Supporting Information

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SI Text: Dose Calculations

Biological effects of ionizing radiation exposure are expressed in nonhuman biota via the radiological dose they absorb from external and/or internal pathways and that correspond to the energy deposited in their bodies. The intensity of this deposit is a function of the radiation's energy, as well as the organism's shape and composition, which determine the penetration of emitted radiation. Focusing on internal exposure to i radioisotopes (^{134}Cs, ^{137}Cs, ^{110m}Ag, ^{210}Po, ^{40}K) for the biota in our study, we determined the corresponding dose conversion coefficients (DCCs), which are specific for each radionuclide–organism combination. These coefficients are used to simplify the calculations, allowing conversion of the activity of a radionuclide in an organism (Bq unit mass^{-1} or unit volume^{-1}) to a dose rate (Gy unit time^{-1}) as shown in Eq. **S1** (1). Calculations, considering the size of the organisms (Table S1) and their composition (Table S2), used the EDEN software to calculate the internal DCCs (Tables S3, S4, S5, and S6)

$$\mathrm{IDR}(i,j) = \mathrm{DCC}_{\mathrm{int}}(i,j) \times C(i,j). \quad \textbf{[S1]}$$

Internal dose rate (IDR) is shown to be a function of the ith radionuclide and jth organism, $\mathrm{DCC}_{\mathrm{int}}(i,j)$ is the dose conversion coefficient for internal exposure of the organism j to radionuclide i [μGy·h^{-1} per Bq·kg^{-1} wet weight] and $C(i,j)$ is the activity concentration of radionuclide i in organism j [Bq·kg^{-1} wet weight, measured as dry and converted into wet by applying the dry to wet ratio of 0.244 for Pacific bluefin tuna (PBFT)].

For PBFT, considering their migration, we calculated (Eq. **S2**) the internal dose these fish received from the beginning of their exposure in Japanese waters to their catch in California, referred to as the internal cumulative dose

$$\mathrm{ICD}(i,\mathrm{PBFT}) = \sum_{t=0}^{120} \mathrm{IDR}(i,\mathrm{PBFT}) \times t. \quad \textbf{[S2]}$$

The internal dose rate IDR (i,PBFT) was calculated for the same time that the fish activity concentration $C(i,$ PBFT) was calculated (i.e., 0, 30, 60, 90, and 120 d before their capture). These back-calculated radioactivity concentrations matched independent field surveys for Japanese coastal waters presented by Ministry of Agriculture, Forestry and Fisheries in Japan (2). We then used the function that best fit the data to calculate the daily internal dose rate (Fig. S1). The fitting process was performed on data up to March 30, 2011 (148 d before capture), to compare the back-calculated results with those of modeling approaches with no direct radioactivity measurements in biota (3). Garnier-Laplace et al.'s (3) modeling estimated biota concentrations of radioactivity by considering the peak seawater concentrations during the 3-wk period just after the accident (330 m offshore the Fukushima Daiichi site on March 30) and using equilibrium-based bioconcentration factors (CFs). These authors then estimated a dose rate of 1.5 $\times 10^3$ μGy·h^{-1} for pelagic fish internally exposed to $^{134+137}$Cs. Activity concentrations in water decreased rapidly with distance due to very high dilution (about 1/1,000 30 km offshore), and Cs concentration factors show a large interspecies variability. Taking into account dilution and their use of equilibrium CFs, this may explain why their dose rates are about two orders of magnitude greater than those derived from actual samples (as calculated in the present study: 1.6–2.5 $\times$ 10^{-2} μGy·h^{-1}). The contribution of internal dose rate due to cesium isotopes is estimated to be about 75% of the total dose rate absorbed by marine organisms from artificial radionuclides measured in seawater on March 30, 2011 (Table S7).

