PACIFIC SCIENCE Early View

A Quarterly Devoted to the
Biological and Physical Sciences of the Pacific Region

In the Wake of Fukushima: Radiocesium Inventories of Selected North Pacific Fish

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Abstract
Thirteen commonly consumed types of fish caught in the North Pacific and locally available in Hawaii were analyzed using gamma spectroscopy to measure Fukushima-derived and historic ^{134}Cs and ^{137}Cs isotopes. All fish samples had detectable ^{137}Cs above 95% Confidence Intervals. Three out of the thirteen samples had ^{134}Cs, an isotope indicative of Fukushima releases, detected above 95% Confidence Intervals. The highest ^{134}Cs and ^{137}Cs concentration in the examined species was in ahi tuna carrying 0.10±0.04 Bq/kg and 0.62±0.05 Bq/kg, respectively. Other samples with ^{134}Cs activities found above their 2-sigma uncertainty were albacore tuna and swordfish. Historic and Fukushima-derived contributions were evaluated and in several samples the Fukushima-derived radiocesium dominated the total radiocesium inventory with up to 61% contribution. All activities were below derived intervention limits of 1200 Bq/kg and the doses to humans from consuming the fish attributable to radiocesium were 0.02-0.2 μSv, in comparison to 6-20 μSv contributed by the natural ^{40}K present in the same fish.

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Pacific Science, vol. 71, no. 2
December 9, 2016 (Early view)

Official Journal of the Pacific Science Association

Introduction

The magnitude 9.0 Tohoku earthquake and tsunami led to significant damage at the Fukushima Daiichi Nuclear Power Plant in March 2011. Following the accident, radioisotopes escaped into the atmosphere and the marine environment (Buesseler et al. 2011, Inomata et al 2016, Tsubono et al 2016). Additionally, over 500 tons of contaminated wastewater was released into the Pacific Ocean (Watabe et al. 2013). The total impact of the releases still remains unclear, as the concentration of cesium isotopes off the coast of Japan remain elevated and release of these isotopes from the terrestrial to marine domain is ongoing (Buesseler et al. 2011, Buesseler 2012).

While radionuclide activities in the immediate vicinity of the damaged reactor were significantly elevated, Fukushima-derived radiocesium activities near the Hawaiian Islands were indistinguishable from the pre-Fukushima baseline. In 2012 the southeastern most boundary of the radiation plume brushed just north of the Midway Island with low activities totaling only about 3-5 Bq/m^3, including the pre-Fukushima concentration of 1-2 Bq/m^3 (Kameník et al. 2013). In 2011, before the accident, the levels of ^{137}Cs in the central Pacific Ocean were around 1.5 Bq/ m^3 from remnants of fallout from the nuclear weapons tests performed in the second half of the 20th century (Smith et al. 2015) and levels of ^{134}Cs were below detection due to its short, two-year half life. Therefore, ^{134}Cs may be used as a Fukushima-only derived tracer used to track the power plant related contamination. Recent estimates of radiocesium oceanic contamination from Fukushima range from 3-4 PBq via direct discharge and 12-15 PBq through atmospheric deposition (Aoyama et al 2015). Even though ocean circulation and dispersion models predicted the radiocesium plume to reach the Hawaiian Islands (Rossi et al. 2013), only the preexisting ^{137}Cs baseline activities were observed on Oahu, Station Aloha (45N, 158W) and the Kona Coast (19N, 156W) with no detectable ^{134}Cs (Kamenik et al. 2013). Therefore, there has not been a systemic study of radiocesium bioaccumulation of marine organisms in the central Pacific region. But Madigan et al. (2012) showed that migrating organisms can transport the Fukushima-signature over significant distances as they showed detectable ^{134}Cs (6.3±1.5 Bq/kg) in Pacific bluefin tuna caught off the California coast only a year after the incident. Another study found 0.23-0.82 Bq/kg of ^{134}Cs in the East Pacific Albacore (*Thunnus alalunga*) in 2012 (Neville et al 2014). Accordingly, while the plume has not necessarily reached the Hawaiian Islands, it did travel within established fishing grounds across major migratory paths northeast of the islands in the Kuroshio and Kuroshio extension currents (Madigan et al. 2012, Kameník et al. 2013). This warranted a closer examination of radiocesium content of fish consumed in the Hawaiian Islands.

