PERSISTENT ORGANIC POLLUTANTS IN THE LIVERS OF MOOSE HARVESTED IN THE SOUTHERN NORTHWEST TERRITORIES, CANADA

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ABSTRACT: Moose (*Alces alces*) are an important traditional and spiritual resource for residents of the southern Northwest Territories and local residents are concerned about contaminants that may be present in the country foods they consume. As part of a larger program looking at contaminants in moose organs, we collected liver samples from moose harvested in two separate but adjoining regions within the Mackenzie River drainage area, the Dehcho and South Slave. We analyzed liver samples for a wide range of persistent organic pollutants (POPs) including polychlorinated biphenyls (PCBs), DDT related compounds, toxaphene, brominated diphenyl ethers (PBDEs) and perfluorinated alkyl substances (PFASs). Overall concentrations of major groups of POPs (total (Σ) PCBs, ΣPBDEs, ΣPFASs were consistently low (generally < 2 ng/g wet weight) in all samples and comparable to the limited data available from moose in Scandinavia. PFASs were the most prominent group with geometric means (range) of 1.3 (0.81–2.5) ng/g ww in the Dehcho and 0.93 (0.63–1.2) ng/g ww in the South Slave region. Decabromodiphenyl ether (BDE-209) was the most prominent PBDE congener, similar to that found in other arctic/subarctic terrestrial herbivores. In general, BDE-209 and PFASs, which are particle-borne and relatively non-volatile, were the predominant organic contaminants.

ALCES VOL. 53: 65–83 (2017)

Key words: Dehcho region, POPs, polychlorinated biphenyls, perfluorooctane sulfonate, persistent organic pollutants, liver, moose, South Slave region, Northwest Territories

INTRODUCTION

Moose (*Alces alces*) are an important source of traditional food and of cultural significance for Canada's northern First Nation communities, and is a frequently consumed food in the southwestern Northwest Territories (NT) (Kuhnlein et al. 1995, Receveur et al. 1997, Berti et al. 1998). The long range transport and deposition of contaminants which often bio-magnify as they move through the food chain are of concern, especially as related to human exposure from consumed country foods (Van Oostdam et al. 2005, Donaldson et al. 2010). Recent studies have documented baseline levels of various heavy metals in the organs of moose from Northern Canada and Alaska (O'Hara et al. 2001, Gamberg et al. 2005a, Gamberg et al. 2005b, Arnold et al. 2006, Landers et al. 2008, Larter et al. 2016); however, information on levels of persistent organic pollutants (POPs) in the tissues of moose is scarce.

Analyses conducted in the early 1970s indicated that moose in Idaho had very low

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levels of organochlorine pesticides (OCPs) (Benson et al. 1973). Liver and muscle of moose from Denali National Park in central Alaska were analysed for a suite of POPs as part of the Western Airborne Contaminants Assessment Project (WACAP) (Landers et al. 2008). Low and variable concentrations of PCBs and hexachlorobenzene (HCB), and OCPs were detected in 3 moose liver and muscle samples; DDT related compounds (*p,p′*- DDD + *p,p′*- DDT, ~34–340 ng/g lipid weight (lw)) and HCB predominated (~0.1–0.72 ng/g lw). Moose muscle, fat, and liver from communities in the Mackenzie Valley of NT that had been cooked/baked had total (Σ) PCB concentrations ranging from 3 to 23 ng/g (Berti et al. 1998), but raw muscle and liver were not analysed. Recent studies in Scandinavia report low concentrations of a wide range of POPs in moose muscle and liver including PCBs, OCPs, polybrominated diphenyl ethers (PBDEs), and polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) (Danielsson et al. 2008, Mariussen et al. 2008, Suutari et al. 2009, Holma-Suutari et al. 2016). A recent assessment of data for POPs in Canadian arctic food webs concluded that PBDEs, particularly decabromodiphenyl ether (BDE-209) and perfluorinated alkyl substances (PFASs), were the predominant halogenated contaminants in caribou (*Rangifer tarandus*) with concentrations typically higher than PCBs or OCPs (Muir et al. 2013); no results for moose were included.

As part of a study assessing baseline concentrations of various contaminants in the organs of moose harvested for consumption by local residents, we analyzed livers to investigate the baseline levels of a wide range of POPs and emerging contaminants of concern including PCBs, OCPs, PFASs, PBDEs, and non-PBDE brominated flame retardants (BFRs). Knowledge of baseline levels of these contaminants in moose is important because comparable data are limited and moose consumption by local residents may increase in future due to the declining availability of caribou as an alternate country food resource.

METHODS

Supporting information regarding more specific description of analytical methods, quality assurance, and raw data is provided in "Supporting Information" at http://alcesjournal.org/index.php/alces. Reference to "Supporting Information" follows throughout, and nomenclature in tables beginning in "S" refers to tables in "Supporting Information."

Study Area

The Dehcho (*ca.* 154,000 km^2) and South Slave (214,000 km^2) are administrative regions of the southern NT located substantially in the northern boreal forest where moose, boreal woodland caribou (*R. t. caribou*), and wood bison (*Bison bison athabascae*) are the dominant ungulates. The samples for this study were collected by local harvesters from Jean Marie River, Hay River, Fort Smith and Fort Resolution (Fig. 1).

Moose samples

First Nation harvesters were requested to provide biological samples and general information from harvested moose. For the purpose of this study, we requested a minimum 5 cm x 5 cm piece of liver, an incisor tooth, and the following information: name of hunter, date and location of harvest, sex, estimated age (calf, yearling, adult), general body condition (excellent, good, fair, poor), and whether pregnant (yes, no) (Table S1). Liver samples (n = 7) from moose harvested in 2006 in the Dehcho and in 2010 in the South Slave (n = 7) were analyzed for 202 individual organohalogen compounds. A first incisor from each moose was forwarded to Matson's Laboratory (Manhattan, Montana, USA) for aging by counting cementum

