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Radiocesium in Pacific Bluefin Tuna *Thunnus orientalis* in 2012 Validates New Tracer Technique

Daniel J. Madigan,*[,†] Zofia Baumann,[‡] Owyn E. Snodgrass,[§] Halim A. Ergül,[∥] Heidi Dewar,[§] and Nicholas S. Fisher[‡]

[†]Hopkins Marine Station, Stanford University, 120 Ocean View Boulevard, Pacific Grove, California 93950, United States

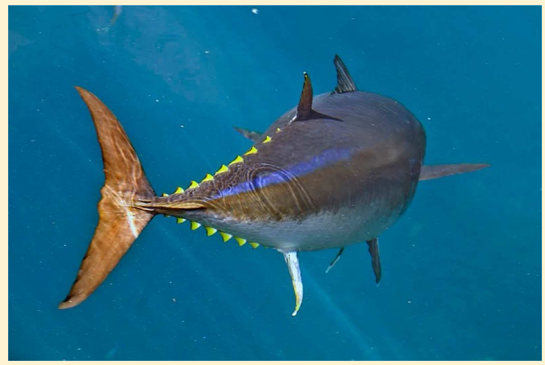
[‡]School of Marine and Atmospheric Sciences, Stony Brook University, Stony Brook, New York 11794, United States

[§]Fisheries Resources Division, Southwest Fisheries Science Center, National Marine Fisheries Service, NOAA, 8604 La Jolla Shores Drive, La Jolla, California 92037, United States

[∥]Department of Biology, Kocaeli University, 41300 Umutteppe, İzmit, Kocaeli, Turkey

Supporting Information

ABSTRACT: The detection of Fukushima-derived radionuclides in Pacific bluefin tuna (PBFT) that crossed the Pacific Ocean to the California Current Large Marine Ecosystem (CCLME) in 2011 presented the potential to use radiocesium as a tracer in highly migratory species. This tracer requires that all western Pacific Ocean emigrants acquire the ^{134}Cs signal, a radioisotope undetectable in Pacific biota prior to the Fukushima accident in 2011. We tested the efficacy of the radiocesium tracer by measuring ^{134}Cs and ^{137}Cs in PBFT (n = 50) caught in the CCLME in 2012, more than a year after the Fukushima accident. All small PBFT (n = 28; recent migrants from Japan) had ^{134}Cs (0.7 ± 0.2 Bq kg^{-1}) and elevated ^{137}Cs (2.0 ± 0.5 Bq kg^{-1}) in their white muscle tissue. Most larger, older fish (n = 22) had no ^{134}Cs and only background levels of ^{137}Cs, showing that one year in the CCLME is sufficient for ^{134}Cs and ^{137}Cs values in PBFT to reach pre-Fukushima levels. Radiocesium concentrations in 2012 PBFT were less than half those from 2011 and well below safety guidelines for public health. Detection of ^{134}Cs in all recent migrant PBFT supports the use of radiocesium as a tracer in migratory animals in 2012.



INTRODUCTION

The discharge of radionuclides into the western Pacific Ocean in 2011 from the failed Fukushima nuclear power plant has led to studies of radionuclide concentrations in seawater and marine biota, both near Japan[1−3] and in migratory marine species.[4] In 2011, radiocesium from Fukushima was detected in Pacific bluefin tuna, *Thunnus orientalis,* that had recently traversed the North Pacific Ocean, suggesting the potential for Fukushima-derived radionuclides to serve as tracers of long-distance migrations by highly migratory species in the Pacific Ocean.[4] While tools such as electronic tags have provided extensive animal movement data prospective from the date of tagging,[5] chemical tracers (such as radiocesium) can provide retrospective migration information that is often uniquely informative.[4,6−8]

Certain conditions are necessary for the reliable use of Fukushima-derived radiocesium to trace migrations. While ^{137}Cs ($t_{1/2}$ = 30.1 years) still exists throughout the Pacific in low, "background" levels as a result of nuclear weapons testing that peaked in the 1960s, the shorter lived ^{134}Cs ($t_{1/2}$ = 2.1 years) from nuclear weapons testing has long since decayed.[2] A point source of anthropogenic radionuclides such as Fukushima is therefore the only substantial source of ^{134}Cs in the Pacific Ocean, and consequently the presence of ^{134}Cs indicates recent migration from the contaminated region. For reliable application of this tracer, all animals migrating from contaminated waters must accumulate and retain measurable levels of ^{134}Cs to accurately identify their status as recent migrants from the western Pacific (to avoid interpretation of recent migrants as residents). Conversely, animals must excrete ^{134}Cs in their new environment at some determinable rate so that absence of ^{134}Cs can be interpreted as residency in the noncontaminated region for an interpretable period of time (to avoid erroneous identification of recent migrants, and to constrain the time range of a "recent" migration). Given that potential transport of Fukushima-derived radiocesium is currently being examined in a variety of migratory animals including whales, turtles, tunas, sharks, and seabirds to infer migratory patterns, it is critical to test these assumptions.

Pacific bluefin tuna (PBFT) are an ideal species for the validation and application of the radiocesium tracer. All PBFT

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Table 1. Catch Date, Size, Estimated Age, and Radionuclide Concentrations in 50 Pacific Bluefin Tuna (PBFT) *Thunnus orientalis*, Captured in the California Current Large Marine Ecosystem in 2012[a]