This is the first study focusing on post-Fukushima radiocesium bioaccumulation in commonly consumed North Pacific fish available in Hawaiian markets. The goal of the study was to directly compare radiocesium inventories and associated committed effective doses to human consumers in fish from pre-Fukushima, historic sources that include nuclear weapons, to the more recent, Fukushima releases. Because of the absence of prior information on bioaccumulation in different species and the uncertainty of the location of their habitat with respect to the radiocesium plume, no one species could be targeted, rather a suite of several fish species was surveyed. For completeness, this study also reports trophic level, as well as general depth and geographic region of habitat, but the low sample size and absence of information on exact fishing ground location does not allow us to draw significant conclusions regarding how these factors influence radiocesium bioaccumulation in fish. For reference, the study also compares radiocesium levels to another beta/gamma emitter ^{40}K, which is a naturally occurring radioisotope and has a very similar chemical behavior in aquatic systems as cesium. This study does not address the total committed effective dose from all radionuclides present in fish, which are usually dominated by natural radionuclides, e.g. ^{210}Po (Fisher et al 2013, Johaneson et al 2015). The study also does not report on dose rates to fish. The significance of the study goes beyond local relevance of informing the public on current levels of radiocesium consumed in fish; it also informs the public and health officials on pre- vs. post-Fukushima levels of radiocesium and provides perspective on the Fukushima releases in comparison to other historic radiocesium sources in the Pacific.

Methods

Thirteen commonly consumed types of fish available in local markets were chosen based on information from suppliers, confirming that they were caught in the North Pacific, not restricted to areas near Hawaii but excluding fish caught below 20°N as the plume was not expected to disperse below this latitude (Kamenik et al 2013). This paper does not report specifically where each sample was caught as some of this information is proprietary to the supplier. Other general characteristics of each species were identified from a referenced general public information base (Fish Base 2015) and incorporated into the study (Table 1). The flesh of samples were frozen, freeze-dried on a benchtop freeze drier, milled, and filled into 120-mL polypropylene cylinder containers (5 cm diameter). Sample masses ranged from 100 to 150 grams dry weight and were each counted for 7 days. The samples were analyzed using gamma spectroscopy (Ortec HPGe, model GEM40, relative efficiency 43 %, resolution 1.76 keV for 1.33 MeV gamma line of ^{60}Co) to measure ^{134}Cs and ^{137}Cs isotopes using their 604 and 661 keV photopeaks, respectively. The gamma spectra were evaluated using Hypermet-PC V5.01 software (Révay et al., 2001). The gamma-spectrometric efficiency was determined using certified reference fish tissue material obtained from the Japanese Society for Analytical Chemistry (Yonezawa 2015). Efficiencies at the two gamma lines of interest were 2.3% and 2.5% for 604 and 661 keV, respectively. The radiocesium activities were decay corrected to time of purchase in March 2015. Final activities were expressed as Bq/kg of wet weight.

<Table 1 about here>

The committed effective dose to the population received from consuming the fish containing the measured radiocesium activities was calculated as:

$$\text{Dose} = C_i * R_{\text{ing}} * \text{DC}$$

Where C_i is the radionuclide concentration (Bq/kg wet weight), DC represents the dose coefficient (nSv/Bq), and R_{ing} is the ingestion rate (Eckerman et al 2012), similarly to the calculation documented in Fisher et al (2013). An ingestion rate of 24.1 kg of seafood/year was used, which represents average consumption for the US population. The dose calculation assumed that only one species of fish was consumed year round.

Results

The sum of ^{134}Cs and ^{137}Cs radiocesium concentrations measured in the fish specimens range from 0.08±0.03 to 0.72 ±0.06 Bq/kg of wet weight (Figure 1, Table 2), far below the United States Food and Drug Administration (FDA) Derived Intervention Level (DIL) for imported foods of 1200 Bq/kg (^{137}Cs + ^{134}Cs combined) and the critical limit set by the FDA for either cesium isotope, which is 370 Bq/kg (FDA, 2015).

