Pacific bluefin tuna transport Fukushima-derived radionuclides from Japan to California

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The Fukushima Dai-ichi release of radionuclides into ocean waters caused significant local and global concern regarding the spread of radioactive material. We report unequivocal evidence that Pacific bluefin tuna, *Thunnus orientalis*, transported Fukushima-derived radionuclides across the entire North Pacific Ocean. We measured γ-emitting radionuclides in California-caught tunas and found ^{134}Cs (4.0 ± 1.4 Bq kg^{-1}) and elevated ^{137}Cs (6.3 ± 1.5 Bq kg^{-1}) in 15 Pacific bluefin tuna sampled in August 2011. We found no ^{134}Cs and background concentrations (~1 Bq kg^{-1}) of ^{137}Cs in pre-Fukushima bluefin and post-Fukushima yellowfin tunas, ruling out elevated radiocesium uptake before 2011 or in California waters post-Fukushima. These findings indicate that Pacific bluefin tuna can rapidly transport radionuclides from a point source in Japan to distant ecoregions and demonstrate the importance of migratory animals as transport vectors of radionuclides. Other large, highly migratory marine animals make extensive use of waters around Japan, and these animals may also be transport vectors of Fukushima-derived radionuclides to distant regions of the North and South Pacific Oceans. These results reveal tools to trace migration origin (using the presence of ^{134}Cs) and potentially migration timing (using ^{134}Cs:^{137}Cs ratios) in highly migratory marine species in the Pacific Ocean.

pelagic | radioactivity | bioaccumulation

The infrequency of nuclear accidents coupled with potentially wide-ranging effects on ecosystems and human health make the dynamics and risks of radionuclide discharge into the environment a relatively poorly understood but highly important area of research (1–4). On March 11, 2011, an earthquake and subsequent tsunami led to flooding of the Fukushima Dai-ichi nuclear power plants in Japan. Coolant pumps failed to operate and the power plant reactors overheated, leading to a release of radionuclides directly into the ocean exceeding that from any previous accident (3). The release of radionuclides produced a 1–2 wk pulse that peaked on April 6, 2011 with ocean concentrations of 68 MBq m^{-3} (3) and an estimated total release of up to 22 × 10^{15} Bq of ^{137}Cs (4) (1 Bq = 1 disintegration s^{-1}). The dominant long-lived γ-emitting radionuclides ^{134}Cesium ($t_{1/2}$ = 2.1 y) and ^{137}Cs ($t_{1/2}$ = 30 y) were released at a consistent ratio of about 1 (0.99 ± 0.03) (3), although the ratio of ^{134}Cs:^{137}Cs would decline slowly over time because of the differential decay rate constants of the two isotopes (Fig. S1). After considerable dilution 2–3 mo after maximum discharge, surface concentrations still exceeded prior concentrations by up to 10,000-fold in coastal waters (3) and up to 1,000-fold over a 150,000 km^2 area of the Pacific up to 600 km east of Japan (4). Before the Fukushima discharge, low concentrations (1.5 mBq L^{-1}) of the long-lived ^{137}Cs (fallout from weapons testing) were detectable in Japanese waters (3), whereas the shorter-lived ^{134}Cs was undetectable in Pacific surface waters and biota.

The Pacific bluefin tuna (PBFT), *Thunnus orientalis*, is a highly migratory fish that inhabits the western and eastern North Pacific Ocean at various life stages (5) (Fig. 1*A*). Mature PBFT spawn in the western Pacific, and some juveniles remain in Japanese waters while others migrate eastward to the California Current Large Marine Ecosystem (CCLME) (Fig. 1*A*), with most migrating late in their first year or early in their second (5). Thus, all bluefin between years 1–2 (here, 2-y-old PBFT) caught during summer in the eastern Pacific must have migrated from the western Pacific within several months of capture. Waters north of the Kuroshio Current (Fig. 1*A*) showed high radionuclide concentrations in spring 2011 (3), and juveniles make extensive use of this region before their eastward migration to the CCLME (6).

We tested the possibility that juvenile PBFT served as biological vectors of radionuclides between two distant ecoregions: the waters off Japan and the CCLME. We analyzed 2-y-old PBFT caught off San Diego, CA, in August 2011, known from size to be recent Japan migrants, for the presence of Fukushima-derived radionuclides. Because Cs accumulates in the muscle tissue of fish (7), we analyzed the white muscle tissue of PBFT in 2011 for concentrations of ^{134}Cs, ^{137}Cs, and various naturally occurring γ-emitting radionuclides. To rule out non-Fukushima sources of radiocesium in fish muscle, we also measured radionuclide concentrations in PBFT collected in California waters before the Fukushima discharge (2008) and in yellowfin tuna (YFT), *T. albacares* (August 2011), in the CCLME where they are highly residential (8, 9).

Results and Discussion

All 15 PBFT collected in 2011 contained ^{134}Cs (4.0 ± 1.4 Bq kg^{-1} dry wt) and ^{137}Cs (6.3 ±1.5 Bq kg^{-1}) in white muscle tissue (Table 1, Table S1, and Fig. 1*A*). At the time of capture, total $^{134+137}$Cs concentrations were about 10 times higher in 2011 PBFT than in PBFT from previous years (Table 1). In contrast, 2008 PBFT and 2011 YFT had no measurable ^{134}Cs and consistent, much lower ^{137}Cs concentrations (consistent with background concentrations from fallout) than the 2011 PBFT (Table 1, Table S2, and Fig. 1*A*). This is unequivocal evidence that Fukushima-derived radionuclides were transported to the CCLME by Pacific bluefin tuna, as no other sources of ^{134}Cs were present in the North Pacific preceding the Fukushima disaster (3, 4). Mean concentrations of the naturally occurring γ-emitting ^{40}K in the 2011 PBFT were 347 ± 49 Bq kg^{-1} (Table 1). Other naturally occurring γ-emitting radionuclides (^{7}Be, ^{211}Bi, and ^{212}Pb) were detectable at extremely low concentrations (Table S3), approximately three orders of magnitude below measured radiocesium concentrations.

