Chapter 8
Detection of ^{131}I, ^{134}Cs, and ^{137}Cs Released into the Atmosphere from FNPP in Small Epipelagic Fishes, Japanese Sardine and Japanese Anchovy, off the Kanto Area, Japan

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Abstract The artificial radionuclides ^{131}I, ^{134}Cs, and ^{137}Cs released from FNPP were detected in Japanese sardine (*Sadinopes melanostictus*) and Japanese anchovy (*Engraulis japonicus*) off the Kanto area of Japan. In the research period from 24 March 2011 to 13 July 2011, the maximum concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs were detected in the internal organs of Japanese anchovy collected on 24 March 2011. The concentration of ^{131}I in the internal organs tended to be higher than that in muscle and the whole body, although no clear tendency was observed for ^{134}Cs and ^{137}Cs; it was thought that that was caused by ^{131}I of the planktonic contents in the internal organs. These radionuclides detected in sardine and anchovy would be derived through the atmospheric pathway from FNPP to off the Kanto area, because these radionuclides were detected before the direct release of contaminated water into the ocean from FNPP.

Keywords Radioiodine • Diocesium • Epipelagic fish • Atmosphere

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8.1 Introduction

Large amounts of artificial radionuclides such as ^{131}I, ^{134}Cs, and ^{137}Cs, were released into the environment by the Fukushima Dai-ichi Nuclear Power Plant (FNPP) accident, which was caused by the Great East Japan Earthquake and tsunami on 11 March 2011. Tokyo Electric Power Company (TEPCO) estimated that 4.7×10^{15} Bq of radioactive materials including ^{131}I, ^{134}Cs, and ^{137}Cs were released directly into the ocean from the FNPP Unit 2 reactor during April 1–6 in 2011 (Nuclear Emergency Response Headquarters 2011), although it was reported that the direct release to the ocean had already occurred on 26 March 2011, and showed the estimation that the total amount of ^{137}Cs directly released was $3.5 \pm 0.7 \times 10^{15}$ Bq from March 26 to the end of May 2011 (Tsumune et al. 2012). The total quantity of ^{131}I and ^{137}Cs released into the atmosphere between 12 March 2011 and 1 May 2011 was estimated to be approximately 2.0×10^{17} Bq and 1.3×10^{16} Bq, respectively. Furthermore, the quantities of ^{131}I and ^{137}Cs deposited on the ocean surface from the atmosphere were estimated as 9.9×10^{16} Bq and 7.6×10^{15} Bq, respectively (Kobayashi et al. 2013).

Monitoring research detected ^{131}I, ^{134}Cs, and ^{137}Cs in marine organisms (Fisheries Agency 2014). The source of the ^{131}I and ^{134}Cs detected in marine organisms clearly originated from FNPP because of the short physical half-lives, 8.02 days for ^{131}I and about 2.06 years for ^{134}Cs. However, it has been unclear whether the radionuclides were released into the atmosphere or directly into the ocean from FNPP. In this report, we focus on the detection of ^{131}I, ^{134}Cs, and ^{137}Cs in small epipelagic fishes, sardine and anchovy, off the Kanto area of Japan. Our results indicate that the ^{131}I, ^{134}Cs, and ^{137}Cs detected in small epipelagic fishes was released into the atmosphere from FNPP.

8.2 Experimental Procedure

Fish samples were commercially collected from 24 March 2011 to 13 July 2011 at regions shown in Fig. 8.1. An individual fish sample contain only small amounts of ^{131}I, ^{134}Cs, and ^{137}Cs, so to determine the concentrations we used multiple fish samples for each measurement specimen. Therefore, we prepared the specimen for measurement consisting of muscle, internal organs, and whole body from multiple samples, a total of 5–10 kg individuals. The previous report showed ^{134}Cs and ^{137}Cs concentrations in most of the fish samples obtained from raw measurement specimens (Takagi et al. 2014). In this report, those samples were re-measured after ashing. On the other hand, ^{131}I concentrations were obtained from the measurement using a raw measurement specimen. For preparation of the ashed measurement specimen, raw samples were dried in an oven at 105 °C for 72–120 h, carbonized in a gas furnace at 350–400 °C for approximately 6 h, and ashed in an electric furnace at 450 °C for 48–72 h. The ashed samples were ground to a fine powder, transferred to a plastic cup, and pressed using a hand press. The concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs were measured using a high-purity germanium (HpGe) semiconductor

