Journal of Environmental Radioactivity 195 (2018) 104–108

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Tritium in fish from remote lakes in northwestern Ontario, Canada

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ARTICLE INFO

Keywords:
Lake Whitefish
Northern Pike
White Sucker
Background tritium levels
Freshwater fishes

ABSTRACT

Tritium is most commonly generated as a by-product of nuclear reactors. As such, environmental concentrations are typically only reported near regions of interest, and background concentrations in areas unaffected by anthropogenic disturbance are not well characterized. To provide information on background levels of tritium in the natural environment, tissue-free water tritium (TFWT) and organically-bound tritium (OBT) were measured in the flesh of 106 fish collected within three lakes located at the IISD-Experimental Lakes Area (ELA) in Ontario, Canada in 2014. For the three ELA lakes studied, water tritium (HTO) activity concentration was determined to be below reliably detectable levels (0.6 Bq/L). Fish TFWT was found to be below 0.7 Bq/L, similar to the surrounding water tritium activity concentration. Fish OBT activity concentrations, at below 5 Bq/L, were also very low. Fish size was significantly related to OBT activity in Lake Whitefish and White Sucker from Lake 302, but not in other lakes. Though we observed significant differences in potential tritium exposure to humans among lakes, the levels of tritium reported here are below the Canadian natural background radiation of 1.8 mSv/y. These results provide information on background levels of tritium in freshwater fishes in Canada.

1. Introduction

Tritium is produced naturally from interactions of cosmic rays with gases in the upper atmosphere and, to a much lesser extent, by processes taking place within the earth crust. Anthropogenic sources include the production of tritium as a by-product of nuclear reactors. Because tritium is routinely released from Canada Deuterium Uranium (CANDU) reactors, tritium studies have been focused in regions close to nuclear reactors (Le Goff et al., 2016; Kořinková et al., 2016; Thompson et al., 2015; Fiévet et al., 2013).

Tritium can pose a health risk only if ingested through drinking water or food, inhaled, or absorbed through the skin. Tritium decays to helium, and in doing so, releases low energy beta radiation. Knowledge of levels of tritium in the environment is used in the assessment of population exposure to environmental tritium (Boyer et al., 2009; Fiévet et al., 2013; Melintescu and Galeriu, 2011).

Currently, baseline fresh water biota tritium data in Ontario (Canada) is very scarce as most measurements have been obtained from samples collected near nuclear facilities. Areas that are unaffected by nuclear sites can provide data on background tritium levels, but are data deficient. These background levels likely originate from nuclear weapons testing and from cosmogenic production (Kim et al., 2016). The IISD-Experimental Lakes Area (ELA) is an internationally unique research station encompassing 58 remote freshwater lakes with highly controlled access, removed from anthropogenic disturbance. The ELA is approximately 1000 km away from the closest nuclear facilities in Ontario, and over 500 km from the closest US facilities in Minnesota. As such, the analysis of biota and water samples collected from the ELA provides both opportunities to report background levels of tritium in the absence of significant nuclear operations and long term effects in fish after tritium tracer experiments. By providing a better description of background levels in the natural environment and long term effects in fish, the results of this study also contribute to a more precise understanding of the tritium footprint of nuclear facilities in Canada and elsewhere.

As part of a collaboration between Canadian Nuclear Laboratories (CNL), Health Canada and the ELA that aimed to study background tritium levels in Canadian foods, a total of 106 fish samples were obtained from three ELA lakes during the summer of 2014. Fish tissue free water tritium (TFWT) and organically bound tritium (OBT) were determined on each of the fish sampled. The TFWT activity concentration in fish reflects tritium activity concentration in water that occurred a few hours or days before sampling. In contrast, OBT activity concentration in fish can reflect environmental tritium levels accumulated

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https://doi.org/10.1016/j.jenvrad.2018.10.003
Received 27 June 2018; Received in revised form 3 October 2018; Accepted 8 October 2018

Table 1
Inventory of ELA fish received by CNL low background laboratory.

Lake	Species	Number	Male (M), Female (F) or Unknown (U)
305	Northern Pike	11	M (8) and F (3)
	Lake Whitefish	18	M (6) and F (12)
302	White Sucker	17	M (0), F (16) and U (1)
	Lake Whitefish	30	M (18), F (10) and U (2)
226	Lake Whitefish	30	M (15), F (14) and U (1)
Total	-	**106**	-

M = males, F = females, U = gender unknown.

over months to years prior to sampling. Species of interest were Northern Pike (*Esox lucius*), Lake Whitefish (*Coregonus clupeaformis*) and White Sucker (*Catostomus commersonii*). Northern Pike and Lake Whitefish are the focus of both recreational and commercial fisheries (DFO, 2010; Kinnunen, 2003), and White Sucker are a species of importance in aboriginal culture (Cooke and Murchie, 2013). Tritium in water (as HTO) was also measured in the same three lakes, to provide context for reported fish concentrations.

2. Material and methods

Fish were collected from Lake 305 (Northern Pike and Lake Whitefish), Lake 302 (White Sucker and Lake Whitefish) and Lake 226 (Lake Whitefish) at the ELA using 3–4.5" mesh gillnets used for Lake 302 and Lake 226 and multimesh experimental gillnets used for Lake 305 during summer (July 4 - August 5) of 2014 (Chen et al., 2015a). Fish were sampled for length and weight, and sex was determined via dissection. Where possible, fillets of at least 100 g were taken from each fish. Fillets were dried and homogenized (Table 1; Chen et al., 2016) and delivered to the CNL Low Background Laboratory.

