Risk Assessment of Dietary Lead (Pb), Cadmium (Cd), and Mercury (Hg) Exposure among First Nations People in Ontario, Canada - a Total Diet Study and Probabilistic Assessment

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Abstract

This thesis quantified risks of lead (Pb), cadmium (Cd), and mercury (Hg) in the diet of First Nations peoples residing on-reserve in the province of Ontario, Canada. Data was obtained from the 2011-2012 First Nations Food, Nutrition, and Environment Study (FNFNES) and Health Canada to construct total diet studies and probabilistic assessments. Results indicated that the majority of the population is at low risk of exceeding the reference values for these contaminants. Average exposures of Pb and Hg were higher than the general Canadian population (1.7 and 1.6 times greater, respectively), whereas Cd was 59% lower than the Canadian average. The upper percentiles of the population exposure distributions were characterized for contributing food items to assist risk management strategies. For cadmium exposures, smokers had elevated exposures compared to non-smokers. Women of childbearing age had lower dietary MeHg exposures than the total population and were largely below the reference value.

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Table of Abbreviations

ATSDR	Agency for Toxic Substances and Disease Registry
CHMS	Canadian Health Measures Survey
CONTAM	The Panel on Contaminants in the Food Chain
EFSA	European Food Safety Authority
FFQ	Food Frequency Questionnaire
FNBI	First Nations Biomonitoring Initiative
FNFNES	First Nations Food, Nutrition, and Environment Study
IARC	International Agency for Research on Cancer
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
JECFA	The Joint FAO/WHO Expert Committee on Food Additives
PTDI	Provisional Tolerable Daily Intake
PTMI	Provisional Tolerable Monthly Intake
PTWI	Provisional Tolerable Weekly Intake
RfD	Reference Dose
SE	Standard Error
TDS	Total Diet Study
UNEP	United Nations Environmental Programme
WHO	World Health Organization

1. Overview

Modern society is dependent on many process and products that are possible only through advances made through industrialization. However, the history of industrialization has not occurred with an equal consideration of environmental impacts to economic and social gains. This has resulted in a legacy of environmental pollution and contamination that has, and will continue to have, impacts on the health and well-being of populations globally. The extent of the health impacts from these releases has yet to be fully realized as the array of chemicals, environments, and populations has a complexity that demands a focused approach that is encumbered by the rapid pace of progress. In 2012, the World Health Organization estimated that approximately 12.6 million deaths (23% of all deaths globally) were attributed to environmental sources which include pollution and contaminants (WHO, 2016). Depending on the properties of contaminants of concern and the characteristics of exposed populations, the evaluation of population health risks needs to be appropriately tailored to support holistic risk management and sustainable development.

Environmental contaminants such as lead (Pb), cadmium (Cd), and mercury (Hg) have a dichotomy in that although they are naturally occurring and released into the environment, they can also be released from anthropogenic sources. The assessment of the risk of these metals to human populations is therefore unique as they will always be present in the environment at some background concentration. Although they are naturally occurring, they carry a notable risk to the health of populations, as they are considered to be "non-essential metals" since they have no biological purpose or benefit. Exposures to metals can occur through inhalation, ingestion, and/or absorption at varying levels of effectiveness to enter systemic circulation in the human body and exert toxicity (ATSDR, 2012, 2007, 1999). In the context of environmental contaminants, exposures are characteristically chronic in nature (occurring throughout a lifetime) although low in concentration. For such contaminants, dietary assessments provide valuable insights on a baseline of exposure, which is an important step in characterizing the complexity of exposures from a population health perspective (WHO, 2005).

In Canada, the national health regulatory agency, Health Canada, conducts routine dietary assessments in the form of a Total Diet Study (TDS) for the general population to assess exposures to not only Pb, Cd, and Hg, but to other metals and organic pollutants (Dabeka and Cao, 2013). Other developed countries such as the United States, the United Kingdom, Belgium, France, Spain, and German conduct similar studies which are routinely contrasted to Canadian results, with findings integrated into risk management and regulatory decisions for protecting population health. The Canadian population is known for its diversity; however at a national assessment level, characterizing this diversity in subpopulations with strong cultures is unmanageable, especially in a dietary context. A population that is routinely excluded from national health assessment surveys for logistical reasons are the aboriginal populations living on reserves across the country.

The aboriginal peoples of Canada– the Inuit, Métis, and the First Nations, are a large and diverse population with multiple languages and traditional practices. First Nations are the most populous of the three, with approximately 850,000 individuals identifying as such, representing 60.8% of the Canadian Aboriginal population according to the National Household survey conducted in 2011 (Statistics Canada, 2013). First Nation peoples reside across Canada, however the majority are south of the 60th parallel North. The province with the largest populations is Ontario with 23.6% of all First Nations, followed by the western provinces of

British Columbia (18.2%) and Alberta (13.7%) (Statistics Canada, 2013). First Nations peoples may live on reserves, which are lands preserved for these populations established through the treaty system during European settlement of Canada (Waldram et al., 1995). In the province of Ontario the on-reserve population of First Nations was 34,548 in 2012 (Statistics Canada, 2013).

First Nations populations have strong cultural connections to the environment. This includes a subsistent orientation whereby communities have relied on ecozone adapted traditional food systems for survival (Waldram et al., 1995). Traditional foods represent cultural, nutritional, and economically important aspects of First Nations life (Johns et al., 1994; Kuhnlein and Chan, 2000). With the expansion of industrialization and resource extraction in remote parts of Canada, traditional practices are being mixed with increasingly accessible western culture and foods in aboriginal populations, the result of which has been a decline in traditional food consumption (Kuhnlein et al., 2004). Furthermore, the sensitivity of traditional foods to the accumulation of environmental pollutants has raised concerns over the safety of their consumption, further contributing to the decline of their consumption (Fitzgerald et al., 2004, 1999; Kuhnlein and Chan, 2000). In 2007, the First Nations Food, Nutrition, and Environment Study (FNFNES) was initiated as a 10-year, systematic project that aimed to collect a national baseline of representative data on the composition, quality, and contaminants in diets of First Nations peoples, with a focus on traditional foods (Chan et al., 2014).

This decline of traditional food consumption has been paralleled by a rise in the prevalence of chronic diseases such as diabetes, obesity, heart disease, and chronic kidney disease, all of which have strong links to dietary patterns (Dyck et al., 2012; Gao et al., 2007; MacMillan et al., 2003; Reading, 2015). In the Ontario FNFNES report, it was observed that 34% of the adult population was overweight, and 49% were obese (Chan et al., 2014). Obesity is a leading risk factor in the development of diabetes, which was self-reported in the FNFNES in 30% of adults (Chan et al., 2014). This is similar to previous reports of First Nation peoples health such as a study in Saskatchewan that observed the prevalence of diabetes to more than double in females to almost half the population, and tripled in males to approximately 40% of the population between 1980 and 2005 (Dyck et al., 2010). Diabetes has been cited as the leading cause of the higher prevalence of end stage renal disease in approximately 60% of cases in Aboriginal populations (Dyck, 2001). The prevalence of sever chronic kidney disease is significantly higher in First Nations populations than non-First Nations populations (Gao et al., 2007) which makes this another critical population health concern (Dyck, 2001; Gao et al., 2007; Samuel et al., 2014; Yeates and Tonelli, 2006). The disparities in health outcomes of First Nations populations have highlighted the need for focused prevention programs and initiatives.

Elevated levels of contaminants have been documented in many traditional foods and observed in biomonitoring sampling of aboriginal populations in Canada (Chan, 1998; Kuhnlein & Chan, 2000). Biomonitoring, where blood and urine samples are collected and analyzed, was also a knowledge gap in First Nations populations that was addressed by the 2011 First Nations Biomonitoring Initiative (FNBI). This study was designed to be directly comparable to Cycle 1 of the Canadian Health Measures Survey (CHMS) (La Corte and Wuttke, 2012). Results indicated exposures to cadmium to be significantly higher, while mercury exposures were similar although highly variable with the Pacific region and males from the Great Lakes regions to have significantly higher total mercury levels than the Canadian population, and lead exposures were significantly lower (Assembly of First Nations, 2013).

The purpose of this thesis was to determine:

1. What is the level of exposure of First Nations peoples living on-reserve in Ontario, Canada to heavy metals (Cd, Pb, Hg) from dietary sources?
2. How do dietary metal exposures in First Nations populations living on-reserve in Ontario, Canada compare to the general Canadian population and published literature in other indigenous populations?
3. What are the main food items contributing to exposures to these contaminants?
4. Do patterns of exposures and associated risk vary seasonally with respect to traditional food consumption?
5. How does the characterization of exposure magnitudes and sources differ within the province on an ecozone level (a grouping based on geographical and cultural population characteristics)?
6. Does the population of women of childbearing age (defined in this study as women between the ages of 19-50) have a different exposure and risk profile compared to the total study population?

To address these research questions, data collected from the FNFNES project was used as a primary source for the characterization of dietary patterns. FNFNES data for the province of Ontario was collected prior to the initiation of this thesis and the enrollment of Ms. Juric at the University of Ottawa. Ms. Juric participated in FNFNES data collection for the Atlantic Region of the study to gain insight on the process and methodologies while completing this thesis. Two complementary methods were used to assess the dietary intakes of metals in the study populations. The first was a Total Diet Study approach to quantify contaminant intakes from all dietary sources based on data collected using a 24-hour recall questionnaire. This method assessed the contribution to the dietary intakes of cadmium, lead, and mercury from both market food and traditional food consumption. The second was a detailed probabilistic exposure assessment using Monte Carlo simulations on traditional food consumption only, but it also included annual and seasonal investigations.

The results of this thesis are limited to the quantification of contaminant exposures from the diet. Exposures from occupational, residential, or other lifestyle practices are beyond the scope of this thesis. Furthermore, limitation in the information collected through the FNFNES project imposed constraints on the ability to characterize conclusions with respect to hunting and fishing practices. Information on the location where traditional food samples were collected/hunted/fished was a source of uncertainty in the data set and therefore was not included in the analysis. Information on the method of hunting/ fishing was not collected and therefore cannot be commented on in relation to contaminant exposures. All traditional food samples were collected in raw, uncooked forms, and therefore the impact of cooking practices was not reflected, and was assumed to be negligible on metal exposures.