PBFT no.	catch date	SL[b] cm	age[c] years	^{134}Cs	^{137}Cs Bq kg^{-1} dry wt	^{40}K	^{134}Cs:^{137}Cs
1	8/4/2012	63.1	1.3	0.64	1.72	385	0.37
2	8/4/2012	63.4	1.3	0.61	2.03	372	0.30
3	8/4/2012	63.5	1.3	0.33	1.61	382	0.20
4	8/4/2012	63.8	1.3	0.67	1.82	456	0.37
5	8/4/2012	64.8	1.4	0.56	2.23	275	0.25
6	8/4/2012	65.6	1.4	0.58	1.51	347	0.38
7	8/4/2012	65.8	1.4	0.61	2.03	455	0.30
8	8/4/2012	65.8	1.4	0.79	1.82	405	0.43
9	8/4/2012	65.9	1.4	0.76	2.45	500	0.31
10	8/4/2012	66.2	1.4	0.50	1.43	364	0.35
11	8/18/2012	66.6	1.4	0.59	2.71	412	0.22
12	8/4/2012	66.6	1.4	0.69	1.89	422	0.37
13	8/4/2012	66.8	1.4	1.30	2.91	446	0.45
14	8/4/2012	67.0	1.4	0.98	2.45	471	0.40
15	8/4/2012	67.1	1.4	0.60	1.93	335	0.31
16	8/4/2012	67.4	1.5	0.49	1.58	388	0.31
17	8/4/2012	67.8	1.5	0.75	1.89	487	0.39
18	8/4/2012	67.9	1.5	0.51	1.50	362	0.34
19	8/4/2012	68.5	1.5	0.54	2.23	310	0.24
20	8/18/2012	68.9	1.5	0.30	1.22	269	0.24
21	8/4/2012	68.9	1.5	0.42	1.13	269	0.37
22	8/4/2012	69.0	1.5	0.75	1.95	459	0.39
23	8/4/2012	69.1	1.5	0.68	1.66	382	0.41
24	8/18/2012	69.4	1.5	0.96	3.12	497	0.31
25	8/4/2012	69.4	1.5	0.75	2.14	324	0.35
26	8/4/2012	69.9	1.6	0.41	1.16	226	0.35
27	8/4/2012	71.8	1.6	0.54	1.88	383	0.29
28	8/22/2012	72.3	1.6	1.14	2.93	702	0.39
29	6/9/2012	73.7	1.7	nd	1.00	399	–
30	6/10/2012	76.5	1.8	0.86	2.29	439	0.38
31	6/9/2012	77.4	1.8	nd	1.33	432	–
32	6/10/2012	78.0	1.9	nd	1.98	534	–
33	8/18/2012	79.2	1.9	nd	0.98	540	–
34	8/18/2012	79.5	1.9	nd	0.94	676	–
35	8/18/2012	80.7	2.0	nd	0.96	588	–
36	8/18/2012	80.7	2.0	1.19	3.14	561	0.38
37	6/9/2012	81.0	2.0	nd	1.69	371	–
38	8/18/2012	81.0	2.0	1.26	3.81	518	0.33
39	8/4/2012	82.8	2.1	nd	0.54	438	–
40	8/18/2012	84.4	2.1	0.61	2.63	349	0.23
41	8/18/2012	87.3	2.2	1.16	4.21	564	0.28
42	6/10/2012	107.7	3.0	nd	1.43	613	–
43	6/10/2012	108.5	3.0	nd	0.86	503	–
44	6/10/2012	109.8	3.1	nd	1.71	711	–
45	6/10/2012	112.7	3.2	nd	1.37	505	–
46	6/10/2012	115.7	3.3	nd	0.36	482	–
47	5/26/2012	117.0	3.4	nd	1.28	429	–
48	6/10/2012	118.3	3.4	nd	0.53	459	–
49	6/10/2012	122.4	3.6	nd	1.08	543	–
50	5/26/2012	132.4	4.0	nd	0.97	558	–

[a]Small PBFT: nos. 1-28; larger PBFT: nos. 29-50, as defined in the text. [b]Estimated from CFL.[21] [c]Estimated from SL.[19] nd: not detected. Dash (–) indicates ^{134}Cs:^{137}Cs ratios that could not be determined due to nondetection of ^{134}Cs.

spawn in the western Pacific Ocean, and juveniles forage in the waters around Japan.[9−11] Juveniles then either remain in the western Pacific or migrate eastward to the California Current Large Marine Ecosystem (CCLME). Most PBFT are thought to migrate late in their first year or early in their second.[9] Thus, the youngest PBFT in the CCLME (approximately 1−1.5 years old) must have migrated from Japan within the preceding year. Previous studies suggest that larger, older PBFT in the CCLME are primarily residents for >1 year.[9] However, some fish migrate from Japan at older ages, and in a given year the proportion of

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Japan migrants to CCLME residents is largely unknown. This information could improve fisheries modeling and management of PBFT, in which severe population declines have recently been reported.[12,13]

For radiocesium to function as a reliable tracer in PBFT, all PBFT in the CCLME below some threshold age (which must be recent migrants from Japan) must assimilate adequate concentrations of ^{134}Cs before their eastward migration and retain measurable concentrations after their trans-Pacific migration to the CCLME. Marine fish have been shown to acquire Cs from both the aqueous phase and from diet.[14] Older fish in the CCLME that are residential (>1 year in the CCLME) must lose ^{134}Cs due to excretion,[4,14,15] so that only recently migrated fish would carry measurable levels of ^{134}Cs, allowing the ^{134}Cs to distinguish recent migrants from >1 year CCLME residents.

We collected 50 samples of PBFT in 2012 and measured muscle tissue for ^{134}Cs and ^{137}Cs to determine if the acquisition of radiocesium by migrating PBFT persisted into the summer of 2012, more than a year after the Fukushima disaster. Radiocesium levels were compared to concentrations of another γ-emitting radionuclide, the naturally occurring ^{40}K, to provide context for observed radiocesium concentrations. For comparison, we also collected 5 samples of yellowfin tuna (*Thunnus albacares*) in 2012, which are known from electronic tagging studies to be residents of the CCLME and do not make migrations from the western Pacific Ocean.[5,16] We used small PBFT, known to be migrants from Japan, to test whether all migrants would demonstrate a measurable radiocesium signal from Fukushima. We examined the radiocesium levels in older fish to determine if residents and migrants could be discerned via the absence of ^{134}Cs (due to excretion during a year or more in CCLME waters) or presence of ^{134}Cs (due to recent migration from Japan). Finally, we use ratios of ^{134}Cs:^{137}Cs to estimate time of departure from Japan in recently migrated PBFT.

MATERIALS AND METHODS

Sampling and Radioanalysis. Tuna tissue samples were collected from PBFT and yellowfin tuna (YFT; *Thunnus albacares;* CCLME residents)[5,16] captured by recreational sport fishermen. Fish were caught within 300 km of San Diego, CA and landed in San Diego where they were filleted and frozen for human consumption. We sampled 250−500 g of white muscle tissue from the dorsal musculature behind the head. Muscle tissue was kept on dry ice (−55 °C) and shipped to Stony Brook University for radioanalysis. Muscle samples were freeze-dried and 65 ± 23 g dry wt of white muscle tissue was blended and combusted at 450 °C for 4 h. Combustion resulted in further compactness of the sample, which assured better counting efficiency during γ-radioanalysis (details below). Loss of Cs during combustion was assumed to be negligible, as shown previously.[17] Ash was further ground with a mortar and pestle to achieve a uniform matrix, placed inside plastic jars, and stored at 60 °C prior to radioanalysis.

