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# Radiocesium contamination of greenlings (*Hexagrammos otakii*) off the coast of Fukushima

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**We measured the radiocesium ($^{134}$Cs and $^{137}$Cs) contamination of 236 greenlings (*Hexagrammos otakii*) off the coast of Fukushima Prefecture in Japan, following the accident at the Fukushima Daiichi Nuclear Power Plant in March 2011. The radiocesium concentrations of greenlings caught approximately 40 km south of the power plant were significantly higher than those of greenlings caught approximately 50 km north of the power plant. The radiocesium concentrations of greenlings caught in southern waters were significantly higher in shallow than in deep waters. Meanwhile, two outlier specimens of greenlings with higher $^{137}$Cs concentrations, 16,000 Bq/kg-wet on 1 August 2012 and 1,150 Bq/kg-wet on 8 May 2013, were caught approximately 20 km from the power plant. Our calculations suggest that the probability of two such outlier specimens being found off the coast of Fukushima is exceedingly low. By contrast, extremely contaminated greenlings were frequently caught in the power plant port (geometric mean of $^{137}$Cs = 17,364 Bq/kg-wet). Our results suggest that the two outlier greenlings with higher $^{137}$Cs concentrations migrated from the power plant port. Continued close monitoring of radiocesium concentrations in the area should be done to ensure the safety of food supplies.**

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident caused by the Japan earthquake and tsunami on 11 March 2011 released a huge amount of anthropogenic radionuclides into the environment. The radionuclides were released into the ocean from atmospheric fallout and through direct release and leakage from the FDNPP. Although most of the short-lived radionuclides soon decayed to a level below the detection limit, two isotopes of radiocesium ($^{134}$Cs and $^{137}$Cs), which have relatively long half-lives (2.07 year for $^{134}$Cs and 30.1 year for $^{137}$Cs), have been continually detected in the environment since the accident. Tsumune *et al.*[1] estimated that 3.5 $\pm$ 0.7 PBq of $^{137}$Cs was released directly into the ocean from 26 March 2011 to the end of May 2011. Previous studies suggested that the highly contaminated seawater discharged from the FDNPP flowed mainly along the southern coastline of Fukushima[2–4].

Extensive monitoring of not only marine products but also seawater and seabed sediments started immediately after the FDNPP accident, with much attention focused on the radiocesium concentrations of marine products, because they are important food resources[5]. The monitoring detected high concentrations of radiocesium in marine products[6,7] and seabed sediments[8] in the southern coastal waters of Fukushima due to the flow of highly contaminated seawater in that direction. On 18 April 2011, 14,400 Bq/kg-wet of radiocesium ($^{134}$Cs + $^{137}$Cs) was detected in a specimen of Japanese sand lance (*Ammodytes personatus*) caught approximately 30 km south of the FDNPP[5]. This planktivorous pelagic fish is valuable to fisheries and an important prey item for piscivorous fish species off the coast of Fukushima[9,10]. This rapid contamination of planktivorous fish by radiocesium was also observed during the initial stage of the Chernobyl accident in 1987[11].

Cesium, which is a biochemical analogue of potassium, shows similar behaviour to potassium in organisms. Cesium absorbed by marine organisms is excreted by the potassium ion transport system during osmoregulation[12,13]. Therefore, the rapid decrease in the radiocesium concentration of seawater would have gradually reduced the contamination of marine organisms, especially coastal pelagic fish species[1–4,6,14]. Accordingly, a marine organisms as highly contaminated as the larvae of the Japanese sand lance was not reported until 1.5 years after 18 April 2011.

On 1 August 2012, however, 25,800 Bq/kg-wet of radiocesium ($^{134}Cs$ = 9,800 Bq/kg-wet, $^{137}Cs$ = 16,000 Bq/kg-wet) was unexpectedly detected in the muscle tissue of two greenlings (*Hexagrammos otakii*) caught approximately 20 km north of the FDNPP[15]. Although Tokyo Electric Power Corporation (TEPCO) had carried out an intensive investigation in the waters, such an extremely contaminated fish had not been caught until that point, and the reason for the high level of contamination in the two greenlings was unclear[15]. In order to ensure the safety of fishery products caught off the coast of Fukushima, it was necessary to reveal the reason for the contamination and to estimate the likelihood that other marine products would be similarly contaminated.