Because bluefin tuna are harvested annually in the Eastern Pacific Ocean (EPO) at 1.7–9.9 × 10^3 metric tonnes (10) (Table S4) for human consumption (2000 to 2010), the possibility of radioactive contamination raises public health concerns. Radiocesium concentrations of post-Fukushima PBFT reported here were more than an order of magnitude below the recently

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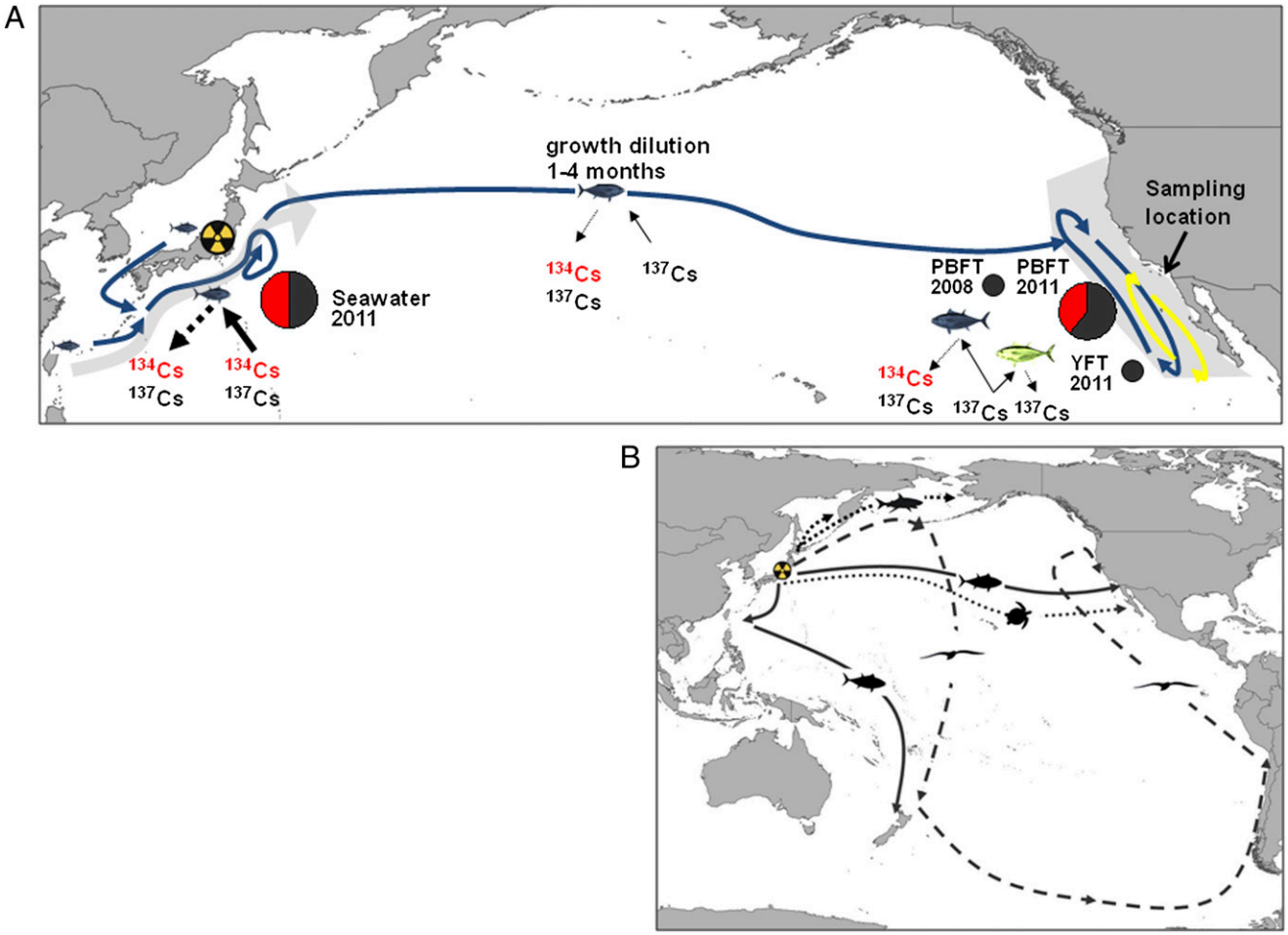

Fig. 1. (*A*) Map of the northern Pacific ocean showing simplified movement patterns for juvenile PBFT (blue arrows) from Japan to the CCLME and juvenile YFT (yellow arrows) in the CCLME. Kuroshio Current (gray arrow east of Japan) and CCLME (gray region west of N. America) are shown. Pie charts show mean concentrations of ^{134}Cs (red) and ^{137}Cs (dark gray) in seawater (Bq m^{3}) (3), and muscle tissue (Bq kg^{-1}) in PBFT after Fukushima, PBFT before Fukushima, and YFT after Fukushima. Pie charts for fish are sized to scale of total radiocesium concentrations. Black arrows show uptake (solid arrows) and efflux (dotted arrows) of radiocesium in different ocean regions (arrow thickness scaled for relative efflux and uptake rates). (*B*) Simplified migration patterns of some highly migratory species in the Pacific that inhabit waters around Japan and make subsequent long distance migrations to distant ecoregions including Kamchatka, the Aleutian Islands, North America, South America, and New Zealand. Migration patterns are shown for salmon sharks (17) (short dashed line), sooty shearwaters (18) (long dashed line), Pacific bluefin tuna (6) (solid lines), and loggerhead turtles (19) (dotted line).

