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Impact of Fukushima-derived radiocesium in the western North Pacific Ocean about ten months after the Fukushima Dai-ichi nuclear power plant accident

Yuichiro Kumamoto [a, *], Michio Aoyama [b], Yasunori Hamajima [c], Akihiko Murata [a], Takeshi Kawano [a]

[a] Research and Development Center for Global Change, Japan Agency for Marine-Earth Science and Technology, 2-15 Natushima-cho, Yokosuka, Kanagawa 237-0061, Japan
[b] Institute of Environmental Radioactivity, Fukushima University, 1-1 Kanayagawa, Fukushima 960-1296, Japan
[c] Low Level Radioactivity Laboratory, Kanazawa University, Wake, Nomi, Ishikawa 923-1224, Japan

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ABSTRACT

We measured vertical distributions of radiocesium (^{134}Cs and ^{137}Cs) at stations along the 149°E meridian in the western North Pacific during winter 2012, about ten months after the Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident. The Fukushima-derived ^{134}Cs activity concentration and water-column inventory were largest in the transition region between 35 and 40°N approximately due to the directed discharge of the contaminated water from the FNPP1. The bomb-derived ^{137}Cs activity concentration just before the FNPP1 accident was derived from the excess ^{137}Cs activity concentration relative to the ^{134}Cs activity concentration. The water-column inventory of the bomb-derived ^{137}Cs was largest in the subtropical region south of 35°N, which implies that the Fukushima-derived ^{134}Cs will also be transported from the transition region to the subtropical region in the coming decades. Mean values of the water-column inventories decay-corrected for the Fukushima-derived ^{134}Cs and the bomb-derived ^{137}Cs were estimated to be 1020 ± 80 and 820 ± 120 Bq m^{-2}, respectively, suggesting that in winter 2012 the impact of the FNPP1 accident in the western North Pacific Ocean was nearly the same as that of nuclear weapons testing. Relationship between the water-column inventory and the activity concentration in surface water for the radiocesium is essential information for future evaluation of the total amount of Fukushima-derived radiocesium released into the North Pacific Ocean.

1. Introduction

The massive Tohoku earthquake and consequent giant tsunamis on 11 March 2011 resulted in serious damage to the Fukushima Dai-ichi Nuclear Power Plant (FNPP1) in eastern Japan (Prime Minister of Japan and His Cabinet, 2011). Radiocesium (^{134}Cs and ^{137}Cs) derived from the damaged FNPP1 caused radioactive contamination of the islands of Japan and the North Pacific Ocean (Yoshida and Kanda, 2012). Most of the Fukushima-derived radiocesium deposited on the Japanese islands has remained in the soil, and measurement of radiocesium activity in the soil (Nuclear Regulation Authority, 2012) has revealed that the radioactivities of ^{134}Cs and ^{137}Cs released from the FNPP1 were approximately equivalent. The estimated total amount of ^{137}Cs (or ^{134}Cs) activity in the islands of Japan is 2.4 PBq (10^{15} Bq), based on soil measurements and the air dose rate derived from airborne monitoring (Morino et al., 2013). In contrast, radiocesium released into the North Pacific Ocean was immediately diluted by water advection and diffusion. As a result, it has been difficult to collect enough seawater samples for a quantitative discussion of the total amount of Fukushima-derived radiocesium in the vast ocean.

The Fukushima-derived radiocesium were released into the North Pacific, mostly in March and April 2011, through two major pathways: direct discharge of radioactive water and atmospheric deposition. In late March 2011, the Tokyo Electric Power Company and the Japanese Government began marine monitoring in the coastal area within a radius of about 30 km from the FNPP1 (Nuclear Regulation Authority, 2011; Oikawa et al., 2013; Tokyo

* Corresponding author. Tel.: +81 46 867 9505; fax: +81 46 867 9835.
E-mail address: kumamoto@jamstec.go.jp (Y. Kumamoto).

