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Radiological impact of the nuclear power plant accident on freshwater fish in Fukushima: An overview of monitoring results

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ABSTRACT

Radionuclide (^{131}I, ^{134}Cs, and ^{137}Cs) concentrations of monitored freshwater fish species collected from different habitats (rivers, lakes, and culture ponds) in Fukushima Prefecture during March 2011–December 2014 (total 16 species, $n = 2692$) were analyzed to present a detailed description of radionuclide contamination after the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, and to elucidate species-specific spatiotemporal declining trends of ^{137}Cs concentration for their respective habitats. Low concentrations of ^{131}I ($\leq$24 Bq kg^{-1}-wet) were detected from only 11 samples collected during March–June 2011, demonstrating that ^{131}I transferred to freshwater fish were not intense. In river and lake fishes, a more gradual decrease and higher radiocesium (^{134}Cs, ^{137}Cs) concentrations were observed than in culture pond fishes, which strongly implied that radiocesium in freshwater fish species was mainly bioaccumulated through the food web in the wild. During 2011–2014, percentages above the Japanese regulatory limit of 100 Bq kg^{-1}-wet for radiocesium in river and lake fish (14.0% and 39.6%, respectively) were higher than in monitored marine fish (9.9%), indicating longer-term contamination of freshwater fish species, especially in lakes. Higher radiocesium concentrations (maximum 18.7 kBq kg^{-1}-wet in *Oncorhynchus masou*) were found in the northwestern areas from the FDNPP with higher deposition. However, radiocesium contamination levels were regarded as 1–2 orders of magnitude less than those after the Chernobyl accident. Lagged increase of ^{137}Cs concentration and longer ecological half-lives (T_{eco}: 1.2–2.6 y in the central part of Fukushima Prefecture) were observed in carnivorous salmonids (*O. masou*, *Salvelinus leucomaenis*), whereas a rapid increase and decrease of ^{137}Cs concentration and shorter T_{eco} (0.99 and 0.69 y) were found in herbivorous and planktivorous osmerids (*Plecoglossus altivelis*, *Hypomesus nipponensis*) with younger age at maturity. Comparison of T_{eco} among salmonids, osmerids, and cyprinids suggests that, in addition to the fish feeding habits and life-cycles, hydraulic conditions in rivers and lakes (e.g., turnover time), which are expected to affect radiocesium concentration in prey items, are an important factor affecting the ^{137}Cs decreasing rate of freshwater fish.

1. Introduction

Some of the huge amount of radioactive fallout released after the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident that occurred on and after 11 March 2011 (Chino et al., 2011) was deposited on land (Mikami et al., 2015; Saito et al., 2015), contaminating the terrestrial and aquatic ecosystems of eastern Japan (e.g., Tazoe et al., 2012; Mizuno and Kubo, 2013; Yamamoto et al., 2014a). Radionuclide contamination, especially that by radiocesium (^{134}Cs and ^{137}Cs), was severe in areas northwest of the FDNPP (Mikami et al., 2015), where high radioactive plume transport and deposition with snowfall occurred during mid-March 2011 (Povinec et al., 2013). Consequently, radiocesium concentrations exceeding the Japanese regulatory limit for foodstuffs (100 Bq kg^{-1}-wet), which were enforced in April 2012, have often been detected from terrestrial and aquatic biota in Fukushima Prefecture and its

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vicinity (MAFF, 2015).

From 30 March 2011, the Fukushima Prefectural Inland Water Fisheries Experimental Station began monitoring radioiodine (^{131}I) and radiocesium concentrations in Fukushima's freshwater products at the instruction of the Fukushima Prefectural Government, as reported already for marine products taken off the coast of Fukushima Prefecture (Wada et al., 2013). The monitoring data, officially released by the Japan Ministry of Agriculture, Forestry and Fisheries (MAFF, 2015), have been used for the evaluation criteria for foodstuff shipments. As of May 2015, eight fish species (Japanese eel *Anguilla japonica*, Ayu *Plecoglossus altivelis*, white-spotted char *Salvelinus leucomaenis* (resident form), masu salmon *Oncorhynchus masou* (resident form), kokanee *Oncorhynchus nerka*, Japanese dace *Tribolodon hakonensis*, common carp *Cyprinus carpio*, crucian carp *Carassius* spp.) and one crustacean species (Japanese mitten crab *Eriocheir japonica*) distributed in contaminated rivers and lakes in Fukushima Prefecture are listed as restricted foodstuffs for shipment by the Japan Ministry of Health, Labour and Welfare (MHLW, 2015).

Some studies using these publicly available data showed that higher radiocesium concentrations were found in freshwater fish caught in areas with high radiocesium deposition (Mizuno and Kubo, 2013; Arai, 2014). One study showed that, in 2011, significantly higher radiocesium concentrations were found in the order of carnivorous Salmonidae, omnivorous Cyprinidae, and herbivorous Plecoglossidae (Mizuno and Kubo, 2013), which implies higher bioaccumulation in higher trophic levels, as observed in the aftermath of the Chernobyl accident (Koulikov, 1996; Sundbom et al., 2003). Another study has demonstrated that radiocesium concentrations in freshwater fish decreased considerably during the three-year period following the FDNPP accident (Arai, 2014). However, no report in the relevant literature, except for one of an ayu study in 2011 (Iguchi et al., 2013), describes inspection of the decreasing rates of radiocesium (expressed by ecological half-life, T_{eco}; Jonsson et al., 1999) in freshwater fish living in different freshwater environments (e.g., lakes and rivers) with varying degrees of contamination in Fukushima Prefecture.