changed Japanese safety limit of 100 Bq kg^{-1} wet wt (about 400 Bq kg^{-1} dry wt) (11). Inferences about the safety of consuming radioactivity-contaminated seafood can be complicated due to complexities in translating food concentration to actual dose to humans (12), but it is important to put the anthropogenic radioactivity levels in the context of naturally occurring radioactivity. Total radiocesium concentrations of post-Fukushima PBFT were approximately thirty times less than concentrations of naturally occurring ^{40}K in post-Fukushima PBFT and YFT and pre-Fukushima PBFT (Table 1). Furthermore, before the Fukushima release the dose to human consumers of fish from ^{137}Cs was estimated to be 0.5% of that from the α-emitting ^{210}Po (derived from the decay of ^{238}U, naturally occurring, ubiquitous and relatively nonvarying in the oceans and its biota (13); not measured here) in those same fish (12). Thus, even though 2011 PBFT showed a 10-fold increase in radiocesium concentrations, ^{134}Cs and ^{137}Cs would still likely provide low doses of radioactivity relative to naturally occurring radionuclides, particularly ^{210}Po and ^{40}K.

However, 2011 PBFT ^{134}Cs and ^{137}Cs concentrations were less than they would have been in Japanese waters because of growth, radioactive decay, and efflux of cesium. To estimate the concentrations of ^{134}Cs and ^{137}Cs in 2011 PBFT at the time they left Japanese waters, we back-calculated radiocesium concentrations in muscle over a range of potential trans-Pacific migration times (30–120 d) using a loss rate of assimilated Cs from fish tissue of 1.9% day^{-1} (14), growth dilution attributable to an increase in body mass during the trans-Pacific migration, and radioactive decay (details in *SI Methods*). We estimated that PBFT in Japan had 1.5–15 times higher radiocesium concentrations (30–120 d before capture, respectively) than concentrations measured at time of capture in California (Fig. 2 and Table S1). Variations in animal size at exposure and growth because exposure may influence Cs concentrations; there was a negative trend between animal size and ^{137}Cs and ^{134}Cs concentrations, although the correlation was not significant (Spearman's ρ; Fig. S2 and *SI Methods*). The Japanese Ministry of Agriculture, Forestry, and Fisheries report post-Fukushima concentrations of $^{134+137}$Cs in PBFT around Japan at 61–168 Bq kg^{-1} dry weight (11). Our estimates of total radiocesium (i.e., ^{134}Cs + ^{137}Cs) concentrations in PBFT for 90 and 120 d before capture are 73–147 Bq kg^{-1}, respectively (Table S1). The similarity between back-calculated values and those measured in Japan PBFT suggests that our

Table 1. Measured ^{134}Cs, ^{137}Cs, and the naturally occurring radionuclide ^{40}K for post-Fukushima bluefin (PBFT 2011), pre-Fukushima bluefin (PBFT 2008), and post-Fukushima yellowfin tuna (YFT 2011) caught in California waters

				Radionuclide concentrations (Bq kg^{-1})				
	SL (cm)*	Body mass (kg dry)†	Age (y)‡	^{134}Cs	^{137}Cs	^{40}K	^{134}Cs:^{137}Cs	$^{134+137}$Cs
PBFT 2011 (n = 15)								
Median	66.5	1.5	1.5	4.3	6.0	367	0.66	10.6
Mean	66.2	1.5	1.5	4.0	6.3	347	0.62	10.3
SD	3.6	0.2	0.1	1.4	1.5	49	0.14	2.9
PBFT 2008 (n = 5)								
Median	66.3	1.5	1.4	0	1.4	266	0	1.37
Mean	66.2	1.5	1.4	0	1.4	258	0	1.11
SD	1.2	0.09	0.05	0	0.2	43	0	0.64
YFT 2011 (n = 5)								
Median	72.3	1.9	1.2	0	1.2	342	0	1.21
Mean	72.3	1.9	1.2	0	1.1	333	0	1.10
SD	2.5	0.2	0.01	0	0.4	78	0	0.35

All radionuclide concentrations are in Bq kg^{-1} dry mass.
*Estimated from CFL [PBFT (20) and YFT (21)].
†Estimated from standard length (SL) (22, 23).
‡Estimated from SL (24, 25).

model performs well in predicting previous concentrations of Cs in tuna captured months after exposure.

Decay-corrected ^{134}Cs:^{137}Cs ratios measured in post-Fukushima bluefin caught off California averaged 0.62 ± 0.14 and ranged from 0.26 to 0.84 (Table 1 and Table S1). Ratios of ^{134}Cs:^{137}Cs have been used to discern sedimentation patterns and water movements in the Irish Sea, which received 15 PBq of discharged ^{137}Cs between 1978 and 1983 from the nuclear fuel-reprocessing plant at Sellafield (15, 16). However, Cs ratios have not been used previously to track biota movements. Assuming that PBFT tissue would reflect the ^{134}Cs:^{137}Cs ratio of 1.0 to which they were exposed near Japan, radiocesium ratios can be used to make inferences about migration and the timing of exposure to contaminated waters. Back-calculated ^{134}Cs:^{137}Cs ratios in PBFT approached 1.0 (0.8 ± 0.2) at 4 mo before capture (Fig. 2), suggesting departure from Japan ∼4 mo before capture off California. This result suggests that the radiocesium levels in California-caught PBFT were the result of <1 mo exposure to contaminated waters near Japan. Because radiocesium release was pulsed, it is possible that back-calculated ratios will always converge on the period of maximum discharge (rather than on the date of PBFT emigration from Japanese waters). The ^{134}Cs:^{137}Cs ratio of seawater near Japan would have changed little from 1.0 within 4 mo of the pulsed radioactivity discharge (1.0–0.9; Fig. S1). However, additional PBFT radiocesium data across longer time scales (e.g., PBFT from 2012) will indicate whether back-calculated ^{134}Cs:^{137}Cs ratios in PBFT consistently converge on 1.0 in April 2011 (period of maximum discharge) or converge on 1.0