Doses received by humans [D_i; (Sv)] from consuming contaminated PBFT can be calculated (Eq. **S3**) as a product of radionuclide concentration in the tuna muscle [C_i (Bq·kg^{-1} wet weight); Table 1], the mass ingested [R (kg); from refs. and5)], and a dose coefficient [DC (Sv·Bq^{-1})]

$$D_i = C_i \times R \times DC. \quad \textbf{[S3]}$$

The wet weight of PBFT was estimated by multiplying the measured dry weight by 0.244 (Table 1). The human ingestion rates for fish and the scenarios developed in this paper are found in the main text. The DC incorporates sophisticated calculations that incorporate aspects of human physiology, radiation physics, and the temporal and spatial deposition of energy absorbed from consuming radionuclide-contaminated foodstuffs, as developed and tabulated by the International Commission on Radiation Protection (6). The DC includes the fraction of ingested material that is absorbed and crosses the wall of the human gastrointestinal tract (i.e., 1.0 for Cs and K isotopes; 0.5 for Po); a tissue weighting factor (W_T) that accounts for differences in probability of stochastic effects occurring among different tissues; a radiation weighting factor (W_R) that accounts for the differences in biological damage from different types of radiation emissions due to the amount of energy deposited [W_R = 1 for gamma and electron emissions and 20 for alpha emissions (6)]; and integration of the dose over time because the irradiation of tissues from ingested radionuclides is time-dependent due to the physical half-life of the radioisotope, as well as the kinetics of the element within the body. For adults, the integration period is 50 y. Combined, these calculations result in what the International Commission on Radiological Protection terms the "committed effective dose" (6).

The scientific data on dose–response relationships at very low doses of ionizing radiation are inconclusive, and unfortunately such scientific uncertainties do little to assure the public and can lead to mistrust. Two opposing views prevail regarding the uncertainties in risk estimates for cancer due to exposure to ionizing radiation at the low doses reported in this paper. One approach argues that until the uncertainties of dose–response relationships at low doses are resolved, it is prudent to endorse a risk model of cancer induction that is linearly proportional to the dose received, even at extremely low levels. The US National Academy of Sciences has stated: "given our current state of knowledge, the most reasonable assumption is that the cancer risks from low doses of x- or gamma-rays decrease linearly with decreasing dose" (7). This approach implies that even if additional doses are smaller than that from environmental background, there would be a proportional increase in cancer rates. The counter argument is that low doses of radiation either produce no additional cancers or do so at an undetectable extent that precludes quantification and renders estimates of increased cancer rates highly questionable. The science is emerging on this issue. For example, recent research on the mechanistic response and repair of DNA in human cells following exposure to low doses of radiation "casts considerable doubt on the general assumption that risk to ionizing radiation is proportional to dose" (8). Further, the United Nations Scientific Committee on the Effects of Atomic Radiation stated that due to the uncertainties in the assessment of risk at low doses, it "does not recommend multiplying low dose by large numbers of individuals to estimate numbers of radiation-induced health effects within a population

exposed to incremental dose at levels equivalent to or below natural background levels" (9). Given that (1) the estimated doses that human consumers of Cs-tainted PBFT would receive are well below those from environmental background radioactivity and (2) considerable uncertainties exist in estimating increased cancer rates from such low doses, estimated human health risks from PBFT consumption should be regarded accordingly.

1. Beaugelin-Seiller K, Jasserand F, Garnier-Laplace J, Gariel JC (2006) Modeling radiological dose in non-human species: Principles, computerization, and application. *Health Phys* 90(5):485–493.
2. Ministry of Agriculture Forestry and Fisheries (2013) Results of the inspection on radioactivity materials in fisheries products. Available at: http://www.jfa.maff.go.jp/e/inspection/index.html. Accessed February 5, 2013.
3. Garnier-Laplace J, Beaugelin-Seiller K, Hinton TG (2011) Fukushima wildlife dose reconstruction signals ecological consequences. *Environ Sci Technol* 45(12): 5077–5078.
4. FAO (2012) FAOSTAT. Available at http://faostat.fao.org/site/610/default.aspx#ancor. Accessed August 3, 2012).
5. Puffer H, Duda M, Azen S (1982) Potential health hazards from consumption of fish caught in polluted coastal waters of Los Angeles County. *North American Journal of Fisheries Management* 2(1):74–79.
6. International Commission on Radiation Protection (2012) Compendium of dose coefficients based on ICRP Publication 60. ICRP Publication 119. *Ann ICRP* (Suppl):1–130.
7. Brenner DJ, et al. (2003) Cancer risks attributable to low doses of ionizing radiation: assessing what we really know. *Proc Natl Acad Sci USA* 100(24):13761–13766.
8. Neumaier T, et al. (2012) Evidence for formation of DNA repair centers and dose-response nonlinearity in human cells. *Proc Natl Acad Sci USA* 109(2):443–448.
9. UNSCEAR (2012) Report of the United Nations Scientific Committee on the effects of atomic radiation. *Proceedings of the Fifty-Ninth Session* (United Nations, New York), pp 1–14.

