

Radioactive contamination of fish in Tokyo Bay and Edogawa River due to the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident

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Abstract

Radioactive contamination due to the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident to fish inhabiting Tokyo Bay and Edogawa River was investigated. More than $100 \text{ Bq} \cdot \text{kg}^{-1}$ of radioactive cesium, which is the regulation value of foods related to radioactive contamination in Japan, were detected from eels collected in Edogawa River in 2013, two years after the FDNPP accident. Radioactive cesium precipitated in a high contamination zone in the eastern part of Tokyo metropolitan and the northern part of Chiba Prefecture flows into Tokyo Bay through Edogawa River. This radioactive cesium was thought to be a source of contamination of eels and other fish. In Tokyo Bay the benthic fish like goby was contaminated with the radioactive cesium. The contamination of fishes inhabiting in Tokyo Bay and Edogawa River due to the radioactive cesium continues even now, about 6 years have passed since the FDNPP accident. However, the organs of the shellfish living in Tokyo Bay were clearly contaminated by radioactive nuclide $^{110\text{m}}\text{Ag}$ immediately after the FDNPP accident. It suggests the possibility that non-volatile radioactive materials such as crushed nuclear fuel particles also flew to the Tokyo metropolitan areas and were precipitated there.

Key Words: Radioactive contamination, Fish, Shellfish, Eel, Goby, Tokyo Bay, Edogawa River, FDNPP accident, $^{110\text{m}}\text{Ag}$, ^{134}Cs , ^{137}Cs

Introduction

About six years have passed since the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident caused by the Great East Japan Earthquake that occurred on March 11, 2011. A large amount of radioactive materials were released into the environment due to the meltdown and explosion of nuclear reactors Nos. 1 to 4 that occurred between March 12 and 15 in the early hours [1-6]. Radioactive nuclides released at this time also flew to the Tokyo metropolitan area on March 16 and 22, and they were washed out by rainfall and radioactively contaminated the ground surface [7-12]. Radioactive contamination in

soil of the Tokyo metropolitan due to the FDNPP accident was analyzed by Ishida and Yamazaki (2017) [13]. According to the results, it is pointed out that 10 to 20% of radioactive cesium deposited in the Tokyo metropolitan area in the 5 years after the accident may flow into Tokyo Bay. On the other hand, spatiotemporal analysis on the radioactive cesium contamination of the Tokyo Bay sediments is also conducted [14]. It was revealed that about 20% of radioactive cesium deposited in a high contaminated area in the eastern Tokyo and the northern Chiba Prefecture flowed out of Edogawa River and deposited in the Old-Edogawa River estuary in the inner part of Tokyo Bay.

The estuary of Edogawa River are the few remaining breeding areas of fish in the Tokyo Bay ecosystem. It is also a source of fresh seafood called "Edo-Mae (It means fish collected in Tokyo Bay)". Therefore, there is a high possibility that fish inhabiting Tokyo Bay is being made radioactive contamination by the FDNPP accident. But radioactive cesium derived from FDNPP continues to flow into Tokyo Bay [14].

In this study, the concentration of radionuclides of fish collected in Tokyo Bay, Old-Edogawa River, and Edogawa River since the FDNPP accident were measured, and the current situation and fluctuation of the radioactive contamination of fish were investigated.

Sampling and Analytical Method

Sample collection

Fish sampled in Tokyo Bay and Edogawa River from August 2011 to July 2017 after the FDNPP accident. Edogawa River is a most important source of the radioactive cesium contamination in Tokyo Bay by the FDNPP accident. Fishes were collected by fishing or netting. A large fish was dissected and its muscle and organs were measured as a specimen. For a small fish, the whole body was measured. The shellfish collected from the coastal beach around Tokyo Bay. A large shellfish living in deep sea was collected by fishermen. The shellfish removed the shell and measured the tissue. Several samples were purchased on the market.