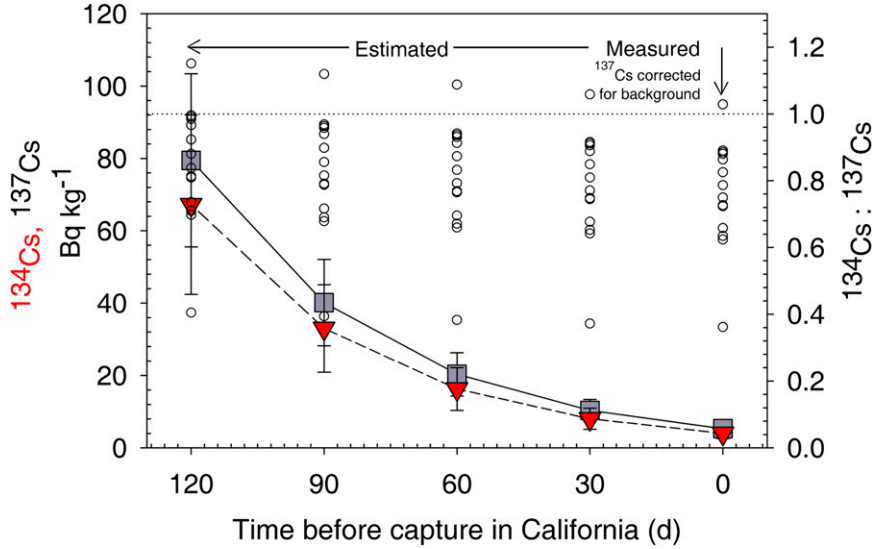

Fig. 2. Measured (corrected for background levels of ^{137}Cs) and back-calculated estimates of radiocesium concentrations in muscle of post-Fukushima Pacific bluefin tuna *Thunnus orientalis* (n = 15). Estimated values of ^{134}Cs and ^{137}Cs concentrations in 2011 PBFT for various times before capture in California (30, 60, 90, 120 d) account for background levels of ^{137}Cs, decreases in tissue concentrations because of growth dilution and efflux rates of radiocesium out of the fish during their migration across the Pacific (detailed in *SI Methods*). Mean concentrations of ^{134}Cs (red triangles) or ^{137}Cs (gray squares) shown on the left y axis. Error bars represent 1 SD. Ratios of ^{134}Cs:^{137}Cs for each individual fish (empty circles) shown with scale on the right y axis. Dotted line represents 1:1 ratio of ^{134}Cs:^{137}Cs, the ratio expected in tuna while in waters off Japan contaminated with radiocesium at a ^{134}Cs:^{137}Cs ratio of 1.0.

over a range of dates after the Fukushima discharge (demonstrating the utility of ^{134}Cs:^{137}Cs ratios as a tool to assess the timing of PBFT migrations).

The total load of radiocesium transported to the CCLME by PBFT can be estimated from catches in the EPO. Catch data varies yearly (10), but assuming PBFT commercial catches in 2011 were within the range of catch from 2000 to 2010, transported and harvested radiocesium in tuna muscle tissue in 2011 could range from ~3–17 × 10^6 Bq (Table S4) or <<1% of total radiocesium released into Japanese waters (4). Catch data represent a portion of the PBFT in the EPO, so total transport of radiocesium by PBFT would likely be higher. Still, this is a small quantity of radiocesium to be introduced to a large pelagic ecosystem, but it is also a conservative estimate based on one species. Other highly migratory species (HMS) (e.g., turtles, sharks, and seabirds) that forage near Japan may assimilate radiocesium and transport it to distant regions of the north and south Pacific (Fig. 1*B*). Tissue concentrations of radiocesium in these species would depend on time spent near Japan, foraging strategies, and timing of migration. The potential for species in Fig. 1*B* and other HMS (e.g., pinnipeds, whales, and billfish) that forage in Japan to transport Fukushima-derived radiocesium is speculative. However, the presence of Fukushima-derived radiocesium in all 2011 PBFT individuals reported here suggests that study of other HMS is warranted. Our results demonstrate that Fukushima-derived radionuclides in animal tissues can serve as tracers of both migration origin (presence or absence of ^{134}Cs) and, potentially, timing (using ^{134}Cs:^{137}Cs ratios) in mobile marine animals, providing valuable complementary movement data to extensive tagging programs in the Pacific (8). Extensive data regarding spatiotemporal variations in Cs concentrations in the west Pacific, and consequent uptake by biota, are forthcoming, which will sharpen the precision of these tracers. The Fukushima disaster, thus, provides an opportunity to examine both the extent of transport of anthropogenic radionuclides by highly migratory species and an unexpected tool for examining migratory origins of apex predators in the Pacific Ocean.

