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A study on the levels of radioactivity in fish samples from the experimental lakes area in Ontario, Canada

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ABSTRACT

To better understand background radiation levels in country foods, a total of 125 fish samples were collected from three lakes (Lake 226, Lake 302 and Lake 305) in the Experimental Lakes Area (ELA) in Ontario of Canada during the summer of 2014. Concentrations of naturally occurring radionuclides (^{226}Ra, ^{210}Pb and ^{210}Po) as well as anthropogenic radionuclides (^{134}Cs and ^{137}Cs) were measured. This study confirmed that ^{210}Po is the dominant contributor to radiation doses resulting from fish consumption. While concentrations of ^{210}Pb and ^{226}Ra were below conventional detection limits, ^{210}Po was measured in almost all fish samples collected from the ELA. The average concentration was about 1.5 Bq/kg fresh weight (fw). None of the fish samples analysed in this study contained any detectable levels of ^{134}Cs. An average ^{137}Cs level of 6.1 Bq/kg fw was observed in freshwater fishes harvested in the ELA, almost twice that of samples measured in the National Capital Region of Canada in 2014 and more than 20 times higher than the levels observed in marine fish harvested from the Canadian west coast in 2013 and 2014. However, it is important to note that the concentrations of ^{137}Cs in fish samples from these inland lakes are considered very low from a radiological protection perspective. The resulting radiation dose for people from fish consumption would be a very small fraction of the annual dose from exposure to natural background radiation in Canada. The results indicate that fishes from inland lakes do not pose a radiological health concern.

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1. Introduction

Releases of radioactive contaminants, especially the long-lived radioactive ^{137}Cs, into the Pacific Ocean following the Fukushima-Daiichi nuclear accident in 2011 have raised public concern about the safety of consuming seafood. To address this, many fish samples harvested from the Canadian west coast have been analysed for radioactive caesium (Health Canada 2015; Chen et al. 2015a). So far, measurements of radioactive caesium in marine fish from the Pacific Ocean have shown that levels of ^{134}Cs and ^{137}Cs were below the detection limit of about 1 Bq/kg fresh weight (fw). Since radioactive caesium can be dispersed over great distances by atmospheric transport (UNSCEAR 2014), it is also in the public interest to know the levels of radioactive caesium in inland freshwater fish and the potential long-range impact from the Fukushima accident.

To address ongoing public concerns for the safety of fish consumption, limited fish samples from inland waterbodies in the National Capital Region (NCR) of Canada were analysed for radioactivity (Chen et al. 2015b). These NCR results showed that none of the fish samples analysed contained any detectable levels of ^{134}Cs (an indicator of recent releases from Fukushima accident). For environmental ^{137}Cs, an average level of 3.7 Bq/kg fw was observed in freshwater fishes. It is well known that large amounts of radioactive caesium were produced during atmospheric nuclear weapons tests conducted in the 1950s and 1960s. As a result of atmospheric testing and radioactive fallout, radioactive caesium was dispersed and deposited worldwide. Due to its long half-life of 30 years, small but measurable levels of ^{137}Cs continue to be reported in various environmental samples decades after the original releases.

To further address the public concerns for radioactivity in fish

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from inland waterbodies, a total of 125 fish samples were collected from three lakes in the Experimental Lakes Area (ELA, northwestern Ontario, Canada) during the summer of 2014 where data of radioactivity concentrations in fish measured 35 years ago were available (Elliot et al. 1981). Northwestern Ontario of Canada, a popular fishing destination for both Canadians and American tourists (DFO 2010). Historical data combined with current levels of radioactive caesium also permitted the estimation of the effective half-life of ^{137}Cs in a northern freshwater environment.

To put radiation doses received from radioactive caesium intake into perspective, naturally occurring radionuclides commonly found in fish were also measured in this and the previous study (Chen et al. 2015b). In the environment, most naturally occurring radionuclides come from the uranium decay series. Among various radionuclides in the uranium decay series, ^{210}Po, ^{210}Pb and ^{226}Ra are commonly found in fish in varying concentrations. The NCR fish study confirmed that ^{210}Po is the main contributor to radiation dose resulting from fish consumption; it also showed that small fish may be more capable of incorporating ^{210}Po into muscle. In the current ELA fish study, activity concentrations of naturally occurring radionuclides (^{226}Ra, ^{210}Pb, and ^{210}Po) were measured across a size range from multiple species to further evaluate the size dependency of ^{210}Po.

The primary objective of this study was to address public concerns for the safety of fish consumption from radiation doses resulting from naturally occurring and anthropogenic radionuclides in fish in Northwestern Ontario, and compare patterns observed (estimated doses and size dependence of radionuclides) to other inland locations. A secondary objective was to estimate the effective half-life of ^{137}Cs in typical Northwestern Ontario lakes.

2. Materials and methods

2.1. Fish samples

Fish were collected from three lakes in the ELA during the summer of 2014: Lake 305 (11 Northern Pike (*Esox lucius*) and 20 Lake Whitefish (*Coregonus clupeaformis*)); Lake 302 (20 White Sucker (*Catostomus commersonii*) and 40 Lake Whitefish); and Lake 226 (34 Lake Whitefish). Information for individual fish from which samples are derived is given in Table A of the appendix. The whitefish from Lake 302 and Lake 226 are of special interest because historical measurements of ^{137}Cs and ^{226}Ra concentrations are available for comparison (Elliott et al. 1981). Samples from lakes 226 and 305 were obtained opportunistically as they coincided with destructive sampling planned on these lakes for other purposes. Otherwise, species were selected as being representative of those present in lakes in the region, and common in the lakes sampled during the current study. Northern Pike and Lake Whitefish are the focus of both recreational and commercial fishing in North America and Canada specifically (DFO 2010; Kinnunen 2003), and White Sucker continue to be an important species of importance in aboriginal fisheries (Cooke and Murchie, 2013).

For the purpose of addressing radiological concerns about fish consumption, only the edible portion of fish was evaluated. Therefore, skin and bone were first separated from the flesh before a homogenised sample was prepared for determination of radioactivity.

