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Arsenic and mercury in lake whitefish and burbot near the abandoned Giant Mine on Great Slave Lake

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ABSTRACT

Contaminant levels in fish are of public concern in northern Canada where they are an important food source. In this study, we investigated the concentration of total arsenic, four arsenic species (arsenite (AsIII), arsenate (AsV), dimethylarsinate (DMA), and monomethylarsonate (MMA)), and total mercury (Hg) in the muscle and liver of lake whitefish (*Coregonus clupeaformis*) and burbot (*Lota lota*) collected at two sites near the abandoned Giant Mine site (Baker Pond and Yellowknife Bay) and two reference sites more than 25 km away (Chitty Lake and southern Great Slave Lake). Total arsenic concentrations were typically higher in fish tissues collected near the mine site, and higher in burbot than lake whitefish. We found lower concentrations of arsenic in the muscle tissue of adult lake whitefish than juveniles. All four arsenic species were only detected in the liver tissues of adult lake whitefish collected from Baker Pond on the mine site, and juvenile lake whitefish from the adjacent Yellowknife Bay. Mercury levels were highest in fish from Chitty Lake, and higher for burbot than lake whitefish, similar with other research reporting elevated mercury in small northern lakes relative to larger waterbodies. However, mercury levels in fish were not elevated beyond consumption guidelines. Elevated arsenic concentrations in the fish tissues collected near the mine site suggest that the area continues to be a source of arsenic to the aquatic food web; therefore, continued monitoring is warranted, particularly with a large portion of the local population harvesting wild food sources.

Introduction

In Canada's Northwest Territories (NWT), fish are an important and culturally valued resource, with approximately 40% of the population hunting and fishing to supplement their diet (GNWT, 2014), and 25% of the total population partaking in recreational angling, which is, per capita, more than most other jurisdictions in Canada (Fisheries and Oceans Canada, 2012). Yellowknife Bay, on the north shore of Great Slave Lake, supports many species of large bodied fish that are used as food by people in the City of Yellowknife and the nearby aboriginal communities of Dettah and N'dilo. Large bodied fish are known to accumulate contaminants and often have elevated concentrations relative to biota lower in the food chain (e.g., Evans et al., 2005; Kidd et al., 2012). Elevated concentrations of certain metals in fish, such as mercury and arsenic, are of public concern because of the well documented health risks associated with consuming fish with high metal burdens (Canadian Council of Ministers of the Environment (CCME), 2000; Canadian Food Inspection Agency (CFIA), 2014; Health Canada, 2012).

Mercury is of particular concern because of its documented toxicity, persistence in the environment, high potential for bioaccumulation in the aquatic food web and ability to biomagnify with increasing trophic levels (CCME, 2000; Kidd et al., 2012; Wiener et al., 2003). Toxic effects on fish include disrupted neurological function and reduced growth, oxygen uptake, reproductive development, sensory abilities, osmoregulation, and digestion (Kidd et al., 2012; Scheuhammer et al., 2015; Wiener et al., 2003). In the aquatic environment, mercury can be converted through biogeochemical interactions to the more toxic organic methylmercury (MeHg) (CCME, 2000; Chetelat et al., 2015; Jensen and Jernelöv, 1969; Winfrey and Rudd, 1990). Fish accumulate MeHg through their diet (Rodgers, 1994). Fish tissues are typically analyzed for total mercury, since it has been demonstrated that the majority of mercury in fish is present as MeHg (Bloom, 1992; Forsyth et al., 2004). Mercury is naturally occurring in the environment (Lockhart et al., 2005), but levels can be exacerbated by the cumulative impacts of natural disturbances like fire (Garcia and Carignan, 1999; Kelly et al., 2006) and anthropogenic activities such as logging (Garcia and Carignan, 1999), mining (Lockhart et al., 2005), flooding for hydroelectric development (Bodaly et al., 1984), or atmospheric inputs from waste incineration and fossil fuel emissions (Kidd et al., 2012).

Point-source contamination from industrial activities into the aquatic environment is a prime public concern. Arsenic contamination is often associated with historic gold mining activity since arsenic commonly occurs in the ore of gold bearing metal sulfide deposits (Cohen

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and Bowell, 2014). The toxicity of arsenic is well known (Bowell and Craw, 2014), being a carcinogen in humans (Kapaj et al., 2006) and causing toxic and biological effects on fish (Pedlar et al., 2002b). It bioaccumulates in the aquatic environment, but concentrations normally decrease with trophic position (Chen and Folt, 2000; Dutton and Fisher, 2011; McIntyre and Linton, 2012). Adverse impacts to fish include effects to growth and reproduction through diminished appetite, altered feeding behavior, increased abnormalities, reductions in gonadal development, spawning success in adults, hatching success of eggs, and overall health and survival during the early life stages (McIntyre and Linton, 2012; Wiener et al., 2003).

Like mercury, arsenic has different species (or forms), and these species have varying toxicities due to different physical and chemical properties. The inorganic forms of arsenic [arsenite (AsIII) and arsenate (AsV)] are generally considered more toxic than organic species [dimethylarsinate (DMA) and monomethylarsonate (MMA)], with the trivalent form generally considered the more toxic of the two inorganic species (McIntyre and Linton, 2012). Arsenic can be absorbed directly through the gills of fish as well as through the gut, where it can be methylated from an inorganic to organic form (McIntyre and Linton, 2012).

Giant Mine on the shore of Yellowknife Bay in Great Slave Lake operated between 1948 and 2004 and was one of the most productive gold mines in Canadian history (Government of Canada, 2011). Gold production at Giant Mine also resulted in the production and subsequent storage of large amounts of arsenic waste onsite, rendering the mine site one of the most contaminated sites in Canada (Federal Contaminated Sites Action Plan (FCSAP), 2014; Office of the Auditor General of Canada (OAG), 2012; Government of Canada, 2011). Close to 260,000 tonnes of arsenic trioxide (As_2O_3) was generated as a byproduct of gold ore processing at Giant Mine. Two hundred thirty seven thousand tonnes of As_2O_3 was captured by emission control technologies over the operating life of the mine and is currently stored underground; however, 20,000 tonnes was not captured and was released to the surrounding landscape via emissions from the roaster stacks employed at the mine (Jamieson, 2014; Wrye, 2008). Over many decades, in addition to the atmospheric fallout from historic roaster stack emissions, Yellowknife Bay received indirect anthropogenic inputs of arsenic through the discharge of mine wastewater via Baker Creek, and the historical deposition and erosion of tailings along the northeast shoreline of Yellowknife Bay (Andrade et al., 2010). In the early years of mine operations (1948–1951), tailings were deposited directly into Yellowknife Bay in a small embayment on the north shore. These tailings have subsequently redistributed within Yellowknife Bay over time (Golder Associates Limited, 2005).

