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ORIGINAL PAPER

Potential health risks posed by polycyclic aromatic hydrocarbons in muscle tissues of fishes from the Athabasca and Slave Rivers, Canada

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Abstract Polycyclic aromatic hydrocarbons (PAHs) are released to the environment from oil sands operations and from natural sources in Alberta, Canada. Concentrations of 16 USEPA priority PAHs were measured in tissues of fishes collected from three locations on the Athabasca River in Alberta and two downstream locations on the Slave River in the Northwest Territories, Canada. A total of 425 individual fish were collected including 89 goldeye (*Hiodon alosoides*), 93 whitefish (*Coregonus clupeaformis*), 104 northern pike/jackfish (*Esox lucius*), 96 walleye (*Sander vitreus*) and 43 burbot/loche mariah/mariah (*Lota lota*). Fish were sampled during the summer and fall of 2011 and spring of 2012. Dorsal muscle of fishes from upstream reaches of the Athabasca River, close to oil sands extraction and upgrading activities, contained greater concentrations of individual PAHs than concentrations in muscle of fishes from further downstream in the Slave River. Concentrations of the sum of USEPA indicator PAHs ($\sum$PAHs) in fishes collected in the vicinity of Fort McKay, closest to oil sands activities, varied among seasons with average concentrations ranging from 11 (burbot, summer) to 1.2×10^2 ng/g, wm (burbot, spring) with a mean of 48 ng/g, wm. Concentrations of $\sum$PAHs in fishes collected in the vicinity of Fort Resolution, the location most distant from oil sands activities, also varied among species and seasons, with average concentrations ranging from 4.3 (whitefish, summer) to 33 ng/g, wm (goldeye, summer) with a mean of 13 ng/g, wm. Significant differences in concentrations of $\sum$PAHs in muscle were observed within goldeye, jackfish, walleye and whitefish among sites. Health risks posed by PAHs to humans were assessed probabilistically using a B[a]P equivalents approach ($B[a]P_{eq}$). The average lifetime risk of additional cancers for humans who consumed fish was deemed to be within an 'acceptable' range of risk (i.e., less than 10^{-6}).

Keywords Muscle · Oil sands · Human health · Daily intake (DI) · *De minimis* · Assessment · PAH · Cancer

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic compounds composed of two or more fused aromatic rings, which are released to the environment by both human activities and natural events. PAHs are contaminants formed during the incomplete combustion of organic material and are especially abundant in petroleum deposits, and can also be released during operations involving the extraction, transport or processing of petroleum (Pampanin and Sydnes 2013).

The Alberta oil sands in Canada contain the second largest proven petroleum hydrocarbon reserves in the world, totaling an estimated 173.2 billion barrels of recoverable oil (Government of Alberta 2013). A large proportion of these reserves are found in deposits of bitumen, which cover approximately 60,000 km^2 (Conly et al. 2002). Global demand for oil was 84.7 million barrels per day in 2008 and is expected to reach 105 million barrels per day by 2030 (International Energy Outlook 2013). Conventional production of crude oil is unable to meet this rising global demand for the readily available energy in petroleum hydrocarbons. Nonconventional oil sources including deposits of oil sands in Canada are required for a safe and secure energy future for North America and for the rest of the world. The global demand for Canadian oil has resulted in economic benefits for the country (Timilsina et al. 2005). The extensive development of the oil sands has also contributed to increased deposition of PAHs to the Athabasca River and its tributaries (Parajulee and Wania 2014). Dissolved polycyclic aromatic compound (PAC) concentrations up to 4.8 μg/L have been reported in melted snow collected from Athabasca River and its tributaries (Kelly et al. 2009). Concentrations of PAHs in sediments from Lake Athabasca and Lake Richardson, in the Peace/Athabasca delta, ranged from 1259 to 1867 μg/kg wet mass (Evans 2002). Total concentrations of PAHs in sediments of the Athabasca River Delta have reportedly increased between 1999 and 2009 at a rate of 0.05 mg/kg/years (Timoney and Lee 2011).

Parts of the deposits of bitumen are in close proximity to the Athabasca River and its tributaries, thereby contributing hydrocarbons to the river. Some residents of downstream communities, especially in Fort Chipewyan Alberta, on the shore of Lake Athabasca, have expressed concern that oil sands activities are contaminating country foods such as fish and game by contributing greater than natural levels of PAHs to the ambient environment (Timoney and Lee 2009; Chen 2009). Since fish is a major cultural and economic resource, the presence of PAHs in fishes of the Athabasca/Slave River system raises issues about potential risks to the health of humans in Aboriginal communities in the area (Usydus et al. 2009). Generally, dietary exposure to elevated concentrations of PAHs has been associated with increased risk of cancer in humans (Stacewicz-Sapuntzakis et al. 2008; Yoon et al. 2007). Some PAHs, such as benzo[a]pyrene, chrysene, indeno(1,2,3-c,d)pyrene and benzo(b)fluoranthene, are known carcinogens. They also produce mutagenic and genotoxic effects in experimental animals (Deutsch-Wenzel 1983; Thyssen et al. 1981). Potential health risks of fish consumption need to be balanced with the proven benefits of the consumption of essential omega-3, unsaturated fatty acids and minerals in fish which have many health benefits including the reduction of coronary heart disease and can lessen hypertension (Sidhu 2003; Berry 1997).

Although there are regulatory and monitoring activities in the Athabasca basin, studies of PAHs in the region are few, making little data available for assessment of baseline concentrations of contaminants or effects on populations of fishes or the people who consume them. Stakeholders have, in past years, expressed the need for the establishment of a comprehensive and transparent monitoring program in the Athabasca River (Dillon et al. 2011; Giesy et al. 2010; Weinhold 2011). Good reasons exist for the call, largely due to the possible effects and continuing expansion of oil and gas exploration and extraction within the basin. Despite the debate surrounding the cause of pollution (Wiklund et al. 2012; Kelly et al. 2010), establishing a monitoring program for PAHs in portions of the basin will provide baseline data in the area so that the status and trends of contamination can be assessed. Furthermore, information on current sources of contaminants such as PAHs is necessary so that appropriate control measures can be implemented. Finally, since PAHs have been naturally released from deposits of bitumen for millennia, it is important to determine the relative proportions emanating due to natural processes and additional releases due to activities of humans, including extraction and upgrading of petroleum hydrocarbons.

The aim of this work is to describe the spatial and seasonal distribution of PAHs in muscle of whitefish (*Coregonus clupeaformis*), northern pike (*Exos luscious*), walleye (*Sander vitreus*), goldeye (*Hiodon alosoides*) and burbot (*Lota lota*) and to apply a probabilistic approach to estimate risks due to exposure to PAHs through fish consumption in the Athabasca/Slave Rivers (Liang et al. 2013). Inter-season comparison and intra- and inter-specific variability of concentrations of PAHs in muscle of fishes were analyzed at Fort McMurray, Fort McKay, Fort Chipewyan, Fort Smith and Fort Resolution. Fishes to be studied were chosen based on their abundance along the basin and their cultural and economic significance to Aboriginal communities. They are therefore of interest in monitoring contaminant levels and assessing the potential for impacts on human health.

Materials and methods

Chemicals and reagents

All solvents used were of HPLC grade (Fisher Scientific, Canada). PAH quantification was calibrated using a five-point standard calibration curve (2, 10, 40, 200 and 800) ng/ml (Wellington Laboratories, Guelph, Canada) containing naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorine (Fl), phenanthrene (P), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]-pyrene (BaP), indeno[1,2,3-c,d]pyrene (InP), dibenzo[a,h]anthracene (DBA) and benzo[g,h,i]perylene (BgP)]. The calibration standards also contained a 100 ng/ml mixture of isotopically labeled deuterated PAH standards (naphthalene-d_8, acenaphthene-d_{10}, fluorene-d_{10}, phenanthrene-d_{10}, anthracene-d_{10}, fluoranthene-d_{10}, pyrene-d_{10}, benz[a]anthracene-d_{12}, chrysene-d_{12}, benzo[b]fluoranthene-d_{12}, benzo[k]fluoranthene-d_{12}, benzo[a]pyrene-d_{12}, indeno[1,2,3-c,d]pyrene-d_{12}, benzo[g,h,i]perylene-d_{12}, dibenz[a,h]anthracene-d_{14} and dibenzo[a, i]pyrene-d_{14}) and three deuterated PAH internal standards (acenaphthylene-d_8, p-terphenyl-d_{14} and benzo[e]pyrene-d_{12}). Recovery standards, containing deuterated PAHs and deuterated PAH internal standards, were also purchased from Wellington Laboratories, Guelph, Canada. The 2000 ng/ml stock solution of the recovery standards was diluted to produce a mixture of 100 ng/ml mixture of surrogate standards. Silica gel (80–100 mesh; Sigma-Aldrich, Canada) and anhydrous sodium sulfate (12–60 mesh, Sigma-Aldrich, Canada) were baked in a muffle furnace at 450 °C overnight before use. Acid- and base-modified silica was made at ratios of 1:2 (98 %) sulfuric acid (EMD, Canada): silica gel and 1:3 (1 N) sodium hydroxide (Sigma-Aldrich, Canada): silica gel, respectively. Modified silica was then mixed on a roller for 3 h and used immediately.

Collection of fish

Fish were collected from five locations along the Athabasca and Slave Rivers in cooperation with First Nations fishers and regional and federal agencies. Selected fishes were collected from locations near Fort McMurray, Fort McKay, Fort Chipewyan, Fort Smith and Fort Resolution (Fig. 1). Sampling was seasonal: June/July (summer), October (fall) 2011 and May (spring) 2012. Fish were collected using gill nets (4.25 inch mesh) and were placed on ice for transport to the field laboratory. After euthanasia, each fish was measured, weighted, photographed and examined for the presence of external lesions or abnormalities. Fish were then opened ventrally from the vent to the pericardium, and the left side of the body was removed to reveal the internal organs for examination. Examinations were compatible with Canada's Environmental Effects Monitoring (EEM) procedures (www.ec.gc.ca/eem). Muscle tissue samples were collected from the mid-body dorsal area and were frozen at −18 °C in pre-cleaned amber jars for PAH analysis.

Extraction procedure

For analysis samples of fish muscle were homogenized and dried with excess anhydrous Na_2SO_4. About 15 g wet mass (wm) of fish was then extracted for 18 h in a Soxhlet apparatus with 250 ml dichloromethane (DCM). Deuterated PAHs (naphthalene-d_8, acenaphthene-d_{10}, fluorene-d_{10}, phenanthrene-d_{10}, anthracene-d_{10}, fluoranthene-d_{10}, pyrene-d_{10}, benz[a]anthracene-d_{12}, chrysene-d_{12}, benzo[b]fluoranthene-d_{12}, benzo[k]fluoranthene-d_{12}, benzo[a]pyrene-d_{12}, indeno[1,2,3-c,d]pyrene-d_{12}, benzo[g,h,i]perylene-d_{12}, dibenz[a,h]anthracene-d_{14} and dibenzo[a,i]pyrene-d_{14}) were added as recovery surrogate standards to all the samples prior to extraction. The extract was concentrated to approximately 1–2 ml by

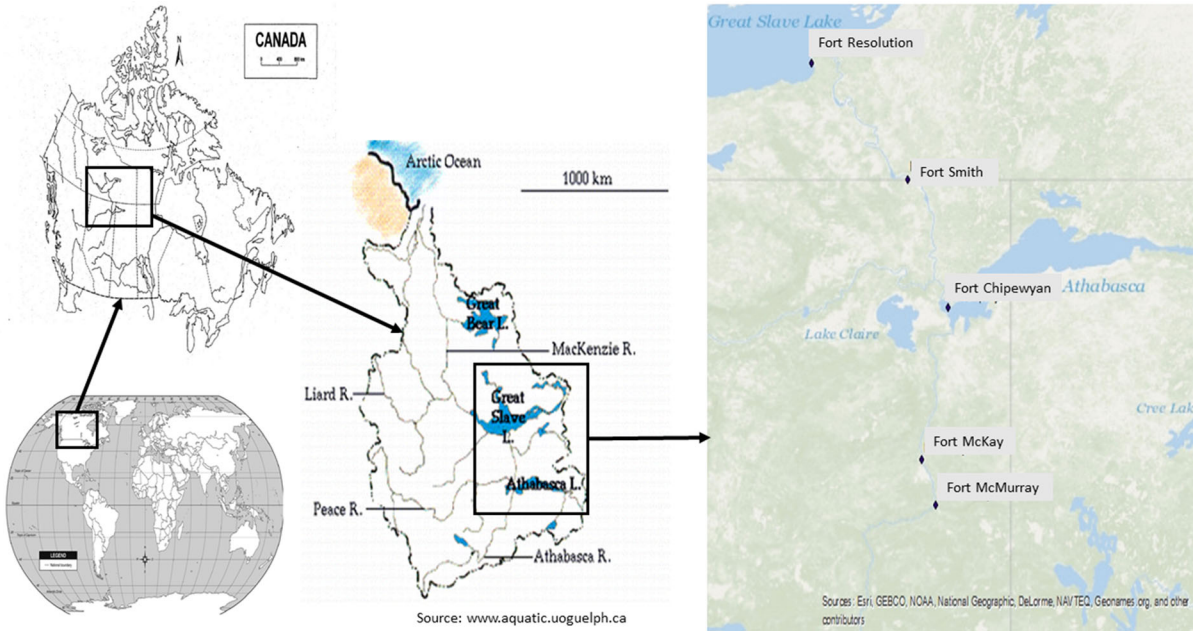

Fig. 1 Locations in the Athabasca/Slave River system, Canada, from which target fishes were collected

rotary evaporation. A mixed bed silica column was used for cleanup. Two grams of basic silica was placed on 1 g of unmodified silica in a glass column (22 cm × 1.5 cm, i.d.). Another 1 g silica was added, and then, 4 g acid–silica was loaded over the basic silica layers. The column was then topped with 2 g of anhydrous sodium sulfate. The column was eluted with 30 ml of n-hexane, which was discarded, before the sample was loaded. The fraction containing PAHs was collected by eluting the column with 150 ml of hexane/dichloromethane (1:1). Extract was then concentrated to 1 ml by rotary evaporation, and 0.1 ml of nonane containing deuterated PAH internal standards (acenaphthylene-d_8, p-terphenyl-d_{14}, benzo[e]pyrene-d_{12}) was then added to the extract before further concentration to 0.1 ml under a gentle stream of nitrogen.