For γ-radioanalysis of ^{134}Cs, ^{137}Cs, and ^{40}K we used high purity germanium detectors (HPGe; Canberra Industries). Sample counting times were adjusted to allow propagated counting errors of <10% for ^{137}Cs (662 keV), <15% for ^{134}Cs (605 keV), and <3% for ^{40}K (1461 keV). Most samples were counted for up to 3 days. Genie 2000 software (Canberra) was used to analyze the peaks in the energy spectrum. The lowest detection limits were 0.1 Bq kg^{-1} for both ^{134}Cs and ^{137}Cs and 0.9 Bq kg^{-1} for ^{40}K. These detection limits were calculated for each individual fish muscle sample using the "well-known blank" method.[18] Counting geometry was taken into consideration by varying the fullness of the jar when testing standards made of known quantities of ^{75}Se, ^{134}Cs, ^{137}Cs, and ^{40}K emitting over a broad energy spectrum (265, 605, 662, and 1461 keV, respectively). ^{134}Cs and ^{137}Cs concentrations in tuna muscle samples were decay-corrected to catch dates in the CCLME (Table 1). No decay correction was required for ^{40}K due to its long half-life ($t_{1/2}$ = 1.2 × 10^9 years).

Age Estimation. Age (age$_{est}$) was estimated from standard length (SL; cm) for PBFT and YFT according to Bayliff et al.[19] and Wild.[20] SL was calculated from curved fork length (CFL; cm) for PBFT according to Farwell[21] and for YFT according to Scida et al.[22] When possible we measured the length from rostrum to operculum (operculum length, or OL, cm) and curved fork length (CFL, cm) of whole fish and calculated a regression for PBFT:

$$\mathrm{CFL} = 2.8373\mathrm{OL} + 12.307 \quad (1)$$

to estimate CFL from OL (Supporting Information (SI) Figure S1). The linear equation had an r^2 value of 0.98 (SI Figure S1).

Back-Calculated Departure Date. The ratio of radioactive Cs isotopes (^{134}Cs:^{137}Cs = R) was used to back-calculate the time at which individual PBFT left the Fukushima-contaminated waters around Japan. This ratio would decrease in PBFT after their departure due to continued exposure to ^{137}Cs at background levels (1 mBq L^{-1}) but no ^{134}Cs in central and eastern Pacific waters, and the faster decay rate of ^{134}Cs. A model based on that from Madigan et al.[4] was used to estimate how ^{134}Cs:^{137}Cs ratios in PBFT would change over time after leaving waters around Japan. Briefly, eq 2 describes the processes that impact the change of ^{134}Cs:^{137}Cs ratio over time t. These processes are: radioactive decay (λ_1 and λ_2 for ^{137}Cs and ^{134}Cs; $t_{1/2}$ = 30.2 and 2.1 years, respectively) and efflux (k_e: 0.02 d^{-1})[15] of previously accumulated Cs.[14] Background ^{137}Cs in PBFT muscle (measured in pre-Fukushima PBFT) is represented by A (here, A = 1 Bq kg^{-1} dry wt), which is the same for YFT never exposed to Fukushima radionuclides, PBFT residents of the CCLME >1 year, and PBFT before Fukushima (2008).[4] Background ^{137}Cs (from nuclear weapons testing fallout) in PBFT is a result of uptake of ^{137}Cs from seawater and food (collectively, k_a) and loss of previously accumulated ^{137}Cs (k_e).[4] Since PBFT concentrations of ^{134}Cs and ^{137}Cs change differently over time due to different decay rates and acquisition of weapons fallout ^{137}Cs but not ^{134}Cs after leaving contaminated waters, estimated time since departure (t) will depend not only on the initial R calculation (R_0) but also on absolute concentrations of ^{134}Cs and ^{137}Cs in PBFT. The contribution of ^{137}Cs from weapons fallout (1.0 Bq kg^{-1} dry wt) will contribute relatively more ^{137}Cs to the total pool of ^{137}Cs in PBFT muscle in PBFT that acquire low amounts of Cs in contaminated waters. Consequently, fish that acquire relatively low amounts of ^{134}Cs and ^{137}Cs in contaminated waters will have R values that decrease more rapidly, with ^{134}Cs decreasing more rapidly than ^{137}Cs. In contrast, PBFT that acquire more ^{134}Cs and ^{137}Cs in contaminated waters will have slower rates of decrease of R, as both ^{134}Cs and ^{137}Cs concentrations will decrease at fairly similar rates until the ^{137}Cs concentration decreases to levels where background ^{137}Cs (A) contributes an appreciable proportion of ^{137}Cs to the total pool of ^{137}Cs in PBFT muscle:

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$$R_t = \frac{[^{134}\mathrm{Cs}]_t e^{-t(\lambda_2+k_e)}}{([^{137}\mathrm{Cs}]_t - A)e^{-t(\lambda_1+k_e+k_a)} + A} \quad (2)$$

Background ^{137}Cs concentration in PBFT (A) is a product of ^{137}Cs concentration in seawater (1 mBq L^{-1}) and the Cs uptake rate constant (k_a) divided by the sum of efflux (k_e), radioactive decay (λ_1) and accumulation (k_a) rate constants:

$$A = \frac{(^{137}\mathrm{Cs}_w)k_a}{\lambda_1 + k_e + k_a} \quad (3)$$

It is assumed that ^{134}Cs and ^{137}Cs have identical rate constants of uptake and loss in fish. In 2012, accumulation rate constants (k_a) and efflux rate constants (k_e) yield A values = 1 Bq kg^{-1} of ^{137}Cs. PBFT that migrate away from Fukushima-contaminated waters will lose previously accumulated Cs from muscle tissue, and the concentration of ^{137}Cs and ^{134}Cs in PBFT muscle will asymptotically approach A (1 Bq kg^{-1} dry wt) and 0, respectively. Dividing A by the concentration of ^{137}Cs in surface seawater in the Pacific yields a dry wt concentration factor in PBFT muscle of 1000, and a wet wt concentration factor of 244, somewhat higher than the value of 100 calculated for generic marine fish.[23]

Back-calculations of departure date from Japan require an assumed initial value for ^{134}Cs:^{137}Cs ratio (R) to solve for time t. R at the date of maximum discharge was approximately 1.[2] For PBFT in 2012, we calculated an assumed value of R for April 6, 2012, one year after peak discharge of radionuclides[2] that would result from the different decay rates of ^{134}Cs and ^{137}Cs. This value (0.73) was used in estimates of departure date from Japan for individual PBFT (SI Table S2).

Statistical Analysis. Mann−Whitney U-tests were used to compare the ^{134}Cs and ^{137}Cs concentrations of small (63.4−72.3 cm SL) PBFT, larger PBFT migrants (^{134}Cs present), larger PBFT residents (^{134}Cs absent), PBFT migrants from 2011,[4] and YFT residents in the CCLME. Mann−Whitney U-test p-values were considered significant at $p = 0.05$. Pearson's linear correlation was used to assess the potential linear correlation between PBFT age and estimated time since migration from waters around Japan (calculated from ^{134}Cs:^{137}Cs ratios, eq 2), with p-values significant at $p = 0.05$ and H_0 = no correlation between variables.

