S1, 30°N, 145°E) with

the Intelligent Operative Net Sampling System. SS larger than $0.45\ \mu\text{m}$ were collected at 10 and 200 m depth at K2 and S1 using an *in situ* filtration system (McLane WTS-6-1-142V). ^{134}Cs and ^{137}Cs radioactivities were measured by low background γ spectrometry for seawater and ultra-low background γ spectrometry using Ge detectors for SS and zooplankton (Hamajima and Komura, 2004). A numerical simulation of radionuclide dispersion in the ocean was conducted with a particle-tracking model using surface currents reproduced by the Japan Coastal Ocean Predictability Experiment 2 (JCOPE2) model (Miyazawa *et al.*, 2009). The radionuclide concentration in each grid is diagnosed from the number of particles in the grid and the radioactive intensity specified for each particle. Initial radioactivity of the particles in the source grid, the closest grid to the FNPP, was determined from the field observations of METI (2011) and TEPCO (2011c). A modified one-way nested global-regional air quality forecasting system (AQF) (Takigawa *et al.*, 2007) was also applied to simulate the time-space variations in the ^{137}Cs aeolian deposition flux over the western North Pacific. The amounts of ^{134}Cs and ^{137}Cs emitted to the atmosphere from the FNPP were obtained by applying

Table 1. ^{134}Cs and ^{137}Cs concentrations and the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio in seawater at each station

