Long-lived radionuclides from the Fukushima nuclear power plant in Japan, and consequences for Pacific ecosystems and seafood consumers

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Brookhaven National Lab, September 2016
Fukushima Daiichi Nuclear power plant

March 11, 2011
Earthquake, Tsunami ...and a failed Nuclear Power Plant

Buesseler et al. 2011. ES&T
SAMPLING & RADIOANALYSIS

smaller mixed zooplankton

deflect mixed zooplankton

and myctophid fish

Anthropogenic radioisotopes:

$^{134}\text{Cs}$, $^{137}\text{Cs}$, $^{110}\text{m Ag}$

Japan

US - Hawaii

US - Stony Brook

Long Island

Photos by Ken Kostel
Background information on radioactive iodine

\(^{131}\text{I} \): 8-day half-life; after 70 days only 0.1% remaining; after 1 year 0.000000000002% remaining

\(^{127}\text{I} \): stable, present at concentrations of ~ 400 nM in seawater

I bioconcentrated about 10,000 times out of seawater by brown macroalgae (active uptake), but relatively little bioaccumulation in most other marine organisms
Background information on radioactive cesium

\(^{137}\text{Cs}: \) 30-year half-life; still detectable in Pacific seawater and trace levels in biota as remnants from nuclear weapons testing in the Pacific, which peaked in the 1960s (fission product)

\(^{134}\text{Cs}: \) 2.1-year half-life: undetectable in Pacific waters and biota (neutron activation product) prior to Fukushima accident

\(^{133}\text{Cs}: \) stable, present at concentrations of ~40 nM in seawater
Cesium

non-essential for all organisms, generally follows K uptake pathways and tissue distributions

shows low uptake in marine phytoplankton (Kds < 100) but much higher in freshwater where there are orders of magnitude lower K, Na concentrations (Kds ~10,000)

modest biomagnification in marine food chains (much less than methylmercury)—high assimilation efficiencies (~70%) in fish and daily loss rates of ~2% from excretion; ~2/3 of Cs from diet

concentrates in muscle tissue (e.g., filet of fish)
Particulate vs. dissolved $^{137}\text{Cs}$ in waters off Japan
Buesseler et al. (PNAS 2012)
### Mean concentrations (Bq kg$^{-1}$ dry) in biota

<table>
<thead>
<tr>
<th></th>
<th>$^{137}$Cs</th>
<th>$^{134}$Cs</th>
<th>$^{110m}$Ag</th>
<th>$^{40}$K</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Copepods</strong></td>
<td>14.8</td>
<td>17.7</td>
<td>8.4</td>
<td>199</td>
</tr>
<tr>
<td>n = 17</td>
<td>CF = 79</td>
<td>CF = 71</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Large zooplankton</strong></td>
<td>11.1</td>
<td>13.7</td>
<td>17.9</td>
<td>217</td>
</tr>
<tr>
<td>(euphausiids, gelatinous)</td>
<td>CF = 16</td>
<td>CF = 22</td>
<td></td>
<td></td>
</tr>
<tr>
<td>n = 4</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Fish</strong></td>
<td>10.7</td>
<td>10.0</td>
<td>bd</td>
<td>168</td>
</tr>
<tr>
<td>n = 3</td>
<td>CF = 16</td>
<td>CF = 13</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
CESIUM ISOTOPIC RATIOS IN BIOTA ~1

Data from Buesseler et al. 2012 PNAS

1:1
**RANGES OF RADIOISOTOPES IN BIOTA**

Data from Buesseler et al. 2012 PNAS

Ratio of mean natural : anthropogenic = \(6 : 1\)

Pre-Fukushima \(^{137}\text{Cs}\) = 37 - 80 x \(10^{-3}\) Bq dry kg\(^{-1}\)

Kaeriyama et al., 2008
Elevated Cs concentration in demersal fish off Fukushima

Changes in total cesium ($^{137}$Cs and $^{134}$Cs in Bq/kg wet) over time in bottom fish in eastern Japan.