RESULTS

We collected 50 Pacific bluefin tuna in 2012 in May, June, and August (Table 1). PBFT ranged from 63.1 to 132.4 cm SL (79.4 ± 18.6 cm), corresponding to estimated ages of 1.3−4.0 years (1.9 ± 0.7 years) (Table 1). ^{134}Cs was detected in 33 fish and concentrations of ^{137}Cs and ^{40}K are reported for all PBFT. All of the small PBFT (63.1−72.3 cm SL; age$_{est}$ = 1.3−1.6 years; n = 28; hereafter "small PBFT") had measurable concentrations of ^{134}Cs (0.7 ± 0.2 Bq kg^{-1}), whereas only 5 of the 22 larger PBFT (73.7−132.4 cm SL; age$_{est}$ = 1.7−4.0 years) had detectable concentrations of ^{134}Cs (1.02 ± 0.27 Bq kg^{-1}) (Tables 1 and S1). When detected, ^{134}Cs concentrations ranged from 0.3−1.3 Bq kg^{-1} dry wt (0.7 ± 0.3 Bq kg^{-1}), approximately 18% of the mean concentration reported in 2011 PBFT (4.0 ± 1.4 Bq kg^{-1}).[4] For PBFT containing ^{134}Cs above the detection limit, the mean ^{134}Cs:^{137}Cs ratio was 0.33 ± 0.06 (Table 1). YFT had no ^{134}Cs and only background levels (0.84 ± 0.12) of ^{137}Cs (Table 2). Concentrations of naturally occurring ^{40}K ranged from 226 to 711 Bq kg^{-1}, and ratios of radioactivity from ^{40}K to $^{134+137}$Cs in PBFT ranged from 99 (PBFT no. 5) to 719 (PBFT no. 34) (Tables 1 and S1).

PBFT sampled in the CCLME are either recent Japan migrants or CCLME residents (Figure 1A), and radiocesium concentrations enabled us to classify PBFT as migrants or residents (Table 1, Figure 1B). All fish ≤ 1.6 years old were migrants, while only 5 of 22 PBFT age 1.7−4 years were

Table 2. Catch Date, Size, and Radionuclide Concentrations in Five Yellowfin Tuna (YFT) *Thunnus albacares*, Captured in the California Current Large Marine Ecosystem in 2012. ^{134}Cs Was Undetectable in All YFT

YFT no.	catch date	SL[a] cm	age[b] years	^{137}Cs Bq kg^{-1} dry wt	^{40}K Bq kg^{-1} dry wt
1	9/22/2012	64.6	1.2	0.90	630
2	9/22/2012	62.7	1.2	0.85	575
3	9/22/2012	66.1	1.2	0.92	636
4	9/22/2012	61.3	1.2	0.90	650
5	9/22/2012	67.0	1.2	0.62	404
average		64.4	1.2	0.84	579
SD		2.4	0	0.12	102

[a]Estimated from CFL.[22] [b]Estimated from SL.[20]

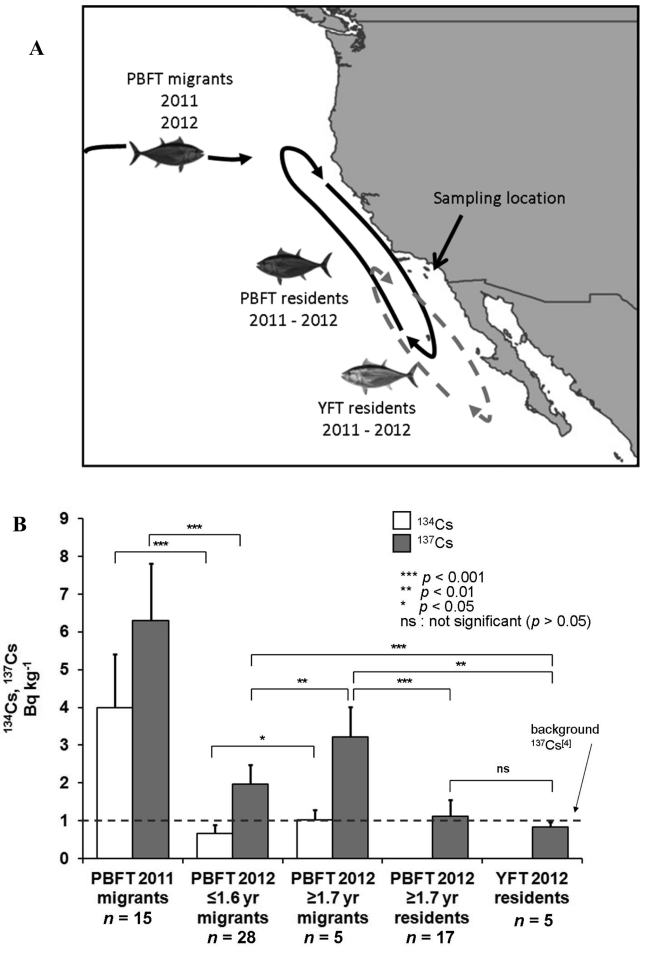

Figure 1. Map of simplified movement patterns (A) and concentrations of ^{134}Cs and ^{137}Cs (B) in Pacific bluefin tuna (migrants and residents) and yellowfin tuna (residents) in the CCLME. Radiocesium concentrations in (B) are mean values (Bq kg^{-1} dry wt) + SD P-values shown are for Mann−Whitney U-tests.

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migrants. Small migrant 2012 PBFT had lower concentrations than larger migrant PBFT of both ^{134}Cs ($p < 0.05$) and of ^{137}Cs ($p < 0.01$) (Figure 1B). Both 2012 migrant groups had lower concentrations than recent migrants sampled in 2011. Larger resident PBFT and YFT had no detectable ^{134}Cs and only background levels of ^{137}Cs (Tables 1 and 2, Figure 1B), and radiocesium levels between these groups was not significantly different (Figure 1B). The smallest fish categorized as a CCLME resident of >1 year was 73.7 cm SL (age_{est} = 1.7 years) (Table 1), establishing this size and age as the threshold in this study between the 100% migrant group (small PBFT) and the mixed group of residents and migrants (larger PBFT) (Figure 2).