Arsenic loading to Yellowknife Bay has long been a concern with several earlier studies identifying elevated levels of arsenic in sediment and surface waters of Yellowknife Bay (Jackson et al., 1996; Mace, 1998; Moore et al., 1978; Mudroch et al., 1989). However, relatively little information is available regarding arsenic in fish in Yellowknife Bay (Jackson et al., 1996; de Rosemond et al., 2008), and none from sites directly on the mine site.

The primary objective of this work was to evaluate arsenic and mercury burdens in fish close to Giant Mine and compare these data with that from fish collected from reference lakes beyond the influence of historic mining activity at Giant. We hypothesize that arsenic concentrations will be highest in the fish tissues collected nearest the mine site and higher in fish with lower trophic status, whereas mercury concentrations will be higher in the tissues of fish collected from small lakes and with higher trophic positioning. We analyzed metal concentrations in the liver and muscle tissues from juvenile and adult lake whitefish (*Coregonus clupeaformis*) and adult burbot (*Lota lota*), two large bodied fish species that occur in Yellowknife Bay. These species were selected as they are commonly harvested fish for human consumption (flesh of both species and the liver of burbot are eaten) and represent different trophic positions. Lake whitefish feed mainly on plankton as juveniles, and benthic invertebrates as adults (Scott and Crossman, 1973). Therefore depending on size, lake whitefish can be classified into different trophic levels. Burbot are top-level predators having an almost exclusively fish-based diet as adults (Amundsen et al., 2003) and occupy a higher trophic position than lake whitefish (Cott et al., 2011).

Methods

Sample sites and fish collections

Lake whitefish and burbot were collected at four locations within 200 km of Giant Mine, Yellowknife, NWT. Lake whitefish collected from southern Great Slave Lake were obtained from local commercial fisherman, and lake whitefish from all other sampling locations were captured using multi-mesh gillnets. Burbot were targeted using longlines baited with cisco (*Coregonus artedi*). All fish were immediately killed upon capture, placed on ice, and frozen until dissection. Fork length and total length (±1.0 mm) were recorded for lake whitefish and burbot, respectively, and total body mass (±1.0 g wet) was measured for both fish species. Tissue samples of skinless white dorsal muscle and liver (±10 g) were collected from each fish, placed in individual small plastic bags and frozen at −20 °C for subsequent analyses.

In December 2010, adult lake whitefish (n = 8) were collected from Baker Pond, a reach of Baker Creek (62° 30′ 28 N 114° 21′ 32 W), which flows through the Giant Mine site into Yellowknife Bay (Fig. 1). Historically, Baker Pond was the receiving environment for Giant Mine's tailings and treated waste water (Fawcett et al., 2015).

Concentrations of arsenic in surface waters and sediments in Baker Pond vary seasonally in association with changes in redox conditions and changing inputs from the catchment. Reported values for arsenic in surface waters and sediments range from 200 to 4000 μg/L and from 2000 to 14,000 mg/kg, respectively (Nash, 2015; Fawcett et al., 2015; Walker et al., 2015). At the time of fish collection there was less than 1 m of water below the ice.

In March 2012, adult (n = 8) and juvenile (n = 8) lake whitefish, and adult burbot (n = 8) were collected from Yellowknife Bay, Great Slave Lake (62° 24′ 40 N 114° 20′ 13 W), approximately 1 km from Giant Mine (Fig. 1). Previous research has shown that this area of Yellowknife Bay (Back Bay) has been impacted by historical mining activities, either through the discharge and redistribution of tailings and wastewater or via roaster emissions (Jackson et al., 1996; Andrade et al., 2010). Concentrations of arsenic in surface waters of Yellowknife Bay vary seasonally and recently reported concentrations range between 0.5 and 10 μg/L (Jackson et al., 1996; Andrade et al., 2010). The lacustrine sediments of Yellowknife Bay act as both a source and a sink of arsenic to overlying waters, dependent on redox conditions and other biogeochemical factors (Andrade et al., 2010). Values of reported concentrations of arsenic in sediments from the main basin of Back Bay range between 53 and 1000 mg/kg (Jackson et al., 1996; Andrade et al., 2010).

In June 2012, adult lake whitefish (n = 9) and burbot (n = 8) were collected from Chitty Lake (62° 42′ 48 N 114° 7′ 54 W), approximately 25 km northeast of Giant Mine in an area expected to be beyond the influence of historic roaster arsenic emissions on water, sediment and aquatic biota (Wagemann et al., 1978) (Fig. 1). Wagemann et al. (1978) report arsenic concentrations in water and sediment as <10 μg/L and 28 mg/kg, respectively. Chitty Lake is primarily surrounded by Archean metasedimentary rocks of the Yellowknife Supergroup. Arsenic concentrations are generally lower in this unit compared to bedrock of the Yellowknife Greenstone Belt (Boyle, 1960; Galloway et al., 2015); therefore, geogenic inputs of arsenic to lake sediments and water are expected to be low.

In July 2012, adult lake whitefish (n = 8) were collected from the south side of Great Slave Lake, approximately 10 km north of Hay River, NWT (60° 59′ 32 N 115° 41′ 10 W). The southern and eastern shores of Great Slave Lake are part of the Western Canadian Sedimentary Basin.