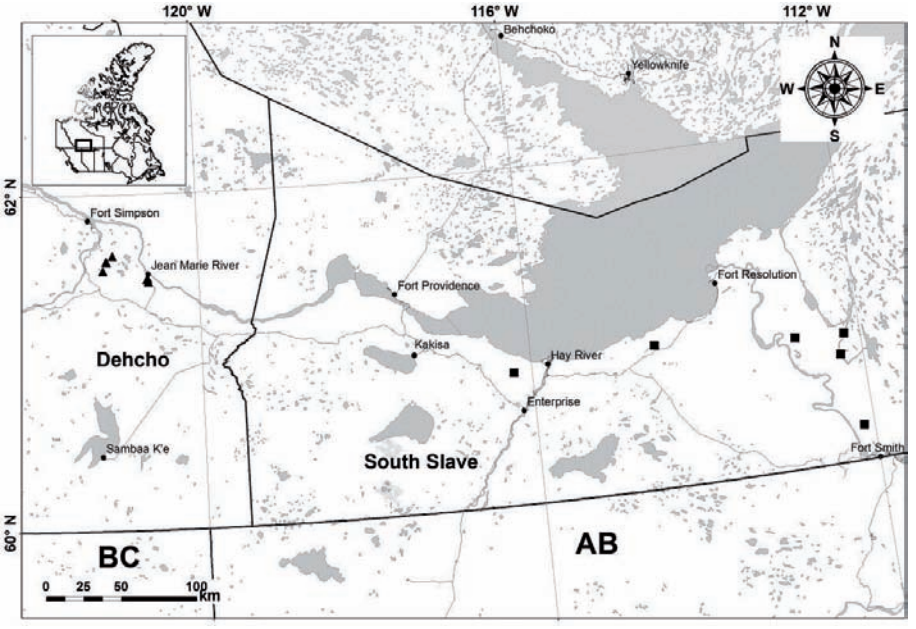

Fig. 1. Locations of moose collected from the eastern Dehcho and South Slave regions of the Northwest Territories showing roads and communities. Moose were harvested in 2016 in Dehcho (▲) and in 2010 in South Slave (■).

annuli from the root of the first incisor; 1 June was used as the birthdate (Matson 1981).

Liver tissue analysis

Liver samples were analyzed for PCBs, organochlorine pesticides (OCPs), and other chlorinated organics (OCOs) following US EPA Method 1699 (US EPA 2007) by ALS Global Laboratories (Burlington, Ontario, Canada), except for 4 samples from the Dehcho which were analysed by Environment Canada (National Laboratory for Environmental Testing [NELT]); both labs are accredited by the Canadian Association for Laboratory Accreditation and ISO 17025 certified. The NLET used previously established methods (Hoekstra et al. 2002, Muir et al. 2006), and 3 samples from the Dehcho were analyzed by both labs (see Quality Assurance section). For both methods, sample preparation was done in a clean room laboratory (positively pressurized with carbon and high-efficiency particulate arresting filters) at the Canada Centre for Inland Waters (CCIW, Burlington, Ontario, Canada). Sample preparation, extraction, and cleanup/isolation of the OCPs, OCOs, and BFRs is described in detail in Supporting Information. Clean extracts were concentrated to a final volume of 40 to 100 L in isooctane prior to analysis by gas chromatography (GC) using either electron capture detection (GC-ECD), GC high-resolution mass spectrometry (GC-HRMS), or GC-low resolution MS (GC-LRMS). A list of individual PCB/OCO/OCP analytes is provided in Supporting Information (Table S2).

The GC-ECD analysis was conducted on 7 Dehcho samples. Final extracts of all samples from South Slave and 3 of 7 from Dehcho were analyzed by GC-HRMS for 31 OCP related compounds (Table S2) using GC-HRMS at ≥10,000 resolution, and for 87 individual + co-eluting PCB congeners using GC-LRMS. Analyses of all PBDEs and other brominated flame retardants (BFRs) toxaphene-related compounds including

22 polychlorinated bornane congeners as well as α- and β-endosulfan and endosulfan sulfate, were measured by GC- electron capture-negative ion mode (ECNI) low resolution mass spectrometry. PFASs were measured in liver samples (0.25–0.30 g) as described by Lescord et al. (2015). Further details of analytical methods for all POPs are provided in Supporting Information.

Quality assurance and data analysis

GC-MS and GC-ECD analysis of the 3 Dehcho samples analysed by both methods indicated that GC-MS yielded 28–75% higher values for PCBs, HCB, and chlordane-related compounds (ΣCHL) and 49% lower values for ΣHCH (Table S3); the discrepancies may reflect the low sample concentrations. Recovery studies for OCPs, PCBs, and BFRs spiked into moose tissue prior to extraction were very good and provided in Tables S4 and S5. Low levels of PCBs and PBDEs were present in lab blanks and therefore all results were blank subtracted. Method detection limits (MDLs) for PCBs, OCP/OCOs, and PBDE/BFRs were calculated for all analytes based on results from 6 laboratory blanks that were analyzed in the same laboratory at approximately the same time, where MDL = 3x standard deviation of the blanks. For analytes with non-detectable blank values, the instrument detection limit (IDL) based on a signal to noise ratio of approximately 10:1 was used for statistical calculations. Further details on quality assurance are provided in Supporting Information.

Preliminary statistical analyses indicated that results for most individual analytes and total (Σ) groups were not normally distributed based on the Shapiro-Wilk statistic <0.05 and coefficients of skewness and kurtosis > 2. Log transformed data generally were normally distributed. Correlations and comparison of means using the Student's *t*-test were conducted with log transformed wet weight data using Systat Version 13 (Systat Software Inc., San Jose, California, USA). Results for males and females were pooled because preliminary analysis showed no significant differences in mean concentrations by sex. Also, mean concentrations of all major analytes were compared between adult moose (n = 8) and calves (≤ 1 yr; n = 6) and no significant differences were found; therefore, only correlations with age were examined. Significance were set at $P \leq 0.05$ for all tests.

RESULTS AND DISCUSSION

Sample and Data Characteristics

The 14 animals sampled were harvested over a wide area and based upon harvester reports, 13 of 14 were considered as excellent or good condition; one calf was rated as fair condition (Table S1). All moose from the Dehcho (n = 7) were harvested near the community of Jean Marie River and likely came from one localized population. South Slave moose were harvested both east and west of the Slave River and were possibly from two localized populations (Fig. 1).

Concentrations of major groups of POPs in moose liver are summarized in Table 1, and results for 202 individual analytes are provided in Tables S6, S7, and S8. A primary indication that low concentrations were common overall is that only 95 of the 202 individual target compounds were detectable (Table S6). Of these, 73 (Dehcho) and 67 (South Slave) were > MDL which is the 99% level of confidence that a given analyte is present (Gomez-Taylor et al. 2003). We report all results in order not to censor the data, but for those analytes <MDL, the results have to be regarded as less certain. This issue is not unique as other studies of POPs in arctic terrestrial herbivores (and vegetation) have faced similar analytical issues. For example, Danielsson et al. (2008) found PBDEs and PFASs were below reporting limits in moose from central Sweden.

