Chapter 3
Temporal Changes in ^{137}Cs Concentration in Zooplankton and Seawater off the Joban–Sanriku Coast, and in Sendai Bay, After the Fukushima Dai-ichi Nuclear Accident

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Abstract The Fukushima Dai-ichi Nuclear Power Plant (FNPP) accident following the Great East Japan Earthquake in 2011 resulted in the release of enormous quantities of anthropogenic radionuclides into the ocean off the east Japanese coast, especially radioactive cesium (^{134}Cs and ^{137}Cs). FNPP-derived radioactive Cs might have consequently accumulated within marine food webs via seawater intake and predator–prey interactions. This study provides evidence of temporal variability in ^{137}Cs concentrations in seawater and zooplankton samples collected off the Joban–Sanriku coast and in Sendai Bay between June 2011 and December 2013. In Sendai Bay, seawater ^{137}Cs concentration was more than 1 Bq/kg in June 2011 and rapidly decreased over the study period. ^{137}Cs concentration in zooplankton was also measured to be as high as high 23 Bq/kg-wet in June 2011, and this concentration decreased at a slower rate than seawater concentrations. The difference in the rate of decrease of ^{137}Cs concentration between seawater and zooplankton resulted in an elevated apparent concentration ratio (aCR) for zooplankton. The observed relationship between ^{137}Cs in seawater and the aCR of zooplankton reflected the progression of ^{137}Cs contamination in zooplankton from the beginning of the FNPP accident to the restoration phase.

Keywords Fukushima Dai-ichi Nuclear Power Plant accident • ^{134}Cs • ^{137}Cs • Zooplankton • Seawater • Dynamic equilibrium • Concentration ratio

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3.1 Introduction

After the magnitude 9.0 Great East Japan Earthquake and subsequent tsunami on March 11, 2011, a loss of electric power at the Fukushima Dai-ichi Nuclear Power Plant (hereafter FNPP) resulted in an overheated reactor and hydrogen explosions. Enormous quantities of radionuclides were then released into the ocean through atmospheric fallout as well as direct release and leaking of the heavily contaminated coolant water (Chino et al. 2011; Buesseler et al. 2011). Because of its relatively long half-life (2.07 years for ^{134}Cs and 30.07 years for ^{137}Cs), evaluation of this radioactive Cs in the marine environment is important for addressing risks both to marine ecosystems and to public health through consumption of fisheries products. The Japanese government conducted intensive monitoring of ^{131}I, ^{134}Cs, and ^{137}Cs concentrations in seawater offshore near the FNPP (Nuclear Regulation Authority 2014) and in fisheries products in a wide area around Japan to ensure the safety of fishery products (Fisheries Agency 2014). In the western North Pacific, the dispersion pattern of FNPP-derived radioactive cesium from just after the FNPP accident was studied by means of direct observations and simulation models (see Chap. 2). The FNPP-derived radioactive Cs was dispersed eastward in the surface seawater in a wide area of the northern Kuroshio Extension, and a part of the FNPP-derived radioactive Cs contamination intruded into the southern area of the Kuroshio Extension with mode water and was transported westward far south of the Japan Islands (see Chap. 2).

Wada et al. (2013) demonstrated the temporal change in ^{134}Cs and ^{137}Cs concentrations as total radioactive cesium (^{134}Cs + ^{137}Cs), which is limited to 100 Bq/kg-wet by Japanese authorities, in numerous species of marine organisms collected around Fukushima Prefecture and clarified the difference in the decrease rate of radioactive cesium among species. The decrease in rates of radioactive Cs in demersal fish was slower than that of pelagic fish (Wada et al. 2013; Iwata et al. 2013; Buesseler 2012), mainly because of a high concentration of FNPP-derived radioactive cesium in the marine sediments offshore near the FNPP (Kusakabe et al. 2013; Chap. 4). Even though temporal changes of many fisheries products were clarified from the monitoring data, the mechanism controlling the concentrations of radioactive Cs in each marine organism is still unknown (Wada et al. 2013; Iwata et al. 2013; Buesseler 2012). One of the most important factors controlling the amount of radioactive Cs in marine organisms is the uptake of radioactive Cs through food (Yoshida and Kanda 2012). Unfortunately, information concerning FNPP-derived radioactive Cs in the prey of fisheries products such as zooplankton and benthos is limited to those of zooplankton collected from the open ocean after the FNPP accident (Buesseler et al. 2012; Kitamura et al. 2013). Before the FNPP accident, several studies reported the concentration of ^{137}Cs in zooplankton around the Japanese coast (Tateda 1998; Kaeriyama et al. 2008a). Kaeriyama et al. (2008a) reported that the concentration of ^{137}Cs in zooplankton collected before the FNPP accident off the coast of Aomori Prefecture ranged from 0.01 to 0.02 Bq/kg-wet.

The concentration ratio (CR) (concentration in organisms relative to that in media) under equilibrium conditions is a useful environmental parameter, used in

mathematical models to estimate the level of radionuclides present in the organisms in comparison to the surrounding environment such as soil, sediments, water, or air (IAEA 2004; Tagami and Uchida 2013; Howard et al. 2013). The recommended CR values for ^{137}Cs in marine zooplankton, fish, and crustaceans are 40, 100, and 50, respectively (IAEA 2004). In this chapter, we did not calculate CR under equilibrium conditions; therefore, the CR value was referred to as the "apparent CR (aCR)" and was compared to the pre-FNPP CR.

