DIETARY MARKERS AND CONTAMINANT EXPOSURES ARE CORRELATED TO WILD FOOD CONSUMPTION IN TWO NORTHERN ONTARIO FIRST NATIONS COMMUNITIES

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Abstract/Résumé

First Nations peoples experience many benefits from eating locally-harvested wild foods, but these benefits must be considered along with the potential risks associated with exposure to environmental contaminants. Unlike store-bought foods, wild foods are an important traditional resource and a significant source of dietary protein, essential minerals and polyunsaturated fatty acids, believed to help in the prevention and treatment of obesity and obesity-related diseases such as type-2 diabetes mellitus. Wild foods continue to be an important and healthy food choice for First Nations peoples; however, they are also a primary source of dietary mercury, polychlorinated biphenyls (PCBs) and other persistent organic pollutants (POPs). To assess the effects of wild food consumption on dietary markers and contaminant accumulation, we grouped individuals from two remote Oji-Cree First Nations communities of north-western Ontario (n=71) according to their level of wild food consumption. In this study, I observed significantly higher organic contaminants in blood and higher mercury concentrations in hair for individuals consuming greater amounts of wild food. Age-adjusted contaminant concentrations were on average 3.5-times higher among high-frequency wild food consumers, with many exceeding federal and international health guidelines for mercury and PCB exposures. Contaminants in these populations approach, and in some cases exceed, threshold levels for adverse effects with potential consequences especially for prenatal development. Here, I also investigated the potential for stable isotope ratios of carbon and nitrogen ($\delta^{13}C$ and $\delta^{15}N$) to serve as dietary markers and found strong positive correlations between stable isotopes and frequency of wild food and fish consumption. Frequency of fish consumption and $\delta^{15}N$ was also shown to be positively correlated with mercury concentrations in hair and PCB concentrations in plasma.

The results of this thesis demonstrate that known differences in dietary behaviour are clearly reflected in stable isotope ratios and contaminant concentrations. The data also show that contaminant exposures to those consuming wild foods in remote Boreal ecosystems is comparable to those associated with serious health effects in industrialized areas, and the problem of contaminants in wild foods is more widespread than the available literature would have led us to believe. These results affect our appreciation of contaminant exposures to First Nations peoples and will have implications for dietary choices, particularly if individuals are encouraged to consume greater amounts of wild foods for their proposed health benefits. We recommend further attention be given to the risks of contaminants in locally-harvested wild foods when promoting the benefits of their consumption to First Nations people as the problem of contaminants in remote communities practicing traditional lifestyles is often underreported and underplayed.

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1.0 Introduction

Traditional wild foods are an inseparable part of First Nations cultural and spiritual identity and are often cited as having important nutritional benefits that contribute to the reduction of obesity and obesity-related diseases such as type-2 diabetes mellitus (T2DM). With increasing global and local concerns over mercury and persistent organic pollutants (POPs) and their migration into northern environments and local food webs, there is a need to assess the potential health risks associated with the consumption of locally-harvested foods when promoting their consumption for proposed health benefits.

POPs and other contaminants are characterized by their ability to be transported into northern and Arctic environments via atmospheric, oceanic (Wania and Mackay, 1996), and biological transport pathways (Blais *et al.*, 2007). Due to their high degree of persistence, relative insolubility in water and lipophilicity, they are known to bioaccumulate and bioconcentrate in the muscle and fatty tissues of organisms and biomagnify through northern and Arctic food webs. Bioaccumulation is the uptake and retention of chemical contaminants, as a function of fugacity, by any pathway such as diet, water or air, whereas bioconcentration is the uptake and retention of contaminants via water only. Biomagnification is the uptake and retention of contaminants through trophic levels, as a function of both fugacities and trophic levels. Biomagnification results in elevated contaminant concentrations in the body tissues of predatory fish, birds, marine mammals and, ultimately, humans. This poses a potential threat to northern First Nations peoples that benefit from consuming locally-harvested wild foods. Many researchers have reported a transition from locally-harvested wild foods to highly processed store-bought foods that are high in starch, fat, and sugar as Northerners became more fearful of

the risks (Dewailly *et al.*, 2002; Kuhnlein, 1992; Kuhnlein *et al.*, 2001) (Gittelsohn *et al.*, 1998; Kuhnlein, 1995; Kuhnlein *et al.*, 2004; Receveur *et al.*, 1997; Sharma *et al.*, 2007). Researchers are also associating this so-called "nutrition transition" with changes in food availability, deteriorating health, increases in non-communicable disease, decreasing physical activity, increasingly sedentary lifestyles, and loss of culture and cultural morale within First Nations communities (Gittelsohn *et al.*, 1998; Harris *et al.*, 1997; Kuhnlein *et al.*, 2001). Lower quality processed foods are also more appealing to Northerners than higher quality fresh meats and produce due to their lower costs, especially to those who rely on social assistance (Kuhnlein *et al.*, 2001). Regardless, traditional lifestyle activities such as hunting, fishing and trapping continue to be very important in many northern First Nations communities.

Unlike store-bought foods, wild foods such as fish and hunted meats are an important traditional resource and a significant source of dietary protein, essential minerals and polyunsaturated fatty acids (Das, 2000; Dewailly and Blanchet, 2003; Kuhnlein *et al.*, 2002). These are believed to help in the prevention and treatment of obesity and obesity-related diseases such as T2DM which continue to increase at alarming rates (Garriguet, 2008; Harris *et al.*, 1997; Young *et al.*, 2000). Beneficial effects of fatty acid intake are also correlated with lower risk of cardiovascular disease (Dewailly *et al.*, 2001, 2002), levels of HDL ("good") cholesterol and triacylglycerols (Dewailly *et al.*, 2003), and infant development (Jacobson *et al.*, 2008). Fish, in particular, continue to be promoted as an important and healthy food choice for First Nations people (Dewailly *et al.*, 2002) however they are also a primary source of dietary mercury and other environmental contaminants associated with important health risks.

Northern Contaminants

POPs are defined as chemicals that persist in the environment, bioaccumulate in food webs and are toxic to life. Certain physical and chemical properties contribute to a chemical's persistence and ability to bioaccumulate, such as water solubility and ability to partition between environmental compartments and fate processes (e.g., octanol-water partition coefficient), etc. The octanol-water partition coefficient (K_{ow}) is the concentration of a chemical in octanol (surrogate for a fat or lipid) over the concentration of a chemical in water at equilibrium. K_{ow} is expressed as log values because of the large range of K_{ow}s and a high log K_{ow} indicates a high affinity for lipid-rich tissues in biota. Bioaccumulation generally occurs for chemicals with log K_{ow} values of 3 to 6 or greater. Where log K_{ow} values are greater than 6, bioaccumulation is reduced because of larger molecular sizes, which is an impediment to the movement of the chemicals across cell membranes, and insolubility in water. Ionic chemicals have log K_{ow}s less than 1 (i.e., do not partition into octanol) and therefore do not bioaccumulate in lipid-rich tissues.

The UN Stockholm Convention on POPs has banned or severely restricted the production and use of certain POPs, yet they continue to be found in northern and Arctic environments at levels that could, in some cases, place Northerners at risk. These include twelve POPs which were originally known as the "Dirty Dozen", along with five additional POPs that have since been added to the list of banned or severely restricted POPs (Table 1.1). As a result of their discontinued production and use in many industrialized countries, decreasing trends have been observed for some POPs such as PCBs and DDT. Nevertheless, they continue to be contaminants of major concern in northern and arctic environments (INAC, 2003a, 2003b, 2009; AMAP, 2009) with many Northerners exceeding guideline levels for mercury, PCBs and other organic contaminants. Recent results of long-term biomonitoring studies in Canada's northern

First Nations communities show 24% and 52% of Inuit mothers from Nunavut and Nunavik, respectively, exceeding Health Canada's 'Level of Concern' of 5 μg/L for PCBs in blood (AMAP, 2009). These same studies also show 32% and 31% of Inuit mothers from Nunavut and Nunavik, respectively, exceeding the US EPA's guideline level of 5.8 μg/L for mercury in blood (AMAP, 2009). Many POPs have a tendency to migrate into colder northern and Arctic regions from southern latitudes, where industrial emissions are greatest. This phenomenon is often referred to as the grasshopper effect or global distillation (Wania and Mackay, 1996). Exposure occurs via the environment or diet and can lead to a wide range of health effects for both wildlife and humans. Consequences are greatest for those that rely on traditional wild food sources due to increased exposures to potentially contaminated fish and wildlife.

Table 1.1: List of banned or severely restricted POPs

POP	Date of Introduction	Definition/Use
Aldrin	1949	Insecticide
Chlordane	1945	Insecticide
DDT/DDE	1942	Insecticide
Dieldrin	1948	Insecticide
Endrin	1951	Rodenticide/Insecticide
Heptachlor	1948	Insecticide
Hexachlorobenzene (HCB)	1945	Fungicide
Mirex	1959	Insecticide
Toxaphene	1948	Insecticide
PCBs	1929	Commercial
Dioxins	1920s	Commercial Byproduct
Furans	1920s	Commercial Byproduct
Hexachlorocyclohexane (HCH), including α-HCH, β-HCH and γ-HCH (Lindane)	1940s	Commercial Byproducts and Insecticide (Lindane)
Chlordecone	1970s	Insecticide
Polybrominated diphenyl ethers (PBDEs)	1970s	Flame Retardant
Pentachlorobenzene	1920s	Commercial
Perflorooctanesulfonic acid (PFOS), its salt and perflorooctanesulfonyl fluoride (PFOSF)	1949	Commercial

(Source: Krümmel, 2006; ATSDR, 2011)

Mercury

Mercury is one of the most toxic elements in the environment and it is found in air, soil, and water. Natural inputs of mercury into the environment include volcanoes, weathering of natural mercury deposits, volatilization from the ocean, and melting of permafrost. Major anthropogenic sources of mercury are coal burning and waste incineration. In northern environments, important sources of mercury pollution are hydroelectric dams, mining, coal-burning, and the melting and flooding of permafrost due to climate warming.

Aside from inhaling particulates of inorganic mercury (Hg^{2+}) or vaporized elemental mercury (Hg^{0}) emitted from amalgam tooth fillings, humans are generally exposed to organic mercury as methylmercury (MeHg) through their diet due to the fact that traces of mercury are found in all foods (Dabeka et al. 2003), particularly edible fish tissues (Clarkson *et al.*, 2007; Health Canada, 2004). Mercury concentrations are quite low in fruits and vegetables as mercury uptake by plants from soil is low, whereas mercury concentrations in fish can be quite high and potentially toxic (Health Canada, 2004; INAC, 2003a; Mergler *et al.*, 2007). Fish can accumulate mercury from water (i.e., bioconcentration) through the gills and the organisms they eat (i.e., bioaccumulation). This is particularly the case for predatory fish species that are higher in aquatic food chains. Similarly, humans tend to accumulate mercury over time and with age despite having the ability to metabolize and excrete mercury (Clarkson *et al.*, 2007; Counter and Buchanan, 2004). Fish continue to be an important and healthy food choice because it is low in saturated fat and an excellent source of high-quality protein and omega-3 fatty acids (Health Canada, 2004) however they are also a primary source of dietary mercury (Chapman and Chan, 2000).

Mercury is a potent neurotoxin that can cross the blood-brain barrier, harming the brain and spinal cord, particularly in the developing nervous system of a fetus or young child (Grandjean *et al.*, 1998; Igata, 1993; Mergler *et al.*, 2007; Shipp *et al.*, 2000). High levels of mercury can be harmful and toxic to developing fetuses (Igata, 1993); even from exposure to low levels (Mergler *et al.*, 2007), with the greatest risk being brain damage. The human body can metabolize mercury and it is possible to reduce body burdens by reducing the amount of mercury in ones diet.

In lakes and oceans, MeHg bioaccumulates in fish and biomagnifies through aquatic food webs leading ultimately up to humans that consume fish, shellfish, fish-eating birds and marine mammals. More than 95% of the mercury measured in fish is methylmercury (Bloom, 1992) and it is easily absorbed in body tissues and generally remains in the bodies of organisms longer than inorganic forms of mercury. Toxic effects will depend on the degree of exposure and can range from a slight impairment to reproductive failure. Methylmercury in fish varies between and within species and the relationship between trophic position and mercury concentrations is well known (Mergler *et al.*, 2007). Piscivorous fish (predatory fish-eating fish) that are feeding at the top of aquatic food chains generally have higher mercury than non-carnivorous fish (Mergler *et al.*, 2007). Mercury concentrations were observed to be higher among those who eat primarily piscivorous fish when compared to those who eat primarily non-piscivorous fish (Chan and Receveur, 2000; Muckle *et al.*, 2001). The frequency of fish consumption will also affect mercury concentrations and contribute to this observed variability (Mergler *et al.*, 2007). Sex and age are also important factors contributing to this variability (Clarkson, 1997). It is therefore prudent and take a best-balanced approach by considering the type, age/size, and amount of fish that one consumes in order to limit ones exposure to dietary mercury, particularly for populations relying on fish as a primarily source of nutrition (i.e. First Nations).

One of the worst cases of mercury poisoning in First Nations occurred in Grassy Narrows, an Ojibwa community in Northern Ontario, where it was discovered in the 1970s that an upstream pulp mill was the source of mercury contamination for fish caught and eaten by the Ojibwa people downstream on the Wabigoon-English River system. First Nations people living in Grassy Narrows first started experiencing neurological symptoms of mercury poisoning in the late 1960s. In the 1970s, researchers measured mercury concentrations in hair from individuals

in Grassy Narrows and in the nearby First Nations community of White Dog that were, in some cases, above 100 ppb (Harada *et al*., 2005).

Due to the bioaccumulation of MeHg in fish and its adverse effects on human health, many government departments such as the United States Environmental Protection Agency (U.S. EPA) and the National Academy of Sciences (NAS, 2000), the World Health Organization (WHO, 1990) and Health Canada (2004) have recommended guidelines for fish consumption. Individuals that are most at risk are pregnant women and children who eat certain types of fish, and people who eat unusually large quantities of fish such as Inuit and northern First Nations peoples.

The U.S. EPA recommends keeping mercury concentrations in hair below 1,000 ppb (or ng/g) (NAS, 2000; Hightower and Moore, 2003) whereas the WHO provides a tolerance limit of 10 μg/g. (WHO, 1990). Health Canada's current guidelines for mercury in hair indicate a 'Normal Acceptable Range' where concentrations are below 6 ppm. An 'Increasing Risk' has been indicated where concentrations are within a range of 6 to 30 ppm and 'At Risk' are individuals with concentrations greater than 30 ppm in their hair (Health Canada, 2004). Legrand et al. (2010) have recently proposed a Health Canada blood guidance value of 8 μg/L for MeHg in blood or 32 μg/g for MeHg in hair when using the internationally accepted standard of hair to blood ratio of 250.

Polychlorinated Biphenyls (PCBs)

PCBs were first manufactured in 1929 and historically added to mineral and silicone oils and used as coolants and insulating fluids to reduce flammability in mechanical and electrical equipment (capacitors, transformers, hydraulic fluids, etc.). There are 209 PCB congeners and approximately 90 of these are detected in environmental samples. By 1977, concern over the

impact of PCBs on the environment led to a North American ban on manufacturing and importing PCBs. PCBs are now being phased out and governments internationally have set strict regulations for the handling, storage, and disposal of PCBs. Potential sources of environmental exposure to PCBs are from previous releases into the environment either from landfills, environmental 'sinks' such as lake sediments, or from global circulation and atmospheric deposition. Accidental releases or improper controls during storage or destruction can also lead to environmental and human exposure. However, as with mercury, human exposure to PCBs is often through the consumption of fish and other fish-eating aquatic birds and mammals. PCBs accumulate in fatty tissues and metabolize at an extremely slow rate, with tissue concentrations increasing with age. Skinning and trimming the fat off of fish can help reduce ones intake of PCBs and other POPs such as organochlorine pesticides (OCPs) and dioxins. Avoiding fish and other animal organs such as the liver and kidneys where higher concentrations of PCBs are often measured, when compared to leaner muscle tissues, is another way of reducing ones exposure to PCBs.

Organochlorine Pesticides (OCPs)

Second-generation OCPs have been widely used since the 1940-50s to the present because of their low cost and high effectiveness. Many OCPs have since been measured in the blood of Northerners from Northwest Territories and Nunavut (Walker *et al.*, 2003) and Inuit from Nunavik (Muckle *et al.*, 2001). Pesticides that are found in northern and Arctic regions are carried from industrialized regions via long-range atmospheric transport, waterways, and oceanic currents (Muckle *et al.*, 2001). Examples of some of the OCPs most commonly measured in the North include: *p,p'*-DDT (*p,p'*-dichlorodiphenyl trichloroethane), *p,p'*-DDE (*p,p'*-dichlorodiphenyl dichloroethylene), Lindane or β-HCH (β -hexachlorocyclohexane), chlordane

(α-chlordane, γ-chlordane) and the components/metabolites of technical chlordane: oxychlordane, *cis*-nonachlore, *trans*-nonachlore, aldrin, hexachlorobenzene (HCB), mirex and toxaphenes parlar 26 and parlar 50.

High levels of PCBs and OCPs can be harmful and toxic to developing fetuses. Children born to exposed mothers might have lower body weight and height, hyperpigmentation of skin, hypertrophy of gums, deformities of nails, increased frequency of bronchitis, lower body control and muscle coordination, lower intelligence, higher frequency of behavioral problems, and higher activity levels (Jacobson and Jacobson, 1993, 1996; Korrick and Sagiv, 2008). PCB and OCP residues in blood have been used extensively as an important marker of environmental and dietary exposures. One limitation of measuring chemical residues in blood is that they represent only the most recent exposures and cannot be used to measure previous exposures unless collected over the long term. Health Canada's maternal blood guidelines for PCBs (as Aroclor 1260, a PCB mixture manufactured by Monsanto) indicate a 'Level of Concern' where concentrations are greater than 5 μg/L (or >5 ppb) for women of child-bearing age and greater than 20 μg/L (or >20 ppb) for men and post-menopausal women. An 'Action Level' has been indicated where concentrations are greater than or equal to 100 μg/L (or ≥100 ppb) (Health Canada, 1986; Van Oostdam *et al.*, 2005).