Station #	Sampling date and time (UTC)	Latitude	Longitude	SST (deg-C)	Salinity	^{134}Cs (Bq kg ⁻¹)	^{137}Cs (Bq kg ⁻¹)	$^{134}\text{Cs}/^{137}\text{Cs}$	Note
Seawater									
2	2011.4.14 13:30	36-09.4N	142-01.6E	18.8	34.74	0.006 ± 0.001	0.009 ± 0.001	0.75 ± 0.14	
3	2011.4.14 17:38	37-05.5N	142-43.2E	6.3	33.18	0.109 ± 0.003	0.117 ± 0.004	0.93 ± 0.04	Off Fukushima (~200 km from FNPP)
4	2011.4.14 21:35	37-51.7N	143-18.5E	5.6	32.94	0.273 ± 0.004	0.284 ± 0.005	0.96 ± 0.02	Off Fukushima (~200 km from FNPP)
5	2011.4.15 9:00	38-12.6N	143-47.1E	9.3	33.76	0.145 ± 0.004	0.148 ± 0.005	0.98 ± 0.04	Earthquake/Tsunami source
6	2011.4.16 0:08	38-06.8N	143-05.0E	8.1	33.60	0.172 ± 0.004	0.174 ± 0.005	0.99 ± 0.04	Earthquake/Tsunami source
7	2011.4.16 11:06	38-59.3N	145-46.9E	11.3	34.15	0.053 ± 0.002	0.061 ± 0.003	0.87 ± 0.06	
9	2011.4.17 4:23	40-57.6N	150-52.1E	9.6	34.06	0.067 ± 0.002	0.072 ± 0.003	0.92 ± 0.05	
10	2011.4.17 10:23	41-57.4N	152-27.7E	3.1	32.95	0.017 ± 0.002	0.019 ± 0.002	0.92 ± 0.14	
11	2011.4.17 16:33	42-58.5N	154-08.3E	3.0	33.00	0.013 ± 0.002	0.014 ± 0.001	0.89 ± 0.14	
12	2011.4.17 21:26	43-59.6N	154-59.6E	2.6	32.98	0.041 ± 0.002	0.043 ± 0.003	0.94 ± 0.08	
13	2011.4.18 8:34	44-57.7N	157-03.0E	3.6	33.06	0.018 ± 0.001	0.021 ± 0.002	0.88 ± 0.08	
14	2011.4.18 15:35	45-58.0N	158-30.2E	1.9	32.98	0.014 ± 0.002	0.016 ± 0.002	0.85 ± 0.15	
15-01	2011.4.21 21:00	47-00.0N	160-00.0E	1.7	33.02	0.008 ± 0.001	0.009 ± 0.001	0.90 ± 0.01	Station K2
15-02	2011.4.21 21:00	47-00.0N	160-00.0E	1.7	33.02	0.010 ± 0.001	0.007 ± 0.001	1.39 ± 0.01	Station K2 (filtration)
15-11	2011.4.21 21:00	47-00.0N	160-00.0E	1.7	33.02	0.008 ± 0.001*	0.007 ± 0.002*	1.19 ± 0.41	Station K2: 10 m
15-12	2011.4.21 21:00	47-00.0N	160-00.0E	1.7	33.02	0.005 ± 0.001*	0.010 ± 0.001*	0.52 ± 0.13	Station K2: 10 m (filtration)
15-21	2011.4.21 21:00	47-00.0N	160-00.0E	3.7	33.88	<LOD	<LOD		Station K2: 200 m
15-22	2011.4.21 21:00	47-00.0N	160-00.0E	3.7	33.88	<LOD	0.004 ± 0.001		Station K2: 200 m (filtration)
16	2011.4.26 2:11	38-05.0N	146-25.0E	11.7	34.27	0.048 ± 0.002	0.053 ± 0.002	0.90 ± 0.05	Station JKEO
17	2011.4.26 11:34	36-59.8N	146-03.1E	13.1	34.39	0.067 ± 0.002	0.067 ± 0.002	1.00 ± 0.04	
18	2011.4.26 17:25	36-01.2N	145-46.1E	18.9	34.74	0.005 ± 0.001	0.009 ± 0.001	0.63 ± 0.15	Kuroshio extension
19	2011.4.26 23:32	35-00.0N	145-24.2E	18.8	34.78	0.004 ± 0.001	0.005 ± 0.001	0.85 ± 0.32	Kuroshio extension
20	2011.4.27 5:19	34-02.4N	145-07.0E	18.6	34.76	0.009 ± 0.001	0.009 ± 0.001	1.06 ± 0.20	
21	2011.4.27 11:22	33-01.0N	144-47.0E	17.7	34.73	0.013 ± 0.001	0.017 ± 0.001	0.76 ± 0.08	
22	2011.4.27 19:23	32-28.3N	144-30.1E	17.6	34.68	0.056 ± 0.002	0.052 ± 0.002	1.09 ± 0.06	Station KEO
23	2011.4.28 4:03	31-02.1N	144-49.0E	18.6	34.70	0.006 ± 0.001	0.007 ± 0.001	0.94 ± 0.24	
25-1	2011.4.28 22:00	30-00.0N	145-00.0E	19.3	34.76	0.014 ± 0.001	0.018 ± 0.001	0.79 ± 0.06	Station SI: 10 m
25-2	2011.4.28 22:00	30-00.0N	145-00.0E	17.1	34.71	0.003 ± 0.001	0.005 ± 0.001	0.56 ± 0.22	Station SI: 200 m
24	2011.5.3 7:01	31-00.8N	143-44.7E	18.6	34.72	0.011 ± 0.001	0.011 ± 0.001	1.01 ± 0.14	
26	2011.5.3 12:28	31-58.0N	142-33.2E	19.4	34.77	<LOD	0.005 ± 0.001		
27	2011.5.3 18:29	33-00.5N	141-16.0E	20.3	34.65	<LOD	0.004 ± 0.001		
28	2011.5.3 23:59	33-57.0N	140-14.9E	21.2	34.74	0.006 ± 0.001	0.006 ± 0.001	1.05 ± 0.22	

^{134}Cs and ^{137}Cs activities were corrected to sampling data and time; the limit of detection (LOD) of the γ -spectroscopy system is 0.002 Bq kg⁻¹.
 *Accuracy of data for sample #15-1 is lower than others because seawater sample of 10L is smaller than others (20 L).