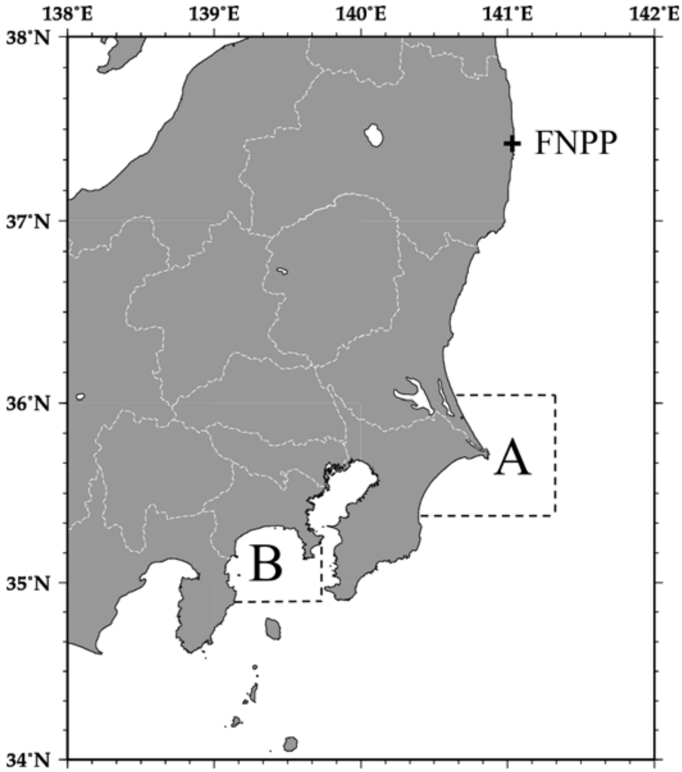

Fig. 8.1 Location of the Fukushima Dai-ichi Nuclear Power Plant (*FNPP*) and sampling regions (*A* and *B*). Respective sampling regions are surrounded by *dashed lines* indicating shorelines

detector with a multichannel analyzer (Seiko EG & G; ORTEC, Oak Ridge, TN, USA). This HpGe semiconductor detector has a resolution of 1.44 keV at a peak of 662.15 keV (^{137}Cs). The counting efficiency of the Ge semiconductor detector was calibrated using volume standard sources (MX033U8PP; Japan Radioisotope Association, Tokyo, Japan). Coincidence summing effects of ^{134}Cs were corrected with ^{134}Cs standard solutions (CZ005; Japan Radioisotope Association, Tokyo, Japan). The counting times were about 7,200 s for the raw specimen and from about 3,000 s to about 7,200 s for ashed specimens. All radionuclide concentrations were corrected for decay from the respective sampling date. The concentration of three standard deviations (σ) from counting error was defined as the detection limit.

8.3 Concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs in Sardine and Anchovy

There was no difference in the ^{134}Cs/^{137}Cs concentration ratio between sardine and anchovy and among the respective measurement specimens. Considering that the half-life for ^{134}Cs is 2.1 years, the ^{134}Cs/^{137}Cs concentration ratio in these small