Radioactivity measurements

Radionuclides in the samples were quantified by connecting a 4096 channel MCA (Labo Equipment, MCA600) to a low energy HPGe detector (ORTEC, LO-AX/30P) shielded in lead 10 cm thick, sealing the specimen inside a plastic container with diameter of 5.5 cm and depth of 2.0 cm, then measuring it via γ ray spectrometry. The HPGe detector calculated the geometric efficiency relative to the sample weight using the American NIST Environmental

Radioactivity Standard, SRM 4350B (River Sediment) and SRM 4354 (Freshwater Lake Sediment), and the efficiency was corrected within the range of 2 to 30 g of the sample weight [15]. The measurement time was set so that the counting error would be less than 5% according to the radioactive intensity of the samples. ^{110m}Ag (γ ray energy: 658 keV, half-life: 252 days), ^{134}Cs (605 keV, 2.06 years), and ^{137}Cs (662 keV, 30.1 years) were quantified in this study. A ^{134}Cs solution with known concentration was used to correct the sum peak effect for ^{134}Cs counting. The detection limit of each radionuclides under appropriate conditions was $0.6 \text{ Bq} \cdot \text{kg}^{-1}$ in fish sample. Radioactivity of the nuclides was indicated by the values of sampling day. Radioactivity concentrations of fish and shellfish are indicated on a wet weight basis.

Results and Discussion

Radioactivity of ^{110m}Ag , ^{134}Cs , and ^{137}Cs of fish inhabiting Tokyo Bay

A radioactive plume flew into the Tokyo metropolitan area on March 16 and 22, 2011 immediately after the FDNPP accident, and Tokyo Bay and its catchment area suffered a large scale radioactive contamination. Radioactive contamination of soil in the Tokyo metropolitan area and sediments in Tokyo Bay has already been reported [7-12]. However, radioactive contamination of fishes and shellfishes inhabiting Tokyo Bay and its inflowing rivers has not yet been reported.

The activity of $^{134+137}\text{Cs}$ of fish and shellfish collected in Tokyo Bay from August 2011 to May 2013 are shown in Table 1 and Figure 1. Since the radioactivity ratio of ^{134}Cs and ^{137}Cs released in the FDNPP accident was almost 1.0 [16-18], the $^{134+137}\text{Cs}$ radioactivity in 2017 will be reduced almost 0.5 times immediately after the accident due to radioactive decay.

Throughout the study period, the radioactive cesium contamination of fish inhabiting Tokyo Bay showed low values. A large amount of radioactive cesium flows in the Tokyo Bay from the catchment area, but

since they are strongly adsorbed by sediments in the estuary area, so far it is considered that the ecosystem of Tokyo Bay has not received much contamination by the radioactive cesium. However, the radioactive cesium of carnivorous bottom fish such as goby, mullet, stingray, and sea bass showed higher concentration than the surface migratory fish. Especially, goby which mainly stuffs small fish and shellfish was high contamination, the activity of $^{134+137}\text{Cs}$ reaches a maximum of about $34 \text{ Bq} \cdot \text{kg}^{-1}$.

A relatively large fish caught in Tokyo Bay was dissected and the distribution of radioactive cesium in each organs was measured. Results are shown in Table 2. The radioactive cesium concentration of the viscera was higher than muscle in one mullet, but the tendency of accumulation of radioactive cesium in specific organs was not observed. Radioactive cesium was accumulated in the muscle in fish inhabiting Tokyo Bay.

A short half-life radionuclide $^{110\text{m}}\text{Ag}$ was detected from the internal organs of several shellfish collected in Tokyo Bay early in the FDNPP accident. The radioactivity of $^{110\text{m}}\text{Ag}$ in the organs of the shellfish after seven months from the accident was 0.4 to $2.5 \text{ Bq} \cdot \text{kg}^{-1}$. Many of the shellfishes have the property of accumulating heavy metals in the internal organs [19], suggesting that radioactive silver that flowed into Tokyo Bay accumulated in the organs of the shellfish. This suggests the possibility that low volatility radioactive materials like crushed nuclear fuel particles flew to the Tokyo metropolitan area more than 200 km from FDNPP [20].

Radioactivity of ^{134}Cs and ^{137}Cs of fish inhabiting Edogawa River

The Edogawa catchment area located in the east of Tokyo, the north of Chiba Prefecture, is the region where it receives the highest radioactive contaminated in the Tokyo metropolitan area due to the FDNPP accident [7-12]. Our previous studies have revealed that radioactive cesium flows into the Edogawa River due to rainfall from this

highly contaminated zone [13-14]. Therefore, there is a high possibility that ecosystems such as fish inhabiting Edogawa River are highly contaminated with radioactive cesium.