GC–MS analysis

PAHs were identified and quantified using a Hewlett Packard (HP) 7890A GC fitted with a 60 m, 0.25 mm i.d. DB-5 silica capillary column and an HP 7683 series autosampler. The injection temperature was 250 °C, and the detector temperature was 280 °C. The temperature ramp was: 60 °C for 2 min, 20 °C/min to 160 °C followed by 5 °C/min to 268 °C and 2 °C/min to 300 °C, where it was held for 10 min to give a total run of 55.5 min. The HP 5975 series mass selective detector was operated in selected ion mode (SIM). A 1 μL sample of extract or standard was injected in split/splitless mode. Mass spectra were acquired in electron impact (EI) mode at 70 eV.

Quality assurance and quality control

All analytical data were subject to strict quality control. Method blanks (solvent) and spiked blanks (standards spiked into solvent and reagent) were used to determine background contamination. Some samples were analyzed in duplicate. Instruments were calibrated frequently with certified standards. PAHs were quantified using the internal calibration method based on five-point calibration curves for individual compounds. The surrogate recoveries averaged 84 ± 16 %. Instrument detection limits ranged from 0.1 to 2.0 ng/g, wet mass (wm).

Dietary exposure estimates

Because health risk criteria are not available for all the individual PAH compounds, the potential carcinogenic risk of PAH mixtures is often expressed using a

toxicity equivalent factor approach, and this is done by relating the potencies of individual PAHs to that of benzo(a)pyrene (B(a)P, which has the greatest potency of the PAHs to cause cancer (Agency for Toxic Substances and Disease Registry (ATSDR) 1996). Toxicity equivalence factors (TEFs) relative to B[a]P have been developed for assessing risks posed by mixtures of PAHs (Table 1) (Nisbet and LaGoy 1992). These TEFs were adopted to calculate the potential toxicity of the PAH mixtures measured in this study as total benzo[a]pyrene equivalents ($B[a]P_{eq}$). This approach has been suggested to be superior for assessing the carcinogenic potency of PAH mixtures (Binelli and Provini 2004; Xia et al. 2010).

Methods for assessment of risks advocated by both Health Canada and the USEPA were used for assessing the carcinogenic risk to humans in the Athabasca/Slave River system due to the consumption of PAHs in fish. To evaluate the potential impacts on the inhabitants of Fort McMurray, Fort McKay, Fort Chipewyan, Fort Smith and Fort Resolution, it was deemed most appropriate to use Canadian population data, based on local populations, who repeatedly consume fish from the same aquatic source, most of their lives. This approach is more focused on local conditions and customs. The per capita consumption of fish to estimate the contribution of PAH to the daily intake (DI) is needed. In this regard, since there are no measured data for fish consumption in the Athabasca/Slave Rivers, estimations were used. The most precise and reliable data on consumption and body weight by various groups in Canada were used (Richardson 1997, 2013) (Appendix 1, 2). In addition, a range of fish consumption rates and representative body mass were used to monitor the potential risk to fish consumers in the sampled locations (Tables 2, 3).

Table 1 PAHs and their toxic equivalent factors (TEFs) (Nisbet and LaGoy 1992)

PAHs	TEFs
Naphthalene (NAP)	0.001
Acenaphthene (ACE)	0.001
Acenaphthylene (ACY)	0.001
Fluorine (FLO)	0.001
Phenanthrene (PHE)	0.001
Anthracene (ANT)	0.01
Fluoranthene (FLA)	0.001
Pyrene (PYR)	0.001
Benz(a)anthracene (BaA)	0.1
Chrysene (CHR)	0.01
Benzo(b)fluoranthene (BbF)	0.1
Benzo(k)fluoranthene (BkF)	0.1
Benzo(a)pyrene (BaP)	1
Dibenz(a,h)anthracene (DahA)	5
Indeno(1,2,3-cd)pyrene (IcdP)	0.1
Benzo(g,h,i)perylene (Bghip)	0.01

Concentrations of BaP_{eq} in fishes and the potential daily intake (DI) of PAHs via consumption of fish for specific populations were estimated (Eqs. 1 and 2), respectively.

$$\mathrm{BEC}_i = \sum_{i=1}^{n} \mathrm{C}i \times \ \mathrm{TEF}_i \tag{1}$$

$$\mathrm{DI} = \sum_{i=1}^{n} \mathrm{BEC}i \times \mathrm{FC} \tag{2}$$

where BECi is the concentration of $B[a]P_{eq}$ in fish (ng/g, wm); C_i is the concentration of PAH congener i in fish; TEFi = TEF of PAH congener i (Table 1) (Nisbet and LaGoy 1992). FC is fish consumption per day (g/d).

The lifetime cancer risk (LCR) of population groups in the Athabasca and Slave Rivers caused by exposure due to fish consumption was calculated (Eq. 3).

$$\mathrm{LCR} = \frac{\mathrm{DI} \ \times \mathrm{ED} \ \times \mathrm{EF}}{\mathrm{BM} \ \times \mathrm{AT}} \times CF \times SFB[a]P \tag{3}$$

where DI is the daily intake of PAHs via fish consumption (ng/g); ED is duration of exposure (years); EF is the exposure frequency (days/year); BM is the average body mass (kg); AT is averaging time (days); CF is the conversion factor (10^{-6} kg/mg); cancer-causing ability of B[a]P was used in the determination of oral slope factor. The oral slope factor for B[a]P is 4.5, 5.9, 9.0 and 11.7 with a geometric mean of 7.3 $(\mathrm{mg/kg/day})^{-1}$. The human population in the region of interest, from which samples of fish were collected, was divided into 4 groups according to age: children (4 to <12 years), teens (12 to <20 years), adults (20 to <65 years) and seniors (≥65 years) (Tables 2, 3, 4).

Table 2 Estimated lognormal probability density functions describing a range of possible fish consumption rates (g/day) for groups in the study locations

Gender	Children			Teens			Adults			Seniors		
	Low	Medium	High	Low	Medium	High	Low	Medium	High	Low	Medium	High
Female	70	128	220	50	150	300	60	300	600	50	200	500
Male	70	128	220	50	190	350	60	400	800	50	300	700

Table 3 Estimated lognormal probability density functions describing a range of body mass (kg) for groups in the study locations

Gender	Children			Teens			Adults			Seniors		
	Low	Medium	High	Low	Medium	High	Low	Medium	High	Low	Medium	High
Female	20	35	50	30	55	75	50	100	200	50	80	150
Male	15	32	45	40	60	80	60	150	250	60	90	160

Data analyses

Differences in the concentration of PAHs among sampling locations, seasons and species were evaluated using the Kruskal–Wallis nonparametric test. All data were log-transformed to approximate a normal distribution for risk assessment. Pearson's correlation analysis was used to test the relationship between fish mass, length, liver somatic index (LSI) and concentrations of PAHs. Box-Whiskers plots were used for descriptive statistical analysis (McGill et al. 1978). All statistical analyses were conducted with Microsoft Excel, SigmaPlot for Windows, version 11.0, or SYSTAT for Windows, version 12.0.

Results

PAHs were detected in all samples of fish muscle collected from the Athabasca and Slave Rivers at each location and during each season (Table 5). Biological parameters for fishes collected at Fort Resolution, Fort Smith, Fort Chipewyan, Fort McKay and Fort McMurray in 2011–2012 are available in Appendix 3. A total of 425 samples of fish muscle among seasons, locations and species were analyzed (Table 6). The mean concentration of $\sum$PAHs in muscle of the 425 samples, averaged across species, seasons and locations, was 30 ng/g, wet mass (wm). Mean concentrations among species, locations and seasons were: $\sum$2-ring (Naphthalene) 5.8 $\pm$ 1.5 ng/g, wm, $\sum$3-ring (acenaphthylene, acenaphthene, fluorene, phenanthrene and anthracene) 11 $\pm$ 2.2 ng/g, wm, $\sum$4-ring (fluoranthene, pyrene, benz(a) anthracene and chrysene) 7.2 $\pm$ 2.7 ng/g, wm, $\sum$5-ring PAHs (benzo(b) fluoranthene, benzo(k) fluoranthene, benzo(a) pyrene and dibenz(ah) anthracene) 4.6 $\pm$ 1.4 ng/g, wm and $\sum$6-ring (indeno(1, 2, 3-cd) pyrene and benzo(ghi) perylene) PAHs 1.4 $\pm$ 0.7 ng/g, wm (Fig. 2). Among all samples, measured concentrations

Table 4 Parameters used in the incremental lifetime cancer risk assessment. The risk for each group was calculated separately

Definition	Units	Children	Teens	Adults	Seniors
Exposure frequency (EF)	Days/year	365	365	365	365
Exposure duration (ED)	Year	70	70	70	70
Averaging time (AT)	Days	25,550	25,550	25,550	25,550

Table 5 Comparison of PAHs in muscles of fish collected at Fort Resolution, Fort Smith, Fort Chipewyan, Fort McKay and Fort McMurray in 2011–2012

Species	Site	Nap	Acy	Ace	Flu	Phe	Ant	Flua
A. Summer								
Burbot	FR	n.d	n.d	1.4 ± 2.8	1.0 ± 2.0	n.d	n.d	0.3 ± 0.2
	FS	n.d	n.d	2.0 ± 3.8	3.3 ± 6.6	n.d	0.9 ± 1.9	0.4 ± 0.4
	FC	5.9 ± 0.5	2.3 ± 2.0	n.d	n.d	n.d	n.d	0.4 ± 0.1
	FM	n.a	n.a	n.a	n.a	n.a	n.a	n.a
	FMU	5.9 ± 0.5	2.3 ± 2.0	n.d	n.d	n.d	n.d	0.5 ± 0.6
Goldeye	FR	11 ± 8.5	4.7 ± 3.8	7.9 ± 5.8	1.3 ± 1.4	n.d	n.d	n.d
	FS	n.d	n.d	n.d	n.d	n.d	0.2 ± 0.2	0.5 ± 0.6
	FC	1.9 ± 1.5	2.3 ± 2.1	1.1 ± 0.8	n.d	n.d	1.1 ± 1.4	1.0 ± 1.0
	FM	2.5 ± 1.9	2.5 ± 1.3	4.3 ± 7.2	3.0 ± 5.1	1.0 ± 1.3	2.3 ± 3.4	1.0 ± 0.9
	FMU	4.5 ± 5.4	9.9 ± 16.5	4.6 ± 4.4	5.9 ± 9.1	1.9 ± 1.8	3.7 ± 4.6	1.6 ± 2.2
Jackfish	FR	n.d	n.d	n.d	n.d	n.d	n.d	0.2 ± 0.2
	FS	1.7 ± 2.2	1.0 ± 0.4	0.8 ± 0.8	1.2 ± 1.6	0.5 ± 0.7	0.7 ± 1.4	n.d
	FC	2.1 ± 1.5	4.8 ± 3.6	1.4 ± 0.9	n.d	n.d	n.d	0.4 ± 0.6
	FM	7.6 ± 8.9	4.2 ± 4.4	2.9 ± 1.5	2.8 ± 4.3	2.0 ± 3.0	1.6 ± 1.7	0.8 ± 0.7
	FMU	14.4 ± 25.0	13.4 ± 15.7	7.7 ± 11.3	6.0 ± 12.0	2.3 ± 4.0	4.7 ± 6.6	1.0 ± 0.9
Walleye	FR	n.a	n.a	n.a	n.a	n.a	n.a	n.a
	FS	n.d	n.d	n.d	n.d	n.d	0.3 ± 0.4	0.7 ± 0.6
	FC	2.8 ± 2.2	3.4 ± 3.4	1.7 ± 1.2	n.d	n.d	0.8 ± 1.1	2.1 ± 2.7
	FM	8.2 ± 6.8	5.3 ± 4.1	4.1 ± 2.9	4.2 ± 7.9	2.4 ± 2.4	3.2 ± 6.0	3.9 ± 53
	FMU	4.3 ± 3.4	4.9 ± 5.9	3.6 ± 4.7	1.8 ± 1.6	0.7 ± 0.7	1.7 ± 1.9	1.5 ± 1.6
White fish	FR	n.d	n.d	n.d	n.d	n.d	n.d	0.3 ± 0.5
	FS	n.d	n.d	1.8 ± 3.1	1.0 ± 2.2	n.d	1.0 ± 1.5	0.3 ± 0.3
	FC	3.3 ± 2.8	3.1 ± 2.6	3.6 ± 4.2	2.7 ± 4.5	0.5 ± 0.7	0.6 ± 0.6	0.7 ± 0.6
	FM	4.2 ± 3.3	5.3 ± 3.6	8.4 ± 13	4.2 ± 4.7	3.0 ± 3.4	1.7 ± 2.3	1.7 ± 1.8
	FMU	n.a	n.a	n.a	n.a	n.a	n.a	n.a
B. Fall								
Burbot	FR	2.6 ± 1.7	1.5 ± 1.3	1.7 ± 1.4	2.1 ± 1.3	3.3 ± 2.6	3.7 ± 4.7	1.4 ± 1.3
	FS	3.1 ± 1.0	1.7 ± 0.5	3.1 ± 0.2	1.9 ± 0.0	11.8 ± 8.2	14.1 ± 6.4	0.5 ± 0.4
	FC	5.2 ± 1.6	2.3 ± 0.5	3.7 ± 1.0	1.8 ± 0.2	2.4 ± 0.9	5.6 ± 0.8	1.9 ± 0.1
	FM	3.0 ± 0.7	1.8 ± 1.1	2.2 ± 2.1	2.4 ± .2	1.5 ± 0.6	2.4 ± 1.4	0.9 ± 0.2
	FMU	n.a	n.a	n.a	n.a	n.a	n.a	n.a
Goldeye	FR	1.1 ± 0.7	2.0 ± 1.6	0.9 ± 0.7	0.9 ± 1.1	n.d	0.7 ± 0.4	0.4 ± 0.4
	FS	2.1 ± 1.3	3.0 ± 2.5	1.0 ± 0.7	n.d	n.d	1.8 ± 2.8	1.5 ± 1.8
	FC	3.2 ± 1.6	1.8 ± 1.3	3.4 ± 3.2	1.4 ± 0.5	1.6 ± 0.7	2.3 ± 2.7	0.6 ± 0.7
	FM	3.9 ± 3.8	2.8 ± 2.9	1.8 ± 1.9	1.6 ± 2.4	1.6 ± 1.4	1.8 ± 1.5	0.6 ± 0.7
	FMU	10.3	1.5	n.d	n.d	n.d	n.d	n.d
Jackfish	FR	n.d	1.5 ± 0.9	n.d	n.d	n.d	n.d	0.4 ± 0.7
	FS	n.d	2.1 ± 0.8	n.d	1.0 ± 1.5	n.d	n.d	0.5 ± 1.2
	FC	5.1 ± 4.6	2.6 ± 1.9	2.4 ± 3.6	2.8 ± 5.7	n.d	0.8 ± 1.6	0.2 ± 0.2
	FM	8.5 ± 6.2	4.9 ± 7.7	11.5 ± 14.5	6.6 ± 6.9	3.3 ± 4.7	3.9 ± 2.1	4.0 ± 4.9
	FMU	n.d	2.9 ± 2.2	2.2 ± 2.1	n.d	1.1 ± 0.2	1.0 ± 0.0	1.0 ± 1.2
Walleye	FR	n.d	n.d	n.d	n.d	n.d	n.d	n.d
	FS	1.6 ± 1.7	1.2 ± 0.5	1.9 ± 1.9	2.1 ± 1.6	5.0 ± 4.2	5.5 ± 5.5	0.3 ± 0.3

