Effects of the nuclear disaster on marine products in Fukushima: An update after five years

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1. Introduction

The Great East Japan Earthquake (moment magnitude 9.0) and the gigantic tsunami waves that struck northeastern Japan on 11 March 2011 (Miura et al., 2011) caused a severe accident at Fukushima Dai-ichi Nuclear Power Plant (FDNPP), owned by the Tokyo Electric Power Company (TEPCO) (IAEA, 2011). As a consequence, large amounts of 134Cs and 137Cs (hereinafter radiocesium) were later released directly into the Pacific Ocean from the FDNPP (Tsumune et al., 2012). The released amount of 137Cs was estimated as 3.5–5.9 PBq (Tsumune et al., 2013; Miyazawa et al., 2013; Aoyama et al., 2015). The leakage of extremely contaminated...
water from a cracked sidewall near the intake channel of Unit 2 in early April 2011 (Tsumune et al., 2012; IAEA, 2015) contributed greatly to radiocesium contamination of the surrounding coastal and offshore waters (Buesseler et al., 2011; Aoyama et al., 2013), although several other processes (e.g., atmospheric deposition to the ocean surface) have contributed to radiocesium contamination in the ocean as a result of the FDNPP accident (Morino et al., 2011; Tsumune et al., 2013; IAEA, 2015). Subsequently, radiocesium with physical half-lives of 2.07 y for $^{134}$Cs and of 30.17 y for $^{137}$Cs, was detected continuously from marine biota collected in the waters off Fukushima Prefecture and its vicinity immediately after the FDNPP accident (Buesseler, 2012; Buesseler et al., 2012; Wada et al., 2013; Sohtome et al., 2014), although the radiocesium concentrations in surface seawater off the coast of Fukushima Prefecture dropped exponentially in 2012, with the exception of concentrations in the FDNPP port (Aoyama et al., 2013; Kanda, 2013; Kaeniyama, 2015). Higher levels of $^{137}$Cs concentrations in seawater near the FDNPP continued until the end of 2015. The radiocesium contamination of marine biota and sediments was severer in shallow coastal waters south of the FDNPP (Kusakabe et al., 2013; Wada et al., 2013; Ambe et al., 2014), probably because the extremely contaminated water flowed mainly southward immediately after leakage from the FDNPP (Tsumune et al., 2012).

Wada et al. (2013) analyzed monitoring data of marine products obtained during April 2011—October 2012. Results showed species-specific declining trends and a wide geographical distribution of radiocesium concentrations in the waters off Fukushima Prefecture. Those results indicated that some specimens of demersal fishes (e.g., flatfishes, rockfishes) caught in coastal shallow waters after the FDNPP accident have often exceeded the Japanese regulatory limit of 100 Bq kg$^{-1}$ wet for foodstuffs (combined $^{134}$Cs and $^{137}$Cs). The results have also shown a more gradual declining trend of radiocesium concentration than those found for pelagic fish species, invertebrates (cephalopods, bivalves, gastropods, and crustaceans), and seaweed of various kinds. Results of several model studies for demersal fishes have implied that, along with the direct uptake of highly contaminated seawater, the gradual food chain transfer of radiocesium introduced to the ecosystem from the initial contamination of the seawater and continuous radiocesium uptake from the benthic food web, are the main causes of the lagged increase and gradual declining trend shown by these demersal fishes (Tateda et al., 2013, 2015, 2016; Kurita et al., 2015; Watanabe et al., 2015).

However, the exponential decreasing trends of $^{137}$Cs concentrations in prey items (e.g., benthic invertebrates) and sediments (Sohtome et al., 2014), and the gradual alteration of generation in many fish species during the five years after the FDNPP accident explain the lowered radiocesium concentrations in marine products, including demersal fishes. Actually, substantially lower radiocesium concentrations in newly born generations that did not contact the extremely contaminated seawater immediately after the FDNPP accident were found in Japanese flounder Paralichthys olivaceus (Kurita et al., 2015) and in Pacific cod Gadus macrocephalus (Narimatsu et al., 2015). In contrast, much higher radiocesium concentrations in fish species collected inside the FDNPP port were reported from several studies using data publicized by TEPCO (Wada et al., 2013; Shigenobu et al., 2014; Fujimoto et al., 2015a, b). These results suggest the necessity of a comprehensive study addressing the time-series trend and recent radiocesium contamination levels in marine products based on the enormous volume of monitoring results. Nevertheless, no report in the relevant literature describes a study that has compiled multi-species data and which has elucidated area-specific and taxon-specific trends of radiocesium concentrations in the waters off Fukushima Prefecture. There, coastal gill net and trawl fisheries started since June 2012 on a trial basis (called “trial fishing operations” after Wada et al., 2013) have gradually expanded the target areas and species according to monitoring results (Shibata et al., 2015; Yagi, 2016). The marine products landed through trial fishing operations are sold through commercial markets and are consumed by the general public (Yagi, 2016).

In this study, to present an updated detailed description of the decreasing trend of radiocesium concentrations after the FDNPP accident, we compiled and analyzed the original detailed data of monitored marine products off Fukushima Prefecture during 2011—2015, which include published data presented by Wada et al. (2013) (169 species, $n = 6462$). Based on those results, we evaluated area-specific and taxon/habitat-specific decreasing trends of radiocesium concentrations for each category, and described the spatiotemporal distribution of radiocesium concentrations in demersal fishes. We also analyzed all marine species data released by TEPCO from 2012 to 2015 to clarify the radiocesium contamination level within a 20 km radius area from the FDNPP, where trial fishing by gill net, trawling, and other fishing methods were not operated as of December 2015. Subsequently, we calculated the ecological half-lives for the respective species to evaluate the decreasing trends inside and outside of the FDNPP port. Finally, we briefly introduce the restoration process and the present situation of trial fishing operations in Fukushima Prefecture and discuss the present obstacles, risk of seafood consumption, and potential benefits of restoring Fukushima’s fisheries in the future.