This work contributes to the ongoing risk assessments of contaminant exposure in First Nations communities and will also be useful for the development of culturally relevant strategies to promoting traditional food use while reducing the body burden of heavy metals. Results of the exposure assessments may be used to provide comprehensive baseline data for communities involved in resource development or other contaminated sites issues including natural events such as impacts of forest fire. Comparison to the baseline levels established though the project could guide risk mitigation activities. Lastly, this work will serve as a pilot methodology for future analysis of contaminant data collected from other regions of Canada to answer national and regional questions. Environmental health researchers and health professionals will also benefit from the results of this study in terms of methodology and best practice in data interpretation.

2. Risk Assessment of Dietary Lead (Pb) Exposure among First Nations People in Ontario, Canada - a Total Diet Study and Probabilistic Assessment

2.1. Abstract

Scientific consensus on lead (Pb) exposures currently states that no threshold exists which is protective for health, therefore Pb exposures should be minimized to as low as achievable to protect population health. Among indigenous populations, the profile of Pb exposures is different than in general population, with higher contribution from traditional food including hunted wild games that can be contaminated with lead-containing ammunition. The objective of this study was to estimate dietary exposure to Pb among First Nations living on-reserve in the province of Ontario, Canada. A total diet study was constructed based on a 24-hour recall and lead concentrations from Health Canada and the First Nations Food, Nutrition, and Environment Study (FNFNES). A probabilistic assessment of annual and seasonal traditional food consumptions was conducted to provide a more detailed characterization of Pb exposures from traditional food consumption. Results were contrasted to exposures in the general Canadian population and estimates of increased population risk expressed as increases in systolic blood pressure were calculated. Results indicated traditional foods to be the primary contributor to the dietary Pb intake (73%), despite representing only 1.8% of the average caloric intake. The average dietary Pb exposure (0.21µg/kg/d) in the First Nations population in Ontario was 1.7 times higher than the dietary Pb exposure in the general Canadian population. The top traditional foods contributing to Pb exposures were moose and deer meat. Given the range of lead concentrations in traditional foods beyond previously characterized background levels, it is presumed that a diet which occasionally includes foods hunted with lead ammunition and shot puts the population at elevated risk of lead toxicity.

Key Words: Lead, Total Diet Study, Dietary Exposure, First Nations, Ontario, Probabilistic Modelling

2.2. Introduction

Lead is a well-studied environmental pollutant with both naturally occurring and anthropogenic sources (ATSDR, 2007). Historically, lead was widely used as an additive in common consumer items such as paints and gasoline as well as in plumbing infrastructure, which resulted in elevated exposures in populations globally (ATSDR, 2007). In North America, the level of lead in these items has been reduced and even been eliminated since the 1970’s due to stringent regulations (Nriagu, 1990; Thomas et al., 1999). Despite this decline, management of lead exposures remains a modern health priority in North America as well as other regions around the world (WHO, 2010a).

Epidemiological studies over the past 10 years have shown conclusive evidence of adverse human health effect despite declining magnitudes of chronic lead exposures in populations; this is most prominently observed with neurodevelopmental impairment among children (Canfield et al., 2003; Hu et al., 2006; Lanphear et al., 2005; Schnaas et al., 2005; Surkan et al., 2007). In adults, the strongest weight of evidence for adverse health effects is supported by the association between chronic low dose lead exposures and coronary heart disease characterized by increases in systolic blood pressure, and chronic kidney disease characterized by a reduction in the glomerular filtration rate (Kopp et al., 1988; Muntner et al., 2003; Navas-Acien et al., 2009, 2007). Reviews of the toxicological mechanisms and health effects of lead have been summarized by international agencies, including WHO (WHO, 1995), IARC (IARC, 2006), and ATSDR (ATSDR, 2007) with the current state of evidence for assessing population health risk reviewed by international organizations such as the Joint FAO/WHO Expert Committee on Food Additives (JECFA) and the European Food Safety Authority (EFSA) (EFSA, 2010; WHO, 2011). Generally, lead interferes with enzymes to disrupt homeostasis though mimicking essential elements such as calcium, magnesium, and zinc (ATSDR, 2007).

Currently, lead exposures in North American populations are predominantly in the inorganic form and come from dietary sources (ATSDR, 2007). In the general Canadian adult population, the average lead intake from dietary sources was 0.12µg/kg/d, as reported by the 2007 Canadian Total Diet Study (TDS) (Health Canada, 2007). One of the limitations of TDSs was that they are general screening tools unable to reflect the dietary exposures in populations with unique dietary compositions, such as the Aboriginal peoples. In Canada, over 1.4 million people identify as Aboriginal (First Nations, Inuit, Metis), representing approximately 4.3% of the Canadian population based on results from the 2011 National Household Survey (Statistics Canada, 2013). The largest group of Aboriginal peoples in Canada are the First Nations accounting for approximately 60% of the Aboriginal population. There are over 600 First Nation bands and communities representing over 50 nations (AANDC, 2015). The largest population of First Nations, more than 200,000 people and 21% of the total population, reside in the province of Ontario, Canada (AANDC, 2015). About 30% of First Nations peoples live on reserves (Statistics Canada, 2013). The diet of First Nations peoples is composed of a mixture of market foods and traditional foods which are obtained locally from the hunting of wildlife and harvesting of plants (Waldram et al., 1995), which makes it distinct from the diet of the general Canadian population.

Lead is found in many foods indirectly because of the ubiquitous nature as a pollutant and naturally occurring metal, however, in traditional foods there is also the potential for direct contamination through hunting with lead shots and ammunition (Bjerregaard et al., 2004; Johansen et al., 2001). The assessment of dietary exposures of lead in First Nations peoples to date has been focusing mainly on specific traditional food items in small population samples (Hanning et al., 2003; L. Tsuji et al., 2008; L. J. S. Tsuji et al., 2008). Although traditional foods are largely unregulated from a food safety and contaminant perspective, they are integral component of health and connectedness in Aboriginal populations on the levels of the individual, family, community, culture, and environment (Laberge Gaudin et al., 2015, 2014). Further benefits of traditional foods include greater nutrient density, increased physical activity during harvesting, and lower costs (Kuhnlein and Receveur, 1996). Aboriginal peoples globally are in a state of dietary transition away from traditional food systems while concurrently experiencing a rise in chronic disease prevalence (Kuhnlein and Receveur, 1996; Kuhnlein and Chan, 2000). Increased consumption of a westernized market food-based diet has taken the place of traditional foods because of availability and convenience whereas traditional food harvesting and preparation require time, skills, and hunting resources and knowledge (Kuhnlein et al., 2004; Laberge Gaudin et al., 2015). This shift has resulted in a diet of poorer nutritional quality, leading to significant increases in prevalence of obesity and diabetes (Johnson-Down et al., 2015; Kuhnlein et al., 2004; Schuster et al., 2011).

Given the importance of traditional foods and their potential for contributing to lead exposures, the characterization of the contribution to dietary sources of lead in First Nations populations is necessary for the development of strategic and culturally relevant risk management messages. Blood lead levels in Canadian First Nations population living on reserve reported by the 2011 First Nations Biomonitoring Initiative (sample size: 252) were similar to the general Canadian population as assessed through Cycle 1 (2007-2009) of the Canadian Health Measures Survey (sample size: 5319). Studies of lead exposures in Aboriginal populations globally have suggested associations between traditional food consumption and blood lead concentrations with varying certainty and explanations of population level variances (Dewailly et al., 2001; Hanning et al., 2003; Johansen et al., 2006). A common observation in adult Aboriginal population biomonitoring studies is males having higher lead exposure than females (Anticona et al., 2011; Dewailly et al., 2001; Laird et al., 2013a; L. Tsuji et al., 2008). This is often interpreted by a paralleled higher consumption of traditional foods among males (Van Oostdam et al., 2005). Although the available biomonitoring results showed that First Nations population have similar blood lead values to the Canadian population, this is not the case for other Aboriginal populations. For instance, the Inuit population of the western arctic had elevated mean levels ranging from 2.2 to 3.8 times higher for the total population and women of childbearing age subpopulation, respectively, compared to the Canadian population (Laird et al., 2013a).

Women of childbearing age and young children have been particularly studied because of the potential risk lead exposures may have on the developing brain. Inuit and Dene/Metis mothers from the Northwest Territories and Nunavut, Canada, were found to have significantly higher blood lead concentrations compared to Caucasian counterparts residing in the region (Butler Walker et al., 2006). Similar results were observed with Inuit mothers from the Inuvik and Baffin Island regions of the Canadian Arctic having elevated blood lead compared to Southern Canadian non-Inuit mother (Curren et al., 2014). A paired survey of blood lead concentrations and dietary patterns in Inuit women from Nunavik region in Northern Quebec observed age, smoking, and the consumption of waterfowl to account for 30% of the variability in blood lead concentrations (Dewailly et al., 2001). In a population of First Nations women in Northern Ontario, a strong correlation was observed between maternal and cord blood samples with total traditional food intakes (Hanning et al., 2003). Investigations into the sources of lead in blood samples through the use of isotopes has indicated lead hunting shot and ammunition to be the greatest contributor to blood lead concentrations in Inuit and First Nations populations (Fillion et al., 2014; Tsuji et al., 2008).

Given the multiple aspects of lead exposures in Aboriginal populations, especially First Nations where there is limited data, the characterization of lead exposures using a total diet approach provides insights that can be integrated into population health programs. Dietary lead intakes is particularly of interest as toxicological reference values for assessing dietary exposures have been revoked by regulatory agencies (EFSA, 2010; Health Canada, 2013a) given the consistent and evolving body of evidence supporting low dose effects. This has culminated in the general scientific consensus that no threshold for lead toxicity exists (WHO, 2011).

The objectives of this study were to (i) characterize and quantify sources of lead in the total diet of First Nations adults in Ontario; (ii) assess the health risk to the population using the new non-threshold approach; and (iii) identify the key contributing food items for lead exposures.