<Figure 1 about here>

Five samples showed the Fukushima tracer ^{134}Cs, present above critical levels and at the 68% confidence interval (CI at 1-sigma uncertainty) but only three of those fish exhibited activities above the range of their 2 sigma uncertainty representing 95% CI. The highest ^{134}Cs concentration was found in the ahi (*Thunnus obesus*) sample but did not exceed 0.10±0.04 Bq/kg. Other species with ^{134}Cs evident above the 95% CI were albacore tuna and swordfish (Table 2). The CL for ^{134}Cs was about 0.005 Bq/kg, and the method was not sensitive enough to account for the very low ^{134}Cs activities due to the fact that by the time of sampling two halflives had passed for the 134 isotope (^{134}Cs $t_{1/2} = 2$ years). All samples had ^{137}Cs present at levels above CL and 95% CI. The maximum ^{137}Cs concentration found was 0.72 ± 0.06 Bq/kg. The highest concentrations resided in onaga (*Etelis coruscans*) and a species known to be migrating between Japan and the central/northeast Pacific (Masuda et al 1984), monchong (*Taractichthys steindachneri*).

<Table 2 about here>

The naturally occurring ^{40}K on the other hand revealed concentration levels ranging from over 42 Bq/kg to 161 Bq/kg. The highest concentration of 161 ± 7 Bq/kg existed in the fatty local game fish opah (*Lampris regius*) sample, while the lowest concentration of 43 ± 2 Bq/kg resided in the Alaskan species, the dover sole (*Microstomus Pacificus*). The CLs for ^{40}K were around 1 Bq/kg falling far below the detected levels in each sample. This study did not analyze other alpha and beta emitting radionuclides and therefore does not account for activities or doses associated to other Fukushima-derived or natural radionuclides that may be present.

Discussion

Fukushima-derived new vs pre-Fukushima historic radiocesium levels

In this study we present individual radiocesium activities decay corrected to the date of purchase in 2015 to calculate the cesium-derived dose to humans by consuming freshly caught fish. The detected radiocesium is the combination of radionuclides from the 2011 Fukushima release (^{134}Cs and ^{137}Cs would be present) and previous historic releases that are present as ^{137}Cs, which is no longer accompanied by ^{134}Cs as the latter isotope decayed. Table 3 shows radiocesium estimates in fish parsed out to radiocesium linked to the Fukushima accident and the historic baseline concentrations from pre-Fukushima sources. This is possible by using the 2011-decay corrected ^{134}Cs and the assumption that ^{134}Cs and ^{137}Cs were released in a 1:1 ratio in 2011.

This way the old, historic, and new, Fukushima-derived, ^{137}Cs can be determined (Table 3) assuming that no mass dependent fractionation occurs during the bioaccumulation process. There were three samples that had new radiocesium contribution equal to or exceeding pre-Fukushima levels at 95% CI , while in the rest of the samples historic fallout dominates the radiocesium inventory in the fish. This finding highlights the fact that the Fukushima-derived radiocesium contribution to fish is minimal or equal to radiocesium levels from historic sources in distant regions from the nuclear accident.

<Table 3 about here>

Committed effective dose to humans from consuming the sampled fish

The US FDA designated derived intervention limit for radiocesium isotopes in fish is 1200 Bq/kg and our data indicate that the tested fish samples had less then 0.1% of this level. To calculate the radiocesium-derived committed effective dose to humans from consuming the sampled fish, radionuclide-specific committed effective dose coefficients for adult human ingestion (DC) were utilized in the calculation. The DC for ^{137}Cs is 13 nSv/Bq and for ^{134}Cs is 19 nSv/Bq (Eckerman et al 2012). The highest concentration of total radiocesium in the tested fish samples is about 0.72 Bq/kg, which corresponds to a dose of 240 nSv/year, assuming a consumption of 24 kg/yr. Eating one 100-g wet weight serving of this fish results in a radiocesium-derived dose of 1 nSv. Other radionuclides not tested in this study would contribute a more significant additional dose, for example the naturally occurring alpha-emitter ^{210}Po has been shown to contribute as much as 90% of the total dose in fish (Johansen et al 2015). Table 4 displays the committed effective dose based on the above listed DCs for radiocesium isotopes and 6.2 nSv/Bq for ^{40}K (Eckerman et al 2012), and ingestion rates of 24.1 kg/yr of fish. The conversion of activities to dose reveals that ^{40}K is responsible for about 100 times higher dose than the two cesium isotopes combined (Table 4). Doses to humans from consuming the fish specifically attributable to radiocesium were 0.02-0.2 μSv/yr, in comparison to 6-20 μSv/yr contributed by the natural ^{40}K present in the same fish. The radiocesium and radiopotassium doses estimated here are comparable to those reported by Fisher et al. (2013). Again, it must be pointed out that radiocesium represents only a fraction of the total dose attributable to other radionuclides present in fish (Johansen et al 2015).