2. Materials and methods

2.1. Sampling and measurement of radiocesium in marine products

Detailed sampling methods and radiocesium measurement procedures in marine products in Fukushima Prefecture were described by Wada et al. (2013). The sampling of marine products comprised two sampling frames: randomized sampling by fishing vessel and fixed sampling by research vessel. The fishing vessel samplings were performed weekly by fishery workers using various fishing methods (Table S1). The detailed locations for each fishing vessel sampling were chosen to cover a broad fishing area with a wide depth range for each fishing method. Fishery workers provided information related to sampling sites (latitude, longitude, and depth) immediately after sampling. Research vessel samplings (mainly by RV Kotaka, 59 t; RV Takusui, 30 t; and RV Iwakimaru, 189 t) were basically performed weekly by the staff of the Fukushima Prefectural Fisheries Experimental Station. They planned to collect samples from same sampling stations at the same depths (e.g., trawling depths of RV Iwakimaru: every 25, 50, or 100 m depth from 75 to 500 m depth) (Table S1). These randomized and fixed sampling frames contributed to coverage of all coastal fishing areas off Fukushima Prefecture. Less frequent samplings near the FDNPP were conducted in 2011 and 2012, when the evacuation zone with a 20 km radius from the FDNPP was set by the Japanese Government until August 2012. Samples were identified and processed at the Fukushima Prefectural Fisheries Experimental Station. During 2011—2015, 180 species ($n = 32,492$) were identified. Primarily, muscle tissues were used for radiocesium measurements, but whole bodies or other parts (gonads, muscle with skin without scales, and bodies without head and internal organs) were used in some cases (Table S1). Basically, one individual was used for radionuclide measurements for each species (70.1% of all samples), but several individuals were mixed and used as one sample if the volume of measured parts for each was small (Table S1).

Gamma rays from $^{134}$Cs and $^{137}$Cs were analyzed using a closed-end coaxial high-purity germanium (HPGe) detector at the...
means that both $^{134}$Cs and $^{137}$Cs concentrations of the sample were lower than the detection limit (month/year) were calculated for each category. Here, a sample was prepared in the plastic cylindrical container ($55 \text{ mm diameter, } 64 \text{ mm height}$). The counting efficiency of these HPGe semiconductor detectors was calibrated using volume standard sources (MX033U8PP; The Japan Radioisotope Association, Tokyo, Japan). The counting time for a sample was 2000 s. Genie 2000 software was used to analyze the respective peaks in the energy spectrum for $^{134}$Cs ($605 \text{ keV}$ and $796 \text{ keV}$) and $^{137}$Cs ($662 \text{ keV}$). A concentration higher than three times the standard deviation from counting statistics was defined as the detection limit concentration, resulting in the respective detection limits of $^{134}$Cs and $^{137}$Cs of $2.7 \pm 0.9$, and $79 \pm 26 \text{ Bq kg}^{-1}$-wet and $2.5 - 39 \text{ Bq kg}^{-1}$-wet, depending on the quantity and density of the sample. 

This study compiled and analyzed detailed original data of all monitored marine products collected from the waters off Fukushima Prefecture, including $^{134}$Cs and $^{137}$Cs concentrations with detection limits, longitude and latitude, and depth (Table S1). Publicly available data released by the Ministry of Agriculture, Forestry and Fisheries of Japan (MAFF, 2016) showed the $^{134}$Cs and $^{137}$Cs concentrations and the municipality, and the offshore waters where marine products were collected, but showed no detailed sampling location (latitude and longitude, and depth).

2.2. Spatiotemporal decreasing trend of radiocesium concentrations for each taxon/habitat

To elucidate taxon-specific and habitat-specific spatiotemporal decreasing trends, radiocesium data of marine products were first compiled for the following nine categories: demersal fishes ($98 \text{ species, } n = 24,750$), pelagic fishes ($28 \text{ species, } n = 2509$), crustaceans ($15 \text{ species, } n = 813$), cephalopods ($10 \text{ species, } n = 2545$), bivalves ($9 \text{ species, } n = 460$), gastropods ($10 \text{ species, } 735$), echinoderms ($3 \text{ species, } n = 545$), ascidians ($1 \text{ species, } n = 17$), and seaweeds ($6 \text{ species, } n = 118$). Aside from demersal/pelagic fishes (including Osteichthyes and Chondrichthyes) and seaweeds (including Phaeophyceae and Ulvophyceae), six other categories were compiled based on the taxonomic class level. Fish habitat categories (demersal/pelagic) followed those described by Wada et al. (2013). Then, data for each category were compiled for 10 areas (Fig. 1), which were divided arbitrarily to ascertain the area-specific contamination trend (Wada et al., 2013). The 10 areas were demarcated by six horizontal lines of latitude ($37^\circ 34'\text{N, } 37^\circ 44', 37^\circ 37', 37^\circ 15', 37^\circ 02', \text{ and } 36^\circ 50'$) and the intersection points of those six lines with a $50-m$ depth isoline. Subsequently, the percentages of samples with radiocesium concentration (combined $^{134}$Cs and $^{137}$Cs) higher than the Japanese regulatory limit of $100 \text{ Bq kg}^{-1}$-wet (hereinafter, $>\text{DL}\%$) and lower than the detection limit (hereinafter, $<\text{DL}\%$) within a certain time period (month/year) were calculated for each category. Here, a sample with radiocesium concentration lower than the detection limit means that both $^{134}$Cs and $^{137}$Cs concentrations of the sample were below the detection limit. As a whole, the arithmetic mean (±standard deviation, SD) detection limit of these samples measured at the Fukushima Agricultural Technology Centre ($n = 22,722$, Table S1) was $8.3 (±1.1)$ Bq kg$^{-1}$-wet for $^{134}$Cs and $7.4 (±1.0)$ Bq kg$^{-1}$-wet for $^{137}$Cs. Although a slightly higher detection limit was observed in 2011, temporal changes of the detection limit were small: annual arithmetic mean (±SD) detection limits for the years of 2011–2015 were, respectively $9.3 (±1.4)$, $8.4 (±1.2)$, $8.2 (±1.1)$, $8.2 (±1.9)$, and $8.2 (±1.1)$ Bq kg$^{-1}$-wet for $^{134}$Cs and $8.6 (±2.5)$, $7.6 (±1.0)$, $7.5 (±0.9)$, $7.4 (±0.9)$, and $7.3 (±0.9)$ Bq kg$^{-1}$-wet for $^{137}$Cs. Consequently, we judged that $<\text{DL}\%$ can be adopted as a useful indicator showing temporal changes of radiocesium concentrations in monitored marine products in Fukushima Prefecture.