Table 1. Mean and range of concentrations of major groups of persistent organic pollutants in moose liver (ng/g wet weight and lipid weight; n = 7 for each region) from the Dehchlo and South Slave regions of Northwest Territories, Canada. See Table S2 in Supporting Information for the full list of analytes represented by each group.

	Dehcho	Dehcho	Dehcho	Dehcho	Dehcho	Dehcho	Dehcho	Dehcho	South Slave	South Slave	South Slave	South Slave	South Slave	South Slave	South Slave	South Slave
	Arith Mean	GM	min	max	Arith Mean	GM	min	max	Arith Mean	GM	min	max	Arith Mean	GM	min	max
Organohalogen group	ng/g ww	ng/g ww	ng/g ww	ng/g ww	ng/g lw	ng/g lw	ng/g lw	ng/g lw	ng/g ww	ng/g ww	ng/g ww	ng/g ww	ng/g lw	ng/g lw	ng/g lw	ng/g lw
% lipid	6.3	6.2	5.3	7.2					5.7	5.7	5.0	6.5				
ΣDDT	0.014	0.014	0.013	0.017	0.22	0.22	0.18	0.26	<0.02		<0.02		<0.20			
ΣCHL	0.104	0.089	0.032	0.150	1.68	1.42	0.47	2.66	0.111	0.093	0.038	0.249	1.89	1.64	0.59	3.82
ΣHCH	0.145	0.138	0.091	0.218	2.36	2.22	1.29	3.86	0.198	0.185	0.122	0.385	3.47	3.27	1.99	5.92
ΣCBz	0.51	0.33	0.11	1.18	8.65	5.32	1.56	21.6	0.83	0.76	0.49	1.50	14.5	13.4	9.28	23.4
ΣPCB	0.65	0.61	0.34	1.08	10.9	9.72	4.71	19.8	0.39	0.36	0.15	0.64	6.87	6.32	3.08	11.5
Σmono-di-CB	0.20	0.05	0.001	0.53	3.34	0.82	0.02	8.78	0.55	0.38	0.04	1.18	9.59	6.76	0.70	21.1
Σtri-CB	0.06	0.01	0.002	0.26	1.00	0.23	0.02	4.52	0.11	0.067	0.020	0.45	1.86	1.18	0.31	6.92
Σtetra-CB	0.12	0.05	0.004	0.42	1.96	0.80	0.06	6.60	0.056	0.045	0.015	0.15	0.98	0.79	0.23	2.68
Σpenta-CB	0.036	0.007	0.001	0.22	0.58	0.11	0.017	3.46	0.010	0.010	0.010	0.010	0.18	0.18	0.15	0.20
Σhexa-CB	0.084	0.045	0.01	0.20	1.34	0.72	0.092	3.77	0.026	0.020	0.015	0.095	0.46	0.35	0.23	1.70
Σhepta-CB	0.057	0.025	0.01	0.12	1.04	0.41	0.075	2.20	<0.002		<0.002		<0.02			
Σocta-CB	0.030	0.027	0.01	0.04	0.48	0.43	0.18	0.82	<0.002		<0.002		<0.02			
Σnona-deca-CB	0.013	0.009	0.003	0.025	0.23	0.15	0.04	0.47	<0.002		<0.002		<0.02			
Σendosulfan	0.044	0.033	0.008	0.10	0.72	0.53	0.15	1.88	0.016	0.014	<0.002	0.036	0.29	0.26	0.16	0.72
ΣPBDEs	0.44	0.26	0.039	1.47	7.33	4.13	0.73	27.0	0.17	0.12	0.05	0.50	2.98	2.20	0.76	8.94
BDE-209	0.15	0.077	0.004	0.54	2.26	1.24	0.08	7.69	na[1]				na[1]			

Table 1 continued . . . .

Table 1 continued

Organohalogen group	Dehcho Arith Mean ng/g ww	Dehcho GM ng/g ww	Dehcho min ng/g ww	Dehcho max ng/g ww	Dehcho Arith Mean ng/g lw	Dehcho GM ng/g lw	Dehcho min ng/g lw	Dehcho max ng/g lw	South Slave Arith Mean ng/g ww	South Slave GM ng/g ww	South Slave min ng/g ww	South Slave max ng/g ww	South Slave Arith Mean ng/g lw	South Slave GM ng/g lw	South Slave min ng/g lw	South Slave max ng/g lw
Toxaphene	0.84	0.64	0.28	2.17	13.6	10.3	4.37	30.7	1.06	0.85	0.17	1.83	18.8	15.0	2.57	32.7
ΣPFCAs	0.75	0.70	0.42	1.38	[2]				0.63	0.62	0.39	0.83	[2]			
ΣPFSAs	0.57	0.52	0.33	1.13					0.30	0.30	0.19	0.37				
ΣPFASs	1.32	1.23	0.81	2.51					0.93	0.92	0.63	1.20				

[1]na = not analysed; [2]Results for PFASs are reported on a wet weight basis only.

Results in Tables 1, S6, and S7 are reported on wet weight and lipid weight basis (except for PFASs) to facilitate comparison with other studies. However, major (Σ) groups of organohalogen compounds and most individual analytes were not positively correlated with % lipid in moose liver (Table S9A). The exceptions were PFOS and hexachlorobutadiene (HCBD) which were significantly positively correlated; α-HCH, *cis*-chlordane, CB 105/127, and CB156 were significantly negatively correlated (Table S9B, S9C); therefore, we concluded that lipid adjustment was not appropriate for most analytes and all statistical analyses were conducted with wet weight data.

Perfluoroalkyl Substances

The major organohalogen contaminants in moose liver were the PFASs (Table 1). Concentrations of total (Σ) PFASs ranged from 0.81 – 2.5 ng/g (ww) in Dehcho and 0.63–1.2 ng/g ww in South Slave. ΣPFCAs (sum C4 to C16 perfluorocarboxylates) were 1.3 to 2-fold higher than ΣPFSAs (sum of C4-C10 perfluoroalkyl sulfonates) and averaged 0.75 ng/g ww in Dehcho and 0.63 ng/g ww in South Slave. Mean concentrations of ΣPFSAs were significantly higher in Dehcho moose due to higher PFBS and PFHxS (Tables S10A, S10B). No significant differences between Dehcho and South Slave were observed for ΣPFCAs; however, PFUnA was significantly higher in Dehcho than South Slave (Tables S10A, S10B). The relative prominence of the PFASs was anticipated based on results for caribou liver from the Canadian Arctic (Table 2) where they are also present at low concentrations (Ostertag et al. 2009, Müller et al. 2011).