<Table 4 about here>

Conclusion

^{134}Cs and ^{137}Cs activities of each specimen were far below the FDA's DIL of 1200 Bq/kg. All samples had less than 1 Bq/kg of total radiocesium activity, and only five samples showed evidence of Fukushima-derived ^{134}Cs. Higher total radiocesium concentrations were found in fish that reportedly inhabit regions between Japan and the central/northeast Pacific. These may have accumulated radiocesium either through migration routes, or may have lived near Japan and were caught and imported from those fishing grounds. The highest total radiocesium activity was found in ahi and onaga. New, Fukushima-derived radiocesium contribution ranges from 0-61% of the total radiocesium inventory in fish, ahi, albacore tuna, and swordfish being the 3 samples with detectable levels. This study suggests that about 40% of fish tested here and are consumed on the islands of Hawai'i were recently exposed to the path of the Fukushima-derived radiocesium plume in the North Pacific Gyre.

The committed effective dose to humans from consuming the fish in this investigation from 2015 is 0.02-0.2 μSv/yr which is significantly lower than the range reported in fish near Japan for the period of 2011-2014 (Buesseler et al. 2015) with a potential committed effective dose of >80 μSv/yr equivalent to 100 Bq/kg set as the intervention limit in Japan. This shows that while the largest anthropogenic deposition of radioactivity into the local marine ecosystem near Japan resulted in fisheries closures in several prefectures near Fukushima, radiocesium became diluted enough throughout the Pacific to not affect consumption recommendations for fish currently caught in the rest of the Pacific. In addition, this study found that committed effective dose to humans from consuming the tested fish is 100 times higher from the naturally occurring ^{40}K than what can be related to total radiocesium inventory. Yet, an even higher dose is expected from naturally occurring alpha-emitters that were not quantified in this study.

This study provides a snapshot of a few selected species sampled in 2015, four years after the accident. Regular sampling of species that show signs of Fukushima-derived radiocesium may better inform us about the variability and spatial distribution of the released radiocesium isotopes.

Acknowledgements

The authors are grateful to K.G.W. Inn for his guidance on data processing. H.A. acknowledges funding from the University of Hawaii Undergraduate Research Opportunities Program. This paper is SOEST publication record number 9855.

Table 1. List of fish species sampled, the species general reported geographic region of habitat and depth, trophic level, usual size, and maximum age as reported in a database Fish Base (Fish Base 2015: Allen 1985, Cohen et al 1990, Collette et al 1983, Eschmeyer et al 1983, Nakamura 1985, Paige et al 1991, Palko et al 1982, Palmer 1986, Smith 1986,).

:

Selected Fish	Species	Geographic Range				Habitat Depth		Trophic Level		Size (cm)		Maximum Age Reported
		N	**S**	**E**	**W**	**Maximum**	**Minimum**		**±**	**Maximum**	**Average**	
Ahi (Bigeye Tuna)	*Thunnus obesus*	45N	43S	180E	180W	250	0	4.5	0	250	180	11
Albacore Tuna	*Thunnus alalunga*	59N	46S	180E	180W	600	0	4.3	0.2	140	100	13
California King Salmon (Chinook)	*Oncorhynchus tshawytscha*	72N	27°N	136E	109W	375	0	4.4	0.7	150	70	9
Alaskan Cod	*Gadus macrocephalus*	63N	31°N	119E	119W	1280	0	4.2	0	119	67	25
Dover Sole	*Microstomus pacificus*	65N	31N	180E	180W	1370	10	3.2	0.1	76	40	56
Alaskan Halibut	*Hippoglosus stenolepis*	73N	42N	138E	123W	1200	0	4.1	0.2	258	-	55
Mahi Mahi (Dorado)	*Coryphaena hippurus*	47N	38S	180E	180W	85	0	4.5	0.6	127	50	4
Monchong (Pomfret)	*Taractichthys steindachneri*	40	36	40E	110W	700	50	4.3	0.5	60	-	8
Onaga (Long-Tail Red Snapper)	*Etelis coruscans*	35 N	32 S	29E	142W	400	0	4.4	0.2	120	50	-
Opakapaka (Crimson Snapper)	*Pristipomoides filamentosus*	35N	26S	31E	144W	400	40	4.2	0.4	100	50	44
Opah (Moonfish)	*Lampris guttatus*	70N	45S	180E	180W	500	100	4.2	0.62	200	120	11
Swordfish	*Xiphias gladius*	61N	50S	180E	180W	800	0	4.5	0.2	455	300	20
Yellowfin Tuna	*Thunnus albacares*	59N	48S	180E	180W	250	0	4.4	0.4	239	150	9

