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

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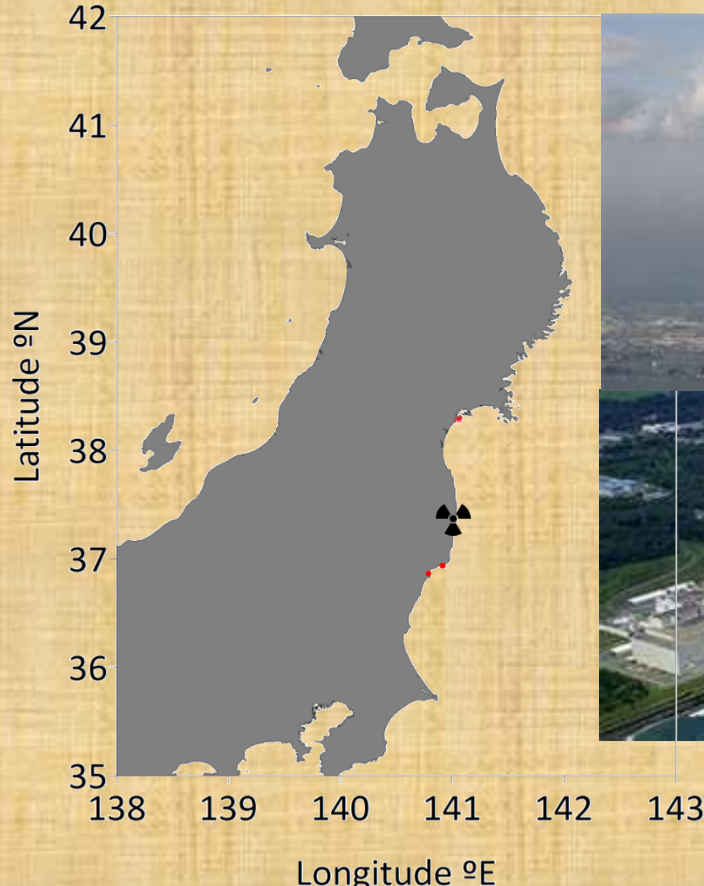
Stony Brook University

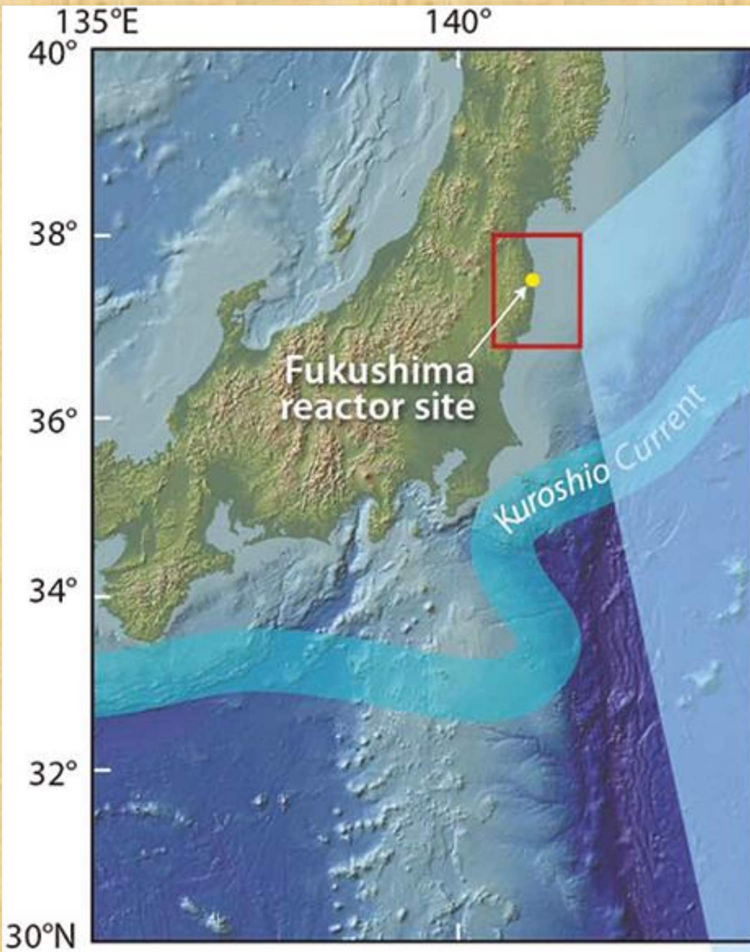
Brookhaven National Lab, September 2016

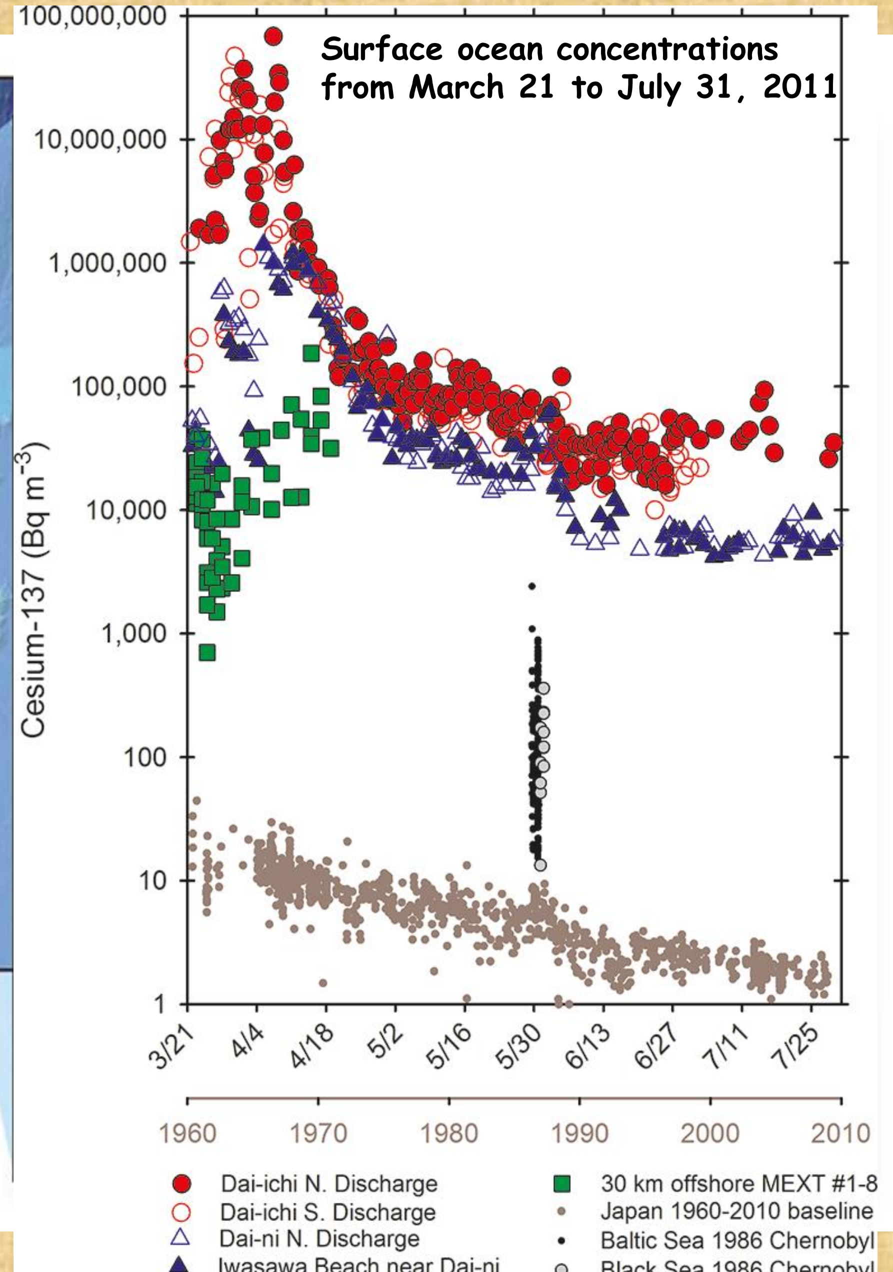
Fukushima Daiichi Nuclear power plant



March 11, 2011

Earthquake, Tsunami …and a failed Nuclear Power Plant



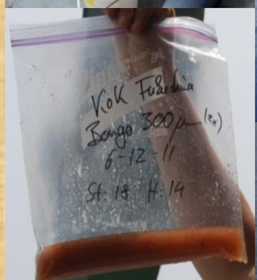


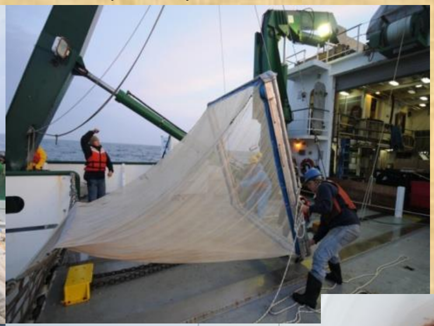
Buesseler et al. 2011. ES&T

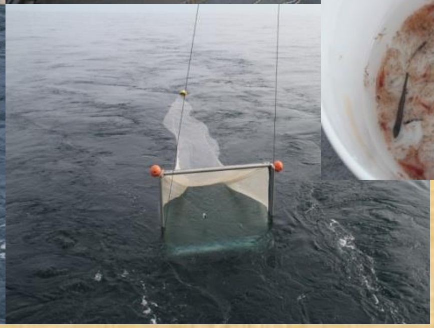
SAMPLING & RADIOANALYSIS

smaller mixed zooplankton

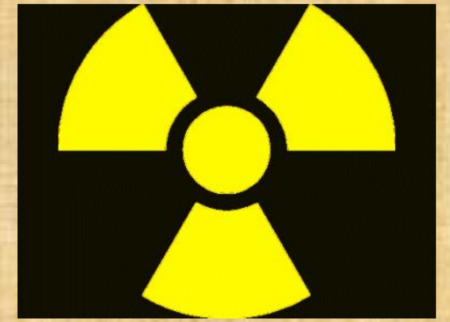
bigger mixed zooplankton and myctophid fish







