Temporal variability of Fukushima-derived radiocesium and its balance at pelagic time-series stations in the western North Pacific

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1. Introduction
On 11 March 2011, the 2011 Tohoku-Oki Earthquake occurred off Miyagi Prefecture, Japan. This earthquake and the tsunami seriously damaged the Fukushima Daiichi Nuclear Power Plant (FNPP1). As a result, large quantities of artificial radionuclides were emitted from FNPP1 by hydrogen explosions, venting, and intentional and accidental discharge of contaminated water. At a pelagic time-series station in the western North Pacific, K2 (47N / 160E, water depth: ca. 5200 m), the transport and distribution of radiocesium, which was derived from the FNPP1 accident, in seawater, zooplankton, sinking particle and seafloor sediment had been observed since the accident. Principal results are as follows:

2. Results and discussion
(1) Transport of radiocesium to K2
In April, 2011, about one month after the accident, radiocesium of sea surface water in the western North Pacific (WNP) was measured. Fukushima-derived radiocesium was detected in broad area of the WNP including K2, which is about 2,000 km from the FNPP1 (Fig.1a). Its mean concentration (~0.048 Bq kg⁻¹) became about 50 times higher than that just before the accident (~0.001 Bq kg⁻¹), hereinafter: background level. Based on the mathematical simulation, Fukushima-derived radiocesium likely arrived at K2 within a week after the accident as an eolian dust (Fig.1b) rather than contaminated water.

(2) Temporal variability in radiocesium
1) Seawater and zooplankton
Concentrations of radiocesium (137Cs) in the surface seawater were at K2 was ca. 0.008 Bq kg⁻¹ in April 2011 (Fig.2). It is suspected that its concentration was ca. 8 times higher than the background level (Nagaya and Nakamura, 1987). After 137Cs increased by ca. 0.018 Bq kg⁻¹ in July 2012, 137Cs decreased with elapsed time (Fig.2). In July 2013, 137Cs was 0.002 Bq kg⁻¹ and close to the background level. Concentrations of 137Cs in zooplankton upper 200 m ranged ca. 13-60 Bq kg⁻¹ in dry weight (dw) in April 2011 (Fig.2). Its concentration was two orders higher than the background level observed off the east coast of Tohoku district (0.09-0.47 Bq kg⁻¹, Kaeriyama et al., 2008). 137Cs concentration decreased with time and 137Cs concentration after summer 2012 was relatively stable around 1.5 Bq kg⁻¹ (dw). The ratio of 137Cs of zooplankton (dw) to that of seawater in summer 2013 ranged from 500 to 1000. Assuming that water contents of zooplankton is 90% (Honda et al., 2012), the ratio of 137Cs of zooplankton in wet weight to that of seawater become one order smaller than that of the above ratio. These are comparable to concentration factor (CF) reported previously (e.g. IAEA, 2004).

2) Sinking particles (sinking velocity)
Fukushima-derived 137Cs was detected at the first time from sinking particles collected between 26 March and April 6 2011 at 500 m and 18 April 2011 at 4810 m (Fig. 3a, b). Concentration of 137Cs of sinking particle ranged from ca. 20 to 400 Bq kg⁻¹ with average of ca. 80 Bq kg⁻¹ (Fig.3c). Maximum concentration was observed in late March 2011 at 500 m while in early June 2011 at 4810 m. Concentrations of radiocesium was below detection limit after February 2012 at 500 m and middle April 2012 at 4810 m. According to the time lag of first detection period between 500 m and 4810 m, sinking velocity of 137Cs was estimated to be larger than ca. 180 m day⁻¹ in previous report (Honda et al., 2013).

3) Removal rate and residence time
Total 137Cs flux (137Cs inventory) at 500 m and 4810 m for about one year was 2.25 and 2.61 Bq m⁻², respectively. Taking into account for the difference in trapping efficiency between sediment traps at two depths, 137Cs inventories were comparable. The input of eolian 134Cs estimated with 137Cs concentrations in surface seawater and at the mixed layer depth in April 2011 was ca. 450 Bq m⁻². Thus removal rate of 137Cs from surface layer are estimated to be 0.5 ~ 0.6% yr⁻¹ and, thus, residence time of 137Cs was estimated to be 170 ~ 200 years. It is also concluded that most of radiocesium is dissolved in the ocean. This result coincided well with the previous reports (e.g. Buesseler et al., 1990).

4) Inventory of radiocesium in seafloor sediment: lateral transport of radiocesium (?)
Seafloor sediment at K2 (water depth: ca. 5200 m) was collected by multiple core sampler in July 2012. 137Cs was detected in only surface 5 mm layer. Using the bulk density, 137Cs inventory in seafloor sediment was estimated to be ca. 20 Bq m⁻². This inventory is several times higher than that observed at 500 m and 4810 m sediment trap (~2.5 Bq m⁻²). It might be indicative of that radiocesium was transported laterally near seafloor (between 4810 m and seafloor). It is noted that 137Cs was not detected from another core sample. It is reported that radiocesium was ubiquitous in the seafloor sediment at least near Fukushima (e.g. Kusakabe et al., 2013). Therefore, there is possibility that detected 137Cs (Fig.4) is not representing the inventory of the seafloor at K2.

Fig.1 (a) Horizontal distribution of 137Cs in surface seawater in April 2011 (b) Simulated accumulated deposition of eolian 137Cs at 13 April 2011 (Honda et al., 2012)

Fig.2 Time-series variability in 137Cs of zooplankton (ZP; triangles) and seawater (SW; circles) from surface (-S) and subsurface (-SS) layers (Kitamura et al., in preparation; Fukuda et al., in preparation). 137Cs of ZP and SW before the accident (BA) are also shown.

Fig.3 137Cs flux at (a) 500m and (b) 4810 m, and (c) 137Cs concentrations at both depths (open circles: 500m, closed circles: 4810 m). Arrows show the 11 March 2011 when the 2011 Tohoku-Oki Great Earthquake occurred (Honda and Kawakami, 2014)

Fig.4 Vertical profile of 134Cs concentration in seafloor sediment at K2.

References