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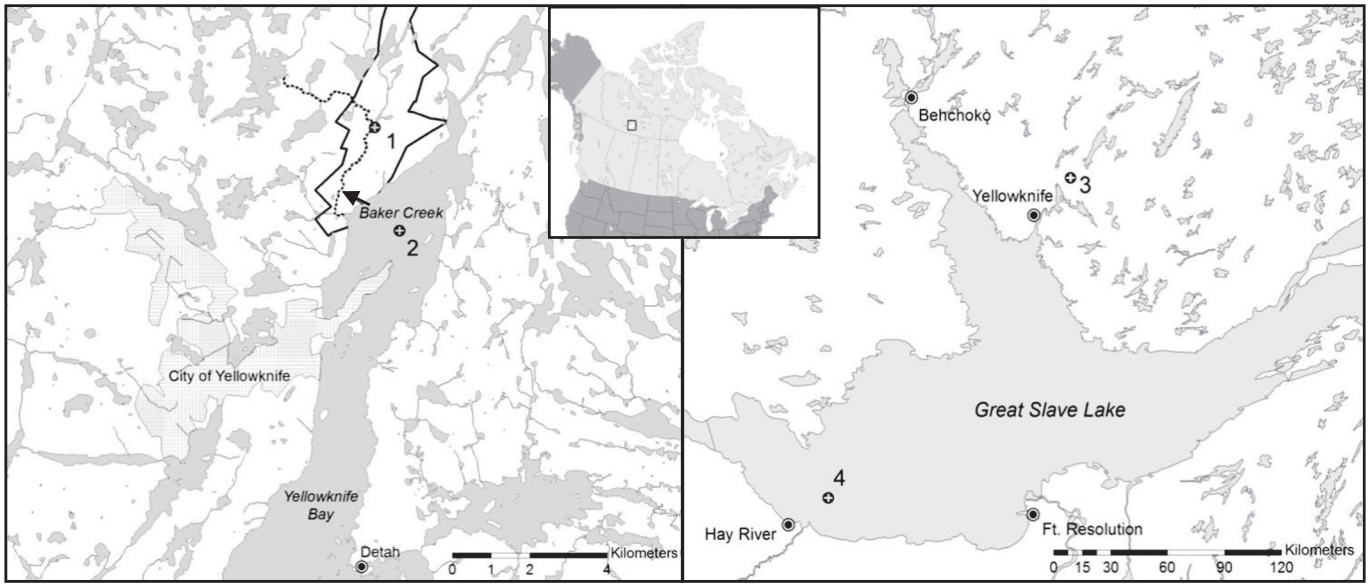

Fig. 1. Locations and proximity to Yellowknife, Northwest Territories, Canada, where fish were collected for arsenic and mercury analyses. 1 = Baker Pond on the Giant Mine Lease Site, 2 = Yellowknife Bay, Great Slave Lake, 3 = Chitty Lake, 4 = Great Slave Lake, near Hay River. The black line left panel denotes the Giant Mine lease area. The gray hashed area represents the urbanized area of the City of Yellowknife. Note that the Giant Mine lease area falls within Yellowknife city limits.

Laboratory analyses

All analyses were performed at the ALS analytical laboratory in Vancouver with the exception of arsenic speciation, which was performed at the ALS laboratory in Sweden. The metal analyses were carried out using methods adapted from US EPA Method 200.3 "Sample Procedures for Spectrochemical Determination of Total Recoverable Elements in Biological Tissues" (1996). Tissue samples were homogenized and subsampled prior to hotblock digestion with nitric and hydrochloric acids, in combination with repeated additions of hydrogen peroxide. Total metal analysis was performed by inductively coupled plasma-mass spectrometry, adapted from US EPA Method 6020A (note only total arsenic and total mercury data are reported here). Both total arsenic and total mercury analyzed had detection limits of 0.01 mg/kg wet weight (ww). Mercury analysis for dry samples was performed by atomic fluorescence spectrophotometry, adapted from US EPA Method 245.7. This method had a detection limit of 0.05 mg/kg dry weight (dw).

Tissue samples used for arsenic speciation were homogenized, and then an ultrasound-assisted extraction into 1 + 1 methanol + water was used to obtain extract which was then filtered (0.45 μm) and diluted 10×. Extracts were analyzed by ion chromatography (Hamilton PRP-X100 column in a Bischoff gradient system) with post column hydride generation for improved sensitivity. Detection was enabled with inductively coupled plasma mass spectrometry (ICP-MS; Thermo Fisher Element 2). The arsenic species analyzed and associated detection limits were as follows: arsenite (AsIII; 0.01 mg/kg dw), arsenate (AsV; 0.04 mg/kg dw), dimethylarsinate (DMA; 0.01 mg/kg dw), and monomethylarsonate (MMA; 0.02 mg/kg dw).

Quality assurance and control were done as per ALS Laboratories' quality control protocol (ALS, 2012). Quality control protocol includes instrument (e.g. verification of initial calibration, second source calibration verification standard, continuing calibration verification, and instrument blanks) and method quality controls (e.g. method blanks, laboratory duplicate samples, and calibration to reference material) (ALS, 2012).

Statistical analysis

The experimental design is an unbalanced one way design. The overall statistical hypothesis of 'no difference among locations' is tested using analysis of variance under the assumption of normality, or a Kruskal Wallis test if the assumption of normality could not be met using a logarithmic transformation. Post-hoc multiple comparisons among all means are conducted using Tukey's test modified for unequal sample sizes (Zar, 1999) whereas multiple comparisons among all medians are conducted following Gao et al. (2008). For both post-hoc tests, alternative hypotheses were two sided and the Holm adjustment for P-values (Holm, 1979) was used to control the overall Type I error rate to account for multiple comparisons. When more than two comparisons were possible, multiple comparisons were conducted only if the null hypothesis of equality of means or medians was rejected using a 5% level of significance. Total arsenic and total mercury analyses were conducted on a ww basis. To account for size and age related variability in metal uptake, fish of similar size (by site and species/maturity) were selected for analysis (see Tables 1 and 2). Box and Whisker plots were used to present the data spread, a robust measure of central tendency, and to flag "aberrant" observations. The bottom, middle and top of the box are the 25th, 50th (median) and 75th percentiles respectively. The upper "whisker" is drawn at the first observation that exceeds the median + 1.5X the interquartile range; observations greater than this are flagged as "aberrant" using a circle. The lower whisker is similarly defined. All statistical analysis were conducted using R (R Core Team, 2014).

Results

Summary statistics of concentrations per species per tissue per site for total arsenic and total mercury are presented in Table 1 and for arsenic species in Table 2.

Total arsenic in fish tissues

The maximum total arsenic concentration found in lake whitefish muscle was from Baker Pond at 0.57 mg/kg ww (Table 1). Total arsenic concentrations were significantly higher in Baker Pond than Yellowknife Bay, Chitty Lake, and southern Great Slave Lake (P = <0.0001, 0.017, and 0.12 respectively), with no difference detected between the other sites (Fig. 2). The concentrations of total arsenic in the livers of lake whitefish were higher than that of the muscle for Baker Pond and Yellowknife Bay, with a maximum concentration of 1.38 mg/kg ww

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Table 1
Total arsenic (As) and total mercury (Hg) concentrations in fish by location, species, and tissue. All fish were collected in the Northwest Territories, Canada within 200 km of Yellowknife. Values preceded by < below detection limits, nd = no data. Both wet weight (ww) and dry weight (dw) are represented. LKWH = lake whitefish, BURB = burbot, j = juvenile.

