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Studying levels of Fukushima-derived radioactivity in sockeye salmon collected on the west coast of Vancouver Island

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ABSTRACT

To investigate potential radioisotope contamination from the Fukushima nuclear accident, measurements of 10 sockeye salmon (*Oncorhynchus nerka*) collected on June 21 and June 31, 2014 in the Alberni Inlet on the west coast of Vancouver Island, British Columbia, Canada were performed using low-background gamma-ray spectroscopy. Activity concentrations of the anthropogenic radioisotopes ^{134}Cs and ^{137}Cs as well as the naturally occurring radioisotope ^{40}K were measured. Detection of ^{137}Cs occurred in half of the sockeye with activity concentrations ranging from 0.23 to 1.43 Bq/kg dry weight. The ^{134}Cs isotope was detected in a single sockeye salmon with activity concentrations ($\pm\sigma$) measured in the two subsamples of 0.31(8) and 0.37(10) Bq/kg dry weight. The dose contribution from each of the measured radionuclides was calculated. In the sockeye salmon with the greatest radiocesium concentrations, the dose contribution from anthropogenic radiocesium (^{134}Cs +^{137}Cs) was found to be 450 times less than the dose from naturally occurring radionuclides in the same sample. In conclusion, the total radiocesium activity concentration in every sample is at least 500 times lower than Health Canada's action levels for radioactively contaminated food following a nuclear emergency. Assuming all seafood has as much radiocesium as the most contaminated sample measured, the added annual dose from radiocesium to an adult individual with an average Canadian level of seafood consumption would be 0.046 μSv per year.

1. Introduction

The Fukushima Nuclear Accident on March 11, 2011 raised global concerns due to the release of fission fragments and nuclear fuel into the atmosphere and Pacific Ocean. Since the initial event, there has been an international effort to both model (Behrens et al., 2012; Rossi et al., 2013; Christoudias and Lelieveld, 2013) and monitor the spread of Fukushima-derived radioisotopes across the Pacific Ocean (Buesseler et al., 2011, 2012). Two gamma-ray emitting radioisotopes commonly used to monitor the fallout from Fukushima are ^{137}Cs ($t_{1/2}$ = 30 y) and ^{134}Cs ($t_{1/2}$ = 2.1 y). The highest concentration of ^{137}Cs measured coming out of the discharge channels from Fukushima Dai-ichi Nuclear Power Plant was 86 MBq per cubic meter of water (Buesseler et al., 2011) with total discharges from the power station estimated to be as high as 22 PBq (Buesseler et al., 2012).

One of the largest persisting concerns from the public is the perceived health risk from consuming Pacific seafood which may be contaminated with Fukushima-derived radionuclides. Models projecting the path and dilution of the radioactive plume from Fukushima crossing the Pacific Ocean have suggested that it will arrive and remain at the northwest American continental shelf from 2014 to 2020. The ^{137}Cs activity concentrations in the plume during this time was calculated to range from 0.01 to 0.03 Bq/L in the ocean waters (Rossi et al., 2013). While 0.01–0.03 Bq/L is well below Canadian action levels for radiocesium contamination in commercial food and beverages (1000 Bq/kg) and public drinking water (100 Bq/L) (Health Canada, 2000) there still exists an intrinsic public fear and concern of radiation in any form.

In an effort to address the concerns over public health and safety, various radioisotope contamination studies in marine biota have been completed (Madigan et al., 2012; Fisher et al., 2013; Neville et al., 2014; Chen et al., 2014, 2015, 2016). To further the international monitoring campaign and continue to address the health concerns of the public, 10 sockeye salmon were collected in the Alberni Inlet on the west coast of Vancouver Island, British Columbia, Canada to measure their degree of Fukushima-derived (^{134}Cs + ^{137}Cs) contamination.