Ecological half-lives ($T_{\text{eco}}$), which were used as an indicator of the decreasing trend of radiocesium in previous studies (Wada et al., 2013, 2016; Sohtome et al., 2014), were not calculated in this session because data collected by the Fukushima Prefectural Government have higher $<\text{DL}\%$ in the later monitoring period (e.g., $<\text{DL}\%$ in demersal fishes in 2015: 86.3%). Moreover, $T_{\text{eco}}$ calculation excluding $<\text{DL}$ data would engender underestimation of the decreasing trend. $T_{\text{eco}}$ calculation was performed for the data published by TEPCO (see Section 2.3.). Instead, spatiotemporal changes in radiocesium activity concentrations in demersal fish species are depicted by bubble charts for each year during 2011–2015 to address the decreasing trend clearly with spatial variations.

2.3. Comparison of radiocesium contamination levels inside and outside of the port of the FDNPP

To compare the radiocesium contamination levels of marine products inside and outside of the FDNPP port, all publicly available data released by TEPCO (TEPCO, 2016) were compiled (87 species, $n = 5458$, Table S2). On a monthly basis, TEPCO has measured the
134Cs and 137Cs concentrations in marine species collected within a 20 km radius from the FDNPP from April 2012, and those inside the port from October 2012. Detection limits of the TEPCO data within a 20 km radius published from December 2013 (mean ± SD in 134Cs and 137Cs: 3.8 ± 1.1 and 3.7 ± 0.5 Bq kg⁻¹-wet, respectively) were lower than those measured by the Fukushima Prefectural Government (8.3 ± 1.1 and 7.4 ± 1.0 Bq kg⁻¹-wet, respectively). Therefore, the TEPCO data were advantageous for evaluating the radiocesium decreasing trends inside and outside the FDNPP port.

To ascertain whether decreasing trends of radiocesium of marine species inside and outside of the port of FDNPP were statistically significant or not, a single-component exponential model fitted for 137Cs concentrations in each species was examined using software BellCurve (Excel ver.2.00; Social Survey Research Information Co. Ltd.). Correlation between the 137Cs concentration (after conversion to natural log scale) and the number of days since the nuclear accident was tested for the dataset considering the detection limit (DL-shifted dataset) as follows. First, the number of data points (N) was counted monthly for each species. Then the top NDL data were omitted from the monthly compiled data. The remaining data were used as the DL-shifted dataset for statistical analyses. When NDL exceeded half the number of data in a certain month, no data were used for the month. Because of the probability that log-transformed 137Cs concentrations of a species in a limited time period are expressed by normal distribution curves, as shown for fat greenling Hexagrammos otakii off the coast of Fukushima Prefecture (Shigenobu et al., 2014), the original dataset excluding the data below the detection limit would have truncated normal distributions, consequently leading to overestimation of the arithmetic mean and underestimation of the fitted slope. In contrast, the DL-shifted dataset with the same median of the original dataset can overcome these problems.

The fitted single-component exponential model is expressed as \( A_t = A_0 e^{-\lambda t} \) where \( A_t \) and \( A_0 \) respectively represent 137Cs concentrations at times \( t \) (d) and 0, and where \( \lambda \) is the decreasing rate constant (d⁻¹) that allows the calculation of the effective ecological half-life \( (T_{eff} = \ln 2 / \lambda) \) or ecological half-life \( (T_{eco} = \ln 2 / \lambda_{eff}) \), which are calculated, respectively, from observed or decay-corrected data. In addition, \( t \) is the number of days from the initial date of the FDNPP accident of 12 March 2011, when the first hydrogen explosion occurred at Unit 1 of the FDNPP (Wakeford, 2011). In the \( T_{eco} \) calculation, 137Cs concentrations were corrected for physical decay from 12 March 2011.

To test statistically significant differences of declining slopes and intercepts inside and outside of the port of FDNPP, regression analysis followed by analysis of covariance (ANCOVA) was applied to the DL-shifted dataset of 12 species. The number of 137Cs data in each area amounted to 10 or more (Table 1).

3. Results

3.1. Monitoring results overview

Fig. 2 shows the radiocesium (134Cs and 137Cs) concentrations in marine products monitored during April 2011–December 2015 (n = 32,492). All detailed data were compiled for presentation in Table S1. Immediately after the FDNPP accident, over half of the samples exceeded the Japanese regulatory limit of 100 Bq kg⁻¹-wet (Fig. 2b); some samples even exceeded 1000 Bq kg⁻¹-wet (Fig. 2a). Thereafter, radiocesium concentrations have decreased continuously and substantially with time. Only four samples exceeded the Japanese regulatory limit in 2015 (Fig. 2a), resulting in the lower > RL% in later years (0.86% in 2014, 0.05% in 2015) (Fig. 2b). In contrast, only a small percentage of samples were found to be below the detection limit immediately after the FDNPP (>DL%: 14.9% in 2011). Results show that >DL% increased gradually with time, reaching 89.5% in 2015.

3.2. Taxon/habitat-specific and area-specific trends

Fig. 3 shows taxon/habitat-specific trends in the values of >RL% and <DL% for each year during 2011–2015. In 2011, >RL% in each category were higher than in the following four years, although the percentages varied greatly from 1.8% in cephalopods to 63.3% in seaweeds. In 2012, a drastic decrease of >RL% was found in all categories except for demersal fish. The percentage became 0 in four categories (crustaceans, seaweeds, cephalopods, and gastropods). The decreasing trend of >RL% in demersal fish was more gradual compared with that in other categories, but it fell below 5% in 2013 and became quite low in 2014 (1.1%) and 2015 (0.06%). In contrast, <DL% in all categories were lower in 2011. They increased gradually in 2012 and later years. Demersal fish showed the slowest increase of <DL%, but the figure reached 86.3% in 2015.