Table 5 continued

Species	Site	Nap	Acy	Ace	Flu	Phe	Ant	Flua
	FC	n.d	1.7 ± 1.0	1.1 ± 0.7	0.9 ± 0.4	1.6 ± 1.9	1.7 ± 1.9	0.4 ± 0.3
	FM	3.8 ± 2.8	3.4 ± 3.0	2.8 ± 2.5	3.4 ± 3.2	3.6 ± 3.1	3.4 ± 4.3	1.5 ± 1.6
	FMU	1.8 ± 0.2	6.6 ± 3.3	3.7 ± 2.3	3.6 ± 2.3	2.5 ± 2.0	5.2 ± 3.0	0.7 ± 0.2
White fish	FR	2.4 ± 1.7	1.5 ± 0.9	1.5 ± 1.5	2.8 ± 3.8	0.5 ± 0.6	0.6 ± 0.5	0.4 ± 0.5
	FS	n.d	n.d	1.6 ± 1.7	1.3 ± 1.5	3.9 ± 4.6	4.9 ± 5.9	0.7 ± 1.1
	FC	2.3 ± 1.8	1.9 ± 1.6	2.4 ± 3.2	1.1 ± 0.9	0.9 ± 1.1	1.3 ± 2.1	1.0 ± 0.9
	FM	4.9 ± 4.9	3.3 ± 3.6	3.2 ± 3.4	3.4 ± 3.2	2.0 ± 1.7	4.3 ± 3.5	2.6 ± 3.5
	FMU	2.9 ± 3.4	2.2 ± 2.2	1.4 ± 1.1	3.5 ± 4.5	1.7 ± 2.3	3.2 ± 6.4	2.1 ± 3.0
C. Spring								
Burbot	FR	1.2 ± 0.7	1.1 ± 0.6	2.3 ± 2.0	1.0 ± 1.3	0.9 ± 1.4	1.0 ± 1.4	n.d
	FS	n.d	n.d	n.d	0.9	n.d	n.d	n.d
	FC	n.a	n.a	n.a	n.a	n.a	n.a	n.a
	FM	n.a	n.a	n.a	n.a	n.a	n.a	n.a
	FMU	25.0 ± 21.4	2.5 ± 0.7	1.2 ± 0.1	5.3 ± 1.9	1.8 ± 1.1	2.9 ± 1.5	4.3 ± 1.9
Goldeye	FR	1.3 ± 0.8	1.0 ± 0.4	0.8 ± 0.6	0.7 ± 0.3	0.2 ± 1.7	0.2 ± 2.2	0.5 ± 0.5
	FS	n.d	1.6 ± 0.3	0.7 ± 0.5	n.d	n.d	n.d	n.d
	FC	n.d	n.d	1.1 ± 1.7	n.d	n.d	n.d	n.d
	FM	20.9 ± 12.6	1.7 ± 1.0	2.1 ± 1.5	2.9 ± 1.9	1.7 ± 2.3	1.9 ± 1.2	1.5 ± 1.8
	FMU	23.0 ± 15.5	1.8 ± 0.7	2.8 ± 3.9	4.6 ± 4.6	1.8 ± 1.0	2.4 ± 1.5	0.8 ± 0.6
Jackfish	FR	1.0 ± 0.7	1.5 ± 0.9	1.4 ± 1.2	1.0 ± 1.0	0.6 ± 0.5	0.4 ± 0.5	0.1 ± 0.2
	FS	1.8 ± 0.7	0.9 ± 0.3	1.5 ± 1.3	1.6 ± 2.5	0.9 ± 0.6	0.6 ± 0.5	0.3 ± 0.3
	FC	4.4 ± 3.0	3.5 ± 4.5	1.2 ± 1.0	1.2 ± 1.4	1.0 ± 1.6	0.6 ± 0.8	0.5 ± 1.0
	FM	26.1 ± 15.9	2.5 ± 0.3	1.7 ± 1.0	4.7 ± 4.0	3.7 ± 1.6	8.3 ± 0.8	1.9 ± 1.3
	FMU	35.4 ± 17.3	2.3 ± 1.2	1.6 ± 1.6	4.1 ± 2.2	3.3 ± 1.3	4.8 ± 2.6	3.4 ± 1.4
Walleye	FR	1.1 ± 1.0	1.5 ± 0.9	0.9 ± 0.7	0.7 ± 0.7	0.6 ± 0.7	0.6 ± 0.7	n.d
	FS	1.0 ± 1.0	n.d	1.9 ± 1.7	1.7 ± 1.7	1.7 ± 1.7	n.d	0.4 ± 0.1
	FC	2.6 ± 2.0	2.3 ± 2.1	2.2 ± 3.4	1.8 ± 3.0	1.8 ± 3.3	1.8 ± 2.9	0.9 ± 0.6
	FM	24.9 ± 18.4	1.7 ± 0.5	1.5 ± 0.9	2.7 ± 1.0	1.7 ± 1.3	3.6 ± 1.0	4.1 ± 5.0
	FMU	11.3 ± 7.0	2.6 ± 1.1	1.2 ± 0.9	1.5 ± 1.2	2.3 ± 1.0	3.1 ± 1.5	3.1 ± 1.2
White fish	FR	2.0 ± 2.7	1.6 ± 1.7	2.4 ± 3.1	2.2 ± 2.7	1.8 ± 1.9	1.4 ± 1.5	0.2 ± 0.3
	FS	1.3 ± 1.0	1.4 ± 1.4	1.2 ± 1.7	0.8 ± 1.0	0.1 ± 0.1	0.3 ± 0.5	0.1 ± 0.1
	FC	2.2 ± 1.8	0.6 ± 0.4	2.3 ± 3.1	0.9 ± 0.6	0.8 ± 0.9	0.6 ± 0.5	0.6 ± 0.8
	FM	29.3 ± 14.5	6.2 ± 5.6	18.3 ± 21.8	4.9 ± 2.8	1.3 ± 0.0	7.5 ± 9.8	0.3 ± 0.3
	FMU	23.0 ± 14.8	6.7 ± 9.3	9.6 ± 16.4	7.1 ± 8.6	3.3 ± 2.7	6.5 ± 10.8	1.9 ± 2.1

Species	Pyr	BaA	Chr	BbF	BkF	BaP	DbahA	Ind	BghiP
A. Summer									
Burbot	0.5 ± 0.5	n.d	n.d	0.2 ± 0.3	n.d	0.2 ± 0.3	1.0 ± 2.6	0.6 ± 1.1	0.2 ± 0.4
	0.2 ± 0.2	0.2 ± 0.2	0.2 ± 0.1	n.d	n.d	0.1 ± 0.2	0.2 ± 0.3	0.2 ± 0.3	0.1 ± 0.1
	0.2 ± 0.3	0.2 ± 0.3	0.15 ± 0.1	0.14 ± 0.1	0.2 ± 0.0	1.36 ± 0.0	n.d	n.d	n.d
	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
	0.7 ± 1.1	0.1 ± 0.2	n.d	0.2 ± 0.1	n.d	0.6 ± 0.7	0.2 ± 0.1	n.d	n.d
Goldeye	n.d	0.14 ± 0.1	0.18 ± 0.2	3.36 ± 4.7	2.6 ± 3.6	0.7 ± 0.4	n.d	n.d	n.d
	0.3 ± 0.3	0.8 ± 1.8	0.4 ± 0.8	0.1 ± 01	n.d	n.d	n.d	0.3 ± 0.4	n.d
	1.5 ± 1.7	1.8 ± 2.9	0.5 ± 0.4	0.5 ± 0.4	0.3 ± 0.5	1.1 ± 1.4	n.d	0.3 ± 0.4	0.1 ± 0.1

