Chapter 9
Radiocesium Concentration of Small Epipelagic Fishes (Sardine and Japanese Anchovy) off the Kashima-Boso Area

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Abstract After the Fukushima Dai-ichi Nuclear Power Plant (FNPP) accident, which occurred in March of 2011, the National Research Institute of Fisheries Science (NRIFS) undertook emergent radioactivity monitoring of 63 samples of small epipelagic fishes (such as sardine and Japanese anchovy) collected by commercial fishery boats off the Kashima-Boso area (located to the south of the Fukushima coast) from 24 March 2011 to 21 March 2013. Fluctuations in the radiocesium concentration in fish muscles synchronized with the decreasing concentration from seawater near the fishing ground; the radiocesium concentration in fish muscles reached a maximum of 31 Bq/kg-wet in July 2011, after which it declined gradually. From 2012 to 2013, the radiocesium concentrations in fish muscles were low (0.58–0.63 Bq/kg-wet). Compared to the ^{137}Cs concentration before the FNPP accident, ^{137}Cs concentration in fish muscles in 2013 was still about 10 times higher, whereas it was about 4.5 times higher in seawater near the fishing ground in 2012.

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9.1 Introduction

Artificial radionuclides were released into the environment as a result of the Tokyo Electric Power Company (TEPCO) Fukushima Dai-ichi Nuclear Power Plant (FNPP) accident that occurred in March of 2011. TEPCO (2012) has estimated that radiocesium (^{134}Cs and ^{137}Cs) at approximately 10 PBq for both ^{134}Cs and ^{137}Cs was released into the atmosphere in March 2011. In addition, it was estimated that ^{134}Cs was released into the atmosphere and the ocean from the port of the nuclear power plant from March to September 2011 at a level of 3.5 PBq and that the level of ^{137}Cs was 3.6 PBq. Some of the released radiocesium was taken into the bodies of marine organisms through the surrounding water and their prey, and an investigation into radioactive substances in marine products, conducted by Fisheries Agency (FA), showed that a relatively high radiocesium concentration (compared to the concentration before the FNPP accident) was detected in some of the fish of the northwest Pacific Ocean (Fisheries Agency 2012, 2013). These results indicate that there is a possibility of long-term residual radiocesium in organisms with strong regional characteristics, including bottom fish (Buesseler 2012). The mechanisms of radioactive substance migration in marine ecosystems need to be understood (Yoshida and Kanda 2012), but this would entail an analysis of the radioactive substance transfer mechanism corresponding to ecological characteristics related to each component of the marine ecosystem. The National Research Institute of Fisheries Science (NRIFS), Fisheries Research Agency (FRA) cooperated with the radioactive substance investigation and conducted an intensive survey of marine products caught in the Kanto region for approximately 6 months immediately after the FNPP accident.

Some of the species of fish that were present during the high fishing season off the Pacific Ocean coast after the FNPP accident included small migratory epipelagic fishes, namely, sardine (*Sardinops melanostictus*) and Japanese anchovy (*Engraulis japonicus*). Sardine and Japanese anchovy actively eat plankton, and they are preyed upon by whales and large fishes, which gives them an important ecological niche in the Pacific Ocean coastal areas of the Tohoku region. Each year, from winter/spring to summer, sardine and Japanese anchovy migrate to Kashima-Boso, temporarily remaining in the area to create a fishing ground, and they are fished in large- to medium-scale round-haul fisheries (Uchiyama 1998; Yasumi 2008; Kubota 2012). Thus, this chapter contains an analysis of fluctuations in the radiocesium concentration of these small epipelagic fishes caught in the fishing grounds off the Kashima-Boso area.