Brominated Flame Retardants (BFRs)

Brominated flame retardants (BFRs) such as polybrominated diphenyl ethers (PBDEs) and polybrominated biphenyls (PBBs) have become ubiquitous environmental contaminants that are measured in indoor and outdoor air, remote Arctic regions, house and office dust, window films, rivers, lakes, sediments, sewage sludge and effluent, foods, and biota (terrestrial and marine mammals, fish, birds, and humans) (Alaee, 2003). There are three commercial mixtures

sold as penta-BDE, octa-BDE, and deca-BDE containing 4-5, 7-8, and 10 bromines, respectively (Alaee *et al.*, 2003). Penta-BDEs are added to mattresses and foam cushioning in upholstery; octa-BDEs are used in business equipment, automobile trim, telephones, and kitchen appliance castings; and deca-BDEs are used in electronic enclosures, such as wire insulation, televisions, and computers. Deca-BDEs are also used as a fabric treatment and coating on carpets and draperies. PBDEs are highly lipophilic and accumulate in the environment as well as in humans, with $\log K_{ow}$s in the range of 5.9-6.2 for tetra-BDEs, 6.5-7.0 for penta-BDEs, 8.4-8.9 for octa-BDEs and 10 for deca-BDEs (Watanabe and Tatsukawa, 1990). Structurally similar to PCBs, and also having 209 congeners following the same nomenclature, PBDEs appear to behave similarly in the environment and are associated with similar health effects in wildlife and humans. The European Union has banned the use of penta-BDEs and octa-BDEs, while the United States has voluntarily stopped using these two mixtures; however deca-BDEs are still in use (Wahl *et al.* 2008). Industries are moving toward the use of other brominated flame retardants (BFRs) such as hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A).

Many PBDE congeners are persistent, bioaccumulative, and capable of long-range atmospheric transport, with less brominated congeners being measured in Arctic regions and higher trophic level-feeding marine biota (Hale *et al.*, 2003; Ikonomou and Addison, 2008). Deca-BDEs (i.e., PBDE-209) tend to be restricted to points of release likely due to their low volatility and water solubility. PBDEs are also structurally similar to the thyroid hormone and, accordingly, are known endocrine disruptors (thyroid and estrogenic effects) contributing to developmental effects (brain and reproductive organs) and possibly cancer (Wahl *et al.*, 2008). They have been detected world-wide in human blood, breast milk, and adipose tissue with higher

levels observed in the United States as compared to Europe and Japan (Alaee, 2003) and higher levels observed in children as compared to adults (Fischer *et al.*, 2006). Due to the lipophilicity of PBDEs and their pervasive presence in consumer products and house dust, suspected routes of exposure for humans include both diet and the indoor environment. Four commonly reported PBDEs congeners are 2,2',4,4'-tetraBDE (BDE-47), 2,2',4,4',5-pentabromodiphenyl ether (BDE-99), 2,2',4,4',6-pentaBDE-100, and 2,2',4,4',5,5'-hexaBDE (BDE-153). Compositionally similar to PBDE-153, another commonly reported BFR is 2,2',4,4',5,5'-hexaBB-153 (PBB-153). PBDEs have been detected in the breast milk from every Canadian province and recent evidence strongly suggests that levels of PBDEs in the Canadian environments are increasing (Alaee *et al.*, 2003).

Northern Ontario First Nations South of 60th Parallel

Fish and other wild foods represent an important part of the diet of northern Ontario First Nations peoples and exposures to POPs through diet are of particular interest not only for governments and community health authorities, but also those who rely on locally-harvested foods. Compared to southern Ontario, there are many high fish-eating populations in north-western Ontario. With fish being available year round, a northern Ontario First Nations' diet consists largely of fish caught from local lakes and rivers and inland fish have been shown to be often more contaminated than sea-run fish (Lockhart *et al.*, 2005; Riget *et al.*, 2004), putting these populations at an elevated risk of exposure to mercury and other POPs. The risks and effects of eating fish and other marine mammals have been well established in northern Aboriginal and Inuit communities (Yukon, Northwest Territories, Nunavut); however few studies have focussed on First Nations south of the 60th parallel in north-western Ontario and this warrants further attention in order to better understand the benefits and risks of eating locally

harvested wild foods in remote northern Ontario First Nations communities. Aside from atmospheric deposition, many First Nations communities in northern Ontario are closer to various point sources of pollution including coal plants, hazardous waste disposal sites and unsafe landfills, hydro-electric dams, mining, pulp and paper, and other industrial activities. Additionally, POPs are believed to fractionate and fall out en route to Arctic regions (Wania and Mackay, 1996); depositing in remote Boreal ecosystems such as the northern reaches of Ontario. Compared to Arctic populations, First Nations south of the 60th parallel are generally neglected and environmental contaminants in these remote communities practicing traditional lifestyles is often underreported and underplayed. They also continue to fall victim to high suicide rates among youth as compared to non-aboriginals, alcohol and drug abuse, poor water quality, substandard housing conditions, low education levels and high unemployment rates (Statistics Canada, 2001).

Variable Diets and Effects of Fish and Wild Food Consumption

Northern epidemiology and nutritional studies are often plagued by many confounding factors, including smoking and drinking, contaminant exposures, nutrient complexities such as the general nutritional status of mothers (folic acid, Ω-3 fatty acids, iron), contaminant-nutrient interactions (Hg-Se, Hg-tropical fruits, Hg-teas, Hg-Ω-3 fatty acids, etc.), genetic factors, lack of suitable controls (i.e., non-fish-eating populations), limited sample sizes, variable diets, and dietary recall error (Becker and Welten, 2001; Canuel *et al.* 2006; Delormier and Kuhnlein, 1999; Fuller *et al.* 2005; Mergler *et al.* 2007; Passos *et al.* 2003; Van Oostdam *et al.* 2005). The list of confounding factors is long, however the focus of this study will be on variable diets and how they can affect dietary markers and contaminant exposures in two remote First Nation communities in north-western Ontario. With the large differences in dietary behaviours

observed within and between contemporary First Nations communities, variable diets will no doubt affect contaminant exposures within and between communities. It is important to note that this does not apply only to First Nations communities in north-western Ontario. Over a million First Nations people located in communities all across northern Canada have in the last 50 years undergone dramatic lifestyle transformations despite their remote locations, and are experiencing alarming rates of obesity and obesity-related diseases (in particular, type-2 diabetes), while at the same time are exposed to environmental contaminants through locally-harvested wild food consumption. When conducting benefit-risk assessments of wild food consumption, it is important to fully understand the specific dietary and cultural behaviours of First Nations populations while also considering environmental contaminants which may affect the health of First Nations communities.

This thesis presents the effects of wild food consumption on abundances of stable isotopes and environmental contaminant concentrations measured in hair and blood from individuals residing in two isolated and remote north-western Ontario First Nations communities who eat varying amounts of locally-harvested wild foods. The main objectives of this thesis were: 1) to establish relationships between dietary behaviour and stable isotopes and environmental contaminant concentrations in First Nations peoples; and, 2) to assess exposures to environmental contaminants in high-frequency and low-frequency wild food consumers from these two First Nations communities.

Specifically, the first objective was to examine the effect of varying frequencies of wild food and fish consumption, as generally practiced in these First Nations communities, on abundances of stable isotopes and environmental contaminant concentrations in body tissues and assess the potential for these to serve as dietary markers. The second objective was to examine

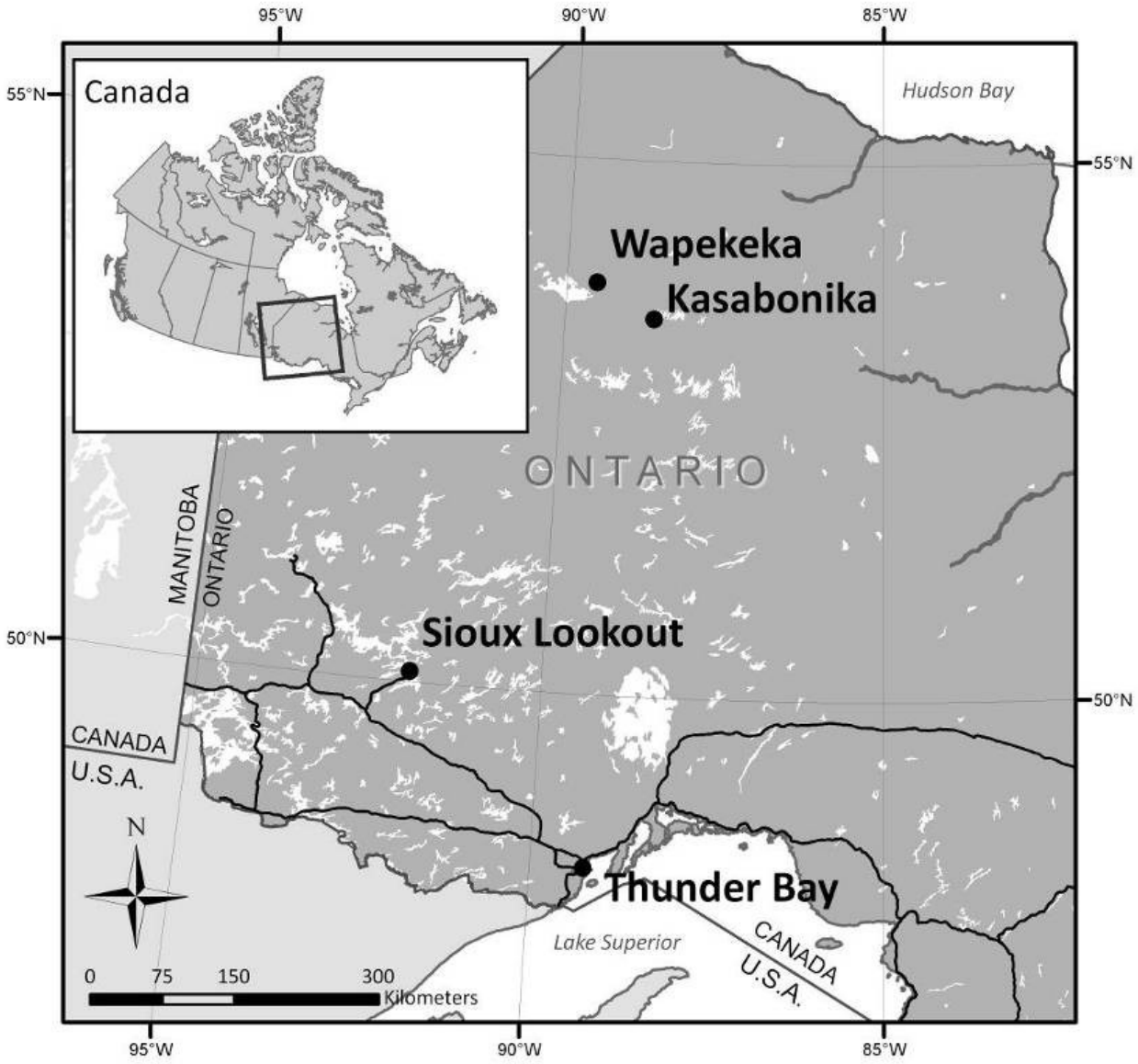
the effect of varying frequencies of wild food consumption, as generally practiced in these First Nations communities, on the presence and concentrations of contaminants in the body tissues of First Nations people residing in these communities.

2.0 Methods

2.1.1 Study Locations and Populations

During the months of August through November 2007, two north-western Ontario (Canada) Ojibwa-Cree First Nations communities (Figure 2.1) were visited by researchers from the University of Ottawa. Wapekeka First Nation (pop. 328) is located at the mouth of the Fawn River on Wapekeka (Angling) Lake, 26 km northwest of Big Trout Lake and 451 km northeast of Sioux Lookout, Ontario, while Kasabonika First Nation (pop. 791) is located on the Ashweig River, 480 km northeast of Sioux Lookout. Kasabonika First Nation is approximately 30 km south-east of Wapekeka First Nation.

Figure 2.1: Map showing Wapekeka and Kasabonika First Nations in north-western Ontario, Canada (inset).



Participant recruitment was conducted in Wapekeka and Kasabonika First Nations using a mixed-method ethnological approach (Hammersley and Atkinson, 1995) in September 2007. A total of 83 individuals were incrementally recruited for this community-based study and interviewed with the assistance of the First Nations band councils, local research coordinators and translators. The target number of participants was 100. Participants were recruited based on self-described dietary preferences, including their reliance on either wild foods and/or market foods. No vegetarians were identified in these populations however this is not to say that there are none. Informed consent was obtained from each adult participant following guidelines approved by the University of Ottawa Research Ethics Board and Health Canada Ethics Board. Of the 83 individuals that were recruited, only 72 participated in the study. Eleven of the recruited individuals were not included in the study because they chose not to participate. The 72 participants were assigned an identification code to ensure anonymity and confidentiality. Demographic and dietary information was collected using a mixed-method ethnological approach via semi-structured interviews, food frequency questionnaires, 3-day dietary records and 24-hour dietary recalls. Further details on participant recruitment and the mixed-method ethnographic approaches are provided in Robidoux *et al.* (2012). To test the effect of dietary behaviour on stable isotopes and environmental contaminants in body tissues, it was necessary to select individuals from the populations and divide them into two groups of food consumers; those that eat relatively higher amounts of wild foods and those that eat relatively higher amounts of market foods. It is important to note that wild food consumption refers to primarily wild meat from fish, mammals, and birds. In this remote region of northern Ontario, a wild food diet is essentially animal-based, consisting primarily of freshwater fish, moose (*Alces alces*), beaver (*Castor canadensis*) and geese (*Branta canadensis*). While wild berries and other edible

plants exist in these study regions, ethnographic observations and dietary records indicated that wild edible plants made up a negligible proportion of the wild food intake for most participants. Dietary records indicated that the relative contribution of food sources as based on the four basic food groups (i.e., vegetables and fruits, grain products, meat and alternatives, and milk and alternatives) was similar among combined high-frequency wild (HW) food consumers and low-frequency wild (LW) food consumers. However, upon further analysis of the meat and alternatives group, it was evident that HW participants replaced a portion of store-bought meat with wild fish and/or hunted meats, whereas LW participants consumed almost exclusively store-bought meats.

Participants from the two communities were grouped based on their frequency of wild food consumption by determining dietary behaviours based on semi-structured individual interviews. Inclusion criteria required that individuals be Aboriginal, over 18 years of age, non-pregnant, and free of type-1 diabetes. None of the participants had type-1 diabetes however 26 of the 72 participants of the participants were diagnosed as type-2 diabetic. The number of individuals participating in each community (39 in Wapekeka and 33 in Kasabonika) represents approximately 9% of the eligible population in Kasabonika and 24% of the eligible population in Wapekeka at the time of the study. One of the 72 participants was excluded from the analyses because of incomplete dietary information. Based on dietary data recorded through ethnographic observations, semi-structured interviews, food frequency questionnaires, 3-day dietary records and 24-hour recalls, individuals were ranked according to the frequency of their wild food consumption. Participants were placed in the HW food consumption group if their WFFI was 60 or higher and they ate more than 2 wild food meals per week. Participants were placed in the LW food consumption group if their WFFI was 40 or lower and they ate less than 1 wild food

meal per month. HW and LW food consumption groups were further divided into three categories of HW and LW food consumers to determine thresholds in which we observed differences in dietary markers, contaminant concentrations and anthropogenic measurements between HW and LW. We used a variable threshold for defining HW food consumption and LW food consumption in 71 of the 72 participants: Category 1: HW1 (n=21), ≥1 wild food meal per day, LW1 (n=50), <1 wild food meal per day; Category 2: HW2 (n=24), ≥1 wild food meal per week, LW2 (n=47), <1 wild food meal per week; Category 3: HW3 (n=43), ≥2 wild food meals per month, LW3 (n=28), <2 wild food meals per month.

Individuals were further grouped in relation to their level of fish consumption because fish is an important locally-harvested wild food source and one of the only wild foods that is widely available throughout the entire year in both Kasabonika and Wapekeka. A Fish Consumption Frequency Index (FCFI) was developed based on the number of fish meals eaten by individuals, as reported in individual interviews and food frequency questionnaires. Those who ate fish meals less than once a month or never were assigned to group 1 (n=35), less than once a week to group 2 (n=14), at least once a week to group 3 (n=12), or more than twice a week to group 4 (n=9).

2.1.2 Sample Collection and Preparation

Blood (n=72) and hair (n=71) samples were collected in October and November 2007 from participants. Blood samples were collected from resting and fasted individuals, upon which they were immediately placed on ice and immediately centrifuged at 3500 revolutions per minute before temporarily freezing plasma at -20°C for safe shipping to the University of Ottawa's Behavioural and Metabolic Research Unit (BMRU) at Montfort Hospital in Ottawa, Ontario. Upon receiving samples at BMRU, plasma samples were stored at -80°C until sample analysis.

Hair samples were collected from the nape of the head as near as possible to the scalp using stainless steel scissors, upon which they were sealed separately in labelled bags for safe transportation to the University of Ottawa. Hair was not collected from one of the participants in Wapekeka, hence the discrepancy between the number of blood (n=72) and hair (n=71) samples. All hair samples (n=71) were cut to 1 centimetre (cm) lengths, starting from the base, which represented approximately one month of recent hair growth. Hair samples were then cleaned by soaking in a 2:1 chloroform:methanol solution to remove any lipid residues and then rinsed several times with distilled water. Samples were thoroughly dried before any analysis was performed.

Anthropometric measurements were collected prior to blood and hair collection and included body weight, height, and waist circumference. Body weight was determined with a standard beam scale, and height and waist circumference were measured with a measuring tape. Height was measured with the participant's bare feet together, with heels, back, and head against a wall, and following normal inspiration. Waist circumference was measured directly on the skin, in duplicate and averaged, at the mid-point between the last floating rib and the top of the iliac crest. Body mass index (BMI) was calculated by dividing body weight (in kilograms, kg) by height (in metres, m) squared.

