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Biological half-life of radioactive cesium in Japanese rockfish *Sebastes cheni* contaminated by the Fukushima Daiichi nuclear power plant accident

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ABSTRACT

Since the Fukushima accident in March 2011 the concentration of radioactive cesium in Japanese rockfish (*Sebastes cheni*) has been decreasing slower than other fish species. The aim of this study was therefore to investigate the possibility of slow elimination rate (i.e., relatively longer T_b) as one of the reasons for the slow decrease in ^{137}Cs concentrations in Japanese rockfish (*S. cheni*). To do this, we reared twenty-three individuals of this species for a period of about 1 year, during which time we measured the ^{137}Cs concentrations and γ-ray spectra 14 times by using a high-efficiency NaI(Tl) scintillator. We then examined the relationship between the ^{137}Cs concentrations and the total length of each individual. We estimated the biological half-life (T_b, day) for each individual using the total number of ^{137}Cs counts in the energy region, and examined the effects of total length and ^{137}Cs concentration on T_b by generalized linear model (GLM). We also examined the effect of sex, total length, seawater temperature, and the ^{137}Cs concentration of seawater on temporal changes in the ^{137}Cs count reduction rate by GLM. There was no clear relationship between the corrected whole-body ^{137}Cs concentrations and the total length in females, however there was a significant positive correlation between these two variables in males. The difference between males and females may be attributable to variation in the degree of dilution because of variable growth of individuals, and suggests that the ^{137}Cs concentrations of small individuals may be greatly diluted because of faster growth. However, there was no significant difference in T_b between sexes. The mean T_b (±SD) in all individuals was 269 (±39) days; this T_b value is 2.7–5.4 times longer than past T_b values (marine fish: 50–100 days), and is thought to be one of the reasons for the slower decrease in ^{137}Cs concentrations in this species than other fish species on the coast of Fukushima. The GLM showed significant effects of both total length and ^{137}Cs concentration on T_b, which may reflect a reduction in the metabolic rate with increased body size (i.e., aging) and gradient of concentration against seawater. The GLM also showed a significant positive effect of seawater temperature on the reduction rate of the ^{137}Cs counts (D, day^{-1}). Therefore, D was clearly related to seasonal variations in the temperature of seawater, and this relationship may be attributable to changes in the metabolic rate that are controlled by variations in the seawater temperature. From these measurements, we examined the processes that control reductions in ^{137}Cs radioactivity.

1. Introduction

The Great East Japan Earthquake and tsunami that struck on 11 March 2011 caused a severe accident at the Tokyo Electric Power Company (TEPCO) Fukushima Dai-ichi Nuclear Power Plant (FDNPP) (IAEA, 2011). As a consequence of the accident, large amount of radioactive cesium (estimates for ^{137}Cs vary from 3.5 PBq according to Tsumune et al. (2012) to 27 PBq reported by Bailly du Bois et al. (2012)) had been released into the ocean from the FDNPP. The released radioactive cesium transported dominantly southward was from direct ocean discharges from the FDNPP (Aoyama et al., 2012; Tsumune et al., 2012), hence relatively high

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concentrations of radioactive cesium (both ^{134}Cs and ^{137}Cs) were also detected in a variety of marine organisms around the southern coast of Fukushima Prefecture after the accident (Nemoto et al., 2012; Shigenobu et al., 2014; Arakawa et al., 2015). Since then, radioactive cesium in seawater has been rapidly spreading and has been diluted (Aoyama et al., 2013a,b). Approximately 4 years have passed since the accident occurred, and the radioactive cesium concentrations in seawater off the coast of Fukushima Prefecture have now dropped so that they are close to pre-accident levels (0.001–0.002 Bq/L) (Kusakabe et al., 2013; Kusakabe, 2014; Oikawa et al., 2013). Concentration reductions have been observed in seaweed, cephalopods, shellfish, and crustaceans, however the concentration reduction rates have varied by taxonomic group. In addition, the concentrations have also declined in fish species that were significantly contaminated (e.g., Japanese rockfish (*Sebastes cheni*), fat greenling, (*Hexagrammos otakii*), and marbled sole (*Pleuronectes yokohamae*)) (Iwata et al., 2013; Wada et al., 2013; Sohtome et al., 2014).