Greenlings are omnivorous, sedentary, demersal fish that are widely distributed along the coastal waters off Japan[9,16]. A previous study of tagged fish suggested that the migration range of greenlings was restricted to approximately 30 km in radius[9]. The main prey items of greenlings are benthic crustaceans and polychaetes. Therefore, it is thought that greenlings take in radiocesium from highly contaminated sediments through the benthic food web[14,17].

Greenlings are one of the most seriously contaminated fish species that have been found off the coast of Fukushima, especially south of the FDNPP. On 15 July 2011, 3,000 Bq/kg-wet of radiocesium was detected in a specimen of greenlings caught approximately 30 km south of the FDNPP[5]. According to the published datasets for radiocesium concentration of greenlings in 2011, 87 of 114 specimens (76.3%) exceeded the Japanese standard limit for contamination in foods (100 Bq/kg-wet, $^{134}Cs + ^{137}Cs$)[5]. Although the radiocesium concentrations of greenlings that were reported in published datasets varied, the temporal trend of the concentrations was slightly declining (ecological half-life: 301 days)[6], like those of other demersal fish species[5].

In the present study, we measured the radiocesium concentration in muscle tissue of individual greenlings caught at different depths in order to determine the concentration variability within this species and to measure area-specific contamination off the coast of Fukushima. In addition, we attempted to calculate the probability that outlier greenlings with higher concentrations would be found off the coast of Fukushima, using the published datasets of the Ministry of Agriculture, Forestry and Fisheries of Japan (MAFF)[5] and TEPCO[15].

## Results

**Radiocesium concentration of greenlings off the coast of Fukushima.** We measured radiocesium contamination of 236 samples of muscle tissue taken from individual greenlings caught in northern (approximately 50 km north of the FDNPP) and southern (approximately 40 km south of the FDNPP) waters from May 2012 to March 2013 (Table 1, Fig. 1). The radiocesium concentrations of individual greenlings showed that the contamination level in the northern waters is low; in this region, the radiocesium concentrations of all individuals caught after October 2012 were lower than the Japanese standard limit (n = 52, geometric mean = 12.0 Bq/kg-wet, range from not-detected to 39.2 Bq/kg-wet). Meanwhile, the radiocesium concentrations of greenlings caught in southern waters in May 2012 varied (n = 47, geometric mean = 120 Bq/kg-wet, range from 2.95 to 1,070 Bq/kg-wet). In addition, the level of contamination in southern waters was significantly higher in shallow (less than 30 m deep) than in offshore (more than 50 m deep) waters (Student's *t*-test, $t = 5.089$, $p < 0.001$). In southern shallow waters, 40 of 68 individuals (58.8%) exceeded the Japanese standard limit, but in the southern offshore waters, only 15 of 84 individuals (17.9%) exceeded the limit (Table 1).

Figures 2 and 3 show the time series trend of radiocesium concentrations for greenlings caught in the FDNPP port[15] and off the coast of Fukushima from May 2011 to December 2013[5,15]. Except for the greenlings caught in the FDNPP port, 76.3% of specimens caught in 2011, 41.2% in 2012, and 15.9% in 2013 exceeded the Japanese standard limit for radiocesium. Therefore, the radiocesium concentrations of greenlings gradually declined over time. According to the published datasets from MAFF[5] and TEPCO[15], there was no significant difference in the radiocesium concentrations of greenlings caught within 20 km of the FDNPP and those caught more than 20 km of the FDNPP in 2012 (Mann-Whitney *U*-test, $u = 14258$, $p = 0.4677$) or 2013 (Mann-Whitney *U*-test, $u = 5511$, $p = 0.3532$). However, the radiocesium concentrations of greenlings caught in the FDNPP port were extremely high (n = 83, geometric mean = 26,242 Bq/kg-wet, range from 480 to 740,000 Bq/kg-wet)[15].