9.2 Collection of Fish and Radioactivity Measurement

NRIFS prepared 63 specimens for radioactivity measurement for each fish species for each sampling date for adult sardine and Japanese anchovy caught mainly in a large- to medium-scale round-haul fishery off the Kashima-Boso area (Fig. 9.1)

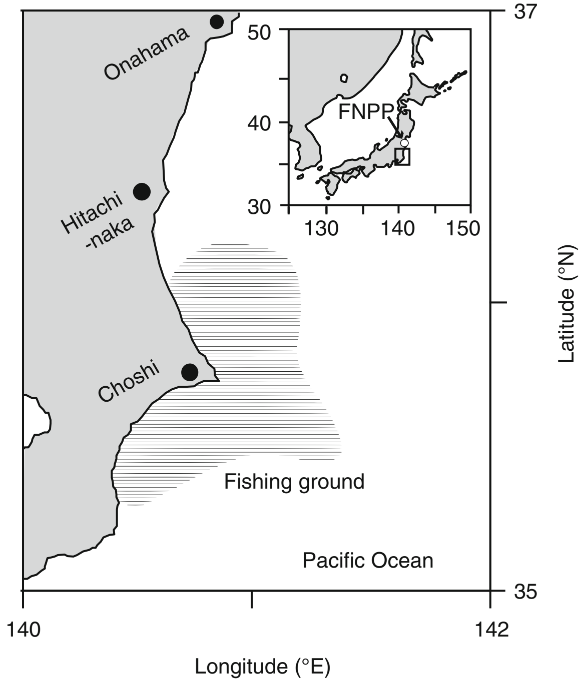

Fig. 9.1 Locations of the Fukushima Dai-ichi Nuclear Power Plant (FNPP), and fishing grounds of fish sampled (sardine and Japanese anchovy) in this chapter (*shaded area*)

from 24 March to 3 November 2011; 27 June and 20 August 2012; and 18 February 2013. Muscles were chosen as the measurement sample for this study. For small epipelagic fishes collected in 2011, 60 raw specimens were prepared, and for small epipelagic fishes collected in 2012 and 2013, 3 ashed specimens were prepared, assuming that the radiocesium concentration would be quite low.

To measure radiocesium concentrations, a germanium semiconductor detector (EG ORTEC Solid-State Photon Detector) and a pulse-height analyzer (SEIKO EG MCA 7600 Multichannel Analyzer) were used. The resolution of the germanium semiconductor detector [full width at half-maximum (FWHM)] was 1.80 keV (^{60}Co, 1,333 keV), and the relative efficiency was 33.0 %. The standard source was the quasi-gamma-ray volume source standard MX033SPS prepared by Japan Radioisotope Association (a special order was placed to obtain source heights of 5, 10, 20, 30, and 50 mm), and the MX033U8PP type prepared by the Association. Nuclides that were objects of measurement were ^{134}Cs (605, 796 keV; without summing the effect correction) and ^{137}Cs (662 keV). For calculation of the targeted nuclide concentrations, we followed the directive 'The gamma-ray spectrometry by germanium semiconductor detector' (Ministry of Education, Culture, Sports, Science and Technology 1992), and we calculated using the Covell method. Sixty specimens of epipelagic fishes collected in 2011 were used as raw samples for 7,200-s measurements, and three specimens of epipelagic fishes collected in 2012 and 2013 were ashed under the assumption that the radiocesium concentration is quite low; they were analyzed by carrying out 40,000-s measurements or longer. Radiocesium data (both ^{134}Cs and ^{137}Cs) were obtained by making attenuation corrections to the sample of small epipelagic fishes for the sampling date.

9.3 Tracking the 2010 Year-Class Fish

We first introduce the distribution ecology based on the fluctuation in catch volumes for both sardine and Japanese anchovy. From March to August 2011, the sardine haul was more than that of Japanese anchovy, and the sardine haul in Chiba Prefecture during this period (66,000 tons) was twice that of Japanese anchovy (33,000 tons) (National Research Institute of Fisheries Science 2011). As for which sardine haul is relatively larger, the main target of the round-haul fishery is the southward migrating group of sardine during winter and spring, but during summer, the target changes to the northward migrating group composed of age 2 fish and older as usual (Fisheries Agency and Fisheries Research Agency 2011). However, in the fishing season of 2011, the 2010 year-class had the highest amount of recruitment to the sardine stock since 2002 (Kawabata et al. 2012). Furthermore, during the fishing seasons from winter/spring to the summer of 2011, the 2010 year-class was widespread in the Boso area. Thus, only a small number of age 2 fish and older (which are older than the 2010 year class) were mixed among the catch (Fisheries Agency and Fisheries Research Agency 2011).