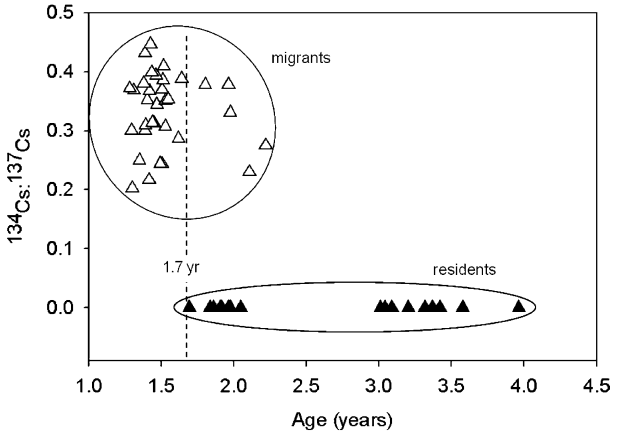

Figure 2. Ratios of ^{134}Cs:^{137}Cs in Pacific bluefin tuna caught in the CCLME from June through August 2012. Open triangles indicate recent Japan migrants, and filled triangles indicate fish that are resident (>1 year) to CCLME. Cs ratios equal to 0 are due to absence of ^{134}Cs in these fish and are only observed in older fish (*x*-axis: ≥1.7 years old). ^{134}Cs:^{137}Cs ratios greater than zero in PBFT >1.7 years represent PBFT that migrated from Japan at older ages.

There was a positive correlation between fish age and time since departure from Japan (estimated from ^{134}Cs:^{137}Cs ratios) ($p < 0.01$, $r^2 = 0.36$; Figure 3A) indicating that larger PBFT left Japan earlier. Estimated departure times from Japan suggest that most migrants left Japan in early mid June, with a few departing in April and July (Figure 3B).

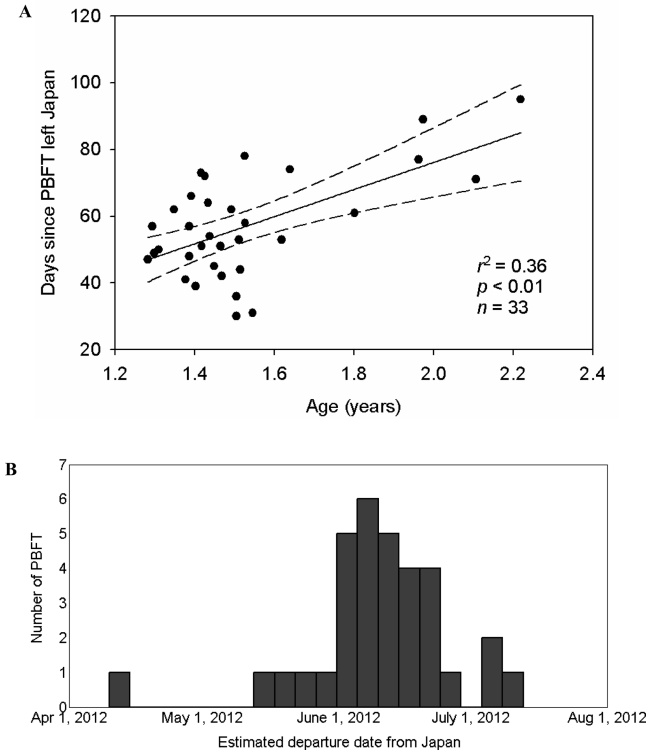

Figure 3. Relationship between PBFT age and estimated time since departure (days) from Japan for the 33 PBFT that contained ^{134}Cs (A) and histogram of estimated departure dates from Japan (B) for recent migrant PBFT. *P*-value and r^2 reported in (A) are for Pearson's linear correlation test. Solid line represents linear fit to data, dashed lines show 95% CI. Departure dates in (B), estimated from ^{134}Cs:^{137}Cs ratios, were centered on early June.

DISCUSSION

Our results demonstrate that PBFT continue to transport Fukushima-derived radiocesium across the Pacific Ocean to the CCLME. These findings support the use of Fukushima-derived radiocesium in Pacific bluefin tuna to determine whether PBFT in the eastern Pacific Ocean have recently migrated from Japan or have been residential in the CCLME for at least one annual cycle, at which point ^{134}Cs is undetectable and ^{137}Cs decreases to background levels.

Prior to this study it was unknown whether concentrations of radiocesium in PBFT in 2012 would be higher or lower than those measured in 2011. Concentrations of Cs in PBFT could decrease over time due to dilution of radiocesium in waters around Japan, the decay of ^{134}Cs, and the potential for decreasing radiocesium concentrations in PBFT prey due to Cs dilution in seawater. However, PBFT captured in the CCLME in 2012 spent more time in contaminated waters than those sampled in 2011. PBFT in 2011 likely spent only 1−3 months in contaminated waters prior to eastward migration, and estimations of departure dates suggest that some may have spent less than one month in contaminated waters before their trans-Pacific migration.[4] Small PBFT in 2012 potentially spent their entire first year in coastal waters around Japan before migrating eastward. There is also the potential for trophic biomagnification of Cs in juvenile PBFT,[14,24] which are mid- to high-trophic level predators that feed on crustaceans, forage fish, and squid.[11] The potential biomagnification of Cs in PBFT presents the possibility of a time lag between maximum radiocesium concentrations in seawater and maximum concentrations in tuna (and other high trophic level predator) muscle tissue. Longer exposure times and potential trophic biomagnification presented the potential for higher concentrations of Cs in 2012 PBFT. Our results show that migrant PBFT concentrations of ^{134}Cs in 2012 dropped to approximately 18% of those in 2011. Total radiocesium ($^{134+137}$Cs) also dropped, from 10.3 ± 2.9 in 2011[4] to 2.9 ± 1.0 Bq kg^{-1} in 2012 (Table 1), suggesting that radiocesium levels declined significantly in PBFT from 2011 to 2012 in the CCLME. This suggests that any enhanced bioaccumulation of $^{134+137}$Cs in PBFT over the longer exposure period was outweighed by the year-long dilution of radiocesium in contaminated waters in 2012. This is in contrast to the report that $^{134+137}$Cs concentrations remained steady (though highly variable) in demersal, coastal fish off Fukushima in 2012.[3] It is likely that differences in radiocesium concentrations in both seawater and prey lead to differences in Cs concentrations between demersal coastal fish and pelagic fish such as PBFT.