2.3. Method

2.3.1. Ethics

Ethics approvals were obtained from the Research Ethics Board of the University of Ottawa and Health Canada.

2.3.2. First Nations Food, Nutrition and Environment Study (FNFNES)

Dietary patterns and contaminant concentrations in traditional foods were obtained through the *First Nations Food, Nutrition, and Environment Study (FNFNES)* Ontario region results collected in 2011-2012 (Chan et al., 2014). The FNFNES was designed to study the diet of First Nations adults across Canada living on-reserve south of the 60th parallel. A total of 18 First Nation communities from the province of Ontario, were selected to participate based on a systematic random sampling method with probability proportional to the size of the community. Community selection was designed to be representative of the First Nations population in the region based on a combined ecozone/ cultural area framework. Three ecozones exist in the province of Ontario: the Boreal Shield, the Hudson Plains, and the Mixedwood Plains; and two cultural areas: Northeast and Subarctic. Using this framework, First Nations communities in Ontario were stratified into 4 strata: Boreal Shield/ Subarctic (Ecozone 1), Boreal Shield/ Northeast (Ecozone 2), Hudson Plains/ Subarctic (Ecozone 3), and Mixedwood Plains/ Northeast (Ecozone 4) (Figure 2-1). A minimum of 4 communities per strata, with a maximum of 6 were allowed. Participating household in each community were randomly selected with a target response rate of 100 household per community in all but two communities which had larger populations and a target of 200 households. At each household, one adult who met the following inclusion criteria was invited to participate: 19 years of age or older; able to provide written informed consent; self identifies as being a First Nation person living on-reserve in Ontario; and whose birthday was next. A total of 1,429 individuals participated.

All participating individual completed a household interview which included the following sections: a 24-hour dietary recall; traditional food frequency questionnaire (FFQ); socio/health/lifestyle questionnaire; and food security questionnaire. The 24-hour dietary recall was conducted through an in-person interview to record the types and quantities of all food and beverages consumed in the previous day. Interviews were conducted by trained community-based research assistants who employed the use of physical visual aids to assist participants in quantifying the amounts of foods and beverages consumed. For the FFQ, participants were asked to retrospectively recall the number of days on which they consumed each food item for the past four seasons, which were defined as 90-day periods. The FFQ Contained a total of 143 traditional food items categorized into 7 categories based on consultation with literature and community representatives. Categories with number of food items were: fish (29), land mammals (22), wild birds (25), wild berries (27), wild plant roots, grains, shoots, and greens (29), tree foods (9), and mushrooms (2). When participants recalled consuming traditional food items not explicitly listed in the questionnaire, open ended responses were recorded as “other” in the food category.

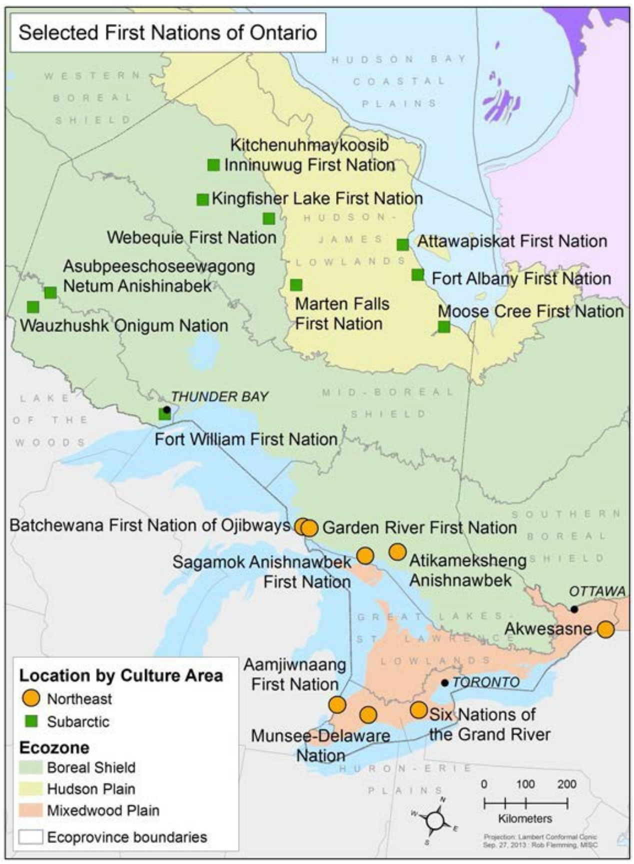


Figure 2-1 - Map of First Nations communities in Ontario participating in the FNFNES 2011-2012

2.3.3. Tap Water Samples & Analysis

In addition to the individual questionnaires, tap water samples were collected from a target of 20 participating households in each community through the FNFNES. Tap water samples were collected after water had been run for five minutes or until the stagnant water was flushed-out of the system. Prior to analysis, water samples were filtered through a 0.45 micron pore filter and digested using nitric acid based on EPA method #200.2. Inductively Coupled Argon Plasma Mass Spectroscopy (ICP/MS) was used to analyse lead concentration based on EPA method #200.8. For QA/QC, the percent recovery of certified reference material ranged from 87.7-106.7%, with the percent recovery of matrix spiked samples ranging similarly from 92.3-105%. The detection limit for lead was 0.005μg/L.

2.3.4. Traditional Food Samples & Analysis

Traditional food samples were collected from participating FNFNES communities based on community identified needs such as commonly consumed food, foods of importance for nutrition or environmental concerns, and foods known to accumulate higher concentrations of contaminants. Communities typically provided up to 30 composite food samples, with each composite comprising tissue from up to 5 replicates. A total of 419 composite food samples comprised of a sum total of 1237 replicates, representing 141 different traditional food items were analyzed for lead content. Lead content was analyzed from homogenized composite samples digested in an open vessel using a combination of nitric acid and hydrogen peroxide based on EPA 200.3/6020A. Inductively coupled plasma mass spectrometry (ICP/MS) was employed to quantify lead concentrations with a limit of detection of 0.004μg/g. Recovery of certified reference material ranged between 70-130%.

Lead concentrations in traditional food was compared to the CODEX alimentarius standard established by the FAO and WHO (Codex Alimentarius, 2015).

2.3.5. Total Diet Study

Total diet studies are conducted to assess the intakes of key nutrients and contaminants in a population (EFSA et al., 2011). Using data from the 24-hour recall survey, the intake of recalled food (expressed as grams per day) was multiplied by the average concentration of lead in that food item in the database of contaminant concentration to determine the dietary lead intake. Lead concentrations of market food were collected in 2011 and 2012 through the Canadian Total Diet Study by Health Canada (Health Canada, 2016). Concentrations of lead in traditional food items were obtained through samples collected through the FNFNES, which were analyzed in uncooked, raw states. Water consumption was included in the calculation of lead intakes as per harmonized total diet study guidance (EFSA et al., 2011), with concentrations in tap water represented by community-specific values measured by FNFNES. Contaminant concentrations values below the limit of detection were represented by an upper-bound approach to provide conservative estimate as the limit of detection varied between different data sources. Dietary lead intakes have been reported as weighted values based survey weights accounting for factors such as design weight (the inverse of the selection probability) and adjustment factors (non-response rate).

2.3.6. Risk Assessment of Dietary Intake

In adult populations, the adverse outcome of chronic lead exposure with the strongest weight of evidence as assessed by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) was an increase in systolic blood pressure (JECFA, 2010). Based on epidemiological evidence, JECFA concluded that a dietary exposure of 1.3μg/kg/d was associated with a 1mmHg increase in systolic blood pressure (JECFA, 2010). Dietary exposures in this study were compared to this exposure level using a margin of exposure (MOE) approach to characterize risk. MOEs less than 1 were considered to present an elevated population risk.

2.3.7. Traditional Food Probabilistic Exposure Assessment

A probabilistic approach was used to estimate lead exposure from annual traditional food consumption. This provided a detailed assessment that reflected the variability in the types and amounts of traditional foods consumed throughout the year and ability to identify and prioritize patterns of exposures in this dietary component. Monte Carlo simulations were constructed in Excel 2010 add-in Crystal Ball (Oracle; version 11.1.2.3). Lead intake for each iteration $_j$ ($\mu g \bullet kg^{-1} \bullet d^{-1}$) was modeled based on the sum of the product of the consumption of each food $_i$ *($g \bullet d^{-1}$)* by lead concentration $_i$ ($\mu g \bullet g^{-1}$), divided by body weight $_j$ (Equation 1).

Equation 1

$$\boldsymbol{Pb\ Intake_j(\mu g/kg/d)} = \sum_{\boldsymbol{i=1}}^{\boldsymbol{69}} \frac{[\boldsymbol{food_i}(\boldsymbol{g/d})]\ \boldsymbol{x}\ [\boldsymbol{Pb}]_{\boldsymbol{i}}(\mu\boldsymbol{g/g})}{\boldsymbol{Body\ Weight_j}(\boldsymbol{kg})}$$

Traditional food consumption distributions were derived from the FFQ conducted through the FNFNES. Consumption frequencies were converted into grams by applying age and sex specific serving size data for food groups reported through 24-hour recall responses. Daily consumption values in grams per day were computed by averaging intakes over a one-year period. Traditional food items were included in the simulation if consumption was reported in more than 5% of the population to limit input parameters with negligible bearing on simulation outputs. The total numbers of traditional food items included were 69 for the province, 47 in Ecozone 1, 63 in Ecozone 2, 41 in Ecozone 3, and 55 in Ecozone 4. Consumption data was parameterized using the custom distribution function in Crystal Ball as the sample data was representative of the provincial and ecozone populations of First Nations in Ontario. Input distributions of lead concentrations in each traditional food item was represented through FNFNES traditional food composite analysis fitted to lognormal distributions represented by the average, the standard deviation derived as an assumed coefficient of variation of 100%, and bounded by LOD/2 and three standard deviations. For traditional food items with no direct lead concentrations represented through the FNFNES samples, surrogate food items were selected from the data set based on trophic level and species similarities. Body weight data was obtained through the FNFNES and represented as an input through a custom distribution function. Simulations were constructed for the total provincial population, ecozone populations, women of child-bearing age subpopulation (n= 562), and seasonal simulations for the total provincial population. Simulations were run for 10 000 iterations with a Monte Carlo sampling method.