Anthropogenic radioisotopes:
^{134}Cs, ^{137}Cs, ^{110m}Ag



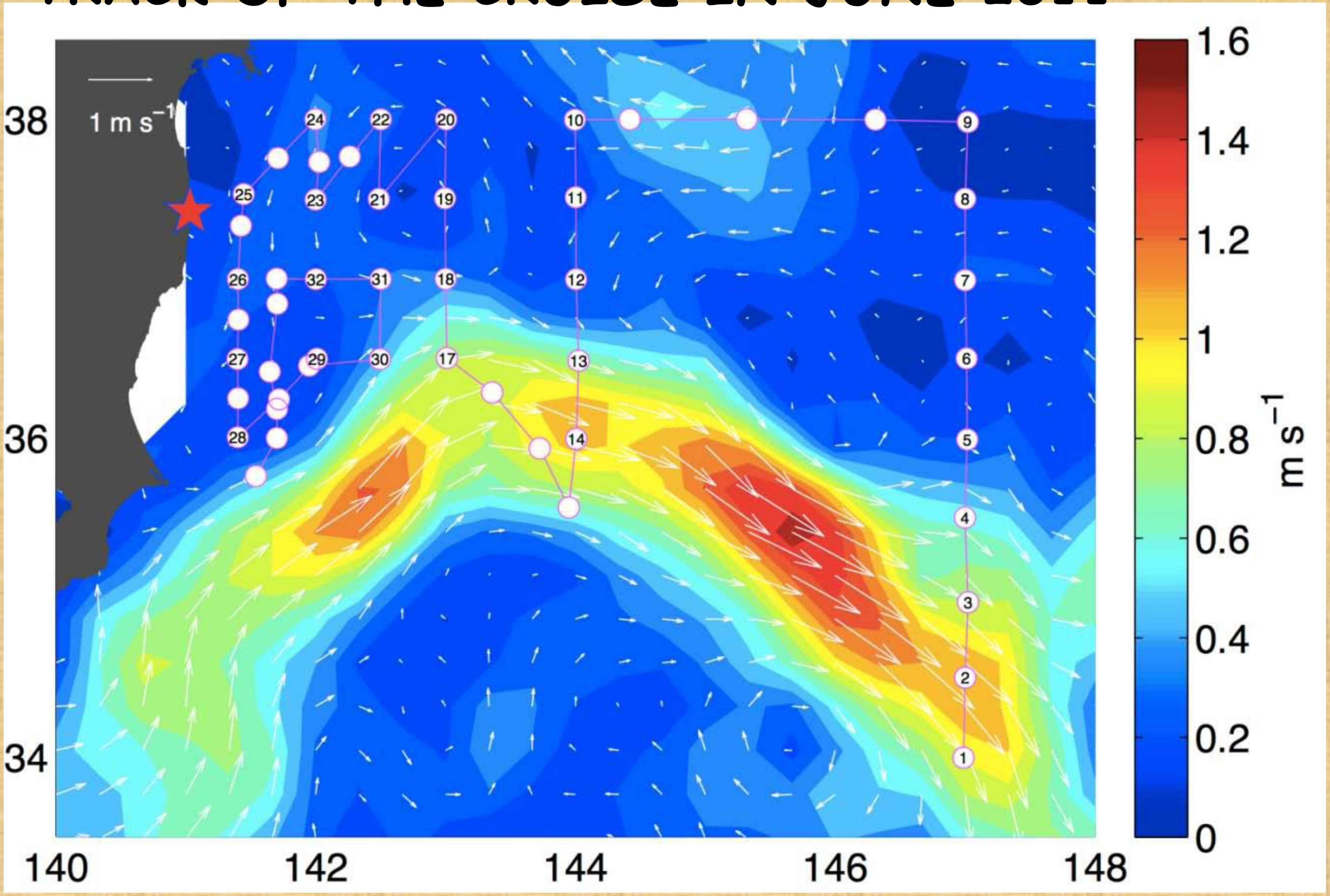
Japan

US - Hawaii

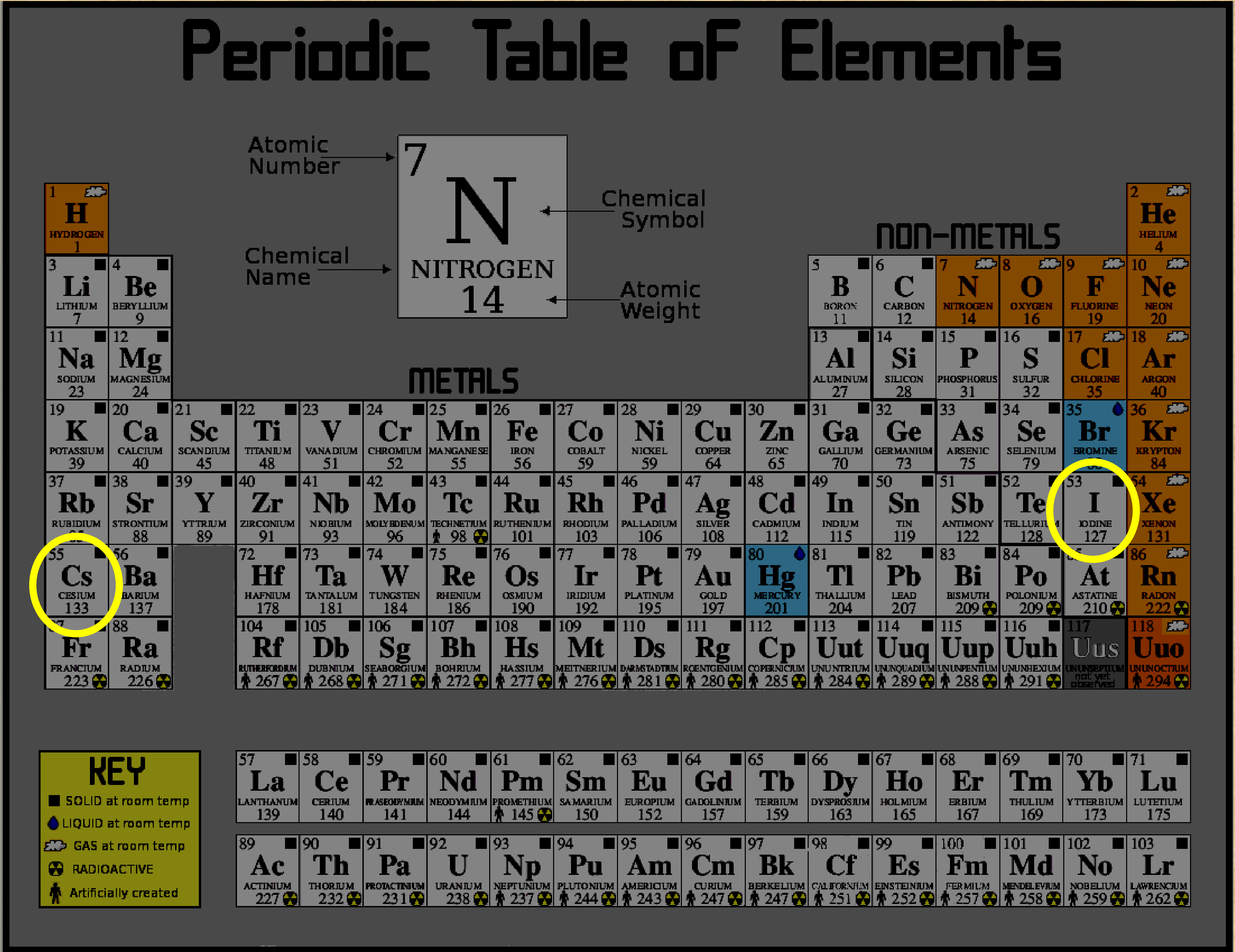
US – Stony Brook
Long Island

Photos by Ken Kostel

TRACK OF THE CRUISE IN JUNE 2011



Buesseler et al. (PNAS 2012)



Periodic Table of Elements
Atomic Number
Chemical Symbol
Chemical Name
Atomic Weight
7 N NITROGEN 14
NON-METALS
METALS
1 H HYDROGEN 1
2 He HELIUM 4
3 Li LITHIUM 7
4 Be BERYLLIUM 9
5 B BORON 11
6 C CARBON 12
7 N NITROGEN 14
8 O OXYGEN 16
9 F FLUORINE 19
10 Ne NEON 20
11 Na SODIUM 23
12 Mg MAGNESIUM 24
13 Al ALUMINUM 27
14 Si SILICON 28
15 P PHOSPHORUS 31
16 S SULFUR 32
17 Cl CHLORINE 35
18 Ar ARGON 40
19 K POTASSIUM 39
20 Ca CALCIUM 40
21 Sc SCANDIUM 45
22 Ti TITANIUM 48
23 V VANADIUM 51
24 Cr CHROMIUM 52
25 Mn MANGANESE 55
26 Fe IRON 56
27 Co COBALT 59
28 Ni NICKEL 59
29 Cu COPPER 64
30 Zn ZINC 65
31 Ga GALLIUM 70
32 Ge GERMANIUM 73
33 As ARSENIC 75
34 Se SELENIUM 79
35 Br BROMINE
36 Kr KRYPTON 84
37 Rb RUBIDIUM
38 Sr STRONTIUM 88
39 Y YTTRIUM 89
40 Zr ZIRCONIUM 91
41 Nb NIOBIUM 93
42 Mo MOLYBDENUM 96
43 Tc TECHNETIUM 98
44 Ru RUTHENIUM 101
45 Rh RHODIUM 103
46 Pd PALLADIUM 106
47 Ag SILVER 108
48 Cd CADMIUM 112
49 In INDIUM 115
50 Sn TIN 119
51 Sb ANTIMONY 122
52 Te TELLURIUM 128
53 I IODINE 127
54 Xe XENON 131
Cs CESIUM 133
56 Ba BARIUM 137
72 Hf HAFNIUM 178
73 Ta TANTALUM 181
74 W TUNGSTEN 184
75 Re RHENIUM 186
76 Os OSMIUM 190
77 Ir IRIDIUM 192
78 Pt PLATINUM 195
79 Au GOLD 197
80 Hg MERCURY 201
81 Tl THALLIUM 204
82 Pb LEAD 207
83 Bi BISMUTH 209
84 Po POLONIUM 209
At ASTATINE 210
86 Rn RADON 222
Fr FRANCIUM 223
88 Ra RADIUM 226
104 Rf RUTHERFORDIUM 267
105 Db DUBNIUM 268
106 Sg SEABORGIUM 271
107 Bh BOHRIUM 272
108 Hs HASSIUM 277
109 Mt MEITNERIUM 276
110 Ds DARMSTADTIUM 281
111 Rg ROENTGENIUM 280
112 Cp COPERNICIUM 285
113 Uut UNUNTRIUM 284
114 Uuq UNUNQUADIUM 289
115 Uup UNUNPENTIUM 288
116 Uuh UNUNHEXIUM 291
117 Uus UNUNSEPTIUM not yet observed
118 Uuo UNUNOCTIUM 294
57 La LANTHANUM 139
58 Ce CERIUM 140
59 Pr PRASEODYMIUM 141
60 Nd NEODYMIUM 144
61 Pm PROMETHIUM 145
62 Sm SAMARIUM 150
63 Eu EUROPIUM 152
64 Gd GADOLINIUM 157
65 Tb TERBIUM 159
66 Dy DYSPROSIUM 163
67 Ho HOLMIUM 165
68 Er ERBIUM 167
69 Tm THULIUM 169
70 Yb YTTERBIUM 173
71 Lu LUTETIUM 175
89 Ac ACTINIUM 227
90 Th THORIUM 232
91 Pa PROTACTINIUM 231
92 U URANIUM 238
93 Np NEPTUNIUM 237
94 Pu PLUTONIUM 244
95 Am AMERICIUM 243
96 Cm CURIUM 247
97 Bk BERKELIUM 247
98 Cf CALIFORNIUM 251
99 Es EINSTEINIUM 252
100 Fm FERMIUM 257
101 Md MENDELEVIUM 258
102 No NOBELIUM 259
103 Lr LAWRENCIUM 262
KEY
SOLID at room temp
LIQUID at room temp
GAS at room temp
RADIOACTIVE
Artificially created

Background information on radioactive iodine

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

^{127}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}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}Cs: 2.1-year half-life: undetectable in Pacific waters and biota (neutron activation product) prior to Fukushima accident

^{133}Cs: stable, present at concentrations of ~40 nM in seawater

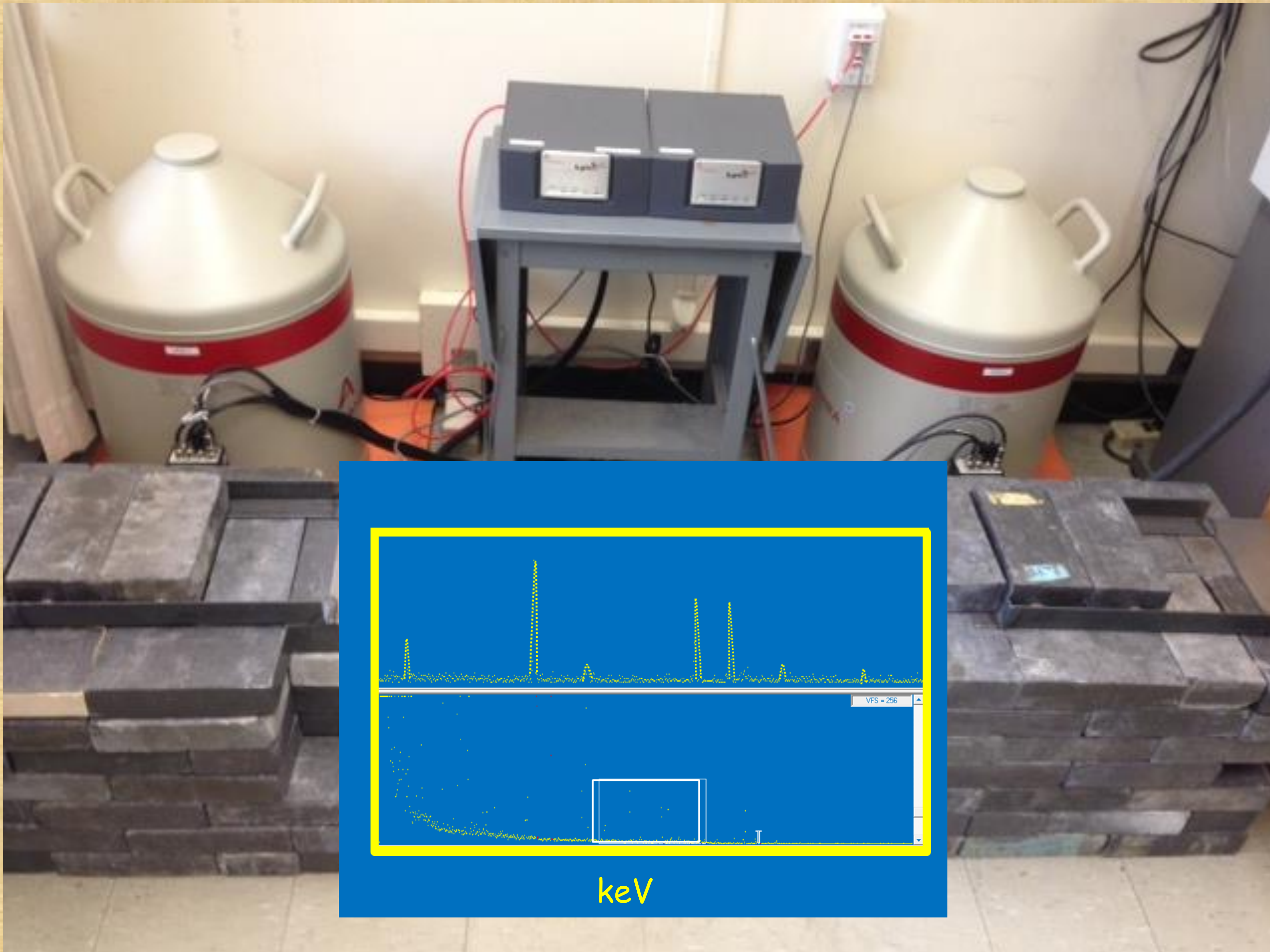
Cesium

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

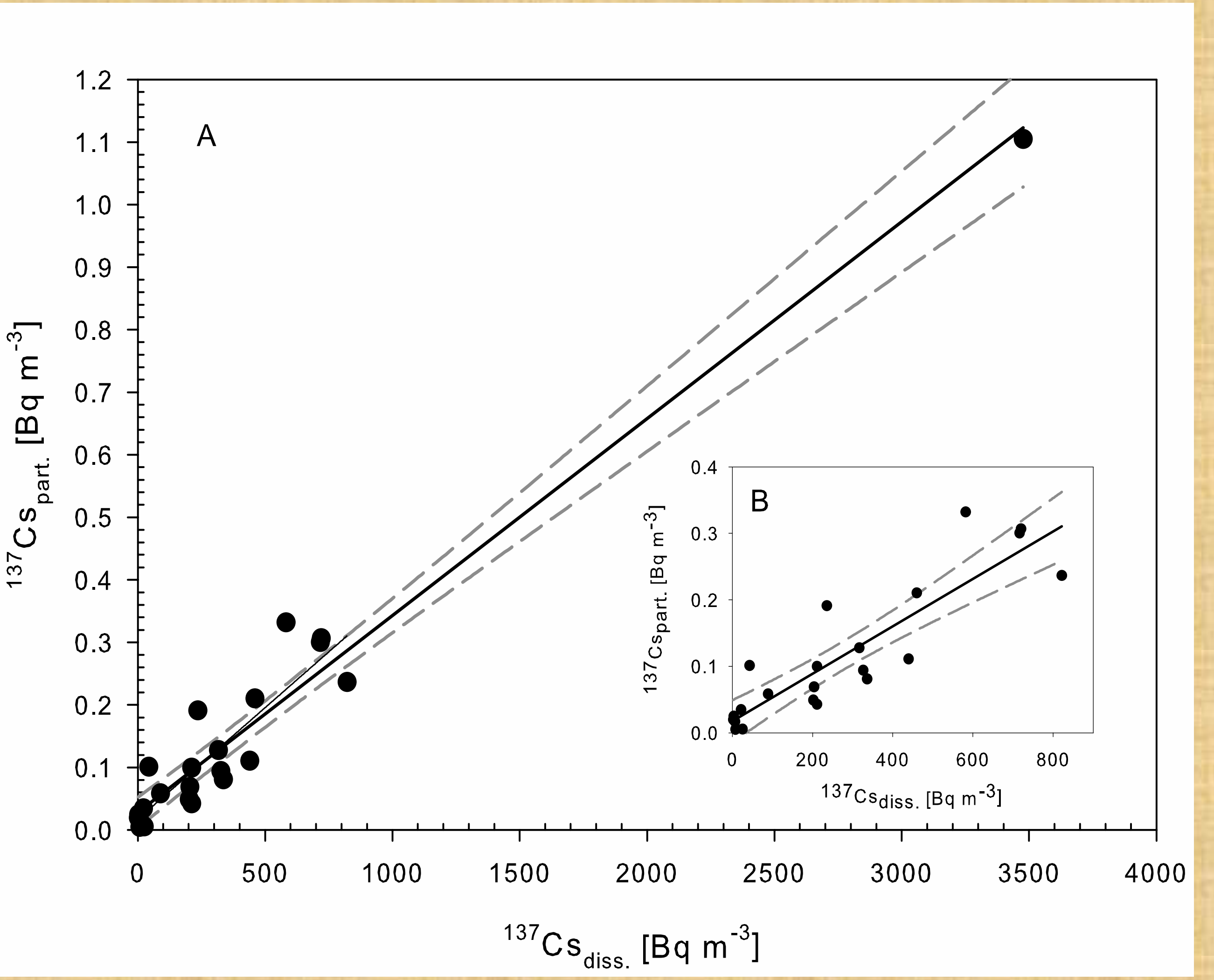
shows low uptake in marine phytoplankton (Kds < 100) but much higher in freshwater where there are orders of magnitude lower K, Na concentrations (Kds ~10,000)