Table 2. ^{134}Cs and ^{137}Cs concentrations and their ratio in suspended solids and zooplankton at stations S1 and K2

Station	Depth	Sampling date and time (UTC)	^{134}Cs (Bq kg-dw $^{-1}$)	^{137}Cs (Bq kg-dw $^{-1}$)	$^{134}\text{Cs}/^{137}\text{Cs}$	Abundance (mg-dw kg $^{-1}$)	^{137}Cs (mBq kg $^{-1}$)	Water content (%)	^{137}Cs (Bq kg-ww $^{-1}$)	Note
Suspended solids										
K2	10 m	2011.4.22 5:30–7:30	38.38 $\pm$ 5.58	32.42 $\pm$ 2.10	1.18 $\pm$ 0.19	1,654	0.054 $\pm$ 0.006	—	—	
K2	200 m	2011.4.22 5:30–7:30	5.11 $\pm$ 1.24	5.92 $\pm$ 0.94	0.86 $\pm$ 0.25	678	0.004 $\pm$ 0.001	—	—	
S1	10 m	2011.5.02 7:00–9:00	3.55 $\pm$ 1.17	3.42 $\pm$ 0.76	1.04 $\pm$ 1.09	892	0.003 $\pm$ 0.001	—	—	
S1	200 m	2011.5.02 7:00–9:00	13.32 $\pm$ 2.25	10.95 $\pm$ 1.15	1.22 $\pm$ 0.24	664	0.007 $\pm$ 0.001	—	—	
Zooplankton										
K2	0–80 m	2011.4.23 10:00–13:00	15.05 $\pm$ 8.64	13.46 $\pm$ 7.87	1.12 $\pm$ 0.92	61.2	0.001 $\pm$ 0.000	87.2	1.72 $\pm$ 1.00	$n = 5$
K2	80–200 m	2011.4.23 10:00–13:00	55.74 $\pm$ 8.78	59.21 $\pm$ 7.48	0.94 $\pm$ 0.19	5.9	0.000 $\pm$ 0.000	94.7	3.16 $\pm$ 0.40	$n = 3$
S1	0–50 m	2011.5.02 10:00–13:00	70.58 $\pm$ 1.39	71.46 $\pm$ 3.91	0.99 $\pm$ 0.06	10.7	0.001 $\pm$ 0.000	94.4	4.01 $\pm$ 0.22	
S1	50–200 m	2011.5.02 10:00–13:00	60.10 $\pm$ 1.22	57.74 $\pm$ 3.36	1.04 $\pm$ 0.06	5.9	0.000 $\pm$ 0.000	92.5	4.31 $\pm$ 0.25	

the SPEEDI reverse method (Chino *et al.*, 2011) and from data in TEPCO reports (2011d). More detail about the above methods are in Appendix.

RESULTS AND DISCUSSION

Cs in seawater, suspended solids and zooplankton

^{137}Cs concentration in surface seawater ranged from 0.004 to 0.284 Bq kg $^{-1}$ (average 0.048 ± 0.066 Bq kg $^{-1}$) (Table 1). Samples from stations off Fukushima (#3–4) and off Miyagi (#5–6) had higher ^{137}Cs concentrations than those at other stations (Fig. 1). ^{137}Cs concentrations east of this region (#16 [JKEO] and 17) were also high. ^{137}Cs concentrations north of 40°N were relatively higher (0.028 Bq kg $^{-1}$ on average) than those south of 35°N (0.013 Bq kg $^{-1}$ on average). The low ^{137}Cs concentration in the eastward flowing Kuroshio extension current (<0.01 Bq kg $^{-1}$ at #18, #19) were unexpected, because we expected the Kuroshio extension to be the main transport of contaminated water and SS to the pelagic ocean. At K2 and S1, subsurface waters from 200 m depth were also collected by a multiple water-sampling system and radioactive Cs were measured (samples #15–21, 15–22, and 25–2). Although at K2 ^{134}Cs was not detected in subsurface water, at S1 ^{134}Cs was detected. This transport may have occurred during the formation of subtropical mode water (Suga *et al.*, 2004) around S1. In the early 1960s, just after the period of frequent atmospheric nuclear testing (Aoyama and Hirose, 2004), the ^{137}Cs concentration in surface seawater in the Pacific was about 0.01–0.02 Bq kg $^{-1}$. Thereafter, the Cs concentration in surface seawater in the northern North Pacific decreased, to 0.003–0.01 Bq kg $^{-1}$ in the early 1970s (Bowen *et al.*, 1980), and more recently to 0.001–0.002 Bq kg $^{-1}$ (MOE, 2009). Compared to the maximum ^{137}Cs concentrations at the north drain outlet (68,000 Bq kg $^{-1}$; TEPCO, 2011a) and at about 30 km off Fukushima (186 Bq kg $^{-1}$; MEXT, 2011), the ^{137}Cs concentration observed in this study were quite low, although they ranged from a few times to two orders of magnitude higher than those measured before FNPP-AC. Note that at almost every station, ^{134}Cs were detected while not detectable before FNPP-AC. In addition, observed $^{134}\text{Cs}/^{137}\text{Cs}$ ratio ranged from about 0.5 to 1 (Table 1). Taking into account for relative decrease of ^{134}Cs to ^{137}Cs because of differences in background of ^{134}Cs and ^{137}Cs (0 and 0.002 Bq kg $^{-1}$, respectively), and half life (about 2 years for ^{134}Cs against about 30 years for ^{137}Cs), original $^{134}\text{Cs}/^{137}\text{Cs}$ ratio was estimated to be 1 within measurement error. The $^{134}\text{Cs}/^{137}\text{Cs}$ ratio of drain water and air from the FNPP was reported to be nearly 1 (TEPCO, 2011e, f). Thus, radionuclides from the FNPP were likely transported to nearly all of the stations.