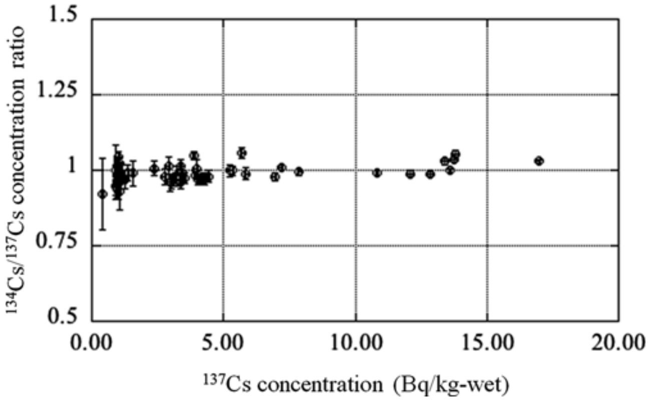

Fig. 8.2 Relationship between $^{134}Cs/^{137}Cs$ concentration ratio and concentration of ^{137}Cs detected in this study. The $^{134}Cs/^{137}Cs$ concentration ratio was calculated from the data decay corrected on 11 March 2011

epipelagic fishes was close to 1.0 (Fig. 8.2, Table 8.1). This ratio is consistent with the $^{134}Cs/^{137}Cs$ concentration ratio already reported in seawater and marine organisms (Aoyama et al. 2012; Wada et al. 2013). The ratio indicated that the ^{134}Cs and most of the ^{137}Cs detected in these small epipelagic fishes originated from the FNPP accident. The concentration of ^{137}Cs in muscle and whole bodies without internal organs of sardines collected off the Kanto area in 2010, before the FNPP accident, was 0.052 ± 0.0038 Bq/kg-wet and 0.030 ± 0.0044 Bq/kg-wet, respectively (Fisheries Research Agency 2012).

The previous report showed the summed concentration of ^{134}Cs and ^{137}Cs in raw measurement specimens for muscle of sardine and anchovy (Takagi et al. 2014). These concentrations were 61.0 % to 155.9 % of the sum concentration of ^{134}Cs and ^{137}Cs in the ashed measurement specimen consisting of the same samples as the raw measurement specimen. Figure 8.3 shows the temporal variation of ^{131}I and ^{137}Cs concentrations in the internal organs of sardine and anchovy. The maximum concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs were 309.08 ± 2.06 Bq/kg-wet, 61.01 ± 0.52 Bq/kg-wet, and 59.63 ± 0.39 Bq/kg-wet, respectively, in the internal organs of anchovy collected 24 March 2011 (Fig. 8.3, Table 8.1). There was no detection of ^{131}I on 26 April 2011 because of the short physical half-life, 8.02 days. The concentrations of ^{131}I in the internal organs of sardine and anchovy until 26 April 2011 decreased to half by 4.4 and 4.6 days, respectively.

The respective concentrations in fishes collected in region B were obviously lower than those in region A. It was clear that the reason was the distance from FNPP to each sampling region. The concentration of ^{131}I in the internal organs tended to be higher than those in other measurement specimens, although no clear tendency was observed for ^{134}Cs and ^{137}Cs (Table 8.1). Although the concentration factor of iodine in fish is from 9 to 10, the factor of iodine in phytoplankton and zooplankton is from 800 to 1,000 (IAEA 2004). The measurement specimen from the internal organs of sardine and anchovy, which are plankton feeders, could include some plankton. Therefore, the higher concentrations of ^{131}I would be detected in the internal organ specimens from sardine and anchovy.

Table 8.1 Concentrations of ^{131}I, ^{134}Cs and ^{137}Cs in sardine and Japanese anchovy