The eel is at the top of the Edogawa River ecosystem's food chain, and the eating habits change from benthic beings such as earthworms and hills to shrimp and fish according to their growth. Ten eels were collected from March 2013 to October 2016 in Edogawa River. In addition, freshwater prawn (Write here as "shrimp") which is the predominant predator of the eel were collected. Radioactivity was measured on eel for edible parts of muscles and shrimp for whole body. The results are shown in Table 3 and Figure 2. Fishes captured in Edogawa River by 2014 was contaminated with higher concentration of radioactive cesium as compared to Tokyo Bay. This is consistent with the time when the Edogawa River water system was the most contaminated [14]. The cesium concentration of the eel is 3 to 5 times that of the predator shrimp, which indicates that the accumulation of radioactive cesium by the food chain is proceeding in the Edogawa River ecosystem.

Relationship between radioactive cesium contamination of eel and its body weight

As shown in Figure 3, a clear correlation was found between the $^{134+137}\text{Cs}$ activity of eel and its body weight. This indicates that radioactive cesium accumulated in the muscle with the growth of eel. The activity of $^{134+137}\text{Cs}$ of shrimp gradually decreases according to radioactive decay of ^{134}Cs . However, even now the activity of $^{134+137}\text{Cs}$ is 6.8 to $13.5 \text{ Bq} \cdot \text{kg}^{-1}$, which indicates that the radioactive cesium recycling process by the food chain is established in the Edogawa River ecosystem. It also suggested that radioactive pollution of eel preying such contaminated food is higher than shrimp and that radioactive cesium may be accumulated in the higher species by the food chain.

In 2013, when the radioactive cesium contamination was the highest in Edogawa River [14], the $^{134+137}\text{Cs}$ activity of four eels exceeded $100 \text{ Bq} \cdot \text{kg}^{-1}$ of the control values of

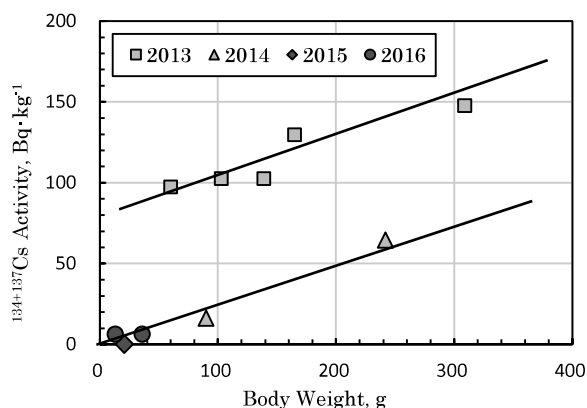


Fig. 3 Relationship between the concentration of radioactive cesium of eel's muscle and their body weight.

radioactivity contamination for the foods by the Japanese Government [21]. The eel that exceeds the radioactivity regulation value was found in the Edogawa River water system in the Tokyo metropolitan area more than 200 km away from FDNPP shows the seriousness of the FDNPP accident. We have found that the radioactive cesium contamination of the sediment of the Old-Edogawa River estuary was the worst in 2013 [14]. This is consistent with radioactive cesium contamination of eel in Edogawa River showing the highest level in 2013. The concentration of radioactive cesium in eel collected after 2014 clearly decreased, but the contamination level of grown specimens did not decrease significantly. This means that the recycling of radioactive cesium through fish by the food chain of the Edogawa ecosystem continues.

Conclusions

Radioactive contamination of sea water and sediment in Tokyo Bay has continued for more than 5 years since the FDNPP accident. The inflow source of radioactive substances flowing into Tokyo Bay is a high contamination zone in the eastern Tokyo and the northern Chiba Prefecture. Even now, the radioactive cesium such as ¹³⁴Cs and ¹³⁷Cs

flows from Edogawa River through Old-Edogawa River into Tokyo Bay, and it is caused by the radioactive contamination of fish and shellfish inhabiting there. The fish most contaminated with the radioactive cesium throughout the study period was eel and the contamination of shrimp which was the predator of eel was also observed. A clear correlation was found between the radioactive cesium contamination of eel's muscle and body weight, and it was presumed that the radioactive cesium accumulated in eel along with growth. Benthic fish such as goby were also contaminated with the radioactive cesium. Nonetheless, the contamination level of radioactive cesium of fish inhabiting Tokyo Bay was lower than the Edogawa River water system. The radioactive cesium contamination of shellfish in Tokyo Bay was not observed despite the sediment contamination. However, in the early of the FDNPP accident, there was a clear contamination of ^{110m}Ag from the internal organs of the shellfish living in the sediment of Tokyo Bay. The fact that the ^{110m}Ag was detected from shellfish in Tokyo Bay, it probably suggests that the non-volatile radionuclides such as uranium and plutonium also flew to the Tokyo metropolitan area due to the FDNPP accident.