2.1.3 Stable Isotope Analysis of Hair Samples

Abundances of stable isotopes of organic carbon and nitrogen in the hair of study participants were measured at the University of Ottawa's G.G. Hatch Stable Isotope Laboratory. Cleaned hair samples (approximately 0.6 milligram) were cut, weighed, and placed into tin capsules (8 x 5 millimetres (mm), Isomass Scientific) for determination of bulk ^{13}C and ^{15}N abundances. The isotopic composition of organic carbon and nitrogen is determined by the

analysis of CO_2 and N_2, produced by flash combustion at 1800 °C on a CE 1110 Elemental Analyzer. This is followed by gas chromatograph separation and on-line analysis by continuous-flow with a DeltaPlus Advantage isotope ratio mass spectrometer coupled with a ConFlo interface. Data were normalized using internal standards previously calibrated with International standards IAEA-CH-6, IAEA-NBS22, IAEA N1, IAEA-N2, USGS-40, USGS-41. Analytical precision is +/-0.2‰. Isotopic ratios are denoted and calculated using equation (1), where X is the heavier isotope (^{13}C, ^{15}N), R_{sample} is the raw ratio of the heavy to light isotope in the tissue sample, and $R_{standard}$ is the raw ratio of the heavy to light isotope in the internationally accepted standard. The standards used for $\delta^{13}C$ and $\delta^{15}N$ include PeeDee Belemite (PDB) carbonate limestone and atmospheric nitrogen (N_2), respectively.

$$\delta X = \frac{R_{sample} - R_{standard}}{R_{standard}} \quad (1)$$

2.1.4 Environmental Contaminant Analysis of Blood and Hair Samples

Organic contaminants in blood (plasma) included here are Aroclor1260, polychlorinated biphenyl (PCB)28, PCB52, PCB99, PCB101, PCB105, PCB118, PCB128, PCB138, PCB153, PCB156, PCB163, PCB170, PCB180, PCB183, PCB187, aldrin, α-chlordane, γ-chlordane, β-hexachlorocyclohexane (β-HCH), *cis*-nonachlor, *trans*-nonachlor, *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE), dichlorodiphenyltrichloroethane (DDT), hexachlorobenzene (HCB), mirex, oxychlordane, polybrominated biphenyl (PBB)153, PBDE47, PBDE99, PBDE100, PBDE153, toxaphene Parlar26, and toxaphene Parlar50. Organic contaminants were measured using an E-446 GC-MS (gas chromatography-mass spectrometry), Chromatograph 6890 (Agilent) using a solid phase extraction followed by gas chromatography coupled to mass detection (Agilent 5973 network). The plasma samples were enriched with

internal standards and denatured with formic acid. The compounds were extracted from the aqueous matrix using solid phase separation and extracts were cleaned using florisil columns prior to analysis. Plasma samples were eluted from columns using methylene chloride-hexane (25:75 vol/vol) and analyzed on gas chromatograph equipped with dual capillary columns. Peaks were identified by relative retention times obtained on the two columns using a computer program developed by the Quebec Toxicology Centre. Generated ions were measured after negative chemical ionization. The concentration of each analyte measured was determined using percent recovery of labelled internal standards. The ECD (electron capture detector; Agilent G2397A) served to verify the detection limits for PCB congeners 28 and 52. To verify results, an interlaboratory comparison was made with the External Quality Assessment Scheme (G-EQUAS), Germany. Data were screened to exclude POPs that were below the detection limit in more than 60% of cases. Individual PCB congeners are not included here, but rather the sum of 12 PCB congener concentrations (PCB99, PCB105, PCB118, PCB128, PCB138, PCB153, PCB156, PCB163, PCB170, PCB180, PCB183, PCB187) is presented. Aroclor 1260 was deemed the most appropriate measurement of exposure to PCBs as it was detected in the blood of every participant and none of Aroclor 1260 concentrations measured here were below the method's detection limit. To reduce bias, random numbers were generated between 0 and the detection limit of each POP included in analysis for cases below the detection limit (Miller & Amrhein, 1995). POPs were considered in the analysis if the detection frequency was greater than 60%. The replacement of non-detects with random numbers did not change the significance of statistical tests used during analysis.

Total mercury in the hair of participants was measured at the Laboratory for the Analysis of Natural and Synthetic Environmental Toxicants (L.A.N.S.E.T.) in the University of Ottawa's

Centre of the Advanced Research in Environmental Genomics (CAREG). For mercury analysis, a 2 to 5 milligram sample of a hair (in 1 cm lengths from the scalp) from 71 of the 72 participants was placed in a Mercury SP-3D analyzer (Nippon Instruments Corporation, Japan) which heated the samples to a maximum temperature of 950°C. Mercury released from the hair was subsequently collected and isolated in a two-stage gold amalgam process before being transferred and detected via cold vapor atomic absorption spectroscopy. Blanks and a standard solution diluted from a stock solution of Fisher Scientific (CSM114-100) Certified Reference Material (1000 ppm for Trace Metals sample) (Dorm-3, National Research Council). Mercury values of the standard solution (n=19) were 51.68 ± 0.15 parts per billion (ppb) standard deviation (SD) compared to the mercury standard value of 56 ± 10 ppb SD. Sample recovery was 92.3% and our results were within the normal range. Mercury values of Dorm-3 (n=5) were 344.4 ± 23.44 ppb SD (milligram/kilogram, dry weight) compared to the certified mercury value of 382 ± 60 ppb SD. Our results were slightly lower than certified values, but within their normal range.

2.1.5 Environmental Contaminant Analysis of Wild Food Samples

Wild food samples were homogenized using a meat grinder. Samples for total mercury analysis were freeze-dried to determine water content and homogenized. Subsamples were analyzed in triplicate on a Mercury SP-3D analyzer (Nippon Instruments Corporation, Japan). A certified reference material (Dorm-3, National Research Council Canada) was analyzed after every 5 to 10 samples to ensure accuracy and reproducibility of the results. Detection limits were 0.01 nanogram (ng) mercury per sample. Dorm-3 certified reference materials met reported concentrations of 382 ± 24 ppb.

Wild food samples for organic contaminant analysis were mixed with Hydromatrix (Varian), spiked with recovery standards (1,3-DBB, 1,3,5-TBB, 1,2,4,5-TTBB, d-HCH, Endrin Ketone, BZ30 and 205, Ultra Scientific). The mixture was extracted following procedures in Dionex Application Note 322 (1996) using an Accelerated Solvent Extractor 200 (Dionex). The sample extracts were cleaned using EPA Method 3640A (1994) to remove lipids. The samples were injected on two Envirogel columns (150 and 300 mm, Waters) connected sequentially to a preparative 1200 HPLC coupled with a photodiode array and fraction collector (Agilent Technologies). The collected sample fractions were evaporated to 1 millilitre in 2,2,4-trimethylpentane (Fisher Scientific) and further fractionated on 8 grams of silica Davisil 635 (Fisher Scientific) packed into a chromaflex column with hexane following methods in EPA Method 3630C (1996). The samples were evaporated to 500 microlitre (µL) in 2,2,4-trimethylpentane for analysis on a 6890 Gas Chromatograph with a micro Electron Capture Detector (Agilent Technologies) following methods in EPA Method 508.1 (1995). One µL was injected in splitless mode on a DB-5MS 60m, 250 micrometre (µm), 0.25 µm column (J&W Scientific).

Chromatographic peaks were interpreted using Agilent Chemstation software (Rev. B.03.01). Compounds were identified by analyzing a linear set of standards and comparing their retention times with those of the sample compounds. From the standard mixtures, approximately 35 PCB congeners and 31 pesticide compounds were confirmed. Quantitative analysis was completed with the use of octachloronaphthalene as an internal standard in the standard mixtures and samples.

Analytical blanks (n=12), comprised Hydromatrix® and recovery standards subjected to the entire extraction and sample clean-up procedures, contained an average of 92 pg of PCBs

based on a sample weight of 10 grams. Recovery was 73.4% ± 21.3% SD for PCB 30 and PCB 205. All samples were blank subtracted and recovery corrected. Standard Reference Material (SRM2977, National Institute of Standards and Technology) was subjected to the extraction and sample clean-up procedures, and values fell within certified limits.

2.1.6 Statistical Analyses

Raw laboratory analytical results are presented in the Appendix. Unless otherwise indicated, data are presented as means ± standard error (SE). Normality and log-normality were assessed using the Shapiro-Wilks test. Accordingly, data were log-transformed prior to statistical analyses. Aside from log(Mirex), log(β-HCH) and log(Parlar26), all data were log normalized. Data were analyzed using analysis of variance (ANOVA) or Student's t-test. Differences between mean values were verified post hoc with Tukey's honest significant difference test. Analysis of covariance (ANCOVA) was used to adjust for age and test the combined effects of age and wild food consumption frequency on contaminant concentrations. Aside from log(Mirex), log(β-HCH) and log(Parlar26), all data were log normalized. Results with a p-value of less than 0.05 were considered statistically significant. All statistical analyses were performed using JMP version 5.1.2 (SAS Institute Inc.) and SigmaPlot version 11.0 (Systat Software, Inc.).

3.0 Stable isotopes and contaminants correlated with dietary preferences: Dietary markers in two remote First Nations communities in Northern Ontario (Canada)

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Timothy Andrew Seabert conducted the field study, sample management and preparation, stable isotope analysis, mercury analysis, data analysis and writing. Shinjini Pal and Eva M. Krümmel participated in the field study and assisted with sample management. Michael A. Robidoux, François Haman, Pascal Imbeault and Jules M. Blais were the principal investigators who proposed and designed the research program.

3.1 Abstract

This study examined the use of stable isotopes as dietary markers in two First Nations communities where there is known variation in the frequency of consumption of traditional country foods among individuals from these communities. Specifically, we investigated whether $\delta^{13}C$ and $\delta^{15}N$ in hair relate to the frequency of wild food and fish consumption and to mercury and PCB concentrations in hair and blood, respectively. We observed significant differences in $\delta^{13}C$ and $\delta^{15}N$ between varying frequencies of high-frequency wild (HW) food consumers and low-frequency wild (LW) food consumers. In addition, $\delta^{15}N$ was positively correlated with mercury in hair and PCBs in blood. $\delta^{15}N$ also increased with higher frequencies of wild food and fish consumption, and with age. Mercury and PCB concentrations also increased significantly with age, due to higher frequency of wild food and fish consumption among older individuals. $\delta^{13}C$ hair decreased with higher frequencies of wild food and fish consumption, and with age. Our results demonstrate that known differences in dietary behaviour among HW and LW food consumption groups from two First Nations communities in Northern Ontario are clearly reflected in stable isotope ratios and that stable isotope ratios are correlated not only to dietary preferences and the trophic level at which an individual is feeding, but also to contaminant concentrations and age.

3.2 Introduction

Throughout northern Canada, First Nations communities have undergone dramatic lifestyle changes in recent decades. The increased availability of low quality foods (Batal *et al.*, 2005) along with lower levels of physical activity (Harris *et al.*, 1997; Katzmarzyk, 2008; Liu *et al.*, 2006) have contributed to the increased prevalence of obesity and obesity-related diseases

observed among Aboriginal peoples in Canada, particularly type-2 diabetes mellitus (T2DM) (Young *et al.*, 2000; Gittelsohn *et al.*, 1998; Harris *et al.*, 1997; Garriguet, 2008). Despite the fairly recent nutrition transition from a primarily traditional-based diet to a market-based diet, fish and other locally-harvested wild foods continue to represent an important part of the diet of northern First Nations peoples. This dietary transition has not been uniform across First Nations communities. The degree of traditional wild food consumption varies greatly between communities and individuals. It is therefore very important to fully understand the specific dietary behaviours between communities and individuals with respect to both traditional wild food and store-bought food consumption when conducting dietary assessments. This is of particular value where there are highly variable diets and significant under-reporting when conducting dietary assessments using conventional techniques such as food frequency questionnaires and dietary recalls. Distinct differences in food consumption can be measured in human biological tissues using dietary markers such as stable isotopes. These are directly related to both food sources and dietary behaviour of a particular region (Nardoto *et al.* 2006).

Naturally-occurring stable isotope ratios of carbon ($^{13}C/^{12}C$ or $\delta^{13}C$) and nitrogen ($^{15}N/^{14}N$ or $\delta^{15}N$) have been used extensively to gather information about historical human diets (Macko *et al.*, 1999; Schoeninger and DeNiro, 1984; van der Merve *et al.*, 2003), and to an increasing extent, the contemporary human diet (O'Connell and Hedges, 1999; Jahren *et al.*, 2006; Petzke *et al.*, 2005a, 2005b; Wilkinson *et al.*, 2007). They have become valuable dietary markers for providing quantitative and objective information about the human diet, particularly sources of nutrition (Petzke *et al.*, 2005a, 2005b; Jahren *et al.*, 2006). Isotopic methods using $\delta^{13}C$ and $\delta^{15}N$ have been developed to quantify the consumption of sugars and sweeteners made from corn and sugar cane (Jahren *et al.*, 2006) and the consumption of protein (Petzke *et al.*

2005a, 2005b). Their abundances have been shown to clearly reflect dietary behaviour (O'Connell and Hedges, 1999; Bol and Pflieger, 2002; Petzke *et al.*, 2005a, 2005b). The use of these methods can be directly applied to assessing dietary preferences within and between First Nations communities. Many of the store-bought foods that are currently available in First Nations communities are highly processed foods with low nutrient density because they are relatively inexpensive when compared to fresh meats and produce and have a long shelf life. Examples of store-bought foods that are typically available in First Nations communities are simple carbohydrates and starches (i.e., pasta, rice and white bread), processed meats, along with foods and drinks containing corn-derived sugar sweeteners such as high fructose corn syrup (HFCS). Based on the enrichment of ^{13}C observed in these foods (Jahren *et al.*, 2006), individuals consuming primarily store-bought foods and having a low-frequency wild food diet will have a higher $\delta^{13}C$ value incorporated into their body tissues than those relying more heavily on locally-harvested wild foods. Similarly, $\delta^{15}N$ values are also an indication of what an individual is eating and the composition of their food, reflecting not only dietary behaviours but also the trophic level at which an individual is feeding (O'Connell and Hedges, 1999; Petzke *et al.*, 2005a, 2005b). Food web studies have shown a step-wise enrichment of $\delta^{13}C$ and $\delta^{15}N$ by approximately 1‰ and 3‰, respectively, with each increasing trophic level (Minawaga and Wada, 1984) demonstrating the biomagnification of $\delta^{13}C$ and $\delta^{15}N$ with increasing trophic levels. Based on the enrichment of ^{15}N in a high wild food diet, individuals relying more heavily on locally-harvested fish (including store-bought fish) and other high trophic level wild foods will have higher $\delta^{15}N$ values incorporated into their body tissues than individuals consuming primarily market foods and having a low-frequency wild food diet.

Despite the dietary shifts occurring in northern First Nations communities, wild fish continue to be an important and healthy food choice for First Nations people as they are low in refined carbohydrates and saturated fats, and high in protein, polyunsaturated fatty acids and other important nutrients. However, fish are also a primary source of mercury exposure (Chapman and Chan, 2000). This is particularly important for individuals that rely on fish as a source of nutrition. Depending on frequency of fish consumption (both store-bought and locally harvested), individuals may accumulate mercury in their body tissues over time despite their ability to metabolize and excrete mercury (Clarkson *et al.* 2007, Counter and Buchanan, 2004). Hair mercury has been shown to be a reliable marker of not only mercury exposure but also fish consumption in high fish-eating populations (Passos *et al.*, 2003; Legrand *et al.*, 2005, 2007). Mercury from fish consumption is rapidly deposited into scalp hair within 3-5 days of a single exposure (Kershaw *et al.*, 1980). As scalp hair grows at a rate of approximately 1 centimeter per month, a chronology of mercury exposure may also be determined from hair strands. Polychlorinated biphenyls (PCBs), which are present in relatively high blood concentrations in individuals consuming fatty fish, are another relevant dietary marker. As with mercury, human exposure to PCBs is mainly through the consumption of fish and other hunted meats. PCBs accumulate in fatty tissues and metabolize at an extremely low rate, with tissue concentrations increasing with age. Blood concentrations have been used extensively as an important marker of environmental and dietary exposures to PCBs and other POPs. The use of blood concentrations is limiting in the sense that they represent only the most recent exposures and cannot be used to measure previous exposures unless collected on a temporal basis. Based on the assumption of contaminant accumulation in locally-harvested wild foods, individuals relying more heavily on fish and other wild foods will have a higher body burden of mercury and PCB concentrations

(i.e., mercury in hair and PCBs in blood, respectively) than those consuming primarily market foods.

The purpose of this study was to investigate whether $\delta^{13}C$ and $\delta^{15}N$ are reliable dietary markers to discriminate between varying frequencies of high-frequency wild (HW) food consumers and low-frequency wild (LW) food consumers in two Ojibwa-Cree First Nations communities of northern Ontario, Canada (Figure 2.1) as described in section 2.2.1. When compared to LW food consumers, we predict that HW food consumers in their respective categories will have higher $\delta^{15}N$ and lower $\delta^{13}C$. We also predict that these isotopic ratios will be correlated to mercury and PCB concentrations.

3.3 Results and Discussion

Age, sex ratios, WFFI, weight, height, waist circumference, and BMI are presented in Table 3.1 for each of the three categories of HW and LW food consumers. Age differences were observed between high-frequency wild food consumers within the Category 1 and Category 2 groups with individuals in the HW1 and HW2 groups significantly older than those in the LW1 and LW2 groups, respectively ($p<0.05$). No age differences were observed between the HW3 and LW3 groups likely due to the fact that younger individuals tend to consume less wild foods, and more market foods, than older individuals; hence, we see younger individuals and more subtle differences in age and wild food consumption among these groups. The combined age range of all participants spanned 23 to 73 years with an average age of 43 years. No age difference was observed between pooled males and females ($p=0.76$). Sex ratios were reasonably matched between HW and LW food consumption groups within each of the three categories. There were more females than males in all groups except for HW1 and HW2 which

had only slightly more males. Body weight, height, waist circumference, and body mass index (BMI) did not differ between any of the HW and LW groups in Categories 1, 2 and 3.