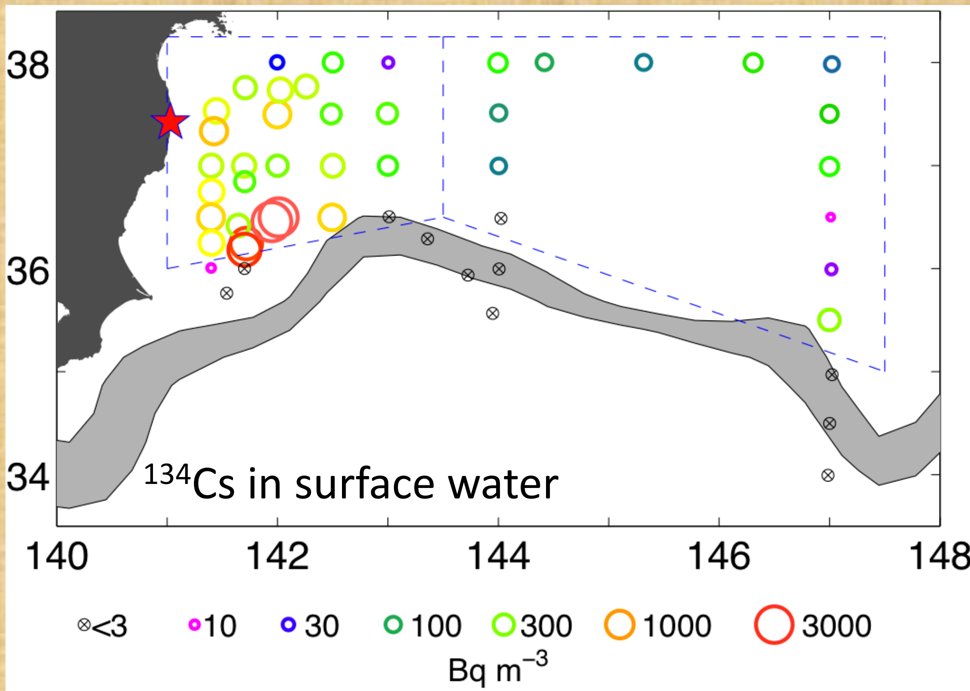
modest biomagnification in marine food chains (much less than methylmercury)—high assimilation efficiencies (~70%) in fish and daily loss rates of ~2% from excretion; ~2/3 of Cs from diet

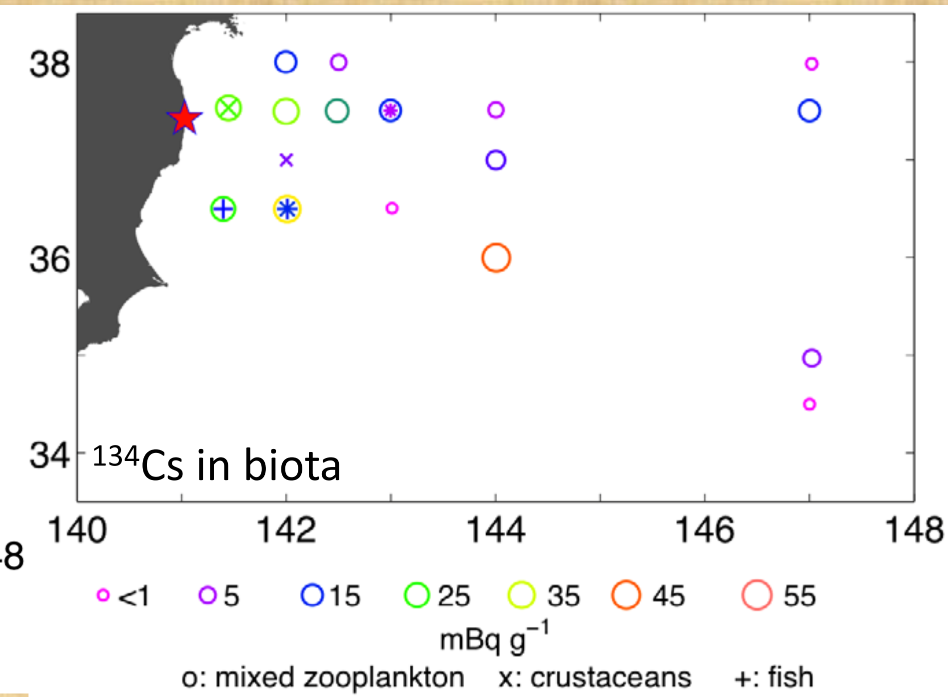
concentrates in muscle tissue (e.g., filet of fish)

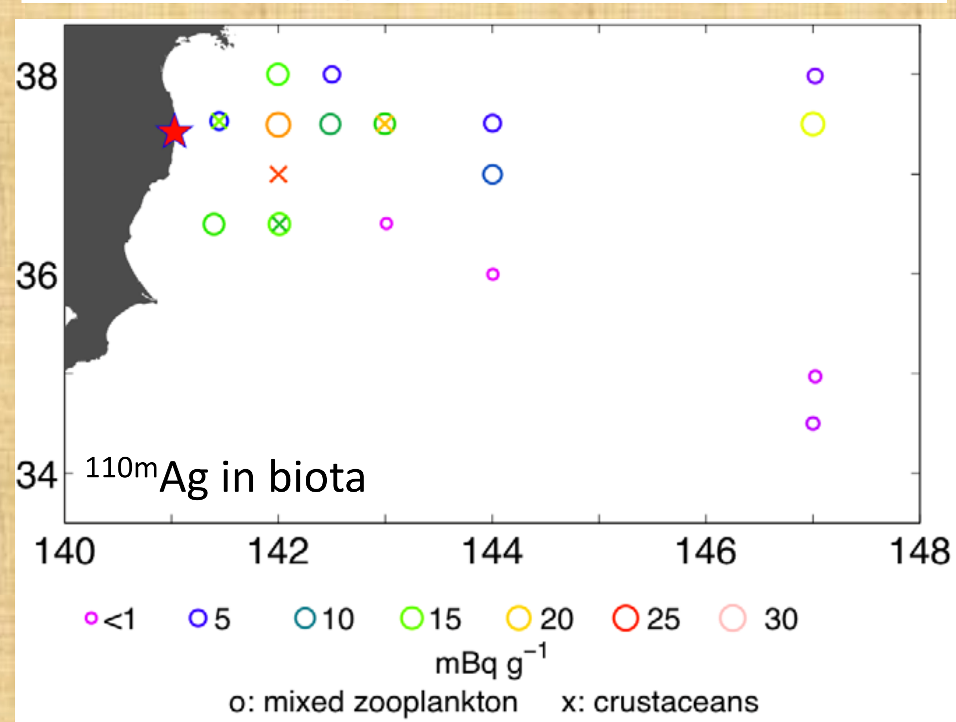

VFS = 256
keV

Particulate vs. dissolved ^{137}Cs in waters off Japan







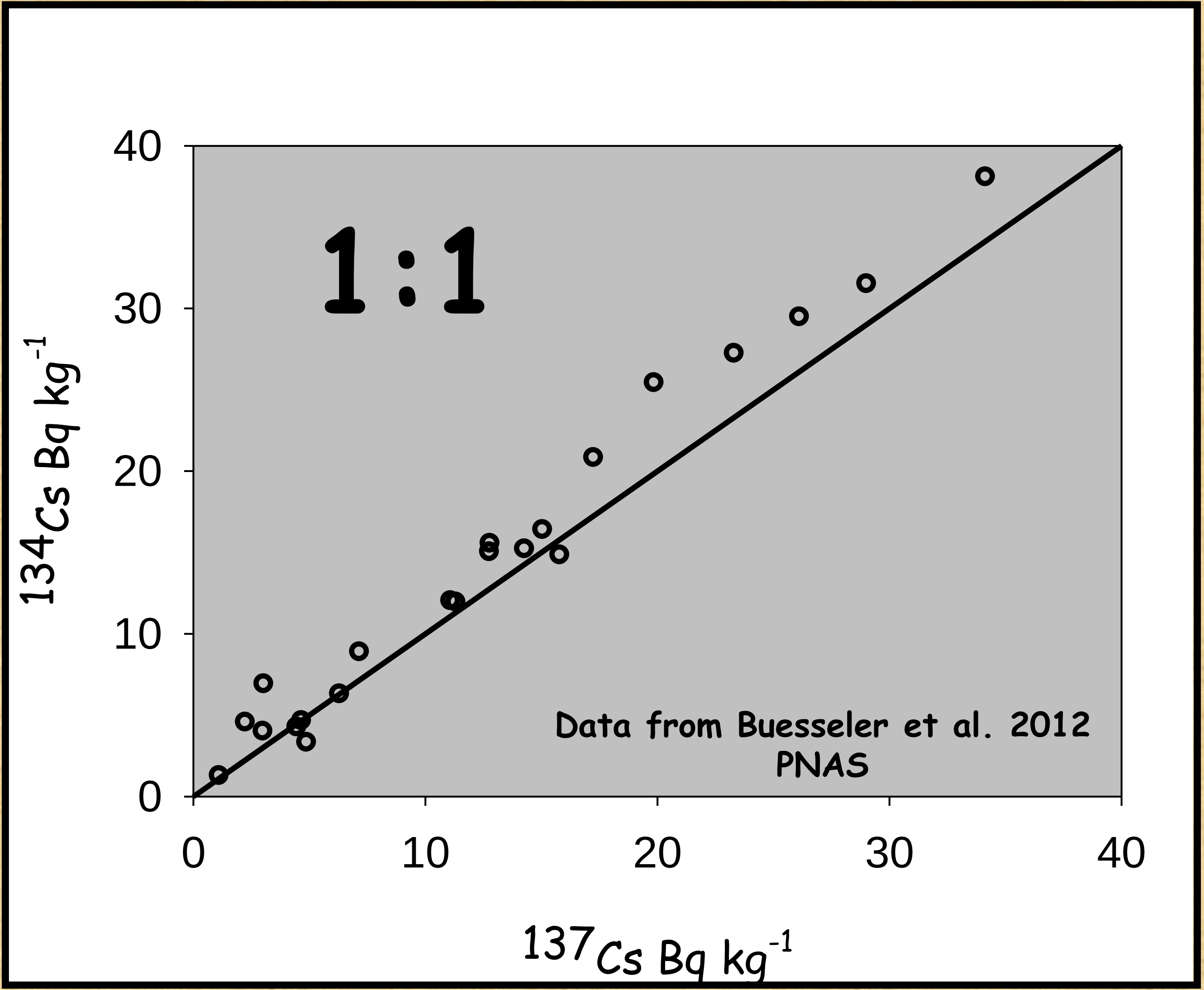


Buesseler et al. (PNAS 2012)

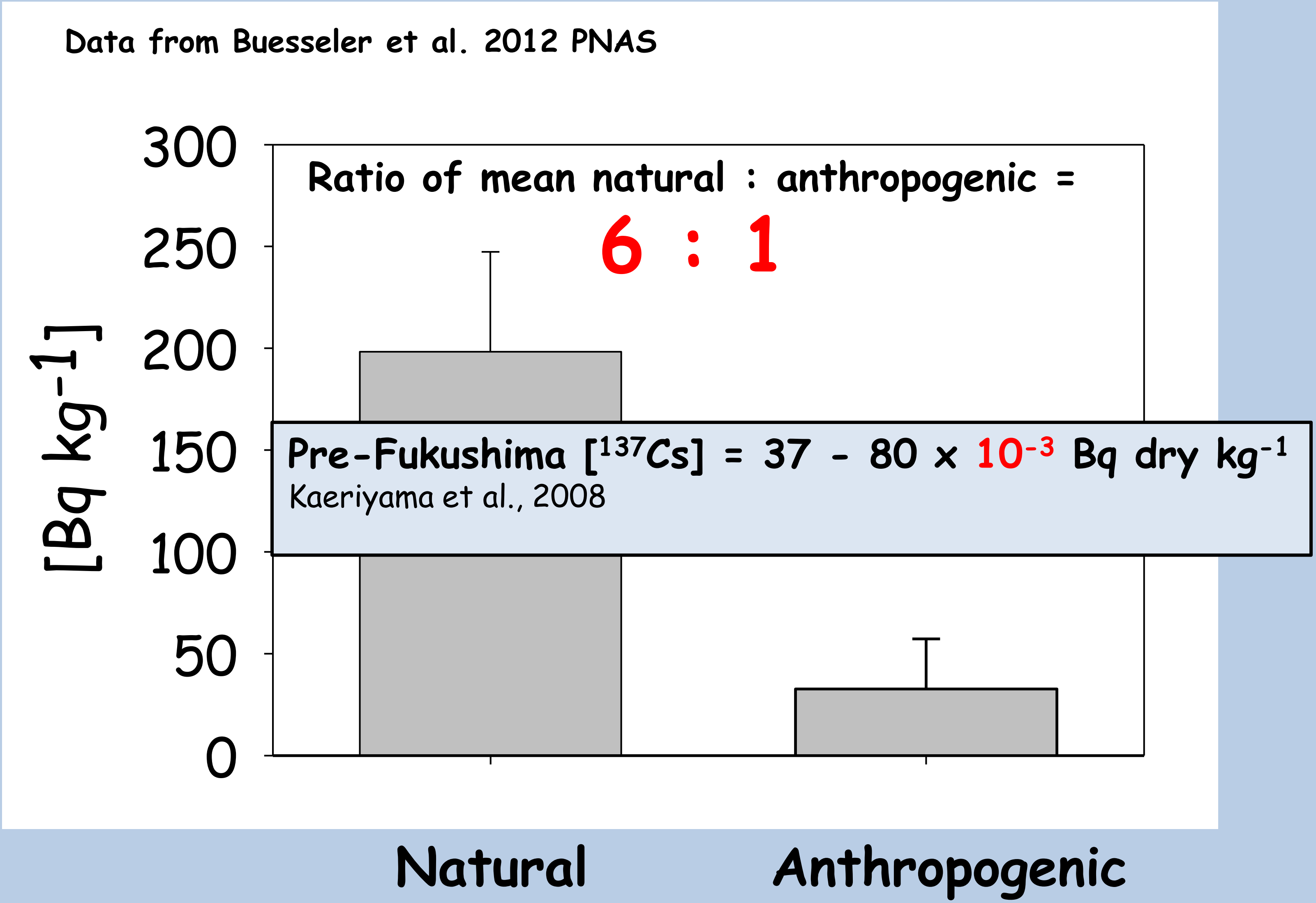
Mean concentrations (Bq kg^{-1} dry) in biota

	^{137}Cs	^{134}Cs	^{110m}Ag	^{40}K
Copepods n = 17	14.8 CF = 79	17.7 CF = 71	8.4	199
Large zooplankton (euphausiids, gelatinous) n = 4	11.1 CF = 16	13.7 CF = 22	17.9	217
Fish n = 3	10.7 CF = 16	10.0 CF = 13	bd	168