Total cesium in five different fish types as a function of time.

Constantly elevated Cs concentration in benthic invertebrates and sediment

Release of $^{137}$Cs from Japanese sediment to overlying seawater
The effect of bioturbation on Cs release from Japanese sediment to overlying seawater
## Comparison of dietary uptake parameters

<table>
<thead>
<tr>
<th>Organism</th>
<th>Diet</th>
<th>Assimilation Efficiency (%)</th>
<th>Efflux rate constant $k_{ef} (d^{-1})$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Polychaete Nereis succinea</strong></td>
<td>Sediment (IO2 station)</td>
<td>15.8 ± 7.6</td>
<td>0.431 ± 0.073</td>
</tr>
<tr>
<td></td>
<td>Sediment (NP1 station)</td>
<td>18.5 ± 7.32</td>
<td>0.425 ± 0.143</td>
</tr>
<tr>
<td><strong>Asian shore crab Hemigrapsus sanguineus</strong></td>
<td>Polychaete exposed to seawater</td>
<td>54.5 ± 10.6</td>
<td>0.138 ± 0.064</td>
</tr>
<tr>
<td></td>
<td>Polychaete exposed to sediment</td>
<td>27.3 ± 5.25</td>
<td>0.059 ± 0.024</td>
</tr>
<tr>
<td><strong>Killifish Fundulus heteroclitus</strong></td>
<td>California black worm</td>
<td>78.5 ± 7.2</td>
<td>0.045 ± 0.013</td>
</tr>
</tbody>
</table>

The assimilation efficiency was highest in killifish, followed by the Asian shore crab, and lowest in polychaetes, while loss rate constants were highest in the polychaetes and lowest in killifish.

Polychaetes feeding on IO2 sediment had similar assimilation efficiencies and loss rate constants of Cs as those feeding on NP1 sediment.

Both the assimilation efficiency and loss rate constant of Cs from killifish were similar to those of 3 fish species: *P. maxima*, *S. auratus*, *S. canicula* (Mathews et al. 2008).
Comparison of aqueous uptake parameters

<table>
<thead>
<tr>
<th>Organism</th>
<th>Uptake rate constant $k_u$ (mL g$^{-1}$ d$^{-1}$)</th>
<th>Efflux rate constant $k_{ew}$ (d$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polychaete <em>Nereis succinea</em></td>
<td>10.3 ± 1.7</td>
<td>0.2 ± 0.01</td>
</tr>
<tr>
<td>Asian shore crab <em>Hemigrapsus sanguineus</em></td>
<td>1.5 ± 0.1</td>
<td>0.1 ± 0.01</td>
</tr>
<tr>
<td>Killifish <em>Fundulus heteroclitus</em></td>
<td>0.6 ± 0.1</td>
<td>0.06 ± 0.008</td>
</tr>
</tbody>
</table>

- Both uptake rate constant and loss rate constant are highest in polychaetes, followed by the Asian shore crab, and lowest in killifish.
### Fraction of $^{137}$Cs from diet

<table>
<thead>
<tr>
<th>Polychaetes eating sediment</th>
<th>Crabs eating worms</th>
<th>Fish eating worms</th>
</tr>
</thead>
<tbody>
<tr>
<td>98%</td>
<td>98%</td>
<td>99%</td>
</tr>
</tbody>
</table>
Conclusions from sediment experiments

1. These experimental results suggest that Cs can desorb from contaminated sediments at rates influenced by bioturbation and can be a source of Cs for marine benthic fauna.

2. Efficient assimilation of Cs from prey can lead to its build-up in benthic food chains. Our findings help explain why bottom fish remain more contaminated by radiocesium than pelagic fish.
Pacific bluefin tuna

Extremely large home range

Trans-oceanic migrations

Discrete spawning area
Pacific bluefin life history

- Born
- Some stay
- Some migrate east
- Return west to spawn
- Growth
Fukushima accident: 2011

Massive release of radiocesium into the ocean

Did bluefin carry it over?