2.3.8. Statistical Analysis

JMP statistical software (version 12.1.0) was used to obtain summary statistics. Output distributions did not conform to assumptions of normality prior and after a logarithmic transformation. Non-parametric statistical tests were used to compare outputs. Tests of statistical differences utilized Kruskal-Wallis test, with significance considered as $p<0.05$.

2.4. Results

2.4.1. Total Diet Study

Based on responses to the 24-hour recall, 13% of the population reported consuming traditional foods. The northern-most ecozones, ecozone 1 and ecozone 3, had greater percentages (19.5% and 23.3% respectively) consuming traditional food compared to the province as a whole. These ecozones represented 37% and 33%, respectively, of the population reporting traditional food consumption. Traditional food sources represent 72.7% of the average lead intake for the population, while representing only 1.8% of the average caloric intake (Supplementary Table A-2).

The dietary lead dose from market food sources in the population who did not consume traditional foods is presented in Table 2-1. In non-traditional food consumers, market foods represented an average 96% of the lead dose for the entire provincial population, with the remaining 4% of the lead dose from tap water consumption. Compared to the average total dietary lead intake of the general Canadian population of 0.12μg/kg/d, the populations of First Nations who did not report traditional food consumption in the past 24-hours had lead intakes that were close to half of the Canadian population.

Table 2-1 - Summary of lead doses (μg/kg/d) from Market Food sources for non-traditional food consumers (n=1239).

	n	Mean Pb Dose (μg/kg/d)	Standard Error	50th	90th	95th	97.5th	99th
Province	1239	0.056	0.0010	0.049	0.095	0.11	0.14	0.18
Ecozone 1	289	0.056	0.0025	0.044	0.097	0.12	0.16	0.19
Ecozone 2	318	0.059	0.0018	0.053	0.095	0.12	0.14	0.18
Ecozone 3	204	0.050	0.0020	0.048	0.086	0.10	0.12	0.15
Ecozone 4	428	0.054	0.0016	0.050	0.097	0.12	0.16	0.20

In the population reporting traditional food consumption in the past 24-hours, market foods represented an average 3% of the lead dose for the province, with 97% of the total lead dose coming from traditional foods. The contribution of traditional foods to the lead dose in this sub regional populations ranged from an average of 93% in ecozones 2 to 97% in ecozones 1 and 3 (northern-most ecozones). The dietary lead dose from traditional food sources in the population with reported traditional food consumption in the 24-hour recall is presented in Table 2-2a. Ecozone 3 had the highest average lead dose from traditional food consumption; however, ecozone 1 had the widest range of lead doses.

The summary of lead doses from annual traditional food consumption, as calculated through a Monte Carlo simulation using consumption data from the FFQ is presented in Table 2-2b. Ecozone 3 has the highest mean lead intakes, which is consistent with findings from lead intakes computed through a total diet study approach (Table 2-2a). Dietary lead exposures expressed as a daily average from annual traditional food consumption (Table 2-2b) are lower than doses estimated through the total diet study (Table 2-2a) as the patterns of traditional food consumption in the later include low traditional food consumers, as well as fluctuations in seasonally available food items.

Table 2-2a - Summary of lead doses (µg/kg/d) from Traditional Food sources in traditional food consumers using data collected from 24-hr recall (n=190)

	n	Mean Pb Dose (µg/kg/d)	Standard Error	50th	90th	95th	97.5th	99th
Province	190	1.5	0.18	0.78	4.4	5.4	8.0	13
Ecozone 1	70	1.4	0.25	0.56	5.0	5.5	12	24
Ecozone 2	26	1.2	0.28	0.39	3.0	3.9	4.3	4.3
Ecozone 3	62	2.5	0.30	1.7	4.8	5.8	7.4	7.8
Ecozone 4	32	1.4	0.60	0.014	2.2	11	12	12

Table 2-2b - Summary of lead doses (µg/kg/d) from traditional food consumption using data collected from Food Frequency Questionnaire and simulated through a Monte Carlo Simulation. (n=10,000 iterations)

	Mean Pb Dose (µg/kg/d)	Standard Error	50th	90th	95th	97.5th	99th
Province	0.18	0.0047	0.051	0.44	0.73	1.2	2.1
Ecozone 1	0.21	0.0033	0.10	0.50	0.77	1.1	1.5
Ecozone 2	0.20	0.0054	0.035	0.48	0.93	1.5	2.5
Ecozone 3	0.24	0.0052	0.11	0.49	0.79	1.2	2.1
Ecozone 4	0.12	0.0054	0.0082	0.24	0.49	0.99	1.8

The average lead intake from market food sources was similar across the four ecozones, although statistically different between ecozones 1 and 2 and ecozone 1 and 4 (Wilcoxon each pair test, $p<0.05$). Community water sources contributed a minor portion of the lead intake, ranging from 0.11% in Ecozone 3 to 1.6% in Ecozone 4, although varied significantly across the ecozones (Kruskal-Wallis Test; $p<0.05$). For the lead dose from water, the only non-significant ecozone comparisons were between the two southern-most ecozones (ecozones 2 and 4) and the two northern-most ecozones (ecozones 1 and 3) (Wilcoxon each pairs test; $p>0.05$).

The top 5 market foods contributing to the total dietary lead dose are presented in Table2-3. For the province, the top 5 market foods reflected 9.8% of the average lead dose. In southern ecozones (ecozones 2 and 4), market foods contributed a greater proportion of the lead dose (16.9% and 13.3%, respectively). The top 5 traditional foods contributing to the total dietary lead dose are presented in Table 2-4a. The traditional food item with highest contribution to the lead dose for the province, as well as ecozones 1, 2, and 3 was moose meat, ranging from contributing 30% of the lead dose in ecozone 2 to 85.2% of the dose in ecozone 3. Deer meat was the second leading traditional food contributor to the lead dose, and the leading contributor in ecozone 4.

Table 2- 4b presents the top 5 traditional foods based on annual traditional food consumption, and therefore reflects seasonal availability, unlike table 4a which was based on a 24-hour recall conducted in the fall season. Despite the temporal differences, moose and deer meat remain the leading traditional foods contributing to the lead dose which suggests both methods capture similar leading sources of dietary lead. Other top foods not highly represented in Table 2-4a include goose, mallard and beaver.

Table 2-5 summarizes the lead concentration in five of the commonly consumed traditional food items, with comparison to Codex alimentarius guidance on the maximum level of lead in the meat of cattle, pig, sheep and poultry (without bones). The Codex threshold (0.1 ug/g) for lead was exceeded the in 24% to 63% of composite samples of these food items (moose and mallard, respectively) (Table 2-6).

Table 2-3 - Top 5 market foods contributing to the total Lead intake with mean dose (ug/kg/d), standard error (SE), and the percent contribution to the total Lead Dose

Province				Ecozone 1				Ecozone 2			
Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose
Coffee	0.0076	0.00024	3.6%	Soft drinks, canned	0.0062	0.00067	2.6%	Coffee	0.0098	0.00053	7.0%
Soft drinks, canned	0.0058	0.00029	2.8%	Coffee	0.0061	0.00039	2.5%	Soft drinks	0.0055	0.00046	3.9%
Luncheon meat, cold cuts	0.0029	0.00023	1.4%	Luncheon meat, cold cuts	0.0030	0.00050	1.3%	Luncheon meat, cold cuts	0.0033	0.00047	2.4%
Bread, white	0.0026	0.000099	1.2%	Cereal, oatmeal	0.0027	0.00035	1.1%	Bread, white	0.0031	0.00020	2.2%
Cereal, oatmeal	0.0018	0.00014	0.9%	Bread, white	0.0023	0.00018	1.0%	Cereal, wheat, rice, bran	0.0020	0.00031	1.4%
Total	0.21	0.024	9.8%	Total	0.24	0.046	8.5%	Total	0.14	0.025	16.9%

Ecozone 3				Ecozone 4			
Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose
Soft drinks, canned	0.0056	0.00052	0.9%	Coffee	0.0082	0.00049	5.1%
Coffee	0.0054	0.00047	0.9%	Soft drinks	0.0057	0.00048	3.6%
Bread, white	0.0030	0.00023	0.5%	Luncheon meat, cold cuts	0.0028	0.00039	1.8%
Pasta	0.0021	0.00044	0.3%	Bread, white	0.0026	0.00019	1.6%
Luncheon meat, cold cuts	0.0019	0.00031	0.3%	Cereal, wheat, rice, bran	0.0019	0.00035	1.2%
Total	0.61	0.094	3.0%	Total	0.16	0.047	13.3%

Table 2-4a - Top 5 traditional foods with mean lead dose (µg/kg/d), standard error (SE), and the percent contribution of the total dietary lead dose from TDS

Province				Ecozone 1				Ecozone 2			
Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose
Moose Meat	0.12	0.017	57.1%	Moose Meat	0.17	0.040	70.8%	Moose Meat	0.042	0.015	30.0%
Deer Meat	0.043	0.018	20.5%	Deer Meat	0.0095	0.024	4.0%	Deer Meat	0.032	0.020	22.9%
Canada goose	0.002	0.0014	1.0%	Trout	0.00071	0.00034	0.3%	Rabbit meat	0.00083	0.00064	0.6%
Trout	0.00031	0.00011	0.1%	Pickerel	0.00047	0.00014	0.2%	Moose Tongue	0.00040	0.00070	0.3%
Pickerel	0.00023	0.000055	0.1%	Whitefish	0.00038	0.00015	0.2%	Moose Liver	0.00024	0.00024	0.2%
Total	0.21	0.024	78.8%	Total	0.24	0.046	75.4%	Total	0.14	0.025	53.9%

Ecozone 3				Ecozone 4			
Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Pb Dose (µg/kg/d)	SE	% of Total Dose
Moose Meat	0.52	0.093	85.2%	Deer Meat	0.097	0.046	60.6%
Canada goose	0.035	0.014	5.7%	Moose Meat	0.0076	0.0050	4.8%
Caribou meat	0.0033	0.0012	0.5%	Maple Syrup	0.00037	0.00017	0.2%
Turkey meat	0.00055	0.00081	0.1%	Perch	0.00018	0.000098	0.1%
Whitefish	0.00019	0.00021	0.0%	Winter squash	0.000091	0.000047	0.1%
Total	0.61	0.094	91.6%	Total	0.16	0.047	65.8%