In June 2011, only 3 months after the FNPP accident, the Fisheries Research Agency initiated a monitoring program to measure the environmental concentration of FNPP-derived radioactive Cs in different marine ecosystems, such as seawater, sediments, zooplankton, benthos, and fishes, in the most severely affected area off the coasts of Fukushima, Miyagi, and Ibaraki Prefectures (hereafter Joban–Sanriku coast) and in Sendai Bay (Fig. 3.1). In this chapter, we describe temporal changes in the concentrations of ^{137}Cs in seawater and zooplankton off the Joban–Sanriku coast and in Sendai Bay that occurred from June 2011 to December 2013 based on data from Kaeriyama et al. (2014). Although ^{134}Cs was also determined, the decreasing trend of ^{134}Cs during more than 2 years was strongly affected by the physical decay of ^{134}Cs. Thus, only ^{137}Cs is presented (^{134}Cs data were reported in

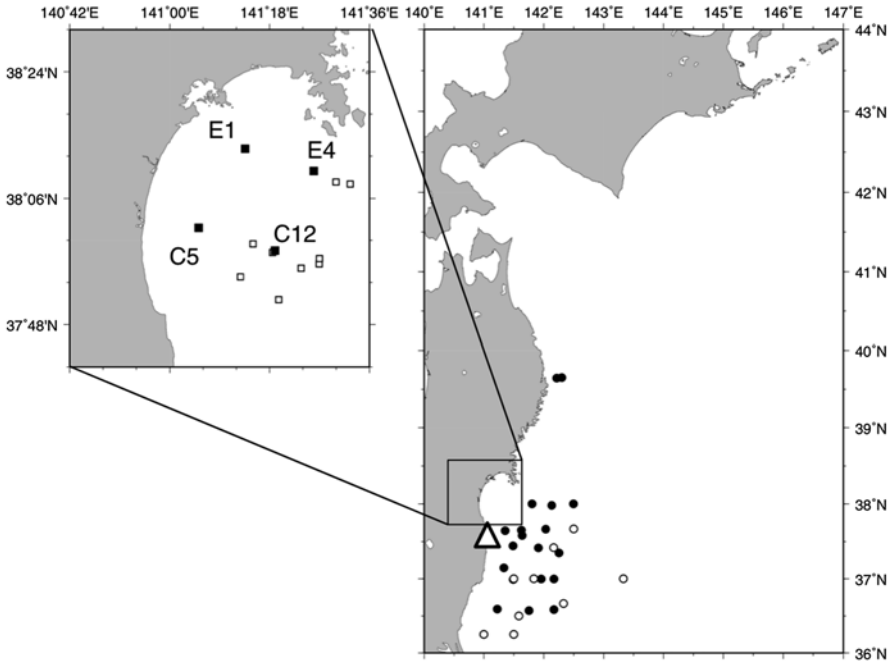

Fig. 3.1 Seawater and zooplankton sampling locations. *Filled and open circles* indicate sampling locations off the Joban–Sanriku coast in 2011 and in 2012, respectively. *Filled and open squares* indicate the repeated sampling stations (E1, E4, C5, C12) and other stations, mostly observed in June 2011 in Sendai Bay. The Fukushima Dai-ichi Nuclear Power Plant is shown as an *open triangle* in the *right panel* (Modified from Kaeriyama et al. 2014)

Kaeriyama et al. 2014). The fate of FNPP-derived radioactive Cs in seawater and zooplankton is also discussed in regard to the atomic ^{137}Cs/stable Cs ratio and the relationship between ^{137}Cs in seawater and ^{137}Cs aCR of zooplankton.

3.2 Temporal Changes of ^{137}Cs in Seawater and Zooplankton

After the FNPP accident, environmental ^{137}Cs concentrations increased in seawater and zooplankton in the area off the Joban–Sanriku coast and in Sendai Bay. Off the Joban–Sanriku coast, the concentration of ^{137}Cs decreased drastically by one order of magnitude between 2011 and 2012 (Fig. 3.2a). Generally, the behavior of cesium is thought to be conservative. Cesium is a soluble substance (<1 % is attached to marine particles) (Buesseler et al. 2011), and it is dispersed primarily by ocean currents. In fact, FNPP-derived radioactive Cs was dispersed eastward rapidly in the North Pacific, with an estimated speed of 8 cm/s, following predominant water currents (Aoyama et al. 2013). According to Kaeriyama et al. (2013), ^{134}Cs and ^{137}Cs concentrations in surface seawaters at 144°E decreased by one or two orders of magnitude between July 2011 and July 2012. The fate of ^{137}Cs off the Joban–Sanriku coast also mainly depends on seawater dilution. In Sendai Bay, the ^{137}Cs monthly average value measured in seawater drastically decreased from 770 mBq/kg in June to 30 mBq/kg in December 2011. Subsequently, the decreasing trend continued, although moderately, until the concentration reached 7 mBq/kg in November–December 2013 (Fig. 3.2a). The residence time of seawater in Sendai Bay has been estimated to be 40 days (Kakehi et al. 2012) even for calm ocean conditions; therefore, the rapid decrease in ^{137}Cs observed during the first year following the FNPP accident might have been influenced by the level of water exchange in this bay. ^{137}Cs peaked in surface waters between June and September 2011 at the E1, E4, and C5 sampling stations, although the vertical differences in ^{137}Cs concentrations were not obvious in December 2011 for the same stations (Fig. 3.3). The depth of the seasonal mixed layer may also influence the seasonal variation observed in the seawater ^{137}Cs vertical profile. In April 2012, the differences observed in ^{137}Cs concentrations between the surface and the middle or bottom waters were reduced in comparison with the differences observed during June and September 2011. During 2011–2012, winter mixing led to a homogeneous vertical distribution of ^{137}Cs in this bay.