**The probability of outlier greenlings with higher concentrations.** Figure 4 shows the probability that contaminated greenlings will be found, inferred from the normal distribution curve of log-transformed concentration values for $^{137}Cs$, which has a long half-life (30.1 years), and using values from the published datasets of greenlings caught within 20 km[15] and more than 20 km away[5] from the FDNPP port. Normality of the log-transformed $^{137}Cs$ concentrations from the datasets for April–December 2012 and April–December 2013 was confirmed. However, the normality of the 2012 and 2013 datasets for individual greenlings was rejected ($p < 0.05$) because a significant difference among radiocesium concentrations was confirmed within the sampling localities. Accordingly, we excluded the datasets of individual greenlings from this analysis.

The arithmetic means ± standard deviations ($\sigma$) of the log-transformed $^{137}Cs$ concentrations for April–December 2012 and April–December 2013 were 1.815 ± 0.4667 and 1.433 ± 0.3609, respectively. The peak of the normal distribution curve for April–December 2013 was a smaller concentration than the peak for April–December 2012. For April–December 2012, the specimen with 16,000 Bq/kg-wet of $^{137}Cs$ concentration (log-transformed value = 4.204) was an outlier on the higher side ($+3\sigma$ from the arithmetic mean = 3.215, $^{137}Cs$ = 1,641 Bq/kg-wet) (Figs. 3 & 4). The probabilities of finding greenlings with concentrations exceeding 1,641 Bq/kg-wet and 16,000 Bq/kg-wet of $^{137}Cs$ from April to December 2012 were below about $1.346 \times 10^{-3}$ and $1.527 \times 10^{-7}$, respectively. For April–December 2013, the specimen with 1,150 Bq/kg-wet of $^{137}Cs$ concentration (log-transformed value = 3.061) was an outlier on the higher side ($+3\sigma$ from the arithmetic mean = 2.516, $^{137}Cs$ = 328 Bq/kg-wet) (Figs. 3 & 4). This specimen of greenlings was caught approximately 20 km south of the power plant (off the coast of Hirono town) on 8 May 2013[5]. The probabilities of finding greenlings with concentrations exceeding 328 Bq/kg-wet and 1,150 Bq/kg-wet of $^{137}Cs$ from April to December 2013 were below about $1.350 \times 10^{-3}$ and $3.239 \times 10^{-6}$, respectively.

## Discussion

Measurements of contamination in individual greenlings reveal the specific area of contamination because the migration range of greenlings is restricted[9]. In southern waters, radiocesium concentrations of greenlings caught in shallow waters were significantly higher than those caught in offshore waters (Table 1). Since September 2012, no individual was caught in southern offshore waters that exceeded the Japanese standard limit for radiocesium, while 18 of 45 individuals (40.0%) from southern shallow waters exceeded the limit. Meanwhile, Figures 3 and 4 showed that the decline of radiocesium concentrations in greenlings was due not only to the decay of $^{134}Cs$ but also to a decline in the concentrations of $^{137}Cs$ in greenlings. This spatiotemporal trend of radiocesium concentrations in greenlings was similar to that in other demersal fish species[6,7,14]. Therefore, the basic process of radiocesium contamination is thought to be the same in greenlings as in other demersal fish species off the coast of Fukushima.

