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Radiocesium in the western subarctic area of the North Pacific Ocean, Bering Sea, and Arctic Ocean in 2013 and 2014

Yuichiro Kumamoto[a,*], Michio Aoyama[b], Yasunori Hamajima[c], Shigeto Nishino[d], Akihiko Murata[a], Takashi Kikuchi[d]

[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, Fukushima 960-1296, Japan
[c] Low Level Radioactivity Laboratory, Kanazawa University, Wake, Nomi, Ishikawa 923-1224, Japan
[d] Institute of Arctic Climate and Environment Research, Japan Agency for Marine-Earth Science and Technology, 2-15 Natushima-cho, Yokosuka, Kanagawa 237-0061, Japan

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ABSTRACT

We measured radiocesium (^{134}Cs and ^{137}Cs) in seawater from the western subarctic area of the North Pacific Ocean, Bering Sea, and Arctic Ocean in 2013 and 2014. Fukushima-derived ^{134}Cs in surface seawater was observed in the western subarctic area and Bering Sea but not in the Arctic Ocean. Vertical profile of ^{134}Cs in the Canada Basin of the Arctic Ocean implies that Fukushima-derived ^{134}Cs intruded into the basin from the Bering Sea through subsurface (150 m depth) in 2014.

1. Introduction

The massive Tohoku earthquake and consequent giant tsunami on March 11, 2011 resulted in release of radiocesium (^{134}Cs and ^{137}Cs) into the North Pacific Ocean from the Fukushima Dai-ichi nuclear power plant, FNPP1 (Yoshida and Kanda, 2012). Evaluation of Fukushima-derived ^{137}Cs (half-life =30.04 y) in the marine environment is necessary to address risks to marine ecosystem and public health because of its relative long residence time in the ocean. In fact ^{137}Cs derived from the nuclear weapon tests mostly in the 1950s and 1960s (bomb-produced ^{137}Cs) was still observed in surface seawater of the North Pacific Ocean to be 1 – 2 Bq m^{-3} just before the FNPP1 accident (Aoyama et al., 2012), which is now masked by Fukushima-derived ^{137}Cs. On the other hand, the bomb-produced ^{134}Cs had decayed to undetectable levels due to its short half-life (2.07 y) by March 2011. Therefore ^{134}Cs is a unique tracer for Fukushima-derived radiocesium. Most of ^{134}Cs/^{137}Cs ratios, which were corrected to the date of the FNPP1 accident for radioactive decay, in soil collected near FNPP1 were about 1 (Saito et al., 2015), suggesting that total amounts of ^{134}Cs and ^{137}Cs released from FNPP1 were equivalent.

During the past five years Fukushima-derived radiocesium was measured in seawater samples collected in the whole area of North Pacific Ocean. Efforts for the measurements just after the accident, in April and May 2011, achieved success in evaluation of the total amount of radiocesium in the North Pacific Ocean. The total amount of ^{137}Cs (or ^{134}Cs) derived from direct discharge of contaminated water and atmospheric deposition in the basin were estimated to be about 3.5 (Tsumune et al., 2012) and 12–15 (Aoyama et al., 2016a) PBq (10^{15} Bq), respectively. In addition, radiocesium measurements along cross-sectional line across the basin revealed horizontal and vertical spreading directions of Fukushima-derived radiocesium. The directly-discharged radiocesium has been transported eastward along surface current in surface mixing layer in the north of the Kuroshio Front (Aoyama et al., 2013; Kumamoto et al., 2016). In the south of the front, subtropical area, Fukushima-derived radiocesium deposited has been transported southward through subsurface layer due to subduction of the subtropical mode water (Kaeriyama et al., 2014; Kumamoto et al., 2014; Yoshida et al., 2015; Aoyama et al., 2016a).

In the Bering Sea, a northern marginal sea adjacent to the North Pacific Ocean, Fukushima-derived ^{134}Cs was observed in surface seawater in summer 2012 (Kumamoto et al., 2016). In the Arctic Ocean, however, Fukushima-derived ^{134}Cs was not detected in 2012 (Smith et al., 2015; Kumamoto et al., 2016). These results suggest that Fukushima-derived ^{134}Cs deposited on the Bering Sea but not on the Arctic Ocean and had not been transported from the Bering Sea to Arctic Ocean by summer 2012, about 1.5 years after the accident. Here we present results of radiocesium measurements in seawater from the western subarctic area of the North Pacific Ocean, Bering Sea, and

* Corresponding author.
E-mail address: kumamoto@jamstec.go.jp (Y. Kumamoto).