Parameter	Tissue	Species	Baker Pond	Yellowknife Bay	Chitty Lake	GSL South
			Mean (range) SD ±	Mean (range) SD ±	Mean (range) SD ±	Mean (range) SD ±
Length (mm)		LKWH	371.6 (315–446) 41.12	366.3 (310–420) 31.77	432.9 (398–449) 17.51	420 (362–472) 33.37
		LKWH (j)	nd	202.3 (195–210) 5.59	nd	nd
		BURB	nd	508.4 (390–605) 71.25	520.5 (456–652) 56.23	nd
Total As (mg/kg ww)	Muscle	LKWH	0.49 (0.31–0.57) 0.09	0.19 (0.12–0.27) 0.06	0.28 (0.18–0.64) 0.15	0.19 (0.1–0.35) 0.11
	Liver	LKWH	0.55 (0.2–1.38) 0.36	0.32 (0.25–0.4) 0.05	0.18 (0.12–0.24) 0.04	nd
	Muscle	LKWH (j)	nd	0.18 (0.11–0.35) 0.08	nd	nd
	Liver	LKWH (j)	nd	0.57 (0.3–0.76) 0.15	nd	nd
	Muscle	BURB	nd	0.29 (0.22–0.37) 0.05	0.3 (0.15–0.42) 0.11	nd
	Liver	BURB	nd	1.36 (0.85–1.82) 0.3	0.36 (0.22–0.53) 0.09	nd
Total As (mg/kg dw)	Muscle	LKWH	1.86 (0.05–02.44) 0.75	0.19 (0.54–1.26) 0.27	1.26 (0.88–2.80) 0.64	0.77 (0.41–1.39) 0.42
	Liver	LKWH	1.79 (0.64–4.68) 1.24	1.27 (0.97–1.51) 0.20	0.69 (0.47–0.91) 0.15	nd
	Muscle	LKWH (j)	nd	1.0 (0.57–1.66) 0.38	nd	nd
	Liver	LKWH (j)	nd	2.0 (0.94–2.91) 0.61	nd	nd
	Muscle	BURB	nd	1.45 (1.14–1.88) 0.24	1.3 (0.68–2.1) 0.57	nd
	Liver	BURB	nd	2.46 (1.45–3.25) 0.61	0.81 (0.5–1.72) 0.40	nd
Total Hg (mg/kg ww)	Muscle	LKWH	0.07 (0.05–0.14) 0.03	0.05 (0.03–0.09) 0.02	0.1 (0.07–0.18) 0.04	0.04 (0.03–0.06) 0.01
	Liver	LKWH	0.09 (0.04–0.18) 0.05	0.09 (0.06–0.15) 0.03	0.23 (0.11–0.42) 0.11	nd
	Muscle	LKWH (j)	nd	0.03 (0.03–0.04) <0.01	nd	nd
	Liver	LKWH (j)	nd	0.04 (0.02–0.08) 0.03	nd	nd
	Muscle	BURB	nd	0.08 (0.05–0.12) 0.02	0.18 (0.06–0.26) 0.07	nd
	Liver	BURB	nd	0.01 (<0.01–0.02) 0.01	0.06 (0.02–0.26) 0.08	nd
Total Hg (mg/kg dw)	Muscle	LKWH	0.3 (0.2–0.56) 0.12	0.24 (0.15–0.45) 0.09	0.47 (0.3–0.81) 0.21	0.18 (0.12–0.25) 0.05
	Liver	LKWH	0.28 (0.12–0.57) 0.15	0.35 (0.23–0.55) 0.13	0.90 (0.38–1.76) 0.45	nd
	Muscle	LKWH (j)	nd	0.19 (0.15–0.32) 0.05	nd	nd
	Liver	LKWH (j)	nd	0.13 (0.05–0.29) 0.1	nd	nd
	Muscle	BURB	nd	0.40 (0.27–0.61) 0.11	0.84 (0.12–1.24) 0.36	nd
	Liver	BURB	nd	0.02 (0.01–0.04) 0.01	0.22 (0.04–1.28) 0.43	nd

from the former location. However, in Chitty Lake concentrations in the muscle were higher than in the liver (Table 1). Total arsenic concentrations in the liver tissue of lake whitefish from Chitty Lake were significantly lower than both Baker Pond (P = <0.0001) and Yellowknife Bay (P = <0.0001) (Fig. 2). The total arsenic levels in the muscle of lake whitefish were similar between adults and juveniles, but was significantly elevated in the livers of juveniles compared to adults (P = <0.0001) (Fig. 3). There was no difference detected between the levels of total arsenic in the muscle of burbot collected from Yellowknife Bay and Chitty Lake. The arsenic in the liver tissues, however, was

Table 2
Arsenic species [arsenite (AsIII), arsenate (AsV), dimethylarsinate (DMA), and monomethylarsonate (MMA)] concentrations in fish by location, species, and tissue. All fish were collected in the Northwest Territories, Canada within 200 km of Yellowknife. Values preceded by < below detection limits, nd = no data. LKWH = lake whitefish, BURB = burbot, j = juvenile.

Parameter	Tissue	Species	Baker Pond	Yellowknife Bay	Chitty Lake	GSL South
			Mean (range) SD ±	Mean (range) SD ±	Mean (range) SD ±	Mean (range) SD ±
Length (mm)		LKWH	371.6 (315–446) 41.12	366.3 (310–420) 31.77	432.9 (398–449) 17.51	420 (362–472) 33.37
		LKWH (j)	nd	202.3 (195–210) 5.59	nd	nd
		BURB	nd	508.4 (390–605) 71.25	520.5 (456–652) 56.23	nd
As (III) (mg/kg dw)	Muscle	LKWH	<0.01	<0.01	<0.01	<0.01
	Liver	LKWH	0.02 (0.01–0.05) 0.02	<0.01	0.01 (0.01) <0.01	nd
	Muscle	LKWH (j)	nd	<0.01	nd	nd
	Liver	LKWH (j)	nd	0.02 (0.02) <0.01	nd	nd
	Muscle	BURB	nd	<0.01	<0.01	nd
	Liver	BURB	nd	<0.01	<0.01	nd
As (V) (mg/kg dw)	Muscle	LKWH	<0.04	<0.04	<0.04	<0.04
	Liver	LKWH	0.04 (0.04–0.05) <0.01	<0.04	<0.04	nd
	Muscle	LKWH (j)	nd	<0.04	nd	nd
	Liver	LKWH (j)	nd	0.08 (0.08) <0.01	nd	nd
	Muscle	BURB	nd	<0.04	<0.04	nd
	Liver	BURB	nd	<0.04	<0.04	nd
MMA (mg/kg dw)	Muscle	LKWH	<0.02	<0.02	<0.02	<0.02
	Liver	LKWH	0.02 (0.02–0.03) <0.01	<0.02	<0.02	nd
	Muscle	LKWH (j)	nd	<0.02	nd	nd
	Liver	LKWH (j)	nd	0.04 (0.04) <0.01	nd	nd
	Muscle	BURB	nd	<0.02	<0.02	nd
	Liver	BURB	nd	<0.02	<0.02	nd
DMA (mg/kg dw)	Muscle	LKWH	<0.01	<0.01	<0.01	<0.01
	Liver	LKWH	0.01 (0.01–0.02) <0.01	<0.01	<0.01	nd
	Muscle	LKWH (j)	nd	<0.01	nd	nd
	Liver	LKWH (j)	nd	0.02 (0.02) <0.01	nd	nd
	Muscle	BURB	nd	0.05 (0.03–0.08) 0.02	0.02 (0.01–0.09) 0.03	nd
	Liver	BURB	nd	0.05 (0.24–0.86) 0.23	0.08 (0.01–0.18) 0.06	nd