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Electric Power Company, 2011). The decay-corrected $^{134}Cs/^{137}Cs$ ratio in those monitoring data was almost 1 and in agreement with the ratio in standing water in the damaged FNPP1 (Nishihara et al., 2012). Thus, the activity concentrations of ^{134}Cs and ^{137}Cs in the radioactive water directly discharged from the FNPP1 were also approximately equivalent. These measurements have facilitated evaluation of the total amount of ^{137}Cs (or ^{134}Cs) derived from the directly discharged radioactive water. Many estimations were in the range between 2 and 6 PBq (Estournel et al., 2012; Kawamura et al., 2011; Miyazawa et al., 2013; Nair et al., 2014; Prime Minister of Japan and His Cabinet, 2011; Tsumune et al., 2012). However, temporal or spatial extrapolating in the calculation resulted in a larger amount ranged from 11 to 27 PBq (Bailly du Bois et al., 2012; Charette et al., 2013; Rypina et al., 2013).

Efforts to obtain data of Fukushima-derived radiocesium in the open ocean have continued, and radiocesium measurements made in both surface water (Aoyama et al., 2013; Honda et al., 2012; Kaeriyama et al., 2013; Kameník et al., 2013; Karasev, 2012) and the ocean interior (Buesseler et al., 2012; Kaeriyama et al., 2014; Kumamoto et al., 2014; Povinec et al., 2013; Ramzaev et al., 2014) have been reported. These data obtained in the open ocean have revealed that the Fukushima-derived radiocesium released into the North Pacific Ocean has been transported eastward by surface currents and conveyed southward across the Kuroshio Extension current in subsurface layers. However, the available data are still insufficient for evaluation of the total amount of radiocesium, including that from atmospheric deposition, in the vast North Pacific Ocean. Atmospheric deposition has been estimated by using atmospheric model simulations (Estournel et al., 2012; Kawamura et al., 2011; Kobayashi et al., 2013; Miyazawa et al., 2013; Morino et al., 2011, 2013; Stohl et al., 2012). The results of these model simulations consistently show that a large portion of the radiocesium released to the atmosphere from the FNPP1 was subsequently deposited on the North Pacific Ocean. However, the simulated total amount of ^{137}Cs (or ^{134}Cs) deposited on the ocean ranges widely, from 5 to 30 PBq.

The two major isotopes in the Fukushima-derived radiocesium are ^{134}Cs and ^{137}Cs. These radiocesium isotopes were also released into the environment before the FNPP1 accident by atmospheric nuclear weapons testing, mainly in the 1950s and 1960s; nuclear fuel reprocessing, mainly in the 1980s; and the Chernobyl accident in 1986. In the North Pacific Ocean the major source of radiocesium was atmospheric deposition due to the nuclear weapons testing (Aoyama et al., 2006). The bomb-derived ^{137}Cs released into the North Pacific was still there just before the accident because its half-life is long, 30.04 y. In the 2000s, ^{137}Cs activity concentration of 1.0–2.5 Bq m^{-3} was widely observed in surface seawaters of the North Pacific Ocean (Aoyama et al., 2012a). After the FNPP1 accident, the Fukushima-derived ^{137}Cs was of course added to the bomb-derived ^{137}Cs. In contrast, the ^{134}Cs released before the accident had disappeared, because its half-life is only 2.07 y, which indicates that ^{134}Cs can be used as a tracer of radiocesium from the FNPP1 accident. The Fukushima-derived radiocesium measurements in the past suggested that the activities of ^{134}Cs and ^{137}Cs released from the FNPP1 were equivalent. Thereby, after the decay-correction, the excess ^{137}Cs activity concentration relative to the ^{134}Cs activity concentration in seawaters collected after the FNPP1 accident represents the bomb-derived ^{137}Cs activity concentration. By measuring both ^{134}Cs and ^{137}Cs activity concentrations, both the Fukushima-derived and the bomb-derived ^{137}Cs activity concentrations can be determined simultaneously. A few studies have already calculated the bomb-derived ^{137}Cs activity concentrations in seawaters of the North Pacific Ocean in this way (Inoue et al., 2012; Kameník et al., 2013; Ramzaev et al., 2014). However, they did not compare the activity concentrations and inventories of the Fukushima-derived and bomb-derived ^{137}Cs quantitatively.

