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Biokinetics of radiocesium depuration in marine fish inhabiting the vicinity of the Fukushima Dai-ichi Nuclear Power Plant

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ABSTRACT

Radiocesium (^{134}Cs and ^{137}Cs) released from the Fukushima Dai-ichi Nuclear Power Plant (1FNPP) accident contaminated the fish inhabiting the port of 1FNPP. Radiocesium concentrations in some fishes, especially rockfish, have still remained at elevated levels, while concentrations in olive flounder have decreased in 2015 to the level which is close to the Japanese regulatory limit for seafood products (0.1 kBq kg-wet^{-1}). In this study a dynamic food chain transfer model was applied to reconstruct radiocesium levels in olive flounder residing around the port area. As a result, the observed ^{137}Cs concentrations in olive flounder collected from the port could be explained by the simulated values in the fish, using the seawater level records at the port entrance. The reconstructed maximum $^{134+137}$Cs concentration in olive flounder inhabiting the port area was 72 kB kg-wet^{-1} in July 2011 and the ecological half-life (EHL) was estimated as being 180 days during the period of 2014–2015. Short term simulation which assumed that the coastal water fish swam into the port during 1 month, demonstrated that the radiocesium level in the olive flounder may become equivalent to the depurated level in the fish which were initially contaminated. This result indicated that the increase of radiocesium levels in wandering fish is unlikely to change total radiocesium concentrations in the olive flounder. In this sense, the radiocesium levels in the olive flounder of the port area can be interpreted as being convergent in 2015, regardless of the differences in their contamination histories. On the other hand, the higher ^{137}Cs concentrations in fat greenling, compared to the olive flounder, can be attributed to a history of exposure to the contaminated seawater and food at the inner area of the port, such as the shallow draft quay and seawall area. As a result of the reconstructed initial higher radiocesium concentration, constrained by exposure history at the inner area of the port, the depurated radiocesium concentration in fat greenling is still likely to be greater than the regulatory level in the port area in 2015.

1. Introduction

Radiocesium ($^{134+137}$Cs) was released to the coastal waters as a result of the accident at the Fukushima Dai-ichi Nuclear Power Plant (1FNPP) Tokyo Electric Power Company (TEPCO) on March 2011 (Tsumune et al., 2013; Katata et al., 2014). The radiocesium introduced into seawater was transferred rapidly to marine fish inhabiting the coastal water in the vicinity of the 1FNPP. As a result, the radiocesium concentration in the port fish was still elevated even four years after the accident. The radiocesium concentrations in fish collected from 1FNPP port area were particularly elevated in dark banded rockfish *Sebastes inermis* (0.5–55 kBq kg-wet^{-1}) and spotbelly rock fish *Sebastes pachycephalus* (0.2–25 kBq kg-wet^{-1}), followed by greenling *Hexagrammos otakii* (0.1–2.5 kBq kg-wet^{-1}), marble flounder *Pleuronectes yokohamae* (0.02–3.8 kBq kg-wet^{-1}), common skete *Okamejei schmidti* (0.03–0.5 kBq kg-wet^{-1}) during a 1 year period between September 2014 and August 2015 (TEPCO, 2015). With regard to olive flounder *Paralichthys olivaceus* from the port, the $^{134+137}$Cs concentrations in the fish (0.007–0.3 kBq kg-wet^{-1}) decreased to levels which were close to the Japanese regulatory level for Seafood Safety of 0.1 kBq kg-wet^{-1} (TEPCO, 2015). On the other hand, the radiocesium concentrations in coastal water

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fish were reduced to levels which were almost below the 0.1 kBq kg-wet^{-1} regulatory level within 30 km radius area, with the exception of occasional cases of higher concentrations in slime flounder *Microstomus achne* and marble flounder. For the coastal olive flounder, its radiocesium levels decreased below the Japanese regulatory level (TEPCO, 2015). However, the movement of contaminated fish out from the port area may contribute to an elevated radiocesium value in the fish which are monitored in coastal water (Povinec and Hirose, 2015; Shigenobu et al., 2015). Similarly, those fish which wander into and then out of the port may elevate the radiocesium level of fishes sampled for monitoring in coastal water (Wada et al., 2013). To clarify the safety of olive flounder as seafood, further scientific understanding of the bio-environmental kinetics of radiocesium in olive flounder which inhabit the vicinity of 1FNPP is necessary (Johansen et al., 2015).

The radioactivity monitoring for fish residing near the 1FNPP only began two years after the accident, thus the time-series record of levels in fish in the port is not well understood. The empirical modelling approach was applied to reconstruct the radioactivity levels in the marine biota (Vives i Batlle et al., 2014, 2016; Fujimoto et al., 2015), however the analysis with food chain transfer was not included in these evaluations. In contrast, for the coastal waters of the Pacific Ocean affected by the Fukushima accident, radiocesium bio-environmental biokinetics in olive flounder was better understood by the dynamic food chain model application, using seawater levels which were reconstructed by the Regional Ocean Model ling System ROMS (Tateda et al., 2015).