Table 2-4b - Top 5 traditional foods with mean lead dose (µg/kg/d), standard error (SE), and mean consumption (g/d) based on annual traditional food consumption as reported in the FFQ and modelled with Monte Carlo Simulation**.**

Province				Ecozone 1				Ecozone 2			
Food	Mean Pb Dose (µg/kg/d)	SE	Mean Intake (g/d)	Food	Mean Pb Dose (µg/kg/d)	SE	Mean Intake (g/d)	Food	Mean Pb Dose (µg/kg/d)	SE	Mean Intake (g/d)
Deer Meat	0.078	0.0040	1.6	Moose Meat	0.099	0.0021	10	Deer Meat	0.13	0.0049	2.6
Moose Meat	0.065	0.0020	6.7	Beaver	0.043	0.0020	0.77	Moose Meat	0.060	0.0022	6.3
Beaver	0.018	0.0020	0.37	Deer Meat	0.034	0.0014	0.70	Deer Liver	0.0033	0.00048	0.22
Goose	0.0078	0.00037	3.6	Goose	0.013	0.00040	5.2	Beaver	0.0015	0.00019	0.030
Mallard	0.0073	0.0010	0.44	Mallard	0.0087	0.00064	0.50	Mallard	0.00062	0.000099	0.030
Total	0.18	0.0047	38	Total	0.216	0.0033	61	Total	0.20	0.0054	32

Ecozone 3				Ecozone 4			
Food	Mean Pb Dose (µg/kg/d)	SE	Mean Intake (g/d))	Food	Mean Pb Dose (µg/kg/d)	SE	Mean Intake (g/d)
Moose Meat	0.11	0.0027	12	Deer Meat	0.11	0.0054	2.3
Beaver	0.045	0.0041	0.87	Moose Meat	0.0070	0.00046	0.75
Goose	0.028	0.00074	12	Black Bear	0.00065	0.000083	0.010
Mallard	0.026	0.00090	1.6	Beaver	0.00050	0.000059	0.010
Teal	0.0093	0.00057	0.55	Mallard	0.00045	0.000043	0.030
Total	0.24	0.0052	53	Total	0.12	0.0054	14

Table 2-5 - Concentrations of Pb with ranges in five common food items consumed across the province

	n (replicates)	n (composites)	Mean (SD)[a] ug/g	GM	Range ug/g	< Codex Threshold [b] Mean (SD) [a] ug/g	% of n	> Codex Threshold [c] Mean (SD) [a] ug/g	% of n
Beaver	30	11	9.4 (28)	0.044	<LOD - 99	0.017 (0.024)	73%	34 (56)	27%
Deer	42	10	4.5 (13)	0.032	<LOD - 42	0.035 (0.024)	70%	22 (24)	30%
Moose	83	17	0.87 (3.0)	0.022	<LOD - 13	0.013 (0.022)	76%	3.7 (6.2)	24%
Canada Goose	28	8	0.39 (0.48)	0.039	<LOD - 1.2	0.0027 (0.0014)	50%	0.78 (0.46)	50%
Mallard	26	8	1.6 (2.7)	0.25	<LOD - 8.5	0.032 (0.031)	38%	2.5 (3.4)	63%

< LOD included as LOD/2; LOD=0.004ug/g

b - Pb [ ] < 0.1ug/g

c - Pb [ ] > 0.1ug/g

The total dietary lead dose for the entire population, as well as the traditional food consuming and non-traditional food consuming portions of the populations are presented in Table 2-6 along with the most recent estimate of dietary lead exposure for the general Canadian population.

Table 2-6 - Summary of total lead doses (market food, traditional food, and water)

		n	Mean Pb Dose (µg/kg/d)	Standard Error	50th	90th	95th	97.5th	99th
Canada Total Diet Study 2007 [a]			0.12						
Total Population	Province	1429	0.21	0.024	0.056	0.15	1.6	2.8	5.0
	Ecozone 1	359	0.24	0.046	0.055	0.58	2.4	4.3	5.5
	Ecozone 2	344	0.14	0.025	0.057	0.11	0.18	1.1	2.9
	Ecozone 3	266	0.61	0.094	0.06	2.0	3.4	4.8	6.3
	Ecozone 4	460	0.16	0.047	0.053	0.11	0.14	0.2	1.8
Traditional Food Consumers	Province	190	1.5	0.18	0.82	4.5	5.5	8.0	13
	Ecozone 1	70	1.4	0.25	0.59	5.0	5.5	12.2	24
	Ecozone 2	26	1.2	0.28	0.47	3.1	4.0	4.4	4.4
	Ecozone 3	62	2.6	0.30	1.7	4.8	5.8	7.4	7.8
	Ecozone 4	32	1.4	0.60	0.076	2.2	12	12	12
non-Traditional Food Consumers	Province	1239	0.059	0.0011	0.052	0.097	0.12	0.15	0.18
	Ecozone 1	289	0.060	0.0025	0.048	0.099	0.12	0.16	0.19
	Ecozone 2	318	0.061	0.0019	0.055	0.097	0.12	0.14	0.18
	Ecozone 3	204	0.050	0.0020	0.049	0.087	0.10	0.12	0.15
	Ecozone 4	428	0.057	0.0016	0.052	0.10	0.12	0.16	0.20

a – (Health Canada, 2011)

2.4.2. Risk Assessment of total dietary lead exposure

The calculated mean (0.21µg/kg/d) and 95th percentile intakes (1.6 µg/kg/d) were below the previous PTWI from Health Canada (25µg/kg/wk) for total dietary lead exposure at the province, as well as for the ecozone level. In light of this reference doses being reviewed, we used an alternative margin of exposure (MOE) approach which is proposed by the European Food Safety Authority (EFSA) CONTAM panel for risk characterization of dietary lead exposures (EFSA, 2010). Lead exposures from the total diet study were compared to the exposure level (1.3µg/kg/d) associated with an increase of 1mmHg in the systolic blood pressure in adults as defined by JECFA (JECFA, 2010). Based on the total diet, the 95th percentile of the provincial population as well as in ecozones 1 and 3 showed a MOE of less than 1 (Table 2-7). However, the population subgroups of individuals who reported no consumption of traditional food in the past 24 hours showed a 20 times reduction in risk (with MOE higher than 1) compared to the population who reported traditional food consumption. This trend remains at the upper percentiles of the lead dose, as the margin of exposure was greater than 10 for the 95th percentile, with exposures at less than 10% of the exposure guidance value. In the subpopulation reporting traditional food consumption in the past 24 hours, all regions except ecozone 2 had a margin of exposure less than 1 between the mean total dietary lead dose and the exposure value, suggesting an elevated risk. For the province as a whole, the lead dose was associated with an estimated average increase in systolic blood pressure of 1.2mmHg, and up to 2mmHg for ecozone 3. At the 95th percentile of the subpopulation reporting traditional food consumption in the past 24 hours, the total dietary lead dose corresponded to an estimated 4.22mmHg increase in systolic blood pressure for the provincial population, with a regional range up to an increase of 8.9mmHg in ecozone 4.

Table 2-7 - Summary of Margin of Exposures (MOE) between total Pb dietary intakes (from 24-hour recall) and the exposure level of 1.3µg/kg/d associated with an increase of 1mmHg in the systolic blood pressure in adults as defined by JECFA. MOEs less than 1 indicate elevated exposure risk, and have been bolded.

		n	Mean	50th	90th	95th	97.5th	99th
Total Population	Province	1429	6.2	23	8.7	**0.81**	**0.46**	**0.26**
	Ecozone 1	359	5.4	24	2.2	**0.54**	**0.30**	**0.24**
	Ecozone 2	344	9.3	23	12	7.2	1.2	**0.45**
	Ecozone 3	266	2.1	22	**0.65**	**0.38**	**0.27**	**0.21**
	Ecozone 4	460	8.1	25	12	9.3	6.5	**0.72**
Traditional Food Consumers	Province	190	**0.87**	1.6	**0.29**	**0.24**	**0.16**	**0.10**
	Ecozone 1	70	**0.93**	2.2	**0.26**	**0.24**	**0.11**	**0.05**
	Ecozone 2	26	1.1	2.8	**0.42**	**0.33**	**0.30**	**0.30**
	Ecozone 3	62	**0.5**	**0.76**	**0.27**	**0.22**	**0.18**	**0.17**
	Ecozone 4	32	**0.93**	17	**0.59**	**0.11**	**0.11**	**0.11**
non-Traditional Food Consumers	Province	1239	22	25	13	11	8.7	7.2
	Ecozone 1	289	22	27	13	11	8.1	6.8
	Ecozone 2	318	21	24	13	11	9.3	7.2

Ecozone 3	204	26	27	15	13	11	8.7
Ecozone 4	428	23	25	13	11	8.1	6.5

2.4.3. Monte Carlo Simulation

To assess trends in lead intake as a function of seasonal traditional food consumption, inputs for the consumption distributions in the simulation were represented by seasonal average (fall, winter, spring, summer). Since seasonality influences the species availability, a moderate trend coinciding with peak hunting seasons was expected. Elevated lead doses were observed in the fall and winter seasons for the province, and at all ecozones but ecozone 3 which had lead intake peaks during the fall and spring/summer which is reflective of a fall game hunt and a spring bird hunt. Seasonal differences were statistically significant for each ecozone (Supplementary Table A-3).

Simulations for women of childbearing age resulted in slight, but significantly ($p<0.0001$) lower mean lead doses at the provincial level, as well as in ecozones 1 and 3 with differences of 0.027, 0.063, 0.102 μg/kg/d, respectively.

2.5. Discussion

Results of the TDS showed that there was a significant and substantial increase in the population risk profile when a small amount of highly contaminated food items were reported consumed in the dietary assessment. The majority of this increased population risk is attributed to traditional foods, particularly the high lead concentration in a number of commonly consumed terrestrial species. The wide variability of lead concentrations observed in these food items is likely the result of direct contamination from lead hunting shots and ammunition. Composite samples of beaver and deer contained the highest concentrations of lead; however, since composites represented pooled samples, and no information was available on the hunting method, it is not possible to definitively conclude lead ammunition or shot to be the source of these elevated concentrations.