Methods

Tuna tissue samples were collected from recreational anglers in San Diego, CA. Muscle samples were freeze-dried and ground with mortar and pestle and analyzed using a low-energy germanium detector. We detected the γ emissions of ^{137}Cs and ^{134}Cs, ^{40}K, and other naturally occurring radionuclides. ^{134}Cs and ^{137}Cs concentrations of post-Fukushima PBFT and YFT samples were decay-corrected to the angler-estimated catch date for all fish. We calculated ^{134}Cs and ^{137}Cs concentrations in 2011 PBFT for various times before capture in California (0, 30, 60, 90, 120 d). We accounted for background ^{137}Cs in tuna muscle (1.0 Bq kg^{-1}) by subtracting 1.0 from total ^{137}Cs values. We then accounted for the radioactive decay of Cs isotopes, important only for the shorter-lived ^{134}Cs ($t_{1/2}$ = 2.1 y) using an exponential decay model. To account for the metabolic efflux of assimilated cesium out of fish, we used an experimentally derived marine fish efflux rate constant k of 0.019 d^{-1} (14). To address growth dilution of the Cs concentrations in muscle, we calculated change in fish body mass (0–120 d before catch) and calculated dilution of Cs concentrations over this range of days due to growth.

We estimated radiocesium transport to the CCLME by PBFT using catch data as a proxy for PBFT biomass in the CCLME. Catch data from 2000 to 2010 were converted to muscle biomass and then multiplied by mean measured radiocesium concentrations to generate a range of estimates of transported radiocesium. Spearman's ρ analyses (two-tailed, nonparametric; α = 0.05) were performed to assess correlation between 2011 PBFT length and radionuclide concentrations.

Equations and further details of the methods are included in *SI Methods*.

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Supporting Information

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SI Methods

Radioanalysis. Tuna tissue samples were collected from recreational anglers in San Diego, CA. Angler-approximated capture date and fish size (CFL in centimeters) was recorded. Approximately 30 g dry weight (29.4 ± 7.8 g) of white muscle tissue was collected from the hypaxial musculature below the first dorsal fin of each fish. All sampled fish were caught in US waters in the tuna fishing grounds off San Diego, CA. Samples were kept on dry ice (−55 °C) and shipped to Stony Brook for radioanalysis. Muscle samples were freeze-dried and then ground with mortar and pestle. Pulverized tissue was placed into 4 oz Nalgene straight-side clear jars. Before radioanalysis, all samples were stored at 60 °C to avoid rehydration.

We used a low energy germanium detector (LEGe) (Canberra; Model GLP 3830 with a 3800 mm^2 active area). Sample counting times were adjusted to allow propagated counting errors of <10% for ^{137}Cs (662 keV), ^{134}Cs (605 keV), and ^{40}K (1,460 keV), when possible. Genie 2000 software was used to analyze the peaks in the energy spectrum. Most post-Fukushima PBFT samples were counted for 2 d but 2008 PBFT samples and 2011 YFT samples were counted for up to 5 d because of lower radioactivity. The lower detection limits were 0.6, 0.3, and 4 Bq kg^{-1} for ^{134}Cs, ^{137}Cs, and ^{40}K, respectively. Naturally occurring ^{212}Pb (239 keV), ^{7}Be (478 keV), and ^{211}Bi (350 keV) were also detected at trace levels in tuna samples. Counting geometry was taken into consideration, and samples were calibrated using known quantities of ^{75}Se, ^{137}Cs, and ^{152}Eu emitting over a broad energy spectrum (265 keV, 662 keV, and 1,408 keV, respectively). We also compared our counts of ^{137}Cs and ^{40}K with those in a certified International Atomic Energy Agency (IAEA) fish standard (standard 414), consisting of freeze-dried fish muscle from the Irish and North Seas (1). The detection limits for each individual radioisotope were calculated for each individual sample of fish muscle using the "well-known blank" method (2).

^{134}Cs and ^{137}Cs concentrations of post-Fukushima PBFT and YFT samples were decay-corrected to August 25th, 2011, which was the angler-estimated catch date for all fish. ^{134}Cs and ^{137}Cs concentrations in PBFT collected in 2008 were decay-corrected to the capture date of August 25th, 2008.

Statistical Analysis. Spearman's ρ (two-tailed) at confidence interval of 95% (α = 0.05) was used to assess correlation between 2011 PBFT length and cesium concentrations. Spearman's rank correlation is nonparametric (does not assume data normality), not highly influenced by outliers, and gives an estimate of monotonic correlation between two variables even if the relationship is not linear. Spearman's correlation coefficient r_s falls between −1 and 1 and gives direction and strength of correlation. P values are reported with α = 0.05. Analysis was performed using MatLab (Version 7.1).

Back-Calculations for 2011 PBFT. We calculated ^{134}Cs and ^{137}Cs concentrations in 2011 PBFT for various times before capture in California (0, 30, 60, 90, 120 d), accounting for background levels of ^{137}Cs, decreases in tissue concentrations attributable to growth dilution, and efflux rates of radiocesium out of the fish during their migration across the Pacific.

We first accounted for the background ^{137}Cs in tuna muscle (1.0 Bq kg^{-1}) by subtracting 1.0 from total ^{137}Cs values. We then accounted for the radioactive decay of Cs isotopes, important only for the shorter-lived ^{134}Cs ($t_{1/2}$ = 2.1 y). Fish growth between departure from Japan and capture in August would dilute the original radiocesium concentrations and add new muscle mass with assimilated background ^{137}Cs. We assumed a growth rate of 0.709 mm day^{-1} for fish >56 cm SL (3). From standard length (SL) we estimated wet body mass (m_{wet}) in wet kg:

$$m_{wet} = SL^{3.095} \times 1.14 \times 10^{-5} \quad (4) \qquad \textbf{[S1]}$$

and wet mass converted to dry mass:

$$m_{dry} = m_{wet} \times 0.244 \quad (5) \qquad \textbf{[S2]}$$

The dry weights are shown in Table 1 and Tables S1–S3. We assumed a constant pool of ^{137}Cs in fish muscle tissue due to observed levels in pre-Fukushima fish (1.0 Bq kg^{-1} ^{137}Cs). We calculated the pool of background (not Fukushima-derived) ^{137}Cs in the muscle of each fish ($^{137}Cs_{bgd,capture}$) at the time of capture:

$$^{137}Cs_{bgd,capture} = m_{capture} \times 1.0 \text{ Bq kg}^{-1} \qquad \textbf{[S3]}$$

and subtracted this value from total measured ^{137}Cs values at the time of capture.