2.2. Gamma counting for radioactive caesium

To determine levels of radioactive caesium in fish samples, gamma counting was used. Homogenised samples of about 170 g were fully packed in PVC vials (60 mm in diameter and 55 mm in height), and then scanned for gamma emitting radionuclides by counting on a Compton suppressed gamma spectrometer (ORTEC) (Zhang et al. 2013). Briefly, the spectrometer consists of an n-type GMX high purity germanium (HPGe) coaxial primary detector, crystal diameter of 66.2 mm and length 69.0 mm, with a carbon fibre end cap and a guard detector consisting of a 9″×9″ NaI(Tl) annulus with four photomultiplier tubes (PMT) and a 3″×3″ NaI(Tl) plug with one PMT.

The list-mode data-acquisition system for the spectrometer utilizes all-digital electronics, based on the X-Ray Instrumentation Associates (XIA LLC) Digital Gamma Finder (DGF)/Pixie-4 software and card package. The use of a list-mode data-acquisition technique enabled simultaneous determination of ^{134}Cs and ^{137}Cs activity concentrations using a single measurement by coincidence and anticoincidence mode respectively.

The fish samples were counted for 24 h. Certified mixed radionuclide standards were used to determine the respective counting efficiencies on the Compton spectrometer. A standard consisting of a Parkway jar containing a solid with a density of 1.15 g/cm^3 simulating water (Eckert and Ziegler Analytics, SRS: 79535-411) was used for the calibrations. The detection limits for radioactive caesium (^{134}Cs and ^{137}Cs) depend on each individual sample, and are lower than a normal non-Compton suppressed gamma spectrometer approximately by a factor of 5.

Due to insufficient sample material for most samples collected and time available for gamma counting, samples were pooled, although a few large samples were counted individually. A pooled sample is a mixture of several fish samples of the same species from the same lake with equal weight contribution from individual fish samples. Table A in the appendix provides information and pooled sample IDs on which individual fish samples were pooled together.

2.3. Estimation of effective half-life of ^{137}Cs in fish

Because contaminants persist globally in the environment that fish occupy, they ingest contaminated food and continue to take in radionuclides. Thus, long term declines in radioactivity concentrations in fish are controlled by or depend on behaviour of radionuclides in the environment, often quantified using the ecological half-life, $T_{1/2}^{eco}$, which is a parameter that integrates all processes (except radioactive decay) that cause a reduction of activity in a specific medium (freshwater in this study) (Sundbom et al 2003; IAEA 2010). Assuming that the decline in radioactivity concentration $C(t)$ from an initial concentration $C(0)$ is exponential:

$$C(t) = C(0)\cdot e^{-(\lambda_r+\lambda_{eco})t}$$

where t is the time elapsed, λ_r the radioactive decay constant, which relates to the physical half-life $T_{1/2}^{r}=ln2/\lambda_r$. The rate of decline, λ_{eco}, is related to the ecological half-life as $T_{1/2}^{eco}=ln2/\lambda_{eco}$. For long-term decline, the effective half-life, $T_{1/2}^{eff}$, of a radionuclide includes the physical decay half-life and the ecological half-life:

$$\frac{1}{T_{1/2}^{eff}} = \frac{1}{T_{1/2}^{eco}} + \frac{1}{T_{1/2}^{r}}$$

where $T_{1/2}^{r}$=30.2 years for ^{137}Cs.

Activity concentrations of ^{137}Cs in whitefish from Lake 302 and Lake 226 were measured in 1979 (Elliott et al. 1981). The average ^{137}Cs concentrations were 57 and 49 Bq/kg (fw) with a standard deviation of 20 and 18 Bq/kg (fw) for whitefish in Lake 302 and 226 at that time, respectively. These averages served as initial activity concentrations $C(0)$ for whitefish in the two lakes. Assuming no significant amount of ^{137}Cs was added to the environment after atmospheric nuclear weapons tests conducted in the 1950s and

1960s, the effective half-life of ^{137}Cs in fish of northern Ontario lakes could be determined with activity concentrations of $C(t)$ $(t = 35$ years$)$ measured in the current study using the above equations.

2.4. Radiochemical analysis for naturally occurring radionuclides

The radioactivity concentration of ^{210}Po was determined for most fish samples individually. Due to the rather large number of samples, radiochemical analysis for ^{210}Po was conducted by two groups: the radiochemical laboratory of the Saskatchewan Research Council (SRC) (422 Downey Road, Saskatoon, Canada), and the radiochemical laboratory at the Radiation Protection Bureau (RPB) of Health Canada with alpha counting and spectra analysis performed by the environmental laboratory of the Canadian Nuclear Safety Commission (CNSC). Methods of radiochemical analysis of ^{210}Po in fish samples performed by SRC and RPB-CNSC are similar. Briefly, for each fish sample analysed by SRC, 20 g of wet sample was acid digested in a microwave digestion system prior to counting by alpha spectrometry for ^{210}Po, with ^{209}Po added as a tracer to determine the chemical recovery for the method. Independent measurements of ^{210}Po concentration in the same samples performed by the two laboratories showed good agreement (Sadi et al. 2016). Due to its short half-life, reported ^{210}Po concentrations were decay corrected from measuring date to the harvest date for each fish sample individually, as commonly being done in the literature and in our recent fish studies (Health Canada 2015, Chen et al 2015a, 2015b).

Radiochemical analyses for ^{210}Pb and ^{226}Ra were analyzed by the SRC. To determine ^{210}Pb and ^{226}Ra concentrations in fish samples, a minimum of 200 g of wet sample was needed in order to maintain the detection limits for ^{210}Po and ^{210} Pb at similar levels (about 0.2 Bq/kg). The samples were thermally dried. For ^{210}Pb, ashed samples were digested with nitric and perchloric acids and held for 30 days to guarantee equilibrium between ^{210}Pb and ^{210}Bi. Lead-210 was determined indirectly by the precipitation and counting of its high energy beta emitting progeny, ^{210}Bi. Bismuth was isolated by solvent extraction and subsequently precipitated as bismuth oxychloride. The precipitate was collected on a filter paper/disk assembly and beta counted in a low background counting system. Radium isotopes in the sample solution were separated by co-precipitation with lead sulfate. The precipitate was re-dissolved and the radium isotopes were separated from the other elements by co-precipitation with barium sulfate. The precipitate was filtered and then mounted on a plastic disk. It was then counted on an alpha spectrometer. The ^{226}Ra alpha energy is distinct and the peak can be clearly identified. Because a minimum of 200 g material was required, only pooled samples were sent to the SRC for the analyses of ^{210}Pb and ^{226}Ra, due primarily to insufficient sample material for most individual samples. Table A in the appendix provides information on which individual fish samples were pooled together.