Fig. 4 portrays area-specific trends in >RL% and <DL% of demersal fish and all species in each year during 2011–2015. As a whole, >RL% and <DL% of demersal fish were, respectively, higher and lower than those of all species in all 10 areas. These trends were particularly evident in the five shallower areas, especially in areas D and E. In 2011, >RL% in demersal fish showed the highest 84.9% in area E, and exceeded 50% in four other areas (B, C, D, and I). In contrast, >RL% were less than 20% in areas F and G. Subsequently, >RL% in demersal fish decreased constantly in 2012 and later. In 2013, the values were less than 3%, except for values of three areas (C, D, and E). The decreasing trends in areas C and D were more gradual, but those decreased respectively to 0.6% and 0.2% in 2015. An opposite trend was found for <DL%. In 2011, the <DL% of demersal fish was less than 5%, except for three offshore areas (F, G, and H), which showed more than 10%. In 2012 and later, <DL% increased gradually in all areas. It exceeded 80% in five offshore areas (F, G, H, I, and J) in 2014. The increasing trend was more gradual in areas C, D, and E, but <DL% exceeded 70% in these areas in 2015.

Fig. 5 depicts the spatial distributions of radiocesium concentrations in demersal fish in each year during 2011–2015. In 2011 and 2012, radiocesium concentrations higher than 100 Bq kg⁻¹-wet were observed in almost all areas off Fukushima, but higher concentrations (>1000 Bq kg⁻¹-wet) were found more frequently in the shallow waters south of the FDNPP. The <DL data were found sporadically in the northern deeper areas off Fukushima Prefecture. In 2013, almost all data over 100 Bq kg⁻¹-wet were detected from the southern shallow waters from the FDNPP. Furthermore, <DL data were observed frequently in all offshore areas. In 2014 and 2015, data over 100 Bq kg⁻¹-wet were found more sporadically in the shallower areas south of the FDNPP, whereas <DL data were frequently observed in all areas, even in areas near the FDNPP.

3.3. Comparison of 137Cs concentrations inside and outside of the FDNPP port

Fig. 6 presents the 137Cs concentrations of 12 species collected inside and outside (within a 20 km radius) of the FDNPP port. They showed statistically significant regression slopes in both areas. All the TEPCO data are shown in Table S2. Extremely higher 137Cs concentrations (maximum 480 kBq kg⁻¹-wet in fat greenling) were observed in fish at the FDNPP port. Higher 137Cs concentrations (>500 Bq kg⁻¹-wet) were observed from a 20 km radius area, but they never exceeded 1100 Bq kg⁻¹-wet, except for one fat greenling sample in August 2012 (16,000 Bq kg⁻¹-wet).

Decreasing trends of 137Cs concentrations were found for almost...
all species both inside and outside of the FDNPP port, reflected in the significantly negative exponential functions depicted in Fig. 6 and Table 1. Statistical results of all species described in Table S3 show that $T_{\text{eco}}$ calculated from the statistically significant regression were 87 d–567 d inside the port (geometric mean: 218 d) and 204 d–1639 d outside the port (geometric mean: 386 d).

Among 12 species (Fig. 6), statistically significant differences of regression slopes were detected for four species (Table 2). Among these, three species (fat greenling, marble flounder Pseudopleuronectes yokohamae, and red stingray Dasyatis akajei) showed a faster decrease in the FDNPP port than in the outside area, resulting in shorter $T_{\text{eco}}$ (88–194 d) than in fish from a 20 km radius area (254–423 d). Only Japanese flounder showed a statistically slower decrease in the FDNPP port, resulting in longer $T_{\text{eco}}$ for fish in the port (321 d) than in the 20 km radius area (241 d). No significant difference of regression slopes calculated for inside and outside areas in the FDNPP port were found for eight species which showed $T_{\text{eco}}$ of 186–605 d. Intercepts ($A_0$) of regressions of all these eight species were statistically higher in the FDNPP port than in the 20 km radius area.

4. Discussion

4.1. Radiocesium contamination levels of marine products, especially for demersal fish

Based on results obtained from a total of 37,950 data measured by Fukushima Prefecture and TEPCO during 2011–2015, it is readily apparent that radiocesium contamination levels of marine products in Fukushima Prefecture have decreased dramatically during the five years following the FDNPP accident (Fig. 2). Several previous reports have described that, compared with pelagic fish and other taxonomic categories (e.g., cephalopods, bivalves, and seaweeds), a more gradual decreasing trend of radiocesium concentrations was found for demersal fish species, especially those distributed in shallow coastal areas south of the FDNPP (Buesseler, 2012; Tateda et al., 2013, 2016; Wada et al., 2013). These trends were supported by our data (Figs. 3 and 4). However, our results added further evidence that, even in demersal fish, contamination levels have decreased significantly during the five years since the FDNPP