We measured 15 fish caught off San Diego…….

Buesseler at al 2011, ES&T

Kitagawa et al 2009, Env Biol Fish
...and all 15 did.

15 Post-Fukushima bluefin: all measurable $^{134}\text{Cs}$ and elevated $^{137}\text{Cs}$
5 Pre-Fukushima (2008) bluefin: background $^{137}\text{Cs}$
5 Post Fukushima yellowfin in eastern Pacific: background $^{137}\text{Cs}$

Madigan et al 2012, PNAS
Measured $^{134}\text{Cs}$, $^{137}\text{Cs}$, and the natural $^{40}\text{K}$ for post-Fukushima bluefin (PBFT 2011), pre-Fukushima bluefin (PBFT 2008), and post-Fukushima yellowfin tuna (YFT 2011)

<table>
<thead>
<tr>
<th>SL</th>
<th>Body mass</th>
<th>Age</th>
<th>$^{134}\text{Cs}$</th>
<th>$^{137}\text{Cs}$</th>
<th>$^{40}\text{K}$</th>
<th>$^{134}\text{Cs}$:$^{137}\text{Cs}$</th>
<th>$^{134}$+137Cs</th>
</tr>
</thead>
<tbody>
<tr>
<td>cm</td>
<td>kg dry</td>
<td>years</td>
<td>Bq kg$^{-1}$ dry mass</td>
<td>Bq kg$^{-1}$ dry mass</td>
<td>Bq kg$^{-1}$ dry mass</td>
<td>Bq kg$^{-1}$ dry mass</td>
<td>Bq kg$^{-1}$ dry mass</td>
</tr>
<tr>
<td>-----</td>
<td>-----------</td>
<td>------</td>
<td>------------------</td>
<td>------------------</td>
<td>---------------</td>
<td>----------------</td>
<td>-------------</td>
</tr>
<tr>
<td>Median</td>
<td>66.5</td>
<td>1.5</td>
<td>1.5</td>
<td>4.3</td>
<td>6.0</td>
<td>367</td>
<td>0.66</td>
</tr>
<tr>
<td>Mean</td>
<td>66.2</td>
<td>1.5</td>
<td>1.5</td>
<td>4.0</td>
<td>6.3</td>
<td>347</td>
<td>0.62</td>
</tr>
<tr>
<td>SD</td>
<td>3.6</td>
<td>0.2</td>
<td>0.1</td>
<td>1.4</td>
<td>1.5</td>
<td>49</td>
<td>0.14</td>
</tr>
<tr>
<td>Median</td>
<td>66.3</td>
<td>1.5</td>
<td>1.4</td>
<td>0</td>
<td>1.4</td>
<td>266</td>
<td>0</td>
</tr>
<tr>
<td>Mean</td>
<td>66.2</td>
<td>1.5</td>
<td>1.4</td>
<td>0</td>
<td>1.4</td>
<td>258</td>
<td>0</td>
</tr>
<tr>
<td>SD</td>
<td>1.2</td>
<td>0.09</td>
<td>0.05</td>
<td>0</td>
<td>0.2</td>
<td>43</td>
<td>0</td>
</tr>
<tr>
<td>Median</td>
<td>72.3</td>
<td>1.9</td>
<td>1.2</td>
<td>0</td>
<td>1.2</td>
<td>342</td>
<td>0</td>
</tr>
<tr>
<td>Mean</td>
<td>72.3</td>
<td>1.9</td>
<td>1.2</td>
<td>0</td>
<td>1.1</td>
<td>333</td>
<td>0</td>
</tr>
<tr>
<td>SD</td>
<td>2.5</td>
<td>0.2</td>
<td>0.01</td>
<td>0</td>
<td>0.4</td>
<td>78</td>
<td>0</td>
</tr>
</tbody>
</table>

Madigan et al., PNAS, 2012
Simplified movement patterns for juvenile Pacific bluefin tuna (blue arrows) from Japan to the California and juvenile yellowfin tuna (yellow arrows)
Cesium concentrations in post-Fukushima bluefin tuna

