Rep. Mar. Ecol. Res. Inst., No.22 (Supplement), 17-25, 2017

Special Report: "Radioactivity in the Marine Environment and in Fisheries Products during the Five Years after the Fukushima Dai-ichi Nuclear Power Plant Accident

Long-term Distribution of Radioactive Cesium in the Coastal Seawater and Sediments of Japan

Hyoe Takata*1§, Masashi Kusakabe*1, Naohiko Inatomi*1, Kazuyuki Hasegawa*1, Takahito Ikenoue*1, Yukihiko Watanabe*2, Teruhisa Watabe*3, Chiyoshi Suzuki*3, Jun Misonoo*3 and Shigemitsu Morizono*3

Abstract: A radioactivity survey was launched in 1983 to determine background levels of artificial radionuclides such as ^{90}Sr and ^{137}Cs in marine environments offshore of nuclear power stations throughout Japan and one nuclear fuel reprocessing plant. In addition, an environmental radioactivity monitoring study, with a particular focus on the waters off Fukushima and nearby prefectures, was initiated shortly after the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. Here, we report on spatiotemporal changes of radiocesium in the waters off FDNPP, as well as on long-term (>30-year) temporal changes in radionuclides in the marine environment off the nuclear power stations and the fuel reprocessing facility. We determined that ^{137}Cs concentrations decreased over the period 1983–2010, with the exception of a spike corresponding to the Chernobyl nuclear accident in 1986. Levels of radiocesium prior to the FDNPP accident (2006–2010) indicated that ^{137}Cs concentrations ranged from 1.3 to 2.5 mBq/L (mean: 1.6 mBq/L). Post-accident monitoring revealed a heavy burden of artificial radionuclides in Japanese coastal areas, as ^{137}Cs concentrations in surface seawaters of the northwestern North Pacific Ocean increased markedly. On the other hand, concentrations of ^{134}Cs, which has a short half-life (around 2 years), decreased sharply due to seawater dispersion by coastal currents. Although concentrations of ^{137}Cs in coastal seawater within 10 km of FDNPP remained high even 5 years after the FDNPP accident, concentrations in seawater >30 km from FDNPP have almost returned to background values.

Concentrations of ^{137}Cs in sediments declined slightly over the period 1983–2010, but maximum values were observed at all monitoring stations near the FDNPP in September 2011 (8.0–580 Bq/kg-dry), several months after the accident; almost all concentrations exceeded pre-accident 5-year averages (2.1 Bq/kg-dry) for Japanese coastal regions. However, five years of monitoring after the accident reveal a trend of exponential decline.

Keywords: seawater, sediment, ^{134}Cs, ^{137}Cs, Fukushima Dai-ichi Nuclear Power Plant

Introduction

The Marine Ecology Research Institute (MERI) has been conducting surveys of anthropogenic radionuclides in marine environments offshore of commercial nuclear power plants throughout Japan since 1983. Because of the large amount of radioactive nuclides released as a result of the accident at the Fukushima Dai-ichi Nuclear Power Plant (hereafter "FDNPP") in March 2011, MERI instituted a survey specifically focusing on the distribution and behavior of man-made radionuclides in seawater and marine sediments at sites close to FDNPP (hereafter "FDNPP monitoring"); results of this survey have previously been reported in extensive detail (Takada and Suzuki, 2003; Kusakabe *et al*., 2013; Watabe *et al*., 2013;

(Received 16 February 2017; accepted 18 May 2017)

∗1 Central Laboratory, Marine Ecology Research Institute, 300 Iwawada, Onjuku-machi, Isumi-gun, Chiba 299-5105, Japan
∗2 Demonstration Laboratory, Marine Ecology Research Institute, 4-7-17 Arahama Kashiwazaki-shi, Niigata 945-0017, Japan
∗3 Head Office, Marine Ecology Research Institute, 347 Yamabuki-cho, Shinjku-ku, Tokyo 162-0801, Japan
§ E-mail: takata@kaiseiken.or.jp