Here we report the vertical distributions of ^{134}Cs and ^{137}Cs activity concentrations at stations along 149 °E between approximately 42 °N and 4 °S in winter 2012, about ten months after the FNPP1 accident. We preliminarily reported the distribution and behavior of the Fukushima-derived ^{134}Cs only prior to the bomb-derived ^{137}Cs along this meridional line between 42 °N and 10 °N (Kumamoto et al., 2014). In the preliminary report, we found that activity concentration of the Fukushima-derived ^{134}Cs in surface water was highest in the transition region between the subarctic and Kuroshio Extension fronts (approximately 35–40 °N), where the FNPP1 is situated (Fig. 1). In addition, a subsurface maximum of the Fukushima-derived ^{134}Cs was found in the subtropical region south of the Kuroshio Extension Current, and southward transportation of the Fukushima-derived ^{134}Cs was evaluated. In this report, we (1) additionally publish ^{134}Cs and ^{137}Cs activity concentrations observed between 10 °N and 4 °S, which was not published in the previous report, (2) compare them between 42 °N and 4 °S to estimate the bomb-derived ^{137}Cs, which was not calculated in the previous report, and (3) discuss the impact of the Fukushima-derived radiocesium on the bomb-derived radiocesium, which had existed since before the FNPP1 accident, in the western North Pacific in winter 2012. In addition, we evaluated the relationship between the activity concentration in surface water and the water-column inventory of the radiocesium. These results will contribute to the eventual determination of the total amount of the Fukushima-derived radiocesium in the North Pacific Ocean.

2. Samples and experimental procedures

Seawater samples for radiocesium measurements (about 20 dm^3 each) were collected during cruise MR11-08 of the Research Vessel *Mirai* from December 2011 to February 2012. This cruise also performed repeat hydrography along one of the observation lines of the World Ocean Circulation Experiment (WOCE) in the western Pacific Ocean (the WOCE-P10/P10N line), which approximately follows the 149 °E meridian. We collected surface seawater using a bucket at 38 stations along the line between 42 °N and 4 °S (Fig. 1). Along this line, the area north of the subarctic front (approximately 40 °N) is defined as the subarctic region, that south of the Kuroshio Extension front (approximately 35 °N) is defined as the subarctic, and the area between the two fronts, in which the FNPP1 is also situated, is designated the transition region (Fig. 1) (Kumamoto et al., 2014). Although the boundary between the subtropical and tropical regions is not clear, the area south of the North Equatorial current (approximately 12 °N) is provisionally regarded as the tropical region. At 18 of the 38 stations, seawater samples from the surface to 800 m depth were collected using 12-Liter polyvinyl chloride bottles (Model 1010X NISKIN-X, General Oceanics Inc.). The seawater was filtered through a 0.45 μm pore size membrane filter (HAWP14250, Millipore) and acidified on board ship by adding 40 cm^3 of concentrated nitric acid (Nitric Acid 70% AR, RCI Labscan, Ltd.) within 24 h after sampling. After the cruise, radiocesium in the seawater sample was concentrated on ammonium phosphomolybdate (Aoyama and Hirose, 2008) in onshore laboratories for measurement of gamma-ray activity.

The radiocesium activity of the ammonium phosphomolybdate was measured in a laboratory of the Mutsu Oceanographic Institute at the Japan Agency for Marine-Earth Science and Technology (MIO/JAMSTEC) or the Low Level Radioactivity Laboratory at Kanazawa University (LLRL/KU). In the MIO/JAMSTEC laboratory, the radiocesium was measured with low-background Ge-detectors (Well-type GCW2022-7915-30-ULB, Canberra Industries, Inc.) that were calibrated with gamma-ray volume sources (Eckert & Ziegler Isotope Products) certified by Deutscher Kalibrierdienst. The averages of the analytical uncertainties (standard deviations) for the