The measured ^{134}Cs/^{137}Cs activity ratio one month after the accident at the Dai-ichi discharge channels was measured to be 0.99(3) (Buesseler et al., 2011) though this ratio is expected to change over time as ^{134}Cs decays more quickly than ^{137}Cs. Because of its slower

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decay rate, one would expect that there should be a larger quantity of ^{137}Cs from Fukushima available for detection making it a better tracer. However, because of it's long half-life, there still remains measurable quantities of ^{137}Cs in the environment as a legacy of atmospheric nuclear weapons testing and the Chernobyl nuclear accident. In contrast, cesium-134 with its 2.1 year half-life will have completely decayed away since these nuclear weapons tests and accident. For this reason, while both ^{134}Cs and ^{137}Cs may originate from Fukushima, the ^{134}Cs isotope is the more useful indicator of contamination from Fukushima as it there is no ambiguity regarding its source.

For a simple comparison to help interpret the measured data for the public and those not accustomed to radioactivity measurements, the naturally occurring radioisotope ^{40}K in each sample was measured to provide a simple reference between the anthropogenic activity and the naturally occurring activity in each sample. For a more in depth investigation of the potential effect of consuming contaminated seafood, an upper limit of the dose contributions from each the previously listed radioisotopes was calculated. To more clearly demonstrate the health risks, the total anthropogenic radiocesium (^{134}Cs + ^{137}Cs) dose was compared with the naturally occurring dose from ^{40}K. Polonium-210, another naturally occurring radioactive material (NORM) found ubiquitously in nature, is responsible for the largest dose to individuals of the NORM isotopes from fish consumption (Chen et al., 2016). As ^{210}Po decays primarily via alpha particle emission, it was not possible to quantify its presence in each sample with the detector used in this study. Thus, data from other monitoring campaigns (Fisher et al., 2013; Chen et al., 2015, 2016) was included in the dose calculations and comparison in this study to have a clearer description of the added risk from the consumption of Pacific seafood contaminated with Fukushima-derived radioisotopes.

2. Methods

2.1. Sample collection

Ten sockeye salmon (*Oncorhynchus nerka*) were collected in July 2014 from First Nations food, social, ceremonial, and commercial fisheries in the Alberni Inlet on the west coast of Vancouver Island. After the collection of each sample, the fillet was removed and freeze-dried to both concentrate and preserve the sample. As the freeze-dried fillet were still quite large, each sample was split into 2 subsamples. Each subsample was then homogenised so that it could take the shape of the measurement vessel.

2.2. Radioisotope detection

To measure anthropogenic ^{134}Cs and ^{137}Cs as well as the NORM radioisotope ^{40}K, low-background gamma-ray spectroscopy was employed. The detector used was the Germanium detector for Elemental Analysis and Radioactivity Studies (GEARS), a low-background shielded, High-Purity Germanium (HPGe) gamma-ray detector at Simon Fraser University. This detector was previously used to monitor ^{131}I levels in rainwater and seaweed in British Columbia in the months immediately following the nuclear accident (Chester et al., 2013). The absolute efficiency of this detector has been well characterised for a point source using the technique described in Rizwan et al. (2015).

The freeze-dried and homogenised sockeye salmon subsamples were measured for 1–4 days in a 600 mL Marinelli beaker designed to surround the GEARS detector thereby increasing the detection efficiency of the measurement. To determine a correction for the absolute efficiency of GEARS while measuring samples in the non-pointlike Marinelli beaker geometry, the GEANT4 Monte Carlo simulation toolkit (Agostinelli et al., 2003; Allison et al., 2006) was utilised via the method described in Chester et al. (2013).

The data acquisition system used with GEARS was an Ortec DSPEC jr. 2.0 multichannel analyzer coupled to a computer running the Ortec Maestro32 software. Analysis of measured spectra was done with the gf3 program in the RadWare software package (Radford, 2000). An example of a measured sample spectrum can be seen in Fig. 1.