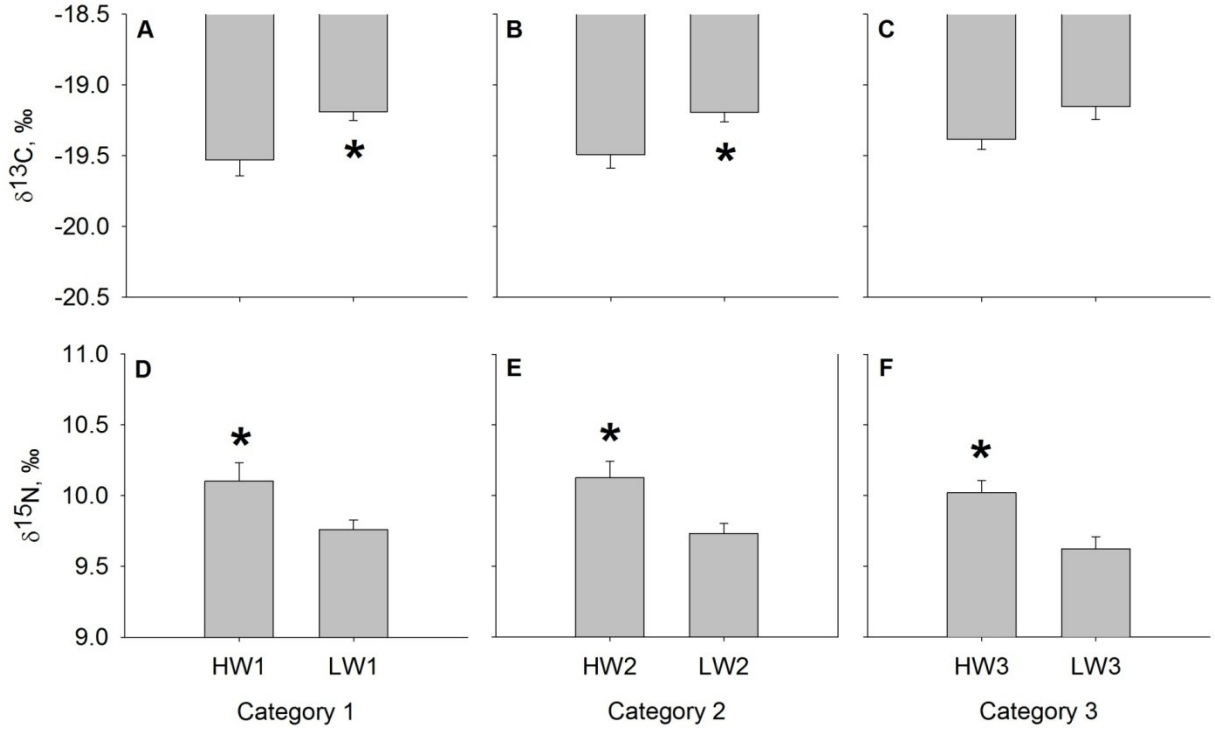
Table 3.1: Group profiles for the three categories of high-frequency wild food consumer (HW) and low-frequency wild food consumer (LW) groups. Category 1: HW1 (n=21), ≥1 wild food meal per day, LW1 (n=50), <1 wild food meal per day; Category 2: HW2 (n=24), ≥1 wild food meal per week, LW2 (n=47), <1 wild food meal per week; Category 3: HW3 (n=43), ≥2 wild food meals per month, LW3 (n=28), <2 wild food meals per month. Means ± SD and selected ranges are presented. A bolded value indicates a difference with a significance of $p<0.01$ between HW and LW groups within each respective category.

	HW1	LW1	HW2	LW2	HW3	LW3
n	**21**	50	**24**	47	**43**	28
Age	**51±16 (23-73)**	41±10 (23-63)	**50±16 (23-73)**	39±9 (23-63)	46±15 (23-73)	39±8 (23-57)
Sex (M/F)	12/9	19/31	13/11	18/29	18/25	13/15
WFFI (0-100)	**82±4**	40±24	**80±6**	38±24	**73±9**	20±10
Weight (kg)	95±18	87±16	92±19	88±16	90±16	88±18
Height (cm)	168±12	167±8	167±12	167±8	167±10	167±8
Waist (cm)	116±10	112±14	115±10	112±14	114±11	112±15
BMI (kg/m^2)	34±4	31±5	33±5	32±5	32±4	31±5

3.3.1 Stable Isotopes

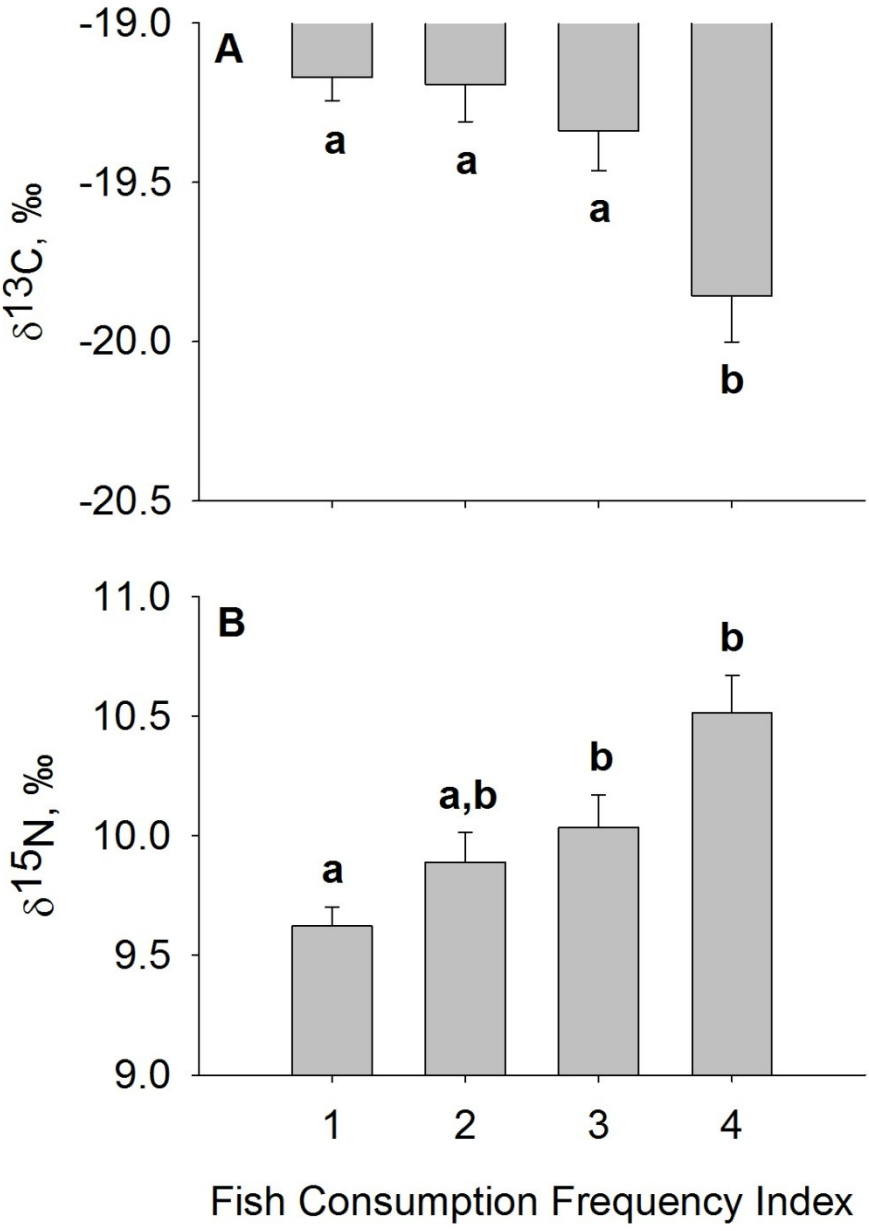
Significant enrichment of ^{13}C was measured in hair from individuals in the LW1 (-19.19 ± 0.06‰) and LW2 (-19.19 ± 0.07‰) food consumption groups when compared to mean $\delta^{13}C$ values from the HW1 (-19.53 ± 0.11‰) and HW2 (-19.49 ± 0.10‰) groups, respectively (Figures 3.1A and 3.1B, ANOVA, $p<0.05$). There was no significant difference in hair ^{13}C enrichment between HW3 (-19.38 ± 0.07‰) and LW3 (-19.15 ± 0.09‰, Figure 3.1C). Fruit and vegetables (C3 plants) which carry a distinctive $\delta^{13}C$ signature are either avoided or unavailable due to their high cost and perishability and are generally lacking in the typical First Nations diet, as observed in these populations. Significant enrichment of ^{15}N was measured in hair from individuals in HW1, HW2 and HW3 food consumption groups when comparing to the LW groups within their respective categories (Figures 3.1D, 3.1E and 3.1F). While there were no significant differences in $\delta^{13}C$ between Kasabonika and Wapekeka First Nations communities ($p=0.41$), $\delta^{15}N$ was significantly higher in Kasabonika ($p<0.05$).

Figure 3.1: Mean $\delta^{13}C$ ± SE (‰) and mean $\delta^{15}N$ ± SE (‰) for Category 1 (HW1 and LW1), Category 2 (HW2 and LW2) and Category 3 (HW3 and LW3) food consumption groups. Category 1: HW1 (n=21), ≥1 wild food meal per day, LW1 (n=50), <1 wild food meal per day; Category 2: HW2 (n=24), ≥1 wild food meal per week, LW2 (n=47), <1 wild food meal per week; Category 3: HW3 (n=43), ≥2 wild food meals per month, LW3 (n=28), <2 wild food meals per month. An asterisks (*) indicates a significantly higher ($p<0.05$) value in individuals from the respective food consumption group.



Mean $\delta^{13}C$ and $\delta^{15}N$ were further plotted against a scale of increasing fish consumption by the study participants using a Fish Consumption Frequency Index (FCFI). Figure 3.2 illustrates that frequency of fish consumption significantly influences $\delta^{13}C$ and $\delta^{15}N$ in hair. Based on individual interviews and food frequency questionnaires, the consumption of store-bought (i.e., canned or frozen fish) was negligible and therefore fish consumption, as practiced in these populations, is defined here as consuming locally-harvested freshwater fish. Mean $\delta^{13}C$ decreased with greater consumption of fish (a ^{13}C depleted food source). Mean $\delta^{13}C$ was significantly higher in fish consumption groups 1 (less than one fish meal per month or never, n=35), 2 (less than one fish meal a week, n=14) and 3 (at least one fish meal per week, n=12) (-19.17 ± 0.07‰, -19.19 ± 0.11‰ and -19.34 ± 0.13‰, respectively) than in group 4 (at least two fish meals per week, n=9) (-19.86 ± 0.15‰) (Figure 3.2A, ANOVA, $p<0.05$). The inverse relationship between abundance of ^{13}C and fish consumption supports the fact that individuals eating less fish (or wild food for that matter) tend to rely more heavily on market foods and subsequently are consuming more ^{13}C enriched food products which is reflected in the higher $\delta^{13}C$ values in individuals with a FCFI of 1, 2 and 3. Mean $\delta^{15}N$ was significantly higher in individuals that consumed fish at least one a month, or having a FCFI of 2, 3 or 4 (9.89 ± 0.13‰, 10.04 ± 0.14‰ and 10.51 ± 0.16‰, respectively) compared individuals that consumed fish less than once a month, or having a FCFI of 1 (9.62 ± 0.08‰) (Figure 3.2B, ANOVA, $p<0.05$), indicating an enrichment of ^{15}N with greater consumption of fish (a ^{15}N enriched food source). $\delta^{15}N$ is often used in wildlife studies as a measure of trophic status and individuals who consume fish tend to be eating higher in the food chain as compared to those eat very little or no fish.

Figure 3.2: Mean $\delta^{13}C \pm SE$ (‰) and mean $\delta^{15}N \pm SE$ (‰) plotted against fish consumption frequency index (FCFI). Indices that do not share a common symbol are significantly different ($p<0.05$). Notes: FCFI 1 = less than one fish meal per month or none (n=35), FCFI 2 = less than one fish meal per week (n=14), FCFI 3 = at least one fish meal per week (n=12), and FCFI 4 = more than two fish meals per week (n=9).

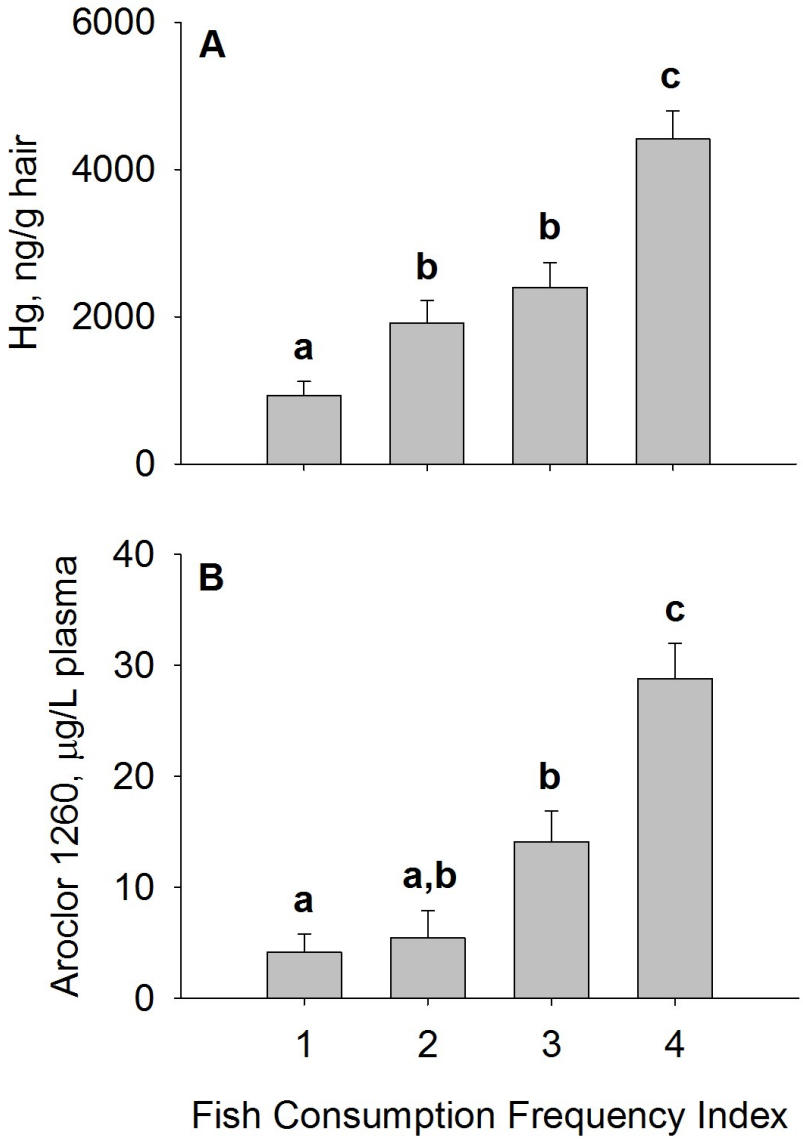


3.3.2 Mercury and PCBs

Frequency of fish consumption was significantly correlated with both mercury in hair and PCBs in blood. Mean mercury concentrations in hair were significantly higher for those who ate fish at least twice a week, i.e., having a FCFI of 4 (4410 ± 386 ng/g hair), when compared to those who ate fish once a week or less, i.e., having a FCFI of 1, 2 or 3 (924 ± 196 ng/g, 1913 ± 309 ng/g and 2398 ± 334 ng/g, respectively) (Figure 3.3A, ANOVA, $p<0.05$). No difference in mean mercury concentrations was observed between those having a FCFI of 2 (fish once a week) and those having a FCFI of 3 (fish once a month). Those that eat fish less than once a month or never (FCFI of 1) have significantly lower mercury concentrations than those who eat fish at least once a month or more, i.e., having a FCFI of 2, 3, or 4 (Figure 3.3A, ANOVA, $p<0.05$).

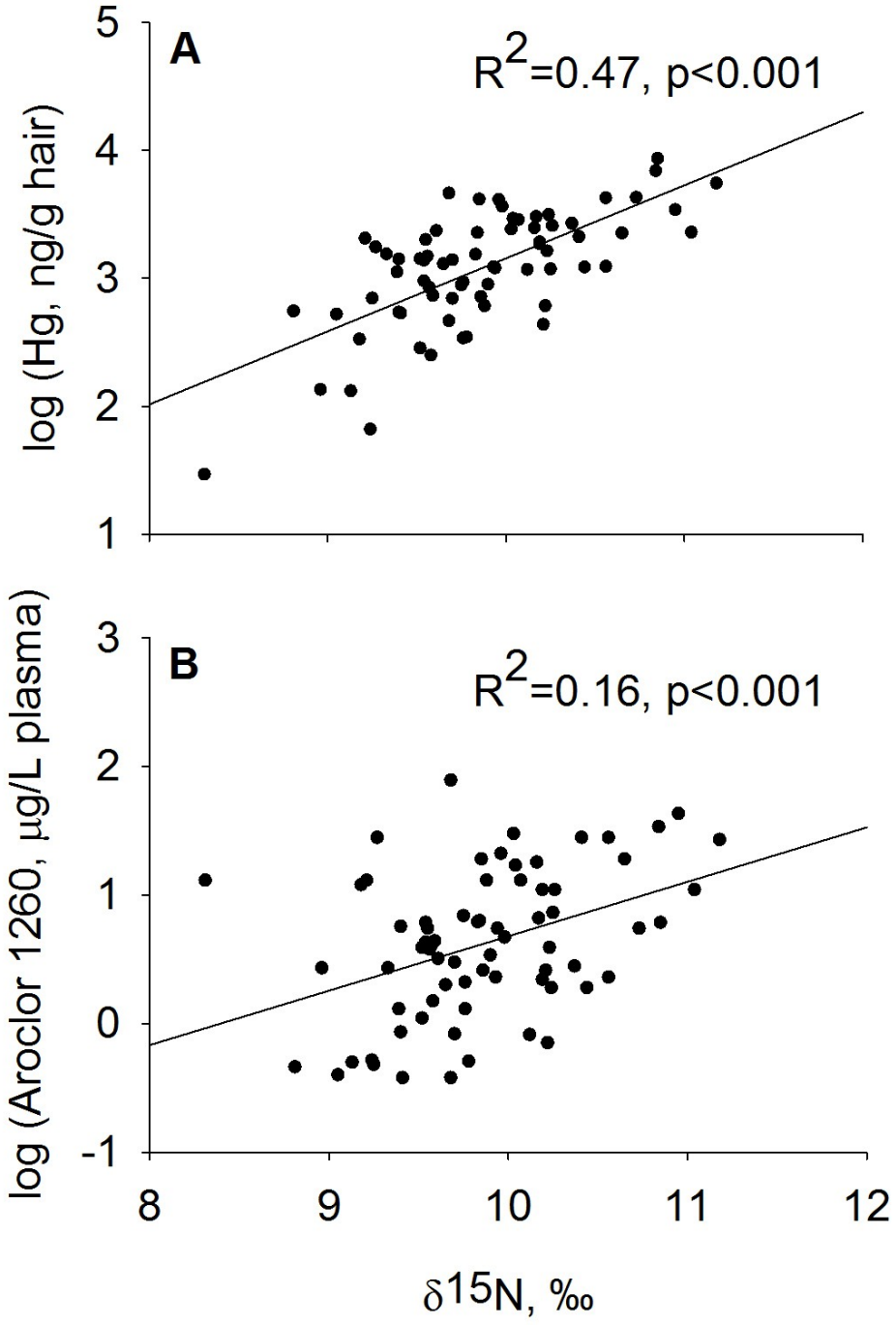
Mean PCB concentrations (as Aroclor 1260) in plasma were also significantly higher for those who ate fish at least twice a week, i.e., having a FCFI of 4 (28.8 ± 3.2 µg/L), when compared to those who ate fish once a week or less, i.e., having a FCFI of 1, 2 or 3 (4.1 ± 1.6 µg/L, 5.4 ± 2.5 µg/L and 14.1 ± 2.8 µg/L, respectively) (Figure 3.3B, ANOVA, $p<0.05$). Although PCB concentrations are lower for those having a FCFI of 2 than those having a FCFI of 3, they are not statistically different, nor are there significant differences in PCB concentrations between FCFI groups 1 and 2. However, those that eat fish less than once a month or never (FCFI of 1) have significantly lower PCB concentrations than those who eat fish at least once a month or more, i.e., having a FCFI of 2, 3, or 4 (Figure 3.3B, ANOVA, $p<0.05$).