Region	Sampling Date	Internal organs			Muscle			Whole body		
		^{131}I	^{137}Cs	^{134}Cs	^{131}I	^{137}Cs	^{134}Cs	^{131}I	^{137}Cs	^{134}Cs
Sardine										
A	2011/3/28	84.06 ± 1.55[a]	13.75 ± 0.12	13.34 ± 0.09	7.90 ± 0.42	3.04 ± 0.05	2.88 ± 0.04	24.47 ± 0.77	3.96 ± 0.09	3.93 ± 0.07
A	2011/4/6		6.96 ± 0.11	6.53 ± 0.08	5.77 ± 0.35	3.55 ± 0.05	3.37 ± 0.04	3.50 ± 0.16	2.90 ± 0.06	2.88 ± 0.05
A	2011/4/11	13.22 ± 0.20	3.69 ± 0.09	3.58 ± 0.07	2.00 ± 0.22	3.36 ± 0.06	3.32 ± 0.04	6.20 ± 0.28	5.24 ± 0.08	5.10 ± 0.06
A	2011/4/13	6.73 ± 0.29	3.43 ± 0.09	3.23 ± 0.07	0.99 ± 0.17	3.41 ± 0.06	3.29 ± 0.04	3.25 ± 0.23	2.75 ± 0.06	2.62 ± 0.04
A	2011/4/25	1.78 ± 0.30	2.28 ± 0.07	2.12 ± 0.05	<0.75	4.28 ± 0.06	3.99 ± 0.04	<0.73	3.39 ± 0.06	3.13 ± 0.04
A	2011/4/26	0.86 ± 0.25			0.74 ± 0.21			0.57 ± 0.15		
A	2011/5/5	<0.79	<0.79		<0.53			<0.59		
A	2011/5/9	<0.69	<0.69		<0.52			<0.56		
A	2011/5/16	<0.60	1.66 ± 0.05	1.52 ± 0.04	<0.56	3.91 ± 0.04	3.64 ± 0.03	<0.59	3.23 ± 0.09	3.00 ± 0.06
A	2011/5/20	<0.64	2.25 ± 0.07	2.11 ± 0.06	<0.56	5.70 ± 0.08	5.67 ± 0.06	<0.54	3.88 ± 0.04	3.83 ± 0.03
A	2011/5/25	<0.61	1.33 ± 0.07	1.12 ± 0.05	<0.60			<0.61	3.15 ± 0.05	2.88 ± 0.03
A	2011/6/2	2.19 ± 0.48	50.20 ± 0.28	44.78 ± 0.20	<0.72	12.86 ± 0.10	11.80 ± 0.07	<0.82	12.09 ± 0.11	11.12 ± 0.08
A	2011/6/4	1.16 ± 0.25	8.64 ± 0.16	8.14 ± 0.12	<0.68	4.25 ± 0.06	3.84 ± 0.05	<0.59	4.44 ± 0.07	4.04 ± 0.05
A	2011/6/22	<0.85	13.37 ± 0.15	12.57 ± 0.12	<0.63	13.42 ± 0.10	12.64 ± 0.08	<0.61		
A	2011/6/29	<0.77	12.92 ± 0.17	12.01 ± 0.13	<0.81	13.78 ± 0.10	13.22 ± 0.08	<0.70	13.76 ± 0.11	12.96 ± 0.08
B	2011/4/11	2.39 ± 0.33	1.15 ± 0.05	1.07 ± 0.03	<0.57	0.94 ± 0.02	0.86 ± 0.02	0.89 ± 0.24	0.92 ± 0.03	0.85 ± 0.02
B	2011/6/6	<0.58	2.29 ± 0.07	2.07 ± 0.05	<0.65			<0.62	1.26 ± 0.03	1.13 ± 0.02
Japanese anchovy										
A	2011/3/24	309.08 ± 2.06	61.01 ± 0.52	59.63 ± 0.39	14.30 ± 0.35			117.46 ± 1.27		
A	2011/4/7	12.05 ± 0.40	8.80 ± 0.18	8.72 ± 0.13	2.20 ± 0.35	2.38 ± 0.04	2.34 ± 0.03			
A	2011/4/14	3.61 ± 0.39			<0.67			2.34 ± 0.28		
A	2011/4/18	1.75 ± 0.09	6.22 ± 0.11	5.95 ± 0.08	<0.54	4.01 ± 0.05	3.77 ± 0.04	1.20 ± 0.16	2.95 ± 0.07	2.74 ± 0.05