Table 1 | Data for individual greenlings caught north and south of the Fukushima Daiichi Nuclear Power Plant (FDNPP)

| Area | Sampling date | Depth (m) | Number of individuals | $^{134}Cs + ^{137}Cs$ concentrations (Bq/kg-wet) Range Max. | Min. | Geometric mean* | Number of specimens in which radiocesium was not detected | Number of individuals in which contamination exceeded the Japanese standard limit** (frequency %) |
|---|---|---|---|---|---|---|---|---|
| Northern waters | 2012.06.26 | 50 | 28 | 189 | 6.70 | 44.3 | 0 | 4 (14.3%) |
| | 2012.08.28 | 70 | 4 | 127 | 2.42 | 8.69 | 0 | 1 (25%) |
| | 2012.10.25 | 70 | 16 | 29.7 | not detected | 12.7 | 1 (4.24 Bq/kg-wet) | 0 |
| | 2012.10.25 | 100 | 5 | 2.2 | 4.00 | 7.79 | 0 | 0 |
| | 2013.01.17 | 50 | 1 | 3.60 | | | 0 | 0 |
| | 2013.02.26 | <30 | 30 | 39.2 | 4.46 | 15.8 | 0 | 0 |
| Southern waters | 2012.05.20 | <30 | 14 | 1070 | 613 | 805 | 0 | 14 (100%) |
| | 2012.05.21 | 50 | 27 | 379 | 2.95 | 52.7 | 0 | 8 (29.6%) |
| | 2012.05.21 | 100 | 6 | 513 | 5.02 | 56.9 | 0 | 3 (50.0%) |
| | 2012.07.20 | 50 | 4 | 442 | 25.7 | 79.6 | 0 | 1 (25.0%) |
| | 2012.07.20 | 100 | 23 | 987 | not detected | 23.1 | 2 (5.88 and 5.89 Bq/kg-wet) | 3 (13.0%) |
| | 2012.07.21 | <30 | 3 | 706 | 291 | 401 | 0 | 3 (100%) |
| | 2012.09.21 | <30 | 12 | 299 | 15.7 | 70.9 | 0 | 3 (25.0%) |
| | 2012.09.21 | 60 | 2 | 19.3 | 14.9 | Arithmetic mean = 17.1 | 0 | 0 |
| | 2012.09.21 | 100 | 11 | 25.5 | not detected | 8.29 | 1 (3.28 Bq/kg-wet) | 0 |
| | 2012.11.26 | <30 | 15 | 145 | 40.3 | 72.2 | 0 | 5 (33.3%) |
| | 2012.11.26 | 100 | 11 | 80.7 | not detected | 14.5 | 0 | 0 |
| | 2013.02.11 | <30 | 18 | 233 | 3.64 | 62.8 | 0 | 10 (55.6%) |
| | 2013.03.01 | <30 | 6 | 169 | 28.6 | 108 | 0 | 5 (83.3%) |

*We used the detection limit to calculate the geometric mean for samples in which radiocesium was not detected.
**The Japanese standard limit for radiocesium ($^{134}Cs + ^{137}Cs$) in foods is 100 Bq/kg-wet.

Previous studies suggested that direct exposure to extremely contaminated seawater from the FDNPP in April 2011[3,4] was one of the main factors that caused serious contamination of marine organisms in southern waters[6]. Especially in shallow waters, demersal fish species and benthic organisms could be directly exposed to the extremely contaminated seawater. In addition to receiving direct contamination, demersal fish species receive radiocesium through the contaminated benthic food web. This process of local contamination probably causes the variability in radiocesium concentrations within fish species off the coast of Fukushima, especially among sedentary demersal fish species.