To address growth dilution of the Cs concentrations in muscle, we calculated fish body mass, m_t (0–120 d before catch) from standard lengths and converted to dry mass. For Pacific bluefin:

$$SL_t = SL_{capture} - \left(t \times 0.0709 \text{ cm day}^{-1}\right) (3) \qquad \textbf{[S4]}$$

where $SL_{capture}$ is length at time of capture, estimated from measured CFL (cm), for t = [0, 30, 60, 90, or 120] days. We then calculated m_t from SL_t:

$$m_t = 1.41 \times 10^{-5} \times SL_t^{3.095} (4) \qquad \textbf{[S5]}$$

We assumed no uptake of Fukushima-derived $^{134+137}$Cs into fish muscle between departure from Japanese waters and capture off San Diego. We assumed total load of Fukushima-derived Cs in PBFT (Cs_F):

$$Cs_F = \text{total measured } ^{134}\text{Cs} + \text{total measured } ^{137}\text{Cs} - ^{137}Cs_{bgd,capture} \qquad \textbf{[S6]}$$

and divided Cs_F by the calculated body mass at 0–120 d before capture. Estimated ^{134}Cs and ^{137}Cs concentrations in fish muscle (i.e., [^{134}Cs] and [^{137}Cs]) increased with each regressive time step because of this growth dilution (Table S1).

To account for the metabolic efflux of assimilated cesium out of fish, we used an experimentally derived marine fish efflux rate constant (k) of 0.019 d^{-1} (6). Using this value, we used an exponential decay model to determine Cs_{Ft} before capture because assimilated metals are lost from marine animals following an exponential function (7):

$$\left[^{134}\text{Cs}\right]_t = \left[^{134}\text{Cs}\right]_{capture} \times e^{kt} \qquad \textbf{[S7]}$$

$$\left[^{137}\text{Cs}\right]_t = \left(\left[^{137}\text{Cs}\right]_{capture} - \left[^{137}\text{Cs}\right]_{bgb,capture}\right) \times e^{kt} \qquad \textbf{[S8]}$$

where t = time before capture in CA (0, 30, 60, 90, or 120 d), $[Cs]_{capture}$ is the measured Cs concentration at day of capture,

and $[Cs]_t$ is the concentration of ^{134}Cs or ^{137}Cs back-calculated for time before capture t.

Estimates of Radiocesium Transport to CCLME. We estimated radiocesium transport to the CCLME by PBFT using catch data as a proxy for PBFT biomass in the CCLME. Catch data from 2000 to 2010 (8) were converted to muscle biomass. We then calculated values of transported radiocesium:

$$\text{muscle biomass} \times \text{dry wt conv.factor}\ (0.244\ (\text{ref.5})) \times \text{mean}\left[^{134+137}\text{Cs kg}^{-1}\right](\text{this study}) \quad \textbf{[S9]}$$

to generate a range of values for transported radiocesium, assuming catch in 2011 fell within the range of catch from 2000 to 2010. Estimated transport = 3–17 × 10^6 Bq.

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4. Itoh T (2001) Estimation of total catch in weight and catch-at-age in number of bluefin tuna *Thunnus orientalis* in the whole Pacific Ocean. *Bull Natl Res Inst Far Seas Fish* 38:83–111.
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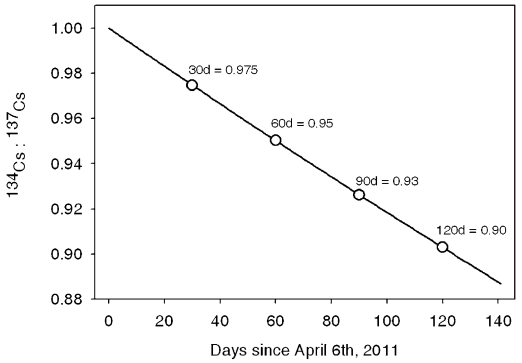

Fig. S1. Ratio of ^{134}Cs:^{137}Cs in seawater contaminated by Fukushima-derived radiocesium. Decrease in ratios is attributable to differences in decay rates of ^{134}Cs ($t_{1/2}$ = 2.1 y) and ^{137}Cs ($t_{1/2}$ = 30 y). Projected ^{134}Cs:^{137}Cs ratios are shown for various time intervals (30, 60, 90, and 120 d) after the peak pulse discharge on April sixth, 2011.

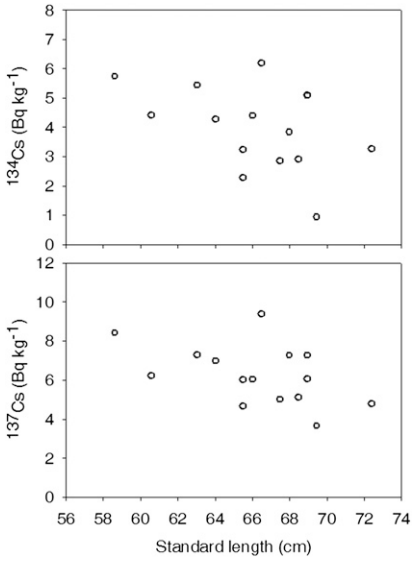

Fig. S2. Relationship of standard length (cm) and ^{134}Cs and ^{137}Cs concentrations in white muscle tissue in Pacific bluefin tuna, *Thunnus orientalis,* sampled in August 2011. Spearman's rank correlation was performed, and there was no significant relationship between Cs concentrations and size [^{134}Cs (n = 15): df = 13, r_s= −0.370, P = 0.17; ^{137}Cs (n = 15), df = 13, r_s= −0.483, P = 0.07].