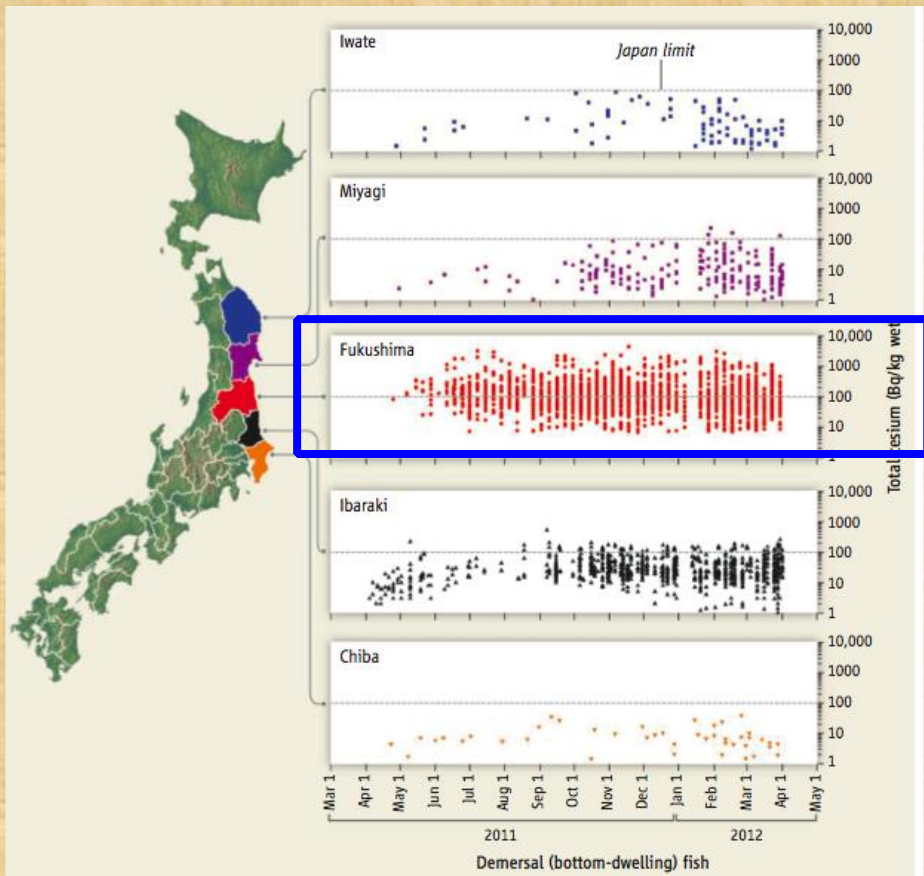
CESIUM ISOTOPIC RATIOS IN BIOTA ~1



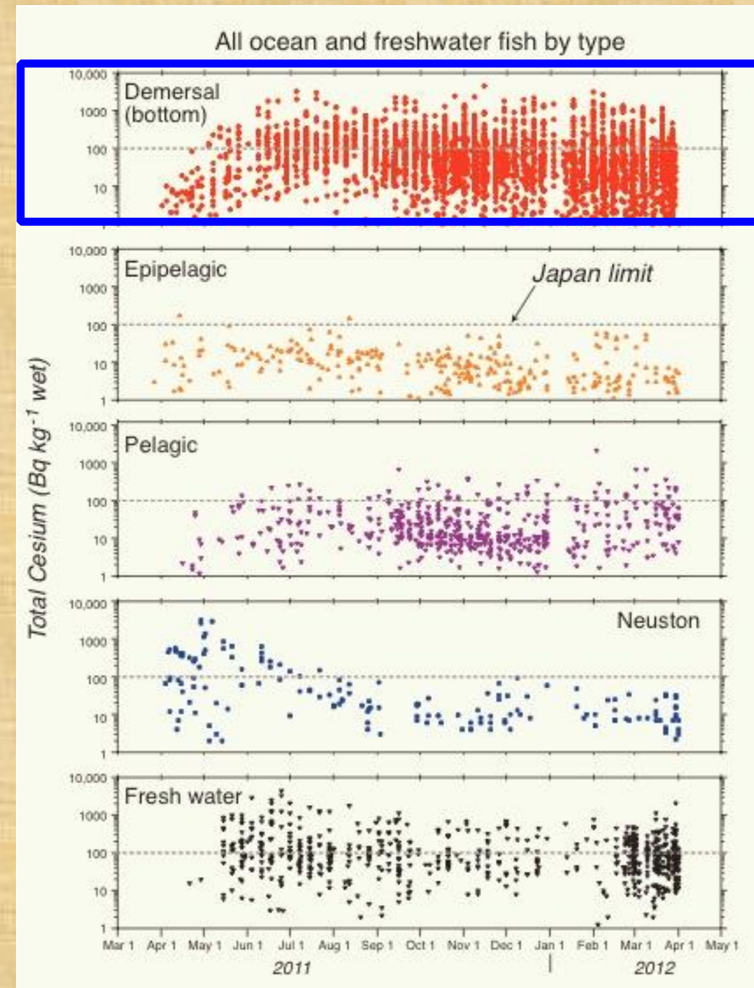
RANGES OF RADIOISOTOPES IN BIOTA



Elevated Cs concentration in demersal fish off Fukushima



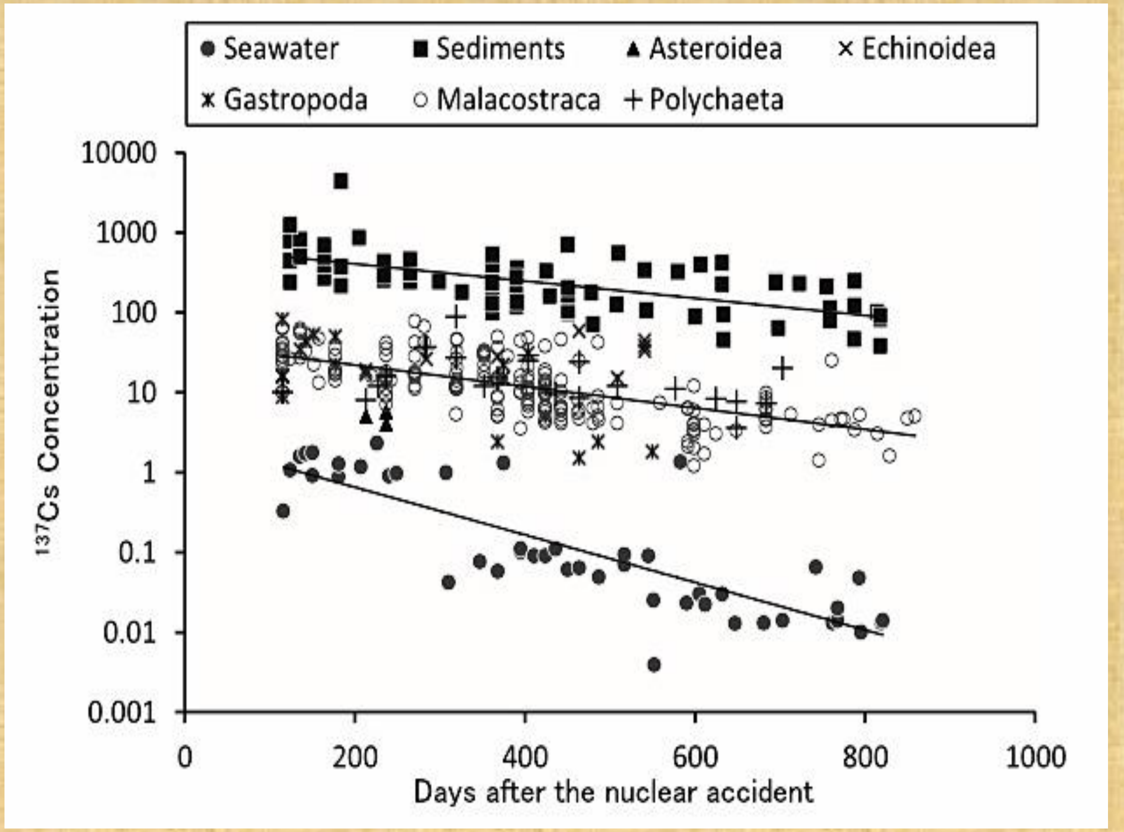
Changes in total cesium (^{137}Cs and ^{134}Cs in Bq/kg wet) over time in bottom fish in eastern Japan.

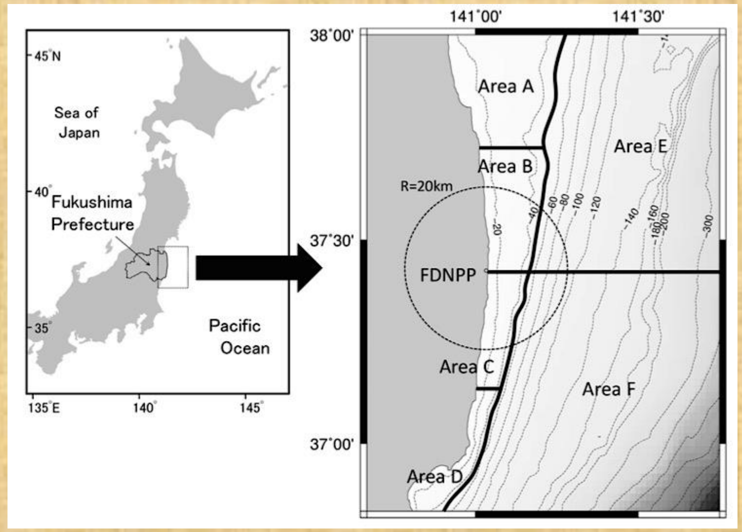


Total cesium in five different fish types as a function of time.

Buesseler, K.O. (2012) Fishing for Answers off Fukushima. ***Science***, 338(6106), 480-482.

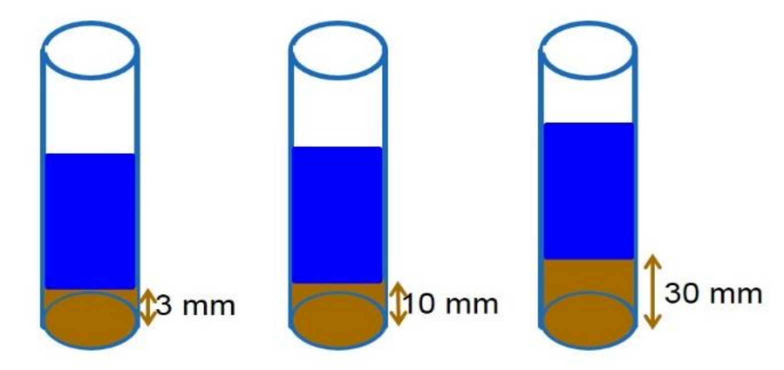
Constantly elevated Cs concentration in benthic invertebrates and sediment

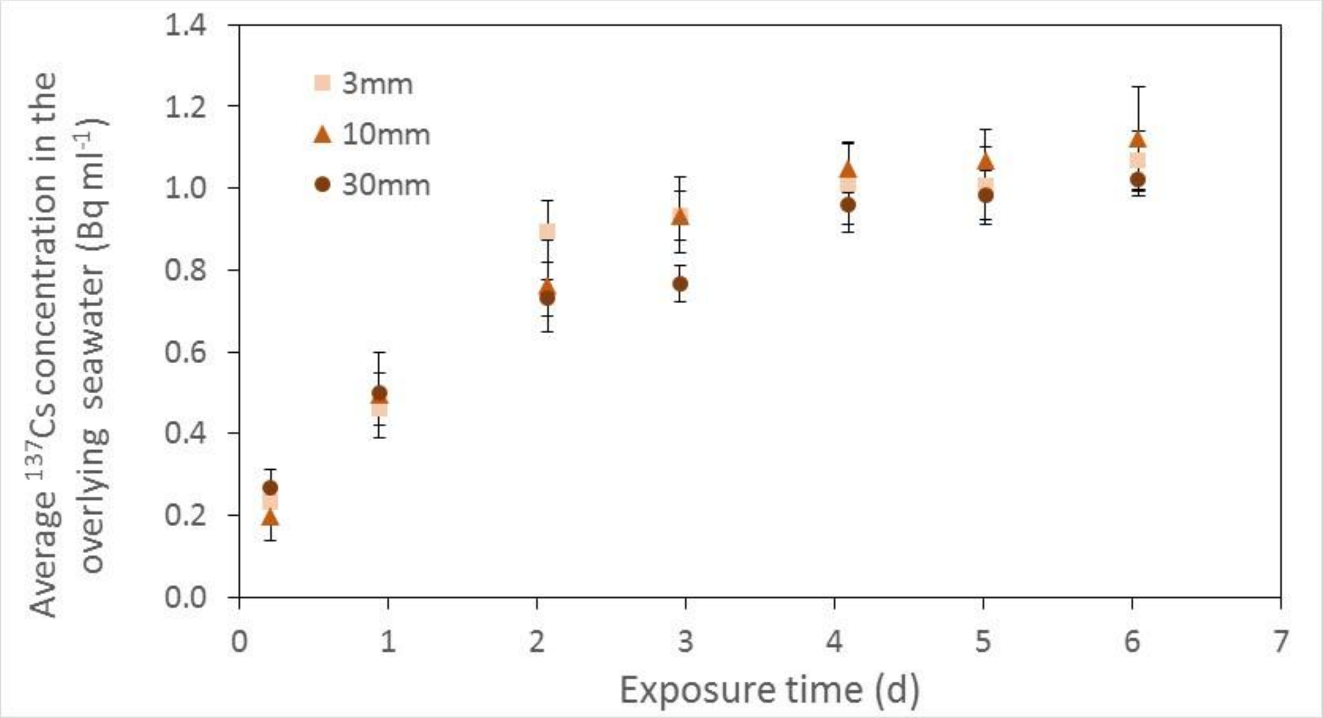




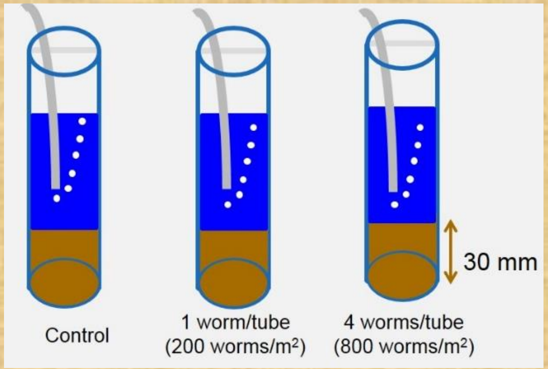
Sohtome, T., et al. (2014) Radiological impact of tepco's fukushima dai-ichi nuclear power plant accident on invertebrates in the coastal benthic food web. ***Journal of Environmental Radioactivity*** 138, 106-115.

