Estimation of ecological half-lives of radiocesium in marine biota at the offshore of Fukushima, Japan

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A large amount of radionuclides was released by TEPCO's Fukushima Daiichi Nuclear Power Plant (FDNPP) into the environment on account of the accident that occurred here in March, 2011, and this included cesium-137 ($T_{1/2}$ = 30 y). The aim of this paper is to report the ^{137}Cs concentrations in marine biota that were contaminated due to the FDNPP accident, and to evaluate the ecological half-life (T_{eco}) in nature. The T_{eco} of ^{137}Cs in the oceanic community along the coastline of Fukushima was calculated on the basis of food monitoring data. The monitoring data were re-organized by the southern and northern sampling areas with respect to the FDNPP site. The regional comparison of ^{137}Cs contamination showed that the concentration in the southern region was higher; however, the calculated T_{eco} for each species was similar in the southern and northern regions. Furthermore, T_{eco} obtained from the field data was compared to literature values of the biological half-life (T_b) etimated under laboratory conditions. The result showed that seaweed and bivalves had similar values of T_{eco} and T_b. On the other hand, demersal fish (which live near, or on the seabed), had longer T_{eco} compared to T_b. It is conceivable that one reason for the longer T_{eco} of demersal fish is prolonged exposure due to diet.

Key Words: *radiocesium, marine environment, biota, ecological half-life, trophic level*

1. Introduction

The Great East Japan Earthquake, a magnitude 9 earthquake, triggered the TEPCO Fukushima Daiichi Nuclear Power Plant (FDNPP) accident of 11th March 2011. The subsequent tsunami caused severe destruction at the site, which led to an atmospheric discharge of radionuclides. Cesium-137 was one of the major radionuclides released into the environment by the accident. The total reported atmospheric discharge of ^{137}Cs was approximately 1.5 x 10^{16} Bq[1)]; furthermore, there was a discharge of contaminated water into the sea, increasing the total amount of ^{137}Cs to approximately 3.5 x 10^{15} Bq[2)].

The initial dispersion of waterborne ^{137}Cs occurred southward due to the (southward) current flowing along the coastline of the Fukushima Prefecture, and was eventually spread eastward by the Kuroshio Current[2)]. As a result, higher concentrations of ^{137}Cs were detected in the ocean surface water to the south of the FDNPP site and north of the Kuroshio Current[3), 4)]. Moreover, several measurements of ^{137}Cs concentrations have shown that the FDNPP accident led to the contamination of marine life[4), 5)].

Due to the long radioactive half-life of ^{137}Cs (30 y), the rate of loss in marine life is inevitably a serious matter to consider for consumers. To estimate the rate of loss in marine organisms, the biological half-life (T_b) of ^{137}Cs has been studied under controlled settings, however, the ecological half-life (T_{eco}) of ^{137}Cs in marine biota, which reflects field data, is not as well studied. Such field data are more practical and useful than controlled laboratory data as they account for the influence of existing ecological and environmental factors.

This study aims to address the dispersion trend and T_{eco} of ^{137}Cs in marine biota found offshore of the Fukushima Prefecture, based on field data. We will also compare and contrast T_b with T_{eco}, and reveal the mechanisms or trends of ^{137}Cs accumulation in the local marine biota.

2. Materials and Methods

(1) Definitions

In this paper the effective half-life in wild biota is referred to as the ecological half-life (T_{eco}), as opposed to the biological half-life (T_b).

a) Ecological half-life (T_{eco})

T_{eco} indicates the half-life of a certain radionuclide in organisms at a given site in which the contamination has occurred; for ^{137}Cs in marine organisms, it is the time required to achieve a 50% reduction in the average ^{137}Cs concentration in tissue. In this case, the rate of uptake is not controlled, and the loss rate reflects all of the possible processes, including biological elimination and radioactive decay. In this study, T_{eco} focuses on half-life in organisms and is distinguished from general environmental half-life concept, which includes non-living components of the environment.

b) Biological half-life (T_b)

T_b is the time period required for a 50% reduction of a radionuclide in a distinct species by biological elimination, as estimated in laboratory experiments or under controlled conditions. In these settings the radionuclide (^{137}Cs) is generally fed to the target organisms in water, or a digestible form. Once the concentrations reach equilibrium, the organisms are transferred to an uncontaminated environment to monitor the loss rate.

(2) Sampling areas and data selection

The concentration data were collected from the food monitoring data provided on the website of Fukushima Prefecture[5)]. The sampling period in this study was 535 days after the TEPCO accident. The monitoring data, used for the present study, were gathered from two sampling areas: the southern (Iwaki-shi), and the northern (Shinchi-cho, Soma-shi, Minami Soma-shi) parts with respect to the FDNPP site.