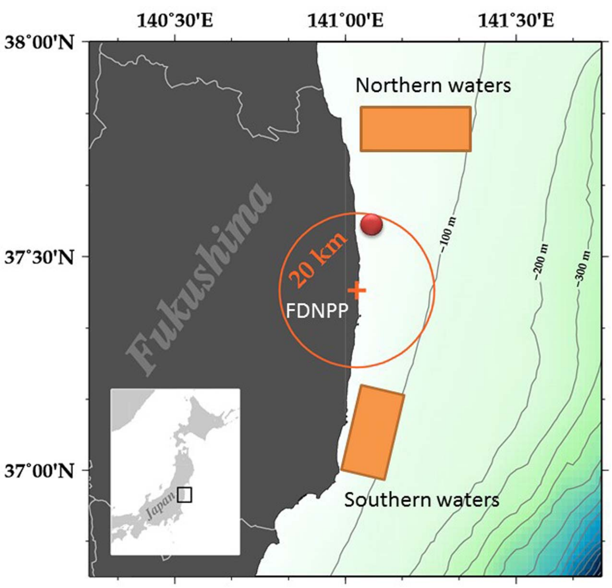


**Figure 1 | Greenlings were collected from several sampling locations.** The larger orange circle marks a 20 km radius around the Fukushima Daiichi Nuclear Power Plant (FDNPP), and the smaller red spot marks the sampling point where the greenlings with 25,800 Bq/kg-wet of radiocesium were caught on 1 August 2012. The map was created using the Generic Mapping Tools (GMT)[26].

Ambe et al.[8] reported radiocesium concentrations of the sediments (0–1 cm layer) collected around our sampling point in southern waters in July 2012. The concentrations of sediments at the depths of 30 m, 50 m, and 100 m were 761 Bq/kg-wet, 742 Bq/kg-wet, and 140 Bq/kg-wet, respectively (dry weight concentrations in the report[8] were converted into wet weight concentrations depending on the percentage of water content that was provided in personal communications of co-authors). These concentrations were equal to or higher than the concentrations measured in greenlings in this study (Table 1). A previous report noted that radionuclide bioavailability from contaminated sediments is typically low, with the transfer factor generally below 1.0[18]. Therefore, we assumed that the strong adsorption of $Cs^+$ to clay minerals would prevent marine organisms from absorbing much radiocesium through the benthic food web[19,20].

Otosaka and Kobayashi[21] calculated that the amount of bioavailable $^{137}Cs$ in sediment (0–3 cm layer) collected in the coastal waters of Ibaraki Prefecture, approximately 70 km south of the FDNPP, was only about 20% of total sedimentary $^{137}Cs$, because more than 75% of the $^{137}Cs$ was incorporated into lithogenic fractions that were not bioavailable to marine organisms. Accordingly, the intake of radiocesium through the benthic food web is expected to be limited for greenlings and other demersal fish species, even if the sediments are highly contaminated. Experimental rearing of greenlings and exposure of test subjects to contaminated sediments should be conducted to elucidate the transfer of radiocesium from contaminated sediments to benthic organisms and demersal fish species.

The probabilities that two outlier specimens of greenlings with higher radiocesium concentrations would be found off the coast of Fukushima were exceedingly low (Fig. 4). These facts suggest that the extremely contaminated greenlings were not contaminated outside