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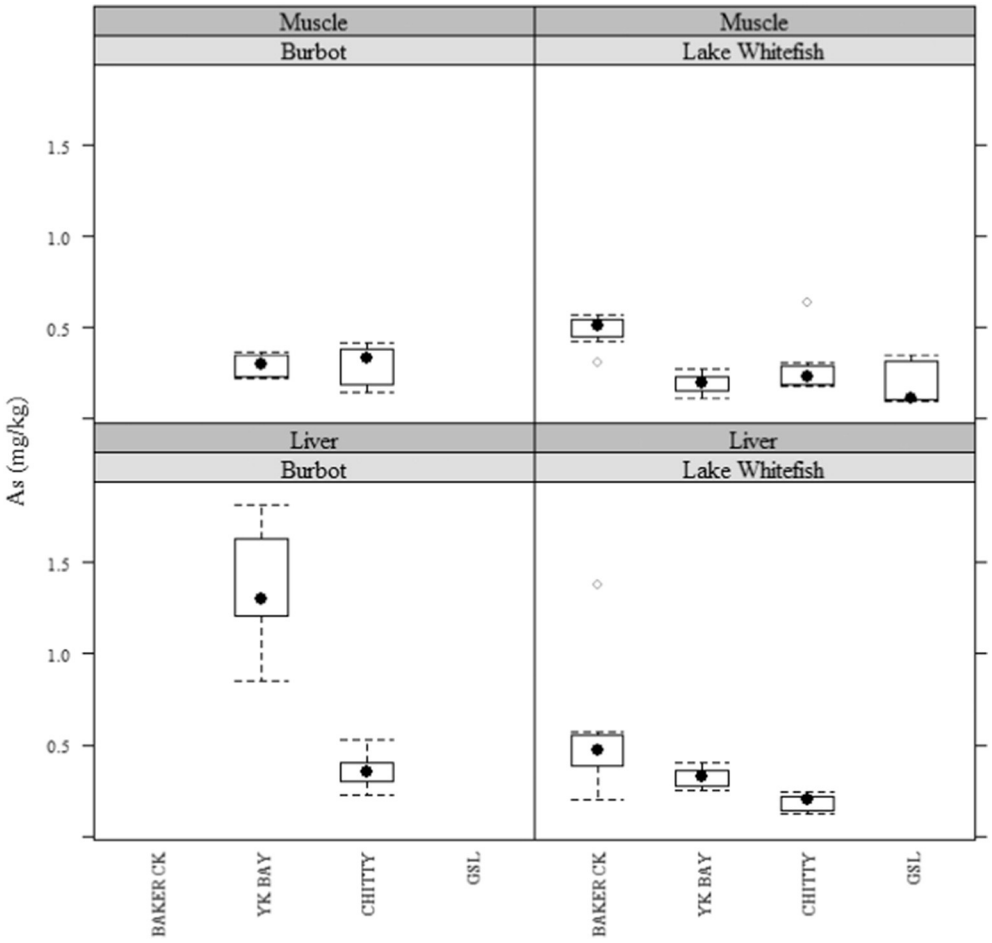

Fig. 2. Total arsenic (As) concentrations in fish by location, species, and tissue. All fish were collected in the Northwest Territories, Canada within 200 km of Yellowknife. Values are expressed as mg/kg (ppm) wet weight. The bottom, middle and top of the box are the 25th, 50th (median) and 75th percentiles respectively. The upper "whisker" is drawn at the first observation that exceeds the median + 1.5 x the interquartile range. Observations greater than this are flagged as "aberrant" using a circle. The lower whisker is similarly defined. Significant pairwise differences were found between all sites for burbot liver, between all sites for lake whitefish liver (with exception of YK Bay–Baker Ck), and between Baker Ck and all sites for lake whitefish muscle. BAKER CK = Baker Pond (a widening of Baker Creek), YK BAY = Yellowknife Bay, CHITTY = Chitty Lake, GSL = the south side of Great Slave Lake (near Hay River).

significantly higher in burbot from Yellowknife Bay (P = <0.0001; Fig. 2). The highest total arsenic concentration was recorded from a Yellowknife Bay burbot liver at 1.82 mg/kg ww (Table 1).

Arsenic species in fish tissues

All four arsenic species were found in the liver tissue of lake whitefish from Baker Pond (Table 2). The inorganic and more toxic trivalent form, As(III), was the most common species detected, found in 25% of the liver samples, followed by As(V) (12.5%), DMA (12.5%) and MMA (12.5%). As(III) was also found at detection limits in lake whitefish liver tissue from Chitty Lake (Table 2), detected in 12.5% of the livers sampled. No arsenic species were above detection limits for any lake whitefish muscle tissues collected, or from the lake whitefish liver tissues collected from Yellowknife Bay (Table 2). The inorganic DMA was the only form of arsenic found in burbot (Table 2), and was detected in 100% of the muscle and liver tissue samples taken from Yellowknife Bay, and 37.5% and 87.5% of the muscle and liver tissue samples collected from Chitty Lake burbot, respectively.