<table>
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<tr>
<th>Family</th>
<th>Species</th>
<th>Area</th>
<th>$n^a$</th>
<th>$R^2$</th>
<th>$P^b$</th>
<th>$A_0$ (Bq kg$^{-1}$ wet)</th>
<th>$\lambda_{y}(d^{-1})$</th>
<th>$T_{\text{eco}}(d)$</th>
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<td>&lt;0.001</td>
<td>2.77 x 10^2</td>
<td>0.00294</td>
<td>235 241</td>
</tr>
<tr>
<td></td>
<td>Marble flounder</td>
<td>FDNPP port</td>
<td>188</td>
<td>0.24</td>
<td>&lt;0.001</td>
<td>4.47 x 10^4</td>
<td>0.00364</td>
<td>190 194</td>
</tr>
<tr>
<td></td>
<td>(Pseudopleuronectes yokohamae)</td>
<td>20 km radius</td>
<td>263</td>
<td>0.48</td>
<td>&lt;0.001</td>
<td>1.81 x 10^2</td>
<td>0.00200</td>
<td>346 357</td>
</tr>
<tr>
<td></td>
<td>Common skate</td>
<td>FDNPP port</td>
<td>45</td>
<td>0.40</td>
<td>&lt;0.001</td>
<td>7.39 x 10^3</td>
<td>0.00282</td>
<td>246 251</td>
</tr>
<tr>
<td></td>
<td>(Okamejei kenojei)</td>
<td>20 km radius</td>
<td>430</td>
<td>0.74</td>
<td>&lt;0.001</td>
<td>6.12 x 10^2</td>
<td>0.00234</td>
<td>296 304</td>
</tr>
<tr>
<td></td>
<td>Red stingray</td>
<td>FDNPP port</td>
<td>36</td>
<td>0.25</td>
<td>0.0020</td>
<td>1.23 x 10^3</td>
<td>0.00797</td>
<td>87 88</td>
</tr>
<tr>
<td></td>
<td>(Dasyatis alajei)</td>
<td>20 km radius</td>
<td>35</td>
<td>0.29</td>
<td>&lt;0.001</td>
<td>1.14 x 10^2</td>
<td>0.00170</td>
<td>408 423</td>
</tr>
</tbody>
</table>

\[ \text{a} \text{ represents the number of samples used for statistical analysis (DL-shifted dataset).} \]

\[ \text{b} \text{ P. probability. All data show statistical significance (}P < 0.05\). \]

Fig. 2. Comprehensive results of monitoring during 2011–2015: (a) radiocesium concentration \(134\text{Cs} + 137\text{Cs}\) in Bq kg$^{-1}$-wet; (b) monthly changes of percentage of samples higher than the Japanese regulatory limit of 100 Bq kg$^{-1}$-wet (open circle) and those below the detection limit (open diamond). Gray and black vertical bars respectively show the numbers of samples below the regulatory limit and above the regulatory limit.
This situation was quite different from that of freshwater fish in areas northwest from the FDNPP, for which radiocesium contamination of longer duration was predicted (Wada et al., 2016) presumably because of the continuous uptake of contaminated food in rivers and lakes (Matsuda et al., 2015; Tsuboi et al., 2015) and less physiological activity to excrete K⁺ (biochemical analog of Cs⁺) during osmoregulation (Watanabe and Kaneko, 2015). Actually, >RL% of demersal fish in areas C, D, and E decreased respectively to 0.6%, 0.2%, and 0% in 2015, although <DL% in areas C–E (73.7%) remained lower than in seven other areas (94.6%) in 2015. Furthermore, it is noteworthy that almost all samples (84.4%) in area C were collected from within the 20 km radius from the plant (Table S1), which indicates that, even within that area, where trial fishing had not been conducted as of December 2015, radiocesium contamination levels in marine products have decreased almost to less than the Japanese regulatory limit. These results agree well with TEPCO data obtained from outside of the FDNPP port, for which only a small fraction of sample organisms (0.38%) exceeded 100 Bq kg⁻¹-wet in 2015. The maximum radiocesium concentration in TEPCO data (260 Bq kg⁻¹-wet in red stingray) outside of the port was comparable to that observed from monitored data (220 Bq kg⁻¹-wet in Japanese rockfish Sebastes cheni) in 2015.

The time-series trends of radiocesium concentrations observed in bivalves, gastropods, seaweeds, and some pelagic larval fish (Wada et al., 2013) were well explained by a dynamic biological compartment model (Tateda et al., 2013), which showed that a quick increase was mainly attributable to the direct uptake of highly contaminated seawater immediately after the FDNPP accident and that a subsequent constant decrease was governed by natural decreasing processes in the ecosystem, primarily driven by the rapid decrease of ¹³⁷Cs concentrations in seawater. In contrast, the cause or mechanism of the slow decreasing trend in demersal fish, which was longer than the reported biological half-lives of several marine fish in Japan (19–55 d, Kasamatsu, 1999), has not been concluded yet, although a lagged increase was explained by...
the direct uptake and gradual food chain transfer of $^{137}\text{Cs}$ introduced into the ecosystem from the highly contaminated seawater immediately after the FDNPP accident (Tateda et al., 2013; Kurita et al., 2015; Watanabe et al., 2015). Several reports have described that the food web transfer of radiocesium through ingestion of contaminated prey items around the sediments (i.e., benthic invertebrates) is a necessary prerequisite to explain the slow decreasing trend in demersal fish (Sohtome et al., 2014; Tateda et al., 2015). Additional potential contamination sources (i.e., contaminated detritus and sediments) taken directly or indirectly through prey items might partially contribute to explaining the results (Tateda et al., 2013, 2015).

As described above, it is true that the trend of decrease in demersal fish was slow, but it is also true that the radiocesium concentrations decreased dramatically during the five years following the FDNPP accident (Figs. 3 and 5). Recently, by conducting rearing experiments using a benthic polychaete *Perinereis aibuhitensis*, Shigenobu et al. (2015) revealed that the concentration ratio between the polychaete and sediments (wet/wet) was low (<0.10). They also inferred that most of the sedimentary $^{137}\text{Cs}$ in the digestive system of benthic organisms would be excreted with their wastes because $^{137}\text{Cs}$ concentration in the polychaete decreased quickly after separation from contaminated sediments taken from near the FDNPP, which had lower bioavailable $^{137}\text{Cs}$ fractions (Ono et al., 2015). In addition, Sohtome et al. (2014) demonstrated that radiocesium concentrations in benthic invertebrates in the coastal waters off Fukushima Prefecture, which were lower than in many demersal fish, have decreased exponentially over time along with