In contrast to the rapid decrease of FNPP-derived radioactive Cs measured in seawater, the concentration of ^{137}Cs in zooplankton showed only a gradual decrease over the course of this study. ^{137}Cs concentration in zooplankton ranged from 0.21 to 23 Bq/kg-wet (Fig. 3.2b). Off the Joban–Sanriku coast, the median ^{137}Cs concentration in zooplankton decreased from 1.4 to 0.39 Bq/kg-wet between July–August 2011 and August 2012 (Fig. 3.2b). Although these data varied considerably among stations, the ^{137}Cs concentrations in zooplankton differed significantly between July–August 2011 and August 2012 (Wilcoxon rank-sum test, $p<0.05$). In Sendai Bay, ^{137}Cs concentrations in zooplankton did not differ significantly between zoo-

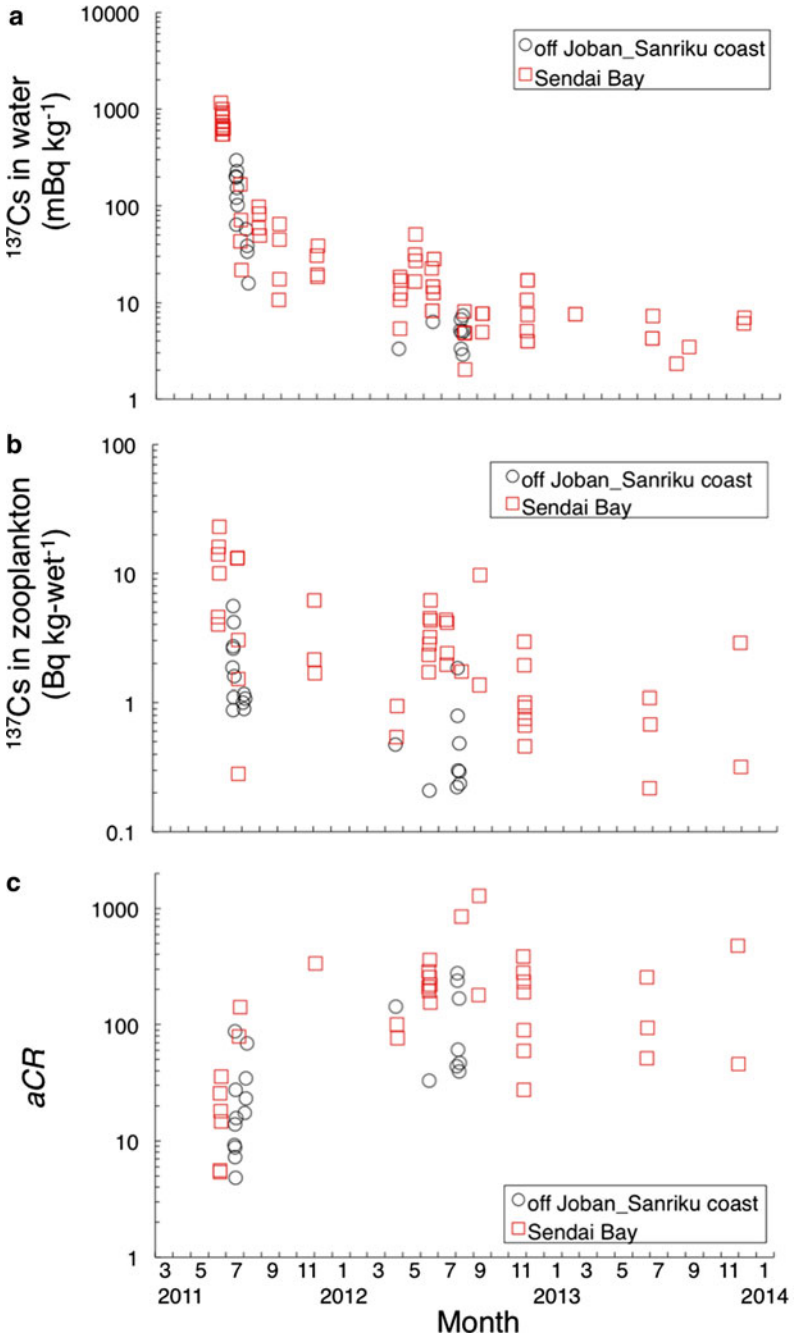

Fig. 3.2 Temporal changes in the concentration of ^{137}Cs in seawater (average value of two or three depth strata) (**a**) and in zooplankton (**b**), and the apparent concentration ratio (*aCR*) for zooplankton (**c**). *Black open circles and red squares* indicate data obtained off the Joban–Sanriku coast and in Sendai Bay, respectively (Modified from Kaeriyama et al. 2014)

