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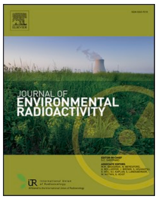

Radioactivity concentration measurements in fish and shellfish samples from the west coast of Canada after the Fukushima nuclear accident (2011–2018)

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ABSTRACT

Radioactive contamination of the Pacific Ocean following the Fukushima nuclear accident has raised public concern about seafood safety, particularly in coastal Indigenous communities. To address this, Health Canada and partners have collected and analyzed a total of 621 samples of commonly consumed salmon, ground fish, and shellfish from the Canadian west coast from 2011 to 2018. While the vast majority of the ^{137}Cs and ^{134}Cs levels were below the Minimum Detectable Concentration (MDC, typically 0.7–1.0 Bq kg^{-1} fw for a 6 h counting), further examination of 19 fish samples revealed ^{137}Cs concentrations of 0.17–0.53 Bq kg^{-1} fw with an average value and uncertainty (k = 1) of 0.29 ± 0.02 Bq kg^{-1} fw. Of these, only two samples were found to have trace levels of ^{134}Cs likely derived from the Fukushima accident. The global fallout contribution from atmospheric nuclear weapons testing to the observed ^{137}Cs in these two samples was determined to be 0.26 ± 0.08 Bq kg^{-1} fw (49 ± 14%) and 0.12 ± 0.02 Bq kg^{-1} fw (24 ± 4%) for collection years 2015 and 2016, respectively. The annual average level of ^{137}Cs in fish and shellfish was also determined by spectral summation for collection years 2014–2018. In fish, ^{137}Cs levels determined through spectral summation were relatively constant (0.18–0.25 Bq kg^{-1} fw) with an average value and uncertainty of 0.21 ± 0.02 Bq kg^{-1} fw. By contrast, 38 shellfish samples (bivalves) were measured and revealed no radiocesium or other anomalies in either tissue or shell. In all, measurements over eight years showed that the radioactivity in fish and shellfish was dominated by natural radionuclides and that the level of anthropogenic radionuclides, as indicated by the radioactive cesium content, remained small. An upper bound for ingested dose from ^{137}Cs was determined to be approximately 0.26 μSv per year, far below the worldwide average annual effective dose of 2400 μSv from exposure to natural background radiation. We can therefore conclude that fish, such as salmon, ground fish, and shellfish from the Canadian west coast are of no radiological health concern despite the Fukushima Dai-ichi nuclear accident of 2011.

1. Introduction

The Fukushima Dai-ichi nuclear accident of 2011 released 100–500 PBq of ^{131}I (half-life 8.02 days) and 6–21 PBq of ^{137}Cs (half-life 30.08 yr) to the atmosphere (UNSCEAR, 2021; Aoyama et al., 2020). In the initial phase, from March to April 2011, 3–6 PBq of ^{137}Cs and ^{134}Cs (half-life 2.07 yr) were released directly into the Pacific Ocean with another 5–15 PBq contributed via deposition from the atmosphere (UNSCEAR, 2021; Aoyama et al., 2020). Owing to its long-half life, the environmentally persistent and pervasive ^{137}Cs becomes the primary radiological concern over time. While subsequent reports would conclude that the majority of ^{137}Cs in the environment would still be derived from historical atmospheric nuclear weapons testing (henceforth referred to simply as global fallout) (UNSCEAR, 2013, 2021), the potential impact of ocean-borne radioactivity was of significant public concern as the prevailing Kuroshio Current brought the plume across the North Pacific to the coastal waters and shores of North America (World Health Organization, 2013).

On Canada's west coast, in the province of British Columbia, concern

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for the potential ramifications of ocean-borne radioactivity was particularly significant. In addition to assuring food safety, adverse economic impact to the seafood industry was a concern, particularly with regard to export. For the Indigenous communities along the coast of British Columbia, there was the additional dimension of cultural impact. This is plainly evident if we consider that, in North America, coastal Indigenous peoples consume an average of 3.1 times more seafood than others in country (Cisneros-Montemayor et al., 2016).

To address public concern, Health Canada has worked collaboratively with other governmental agencies and academic groups to directly assess the radiological impact to consumers of fish and shellfish that are very much representative of commerce and consumption along Canada's west coast. The majority of this assessment came through our participation in the Integrated Fukushima Ocean Radionuclide Monitoring (InFORM) project (https://fukushimainform.ca/). Funded by the Marine Environmental Observation, Prediction and Response (MEOPAR) network, this was a collaborative radiation monitoring initiative aimed at assessing and communicating the potential environmental impact of the Fukushima Dai-ichi nuclear accident to Canada's Pacific and Arctic Oceans. In all, from 2011 to 2018, Health Canada has analyzed 621 fish and shellfish samples from the west coast of Canada for radioactive content (Government of Canada, 2018, 2019). Those results are presented here, with focus on the radiocesium (i.e. ^{134}Cs + ^{137}Cs) content and the subsequent health impact assessment.