Table S1. Measured concentrations of ^{134}Cs, ^{137}Cs, and the naturally occurring radionuclide ^{40}K and back-calculated concentrations of ^{134}Cs and ^{137}Cs for individual post-Fukushima bluefin (PBFT 2011) caught in California waters

				Measured (Bq kg^{-1} dry)‡					Calculated (Bq kg^{-1} dry)									
									0 d		30 d		60 d		90 d		120 d	
PBFT 2011	SL (cm)*	Body mass (kg dry)†	Age (y)†	^{134}Cs	^{137}Cs	$^{134+137}$Cs	^{40}K	^{134}Cs:^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs
1	72.4	2.0	1.7	3.3	4.8	8.0	367	0.68	3.3	3.8	6.5	7.3	13.0	14.3	26.1	27.8	52.5	54.4
2	64.0	1.3	1.4	4.3	7.0	11.3	382	0.61	4.3	6.0	8.6	11.8	17.5	23.2	35.5	45.8	72.6	91.1
3	60.6	1.1	1.3	4.4	6.2	10.6	288	0.71	4.4	5.2	9.0	10.3	18.3	20.5	37.5	40.8	77.1	81.6
4	68.0	1.6	1.6	3.8	7.3	11.1	344	0.53	3.8	6.3	7.7	12.2	15.5	23.9	31.3	47.0	63.5	92.7
5	69.0	1.7	1.6	5.1	6.0	11.1	368	0.84	5.1	5.1	10.2	9.9	20.5	19.3	41.4	37.8	83.7	74.3
6	69.0	1.7	1.6	5.1	7.3	12.3	281	0.70	5.1	6.3	10.2	12.2	20.4	23.9	41.2	46.8	83.4	92.2
7	69.5	1.7	1.6	0.9	3.7	4.6	401	0.26	0.9	2.7	1.9	5.2	3.7	10.1	7.6	19.8	15.3	39.0
8	66.5	1.5	1.5	6.2	9.4	15.6	297	0.66	6.2	8.4	12.5	16.4	25.1	32.2	50.9	63.5	103.5	125.5
9	58.6	1.0	1.2	5.7	8.4	14.2	424	0.68	5.7	7.4	11.7	14.7	24.0	29.3	49.4	58.7	102.0	118.0
10	66.0	1.5	1.5	4.4	6.0	10.4	337	0.73	4.4	5.0	8.8	9.9	17.9	19.4	36.2	38.2	73.7	75.6
11	67.5	1.6	1.5	2.8	5.0	7.9	259	0.57	2.8	4.0	5.7	7.8	11.5	15.4	23.3	30.2	47.2	59.6
12	68.5	1.7	1.6	2.9	5.1	8.0	393	0.57	2.9	4.1	5.8	8.0	11.7	15.7	23.7	30.8	47.9	60.7
13	65.5	1.4	1.5	3.2	6.0	9.2	321	0.54	3.2	5.0	6.5	9.8	13.2	19.3	26.7	38.1	54.4	75.5
14	63.1	1.3	1.4	5.4	7.3	12.7	369	0.75	5.4	6.3	11.0	12.4	22.3	24.5	45.5	48.5	93.2	96.5
15	65.5	1.4	1.5	2.3	4.7	6.9	376	0.49	2.3	3.7	4.6	7.2	9.2	14.1	18.8	27.9	38.2	55.3
Median	66.5	1.5	1.5	4.3	6.0	10.6	367	0.66	4.3	5.1	8.6	9.9	17.5	19.4	35.5	38.2	72.6	75.6
Mean	66.2	1.5	1.5	4.0	6.3	10.3	347	0.62	4.0	5.3	8.0	10.3	16.3	20.3	33.0	40.1	67.2	79.5
SD	3.6	0.2	0.1	1.4	1.5	2.9	49	0.14	1.4	1.5	2.9	3.0	5.9	6.0	12.1	11.9	24.8	23.9

Estimated concentrations of ^{134}Cs and ^{137}Cs for individual fish (n = 15) are shown for a range of days (0, 30, 60, 90, and 120) before capture for post-Fukushima bluefin, taking into account background levels of ^{137}Cs, radiocesium efflux during transit, growth dilution, and radioactive decay.
*Estimated from CFL (1).
†Estimated from SL (2, 3).
‡Decay-corrected to catch date.

1. Farwell CJ (2011) *Based on a Regression by C. Farwell* (Tuna Research and Conservation Center, Pacific Grove, CA).
2. Itoh T (2001) Estimation of total catch in weight and catch-at-age in number of bluefin tuna *Thunnus orientalis* in the whole Pacific Ocean. *Bull Natl Res Inst Far Seas Fish* 38:83–111.
3. Bayliff WH, Ishizuka Y, Deriso R (1991) Growth, movement, and attrition of northern bluefin tuna, *Thunnus thynnus*, in the Pacific ocean, as determined by tagging. *Inter-Am Trop Tuna Comm Bull* 20:3–94.