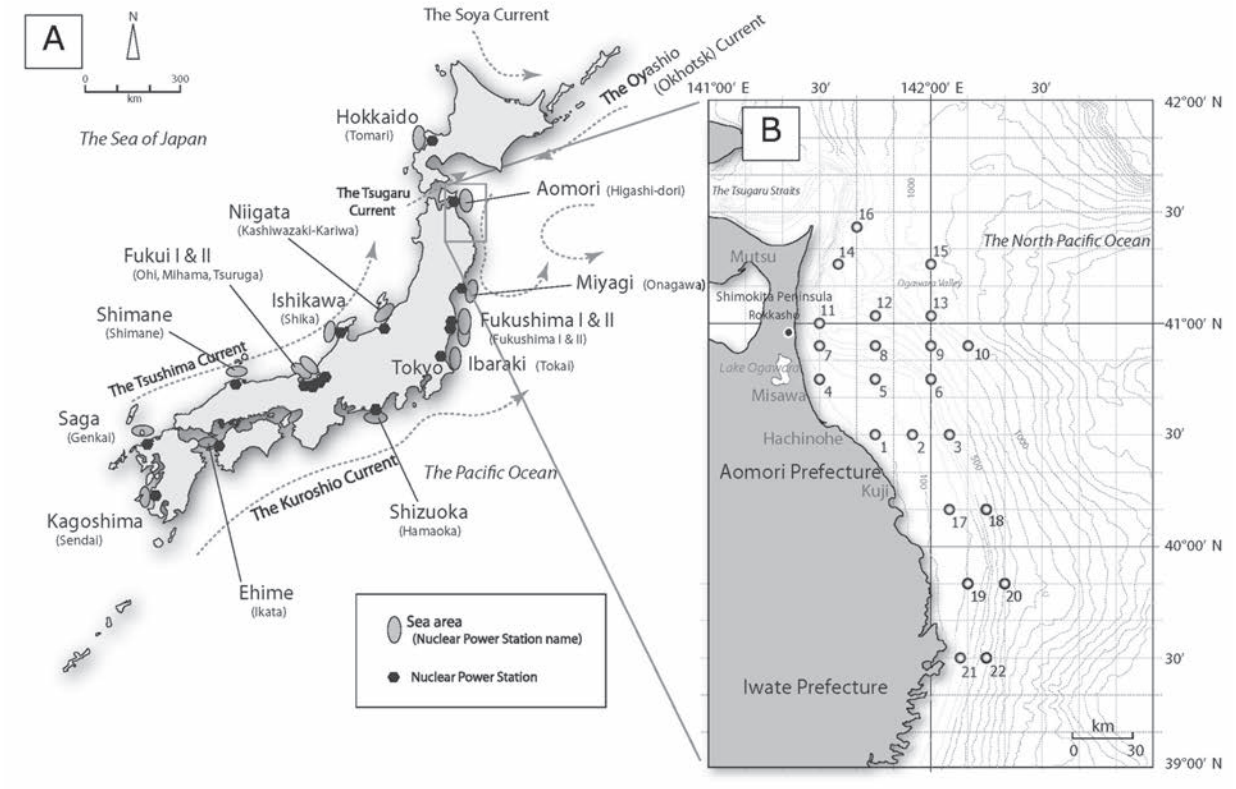

Fig.1 Location of the marine areas included in this study. A: 15 coastal marine regions (four sampling stations in each region) adjacent to commercial nuclear power plants. B: 22 offshore sampling stations in waters adjacent to the nuclear fuel reprocessing facilities.

Oikawa *et al*., 2013a, b, 2014; Kusakabe, 2014; Takata *et al*., 2013, 2015, 2016; Isono *et al*., 2015a, b). Here, we provide a summary of the trends in the concentrations of artificial radioactive nuclides in Japanese coastal waters and sediments since 1983 including the Nuclear Regulation Authority's monitoring data for radioactive Cs (^{134}Cs, ^{137}Cs) concentrations in marine systems over the period 1983–2015.

Methods

Sampling

Sampling procedures have been summarized in detail elsewhere (Oikawa *et al*., 2014; Isono *et al*., 2015a). Briefly, we collected seawater and sediment samples from 15 coastal regions in which commercial nuclear power plants are located, with four sampling stations established within each region (Fig. 1A). Since 1991, prior to regular operation of the nuclear fuel reprocessing facilities in the village of Rokkasho, Aomori Prefecture, surveys involving 22 sampling stations have been conducted in the waters off Aomori and Iwate Prefectures (Fig. 1B). In addition, there has been an ongoing marine environmental monitoring project in the waters offshore of FDNPP since March 2011, shortly after the nuclear accident. As of 2015, the FDNPP monitoring area included 32 sampling stations in coastal areas 30–90 km from FDNPP, along with 10 stations in the open ocean (Fig. 2A) and seven stations within a 10-km radius of FDNPP (Fig. 2B).

Seawater samples were collected annually in May/June; samples consisted of 60–200 L of seawater collected with Van Dorn or Niskin water samplers. Water samples were collected from each station in the surface layer (1-m depth), from a depth of 50–100 m, and from 10–40 m above the seafloor. Seawater samples were not filtered. Two milliliters of 6 M hydrochloric acid or 15 M nitric acid were added per liter of seawater