Before preparing the radioactivity measurement samples, ten specimens were randomly selected, and the standard length (SL, mm) was measured to calculate the average length of each sample. The average values of SL of small epipelagic fish samples were 132–200 and 99–124 mm for sardine and Japanese anchovy, respectively (Fig. 9.2). The average length of fishes used as samples generally matched the mode of length composition of the catch in the Joban-Boso area from March to August 2011 (National Research Institute of Fisheries Science 2011). Thus, it was determined that both the sardine and Japanese anchovy used for this study were the results of continuously tracking the temporal fluctuations in radiocesium concentration of age 1 fish (the 2010 year-class) in the surveyed area in the ocean. It was also determined from the average length of the sample that we tracked the 2010 year-class until August 2012, and the February 2013 sample was the result of tracking age 1 fish from the 2012 year-class. Whitebait (Japanese anchovy) of coastal areas off Fukushima Prefecture is known as an example of the turnover of fish schools affecting the fluctuations of radiocesium concentration in fish (Wada et al. 2013), but it was considered to be fluctuation of radiocesium concentration in the 2010 year-class sardine that was tracked during 2011 in this chapter. Subsequently, the main catch for Japanese anchovy during April to June was age 1 fish (the 2010 year-class) (Fisheries Agency and Fisheries Research Agency 2011), similar to sardine.

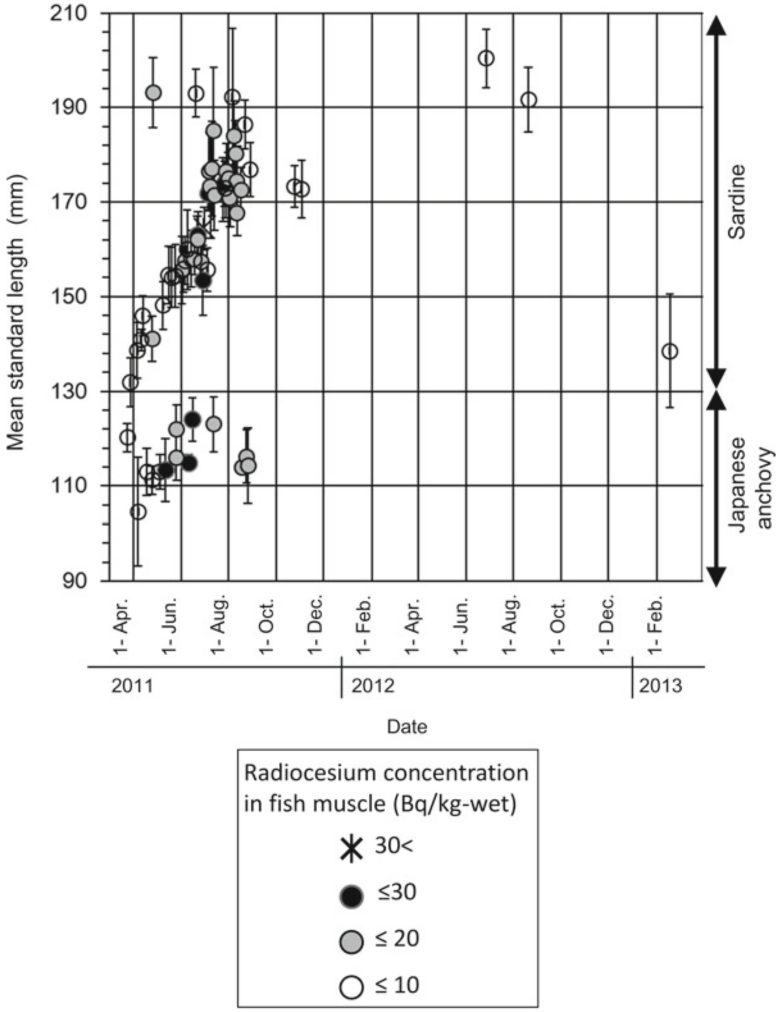

Fig. 9.2 Mean standard length of fish samples (sardine and Japanese anchovy) used in this chapter. *Vertical bar* indicates standard deviation