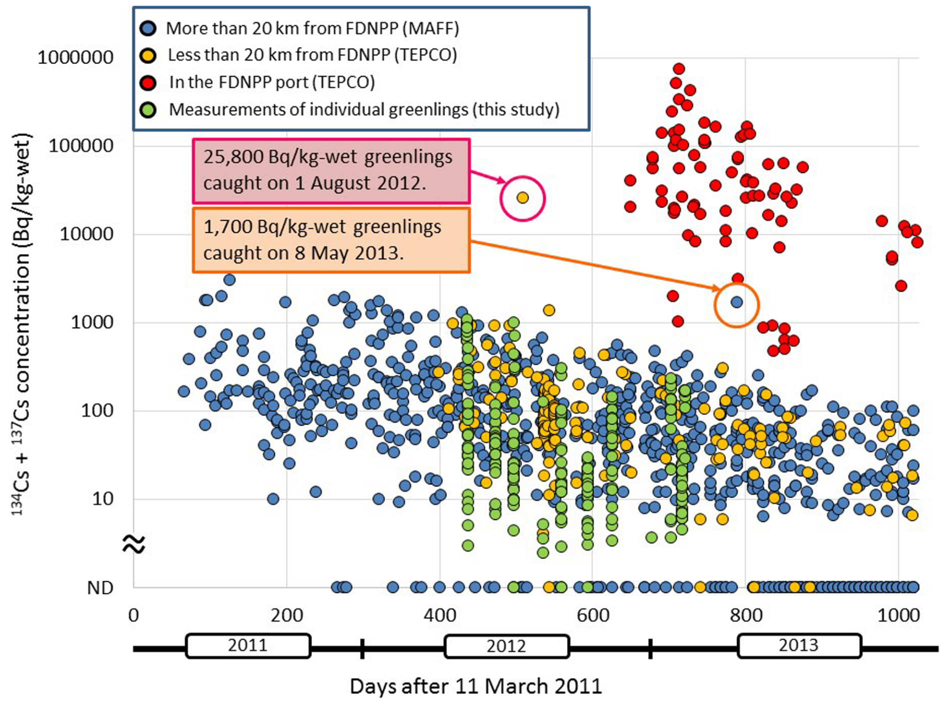


**Figure 2 | Temporal trends of radiocesium concentrations ($^{134}Cs$ + $^{137}Cs$) for greenlings caught within and more than 20 km from the Fukushima Daiichi Nuclear Power Plant (FDNPP) port are shown.** Tokyo Electric Power Corporation (TEPCO) has been monitoring marine products within 20 km of the FDNPP since April 2012.

the FDNPP port. Kanda[22] reported that radiocesium from the FDNPP was continuing to enter the sea after the accident on 11 March 2011. The averages of $^{137}Cs$ concentrations in seawater collected from the unit 1–4 intake canal of the FDNPP for June–August 2011 and April–September 2012 were calculated as 305 to 1,650 Bq/L and 17.1 to 209 Bq/L, respectively[22]. The concentration ratio (CR) for $^{137}Cs$ between the 25,800 Bq/kg-wet ($^{137}Cs$ = 16,000 Bq/kg-wet) specimen and seawater around the unit 1–4 intake canal for June–August 2011 and April–September 2012 ranged from 9.70 to 52.5 and from 76.6 to 936, respectively. The range of CR for June–August

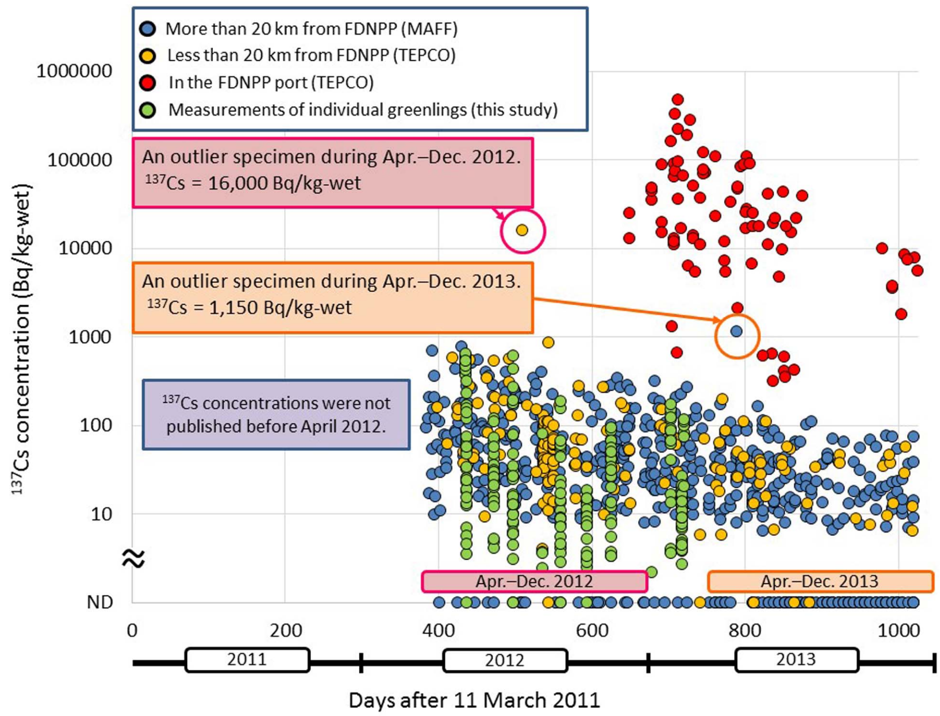


**Figure 3 | Temporal trends of $^{137}Cs$ concentrations for greenlings caught within and more than 20 km from the Fukushima Daiichi Nuclear Power Plant (FDNPP) port are shown.** The dataset of $^{137}Cs$ concentrations was not published before April 2012.