While PFOS was significantly correlated with % lipid, other PFSAs and PFCAs were not (Table S9B). ΣPFCAs, ΣPFSAs, and major individual PFASs were also not correlated with age of the moose (Table S9A, S9B). The correlation with % lipid

Table 2. Comparison of mean (± standard deviation if available) organohalogen concentrations in other studies on moose, caribou, and mountain goat tissues.

Species	Tissue	Region	Unit[1]	Statistic[2]	ΣDDT	ΣCHL	ΣHCH	ΣCBz	ΣPCB	Σendo-sulfan	ΣPBDEs	Toxa-phene	PFOS	ΣPFCAs	Refer-ence
Moose	liver	Dehcho & South Slave	lw	Mn ± SD	0.22 ± 0.04	0.70 ± 0.62	3.0 ± 1.3	11.8 ± 7.8	8.7 ± 4.5	0.49 ± 0.47	5.0 ± 6.7	16 ± 11	0.34 ± 0.22	0.69 ± 0.24	This study
Moose	liver	Norway	lw	Mn ± SD	-	-	-	-	-	-	0.42 – 9.4	-	-	-	[1]
Moose	muscle	Finland	lw	Mn ± SD	-	-	-	-	5.4 ± 2.8	-	-	-	-	-	[2]
Moose	muscle	Finland	lw	Mn ± SD							1.24 ± 0.94				[3]
Moose	muscle (baked)	Mackenzie R. valley	ww	Mn	0.30	0.30	1.0	0.20	3.0	-	-	<0.1	-	-	[4]
Moose	liver (baked)	Mackenzie R. valley	ww	Mn	-	-	-	-	23	-	-	1.0	-	-	[4]
Moose	liver	Denali, Alaska	lw	Mn	<0.1–340	-	-	0.72	0.025	<0.1	-	-	-	-	[5]
Moose	muscle	Denali, Alaska	lw	Mn	<0.1–630	-	-	<0.1	0.48	<0.1	-	-	-	-	[5]
Moose	muscle	Sweden	lw	Mn	42	0	9	15	896	-	2	<1	-	-	[6]
Moose	muscle	Sweden	lw	Mn	1.1	<0.1	3	16	9[3]	<1	<0.1	-	<0.1	<1.3–3.3	[7]
caribou	liver	Bathurst herd	ww	Mn	-	-	-	-	-	-	-	-	2.2	9.8	[8]
caribou	fat	Bathurst herd	lw	Mn ± SD	1.9 ± 0.57	0.81 ± 0.17	9.7 ± 1.1	-	11 ± 2.0	-	-	-	-	-	[9]
caribou	liver	Beverly herd	lw	Mn	<1.0	22.0	31.7	36.6	12.2	-	-	-	-	-	[10]
caribou	liver	Bathurst herd	lw	Mn ± SD							77 ± 18				[11]
caribou	liver	Nunavut	ww	Mn	-	-	-	-	-	-	-	-	2.7	3.5	[12]
caribou	liver	West Greenland	ww	Mn ± SD	0.02 ± 0.01	0.88 ± 0.23	0.21 ± 0.04	-	0.96 ± 1.2	-	-	0.1	-	-	[13]
caribou	liver	West Greenland	lw	Mn	-	-	-	6.3	-	0.03	-	-	1.4	1.6	[14,15]

Table 2 continued . . . .

Table 2 continued

Species	Tissue	Region	Unit[1]	Statistic[2]	ΣDDT	ΣCHL	ΣHCH	ΣCBz	ΣPCB	Σendo-sulfan	ΣPBDEs	Toxa-phene	PFOS	ΣPFCAs	Refer-ence
Mt goat	liver	Mackenzie Mts.	lw	Mn ± SD	41 ± 31	<9.0	13 ± 13	44 ± 7.3	4.9 ± 2.3	<29	<1	54 ± 47	-	-	[16]

[1]lw = lipid weight; ww = wet weight. Results for PFCAs and PFOS are all wet weight. [2]Mn (Mean) ± SD = arithmetic mean ± standard deviation.[3]Sum of CB118, 138, 153 and 180.

References: [1] Mariussen et al. (2008); [2] Suutari et al. (2009); [3] Holma-Suutari et al. (2016); [4] Berti et al. (1998); [5] Landers et al. (2008); [6] Jansson et al. (1993); [7] Danielsson et al. (2008); [8] Müller et al. (2011) ; [9] Elkin and Bethke (1995); data also in Kelly and Gobas (2001); [10] Elkin Unpublished. Reported in the AMAP POPs assessment annex (De March et al. 1998); [11] Morris et al. (2016); [12] Ostertag et al.(2009); [13] Johansen et al. (2004); [14] Vorkamp et al. (2004); [15] Bossi et al. (2015); [16] Larter et al. (2015).

was unexpected because as an ionizable substance, PFOS is usually associated with proteins; positive correlations have been reported with plasma cholesterol and lipid in humans (Starling et al. 2014, Zeng et al. 2015), and with % lipid in fish fillets (Jm et al. 2015).

Müller et al. (2011) measured PFASs in willow collected from the Bathurst caribou summer range and PFAS concentrations in precipitation collected at Snare Rapids, NT about 140 km north of Yellowknife (Scott et al., unpublished data in Müller et al. 2011) (Fig. 2). Although these samples were collected about 300 and 600 km, respectively, northeast of the moose sampling sites in Dehcho and South Slave regions, they are the closest locations with PFAS sampling in northern Canada and are sites not impacted by local sources. The pattern of PFASs in willow and precipitation is relatively similar with higher proportions of PFHpA to PFNA, as well as PFOSA, than in moose liver. The pattern of PFCAs in moose liver is shifted to longer chain, more bioaccumulative PFASs (PFNA, PFDA, PFUnA) with PFOS more prominent due to its greater bioaccumulation than shorter chain PFSAs, PFHxS, and PFHpS. Also, PFOSA is a known precursor of PFOS in biota (Xu et al. 2004, Brandsma et al. 2011), and another source of PFOS. The pattern of individual PFASs (Fig. 2) in moose liver is dominated by 9 to 11 carbon perfluorocarboxylates, PFNA-PFUnA similar to what is found in caribou liver (Müller et al. 2011). However, one notable difference is that caribou have an "even-odd" carbon chain pattern with higher PFUnA than PFDA, a pattern not apparent in moose.