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Table 1 Concentration of radioactive cesium in fish and shellfish inhabiting Tokyo Bay.

Area	Species	Sampling Date	Activity, Bq·kg ⁻¹		
			¹³⁴ Cs	¹³⁷ Cs	¹³⁴⁺¹³⁷ Cs
Tokyo Bay	Yellowfin goby (<i>Acanthogobius flavimanus</i>)	2011/10/21	6.5	6.8	13.3
		2011/12/7	4.4	5.3	9.7
		2011/12/18	nd	0.2	0.2
		2011/12/18	14.6	19.2	33.8
		2011/12/22	1.2	1.4	2.6
	Flathead mullet (<i>Mugil cephalus</i>)	2011/8/20	1.3	1.5	2.8
		2011/12/20	1.2	1.7	2.9
		2011/12/20	5.0	8.4	13.4
	Stingray (<i>Batoidea</i>)	2011/12/7	4.8	5.7	10.5
	Marbled sole (<i>Pseudopleuronectes yokohamae</i>)	2012/4/10	2.5	1.1	3.6
		2013/5/10	nd	nd	nd
	Gurnard (<i>Chelidonichthys spinosus</i>)	2012/4/10	1.3	2.0	3.3
	Surfperch (<i>Ditrema temmincki</i>)	2013/5/31	nd	3.7	3.7
	Japanese rockfish (<i>Sebastes inermis</i>)	2013/5/10	0.6	1.0	1.6
		2013/5/10	nd	nd	nd
	Japanese sea bass (<i>Lateolabrax japonicus</i>)	2011/12/22	2.1	3.3	5.4
		2011/12/22	1.5	3.7	5.2
		2012/11/25	1.2	1.7	2.9
		2012/11/25	nd	nd	nd
		2012/11/25	nd	2.3	2.3
		2013/5/10	0.7	1.3	2.0
		2013/5/12	nd	nd	nd
	Gafftopsail goby (<i>Myersina filifer</i>)	2012/6/27	nd	nd	nd
	Japanese whiting (<i>Sillago japonica</i>)	2012/6/28	nd	0.7	0.7
		2013/5/31	nd	nd	nd
	Big-eyed flathead (<i>Suggrundus meerdervoortii</i>)	2012/6/27	nd	nd	nd
		2013/5/31	nd	nd	nd
	Red barracuda (<i>Sphyraena pinguis</i>)	2011/10/24	2.3	3.2	5.5
		2013/5/10	nd	nd	nd
		2013/5/10	nd	0.3	0.3
	Chub mackerel (<i>Scomber japonicus</i>)	2011/10/23	0.2	0.5	0.7
		2013/5/10	nd	0.1	0.1
	Spotnape ponyfish (<i>Nuchequula nuchalis</i>)	2012/6/28	nd	nd	nd
Japanese sardine (<i>Sardinops melanostictus</i>)	2012/4/10	nd	nd	nd	
Japanese jack mackerel (<i>Trachurus japonicus</i>)	2012/9/8	nd	nd	nd	
Japanese anchovy (<i>Engraulis japonicus</i>)	2012/9/8	nd	nd	nd	
Area	Species	Sampling Date	Activity, Bq·kg ⁻¹		
			¹³⁴ Cs	¹³⁷ Cs	¹³⁴⁺¹³⁷ Cs
Tokyo Bay	Swimming crabs (<i>Portunidae</i>)	2012/4/2	2.8	2.9	5.7
	Manila clam (<i>Ruditapes philippinarum</i>)	2011/10/1	nd	nd	nd
		2012/4/10	nd	nd	nd
	Hard clam (<i>Mercenaria mercenaria</i>)	2011/8/20	nd	nd	nd
		2011/10/30	0.5	1.0	1.5
		2012/4/2	nd	nd	nd
	Mirugai (<i>Tresus keenae</i>)	2011/10/1	nd	nd	nd
		2011/10/1	0.5	0.7	1.2
	Pen shell (<i>Atrina pectinata</i>)	2011/10/1	nd	nd	nd

Table 2 Concentration of radionuclides in the organs of fish and shellfish inhabiting Tokyo Bay.