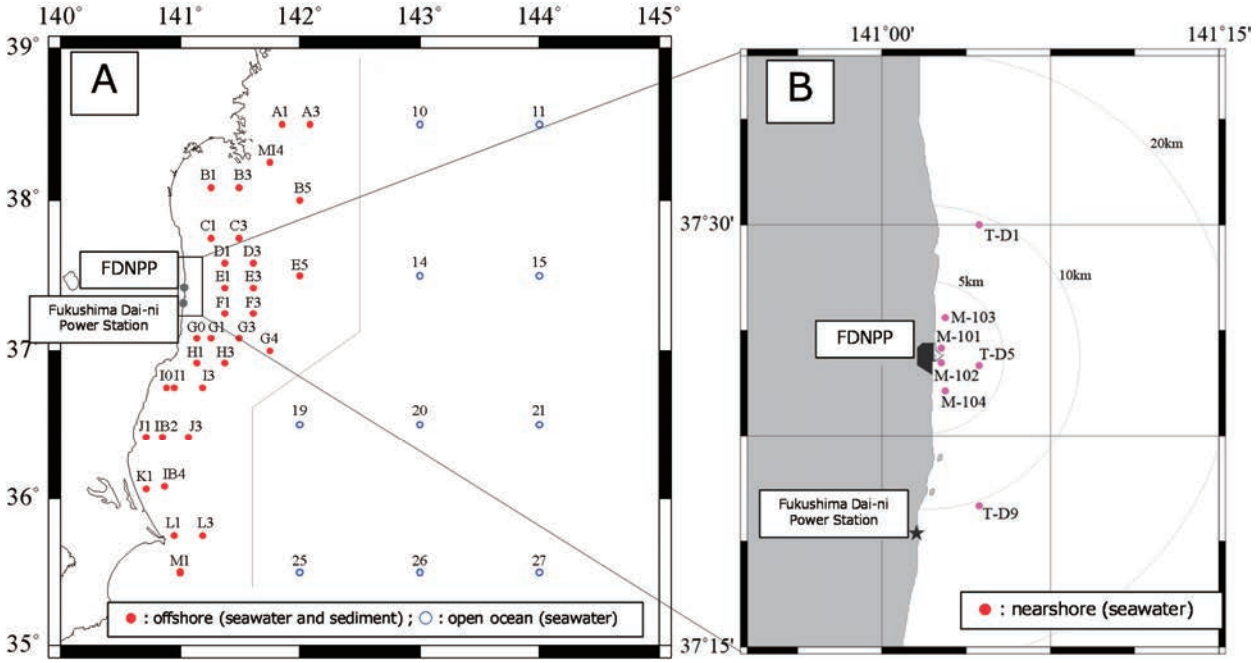

Fig.2 Monitoring area. A: 32 sampling stations in the coastal area located 30-90 km from the FDNPP and 10 stations in the open ocean. B: seven nearshore sampling stations within a 10-km radius of the FDNPP.

to each sample, after which samples were stored under cool, dark conditions until analysis. Surface sediment samples were collected using an Ekman–Birge grab sampler or a box corer (Rigo stainless steel box [45-cm sides] capable of taking pillar-shaped samples at sediment depths up to about 40 cm) or a multiple corer (Rigo acrylic pipe with an inside diameter of 8 cm [eight rows]). We collected about 2.5 kg of sediment from the layer about 3 cm below the surface. The sediment samples were either refrigerated or frozen until analysis.

Analysis

Cesium-137 in the seawater samples was chemically separated and quantified by gamma-ray spectrometry. After drying, measured quantities of seabed sediment were placed in plastic containers, and ^{137}Cs concentrations were determined via gamma-ray spectrometry. The lower detection limits for both ^{134}Cs and ^{137}Cs in all samples were 0.3–1.0 mBq/L for seawater and about 0.7 Bq/kg-dry for seabed sediments. See Oikawa *et al*. (2013a) for additional details of the analytical methods.

Results and Discussion

Cs-137 in seawater offshore of commercial nuclear power plants

Temporal changes in ^{137}Cs concentrations in both surface and bottom layers of seawater in the 15 coastal regions offshore of commercial nuclear power plants for 1983–2015 are presented in Figure 3. Prior to the FDNPP accident, low levels of ^{137}Cs in seawater were detected using beta-ray measurement following sample pre-concentration. However, ^{134}Cs, with a half-life of 2 years, was also released in the accident; the activation product of ^{134}Cs has approximately the same radiation level (activity ratio ≈ 1) as that of ^{137}Cs, which is a fission product. Because beta rays of ^{134}Cs and ^{137}Cs cannot be distinguished, gamma-ray spectrometry was applied to all samples collected since 2012, as this approach can differentiate ^{134}Cs and ^{137}Cs. Cesium levels in some of the samples from 2011, for which the beta-ray approach was used, are reported as $^{134+137}$Cs.

Although ^{137}Cs, which originated primarily from atmospheric nuclear-weapons testing in the Northern Hemisphere between 1945 and 1965, continues to be detected in the surface layers of the North Pacific

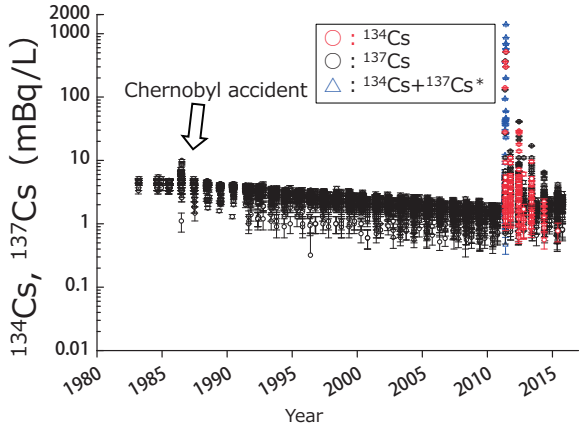

Fig.3 Concentrations of ^{137}Cs in surface waters of the marine areas adjacent to nuclear power plants over the period 1983–2015. Samples based on beta measurements in 2011 included ^{134}Cs in addition to ^{137}Cs; thus, these data are shown as "$^{134+137}$Cs". Vertical bars indicate counting errors (1σ). ND = not detected and therefore not plotted.

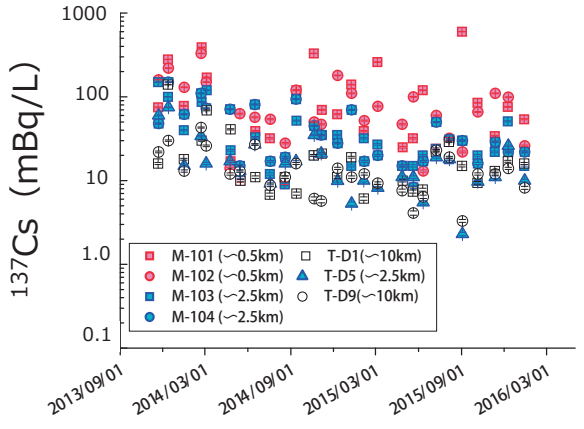

Fig.4 Temporal variation in ^{137}Cs concentrations in seawater in nearshore areas within 10 km of the FDNPP over the period November 2013 to February 2016. Vertical bars indicate counting errors (1σ). Figures in quotation marks next to measurement points indicate distance from the FDNPP.