Table 2. Radiocesium isotope and 40K activities measured in fish samples in this study. Radionuclide activities are given in Bq/kg of wet weight, their reported uncertainty is 2 sigma, also provided are critical levels that express the a posteriori decision level of the minimum measured concentration that must be present in a sample for detecting the analyte with a tolerable error of false detection (α=5%). All radionuclides are decay corrected to the date of purchase in the March 2015. "bd" indicates that the sample countrate was statistically not different from background.

	Radionuclide Concentrations (Bq/kg)										
Species	^{134}Cs	2 σ	CL	^{137}Cs	2 σ	CL	$^{134}Cs+^{137}Cs$	2 σ	^{40}K	2 σ	CL
Ahi	0.10	0.04	0.005	0.62	0.05	0.004	0.72	0.06	136	6	1
Albacore Tuna	0.05	0.03	0.005	0.32	0.05	0.004	0.37	0.06	120	5	1
King Salmon	0.00	0.01	0.004	0.18	0.05	0.003	0.18	0.06	156	7	1
Cod	bd	-	0.006	0.15	0.06	0.004	0.15	0.06	100	4	1
Dover Sole	0.00	0.01	0.005	0.56	0.11	0.004	0.56	0.11	43	2	1
Halibut	0.02	0.05	0.005	0.21	0.04	0.004	0.23	0.07	127	5	1
Mahi Mahi	0.01	0.02	0.004	0.15	0.06	0.003	0.16	0.07	110	9	1
Monchong	bd	-	0.006	0.70	0.15	0.005	0.70	0.15	97	4	1
Onaga	bd	-	0.007	0.72	0.06	0.005	0.72	0.06	118	5	1
Opah	0.00	0.01	0.004	0.08	0.03	0.003	0.08	0.03	161	7	1
Opakapaka	0.00	0.02	0.005	0.16	0.02	0.004	0.16	0.03	49	2	1
Swordfish	0.07	0.06	0.005	0.49	0.05	0.004	0.55	0.08	116	5	1
Yellowfin Tuna	bd	-	0.005	0.22	0.05	0.004	0.22	0.05	124	18	1

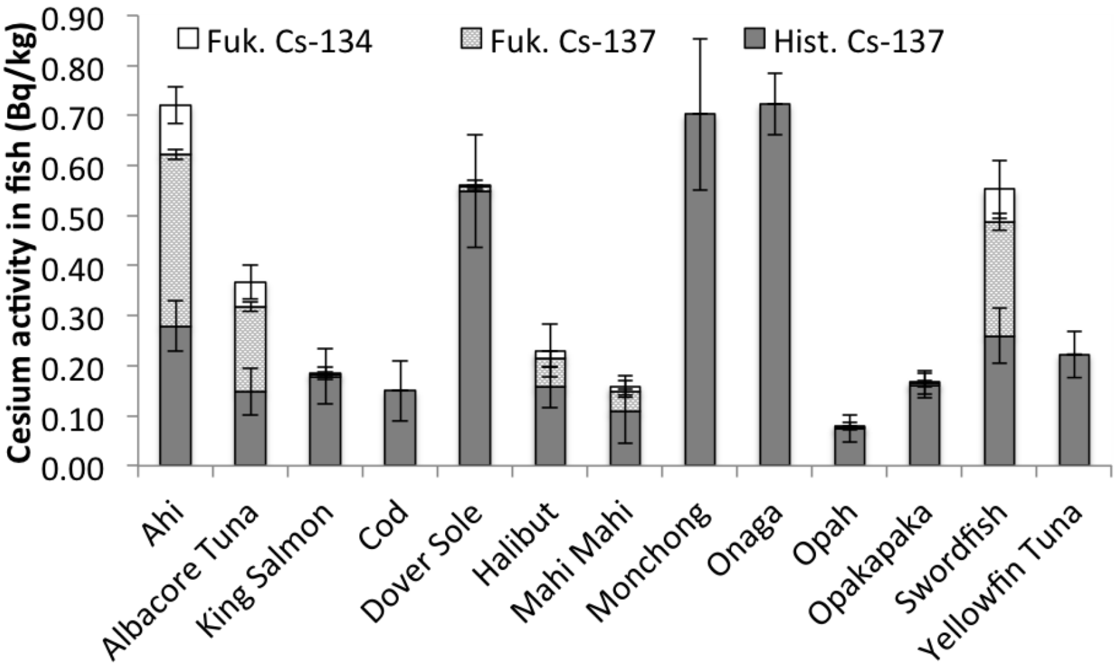
Table 3. Radiocesium levels in March 2015 (Bq/kg wet weight) and contribution from calculated historic (137Cs only), Fukushima- derived (Fuk. 137Cs and Fuk. 134Cs), and total 137Cs and 134Cs in fish. All uncertainties are 2 sigma propagated errors, “bd” indicates that the sample countrate was statistically not different from background.