Table 5 continued

Species	Pyr	BaA	Chr	BbF	BkF	BaP	DbahA	Ind	BghiP
	3.9 ± 8.1	2.2 ± 3.8	0.8 ± 0.6	0.9 ± 1.0	0.7 ± 0.6	1.3 ± 1.2	1.5 ± 2.0	1.1 ± 1.3	0.5 ± 1.2
	5.2 ± 12.6	2.8 ± 3.5	1.4 ± 2.1	1.4 ± 0.9	1.2 ± 0.9	2.7 ± 2.2	1.7 ± 2.8	1.0 ± 2.1	0.4 ± 0.5
Jackfish	0.2 ± 0.2	0.5 ± 1.1	0.3 ± 0.7	0.3 ± 0.5	0.2 ± 0.4	0.6 ± 1.5	0.3 ± 0.6	0.2 ± 0.3	0.07 ± 0.1
	0.1 ± 0.1	1.3 ± 2.4	0.6 ± 1.5	0.1 ± 0.1	0.2 ± 0.3	0.3 ± 0.5	0.3 ± 0.6	0.2 ± 0.3	.2 ± 0.3
	1.0 ± 1.4	03 ± 0.3	0.1 ± 0.1	0.1 ± 0.3	n.d	0.1 ± 0.1	01 ± 0.1	0.2 ± 0.5	0.1 ± 01
	1.6 ± 2.5	1.7 ± 2.5	1.7 ± 2.2	1.1 ± 1.3	1.0 ± 1.0	0.3 ± 0.4	2.0 ± 4.1	3.2 ± 4.2	0.6 ± 0.8
	4.4 ± 4.4	0.9 ± 1.1	0.4 ± 0.3	0.5 ± 0.4	0.4 ± 0.4	1.2 ± 0.9	08 ± 2.1	0.1 ± 0.1	0.2 ± 0.2
Walleye	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
	0.8 ± 0.9	0.8 ± 0.9	0.3 ± 0.3	0.1 ± 0.1	0.1 ± 0.1	0.2 ± .3	0.1 ± 0.1	n.d	0.1 ± 0.2
	12.2 ± 10.0	0.6 ± 0.6	0.5 ± 0.5	0.4 ± 0.4	0.4 ± 0.6	0.6 ± 0.3	0.3 ± 0.3	0.6 ± 0.1	0.2 ± 0.4
	5.0 ± 4.6	2.9 ± 3.2	0.9 ± 1.0	1.7 ± 2.1	0.9 ± 1.3	0.7 ± 0.9	0.2 ± 0.3	0.3 ± 0.4	0.3 ± 0.4
	2.3 ± 3.3	1.8 ± 2.3	1.1 ± 1.5	1.1 ± 0.9	0.6 ± 0.4	0.2 ± 0.3	0.9 ± 2.0	0.5 ± 0.7	0.4 ± 0.5
White fish	0.6 ± 0.7	0.5 ± 0.7	0.2 ± 0.4	0.1 ± 0.1	n.d	0.1 ± 0.1	0.2 ± 0.5	0.1 ± 0.2	n.d
	0.2 ± 0.3	1.8 ± 2.8	1.1 ± 2.0	n.d	0.2 ± 0.3	0.1 ± 0.2	0.2 ± 0.3	0.3 ± 0.6	0.1 ± 0.2
	5.6 ± 5.9	1.6 ± 1.3	0.8 ± 0.5	2.3 ± 4.5	1.3 ± 1.9	4.7 ± 8.6	0.6 ± 0.9	0.8 ± 0.9	0.2 ± 0.2
	4.5 ± 7.8	6.6 ± 11.3	5.6 ± 7.5	3.4 ± 3.6	2.1 ± 3.3	3.9 ± 4.7	2.6 ± 2.5	3.5 ± 3.8	1.6 ± 3.2
	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
B. Fall									
Burbot	0.7 ± 0.6	3.4 ± 1.3	1.8 ± 0.9	0.9 ± 0.5	1.1 ± 1.1	1.0 ± 0.9	1.2 ± 1.3	0.8 ± 0.6	0.8 ± 0.6
	0.5 ± 0.4	6.3 ± 8.2	4.1 ± 3.0	2.1 ± 1.4	1.1 ± 1.6	1.9 ± 0.7	2.1 ± 3.0	n.d	0.2 ± 0.3
	2.6 ± 0.4	5.5 ± 1.6	2.7 ± 1.0	4.8 ± 0.8	3.1 ± 0.4	1.6 ± 0.4	2.4 ± 0.4	n.d	1.2 ± 0.9
	1.9 ± 2.1	3.7 ± 2.6	1.3 ± 0.4	4.1 ± 3.0	2.8 ± 1.9	3.6 ± 0.0	3.5 ± 4.7	2.1 ± 2.3	0.4 ± 0.4
	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
Goldeye	1.4 ± 1.9	0.4 ± 0.5	0.5 ± 0.4	2.1 ± 3.3	1.7 ± 2.7	0.6 ± 0.3	3.1 ± 5.3	2.1 ± 1.6	0.3 ± 0.3
	0.2 ± 0.2	8.9 ± 6.0	4.4 ± 3.5	1.9 ± 1.4	0.9 ± 0.4	0.7 ± 0.4	5.8 ± 4.6	0.4 ± 0.3	0.9 ± 0.9
	1.4 ± 1.4	3.5 ± 2.1	0.5 ± 0.4	1.4 ± 1.8	1.2 ± 1.3	0.6 ± 0.4	0.4 ± 0.4	0.4 ± 0.4	0.3 ± 0.3
	1.9 ± 2.7	4.7 ± 5.7	3.1 ± 3.7	2.2 ± 1.9	2.2 ± 1.5	2.6 ± 3.8	2.7 ± 5.8	2.3 ± 2.7	0.7 ± 1.0
	n.d	n.d	0.2	0.1	0.1	0.9	n.d	n.d	n.d
Jackfish	n.d	1.0 ± 1.0	0.5 ± 0.3	0.4 ± 0.6	0.4 ± 0.4	0.3 ± 0.3	0.6 ± 0.6	1.1 ± 1.0	0.5 ± 0.4
	0.3 ± 0.8	0.5 ± 1.2	1.1 ± 2.1	0.8 ± 1.3	1.7 ± 2.6	0.3 ± 0.3	0.8 ± 1.4	1.2 ± 1.2	1.2 ± 1.6
	0.2 ± 0.3	0.1 ± 0.1	0.1 ± 0.1	0.3 ± 0.3	0.3 ± 0.3	0.6 ± 0.6	n.d	n.d	0.1 ± 0.1
	3.6 ± 2.0	3.0 ± 3.4	2.1 ± 2.7	0.7 ± 0.6	0.8 ± 0.6	3.3 ± 3.2	2.1 ± 3.3	2.3 ± 3.4	0.6 ± 0.6
	0.3 ± 0.1	5.2 ± 1.1	2.6 ± 0.1	0.3 ± 0.2	0.3 ± 0.0	0.9 ± 1.1	0.9 ± 0.8	1.3 ± 1.3	0.6 ± 0.6
Walleye	n.d	1.03 ± 1.9	0.4 ± 0.1	0.2 ± 0.2	n.d	0.2 ± 0.2	0.2 ± 0.3	1.3 ± 1.1	0.9 ± 0.8
	0.4 ± 0.5	5.1 ± 3.9	2.3 ± 2.2	3.2 ± 2.6	2.2 ± 1.7	2.1 ± 2.1	2.3 ± 2.7	1.9 ± 2.0	1.4 ± 2.0
	0.4 ± 0.3	1.5 ± 0.9	0.9 ± 0.8	1.3 ± 0.7	0.7 ± 0.6	1.5 ± 1.6	2.8 ± 2.9	2.1 ± 1.9	1.0 ± 0.3
	1.4 ± 1.7	3.9 ± 3.9	2.5 ± 3.2	0.9 ± 1.2	1.1 ± 1.1	1.0 ± 0.8	1.3 ± 1.7	3.1 ± 2.4	0.7 ± 0.9
	0.7 ± 0.6	0.4 ± 0.0	0.5 ± 0.5	0.6 ± 0.6	0.5 ± 0.5	1.0 ± 1.0	1.5 ± 1.7	3.6 ± 4.4	2.8 ± 2.1
White fish	0.1 ± 0.1	1.6 ± 1.7	1.0 ± 1.3	0.6 ± 0.9	0.8 ± 1.3	0.6 ± 0.5	1.2 ± 2.1	0.8 ± 0.7	0.9 ± 1.5
	0.5 ± 1.1	4.0 ± 4.7	1.3 ± 1.2	0.2 ± 0.2	0.1 ± 0.1	0.4 ± 0.4	0.1 ± 0.1	0.7 ± 1.1	0.4 ± 0.4
	3.0 ± 3.3	3.5 ± 3.4	2.3 ± 2.1	1.1 ± 0.7	1.4 ± 0.9	3.3 ± 6.6	1.9 ± 2.1	5.1 ± 8.2	0.6 ± 0.6
	2.6 ± 3.9	4.0 ± 6.6	3.3 ± 5.3	2.6 ± 2.6	1.9 ± 1.9	3.4 ± 7.1	0.9 ± 1.2	1.8 ± 2.2	0.5 ± 0.6
	2.8 ± 3.6	2.3 ± 2.1	2.6 ± 2.4	1.5 ± 1.5	1.3 ± 1.4	1.1 ± 1.3	1.0 ± 1.6	0.6 ± 0.8	0.5 ± 0.7

Table 5 continued

Species	Pyr	BaA	Chr	BbF	BkF	BaP	DbahA	Ind	BghiP
C. Spring									
Burbot	1.3 ± 2.3	n.d	n.d	0.1 ± 01	0.1 ± 0.1	0.1 ± 0.0	n.d	n.d	0.1 ± 0.0
	0.1	0.3	0.3	n.d	n.d	0.1	0.5	1.0	n.d
	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
	2.0 ± 1.2	36.4 ± 32.0	22.3 ± 13.0	6.8 ± 3.9	2.9 ± 3.3	1.2 ± 1.0	n.d	0.3 ± 0.2	1.5 ± 0.9
Goldeye	0.8 ± 0.9	0.3 ± 0.3	0.1 ± 0.1	0.8 ± 0.6	0.5 ± 0.3	1.1 ± 0.9	0.2 ± 0.3	1.3 ± 1.4	0.1 ± 0.2
	n.d	0.3 ± 0.3	0.2 ± 0.2	0.8 ± 1.5	0.6 ± 1.0	0.2 ± 0.2	0.3 ± 0.2	0.5 ± 0.3	0.9 ± 0.9
	0.8 ± 1.5	0.3 ± 0.1	0.2 ± 0.1	0.2 ± 0.3	0.2 ± 0.3	0.4 ± 0.5	2.5 ± 3.8	1.5 ± 2.5	0.2 ± 0.4
	1.2 ± 1.2	5.9 ± 4.4	3.8 ± 3.3	9.0 ± 7.3	5.5 ± 3.4	4.8 ± 2.6	0.1 ± 0.1	0.7 ± 0.8	0.7 ± 0.8
	0.8 ± 1.2	6.9 ± 6.8	3.1 ± 3.1	7.2 ± 5.0	4.1 ± 4.2	3.3 ± 4.1	0.5 ± 0.8	0.5 ± 0.9	0.7 ± 0.6
Jackfish	0.4 ± 1.0	0.1 ± 0.1	0.1 ± 0.1	0.2 ± 0.2	0.2 ± 0.1	0.4 ± 0.4	0.6 ± 1.1	1.2 ± 0.8	0.2 ± 0.1
	0.2 ± 0.1	0.2 ± 0.2	0.2 ± 0.1	0.2 ± 0.2	0.3 ± 0.3	0.1 ± 0.1	0.4 ± 0.4	0.1 ± 0.2	0.1 ± 0.1
	0.1 ± 0.1	1.7 ± 2.5	0.8 ± 1.5	0.3 ± 0.3	0.4 ± 0.3	0.8 ± 1.7	0.8 ± 1.7	2.1 ± 2.4	0.5 ± 0.6
	0.7 ± 0.6	15.1 ± 6.1	8.3 ± 7.0	2.1 ± 2.3	0.9 ± 0.5	1.0 ± 0.4	0.3 ± 0.3	2.1 ± 3.8	1.1 ± 1.5
	1.2 ± 0.6	5.5 ± 2.9	3.0 ± 1.3	5.3 ± 3.7	3.4 ± 2.7	1.6 ± 0.4	0.4 ± 0.3	0.5 ± 0.5	1.0 ± 0.9
Walleye	0.6 ± 0.9	0.5 ± 0.8	0.2 ± 0.4	0.4 ± 0.4	0.5 ± 0.5	0.9 ± 0.9	0.2 ± 0.2	2.1 ± 2.1	0.3 ± 0.3
	0.1 ± 0.1	0.2 ± 0.3	0.4 ± 0.8	0.2 ± 0.4	0.2 ± 0.1	0.3 ± 0.3	n.d	0.1 ± 0.2	0.1 ± 0.1
	5.1 ± 8.9	0.1 ± 0.1	0.1 ± 0.0	0.4 ± 0.2	0.3 ± 0.1	0.1 ± 0.1	n.d	n.d	0.1 ± 0.2
	1.5 ± 1.3	11.2 ± 5.1	9.9 ± 6.9	8.3 ± 8.1	7.4 ± 4.7	1.9 ± 1.4	0.1 ± 0.1	0.3 ± 0.3	0.5 ± 0.7
	1.1 ± 0.3	2.5 ± 0.4	1.8 ± 0.3	3.0 ± 1.2	1.6 ± 1.4	2.3 ± 0.9	n.d	n.d	n.d
White fish	n.d	0.1 ± 0.2	0.1 ± 0.1	0.3 ± 0.4	0.3 ± 0.4	0.4 ± 0.5	0.1 ± 0.1	0.9 ± 0.9	0.2 ± 0.3
	1.8 ± 3.0	0.1 ± 0.1	0.6 ± 0.9	0.3 ± 0.3	0.1 ± 0.1	0.7 ± 0.9	n.d	0.2 ± 0.4	0.1 ± 0.2
	1.0 ± 1.5	2.4 ± 3.1	1.4 ± 1.9	0.1 ± 0.1	0.1 ± 0.2	0.1 ± 0.2	0.2 ± 0.2	1.5 ± 2.2	0.3 ± 0.4
	2.1 ± 2.9	1.0 ± 1.3	0.8 ± 0.9	3.0 ± 4.1	0.7 ± 0.7	0.7 ± 0.5	n.d	0.3 ± 0.3	0.5 ± 0.6
	1.2 ± 0.6	8.6 ± 7.2	6.0 ± 4.6	4.5 ± 3.6	2.3 ± 2.9	0.8 ± 0.6	0.4 ± 0.5	0.2 ± 0.2	0.5 ± 0.7

All values are in ng/g, wet mass (wm)

n.a. no specimens available at this location/season, *n.d.* below detection limit

Table 6 Number of fish collected during the sampling period

	Fort Resolution	Fort Smith	Fort Chipewyan	Fort McKay	Fort McMurray	Total
Burbot	22	8	5	2	6	43
Goldeye	12	18	16	28	15	89
Jackfish	24	19	20	20	21	104
Walleye	17	23	15	23	18	96
Whitefish	26	18	18	20	11	93
Total	101	86	74	93	71	425

ranged from 1.7 to 81 ng/g, wm for 2-ring PAHs and from less than the limit of detection (<LOD) to 43 ng/g for 3-ring PAHs, from <LOD to 73 ng/ml for 4-ring PAHs, from <LOD to 26 ng/g for 5-ring PAHs and from <LOD to 26 ng/g for 6-ring PAHs. The 16 USEPA priority PAHs were observed in muscle from fishes collected at all sampling locations and were greater in fishes from the Athabasca River than from the Slave River. Concentrations of $\sum$PAHs measured in fish muscle of the Athabasca/Slave Rivers are

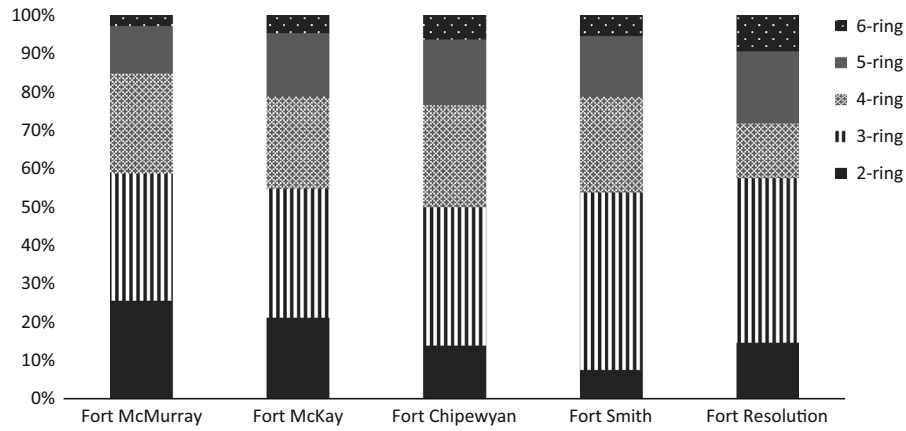

Fig. 2 Distributions of 2-, 3-, 4-, 5-, 6-ring PAHs in the muscle tissues of whitefish, goldeye, burbot, walleye and jackfish from the Athabasca/Slave Rivers

similar to those found in other oil-producing areas (Nkpaa et al. 2013; Al-Yakoob et al. 1994). Concentrations of ∑PAHs in muscle were slightly greater than those measured in fishes from non-oil producing areas (Ramalhosa et al. 2012).