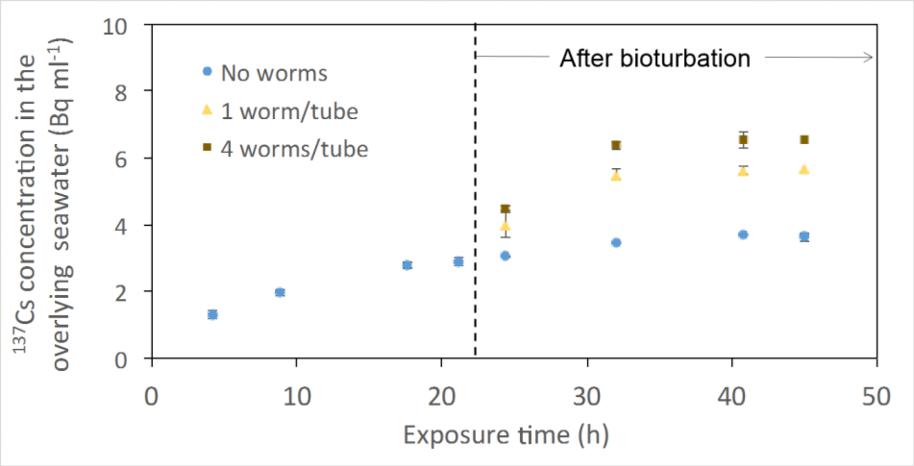
Release of ^{137}Cs from Japanese sediment to overlying seawater

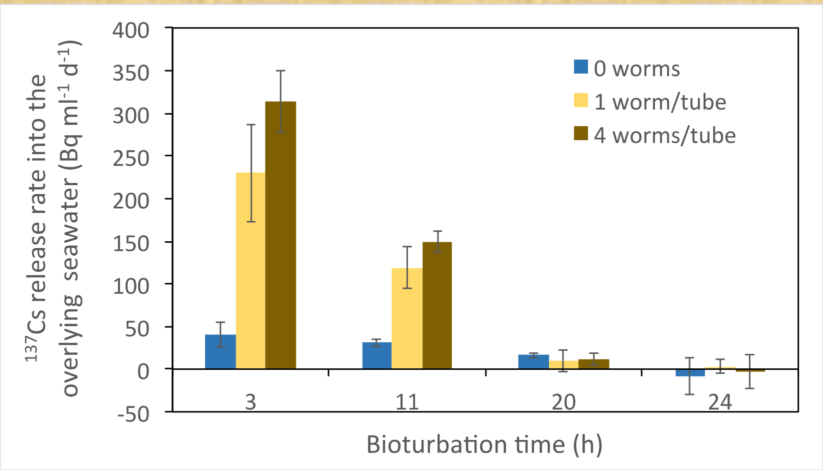




The effect of bioturbation on Cs release from Japanese sediment to overlying seawater







Comparison of dietary uptake parameters

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

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



Organism	**Uptake** rate constant k_u (mL g^{-1} d^{-1})	Efflux rate constant k_{ew} (d^{-1})
Polychaete *Nereis succinea*	10.3 ± 1.7	0.2 ± 0.01
Asian shore crab *Hemigrapsus sanguineus*	1.5 ± 0.1	0.1 ± 0.01
Killifish *Fundulus heteroclitus*	0.6 ± 0.1	0.06 ± 0.008

- 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

Polychaetes eating sediment	Crabs eating worms	Fish eating worms
98%	98%	99%

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.**

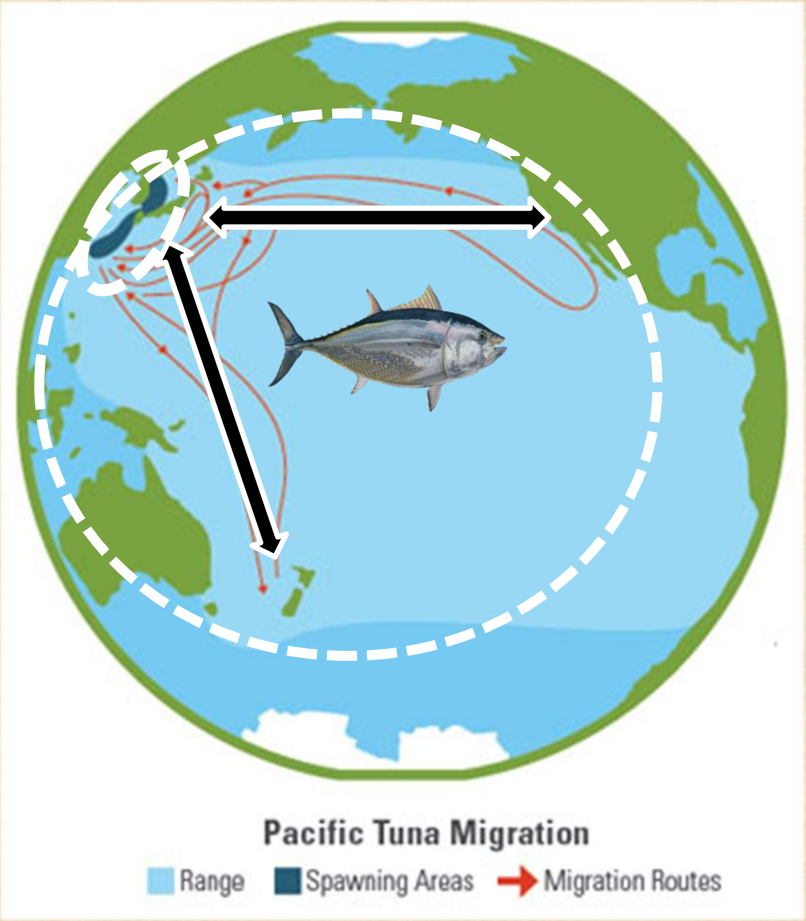

祝
初荷
東都水産

Pacific bluefin tuna

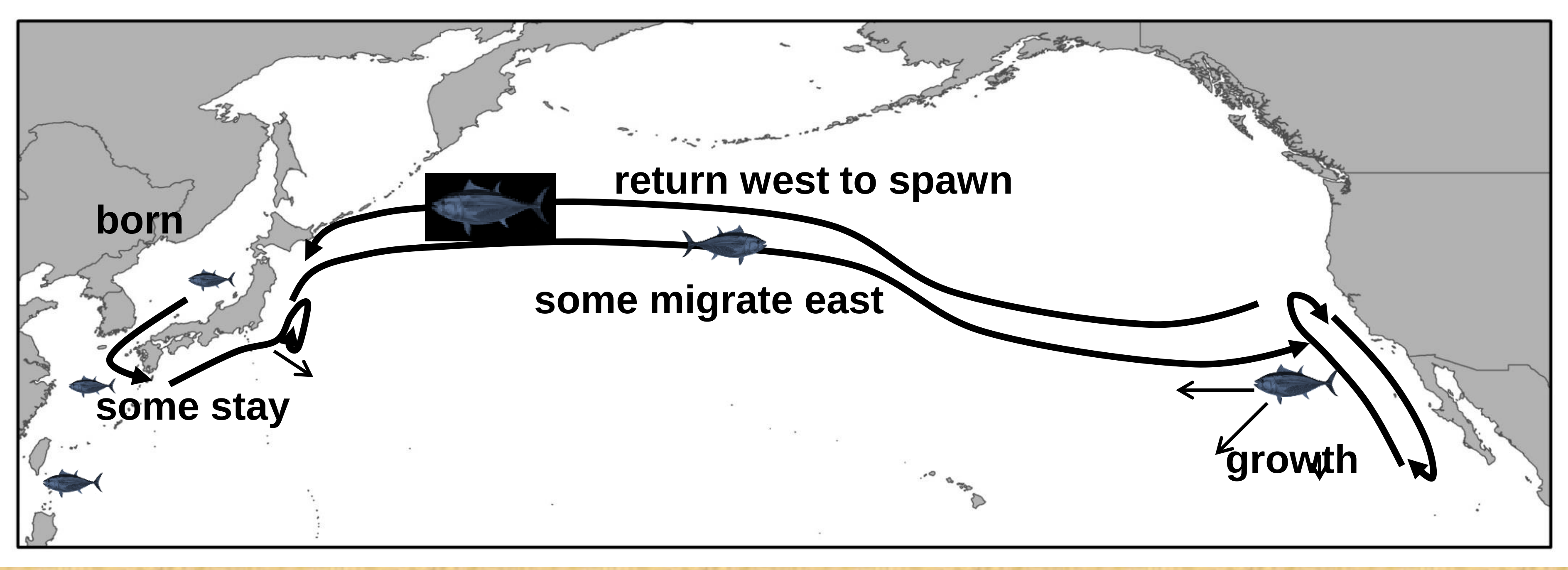
Extremely large home range

Trans-oceanic migrations

Discrete spawning area



Pacific bluefin life history

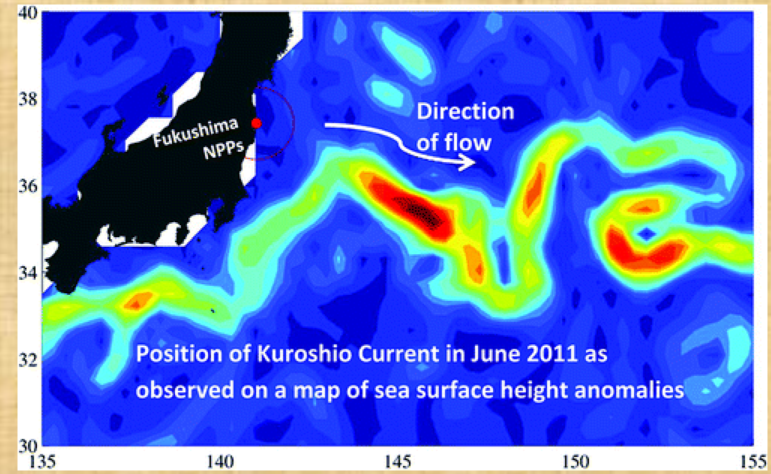


Fukushima accident: 2011

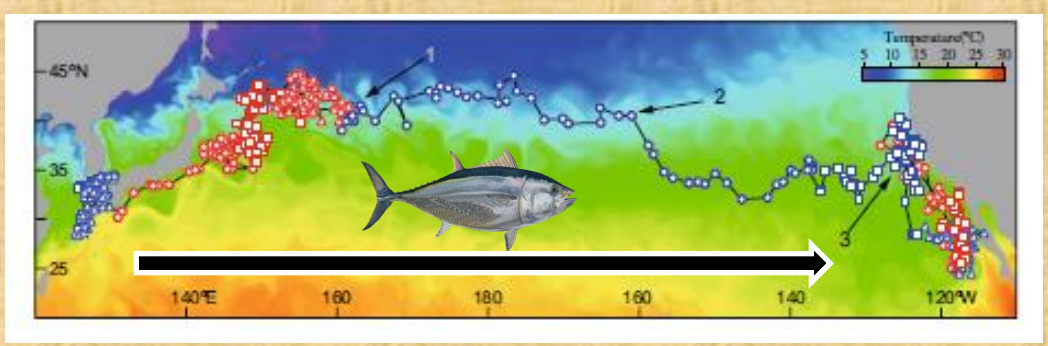
Massive release of radiocesium into the ocean

Did bluefin carry it over?

We measured 15 fish caught off San Diego……..

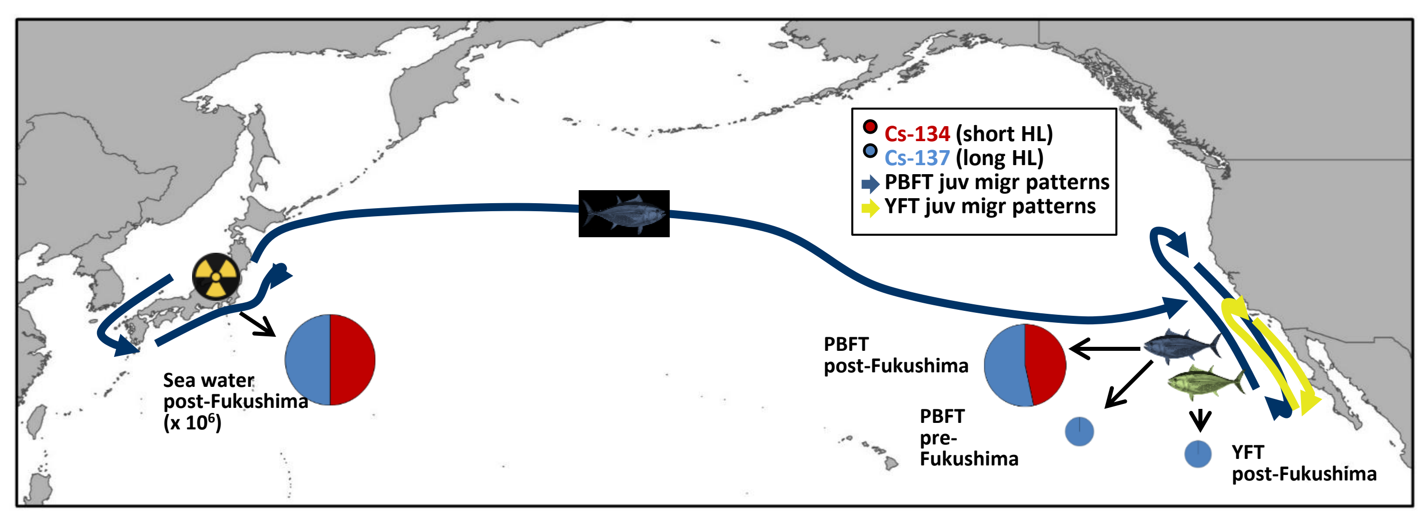


Buesseler at al 2011, ES&T



Kitagawa et al 2009, Env Biol Fish

…and all 15 did.



15 Post-Fukushima bluefin: all measurable ^{134}Cs and elevated ^{137}Cs

5 Pre-Fukushima (2008) bluefin: background ^{137}Cs

5 Post Fukushima yellowfin in eastern Pacific: background ^{137}Cs

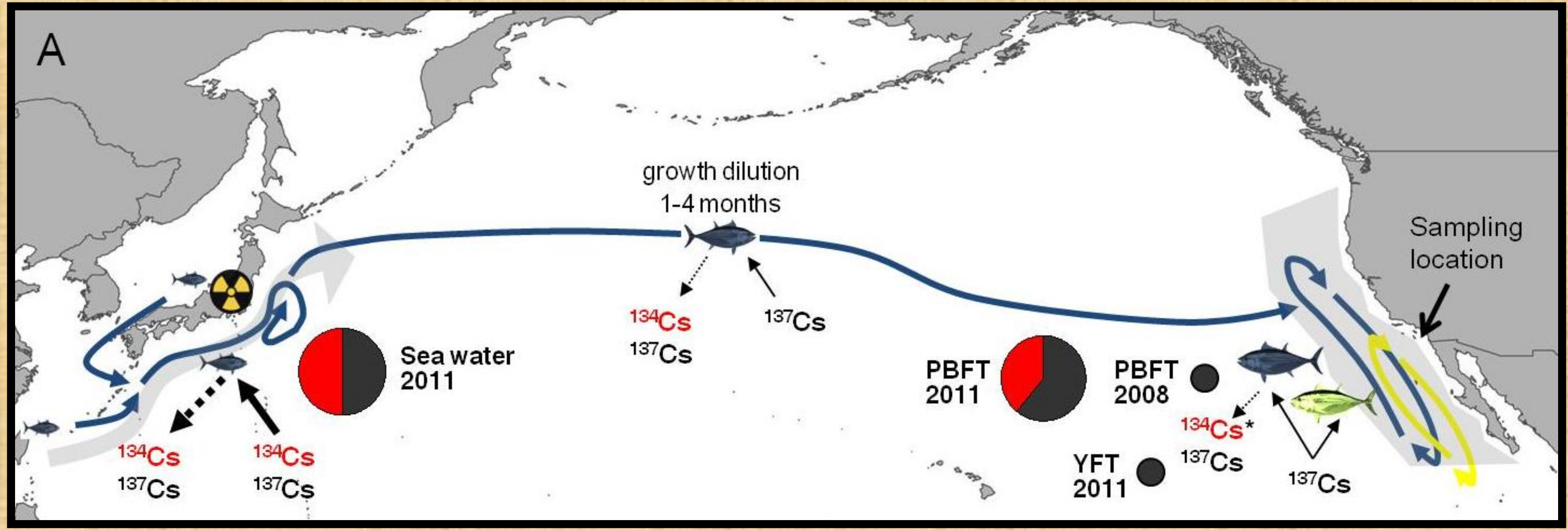
Madigan et al 2012, PNAS

Measured ^{134}Cs, ^{137}Cs, and the natural ^{40}K for post-Fukushima bluefin (PBFT 2011), pre-Fukushima bluefin (PBFT 2008), and post-Fukushima yellowfin tuna (YFT 2011)