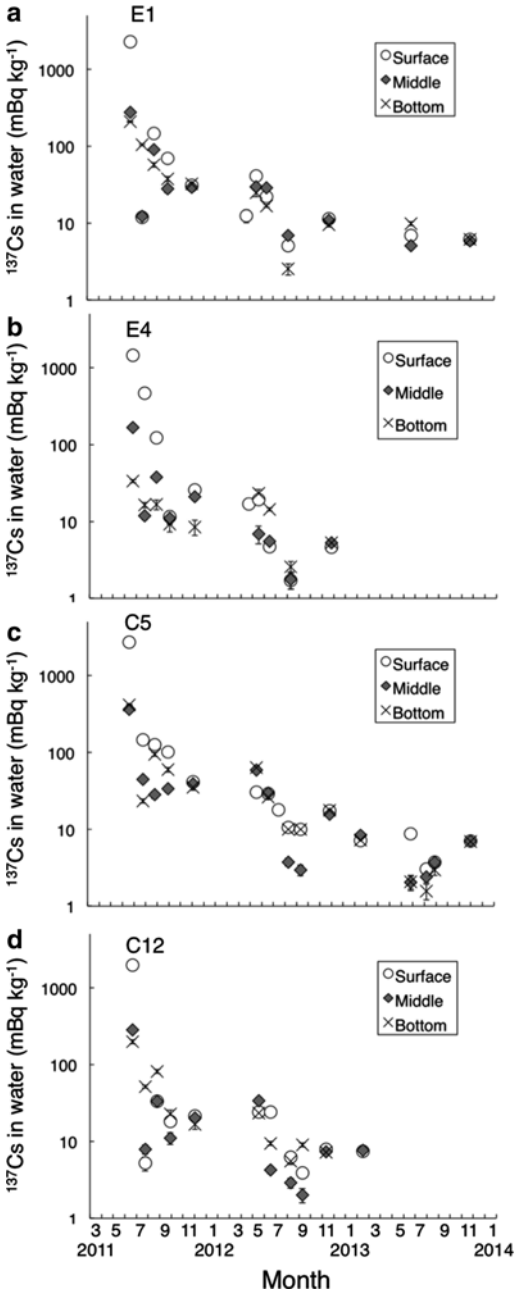

Fig. 3.3 Temporal changes in ^{137}Cs concentration in seawater at E1 (**a**), E4 (**b**), C5 (**c**), and C12 (**d**) in Sendai Bay. *Open circles, filled diamonds, and crosses* indicate the concentration of ^{137}Cs in surface, middle, and bottom waters, respectively (Modified from Kaeriyama et al. 2014)

plankton collected using a Bongo net and a sledge net (Wilcoxon rank-sum test, $p > 0.05$). The temporal change in the ^{137}Cs concentration of zooplankton, in terms of the median value calculated for each sampling period, clearly decreased from June 2011 to April 2012, slightly increased and fluctuated between June and September 2012, and then decreased again between September 2012 and June 2013 (Fig. 3.2b). The median ^{137}Cs value measured in zooplankton in November 2013 was 13 % of that measured in June 2011.

The concentration of radioactive Cs in marine organisms is mainly influenced by the rate of excretion of the organism and its intake of radioactive Cs from the prey and the surrounding seawater. Iwata et al. (2013) estimated the "ecological half-life" (T_{eco}) for marine organisms collected off the Fukushima prefecture. T_{eco} is defined as the time required for the radionuclides concentration to decline by 50 % in a natural population. This value is influenced by both abiotic factors (such as temporal changes in the concentration of radioactive Cs in seawater, extension of the contaminated area, temperature, and salinity) and biotic factors (such as life stages, feeding habitat, and population migration range). The T_{eco} for the zooplankton samples collected in Sendai Bay and off the Joban–Sanriku coast was estimated to be 263 ± 48 days ($T_{eco} \pm$ SE, $p < 0.0001$) and 178 ± 31 days ($p < 0.0001$), respectively. The difference in T_{eco} values between Sendai Bay and the Joban–Sanriku coast may result from the difference in the decreasing rate of ^{137}Cs in the surrounding seawater. The time required for a 50 % decline of ^{137}Cs in seawater in Sendai Bay (122 ± 10 days, $p < 0.0001$) was longer than that of the Joban–Sanriku coast (85 ± 8 days, $p < 0.0001$). The ratios of T_{eco} of zooplankton to the time required for 50 % decline in seawater in Sendai Bay and off the Joban–Sanriku coast are almost comparable (2.2 vs. 2.1), suggesting that the decreasing rate of ^{137}Cs in zooplankton was strongly affected by the decreasing rate of ^{137}Cs in ambient seawater.

3.3 Dynamic Equilibrium of Radioactive Cs Between Zooplankton and Seawater

The concentration of radioactive Cs in marine organisms is mainly influenced by the rate of uptake of radioactive Cs from prey and the surrounding seawater and the excretion rate from the organism, which comes down to the dynamic equilibrium of radioactive Cs between organisms and the surrounding seawater. The atomic ratio of radioactive Cs and stable Cs in organisms and seawater is a good indicator of whether dynamic equilibrium between the organism and seawater has been reached (Tateda and Koyanagi 1994, 1996; Tateda 1998). The range of stable Cs concentrations in this study (16–190 ng/g-dry; Table 3.1) is comparable to the reported values of zooplankton collected around the Japan Islands before the FNPP accident (12–447 ng/g dry; Kaeriyama et al. 2008b; Masuzawa et al. 1988; Marumo et al. 1998; Tateda 1998). The atomic ^{137}Cs/Cs ratio in zooplankton ($0.063–5.1 \times 10^{-7}$; Table 3.1) was one or two order of magnitudes higher than previously reported ($2.7 \pm 2.0 \times 10^{-9}$ (Tateda 1998). Furthermore, the atomic ^{137}Cs/Cs ratio fluctuated with time, and high

Table 3.1 Concentrations of ^{137}Cs and stable Cs in zooplankton and the atomic ^{137}Cs/Cs ratios in zooplankton and seawater