PFOS and ΣPFCAs in moose liver were lower than in livers of caribou sampled from the Bathurst herd in 2008 (Müller et al. 2011), from various communities in Nunavut in the late 1990s (Ostertag et al. 2009), and in reindeer livers from southern Greenland (Bossi et al. 2015). Danielsson et al. (2008) determined a suite of PFASs similar to ours

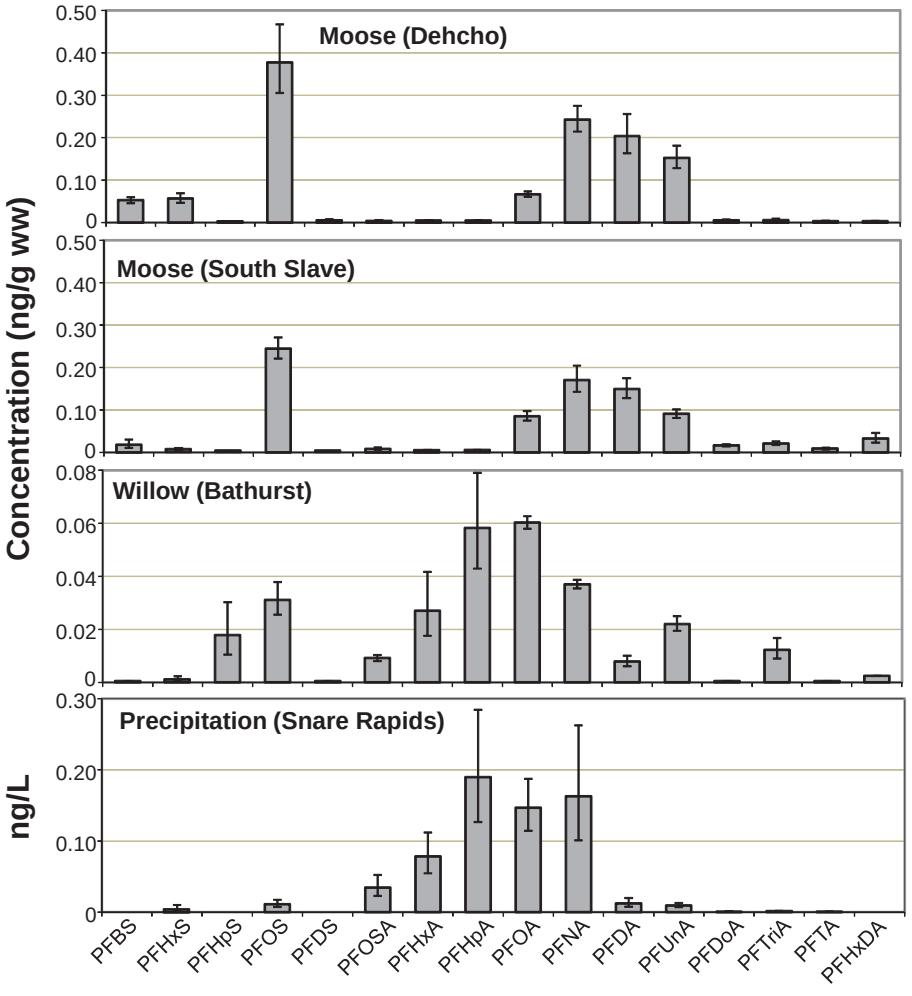

Fig. 2. Geometric mean concentrations of perfluorinated alkyl substances in moose liver (N = 7 at each location) compared with willow (N = 3) and precipitation (N = 4) collected elsewhere in the NWT, Canada. Bars represent standard errors of log transformed data. An explanation of the abbreviations of the individual PFASs is given in Table S2.

in moose muscle from Grimsö in south central Sweden; PFOSA and PFOS were generally <0.1 ng/g ww while individual PFCAs were <0.05 to <1.4 ng/g ww. To our knowledge, these are the only other published data available for PFASs in moose.

PCBs and OCOs

Chlorinated organics, PCBs, and OCP/OCOs represent the next most prominent group of contaminants in moose liver (Table 1). ΣPCBs were significantly higher in Dehcho averaging 0.65 ng/g ww (range = 0.34 – 1.08) compared to 0.39 ng/g ww (range = 0.15 – 0.64) in South Slave. Di-, tri-, and tetrachlorobiphenyls were the major homolog groups in moose liver and CB8/5, 15, 17, 18, 20/33/21, 31/28, and 52 were among the most prominent congeners in samples from both regions (Table S6). Penta-, hexa-, and heptachloro- congeners were detectable in samples from Dehcho, and generally at or near MDLs in South Slave (Table 2). Kelly and Gobas (2001) also noted

higher proportions of tri- (CB28/31) and tetrachloro congeners (CB 52) in caribou fat and liver, but the latter study did not include congeners lower than CB28 which precluded a full comparison. Suutari et al. (2009) reported PCBs in moose muscle from northern Finland (Table 2) and found ΣPCB concentrations (average = 5.4 ng/g lipid) that were based on 37 congeners. Using their congener list, we found Σ_{37}CB values for the Dehcho and South Slave moose liver averaged 5.1 ± 1.6 ng/g lw and 2.5 ± 2.1 ng/g lw, respectively. Suutari et al (2009) found that 6 indicator PCBs (CB 28/31, 52, 101,138, 153, 180) represented an average of 48% of the 37 congeners they analyzed in moose muscle, whereas in this study, 6 congeners averaged 37% (Dehcho) and 30% (South Slave) of Σ_{37}CB. Danielsson et al. (2008) found that CB28, CB52, and CB101 were all < MDL in moose from central Sweden with CB118, CB153, CB138, and CB180 (ΣPCBs ~ 9 ng/g lw) detected in all samples; Janssen et al. (1993) found much higher ΣPCBs (N = 13, 896 ng/g lw) in moose muscle collected in the same study area in the 1980s. Landers et al. (2008) found very low concentrations of PCBs in moose liver and muscle from Denali National Park in Alaska, reporting only CB153 (0.025 ng/g lw in liver and 0.48 ng/g lw in muscle).