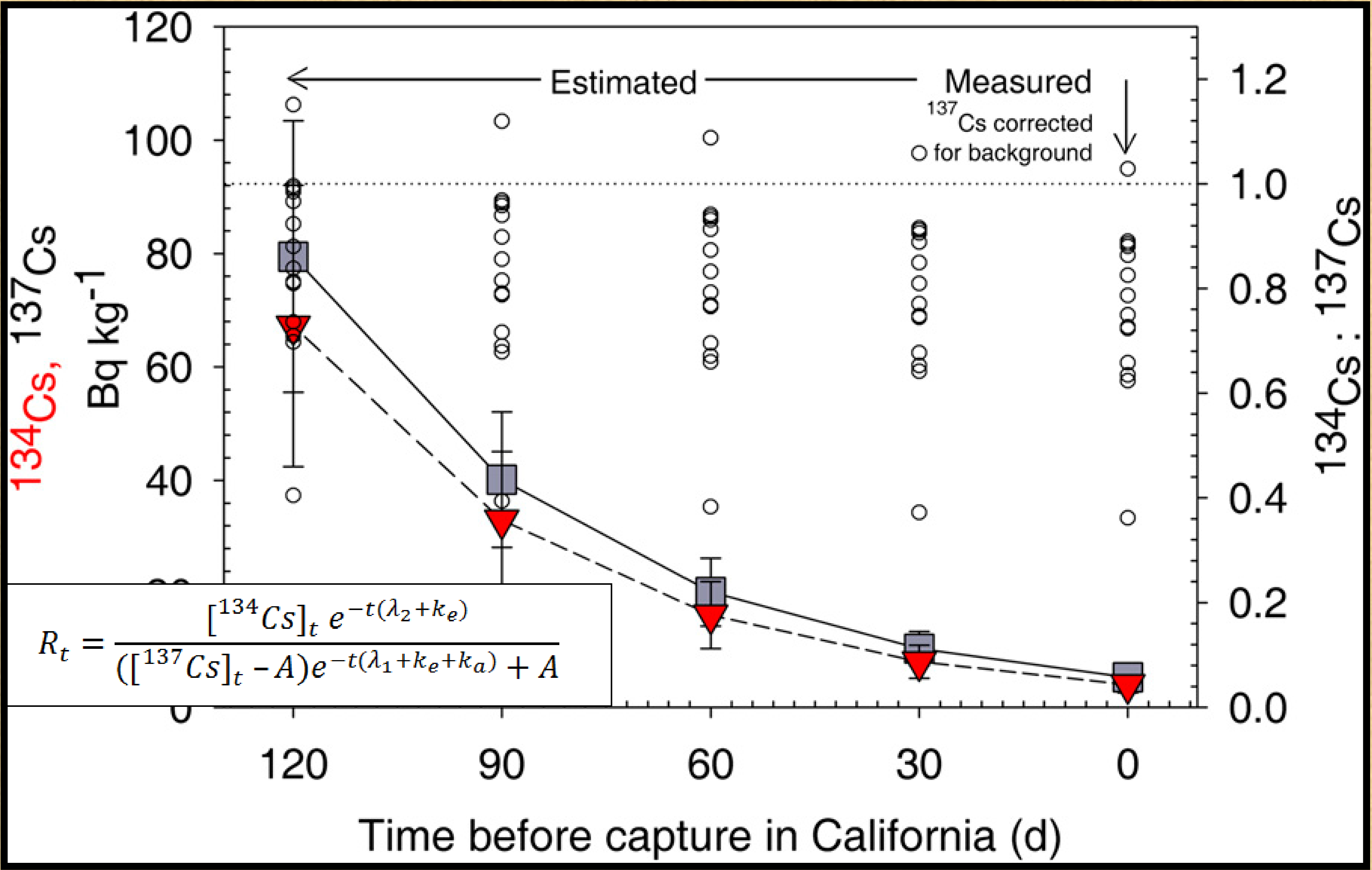
		SL	Body mass	Age	^{134}Cs	^{137}Cs	^{40}K	^{134}Cs:^{137}Cs	$^{134+137}Cs$
					Radionuclide concentrations Bq kg^{-1} dry mass				
		cm	kg dry	years		Bq kg^{-1}			
PBFT 2011 n=15	Median	66.5	1.5	1.5	4.3	6.0	367	0.66	10.3
	Mean	66.2	1.5	1.5	4.0	6.3	347	0.62	10.3
	SD	3.6	0.2	0.1	1.4	1.5	49	0.14	2.9
PBFT 2008 n=5	Median	66.3	1.5	1.4	0	1.4	266	0	1.4
	Mean	66.2	1.5	1.4	0	1.4	258	0	1.4
	SD	1.2	0.09	0.05	0	0.2	43	0	0.6
YFT 2011 n=5	Median	72.3	1.9	1.2	0	1.2	342	0	1.2
	Mean	72.3	1.9	1.2	0	1.1	333	0	1.1
	SD	2.5	0.2	0.01	0	0.4	78	0	0.3

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



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

^{134}Cs:^{137}Cs in myctophid fish off Japan = 0.9; in plankton = 1.1; in water = 1.0

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

U.S.

THE WALL STREET JOURNAL. | HEALTH

U.S. Tuna Has Fukushima Taint

Forbes

Fukushima Radiation May Actually Save Bluefin Tuna

Los Angeles Times

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

npr

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

Washington DC

THE HILL

Democrat worried about tainted seafood from Japan's nuclear meltdown

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
A notícia em primeira mão.

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

EM 03 DE JUNHO DE 2012 ÀS 13H08

East Asia

The Asian Tiger

Consequences Fukushima nuclear disaster persist

Argentina

Bariloche2000
DIARIO DIGITAL INTERACTIVO

Fukushima y la vida marina

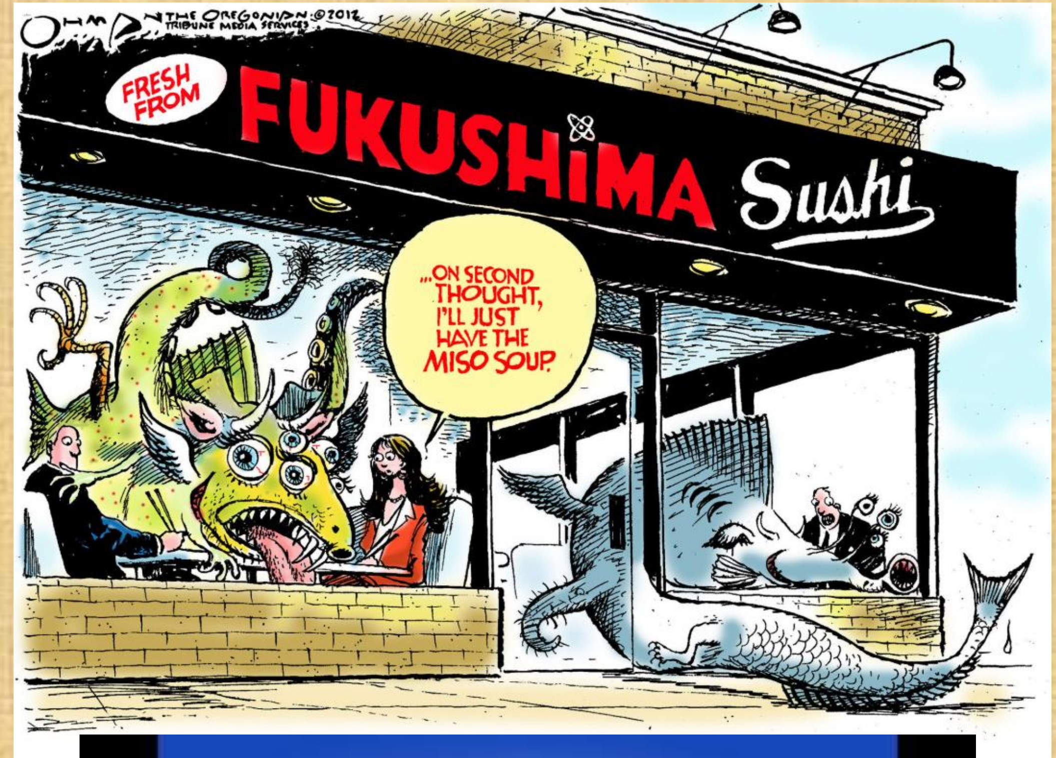
DOMINGO 03 DE JUNIO DE 2012 12:32 - 2504 LECTURAS.

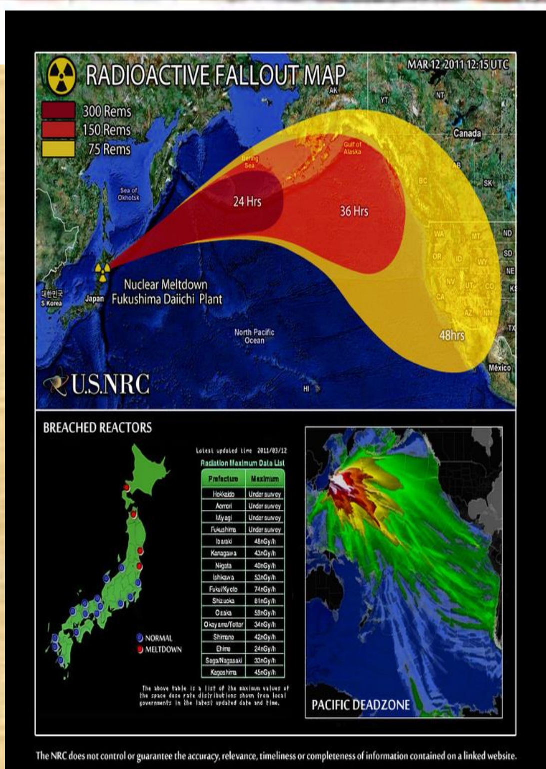
Estonia

EKSPRESS.EE

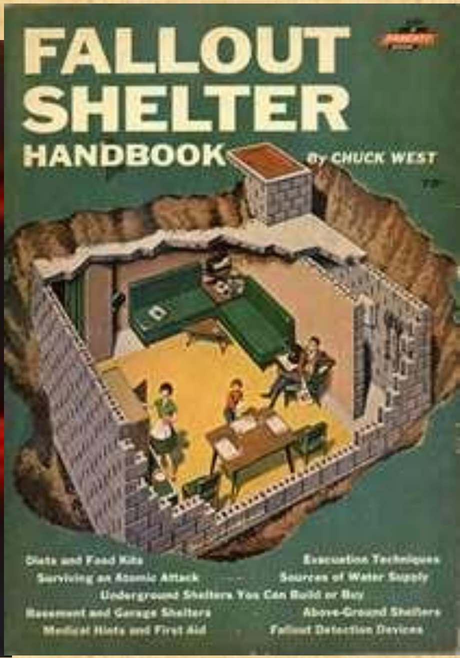
Pärast põgusat kohtumist **Jaapaniga** võib pahatihti jääda mulje, nagu oleks tegu ühiskonnaga, kus kõik alluvad vaikides grupile ning iseseisvat indiviidi ei eksisteerigi.


HOW SCARY IS THAT RADIOACTIVITY FROM JAPAN?!!


OHMAN THE OREGONIAN ©2012 TRIBUNE MEDIA SERVICES
FRESH FROM
FUKUSHIMA Sushi
...ON SECOND THOUGHT, I'LL JUST HAVE THE MISO SOUP.

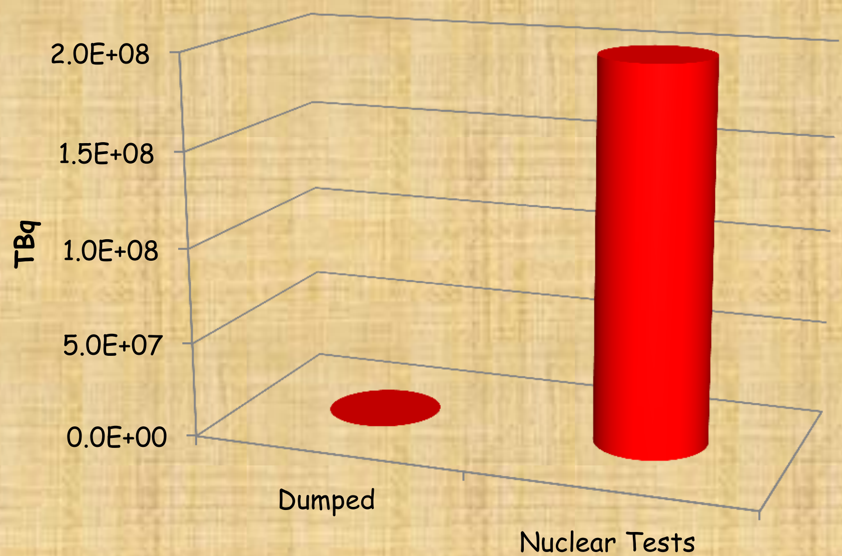

RADIOACTIVE FALLOUT MAP
MAR 12 2011 12:15 UTC
300 Rems
150 Rems
75 Rems
24 Hrs
36 Hrs
48hrs
Canada
Nuclear Meltdown Fukushima Daiichi Plant
Japan
North Pacific Ocean
U.S.NRC
BREACHED REACTORS
NORMAL
MELTDOWN
PACIFIC DEADZONE
The NRC does not control or guarantee the accuracy, relevance, timeliness or completeness of information contained on a linked website.


IMPORTED FROM JAPAN
TSUNAMI BRAND
RADIOACTIVE
BLUE FIN TUNA
TSUNAMI BRAND RADIOACTIVE TUNA
EXTRA-LONG SHELF-LIFE
RICH IN CESIUM 134 AND CESIUM 137
NO NEED TO RE-HEAT - IT'S ALREADY WARM
GREAT FOR MIDNIGHT SNACKS
IT GLOWS IN THE DARK
©RLMMMXII
SANTA MARIA SUN


FALLOUT SHELTER
HANDBOOK
By CHUCK WEST
Diets and Food Kits
Evacuation Techniques
Surviving an Atomic Attack
Sources of Water Supply
Underground Shelters You Can Build or Buy
Basement and Garage Shelters
Above-Ground Shelters
Medical Hints and First Aid
Fallout Detection Devices

Historical Anthropogenic Radionuclide Input to the Oceans

- Total radioactivity, dumped into oceans: 6 x 10^4 TBq
- Total radioactivity, atmospheric nuclear testing: 2 x 10^8 TBq
- Total radioactivity, naturally in the ocean: 2 x 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})

	Tuna: from $^{134+137}Cs$	Tuna: from ^{40}K	Banana: from ^{40}K	Dental x-ray	1 flight NY to LA (cosmic rays)
Consumption of:	200 g	200 g	1 banana		
Dose (μSv)	0.008	0.1	0.1	5	40
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}.