Mercury in fish tissues

The highest recorded total mercury concentration for lake whitefish muscle tissue (0.18 mg/kg ww) was detected from Chitty Lake (Table 1), where concentrations were significantly higher than Baker Pond (P = 0.023), Yellowknife Bay (P < 0.001), and southern Great Slave Lake (P = 0.001). Mercury concentrations in lake whitefish muscle tissue from Baker Pond were significantly higher than in Yellowknife Bay (P = 0.001), and southern Great Slave Lake (P = 0.023) (Fig. 4). The results for lake whitefish livers were similar, with the maximum recorded mercury concentration (0.42 mg/kg ww) detected in Chitty Lake (Table 1) and significantly elevated mercury concentrations in samples from Chitty Lake compared to those from Baker Pond (P < 0.0001) or Yellowknife Bay (P < 0.0001) (Table 2, Fig. 4). The total mercury levels in the muscle and liver tissues of adult lake whitefish from Yellowknife Bay were significantly higher than juveniles (P = 0.004 and 0.022 respectively) (Table 1, Fig. 3). The concentration of total mercury was significantly higher in burbot collected from Chitty Lake compared to Yellowknife Bay for both muscle (P < 0.0001) and liver (P < 0.0001) tissues (Fig. 4). The maximum total mercury concentration found in Chitty Lake burbot was 0.26 mg/kg ww, for both muscle and liver (Table 1).

Discussion

Total arsenic

Arsenic concentrations in adult lake whitefish were highest in fish collected on the Giant Mine property, an area known to have arsenic laden sediments. Sediments from the mouth of Baker Creek have been shown to have arsenic concentrations as high as 2550 mg/kg dw (Jackson et al., 1996), submerged tailings in Yellowknife Bay as high as 3685 mg/kg dw (EBA Engineering Consultants, 2001), and surface

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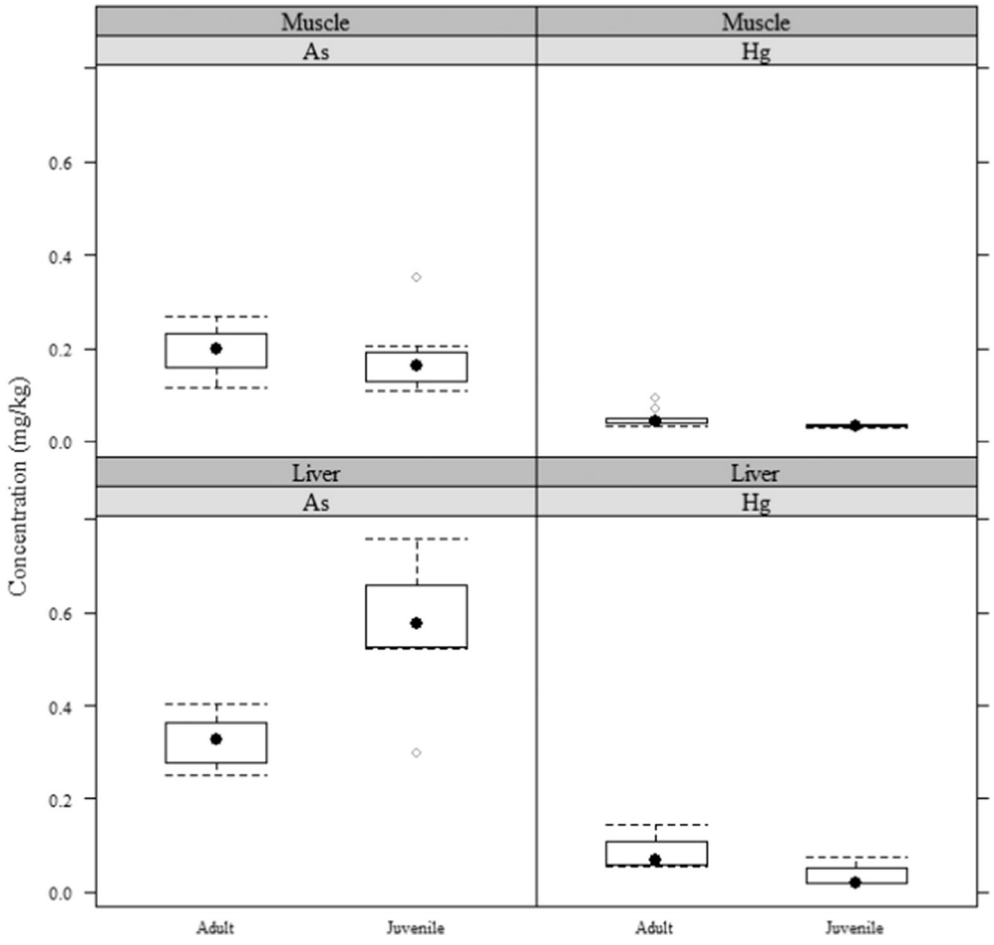

Fig. 3. A comparison of total arsenic (As) and total mercury (Hg) concentrations in the muscle and liver tissues between juvenile and adult lake whitefish in Yellowknife Bay, near Giant Mine, Northwest Territories, Canada. Values are expressed as mg/kg (ppm) wet weight. The bottom, middle and top of the box are the 25th, 50th (median) and 75th percentiles respectively. The upper "whisker" is drawn at the first observation that exceeds the median + 1.5 x the interquartile range. Observations greater than this are flagged as "aberrant" using a circle. The lower whisker is similarly defined. Significant pairwise differences were found between juvenile and adults for all Hg concentrations in the liver and muscle, and As concentrations in the muscle.

sediments in Back Bay up to 1050 mg/kg dw (Andrade et al., 2010). Although arsenic can be absorbed directly from water through the gills (McIntyre and Linton, 2012), arsenic concentrations in sediments and water are not as good of an indicator of bioavailability or toxicity to fish as dietary exposure (Pedlar and Klaverkamp, 2002). Dietary sources of arsenic are absorbed through the gastrointestinal tract and circulated to the other tissues. The liver is known to be a focal organ for arsenic toxicity as it actively accumulates and excretes arsenic in its detoxification role. This is typically reflected by the higher concentrations found in the liver than in other tissues in exposed fish (Pedlar and Klaverkamp, 2002; Sorensen, 1991). Although previous research has shown that fish muscle tissue is not a major repository for arsenic (Pedlar et al., 2002; Pedlar and Klaverkamp, 2002; Sorensen, 1991), our research clearly shows that lake whitefish from the Baker Pond site (an area directly impacted by tailings) (Golder Associates Limited, 2011; Indian and Northern Affairs Canada (INAC), 2010) accumulated arsenic in their muscle tissue. It is unknown how long these lake whitefish were exposed at this site, but the elevated arsenic levels in the fish tissues suggest that it was for an extended period. In Yellowknife Bay, mean total arsenic concentrations measured in lake whitefish muscle (0.19 mg/kg ww and 0.91 mg/kg dw) and liver (0.32 mg/kg ww and 1.27 mg/kg dw) tissues from our study were similar to those reported by de Rosemond et al. (2008) collected at the same location; 0.77 and 1.07 mg/kg dw for muscle and liver tissues respectively. Concentrations of arsenic were generally lower in the muscle than liver tissue, as was reported by Jackson et al. (1996) and lowest (regardless of species or tissue) at sites that were more than 25 km from Giant Mine. These results are consistent to those of Jackson et al. (1996) and Lafontaine (1997) who also showed deceasing arsenic concentrations away from immediate mining impacts.