A total of 8 species were investigated, all of which were collected in the southern part, whereas only 5 species were collected in the northern part (see Table 1). Data values below detection limits were omitted from the present data. In addition, small portions of data which were sampled discontinuously in time were also omitted—for sand crab, data after the 400th day were excluded due to a break of approximately 200 days, for which the trend data was uncertain.

(3) Calculation and statistical analysis

To calculate each T_{eco}, the data were plotted on a logarithmic scale where an exponential trend results in a linear best-fit. The data were plotted with the number of days after the accident along the x-axis, and the ^{137}Cs activity along the y-axis. Using the calculated linear fits, T_{eco} was estimated.

(4) Data comparisons

In order to compare the ^{137}Cs contamination in the southern and the northern areas, the ^{137}Cs concentrations on the 100th day were estimated for each of the 8 species using the computed exponential equation; the 100th day was chosen as an arbitrary point of comparison.

In addition, T_{eco} and T_b from references were compared among organisms of similar groups, including seaweeds, bivalves, and demersal fish. For

Table 1 Sample details and ecological half-lives of ^{137}Cs for each species caught offshore of Fukushima.

Species	Area*	Number of samples	Correlation coefficient (r)	Correlation p-value	T_{eco} (days)
Arame	South	19	-0.854	<0.0001	50
	North	ND	----	----	----
Sakhalin Surf clam	South	43	-0.942	<0.0001	85
	North	ND	----	----	----
Sand crab	South	4	-0.949	0.0515	52.8
	North	4	-0.956	0.0445	66
Whitebait	South	39	-0.729	<0.0001	53
	North	ND	----	----	----
White rockfish (*Shiromebaru*)	South	47	-0.415	0.0037	258
	North	25	-0.237	0.2546	435
Japanese jack mackerel	South	21	-0.604	0.0037	226
	North	19	-0.120	0.6252	1028
Marbled sole	South	92	-0.229	0.0282	555
	North	113	-0.293	0.0016	438
Japanese common skate	South	100	-0.173	0.0849	648
	North	78	-0.162	0.1560	1007

ND: Not available or insufficient data

*Sampling area with respect to the FDNPP site

Table 2 Regional comparison of ^{137}Cs concentrations in marine life using the computed exponential equations, and the ecological half-life of ^{137}Cs between the common species.

Species	^{137}Cs concentration on the 100th day (Bq/kg-wet)			T_{eco} (days)		
	South	North	Ratio	South	North	Ratio
Sand crab	189.2	29.5	6.4	53	66	0.80
White rockfish	422.0	105.8	4.0	258	435	0.59
Japanese jack mackerel	49.8	27.5	1.8	226	1028	0.22
Marbled sole	122.9	49.1	2.5	555	438	1.27
Japanese Common skate	236.3	62.9	3.8	648	1007	0.64

example, Arame (a species of kelp) was compared with *Fucus* (a brown algae) as both are classed as seaweeds.

3. Results and discussion

The calculated T_{eco} and p-values for correlations between the ^{137}Cs concentrations (log converted) and time are given in Table 1. Using these data, further analysis was carried out as follows.

(1) Area comparison of the ^{137}Cs concentrations

The area comparison of the ^{137}Cs concentrations on the 100th day showed that the ratio of the south and north values ranged from 1.8 to 6.4 (see Table 2). Based on these 5 comparisons the marine life in the south contained higher ^{137}Cs concentrations than those in the north. It should be noted that the strength of the correlation of the exponential trend lines varied among species and between the two regions. Consequently, the regionally different ^{137}Cs concentrations in marine biota tended to correspond to the ^{137}Cs dispersion trend in seawater.

(2) Area comparison of the T_{eco} values

A comparison of the T_{eco} results indicate that the ratio of the south and north concentrations ranged from 0.22 to 1.27 (see Table 2). Marbled sole (a flatfish species), and sand crab exhibited south/north ratios of 0.80 and 1.27 respectively, indicating no regional difference in T_{eco} for the same species. There may not be much difference in the environment between the two areas. As the correlation became weaker, the ratio values varied more.

(3) Comparison between T_{eco} and T_b

Seaweed and bivalve data in the field (T_{eco}) showed similar values to T_b data presented in the literature, and the ratios of the field data to the reference data[6), 7)] were 0.93 for seaweed and 1.1 for the bivalves (see Table 3). The probable causes for these results are nonmetabolic ^{137}Cs adsorption for seaweed, uptake routes, and a high turnover rate for bivalves[8), 9)].

In a previous study, ^{137}Cs uptake in seaweed was driven directly by the ^{137}Cs concentration in the surrounding water, although a variation in the ^{137}Cs uptake and T_b among species was observed[8)]. The concentration of ^{134}Cs in the seawater after the accident showed a 1000-fold decrease (from 10^8 to 10^5 Bq/m^3) from April to May 2011, where the activity ratio of ^{134}Cs to ^{137}Cs was close to 1.0[3)]. The ^{134}Cs concentration in the seawater six months after the accident was insignificant in comparison to the peak concentration—e.g. a 10^4~10^5-fold decrease at the FDNPP site[3)]. ^{137}Cs concentration in tissue may have reached equilibrium concentrations for a given concentration in seawater, so that the T_{eco} value became similar to the T_b determined from laboratory experiments.