Station ID	Latitude	Longitude	Sampling date	Days from March 11 2011	^{137}Cs (Bq/kg-wet)	Stable Cs (ng/g-dry)	Atomic ^{137}Cs/Cs ratio	
							Zooplankton ($\times 10^{-7}$)	Seawater[a] ($\times 10^{-9}$)
Off Joban–Sanriku coast								
F250	37°34.8′N	141°38.37′E	2012/4/19	405	0.47	154	0.063	3.5
F250	37°34.8′N	141°38.37′E	2012/6/16	463	0.21	63	0.10	6.6
SY20	37°00.0′N	141°30.0′E	2012/8/3	511	1.8	34	1.1	3.4
SY21	37°00.0′N	141°50.0′E	2012/8/3	511	0.79	34	0.66	6.9
SY22	37°00.0′N	143°50.0′E	2012/8/4	512	0.30	27	0.45	5.1
SY16	36°15.0′N	141°00.0′E	2012/8/6	514	0.29	34	0.17	3
SY17	36°15.0′N	141°30.0′E	2012/8/6	514	0.49	79	0.18	7.7
F250	37°34.8′N	141°38.37′E	2012/8/7	515	0.24	41	0.092	5.3
Sendai Bay								
C16	37°56.6′N	141°26.9′E	2011/7/22	133	0.28	34	0.18	NS[b]
C10	37°59.5′N	141°15.0′E	2011/12/3	267	1.7	53	0.62	NS
E1	38°13.1′N	141°13.1′E	2012/4/22	408	0.95	54	0.25	13
E4	38°09.9′N	141°26.0′E	2012/6/15	462	2.3	16	5.1	8.5
C5	38°01.8′N	141°05.2′E	2012/6/18	465	6.2	126	2.0	29

C5	38°01.8′N	141°05.2′E	2012/7/14	491	4.3	64	1.8	NS
C10	37°59.5′N	141°15.0′E	2012/7/14	491	2.4	42	2.1	NS
C16	37°56.6′N	141°26.9′E	2012/7/15	492	4.1	35	3.1	NS
C22	37°53.6′N	141°39.0′E	2012/7/15	492	2.0	30	1.6	NS
E4	38°09.9′N	141°26.0′E	2012/8/10	518	1.7	127	0.32	2.2
C5	38°01.8′N	141°05.2′E	2012/9/9	548	1.4	33	1.3	7.9
C5	38°01.8′N	141°05.2′E	2012/9/10	549	9.7	77	3.4	7.9
E1	38°13.1′N	141°13.1′E	2012/11/10	610	3.0	71	1.8	1.8
E4	38°09.9′N	141°26.0′E	2012/11/10	610	1.9	179	0.37	1.8
C5	38°01.8′N	141°05.2′E	2012/11/10	610	1.0	69	0.65	11
C5	38°01.8′N	141°05.2′E	2012/11/10	610	0.46	72	0.28	5.3
E1	38°13.1′N	141°13.1′E	2013/6/15	827	0.68	58	0.28	4.4
C5	38°01.8′N	141°05.2′E	2013/6/15	827	1.1	75	0.35	4.4
C5	38°01.8′N	141°05.2′E	2013/6/15	827	0.22	42	0.12	7.6
E1	38°13.1′N	141°13.1′E	2013/11/15	980	2.9	189	0.50	7.3
C5	38°01.8′N	141°05.2′E	2013/11/15	980	0.32	49	0.26	6.3

Source: Modified from Kaeriyama et al. (2014)

[a]The concentration of stable Cs in seawater was assumed to be 0.29 μg/l (Tateda and Koyanagi 1996)

[b]*NS* no sample

values were observed between June and November 2012 (Table 3.1). According to Tateda and Koyanagi (1996), the mean concentration of stable Cs in Japanese coastal waters was 0.29 μg/l. From this value and the ^{137}Cs concentration in seawater obtained in this study, the atomic ^{137}Cs/Cs ratio of seawater was also calculated (Table 3.1). The geometric mean of the atomic ^{137}Cs/Cs ratio in seawater was 5.6×10^{-9} with a range of $2.2–29 \times 10^{-9}$. The geometric mean is comparable with that obtained before the FNPP accident ($3.5–6.9 \times 10^{-9}$; Tateda and Koyanagi 1996). A high atomic ^{137}Cs/Cs ratio of seawater ($11–29 \times 10^{-9}$) was observed at station E1 in April 2012 and at station C5 in June and November 2012. One of the possible explanations for the temporal and spatial variations in the atomic ^{137}Cs/Cs ratios of seawater and zooplankton may be the pulse input of FNPP-derived ^{137}Cs from land to ocean caused by heavy rain during the typhoon season or ice melt during thaw season. Actually, Nagao et al. (2013) reported that the export flux of ^{137}Cs from land to ocean during the heavy rain season (September 2011) through rivers located in the Fukushima Prefecture contributed 50 % of their annual export flux in 2011 (see also Sect. 2.5). The input of FNPP-derived ^{137}Cs from land to ocean is one of the most important processes affecting the coastal environment and needs further investigation to understand the long-term effects of the FNPP accident on the coastal region. Another possible input source of FNPP-derived radioactive Cs is continuing release from the FNPP harbor; the estimated average release rate of ^{137}Cs was 93 GBq day^{-1} in the summer of 2011 and 8.1 GBq day^{-1} in the summer of 2012 (Kanda 2013). However, as this radioactive Cs would be diluted offshore near the FNPP harbor, the elevation of radioactive Cs concentration in seawater and zooplankton would be almost negligible within the present study area. Judging from the atomic ^{137}Cs/Cs ratio, which was higher than before the FNPP accident in zooplankton but constant in seawater, ^{137}Cs dynamic equilibrium between zooplankton and the surrounding seawater was not attained during the study period.