Previous studies have observed terrestrial game, waterfowl, and birds hunted with lead to have elevates lead concentrations in homogenized tissue samples due to fragmentation of bullets and shots (Dobrowolska and Melosik, 2007; Hunt et al., 2009; Johansen et al., 2001; Rodrigue et al., 2005). Concentration of lead in moose, deer, and goose hunted with non-leaded shots and ammunition have been previously documented to have background lead concentrations below the Codex maximum contaminant level (Fachehoun et al., 2015; Horak et al., 2014; Tsuji et al., 2009). In water fowl, the background concentrations of lead is complicated to discern due to potential ingestion of legacy and stray lead shots may become a source of internal exposure to elevate tissue lead concentrations. The elevated lead concentrations observed in mallard samples in this study may be the result of internal distribution from such sources. In 1999 the Canadian government implemented a ban on the use or lead shots for hunting waterfowl and migratory birds within 200m of a water basin, which resulted in significant decrease in background concentrations of lead in many of the high-risk species (Stevenson et al., 2005). The impact of this ban was assessed in the Inuit peoples of Nunavik, Quebec who consumed avian food by comparing blood lead levels pre and post regulation to find significant decreases in cord blood samples of infants and blood levels in children and adults (Couture et al., 2012).

Since foods hunted with lead shots and ammunition are not necessarily contaminated, or if they are, often it is not in a uniform manner, the observed variation in the lead concentrations of food samples is likely reflective of the true dietary profile in foods that may be hunted with lead shot or ammunition. Results from the probabilistic assessments are in agreement with the conclusion of the total diet assessment in that the upper percentiles of the population distribution are at risk of adverse effect when traditional foods harvested by mixed method are consumed. Previous studies in Indigenous populations corroborate the conclusion that foods hunted with lead ammunition and shot elevates blood lead values, especially consumption of birds and waterfowl (Bjerregaard et al., 2004; Dewailly et al., 2001; Johansen et al., 2006). The consumption of game meats, primarily deer and moose, hunted with lead shots have also been associated with increased blood lead concentrations in general hunting populations (Iqbal et al., 2009; Meltzer et al., 2013). In comparison to blood lead levels in southern urban population, previous studies have indicated the sources of lead in the Inuit and First Nations populations to be predominantly from lead hunting shots and ammunition based on the analysis of isotopic ratios (Fillion et al., 2014; Hanning et al., 2003; Lévesque et al., 2003; L. J. S. Tsuji et al., 2008).

The risks of lead exposure from dietary items may be underestimated; since lead shots and ammunition can be ingested as whole pellets, and they may become an internal source of lead if lodged in the gastrointestinal tract or appendix (Carey, 1977; Madsen et al., 1988; Reddy, 1985; Tsuji and Nieboer, 1997). A study of Northern Ontario First Nations suggested approximately 15% of individuals have ingested lead shots lodged in abdominal regions based on a review of abdominal x-rays (Tsuji and Nieboer, 1997). Despite these considerations, national biomonitoring results from the 2011 FNBI indicate blood lead values in First Nations to be significantly lower than the general Canadian population assessed in Cycle 1 (2007-2009) of the CHMS (Assembly of First Nations, 2013; Health Canada, 2010). However, in comparison to CHMS Cycle 2 (2009-2011) results, FNBI blood lead levels appear to be in closer agreement with the Canadian population average although the significance of the trend is unknown and warrants further investigation (Health Canada, 2013b). Limitations in the FNBI include limited assessments of lifestyle or dietary parameters relevant to lead exposures and therefore limited profiling and insights on the traditional food consumption of participants is able to be concluded (La Corte and Wuttke, 2012).

A small-scale study of a First Nations population in Northern Ontario engaged in hunting and consuming birds harvested with lead shots found a significant increase in blood lead concentrations after the hunting season compared to individuals who had only consumed traditional food items, suggesting engagement in hunting activities increased lead exposures (L. J. S. Tsuji et al., 2008). The firing of lead shots and ammunition has been documented to increase blood lead burden through inhalation exposure of discharged dust and particulate. For instance, occupational exposures to firearm instructors and law enforcement officials have demonstrated significant increases in blood lead concentrations following the use of leaded ammunition compared to levels when non-leaded alternatives where used (Rocha et al., 2014; Tripathi et al., 1991, 1990). Similar findings have been reported for recreational shooter in both indoor and outdoor environments (Demmeler et al., 2009; Goldberg et al., 1991; Gulson et al., 2002). The quantification of lead exposure from hunting outdoors with lead ammunition and shots warrants additional study to characterize the risk of this activity.

Further to the direct human health risk implications of hunting with lead shots, lead ammunition is believed to be one of the largest emission releases of lead to land in Canada (Health Canada, 2013a). This has ecological implications as ingestion of spent lead shots is likely the leading cause of lead poisoning in waterfowl, avian, terrestrial game species (Rogers et al., 2012; Stevenson et al., 2005). Non-toxic alternative to lead shots and ammunition are abundantly available, however they come at an increased cost. In the state of California, USA, legislation was initiated requiring all hunting to be conducted with non-toxic ammunition and shots to primarily prevent the extinction of the Californian Condor, a predatory and scavenging raptor species (Church et al., 2006). Compliance with hunting ammunition and shot regulations are difficult to quantify and may be subject to reporting bias from surveying hunters, and the sale of ammunition and shots is a poor indicator due to the large market of recreational shooters who purchase large quantities of low-cost ammunition and shot. Given the documented adverse effects of low level lead exposure of IQ decline in children and a rise in blood pressure in adults, the costs of switching to non-toxic alternatives may be more economical than managing the costs associated with lead toxicity. Further study into the economic and health policy implication of this in the context of indigenous health is needed.

The prevalence of chronic diseases linked to dietary quality such as cardiovascular disease (CVD), diabetes, and obesity are significantly higher in many Canadian Aboriginal populations than the general Canadian population, with the rates increasing over the past decades. For instance, between 1980 and 2005, in a study of First Nations people residing in Saskatchewan the prevalence of diabetes more than double for females to almost half the population and tripled for males to approximately 40% of the population (Dyck et al., 2010). Elevated blood pressure (hypertension), like diabetes, is considered a risk factor for developing cardiovascular disease, of which coronary heart disease and stroke are the leading observed outcomes (Yeates et al., 2015). The prevalence of hypertension in North American First Nations populations was concluded to be approximately 33% in a systematic review by Foulds and Warburton (2014) which was lower than the observed prevalence of 43% in First Nations in Manitoba (Bruce et al., 2010), but higher than the average 22.6% observed by (MacMillan et al., 2003) in First Nations in Ontario. In Ontario, the rate of hospitalization for coronary heart disease increased for Aboriginal peoples during the 1980s and 1990s, while remaining stable and even declining in non-Aboriginal populations (Shah et al., 2000). Furthermore, in Ontario, hypertension was among the most reported health conditions the Ontario First Nations Regional Health Survey, with rated significantly higher than the comparable general population (MacMillan et al., 2003).

Based on the total diet study results, traditional foods are the primary dietary contributor to the lead intake in the First Nations population in Ontario. Compared to dietary lead exposure of the general Canadian population (Health Canada, 2011), the provincial First Nations population has an average dietary lead exposure 1.7 times greater. However, in First Nations individuals with no reported traditional food consumption in the past 24-hours, the total dietary lead exposures is approximately half of the Canadian average, whereas for traditional food consumers, the total dietary lead intake is more than 7 times greater than the Canadian average. Similar patterns were observed with mean lead intakes generated by the probabilistic assessment where levels were similar to the Canadian average in ecozone 4, but elevated between 1.5 to 2 times in the total province and ecozone 3, respectively. This is reflective of higher traditional food consumption in northern ecozones, whereas southern ecozones consume less traditional foods. The consumption of different types of traditional food items (i.e. game vs. bird vs. fish) has been hypothesized to be closely correlated complicating the identification of key contributing sources to the blood lead burden in subsistence populations. Across the province of Ontario, this study found a strong correlation between the consumptions of moose and Canadian goose ($\rho=0.6421$, $p<0.0001$). In regional ecozones, this correlation was strongly observed only in ecozone 1, ($\rho=0.6485$, $p<0.0001$). However, the inclusion of correlation coefficients for strongly correlated variables in the Monte Carlo simulation did not significantly change the output distribution of total lead intake, and therefore were not included.

Despite contributing to the lead intake, traditional foods are an important part of health in First Nations peoples. The FNFNES regional analysis indicated the quality of the total diet improves in terms of essential nutrient intake and a reduction in saturated fats, sugars, and sodium on days when traditional foods are consumed (Chan et al., 2014), consistent with findings from previous studies in Canadian Indigenous populations (Kuhnlein et al., 2004).

2.6. Conclusion

This study comprehensively quantifies the dietary lead exposure in First Nations people living on reserve in Ontario for the first time in a total diet context as well as more detailed characterization of annual traditional food consumption patterns. Results indicate the variability in lead concentration in traditional food items to be the most sensitive input and predictor of risk. Given the range of lead concentrations in traditional foods beyond previously characterized background levels, it is presumed that a diet which occasionally includes foods hunted with lead ammunition and shot puts the population at elevated risk of lead toxicity. Future studies on dietary lead exposures in this population should include an assessment of the hunting method as well as blood lead concentrations, and economic and social implications of a lead shot policy.