Juvenile lake whitefish feed on plankton while adults feed more on benthos (Scott and Crossman, 1973). We found that total arsenic levels were higher in juvenile lake whitefish compared to adults, and this difference is most pronounced in the liver tissues. Arsenic can bioaccumulate in fish, but concentrations generally decrease with trophic position (Chen and Folt, 2000; Dutton and Fisher, 2011; McIntyre and Linton, 2012). Differences in arsenic concentrations among species can reflect variations in feeding habits (Sorensen, 1991). In a contaminated watershed, planktivorous species that fed directly on metal enriched zooplankton were shown to have higher arsenic concentrations than piscivorous species (Chen and Folt, 2000).

The notion that arsenic decreases with trophic position appears to be conflicting. In northern lakes, burbot are at a significantly higher trophic position than lake whitefish (Cott et al., 2011). We found the higher arsenic concentrations in the liver and muscle tissues of burbot from Yellowknife Bay compared to lake whitefish. Jackson et al. (1996) found higher concentrations of arsenic in burbot livers compared to lake whitefish, but found the inverse was true for muscle tissue.

Previous studies have identified a potential pattern in arsenic accumulation over long-term exposure, where initial concentrations in tissues are higher followed by decreased accumulation (Oladimeji et al., 1984; Pedlar and Klaverkamp, 2002; Rankin and Dixon, 1994),

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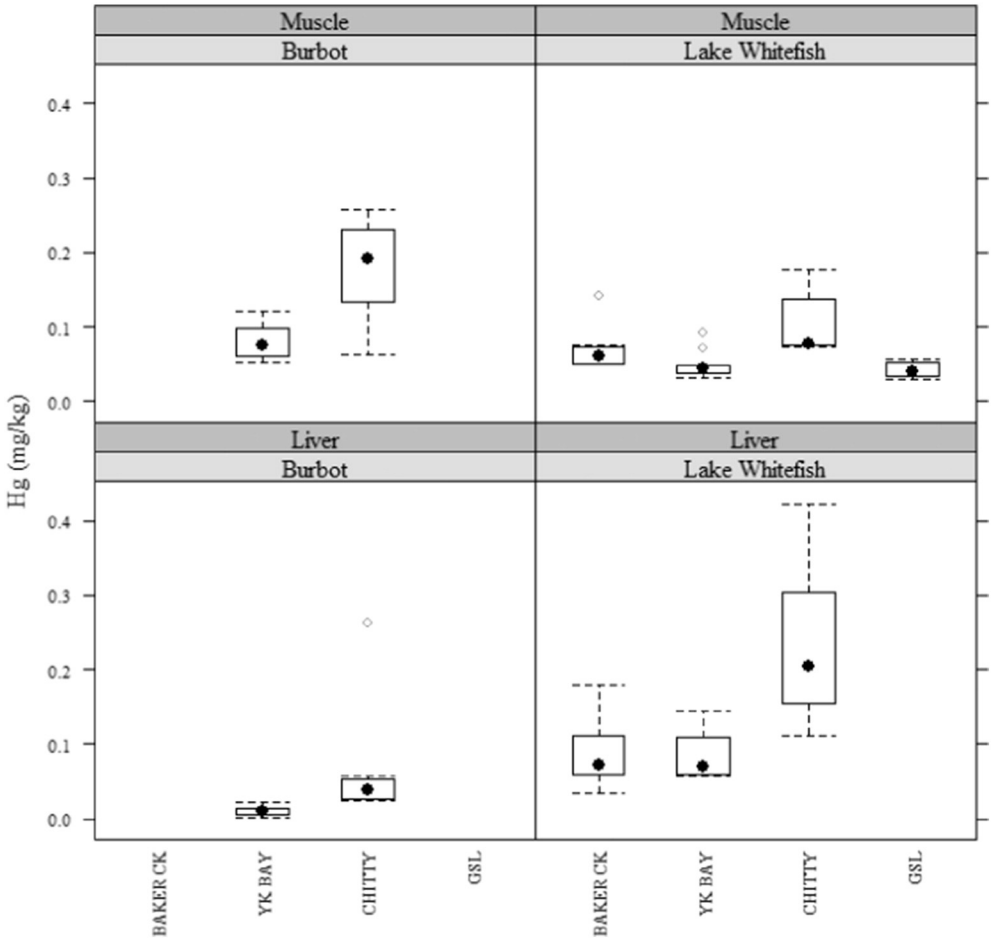

Fig. 4. Total mercury (Hg) concentrations in fish by location, species, and tissue. All fish were collected in the Northwest Territories, Canada within 200 km of Yellowknife. Values are expressed as mg/kg (ppm) wet weight. The bottom, middle and top of the box are the 25th, 50th (median) and 75th percentiles respectively. The upper "whisker" is drawn at the first observation that exceeds the median + 1.5 x the interquartile range. Observations greater than this are flagged as "aberrant" using a circle. The lower whisker is similarly defined. Significant pairwise differences were found between sites for burbot liver, between all sites for lake whitefish liver (with exception of YK Bay–Baker Ck), and between all sites for lake whitefish muscle (except GSL–YK Bay). BAKER CK = Baker Pond (a widening of Baker Creek), YK BAY = Yellowknife Bay, CHITTY = Chitty Lake, GSL = the south side of Great Slave Lake (near Hay River).

indicating that lower concentrations may be related to more efficient excretion of arsenic with continued exposure (Dixon and Sprague, 1981). In Yellowknife Bay, the winter spawning period for burbot occurs in the first three weeks of February (Cott et al., 2014); therefore, it could be that these burbot had more recently migrated into the area closer to Giant Mine for spawning purposes, reducing exposure time compared to the lake whitefish. Burbot likely had had much more direct exposure to the arsenic contaminated sediments in this area of Yellowknife Bay as a result of forming writhing spawning balls of many individuals on the lake bed (McPhail and Paragamian, 2000). Also, burbot are also known to burrow in soft sediments (Boyer et al., 1989), and so would likely have a higher exposure to contaminated pore water and buried contaminated sediments where arsenic concentrations are highest (Andrade et al., 2010), potentially accounting for the higher concentrations of arsenic in burbot from Yellowknife Bay compared to Chitty Lake.