In contrast to T_{eco} (see Sect. 3.2), the biological half-life (T_b) of zooplankton was reported as 13 days (Vives i Batlle et al. 2007). The T_b of zooplankton strongly suggests that dynamic equilibrium should have been attained during this study. Because the zooplankton samples contained multiple species (such as copepods, euphausiids, amphipods, chaetognath), including those with gut contents, the concentration of radioactive Cs in zooplankton may have been affected by interspecies variability in radioactive Cs concentrations in this study. The species-specific difference in stable Cs content was less than one order of magnitude (Kaeriyama et al. 2008b; Masuzawa et al. 1988; Marumo et al. 1998). Thus, the difference in species composition should not be a major factor influencing radioactive Cs in zooplankton. The gut contents of zooplankton may contain suspended particles and/or clay particles; clay particles have higher radioactive Cs than organic particles such as phytoplankton (Kusakabe et al. 2013). In addition, high concentrations of ^{134}Cs and ^{137}Cs were observed in fecal pellets of zooplankton soon after the Chernobyl accident (Fowler et al. 1987). The stable Cs contents in this study were almost comparable with previous studies based on samples containing gut contents (Kaeriyama et al. 2008b). Thus, the high radioactive Cs in gut contents likely did not affect the concentration of radioactive Cs in zooplankton. At present, it is difficult to determine the reason

for the slow decrease in the rate of ^{137}Cs in zooplankton observed in this study. Laboratory experiments on the uptake and excretion of radioactive Cs by zooplankton under unstable conditions, such as radioactive Cs in seawater/prey that increases/decreases with time, would provide insights on the time-dependent concentration of radioactive Cs in seawater and the corresponding time-dependent concentration of radioactive Cs in zooplankton.

3.4 Temporal Changes of the ^{137}Cs Apparent Concentration Ratio (aCR) of Zooplankton

The ^{137}Cs aCR in zooplankton collected off the Joban–Sanriku coast varied from 5 to 276, and the median value increased with time from 12, measured in July 2011, to 29, measured in August 2011, and to 115, measured in August 2012 (Fig. 3.2c). In Sendai Bay, the aCR varied between 5 and 1,280 throughout the study period. Because of the large variation in ^{137}Cs concentrations among zooplankton samples, aCR also varied within each sampling period in Sendai Bay. The aCR monthly median value increased from 16, measured in June 2011, to 335 in December 2011 and fluctuated by more than 80, up to 854 in August 2012 and 730 in September 2012. The ^{137}Cs aCR of zooplankton increased over time, although it varied significantly between months (Fig. 3.2c). In November–December 2013, the median aCR value (262) was more than one order of magnitude higher than *CR* values obtained before the FNPP accident, which ranged from 6 to 14 (Kaeriyama et al. 2008a). The increase in aCR was mainly associated with differences in the rate of decrease of ^{137}Cs in seawater and zooplankton, as was clearly observed in Sendai Bay. The continuous uptake of ^{137}Cs by zooplankton may lead to a slow rate of decrease of ^{137}Cs in zooplankton.

Figure 3.4a conceptually shows the temporal change in ^{137}Cs expected in seawater and zooplankton following a release of large quantities of ^{137}Cs, similar to the FNPP accident. The concentration of ^{137}Cs in seawater is expected to increase soon after the release, and the increase in ^{137}Cs in zooplankton is observed after that (phase I). A sharp peak of ^{137}Cs is observed in seawater samples, followed by an exponential decrease with time (phase II). On the other hand, the maximum concentration of ^{137}Cs in zooplankton is expected to be delayed from the peak of ^{137}Cs concentration in seawater and to gradually decrease with time (phase III). A time lag in the ^{137}Cs concentration between seawater and zooplankton leads to temporal changes in aCR observed in zooplankton (Fig. 3.4b). Eventually, the rate of decrease of ^{137}Cs in seawater and zooplankton equalizes, and the zooplankton aCR reaches the same level as the CR before the release of ^{137}Cs to the environment (phase IV). The dynamic equilibrium of ^{137}Cs between zooplankton and the surrounding seawater is attained during phase IV. Figure 3.4c shows the relationship between seawater ^{137}Cs and aCR in zooplankton resulting from the temporal changes shown in Fig. 3.4a, b. The relationship between seawater ^{137}Cs concentration and ^{137}Cs zooplankton aCR in this study along with those obtained from previous studies con-