2. Materials and methods

2.1. Sample collection

From 2011 to 2012, fish samples including Pacific hake (*Merluccius productus)* and Alaska pollock (*Gadus chalcogrammus*) were collected in collaboration with the Canadian Food Inspection Agency (CFIA) (Government of Canada, 2018). In 2013, fish samples including Pacific halibut (*Hippoglossus stenolepis*), sablefish (*Anoplopoma fimbria*) and spiny dogfish (Squalus acanthias) were collected in collaboration with Fisheries and Oceans Canada (Chen et al., 2015). From 2014 to 2018, fish species including Chinook salmon (*Oncorhynchus tshawytscha*), Chum salmon (*Oncorhynchus keta*), Coho salmon (*Oncorhynchus kisutch*), Pink salmon (*Oncorhynchus gorbuscha*), Sockeye salmon (*Oncorhynchus nerka*), Steelhead trout (*Oncorhynchus mykiss*) and Atlantic salmon (*Salmo salar*) along with shellfish samples including mussels (*Mytilus edulis*, *Mytilus galloprovincialis x Mytilus edulis*), oysters (*Crassostrea gigas*), clams (*Venerupis philippinarum*) and scallops (*Patinopecten yessoensis*) were collected through our participation with the InFORM project. Most of the InFORM fish and shellfish samples were collected and donated by 13 Indigenous communities along the west coast of Canada.

Adult salmon and steelhead trout were collected by net during their upstream migration in their natal rivers and lakes by Indigenous communities ranging from southern British Columbia to the Yukon River in the Yukon (Fig. 1a). As these fish gain the bulk of their mass (>95%) in the North Pacific Ocean and the Bering Sea and cease to feed during their subsequent freshwater migration (Groot and Margolis, 1991), it is assumed that radiocesium was primarily accumulated during their marine life. Shellfish (mussels, oysters, clams and scallops) were collected by hand from six sites in 2016 and three sites in 2017 across British Columbia, from Prince Rupert to the west and east coasts of Vancouver Island (Fig. 1b).

2.2. Sample preparation

For the purpose of addressing radiological concerns about consumption, only the edible portion of collected samples was of interest. Samples were received and processed at the Pacific Biological Station and St. Andrews Biological Station of Fisheries and Oceans Canada as well as the University of Ottawa. Fish samples were processed for the removal of skin and bone. Whole-body tissues of mussels, oysters, and clams were processed as typical edible portions of shellfish, whereas for scallops only the typically consumed adductor muscle was processed. In addition, shells of the individual bivalves sampled were processed for analysis to determine if radiocesium had accumulated in the shell structure. Processed samples were sent to Health Canada's Radiation Protection Bureau and measured individually for radioactive content using gamma spectrometry.

Fresh fish/shellfish were packed into Parkway jars (58.0 × 66.6 mm clarified polypropylene container, Parkway Plastics, Inc.). The typical fish/shellfish tissue sample for analysis ranged from 0.10 to 0.17 kg (fresh weight, fw) with a water content of 70–80%. Selected fish samples, based on the potential observance of ^{137}Cs in abbreviated measurements, were freeze-dried (Labconco FreeZone) over several days. The dried fish samples were subsequently homogenized using a food processor while shells were homogenized using a jaw crusher (Retsch BB50). Once homogenized, these samples were packed into Parkway jars (noted above) to render well-defined sample geometries. Both the food processor and jaw crusher were decontaminated (Radiacwash, Biodex Medical Systems) and rinsed thoroughly between sample treatments.

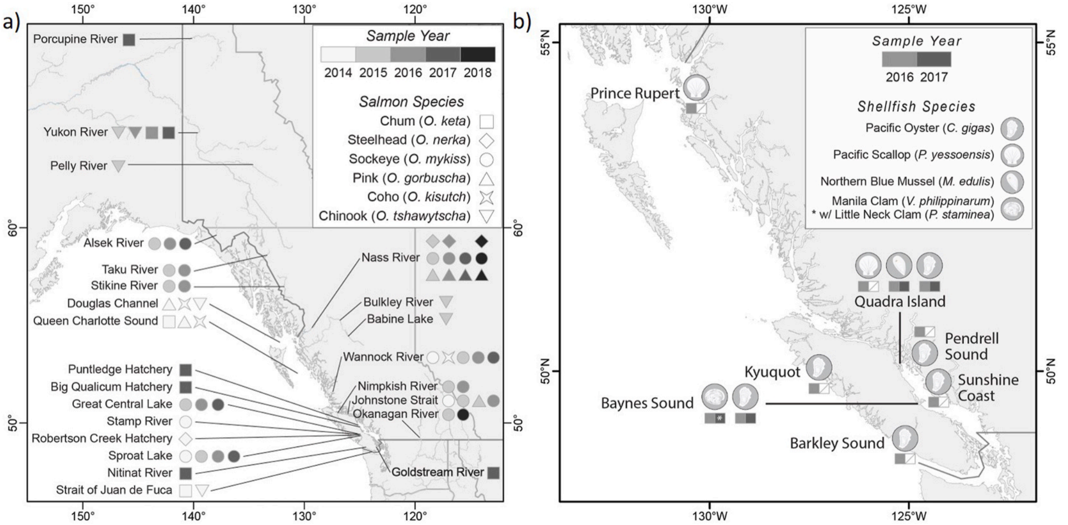

Fig. 1. (a) Map of fish samples collected from 2014 to 2018 through the InFORM project. (b) Map of shellfish (bivalves) collected in 2016 and 2017.