Ocean, its concentration has gradually declined throughout the survey regions, with theexception of a spike in May–June 1986 due to the April 1986 Chernobyl nuclear power-plant accident. Because the beta-ray measurement method was used at that time, the presence of ^{134}Cs was unconfirmed; however, surveys in the following year (1987) showed that levels of ^{137}Cs were the same as those prior to the Chernobyl

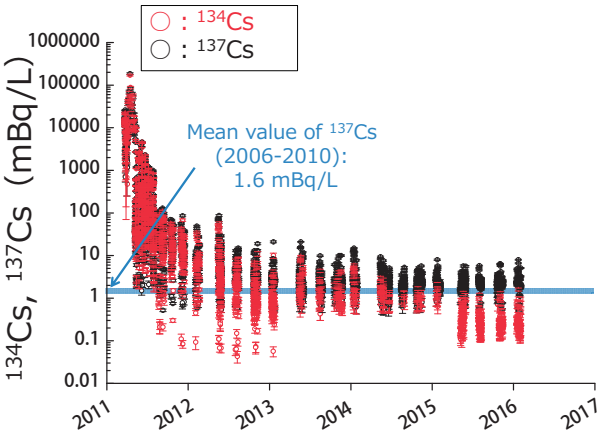

Fig.5 Temporal variations in ^{137}Cs concentrations in seawater offshore of sites over the period March 2011 to February 2016. Vertical bars indicate counting errors (1σ).

accident, declining thereafter at a rate similar to the pre-accident rate. In addition, the mean concentration of ^{137}Cs in seawater based on all data over the 5-year period from 2006 to 2010 (i.e. before the FDNPP accident) was 1.6 mBq/L (range: 1.1–2.5 mBq/L), and the effective environmental half-life was estimated to be 12–20 years, which is shorter than the physical half-life of ^{137}Cs (30.1 years) (Oikawa *et al.*, 2013b). Concentrations of ^{137}Cs increased in the waters off Miyagi, Fukushima Dai-ichi, Fukushima Dai-ni, and Ibaraki monitoring areas in 2011 due to the FDNPP accident, but subsequently decreased exponentially between 2011 and 2015.

Cs-137 in seawater following the FDNPP accident

Temporal variations in ^{137}Cs concentrations in surface seawater within a 10-km radius of the FDNPP are shown in Figure 4. Concentrations of ^{137}Cs in surface seawater ranged from 5.3 mBq/L (station T-D5) to 140 mBq/L (station M-101) in January 2015 and from 8.2 mBq/L (station T-D9) to 54 mBq/L (station M-101) in January 2016, or about one to two orders of magnitude greater than pre-accident levels (1.3–2.5 mBq/L).

Temporal variations in ^{137}Cs concentrations in coastal areas 30–90 km from FDNPP are shown in Figure 5; the unbroken line in this figure represents the mean ^{137}Cs concentration (1.6 mBq/L) in the waters

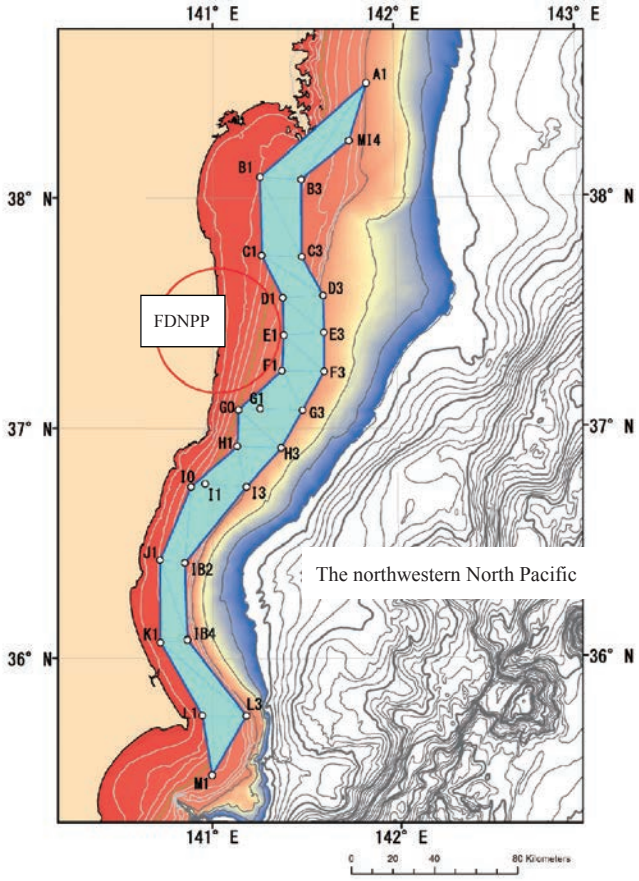

Fig.6 Area used to calculate the ^{137}Cs inventory in seawater (light blue area); circles indicate the 27 stations used for this calculation. Data taken from a report by Takata *et al*. (2016).