9.4 Temporal Fluctuations in Radiocesium Concentration of Small Epipelagic Fishes

Because there was no significant difference between radiocesium concentrations of sardine and Japanese anchovy collected during the research period (Fig. 9.3), these two species were considered together as 'small epipelagic fishes' for the analysis. From March to May 2011, the radiocesium concentration in the muscle of small epipelagic fishes exhibited relatively high concentrations on certain occasions, such as 13 Bq/

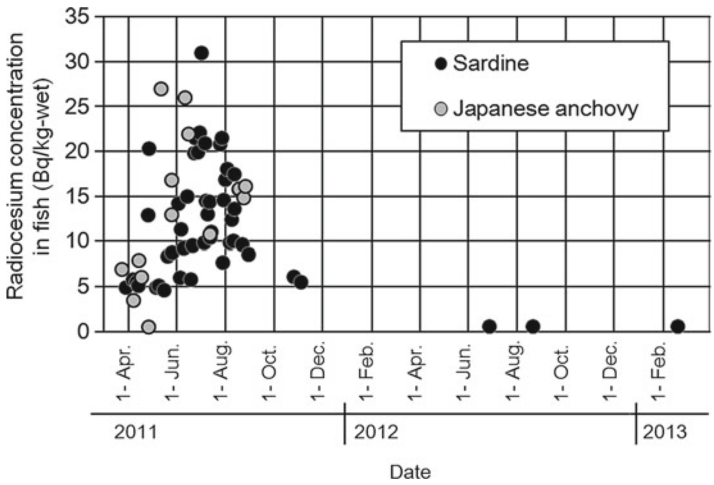

Fig. 9.3 Temporal variations of radiocesium ($^{134}Cs + ^{137}Cs$) concentration in small epipelagic fish (sardine and Japanese anchovy) caught off the Kashima-Boso area

kg-wet on 25 April, 21 Bq/kg-wet on 26 April, and 27 Bq/kg-wet on 12 May; however, the concentrations generally remained at 9 Bq/kg-wet or lower (Fig. 9.3). Concentrations mostly ranged from 9 to 22 Bq/kg-wet from June to August (Fig. 9.3), and these values were significantly higher than the concentrations detected from March to May (Mann–Whitney *U* test, $p<0.01$). After detecting the maximum value of 31 Bq/kg-wet in early July, none of the specimens had a value greater than 20 Bq/kg-wet in August, and 6 and 5.5 Bq/kg-wet were recorded on 25 October and 3 November, respectively, which was after the summer fishing season (Fig. 9.3). Concentrations have continued to decrease since 2012, and levels as low as 0.58–0.63 Bq/kg-wet were detected (Fig. 9.3).

9.5 Decreasing Trend of Radiocesium Concentration of Small Epipelagic Fishes

Measurement results of small epipelagic fishes by NRIFS used in this research were mainly conducted until August 2011. To complete the time-series data, data for 200 specimens collected in the same area (Fig. 9.4a) as this study were referenced from radiocesium concentration data (for both ^{134}Cs and ^{137}Cs) of sardine and Japanese anchovy reported on the FA website from 24 March 2011 to 21 March 2013 (Fig. 9.4b) (Fisheries Agency 2012, 2013). These data were mostly obtained as raw samples through 7,200-s measurements by local municipalities in the same manner as the NRIFS data. However, information regarding the length of fish from which samples were taken was not made public. In addition to muscle samples, samples prepared from the whole fish are included in the measurement samples (in this chapter, whitebait and samples that are labeled as processed goods were excluded).