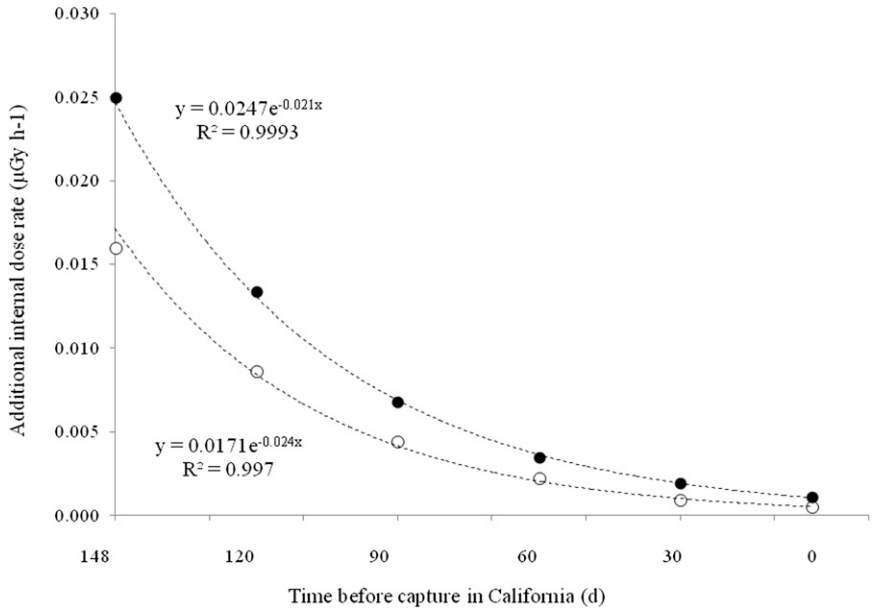

Fig. S1. Fitted curves on $^{134+137}$Cs internal dose rates calculated for PBFT at different times before their capture off California (upper curve, radiation-weighted values; lower curve, nonweighted values).

Table S1. Dimensions and weights of organisms used for the dose calculations

Organisms	Length (cm)	Width (cm)	Height (cm)*	Weight (g)	Dry: wet weight ratio
Euphausiids	2.00	2.00	2.00	0.07	0.20
Copepods	0.001	0.001	0.001	0.05	0.17
Fish	8.00	1.00	7.00	30.0	0.30
Jellyfish	5.00	5.00	7.60	100	0.04
PBFT 1	72.4	7.20	29.29	8,040	0.24
PBFT 2	64.0	6.40	25.60	5,498	0.24
PBFT 3	60.6	6.10	24.10	4,633	0.24
PBFT 4	68.0	6.80	27.33	6,613	0.24
PBFT 5	69.0	6.90	27.76	6,914	0.24
PBFT 6	69.0	6.90	27.76	6,914	0.24
PBFT 7	69.5	6.90	27.98	7,068	0.24
PBFT 8	66.5	6.70	26.68	6,178	0.24
PBFT 9	58.6	5.90	23.24	4,182	0.24
PBFT 10	66.0	6.60	26.47	6,038	0.24
PBFT 11	67.5	6.70	27.11	6,466	0.24
PBFT 12	68.5	6.80	27.55	6,762	0.24
PBFT 13	65.5	6.60	26.25	5,899	0.24
PBFT 14	63.1	6.30	25.17	5,240	0.24
PBFT 15	65.5	6.60	26.25	5,899	0.24

*Deduced from the equivalent ellipsoid volume and weight considering a body density of 1 g·cm^{-3}.

Table S2. Elemental composition (% of total mass) of seawater and animals (1)

Element	Seawater	Animal
Br	6.70E−03	—
C	2.80E−03	1.43E+01
Ca	4.00E−02	—
Cl	1.94E+00	1.00E−01
H	1.08E+01	1.02E+01
K	4.00E−02	1.00E−01
Mg	1.29E−01	—
N	—	3.40E+00
Na	1.08E+00	1.00E−01
O	8.58E+01	7.10E+01
P	—	2.00E−01
S	9.10E−02	3.00E−01

Hyphen denotes negligible quantity.