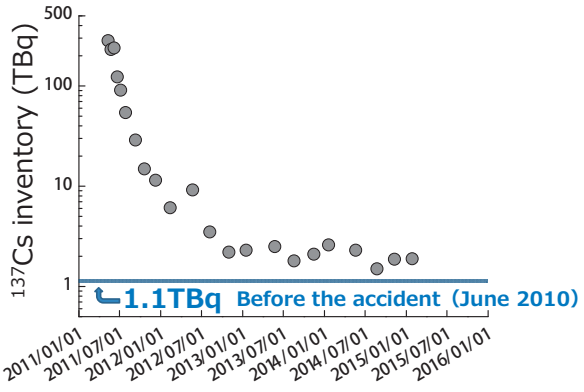

Fig.7 Temporal changes in ^{137}Cs inventories in offshore waters. Data taken from a report by Takata *et al*. (2016).

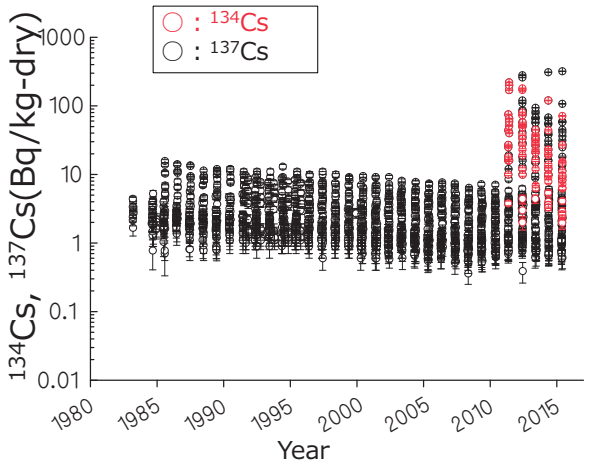

Fig.8 Cs-137 concentrations in sediment samples (0–3 cm depth) collected from 15 coastal marine regions (including each of the four sampling stations in each region) adjacent to commercial nuclear power plants over the period 1983–2015. ND = not detected and therefore not plotted. Vertical bars indicate counting errors (1σ).

offshore of Miyagi, Fukushima, and Ibaraki prefectures over the 5-year period preceding the accident (2006–2010). The highest concentration of ^{137}Cs in the surface seawater after the accident, recorded on 15 April 2011, was 186,000 mBq/L, or approximately five orders of magnitude greater than pre-accident levels. Concentrations then decreased exponentially over time, falling to about 10 mBq/L within 2 years, and maximum levels have not exceeded 10 mBq/L since 2014. Annual mean values of ^{137}Cs concentrations in seawater during the FDNPP monitoring were 2.44 mBq/L in 2014 and 2.35 mBq/L in 2015, and they continue to exhibit a downward trend towards the mean pre-accident background level (1.6 mBq/L).

We conducted a water-column inventory of ^{137}Cs (Bq/m) by measuring ^{137}Cs concentrations at various depths from the surface to the sea bottom at 27 coastal stations (Fig. 6). On the basis of the estimated surface area (6160 km^2) and volume (753 km^3) of the offshore waters on which we focused (30–90 km from the FDNPP), we examined the temporal changes in ^{137}Cs inventories in this area; details of this calculation are described by Takata *et al*. (2016). In May 2011 (2 months after the accident), we estimated a ^{137}Cs inventory of 283 TBq in this region, equivalent to