^{137}Cs concentration in SS from surface and subsurface waters at stations K2 and S1 ranged from 5.92 to

Fig. 2. Simulated horizontal distribution of ^{137}Cs in surface waters of the western North Pacific during 14–26 April 2011.

32.42 Bq kg⁻¹ on a dry weight basis (kg-dw⁻¹) (Table 2). ^{134}Cs was also detected in all SS from both surface and subsurface waters, and the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio was nearly 1 within measurement error. Thus, it is likely that SS were contaminated with radionuclides from the FNPP. From these data and ^{137}Cs concentration in seawater (0.004–0.018 Bq kg⁻¹ in surface and subsurface waters at K2 and S1, Table 1), we estimated the SS-seawater distribution coefficient (K_d s: Bq kg-dw⁻¹ for SS/Bq kg⁻¹ for seawater) to range from 200 to 4,400, with an average value of 2,100. This average K_d s are comparable to the recommended K_d s for open ocean (2,000) reported previously (IAEA, 2004). The total amount of ^{137}Cs in SS per unit weight of seawater ranged from 0.003 to 0.054 mBq kg⁻¹, about three orders of magnitude lower than ^{137}Cs concentration in seawater. This observation is consistent with the finding that ^{137}Cs concentrations in filtered seawater samples (samples #15-02, 15-12 and 15-22) were not significantly different from those in unfiltered samples (samples #15-01, 15-11 and 15-21) (Table 1). However Cs in SS or particulate Cs is noteworthy because Cs

tends to be adsorbed to clay mineral and this is transported to seafloor quickly and deposited for a long period enough to affect benthos (e.g., Børretzen and Salbu, 2002).

At station K2, the ^{137}Cs concentration in zooplankton from the surface mixed layer (upper 80 m) was 13.46 Bq kg-dw⁻¹ and that in zooplankton from the subsurface layer (80–200 m) was 59.21 Bq kg-dw⁻¹ (Table 2). The corresponding values at S1 were 71.46 Bq kg-dw⁻¹ in the surface mixed layer (upper 50 m) and 57.74 Bq kg-dw⁻¹ in the subsurface layer (50–200 m). The ^{137}Cs concentration in zooplankton tended to be higher than those in SS, possibly because (1) some SS might consist of old particulate materials that had not been radioactively contaminated, whereas fresh particulate materials with higher radioactive Cs might have been taken up by or been adsorbed onto zooplankton; or because (2) SS might include phytoplankton, which is a lower trophic level than zooplankton, and the Cs might have become more concentrated in the higher trophic level (Kasamatsu, 1999). However, at this time, the reason remains unknown. ^{134}Cs

Fig. 3. Modelled cumulative ^{137}Cs aeolian input through 1 April 2011.