(continued)

Table 8.1 (continued)

Region	Sampling Date	Internal organs			Muscle			Whole body		
		^{131}I	^{137}Cs	^{134}Cs	^{131}I	^{137}Cs	^{134}Cs	^{131}I	^{137}Cs	^{134}Cs
A	2011/4/26	2.42 ± 0.38			<0.51			1.10 ± 0.17		
A	2011/5/12	<0.72	13.31 ± 0.20	13.16 ± 0.15	<0.79	16.96 ± 0.08	16.54 ± 0.06	<0.75		
A	2011/5/18	<0.65	8.29 ± 0.11	7.60 ± 0.08		13.61 ± 0.07	12.82 ± 0.05	<0.61		
A	2011/5/26	<0.58	8.92 ± 0.11	8.59 ± 0.08	<0.67	10.84 ± 0.06	10.06 ± 0.04	<0.55	7.88 ± 0.07	7.34 ± 0.05
A	2011/5/26	<0.68	8.51 ± 0.12	7.79 ± 0.08	<0.59			<0.69	6.93 ± 0.07	6.34 ± 0.05
A	2011/7/13	<0.57	3.49 ± 0.05	3.06 ± 0.04	<0.70	7.18 ± 0.05	6.52 ± 0.04	<0.57	5.31 ± 0.06	4.77 ± 0.05
B	2011/3/16	18.56 ± 0.60	2.37 ± 0.16	2.37 ± 0.12	2.62 ± 0.13	1.07 ± 0.05	1.00 ± 0.04	4.88 ± 0.22	0.40 ± 0.04	0.37 ± 0.03
B	2011/3/29	13.65 ± 0.70	3.25 ± 0.08	3.23 ± 0.06	1.46 ± 0.35	1.39 ± 0.03	1.35 ± 0.02	4.82 ± 0.34	1.56 ± 0.05	1.52 ± 0.04
B	2011/6/6	<0.77	2.34 ± 0.13	2.11 ± 0.11	<0.50	5.87 ± 0.09	5.38 ± 0.07			

[a]Value shows 1 σ counting error

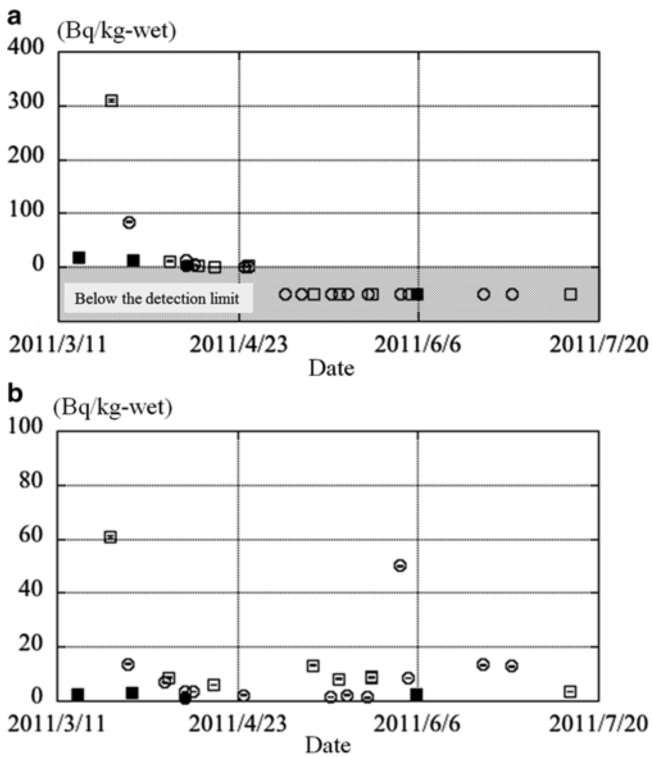

Fig. 8.3 Temporal variation in the concentration of ^{131}I (**a**) and ^{137}Cs (**b**) in internal organs of sardine and anchovy. *Circles* and *square symbols* indicate data for sardine and anchovy, respectively. *Open* and *closed symbols* indicate data for samples collected in regions *A* and *B*, respectively. *Error bar* shows 1 σ value derived from counting statistics. Errors for many of the data are too small to show an error bar