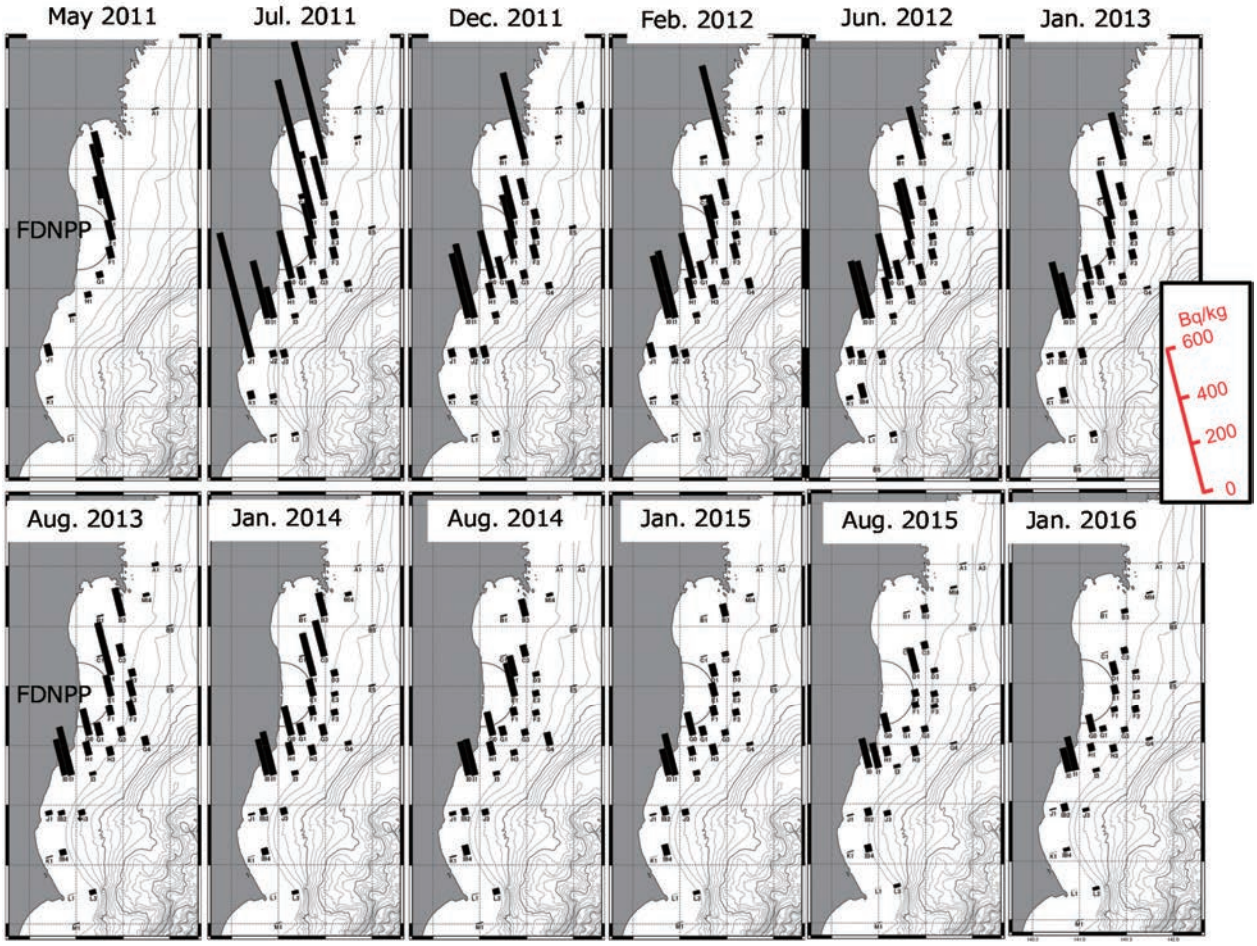

Fig.9 Cs-137 concentrations in sediment samples (0–3 cm depth).

approximately 1.5–1.8% of the total amount of ^{137}Cs that entered the ocean as a result of the accident (approximately 15.5–18.5 PBq; Aoyama *et al*., 2016) (Fig. 7). Our estimated inventory of 283 TBq is over two orders of magnitude higher than the pre-accident inventory of 1.1 TBq, but the inventory declined by an order of magnitude to 14.9 TBq over the subsequent six months (May–October 2011). From October 2011 to February 2015, the inventory fell to 1.89 TBq, thus approaching the pre-accident level; however, relatively high concentrations of ^{134}Cs and ^{137}Cs are still being recorded at nearshore stations, and thus continuous monitoring remains a priority.

Cs-137 in sediments offshore of commercial nuclear power plants

Temporal variations in ^{137}Cs concentrations in sediments over the period 1983–2015 are presented in Figure 8. Concentrations of ^{137}Cs are highly heterogeneous in sediments because of the changes in range of grain sizes and the surface areas of sediment particles; for example, Cs tends to accumulate more in muddy sediments than in sandy sediments. As such, ^{137}Cs concentrations were slightly higher in the predominately muddy sediments off Hokkaido, Niigata, and Fukui prefectures than in the sandy sediments that are more common in other regions (Oikawa *et al*., 2013b). Cesium-137 levels in sediments have, however, generally declined slightly since 1983; even the Chernobyl accident, which raised ^{137}Cs concentrations in surface waters for a year, had no effect on sediment concentrations (Oikawa *et al*., 2013b). We estimated the 5-year (2006–2010) average ^{137}Cs concentration in sediments to be 2.1 Bq/kg-dry (range: 0.72–8.0 Bq/kg-dry).