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The radionuclides in PBFT from the Fukushima nuclear power plant accounted for an even smaller fraction of the total radioactivity in 2012 PBFT than in 2011 PBFT[4] and remained well below safety limits set by the most stringent government regulations (100 Bq kg^{-1} wet wt, or about 400 Bq kg^{-1} dry wt). The radioactivity from the naturally occurring ^{40}K exceeded that of radiocesium by 2−3 orders of magnitude. Another naturally occurring radionuclide, ^{210}Po, is present in tuna muscle provides a radioactive dose to seafood consumers that is orders of magnitude above the dose resulting from the Fukushima radionuclides.[25,26]

The differences between migrant and resident PBFT (Figure 1B) demonstrate that recent migrants to the CCLME can easily be discerned from CCLME residents of at least one year using radiocesium concentrations; this is particularly apparent in concentrations of ^{134}Cs. ^{134}Cs was detected in only 23% (5 of 22 individuals) of older PBFT (Table 1, Figures 1 and 2). This is an expected result, as many older PBFT in the CCLME are assumed to have migrated during their first year, although some variable proportion migrates in subsequent years.[19] The 5 older fish with detectable ^{134}Cs demonstrate the utility of the radiocesium tracer to discern recent migrants from residents in older fish, for which migration status is unknown.[9,19] The higher concentrations of ^{134}Cs and ^{137}Cs in larger vs smaller migrants may be the result of trophic biomagnification, as young PBFT off Japan show a diet shift to higher trophic level prey during their first year, and larger fish would thus be feeding on higher trophic levels in waters around Japan for a longer time period.[11] Efflux rate constants for Cs may also be greater in smaller versus larger fish, though this has not been measured.

Radiocesium levels in older resident PBFT were not significantly different from those in YFT, which do not migrate from the western Pacific and are residential to the CCLME[5,16] and show no detectable ^{134}Cs and only background levels of ^{137}Cs (Table 2) over two years of data analysis.[4] This suggests that older PBFT which carried radiocesium in 2011 eliminated ^{134}Cs to nondetectable and ^{137}Cs to background (~1 Bq kg^{-1}) levels within one year of residency in the CCLME. Therefore, all PBFT in the CCLME for one year or more can be expected to carry radiocesium levels comparable to pre-Fukushima conditions. This makes identification of CCLME residents particularly straightforward, as presence or absence of ^{134}Cs discerns migrants from residents. All PBFT that are 3−4 years old carried no ^{134}Cs, distinguishing all of these PBFT (n = 9) as >1 year CCLME residents (Figure 2).

The significant linear relationship between PBFT age and time since departure from Japan (Figure 3A) suggests that older fish may have migrated from the western Pacific Ocean before younger fish. Older fish had similar ^{134}Cs:^{137}Cs ratios to smaller fish, and greater estimated time since departure in older fish is a consequence of the greater concentrations (Figure 1B) of ^{134}Cs and ^{137}Cs in older fish than those in younger fish (see description of eq 2 in Materials and Methods). Estimated time since Japan departure, based on ^{134}Cs:^{137}Cs ratios, ranged from 30 to 95 days (SI Table S2) with an average time since departure of approximately two months (57 ± 16 days). This average matches the crossing time (65 days) of an electronically tagged juvenile PBFT making its first trans-Pacific migration, and the shortest time since departure in our study (30 days) is possible given the daily swimming speed (172.3 ± 41.7 km d^{-1}) in that study.[27] If our lowest estimate of 30 d is accurate, it is the fastest reported trans-Pacific migration by a PBFT.

Time since departure for larger PBFT (>1.7 years) was nearly a month longer (79 ± 14 day) (SI Table S2). Variability in the timing of offshore migration has been demonstrated in conventional tagging studies,[19] and it is possible that older, larger fish follow different cues for the initiation of offshore migration. Larger fish would have a higher tolerance for colder waters and would potentially be less affected by oceanographic conditions that may limit migration ability during periods of cooler water before spring and early summer.

Departure from Japan began in late spring, centered on early June (Figure 3B), corresponding with conventional tagging studies that show many PBFT initiate their eastward migration during spring-summer.[19] This early summer departure from Japan may explain why most recent Japan migrants to the CCLME were captured in August and not in earlier summer months in the CCLME (Table 1). Departure-date estimates rely on a predictable and consistent ^{134}Cs:^{137}Cs ratio in PBFT muscle before leaving Japan. Our estimate of an initial ^{134}Cs:^{137}Cs ratio in tuna muscle of 0.73 (SI Table S2) was based on the initial release ratio of 1[2] and the different decay rates of ^{134}Cs and ^{137}Cs; more consistent and publicly accessible measurements of PBFT off Japan would provide the actual data to validate or modify this estimate. However, the average ^{134}Cs:^{137}Cs ratio for coastal and pelagic predatory fish species caught on April 9th, 2012, approximately one year after maximum discharge of radiocesium, was approximately equal (0.74 ± 0.18, n = 10 fish species; for more information see SI).[28] These data support our use of this initial ^{134}Cs:^{137}Cs ratio value (0.73) in PBFT off Japan.

The approach used here, especially applied to large data sets or used to interpret data from other chemical tracers, can improve our understanding of movement patterns of PBFT in the North Pacific Ocean. Understanding the relative proportions of migrants to CCLME residents in relation to oceanographic (e.g., ENSO events) or biological (e.g., sardine or anchovy abundance) conditions can help identify the drivers of the eastward migration of PBFT from Japan to the CCLME. Such studies could be applied to fisheries models which can benefit from migratory information.[12]

One source of error in this study is the use of length as a proxy for age, as there is significant variability in this relationship.[19] Aging using otoliths is a more precise (albeit time-intensive) method,[29] although otoliths were not available for every individual sampled for white muscle tissue. Data for fisheries models in particular may require aging using otoliths. It is also possible that time of departure from Japan is more representative of the time of departure from a broader area now contaminated by Fukushima radiocesium due to dispersal. It has recently been shown that Fukushima-derived radioactive Cs is dispersing further away from the Japanese shoreline and could be reaching waters as far as ~170°E in 2012.[30]

The applicability of our results and approach to other Pacific species that migrate from Japan to distant ecoregions is important. Albacore tuna *Thunnus alalunga*,[31] blue sharks *Prionace glauca*,[32] Pacific loggerhead sea turtles *Caretta caretta*,[33] sooty shearwaters *Puffinus griseus*,[34] salmon sharks *Lamna ditropis*,[35] common minke whales *Balaenoptera acutorostrata*,[36] and other highly migratory species are all known to forage in the Kuroshio Current off eastern Japan and subsequently migrate to regions such as the Okhotsk Sea, the Aleutian Islands, the CCLME, and regions of the South Pacific Ocean. Detection of ^{134}Cs and elevated ^{137}Cs in 100% of small, recently migrated PBFT in 2012 suggests that other species that

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forage near Japan have a high probability of acquiring ^{134}Cs and ^{137}Cs, and the model in eq 2 could be applied to radiocesium data in other migratory species. However, the movement patterns and feeding habits of each study species while near Japan should be taken into account when utilizing this tracer, as PBFT are known to feed specifically in waters near Japan during their juvenile stage, making use of the Kuroshio Current before migrating eastward.[10,11,27] Complementary data (e.g., known life history patterns, electronic tagging, or other chemical tracers) may help interpret radiocesium tracer data in other highly migratory taxa.