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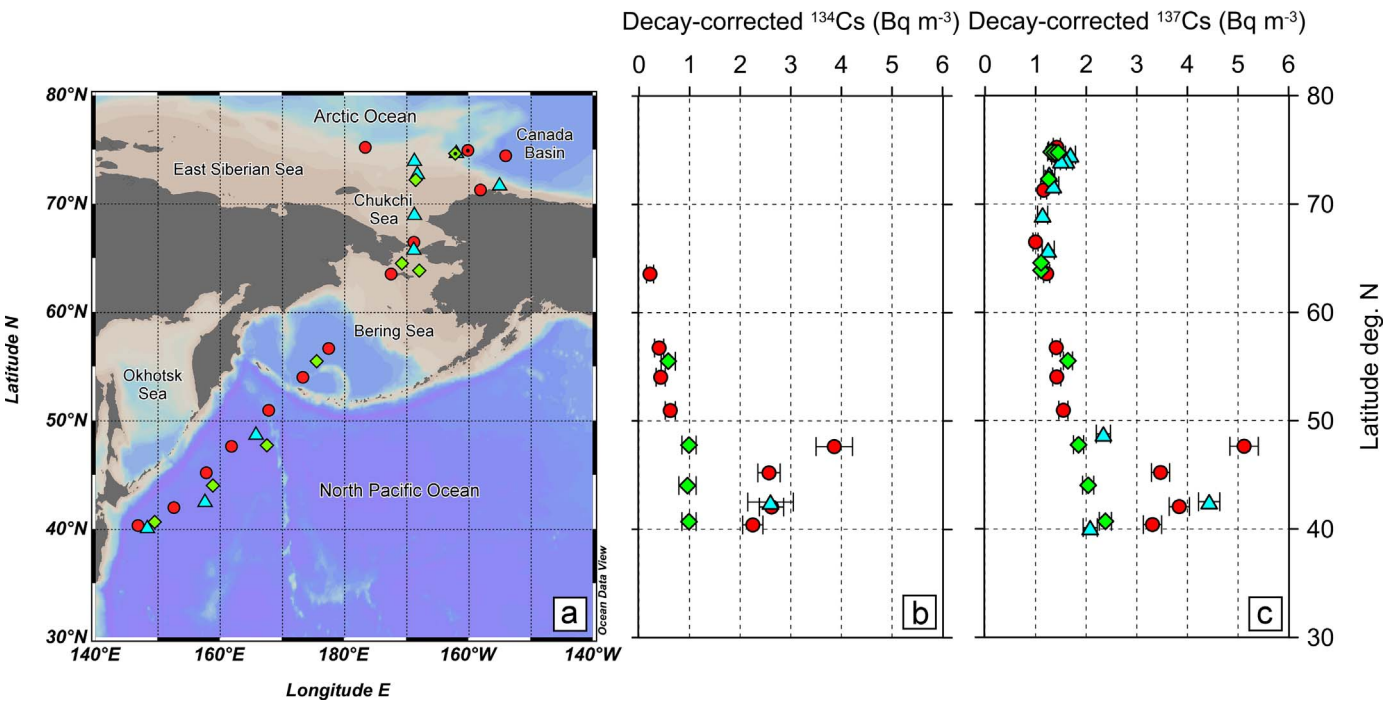

Fig. 1. (a) Sampling stations of surface seawater for radiocesium measurement during MR13-06 (triangles, September-October 2013) and MR14-05 (diamonds, September-October 2014) cruises. To compare with these data, sampling stations and radiocesium data obtained during MR12-E03 (Kumamoto et al., 2016) are also shown in this figure (circles, September-October 2012). Symbols with a small dot in the Canada Basin of the Arctic Ocean indicate stations where seawater samples were also collected from 25 to 800 m depth. A map in this figure was drawn using Ocean Data View software (Schlitzer, 2016). (b) Activity concentration of ^{134}Cs decay-corrected to the date of FNPP1 accident, 11 March 2011 (Bq m^{-3}) in surface seawater. Symbols are same as those in (a). (c) Same as (b) but for ^{137}Cs.

Arctic Ocean in 2013 and 2014 and discuss transportation of Fukushima-derived radiocesium from the Bering Sea to Arctic Ocean during about 3.5 years after the accident.