Classification	Species	Sampling Date	Organs	Activity, Bq·kg ⁻¹			
				¹³⁴ Cs	¹³⁷ Cs	¹³⁴⁺¹³⁷ Cs	^{110m} Ag
Fish	Flathead mullet	2011/8/20	musscle	1.3	1.5	2.8	nd
			viscera	12.9	14.6	27.5	nd
			gill	1.0	1.3	2.3	nd
			skin	nd	nd	nd	nd
	Flathead mullet	2011/12/20	musscle	4.9	8.4	13.3	nd
			viscera	2.9	3.4	6.3	nd
	Stingray	2011/12/7	musscle	4.8	5.7	10.5	nd
			viscera	1.2	1.2	2.4	nd
			liver	0.2	0.3	0.5	nd
			gill	1.2	1.3	2.5	nd
	Red barracuda	2011/10/24	skin	4.0	4.7	8.7	nd
			musscle	2.3	3.2	5.5	nd
			viscera	0.2	0.1	0.3	nd
			gill	nd	nd	nd	nd
			bone	0.7	0.9	1.6	nd
Shellfish	Mirugai	2011/10/1	adductor musscle	1.7	2.2	3.9	nd
			incurrent siphon	nd	nd	nd	1.0
	Mirugai	2011/10/1	adductor musscle	0.5	0.7	1.2	nd
			viscera	0.9	1.0	1.9	1.0
			incurrent siphon	nd	nd	nd	nd
	Pen shell	2011/10/1	adductor musscle	nd	nd	nd	0.4
			viscera	nd	nd	nd	2.5

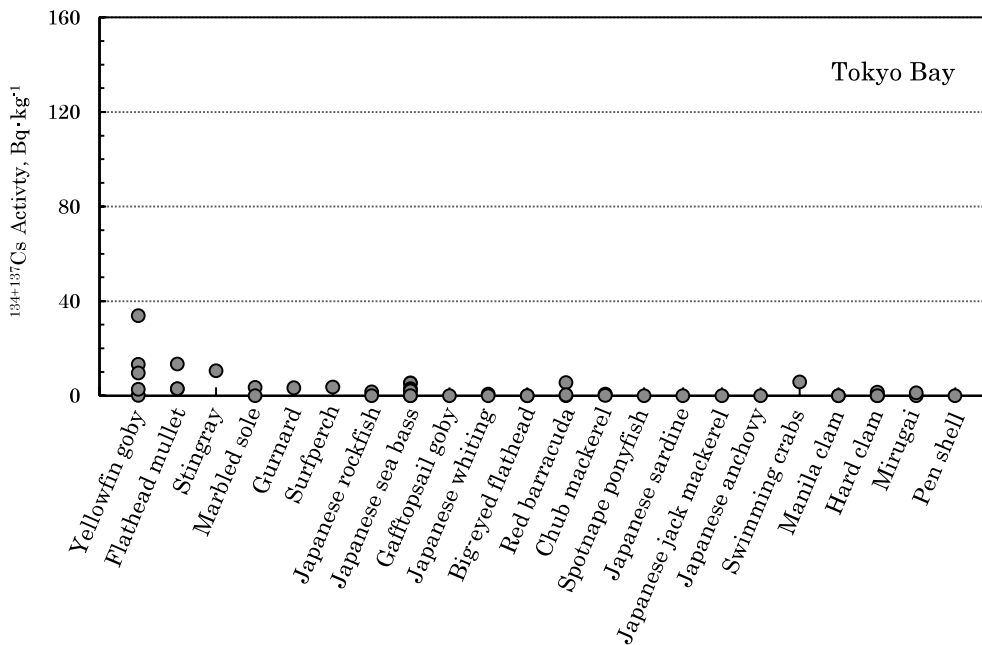


Fig. 1 Activity of ¹³⁴⁺¹³⁷Cs in fish and shellfish inhabiting Tokyo Bay.