Cs-137 in sediments following the FDNPP accident

Sampling of the seabed began in May 2011, two

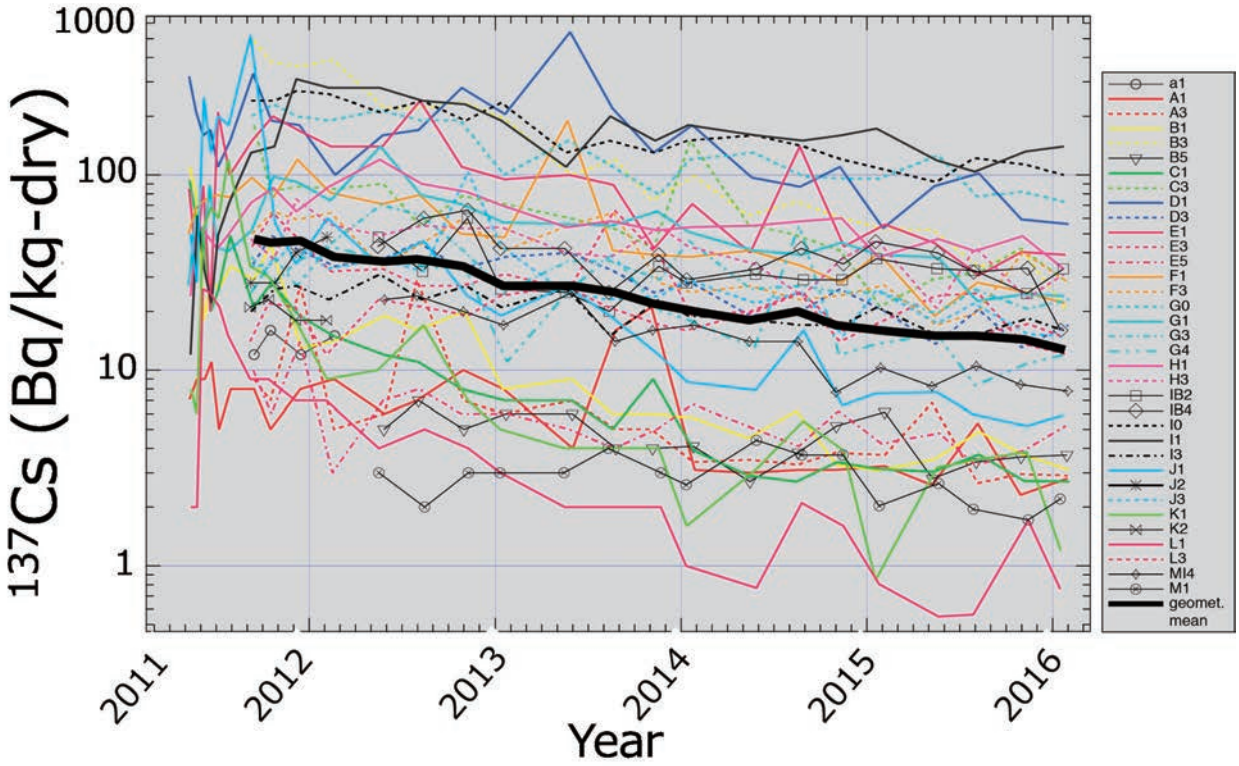

Fig.10 Temporal variations in ^{137}Cs concentrations in sediment samples (0-3 cm depth) collected from the seabed of the marine monitoring area offshore of the FDNPP. Bold (black) line indicates the geometric mean value of all measurement points collected during each sampling cruise.

months after the FDNPP accident, with subsequent sampling conducted at 2- to 3-month intervals. Concentrations of ^{137}Cs were highest at each station in September 2011 (8.0–580 Bq/kg-dry), with almost all samples having concentrations exceeding the 5-year pre-accident average (2.1 Bq/kg) for coastal regions of Japan. However, concentrations declined exponentially during 2011–2016.

Spatiotemporal variations in ^{137}Cs concentrations from May 2011 to January/February 2016 are shown in Figures 9 and 10. Concentrations varied considerably with sampling date over the first half-year, but there was generally much less temporal variation in concentrations. There was no obvious correlation between ^{137}Cs concentration and the proximity of the sampling location to the accident site; for example, the lowest ^{137}Cs concentrations occurred off Miyagi Prefecture in the north and off Chiba Prefecture in the south (stations A1, A3, L1 and L3). Close proximity of a sampling station to FDNPP did not necessarily result in a high ^{137}Cs concentration; between May and July 2011, when sampling was restricted to areas relatively close to the coast, concentrations were higher at the northern stations (e.g., B1, C1 and D1) than at the other stations, and they were also consistently high at stations D1, E1, G0, I0, I1 and J1 (Kusakabe *et al*., 2013). High concentrations (100–200 Bq/kg-dry) were subsequently observed at stations D1, E1, G0, I0 and I1. In contrast, concentrations at station B3, in Sendai Bay, exhibited a decreasing trend, thought to be caused by the lateral transport of surface-layer ^{137}Cs down through the water column and into the bottom sediments, and/or because of bioturbation of sediments (Kusakabe, 2014).

Despite sporadic peaks at each station, the geometric mean of the ^{137}Cs concentration has decreased steadily, from 47 Bq/kg in September 2011 to 13 Bq/kg in January/February 2016. Cs-137 penetrates sediments to depths beyond 3 cm from the sediment surface, and thus may have been transported from the surface layers (Black and Buesseler, 2014; Buesseler, 2014; Otosaka

and Kato, 2014). Desorption/dissolution of ^{137}Cs from sediments is believed to occur at a much lower rate than adsorption does; concentrations would therefore decrease more slowly in sediments than in seawater. For this reason, monitoring of radiocesium levels in marine sediments should remain a top priority.

Acknowledgments

We are grateful to J.F. Zengyoren, the prefectural Federation of Fisheries Co-operative Associations, and the local fishing industry cooperatives for their assistance. We thank the captains and crew of the research vessels of the Sanyo Techno Marine Co. and Kaiyo Engineering Co. Ltd. for their help with sampling. We also thank KANSO Technos, the Kyushu Environmental Evaluation Association, and the Japan Chemical Analysis Center for assistance in seawater radiocesium analyses. The marine environmental radioactivity survey is part of a research project supported by the Japanese Ministry of Education, Culture, Sports, Science, and Technology (May 2011–March 2013) and the Secretariat of the Nuclear Regulation Authority (April 2013–present).

References

Aoyama, M., Kajino, M., Tanaka, T. Y., Sekiyama, T.T., Tsumune, D., Tsubono, T., Hamajima, Y., Inomata, Y. and Gamo, T. (2016). ^{134}Cs and ^{137}Cs in the North Pacific Ocean derived from the TEPCO Fukushima Dai-ichi Nuclear Power Plant accident, Japan in March 2011. II: Estimation of ^{134}Cs and ^{137}Cs inventories in the North Pacific Ocean. *J. Oceanography*, **72**, 67-76.

Black, E.E. and Buesseler, K.O. (2014). Spatial variability and the fate of cesium in coastal sediments near Fukushima, Japan. *Biogeosciences*, **11**, 5123-5137.