![Fig. 4. Yearly changes of percentage of demersal fish samples with radiocesium concentrations ($^{134}\text{Cs} + ^{137}\text{Cs}$ in Bq kg$^{-1}$-wet) above the Japanese regulatory limit of 100 Bq kg$^{-1}$-wet (closed circle) and those below the detection limit (open diamond) for each area. Dotted lines with cross and x symbols respectively show the same data for all species.](image-url)
those in sediments. These results imply that the transfer of radiocesium from benthic preys to demersal fish is declining drastically in accordance with the decrease of radiocesium concentrations in sediments that are expected to be the major continuing contamination source. Actually, sediment $^{137}$Cs concentrations off the coast of Iwaki City (20 m depth, 37°49′59″N), where higher radiocesium concentrations of demersal fish were found (Fig. 5), decreased considerably from 4680 Bq kg$^{-1}$-dry in May 2011 to around 50 Bq kg$^{-1}$-dry in 2013 (Sohtome et al., 2014). In addition to the decreased $^{137}$Cs transfer intensity in the benthic food web, other factors such as alteration of generations (Kurita et al., 2015; Narimatsu et al., 2015), dilution of radiocesium concentrations through growth, especially in young individuals (“growth effect” after Fujimoto et al., 2015b), and seasonal migration of fish (Wada et al., 2015) also contribute to the observed decrease.

Fig. 5. Spatiotemporal changes of radiocesium concentrations ($^{134}$Cs + $^{137}$Cs in Bq kg$^{-1}$-wet) of demersal fish expressed by the area of circles in each year of 2011–2015. Red and blue circles respectively show data of >100 Bq kg$^{-1}$-wet and ≤100 Bq kg$^{-1}$-wet. Black dots show data below the detection limit. Closed stars denote the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) location. Isolines show depth.
et al., 2012; Narimatsu et al., 2015; Shibata et al., 2015) are expected to have contributed synergistically to lower radiocesium concentrations in demersal fish during the last five years.

Future studies using spatiotemporal radiocesium data as a tracer are expected to be necessary to reveal the contamination history and migration range/pattern for each species. For example, much higher > RL% values in the area south from the FDNPP than in the northern area were found for marbled flounder (> RL%: south, 71.2%; north, 1.3% in 2012) and slime flounder (> RL%: south, 46.3%; north, 4.8%), although a less-biased distribution was observed in Japanese flounder (> RL%: south, 33.7%; north, 24.8%), suggesting that the migration range of Japanese flounder is fundamentally longer, although in slime flounder, the existence of a subpopulation conducting a long spawning migration (over several hundred kilometers) as shown in barfin flounder *Verasper moseri* (Wada et al., 2014) was implied by results of a tag-recapture survey (Ishito, 1962). In addition, our data will help develop and validate models simulating the contamination history of demersal fishes in the waters off Fukushima Prefecture.

4.2. Comparison of contamination levels inside and outside of the FDNPP port

Extremely contaminated fish were found in the FDNPP port (Fig. 6), as described in previous reports (Wada et al., 2013; Shigenobu et al., 2014; Fujimoto et al., 2015a, b). Actually, some...
in short, which would migrate from the outside area after the accident and "outside of the FDNPP provided further evidence that 137Cs concentrations in marbled fish outside of the FDNPP port might indirectly reflect the continuous recruitment of young and adult individuals from outside the FDNPP port. Actually, repeated migration from the outer ocean to the shallow lagoon in Fukushima Prefecture has been reported for marbled flounder (Wada et al., 2011).

In contrast, smaller variation of 137Cs concentrations (Fujimoto et al., 2015b) and longer T_{eco} (264 d) were observed for Japanese rockfish, which showed higher 137Cs concentrations in 2015. These results strongly suggest that migrations between areas inside and outside of the port were less frequent for the rockfish, although lower 137Cs concentration (66 Bq kg⁻¹-wet) in July 2013 indirectly suggests recruitment from outside areas. The sedentary life-style with strong site-fidelity of Japanese rockfish was elucidated using acoustic telemetry (Mitamura et al., 2009, 2012). In addition, the longer life span (max. 13 years old) and slower growth of Japanese rockfish (reaching 25 cm total length in 5 years and 30 cm in 10 years; Nemoto and Ishida, 2006) than that of other fish species (Kusakabe et al., 1994; Nemoto et al., 2007), these effects might be more conspicuous in some species in the port. A similar process can be applicable to marble flounder (and probably to black rockfish Sebastes schlegelii, black seabream Acanthopagrus schlegelii, and nibe croaker Nibea mitsukurii) because the large variation of 137Cs concentrations in marbled flounder from the FDNPP port might indirectly reflect the continuous recruitment of young and adult individuals from outside the FDNPP port. Actually, repeated migration from the outer ocean to the shallow lagoon in Fukushima Prefecture has been reported for marbled flounder (Wada et al., 2011).

Table 2

Statistical results of analysis of covariance of 12 species collected inside and outside of the FDNPP port.

<table>
<thead>
<tr>
<th>Species</th>
<th>Statistical comparison</th>
<th>Type III sum of squares</th>
<th>F value</th>
<th>P</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conger myriaster</td>
<td>Regression slope</td>
<td>0.156</td>
<td>0.333</td>
<td>0.57</td>
</tr>
<tr>
<td>Sebastes cheni</td>
<td>Regression slope</td>
<td>0.0567</td>
<td>0.0376</td>
<td>0.85</td>
</tr>
<tr>
<td>Sebastes schlegelii</td>
<td>Regression slope</td>
<td>423</td>
<td>282</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Hexagrammos otakii</td>
<td>Regression slope</td>
<td>348</td>
<td>146</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Hemitrupites villosus</td>
<td>Regression slope</td>
<td>28.3</td>
<td>0.064</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Lateolabrax japonicus</td>
<td>Regression slope</td>
<td>35.4</td>
<td>35.2</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Acanthopagrus schlegelii</td>
<td>Regression slope</td>
<td>1.24</td>
<td>0.664</td>
<td>0.42</td>
</tr>
<tr>
<td>Nibea mitsukurii</td>
<td>Regression slope</td>
<td>52.4</td>
<td>28.2</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Paralichthys olivaceus</td>
<td>Regression slope</td>
<td>3.33</td>
<td>3.99</td>
<td>0.0047</td>
</tr>
<tr>
<td>Pseudopleuronectes yokohama</td>
<td>Regression slope</td>
<td>27.8</td>
<td>16.1</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Okameiji kenojii</td>
<td>Regression slope</td>
<td>2.932</td>
<td>2.49</td>
<td>0.12</td>
</tr>
<tr>
<td>Dasypis okajie</td>
<td>Regression slope</td>
<td>163</td>
<td>439</td>
<td>&lt;0.001</td>
</tr>
</tbody>
</table>

a Statistical comparison for intercept (A₀) examined by analysis of covariance was applied when no significant difference of regression slopes between inside and outside of the FDNPP port was detected by regression analysis (P ≥ 0.05).

b F: Fisher—Snedecor (F) value.

c: P: probability. Boldface denotes statistical significance (P < 0.05).