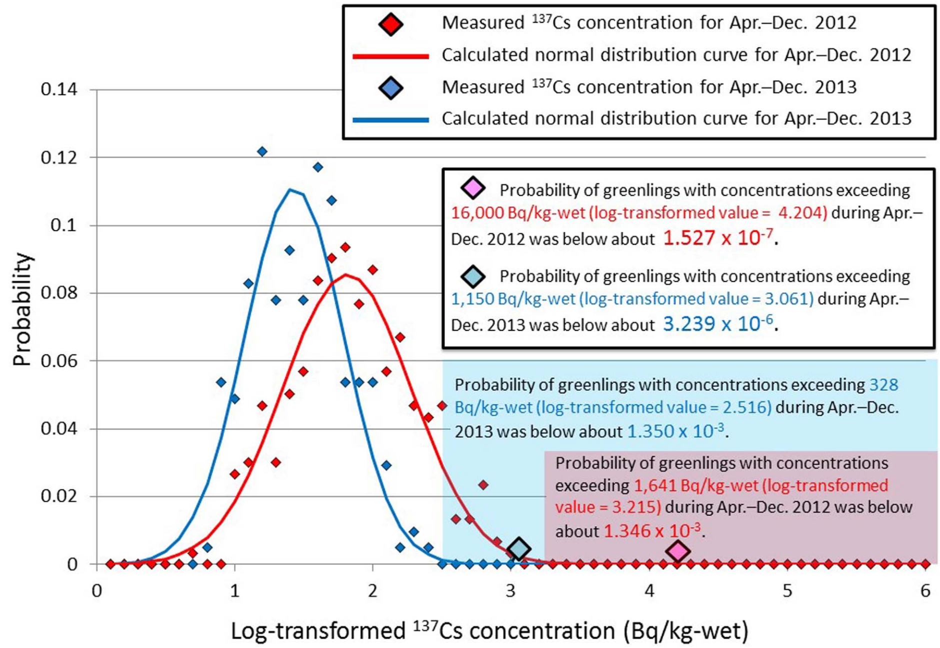


**Figure 4 | Normal distribution curves were calculated from the dataset of log-transformed $^{137}Cs$ concentrations for April–December 2012 (red) and April–December 2013 (blue).**

2011 is consistent with previous findings, in which the range of the CR for $^{137}Cs$ between demersal fish species and seawater around Japan was 15 to 54[23]. Accordingly, the radiocesium concentration in seawater within the FDNPP port before the summer of 2012 was sufficient to contaminate greenlings at a level of 25,800 Bq/kg-wet.

The CR for $^{137}Cs$ between the 1,700 Bq/kg-wet ($^{137}Cs$ = 1,150 Bq/kg-wet) specimen and seawater around the unit 1–4 intake canal for June–August 2011 and April–September 2012 ranged from 0.697 to 3.77 and from 5.50 to 67.3, respectively. In addition, the average of $^{137}Cs$ concentrations in seawater collected from the unit 1–4 intake canal in May 2013 was 9.35 to 107 Bq/L[15]. The CR for $^{137}Cs$ between the 1,700 Bq/kg-wet ($^{137}Cs$ = 1,150 Bq/kg-wet) specimen and seawater around the unit 1–4 intake canal in May 2013 ranged from 10.7 to 123. Accordingly, the radiocesium concentration in seawater within the FDNPP port after 2011 was sufficient to contaminate greenlings at a level of 1,700 Bq/kg-wet.

Both of the two outlier specimens were caught approximately 20 km from the FDNPP[5,15] port. The location where they were caught lies within the migration range of greenlings[9]. Therefore, we assume that the two outlier specimens of greenlings were contaminated within the FDNPP port.