2. Methods

Seawater samples for radiocesium measurement were collected during cruises of research vessel "MIRAI", MR13-06 (September-October 2013) and MR14-05 (September-October 2014). Fig. 1a shows location of sampling stations. Surface seawater were sampled using a 10-L bucket (0 m depth) or pump (about 4 m depth). Temperature and salinity in surface water in the bucket were measured using a calibrated mercurial thermometer and the salinometer (Autosal model 8400, Guildline Instruments), respectively. Water temperature and salinity of the pumped-up surface waters were estimated from data from a CTD system (SBE-11plus, Sea-Bird Electronics, Inc.). The salinity sensor on the CTD was calibrated using results of the bottle salinity measurement. Deeper seawater from 25 m to 800 m depths were collected using 12-Liter Niskin-X bottles (General Oceanics Inc.) equipped with another C TD system (SBE-11plus, Sea-Bird Electronics, Inc.) calibrated using the salinometer. We collected about 20 L of seawater from each depth. Unfiltered seawater was acidified to pH 1.6 by 40 cm^3 of concentrated nitric acid after the sampling.

After the cruises, in laboratories of Mutsu Institute for Oceanography / Japan Agency for Marine-Earth Science and Technology (MIO/JAMSTEC) or Japan Marine Science Foundation, radiocesium in the seawater sample was concentrated using an improved ammonium phosphomolybdate (AMP) method (Aoyama and Hirose, 2008). Radiocesium was quantitatively separated from seawater by coprecipitation with AMP/Cs compound by adding 4 g of AMP and a pipetted aliquot of CsCl solution containing 0.26 g cesium as carrier. Yield of AMP/Cs compound was more than 98%. Radiocesium activity in AMP/Cs samples from the MR13-06 and MR14-05 cruises were measured at MIO/JAMSTEC and Low Level Radioactivity Laboratory / Kanazawa University (LLRL/KU), respectively, using low background Ge-detectors, which were calibrated with gamma-ray volume sources (Eckert & Ziegler Isotope Products) certificated by Deutscher Kalibrierdienst (DKD). ^{134}Cs and ^{137}Cs activities were evaluated from gamma-ray peaks at 605/796 and 661 keV, respectively. Averaged detection limits of the ^{134}Cs and ^{137}Cs measurements at MIO/JAMSTEC for the MR13-06 samples were about 0.5 and 0.2 Bq m^{-3}, respectively. Because the Ge-detectors of LLRL/KU were placed in an underground laboratory (Hamajima and Komura, 2004), averaged detection limit of the ^{134}Cs and ^{137}Cs at LLRL/KU for the MR14-05 samples were about 0.1 and 0.05 Bq m^{-3}, respectively. In order to compare these data directly all the radiocesium activity concentration were decay-corrected to the date of FNPP1 accident, 11 March 2011. Averaged detection limits of the decay-corrected ^{134}Cs and ^{137}Cs measured at MIO/JAMSTEC (LLRL/KU) were about 1.2 and 0.5 Bq m^{-3} (0.4 and 0.2 Bq m^{-3}), respectively.

3. Results

All the radiocesium, temperature, and salinity data obtained in 2013 and 2014 are listed in a supplementary table. In order to discuss temporal change we also show radiocesium data obtained in 2012 (Kumamoto et al., 2016) in areas where we measured radiocesium activity concentration in 2013 and 2014 (Fig. 1).

Fukushima-derived ^{134}Cs in surface water was observed in the subarctic area and Bering Sea in 2013 and 2014 (Fig. 1b). The decay-corrected activity concentration at 42.5°N in the subarctic area in 2013 (2.6 Bq m^{-3}) was close to those observed in 2012. The activity concentrations at the other two stations in the subarctic area in 2013 were below the detection limit (about 1.2 Bq m^{-3} in decay-corrected activity concentration). In contrast, decay-corrected activity concentrations of ^{137}Cs at these stations in 2013 were in same range as those at nearby stations in 2014 (2.0 – 2.5 Bq m^{-3}), where activity concentration of ^{134}Cs was about 1.0 Bq m^{-3} in 2014 (Fig. 1c). Therefore decay-corrected activity concentration of ^{134}Cs in the subarctic area in 2013 and 2014 was probably higher than 1.0 Bq m^{-3}. ^{134}Cs was also