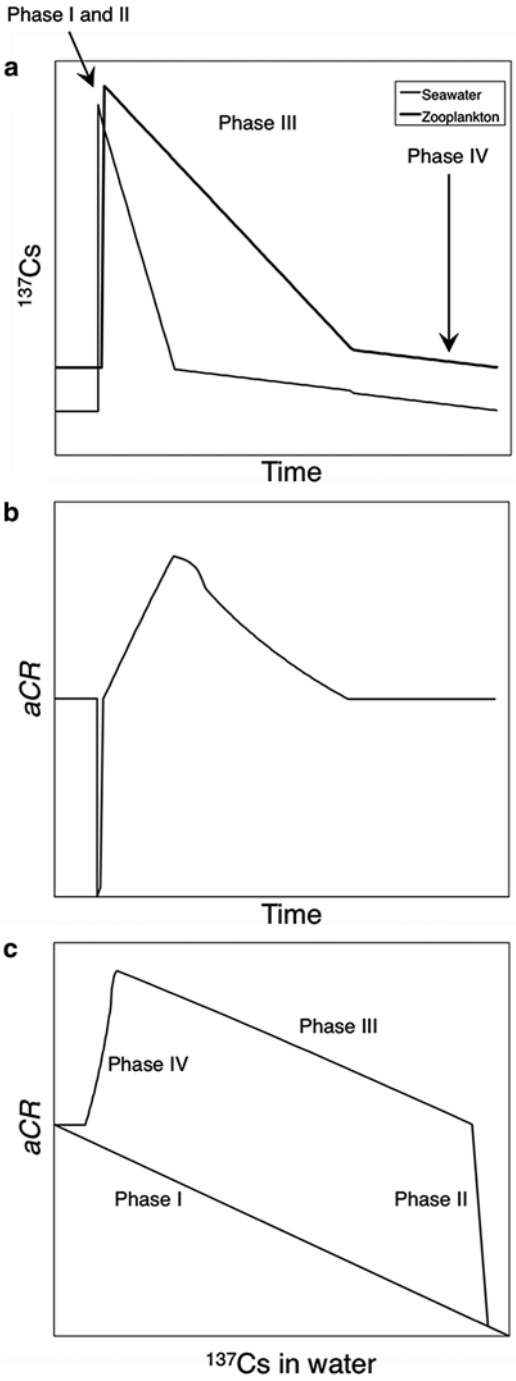

Fig. 3.4 Conceptual temporal variation in ^{137}Cs concentration in seawater (*thin lines*) and in zooplankton (*bold lines*) (**a**), *aCR* for zooplankton (**b**), and a *scatter plot* between ^{137}Cs concentrations in seawater and *aCR* for zooplankton (**c**). The temporal variation of ^{137}Cs is defined as the time-course phase from I to IV (Modified from Kaeriyama et al. 2014)

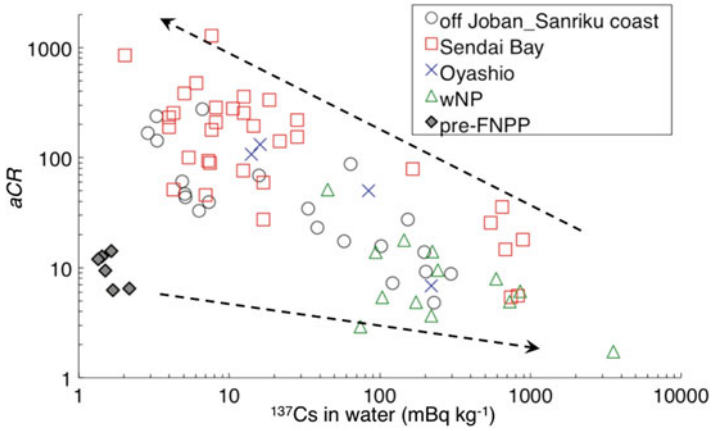

Fig. 3.5 Scatter plot between ^{137}Cs concentration in seawater and *aCR* for zooplankton off the Joban–Sanriku coast (*black open circles*) and in Sendai Bay (*red open squares*) from this study compared to those obtained in June 2011 in the western North Pacific (wNP) (*green open triangles*; Buesseler et al. 2012). The scatter plot between ^{137}Cs concentrations in seawater and the concentration ratio (*CR*) for zooplankton collected off Aomori Prefecture during October 2005 and June 2006, before the FNPP accident, is shown as *black filled diamonds* (data from Kaeriyama et al. 2008a). *Arrows* indicate flow of time (Modified from Kaeriyama et al. 2014)

ducted off the east of Japan in June 2011 (Buesseler et al. 2012) revealed that the pattern observed in Fig. 3.4c corresponds with the aCR increasing phase under dynamic nonequilibrium conditions (phase III; Fig. 3.5). Figure 3.5 also shows data obtained under dynamic equilibrium conditions before the FNPP accident (Kaeriyama et al. 2008a). The time lag expected during the elevation phase (phase I and II) should have occurred during the few months following the FNPP accident; however, this phase is not shown in Fig. 3.5 because these data were not available. On the other hand, the fate of the FNPP-derived ^{137}Cs in seawater and zooplankton varied throughout the 3 years between the FNPP accident and this study, which resulted in the negative correlation shown in Fig. 3.5. Although the ^{137}Cs aCR in zooplankton has steadily increased, the concentration of ^{137}Cs in seawater has remained nearly constant since before the FNPP accident (Fig. 3.5; 1–2 mBq/kg). If no more ^{137}Cs is added to the environment, the aCR in zooplankton would reach the decreasing phase (phase IV), and ^{137}Cs concentration in zooplankton would reach pre-FNPP accident levels in the near future. Based on the T_{eco} of zooplankton off the Joban–Sanriku coast, the ^{137}Cs concentration in zooplankton will reach the pre-FNPP accident level (0.015 Bq/kg-wet) after 2.6 years. Although the data were limited, the observed relationship between ^{137}Cs concentration in seawater and the aCR value measured in zooplankton accurately describes the progression of ^{137}Cs contamination in zooplankton from the beginning of the FNPP accident (dynamic nonequilibrium state) to the restoration phase (dynamic equilibrium state).

3.5 Possible Application of the Relationship Between Seawater ^{137}Cs and aCR to Pelagic Fishes

The concept just mentioned could also be applicable to other marine organisms, in particular to pelagic fishes that prey on zooplankton. Figure 3.6a shows the temporal changes in ^{137}Cs concentration in pelagic fish collected from Sendai Bay and off the Miyagi Prefecture (Fisheries Agency 2014) compared to the seawater and zooplankton concentrations in Sendai Bay shown in Fig. 3.2a, b. The two planktivorous fishes, the sand lance *Ammodytes personatus* and the Japanese anchovy *Engraulis japonica*, together with two carnivorous fishes, the chub mackerel *Scomber japonicus* and the Japanese sea bass *Lateolabrax japonicas*, were selected for this analysis. Figure 3.6b shows the scatter plots between ^{137}Cs in seawater and the aCRs of four fish species in relationship to the ^{137}Cs concentrations measured in zooplankton from Sendai Bay. The concentrations of radioactive Cs in fish published by the Fisheries Agency in 2011 were the total of two radionuclides, ^{134}Cs and ^{137}Cs. The activity ratio of ^{134}Cs to ^{137}Cs just after the FNPP accident is considered to be approximately 1.0 (Chino et al. 2011; Buesseler et al. 2011), and the concentrations of ^{137}Cs, including physical decay, in fish in 2011 were estimated from this ratio. To calculate the ^{137}Cs aCR in fish, the concentration of ^{137}Cs in seawater was estimated from the exponential relationship between the concentrations of ^{137}Cs measured in Sendai Bay and the days since March 11, 2011 (Fig. 3.6a).