Figure 3.3: Mean mercury (Hg) ± SE (ng/g hair) and mean PCBs (as Aroclor 1260) ± SE (µg/L plasma) plotted against fish consumption frequency index (FCFI). Indices that do not share a common symbol are significantly different ($p<0.05$). Notes: FCFI 1 = less than one fish meal per month or none (n=35), FCFI 2 = less than one fish meal per week (n=14 for Hg; n=15 for PCBs), FCFI 3 = at least one fish meal per week (n=12), and FCFI 4 = more than two fish meals per week (n=9).



Because $\delta^{15}N$, mercury (Hg) and PCBs have been shown here increase significantly with fish consumption (i.e., FCFI) we plotted and log Hg and log PCBs (as Aroclor 1260) against $\delta^{15}N$ (Figure 3.4) to test for correlation between $\delta^{15}N$ and mercury and PCBs, both of which are often found in higher concentrations among high-frequency fish consumers. Significant positive correlations between $\delta^{15}N$ and mercury (log Hg vs. $\delta^{15}N$, $R^2 = 0.47$, $p<0.001$) (Figure 3.4A) and $\delta^{15}N$ and PCBs (log Aroclor 1260 vs. $\delta^{15}N$, $R^2 = 0.16$, $p<0.001$) (Figure 3.4B) were observed and this is further evidence that high $\delta^{15}N$ values are attributed to a high-frequency fish diet.

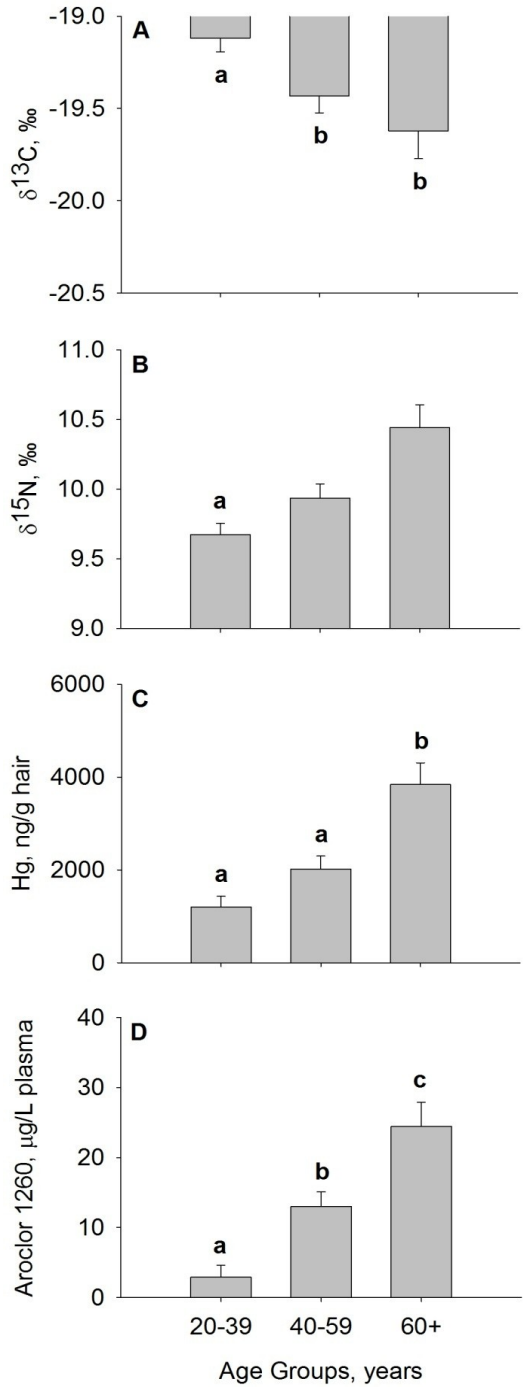
Figure 3.4: Log mercury (Hg) and log PCBs (as Aroclor 1260) concentrations in hair (ng/g) and blood (µg/L), respectively, plotted against hair $\delta^{15}N$ (‰) values for study participants (n=70 for Hg and n=71 for PCBs).



Age Effect

With differences in age observed between individuals in the HW1 and HW2 groups when compared to the LW1 and LW2 groups, respectively, participants were further separated into age groups (20-39, 40-59 and 60+ years) and mean $\delta^{13}C$, $\delta^{15}N$, mercury and PCBs levels were plotted for each of the three age groups (Figure 3.5). Here we observed a significant enrichment of ^{13}C among younger individuals aged 20 to 39 years of age (-19.12 ± 0.07‰) when compared to older individuals in age groups 40-59 and 60+ (-19.43 ± 0.09‰ and -19.62 ± 0.15‰) (Figure 3.5A, ANOVA, $p<0.05$). No difference in mean $\delta^{13}C$ was observed between age groups 40-59 and 60+. Mean $\delta^{15}N$ was significantly higher in individuals aged 60 years or older (10.44 ± 0.16‰) when compared to younger individuals in age groups 20-39 and 40-59 (9.67 ± 0.08‰ and 9.94 ± 0.10‰, respectively) (Figure 3.5B, ANOVA, $p<0.05$). No difference in mean $\delta^{15}N$ was observed between age groups 20-39 and 40-59. Similar to $\delta^{15}N$, mean mercury concentrations were significantly higher in the oldest cohort (3844 ± 463 ng/g) when compared to the younger 20-39 and 40-59 cohorts (1204 ± 228 ng/g and 2018 ± 283 ng/g, respectively) (Figure 3.5C, ANOVA, $p<0.05$). No difference in mean mercury was observed between age groups 20-39 and 40-59. PCBs (as Aroclor 1260) also increased with age with significantly different Aroclor 1260 concentrations observed between all age groups (20-39 years: 2.9 ± 1.7 µg/L; 40-59 years: 13.0 ± 2.1 µg/L; 60+ years: 24.5 ± 3.4 µg/L) (Figure 3.5D, ANOVA, $p<0.05$).

Figure 3.5: Mean $\delta^{13}C$, $\delta^{15}N$, mercury (Hg), and PCBs (as Aroclor 1260) values ± SE for 20-39 (n=37), 40-59 (n=24 for $\delta^{13}C$, $\delta^{15}N$ and Hg; n=25 for PCBs) and 60+ (n=9) age groups (years). Groups that share a common symbol do not differ significantly ($p<0.05$).



3.4 Discussion

3.4.1 Stable Isotopes

The traditional First Nations diet continues to be undermined by the access to low quality market foods in northern Canadian First Nations communities (Batal *et al.*, 2005). The high calorie market foods that are presently available in Kasabonika and Wapekeka are rich in carbohydrates and saturated fats with a significant proportion of foods derived from corn that are easily identified by their conspicuously high natural abundance of ^{13}C (Schoeller *et al.*, 1980; Jahren *et al.*, 2006), whereas a traditional hunter-gatherer diet consists of C_3 plant sources with C_3 plant sources having a lower natural abundance of ^{13}C. Many have advocated the use of carbon isotope ratios ($^{13}C/^{12}C$) as a means to differentiate dietary preferences/behaviours (O'Connell and Hedges, 1999; Petzke *et al.*, 2005a, 2005b; Jahren *et al.*, 2006). Our results indicate that it is possible to measure significantly higher carbon isotope ratios among low-frequency wild food consumers when compared to higher frequency wild food consumers that consume at least 1 wild food meal per day (HW1) or at least 1 wild food meal per week (HW2). However, discriminating between HW and LW food consumers when using a lower threshold of wild food consumption (at least 2 wild food meals per month vs. less than 2 wild food meals per month) using hair $\delta^{13}C$ was not possible here. The similarity of $^{13}C/^{12}C$ ratios among HW3 and LW3 food consumption groups may be due to the similar dietary behaviours regarding consumption of market foods by the majority of individuals in both groups. The fact that no differences in $\delta^{13}C$ were observed between participants from Kasabonika and Wapekeka First Nations is indicative of the similarity in market food products available between the two communities. The use of carbon isotope ratios as a reliable dietary marker may therefore be

problematic in First Nations communities where the frequency of wild food consumption is low, traditional foods are often complemented with market foods that are heavily enriched in ^{13}C, and where the variety of market foods is often limited. Wild food consumption in both Kasabonika and Wapekeka is often complemented with low quality store-bought foods that are heavily enriched in ^{13}C such as corn-based food products, HFCS sweetened beverages, and other foods derived from C4 plants, particularly among LW food consumers, and the similar $\delta^{13}C$ values among the, LW1, LW2, HW3 and LW3 groups is indicative of the significant influence of market foods on hair $\delta^{13}C$. Therefore, our results indicate that it is possible to discriminate between HW and LW food consumers using hair $\delta^{13}C$, particularly when using a higher threshold of wild food consumption such as one wild food meal per day or one wild food meal per week.

Nitrogen isotope ratios ($^{15}N/^{14}N$) have proven here to be a powerful dietary marker for wild food and fish consumption among individuals in these First Nations communities. This outcome corroborates several other studies that have shown hair $\delta^{15}N$ to track animal protein consumption among vegans, vegetarians, and omnivores (O'Connell and Hedges, 1999; Petzke *et al.*, 2005a, 2005b). Omnivores are feeding at the highest trophic level and thus have a greater enrichment of ^{15}N in their hair due to higher animal protein intake. Here, individuals consuming greater amounts of wild fish and hunted meats with significantly higher hair $\delta^{15}N$ values were significantly older (HW1 and HW2) than individuals consuming wild foods less frequently (LW1 and LW2). This is due to the fact that older individuals in these communities tend to consume wild foods in greater amounts, as a result of increased frequency, than younger individuals. With no age difference between the HW3 and LW3 food consumers, hair $\delta^{15}N$ values remained significantly higher among HW3 food consumers.

The high $\delta^{15}N$ values observed in the HW food consumption groups is likely attributed to the high consumption of fish among HW food consumers and their consequently higher trophic status. We observed that frequency of fish consumption, expressed as a FCFI, significantly influences $\delta^{15}N$ in hair. Enrichment of ^{15}N and mercury in hair, along with Aroclor 1260 in blood plasma, were all positively correlated with fish consumption. The significant positive correlations between $\delta^{15}N$ and mercury and $\delta^{15}N$ and PCBs observed here is further evidence that high $\delta^{15}N$ values are attributed to a high-frequency fish diet. Strong correlations between fish consumption frequency and $\delta^{15}N$, mercury and PCBs are also indicative of contaminant exposures linked to fish consumption. It is well established that mercury in hair is significantly correlated with fish consumption (Elhamri *et al.*, 2007) and many studies have used total mercury concentrations in hair as a reliable dietary marker of mercury exposure via fish consumption (Berglund *et al.*, 2005). The next chapter explores elevated contaminant concentrations including, but not limited to, mercury and PCBs among high-frequency wild food consumers.

Here, hair samples from older (60+ years) and middle-aged (40-59 years) individuals were depleted in ^{13}C relative to younger individuals (20-39 year); likely indicating higher corn-based sugar (market food) intake among younger community members. Enrichment of ^{15}N among older individuals implies higher fish consumption rates by community elders – consistent with interviews and food frequency questionnaires. Mercury in hair and Aroclor 1260 in plasma differ significantly among age groups, and both increase significantly with age. It is important to note that many of the older individuals tend to follow a more traditional diet and consume a greater amount and diversity of fish, along with organs by certain HW individuals, which would

likely result in higher mercury and PCB input rates than excretion rates and subsequent age-related accumulation.

The significant depletion of ^{13}C in the hair among the 40-59 and 60+ year age groups implies relatively higher wild food or C3 plant-based food intake by older individuals, whereas the significant enrichment of ^{13}C in hair among the youngest age group implies relatively less wild food consumption and higher C4 plant-based market food intake (market meats and corn-based and sugarcane-based food products). The distinctive $\delta^{13}C$ signature of the highly-consumed sugar sweetened soda (Jahren *et al.*, 2006) likely has much to do with the similarity in hair $\delta^{13}C$ between food preference/behaviour groups. Wilkinson *et al.* (2007) have similarly shown that known dietary differences between age groups in an Alaskan Native Yup'ik population were also reflected in stable isotopic differences of red blood cells.

The high reliance on fish and other wild foods by older individuals was also clearly shown by their elevated $\delta^{15}N$ and mercury in hair. This result corroborated other studies that show a higher $\delta^{15}N$ and mercury in hair in high-frequency fish consumers (both humans and animals). Kidd *et al.* (1995, 1998a, 1998b, 1999, 2001) showed significant correlation between $\delta^{15}N$ (trophic position) and mercury and organochlorine concentrations in freshwater food webs, indicating that $\delta^{15}N$ can be used to predict accumulation of contaminant concentrations in freshwater biota. These relationships between trophic position (as $\delta^{15}N$) and environmental contaminants have been demonstrated in many food web studies, but to our knowledge, the relationship between $\delta^{15}N$ and contaminants such as mercury and PCBs in humans or First Nations people has not been demonstrated. These results demonstrate the general feasibility of using $\delta^{15}N$ to predict accumulation of mercury and PCB s in human subjects.

3.5 Conclusion

Stable isotopes and environmental contaminants as dietary markers have been used widely in both human and animal dietary studies, however none have successfully applied these methods to assessing well-defined dietary behaviours in northern Canadian First Nations communities. With large differences in dietary behaviours observed within First Nations communities, analysis of stable isotopes and environmental contaminants in human hair and blood, can provide quantitative information about an individual's diet and can contribute greatly to Aboriginal health and nutrition studies by providing a "non-invasive, simple, yet powerful tool for monitoring dietary pattern" (Wilkinson *et al.*, 2007). Analysis of hair $\delta^{15}N$ can also contribute greatly to assessing benefits and risks of consuming traditional wild foods such as wild fish and hunted meats by predicting contaminant accumulation in individuals who consume greater amounts of wild food.

Current methods used to assess dietary intake often rely on self-reporting by participants and this has obvious limitations. Dietary assessment tools such as individual interviews, food frequency questionnaires, and dietary recalls are essential to understanding dietary behaviours, however they can be problematic due to, among other things, under-reporting (Vessby, 2000), pride/bias, cost, and time required of both participants and researchers (Becker and Welten, 2001; Kipnis *et al.*, 2003; Shai *et al.*, 2005; Subar *et al.*, 2003; Wilkinson *et al.*, 2008). Although dietary marker-based methods provide less qualitative information, they are relatively free of any bias (Willett, 1991) and can help to validate dietary surveys by identifying inconsistencies between tissue concentrations and dietary intake. Although dietary marker-based methods provide less qualitative information, they are considered to be objective and therefore important in validating dietary surveys by accounting for differences in dietary behaviours among

individuals and identifying inconsistencies between isotopic tissue composition and reported dietary intake (Willett, 1991). This is particularly important when conducting Aboriginal health and nutrition studies and benefit-risk assessments related to traditional First Nations dietary behaviours. Linking dietary behaviours to health risks is often a very difficult task, particularly in northern First Nations communities where there are numerous other health risk factors to consider. Further studies are currently underway to compare stable isotope ratios and dietary preferences with polyunsaturated fatty acids (PUFAs) data for these populations and to assess the use of stable isotope ratios and PUFAs for assessing the risks and benefits of wild food consumption. In summary, our study demonstrates that stable isotopes and environmental contaminants can be used as dietary markers when conducting nutritional studies in not only northern First Nations communities but also numerous other populations by providing quantitative information about an individual's diet.

4.0 Elevated contaminants in wild food consumers from two remote First Nations communities

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Timothy Andrew Seabert conducted the field study, sample preparation, mercury analysis, data analysis, and led the writing. Shinjini Pal and Eva M. Krümmel participated in the field study and contributed to the writing. Linda E. Kimpe led the analysis of organic contaminants in wild food samples and contributed to the writing. Michael A. Robidoux, François Haman, Pascal Imbeault, and Jules M. Blais proposed and designed the study, and contributed to the writing.

