福島第一原子力発電所事故による海洋無脊椎動物の放射性物質汚染に関する研究

[課程博士・論文博士共通]

博士学位論文要約
Summary

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論文題目 Title	Studies on radionuclide contamination of marine invertebrates by Fukushima Daiichi Nuclear Power Station accident		

The Fukushima Daiichi Nuclear Power Station (FDNPS), operated by the Tokyo Electric Power Company (TEPCO) was severely affected due to the Great East Japan Earthquake (Mw 9.0) and subsequent devastating tsunami on 11 March 2011. This accidental release of anthropogenic radionuclides (mostly iodine-131, cesium-134 and -137; ^{131}I, ^{134}Cs and ^{137}Cs) resulted in severe elevations of these radionuclides in marine organisms in the coastal areas of Fukushima and adjacent prefectures. Since the FDNPS accident, there are many studies have been conducted on the contamination of radiocesium to marine biota, seawater and sediments. Higher ^{137}Cs concentrations in the ocean surface and sediments were detected in the area to the south of the FDNPS site. However, a very few studies have been focused on invertebrates (e.g., echinoderms) contamination by radiocesium and the data are insufficient yet. Therefore, the study was aimed to investigate the radiocesium contamination in invertebrates using not only field data but also laboratory data simultaneously to improve the understanding of the contamination relevant mechanisms.

In Chapter 2, the study was aimed to estimate the spatial-temporal variation of radiocesium contamination in invertebrates caught offshore of Fukushima Prefecture over the period from 426–2726 days after the FDNPS accident. Besides, an attempt was taken to ascertain ecological half-life (T_{eco}) of ^{137}Cs and ^{134}Cs in different species of echinoderms. Seven groups of invertebrates (Arthropoda, Mollusca, Porifera, Cnidaria, Annelida, Tunicate, and Echinoderm) were collected from two locations (Yotsukura coast and Ena rocky coast) in Fukushima near FDNPS. A total of 96 species were identified among the seven groups of invertebrates. In Arthropoda, 13 species (Kishi velvet shrimp *Metapenaeopsis dalei,* Southern rough shrimp *Trachysalambria curvirostris,* Sand shrimp *Crangon uritai,* Atlantic blue crab *Callinectes sapidus,* Hermit crab *Dardanus calidus,* Crab of Japan *Philyra syndactyla,* Granulated mask crab *Paradorippe granulata,* Japanese spiny lobster *Panulirus japonicus,* Japanese Sand Shrimp *Crangon affinis*, Japanese mantis shrimp *Oratosquilla oratoria*, Japanese hermit crab *Pagurus gracilipes,* Japanese tiger prawn *Marsupenaeus japonicus* and crab *Plagusia dentipes*) were harvested. Among the marine echinoderms, six species *(*Sea urchin *Mesocentrotus nudus,* Sea potato *Echinocardium cordatum,* Spiny sand seastar *Luidia quinaria,* Starfish of Japan *Distolasterias nipon,* Basket star *Astrocladus coniferus* and Northern Pacific seastar *Asterias amurensis)* were collected. The radiocesium (^{137}Cs and ^{134}Cs) concentrations in the invertebrate samples (483 specimens) were measured using a germanium (Ge) semiconductor detector. Maximum ^{137}Cs activity concentration was observed in Arthropoda (128 Bq/kg–ww in *M. dalei*) at Yotsukura coast, whereas the concentration was highest at Ena rocky coast in Cnidaria (292 Bq/kg–ww). Among the 7 species of Arthropoda, lowest ^{137}Cs activity concentration was observed in *C. sapidus* (4.62 Bq/kg-ww). Due to the inconsistent changing trends of ^{137}Cs activity concentrations over the period, T_{eco} was not determined for 6 groups of invertebrates except echinoderms. Among the Echinoderm species, both radionuclides (^{137}Cs and ^{134}Cs) concentrations were much higher in *E. cordatum* (318 and 165 Bq/kg-ww, respectively) at Yotsukura station after 680 days of the FDNPP accident. Then, lowest concentrations were observed in *M. nudus* (3.26 Bq/kg-WW of ^{137}Cs) and *L. quinaria* (4.45 Bq/kg-WW of ^{134}Cs) after 1173–1174 days of the accident, and by 2726 days after the FDNPS accident,

radiocesium were not detected in *M. nudus* collected from Yotsukura coast. T_{eco} values of ^{137}Cs in *E. cordatum* and *M. nudus* were 358 days and 181–423 days, respectively. The results revealed that variations of spatial environment and food habit may affect on the radiocesium concentrations and ecological half-life.