2.5. Statistical analyses

Relationships between body size and activity concentration of ^{210}Po in fish were evaluated by linear regression and exponential fits of the form $y = ax^b$ using non-linear least squares. Differences in radionuclide activities among lakes were evaluated using ANOVA and post-hoc comparisons were conducted using a Tukey's test. Where appropriate, t-tests were conducted for comparisons between two groups and employed a Welch correction on degrees of freedom to account for heterogeneous variance. All analyses were performed using R (R core team, 2015).

3. Results and discussion

The radioactivity concentrations of ^{210}Po for individual fish samples are given in Table 1. Detailed results of radiochemical analyses for ^{210}Pb and ^{226}Ra and gamma counting results for ^{137}Cs are given in Table 2. Summary results for fish species from three lakes in the ELA are given in Table 3. The results for naturally occurring radionuclides are discussed first, then contamination levels of radioactive caesium, followed by radiation dose estimation from fish consumption.

3.1. Results for naturally occurring radionuclides

Activity concentrations of ^{210}Po were below the detection limit in 6 out of 125 samples (Table 1). Since ^{210}Po is assumed to be always present in fish, a practical calculation was applied to the few fish samples with ^{210}Po concentration below the detection limit, by reporting the ^{210}Po concentration at half of the detection limit (DL = 0.2Bq/kg), i.e. DL/2 in order to permit numerical analysis and estimation of means. Among the 125 freshwater fish samples tested in this study, the overall average concentration of ^{210}Po was 1.5 Bq/kg fresh weight (fw) (Table 3), similar to the ^{210}Po levels in Pacific salmon and trout (Health Canada 2015; Chen et al. 2015a). The average ^{210}Po concentrations were 1.7, 1.6 and 1.4 Bq/kg fw for Northern Pike, White Sucker and Lake Whitefish, respectively. Lake Whitefish ^{210}Po was significantly different among the 3 lakes sampled (ANOVA, $F_{2,91} = 16.91$, $P<0.0001$). Concentrations of ^{210}Po were similar between lakes 305 and 226 (Tukey's HSD, $P = 0.9$), both of which were more than four times greater than concentrations measured in Lake 302 (Table 3, Tukey's HSD, $P<0.0001$ for both comparisons).

Concentrations of ^{210}Pb concentration in fish muscle were significantly lower than ^{210}Po. While ^{210}Po was detectable in almost all fish samples, its parent, ^{210}Pb was below the detection limit (about 0.2 Bq/kg) in almost all samples collected in this study (Table 2). This is consistent with findings in the literature, such as reported by the Norwegian Radiation Protection Authority (Gjelsvik et al. 2012) that ^{210}Pb concentrations were, respectively, 9.5 and 5.2 times lower than ^{210}Po concentrations in pike and whitefish samples from Lake Iso-Ahvenainen in Finland.

Levels of ^{226}Ra were most frequently below detection limit (about 0.06 Bq/kg) in our samples (Table 2). In a report published in 1981 (Elliot et al. 1981), 10 whitefish samples from Lake 302 were analysed for ^{226}Ra. The ^{226}Ra concentrations varied from 0.033 to 0.16 Bq/kg with an average of 0.086 Bq/kg. In the current study, 40 whitefish samples of Lake 302 were mixed into 3 pooled samples (each containing more than 10 individual whitefish) for ^{226}Ra analysis, and the results were below the detection limit of about 0.06 Bq/kg. However, one individual whitefish (A029) from Lake 305 was measured with ^{226}Ra above the detection limit, and the ^{226}Ra concentration in one pooled whitefish sample (WF226b) from Lake 226 was 0.1 Bq/kg averaged over 12 individual whitefish (Table 2). Given that the mean concentration in the 1981 study is very close to the detection limit from the current analysis, it is difficult to conclusively evaluate changes in this radionuclide over time.

3.2. Potential size dependency of ^{210}Po concentration in fish

In this study, significant negative relationships of ^{210}Po with body mass among fishes from two different lakes were detected. The plots of data suggest possible negative relationships for whitefish generally. Northern Pike ^{210}Po scaled negatively with body size (linear regression, $F_{1,9} = 23.5$, $P<0.0001$, $R^2 = 0.72$; Fig. 1). Residual plots of the linear regression suggested the potential for a

Table 1
Results of ^{210}Po concentrations for individual fish samples.