Appendix A- *Supplemental Tables*

Supplementary Table A-1 - Average amount of market and traditional food consumed (g/d). Mean, Standard Error (SE), and percentage of total are presented

		Market Food			Traditional Food			Water			Total	
	n	Mean (g/d)	SE	%	Mean (g/d)	SE	%	Mean (g/d)	SE	%	Mean (g/d)	SE
Province	1429	2271	30	85.1%	24	2.4	0.9%	373	15.8	14.0%	2668	32
Ecozone 1	359	2195	51	84.3%	36	5.6	1.4%	374	29.7	14.4%	2605	55
Ecozone 2	344	2194	58	83.4%	12	3.5	0.5%	425	37.3	16.2%	2632	66
Ecozone 3	266	2271	66	90.3%	78	12.0	3.1%	166	22.9	6.6%	2514	71
Ecozone 4	460	2406	62	86.2%	9	2.1	0.3%	375	27.7	13.4%	2791	63

Supplementary Table A-2 - Average calorie intake. Mean, Standard Error (SE), and percentage of total are presented

		Market Food			Traditional Food			Water			Total	
	n	Mean (KCal)	SE	%	Mean (KCal)	SE	%	Mean (KCal)	SE	%	Mean (KCal)	SE
Province	1429	1960	26	98.2%	36	3.7	1.8%	0	0.0	0.0%	1995	26
Ecozone 1	359	1969	49	97.4%	53	8.7	2.6%	0	0.0	0.0%	2022	49
Ecozone 2	344	1978	54	99.0%	19	5.5	1.0%	0	0.0	0.0%	1997	55
Ecozone 3	266	2050	63	94.7%	115	17.5	5.3%	0	0.0	0.0%	2164	63
Ecozone 4	460	1923	46	99.4%	12	2.8	0.6%	0	0.0	0.0%	1935	46

Supplementary Table A-3 - Summary of mean lead dose (µg/kg/d) with standard error across seasons.

	Fall	Spring	Summer	Winter
Province	0.24 (0.0068)	0.15 (0.0046)	0.16 (0.0045)	0.19 (0.0061)
Ecozone 1	0.26 (0.0056)	0.11 (0.0025)	0.20 (0.0035)	0.18 (0.0037)
Ecozone 2	0.29 (0.0098)	0.23 (0.0073)[a]	0.15 (0.0056)	0.23 (0.0073)[a]
Ecozone 3	0.32 (0.0069)	0.25 (0.0065)[b]	0.22 (0.0041)[b]	0.17 (0.0061)
Ecozone 4	0.15 (0.0068)	0.085 (0.0051)	0.12 (0.0054)	0.16 (0.0075)

a, b - Seasonal comparisons that did not differ with statistical significant based on Tukey-Kramer HSD; p>0.05

3. Risk Assessment of Dietary Cadmium (Cd) Exposure among First Nations People in Ontario, Canada - a Total Diet Study and Probabilistic Assessment

3.1. Abstract

Biomonitoring studies in First Nations populations in Canada have indicated elevated cadmium exposures compared to the general Canadian population. The primary routes of cadmium exposures are through smoking and diet. The objective of this study was to estimate dietary exposure to cadmium among First Nations living on-reserve in the province of Ontario, Canada. A total diet study was constructed based on a 24-hour recall and contaminant concentrations from Health Canada and the First Nations Food, Nutrition, and Environment Study (FNFNES). A probabilistic assessment of annual and seasonal traditional food consumptions was conducted. Results were compared to exposures in the general Canadian population and reference values established by the FAO/WHO. Results indicated market foods to be the primary contributor to the dietary cadmium intake (93%), with similar exposure magnitudes between regional ecozones within the province. The average dietary cadmium exposure in the First Nations population in Ontario (0.13μg/kg/d) is less than the general Canadian population (0.22 μg/kg/d), and below the reference value. However, when traditional foods were consumed, there was an elevation in the cadmium exposure that brought these individuals to an exposure of similar magnitude to the general Canadian population. The top traditional foods contributing to the cadmium exposure are the kidney and liver of large terrestrial game. This study also found that smokers had higher dietary cadmium exposures than non-smokers, although dietary exposures were below reference doses. If a heavy consumer of organ meats (upper 5^{th} percentile of Cd exposure; 1.7μg/kg/d) smoked 20 cigarettes per day, the PTDI would be exceeded 5.6 - 6.9 times, and this exceedance would be primarily attributed to cadmium exposures from smoking (contributing a Cd dose of 2.8-3.8μg/d).

Key Words: Cadmium, Total Diet Study, Dietary Exposure, First Nations, Ontario, Probabilistic

3.2. Introduction

Cadmium is a naturally occurring metal found as a co-contaminant in zinc, lead, and copper ores (UNEP, 2010). Although naturally released through processes such as weathering of rock, volcanic eruptions, and forest fires, anthropogenic activities can release cadmium to the air, water, and soils (ATSDR, 2012). According to arctic ice cores, the advent of industrialization marked the beginning of a trend of substantially increasing cadmium emissions until the 1960s when levels peaked (Boutron et al., 1991). Atmospheric releases are primarily from the mobilization as a by-product in non-ferrous metal refinement (i.e. zinc and copper extraction) and municipal waste incineration. Atmospheric releases eventually either dry or wet deposit onto soils and contribute to releases to land which also include mine tailings, residues from combustion, application of phosphate-based fertilizers, and also direct disposal of nickel cadmium batteries – one of the largest modern uses of cadmium (UNEP, 2010). Emissions of cadmium are a public health concern as cadmium can be readily taken up by plants from contaminated soils to enter into the food web with negative consequences for human and animal populations (ATSDR, 2012).

Cadmium is a non-essential metal which humans are exposed to primarily through oral ingestion and inhalation (ATSDR, 2012). Cadmium toxicity is most significantly and consistently documented for inducing adverse effects on the renal and skeletal systems (ATSDR, 2012). Cadmium is also considered a carcinogen based on observations of lung cancer following cases of elevated occupational exposures in refineries and nickel-cadmium battery manufacturers (IARC, 1993). Following ingestion, cadmium is primarily distributed to the liver and kidneys which account for more than half the total body burden in humans (ATSDR, 2012). Although cadmium does not undergo any degradation once absorbed, the ionic form readily binds to proteins such as metallothionein (Satarug et al., 2010). The cadmium-metallothionein complex is redistributed to the kidneys following complexation, where nephrotoxicity ensues as this bond degrades to release cadmium in the proximal tubual and result in proteinuria by creating lesions in tubular and glomeruli cells that restricts reabsorption of essential proteins (ATSDR, 2012; Satarug et al., 2010). The early signs of cadmium induced kidney dysfunction are observable in the urinary excretion of low molecular weight proteins, which precedes cadmium induced skeletal defects such as osteoporosis and decreased bone mineral density (Johri et al., 2010). Due to the long half-life of cadmium (20-30 years), biomonitoring may reflect different periods of exposure depending on the sample medium (ATSDR, 2012). The two common indices are blood and urine, with the former more reflective of recent exposures, and the latter of chronic exposures (Satarug et al., 2010).

In non-occupationally exposed and non-smoking populations, diet is the leading source of cadmium exposures (Järup et al., 1998; McElroy et al., 2007; Olsson et al., 2002; Satarug et al., 2009). In 2010, the Food and Agriculture Organization (FAO) and World Health Organization (WHO) Joint Expert Committee on Food Additives (JECFA) revised the exposure level for intakes of dietary cadmium that would be protective for populations. Previously, a provisional tolerable weekly intake (PTWI) was set at 7μg/kg/week, which represented the amount of cadmium that could be ingested weekly over a lifetime without appreciable health risk. The detection of β2-microglobulin (β2MG) was considered to be an early and sensitive biomarker of acute, yet reversible, kidney injury (EFSA, 2009). Relating the excretion of this protein to cadmium exposures resulted in a revised Provisional Tolerable Daily Intake (PTDI) guidance value of 0.8μg/kg/d. However, given the long half-life of cadmium, a provisional tolerable monthly intake (PTMI) of 25μg/kg was advised (WHO, 2011). The European Food Safety

Authority (EFSA) also re-evaluated the dietary intake guidance for cadmium, and advised a tolerable weekly intake value of 2.5µg/kg body weight (EFSA, 2009).

In population biomonitoring studies, cigarette smokers have significantly higher body burdens of cadmium than non-smokers, with dietary sources contributing a minor amount to the cadmium burden compared to smoking. In the Canadian Health Measures Survey (CHMS), smoking was observed to explain the greatest amount of variation in blood cadmium levels, and was the second leading contributing factor to urinary cadmium levels after age (Garner and Levallois, 2016). However, despite the CHMS being a national survey, First Nation peoples living on-reserve are not included in scope of this recurring biomonitoring assessment due to logistical constraints. In 2011, the First Nations Biomonitoring Initiative (FNBI) sought to address this gap by collecting biological specimens from First Nations adults across the country in a comparable survey to the CHMS (n=252) (La Corte and Wuttke, 2012). The study found cadmium levels in First Nations living on reserve in Canada to be significantly higher than comparable CHMS biomonitoring data, with blood cadmium two-fold greater and urine approximately 1.5 times greater (Assembly of First Nations, 2013). Within the FNBI results, the Great Lakes region, which included parts of the province of Ontario had among the highest average cadmium levels in both urine and blood results (Assembly of First Nations, 2013). Due to constraints in the FNBI sampling methodology and resources limited assessment was conducted on these results to characterize the determining factors of exposures in contrast to the general Canadian population.

First Nations are the largest Aboriginal population in Canada (Statistics Canada, 2013) with a rich culture that is distinct from the general Canadian population. Diet is one way in which the two populations differ, as First Nations consume locally obtained traditional foods from surrounding areas in additional to commercially available market foods. Relevant to cadmium exposures, the smoking rate in First Nations (51% in First Nations in Ontario self-reported smoking in the FNFNES (Chan et al., 2014)) is higher than in the general Canadian population (14.6%) (Reid et al., 2015). In recent years, a shift has been underway in the dietary composition of many indigenous populations globally, including the First Nations in Canada (Kuhnlein et al., 2004). As the diet has shifted towards more market-based food, there has been a paralleled rise in the rates of chronic health conditions such as diabetes and obesity (Haman et al., 2010; Kuhnlein et al., 2004; Philibert et al., 2009). A diet with traditional food consumption has been documented to be higher in nutrients, in addition to contributing to benefits from the cultural, social, and spiritual aspects associated with their collection, preparation, and consumption (Johns et al., 1994; Kuhnlein and Chan, 2000; Laberge Gaudin et al., 2014; Sheikh et al., 2011). Some traditional foods have been documented to have high cadmium concentrations, most notably the liver and kidney of large terrestrial game such as moose and caribou (Archibald and Kosatsky, 1991; Jin and Joseph-Quinn, 2004; Kim et al., 1998).