Specimens of sedentary rockfishes (Sebastes cheni, Sebastes oblongus, and Sebastes pachycephalus) still showed high 137Cs concentrations of more than 10 kBq kg⁻¹-wet in 2015 (Table S2). The individuals that would have accidentally encountered the highly contaminated water immediately after the accident within or near the FDNPP port might show these higher radioactivity concentrations (Fujimoto et al., 2015a). However, it is noteworthy that the geometric mean of 137Cs concentrations decreased markedly from 3190 Bq kg⁻¹-wet in 2012 to 129 Bq kg⁻¹-wet in 2015. No highly contaminated fish (>1100 Bq kg⁻¹-wet) was collected from the 20 km radius area except for a greenling sample in August 2012. A multi-species comparison of 137Cs concentrations inside and outside of the FDNPP port was detected by regression analysis (Fujimoto et al., 2015a). However, it is noteworthy that the rates of decrease were roughly comparable inside and outside the port. A similar process can be applicable to marbled flounder and smaller variation of 137Cs concentrations in muscle tissues. These effects can also affect the rate of decrease of radiocesium concentrations in fish outside of the FDNPP port. However, because it is documented that shallower coastal areas including fishing ports can serve as a nursery habitat for some coastal fish species and because smaller and younger individuals tend to migrate and inhabit these shallow areas (Kusakabe et al., 1994; Nemoto et al., 2007), these effects might be more conspicuous in some species in the port. A similar process can be applicable to marble flounder and probably to black rockfish Sebastes schlegelii, black seabream Acanthopagrus schlegelii, and nibe croaker Nibea mitsukurii because the large variation of 137Cs concentrations in marbled flounder from the FDNPP port might indirectly reflect the continuous recruitment of young and adult individuals from outside the FDNPP port. Actually, repeated migration from the outer ocean to the shallow lagoon in Fukushima Prefecture has been reported for marbled flounder (Wada et al., 2011).

In contrast, smaller variation of 137Cs concentrations (Fujimoto et al., 2015b) and longer T_{eco} (264 d) were observed for Japanese rockfish, which showed higher 137Cs concentrations in 2015. These results strongly suggest that migrations between areas inside and outside of the port were less frequent for the rockfish, although lower 137Cs concentration (66 Bq kg⁻¹-wet) in July 2013 indirectly suggests recruitment from outside areas. The sedentary life-style with strong site-fidelity of Japanese rockfish was elucidated using acoustic telemetry (Mitamura et al., 2009, 2012). In addition, the longer life span (max. 13 years old) and slower growth of Japanese rockfish (reaching 25 cm total length in 5 years and 30 cm in 10 years; Nemoto and Ishida, 2006) than that of other fishes in Fukushima Prefecture (e.g., fat greenling: max. 9 years old, reaching 30 cm in 2 years and 40 cm in 4–5 years; Izumi, 1999) are expected to engender lower frequency of alteration of generations and lower "growth effect", thereby producing a more gradual decreasing trend. A similar process is applicable to spottbally rockfish S. pachycephalus. The strong site-fidelity of white-spotted conger in shallow coastal areas during feeding season was also revealed by acoustic telemetry (Wada et al., unpublished data), which might explain the similar trend to that of Japanese rockfish.
In contrast to the species described above, differences of $^{137}$Cs concentrations inside and outside of the FDNPP port were small in common skate Okamejei kenojai and Japanese seabass Lateolabrax japonicus (Fig. 6), although significant differences of intercepts ($A_0$) were detected (Table 2). Additional important features of these fish are that the percentages of samples collected within the FDNPP port were much smaller (9.4% and 1.7%, respectively) than those of sedentary rockfishes (S. cheni, 85.7%; S. pachycephalus, 98.0%; and S. oblongus, 100%). These results probably reflect the fact that common skate and Japanese seabass are usually distributed in open coastal waters. A few individuals opportunistically invaded into the FDNPP port and were contaminated thereafter within a short time.

A significantly more gradual decrease in the FDNPP port than in the 20 km radius area was detected only for Japanese common skate Hemitripterus villosus, although significant differences between slopes were not detected. Common traits of these fish are that their main prey items are fish (Fujita et al., 1995; Tomiyama and Kurita, 2011) and that they conduct seasonal coastal–offshore migration in association with their reproductive cycle (Shibata et al., 2015; in press). It is noteworthy that the seasonality of catchment for these fish was more evident in the port area and that it agreed well with their spawning seasons (Japanese flounder, summer; Sea raven, winter). The higher percentages of samples collected within the FDNPP port (24.2% and 36.8%, respectively) compared with previously described common skate and Japanese seabass further support the occasional or seasonal migration and inhabitation of these fish in the port. From these results, it can be speculated that these species can use the FDNPP port as a feeding habitat mainly around the spawning season. Therefore, they frequently feed upon contaminated fish, subsequently leading to the elevated $^{137}$Cs concentrations in the port compared with the outer ocean. Although no data were available for $^{137}$Cs concentrations in prey fish species (mainly Japanese anchovy Engraulis japonicus for Japanese flounder in the coastal waters off Fukushima Prefecture; Tomiyama and Kurita, 2011) in the port, the rapid contamination of migratory fish within the FDNPP port was evident from the fact that $^{137}$Cs in pelagic fish such as gizzard shad Kwonosirus punctatissimus and chum salmon Oncorhyncus keta was only detected within the port (Table S2). In contrast, the radiocesium contamination levels of pelagic fish were almost all below the detection level in the open ocean (Tables S1 and S2). For that reason, the fish feeding habits of these fish might have contributed to the more rapid $^{137}$Cs decrease and subsequently shorter $T_{reo}$ in the 20 km radius area than those of other omnivorous benthic feeders such as marbled flounder, which ingested $^{137}$Cs continuously from benthic organisms (Kasamatsu and Ishikawa, 1997; Sohtome et al., 2014).