4.1 Abstract

Aboriginal peoples in Boreal environments rely on traditional wild foods as an affordable means to sustain healthy dietary practices. Here, we show a strong association between the frequencies of wild food consumption in two remote First Nations communities of Northern Ontario and environmental contaminants in blood and hair. We observed that persistent organic pollutants (POPs) in plasma and mercury in hair were typically 3.5-times higher among high-frequency wild food consumers than low-frequency wild food consumers, with many high-frequency wild food consumers exceeding health guidelines for polychlorinated biphenyl (PCB) and mercury exposures. These results challenge the viability of unrestricted traditional dietary choices in Boreal ecosystems, where individuals are being encouraged to consume greater amounts of wild foods for their proposed health benefits.

4.2 Introduction

Northern First Nations peoples are increasingly shifting away from local wild food sources and consuming greater amounts of modern, less nutritious, store-bought foods often high in caloric content, carbohydrates and saturated fats. This dietary transition has been identified as contributing to the high prevalence of obesity and type-2 diabetes among First Nations peoples (Young et al., 2000; Green et al., 2003). Land-based foods are an inseparable part of First Nations cultural identity, and are often cited as having important nutritional benefits that contribute to the reduction of obesity and obesity-related diseases (i.e., type-2 diabetes mellitus). As a result, the consumption of local wild food sources is often promoted as an important means of combating these deleterious health issues (Receveur and Kuhnlein, 1998; Kuhnlein *et al.*, 2001; Donaldson *et al.*, 2010).

Elevated contaminants in wild foods have been well documented in some aboriginal communities, particularly in northern Inuit communities, where elevated contaminant exposures are related primarily to the consumption of marine mammals (INAC, 2009). Environmental contaminants in land-based foods of remote northern Boreal regions are generally considered to be low, with the exception of mercury which can occasionally reach elevated concentrations, especially in aquatic biota residing in acidic, dystrophic lakes (INAC, 2009).

Here we report surprisingly elevated contaminants in individuals relying on wild foods in very remote Boreal environments of northern Ontario. Populations were divided into distinctive dietary groupings: those that drew extensively from local wild food sources versus those that relied more on store-bought foods (see Section 2.0).

4.3 Results and Discussion

Organic contaminant and lipid analysis was performed in plasma, and mercury was determined in hair of participants. No significant correlation between lipids and contaminant concentrations in plasma was observed likely because of low variability in lipid concentrations in plasma among study participants. The contaminant profiles of the two populations represent individuals who ate varying amounts of wild foods along with imported store-bought foods. Based on dietary data recorded through ethnographic observations, wild food consumption frequency index, 3-day dietary records and 24-hour recalls, 71 adult participants were separated into three categories of high-frequency wild food consumers (HW) and low-frequency wild food consumers (LW) to determine thresholds in which we observed differences in contaminant concentrations between HW and LW. We used a variable threshold for defining high wild food consumption (HW) and low wild food consumption (LW) in the 71 participants: Category 1:

HW1 (n=21), ≥1 wild food meal/day, LW1 (n=50), <1 wild food meal/day; Category 2: HW2 (n=24), ≥1 wild food meal per week, LW2 (n=47), <1 wild food meal per week; Category 3: HW3 (n=43), ≥2 wild food meals per month, LW3 (n=28), <2 wild food meals per month. We observed significantly higher ($p<0.05$) contaminant concentrations in HW1 participants when compared to LW1 participants for 9 of the 13 measured contaminants (Aroclor 1260, ΣPCBs, *p,p'*-DDE, mercury, oxychlordane, *cis*-nonaclor, *trans*-nonaclor, HCB and toxaphene parlar 50) (Table 4.1A) and the same was observed for HW2 when compared to LW2 (Table 4.1B). For the bi-weekly threshold, we observed more subtle differences with significantly higher ($p<0.05$) contaminant concentrations for only mercury, *cis*-nonachlor, HCB and toxaphene parlar 26 in HW3 when compared to LW3 participants (Table 4.1C).

Table 4.1: Differences in age-adjusted contaminant concentrations in plasma between **(A)** HW1 (≥1 wild food meal/day, n=21) and LW1 (<1 wild food meal/day, n=50), **(B)** HW2 (≥1 wild food meal/week, n=24) and LW2 (<1 wild food meal/week, n=47) and **(C)** HW3 (≥2 wild food meals per month, n=43) and LW3 (<2 wild food meals per month, n=28) groups. A bolded value indicates $p<0.05$ and an asterisk indicates $p<0.1$.

(A)

Contaminant	Age (years)			Wild Food Group		Age x Wild Food Group	
	DF	F ratio	p-value	F ratio	p-value	F ratio	p-value
log(Aroclor1260)	1	79.8664	**<.0001**	5.82	**0.0186**	0.1858	0.6678
log(ΣPCBs)	1	89.8342	**<.0001**	5.5745	**0.0211**	0.2111	0.6474
log(p,p'-DDE)	1	71.6829	**<.0001**	6.1706	**0.0155**	0.0066	0.9356
log(Hg)	1	7.8954	**0.0065**	8.7923	**0.0042**	0.0028	0.9578
log(Mirex)	1	85.154	**<.0001**	1.2924	0.2597	1.4335	0.2354
log(Oxychlordane)	1	82.751	**<.0001**	7.9827	**0.0062**	0.0031	0.956
log(cis-Nonachlor)	1	64.6522	**<.0001**	11.6338	**0.0011**	0.0624	0.8034
log(trans-Nonachlor)	1	94.0429	**<.0001**	7.8476	**0.0066**	0	0.9955
log(HCB)	1	35.8465	**<.0001**	6.0317	**0.0167**	0.3289	0.5682
log(β-HCH)	1	5.9173	**0.0177**	0.4346	0.512	0.9026	0.3455
log(PBDE47)	1	1.0590	0.3071	0.1153	0.7353	3.6430	0.0606*
log(Parlar26)	1	25.9891	**<.0001**	3.4618	0.0672*	5.1255	**0.0268**
log(Parlar50)	1	26.3738	**<.0001**	8.3051	**0.0053**	2.7292	0.1032

(B)

Contaminant	Age (years)			Wild Food Group		Age x Wild Food Group	
	DF	F ratio	p-value	F ratio	p-value	F ratio	p-value
log(Aroclor1260)	1	76.0078	**<.0001**	7.0191	**0.0101**	0.2065	0.651
log(ΣPCBs)	1	85.5539	**<.0001**	7.1863	**0.0092**	0.2353	0.6292
log(p,p'-DDE)	1	69.5366	**<.0001**	5.7693	**0.0191**	0.2581	0.6131
log(Hg)	1	7.5815	**0.0076**	9.9427	**0.0024**	0.0004	0.9832
log(Mirex)	1	83.2752	**<.0001**	1.9454	0.1677	1.8441	0.179
log(Oxychlordane)	1	79.0032	**<.0001**	6.7678	**0.0114**	0.0655	0.7988
log(cis-Nonachlor)	1	59.5557	**<.0001**	6.4662	**0.0133**	0.1947	0.6605
log(trans-Nonachlor)	1	88.7798	**<.0001**	6.0949	**0.0161**	0.0041	0.9494
log(HCB)	1	35.5356	**<.0001**	5.534	**0.0216**	0.6219	0.4331
log(β-HCH)	1	5.2445	**0.0252**	0.3212	0.5728	0.7853	0.3787
log(PBDE47)	1	1.5237	0.2214	0.0002	0.9899	1.9121	0.1713
log(Parlar26)	1	25.6276	**<.0001**	0.7068	0.4035	0.9541	0.3322

Contaminant	Age (years)			Wild Food Group		Age x Wild Food Group	
	DF	F ratio	p-value	F ratio	p-value	F ratio	p-value
log(Parlar50)	1	23.4182	**<.0001**	7.5532	**0.0077**	0.8642	0.3559

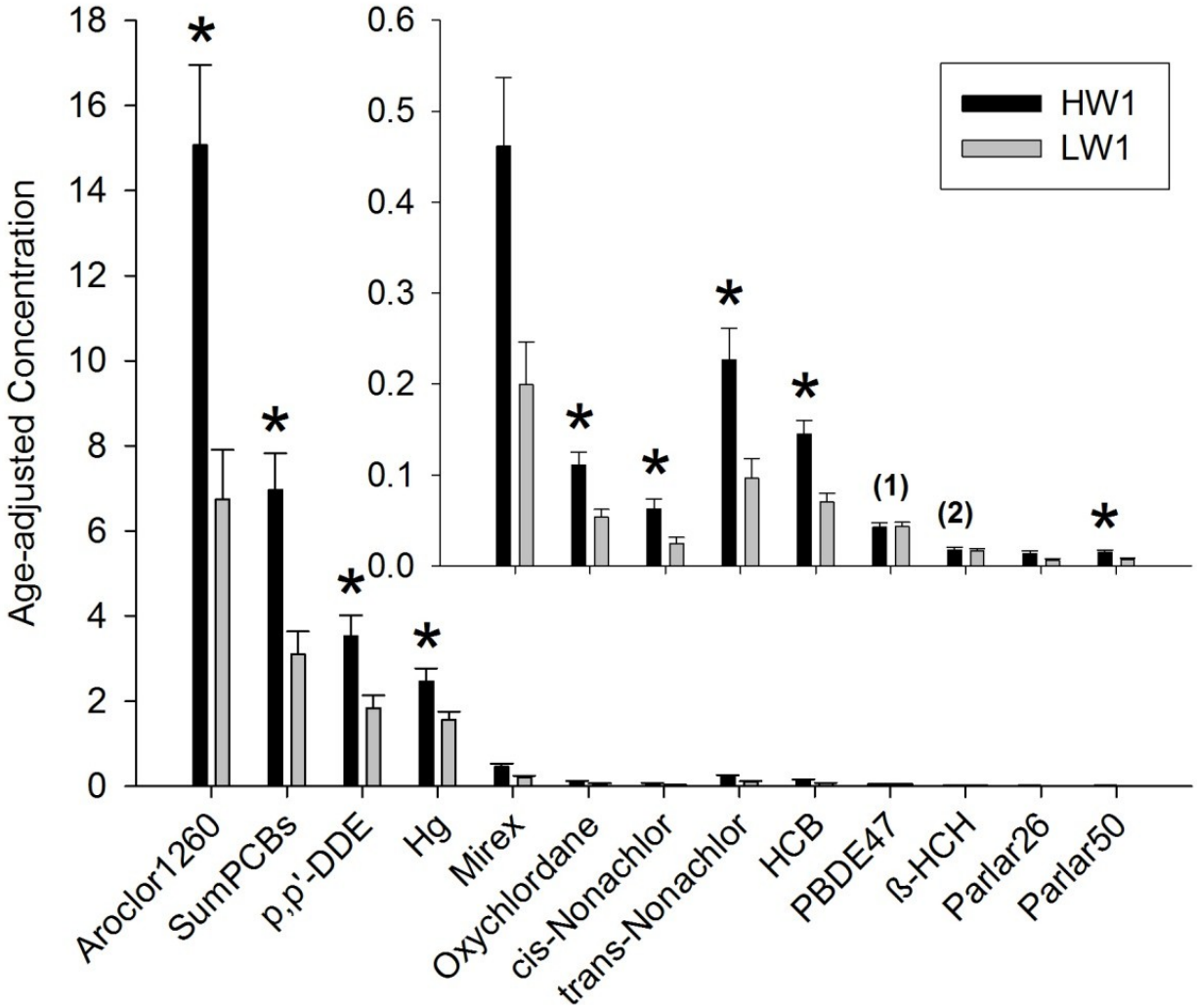
(C)

Contaminant	Age (years)			Wild Food Group		Age x Wild Food Group	
	DF	F ratio	p-value	F ratio	p-value	F ratio	p-value
log(Aroclor1260)	1	59.8763	**<.0001**	0.2866	0.5942	0.3248	0.5706
log(ΣPCBs)	1	68.8864	**<.0001**	0.1755	0.6766	0.5931	0.4439
log(p,p'-DDE)	1	51.1128	**<.0001**	0.0324	0.8577	0.0071	0.9329
log(Hg)	1	2.0172	0.1602	32.0232	**<.0001**	3.4779	0.0666*
log(Mirex)	1	85.6912	**<.0001**	0.0381	0.8458	6.2803	**0.0146**
log(Oxychlordane)	1	54.3588	**<.0001**	0.2897	0.5922	0.0674	0.7959
log(cis-Nonachlor)	1	34.6406	**<.0001**	8.1193	**0.0058**	1.6338	0.2056
log(trans-Nonachlor)	1	58.352	**<.0001**	1.2541	0.2668	0.2067	0.6508
log(HCB)	1	28.4698	**<.0001**	5.7079	**0.0197**	0.2882	0.5932
log(β-HCH)	1	2.6063	0.1111	0.1846	0.6689	1.3211	0.2545
log(PBDE47)	1	1.2075	0.2758	0.0736	0.7870	0.2859	0.5946
log(Parlar26)	1	9.695	**0.0027**	4.6164	**0.0353**	3.6801	0.0593*
log(Parlar50)	1	9.6142	**0.0028**	6.2602	0.0148	5.2936	0.0245

Aside from PBDE47, contaminant concentrations were significantly correlated to age ($0.12 \leq R^2 \leq 0.65$, $p<0.01$). After adjusting for the effect of age, mean mercury in hair and most POPs in plasma from daily wild food consumers (HW1) were significantly higher than those who did not consume wild food on a daily basis (LW1) (Figure 4.1, ANCOVA, p=0.021 to p=0.0011). Toxaphene parlar 26 concentrations were higher in HW1 with marginal significance (p=0.067) and we did not see a difference in PBDE47 (unrelated to age), Mirex and β-HCH concentrations between HW1 and LW1 (Table 4.1A). No differences in contaminant concentrations were observed among males and females (p-values ranged from 0.0994 for PBDE47 to 0.8655 for Aroclor1260), even after adjusting for age (p-values ranged from 0.1218 for β-HCH to 0.9247 for Aroclor1260). The uniform distribution of PBDE47 among HW and LW for all categories may be due to its ubiquity in indoor environments and its widespread presence in consumer products and household dust (Wu *et al.*, 2007).

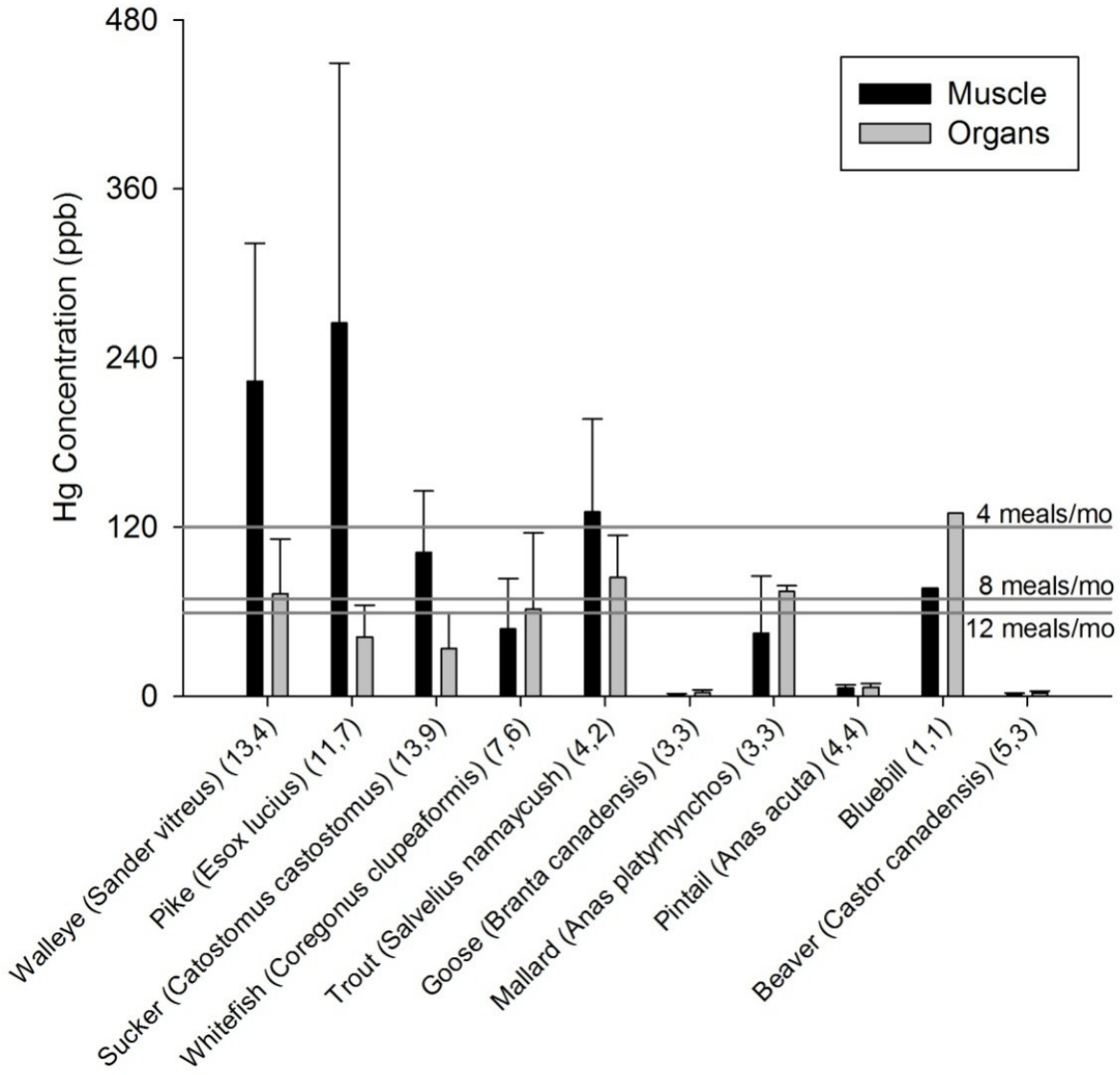
Mercury in hair was over 2-times higher in HW1 participants (2.9±0.3 μg/g; range: 0.85-6.91 μg/g) than in LW1 participants (1.3±0.2 μg/g; range: 0.03-8.63 μg/g) likely due to greater consumption of locally-harvested fish among individuals from the HW1 group. Mercury was significantly correlated with age for HW1 participants (log Hg vs. age; R^2=0.52, p=0.0002), however not in LW1 participants (log Hg vs. age; R^2=0.057, p=0.098). Age-adjusted mercury concentrations in the HW1 group were also significantly higher than the LW1 group (Figure 4.1, ANCOVA, $p<0.0015$).