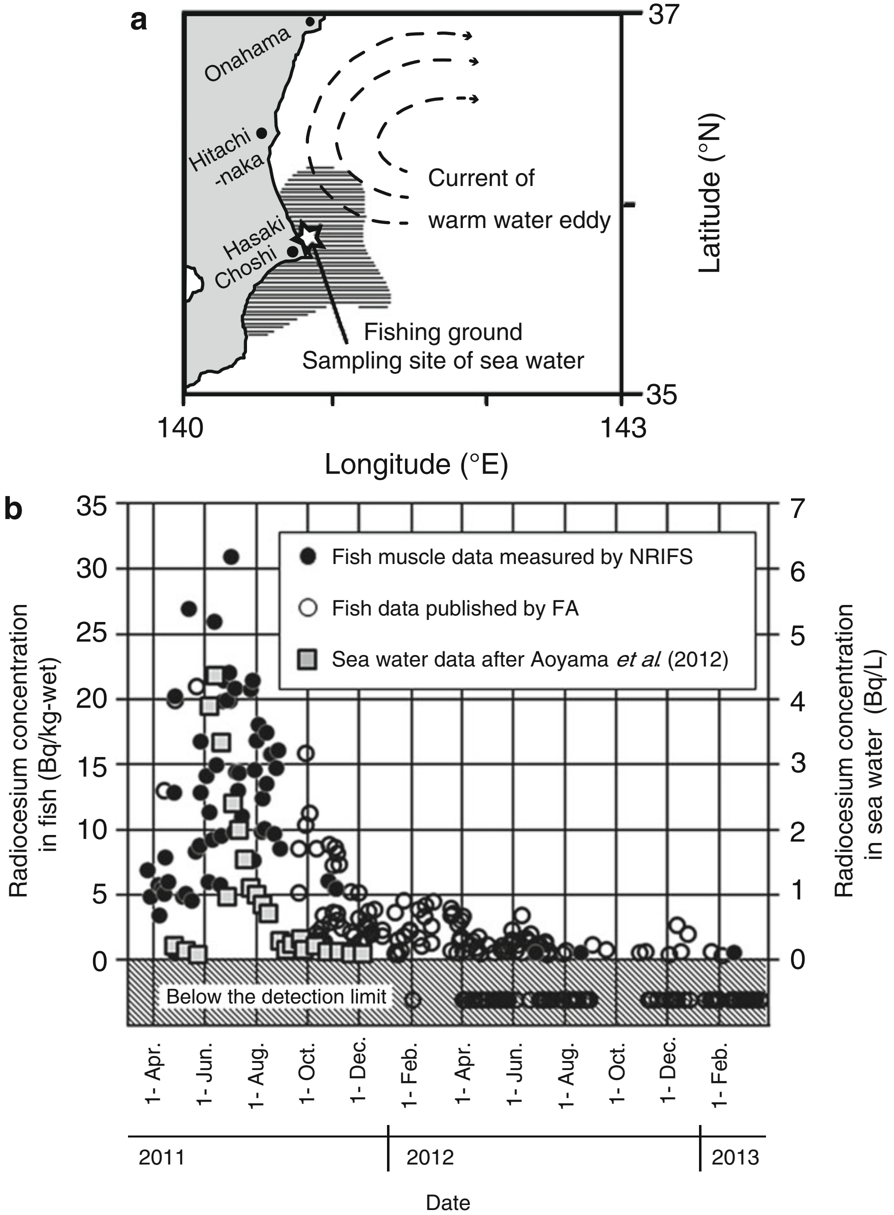

Fig. 9.4 (**a**) Locations of fishing grounds of fish sampled (sardine and Japanese anchovy) in this chapter (*shaded area*), and sampling site of seawater (*star*) (after Aoyama et al. 2012). *Thin curved arrows* indicate current of warm water eddy (after Aoyama et al. 2012). (**b**) Temporal variations of radiocesium ($^{134}Cs + ^{137}Cs$) concentration in small epipelagic fish (sardine and Japanese anchovy) caught off the Kashima-Boso area and seawater (after Aoyama et al. 2012). The National Research Institute of Fisheries Science (NRIFS) measured radiocesium concentration of fish muscle. Data from the Fisheries Agency (FA) indicate radiocesium concentrations of both muscle and whole body in fish. Plot below the *x*-axis indicates the existence of data indicating a radiocesium concentration below the detection limit