Freshwater fish are well known to maintain higher plasma osmolality and consequently to have higher monovalent and divalent ion (e.g., Na^+, K^+, Ca^{2+}, Mg^{2+}) concentrations than surrounding water environments (Hickman and Trump, 1969). For that reason, they reportedly show longer biological half-lives of radiocesium (Ugedal et al., 1992) than marine teleosts (Kasamatsu, 1999), which actively excrete Cs^+ (a biochemical analog of K^+) through gill chloride cells during osmoregulation (Furukawa et al., 2012). These physiological characteristics, in association with radiocesium recycling or remobilization within a freshwater ecosystem (Comans et al., 1989) engendered long-term contamination of freshwater fish after the Chernobyl accident, especially fish living in closed lake ecosystems (Bulgakov et al., 2002). These results underscore the necessity of examining the area-specific and habitat-specific decreasing rates of radiocesium activity in freshwater fish species to predict long-term trends of radiocesium contamination and also to detect factors affecting the contamination of freshwater fish in Fukushima Prefecture. This information is expected to be useful to restart the local freshwater fisheries and recreational fishing reliably and to prevent harmful rumors in the future.

This study analyzes the radionuclide concentrations (^{131}I, ^{134}Cs, and ^{137}Cs) by monitoring surveys during 2011–2014 ($n = 2692$, including unpublished ^{134}Cs and ^{137}Cs concentrations in 2011) to present an overview of the contamination of freshwater fish of different species collected from different areas and habitats (rivers, lakes, and culture ponds) in Fukushima Prefecture, and to show species-specific spatiotemporal declining trends of radiocesium concentrations by calculating the ecological half-lives for their respective habitats. Based on the obtained results, the factors potentially affecting radiocesium concentrations in freshwater fish in Fukushima Prefecture are discussed.

2. Materials and methods

2.1. Sample collection, processing and measurement of radionuclides

Freshwater fish individuals were caught weekly by fishery workers of local fishery cooperatives for each river and lake, and by owners of fish culture ponds in Fukushima Prefecture (Table S1). Samples were identified and processed at the Fukushima Prefectural Inland Water Fisheries Experimental Station. Primarily, whole bodies (Ayu, pond smelt *Hypomesus nipponensis*, willow gudgeon *Gnathopogon caerulescens*, topmouth gudgeon *Pseudorasbora parva*, weather loach *Micropterus dolomieu*) or bodies without the head and internal organs (Japanese eel, white-spotted char, masu salmon, kokanee, rainbow trout *Oncorhynchus mykiss*, Japanese dace, barbel steed *Hemibarbus barbus*, common carp, crucian carp, smallmouth bass *Misgurnus anguillicaudatus*) were used for radionuclide measurements, but in some cases, especially when individual weights exceeded ca. 200 g, only muscle tissues were used (Japanese eel, white-spotted char, masu salmon, kokanee, rainbow trout, Japanese dace, barbell steed, common carp, crucian carp, smallmouth bass, and cultured peled *Coregonus peled*). These samples were processed according to the standard method recommended by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT, 2003). The processed samples were wrapped in plastic bags and were transported to the Fukushima Agricultural Technology Centre (total of 2586 samples during June 2011–December 2014). They were minced, packed tightly into plastic cylindrical containers (55 mm diameter, 64 mm height), weighed, measured for height, calculated for density, and wrapped in polyethylene bags.

Gamma rays from ^{131}I, ^{134}Cs, and ^{137}Cs were detected using a closed-end coaxial high-purity germanium (HPGe) detector (GC3020 with Multi Channel Analyzer Lynx system; Canberra, Meriden, U.S.A.). The counting efficiency of the HPGe semiconductor detector was calibrated using volume standard sources (MX033U8PP; Japan Radioisotope Association, Tokyo, Japan). The counting time for each sample was 2000 s. Genie 2000 software was used to analyze the respective peaks in the energy spectrum for ^{131}I (364 keV), ^{134}Cs (605 keV and 796 keV), and ^{137}Cs (662 keV). The concentration of three times the standard deviation from counting statistics was defined as the detection limit concentration, resulting in respective detection limits of ^{131}I, ^{134}Cs, and ^{137}Cs of 2.4–49 Bq kg^{-1}-wet, 4.2–14 kg^{-1}-wet, and 3.4–11 kg^{-1}-wet, depending on the sample quantity and density.

Immediately after the FDNPP accident, some samples (106 samples in March–June 2011) were measured at the Japan Chemical Analysis Center using the procedures described above. The counting efficiency of the detectors (Model GX2518; Canberra, Meriden, U.S.A.) was calibrated using volume standard sources (MX033U8PP; Japan Radioisotope Association, Tokyo, Japan).

This study analyzed the original data of all monitored freshwater fish species (^{131}I, ^{134}Cs, and ^{137}Cs concentrations with detection limits, Table S1). Publicly available data released by the Ministry of Agriculture, Forestry and Fisheries of Japan (MAFF, 2015) show only the total radiocesium concentrations for 2011. The activity concentrations of ^{134}Cs and ^{137}Cs and detection limits of ^{131}I, ^{134}Cs, and ^{137}Cs were not publicized in 2011.