The chlorobenzenes (ΣCBz consisting of 1,2,4,5- and 1,2,3,4-tetrachlorobenzene [TeCBz], pentachlorobenzene [PeCBz] and HCB) were the most prominent group among the OCOs (Table 1, Table S2). Average concentrations of ΣCBz in South Slave (average = 0.82, range = 0.49–1.5 ng/g ww) were significantly higher than in Dehcho samples (average = 0.51, range = 0.11–1.2 ng/g ww). This difference was mainly due to 1245-TeCBz, 1234-TeCBz, and PeCBz which were greater in South Slave than Dehcho samples, although the differences were not significant (P = 0.07). A related chlorinated aromatic, pentachloroanisole (PCA), was detectable in Dehcho (0.005–0.15 ng/g ww) and South Slave samples (0.004–0.070 ng/g ww). PCA is a degradation product of pentachlorophenol (PCP) and pentachloronitrobenzene (PCNB) which is a prominent OCP in arctic air (Su et al. 2008), but may also be present in moose liver as a result of degradation of HCB to pentachlorophenol, which can be biomethylated to PCA (UNEP 2013); however, PCNB was not detectable (<0.002 ng/g ww) in moose liver.

HCB is one of the most commonly reported OCOs in arctic herbivores. It was detected in moose muscle and liver from Denali National Park (Landers et al. 2008), in tissues of caribou and muskoxen (*Ovibos moschatus*) from West Greenland (Johansen et al. 2004), and in caribou in the Canadian Arctic (Elkin and Bethke 1995, Pollock et al. 2009). Concentrations of HCB plus PeCBz in moose muscle from central Sweden (~16 ng/g lw in 2005) were similar to those in this study (Table 2; Danielsson et al. 2008). HCB concentrations in moose from central Sweden declined significantly at a rate of 6.7% annually over the period 1986 to 2005 (Danielsson et al. 2008).

Hexachlorobutadiene (HCBD), a by-product of chlorinated solvent manufacturing (UNEP 2012), was detected in all samples at concentrations ranging from 0.007–0.014 ng/g ww in Dehcho and 0.003–0.011 ng/g ww in South Slave (Table S6); no significant differences between regions were found (Table S10B). HCBD concentrations were correlated with % lipid (Table S9B). Because HCBD is hydrophobic (log Kow = 4.8; UNEP 2012), a positive correlation with lipid would be expected; however, it is interesting that most other chlorinated organics were not correlated or negatively correlated (Table S9B). HCBD was below detection limits (<0.1 ng/g ww) in moose muscle from central Sweden (Danielsson et al. 2008). Other OCO byproducts in the analytical list

including octachlorostyrene and 3,4,5,6-tetrachloro-veratrole were not detected, although they have been detected consistently in arctic air sampled at Alert (NU) at low pg/m^3 concentrations (Su et al. 2008).

Organochlorine Pesticides

Toxaphene was the most prominent OCP in moose liver with concentrations ranging from 0.28–2.17 ng/g ww in Dehcho and 0.17–1.83 ng/g ww in South Slave (Table 1); mean concentrations were not significantly different between the regions (Table S10A). Toxaphene was mainly present as hepta-, octa-, and nonachlorobornanes (Table S6) when expressed as "total" toxaphene using the technical mixture (Glassmeyer et al. 1999); however, individual chlorobornane congeners were <MDL. Thus, while chlorobornanes appear to be prominent in moose liver, the proportions of individual congeners remain uncertain. Berti et al. (1998) also reported low levels of toxaphene in baked moose liver and muscle from the Mackenzie Valley region (Table 2), and Jansson et al. (1993) did not detect toxaphene (<~10 ng/g lw) in moose livers from central Sweden; neither study included congener analysis. Johansen et al. (2004) found low concentrations of total toxaphene (0.1 ng/g ww) in caribou liver and kidney, and non-detect levels in other grazing animals in Greenland using the same method as here, along with the toxaphene congeners P26, P50, and P62; however, as similar to this study, the congeners were <MDLs. Larter et al. (2015) found higher concentrations of toxaphene in liver of mountain goats (*Oreamnos americanus*) (n = 3, 54 ± 47 ng/g lw) from the Mackenzie Mountains west of the Dehcho region; however, P26, P50, and P62 were also <MDLs. In general, chlorobornanes with 2-endo, 3-exo, 5-endo, 6-exo chlorine substitution are thought to be most resistant to biotransformation (Vetter and Scherer 1999), although most empirical evidence is from marine mammals, with herbivores studied minimally.

ΣHCH was the next most prominent OCP averaging 0.145 ng/g ww (range = 0.091–0.218) in Dehcho and 0.198 ng/g ww (range = 0.12–0.385) in South Slave. No significant differences in mean concentrations between regions were found for ΣHCH or the 3 HCH isomers (Table S10A, S10B); α- and γ-HCH concentrations were significantly correlated with other semi-volatile organochlorines, 1,2,4,5- TeCBz, 1,2,3,4- TeCBz, PeCBz, and PCA whereas β-HCH was not (Table S9B). β-HCH was the major HCH isomer in moose liver averaging 59% and 39% in Dehcho and South Slave, respectively. The β-isomer is known to predominate in mammals apparently due to its more stable molecular configuration (Willett et al. 1998). The β-HCH/ΣHCH ratio in caribou liver from the Bathurst range (Kelly and Gobas 2001) ranged from 2–4%, lower than in moose liver with the α-isomer predominant, and Johansen et al. (2004) found the proportion in caribou averaged 11% in liver and 27% in fat.

ΣHCH concentrations in the combined Dehcho-South Slave data were similar to those reported for moose from central Sweden (Table 2) where Danielsson et al. (2008) found rapidly declining concentrations of α-HCH in moose muscle (22% year) during the period 1986–2000. From 2000 to 2005, α-HCH was <MDL in muscle samples from the same area while β-HCH remained relatively constant averaging 2.9 ng/g lw.