Figure 4.1: Age-adjusted concentrations of POPs in blood (µg/L) and mercury (Hg) in hair (µg/g) for daily high-wild food consumers (HW1) (n=21 for POPs; n=20 for Hg) and non-daily low-wild food consumers (LW1) (n=50). Inset is an enlargement of the age-adjusted concentrations for the nine contaminants on the right side of the x-axis (Mirex through Parlar50). Data are presented as least square means ± SE. Notes: An asterisks (*) indicates a significantly higher ($p<0.05$) concentration, once adjusted for age; (1) PBDE47 concentrations were not adjusted for age; and, (2) indicates $p<0.1$.



We observed 95% of HW1 and 50% of LW1 participants exceeding the U.S. EPA's safety criterion (Rice *et al.*, 2003; NRC, 2000) of 1.0 μg/g mercury in hair. Mercury concentrations were sufficiently high in many locally-harvested fish and game from Kasabonika and Wapekeka (Figure 4.2) that HW1 participants from these communities often exceeded the U.S. EPA's monthly fish consumption limits for MeHg (U.S. EPA, 2000), assuming that more than 95% of mercury in fish was MeHg (Bloom, 1992). MeHg is a potent neurotoxin that can cross the blood-brain and placental barriers, harming the brain and nervous system, even at low exposure levels, particularly in the developing nervous system of a fetus or young child (Mergler *et al.*, 2007). Among the study populations, maternal hair mercury concentrations averaged 1.3±0.30 μg/g (range: 0.03-8.6 μg/g) with 48% of women of reproductive age (23-49 years) above 1.0 μg/g (n=29). PCB concentrations (expressed as Aroclor1260) were over 4-times higher in HW participants (19.7±4.0 μg/L) than in LW participants (4.8±0.7 μg/L). Aroclor1260 was significantly correlated with age for both HW (log Aroclor1260 vs. age; R^2=0.76, p<0.0001) and LW participants (log Aroclor1260 vs. age; R^2=0.43, p<0.0001) and age-adjusted Aroclor1260 concentrations were also significantly higher in HW1 than LW1 groups (Figure 4.1, ANCOVA, p=0.019). Health Canada's PCB guideline indicates that the 'Level of Concern' (LoC) for Aroclor1260 in maternal blood is 5 μg/L and the LoC for Aroclor1260 in the blood of men and post-menopausal women is 20 μg/L (Health Canada, 1986). Of the combined study populations, 43% of the HW1 participants and 2% of the LW1 participants were above the LoC of 20 μg/L. Mean Aroclor1260 among HW1 participants was 19.7±4.00 μg/L compared to 4.8±0.67 μg/L among LW1 participants. Maternal Aroclor1260 concentrations in blood averaged 3.7±0.73 μg/L, which surpassed the lowest-observed-effect-level for neurological development in children (Jacobson *et al.*, 1985; Jacobson *et al.* 1992).

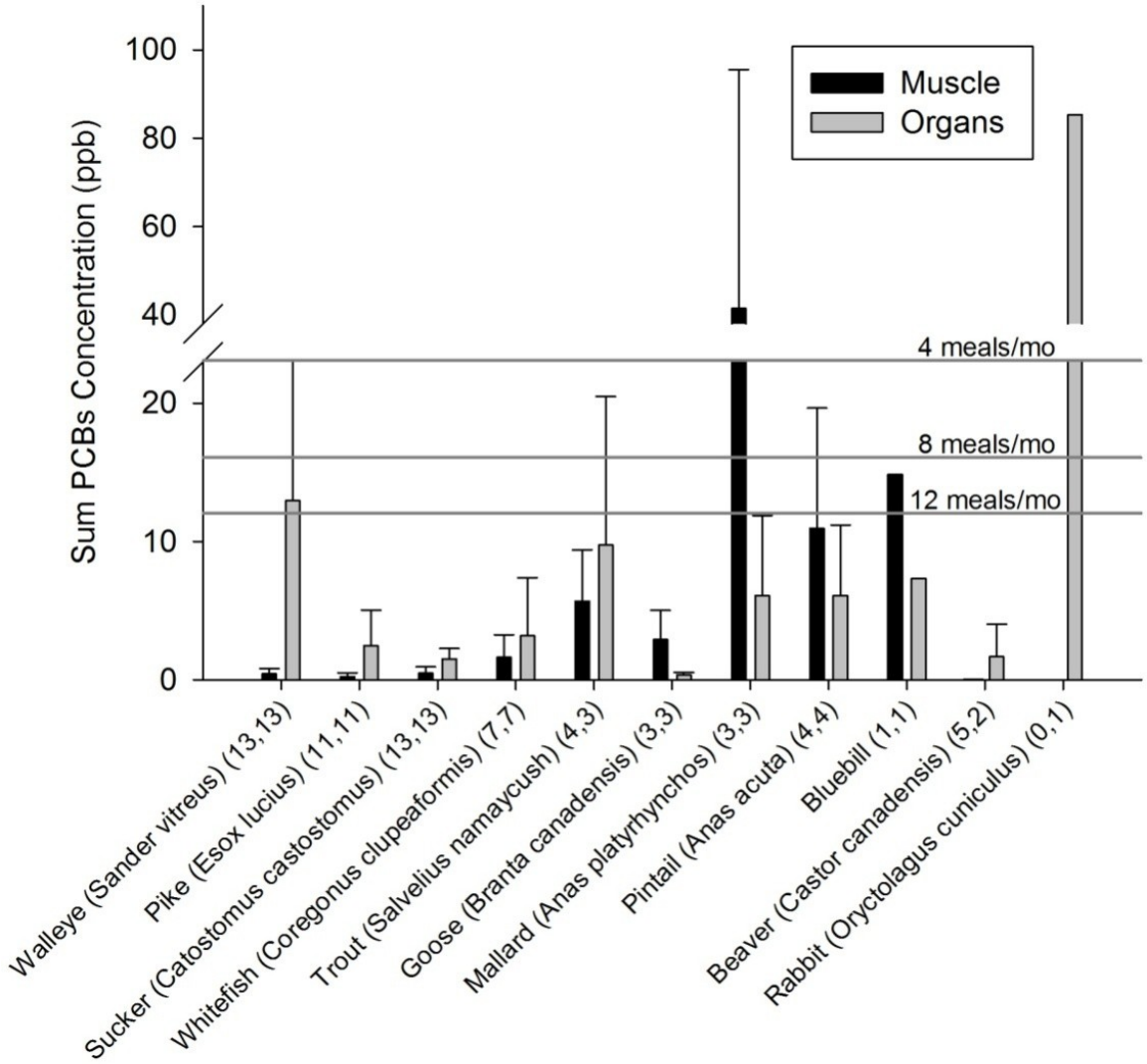
Figure 4.2: Mercury (Hg) concentrations (ppb or ng/g fresh weight) in muscle and organs of various locally-harvested wild foods from Kasabonika and Wapakeka regions. Data are presented as means ± SD. Sample sizes muscle and organ tissues are indicated in parentheses following of each wild food type, respectively. Horizontal lines indicate monthly fish consumption limits for MeHg based on the U.S. EPA's "Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories" (U.S. EPA, 2000).



Mothers and their newborns are particularly vulnerable to PCB exposures due to possible neurodevelopment deficits and other abnormalities resulting from prenatal exposures (Jacobson *et al.*, 1985; Jacobson *et al.*, 1992; Jacobson and Jacobson, 1996). The proportion of women of reproductive age above the LoC is particularly noteworthy because epidemiological evidence has shown that women with comparable PCB concentrations in cord serum gave birth to children with measurable developmental and neurobehavioral deficits (Jacobson *et al.*, 1992; Jacobson and Jacobson, 1996).

PCBs in locally-harvested fish and game from Kasabonika and Wapekeka were sufficiently high (Figure 4.3), that HW1 and HW2 participants from these communities exceeded the U.S. EPA's monthly consumption limits for PCBs (U.S. EPA, 2000). PCBs in locally-harvested rabbit (*Oryctolagus cuniculus*) and waterfowl species (*Anas platyrhynchos, Anas acuta*, *Aythya affinis*) were higher than piscivorous fish such as lake trout (*Salvelinus namaycush*) (Figure 4.3) suggesting the influence of local contamination source(s) such as landfills or refuse incineration practices which are widely used in these Boreal communities. All participants were below the WHO guideline of 200 μg/L *p,p'*-DDE in blood.

Figure 4.3: ΣPCBs concentrations (ppb or ng/g fresh weight) in muscle and organs of various locally-harvested wild foods from Kasabonika and Wapakeka regions. Data are presented as means ± SD. Sample sizes for muscle and organ tissues are indicated in parentheses following of each wild food type, respectively. Horizontal lines indicate monthly fish consumption limits for polychlorinated biphenyls (PCBs) based on the U.S. EPA's "Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories" (U.S. EPA, 2000).



4.4 Conclusion

PCBs in these populations often exceeded threshold levels for adverse effects, with potential consequences particularly for prenatal and early childhood development. Based on these findings, women of reproductive age and children should be assessed for dietary exposures to PCBs and advised about healthy wild food choices and frequency of wild food consumption. This study also highlights a need to increase efforts to reduce contaminant emissions to the environment, particularly for mercury and PCBs, and curb their transfer to remote northern environments. Mercury and POPs measured in fish from seemingly pristine regions south of these communities have been shown to be primarily the result of atmospheric deposition derived from anthropogenic sources (Wiener *et al.*, 2006) and recent increases in MeHg and PCBs in some aquatic food webs of northern latitudes are believed to occur as a result of recent climate warming (Carrie *et al.*, 2010; Macdonald *et al.*, 2005). Further research to investigate the presence of potential local contamination source(s) such as the local landfills and waste disposal sites in northern Boreal communities is also warranted.

Based on the contaminant concentrations measured here, the extent of contamination in locally-harvested wild foods may have serious implications for First Nations peoples who rely on wild foods and value their traditional lifestyles, particularly if individuals were encouraged to consume greater amounts of wild foods for their proposed health benefits. There is renewed interest in land-based food solutions as a cheaper, more nutritious alternative to market foods. On the surface, such solutions make sense because of potential beneficial effects associated with the consumption of wild foods, including higher percentages of energy as protein and micronutrients when compared to store-bought foods which contribute to dietary quality (Kuhnlein and Receveur, 2007).

The potential benefits associated with wild food diets must be considered within regional, cultural and environmental contexts. The tremendous diversity of food sources, modes of preparation, and food selection among First Nations/Aboriginal groups (Wilson, 2003; Willows, 2005), must be acknowledged if health benefits are to be assigned to wild foods. In this remote region of northern Ontario, a wild food diet is essentially animal-based, consisting primarily of freshwater fish, moose (*Alces alces*), beaver (*Castor canadensis*) and geese (*Branta canadensis*). Although wild berries and other edible plants exist in these study regions, ethnographic observations and dietary records indicated that wild edible plants made up a negligible proportion of the wild food intake for most participants. Dietary records indicated that the relative contribution of food sources as based on the four basic food groups (i.e., vegetables and fruits, grain products, meat and alternatives, and milk and alternatives) was similar among HW and LW participants. Upon further analysis of the meat and alternatives group, it was evident that HW participants simply replaced a portion of store-bought meat with wild fish and/or hunted meats, whereas LW participants consumed almost exclusively store-bought meats. Based on the existing portion of wild foods that comprised the diets of HW1 participants, and their relatively high contaminant body burdens compared to LW1 participants, the added risk of exposures to contaminants must be considered alongside any potential health benefits associated with wild food consumption as practised in these communities.

This study demonstrates that the benefits of local wild food consumption must be considered alongside the measurable risks associated with their regular consumption. Advocating the consumption of local wild food resources without acknowledging the regional, cultural and environmental diversities of contemporary First Nations populations potentially exposes individuals to health risks.

5.0 Overall Conclusions

Aside from the underlying social inequalities that are currently observed throughout the majority of Canada's First Nations communities, obesity and T2DM are major threats to the health of many northern Canadian First Nations people. There are approximately 700,000 First Nations people in Canada and about 2.4 million Native Americans in the United States, many of whom live on reservations that rely on locally-harvested wild foods. However, recent changes in dietary sources are believed to be causing significant increases in rates of obesity and chronic disease in some communities, which underscores a need to determine the viability and risks associated with different food sources. This dietary transition has not been uniform across First Nations communities. The degree of traditional wild food consumption varies greatly between communities and individuals. It is therefore very important to fully understand the specific dietary behaviours/preferences between communities and individuals with respect to both traditional wild food and store-bought food consumption when conducting dietary assessments while also understanding regional differences in wild food sources. Dietary markers such as $\delta^{13}C$ and $\delta^{15}N$ can contribute greatly to Aboriginal health studies and risk assessments by providing non-invasive, simple, yet powerful tools for monitoring dietary behaviours. Consideration should be given for their use when conducting dietary assessments and benefit-risk assessments of consuming locally-harvested traditional wild foods and imported store-bought foods. Analysis of hair $\delta^{13}C$ and $\delta^{15}N$ can provide quantitative information about an individual's diet and help to validate diet surveys by addressing error and bias. Further studies are currently underway to compare stable isotope ratios and dietary preferences with polyunsaturated fatty acids (PUFAs) data for these communities and to assess the use of stable isotope ratios and PUFAs for assessing the benefits of wild food consumption. Environmental

contaminants, which may also play a leading role in affecting health of First Nations communities, must also be considered in these assessments of the benefits and risks of consuming traditional wild foods.

In this study, we analyzed stable isotopes and contaminants in hair and blood collected from human subjects who rely to varying degrees on locally-harvested wild foods, and found a strong positive relationship between the frequency of wild food consumption and stable isotopes and contaminants in blood and hair. Our data show that contaminant exposure to those consuming country foods in remote Boreal ecosystems is comparable to those associated with serious health effects in industrialized areas, and the problem of contaminants in traditional foods is more widespread than any of the available literature would have led us to believe. Our results will dramatically affect our appreciation of contaminant exposures to First Nations peoples and will have implications for dietary choices, particularly if individuals were encouraged to consume greater amounts of wild foods for their proposed health benefits.

Contaminants in First Nations are known to be elevated in industrialized areas and in Inuit communities where contaminated marine mammals are consumed, but the two communities considered here are in remote terrestrial Boreal ecosystems with no industrial sources. This study highlights the importance of long range transport of contaminants, as well as the possibility of local contamination from landfills and waste burning, which are widely practiced throughout these remote northern First Nations communities. These results have broader significance for the hundreds of thousands of First Nations people living in remote Boreal ecosystems in Canada alone, and likely elsewhere. There are millions of Aboriginal peoples in Asia following similar practices.

We believe that our study will give needed attention to this important issue because the problem of contaminants in remote communities practicing traditional lifestyles is often underreported and underplayed.

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Table 1: Participant Data used for Statistical Analyses