Exposure associated with species, seasons and locations

The Kruskal–Wallis nonparametric test was used to compare within species for all locations and seasons and then among species by location and season (Table 7). Significant differences in concentrations of ∑PAHs in muscle were observed for goldeye, jackfish, walleye and whitefish among sites ($p < 0.001$). Tests to compare within species for seasonal variation only showed significant differences in concentrations of ∑PAHs in muscle of burbot ($p < 0.001$). There were no significant differences among species by site and by season ($p < 0001$). Analysis by season for locations showed statistically significant differences in ∑PAHs within summer, fall and spring. In general, greater concentrations of ∑PAH were detected in fishes collected from the Athabasca River relative to the Slave River (Fig. 3). The concentration of ∑PAHs in muscle of fishes from near Fort McMurray ranged from 11 ng/g, wm (burbot, summer) to 116 ng/g, wm (burbot, spring) with a mean concentration of 48 ng ∑PAHs/g, wm. The concentration of ∑PAHs in muscle of fishes from near Fort McKay ranged from 29 ng/g, wm (goldeye, summer) to 81 ng/g, wm (walleye, spring) with mean value of 53 ng/g, wm. The concentration of ∑PAHs in muscle of fishes from near Fort Chipewyan varied

Table 7 Kruskal–Wallis nonparametric one-way analysis of variance test showing differences within species for all sites and seasons

Species	Locations		Seasons	
	Statistic	*p* value	Statistic	*p* value
Walleye	41.29	0.000	0.08	0.961
Goldeye	36.45	0.000	4.64	0.098
Jackfish	52.47	0.000	0.36	2.062
Burbot	8.41	0.078	20.00	0.000
Whitefish	37.44	0.000	8.50	0.014

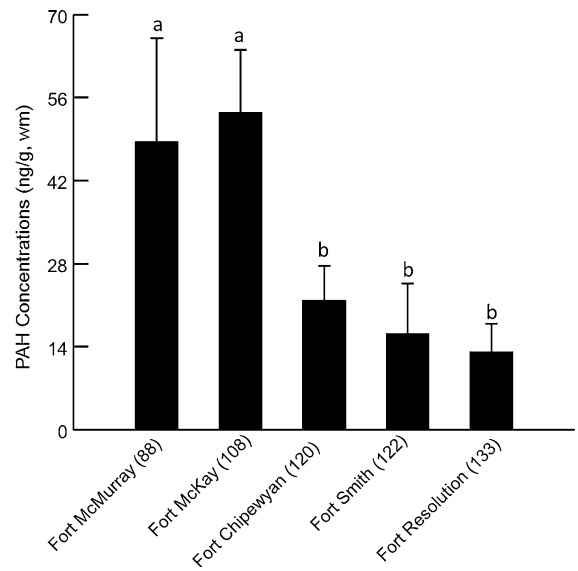

Fig. 3 Levels of PAH values in muscles of the five collected species. Statistical differences between pairs of seasons ($p < 0.05$) are indicated by different letters

from 11 ng/g, wm (burbot, summer) to 47 ng/g, wm (burbot, fall) with mean value of 22 ng/g, wm. The concentration of ∑PAHs in muscle of fishes from near Fort Smith ranged from 3.8 ng/g, wm (burbot, spring) to 55 ng/g, wm (burbot, fall) with mean concentration of 16 ng/g, wm, while the concentration of ∑PAHs in muscle from fish collected near Fort Resolution ranged from 4.3 ng/g, wm (whitefish, summer) to 33 ng/g, wm (goldeye, summer) with a mean concentration of 13 ng/g, wm. The greatest concentration of ∑PAHs was observed in muscle of fishes collected during spring sampling (Fig. 4).

Concentrations of PAHs in Northern Pike

Since some of the collected species migrate seasonally. They might be exposed to different sources of contaminants during different seasons. In contrast, northern pike (*Esox luscious*; jackfish) rarely travel significant distances, and this territorial behavior makes them a more suitable indicator species for localized contamination (Fig. 5). Concentrations of ∑PAHs in pike were greatest at Fort McMurray and least at Fort Resolution. Concentrations of ∑PAHs in pike from Fort Resolution ranged from 1.8 ng/g, wm (summer) to 17.2 ng/g, wm (summer) with a mean value of 7.8 ng/g wm. Concentrations of ∑PAHs in pike from Fort Smith ranged from 2.5 ng/g, wm (summer) to 38.8 ng/g, wm (fall) with a mean of 10.8 ng/g, wm. Concentrations of ∑PAHs in pike from Fort Chipewyan ranged from 2.4 ng/g, wm (summer) to 41.7 ng/g, wm (spring) with a mean of 15.9 ng/g, wm. Concentrations of ∑PAHs at Fort McKay ranged from 4.3 ng/g, wm (fall) to 100.2 ng/g, wm (spring) with a mean of 44 ng/g, wm. Concentrations of ∑PAHs at Fort McMurray ranged from 15.6 ng/g, wm (summer) to 241 ng/g, wm (summer) with mean value of 45 ng/g, wm. The results are consistent with previous findings (Ohiozebau et al., 2015), there being greater concentrations of PAHs in fish bile in the Athabasca River, relative to the Slave River.

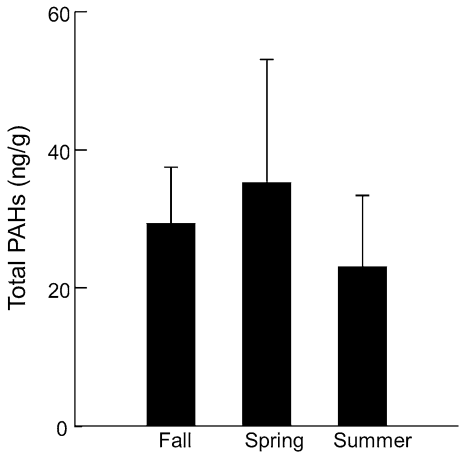

Fig. 4 Levels of PAH values during the sampling seasons

Human health risk assessment

Risks of adverse effects to humans, associated with PAH exposure, can be determined by comparing measurable concentrations to health-based regulatory limits. Averaged concentrations of BaP_{eq} in various fishes are presented in Table 8. The predicted concentration of BaP_{eq} in fish was consistent with the spatial trends in concentrations of PAHs. Nevertheless, Fort Chipewyan, where concentrations of PAHs were less than those at Fort McMurray, had a greater BaP_{eq} concentration than that of Fort McMurray (Fig. 6). This is because of greater concentrations of PAHs with larger TEF values, such as benzo(a)anthracene, benzo(b)fluoranthene and benzo(k)fluoranthene. The least concentration of BaP_{eq} (1.56 ng/g wm) was measured in walleye from Fort Resolution, while the greatest concentration (11.9 ng/g wm) was measured in burbot from Fort McKay.

Minimal risk levels (MRLs)

To develop an understanding of the potential risk to human health based on PAH intake via fish consumption, it was necessary to evaluate risk based on the most sensitive PAH-induced endpoint of relevance to humans. Minimal risk levels (MRLs) are screening levels for estimating the daily acceptable human exposures to dangerous substances, based on non-cancer health effects. MRLs are determined from studies on animals and humans, using the NOEL/uncertainty factor approach. MRLs are reference values to evaluate the toxicity of PAHs based on acute (1–14 days), intermediate (14–365 days) and chronic (365 days and longer) oral exposures (Table 9). It was possible to use the daily rate of consumption of fish to calculate an intermediate oral exposure. In this case, possible human exposures were less than MRL values, thus presenting no remarkable

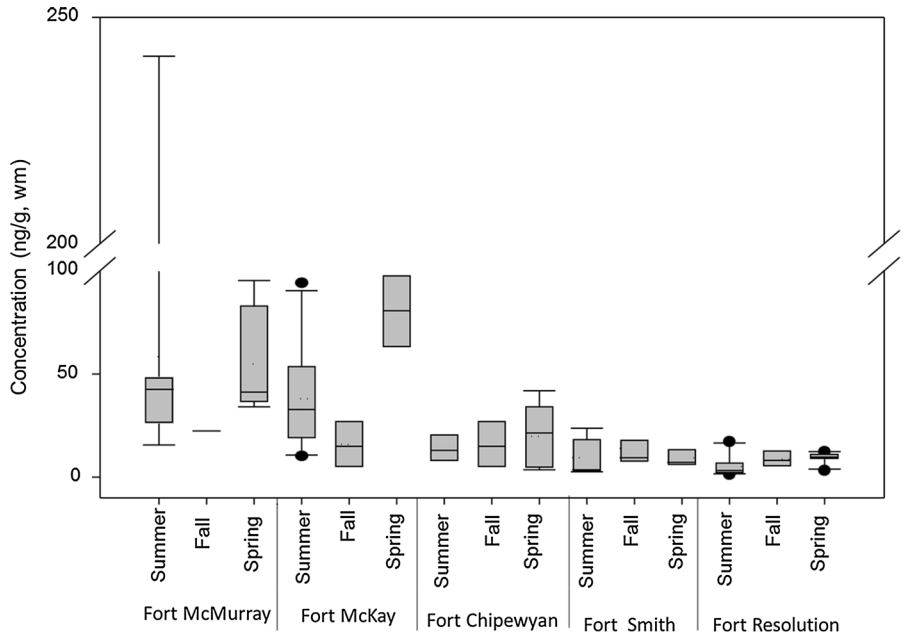

Fig. 5 *Box plots* showing the spread of concentrations (ng/g, wm) of PAH levels in muscle of northern pike from the five locations, during three seasons. Confidence interval is 95 %. Thick line is the median. The width of the box shows the interquartile range. The top 50 % of the concentration are represented by everything above the median. The top 25 % concentrations are shown by the top whisker

Table 8 Average concentration of the total PAHs found in collected fish and the relative TEBaP values

Location	Average PAHs (ng/g wm)	Average PAHs (ng TE BaP/g wm)	Average PAHs (ng/g fm)	Average PAHs (ng TE BaP/g f.m)
Fort Resolution	12.7	3.8	9.3E−3	2.8E−3
Fort Smith	17.2	5.4	14E−3	4.4E−3
Fort Chipewyan	22.2	6.2	16E−3	4.5E−3
Fort McKay	48.7	9.8	41E−3	8.4E−3
Fort McMurray	50.1	5.6	40E−3	4.5E−3

wm wet mass; *fm* fish mass

risk to humans. For example, the DI of PAHs due to consumption of fish at Fort McMurray was 8 % of the MRL for an intermediate exposure. Therefore, it is unlikely that PAHs derived from consumption of fishes in the Athabasca/Slave Rivers would cause intermediate-level adverse effects to humans. Furthermore, the reference value was based on USEPA assumptions of daily consumption of 227 g of fish from the same location over a 70-year life span (USEPA 1991a). Using this consumption value, the estimated daily intakes (DI) for Fort Resolution, Fort Smith, Fort Chipewyan, Fort McKay and Fort McMurray were 12.2, 17.6, 19.9, 31.7 and 18.2 ng BaP_{eq}/kg body mass (bm) per day, respectively. The result obtained for an acute exposure was even less than that for MRL values by several orders of magnitude. As a conservative approximation, the greatest observed concentration for each species and a range of possible fish consumption rates were used to calculate a DI for each species (Table 10). The potential cancer risk due to PAHs from consumption of even the most contaminated fish species in the study area is extremely small.