1. International Commission on Radiation Units and Measurements (1992) *Photon, Electron, Proton and Neutron Interaction Data for Body Tissues* (Nuclear Technology Publishing, Ashford, UK), ICRU Report 46.

Table S3. Additional internal dose rates attributable to radioactive Cs in PBFT calculated at different times before their capture in waters off California in August 2011

	Dose rate ($\mu Gy \cdot h^{-1}$)	
Days before capture	Nonweighted	Radiation-weighted
148	1.6E−02	2.5E−02
120	8.6E−03	1.3E−02
90	4.4E−03	6.8E−03
60	2.3E−03	3.4E−03
30	9.2E−04	1.9E−03
0	5.2E−04	1.1E−03

Table S4. Internal DCCs (10^{-4} $\mu Gy \cdot h^{-1}$ per $Bq \cdot kg^{-1}$) for 15 individual PBFT samples for ^{134}Cs, ^{137}Cs, ^{210}Po, and ^{40}K (radiation-weighted and nonweighted DCCs are shown)

Radionuclide	PBFT 1	PBFT 2	PBFT 3	PBFT 4	PBFT 5/6*	PBFT 7	PBFT 8	PBFT 9	PBFT 10	PBFT 11	PBFT 12	PBFT 13/15*	PBFT 14	Mean
Nonweighted DCCs														
^{134}Cs	2.48	2.30	2.25	2.38	2.41	2.41	2.36	2.20	2.35	2.37	2.39	2.34	2.29	2.35
^{137}Cs	1.02	1.01	1.01	1.01	1.01	1.01	1.01	1.01	1.01	1.01	1.01	1.01	1.01	1.01
^{137m}Ba	0.98	0.91	0.89	0.94	0.95	0.95	0.94	0.88	0.93	0.94	0.95	0.93	0.90	0.93
^{210}Po	29.1	29.1	29.1	29.1	29.1	29.1	29.1	29.1	29.1	29.1	29.1	29.1	29.1	29.1
^{40}K	0.14	0.13	0.12	0.13	0.13	0.14	0.13	0.12	0.13	0.13	0.13	0.13	0.12	0.13
Weighted DCCs														
^{134}Cs	4.17	4.01	3.95	4.09	4.12	4.12	4.07	3.91	4.05	4.08	4.09	4.05	4.00	4.06
^{137}Cs	3.05	3.04	3.04	3.05	3.05	3.05	3.05	3.04	3.04	3.05	3.05	3.04	3.04	3.04
^{137m}Ba	1.67	1.60	1.58	1.63	1.65	1.65	1.63	1.56	1.62	1.63	1.63	1.62	1.60	1.62
^{210}Po	291	291	291	291	291	291	291	291	291	291	291	291	291	291
^{40}K	0.14	0.13	0.12	0.13	0.13	0.14	0.13	0.12	0.13	0.13	0.13	0.13	0.12	0.13

The daughter product from the decay of ^{137}Cs (^{137m}Ba) is also shown.
*PBFT individuals of same size.

Table S5. Development over time for the DCCs (μGy·h^{-1} per Bq·kg^{-1}) for 15 individual PBFT for 0–148 d before their catch off California

Radionuclide	148 d	120 d	90 d	60 d	30 d	0 d
Nonweighted values						
^{134}Cs	2.14E−04	2.18E−04	2.23E−04	2.27E−04	2.30E−04	2.35E−04
^{137}Cs	1.01E−04	1.01E−04	1.01E−04	1.01E−04	1.01E−04	1.01E−04
^{137m}Ba	8.46E−05	8.63E−05	8.83E−05	8.96E−05	9.13E−05	9.29E−05
^{210}Po	2.91E−03	2.91E−03	2.91E−03	2.91E−03	2.91E−03	2.91E−03
^{40}K	1.11E−05	1.15E−05	1.19E−05	1.22E−05	1.26E−05	1.30E−05
Radiation-weighted values						
^{134}Cs	3.84E−04	3.88E−04	3.93E−04	3.98E−04	4.01E−04	4.06E−04
^{137}Cs	3.04E−04	3.04E−04	3.04E−04	3.04E−04	3.04E−04	3.04E−04
^{137m}Ba	1.53E−04	1.55E−04	1.57E−04	1.59E−04	1.60E−04	1.62E−04
^{210}Po	2.91E−02	2.91E−02	2.91E−02	2.91E−02	2.91E−02	2.91E−02
^{40}K	1.11E−05	1.15E−05	1.19E−05	1.22E−05	1.26E−05	1.30E−05