Participant Code	Age (years)	Sex	Height (cm)	Weight (kg)	BMI (kg/m^2)	Waist (cm)	WFFI	FCFI	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	Mercury (ng/g)	Aroclor1260 (µg/L)	ΣPCBs (µg/L)	p,p'-DDE (µg/L)	Mirex (µg/L)	Oxychlordane (µg/L)	cis-Nonachlor (µg/L)	trans-Nonachlor (µg/L)	HCB (µg/L)	ß-HCH (µg/L)	PBDE47 (µg/L)	PBDE153 (µg/L)	Parlar26 (µg/L)	Parlar50 (µg/L)	Total Lipids (g/L)
K HW 00	36	F	165.3	78.4	28.6926	108.1	60	2	-19.02	10.23	1637.50	3.9	1.762	0.83	0.09	0.026	0.010	0.041	0.055	0.012	0.0253	0.01559	0.00097	0.00384	5.7
K HW 01	56	F	157	88	35.7012	106.9	80	4	-19.72	10.41	2113.51	28	12.6	8.6	0.67	0.17	0.099	0.340	0.21	0.024	0.04	0.00256	0.015	0.012	7
K HW 02	62	M	170	86	29.7578	109.5	80	4	-19.04	10.03	2422.77	30	16.99	13	1.1	0.18	0.074	0.360	0.28	0.027	0.03	0.03	0.016	0.017	8
K HW 03	60	M	177.5	66.8	21.2021	107	60	2	-19.28	10.26	2567.50	11	6.378	0.93	0.52	0.063	0.030	0.130	0.091	0.01	0.04	0.01662	0.00017	0.00247	4.2
K HW 04	73	M	171.3	104.3	35.5443	126.2	80	4	-19.95	10.04	2938.00	17	8.159	4.3	0.37	0.13	0.056	0.220	0.13	0.031	0.03	0.00233	0.011	0.012	6.4
K HW 05	68	F	159.2	89.7	35.3921	117.5	80	4	-20.05	11.04	2284.17	11	4.75	2.4	0.27	0.07	0.051	0.160	0.14	0.018	0.04	0.01497	0.012	0.014	6.8
K HW 08	72	F	161.5	94.2	36.1165	125.1	90	4	-20.34	9.68	4621.05	78	35.33	18	2.6	0.6	0.460	1.500	0.54	0.042	0.0244	0.048	0.12	0.12	8.7
K HW 09	66	F	162.5	65.8	24.9183	91.4	80	4	-19.35	11.18	5524.71	27	12.28	4.9	0.8	0.21	0.120	0.440	0.31	0.013	0.0198	0.03	0.028	0.031	6.9
K HW 10	57	F	149.5	79.8	35.7043	110.8	80	4	-20.15	10.56	4250.00	28	13.02	5	0.81	0.16	0.098	0.270	0.27	0.011	0.0144	0.04	0.017	0.015	4.9
K HW 11	58	M	181	104.2	31.8061	120.1	80	3	-19.56	10.95	3436.86	43	19.11	5.3	1.6	0.18	0.098	0.440	0.15	0.017	0.05	0.04	0.017	0.019	7.3
K HW 12	48	F	162	56.5	21.5287	81.9	60	3	-19.81	10.16	2476.86	18	8.187	4.9	0.37	0.1	0.031	0.160	0.14	0.018	0.03	0.01699	0.0057	0.0055	5.2
K LW 15	56	F	153	59.6	25.4603	90.1	10	1	-19.58	9.18	333.78	12	5.004	2.9	0.24	0.075	0.010	0.120	0.059	0.014	0.05	0.00225	0.00115	0.00212	6.2
K HW 17	52	M	172	84.6	28.5965	104	80	3	-19.74	9.27	1751.39	28	13.25	3.8	1.7	0.14	0.082	0.320	0.13	0.018	0.0191	0.03	0.011	0.014	5.9
K HW 19	55	M	168.5	100.8	35.5026	124.8	80	3	-19.54	10.65	2249.09	19	7.937	3	0.7	0.14	0.077	0.330	0.2	0.025	0.04	0.02	0.018	0.015	7
K HW 20	33	F	165.5	83.6	30.5218	118.8	60	2	-19.18	9.86	718.82	2.6	0.985	0.85	0.043	0.034	0.008	0.042	0.049	0.022	0.09	0.00103	0.00192	0.00471	6.6
K HW 21	36	M	175.5	89	28.8959	93.6	60	1	-19.88	9.59	730.65	4.4	2.023	1.3	0.13	0.036	0.010	0.060	0.055	0.018	0.0186	0.01065	0.00165	0.00134	6.6
K HW 22	36	F	165.5	85.5	31.2155	109.1	60	1	-20.04	9.41	533.71	0.38	0.203	0.1	0.028	0.005	0.004	0.006	0.019	0.003	0.0154	0.0116	0.00045	0.00411	6.3
K HW 24	38	F	165	109.8	40.3306	140.2	10	1	-19.22	10.57	2510.61	5.9	2.319	1.7	0.065	0.044	0.027	0.098	0.087	0.023	0.13	0.00628	0.0098	0.01	8.4
K LW 25	57	M	173.5	67.5	22.4236	88.45	30	1	-19.58	9.54	953.17	6.1	4.255	1.4	1	0.041	0.010	0.063	0.059	9E-04	0.0181	0.01858	0.00278	0.0045	6.6
K LW 26	49	F	164.5	87.2	32.2244	126.8	30	1	-19.23	10.25	1181.72	7.3	3.083	2.1	0.12	0.067	0.022	0.100	0.076	0.018	0.03	0.0159	0.0081	0.0052	8.3
K HW 27	42	M	172	83.7	28.2923	102	70	3	-18.66	10.19	1919.41	2.2	0.986	1	0.034	0.039	0.009	0.058	0.044	0.03	0.09	0.02	0.00409	0.00028	8.5
K LW 30	42	F	165	93.6	34.3802	109.6	40	1	-19.25	9.75	883.85	6.9	3.221	2.2	0.15	0.038	0.010	0.067	0.06	0.019	0.07	0.01405	0.00106	0.006	6.8
K LW 31	36	M	176	98.7	31.8634	119.5	10	1	-19.02	10.56	1239.71	2.3	0.918	1	0.035	0.029	0.020	0.059	0.047	0.015	0.05	0.00566	0.0056	0.0055	5.3
K LW 32	40	M	187.5	122.8	34.9298	124.3	20	1	-19.42	10.21	436.06	2.6	1.092	0.73	0.027	0.025	0.005	0.035	0.034	0.013	0.05	0.01862	0.00173	0.00122	5.5
K LW 33	32	M	176.8	125	39.9895	130	20	1	-18.54	9.78	347.61	0.51	0.201	0.72	0.005	0.022	0.003	0.028	1E-04	0.042	0.03	0.01627	0.00269	0.00027	7.3
K HW 35	34	M	181.5	123	37.3381	123.1	80	3	-18.38	10.24	3132.56	1.9	0.786	0.7	0.02	0.032	0.010	0.045	0.013	0.017	0.06	0.00416	0.00325	0.00496	5.6
K LW 36	37	M	175	89	29.0612	102.9	35	2	-18.52	9.76	340.77	1.3	0.673	0.45	0.065	0.02	0.001	0.024	0.044	0.015	0.12	0.01681	0.00033	0.00149	6.7
K LW 37	44	F	153.7	72.2	30.5625	102.4	5	1	-18.8	9.58	250.91	1.5	0.725	0.43	0.069	0.021	0.001	0.025	0.042	0.01	0.002	0.0033	0.00322	0.00429	5.8
K LW 39	39	F	160.5	108.8	42.2356	130	10	1	-18.88	9.52	284.29	3.9	1.716	1.4	0.065	0.031	0.009	0.042	0.025	0.021	0.08	0.0002	0.00341	0.00371	5.6
K LW 40	39	F	166.3	79.8	28.8548	103.3	10	1	-19.92	9.9	898.33	3.4	1.464	0.89	0.1	0.022	0.008	0.033	0.012	0.01	0.014	0.00123	0.00497	0.00453	5.8
K LW 41	46	M	176	89.2	28.7965	107.8	20	1	-18.8	9.88	607.71	13	5.807	2.3	0.48	0.096	0.026	0.180	0.054	0.013	0.08	0.03	0.0004	0.00477	10
K HW 43	33	F	174	101.4	33.4919	121.7	60	2	-18.67	9.76	938.18	2.1	0.889	0.53	0.046	0.019	0.007	0.030	0.031	0.011	0.0039	0.01449	0.0018	0.00352	5.9
K LW 45	30	F	162.5	78.3	29.6521	152.7	40	1	-19.12	9.68	465.00	0.38	0.165	0.2	0.009	0.007	0.001	0.009	0.007	0.005	0.05	0.00569	0.00294	0.00371	5.8
W HW 50	63	F	157.5	94	37.8937	125	65	2	-19.01	10.73	4289.69	5.5	2.344	2	0.14	0.1	0.070	0.190	0.17	0.018	0.07	0.01356	0.026	0.022	7.6
W HW 51	35	M	179	97	30.2737	111	65	1	-19.06	9.55	2001.56	5.5	2.076	2	0.067	0.064	0.021	0.094	0.058	0.014	0.0256	0.01894	0.00323	0.00191	6.9
W HW 52	42	F	162	84	32.0073	104	70	4	-20.34	10.85	8629.70	6.1	2.649	1.3	0.14	0.058	0.075	0.160	0.21	0.006	0.05	0.00583	0.015	0.016	5.5
W HW 53	23	M	194.5	150	39.6508	141	80	3	-19.19	10.12	1167.50	0.82	0.3	0.48	0.006	0.026	0.010	0.039	0.055	0.011	0.06	0.02	0.0025	0.00411	8.4
W HW 54	53	F	153.5	66	28.0109	102	75	2	-	-	-	21	8.636	6.6	0.51	0.077	0.037	0.140	0.1	0.021	0.06	0.00914	0.007	0.01	5.6
W LW 55	41	M	172	97	32.788	105.5	30	1	-19.14	10.19	1855.00	11	5.226	2.3	0.46	0.099	0.050	0.180	0.14	0.023	0.03	0.03	0.0098	0.013	7.4
W HW 57	33	M	182	97.5	29.4349	122.5	80	3	-19.04	9.39	1119.44	1.3	0.769	0.92	0.02	0.02	0.005	0.025	0.04	0.01	0.05	0.03	0.0014	0.00371	7.8
W HW 60	63	F	155	75	31.2175	116	70	2	-19.82	10.17	3039.19	6.6	2.879	2.3	0.21	0.062	0.043	0.120	0.11	0.018	0.04	0.00389	0.015	0.013	5.4
W HW 61	33	F	158	73	29.2421	110	70	1	-19.48	9.54	1386.36	4.3	1.65	1.3	0.053	0.037	0.018	0.072	0.06	0.012	0.11	0.01116	0.00307	0.00117	7.5
W HW 62	59	M	166.5	92	33.1863	108	85	3	-19.89	9.96	4100.54	21	8.575	3.9	0.49	0.21	0.130	0.470	0.25	0.014	0.1	0.00978	0.027	0.023	5.9
W LW 63	34	M	177	105	33.5153	119	20	1	-19.28	9.24	66.07	0.52	0.194	0.5	0.01	0.029	0.002	0.033	0.04	0.11	0.05	0.01163	0.0056	0.0069	4.2
W HW 64	45	F	167.5	83	29.5834	115	65	2	-19.21	10.37	2682.06	2.8	1.326	0.79	0.12	0.024	0.021	0.054	0.072	8E-04	0.04	0.00601	0.005	0.0053	6.8
W HW 65	38	F	158	90	36.0519	126	65	2	-18.74	8.81	553.71	0.46	0.256	0.2	0.02	0.006	0.001	0.010	0.032	0.007	0.0265	0.01385	0.00272	0.00181	8.3
W LW 67	46	M	172	105	35.4922	118	20	1	-19.17	9.7	693.42	3	1.423	1.7	0.048	0.037	0.008	0.050	0.03	0.025	0.04	0.01508	0.00432	0.00223	7.8
W HW 68	70	F	153.5	70.3	29.8359	112.5	95	4	-19.76	10.84	6907.75	34	17.89	11	1.2	0.37	0.170	0.680	0.46	0.044	0.03	0.03	0.022	0.029	6.5
W HW 70	27	F	168.5	103	36.2775	132	70	1	-18.84	10.22	609.00	0.71	0.309	0.36	0.004	0.01	0.005	0.020	0.013	0.011	0.04	0.01289	0.00397	0.00456	5.8
W HW 71	28	M	178.5	106	33.2682	119	70	1	-18.84	9.7	1390.29	0.83	0.392	0.28	0.02	0.01	0.004	0.010	0.021	0.004	0.0098	0.02	0.00276	0.00329	6.2
W HW 72	37	M	175.5	98	31.8179	110	80	2	-19.28	9.98	3654.85	4.7	2.149	1.4	0.081	0.055	0.026	0.098	0.11	0.014	0.0067	0.01054	0.00406	0.0079	8.8
W HW 73	36	F	168	100	35.4308	127.5	65	1	-18.58	9.52	1416.88	1.1	0.502	0.36	0.02	0.01	0.010	0.029	0.04	0.004	0.04	0.01171	0.00343	0.00223	7.6
W HW 74	33	F	162	85	32.3884	114	80	2	-19.92	9.65	1299.73	2	0.947	0.61	0.058	0.01	0.005	0.010	0.027	0.008	0.03	0.0025	0.00424	0.0018	6.2
W HW 75	30	M	173.5	101	33.5523	112	80	2	-19.45	9.56	1488.24	3.8	1.593	0.9	0.046	0.032	0.007	0.041	0.048	0.013	0.06	0.00276	0.00124	0.00192	6.3
W HW 76	50	M	166	97.5	35.3825	116	60	2	-19.58	10.07	2867.00	13	5.955	2.9	0.31	0.077	0.043	0.150	0.049	0.018	0.06	0.00769	0.016	0.016	5.7
W LW 78	37	M	176	86	27.7634	98	20	1	-19.17	9.84	2278.38	6.3	2.834	1.6	0.11	0.058	0.034	0.110	0.14	0.012	0.11	0.03	0.0058	0.0061	8.3
W HW 79	29	F	162	79	30.1021	104	65	2	-19.02	9.25	698.46	0.48	0.247	0.24	0.01	0.009	0.002	0.010	0.008	0.005	0.0203	0.00209	0.00439	0.00234	9.1
W HW 80	26	F	157.5	76	30.6374	113	75	3	-18.91	10.44	1218.05	1.9	0.928	0.65	0.034	0.02	0.001	0.020	0.032	0.008	0.011	0.00068	0.00147	0.00495	6.2
W LW 81	48	F	160.5	59	22.9035	86	5	1	-19.29	8.31	29.30	13	5.395	5	0.12	0.085	0.018	0.110	0.07	0.025	0.06	0.01393	0.00252	0.00071	7.8
W LW 82	23	F	162	72	27.4348	108	15	1	-19.27	9.33	1541.56	2.7	1.186	0.68	0.043	0.024	0.005	0.027	0.019	0.002	0.0015	0.00567	0.00067	0.00337	5.4
W LW 83	31	F	163	99	37.2615	134	20	1	-19.22	9.4	544.72	0.86	0.386	0.35	0.001	0.01	0.006	0.021	0.008	0.014	0.04	0.01049	0.00023	0.00098	5.1
W LW 84	40	F	161.5	85	32.5892	107	30	1	-19.16	9.93	1223.24	2.3	1.089	0.77	0.058	0.02	0.010	0.030	0.052	0.007	0.03	0.00243	0.00333	0.00268	5.7
W LW 85	35	F	161.5	90	34.5062	117	5	1	-19.4	8.96	135.00	2.7	1.229	0.54	0.03	0.035	0.008	0.037	0.02	0.019	0.0045	0.01782	0.00479	0.00255	11
W LW 86	39	M	169	90	31.5115	109	25	1	-19.63	9.94	1200.29	5.5	2.989	1	0.15	0.033	0.009	0.044	0.018	0.02	0.05	0.01696	0.00238	0.005	6.7
W LW 88	42	M	165	69	25.3444	97	30	1	-19.3	9.4	1414.86	5.7	2.821	2	0.17	0.093	0.037	0.160	0.073	0.026	0.04	0.02	0.0085	0.0092	6.8
W LW 89	32	M	178	78	24.6181	100	20	1	-18.54	9.05	523.89	0.4	0.229	0.25	0.003	0.005	0.002	0.009	0.013	0.003	0.04	0.00358	0.00314	0.00179	4.4
W LW 90	30	F	164	105	39.0393	125	15	1	-18.76	9.61	2356.86	3.2	1.351	1.1	0.058	0.03	0.010	0.044	0.041	0.018	0.03	0.00501	0.00148	0.0054	7.4
W LW 91	35	F	159.5	103	40.487	120	15	1	-19.84	9.83	1535.94	6.2	2.778	1.7	0.14	0.032	0.010	0.055	0.03	0.009	0.06	0.00936	0.00143	0.00298	5.8
W LW 92	25	F	156	62	25.4767	97	5	1	-18.49	9.13	131.62	0.5	0.202	0.31	9E-05	0.01	0.003	0.010	0.034	0.005	0.05	0.01975	0.00383	0.00411	6.5
W HW 93	53	F	152.5	100	42.9992	125	85	3	-19.9	9.85	4148.10	19	8.9	5.5	0.26	0.18	0.100	0.290	0.3	0.036	0.06	0.00029	0.036	0.043	7.8
W HW 94	52	F	153.5	86	36.4991	121.5	80	3	-19.44	9.21	2053.00	13	5.889	2.9	0.27	0.1	0.048	0.160	0.14	0.019	0.08	0.00748	0.012	0.014	8.6
W HW 95	25	M	175.5	97	31.4933	111	85	1	-18.44	9.57	847.92	4	1.589	1	0.037	0.024	0.020	0.052	0.043	0.003	0.05	0.01769	0.0003	0.00256	5.3

Table 2: Mercury and ΣPCB concentrations in muscle and organs from locally-harvested traditional wild foods from Kasabonika and Wapekeka

Contaminant in Tissues	n	Measured contaminant concentrations (ppb or ng/g fresh weight)												
Hg (ppb) in muscle														
Walleye	13	188.63	136.15	212.11	232.85	365.31	33.21	207.45	248.45	290.23	332.55	366.11	140.71	151.78
Pike	11	770.48	238.26	262.80	317.36	267.27	119.65	305.71	114.47	237.21	184.99	94.25		
Sucker	13	96.55	70.96	141.94	41.73	45.41	91.03	131.20	184.20	125.60	133.73	116.00	106.97	40.03
Whitefish	7	27.57	31.34	40.75	19.26	126.03	47.68	40.36						
Trout	4	55.86	213.00	109.01	144.76									
Goose	3	1.27	1.71	1.75										
Mallard	3	89.74	32.75	11.19										
Pintail	4	2.87	6.33	4.91	8.47									
Bluebill	1	76.71												
Beaver	5	0.68	0.96	2.62	0.55	0.71								
Hg (ppb) in organs														
Walleye	4	116.06	95.01	39.07	40.31									
Pike	7	72.47	14.37	33.99	24.51	72.45	42.31	33.95						
Sucker	9	52.12	12.63	18.84	12.27	18.67	74.53	74.27	20.29	20.78				
Whitefish	6	11.99	63.95	44.27	166.93	44.70	38.19							
Trout	2	105.48	63.03											
Goose	3	1.19	4.88	0.98										
Mallard	3	70.17	75.17	77.78										
Pintail	4	6.53	9.54	5.93	3.42									
Bluebill	1	129.68												
Beaver	3	3.60	2.23	1.06										
ΣPCB (ppb) in muscle														
Walleye	13	0.93548	0.23999	0.68366	1.07334	0.10571	0.16987	0.07198	0.10323	0.35738	1.06624	0.39335	0.18502	0.14705
Pike	11	0.24126	0.44486	0.44824	0.05813	0.89933	0.01017	0.01620	0.05356	0.08399	0.14598	0.14598		
Sucker	13	0.28653	0.16670	0.08045	0.07533	0.09201	0.15692	1.08635	0.85784	1.27073	1.10775	0.54258	0.37151	0.07516
Whitefish	7	0.43994	0.37504	2.07988	1.93310	4.97261	0.99169	0.55339						
Trout	4	0.13960	7.26987	7.81293	7.53224									
Goose	3	5.33814	1.58993	1.75172										
Mallard	3	11.83175	8.66109	103.84024										
Pintail	4	22.95979	11.57744	5.70411	3.52958									
Bluebill	1	14.83083												
Beaver	5	0.03813	0.02659	0.01592	0.00000	0.00000								
ΣPCB (ppb) in organs														
Walleye	13	23.1652	12.5972	7.9475	15.2048	0.5025	0.6977	0.1776	0.6993	37.9596	19.0429	24.5774		
Pike	11	1.8660	5.6630	1.8426	0.0000	5.8398	0.3022	0.4967	0.0000	7.2224	1.8996	1.8996		
Sucker	13	2.7502	0.9076	0.2715	1.5881	0.5035	1.7101	2.1647	1.5605	2.2284	1.2030	1.1674	1.0366	2.4992
Whitefish	7	1.1007	1.1193	1.0603	1.8794	1.3645	12.5705	3.0929						
Trout	4	1.2974	16.1154	21.5390	0.0000									
Goose	3	0.2149	0.3236	0.5360										
Mallard	3	11.7038	6.4176	0.1227										
Pintail	4	10.1182	10.8781	1.4991	1.7800									
Bluebill	1	7.3273												
Beaver	2	3.3187	0.0000											
Rabbit	1	85.2396												