2.3. Sample measurement - gamma spectrometry

Fresh fish/shellfish samples were individually counted on a Gamma Analyst spectrometer equipped with a high-purity germanium (HPGe) detector (model BE5030, Mirion Technologies, Inc.) and cryostat (model 7915-GA/ULB, Mirion Technologies, Inc.). Counting times were typically 6 h, but were of shorter duration (2–4 h) in earlier years. These counting times were selected to effectively screen samples for the presence of ^{137}Cs, as those with detectable ^{137}Cs would be the best candidates to detect ^{134}Cs upon further analysis. The HPGe crystal dimensions were 80 mm (diameter) by 30 mm (length) with an energy resolution (full-width half-maximum, FWHM) of 2.2 keV at 1332.5 keV. The cryostat window was 0.6 mm thick and made of carbon composite material while the endcap and crystal holder were composed of high-purity aluminum and copper, respectively.

Fish samples with detectable ^{137}Cs on initial screening were subsequently freeze-dried, homogenized and recounted for 336 h (two weeks) using an HPGe detector (model BE5030P, Mirion Technologies, Inc.) with cryostat (model CP5-PLUS-F-RDC-4-ULB, Mirion Technologies, Inc.). Crystal dimensions were 70.5 mm (diameter) by 31.4 mm (length) with a resolution (FWHM) of 1.7 keV at 1332.5 keV. The material and dimensions of the cryostat window were the same as described above. Customized passive shielding for this detector was provided by Gamma Products Inc. (p/n G-4.2.1, Palos Hills, IL, USA). This consisted of an outer 4-inch layer of lead with a ^{210}Pb concentration of < 30 Bq·kg^{-1} (DOE Run Company), a 2-inch middle-layer of borylated (2%) polyethylene, and an inner 1-inch layer of low-background lead with a ^{210}Pb concentration of < 5 Bq·kg^{-1}.

Data was collected from 0 to 2800 keV over 16384 channels using Genie 2k v.3.4.1 (Mirion Technologies, Inc.) as the acquisition software. The analysis software used to quantify radionuclide content was UniSAMPO (v.2.67) – Shaman (v.1.2) from Baryon Oy, Finland. The true coincidence summation correction for ^{134}Cs was calculated using this software with provision of photopeak and total counting efficiencies as inputs. Spectral summation was performed according to energy with the Advanced Analysis Tool for Assessment of Monitoring Information (Aatami, v. 4.11, CTBTO) software, the output of which was analyzed using the UniSAMPO – Shaman analysis software. Spectral summation was performed for all fish and shellfish samples in a given collection year from 2014 to 2018 where the total counting time over all samples was used along with the average sample mass to derive activity concentrations for a particular sample collection period. Likewise, integrated collection times and averaged samples masses were input to the analysis software to produce corresponding Minimum Detectable Concentration (MDC) values. Background spectra were recorded (one week count time) at regular intervals to ensure that there was no detector contamination, particularly from ^{137}Cs. All spectra were corrected for background contribution using the most recent spectral file.

All calibrations were derived from, or validated with, certified reference material (CRM). Energy and peak shape (full width at half maximum) calibrations were performed prior to each sample batch counting using a multi-line CRM (Eckert & Ziegler, SRS 99616) containing 37 kBq (total) of ^{125}Sb, ^{154}Eu, and ^{155}Eu on a 60 mm diameter polypropylene filter disc. Owing to the high water content, counting efficiencies for wet samples were derived from a geometry-matched, mixed radionuclide standard in simulated water (Eckert and Ziegler, SRS 79535–411). A counting efficiency (photopeak and total) for each dried sample was simulated using Virtual Gamma Spectroscopy Laboratory software (v.1.2, CTBTO). The models developed from these simulations were validated against CRM material (dried fish meat and bone) received from the Japan Society for Analytical Chemistry through our participation in an international inter-comparison exercise to validate CRM materials from Japan for ^{90}Sr, ^{134}Cs, ^{137}Cs and ^{40}K (Miura et al., 2018).

All activities are decay corrected to the time of sample collection and are presented with an accompanying experimental uncertainty that was derived by combining, in quadrature, contributions from counting statistics, counting efficiency, and sample weighing. The experimental uncertainty in this manuscript is reported at a coverage factor of $k = 1$.

3. Results

3.1. Sample screening and short-duration gamma spectrometric analysis

As summarized in Table 1, the vast majority of the ^{137}Cs and ^{134}Cs levels were found to be below the MDC which was typically 0.7–1.0 Bq·kg^{-1} fw for a 6 h counting. For all samples collected from 2014 to

Table 1

Radiocesium and ^{40}K concentrations (averaged, fresh weight) in fish and shellfish samples collected from 2011 to 2018 along the west coast of Canada.

Year	Sample number	Sample	^{137}Cs (Bq·kg^{-1})	^{134}Cs (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})
2011	12	Salmon[a]	ND	ND	137 ± 25
2012	20	Hake, pollack[a]	ND	ND	148 ± 19
2013	28	Halibut, sablefish, spiny dogfish[a]	ND	ND	212 ± 29
2014	89	Chinook, chum, coho, pink, sockeye, steelhead[b]	ND	ND	139 ± 22
2015	156	Chinook, chum, pink, sockeye, steelhead[b]	0.51 ± 0.22 (7)[a]	ND	125 ± 19
2016	123	Chinook, chum, pink, sockeye, steelhead[b]	0.53 ± 0.20 (9)[a]	ND	118 ± 14
	10	Shellfish – tissue: northern blue mussel, blue mussel hybrid, pacific oyster, pacific scallop, manila clam[b]	ND	ND	88 ± 27
	10	Shellfish – shells: northern blue mussel, blue mussel hybrid, pacific oyster, pacific scallop, paradise oyster, manila clam[c]	ND	ND	ND
2017	115	Chum, pink, sockeye[b]	0.84 ± 0.28 (3)[a]	ND	127 ± 14
	12	Shellfish – tissue: little neck clam, manila clam, northern blue mussel, pacific oyster[b]	ND	ND	81 ± 7
	6	Shellfish – shells: little neck clam, manila clam, northern blue mussel, pacific oyster[c]	ND	ND	8.1 ± 3.1 (2)[a]
	8	Atlantic salmon[b,d]	ND	ND	115 ± 7
2018	40	Pink, sockeye, steelhead[b]	ND	ND	145 ± 9

Error was derived by combining, in quadrature, contributions from counting statistics, counting efficiency, and sample weighing. It is presented as an averaged value, at a coverage factor of $k = 1$.