Radionuclide	PBFT source	μSv annual consumption
^{134}Cs	USA 8/2011	0.4
^{137}Cs	USA 8/2011	0.5
^{40}K	USA 8/2011	12.7
^{210}Po	USA 8/2011	558
^{134}Cs	Japan 4/2011	15.7
^{137}Cs	Japan 4/2011	16.9
^{40}K	Japan 4/2011	29.7
^{210}Po	Japan 4/2011	1310

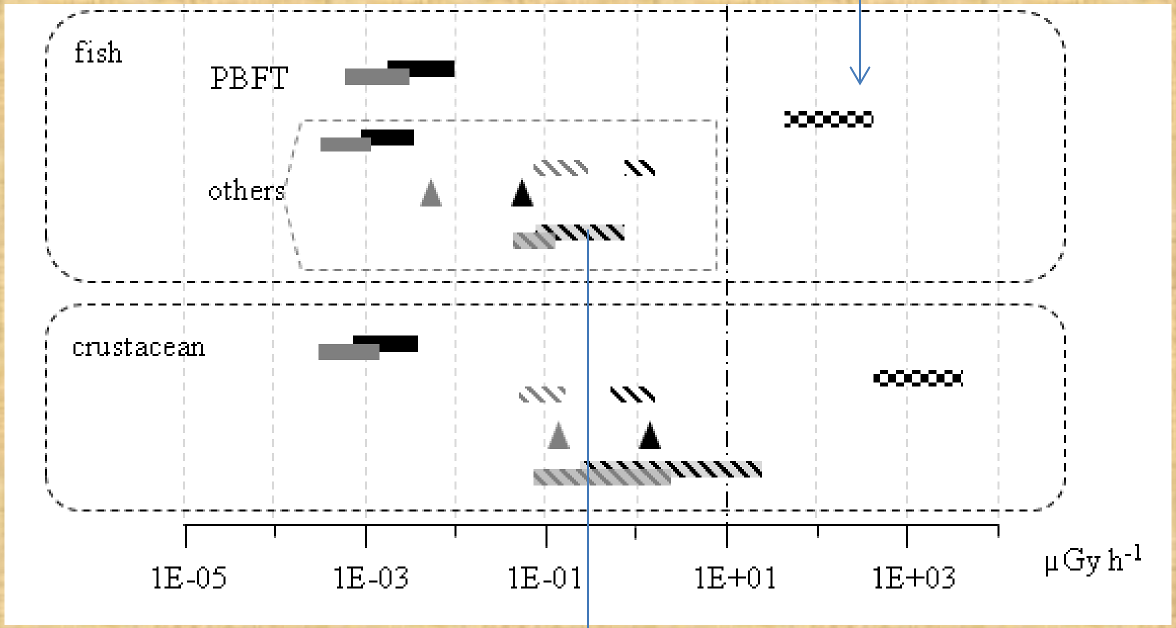
Internal absorbed dose rates to zooplankton and bluefin tuna (nGy h^{-1}); ICRP benchmark = 10,000 nGy h^{-1})

	^{110m}Ag	^{134}Cs	^{137}Cs	^{210}Po	^{40}K	Natural : anthropogenic
Zooplankton (near Japan)	0.76	1.5	1.8	1700	0.1	**420**
Tuna (in California)	n.d.	0.6	1.1	600	1.3	**354**
Tuna (near Japan)	n.d.	9	16.5	600	1.3	**24**

(1 Gy = absorption of 1 joule kg^{-1} of tissue from ionizing radiation)
nd: not detected (Fisher et al., PNAS, 2013)

Radiocesium doses

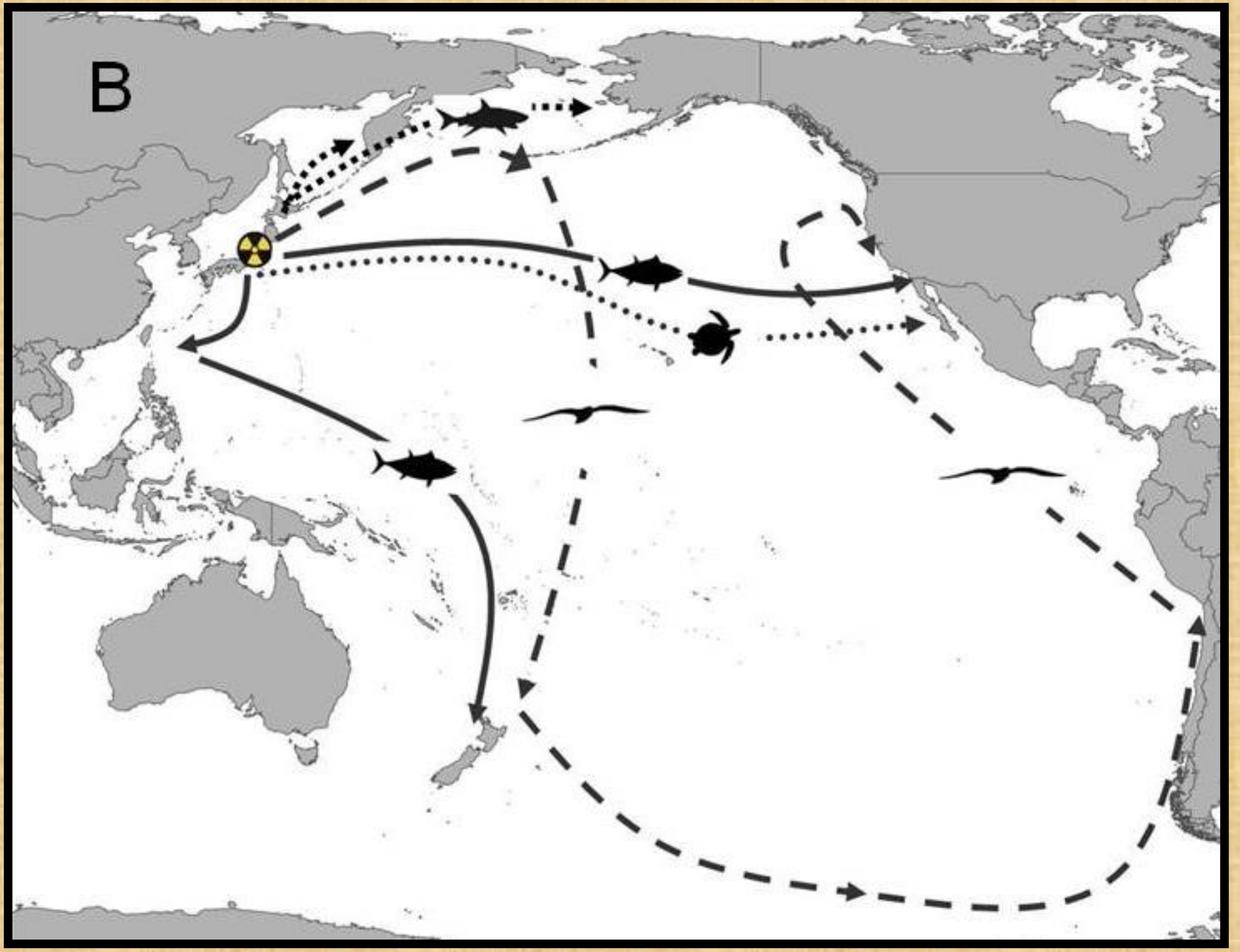
Benchmark safety levels for wildlife



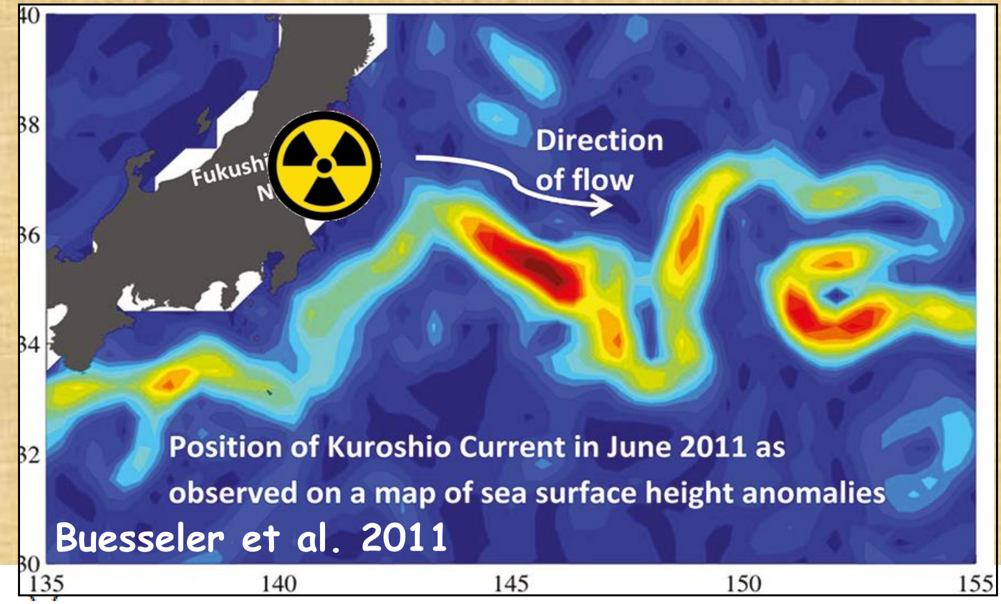
^{210}Po dose

(Fisher et al., PNAS, 2013)

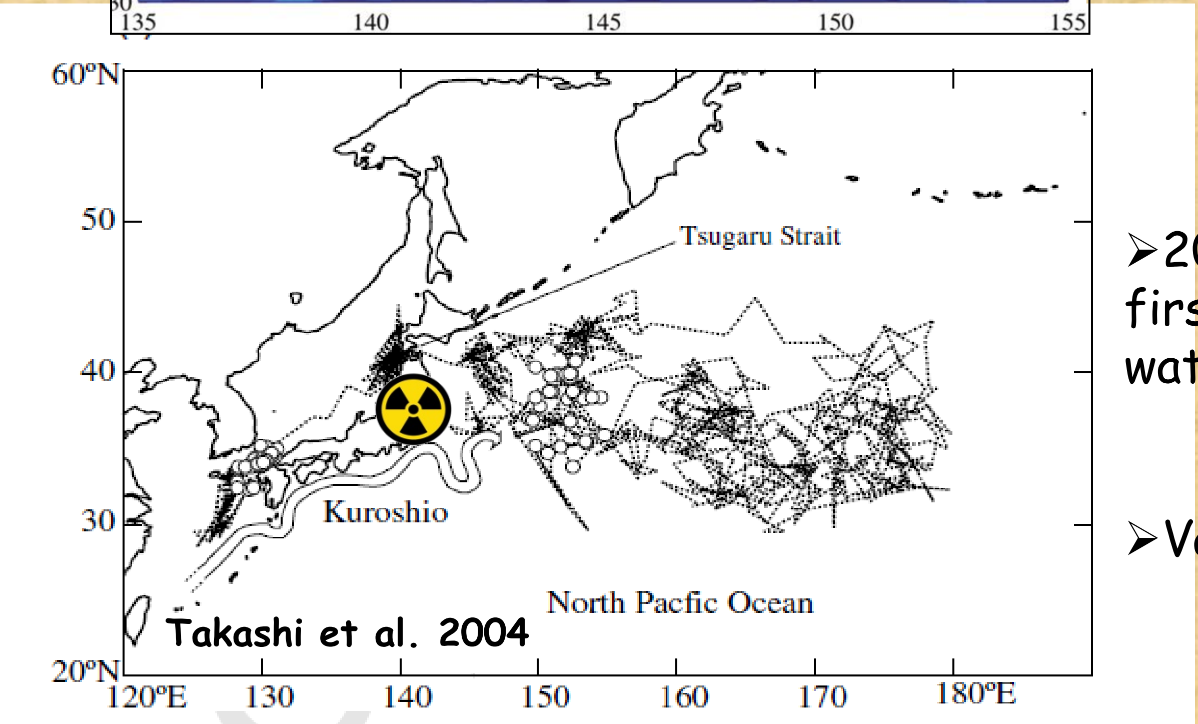
Simplified migration patterns of some highly migratory species in the Pacific



1. What is the Cesium load of PBFT in 2012 and 2013?



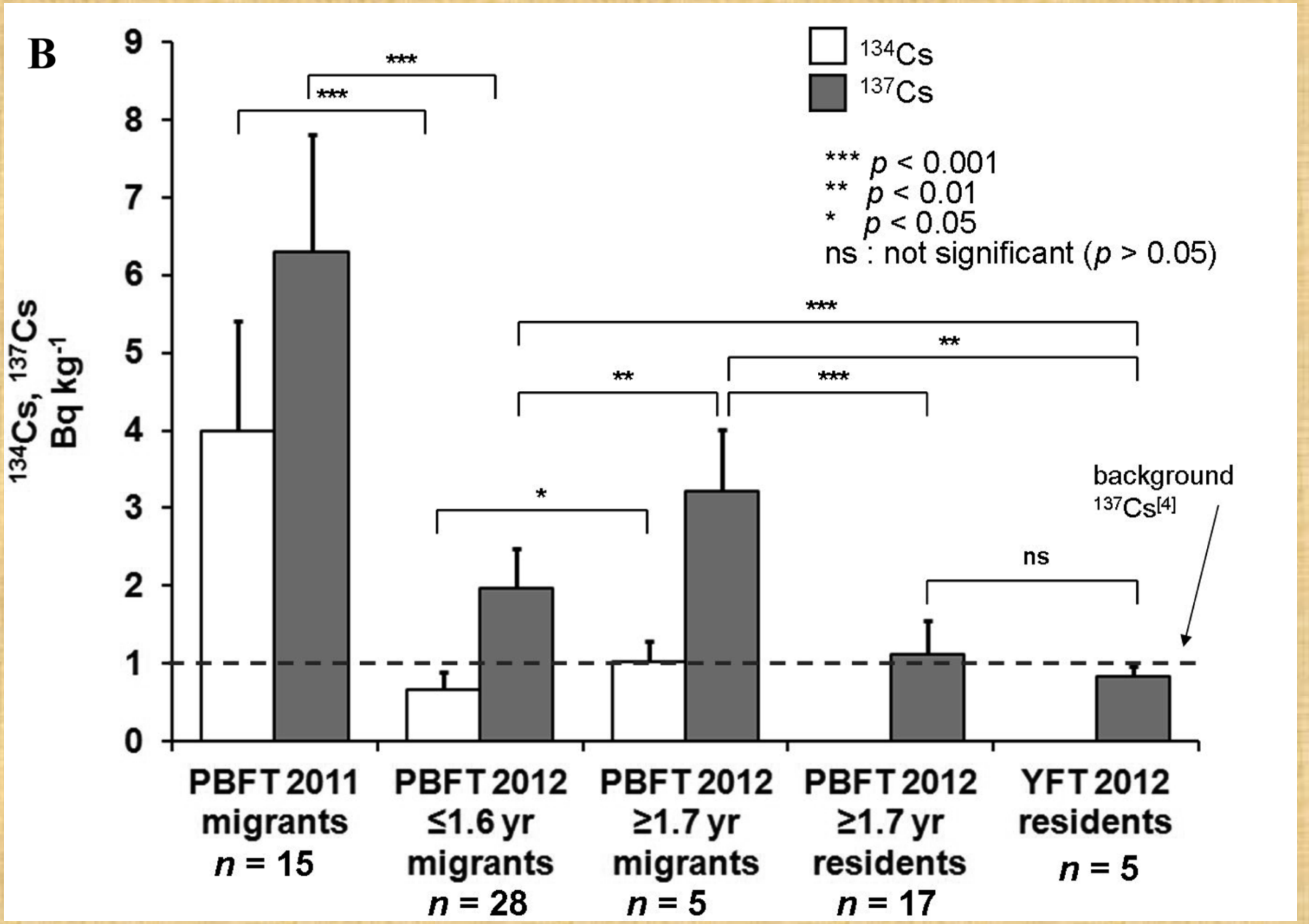
- Our 2011 data are from fish that spent ~1 month in contaminated waters