Previous studies that measured radionuclides from the Bikini atoll weapons tests in the 1940s−1960s attempted to infer migration patterns of pelagic animals, including yellowfin tuna *T. albacares*[6] and albacore tuna *Thunnus alalunga*.[37−39] However, nuclear detonations led to extensive, widespread fallout from the atmosphere,[40] and results of those pioneering studies could only provide general, highly inferential results. In contrast, the Fukushima plant failure represents a more discrete point source of radionuclides. This unique attribute of Fukushima together with the unique migratory biology of PBFT (i.e., that all small PBFT in the CCLME must be migrants from Japan) provides an unprecedented opportunity to test and apply concentrations of anthropogenic radionuclides to trace animal movement.

Despite the assumptions and gaps in knowledge that result from the Fukushima accident and observations of radioisotopes in marine biota, these radiocesium data present a clear and coherent picture of PBFT migrations that can be interpreted unequivocally. The data shown here are the strongest results possible for the evaluation of radiocesium as a tracer in PBFT and validate its use for the study of large-scale Pacific migrations, as was suggested in 2011.[4] We expect this new tool to produce novel data on the recent migration patterns of keystone pelagic predators, which have been historically difficult to obtain.

■ ASSOCIATED CONTENT

Supporting Information

A description of the fish species (sampled near Japan) used to estimate initial ^{134}Cs:^{137}Cs ratios in fish off Japan around April 6th, 2012 and mean ^{134}Cs:^{137}Cs values of these fish, Table S1 showing error values for ^{134}Cs, ^{137}Cs, and ^{40}K measurements and calculated ^{134}Cs:^{137}Cs ratios, Table S2 with estimated time since departure for individual PBFT, and Figure S1 showing relationship between operculum length (OL) and curved fork length (CFL) used to estimate CFL from OL in PBFT. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Phone: (831) 655-6240; fax: (831) 375-0793; e-mail: dmadigan@stanford.edu.

Notes

The authors declare no competing financial interest.

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■ REFERENCES

(1) Buesseler, K. O.; Jayne, S. R.; Fisher, N. S.; Rypina, I. I.; Baumann, H.; Baumann, Z.; Breier, C. F.; Douglass, E. M.; George, J.; Macdonald, A. M.; Miyamoto, H.; Nishikawa, J.; Pike, S. M.; Yoshida, S. Fukushima-derived radionuclides in the ocean and biota off Japan. *Proc. Natl. Acad. Sci. U.S.A.* **2012**, *109* (16), 5984−5988.

(2) Buesseler, K. O.; Aoyama, M.; Fukasawa, M. Impacts of the Fukushima nuclear power plants on marine radioactivity. *Environ. Sci. Technol.* **2011**, *45* (23), 9931−9935.

(3) Buesseler, K. O. Fishing for answers off Fukushima. *Science* **2012**, *338* (6106), 480−482.

(4) Madigan, D. J.; Baumann, Z.; Fisher, N. S. Pacific bluefin tuna transport Fukushima-derived radionuclides from Japan to California. *Proc. Natl. Acad. Sci. U.S.A.* **2012**, *109* (24), 9483−9486.

(5) Block, B. A.; Jonsen, I. D.; Jorgensen, S. J.; Winship, A. J.; Shaffer, S. A.; Bograd, S. J.; Hazen, E. L.; Foley, D. G.; Breed, G. A.; Harrison, A. L.; Ganong, J. E.; Swithenbank, A.; Castleton, M.; Dewar, H.; Mate, B. R.; Shillinger, G. L.; Schaefer, K. M.; Benson, S. R.; Weise, M. J.; Henry, R. W.; Costa, D. P. Tracking apex marine predator movements in a dynamic ocean. *Nature* **2011**, *475*, 86−90.

(6) Suzuki, Z.; Tomlinson, P. K.; Honma, M. Population structure of Pacific yellowfin tuna. *Inter-Am. Trop. Tuna Comm., Bull.* **1978**, *17*, 277−358.

(7) Rooker, J. R.; Secor, D. H.; Metrio, G. D.; Schloesser, R.; Block, B. A.; Neilson, J. D. Natal homing and connectivity in Atlantic bluefin tuna populations. *Science* **2008**, *322* (5902), 742−744.

(8) Ramos, R.; González-Solís, J. Trace me if you can: The use of intrinsic biogeochemical markers in marine top predators. *Front. Ecol. Environ.* **2012**, *10* (5), 258−266.

(9) Bayliff, W. H. A review of the biology and fisheries for northern bluefin tuna, *Thunnus thynnus*, in the Pacific Ocean. *FAO Fish. Tech. Pap.* **1994**, *336*, 244−295.

(10) Inagake, D.; Yamada, H.; Segawa, K.; Okazaki, M.; Nitta, A.; Itoh, T. Migration of young bluefin tuna, *Thunnus orientalis* Temminck et Schlegel, through archival tagging experiments and its relation with oceanographic conditions in the Western North Pacific. *Bull. Natl. Res. Inst. Far Seas Fish.* **2001**, *38*, 53−81.

(11) Shimose, T.; Watanabe, H.; Tanabe, T.; Kubodera, T. Ontogenetic diet shift of age-0 year Pacific bluefin tuna *Thunnus orientalis*. *J. Fish Biol.* **2012**, DOI: 10.1111/j.1095-8649.2012.03483.x.

(12) Whitlock, R. E.; McAllister, M. K.; Block, B. A. Estimating fishing and natural mortality rates for Pacific bluefin tuna (*Thunnus orientalis*) using electronic tagging data. *Fish. Res.* **2012**, *119−120*, 115−127.

(13) Pacific Bluefin Stock Assessment; International Scientific Committee for Tuna and Tuna-like Species in the North Pacific Ocean, 2012. http://isc.ac.affrc.go.jp/pdf/Stock_assessment/Final_Assessment_Summary_PBF.pdf (accessed December 21, 2012).

(14) Mathews, T.; Fisher, N. S. Dominance of dietary intake of metals in marine elasmobranch and teleost fish. *Sci. Total Environ.* **2009**, *407* (18), 5156−5161.