was also detected in all zooplankton, with $^{134}\text{Cs}/^{137}\text{Cs}$ ratios of nearly 1 within measurement error. At station K2, which is characterized by a strong thermocline and where no ^{134}Cs was detected in subsurface seawater (Table 2), particulate materials might transport artificial radionuclides to the subsurface layer. Using the water contents of respective zooplankton samples (about 90%, Table 2), we estimated the ^{137}Cs concentrations in zooplankton on a wet weight basis as 1.72 (4.01) and 3.16 (4.31) Bq kg-ww⁻¹ in surface and subsurface from K2 (S1), respectively. Reported ^{137}Cs concentration in zooplankton in at the southern Baltic Sea and in the northern Adriatic Sea one month after the Chernobyl accident were 0.4–2.1 Bq kg-ww⁻¹ (Knapinska-Skiba *et al.*, 2003) and about 5 Bq kg-ww⁻¹ (Marzano and Triulzi, 2003), respectively. These concentrations are comparable to those observed in this study. In contrast, the ^{137}Cs concentration of zooplankton around Japan was less than 0.1 Bq kg-ww⁻¹ during the last decade (~ 0.08 Bq kg-ww⁻¹; Kasamatsu and Ishikawa, 1997, 0.01–0.02 Bq kg-ww⁻¹; Kaeriyama *et al.*, 2008). Thus, the observed concentrations were two orders higher than before 11 March. Previously reported values of the concentration factor (CF), the ratio of the Cs concentration of zooplankton to that of ambient seawater (Bq kg-ww⁻¹/Bq kg⁻¹), range from

10 to 100 (IAEA, 2004; Kasamatsu and Ishikawa, 1997; Kaeriyama *et al.*, 2008), whereas we estimated the CF for zooplankton in this study to range from 200 to 840, an order of magnitude higher than previous observations. The above CF have been estimated under steady state. Thus these might not be applied to our case that Cs concentration of seawater and zooplankton were measured one month after FNPP-AC and both concentrations were still transient. Moreover a possible explanation to this enrichment is that particulate materials with high ^{134}Cs and ^{137}Cs that originated in the FNPP were adsorbed onto the zooplankton, rather than taken up by them, and collected along with the zooplankton.

Numerical simulation

To elucidate how these particulate materials might have been transported to the pelagic western North Pacific and to explain the horizontal distribution of ^{134}Cs and ^{137}Cs in seawater, we carried out numerical simulations with a particle-tracking model based on JCOPE2 (PT-JCOPE2). The results showed that plumes of radioactively contaminated water extended north-eastward near the coast to 40°N and eastward along the northern flank of the Kuroshio extension about one month after the FNPP-AC (Fig. 2). The high ^{137}Cs concentra-

tion observed at #3 to 6 can be qualitatively explained by dispersion of the water discharged directly into the ocean from the FNPP. In contrast, the PT-JCOPE2 result showed no detectable ^{137}Cs north-east of the FNPP beyond 40°N , 150°E , despite relatively large values observed there during the cruise, suggesting that radionuclides were transported to distant locations to the north-east by another mechanism. A numerical simulation of the aeolian deposition flux (AQF) indicated that ^{137}Cs from the FNPP arrived at stations K2 and S1 no later than a week after the FNPP-AC. Simulation of the cumulative aeolian ^{137}Cs input to the ocean by the beginning of April revealed that ^{137}Cs emitted from the FNPP would have been deposited over a wide area of the western North Pacific (Fig. 3). Therefore, the high ^{137}Cs observed in surface waters north of 40°N can be attributed to aeolian ^{137}Cs input. This AQF model result also showed high ^{137}Cs deposition around JKEO (#16) and KEO (#22) stations, which is supported by observation. ^{134}Cs and ^{137}Cs were mainly emitted to the atmosphere between 12 and 15 March. During this period, a low-pressure system passed through eastern Japan (Takemura *et al.*, 2011). The emitted radionuclides may have ridden an updraft and been largely transported to the western North Pacific. The observed high ^{134}Cs and ^{137}Cs concentration in seawater, SS and zooplankton might be attributable mainly to this aeolian input of particles with high ^{134}Cs and ^{137}Cs concentration. However, the AQF model did not always reproduce the observed ^{137}Cs south of 35°N , which were low. The reason for this discrepancy might be that the turnover time of surface water in southern area is short so that the aeolian input of ^{137}Cs was transported eastward away from the surface in this southern area. In addition, estimated deposition of ^{137}Cs into the sea surface via atmosphere this study (0.18 PBq) was smaller than others' estimate (e.g., 5 PBq, Kawamura *et al.*, 2011). Because of a lot of uncertainties in numerical simulation at this moment such as scenario of direct/indirect discharge of Cs, mixed layer depth, meso-scale eddy and biological activity, direct comparison of simulated values with observed data or quantitative discussion will be conducted in future.