Total chlordane (ΣCHL) concentrations were very low in moose liver averaging 0.10 ng/g ww (range = 0.032–0.15) in Dehcho and 0.11 ng/g ww (range = 0.038–0.25) in South Slave. There were no significant differences in mean concentrations of ΣCHL or for individual chlordane related compounds (Tables S10A, S10B). Two major chlordane-related transformation products, heptachlor epoxide and oxychlordane, represented an average of 73 and 76% of ΣCHL in moose

liver in Dehcho and South Slave samples, respectively. Heptachlor epoxide was correlated with *cis*-chlordane, and several chlorobenzene related compounds (1,2,3,4-TeCBz, PeCBz, and PCA) had weak but non-significant (P = 0.05–06) relationships with 1,2,4,5-TeCBz and α-HCH; oxychlordane was not correlated with any other OCPs (Table S9B).

Danielsson et al. (2008) found chlordane isomers and trans-nonachlor were all <MDL in moose from central Sweden but did not measure oxychlordane. Similarly, Janssen et al. (1993) found non-detect concentrations of chlordanes and nonachlors (<2 ng/g lw) in the same moose population. Kelly and Gobas (2001) reported oxychlordane as the only detectable chlordane compound in caribou liver, and the mean concentration of ΣCHL in caribou liver from West Greenland was 0.88 ± 0.23 ng/g ww; oxychlordane was <0.01 ng/g (Johansen et al. 2004).

ΣDDT was essentially non-detectable in moose liver with concentrations <0.02 ng/g ww in South Slave and ranging from 0.013–0.017 ng/g ww in the Dehcho (Table 1). Although the only other data from northern Canada are for baked moose muscle (Berti et al. 1998), it is noteworthy that ΣDDT was present at similar concentrations as ΣCHL (Table 2). ΣDDT concentrations in moose from Denali NP in Alaska had wide variation due to relatively high concentrations in 1 of 3 animals (*p,p*'-DDD in liver = 340 ng/g lw; *o,p*'-DDT in muscle = 630 ng/g lw) (Landers et al. 2008). In their time trend study, Danielsson et al. (2008) found *p,p*'-DDT and *p,p*'-DDD were consistently < MDL while concentrations of *p,p*'-DDE were detected in all years and declined significantly (3.5% annually) from 1986 to 2005. ΣDDT was readily detectable in caribou in the Canadian Arctic where Elkin and Bethke (1995) reported a mean level of 1.9 ± 0.6 ng/g lw in the Bathurst herd sampled in summer in the early 1990s. Johansen et al. (2004) found very low concentrations of ΣDDT in caribou liver (0.02 ± 0.01 ng/g ww) from West Greenland and non-detect levels of *p,p*'-DDE. DDT related compounds were present at higher concentration in mountain goat liver from the Mackenzie Mountains (41 ± 31 ng/g lw; Larter et al. 2015) than in caribou or moose liver (Table 2).

Endosulfan isomers (α, β) and the degradation product endosulfan sulfate were detected at low concentrations in all samples; Σendosulfan ranged from 0.008–0.10 ng/g ww in Dehcho and <0.002–0.036 ng/g in South Slave. Mean concentrations of Σendosulfan were significantly greater in Dehcho samples due to higher endosulfan sulfate which represented 50% of Σendosulfan in Dehcho samples (Tables S10A, S10B). Landers et al. (2008) analysed endosulfan in moose from Denali NP but found non-detect concentrations, and Danielsson et al. (2008) found non-detect Σendosulfan in moose from Sweden. Vorkamp et al. (2004) reported low ng/g concentrations of α- and β-endosulfan in caribou liver (0.03 ng/g lw) and tissues of other herbivorous mammals from West Greenland. Σendosulfan was <MDL in mountain goat livers, although the detection limit was relatively high (29 ng/g lw) compared to other studies (Larter et al. 2015).

Mirex was detected in 50% of the moose liver samples with concentrations similar between regions ranging from <0.002 to 0.014 ng/g ww in Dehcho and from <0.002 to 0.013 ng/g ww in South Slave; concentrations were not correlated with other OCPs/OCOs. In general, mirex has rarely been measured in arctic herbivores. Pollock et al. (2009) found mirex was <MDLs in peri-renal fat of caribou from the George River herd in Labrador, and it was not identified in other studies of caribou in the NT or Nunavut. Mirex was detected in West Greenland caribou (mean concentration ranged from <0.1 ng/g lw in kidney to 1.2 ng/g lw in liver) and in other terrestrial herbivores including

muskox, sheep (*Ovis sp.*), and hares (*Lepus arcticus*) (Vorkamp et al. 2004).

Brominated Flame Retardants

Σ_{13}PBDEs in moose liver (sum of 13 tri-bromo to heptabromo- congeners; Table S6) ranged from 0.04–1.5 ng/g ww in Dehcho and 0.05–0.50 ng/g ww in South Slave (Table 1); mean concentrations were not significantly different between regions (Table S10A). Σ_{13}PBDEs were significantly correlated with ΣCHL but not with other major PCB organohalogen groups (Table S9A). BDE 47 and 99 were the most prominent PBDE congeners, together averaging 61% of Σ_{13}PBDE in Dehcho and 68% in South Slave (Table S7). BDE 47 was correlated with semi-volatile OCP/OCOs including TeCBz, PeCBz, PCA, and heptachlor epoxide (Table S9B).

Decabromodiphenyl ether (BDE-209) was present at similar concentrations as BDE 47 and 99 in Dehcho moose livers. No BDE-209 data were available for South Slave because the silica column cleanup of the sample differed from the Dehcho samples. BDE-209 eluted with the PCB containing fraction, while all other BDEs were in a more polar elution used for OCPs. The PCB fraction was not archived for BDE analysis.

Morris et al. (2016) found BDE-209 the predominant BDE in moss, lichen, willow, and grasses from the Bathurst caribou summer range in western Nunavut, as well as in caribou muscle (44 ± 23 ng/g lw) and liver (39 ± 12 ng/g lw). BDE-209 was also the major congener in moose muscle from Finland with concentrations up to 177 ng/g lw and wide variability among animals (Holma-Suutari et al. 2016). BDE-209 was also detected at concentrations ranging from < MDL to 21 ng/g lw and the predominant congener in moss from the same regions.

Concentrations of Σ_{13}PBDE in the Dehcho/South Slave moose were similar to those found in moose from Norway and Finland, excluding BDE-209. Mariussen et al. (2008) found that Σ_7PBDE levels in moose from northern Norway were low, ranging from <MDL to 9.4 ng/g lw, and the mean concentration of Σ_{15}PBDE was 1.24 ± 0.94 ng/g lw in moose muscle from Finland (Holma-Suutari et al. 2016). However, Danielsson et al. (2008) found PBDEs (BDE 47,85,99,100,209) were <MDL (0.01 ng/g except for BDE-209 [<0.1 ng/g lw]) in moose muscle from central Sweden.