Potential for risks to local populations

The daily intake (DI) to PAHs due to consumption of fish, for each population group at Fort Resolution, Fort Smith, Fort Chipewyan, Fort McKay and Fort McMurray, was calculated (Fig. 7a, b). The median $B[a]P_{eq}$ daily intakes due to fish consumption for male groups were estimated to be 748, 1093, 1285 and

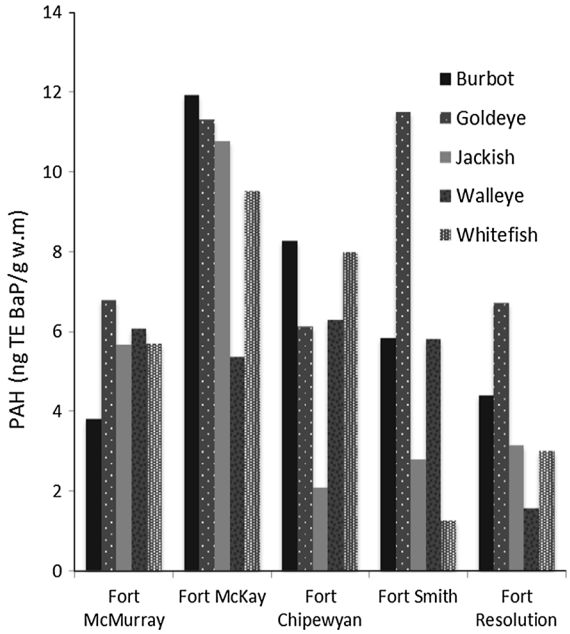

Fig. 6 The concentrations of $TEBaP_{eq}$ (ngg^{-1} wm) values of different fish species, in respective to their sampling locations

1046 ng/g/d, respectively, and that for females at the same locations were 744, 619, 1577 and 1041 ng/g/d for children, teenagers, adults and seniors, respectively. The intake of $B[a]P_{eq}$ increased in the order for males: children, teen, senior and adults. For females, the increasing order was: teens, children, seniors and adults. Based on our estimates, the female adults of Fort McKay have greater potential for exposure (3218 ng/d) to $B[a]P_{eq}$ from consuming fish meals, while female teens in Fort Smith have the least exposure (133 ng/d). In general, across all age groups, males were predicted to have slightly greater daily exposure (1097 ng/d) than did females (1051 ngd^{-1}). This result is similar to other studies (Xia et al. 2010; Martí-Cid et al. 2008). We used a wide range of possible fish consumption rates and body mass values (low, medium, high) to calculate possible risks to consumers based on the measurable PAH values in the sampled locations (Tables 2, 3). None of the values presents appreciable risk to human consumers in the areas. The cumulative probability distributions of the calculated LCR are presented in Table 11. The average values of LCR for all population groups were lower than the range of one in a million (10^{-6}) chance of additional human cancers over a 70-year lifetime (LCR = 10^{-6}).

Discussion

The relatively small concentrations of individual PAHs observed in the fish muscle tissues are clearly related to the relatively rapid depuration of these contaminants in fish (Ahokas and Pelkonen 1984). Complex phenomena, mainly ecology, such as preferred habitat, and bioavailability of individual compounds influence exposure to PAHs (Simonin et al. 2008). Physical characteristics like temperature, turbidity (Kerkhoven and Gan 2011) and acidity of systems also affect organic contaminant distribution in aquatic biota (Schindler et al. 1995). The Athabasca and Slave Rivers are hard water rivers with relatively great concentrations of mainly bicarbonate salts of calcium.

The fishes studied were assigned to trophic levels ranging from 2 to 4. Lake whitefish is a first-order carnivore (Scott and Crossman 1979; Nelson and Paetz 1992). Burbot, walleye and northern pike are piscivores (Braune 1999). Muscle concentrations are greater in lower tropic-level species as were concentrations of PAHs in bile (Ohiozebau et al. 2015). This may in part be due to biodiminution between trophic

Table 9 Minimal risk level (MRL) for different PAHs formulated by the Agency for Toxic Substances and Disease Registry (ATSDR) (1996) according to the duration of oral exposure (Agency for Toxic Substances and Disease Registry (ATSDR) 1996, 347)

Compound	Duration	MRL (mg/kg/d)	Factor of uncertainty	Endpoint
Anthracene	Interm.	10	100	Hepatic
Fluoranthene	Interm.	0.4	300	Hepatic
Fluorene	Interm.	0.4	300	Hepatic
Naphthalene	Acute	0.05	1000	Neurol.
	Interm	0.02	300	Hepatic

Table 10 Daily intakes (DI) of PAHs (ng BaP_{eq}/kg body weight per day) at different body masses using the highest observed TEBaP concentration for each species, based on different daily consumption of fish for female and male groups

Species	Max. TEBaP	Children			Teens			Adults			Seniors		
		Low	Medium	High	Low	Medium	High	Low	Medium	High	Low	Medium	High
A. Females													
Burbot	11.9	6.0	43.5	52.4	19.8	32.5	47.6	14.3	35.7	35.7	11.9	29.8	39.7
Goldeye	11.5	5.8	42.1	50.6	19.2	31.4	46.0	13.8	34.5	34.5	11.5	28.8	38.3
Jackfish	10.8	5.4	39.5	47.5	18.0	29.5	43.2	13.0	32.4	32.4	10.8	27.0	36.0
Walleye	6.3	3.2	23.0	27.7	10.5	17.2	25.2	7.6	18.9	18.9	6.3	15.8	21.0
Whitefish	9.5	4.8	34.7	41.8	15.8	25.9	38.0	11.4	28.5	28.5	9.5	23.8	31.7
B. Males													
Burbot	11.9	7.9	47.6	58.2	14.9	37.7	52.1	11.9	31.7	38.1	9.9	39.7	52.1
Goldeye	11.5	7.7	46.0	56.2	14.4	36.4	50.3	11.5	30.7	36.8	9.6	38.3	50.3
Jackfish	10.8	7.2	43.2	52.8	13.5	34.2	47.3	10.8	28.8	34.6	9.0	36.0	47.3
Walleye	6.3	4.2	25.2	30.8	7.9	20.0	27.6	6.3	16.8	20.2	5.3	21.0	27.6
Whitefish	9.5	6.3	38.0	46.4	11.9	30.1	41.6	9.5	25.3	30.4	7.9	31.7	41.6

levels. PAHs have relatively short metabolic half-lives and as such do not show a tendency to biomagnify. They are readily degradable compounds that are subject to metabolic clearance at lower trophic levels, reducing their potential to be passed along food chains (Walker et al. 2012).

Whitefish had the greatest concentrations of PAHs of the collected species from all sites and seasons. Due to their lipophilic nature the availability of PAHs decreases in open water relative to the benthic zone, thus affecting bottom dwelling organisms (Borga 2011). Species with a preference for benthic habitats are more likely to have greater exposures to PAH in a polluted environment than those with a preference for pelagic environments. Whitefish are occasionally pelagic but mainly feed on benthos (Muir et al. 2010; Scott and Crossman 1979). In contrast burbot is mainly benthic, while northern pike prefer shallow, vegetation-rich habitats. Walleye are primarily a littoral zone species but can be found in waters as deep as 20 m (Scott and Crossman 1979). Goldeye occurs in turbid slow-moving waters of rivers, ponds and marshes. They are also found in muddy shallow areas of lakes but frequent deeper areas over winter. These trends in species tissue concentrations are also consistent with previously measured concentrations of PAHs, reported as fluorescently active compounds (FACs) in bile (Ohiozebau et al. 2015).

Total concentrations of PAHs in fish from Fort Mckay and Fort McMurray were significantly greater ($p < 0.01$) than those in fish from Fort Smith and Fort Resolution indicating greater concentrations of PAHs in the Athabasca River than in the Slave River (Lanfranchi et al. 2007). Concentrations of PAHs in fishes collected from Fort McMurray can be attributed to natural incision of the river into petroleum deposits, aerial deposition from operations located downstream, operations around Fort McMurray and in the clear-water river catchment and finally from general human activity in this increasingly urbanized area.

Many sources may be responsible for the observed PAHs in the collected species. PAHs are generally classified as low molecular weight PAHs (LMW-PAHs; 2- and 3-ring PAHs) compared to larger molecular weight PAHs (HMW-PAHs; 4–6-ring PAHs). The LMW-PAH/HMW-PAH ratios observed in the five species, and seasons from the sampling locations were >1, indicating mainly petrogenic sources (Rocher et al. 2004). 2- and 3-Ring PAHs dominated the distribution at all sampling sites, species and seasons and accounted for 19.4 and 36.2 % of $\sum$PAHs, respectively (Fig. 2). Naphthalene was the compound accumulated to the greatest concentration possibly due to its lesser affinity for particles and greater water solubility. Phenanthrene is a principal PAH component and was the second

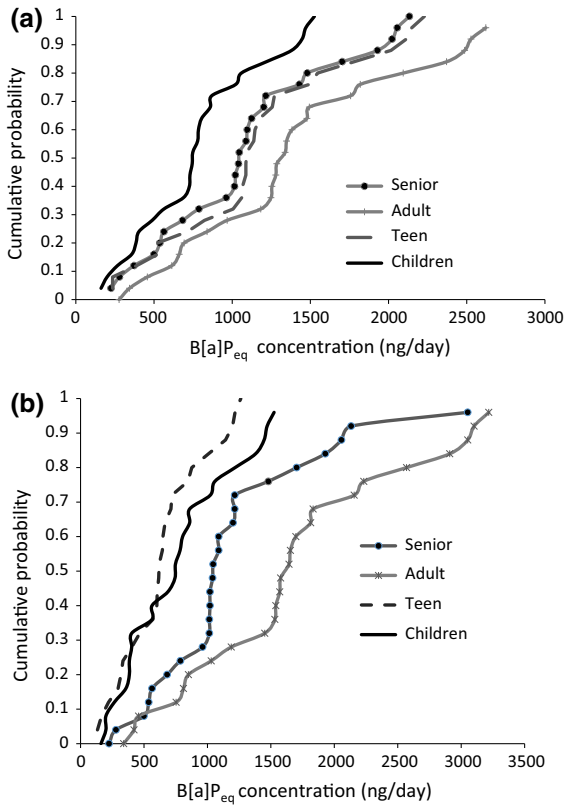

Fig. 7 **a** Probability distributions of daily dietary B[a]Peq exposure for male population groups in the Athabasca/Slave Rivers. **b** Probability distributions of daily dietary B[a]Peq exposure for female population groups in the Athabasca/Slave Rivers

Table 11 Life time cancer risk for combined male and female population groups in Athabasca/Slave Rivers

Location	Children	Teens	Adults	Seniors
Fort Resolution	1.1E−10	6.6E−11	7.1E−11	7.2E−11
Fort Smith	1.5E−10	9.6E−11	1.0E−10	1.0E−10
Fort Chipewyan	1.8E−10	1.0E−10	1.2E−10	1.2E−10
Fort McKay	2.8E−12	1.7E−10	1.8E−10	1.9E−10
Fort McMurray	1.6E−12	9.9E−11	1.1E−12	1.1E−10

most prevalent compound ($\sum$178.8 ng/g) in this study. This is a similar profile of PAH compounds to that generated by petrogenic pollution (Al-Yakoob et al. 1994). Chrysene is normally produced through combustion and was present at a mean concentration of 1.8 ng/g, wm. 4-Ring PAHs accounted for 24.2 % of $\sum$PAHs. The potentially carcinogenic 5- and 6-ring PAHs were lesser in concentration, accounting for only 15.4 and 4.8 % of $\sum$PAHs, respectively. This result is similar to previous fish studies from similar areas in other parts of the world (Ramalhosa et al. 2012; Nkpaa et al. 2013).

Diet is a major route of human exposure to PAHs (Cheung et al. 2007). In this study the estimated exposure to PAHs through fish consumption does not represent a significant additional risk to human consumers. The foregoing risk assessment does not assess other food sources nor other nondietary routes to PAH exposure but addresses only additional risk associated with fish consumption. Furthermore, intake of contaminants such as PAHs should not be the only criterion for consideration when assessing the potential risk to human health exposure, and time and intensity of exposure should also be considered (Binelli and Provini 2004). Therefore, it is unlikely that PAHs derived from fish consumption in the Athabasca/Slave Rivers would be causing adverse-level acute or intermediate effects in humans.

Average values for LCRs for all population groups were less than one in a million chances of additional human cancer over a 70-year lifetime (ILCR $= 10^{-6}$). From this result, it seems unlikely that PAHs derived from fish collected from the locations in the Athabasca/Slave Rivers would be causing adverse effects in First Nations communities in the areas. However, an individual can be exposed daily to a wide range of contaminants through dietary exposure (Pompa et al. 2003; Wei et al. 2011). Contaminants like heavy metals, PAHs and naphthenic acids have been reported in air, land and Athabasca River (Kelly et al. 2009, 2010). Cumulatively, the additive effects may make the HR and LCR values of PAH in fish more significant even if it is less than 1.0 and more than 1 in a million, respectively. The cumulative and possible interactive effects of these different contaminant groups also need to be considered when assessing risk.

It is difficult to absolutely assess the carcinogenic risk of PAHs because of the inherent uncertainties in risk assessment. For example, different cooking methods could affect the concentration of PAHs in cooked fish (Wretling et al. 2010). Also, possible synergistic and/or antagonistic effect might occur among the observed PAHs that might not have been accounted for during risk assessment. The B[a]P$_{eq}$-based approach does not account for the toxicity of all PAHs, e.g., alkylated compounds, to which the

population of interest may be exposed. Also, concentrations of B[a]P_{eq} used in this study to estimate risk were extrapolations from animal toxicity studies, although this risk assessment followed best practice and these values are recommended by the USEPA and Health Canada; nevertheless, they may not totally reflect the carcinogenic potential of these compounds in humans. Despite its inherent challenges, risk assessment provides a useful framework to evaluate the potential effects of environmental contaminants to humans. In the Athabasca/Slave Rivers, health risk assessment of pollutants, especially from the rapid economic development, is necessary to monitor human and ecological impact in the area. This study, which evaluated the carcinogenic risk level for different population groups in Fort McMurray, Fort McKay, Fort Chipewyan, Fort Smith and Fort Resolution, was an essential first step for a long-term risk assessment in the area. While there are some uncertainties, the overall conservative approach we have taken indicates that there is *de minimis* risk to people from PAHs in fishes that they might consume and thus the fish are safe to eat. To do otherwise would deprive individuals of the positive health benefits on neurobehavioral development and prevention of cardiovascular disease of eating fish.