8.4 Detection of ^{131}I, ^{134}Cs, and ^{137}Cs Released into the Atmosphere from FNPP

The ^{137}Cs concentration in sardine gradually decreased until the end of May in 2011, but the concentration suddenly increased in the first week of June in 2011 (Fig. 8.3b, Table 8.1). It was considered that this sudden increase was caused by the disappearance on 30 May 2011 of a warm water eddy, the center of which was located off Iwaki between Onahama and Hasaki from the middle of May (Takagi et al. 2014; Fig. 9.4 in Chap. 9). The warm water eddy prevented the seawater, including ^{131}I, ^{134}Cs, and ^{137}Cs, from moving southward to sampling region A (Aoyama et al. 2012). In this time, ^{131}I was again detected in the internal organs of sardine, although there had come to be no detection of ^{131}I on 26 April 2011. This detection of ^{131}I also could indicate the southward movement of contaminated seawater.

^{134}Cs and ^{137}Cs were detected in sardine and anchovy collected in sampling region A before the southward movement of contaminated seawater. According to the previous report, the reason for these detections was considered to be that the contaminated sardine and anchovy migrated southward to the region earlier than the southward movement of contaminated seawater (Takagi et al. 2014). It is well known that the radioactively contaminated fishes are able to migrate from a contaminated area to a noncontaminated area. Radionuclides were transported from Russia to Japan by walleye pollock and from Japan to the United States of America by Pacific bluefin tuna (Morita et al. 2007; Madigan et al. 2012). However, in the large amount of ^{131}I, ^{134}Cs, and ^{137}Cs deposited on the ocean surface off the Kanto area (Kobayashi et al. 2013), it would be difficult to distinguish between directly released and atmospheric pathway radionuclides.

The $^{131}I/^{137}Cs$ concentration ratio in the internal organs of sardine and anchovy that were collected during from 16 March 2011 to 29 March 2011 in regions A and B was from 4.2 to 7.8. The $^{131}I/^{137}Cs$ concentration ratio of the radionuclides that were released directly from FNPP to 30 km offshore from 26 March to 6 April 2011 agreed with the radioactive decay curve of ^{131}I (Tsumune et al. 2012). However, it was unclear whether this agreement applied to the region A. In addition, the $^{131}I/^{137}Cs$ concentration ratio shows variations during atmospheric transport (Kinoshita et al. 2011) because of differences in the wet deposition rate depending on the size of particles (Hirose et al. 1993), whereas the simulation estimated that the $^{131}I/^{137}Cs$ ratio deposited in the ocean during 22 March 2011 to 24 March 2011 around region A was 6.7–40.4 (T. Kobayashi, personal communication). Therefore, it was also difficult to determine the route (as direct release or via the atmospheric pathway) of these radionuclides by the $^{131}I/^{137}Cs$ concentration ratio because of the range variation in the estimated ratio and the difference in incorporation rate into the internal organs between ^{131}I and ^{137}Cs. On the other hand, it was estimated that the direct release of the contaminated water from FNPP into the ocean occurred from 26 March 2011 (Tsumune et al. 2012). We detected ^{134}Cs and ^{137}Cs in Japanese anchovy collected on 24 March 2011 in region A and on 16 March 2011 in region B. Consequently, it was clear that these radionuclides were released into the atmosphere from FNPP; these would deposit on the surface water in this region through the atmospheric pathway. In addition, we also detected ^{134}Cs and ^{137}Cs in sardines collected on 28 March 2011 in region A and in both sardine and anchovy collected on 29 March 2011 in region B. Considering the distance between the FNPP and these sampling regions, these radionuclides were clearly released into the atmosphere from FNPP.

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