Moose liver samples were also analysed for 16 alternative brominated flame retardants as well as syn- and anti-dechlorane (Table S7). Only allyl 2,4,6-tribromophenyl ether (TBP-AE) was detected (<0.002 – 0.20 ng/g ww) in 4 of 7 samples from South Slave. TBP-AE was the major alternative BFR identified in vegetation samples from the Bathurst region of Western Nunavut (Morris et al. 2016). While it is an alternative BFR, TBP-AE can also be formed by both photolytic and anaerobic debromination of another tribromophenoxy- BFR, TBP-DBPE (Ma et al. 2012).

The concentrations of most halogenated organics measured in this study are much lower than those measured in caribou or mountain goats from the Canadian Arctic/subarctic. During winter moose consume birch (*Betula spp.*), willow (*Salix spp.*), and aspen (*Populus spp.*) twigs, switching to horsetail (*Equisetum spp.*), pond weeds, and grasses in the spring, and to leaves of birch, willow, and aspen and forbs in summer (Arnold et al. 2006). In contrast, caribou and reindeer use lichen as an important food resource (Aagnes et al. 1995, Bergerud et al. 2007), and Müller et al. (2011) found *ca.* 3-fold higher levels of PFCAs in lichen than in willow. Similarly, Morris et al. (2016) found 1.5 to 2-fold higher ΣPBDEs in lichen than in willow or grasses, suggesting that

dietary lichen may explain the higher levels of halogenated organics in caribou than moose.

Dietary differences among animals at the same trophic level add complexity to assessments of biomagnification of contaminants in terrestrial food webs (van den Brink et al. 2016). Mariussen et al. (2008) found an order of magnitude difference in PBDE concentrations between moose and lynx (*Lynx canadensis*). There is also an order of magnitude higher concentration of ΣPBDEs in caribou liver than moose liver (Table 2; Morris et al. 2016). Suutari et al. (2009) found that lipid-based concentrations of PCBs were lower in liver from moose calves than reindeer calves, whereas concentrations in adult moose were similar to those in adult reindeer from the same region. Vorkamp et al. (2004) found 20-fold higher concentrations of HCB and ΣCBz in muskoxen liver and muscle compared to caribou and sheep from the same region, although not for endosulfan, dieldrin, or mirex. Relative biotransformation capacity likely explains these species-specific differences.

CONCLUSIONS

We hypothesized that concentrations of POPs in moose would not be significantly different between the Dehcho and the South Slave regions because they are adjacent, have low extent of urbanization, and the sampling locations were generally remote from towns/villages (Fig. 1). However, significant differences were found as samples from Dehcho had higher concentrations of endosulfan sulfate, HCBD, ΣPFSAs, PFHxS, PFUNA, and ΣPCB; South Slave had higher γ-HCH, octachlorobornanes, and Σmono-di- PCBs (Tables S10A, S10B). The reasons for these differences are not entirely clear. For example, biological factors such as age, % lipid, and male:female ratios that often explain variation in concentrations of POPs in mammals were all similar between regions. The higher concentrations of PCBs, HCBD, and PFSAs could possibly be related to animals foraging consistently near towns and associated infrastructure such as airports, electricity generation (diesel generators), and vehicle repair/maintenance sites that are often associated with past use of PCBs, PFOS, and chlorinated solvents. Although present in most towns, these sites are highly localized and moose rarely forage close by; therefore, we expect negligible impact of such on our samples or concentrations we report. Falk et al. (2012) reported higher concentrations of PFASs in roe deer (*Capreolus capreolus*) from natural terrestrial ecosystems than agrarian or near urban sites in Germany, and concluded that bioaccumulation via the terrestrial food chain, combined with atmospheric deposition, was the most likely exposure pathway.

The time of sample collection may have a greater impact on regional differences in POPs than any point source of contamination. All Dehcho samples were collected in late February through late March, whereas South Slave samples were collected from September to February (Table S1). Moose harvested in February and March would have consumed a winter browse diet (birch, willow, and aspen twigs) longer than those harvested in other months. Although we lack samples of dietary items from the Dehcho or South Slave, willow collected from the Bathurst caribou range had higher PFASs than sedge or grass (Müller et al. 2011); however, in this same area ΣPBDEs in willow and grasses were of similar concentration (Morris et al. 2016). Concentrations of other contaminants such as PCBs and HCBD in vegetation from this region are unknown precluding any dietary comparisons. Kelly and Gobas (2003) predicted a greater accumulation of PCBs in caribou during spring due to higher levels in lichen from melting snow and gas exchange, based on changes in lipid content, body size, and/or milk excretion for females.

Further study is needed to assess the impact of seasonal/local differences in diet and exposure on regional differences in POP concentrations.

Our study provides baseline information for a wide range of halogenated organic contaminants in moose collected by local hunters. Although our sample size was small compared to previous studies on heavy metals in moose (Larter and Kandola 2010), it nevertheless provides risk assessors and resident harvesters with exposure information concerning an important subsistence food resource. Concentrations of cadmium found in the organs of moose from this region resulted in public health advisories to limit the consumption of kidneys and livers (http://www.hss.gov.nt.ca/sites/www.hss.gov.nt.ca/files/resources/moose-organ-consumption-notice.pdf). However, levels of other heavy metals are of minimal public health concern, and the minimal concentrations of POPs reported here provide no reason to discourage the consumption of moose as a healthy food choice.

ACKNOWLEDGEMENTS

We are indebted to all of the traditional harvesters who supplied samples for this study *Mahsi cho*. Matson's Laboratory (www.matsonslab.com, Manhattan, Montana, USA) aged moose teeth. We thank W. Davis and R. McLeod at ALS Global Laboratories (Burlington, Ontario, Canada) for their analytical support and E. Barresi and staff at NLET (Environment Canada, Burlington, Ontario, Canada) for analysis of BFRs, toxaphene, and endosulfan. We thank M. Williamson (Environment Canada, Burlington, Ontario, Canada) for PFAS analyses and A. De Silva for a helpful review of the manuscript. Funding for this program came from the NWT Western Biophysical Program (GNWT), the Department of Environment & Natural Resources, and the Northern Contaminants Program.

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