(15) Mathews, T.; Fisher, N. S.; Jeffree, R. A.; Teyssie, J. L. Assimilation and retention of metals in teleost and elasmobranch fishes following dietary exposure. *Mar. Ecol.: Prog. Ser.* **2008**, *360* (1), 1−12.

(16) Schaefer, K.; Fuller, D.; Block, B. Movements, behavior, and habitat utilization of yellowfin tuna (*Thunnus albacares*) in the northeastern Pacific Ocean, ascertained through archival tag data. *Mar. Biol.* **2007**, *152* (3), 503−525.

(17) Buesseler, K. O.; Casso, S. A.; Hartman, M. C.; Livingston, H. D. Determination of fission-products and actinides in the Black Sea following the Cherynobyl accident. *J. Radioanal. Nucl. Chem.* **1990**, *138* (1), 33−47.

(18) Currie, L. A. Limits for qualitative detection and quantitative determination. Application to radiochemistry. *Anal. Chem.* **1968**, *40* (3), 586−593.

(19) Bayliff, W. H.; Ishizuka, Y.; Deriso, R. Growth, movement, and attrition of northern bluefin tuna, *Thunnus thynnus*, in the Pacific Ocean, as determined by tagging. *Inter-Am. Trop. Tuna Comm., Bull.* **1991**, *20*, 3−94.
(20) Wild, A. A review of the biology and fisheries for yellowfin tuna, *Thunnus albacares*, in the Eastern Pacific Ocean. *FAO Fish. Tech. Pap.* **1994**, *336*, 52−107.
(21) Farwell, C. J. Based on a regression by C. Farwell. *Tuna Research and Conservation Center* **2000**, 1.
(22) Scida, P.; Rainosek, A.; Lowery, T. Length conversions for yellowfin tuna (*Thunnus albacares*) caught in the western North Atlantic Ocean. *Collect. Vol. Sci. Pap. ICCAT* **2001**, *52* (2), 528−532.
(23) IAEA. *Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment*. International Atomic Energy Agency: Vienna, Austria, 2004.
(24) Wang, W.-X.; Ke, C.; Yu, K. N.; Lam, P. K. S. Modeling radiocesium bioaccumulation in a marine food chain. *Mar. Ecol.: Prog. Ser.* **2000**, *208*, 41−50.
(25) Yamamoto, M.; Abe, T.; Kuwabara, J.; Komura, K.; Ueno, K.; Takizawa, Y. Polonium-210 and lead-210 in marine organisms: Intake levels for Japanese. *J. Radioanal. Nucl. Chem.* **1994**, *178* (1), 81−90.
(26) Fisher, N. S., Beaugelin-Seiller, K., Hinton, T. G., Baumann, Z., Madigan, D. J., &Garnier-LaPlace, J. An evaluation of radiation doses and associated risk from the Fukushima nuclear accident to marine biota and human consumers of seafood. *Proc. Natl. Acad. Sci. U.S.A.*, in review.
(27) Kitagawa, T.; Kimura, S.; Nakata, H.; Yamada, H.; Nitta, A.; Sasai, Y.; Sasaki, H. Immature Pacific bluefin tuna, *Thunnus orientalis*, utilizes cold waters in the Subarctic Frontal Zone for trans-Pacific migration. *Environ. Biol. Fish.* **2009**, *84* (2), 193−196.
(28) *Results of the Inspection on Radioactivity Materials in Fisheries Products*; Japanese Ministry of Agriculture, Forestry, and Fisheries: Tokyo, 2011; http://www.jfa.maff.go.jp/e/inspection/index.html.
(29) Shimose, T.; Tanabe, T.; Chen, K.-S.; Hsu, C.-C. Age determination and growth of Pacific bluefin tuna, *Thunnus orientalis*, off Japan and Taiwan. *Fish. Res.* **2009**, *100* (2), 134−139.
(30) Nakano, M.; Povinec, P. P. Long-term simulations of the 137Cs dispersion from the Fukushima accident in the world ocean. *J. Environ. Radioact.* **2012**, *111*, 109−115.
(31) Childers, J.; Snyder, S.; Kohin, S. Migration and behavior of juvenile North Pacific albacore (*Thunnus alalunga*). *Fish. Oceanogr.* **2011**, *20* (3), 157−173.
(32) Nakano, H. Age reproduction and migration of blue shark in the North Pacific Ocean. *Bull. Natl. Res. Inst. Far Seas Fish.* **1994**, *31*, 141−256.
(33) Peckham, S. H.; Diaz, D. M.; Walli, A.; Ruiz, G.; Crowder, L. B.; Nichols, W. J. Small-scale fisheries bycatch jeopardizes endangered Pacific loggerhead turtles. *PLoS ONE* **2007**, *2* (10), e1041.
(34) Shaffer, S. A.; Tremblay, Y.; Weimerskirch, H.; Scott, D.; Thompson, D. R.; Sagar, P. M.; Moller, H.; Taylor, G. A.; Foley, D. G.; Block, B. A.; Costa, D. P. Migratory shearwaters integrate oceanic resources across the Pacific Ocean in an endless summer. *Proc. Natl. Acad. Sci. U.S.A.* **2006**, *103* (34), 12799−12802.
(35) Nagasawa, K. Predation by salmon sharks (*Lamna ditropis*) on Pacific salmon (*Oncorhynchus* spp.) in the North Pacific Ocean. *North Pac. Anadr. Fish Comm. Bull.* **1998**, *1*, 419−433.
(36) *Report of the Sub-Committee on Minke Whales*; International Whaling Commission: Cambridge, 1983; http://www.iwcoffice.org/annual-reports.
(37) Hodge, V. F.; Folsom, T. R.; Young, D. R. Retention of fall-out constituents in upper layers of the Pacific Ocean as estimated from studies of a tuna population. In *IAEA Symposium on the Interaction of Radioactive Contaminants with the Constituents of the Marine Environment*, Seattle, Washington, 1972.
(38) Young, D. R.; Folsom, T. R.; Hodge, V. F. ^{137}Cs and ^{40}K in the flesh of Pacific albacore, 1964−1974. *Health Phys.* **1975**, *29* (5), 689−694.
(39) Suda, A. Hoshano maguro (Tunas contaminated by radioactivity) (in Japanese). *Suisan Kenkyusha Kyogikai, Suisan Kagaku* **1956**, *6* (1), 16−22.
(40) Volchok, H. L., Bowen, V. T., Folsom, T. R., Broecker, W. S., Schuert, E. A., &Bien, G. S. Oceanic distributions of radionuclides from nuclear explosions. In *Radioactivity in the Marine Environment*; National Academy of Sciences: Washington, DC, 1971; pp 42−89.