Table S2. Measured concentrations of ^{134}Cs, ^{137}Cs, and the naturally occurring radionuclide ^{40}K for pre-Fukushima bluefin (PBFT 2008) and post-Fukushima yellowfin tunas (YFT 2011) caught in California waters

				Bq kg^{-1} dry		
Individual fish	SL (cm)*	Body mass (kg dry)†	Age (y)‡	^{134}Cs	^{137}Cs	^{40}K
PBFT 2008						
1	67.5	1.6	1.5	0	<0.8	298
2	65.0	1.4	1.4	0	1.61	266
3	66.5	1.5	1.4	0	1.38	298
4	64.5	1.4	1.3	0	1.37	225
5	66.0	1.5	1.4	0	1.18	202
Median	66.0	1.5	1.4	0	1.4	266
Mean	65.9	1.5	1.4	0	1.4	258
SD	1.2	0.1	0.05	0	0.2	43
YFT 2011						
1	69.0	1.6	1.2	0	1.21	412
2	72.3	1.9	1.2	0	0.49	406
3	74.7	2.1	1.2	0	1.40	342
4	70.9	1.8	1.2	0	1.17	247
5	74.7	2.1	1.2	0	1.22	260
Median	72.3	1.9	1.2	0	1.2	342
Mean	72.3	2.1	1.2	0	1.1	333
SD	2.5	0.2	0.01	0	0.4	78

Estimated concentrations of ^{134}Cs and ^{137}Cs are for 5 individual fish for each species. Reported concentrations of both Cs radioisotopes are decay-corrected to date of capture.
*Estimated from CFL: PBFT (1) YFT (2).
†Estimated from SL (3, 4).
‡Estimated from SL (5, 6).

1. Farwell CJ (2011) *Based on a Regression by C. Farwell* (Tuna Research and Conservation Center, Pacific Grove, CA).
2. Scida P, Rainosek A, Lowery T (2001) Length conversions for yellowfin tuna (*Thunnus albacares*) caught in the western North Atlantic Ocean. *Collect Vol Sci Pap ICCAT* 52:528–532.
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Table S3. Measured concentrations of naturally occurring radionuclides ^{7}Be, ^{211}Bi, and ^{212}Pb and for *n* = 15 post-Fukushima Pacific bluefin tuna (PBFT 2011) caught in California waters

				mBq kg^{-1}			
PBFT 2011	SL (cm)*	Body mass (kg dry)†	Age (years)‡	^{7}Be	^{211}Bi	^{212}Pb	Total
1	72.4	2.0	1.7	0.5	ND	2.5	ND
2	64.0	1.3	1.4	0.4	54.2	1.4	56.0
3	60.6	1.1	1.3	<0.12	35.6	0.8	36.4
4	68.0	1.6	1.6	<0.12	40.0	0.8	40.8
5	69.0	1.7	1.6	<0.12	54.7	0.2	54.9
6	69.0	1.7	1.6	<0.12	42.4	0.9	43.3
7	69.5	1.7	1.6	<0.12	49.3	1.6	50.9
8	66.5	1.5	1.5	<0.12	38.5	0.9	39.4
9	58.6	1.0	1.2	<0.12	43.2	1.5	44.7
10	66.0	1.5	1.5	1.1	49.3	0.9	51.3
11	67.5	1.6	1.5	0.2	41.8	1.2	43.2
12	68.5	1.6	1.6	14.2	53.0	1.1	68.3
13	65.5	1.4	1.5	2.9	58.0	0.4	61.3
14	63.1	1.3	1.4	<0.12	65.4	1.4	66.8
15	65.5	1.4	1.5	1.5	59.8	1.4	62.7
Median	66.5	1.5	1.5	1.1	49.3	1.1	51.1
Mean	66.2	1.5	1.5	3.0	48.9	1.1	51.4
SD	3.6	0.2	0.1	5.0	9.0	0.5	10.5

Note that units (mBq kg^{-1}) are three orders of magnitude less than those for ^{134}Cs, ^{137}Cs, and ^{40}K. ND, not determined.
*Estimated from CFL (1).
†Estimated from SL (2).
‡Estimated from SL (3).

1. Farwell CJ (2011) *Based on a Regression by C. Farwell* (Tuna Research and Conservation Center, Pacific Grove, CA).
2. Itoh T (2001) Estimation of total catch in weight and catch-at-age in number of bluefin tuna *Thunnus orientalis* in the whole Pacific Ocean. *Bull Natl Res Inst Far Seas Fish* 38:83–111.
3. Bayliff WH, Ishizuka Y, Deriso R (1991) Growth, movement, and attrition of northern bluefin tuna, *Thunnus thynnus*, in the Pacific ocean, as determined by tagging. *Inter-Am Trop Tuna Comm Bull* 20:3–94.

Table S4. Pacific bluefin tuna, *Thunnus orientalis*, catch data from 2000 to 2010 in the EPO (1) and total harvested muscle biomass in the EPO

Year	Catch (×10^{3} mt)	Muscle biomass (×10^{6} kg)*
2000	4.2	3.0
2001	1.7	1.2
2002	2.5	1.8
2003	3.7	2.6
2004	9.0	6.4
2005	4.8	3.4
2006	9.9	7.0
2007	4.2	3.0
2008	4.5	3.2
2009	3.6	2.6
2010	7.9	5.6
Median	4.2	3.0
Mean	5.1	3.6
SD	2.6	1.9

*Catch × 0.71 [ratio of muscle tissue to total body mass (2)].

1. Inter-American Tropical Tuna Commission (2010) *Fishery Status Report* (Inter-American Tropical Tuna Commission, La Jolla, CA).
2. Deguara S, Caruana S, Agius C (2010) Product conversion factors in Atlantic bluefin tuna, *Thunnus thynnus* L. *Collect Vol Sci Pap ICCAT* 65:770–775.