The published data on the aforementioned FA website do not state the collection date; thus, in this study we substituted the published date as the collection date for each datum. According to this, even after August 2011, the gradual decreasing trend of the concentration continued. After December 2011, the concentration decreased below 5.0 Bq/kg-wet, and by April 2012, many specimens had concentrations below the lower limit of detection. The detection limit value since April 2012 was 0.54 Bq/kg-wet on average for ^{137}Cs (range, 0.29–0.76). The concentrations of ^{137}Cs obtained from the measurement of ashed samples by NRIFS in June 2012, August 2012, and February 2013 were 0.38, 0.42, and 0.42 Bq/kg-wet, respectively, which were lower than the aforementioned average detection limit values.

9.6 Radiocesium Concentration in Seawater of the Fishing Ground

Aoyama et al. (2012) measured the radiocesium concentration (Fig. 9.4b) of the seawater collected in Hasaki (Fig. 9.4a) near the fishing ground from 25 April to 5 December 2011 after the FNPP accident. According to these results, the radiocesium concentration of seawater was less than 1.0 Bq/l from April to May 2011, and then it suddenly increased in June, reaching an average of 3.9 Bq/l in early June. After attaining the maximum value of 4.4 Bq/l in mid-June, concentration gradually decreased in late June to an average of 3.4 Bq/l. The concentration continued to decrease and reached an average of 1.1 Bq/l in late July, and in late August the average concentration was less than 1.0 Bq/l (Fig. 9.4b) (Aoyama et al. 2012). Differing from the fluctuations in the radiocesium concentration of small epipelagic fishes in the same marine area (Figs. 9.3 and 9.4b), the radiocesium concentration of seawater spiked in June, showing values significantly higher than the concentrations in April to May and July to August (Mann–Whitney *U* test, $p<0.05$). Aoyama et al. (2012) analyzed this situation as a reflection of the temporary inhibition of southward flow of the seawater (strongly affected by the FNPP accident) because of the presence of a warm eddy (Fig. 9.4a) and its arrival to Hasaki in early June. According to the survey conducted by FRA in August 2012, it has been found that the radiocesium concentration near the fishing ground (36°15′N–141°00′E) has decreased to 16 mBq/l (10 mBq/l for ^{137}Cs only) (Fisheries Research Agency 2013).

9.7 Fluctuations of Radiocesium Concentration of Small Epipelagic Fishes Associated with Their Migration Patterns

Compared to spiking fluctuations of the radiocesium concentration of the seawater, the radiocesium concentration in the muscle of small epipelagic fishes showed a relatively mild increase and decrease (Fig. 9.4b). Because radiocesium is incorporated into the bodies of fish from the environment and remains there for some time, the radiocesium concentration of small epipelagic fishes gradually decreased following the fluctuations of radiocesium concentration in seawater. However, because samples were prepared from the catch for this study, fluctuations in concentration in the bodies of fish might be affected by the distribution condition of small epipelagic fishes in the fishing ground. Sardine and Japanese anchovy are widespread in the Sanriku-Boso area during winter and spring, and wintering age 0 fish are known to migrate southward from Sanriku to Boso (Uchiyama 1998; Yasumi 2008; Kubota 2012). From the coast off Hokota City, which is located just to the north of Kashima City, to the coast off Kitaibaraki City, the concentrations detected from April to May 2011 for sardine and Japanese anchovy were 40 and 30–170 Bq/kg-wet, respectively (Fisheries Agency 2012). Based on these results, the reason that the radiocesium concentration of small epipelagic fishes increased before the concentration in the seawater of Hasaki increased, and showed some variability, may be that the school of fish from the northern ocean with a radiocesium concentration higher than that of Hasaki has migrated into the fishing ground off the Kashima-Boso area.

9.8 Comparison of Situations Before and After the FNPP Accident

From ^{134}Cs and ^{137}Cs released as a result of the FNPP accident, we will use the ^{137}Cs nuclide with a relatively long half-life (30.1 years), which can be compared to the pre-FNPP accident conditions, to continue this discussion.