CONCLUDING REMARKS

It was qualitatively verified that Cs from FNPP was dispersed to the broad area in the western North Pacific about one month after FNPP-AC via not only ocean, but also atmosphere. After reducing uncertainties in numerical simulation, application of the PT-JCOPE2 model coupled with an AQF model for aeolian input along with time-series observations of vertical profiles of caesium in suspended and sinking particles should clarify the precise spatial and temporal variations in ^{137}Cs observed in the western North Pacific.

Acknowledgments—This observation would not have been possible without the field support obtained from the Captain and crew of R/V *Mirai* from the Global Ocean Development, Inc. and marine technicians from the Marine Works Japan, Inc. We appreciate Dr. Tsumune of Central Research Institute of Electric Power Industry and anonymous reviewers for their fruitful comments. A part of this work is contribution of the JAMSTEC's rapid response research project for the Tohoku-oki earthquake. This observation is also a part of activities of the Great East Japan Earthquake Working Group organized by the Oceanographic Society of Japan (http://www.kaiyogakkai.jp/sinsai_eng/).

REFERENCES

- Aoyama, M. and Hirose, K. (2004) Artificial radionuclides database in the Pacific Ocean: HAM database. *The Scientific World Journal* **4**, 200–215.
- Børretzen, P. and Salbu, B. (2002) Fixation of Cs to marine sediments estimated by a stochastic modeling approach. *J. Environ. Radioact.* **61**, 1–20.
- Bowen, V. T., Noshkin, V. E., Livingston, H. D. and Volchok, H. L. (1980) Fallout radionuclides in the Pacific Ocean: Vertical and horizontal distribution, largely from GEOSECS stations. *Earth Planet. Sci. Lett.* **49**, 411–434.
- Chino, M., Nakayama, H., Nagai, H., Terada, H., Katata, G. and Yamazawa, H. (2011) Preliminary estimation of release amounts of ^{131}I and ^{137}Cs accidentally discharged from the Fukushima Daiichi nuclear power plant into the atmosphere. *J. Nuclear Sci. Tech.* **48**, 1129–1134.
- Hamajima, Y. and Komura, K. (2004) Background components of Ge detectors in Ogoya underground laboratory. *Appl. Radiat. Isotopes* **61**, 179–183.
- Hirose, K., Aoyama, M., Igarashi, Y. and Komura, K. (2008) Improvement of ^{137}Cs analysis in small volume seawater samples using the Ogoya underground facility. *J. Radioanal. Nucl. Chem.* **276**, 795–798.
- IAEA (International Atomic Energy Agency) (2004) Technical reports series 422, 95 pp.
- Kaeriyama, H., Watabe, T. and Kusakabe, M. (2008) ^{137}Cs concentration in zooplankton and its relation to taxonomic composition in the western North Pacific Ocean. *J. Environ. Radioact.* **99**, 1838–1845.
- Kasamatsu, F. (1999) Marine organisms and radionuclides—With special reference to the factors affecting concentration of ^{137}Cs in marine fish—. *Radioisotopes* **48**, 266–282 (in Japanese).
- Kasamatsu, F. and Ishikawa, Y. (1997) Natural variation of radionuclide ^{137}Cs concentration in marine organisms with special reference to the effect of food habits and trophic level. *Mar. Ecol. Prog. Ser.* **160**, 109–120.
- Kawamura, H., Kobayashi, T., Furuno, A., In, T., Ishikawa, Y., Nakayama, T., Shima, S. and Awaji, T. (2011) Preliminary numerical experiments on oceanic dispersion of ^{131}I and ^{137}Cs discharged into the ocean because of the Fukushima daiichi nuclear power plant disaster. *J. Nucl. Sci. Tech.* **48**(11), 1349–1356.
- Knapinska-Skiba, D., Bojanowski, R. and Piekos, R. (2003) Activity concentration of caesium-137 in seawater and

plankton of the Pomeranian Bay (The southern Baltic Sea) before and after flood in 1997. *Mar. Pollut. Bull.* **46**, 1558–1562.