Japan safety limit: 100 Bq kg\(^{-1}\) wet wt (~410 Bq kg\(^{-1}\) dry wt)

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

\(^{134}\text{Cs}:^{137}\text{Cs}\) in myctophid fish off Japan = 0.9; in plankton = 1.1; in water = 1.0
U.S. Tuna Has Fukushima Taint

Fukushima Radiation May Actually Save Bluefin Tuna

Bluefin tuna carried a little radiation from Japan to California, study says

Nuclear Tuna Is Hot News, But Not Because It's Going To Make You Sick

Democrat worried about tainted seafood from Japan's nuclear meltdown

Parast põgusat kontumist Jaapaniga võib panati mitte jaada mulle, nagu oleks tegu ühiskonnaga, kus kõik alluvad valikides grupile ning iseajal on individu eile eksisteeriti.

U.K.

BBC NEWS SCIENCE & ENVIRONMENT

Bluefin tuna record Fukushima radioactivity

CNN México

Científicos hallan cesio de Fukushima en atún capturado en Estados Unidos

Japan

DAILY YOMIURI ONLINE THE DAILY YOMIURI

Low-level cesium found in tuna off U.S.

Portugal

Expresso MT

Atum com radiação de Fukushima cruza o Pacífico e chega aos EUA

East Asia

The Asian Tiger

Consequences Fukushima nuclear disaster persist

Argentina

Bariloche

Fukushima y la vida marina

Estonia

Ekspresse.ee

Parast põgusat kontumist Jaapaniga võib panati mitte jaada mulle, nagu oleks tegu ühiskonnaga, kus kõik alluvad valikides grupile ning iseajal on individu eile eksisteeriti.
Historical Anthropogenic Radionuclide Input to the Oceans

- Total radioactivity, dumped into oceans: $6 \times 10^4$ TBq
- Total radioactivity, atmospheric nuclear testing: $2 \times 10^8$ TBq
- Total radioactivity, naturally in the ocean: $2 \times 10^{10}$ TBq, about 90% of which is $^{40}$K
Risks to humans

• Excess relative risk of fatal cancer above natural incidence of the disease = 4.1-4.8% per Sv.

• Statistically significant elevations in cancer risk are observed at doses >100-200 mSv.
## Doses to human consumers

(1 Sv = 100 rem = 1 joule kg\(^{-1}\))

<table>
<thead>
<tr>
<th>Consumption of:</th>
<th>Tuna: from (^{134+137}Cs)</th>
<th>Tuna: from (^{40}K)</th>
<th>Banana: from (^{40}K)</th>
<th>Dental x-ray</th>
<th>1 flight NY to LA (cosmic rays)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>200 g</td>
<td>200 g</td>
<td>1 banana</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dose (µSv)</td>
<td>0.008</td>
<td>0.1</td>
<td>0.1</td>
<td>5</td>
<td>40</td>
</tr>
</tbody>
</table>

Top 5% of recreational fishermen in California (µSv y\(^{-1}\))

|                 | 4.7                          | 61.8                 | = 47 bananas/y         |

Average seafood consumer in US (µSv y\(^{-1}\))

|                 | 0.93                         | 12.7                 | = 9.3 bananas/y        |

Average seafood consumer in Japan (µSv y\(^{-1}\))

|                 | 32.7                         | 31.5                 | = 346 bananas/y        |

Avid seafood eaters would get doses >5 orders of magnitude lower than minimum levels leading to cancer! (Fisher et al., PNAS, 2013)
Fukushima-derived radionuclide doses to American and Japanese consumers; assumes annual fish consumption rates in the US = 24.1 kg y\(^{-1}\) and in Japan = 56.6 kg y\(^{-1}\).