Measurement values from May to November 2011 (for which radiocesium concentration data of seawater in Hasaki are complete from the early part of a month to the later part) were used to compare the fluctuations in ^{137}Cs concentration of seawater and small epipelagic fishes in 2011 for monthly average values (Fig. 9.5). The average concentrations in small epipelagic fishes were 6.1 ± 4.3, 8.8 ± 3.8, 8.9 ± 3.9, and 7.9 ± 2.0 Bq/kg-wet for May, June, July, and August, respectively; the August average value was close to those of June and July, but it had relatively small deviations (Fig. 9.5). The values decreased to approximately half the value recorded in May by October and November at 3.2–3.4 Bq/kg-wet. In comparison, the concentrations in seawater were 0.07 ± 0.02 Bq/l in May, with a maximum value of 1.65 ± 0.77 Bq/l in June. It then decreased to 0.95 ± 0.30 Bq/l in July, and the decreasing trend continued; by November, the value was similar to that from May at 0.07 ± 0.01 Bq/l. As these results show, seawater concentrations quickly decreased after peaking in June, but the concentrations of small epipelagic fishes remained relatively high until August. Thus, this clearly indicates a delayed decreasing trend compared to the seawater. Assuming that the small epipelagic fishes and environmental water are in equilibrium, we calculated the concentration coefficient (biological concentration/seawater concentration) of ^{137}Cs in the muscle of small pelagic fish from the average monthly values of radiocesium concentration already

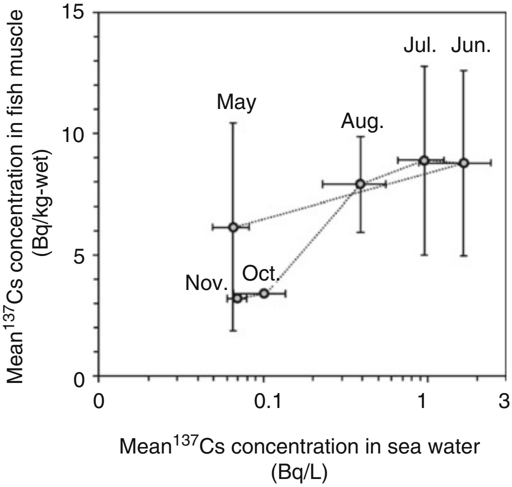

Fig. 9.5 Relationship in mean ^{137}Cs concentration per month between seawater (after Aoyama et al. 2012) and small epipelagic fish (sardine and Japanese anchovy) measured by National Research Institute of Fisheries Science (NRIFS) during May to November 2011. *Horizontal and vertical bars* indicate standard deviation

described. The results showed that the coefficient varied from 5 to 94 in 2011, with October and November having values of 34 and 46, respectively. Based on the survey results by FRA (2013), the concentration coefficient of ^{137}Cs of seawater concentration near the fishing ground in August 2012 was obtained, and the result was 42. The concentration coefficient of ^{137}Cs before the FNPP accident was 10–100 for all types of fishes, but 20–40 for sardine (Yoshida 1999); thus, the concentration coefficient of small epipelagic fishes collected in the target marine area since the fall of 2011 appears to be returning to this range. In contrast to the ^{137}Cs concentration in the muscles of sardine collected in the same marine area in June 2009, 0.038 Bq/kg-wet (Ministry of Agriculture, Forestry and Fisheries Agriculture 2011), concentrations approximately 10 times higher than those from before the FNPP accident (0.42 Bq/kg-wet) were detected in August 2012 and February 2013 during the course of this study. Meanwhile, as the ^{137}Cs concentration in seawater was 2.2 mBq/l in June 2009 (Japan Coastal Guard 2010), the value in August 2012 was approximately 4.5 times higher than that from before the FNPP accident. Therefore, it has been indicated that the seawater concentration decreases before the radiocesium concentration of small epipelagic fishes decreases. Based on these results, although the rapid fluctuations in radiocesium concentrations are decreasing, the radiocesium concentration still remains higher than that before the FNPP accident. It is necessary to continue long-term monitoring to track the decreasing process of radiocesium in small epipelagic fishes and analyze the behaviour of radiocesium in marine ecosystems.

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