- Marzano, F. N. and Triulzi, C. (2003) A radiological survey of northern and middle Adriatic Sea before and after the Chernobyl event. *Mar. Pollut. Bull.* **28**, 244–253.
- METI (Ministry of Economy, Trade and Industry) (2011) Available at <http://www.nisa.meti.go.jp/english/files/en20110425-3.html>
- MEXT (Ministry of Education, Culture, Sports, Science and Technology) (2011) Available at <http://www.mext.go.jp/english/incident/1304192.htm>
- Miyazawa, Y., Zhang, R., Guo, X., Tamura, H., Ambe, D., Lee, J.-S., Okuno, A., Yoshinari, H., Setou, T. and Komatsu, K. (2009) Water mass variability in the western North Pacific detected in a 15-year eddy resolving ocean reanalysis. *J. Oceanogr.* **65**, 737–756.
- MOE (Ministry of the Environment) (2009) Available at <http://www.kankyo-hoshano.go.jp/en/01/0101flash/01011322.html>
- NERH (Nuclear Emergency Response Headquarters, Government of Japan) (2011) Report of Japanese Government to the IAEA Ministerial Conference on Nuclear Safety—The Accident at TEPCO's Fukushima Nuclear Power Stations—. Available at http://www.kantei.go.jp/foreign/kan/topics/201106/iaea_houkokusho_e.html
- Suga, T., Motoki, K., Aoki, Y. and Macdonald, A. M. (2004) The North Pacific climatology of winter mixed layer and mode waters. *J. Phys. Oceanogr.* **34**, 3–22.
- Takemura, T., Nakamura, H., Takigawa, M., Kondo, H., Satomura, T., Miyasaka, T. and Nakajima, T. (2011) A numerical simulation of global transport of atmospheric particles emitted from the Fukushima Daiichi nuclear power plant. *Sci. Online Lett. Atmos. (SOLA)* **7**, 101–104.
- Takigawa, M., Niwano, M., Akimoto, H. and Takahashi, M. (2007) Development of a one-way nested global-regional air quality forecasting model. *Sci. Online Lett. Atmos. (SOLA)* **3**, 81–84.
- TEPCO (Tokyo Electricity Power Corporation) (2011a) Available at <http://www.tepco.co.jp/en/press/corp-com/release/11040815-e.html>
- TEPCO (2011b) Available at http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110603e12.pdf
- TEPCO (2011c) Available at <http://www.tepco.co.jp/en/press/corp-com/release/11041507-e.html>
- TEPCO (2011d) Available at http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110719e6.pdf
- TEPCO (2011e) Available at http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110322e1.pdf
- TEPCO (2011f) Available at http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110322e1.pdf
- Tsumune, D., Tsubono, T., Aoyama, M. and Hirose, K. (2011) Distribution of oceanic ^{137}Cs from the Fukushima Daiichi Nuclear Power Plant simulated numerically by a regional ocean model, accepted by *J. Environ. Radioact.* (in press).
- WHO (World Health Organization) (1989) Health hazards from radiocaesium following the Chernobyl nuclear accident. Report on a WHO working group. *J. Environ. Radioact.* **10**, 257–259.

APPENDIX: METHODS

Sampling and measurement

Surface seawater, zooplankton and suspended solids were sampled during R/V *Mirai* cruise MR11-03 (14 April to 5 May 2011) (<http://www.godac.jamstec.go.jp/cruisedata/mirai/e/MR11-03.html>). Surface seawater was collected with an underway surface pump at each about 1° of latitude along the cruise track (Yokohama - station K2 - station JKEO - station KEO - station S1 - Yokohama) (Fig. 1). The seawater was not basically filtered. Ammonium phosphomolybdate (AMP) was added to the pH-adjusted seawater and AMP/Cs compounds were precipitated. In the laboratory on land, ^{134}Cs and ^{137}Cs radioactivities in the AMP/Cs compounds were measured by low background γ spectrometry using Ge detectors. The measurement time was 6–72 hours, and the measurement error (statistical counting error) ranged from 2% to 30%, about 10% on average.