Buesseler, K.O. (2014). Fukushima and ocean radioactivity. *Oceanography*, **27**, 92-105.

Isono, R.S., Takata, H., Yamada, H., Inatomi, N., Watabe, T., Suzuki, C., Misonoo, J., Morizono, S. and Kusakabe, M. (2015a). ^{137}Cs and ^{90}Sr concentration changes in seawater and bottom sediments collected from 2011 to 2013 around Japan after the Fukushima Dai-ichi Nuclear Power Plant accident. *Rep. Mar. Ecol. Res. Inst*., **20**, 57-65.

Isono, R.S., Inatomi, N., Takata, H., Hasegawa, K., Ikenoue, T., Watanabe, Y., Yamada, H., Watabe, T., Suzuki, C., Misonoo, J., Morizono, S. and Kusakabe, M. (2015b). ^{137}Cs and ^{90}Sr concentration changes in seawater and bottom sediments collected from 2011 to 2014 around Japan after the Fukushima Dai-ichi Nuclear Power Plant accident. *Rep. Mar. Ecol. Res. Inst*., **21**, 23-32.

Kusakabe, M., Oikawa, S., Takata, H. and Misonoo, J. (2013). Spatiotemporal distributions of Fukushima-derived radionuclides in nearby marine surface sediments. *Biogeosciences*, **10**, 5019-5030.

Kusakabe, M. (2014). Temporal change of radionuclides in seawater and bottom sediments in the waters off Fukushima and nearby Prefectures. *Aquabiology*, **36**, 277-282.

MEXT (Ministry of Education, Culture, Sports, Science and Technology)(1976). Radioactivity measurement series No. 3, available at: http://www.kankyo-hoshano.go.jp/series/lib/No3.pdf (accessed September 17, 2016), (in Japanese).

MEXT(1992). Radioactivity measurement series No. 7, available at: http://www.kankyo-hoshano.go.jp/series/lib/No7.pdf (accessed September 17, 2016), (in Japanese).

Nuclear Regulatory Agency (NRA). Database of environmental radiation. http://search.kankyo-hoshano.go.jp/servlet/search.top (accessed September 17, 2016).

Oikawa, S., Takata, H., Watabe, T., Misonoo, J. and M. Kusakabe (2013a). Distribution of the Fukushima-derived radionuclides in seawater in the Pacific off the coast of Miyagi, Fukushima, and Ibaraki Prefectures, Japan. *Biogeosciences*, **10**, 5031-5047.

Oikawa, S., Takata, H., Isoyama, N., Inatomi, N., Watabe, T., Suzuki, C., Misonoo, J., Morizono, S. and Kusakabe, M. (2014). Temporal changes of ^{90}Sr and ^{137}Cs concentration in the waters around Japanese archipelago from 1983 to 2012. *Rep. Mar. Ecol. Res. Inst*., **19**, 1-15.

Oikawa, S., Watabe, T., Takata, H., Suzuki,C., Nakahara, M. and Misonoo, J. (2013b). Long term temporal changes of ^{90}Sr and ^{137}Cs in seawater, bottom sediment and marine organism samples. From the Chernobyl accident to immediately after the Fukushima accident. *Bunseki Kagaku*, **62**, 455-474.

Otosaka, S. and Kato, Y. (2014). Radiocesium derived from the Fukushima Dai-ichi Nuclear Power Plant accident in seabed sediments: initial deposition and inventories.

Environ. Sci. Process. Impacts, **16**, 978–990.

Takada, K. and Suzuki, Y. (2003). Long-term trends of ^{137}Cs concentrations in marine biota, sediments and seawater in the coastal seas in the vicinity of nuclear power plants in Japan. Jap. *J. Health Phys*., **38**,128-139.

Takata, H. (2016). The contribution of desorption to riverine radiocesium flux to the marine environment. *Aquabiology*, **38**, 315-320.

Takata, H., Hasegawa, K., Oikawa, S., Kudo, N., Ikenoue, T., Isono, R.S. and Kusakabe, M. (2015). Remobilization of radiocesium on riverine particles in seawater: The contribution of desorption to the export flux to the marine environment. *Mar. Chem*., **176**, 51-63.

Takata, H., Kaeriyama, H., Kitamura, M., Yokota, M., Ikenoue, T. and Kusakabe, M. (2015). Spatial and temporal change of radiocesium in zooplankton in the marine environment after the Fukushima Dai-ichi Nuclear Power Plant accident. *Aquabiology*, **37**, 310-318.

Takata, H., Kusakabe, M., Inatomi, N., Ikenoue, T. and Hasegawa, K. (2016). The contribution of sources to the sustained elevated inventory of ^{137}Cs in offshore waters east of Japan after the Fukushima Dai-ichi Nuclear Power Station accident. *Environ. Sci. Technol*., **50**, 6957-6963.

Takata, H., Oikawa, S., Watabe, T., Kikkawa, T., Misonoo, J. and Kusakabe, M. (2013). Temporal change of radioactivity in the waters off Fukushima Prefecture. *Aquabiology*, **36**, 269-282.

Watabe, T., Oikawa, S., Isoyama, N., Suzuki, C., Misonoo, J. and Morizono, S. (2013). Spatiotemporal distribution of ^{137}Cs in the sea surrounding Japanese Islands in the decades before the disaster at the Fukushima Dai-ichi Nuclear Power Plant in 2011. *Sci. Total Environ*., **463-464**, 913-921.