Concentration reductions in fish mainly occur when radioactivity is eliminated from the gills and digestive tracts of fish (Fowler and Fisher, 2004; Kaneko et al., 2013). Furukawa et al. (2012a) recently investigated elimination from gills at the cellular level. Cs^+ is also excreted from the ionocyte of gills by the same mechanisms by which K^+ is excreted (Furukawa et al., 2012b). In contrast to freshwater fish, marine fish always excrete ions by osmoregulation, and Cs^+ may be also excreted by marine fish by this mechanism (Furukawa et al., 2012b). The difference in osmoregulation between freshwater fish and marine fish might explain the shorter biological half-life (T_b, measured in days) in marine fish relative to that for freshwater fish (Furukawa et al., 2012b). Buesseler (2012) was concerned about the long-term retention of radioactivity in demersal fish on the Fukushima coast (including Japanese rockfish, (*S. cheni*), greenling (*H. otakii*), and marbled sole (*P. yokohamae*)). Then, Fujita et al. (2014) and Watanabe (2014) have confirmed the slow reduction in concentrations in these fish. Tateda et al. (2013) and Johansen et al. (2014) suggested that the persistent elevation of ^{137}Cs in these species is likely mainly due to their sediment-associated food chains and feeding behaviors. On the other hand, previous studies have reported T_b values which is an index of the excretion rate of radioactivity in marine fish of 50–100 days (Baptist and Price, 1962; Kimura, 1984; Zhao et al., 2001). However, these T_b values may vary depending on the size of the experimental individuals and conditions, and should therefore be interpreted in the context of the experimental conditions. When the T_b is estimated by the lethal measurement method (Ishikawa et al., 2003), it should be interpreted in the context of dilution in growth and individual differences (Thomann, 1981). We need to examine temporal changes in the ^{137}Cs radioactivity in living individuals of various sizes to understand the factor of the slow reduction of ^{137}Cs concentrations in Japanese rockfish contaminated by the Fukushima accident. Johansen et al. (2014) showed that near the FDNPP, *Sebastes* spp. had higher sustained ^{134,137}Cs concentrations than almost all other fish species. This suggests that it is important to monitor *Sebastes* spp. for the health of marine species and for human consumption concerns (Hamada and Ogino, 2012).

The aim of this study was therefore to investigate the possibility of slow elimination rate (i.e., relatively longer T_b) as one of the reasons for the slow decrease in ^{137}Cs concentrations in Japanese rockfish (*S. cheni*). To do this, we reared individuals of this species and measured the ^{137}Cs concentrations and γ-ray spectra for the duration of the experimental period. We then determined the relationship between the ^{137}Cs concentration and the total length of each individual. We estimated the T_b for each individual, and examined the effects of sex, total length, and initial ^{137}Cs counts on the T_b by generalized linear model (GLM). We also examined the effect of sex, total length, seawater temperature, and the ^{137}Cs concentration of seawater on the temporal changes in the reduction rates of ^{137}Cs counts using a GLM.

2. Materials and methods

2.1. Collection of sample individuals and rearing conditions

Thirty-five individuals of Japanese rockfish (*S. cheni*) were collected in the reef area off the Tomioka coast, Fukushima Prefecture (within a 2 km radius from 37° 22′ 47″ N, 141° 05′ 49″ E, and at a depth of 20–30 m) in December 2013 (Fig. 1). Table 1 shows the number of survived individuals, total length, standard length, weight, and condition index (K) at the time of first measurement. The condition index (K) was calculated using the initial and final values of the weight (W, g) and the standard length (SL, cm) using equation of Thompson (1942) (1).

$$K = W \Big/ SL^3 \times 10^3 \quad (1)$$

In this experiment, the female individuals exhibiting more than 40 of condition index, K were egg-bearing. Then, we used the post-spawning values (total length, weight and ^{137}Cs concentration) in the following analyses. Nemoto and Ishida (2006) estimated the growth curve for individuals of this species obtained from the coast of Fukushima. By fitting the total length of the individuals tested in this experiment in this growth curve, the females were assumed to be at least 5–10 years old or older while the males were thought to be 6–10 years old or older.

To avoid damage to the bodies of the fish, all the individuals were collected by bait fishing from a small fishing boat (5 GT, gross tonnage). The 35 Japanese rockfish were immediately transported to the Fukushima Prefectural Fisheries Experimental Station, where individual identification tags were attached. All the individuals were placed into a circular tank (1940 mm diameter × 800 mm depth). Seawater for rearing was pumped from the sea at the Fukushima Prefectural Fisheries Experimental Station (Shimokajiro, Iwaki City, Fukushima), filtered through sand, and poured into the tank at a rate of about 20–30 L/min. The fish were fed Antarctic krill three times a week (about 20 g per fish on Monday, about 10 g per fish on Wednesday, and Friday). The ^{137}Cs concentrations in these krill were lower than the detection limit. The seawater temperature of the tank was measured each weekday morning at 9:00 to an accuracy of at least 0.1 °C using a calibrated digital handy thermometer (MC1000-000, CHINO Co., Tokyo). After a twenty-day acclimation period in same tank, ^{137}Cs radioactivity measurements were initiated and the rearing continued for about 1 year.

2.2. Anesthesia treatment and radioactivity measurement

In this study, anesthesia treatment was carried out to measure the ^{137}Cs radioactivity in the living Japanese rockfish (*S. cheni*) individuals. Each fish was individually transferred from the rearing tank to a 20 L plastic bucket filled with seawater to which 2-phenoxyethanol was added (500 μL/L) and anesthetized. The fish was immersed in 2-phenoxyethanol seawater until only the gills could move voluntarily. The immersion times in the 2-phenoxyethanol seawater were approximately 5 min and ranged from 4 to 7 min. After the anesthesia treatment, each individual was removed from the bucket and the sex was determined. The total length, standard length, width, height, and weight of each fish were measured, after which each fish was stored in a plastic sealed container (230 mm long × 308 mm wide × 104 mm high) with a moistened sponge (Fig. 2). The ^{137}Cs radioactivity was