Meso- and macro-zooplankton samples were collected with the Intelligent Operative Net Sampling System (IONESS) from the Japan Agency for Marine-Earth Science and Technology (JAMSTEC) time-series stations in the western North Pacific sub-arctic gyre (K2, 47°N, 160°E; ~1900 km from the FNPP) and sub-tropical gyre (S1, 30°N, 145°E; ~900 km from the FNPP). Using CTD data, we defined the surface mixed layer depths as 80 m at K2 and 50 m at S1, and collected samples at the following discrete depth intervals: 0–80 m and 80–200 m at K2, and 0–50 m and 50–200 m at S1. The total duration of the net deployment was ~3 hours. The average volume of water filtered by the net (mesh size 0.33 mm) in a single depth interval was about 1750 m³ and about 325 g zooplankton, wet weight (g-ww), was collected on average. Zooplankton samples were frozen at –40°C onboard. In the laboratory on land, the samples were thawed and dried. Next, the samples were pulverized with an agate pestle and mortar and encased in a V7 vessel. Finally, ^{134}Cs and ^{137}Cs activities of the samples were measured with ultra-low background γ spectrometry using Ge detectors in an underground facility (Ogoya Underground Facility of Kanazawa University) (Hamajima and Komura, 2004). Measurement duration was about 4 hours, and the measurement error (statistical counting error and repeatability) was 25–80% for the K2 samples ($n = 3$ –5) and 2–7% for the S1 sample ($n = 1$).

Suspended solids (SS) at 10 and 200 m depth at K2 and S1 were collected by using an *in situ* filtration system (McLane WTS-6-1-142V). About 185 L of seawater at 10 m and 330 L at 200 m were filtered through a membrane filter (pore size 0.45 μm). ^{134}Cs and ^{137}Cs radioactivities of these samples were measured with ultra-low background γ spectrometry using Ge detectors in the underground facility (9). Measurement duration was

7–19 days. The measurement error (statistical counting error and repeatability) for zooplankton was 25–80% for the K2 samples ($n = 3–5$) and 2–7% for the S1 sample ($n = 1$) (Table 2). The average error for suspended particles was about 19% for ^{134}Cs and 13% for ^{137}Cs .

Numerical simulation model

1) JCOPE2 particle tracking model

The Japan Coastal Ocean Predictability Experiment 2 (JCOPE2) particle tracking model simulates the movement of particles by advection and diffusion by ocean currents, and it takes into account the half-life of each radionuclide. The surface current data reproduced by the JCOPE2 operational ocean forecast system were used to determine the advection. JCOPE2 is a data-assimilative ocean general circulation model with a horizontal resolution of $1/12^\circ$. Diffusion is calculated by the random-walk method based on a Smagolinsky-type parameterization. The radionuclide concentration in each grid is diagnosed from the number of particles in the grid and the radioactive intensity specified for each particle. Initial radioactivity of the particles in the source grid, the closest grid to the FNPP, was determined from the field observations of METI (2011) and TEPCO (2011c). Our source term represents over $1,000 \text{ Bq L}^{-1}$ of the source grid concentration on the first simulation day (21 March 2011), maximum of over $20,000 \text{ Bq L}^{-1}$ for the period

from the end of March to the beginning of April, and gradual decrease to approximately 100 Bq L^{-1} level during May, suggesting 3.7 PBq of total amount by direct emission to ocean for the period from 21 March to 6 May 2011.

2) AQF

The AQF system consists of global and regional chemical transport models (CTMs). The global CTM is based on CHASER (Chemical Atmospheric General Circulation Model for the Study of Atmospheric Environment and Radiative Forcing), whereas the regional CTM is based on the WRF (Weather Research and Forecasting)/Chem model. As the background level of ^{137}Cs is quite low, only the regional part of AQF was applied in this study. The system is driven by meteorological data from the National Centers for Environmental Prediction (NCEP) final operational global analysis data set. The precipitation intensity is predicted by the model and used for the estimation of the wet deposition flux at every model time step (1 minute). The model domain covers the western Pacific with a horizontal resolution of 10 km (820×660 grids). The emissions from FNPP through 6 April 2011 were obtained by applying the SPEEDI reverse method (Chino *et al.*, 2011). The emissions from 7 April to 3 May were linearly interpolated using estimations by TEPCO (2011d).