*()denotes the number of samples comprising the averaged value. Where absent, the value pertains to all samples for the given year and sample type.

[a] MDC (2–4 h counting, Bq·kg^{-1} fw): 1.5–2.7, ^{137}Cs; 1.5–3.0, ^{134}Cs; 15–40, ^{40}K.

[b] MDC (6 h counting, Bq·kg^{-1} fw) 0.7–1.5, ^{137}Cs; 0.6–1.5, ^{134}Cs; 10–24, ^{40}K.

[c] MDC (6hr counting, Bq·kg^{-1} fw) 0.5–0.9, ^{137}Cs; 0.5–1.0, ^{134}Cs; 6–13, ^{40}K.

[d] Atlantic salmon was harvested from the Miramichi River in New Brunswick, Canada.

2018 through the InFORM project, ^{137}Cs was detected in a total of 19 fish samples. These samples were further investigated to attain both a higher-fidelity measurement of ^{137}Cs and to reveal the potential presence of the shorter-lived, and hence Fukushima derived, ^{134}Cs.

For shellfish samples harvested in 2016 and 2017, both meat and shells were separately measured; however, no radiocesium, or any other anthropogenic gamma-emitting radionuclides, were found. In fact, the naturally occurring ^{40}K was detected in only two shell samples, albeit with tenuous identifications owing to their proximity to the MDC and the potential for unaccounted background fluctuations of ^{40}K.

Finally, for reference, eight Atlantic salmon samples were collected from Canada's east coast (Miramichi River, New Brunswick) and analyzed in 2017. Individually, no radiocesium or other anthropogenic radionuclides were detected, as was the case for the vast majority of samples (621) screened from the west coast of Canada.

3.2. Extended gamma spectrometric analysis

Radiocesium (^{134}Cs and ^{137}Cs) concentrations are given in Table 2 on both fresh and dry weight (dw) bases, along with ^{40}K concentrations on a fresh weight basis, for the 19 fish samples with detectable ^{137}Cs on initial screening that were subsequently freeze-dried, homogenized and recounted for an extended time (336 h). The water content of fish and shellfish tissue samples was typically between 70 and 80%. The average ^{137}Cs content and associated uncertainty for these 19 samples was found to be 0.29 ± 0.02 Bq·kg^{-1} fw, spanning the range of 0.17–0.53 Bq·kg^{-1} fw, while the average content of ^{40}K was 128 ± 6 Bq·kg^{-1} fw. Of these 19 samples, only two showed trace levels of contamination from the Fukushima accident, evidenced by the detection of the relatively short-lived ^{134}Cs (half-life 2.07 yr). These samples were collected from the Okanagan and Nass Rivers of British Columbia on July 12, 2015 and August 23, 2016 and were found to contain 0.07 ± 0.02 Bq·kg^{-1} fw and 0.07 ± 0.01 Bq·kg^{-1} fw of ^{134}Cs, respectively. Notably, these samples also had the highest levels of ^{137}Cs, with 0.53 ± 0.04 Bq·kg^{-1} fw and 0.51 ± 0.03 Bq·kg^{-1} fw observed for collection years 2015 and 2016, respectively. Given that the ratio of ^{134}Cs–^{137}Cs upon release from the Fukushima Dai-ichi accident was 1:1, as measured in the atmosphere (Hoffman et al., 2013; Friese et al., 2013; Health Canada, 2015), soil (Tagami et al., 2011; Yamaguchi, 2011), water (Buesseler et al., 2012; Honda et al., 2012; Kamenik et al., 2013; Sakaguchi et al., 2012) and vegetation (Cook et al., 2015), we can estimate that the global fallout contribution to the measured ^{137}Cs in these two samples was 0.26 ± 0.08 Bq·kg^{-1} fw (2015) and 0.12 ± 0.02 Bq·kg^{-1} fw (2016) at the time of sample collection. These represent, respectively, 49 ± 14% and 24 ± 4% of the measured ^{137}Cs content. With respect to the size of the fish in which ^{134}Cs was detected, the fork length (distance from the snout to the fork of the tail) of the Sockeye salmon harvested in 2015 was 465 mm while the ten Sockeye salmon samples comprising the collected batch had a mean fork length of 487 mm and range of 455–525 mm. For the Pink salmon harvested in 2016, the fork length was found to be 520 mm while the ten Pink salmon samples comprising the collected batch had a mean fork length of 514 mm and range of 480–545 mm.

3.3. Gamma spectral summation analysis

To expand greatly the detection sensitivity of the initial screening measurements for fresh samples (6 h each), a spectral summation (Hoffman, 2016) was performed for all fish and shellfish samples in a given collection year from 2014 to 2018. The spectral summation data for fish harvested from the west coast is presented in Table 3, while spectral summation data for shellfish (west coast) and Atlantic salmon is presented in Table 4. The summation spectra for all fish samples collected from the west coast of Canada are depicted in Fig. 2, where each has been normalized to the number of samples collected for the corresponding collection period.