For filter-feeding bivalves, phytoplankton is another source of ^{137}Cs uptake besides water. ^{137}Cs uptake in bivalves was caused largely by phytoplankton and this absorption from food sources led to a longer retention time[7)]. However, previous studies found a very low bioconcentration factor of phytoplankton (42 to 49)[7)]; significantly high turnover rates and a short T_b (ranging from 4.5 to 5.2 days)[7)]; and a low assimilation efficiency (ranging from 0.4 to 10%)[9)], of ^{137}Cs in mussels. When the ^{137}Cs concentration in seawater drops drastically, as with the present case, T_{eco} appears to be as short as T_b offshore of the Fukushima Prefecture. The ^{137}Cs concentration in tissue of the Sakhalin surf clam may also have reached equilibrium concentrations for a given concentration in phytoplankton and seawater. Thus, the ^{137}Cs concentration in seawater seems to be the apparent driving force in the ^{137}Cs uptake in seaweed and bivalves.

On the other hand, T_{eco} was much longer than T_b for demersal fish, with a T_{eco}/T_b ratio ranging from 6.7 to 8.5[6]. The result may be attributed to the excessively high assimilation rate (ranging from 78 to 95%)[10], and food habits[11]. The ^{137}Cs uptake from surrounding water was directly proportional to the concentration in the water. If ^{137}Cs competes with coexisting elements such as K^+ for the uptake routes, the uptake rate should also relate to the concentration of K^+. Therefore, the K^+ active transport channel is not likely used for ^{137}Cs uptake and dietary uptake is the main process in marine fish[10]. There is strong correlation between food habit (trophic levels) and concentrations of ^{137}Cs in marine fish. As trophic levels increase, ^{137}Cs concentrations in prey increase, which leads to a higher intake rate for predators. In addition, fish at a higher trophic level have a relatively slow elimination rate[11]. Consequently, the longer biological retention time of ^{137}Cs in prey leads to a longer ^{137}Cs intake period and subsequently longer T_{eco} among predatory fish, including benthic fish.

It is important to note that our 535 day sampling period may not be sufficient to determine T_{eco} for all the marine life, possibly resulting in the low correlations between time and ^{137}Cs concentration for some of the predatory fish in the present study. After the Chernobyl accident, a time lag of one to two years was observed between the peak concentration of ^{137}Cs in seawater and in fish (Japanese sea perch and cod); the concentration in seawater had decreased to the original level after half a year[12]. It was found that T_b of ^{137}Cs was longer in larger, or adult fish, due to a rate of food consumption exceeding the elimination rate (per kg-body weight), with some variations among different fish species[11]. These factors may lead to a longer time lag for predatory fish. In addition, the contamination degree and T_{eco} of migratory fishes may not represent the studied biota, depending season and migratory area. Limited sampling period possibly shades the irregularity and accurate estimiates. Thus, in order to determine representative T_{eco}, long-term field data are indispensable.

4. Conclusions

Overall, marine life had higher ^{137}Cs contaminations in the southern area (with respect to the FDNPP site) than in the northern area. This result corresponds to the ^{137}Cs dispersion trend found in previous studies. T_{eco} values of common marine species in the south and the north tend to remain in the same range despite regional differences in the ^{137}Cs contamination. T_{eco} corresponds to T_b for some seaweeds and bivalves. Conversely, T_{eco} was considerably longer than T_b among benthic fish. In the present study, T_{eco} of predatory fish tended to have low correlations, possibly due to the sampling period available to-date since the accident.

As it likely to take longer to reach peak concentrations of ^{137}Cs in larger fish, it is important to obtain data for longer sampling periods. Consequently, to understand the dynamics of ^{137}Cs at the ecosystem level, and to quantify the rate of loss of ^{137}Cs in the spectrum of marine biota that are important in the human diet, continued field data analysis is imperative.

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Table 3 Comparison of field data (ecological half-life) and laboratory data (biological half-life)

Type of marine species	Species	Parameter type	Half-life (days)	Ratio (T_{eco} / T_b)	Ref.
Seaweed	Arame (Eisenia*)	T_{eco}	50	0.93	This study
	Bladder wrack (Fucus*)	T_b	54	----	(6)
Shellfish (Bivalves)	Sakhalin surf clam	T_{eco}	85	1.13	This study
	Scallop	T_b	75	----	(7)
Demersal fish	Marbled sole (south)**	T_{eco}	555	8.54	This study
	Marbled sole (north)**	T_{eco}	438	6.73	This study
	Plaice**	T_b	65	----	(6)

*Genus of brown algae
**Belongs to Genus Pleuronectes

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