In 2015, TEPCO covered almost the entire seafloor of the FDNPP port with mud containing cement (TEPCO, 2016). This countermeasure, as well as gill net settings at the entrance of the FDNPP port (Fujimoto et al., 2015a), can help decrease the distribution of fish and their biomass, and can also prevent continuous contamination from the benthic food web in the port. In addition, TEPCO reported the decrease of $^{137}$Cs concentrations in seawater in the FDNPP port after the completion of construction of an impervious seawall for groundwater in October 2015 (TEPCO, 2016). These improved circumstances might accelerate the trend of decreasing radiocesium concentrations in fish in the FDNPP port area.

4.3. Restoration process of Fukushima’s coastal fisheries and prospects

Since June 2012, trial fishing operations have resumed in the Soma area, 50 km north of the FDNPP (Wada et al., 2013). Some bottom-trawling vessels started catching octopus species of two kinds (Enteroctopus dofleini and Octopus conispadiceus) and a whelk (Buccinum isotakii) from offshore waters (>150 m depth) in areas F and G. These three species were selected because radioceium had not been detected in them at that time (Wada et al., 2013). From September 2012 and thereafter, other species were added as targets for trial fishing operations. Subsequently, the number of target species included in the positive list increased gradually to 72 species as of December 2015 (Fig. 7, Table S4). These species constitute 40% of monitored marine products in Fukushima Prefecture (180 species: potential target species for full-scale fishing). In addition, since October 2012, target fishing areas have expanded southward. All areas deeper than 150 m depth off the coast of Fukushima Prefecture have become target areas in August 2013. Subsequently, target areas have expanded to shallower areas (Shibata et al., 2015; Yagi, 2016). As of December 2015, all areas except for the 20 km radius area were target areas for gill net and boat seine fishing. All areas deeper than 90 m except for the 20 km radius area were target areas for trawl fishing. As a result, landed amounts of coastal fisheries (trawl, gill net, and boat seine) through trial fishing operations increased gradually from 76 metric tons in 2012 to 1436 metric tons in 2015, equivalent to 6.2% of the landed amount of these fisheries in 2010 (23,178 metric tons) (Fishery statistics of the Fukushima Prefectural Government, 2010–2015). The addition of target species and expansion of areas for trial fishing operations were proposed when the monitoring results fell below the regulatory limit “in a stable manner”. These proposals were finally decided by a monthly conference of directors of fisheries cooperatives in Fukushima Prefecture, which followed two-step meetings for the restoration of local fisheries (Inoue et al., 2015).

In addition to this careful three-step decision-making process, the establishment of a no-take zone within 20 km from the FDNPP and screening measurements of radiocesium concentrations by fishery cooperative staff (at least one specimen for each species/area for each operation date) before fish market auctions have ensured the safety of marine products landed through trial fishing operations. Consumers can readily access all these data through the web site of the fishery cooperative association of Fukushima Prefecture.

However, there exists a negative list for prohibited species for shipment, compiled by the Japanese Government (Table S4). Except for larval sand lance Ammodytes japonicus, which was prohibited not only for shipment but also for intake from April 2011 through June 2012 because of the high radiocesium concentrations

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**Fig. 7.** Monthly changes of number of target species for trial fishing operations (black line) and of species prohibited from shipment (gray line).
immediately after the accident (Wada et al., 2013), all species were included in the negative list for shipment when one or more samples of a species measured in 2012 or later exceeded the Japanese regulatory limit of 100 Bq kg\(^{-1}\). Recently, Okamura et al. (2016) showed using statistical methods that the probability of occurrence of demersal fish samples exceeding the strict Japanese regulatory limit of 100 kg\(^{-1}\)-wet is extremely low in 2015, as our results have validated. These results suggest strongly that the present risk of consumption of fish in Fukushima Prefecture is quite low or negligible, and that consumption of marine products landed through trial fishing, almost all of which were below the detection limit of <10 Bq kg\(^{-1}\)-wet for radiocesium, poses no additional radiological effects for consumers. As pointed out by Matsuizaki et al. (2016), the development of scientific/statistical framework that can support decision-makers to facilitate cancellation of restriction will be desirable to accelerate the restoration of Fukushima’s fisheries reliably.

Radiocesium concentrations of marine products in Fukushima Prefecture have decreased drastically during the last five years. Therefore, the scale of trial fishing operations will certainly expand in the near future. Some reports of studies have described that the waters off Fukushima Prefecture have been serving effectively as a marine protected area after the FDNPP accident because of the drastic decrease of operation intensities (Shibata et al., 2015; in press). Reportedly, when fishing operation intensity (operation hours) decreased to less than 3% in 2012, the catch per unit effort of all marine products by bottom trawling in a target area for trial fishing operations was 3.1 times higher than before the FDNPP accident (2007–2009) (Yamada et al., 2014). Increased biomass of marine products presents a hugely important advantage for reconstruction of Fukushima’s coastal fisheries. Expansion of trial fishing operations with better resource management measures is expected to be a key to restoring and developing Fukushima’s fisheries effectively. Careful monitoring must be continued to ensure the safety of marine products in Fukushima Prefecture and to forestall and contradict harmful rumors.

Acknowledgements

We thank all fishery workers in Fukushima Prefecture who caught marine products for monitoring. We acknowledge Dr. K. Namba for his assistance publicizing the monitoring data.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2016.06.028.

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T. Wada et al. / Journal of Environmental Radioactivity 164 (2016) 312–324 323
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