ID	Po-210 (Bq/kg fw)	ID	Po-210 (Bq/kg fw)	ID	Po-210 (Bq/kg fw)	ID	Po-210 (Bq/kg fw)
A001	0.5 (±0.2)	A033	0.3 (±0.1)	A065	0.4 (±0.1)	A097	2.2 (±0.2)
A002	1.6 (±0.6)	A034	0.7 (±0.4)	A066	0.4 (±0.1)	A098	3.2 (±0.2)
A003	3.3 (±0.9)	A035	0.3 (±0.1)	A067	2.9 (±0.7)	A099	1.4 (±0.7)
A004	1.5 (±0.6)	A036	< 0.2	A068	2.2 (±0.2)	A100	1.0 (±0.1)
A005	1.7 (±0.7)	A037	0.5 (±0.1)	A069	0.5 (±0.1)	A101	3.9 (±1.0)
A006	1.6 (±0.6)	A038	0.4 (±0.1)	A070	0.3 (±0.1)	A102	3.7 (±0.2)
A007	4.0 (±1.0)	A039	0.2 (±0.1)	A071	0.5 (±0.2)	A103	1.2 (±0.6)
A008	< 0.2	A040	0.7 (±0.2)	A072	0.4 (±0.1)	A104	1.7 (±0.1)
A009	0.7 (±0.2)	A041	2.2 (±0.4)	A073	0.3 (±0.1)	A105	2.3 (±0.2)
A010	1.1 (±0.5)	A042	1.0 (±0.5)	A074	< 0.2	A106	1.3 (±0.1)
A011	2.3 (±0.9)	A043	1.0 (±0.5)	A075	3.7 (±0.9)	A107	3.0 (±0.8)
A012	0.5 (±0.2)	A044	1.3 (±0.3)	A076	0.8 (±0.1)	A108	1.3 (±0.1)
A013	1.1 (±0.4)	A045	1.6 (±0.3)	A077	2.2 (±0.9)	A109	0.7 (±0.1)
A014	0.8 (±0.5)	A046	1.2 (±0.5)	A078	1.0 (±0.1)	A110	< 0.2
A015	1.2 (±0.2)	A047	1.8 (±0.8)	A079	0.5 (±0.3)	A111	0.7 (±0.1)
A016	0.7 (±0.2)	A048	1.9 (±0.3)	A080	4.9 (±0.3)	A112	0.5 (±0.2)
A017	5.3 (±0.8)	A049	1.0 (±0.2)	A081	0.8 (±0.1)	A113	0.5 (±0.1)
A018	1.6 (±0.3)	A050	2.4 (±0.4)	A082	3.7 (±0.6)	A114	0.5 (±0.1)
A019	0.6 (±0.4)	A051	1.0 (±0.2)	A083	3.9 (±0.2)	A115	0.3 (±0.3)
A020	4.1 (±0.9)	A052	0.5 (±0.1)	A084	5.8 (±0.4)	A116	0.5 (±0.1)
A021	1.1 (±0.5)	A053	0.8 (±0.1)	A085	1.4 (±0.1)	A117	1.0 (±0.1)
A022	2.7 (±0.6)	A054	0.5 (±0.2)	A086	3.3 (±0.3)	A118	0.5 (±0.1)
A023	4.4 (±0.5)	A055	1.6 (±0.2)	A087	1.4 (±0.6)	A119	0.4 (±0.3)
A024	9.6 (±2.1)	A056	1.7 (±0.4)	A088	2.6 (±0.2)	A120	0.5 (±0.1)
A025	1.3 (±0.6)	A057	0.8 (±0.5)	A089	1.4 (±0.1)	A121	0.5 (±0.1)
A026	1.0 (±0.2)	A058	1.3 (±0.6)	A090	2.2 (±0.8)	A122	1.0 (±0.1)
A027	1.2 (±0.2)	A059	1.1 (±0.3)	A091	0.3 (±0.3)	A123	1.0 (±0.1)
A028	0.2 (±0.2)	A060	2.1 (±0.8)	A092	1.0 (±0.1)	A124	1.0 (±0.1)
A029	5.2 (±0.7)	A061	2.1 (±0.2)	A093	2.7 (±0.2)	A125	< 0.2
A030	0.4 (±0.2)	A062	0.5 (±0.1)	A094	1.7 (±0.1)		
A031	0.5 (±0.2)	A063	0.6 (±0.1)	A095	1.2 (±0.5)		
A032	0.6 (±0.2)	A064	< 0.2	A096	0.6 (±0.1)		

Table 2
Results of sample moisture (percentage of water content) and activity concentrations for ^{210}Pb, ^{226}Ra and ^{137}Cs in individual and pooled fish samples. See Appendix Table A for details on which samples were combined for pooled analyses.

Lake	Species	Sample ID	Moisture	Pb-210 (Bq/kg fw)	Ra-226 (Bq/kg fw)	Cs-137 (Bq/kg fw)
305	Northern Pike	A001	80.12%	< 0.2	< 0.07	12.7 (±0.7)
		A008	79.53%	< 0.2	< 0.06	17.5 (±0.9)
		A011	79.11%	0.2 (±1.0)	< 0.06	13.9 (±0.7)
		P305				15.6 (±0.8)
	Lake Whitefish	A012				4.0 (±0.2)
		A025	78.50%	< 0.3	< 0.07	
		A029	78.99%	< 0.2	0.1 (±0.1)	
		A031	76.22%	0.4 (±1.0)	< 0.06	4.5 (±0.3)
		A046				3.6 (±0.2)
		WF305				4.9 (±0.3)
302	Lake Whitefish	WF302a	77.30%	< 0.3	< 0.07	6.4 (±0.4)
		WF302b	78.44%	< 0.3	< 0.07	5.8 (±0.3)
		WF302c	79.54%	< 0.3	< 0.06	5.0 (±0.3)
	White Sucker	WS302a	82.03%	< 0.2	< 0.06	4.5 (±0.2)
		WS302b	82.27%	0.7 (±1.0)	< 0.06	3.5 (±0.2)
226	Lake Whitefish	WF226a	78.47%	< 0.2	< 0.06	5.1 (±0.3)
		WF226b	78.36%	< 0.2	0.1 (±0.1)	6.1 (±0.3)
		WF226c	79.53%	< 0.2	< 0.06	5.2 (±0.3)

Table 3
Mean concentrations (standard deviation or range in bracket) of radionuclides in fish samples from three lakes at the Experimental Lakes Area.

LAKE	Species	Samples	Moisture	Po-210 (Bq/kg fw)	Pb-210 (Bq/kg fw)	Ra-226 (Bq/kg fw)	Cs-137 (Bq/kg fw)
305	Pike (*Esox Lucius*)	11	80%	1.7 (±1.2)	< 0.2 (<DL, 0.2)	< 0.06	15.6 (±2.5)
302	White Sucker (*Catostomus commersonii*)	20	82%	1.6 (±0.6)	0.4 (<DL, 0.7)	< 0.06	4.0 (±0.7)
305	Whitefish (*Coregonus clupeaformis*)	20	78%	2.2 (±2.4)	< 0.2 (<DL, 0.4)	< 0.06 (<DL, 0.1)	4.9 (±0.4)
226	Whitefish (*Coregonus clupeaformis*)	34	79%	2.1 (±1.3)	< 0.2	< 0.06 (<DL, 0.1)	5.5 (±0.6)
302	Whitefish (*Coregonus clupeaformis*)	40	78%	0.5 (±0.3)	< 0.2	< 0.06	5.7 (±0.6)
	all above	125	79%	1.5	< 0.2	< 0.06	6.1

curvilinear (exponential decline) relationship for this species, which was also significant (non-linear least squares, $P = 0.0003$). As