For fish samples, the average ^{137}Cs content determined by spectral summation was 0.21 ± 0.02 Bq·kg^{-1} fw, spanning a range of 0.18–0.25 Bq·kg^{-1} fw. While there was no discernible ^{134}Cs from spectral summation, the levels of ^{137}Cs were found to be essentially invariant, within experimental error. No other anthropogenic radionuclides characterized by gamma emission were detected. Naturally occurring, gamma-emitting nuclides were dominated by ^{40}K, with an average content in fish of 133 ± 5 Bq·kg^{-1} fw.

For shellfish harvested from Canada's west coast in 2016 and 2017, spectral summation of fresh weight samples (tissue and shell, respectively) yielded no detectable radiocesium or any other anthropogenic anomalies. However, it is notable that spectral summation, with background correction, gave no observable ^{40}K in the shells measured. This

Table 2
Radiocesium and ^{40}K concentrations (dry and fresh weight) determined from long counting (336 h) in 19 fish samples with detectable ^{137}Cs upon initial screening.

Sample	Date collected	location[d]	^{137}Cs (Bq·kg^{-1}dw)[a]	^{134}Cs (Bq·kg^{-1} dw)[a]	^{137}Cs (Bq·kg^{-1} fw)[b]	^{134}Cs (Bq·kg^{-1} fw)[b]	^{40}K (Bq·kg^{-1} fw)[b]
Steelhead trout	2015-08-20	Nass River	0.54 ± 0.05	ND	0.19 ± 0.02	ND	136 ± 7
Steelhead trout	2015-08-20	Nass River	0.73 ± 0.07	ND	0.25 ± 0.02	ND	128 ± 6
Sockeye salmon	2015-09-13	Alsek River	0.63 ± 0.04	ND	0.20 ± 0.01	ND	129 ± 6
Chinook	2015-08-28	Yukon River	0.93 ± 0.11	ND	0.17 ± 0.02	ND	111 ± 6
Sockeye salmon	2015-07-12	Okanagan River	1.96 ± 0.13	0.26 ± 0.07	0.53 ± 0.04	0.07 ± 0.02	130 ± 6
Sockeye salmon	2015-08-20	Nass River	0.71 ± 0.08	ND	0.19 ± 0.02	ND	129 ± 6
Chum	2015-09-05	Yukon River	1.37 ± 0.10	ND	0.37 ± 0.03	ND	132 ± 8
Sockeye salmon	2016-07-27	Sproat Lake	1.11 ± 0.08	ND	0.28 ± 0.02	ND	123 ± 6
Pink salmon	2016-08-23	Nass River	1.78 ± 0.11	ND	0.38 ± 0.02	ND	118 ± 6
Pink salmon	2016-08-23	Nass River	2.35 ± 0.14	0.31 ± 0.05	0.51 ± 0.03	0.07 ± 0.01	125 ± 6
Pink salmon	2016-08-23	Nass River	1.31 ± 0.09	ND	0.39 ± 0.03	ND	143 ± 7
Steelhead trout	2016-08-23	Nass River	0.78 ± 0.06	ND	0.28 ± 0.02	ND	148 ± 3
Steelhead trout	2016-08-23	Nass River	1.61 ± 0.10	ND	0.44 ± 0.03	ND	149 ± 3
Sockeye salmon	2016-08-05	Alsek River	0.83 ± 0.07	ND	0.18 ± 0.02	ND	127 ± 6
Sockeye salmon	2016-08-04	Stikine River	1.01 ± 0.08	ND	0.23 ± 0.02	ND	123 ± 6
Sockeye salmon	2016-07-28	Wannock River	0.94 ± 0.06	ND	0.26 ± 0.02	ND	127 ± 6
Sockeye salmon	2017-09-20	Wannock River	0.74 ± 0.07	ND	0.20 ± 0.02	ND	134 ± 7
Chum	2017-07-30	B.Q. Hatchery[c]	1.52 ± 0.12	ND	0.27 ± 0.02	ND	100 ± 5
Pink salmon	2017-08-18	Nass River	1.18 ± 0.09	ND	0.27 ± 0.02	ND	113 ± 6

Error was derived by combining, in quadrature, contributions from counting statistics, counting efficiency, and sample weighing. It is presented at a coverage factor of $k = 1$.

[a] MDC (Bq·kg^{-1} dw): 0.19–0.51, ^{137}Cs; 0.17–0.55, ^{134}Cs; 2.4–5.6, ^{40}K.

[b] MDC (Bq·kg^{-1} fw) 0.05–0.14, ^{137}Cs; 0.05–0.14, ^{134}Cs; 0.6–1.1, ^{40}K.

[c] Big Qualicum Hatchery.

[d] British Columbia, Canada.

Table 3
Spectral summation results for radioactive content of all fish samples per collection year.