Conclusion

This study analyzed 16 PAHs in edible parts of selected fish species and presents a general model for the probabilistic risk assessment due to PAH intake through consumption of fishes in the Athabasca and Slave Rivers. Measurable concentrations of PAHs were detected across spatial and seasonal studies. The profile was dominated by 2–3-ring PAHs, and 4-ring PAHs were also abundant. The spatial distribution of PAHs varied significantly at different sampling locations with the highest concentration in fishes from Fort McKay. Seasonal variations were also observed. Concentrations of $\sum$PAHs were greater in whitefish than in other species. A probabilistic approach is used to characterize the uncertainty of PAH content in fishes and the daily intake. The results show that the contamination with PAHs detected in the various fishes of the Athabasca/Slave Rivers is likely not a health risk to human consumers in the area. Fresh fish from the Athabasca/Slave Rivers are probably a minor dietary source of PAHs. Emphasis should be placed on science-based monitoring in the Athabasca/Slave River system as a whole. It is desirable therefore that a monitoring program in water, sediments and biota be in place and extend to the entire Athabasca/Slave basin to detect the presence of contaminants and mitigate their potential human and ecological effects. It is not the aim of this paper to diminish the concerns that First Nations communities have expressed about contamination of fish as a valuable economic and cultural resource. While this paper may assuage some concerns relative to immediate and direct health effects, it does not diminish concerns relative to the societal and cultural value of these resources.

Acknowledgments The authors would like to appreciate First Nations and Métis communities of Fort Resolution, Fort Smith, Fort Chipewyan, Fort McKay and Fort McMurray and numerous Provincial and Federal agencies for their assistance during the sampling. The Slave River and Delta Partnership provided invaluable assistance in the coordination of collection and assessment activities in the Slave River and Delta. Portions of this work were funded by the Boreal Songbird Initiative (BSI); Aboriginal Affairs and Northern Development Canada (AANDC); and the Government of the Northwest Territories (GNWT). EO was supported by a New Faculty Scholarship to PDJ from the University of Saskatchewan. Prof. Giesy was supported by the Canada Research Chair program and the program of 2014 'High Level Foreign Experts' (#GDT20143200016) funded by the State Administration of Foreign Experts Affairs, the PR China to Nanjing University and the Einstein Professor Program of the Chinese Academy of Sciences.

Appendix 1

Lognormal probability density functions describing daily fish consumption (g/day) for Canadian Aboriginal fish 'eaters only.' Individuals reporting no fish consumption were excluded. Values were rounded to two significant digits. Values represent arithmetic mean ± standard deviation for definition of lognormal distributions. Different values for males and females are indicated only where statistically significant differences were observed between the sexes in the data. Values represent, respectively, the arithmetic mean ± standard deviation (ARITH), the arithmetic mean and standard deviation of the log-transformed data (LN-TRANS), the geometric mean and geometric standard deviation (GEOMET) (Richardson 1997, 2013).

Gender	Children	Teens	Adults	Senior
Females				
ARITH	170 ± 150	150 ± 150	180 ± 140	250 ± 240
LN-TRANS	4.85 ± 0.76	4.66 ± 0.83	4.96 ± 0.69	5.19 ± 0.81
GEOMET	128 ± 2.1	106 ± 2.3	143 ± 2.0	179 ± 2.2
Males				
ARITH	170 ± 150	260 ± 250	270 ± 190	250 ± 240
LN-TRANS	4.85 ± 0.76	5.23 ± 0.81	5.40 ± 0.63	5.19 ± 0.81
GEOMET	128 ± 2.1	187 ± 2.2	221 ± 1.9	179 ± 2.2
Sexes combined				
ARITH	170 ± 150	200 ± 200	220 ± 160	250 ± 240
LN-TRANS	4.85 ± 0.76	4.95 ± 0.83	5.18 ± 0.65	5.19 ± 0.81
GEOMET	128 ± 2.1	141 ± 2.3	178 ± 1.9	179 ± 2.2

Appendix 2

Proposed probability density functions describing body weight (kg) in the Canadian population. In all cases, PDFs should be defined as lognormal. Values represent, respectively, the arithmetic mean ± standard deviation (ARITH), the arithmetic mean and standard deviation of the log-transformed data (LN-TRANS), the geometric mean and geometric standard deviation (GEOMET) (Richardson 1997, 2013).

Age group	Distribution	Females	Males	Sexes combined
Infants (0–6 months)	Arth			8.2 ± 2.9
	Ln-Trans	–	–	2.05 ± 0.34
	Geomet	–	–	7.8 ± 1.4
Toddlers (7 months–4 years)	Arth	16.4 ± 4.5	16.5 ± 4.6	16.5 ± 4.5
	Ln-Trans	2.76 ± 0.27	2.77 ± 0.27	2.77 ± 0.27
	Geomet	15.8 ± 1.3	16.0 ± 1.3	16.0 ± 1.3
Children (5–11 years)	Arth	33.6 ± 9.3	32.2 ± 8.0	32.9 ± 8.9
	Ln-Trans	3.48 ± 0.27	3.44 ± 0.24	3.46 ± 0.27
	Geomet	32.5 ± 1.3	31.2 ± 1.3	31.8 ± 1.3
Teens (12–19 years)	Arth	56.2 ± 10.2	63.1 ± 15.3	59.7 ± 13.5
	Ln-Trans	4.01 ± 0.18	4.12 ± 0.24	4.06 ± 0.22
	Geomet	55.1 ± 1.2	61.6 ± 1.3	58.0 ± 1.2
Adults (20–59 years)	Arth	63.1 ± 11.9	78.8 ± 12.3	70.7 ± 14.4
	Ln-Trans	4.13 ± 0.18	4.35 ± 0.16	4.24 ± 0.20
	Geomet	62.2 ± 1.2	77.5 ± 1.2	69.4 ± 1.2
Seniors (60+ years)	Arth	63.4 ± 11.6	78.9 ± 14.2	70.6 ± 15.0
	Ln-Trans	4.13 ± 0.18	4.35 ± 0.18	4.23 ± 0.21
	Geomet	62.2 ± 1.2	77.5 ± 1.2	68.7 ± 1.2
Adults (20+ years)	Arth	63.1 ± 11.8	78.8 ± 12.6	70.7 ± 14.5
	Ln-Trans	4.13 ± 0.19	4.35 ± 0.16	4.24 ± 0.20
	Geomet	62.2 ± 1.2	77.5 ± 1.2	69.4 ± 1.2

Appendix 3

Mean (± SD) values for parameters, including: length (cm), mass (g)and liver somatic index (LSI) of fishes collected at Fort Resolution, Fort Smith, Fort Chipewyan, Fort McKay and Fort McMurray in 2011–2012 in (A) summer, (B) fall, (C) spring. Number of individual fish collected indicated in brackets (*n*). *n.a* = no specimen available this location/season. *F* = Fort.

Fish species		F. McMurray	F. McKay	F. Chipewyan	F. Smith	F. Resolution
3a. Summer						
Burbot	Length	41 ± 3.4 (3)	n.a	42 ± 3.4 (2)	50 ± 9.2 (5)	62 ± 4.4 (10)
	Mass	420 ± 104(3)	n.a	693 ± 104 (2)	577 ± 320 (5)	1591 ± 341 (10)
	LSI	6.9 ± 1.5 (3)	n.a	5.1 ± 1.5(2)	2.0 ± 0.2 (5)	13 ± 21 (10)
Goldeye	Length	35 ± 4.5 (10)	38 ± 2.7 (10)	37 ± 1.1 (10)	29 ± 3.5 (10)	38 ± 1.8 (2)
	Mass	489 ± 154 (10)	685 ± 140 (10)	573 ± 55 (10)	221 ± 95 (10)	646 ± 153 (2)
	LSI	1.2 ± 0.3 (10)	1.5 ± 0.2 (10)	1.2 ± 0.3 (10)	0.7 ± 0.2 (10)	1.1 ± 0.1 (2)
Jackfish	Length (cm)	61 ± 22 (10)	62 ± 10 (10)	66 ± 5.1 (10)	68 ± 505 (10)	64 ± 4.2 (10)
	Mass (g)	1610 ± 1369 (10)	1938 ± 1172 (10)	2178 ± 1102(10)	2457 ± 981 (10)	1976 ± 1276 (10)
	LSI	1.4 ± 0.7 (10)	1.8 ± 0.4 (10)	0.8 ± 0.3 (10)	1.4 ± 0.6 (10)	3.3 ± 4.8 (10)
Walleye	Length	5.8 ± 10 (10)	45 ± 13 (10)	51 ± 3.4 (10)	40 ± 7.6 (10)	n.a
	Mass	1347 ± 646 (10)	1003 ± 566 (10)	1365 ± 247(10)	644 ± 364 (10)	n.a
	LSI	1.1 ± 0.3 (10)	1.0 ± 0.3 (10)	1.1 ± 0.4 (10)	0.8 ± 0.2 (10)	n.a
Whitefish	Length (cm)	n.a	42 ± 4.2(10)	41 ± 3.4 (10)	41.1 ± 3.7 (8)	39 ± 1.9 (10)
	Mass	n.a	1281 ± 323 (10)	1177 ± 324 (10)	864 ± 145 (8)	685 ± 223 (10)
	LSI	n.a	1.0 ± 0.2 (10)	1.2 ± 0.3 (10)	0.8 ± 0.3 (8)	1.9 ± 3.2 (10)
3b. Fall						
Burbot	Length	n.a	55 ± 0.9 (2)	59 ± 2.8 (3)	61 ± 5.1 (3)	61 ± 5.0 (10)
	Mass	n.a	1075 ± 7.1 (2)	1387 ± 74 (3)	1335 ± 158 (3)	1662 ± 404 (10)
	LSI	n.a	2.1 ± 0.1 (2)	3.0 ± 0.4 (3)	2.9 ± 0.4 (3)	3.2 ± 1.4 (10)
Goldeye	Length	39 ± 0.0 (1)	36 ± 1.4 (10)	37 ± 2.7 (10)	36 ± 1.3 (10)	36 ± 0.9 (10)
	Mass	700 ± 0.0 (1)	537 ± 47 (10)	627 ± 95 (10)	552 ± 66 (10)	546 ± 65 (10)
	LSI	1.4 ± 0.0 (1)	1.3 ± 0.1 (10)	1.5 ± 0.5 (10)	2.1 ± 3.1 (10)	1.3 ± 0.2 (10)
Jackfish	Length	72 ± 14 (3)	63 ± 8.8 (9)	76 ± 2.5 (10)	67 ± 8.4 (10)	69 ± 11 (10)
	Mass	3287 ± 1454(3)	2531 ± 1415 (9)	4220 ± 1157 (10)	1390 ± 522 (10)	1266 ± 538 (10)
	LSI	1.9 ± 0.4 (3)	1.9 ± 0.3 (9)	1.7 ± 0.2 (10)	1.1 ± 0.5 (10)	1.2 ± 0.4 (10)
Walleye	Length	42 ± 11 (3)	49 ± 4.8 (10)	50 ± 2.5 (5)	49 ± 5.5 (10)	47 ± 6.9 (10)
	Mass	940 ± 588(3)	1356 ± 408 (10)	4220 ± 1157 (5)	1390 ± 522 (10)	1266 ± 538 (10)
	LSI	1.9 ± 0.1 (3)	1.5 ± 0.5 (10)	1.7 ± 0.2 (5)	1.3 ± 0.4 (10)	2.4 ± 1.2 (10)
Whitefish	Length	42 ± 3.4 (10)	40 ± 2.2 (10)	39 ± 3.1 (10)	41 ± 1.8 (10)	44 ± 3.5 (10)
	Mass	1042 ± 235 (10)	1020 ± 150 (10)	1072 ± 200 (10)	1019 ± 125(10)	1296 ± 38 (10)
	LSI	0.8 ± 0.1 (10)	0.8 ± 0.2 (10)	1.4 ± 0.4 (10)	0.8 ± 0.2 (10)	0.9 ± 0.2 (10)
3c. Spring						
Burbot	Length	39 ± 2.6 (3)	n.a	n.a	38 ± 0.0 (1)	63 ± 3.3 (6)
	Mass	420 ± 87 (3)	n.a	n.a	750 ± 0.0 (1)	1623 ± 632 (6)
	LSI	5.2 ± 1.9 (3)	n.a	n.a	1.1 ± 0.0 (1)	7.5 ± 3.7 (6)
Goldeye	Length	34 ± 2.9 (10)	27 ± 5.1 (10)	35 ± 3.1 (10)	37 ± 1.9 (10)	35 ± 3.8 (10)
	Mass	524 ± 113 (10)	285 ± 186 (10)	490 ± 109 (10)	570 ± 100(10)	554 ± 166 (10)
	LSI	1.1 ± 0.2 (10)	1.4 ± 0.2 (10)	1.5 ± 0.6 (10)	1.3 ± 0.2 (10)	1.3 ± 0.2 (10)

continued

Fish species		F. McMurray	F. McKay	F. Chipewyan	F. Smith	F. Resolution
Jackfish	Length	63 ± 9.1 (10)	60 ± 7.2 (5)	63 ± 8.0 (10)	69 ± 11 (10)	69 ± 5.8 (10)
	Mass	3389 ± 1209 (10)	1862 ± 1425 (5)	1653 ± 468. (10)	3237 ± 1508 (10)	2272 ± 1020 (10)
	LSI	1.7 ± 0.6 (10)	1.4 ± 0.5 (5)	1.2 ± 0.5 (10)	1.4 ± 0.2 (10)	2.6 ± 4.4 (10)
Walleye	Length	48 ± 6.8 (10)	44 ± 2.6 (10)	50 ± 6.6 (10)	51 ± 8.7 (10)	46 ± 13 (10)
	Mass	1740 ± 870 (10)	1092 ± 148(10)	1367 ± 398 (10)	1623 ± 771 (10)	1180 ± 712 (10)
	LSI	1.2 ± 0.4 (10)	1.2 ± 0.3 (10)	1.4 ± 0.3 (10)	1.6 ± 0.5 (10)	1.5 ± 0.4 (10)
Whitefish	Length	42 ± 2.0 (4)	38 ± 1.8 (2)	43 ± 5.8 (10)	41 ± 1.3 (5)	39 ± 2.6 (10)
	Mass	1278 ± 315 (4)	1025 ± 35(2)	1384 ± 392 (10)	990 ± 115.3 (5)	807 ± 197 (10)
	LSI	1.2 ± 0.1 (4)	1.0 ± 0.0 (2)	1.3 ± 0.2 (10)	0.9 ± 0.2 (5)	1.1 ± 0.3 (10)

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Appendix 1

Log-normal probability density functions describing daily fish consumption (g/day) for Canadian Aboriginal fish 'eaters only'. Individuals reporting no fish consumption were excluded. Values were rounded to two significant digits. Values represent arithmetic mean ± standard deviation for definition of log-normal distributions. Different values for males and females are indicated only where statistically significant differences were observed between the sexes in the data. Values represent, respectively, the arithmetic mean ± standard deviation (ARITH), the arithmetic mean and standard deviation of the log-transformed data (LN-TRANS), the geometric mean and geometric standard deviation (GEOMET) (Richardson 1997 & 2013).