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>PBFT source</th>
<th>(\mu\text{Sv annual consumption})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{134}\text{Cs})</td>
<td>USA 8/2011</td>
<td>0.4</td>
</tr>
<tr>
<td>(^{137}\text{Cs})</td>
<td>USA 8/2011</td>
<td>0.5</td>
</tr>
<tr>
<td>(^{40}\text{K})</td>
<td>USA 8/2011</td>
<td>12.7</td>
</tr>
<tr>
<td>(^{210}\text{Po})</td>
<td>USA 8/2011</td>
<td>558</td>
</tr>
<tr>
<td>(^{134}\text{Cs})</td>
<td>Japan 4/2011</td>
<td>15.7</td>
</tr>
<tr>
<td>(^{137}\text{Cs})</td>
<td>Japan 4/2011</td>
<td>16.9</td>
</tr>
<tr>
<td>(^{40}\text{K})</td>
<td>Japan 4/2011</td>
<td>29.7</td>
</tr>
<tr>
<td>(^{210}\text{Po})</td>
<td>Japan 4/2011</td>
<td>1310</td>
</tr>
</tbody>
</table>
Internal absorbed dose rates to zooplankton and bluefin tuna (nGy h\(^{-1}\)); ICRP benchmark = 10,000 nGy h\(^{-1}\))

<table>
<thead>
<tr>
<th></th>
<th>(^{110}\text{mAg})</th>
<th>(^{134}\text{Cs})</th>
<th>(^{137}\text{Cs})</th>
<th>(^{210}\text{Po})</th>
<th>(^{40}\text{K})</th>
<th>Natural : anthropogenic</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zooplankton (near Japan)</td>
<td>0.76</td>
<td>1.5</td>
<td>1.8</td>
<td>1700</td>
<td>0.1</td>
<td>420</td>
</tr>
<tr>
<td>Tuna (in California)</td>
<td>n.d.</td>
<td>0.6</td>
<td>1.1</td>
<td>600</td>
<td>1.3</td>
<td>354</td>
</tr>
<tr>
<td>Tuna (near Japan)</td>
<td>n.d.</td>
<td>9</td>
<td>16.5</td>
<td>600</td>
<td>1.3</td>
<td>24</td>
</tr>
</tbody>
</table>

(1 Gy = absorption of 1 joule kg\(^{-1}\) of tissue from ionizing radiation)

nd: not detected (Fisher et al., PNAS, 2013)
Benchmark safety levels for wildlife

$^{210}$Po dose

(Fisher et al., PNAS, 2013)
Simplified migration patterns of some highly migratory species in the Pacific
1. What is the Cesium load of PBFT in 2012 and 2013?

- Our 2011 data are from fish that spent ~1 month in contaminated waters.
- 2012’s cohort spent their first year in contaminated waters.
- Values may be much higher.
Comparison of 2011 and 2012 tuna

Madigan et al., Environ. Sci. Technol., 2013
Radiocesium in bluefin in 2012 validates new tracer technique

Remember smallest bluefin must have migrated recently....

Every PBFT less than 1.7 years had signal

Most PBFT years 1.7 – 4 years old were CCLME residents (17 of 22)

Madigan et al 2013, ES&T
Post-Fukushima changes in cesium activity in Pacific bluefin tuna that have crossed the Pacific to California.

black bars: $^{134}\text{Cs}$; gray bars: $^{137}\text{Cs}$; triangles: $^{134}\text{Cs}:^{137}\text{Cs}$
NOT BLUEFIN TUNA: All samples (n = 91) had undetectable $^{134}$Cs (2012-2015)
Still a work in progress—stay tuned…

Conclusions so far:

1. $^{134}\text{Cs}$ and $^{137}\text{Cs}$ accumulate in bluefin tuna in waters off Japan and are retained by tuna during their migration across the Pacific;

2. Yellowfin tuna which are residential species show no evidence of Fukushima radioactivity;

3. Radioactivity clearly detectable in tuna in California coastal waters, but at low concentrations compared to natural radioactivity, and doses to marine biota and to human consumers are low;

4. Cs isotopes are useful in tracing migration of some fish, mammals, turtles, birds.
Thanks!

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