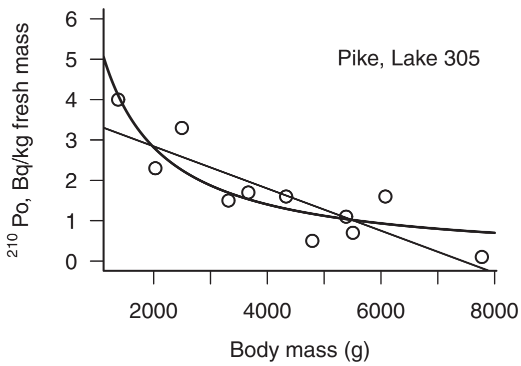

Figure 1. Activity concentrations of ^{210}Po in pike samples from Lake 305 as a function of total body mass. Dark solid line is a non-linear exponential model; solid line is a linear model. Both are significant at $\alpha = 0.05$.

the linear fit crossed zero ^{210}Po concentration at a body weight of about 7400 g, an exponential fitting may be a more reasonable choice (Fig. 1), if characteristics of the dependency were further examined. This observation is supported by a previous study (Chen et al 2015b), where it was observed that concentrations of ^{210}Po in Largemouth Bass (*Micropterus salmoide)* and Northern Pike decreased with increasing fish size, suggesting that small fish may be more capable of incorporating ^{210}Po into muscle. Further, the data presented in Chen et al. (2015b) strongly suggests a curvilinear relationship over both species, which is also supported by our findings for pike over a larger size range than the previous study. Though the shape of the relationships are similar and begin at similar concentrations (between 4 and 6 Bq/kg), it is interesting to note that ^{210}Po levels in Northern Pike in the NCR region were below 1 Bq/kg by approx. 1300 g, but remained above that level in our Pike population until approx. 5 kg.

The dependency of ^{210}Po concentration on fish size was weaker in Lake Whitefish among all three lakes compared with Northern Pike, but still generally negative with body size (Fig. 2; Linear regression, Lake 305, $F_{1,18} = 0.4$, $P = 0.5$; Lake 226, $F_{1,32} = 3.21$, $P = 0.08$; Lake 302, $F_{1,38} = 9.9$, $P = 0.003$). Non-linear exponential fits were not significant for either lake 226 or 305, but was significant for lake 302 ($P = 0.002$; Fig. 2). The relationship with body size was only significant among Lake 302 Whitefish, but was only marginally insignificant among fish in lake 226 at $\alpha = 0.05$. Given results from a previous study (Chen et al. 2015b) and the observation that the population with the greatest sample size was the only one to produce significant relationships between ^{210}Po and body size, it is possible that the lack of statistical significance in the other two lakes reflects insufficient sampling effort rather than the lack of a relationship.

Twenty white sucker samples were harvested from Lake 302. The ^{210}Po concentrations in those white sucker samples showed no significant relationship with fish mass (linear regression, $F_{1,18} = 0.4$, $P = 0.5$; Fig. 3).

While we observed an apparent consistency of a negative (perhaps negative exponential) relationship ^{210}Po and body mass between the ELA and NCR regions in Northern Pike, and within the ELA between Northern Pike and Lake Whitefish, differences in the relationships among species and regions suggest that body mass dependencies may be geographically and species-dependent. For instance, ^{210}Po appears to decrease with body size more rapidly for Northern Pike in the NCR region (Chen et al. 2015b), compared with ELA. Similarly, the more rapid decline in Lake Whitefish with body size compared with Northern Pike in the current study may relate to the different trophic levels each occupies. Both Largemouth Bass and Northern Pike are at higher trophic level than Lake Whitefish

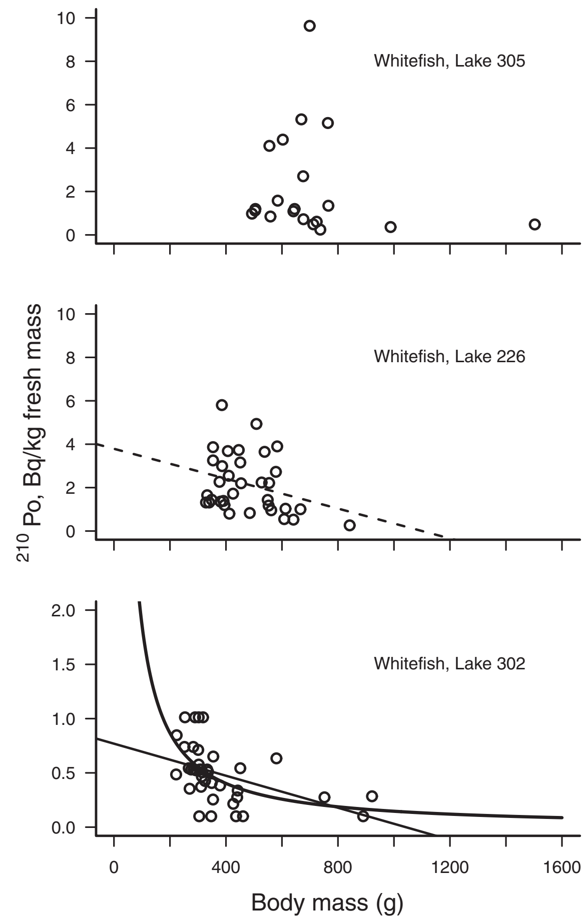

Figure 2. Activity concentrations of ^{210}Po in whitefish samples from Lake 305 (upper panel), Lake 226 (middle panel) and Lake 302 (lower panel) as a function of total body weight. Dark solid lines are non-linear exponential models significant at $\alpha = 0.05$; solid lines are linear models significant at $\alpha = 0.05$; dashed lines are for linear models significant at $\alpha = 0.1$. Note differences in y-axis scales among panels.

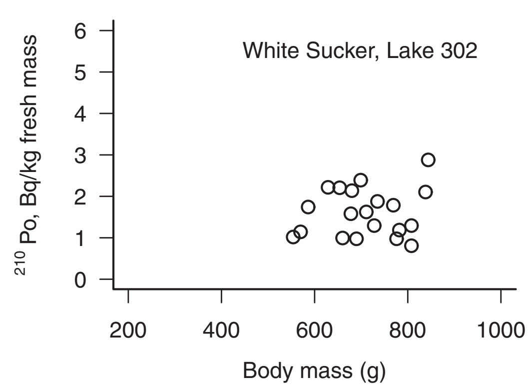

Figure 3. Activity concentrations of ^{210}Po in white sucker samples from Lake 302 as a function of total body weight.

and White Sucker (Rowan and Rasmussen 1994). The greater ^{210}Po levels at similar sizes for higher-tropic species (Northern Pike) reported in the current study suggest that trophic level may affect the size dependency of ^{210}Po in fish. This preliminary finding warrants further investigation across a broader range of geography and trophic levels.