Table 3 Concentration of radioactive cesium in fish inhabiting Edogawa River.

Area	Species	Sampling Date	Activity, Bq·kg ⁻¹		
			¹³⁴ Cs	¹³⁷ Cs	¹³⁴⁺¹³⁷ Cs
Edogawa River	Japanese eel (<i>Anguilla japonica</i>)	2013/3/9	54.5	93.0	147.5
		2013/4/28	33.2	69.3	102.5
		2013/4/28	29.1	68.3	97.4
		2013/4/29	44.7	84.9	129.6
		2013/5/4	35.6	66.9	102.5
		2014/5/4	17.0	47.4	64.4
		2014/8/19	4.5	11.7	16.2
		2015/5/5	nd	nd	nd
		2016/10/2	0.9	5.4	6.3
		2016/10/2	1.2	5.2	6.4
	Freshwater prawn (<i>Macrobrachium</i>)	2012/6/20	10.1	14.0	24.1
		2013/6/6	8.8	18.0	26.8
		2013/6/13	12.3	22.2	34.5
		2014/6/18	3.5	9.5	13.0
		2014/6/22	3.0	8.8	11.8
		2015/6/7	2.8	10.7	13.5
		2017/7/9	0.8	6.0	6.8
	Yellowfin goby (<i>Acanthogobius flavimanus</i>)	2013/6/6	12.8	22.9	35.7
		2013/6/13	14.0	25.3	39.3
		2015/5/5	nd	nd	nd
		2015/5/29	nd	4.8	4.8
		2015/9/22	nd	nd	nd
		2015/9/27	3.2	10.7	13.9
		2016/10/2	1.2	4.8	6.0
		2016/10/2	1.0	4.8	5.8
	Channel catfish (<i>Ictalurus punctatus</i>)	2014/5/2	7.8	25.1	32.9
	Japanese common catfish (<i>Silurus asotus</i>)	2014/8/19	6.9	14.8	21.7
	Japanese sea bass (<i>Lateolabrax japonicus</i>)	2015/8/28	1.7	7.9	9.6
		2016/10/2	1.9	11.2	13.1
		2016/10/2	1.7	9.9	11.6
Japanese sardinella (<i>Sardinella zunasi</i>)	2015/8/8	nd	nd	nd	
Japanese barbel (<i>Hemibarbus barbus</i>)	2016/9/10	0.9	5.4	6.3	

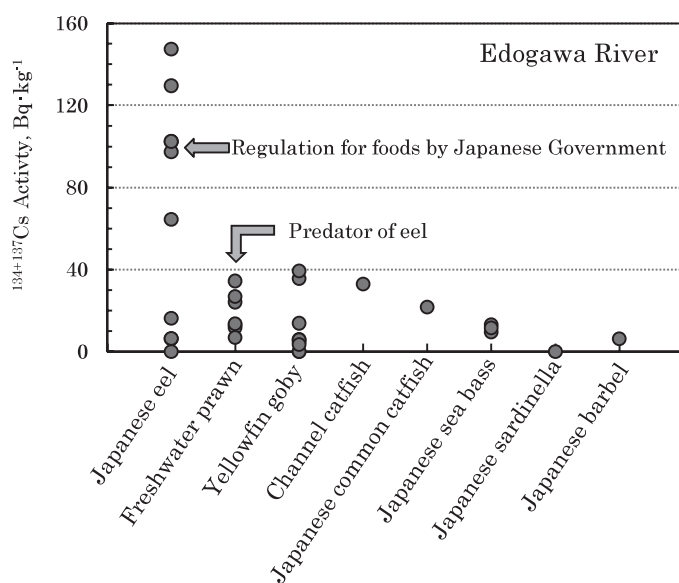


Fig. 2 Activity of ¹³⁴⁺¹³⁷Cs in fish inhabiting Edogawa River.

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Additional notes: The authors notified the Fisheries Agency of Japan about radioactive contamination of eels inhabiting Edogawa River according to this study. It was also confirmed by the Fisheries Agency that eels' radioactive cesium activity exceeded the regulation values of radiation contamination of Japanese foods. As a result, capture of the eel living in the Edogawa River ecosystem was prohibited by Tokyo and Chiba prefectures. This was also widely reported in various media in Japan.