Arsenic species

Inorganic arsenic can be reduced and methylated in the liver of fish to organic forms of arsenic, including MMA and DMA (McIntyre and Linton, 2012). The relative proportion of inorganic arsenic is normally far less than organic arsenic (de Rosemond et al., 2008; McIntyre and Linton, 2012). In our study, of the four arsenic species sampled for in burbot liver from Yellowknife Bay and Chitty Lake, only the organic DMA was above detection levels. However, in more contaminated sites this proportion can be reversed (McIntyre and Linton, 2012). We found this to be the case with fish collected around Giant Mine, with higher relative concentrations of inorganic As(III) than the other species. Also, inorganic arsenic was higher in the livers of juvenile lake whitefish in Yellowknife Bay than in the muscle tissue. We note that there are other species of arsenic (such as arsenobetaine and trimethylated arsenic) that were not detectable by the analytical methods used, and these unidentified arsenic species may also be important. More research is needed on determining arsenic species occurrence in freshwater fish, including background and impacted sites so critical tissue concentration thresholds can be established (McIntyre and Linton, 2012). Guidelines do not currently exist for arsenic (total or arsenic species) in fish tissue for either human (Health Canada, 2012) or for wildlife (CCME, 2000) consumption.

Mercury

Consumption guideline related to human health risk for total mercury in fish is 0.5 mg/kg wet weight (0.5 ppm) "in the edible portion of all retail fish" (Health Canada, 2012). None of the tissue samples analyzed from either burbot or lake whitefish from any of the sites exceeded this consumption guideline. In northern Canada, studies have shown that total mercury concentrations in lake whitefish muscle are quite low, rarely exceeding guideline limits (Lockhart et al., 2005). Consistent with past studies (e.g. Lafontaine, 1997), our research shows that the

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concentrations of mercury in burbot tissues were higher than lake whitefish. The concentrations of mercury in fish tissues were highest at Chitty Lake, which is a relatively productive, small boreal shield lake (Cott et al., 2011). The concentrations of mercury in burbot livers were lower than in the muscle, but the opposite for lake whitefish.

Higher concentrations of mercury in predatory fish from small northern lakes may be a wide spread phenomenon in the north (see Evans et al., 2005; Lafontaine, 1997; Lockhart et al., 2005, and H.K. Swanson, *unpublished data* in support of the 2015 NWT State of the Environment report) and underscore the need to establish ecological baseline conditions in order to distinguish anthropogenic effects from natural occurring sources. In general, mercury concentrations increase with size, age, and trophic position of the fish (Evans et al., 2005; Kidd et al., 2012), but is known to be variable among sites for lake whitefish and burbot (Depew et al., 2013; Lockhart et al., 2005). Many factors are thought to contribute to variation in mercury concentrations among waterbodies, including underlying bedrock geochemistry (Evans et al., 2005; Lockhart et al., 2005), forest disturbance (Garcia and Carignan, 1999; Kelly et al., 2006), lake size (Evans et al., 2005; Bodaly et al., 1993), lake temperature (Bodaly et al., 1993), and dissolved organic carbon (Evans et al., 2005; French et al., 2014).

Although gold mining operations which use mercury amalgamation extraction processes are known to be sources of mercury contamination to fish (Lockhart et al., 2005), different gold extraction processes (i.e. ore roasting) were used at Giant Mine which did not result in significant mercury contamination to Yellowknife Bay. Mercury contamination in fish is rarely from direct exposure to mine effluent, but rather from biomagnification through the aquatic food web (Wiener et al., 2003).

Mercury concentrations in burbot have been increasing significantly over the past two decades in Great Slave Lake (Evans et al., 2013) and the lower Mackenzie River (Carrie et al., 2009). Several factors have been implicated for this increase including warming temperatures, increasing primary production, and increased industrial emissions in Asia. Regardless of the source, continued collection of standardized data on contaminants in the aquatic environment will allow for monitoring of the situation over time.

Conclusions

As anticipated, we found that arsenic levels in burbot and lake whitefish were typically highest at or near Giant Mine compared to sites geographically situated outside of the impacted mine footprint and immediate surrounding area. Detection of the inorganic As species As(III) in lake whitefish was most common at the Giant Mine site. This suggests that the abandoned mine site continues to be a source of arsenic to the aquatic food web. Total mercury concentrations were below the Health Canada consumption guideline for all species and tissues sampled, from all locations. Mercury concentrations were higher in fish from the inland lake sampled compared to those from Great Slave Lake and showed greater concentrations in fish with a higher trophic position in the food web at all locations.

Giant Mine is an extremely expensive legacy issue, with remediation and perpetual maintenance costs approaching $1 billion CAD (Aboriginal Affairs and Northern Development Canada, 2012). The focus of monitoring and remediation has been on the Giant Mine site lease area for which the Government of Canada is responsible (INAC, 2007, 2010). The scope of the Environmental Assessment for the remediation project was also limited to the lease area, including environmental impact analysis and supporting studies (Mackenzie Valley Environmental Iimpact Review Board, 2008). A major limitation with monitoring of fish and relating it to contaminated sites on large lakes or rivers is that fish move, making it a challenge to determine their actual exposure period to contamination from that site. The high costs and time associated with conducting long-term ecological monitoring programs can minimize the scope or otherwise deter such programs from being initiated (Caughlan and Oakley, 2001; Clark et al., 2010). However, from an environmental and human health perspective it is important to monitor contaminant levels in biota, particularly fish species that are harvested for human consumption, especially in areas of known contamination. While still costly (e.g. arsenic speciation analysis is $250–$390/sample), our approach of using standard, repeatable, techniques on exposure and reference sites using key fish species and targeted tissues will contribute to baseline understanding, allow for refinement of future research projects and monitoring plans and inform the overall remediation plan. Also, this type of information collected in a standard way can add to the collective body of knowledge and be used for larger temporal or regional meta-analysis studies (e.g. Evans et al., 2005; Evans et al., 2013; Depew et al., 2013; Lockhart et al., 2005). There is nevertheless a clear need for additional research, including: 1) establishing the spatial extent and gradients of arsenic concentrations in water, sediment and fish away from Giant Mine; 2) identifying the contaminants from anthropogenic sources and distinguishing those from potentially elevated, but otherwise natural background conditions in the Yellowknife area; 3) investigating the variation in exposure to contaminants though differences in diet or habitat, including intra-species analysis among life stages, growth/age, and between sexes (e.g., Madenjian et al. (2015) found that male burbot had lower levels of mercury than females due to testosterone mediated mercury depletion); 4) developing standardized analytical methods that can consistently detect a greater suite of arsenic species; 5) investigating the potential pathways for contaminant uptake into biota in order to pinpoint remediation efforts; and 6) monitoring of contaminants in fish from populations that are harvested for human consumption.

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