3.3. Results of radioactive caesium

In all samples, ^{134}Cs was below the detection limit (similar to the average detection limit of 0.3 Bq/kg for ^{137}Cs). While ^{134}Cs was not detectable, ^{137}Cs was significantly above the detection limit for all samples measured. On average, the ^{137}Cs concentration was 6.1 Bq/kg fw in fish harvested from the ELA. Significant differences existed for ^{137}Cs among species examined in this study (ANOVA, $F_{2,14} = 93.2$, $P< 0.0001$; Table 3). Across all lakes, it was observed that the ^{137}Cs levels were comparable among White Sucker and Lake Whitefish (Tukey's HSD, $P = 0.6$); similarly, ^{137}Cs was not significantly different between White Sucker and Lake Whitefish in Lake 302 (t-test, $t_{4.8} = 1.7$, $P = 0.15$). However, ^{137}Cs was approximately three times higher in Northern Pike compared with Lake Whitefish (Tukey's HSD, $P<0.0001$) and White Sucker (Tukey's HSD, $P<0.0001$). The relatively higher concentration of ^{137}Cs in pike reflects the fact that pike is at a higher trophic level than the other two species. The observed ratio of ^{137}Cs in pike to that in whitefish in our study is within the range of ratios reported by Rowan and Rasmussen (1994) of 2.7 in Lake Ontario, 1.7 in Lake Huron, 2.8 in the Ottawa River, 2.4 and 3.6 in Christie Bay and Western Basin of the Great Slave Lake, 3.3 in Red Lake of USA, 2.8 and 4.9 in Inarinjarvi and Melkutin of Finland, respectively. This finding is also in agreement with the observation reported by Sundbom et al. (2003) that the steady−state activity concentrations of ^{137}Cs in fish increased significantly by trophic level.

The average ^{137}Cs concentration of 6.1 Bq/kg in freshwater fish reported here is almost twice the average level of 3.7 Bq/kg observed in 20 fish samples from the NCR (Chen et al. 2015b) and more than 20 times higher than the 0.24 Bq/kg observed in 45 Pacific Salmon samples harvested in 2014 from the western coast of Canada (Health Canada 2015). Considering that the physical properties of caesium most closely resemble those of potassium, and that there is an overwhelming excess of stable potassium in marine waters relative to radioactive caesium, it is understandable that, for the same environmental level of ^{137}Cs contamination, marine fish are usually much lower in ^{137}Cs than are freshwater fish. The large excess of potassium in seawater serves to block the uptake of radioactive caesium by marine organisms (Fesenko et al. 2010, 2011).

Cs-137 has proven to be the most important long-term anthropogenic contributor to the environmental radiation exposure to the public. The primary source of ^{137}Cs in the biosphere is atmospheric nuclear weapons testing during the 1950s and 1960s. Of the roughly 1 EBq (10^{18} Bq) of ^{137}Cs released to the biosphere, about 90% was produced by atmospheric nuclear testing (NCRP, 2006). Approximately 6% was produced by the Chernobyl accident in 1986 and roughly 4% by nuclear fuel reprocessing facilities. Since ^{134}Cs was not measured even at a detection limit of 0.3 Bq/kg and ^{137}Cs was clearly detectable with an average concentration over all species of 6.1 Bq/kg, it can be concluded that levels of ^{137}Cs in fish samples studied here are primarily normal environmental levels resulting from atmospheric nuclear weapons tests, the Chernobyl nuclear accident and other historical sources. The contribution to environmental ^{137}Cs from the recent Fukushima accident is negligibly small.

As reported in a previous study (Elliot et al. 1981), 15 whitefish samples from the Lake 302 and 22 Lake Whitefish samples from the Lake 226 were collected in the fall of 1979. The average ^{137}Cs concentrations were 57 ± 20 and 49 ± 18 Bq/kg (fw) for whitefish in Lake 302 and 226, respectively. The overall average ^{137}Cs concentration from a total of 37 Lake Whitefish samples was 52 ± 19 Bq/kg (fw) in 1979. In current study, the overall average ^{137}Cs concentration from a total of 74 Lake Whitefish samples of Lake 302 and Lake 226 was 5.6 Bq/kg (fw) with concentrations in samples from Lake 302 still being slightly higher than in samples from Lake 226 (Table 3). Since no major release of ^{137}Cs into the environment has occurred since the study by Elliot et al., one can assume ^{137}Cs levels in fish are equilibrated to ambient caesium concentration, i.e. steady state concentration. Therefore, the ecological half-life of ^{137}Cs in whitefish of the ELA was estimated to be 17.0 years (16.2 and 17.5 years for Lake 302 and Lake 226, respectively). The long-term effective half-life of ^{137}Cs in whitefish of the ELA, including physical decay half-life and ecological half-life resulting from various environmental processes, was estimated to be 10.9 years (the effective half-lives of ^{137}Cs in whitefish were estimated to be 10.5 and 11.1 years for Lake 302 and Lake 226, respectively). The results are comparable to other studies reported in the literature. For example, the long-term effective half-lives for ^{137}Cs in Savannah River fish were determined to be 7.4 years and 8.1 years for upriver site and downstream site, respectively (Paller et al. 2014). The longer effective half-life in the current study compared with those reported in the literature may be due to a comparison of lake vs. river systems, as lakes will have longer residence times than river systems.

3.4. Estimated radiation dose from fish consumption

While ^{226}Ra and ^{210}Pb concentrate in less edible bone tissues, ^{210}Po generally binds to protein and sequesters in edible soft tissues. Caesium-137 is metabolically similar to the essential nutrient, potassium, and has a great affinity for edible muscle. Therefore, levels of ^{210}Po and ^{137}Cs were considered for the estimation of radiation doses resulting from fish consumption. Doses were calculated for average concentrations of ^{210}Po and ^{137}Cs, and for one fish meal of 150 g and several fish meals up to 1 kg. Results are summarised in Table 4 for children (5-y old) and adults. The majority of the radiation dose from fish consumption was due to ^{210}Po. On average, doses from naturally occurring ^{210}Po are more than 100 times higher in children and more than 20 times higher in adults compared to the doses from ^{137}Cs at the levels observed in this study.