In Chapter 3, the purpose of this study was to examine the temporal variations in the ^{137}Cs contamination of *M. nudus*, a sea urchin primary consumer, in the period since the FDNPS accident. To achieve this, ecological half-life (T_{eco}) of radioactive cesium in *M. nudus* were investigated over the period from 426–2726 days after the accident. To measure the T_{eco} of radiocesium (^{137}Cs and ^{134}Cs) in wild *M. nudus*, live samples were collected from 4 locations (Yotsukura coast, Yotsukura offshore, Ena rocky coast, and Ena rocky offshore) in Iwaki City, Fukushima Prefecture, that were between 35 and 50 km from the FDNPS. About 20 individual sea urchins/gonads were crushed and collected in one 100-mL plastic container to measure radioactivity using a germanium (Ge) semiconductor detector (86 specimens). Between 500 and 600 days after the FDNPS accident, the radiocesium (^{137}Cs and ^{134}Cs) activity concentrations in the gonads of sea urchins collected from the Yotsukura coastal area peaked at 139 and 105 Bq/kg-WW, respectively. Between 2012 and 2014, the decrease in the radiocesium (^{137}Cs and ^{134}Cs) activity concentrations in the gonads of the sea urchins was greater in those from the coast areas than in those from the offshore areas. The radiocesium (^{137}Cs and ^{134}Cs) activity concentrations in the gonads of sea urchins collected from the Yotsukura coast and Ena rocky coast decreased significantly (p < 0.05). By 2726 days after the FDNPP accident, the ^{137}Cs activity concentrations were 0.13–0.17 Bq/kg-DW and only ^{137}Cs was detected in the gonads of sea urchins collected from the Yotsukura coast site. T_{eco} of ^{137}Cs in sea urchin was observed as 181-423 days collected from Yotsukura and Ena rocky coast stations, respectively.

In Chapter 4, the purpose of this study was to examine the temporal variations in the ^{137}Cs contamination of *M. nudus* in a controlled environment. To achieve this, the biological half-life (T_{bio}) of ^{137}Cs in *M. nudus* were investigated for three months rearing at laboratory in 2013. During the rearing period from May 2013 to August 2013, 5 sea urchins were reared successfully and survived for between 77 and 91 days. The ^{137}Cs counts in the sea urchins were determined using gamma spectrometry in a closed-end coaxial high-purity germanium (HPGe) semiconductor detector at the Fukushima Prefectural Fisheries Experimental Station. The ^{137}Cs counts in the sea urchins were high between 29 and 44 at the start of the rearing period and then decreased gradually until days 77–91. The decreasing slopes of the ^{137}Cs counts over time were significant in samples C and D. The decreases in the ^{137}Cs counts for samples A (p = 0.220), B (p = 0.324), and E (p = 0.218) were not significant. The T_{bio} of ^{137}Cs in individual sea urchin was observed as 121-157 days in laboratory condition. When all 5 samples were plotted on a single plot, then the T_{bio} of ^{137}Cs was calculated as 181 days. The results from the generalized linear model (GLM) showed that the specific growth rate (SGR) of the sea urchins was significantly related to the rate of decrease in the ^{137}Cs counts (D, day^{-1}).

In Chapter 5, to understand the uptake mechanism of radioactive materials by sea urchin, the apparent concentration factor (ACF) was determined. The ACF measured in our study was much higher (123.1–2648) than the values reported in previous studies (ACF=10–20) indicates the lower uptake of Cs through water and thus, seawater Cs had very small effect on sea urchin contamination. Then, the ^{137}Cs activity concentration in the food of *M. nudus* in 1 day was calculated and the ^{137}Cs activity concentrations in the food of sea urchins at the Yotsukura station up to 700 days after the accident and at about 1000 days after the accident were estimated as 1.16×10^2–1.90×10^4 Bq/kg and 4.48×10^3–3.14×10^4 Bq/kg, respectively. These results therefore suggest that the changes in the ^{137}Cs activity concentrations in the sea urchins reflected the decrease in the ^{137}Cs concentrations in their food rather than the decrease in the seawater concentrations. Finally, to reduce the concentrations of radioactive materials in marine organisms in the Fukushima area, the concentrations of radioactive materials in ecosystems, including those in prey organisms, must first decrease.