2.1. HTO measurements in lake water

Nine lake water samples (3 for each of the lakes) were sampled under ice in March of 2015. Tritium activity concentrations were measured following ISO 9698 (2010) using an ALOKA (LSC LB-7, Hitachi Aloka Medical Ltd., Japan) liquid scintillation counter (LSC). Prior to being placed in the counter, 70 mL of water sample was mixed with Ultima Gold LLT (PerkinElmer) cocktail using a 1:1 ratio. The samples were prepared for counting in a 145 mL low-diffusion polyethylene vial using the ALOKA system (Zinsser Analytic Ltd. Germany). The counting efficiency was determined by using a tritium standard (NIST, USA). Each water sample was counted for 400 min and the minimum detectable activity (MDA) achieved was less than 0.6 Bq/L. Although most LSC use a 20 mL vial to hold 10 mL of water sample, we used a larger water sample in order to achieve the lower detection limit used in our study.

2.2. Fish TFWT measurements

Fish samples were frozen and kept at −20 °C until analyzed. The TFWT was extracted using a laboratory designed freeze-drying system (Kim and Roche, 2013). The frozen fish flesh samples (approximately 100 g) were loaded into a vacuum flask and the extracted tissue-free water was collected under vacuum over 48 h. The extracted water was collected on a liquid nitrogen trap. For each sample, the collected water activity concentration was determined by mixing 50 mL of extracted water from fish tissues with 50 mL of Ultima Gold LLT. Each sample was counted for 400 min using the ALOKA LB-7 LSC. The MDA was approximately 0.7 Bq/L and the overall uncertainty of the measurements was estimated to be less than 10%. The TFWT per g fresh fish tissue was estimated as the measured Bq/L in extracted water multiplied by the moisture content of the fish. Where the measured activity was below detection (0.7 Bq/L), the detection limit of 0.7 Bq/L was used in TWFT calculations.

2.3. Fish OBT measurements

The freeze-dried fish tissue remaining after the extraction of the TFWT measurements was dried at 55 °C for 24 h. The completely dried samples were ground using a small mixer and combusted using a Parr apparatus (model 1121, USA) with pressurized oxygen (Kim and Roche, 2013). The combustion water was then distilled and diluted to 10 mL with tritium-free water, and mixed with 10 mL of Ultima Gold LLT. The OBT activity concentration of the samples was determined using a Quantulus LSC. The counting time was 360 min. In this study, as almost 10 mL of combustion water was collected for each fish sample, the MDA achieved was about 2.5 Bq/L. The OBT value is considered here as total OBT activity (Exchangeable OBT + Non-exchangeable OBT). The overall uncertainty of the OBT measurement was estimated to be less than 25%. The OBT of fish in fresh tissues was estimated by multiplying the measured Bq/L by the ratio of dry mass to fresh mass analyzed and a water equivalence factor of 0.6 (Kim and Stuart, 2015). In order to compare Quantulus measurements with another method, ALOKA LSC (LB-7, Hitachi Aloka Medical Ltd., Japan) were used. To do so, we created a composite large volume sample (obtained by combining 7 Quantalus vials) that was re-counted using the ALOKA LSC system.

2.4. Dose calculations

We used ERICA, a software program that allows the assessment of environmental risks arising from radionuclide exposure (Larsson, 2008; Brown et al., 2016). Based on the measured tritium activity concentration in ELA fishes, the total tritium dose rate was calculated for each fish. In addition, the potential fish tritium dose concentration to humans consuming these fish was calculated based on the Canadian fish products consumption (Fisheries and Oceans Canada, 2013).

2.5. Data analysis

We evaluated the effect of body size (mass, g) on TFWT measurements (Bq/kg fresh weight of fish) of fish using linear regression. We similarly evaluated OBT measurements against body size for all species in all lakes. To determine the effect of lake and sex (male, female, undetermined) on estimated ^3H dose exposure, we examined differences among Lake Whitefish (common species among all lakes) using 2-way ANOVA. Differences among species within lakes were compared using *t*-tests with a Welch correction for heterogeneous variance. Data were log-transformed where necessary to normalize data and residuals. Data were analyzed using R version 3.4.2 (R Core Team, 2017).

3. Results and discussion

3.1. HTO activity concentrations

The measured HTO activity concentrations for lakes 305, 302 and 226 from the ELA in 2015 were similarly below reliably detectable levels (0.6 Bq/L) in all lakes (Table 2). The levels measured were found to be much lower than the levels found in the Ottawa River water, which in 2016 were determined to be approximately 3–5 Bq/L near

Table 2
Water tritium (HTO) activity concentration of lake water.

Sampling Date	Lake	Number of Samples	Activity Concentrations
2014 October 26	302	3	< MDA[a] (0.6 Bq/L)
2014 October 27	305	3	< MDA[a] (0.6 Bq/L)
2014 October 27	226	3	< MDA[a] (0.6 Bq/L)

[a] Less than the Minimum Detectable Activity of 0.6 Bq/L.