Gender		Children	Teens	Adults	Senior
Females	ARITH	170±150	150±150	180±140	250±240
	LN-TRANS	4.85±0.76	4.66±0.83	4.96±0.69	5.19±0.81
	GEOMET	128±2.1	106±2.3	143±2.0	179±2.2
Males	ARITH	170±150	260±250	270±190	250±240
	LN-TRANS	4.85±0.76	5.23±0.81	5.40±0.63	5.19±0.81
	GEOMET	128±2.1	187±2.2	221±1.9	179±2.2
Sexes	ARITH	170±150	200±200	220±160	250±240
combined	LN-TRANS	4.85±0.76	4.95±0.83	5.18±0.65	5.19±0.81
	GEOMET	128±2.1	141±2.3	178±1.9	179±2.2

Appendix 2

Proposed probability density functions describing body weight (kg) in the Canadian population. In all cases, PDFs should be defined as log-normal. Values represent, respectively, the arithmetic mean ± standard deviation (ARITH), the arithmetic mean and standard deviation of the log-transformed data (LN-TRANS), the geometric mean and geometric standard deviation (GEOMET) (Richardson 1997 & 2013).

Age Group	Distribution	Females	Males	Sexes Combined
Infants (0-6 Months)	Arth			8.2 ± 2.9
	Ln-Trans	-	-	2.05 ± 0.34
	Geomet	-	-	7.8 ± 1.4
Toddlers (7m-4yrs)	Arth	16.4 ± 4.5	16.5 ± 4.6	16.5 ± 4.5
	Ln-Trans	2.76 ± 0.27	2.77 ± 0.27	2.77 ± 0.27
	Geomet	15.8 ± 1.3	16.0 ± 1.3	16.0 ± 1.3
Children (5yrs-11yrs)	Arth	33.6 ± 9.3	32.2 ± 8.0	32.9 ± 8.9
	Ln-Trans	3.48 ± 0.27	3.44 ± 0.24	3.46 ± 0.27
	Geomet	32.5 ± 1.3	31.2 ± 1.3	31.8 ± 1.3
Teens (12-19 Yrs)	Arth	56.2 ± 10.2	63.1 ± 15.3	59.7 ± 13.5
	Ln-Trans	4.01 ± 0.18	4.12 ± 0.24	4.06 ± 0.22
	Geomet	55.1 ± 1.2	61.6 ± 1.3	58.0 ± 1.2
Adults (20-59 Yrs)	Arth	63.1 ± 11.9	78.8 ± 12.3	70.7 ± 14.4
	Ln-Trans	4.13 ± 0.18	4.35 ± 0.16	4.24 ± 0.20
	Geomet	62.2 ± 1.2	77.5 ± 1.2	69.4 ± 1.2
Seniors (60+ Yrs)	Arth	63.4 ± 11.6	78.9 ± 14.2	70.6 ± 15.0
	Ln-Trans	4.13 ± 0.18	4.35 ± 0.18	4.23 ± 0.21
	Geomet	62.2 ± 1.2	77.5 ± 1.2	68.7 ± 1.2
Adults (20+Yrs)	Arth	63.1 ± 11.8	78.8 ± 12.6	70.7 ± 14.5
	Ln-Trans	4.13 ± 0.19	4.35 ± 0.16	4.24 ± 0.20
	Geomet	62.2 ± 1.2	77.5 ± 1.2	69.4 ± 1.2

Appendix 3

Mean (± SD) values for parameters, including: length (cm) mass (g),and liver-somatic index (LSI) of fishes collected at Fort Resolution, Fort Smith, Fort Chipewyan, Fort McKay, and Fort McMurray in 2011-2012 in A) Summer, B) Fall, C) Spring. Number of individual fish collected indicated in brackets (n). n.a = no specimen available this location/season. F= Fort.

3a) Summer

FISH SPECIES		F. MCMURRAY	F. MCKAY	F. CHIPEWYAN	F. SMITH	F. RESOLUTION
Burbot	Length	41 ±3.4 (3)	n.a	42 ± 3.4 (2)	50 ± 9.2 (5)	62 ± 4.4 (10)
	Mass	420 ± 104(3)	n.a	693 ± 104 (2)	577 ± 320 (5)	1591 ± 341 (10)
	LSI	6.9 ± 1.5 (3)	n.a	5.1 ± 1.5(2)	2.0 ± 0.2 (5)	13 ± 21 (10)
Goldeye	Length	35 ± 4.5 (10)	38 ± 2.7 (10)	37 ± 1.1 (10)	29 ± 3.5 (10)	38 ± 1.8 (2)
	Mass	489 ± 154 (10)	685 ± 140 (10)	573 ± 55 (10)	221 ± 95 (10)	646 ± 153 (2)
	LSI	1.2 ± 0.3 (10)	1.5 ± 0.2 (10)	1.2 ± 0.3 (10)	0.7 ± 0.2 (10)	1.1 ± 0.1 (2)
Jackfish	Length (cm)	61 ± 22 (10)	62 ± 10 (10)	66 ± 5.1 (10)	68 ± 5.5 (10)	64 ± 4.2 (10)
	Mass (g)	1610 ± 1369 (10)	1938 ± 1172 (10)	2178 ± 1102(10)	2457 ± 981 (10)	1976 ± 1276 (10)
	LSI	1.4 ± 0.7 (10)	1.8 ± 0.4 (10)	0.8 ± 0.3 (10)	1.4 ± 0.6 (10)	3.3 ± 4.8 (10)
Walleye	Length	5.8 ± 10 (10)	45 ± 13 (10)	51 ± 3.4 (10)	40 ± 7.6 (10)	n.a
	Mass	1347 ± 646 (10)	1003 ± 566 (10)	1365 ± 247(10)	644 ± 364 (10)	n.a
	LSI	1.1 ± 0.3 (10)	1.0 ± 0.3 (10)	1.1 ± 0.4 (10)	0.8 ± 0.2 (10)	n.a
Whitefish	Length (cm)	n.a	42 ± 4.2(10)	41 ± 3.4 (10)	41.1 ± 3.7 (8)	39 ± 1.9 (10)
	Mass	n.a	1281 ± 323 (10)	1177 ± 324 (10)	864 ± 145 (8)	685 ± 223 (10)
	LSI	n.a	1.0 ± 0.2 (10)	1.2 ± 0.3 (10)	0.8 ± 0.3 (8)	1.9 ± 3.2 (10)

3b) Fall

Species		F. MCMURRAY	F. MCKAY	F. CHIPEWYAN	F. SMITH	F. RESOLUTION
Burbot	Length	n.a	55 ± 0.9 (2)	59 ± 2.8 (3)	61 ± 5.1 (3)	61 ± 5.0 (10)
	Mass	n.a	1075 ± 7.1 (2)	1387 ± 74 (3)	1335 ± 158 (3)	1662 ± 404 (10)
	LSI	n.a	2.1 ± 0.1 (2)	3.0 ± 0.4 (3)	2.9 ± 0.4 (3)	3.2 ± 1.4 (10)
Goldeye	Length	39 ± 0.0 (1)	36 ± 1.4 (10)	37 ± 2.7 (10)	36 ± 1.3 (10)	36 ± 0.9 (10)
	Mass	700 ± 0.0 (1)	537 ± 47 (10)	627 ± 95 (10)	552 ± 66 (10)	546 ± 65 (10)
	LSI	1.4 ± 0.0 (1)	1.3 ± 0.1 (10)	1.5 ± 0.5 (10)	2.1 ± 3.1 (10)	1.3 ± 0.2 (10)
Jackfish	Length	72 ± 14 (3)	63 ± 8.8 (9)	76 ± 2.5 (10)	67 ± 8.4 (10)	69 ± 11 (10)
	Mass	3287 ± 1454(3)	2531 ± 1415 (9)	4220 ± 1157 (10)	1390 ± 522 (10)	1266 ± 538 (10)
	LSI	1.9 ± 0.4 (3)	1.9 ± 0.3 (9)	1.7 ± 0.2 (10)	1.1 ± 0.5 (10)	1.2 ± 0.4 (10)
Walleye	Length	42 ± 11 (3)	49 ± 4.8 (10)	50 ± 2.5 (5)	49 ± 5.5 (10)	47 ± 6.9 (10)
	Mass	940 ± 588(3)	1356 ± 408 (10)	4220 ± 1157 (5)	1390 ± 522 (10)	1266 ± 538 (10)
	LSI	1.9 ± 0.1 (3)	1.5 ± 0.5 (10)	1.7 ± 0.2 (5)	1.3 ± 0.4 (10)	2.4 ± 1.2 (10)
Whitefish	Length	42 ± 3.4 (10)	40 ± 2.2 (10)	39 ± 3.1 (10)	41 ± 1.8 (10)	44 ± 3.5 (10)
	Mass	1042 ± 235 (10)	1020 ± 150 (10)	1072 ± 200 (10)	1019 ± 125(10)	1296 ± 38 (10)
	LSI	0.8 ± 0.1 (10)	0.8 ± 0.2 (10)	1.4 ± 0.4 (10)	0.8 ± 0.2 (10)	0.9 ± 0.2 (10)

3c) Spring

Species		F. MCMURRAY	F. MCKAY	F. CHIPEWYAN	F. SMITH	F. RESOLUTION
Burbot	Length	39 ± 2.6 (3)	n.a	n.a	38 ± 0.0 (1)	63 ± 3.3 (6)
	Mass	420 ± 87 (3)	n.a	n.a	750 ± 0.0 (1)	1623 ± 632 (6)
	LSI	5.2 ± 1.9 (3)	n.a	n.a	1.1 ± 0.0 (1)	7.5 ± 3.7 (6)
Goldeye	Length	34 ± 2.9 (10)	27 ± 5.1 (10)	35 ± 3.1 (10)	37 ± 1.9 (10)	35 ± 3.8 (10)
	Mass	524 ± 113 (10)	285 ± 186 (10)	490 ± 109 (10)	570 ± 100(10)	554 ± 166 (10)
	LSI	1.1 ± 0.2 (10)	1.4 ± 0.2 (10)	1.5 ± 0.6 (10)	1.3 ± 0.2 (10)	1.3 ± 0.2 (10)
Jackfish	Length	63 ± 9.1 (10)	60 ± 7.2 (5)	63 ± 8.0 (10)	69 ± 11 (10)	69 ± 5.8 (10)
	Mass	3389 ± 1209 (10)	1862 ± 1425 (5)	1653 ± 468. (10)	3237 ± 1508 (10)	2272 ± 1020 (10)
	LSI	1.7 ± 0.6 (10)	1.4 ± 0.5 (5)	1.2 ± 0.5 (10)	1.4 ± 0.2 (10)	2.6 ± 4.4 (10)
Walleye	Length	48 ± 6.8 (10)	44 ± 2.6 (10)	50 ± 6.6 (10)	51 ± 8.7 (10)	46 ± 13 (10)
	Mass	1740 ± 870 (10)	1092 ± 148(10)	1367 ± 398 (10)	1623 ± 771 (10)	1180 ± 712 (10)
	LSI	1.2 ± 0.4 (10)	1.2 ± 0.3 (10)	1.4 ± 0.3 (10)	1.6 ± 0.5 (10)	1.5 ± 0.4 (10)
Whitefish	Length	42 ± 2.0 (4)	38 ± 1.8 (2)	43 ± 5.8 (10)	41 ± 1.3 (5)	39 ± 2.6 (10)
	Mass	1278 ± 315 (4)	1025 ± 35(2)	1384 ± 392 (10)	990 ± 115.3 (5)	807 ± 197 (10)
	LSI	1.2 ± 0.1 (4)	1.0 ± 0.0 (2)	1.3 ± 0.2 (10)	0.9 ± 0.2 (5)	1.1 ± 0.3 (10)