DCCs are for ^{134}Cs, ^{137}Cs, ^{210}Po, and ^{40}K (radiation-weighted and nonweighted DCCs are shown). The daughter product from the decay of ^{137}Cs (^{137m}Ba) is also shown. Calculations were made on the basis of mean characteristics of the 15 individuals (*cf* Table S1).

Table S6. DCCs (μGy·h^{-1} per Bq^{-1}·kg^{-1}) for organisms other than PBFT for ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{210}Po, and ^{40}K (radiation-weighted and nonweighted DCCs are shown)

Radionuclide	Euphausiids	Copepods	Deep-sea fish	Jellyfish
Nonweighted values				
^{110m}Ag	7.13E−05	1.61E−06	8.08E−05	1.36E−04
^{110}Ag	4.96E−04	5.67E−08	4.88E−04	5.92E−04
^{134}Cs	1.04E−04	1.12E−06	1.10E−04	1.44E−04
^{137}Cs	9.88E−05	9.04E−07	9.88E−05	1.01E−04
^{137m}Ba	4.03E−05	1.96E−08	4.25E−05	5.71E−05
^{210}Po	2.90E−03	1.85E−04	2.90E−03	2.91E−03
^{40}K	1.89E−06	9.58E−10	2.37E−06	5.21E−06
Radiation-weighted values				
^{110m}Ag	1.41E−04	7.33E−04	1.51E−04	2.07E−04
^{110}Ag	1.48E−03	1.16E−04	1.47E−03	1.77E−03
^{134}Cs	2.70E−04	5.33E−04	2.76E−04	3.14E−04
^{137}Cs	2.96E−04	1.01E−04	2.96E−04	3.03E−04
^{137m}Ba	1.04E−04	2.20E−04	1.06E−04	1.25E−04
^{210}Po	2.90E−02	1.85E−03	2.90E−02	2.91E−02
^{40}K	1.89E−06	9.58E−10	2.37E−06	5.21E−06

The daughter products from the decay of ^{137}Cs and ^{110m}Ag (^{137m}Ba and ^{110}Ag, respectively) are also shown.

Table S7. Internal and total dose rates absorbed by a hypothetical pelagic fish, estimated from seawater radioactivity values using bioconcentration factors for March 30, 2011, recalculated from data of Garnier-Laplace et al. (1)

Parameter	^{140}Ba	^{134}Cs	^{136}Cs	^{137}Cs	^{131}I	^{140}La
Water concentration (Bq L^{-1})	7.3E+03	4.7E+04	4.2E+03	4.7E+04	1.8E+05	3.6E+03
Aggregated DCC* (μGy·h^{-1} per Bq L^{-1})	8.0E−03	1.7E−02	1.8E−02	1.6E−02	7.0E−04	5.2E−02
Total dose rate/radionuclide (μGy·h^{-1})	6.1E+01	8.1E+02	7.7E+01	7.4E+02	1.2E+02	1.9E+02
Internal DCC/radionuclide (μGy·h^{-1} per Bq kg^{-1})	7.0E−04	2.0E−04	2.0E−04	2.0E−04	1.0E−04	4.0E−04
Concentration ratio (L·kg^{-1})	1.0E+01	8.6E+01	8.6E+01	8.6E+01	1.0E−04	1.2E+02
Internal dose rate (μGy·h^{-1})	5.0E+01	7.7E+02	7.2E+01	7.3E+02	3.0E−03	1.8E+02

*aggregated DCC includes both internal and external exposure, giving in a single value the total estimated dose rate per water concentration.

1. Garnier-Laplace J, Beaugelin-Seiller K, Hinton TG (2011) Fukushima wildlife dose reconstruction signals ecological consequences. *Environ Sci Technol* 45(12):5077–5078.