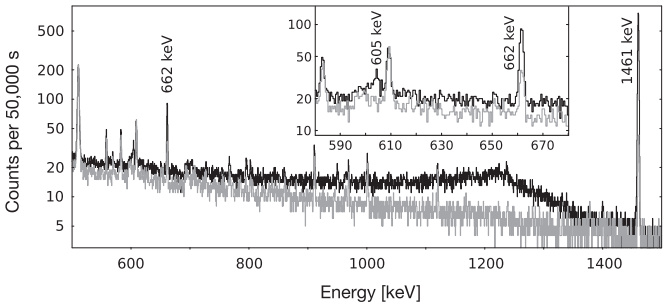

Fig. 1. A measured gamma-ray spectrum of sockeye salmon subsample 10-1 using GEARS (black) with the time scaled background spectrum (grey). The measured spectrum ranging from 500 keV to 1500 keV shows the relative size of the man-made ^{137}Cs gamma-ray peak (662 keV) relative to the NORM ^{40}K gamma-ray peak (1461 keV). The inset spectrum shows the region of interest where the ^{134}Cs (605 keV) and the ^{137}Cs (662 keV) gamma-ray peaks can clearly be observed over the background.

3. Results

The results of the radioactivity monitoring campaign are summarised in Table 1. In the cases where a radionuclide was not detected, the minimum detection limit (MDL) was calculated as defined by Cooper (1970). The measured activity concentration of naturally occurring ^{40}K ranged from 293 to 392 Bq/kg dry weight (dw) in the sockeye salmon samples. This is consistent with ^{40}K levels in other marine biota measurements (Madigan et al., 2012; Chen et al., 2015; Jenkins, 1969) validating the methods used during sample preparation and sample measurement. Detection of ^{137}Cs occurred in half of the sockeye salmon samples with observed activity concentrations decay corrected to the sample collection date, ranging from 0.23 to 1.45 Bq/

Table 1
The measurement livetime, sample mass post freeze-drying, and activity concentrations for ^{40}K, ^{134}Cs, and ^{137}Cs measured in each of the sockeye salmon subsamples. The activity concentrations ($\pm\sigma$) listed below have been decay corrected to the date of sample collection. When a radionuclide could not be detected, the minimum detection limit (MDL) was calculated via the method described in Cooper (1970) to provide an upper limit for the activity concentration of the radionuclide in the sample. MDL values are presented following a less than " < " symbol.

Sample	Livetime (s)	Mass (kg)	Activity Concentration (Bq/kg dw)		
			^{40}K	^{134}Cs	^{137}Cs
Sockeye 1–1	87,453	0.1458265(1)	375(6)	<0.26	0.39(14)
Sockeye 1–2	94,012	0.1014464(1)	360(7)	<0.35	0.57(20)
Sockeye 2–1	155,435	0.0913364(1)	372(6)	<0.30	0.37(15)
Sockeye 2–2	105,383	0.0664156(1)	348(8)	<0.52	<0.45
Sockeye 3–1	96,891	0.1692489(1)	354(5)	<0.24	0.23(11)
Sockeye 3–2	244,682	0.1829204(1)	307(3)	<0.13	<0.12
Sockeye 4–1	147,090	0.0787534(1)	293(6)	<0.32	<0.28
Sockeye 4–2	92,417	0.0960321(1)	327(7)	<0.37	<0.32
Sockeye 5–1	87,542	0.1016158(1)	298(7)	<0.32	<0.29
Sockeye 5–2	185,935	0.0886141(1)	298(5)	<0.26	<0.24
Sockeye 6–1	86,343	0.1177219(1)	336(7)	<0.32	<0.27
Sockeye 6–2	87,047	0.0989486(1)	310(7)	<0.38	<0.32
Sockeye 7–1	158,162	0.0978163(1)	326(6)	<0.29	<0.27
Sockeye 7–2	93,065	0.1008913(1)	352(7)	<0.37	<0.32
Sockeye 8–1	179,718	0.1012463(1)	362(5)	<0.26	0.64(15)
Sockeye 8–2	87,040	0.0940802(1)	354(8)	<0.43	0.58(20)
Sockeye 9–1	164,214	0.1002187(1)	302(5)	<0.24	<0.22
Sockeye 9–2	86,402	0.1128507(1)	324(7)	<0.35	<0.31
Sockeye 10–1	319,767	0.1062626(1)	392(5)	0.31(8)	1.45(12)
Sockeye 10–2	271,032	0.0899201(1)	391(6)	0.37(10)	1.36(15)