In this study, a dynamic biological compartment model of food chain transfer (Tateda et al., 2013) was applied to simulate the radiocesium levels in fish residing in the vicinity of the 1FNPP in order to reconstruct their ^{137}Cs concentrations from the beginning of the accident. The simulation, particularly for olive flounder in which the radiocesium bio-environmental kinetics is better understood, was carried out to clarify the factors which determined the maximum level and the rapidity of the depuration in this fish. Short term exposure simulation for the fish wandering into and out of the port was also carried out. The history of radiocesium levels for greenling in the port area was also simulated for comparative purposes.

2. Material and methods

2.1. Models used

2.1.1. Regional ocean model

The ^{137}Cs levels in seawater were simulated using the Regional Ocean Model System: ROMS. The input sources are atmospheric deposition (12 PBq) to the area under consideration (35°54′N – 40°00′N, 139°54′E – 147°00′E), and direct leakage (3.6 PBq) with continuous release from the 1FNPP (Tateda et al., 2015).

2.1.2. Biological compartment model

The dynamic biological compartment model has 12 biological compartments with a detritus sub-model consisting of four sub-compartments. In the food chain model, olive flounder is categorized as a piscivorous fish and its main food is a planktivorous fish (Kasamatsu, 1999; Tateda et al., 2015). In the biological simulation, daily ^{137}Cs concentrations in planktivorous fish, cephalopod, demersal fish as prey organisms, and olive flounder were determined by using the daily ^{137}Cs concentrations in seawater. For fat greenling, daily ^{137}Cs concentrations in planktivorous fish, invertebrate, cephalopod, and small demersal fish were determined to calculate ^{137}Cs level in this fish.

2.2. Simulation for long term trends

2.2.1. Seawater

The ^{137}Cs levels in seawater were simulated at the port mouth, the shallow draft quay, and the water intake open conduit unit area within the port of 1FNPP, and at St. T-3 11 km south coastal water (N 37°19′20″, E 140°01′35″) for a period extending from 1 March 2011 until 31 December 2014.

2.2.2. Organisms

In this study, the prey organism compositions for olive flounder and fat greenling (Table S1) were applied (Kasamatsu, 1999; Tateda et al., 2016). Cesium-137 levels were also simulated for fat greenling as a reference for fish collected from the port at the same time and under the same conditions.

2.2.3. Published data sets of concentrations in seawater and organisms

After simulation, temporal ^{137}Cs concentrations in seawater at each point were reconstructed from correction by fitting the simulated values to the measured concentrations obtained from the environmental monitoring programs and published studies. For example, ^{137}Cs concentrations in seawater at the port entrance, shallow water quay, water intake open conduit unit, and St. T-3 were selected from the monitoring report by TEPCO (2015); however, those values for the period September 2011 – March 2012 were not used because of the detector contamination in the TEPCO monitoring activity (NRA, 2013). The reported occasions of values which were below detection limit (BDL) in the port area were treated by giving them a possible value which was the median between the BDL value and the simulated value at the port entrance, and 0.0015 Bq l^{-1} for the St. T-3 as references. Measured data also taken from the coastal site St. T-3 were carried out by Aono (unpublished data).

The measured radiocesium levels in olive flounder and fat greenling collected from the port and coastal waters T-S4 (off 3 km 1FNPP) and T-S7 (off 2 km, 11 km south from 1FNPP) for routine environmental monitoring (TEPCO, 2015) were used to verify the compatibility of a theoretical explanation for the observed depuration in these species.

2.3. Simulation for the fish movement scenario

2.3.1. Seawater

To elucidate the temporal changes of the ^{137}Cs levels in olive flounder which swim into the port from adjacent coastal waters, the simulated seawater levels fitted to observed concentrations at the port entrance (starting from 1 September 2014) were used, as discussed below. For fat greenling, those seawater values at the shallow draft quay area were used. As the reference for the ^{137}Cs levels in coastal water, those of St. T-3 were used for biological simulation.

2.3.2. Organisms

For the biological simulation of olive flounder entering into the port, the ^{137}Cs uptake fluxes were derived based on the food organisms inhabiting the port entrance area. The periods of residence in the port were given as 5, 10 and 30 days for the two scenarios of 1) ingestion of food inhabiting the port and 2) without ingestion of food during their residence within the port. After their residence within a port area, the simulation was continued as the fish exited the port and moved to inhabit the St. T-3 for a period of nine months. For fat greenling, the same simulation was carried out using those values for the shallow draft quay area. The initial cesium-137 levels in fishes swimming in from outside of the port