Statistics of fish consumption in countries such as Canada fluctuate over the years. Based on the statistics given by Fisheries and Oceans Canada (2013), averaged over the past 23 y (1988−2010), the Canadian consumption rate for fish products is 8.8 kg (edible weight) per person per year. This includes fresh and frozen sea fish, processed sea fish, total shellfish and freshwater fish. Assuming a child consumed 8.8 kg of fish in a year and all consumed fish contain the average levels of ^{137}Cs and ^{210}Po measured in this study, the resulting radiation dose would be 0.06 mSv, a small fraction of the annual dose from exposure to various types of natural background radiation (2.4 mSv per year) (UNSCEAR 2000).

4. Conclusions

None of the fish samples analysed in this study contained any detectable levels of ^{134}Cs (an indicator of recent releases from the Fukushima accident of 2011). For environmental ^{137}Cs resulting mainly from the fallout of atmospheric nuclear weapons tests in the 1950s and 1960s and accidental releases such as from the Chernobyl accident in 1986, an average level of 6.1 Bq/kg fw was observed in freshwater fishes harvested from the ELA lakes. Our analyses indicate that the average ^{137}Cs concentration in fish from inland lakes is more than 20 times higher than the levels observed in marine fish harvested from the Canadian west coast in 2013 and 2014 (Health Canada 2015; Chen et al. 2015a). However, it should be

Table 4
The estimated radiation doses for children (5 y old) and adults due to intake of one fish meal (150 g) and several fish meals up to 1 kg, assuming fish contains the average ^{137}Cs concentration of 6.1 Bq/kg fw and ^{210}Po concentration of 1.5 Bq/kg fw. (DC is the dose conversion coefficient in the unit nSv/Bq).

	concentration	DC	(nSv/Bq)	μSv	(from 150 g)	μSv	(from 1 kg)
	Bq/kg	children	adults	children	adults	children	adults
^{137}Cs	6.1	9.6	13	0.0088	0.012	0.059	0.079
^{210}Po	1.5	4400	1200	0.99	0.27	6.6	1.8
Sum				1.0	0.28	6.7	1.9

mentioned that the relatively high concentrations of ^{137}Cs in fish samples from these inland lakes are still considered very low from a radiological protection perspective.

This study confirmed that ^{210}Po is the dominant contributor to radiation doses resulting from fish consumption. While concentrations of ^{210}Pb and ^{226}Ra were below conventional detection limits, ^{210}Po was measured in almost all fish samples collected from the ELA in 2014. The average concentration was about 1.5 Bq/kg fw. The resulting radiation dose from consumption of 1 kg of fish containing 1.5 Bq of ^{210}Po would be 2 μSv for adults, a very small fraction (< 1/1000) of the annual dose from exposure to various types of natural background radiation (2.4 mSv per year). The results indicate that fishes from inland lakes pose practically no radiological health concern.

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Appendix

Table A
Information for individual fish samples harvested from three lakes in the ELA, summer 2014, and information on which fish being pooled together and under which pooled sample ID (pooled samples are separated by horizontal lines. For example, the ID P305 contains individual samples from A001 to A011, and the ID WF305 contains individual fish samples from A012 to A031).

LAKE	Species	ID	Sex	Total length (mm)	Total weight (g)	Pooled sample ID
305	pike	A001	M	902	4793	P305
		A002	M	1040	6079	
		A003	M	736	2497	
		A004	F	832	3317	
		A005	F	854	3667	
		A006	M	825	4331	
		A007	M	568	1375	
		A008	M	1074	7776	
		A009	M	956	5505	
		A010	F	970	5387	
		A011	M	675	2030	
	whitefish	A012	F	427	712	WF305
		A013	F	372	504	
		A014	M	383	559	
		A015	M	367	505	
		A016	F	411	677	
		A017	M	400	669	
		A018	M	387	585	
		A019	F	423	724	
		A020	F	390	555	
		A021	M	396	641	
		A022	F	408	676	
		A023	F	403	603	
		A024	F	421	699	
		A025	F	430	766	
		A026	F	371	493	
		A027	F	415	645	
		A028	M	418	737	
		A029	F	418	764	
		A030	F	462	988	
		A031	F	506	1503	
302	whitefish	A032	-	402	580	WF302a
		A033	M	452	752	
		A034	F	342	301	
		A035	M	466	922	
		A036	M	354	347	
		A037	-	340	293	
		A038	-	366	379	
		A039	F	372	426	

Table A (*continued*)

LAKE	Species	ID	Sex	Total length (mm)	Total weight (g)	Pooled sample ID
		A040	-	361	355	
		A052	F	347	314	
		A053	F	327	224	
		A054	-	321	222	
		A062	M	322	267	WF302b
		A063	F	351	303	
		A064	F	380	436	
		A065	M	348	311	
		A066	M	352	325	
		A069	M	350	316	
		A070	M	381	442	
		A071	M	382	451	
		A072	F	335	270	
		A073	M	381	440	
		A074	M	457	890	
		A109	M	325	252	
		A110	M	345	305	
		A111	M	333	283	
		A112	F	354	336	WF302c
		A113	F	354	333	
		A114	F	328	276	
		A115	M	357	354	
		A116	F	357	312	
		A117	M	343	303	
		A118	F	341	285	
		A119	M	357	349	
		A120	F	354	285	
		A121	F	344	306	
		A122	M	357	318	
		A123	M	330	254	
		A124	M	341	290	
		A125	M	373	461	
	white sucker	A041	-	437	654	WS302a
		A042	-	425	690	
		A043	F	433	776	
		A044	F	433	808	
		A045	F	414	678	
		A046	F	438	782	
		A047	F	427	769	
		A048	F	422	735	
		A049	F	388	554	
		A050	F	435	699	
		A051	F	428	660	WS302b
		A055	F	433	711	
		A056	F	405	586	
		A057	F	444	808	
		A058	F	442	728	
		A059	F	409	570	
		A060	F	438	838	
		A061	F	416	680	
		A067	F	445	844	
		A068	F	410	629	
226	whitefish	A075	M	373	538	WF226a
		A076	F	373	485	
		A077	M	388	527	
		A078	F	405	613	
		A079	F	426	641	
		A080	M	373	509	
		A081	F	363	412	
		A082	M	365	446	
		A083	F	324	354	
		A084	M	343	385	
		A085	F	353	391	
		A086	F	344	354	WF226b
		A087	M	349	381	
		A088	F	356	410	
		A089	F	341	349	
		A090	F	390	554	
		A091	-	439	842	
		A092	F	398	562	
		A093	F	393	578	
		A094	M	357	425	
		A095	M	394	552	
		A096	M	405	608	