The concentration of ^{137}Cs and aCR of planktivorous fishes, sand lance, and Japanese anchovy were similar to those measured for zooplankton. On the other hand, Japanese sea bass showed a higher concentration of ^{137}Cs and aCR than other fish and zooplankton. The species-specific difference in utilization of the environment, both for pelagic and benthic food webs and those from brackish environments in the case of the Japanese sea bass (Kosaka 1969), may have led to the observed difference in ^{137}Cs concentrations and aCRs for the Japanese sea bass and other fish and zooplankton. At present, understanding of the relationship between ^{137}Cs in seawater and the aCR in fish and their change with time is limited. Further analysis that includes ^{137}Cs data from prey items such as benthic organisms and seawater samples covering broader areas is required to completely understand the evolution of ^{137}Cs concentrations in food webs. In addition, ecological/biological features of target fish species, including spatiotemporal distribution, life cycles, and feeding habitats, would provide further insights regarding the effect of the FNPP accident on pelagic ecosystems in coastal areas off the FNPP.

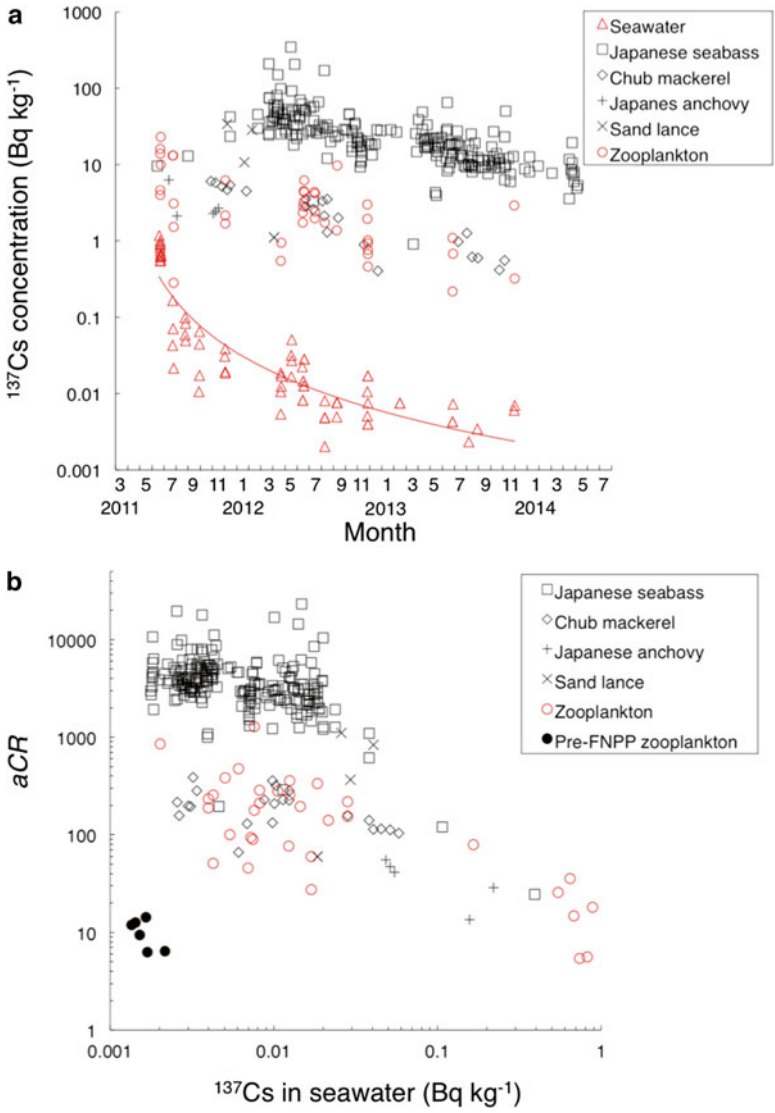

Fig. 3.6 (**a**) Temporal changes in ^{137}Cs concentrations in seawater (*red open triangles*), zooplankton (*red open circles*), sand lance (*black crosses*), Japanese anchovy (*black plus symbols*), chub mackerel (*black open diamonds*), and Japanese sea bass (*open squares*) in Sendai Bay and off the coast of the Miyagi prefecture (Fisheries Agency 2014). (**b**) *Scatter plots* showing the relationship between ^{137}Cs concentration in Sendai Bay seawater and the *aCR* in zooplankton (*red open circles*), sand lance (*black crosses*), Japanese anchovy (*black plus symbols*), chub mackerel (*black open diamonds*), and Japanese sea bass (*black open squares*) in Sendai Bay and off the coast of Miyagi Prefecture. The scatter plot between ^{137}Cs concentration in seawater and the *CR* for zooplankton off Aomori Prefecture, obtained during October 2005 and June 2006 before the FNPP accident, is also shown as *filled circles* (data from Kaeriyama et al. 2008a) (Modified from Kaeriyama et al. 2014)

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