	Radionuclide Concentrations (Bq/kg)										
Species	Historic ^{137}Cs	2 σ	Fuk.^{137}Cs	2 σ	Fuk.^{134}Cs	2 σ	Fuk. $^{134+137}$Cs	2 σ	Total 2015	2 σ	%Fuk.
Ahi	0.28	0.05	0.34	0.01	0.10	0.04	0.44	0.04	0.72	0.06	61
Albacore Tuna	0.15	0.05	0.17	0.01	0.05	0.03	0.22	0.04	0.37	0.06	60
King Salmon	0.18	0.05	0.00	0.00	0.00	0.01	0.01	0.01	0.18	0.06	3
Cod	0.15	0.06	bd	-	bd	-	bd	-	0.15	0.06	0
Dover Sole	0.55	0.11	0.01	0.00	0.00	0.01	0.01	0.01	0.56	0.11	2
Halibut	0.16	0.04	0.06	0.02	0.02	0.05	0.07	0.06	0.23	0.07	32
Mahi Mahi	0.11	0.06	0.04	0.01	0.01	0.02	0.05	0.02	0.16	0.07	32
Monchong	0.70	0.15	bd	-	bd	-	bd	-	0.70	0.15	0
Onaga	0.72	0.06	bd	-	bd	-	bd	-	0.72	0.06	0
Opah	0.07	0.03	0.00	0.00	0.00	0.01	0.01	0.01	0.08	0.03	6
Opakapaka	0.16	0.02	0.00	0.01	0.00	0.02	0.01	0.02	0.17	0.03	3
Swordfish	0.26	0.05	0.23	0.02	0.07	0.06	0.29	0.06	0.55	0.08	53
Yellowfin Tuna	0.22	0.05	bd	-	bd	-	bd	-	0.22	0.05	0

Table 4. Committed effective dose received by humans by consuming each investigated sample of fish based on a 24.1 kg/year US average annual consumption of seafood. Listed are individual isotope contributions to the dose from calculated historic (137Cs only), Fukushima-derived (Fuk. 137Cs and Fuk. 134Cs), and the natural 40K. Dose was calculated based on activity per wet weight of each sample and assumed the consumption of solely a single species of fish throughout the year.

	Committed Effective Dose (nSv/yr)							
Species	Historic ^{137}Cs	2 σ	Fuk.^{137}Cs	2 σ	Fuk.^{134}Cs	2 σ	^{40}K	2 σ
Ahi	87	16	107	3	45	17	20,300	500
Albacore Tuna	46	15	53	3	22	16	18,000	400
King Salmon	56	17	1	1	1	6	23,400	500
Cod	47	19					14,900	400
Dover Sole	172	35	3	1	1	4	6,500	200
Halibut	49	13	18	5	7	24	19,000	400
Mahi Mahi	34	20	12	2	5	10	16,500	700
Monchong	220	47					14,500	400
Onaga	226	19					17,700	400
Opah	23	9	1	1	1	3	24,100	500
Opakapaka	50	8	1	2	1	11	7,400	200
Swordfish	81	17	71	5	30	27	17,300	400
Yellowfin Tuna	70	15					18,600	1,400

Figure 1. Stacked bar graph of historic ^{137}Cs (Hist. Cs-137), Fukushima-derived ^{137}Cs (Fuk. Cs-137) and Fukushima-derived ^{134}Cs (Fuk. Cs-134) activities in Bq/kg of wet weight in each sample of analyzed fish species. Error bars represent 2 sigma for each component..



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