Year	^{137}Cs (Bq·kg^{-1})	MDC (Bq·kg^{-1})	^{134}Cs (Bq·kg^{-1})	MDC (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})	MDC (Bq·kg^{-1})
2014	0.21 ± 0.02	0.08	ND	0.09	137 ± 4	1
2015	0.18 ± 0.02	0.06	ND	0.05	125 ± 8	1
2016	0.19 ± 0.01	0.05	ND	0.05	123 ± 4	1
2017	0.25 ± 0.02	0.09	ND	0.07	137 ± 4	1
2018	0.20 ± 0.03	0.13	ND	0.12	145 ± 5	2

Error was derived by combining, in quadrature, contributions from counting statistics, counting efficiency, and sample weighing. It is presented at a coverage factor of $k = 1$.

Table 4
Spectral summation results for radioactive content of shellfish and Atlantic salmon[a].

Sample	Year	^{137}Cs (Bq·kg^{-1})	MDC (Bq·kg^{-1})	^{134}Cs (Bq·kg^{-1})	MDC (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})	MDC (Bq·kg^{-1})
Shellfish (tissue)	2016	ND	0.23	ND	0.20	119 ± 4	3
	2017	ND	0.20	ND	0.17	87 ± 3	3
Shellfish (shells)	2016	ND	0.14	ND	0.13	ND	2
	2017	ND	0.20	ND	0.18	ND	2
Atlantic salmon	2017	0.20 ± 0.06	0.26	ND	0.20	119 ± 5	3

[a] Error was derived by combining, in quadrature, contributions from counting statistics, counting efficiency, and sample weighing. It is presented at a coverage factor of $k = 1$.

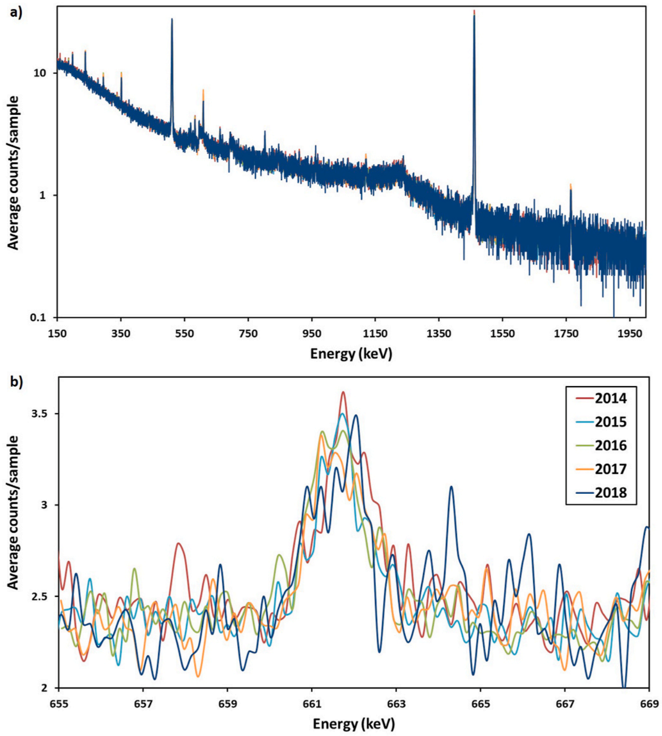

Fig. 2. Spectral summation of fish samples from 2014 to 2018, normalized to sample number. (a) Overlay, 150–2000 keV (b) Overlay with focus on the principal emission line for ^{137}Cs at 661.7 keV.

serves as a footnote to the two tenuous observations of ^{40}K in shells from individual sample analysis (Table 1). The ^{40}K found in tissue samples (119 ± 4 and 87 ± 3 Bq·kg^{-1} fw for 2016 and 2017, respectively) was comparable to that observed in fish samples.

Spectral summation of the eight Atlantic salmon samples received from Canada's east coast (Miramichi River) indicated that the level of ^{137}Cs (0.20 ± 0.06 Bq·kg^{-1} fw, Table 4) was quite similar to that determined in fish harvested from the west coast (Table 3), considering the difference in the determined measurement uncertainty and detection limit owing to the relatively small sample size. The average ^{40}K content was found to be 119 ± 5 Bq·kg^{-1} fw, consistent with that found in both fish and shellfish tissue samples from Canada's west coast.

4. Discussion

4.1. Radiocesium in fish

After the nuclear disaster of 2011 at the Fukushima Dai-ichi power plant, oceanic dispersion models predicted that the subsequent waterborne, radioactive plume would arrive in Canadian coastal waters, off the coast of British Columbia, between years 2013–2014 with peak ^{137}Cs concentrations reaching 3–5 Bq·m^{-3} between years 2015–2017, similar to observed levels in 1980 (Rossi et al., 2013). By contrast, time series measurements of seawater at points extending 1500 km from the coast of British Columbia revealed that the plume arrived in June 2013 with ^{137}Cs concentrations in surface water (i.e. upper 150 m of the water column) reaching ~2 Bq·m^{-3} by February 2014, roughly double that from global fallout contributions (Smith et al., 2015). By 2015 and into early 2016, the ^{137}Cs in surface water reached a maximum of 6–8 Bq·m^{-3} followed by a declination to global fallout levels of ^{137}Cs observed in the 1970's (Smith et al., 2017).