TEPCO has been removing fish from the FDNPP port using gill nets and pods since 10 October 2012. The concentrations of radiocesium in fish caught in the port showed great variation even within the same species[15], indicating that less-contaminated fish were continuing to enter the port. Therefore, in order to prevent fish from moving in and out of the port, TEPCO installed partition nets around the breakwaters and a block fence with nets at the entrance of the port on 12 July 2013[24]. In addition, TEPCO began covering the seabed soil, which included high concentration of radiocesium, with solidified soil on 21 February 2012[25]. In order to ensure the safety of fishery products, it is necessary to continue carefully monitoring marine products that are caught off the coast of Fukushima, especially around the FDNPP.

## Methods

**Sampling and radiocesium measurements of individual greenlings.** We used greenlings caught by fishery workers in Fukushima Prefecture using fishing gear from May 2012 to March 2013. Sampling information (date, location, and depth) was provided by the fishery workers. Fish capture permission for scientific use was granted by Fukushima Prefectural Government office (permission No. 13 and 20 in 2012). Our protocol, including fish sacrifice, was in accordance with a guide for animal experimentation at National Research Institute of Fisheries Science (NRIFS); fish handling approval was granted by the animal experiment committee of NRIFS. We measured contamination of 236 samples of muscle tissue of individual greenlings (Table 1). The processed specimens were packed tightly into plastic cylindrical containers and specific gamma rays of $^{134}Cs$ (605 and 796 keV) and $^{137}Cs$ (662 keV) were measured with a high-purity germanium (HPGe) semiconductor detector (ORTEC, GEM30–70–LB–C, 1.85 keV/1.33 MeV of resolution) with a multichannel analyser on the ground. The energy-dependent efficiency calibration for the HPGe semiconductor detector was conducted using five gamma ray reference sources (Japan Radioisotope Association). These reference sources contained quantified concentrations of the gamma ray radionuclides of $^{54}Mn$, $^{57}Co$, $^{60}Co$, $^{88}Y$, $^{109}Cd$, $^{137}Cs$, and $^{139}Ce$, and their radioactive concentrations and heights differed from one another. The radiocesium concentration at three standard deviations outside the norm was defined as the detection limit concentration. The measurement time was set at 7,200 seconds. The range of the detection limit for samples in which radiocesium was not detected was 1.15 to 2.48 Bq/kg-wet in $^{134}Cs$ and 2.13 to 3.82 Bq/kg-wet in $^{137}Cs$. The measurement error associated with the detection limit was caused by different volumes of specimens. The concentrations of $^{134}Cs$ and $^{137}Cs$ were corrected for decay from the sampling date.

**Data analysis.** The published datasets of MAFF[5] and TEPCO[15] were employed to reveal a time-series trend in radiocesium concentrations in greenlings caught off the coast of Fukushima. Information in published datasets came from single or multiple greenling individuals caught at the same time. The datasets from MAFF[5] contained information about greenlings caught more than 20 km from the FDNPP. Using the normal distribution curves of log-transformed $^{137}Cs$ concentrations for greenlings, we calculated the probability that outlier specimens on the higher concentration side (over $+ 3\sigma$ from the arithmetic mean of log-transformed $^{137}Cs$ concentration) would be caught outside the FDNPP port during April–December 2012 and April–December 2013. Values below the detection limit of radiocesium were excluded from this analysis in order to generate a more conservative estimate. Statistical tests and normality of log-transformed $^{137}Cs$ concentrations for each dataset, April–December 2012 and April–December 2013, were tested using the Excel Statcel 3 software program (OMS publishing Co., Saitama, Japan).


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## Author contributions
Y.S., T.O., K.M., K.N., T.M. and T.W. designed this study. Y.S. and T.M. wrote the main manuscript. Y.S., K.F., D.A. and H.K. carried out measurement and sample preparations. Y.S., D.A. and T.W. prepared figures. All authors reviewed the manuscript.

## Additional information

**Competing financial interests:** The authors declare no competing financial interests.

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