- 2012's cohort spent their first year in contaminated waters

- Values may be much higher

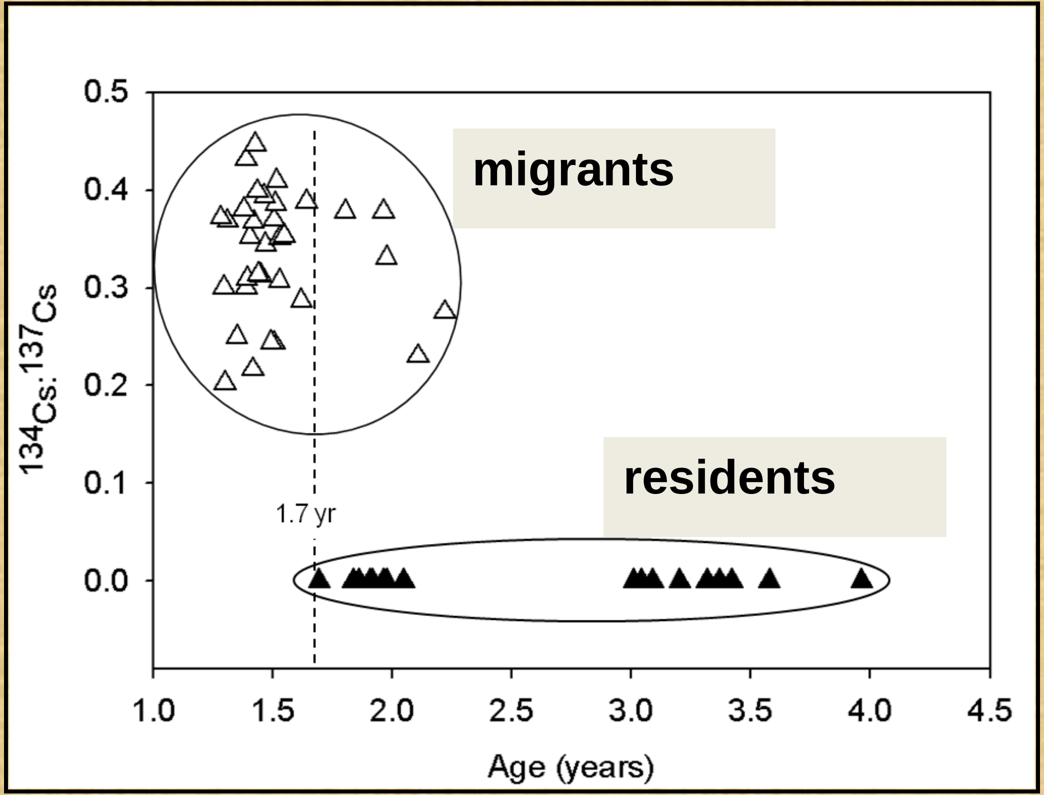
Comparison of 2011 and 2012 tuna



Madigan et al., Environ. Sci. Technol., 2013

Radiocesium in bluefin in 2012 validates new tracer technique

Remember smallest bluefin must have migrated recently….

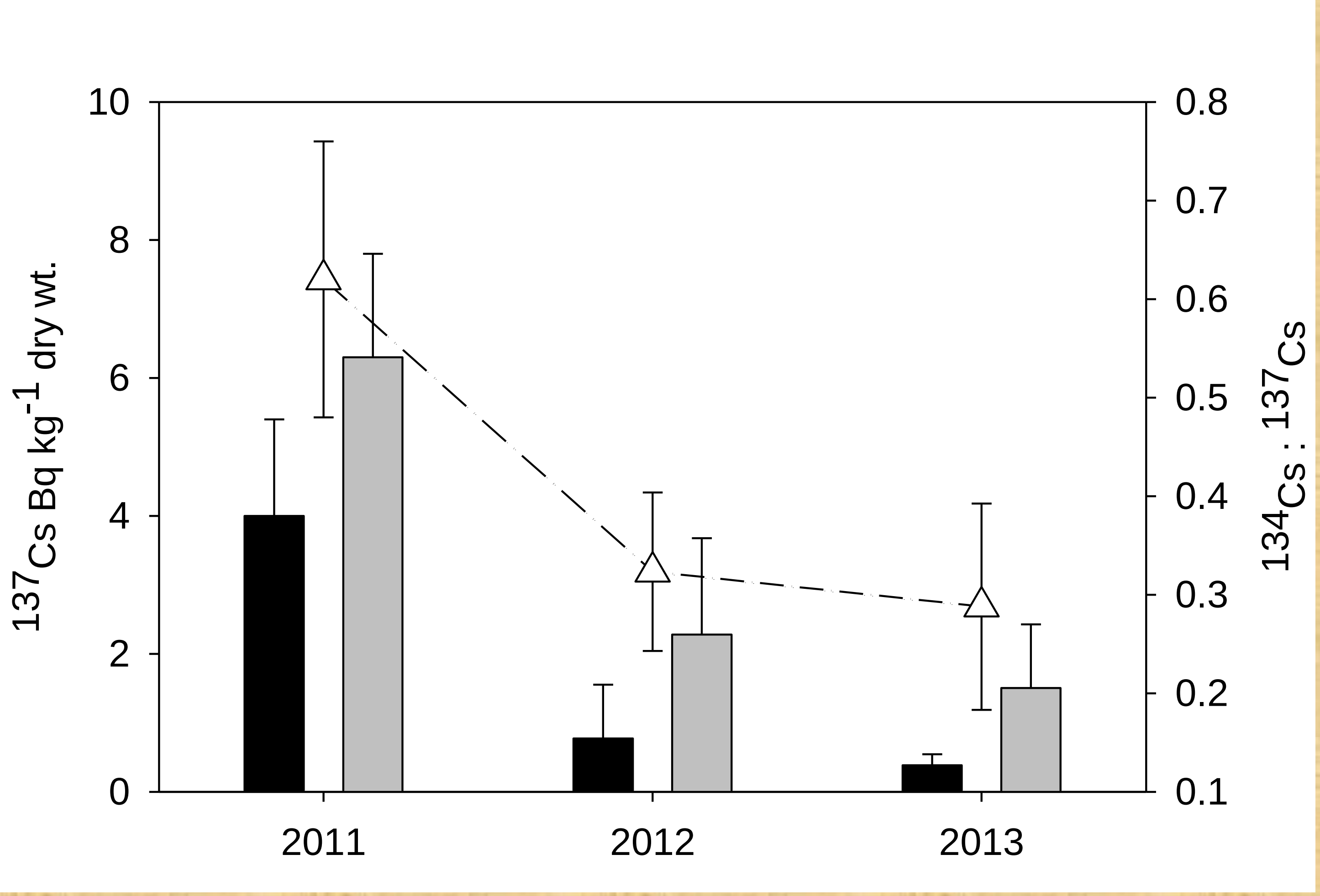


Every PBFT less than 1.7 years had signal

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

Madigan et al 2013, ES&T

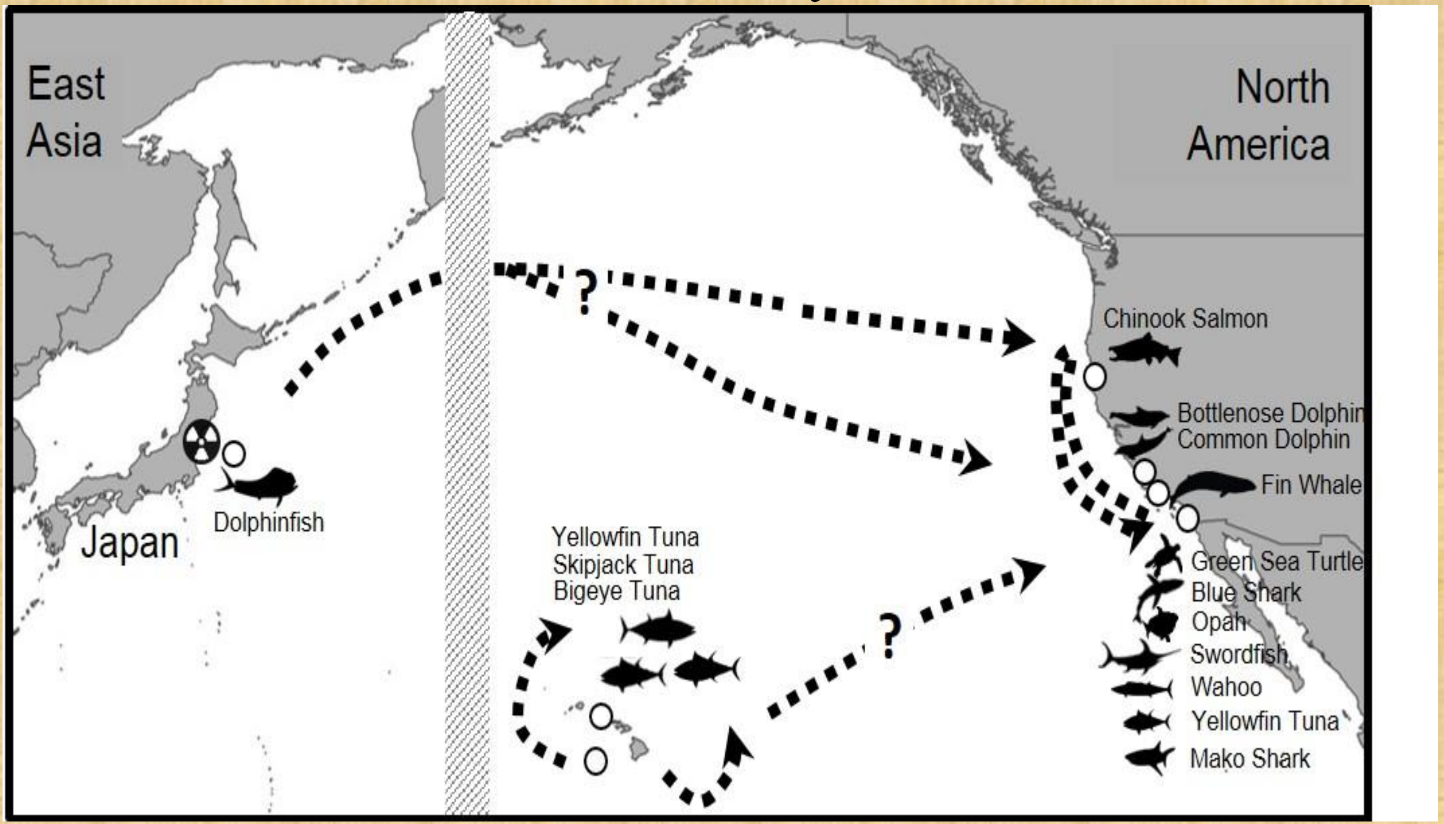
Post-Fukushima changes in cesium activity in Pacific bluefin tuna that have crossed the Pacific to California



n=15 n=93 n=14

black bars: ^{134}Cs; gray bars: ^{137}Cs; triangles: ^{134}Cs:^{137}Cs

NOT BLUEFIN TUNA: All samples (n = 91) had undetectable ^{134}Cs (2012-2015)



Still a work in progress—stay tuned….

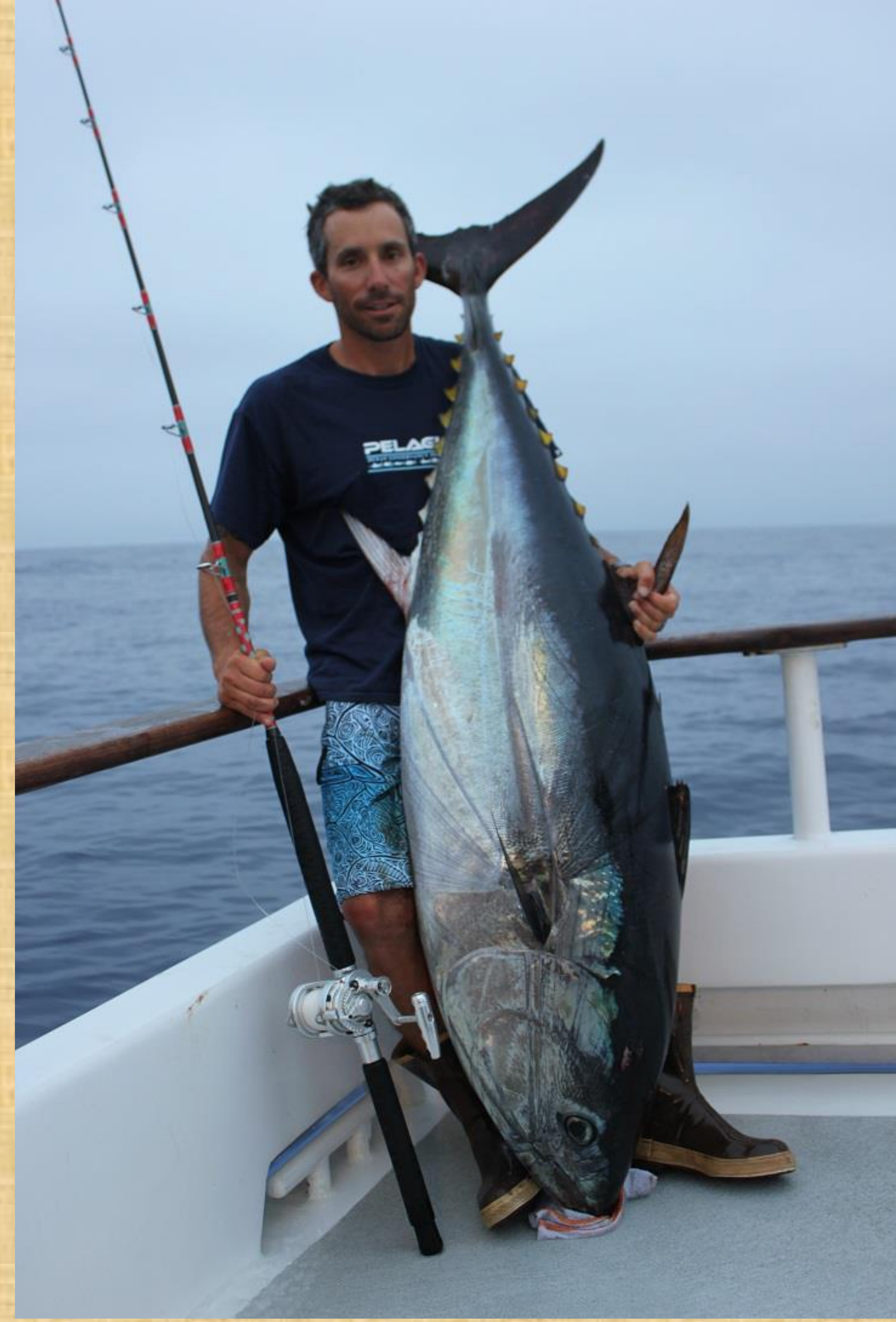
Conclusions so far:

1. ^{134}Cs and ^{137}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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