Geographic variations in ^{137}Cs are generally reflected in fish (Heldal et al., 2015, 2019). However, despite the changes in ^{137}Cs observed in the waters of the North Pacific since the Fukushima accident, it is apparent from this study that the radiocesium content of migratory fish from the eastern North Pacific has remained relatively invariant. This is perhaps a reflection of the vast distances travelled as these fish fed and developed before heading inland to spawn, such that their integrated exposure is relatively constant compared to local variations as the radioactive ocean plume migrated eastward from Japan. Long gamma-counting measurements of 19 homogenized samples flagged upon screening yielded a mean ^{137}Cs content of 0.29 ± 0.02 Bq·kg^{-1} fw while the mean annual concentration of ^{137}Cs from spectral summation (2014–2018) was found to be 0.21 ± 0.02 Bq·kg^{-1} fw. With respect to ^{40}K content, these approaches yielded mean values of 128 ± 6 Bq·kg^{-1} fw and 133 ± 5 Bq·kg^{-1} fw, respectively. That there is good agreement in the determined ^{40}K content lends confidence to the respective methodologies employed. These observations are, in turn, consistent with the findings of earlier reports from Domingo et al. for relatively small sample sizes of salmon harvested from Canada's west coast. Of thirteen salmon harvested from two locations, Domingo et al. (2018) had detected ^{137}Cs in seven Chinook salmon samples with a mean ^{137}Cs content of 0.20 ± 0.03 Bq·kg^{-1} fw and 0.23 ± 0.03 Bq·kg^{-1} fw (±1σ) in 2013 and 2014, respectively, and with an average ^{40}K content of 106 ± 13 Bq·kg^{-1} fw (±1σ). Likewise, Domingo et al. (2017) had detected ^{137}Cs in half of the ten Sockeye salmon harvested from a single location in 2014, with activity ranging from 0.23 to 1.43 Bq·kg^{-1} dw. This range is reasonably consistent with that presented in Table 2 (0.54–1.96 Bq·kg^{-1} dw) for the dried and homogenized samples which underwent extended counting, bearing in mind that they were selected upon screening for ^{137}Cs content and that they comprise a larger number of samples collected over a greater time span (2015–2017) and diversity of collection sites.

The direct observation of the relatively short lived, Fukushima-derived ^{134}Cs was made on only two occasions in the migratory fish sampled (Table 2). These fish samples also had the highest concentrations of ^{137}Cs. This is unsurprising, and in fact reassuring, considering that both isotopes are chemically identical, that both isotopes were released from the Fukushima Dai-ichi accident in nearly equal quantities, and that ^{137}Cs from global fallout is homogenously distributed in the surface water of the North Pacific (Hirose and Aoyama, 2003). Physiologically, there is nothing obviously remarkable about these two fish; their sizes, as indicated by fork length measurements, were typical of their respective type harvested at the same time and location. However, it is noteworthy that these observations, along with the majority of fish samples found to contain radiocesium, come from the 2015 and 2016 sample collection period that coincided with the observed ^{137}Cs peak in coastal waters. Perhaps more importantly, these detections enabled the estimation of global fallout contributions to the measured ^{137}Cs in these samples, with 0.26 ± 0.08 Bq·kg^{-1} fw (49 ± 14%) and 0.12 ± 0.02 Bq·kg^{-1} fw (24 ± 4%) found for samples collected on July 12, 2015 and August 23, 2016, respectively. To add confidence to these estimations, we can derive the corresponding ^{137}Cs concentration in seawater from global fallout for comparison to established values using an appropriate concentration factor (CF). For ^{137}Cs in marine fish, this is simply the ratio of ^{137}Cs in the tissue of the fish (Bq·kg^{-1}, fresh weight) to the ^{137}Cs in seawater (Bq·L^{-1}). A CF value of 100 is recommended by the IAEA for cesium in marine, surface water fish (IAEA, 2004). Applying this factor to the determined global fallout contributions above, and correcting to the date of the Fukushima accident (March 11, 2011), we derive seawater ^{137}Cs concentrations of 2.9 ± 0.8 Bq·m^{-3} and 1.4 ± 0.2 Bq·m^{-3}. These are in good agreement with the known ^{137}Cs concentration of surface water in the North Pacific (1.5–2.0 Bq·m^{-3}) just prior to the Fukushima accident (Kaeriyama, 2015; Hirose and Aoyama, 2003). Bearing in mind that there were only two detections of ^{134}Cs, the subsequent estimations of contribution from global fallout ^{137}Cs are nevertheless distinct. This can be reconciled by considering that fish can have different oceanic journeys that lead to varying exposures to ocean borne radioactivity, particularly where different fish species, collected at different times and locations, are concerned.

The determined radiocesium content in migratory fish from Canada's west coast is seemingly comparable to ^{137}Cs levels detected in Atlantic salmon collected from Canada's east coast in 2017. Although consideration must be given to the comparatively small sample size and to the limitations that imposes, this data does agree well with an earlier report from Carvalho et al. (2011) on the radionuclide content of deep-sea and coastal fish harvested from the North Atlantic. In that report, fish at a similar trophic level to those reported here (e.g. cod, red fish, halibut, plaice) were found to have ^{137}Cs in the range of 0.10–0.26 Bq·kg^{-1} fw, decay corrected to the collection time reported here for the Atlantic salmon (2017). Further to this point is a recent report from Heldal et al. (2019) that found farmed salmon to contain ^{137}Cs at a level (0.05–0.25 Bq·kg^{-1} fw) similar to that determined in wild fish harvested from Norwegian waters from 2015 to 2017.

While the numbers derived from spectral summation and high-fidelity, extended counting experiments pertain to collection years 2014–2018, we expect that the radiocesium content of fish prior to 2014 (and post-Fukushima accident) to be consistent with our findings. This is supported by the measurement of 60 samples from 2011 to 2013 (Table 1) under similar to identical conditions that returned no observed anthropogenic radionuclide detections. Further to this point, we have insight into the radioactive content of marine fish in Canada prior to the Fukushima accident from our participation in the Canadian Total Diet Study (Health Canada). From its start in 2000–2010, radiocesium was not observed under measurement conditions similar or same to those employed here.