(*continued on next page*)

Table A (*continued*)

LAKE	Species	ID	Sex	Total length (mm)	Total weight (g)	Pooled sample ID
		A097	F	362	454	
		A098	M	365	451	WF226c
		A099	M	391	549	
		A100	M	444	666	
		A101	M	394	583	
		A102	M	358	407	
		A103	F	356	395	
		A104	M	338	333	
		A105	M	350	377	
		A106	F	342	329	
		A107	F	353	386	
		A108	M	343	340	

References

Chen, J., Cooke, M.W., Mercier, J.F., Ahier, B., Trudel, M., Workman, G., Wyeth, M., Brown, R., 2015a. A report on radioactivity measurements of fish samples from the west coast of Canada. Radiat. Prot. Dosim. 163, 261–266.

Chen, J., Zhang, W., Sadi, B., 2015b. A report of radioactivity measurements of freshwater fish samples from the national capital region of Canada. J. Environ. Radioact. 144, 175–178.

Cooke, S.J., Murchie, K.J., 2013. Review - status of aboriginal, commercial and recreational inland fisheries in North America: past, present and future. Blackwell Publishing Ltd. Available at Fish. Manag. Ecol. http://www.fecpl.ca/wp-content/uploads/2013/04/FME-Cooke-Murchie-20131.pdf.

Fisheries and Oceans Canada (DFO), 2010. Survey of Recreational Fishing in Canada. Available on. http://www.dfo-mpo.gc.ca/stats/rec/can/2010/RECFISH2010_ENG.pdf.

Elliott, S.E.M., Burns-Flett, C., Hesslein, R.H., Brunskill, G.J., Lutz, A., 1981. Cesium-137, Radium-226, Potassium-40 and Selected Stable Elements in Fish Populations from Great Slave Lake (N.W.T.), Louis Lake (Saskatchewan), Lake Winnipeg (Manitoba), and Experimental LakeS Area (Northwestern Ontario). Canadian Data Report of Fisheries & Aquatic Sciences No. 293.

Fesenko, S., Fesenko, E., Titov, I., Karpenko, E., Sanzharova, N., Gondin Fonseca, A., Brown, J., 2010. Radionuclide transfer to marine biota species: review of Russian language studies. Radiat. Environ. Biophys. 49, 531–547.

Fesenko, S., Fesenko, J., Sanzharova, N., Karpenko, E., Titov, I., 2011. Radionuclide transfer to freshwater biota species: review of Russian language studies. J. Environ. Radioact. 102, 8–25.

Fisheries and Oceans Canada (DFO), 2013. Commercial Fisheries–consumption. Statistics Canada—Table 002–0011. Available on. http://www.dfo-mpo.gc.ca/stats/commercial/consumption-eng.htm.

Gjelsvik, R., Brown, J., Roos, P., Saxen, R., Outola, L., 2012. Polonium-210 and Other Radionuclides in Terrestrial, Freshwater and Brackish Environments. Report 2012:3. Norwegian Radiation Protection Authority, Østerås, Norway.

Health Canada, 2015. Health Canada Analyses of the Radioactive Content of Fish Samples from Canada's West Coast. 2015. Available at. http://registry.data.gc.ca/en/dataset/d1b39de7-e525-4cfa-a605-af38bf174555.

International Atomic Energy Agency (IAEA), 2010. Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments. Vienna.

Kinnunen, R.E., 2003. Great Lakes commercial Fisheries. Available at. http://www.glerl.noaa.gov/seagrant/GLFLI/Notebook/Curriculum/Projects/GLCommercialFinal.pdf.

National Council on Radiation Protection and Measurements, 2006. Cesium-137 in the Environment: Radioecology and Approaches to Assessment and Management. NCRP Report 154. Bethesda.

Paller, M.H., Jannik, G.T., Baker, R.A., 2014. Effective half-lives of ^{137}Cs in various environmental media at the Savannah River site. J. Environ. Radioact. 131, 81–88.

R Core Team, 2015. R: a Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria. https://www.R-project.org/.

Rowan, D.J., Rasmussen, J.B., 1994. Bioaccumulation of radiocesium by fish: the influence of physicochemical factors and trophic structure. Can. J. Fish. Aquat. Sci. 51, 2388–2410.

Sadi, B., Chen, J., Kochermin, V., Tung, G., Chiorean, S., 2016. An in-house method for determination of polonium-210 in fish samples. J. Radiochem Nucl. Chem. http://dx.doi.org/10.1007/s10967-015-4575-6. In press.

Sundbom, M., Meili, M., Andersson, E., Ostlund, M., Broberg, A., 2003. Long-term dynamics of Chernobyl 137Cs in freshwater fish: quantifying the effect of body size and trophic level. J. Appl. Ecol. 40, 228–240.

United Nations Scientific Committee on the Effects of Atomic Radiation, 2000. Sources and Effects of Ionizing Radiation. UNSCEAR 2000 Report, vol. I sources. United Nations, New York.

United Nations Scientific Committee on the Effects of Atomic Radiation, 2014. Levels and Effects of Radiation Exposure Due to the Nuclear Accident After the 2011 Great East-Japan Earthquake and Tsunami. UNSCEAR 2013 Report, vol. I. United Nations, New York.

Zhang, W., Keeshan, B., Mekarski, P., Yi, J., Ungar, K., 2013. Performance evaluation and optimization for a newly developed digital list-mode data acquisition Compton suppression spectrometer. Appl. Radiat. Isot. 81, 96–102.