4.2. Radiocesium in shellfish

Shellfish, specifically bivalves such as scallops, mussels, and oysters have been used as sensitive markers for environmental impact assessment from pollution, including radionuclides, for some time. This is due to the large volume of water that these filter-feeders process, their sedentary nature, and the dual-mechanistic manner in which pollutants are absorbed, i.e. directly through dissolved material or through filtered particulate matter (Baltas et al., 2021; Murakami-Sugihara et al., 2021; Apeti et al., 2010; Ramu et al., 2007; Briant et al., 2017). In this way, detailed information can be obtained about the contamination of a specific locale, as has been utilized in Japan post-Fukushima (Murakami-Sugihara et al., 2021).

In this study, bivalves obtained from aquaculture and traditional harvest locations from Canada's west coast in 2016 and 2017 had no detectable radiocesium in either the shell or flesh components. This is quite reasonable if we consider the steady-state concentration factor (CF) for ^{137}Cs in mussels, as a representative, where the CF is the ratio of ^{137}Cs in the tissue of the shellfish (Bq·kg^{-1}, fresh weight) to the ^{137}Cs in seawater (Bq·L^{-1}). Under laboratory conditions, Baltas et al. (2021) have determined the CF for large and small mussels to be 16.2 ± 4.4 and 19.5 ± 1.2, respectively. By applying the extremes of these CF values, obtained in consideration of their respective uncertainties (i.e. 11.8 and 20.7), to the range of coastal water ^{137}Cs concentration from 2014 to its peak in 2016, i.e. from ~2 to 8 Bq·m^{-3}, we can estimate that the ^{137}Cs content in the bivalves measured was in the range of 0.02–0.17 Bq·kg^{-1} fw, again assuming mussels to be representative. Only the upper limit of this range sits at the MDC obtained through spectral summation of the tissue samples provided (Table 4).

4.3. Health risk assessment and perspective

The Health Canada recommended action levels for food contaminated with ^{134}Cs or ^{137}Cs are 100 Bq·kg^{-1} for public drinking water, 300 Bq·kg^{-1} for fresh milk, and 1000 Bq·kg^{-1} for other commercial foods and beverages. By these measures, there is no radiological concern based on the measurements made.

To provide more reference, the average annual effective dose from ingestion of a unit weight (1 kg fw) of food due to ^{210}Po, a naturally occurring radionuclide, was determined to be 0.88 μSv for salmon harvested on Canada's west coast from 2013 to 2016 (Chen et al., 2019). From long-counting experiments and spectral summation, the average ^{137}Cs concentration in fish was found to be 0.29 ± 0.02 and 0.21 ± 0.02 Bq·kg^{-1} fw, respectively. Applying the committed effective dose coefficient of 0.013 μSv Bq^{-1} for ^{137}Cs (ICRP, 2012), we find corresponding annual dose estimates of 0.0038 and 0.0027 μSv from ingestion of 1 kg fw of fish, respectively. For perspective, these dose estimates are, respectively, 232 and 326 times less than the ingestion dose received from the naturally occurring ^{210}Po. The latter estimate is more representative of average exposure since it is derived from all fish samples measured.

With regard to shellfish, we have previously derived an upper limit for ^{137}Cs content of 0.17 Bq·kg^{-1} fw that also corresponds to the MDC from spectral summation. Applying the same committed dose coefficient, we find a corresponding annual dose of 0.0022 μSv from ingestion of 1 kg fw of shellfish, which is 400 times less than the dose received per kilogram from the naturally occurring ^{210}Po in fish.

From the perspective of yearly consumption, North American coastal Indigenous communities have been estimated to consume, on a per capita basis, 69 kg fw y^{-1} of seafood (Cisneros-Montemayor et al., 2016). Applying the larger determined value of ^{137}Cs in fish (0.29 ± 0.02 Bq·kg^{-1} fw) and assuming that seafood would equate solely to fish, we estimate the upper bound for annual ingestion dose to be approximately 0.26 μSv. Using the more representative value of 0.21 ± 0.02 Bq·kg^{-1} fw obtained from spectral summation, we find a corresponding annual ingestion dose of about 0.19 μSv. For perspective, the worldwide average annual effective dose from exposure to natural background radiation is 2400 μSv (UNSCEAR 2000).

5. Conclusion

From 2011 to 2018, radioactivity measurements by Health Canada of 621 fish and shellfish samples harvested from Canada's west coast were shown to be largely consistent in their radiocesium content based upon sample measurement in the 2–6 h counting time regime. To investigate more deeply in the years coinciding with and subsequent to the advent of the oceanic plume of Fukushima radioactivity to coastal waters, spectral summation has revealed that the representative concentration of ^{137}Cs in the tissue of marine fish has been essentially invariant (0.18–0.25 Bq·kg^{-1} fw) from 2014 to 2018 while that of shellfish was undetectable. Relative to the natural abundance of ^{210}Po in the same fish samples or to the annual dose exposure due to naturally occurring background radiation, it is abundantly clear that, by any metric, the radiocesium content of fish and shellfish from Canada's west coast does not constitute a health risk, despite the Fukushima Dai-ichi nuclear accident of 2011.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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