The province of Ontario has the largest First Nations population in Canada (Statistics Canada, 2013), however limited information is available on the dietary cadmium exposures in this population. Given the elevated biomonitoring results compared to the general Canadian population and the revised dietary guidance value for cadmium exposure, this study aims to assess and characterize the risk of cadmium exposure in the total diet including a detailed assessment of traditional food sources of adult First Nations peoples residing in the province of Ontario, Canada. Results of this study will support public health initiatives to reduce risks of cadmium exposures and serve as a baseline for future work in profiling cadmium exposures in this population.

3.3. Methodology

Details on ethics, the First Nations Food, Nutrition and Environment Study (FNFNES), total diet study methodology, and probabilistic risk assessment methodology are presented in Chapter 2 of this thesis.

3.3.1. Tap Water Samples & Analysis

The methodology for the collection of community specific tap water samples has been provided in Chapter 2 of this thesis. Prior to analysis, water samples were filtered through a 0.45 micron pore filter and digested using nitric acid based on EPA method #200.2. Inductively Coupled Argon Plasma Mass Spectroscopy (ICP/MS) was used to analyse cadmium concentration based on EPA method #200.8. For QA/QC, the percent recovery of certified reference material ranged from 95.8-106.8%, with the percent recovery of matrix spiked samples ranging similarly from 89-107%. The detection limit for cadmium was 0.005μg/L.

3.3.2. Traditional Food Analysis

The methodology for the collection of traditional food samples has been provided in Chapter 2 of this thesis. Cadmium content was analyzed from homogenized composite samples digested in an open vessel using a combination of nitric acid and hydrogen peroxide based on EPA 200.3/6020A. Inductively coupled plasma mass spectrometry (ICP/MS) was employed to quantify cadmium concentrations with a limit of detection of 0.001μg/g. Recovery of certified reference material ranged between 70-130%.

3.3.3. Risk Assessment of Dietary Intake

Dietary intakes of cadmium were compared to the provisional tolerable monthly intake (PTWI) set by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) of 25µg/kg, converted to a daily intake value of 0.8µg/kg/d to assess the population exposure risk.

3.3.4. Statistical Analysis

JMP statistical software (version 12.1.0) was used to obtain summary statistics. Cadmium exposure distributions were not normally distributed, and therefore non-parametric statistical tests were used to evaluate the significance of trends. Significance was considered to be $p<0.05$.

3.4. Results

The Total Diet Study (TDS) indicated that the majority of dietary cadmium exposures were derived from market food sources with 93% for the province and ranging between 88.6 - 99.5% at the ecozone level (ecozones 3 and 4, respectively) (

Supplementary Table B-1). The average cadmium exposures from the total diet, as well as the market food component of the diet did not significantly differ between ecozones, which indicate similar exposure magnitudes across the province. Furthermore, between traditional food consumers and non-traditional food consumers, the cadmium exposure from market foods did not significantly differ (

Supplementary Table B-2). However, cadmium exposures from traditional food significantly differed between ecozones (Kruskal Wallis test; $p<0.0001$) with significant difference in all ecozones comparisons (Wilcoxon each pair test; $p<0.0001$) except between the two northern-most ecozones (ecozones 1 and 3), and the between the two southern-most ecozones (ecozones 2 and 4).

Table 3-1 presents the total dietary cadmium exposures for the population, as well as for the subpopulations based on reported traditional food consumption. For the entire population, the average dietary cadmium exposure was slightly less than half of the general Canadian population. However, in traditional food consumers, cadmium exposures are closer to that of the general Canadian population. The significance of these trends is unknown and presents a limitation in the analysis.

Table 3-1 - Summary of total dietary cadmium intake (µg/kg/d) from all sources based on consumption data from the 24-hour recall. Results presented with population weights applied. n=1429

		n	Mean Cd Dose (µg/kg/d)	SE	50th	90th	95th	97.5th	99th
Canadian Total Diet Study 2007[a]			0.22						
Total Population	Province	1429	0.13	0.0043	0.11	0.25	0.32	0.38	0.54
	Ecozone 1	359	0.14	0.0087	0.11	0.23	0.33	0.40	0.66
	Ecozone 2	344	0.14	0.013	0.11	0.26	0.36	0.46	0.59
	Ecozone 3	266	0.14	0.0058	0.11	0.27	0.33	0.36	0.39
	Ecozone 4	460	0.12	0.0047	0.10	0.24	0.31	0.38	0.54
Traditional Food Consumers	Province	190	0.20	0.028	0.15	0.33	0.37	0.44	2.4
	Ecozone 1	70	0.20	0.040	0.13	0.27	0.37	0.82	2.3
	Ecozone 2	26	0.34	0.16	0.16	0.35	2.7	3.9	3.9
	Ecozone 3	62	0.19	0.012	0.19	0.34	0.37	0.46	0.52
	Ecozone 4	32	0.14	0.022	0.13	0.25	0.44	0.54	0.54
Non-Traditional Food Consumers	Province	1239	0.13	0.0030	0.10	0.24	0.31	0.38	0.53
	Ecozone 1	289	0.13	0.0069	0.10	0.22	0.31	0.41	0.66
	Ecozone 2	318	0.13	0.0060	0.11	0.26	0.34	0.46	0.58
	Ecozone 3	204	0.12	0.0062	0.095	0.22	0.27	0.33	0.37
	Ecozone 4	428	0.12	0.0047	0.10	0.24	0.31	0.38	0.53

a - (Health Canada, 2011)

Table 3-2a presents the cadmium dose from traditional foods in traditional food consumers. Cadmium exposures from traditional food differed significantly between ecozones 1 and 3, and ecozones 2 and 3 (Wilcoxon each pair test; $p<0.05$). Table 3-2b presents the summary of cadmium exposures from traditional foods based on annual consumption patterns as collected through the FFQ. Patterns of cadmium exposures from traditional foods varied between the two methods, largely due to the types of foods captured in both methods.

Table 3-2a - Summary of dietary cadmium exposures (µg/kg/d) from **traditional food sources** in traditional food consumers based on 24-hour dietary recall data. n=190

	n	Mean Cd Dose (µg/kg/d)	SE	50th	90th	95th	97.5th	99th
Province	190	0.084	0.027	0.025	0.12	0.15	0.20	2.3
Ecozone 1	70	0.094	0.040	0.031	0.14	0.15	0.68	2.2
Ecozone 2	26	0.21	0.16	0.019	0.10	2.5	3.8	3.8
Ecozone 3	62	0.070	0.0080	0.050	0.13	0.16	0.20	0.21
Ecozone 4	32	0.0073	0.0018	0.005	0.02	0.032	0.057	0.057

Table 3-2b - Summary of dietary cadmium exposure (µg/kg/d) from annual traditional food consumption based on reported consumption in the Food Frequency Questionnaire (FFQ) and simulated used 10,000 iterations of a Monte Carlo simulation.

	Mean Cd Dose (µg/kg/d)	SE	50th	90th	95th	97.5th	99th
Province	0.11	0.0083	0.0036	0.092	0.31	0.78	2.2
Ecozone 1	0.28	0.013	0.014	0.49	1.2	2.7	5.6
Ecozone 2	0.018	0.0025	0.0021	0.015	0.034	0.073	0.20
Ecozone 3	0.16	0.0084	0.011	0.24	0.61	1.2	3.0
Ecozone 4	0.0010	0.000024	0.00037	0.0023	0.0038	0.0063	0.011

The top 5 market foods contributing to the total dietary cadmium exposure are presented in Table 3-3. For the province, the top 5 market foods accounted for 44% of the average cadmium dose and were mainly a combination of cereals, grains, and potatoes (Table 3-3). Similar trends were observed at the ecozone level with 44-48% of cadmium intakes from market food being represented by the top five food items which were variations of cereals, grains, and potatoes. The top 5 traditional foods contributing to the cadmium exposure from the total diet (Table 3-4) accounted for 6.3% of the total mean cadmium exposure at the provincial level. Most notably, moose liver was considered the top contributing traditional food for the province, and ecozones 1 and 2, contributing close to double the cadmium intake than the second leading food (moose meat).

Table 3-5a presents the top 10 foods for mean cadmium exposures based on annual traditional food consumption, with Table 3-5b presenting the top 10 foods for the cadmium exposure of the upper 5th percentile of the population. In both top food lists, the kidney and liver of large terrestrial game are among the leading foods for the cadmium dose. Supplemental tables B 4-13 provide details on the cadmium dose from the top 20 foods in each ecozone, the intake (g/d), standard error, and the percent contribution to the variance in the dose.

Table 3-7 presents the cadmium concentration in the meat, kidney, and liver samples of deer and moose as analyzed through the FNFNES. Moose kidney samples had the highest average cadmium concentration (μg/g) followed by deer kidney.

Few statistically significant trends were observed on total dietary cadmium intake per population characteristics (Table 3-6). Although males had higher cadmium doses than females, this trend was not statistically significant. For age, only ecozone 3 had a significant difference between age groups (p=0.0357), with differences between the 19-30, and 31-51 year olds (p=0.0356), and between 31-50 and 51-70 year olds (p=0.033). Furthermore, there was no statistical difference within each age group across the ecozones, indicating similar cadmium doses for each age demographic. Smokers had significantly higher cadmium doses (p=0.30221) than non-smokers at the provincial level, and although this trend persisted at the ecozone level, it was not statistically significant.

Table 3-3 - Top 5 market foods for average Cadmium dose (ug/kg/d) with percentage of total Cadmium dose, calculated through the total diet study.

Province				Ecozone 1				Ecozone 2			
Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose
Potatoes	0.024	0.0013	18%	Potatoes	0.022	0.0022	16%	Potatoes	0.025	0.0030	17%
Pasta, plain	0.011	0.00085	8.6%	Pasta, plain	0.013	0.0018	9.6%	Pasta, plain	0.011	0.0018	7.5%
French fries	0.0097	0.00065	7.3%	French fries	0.012	0.0014	9.0%	Bread, white	0.0087	0.00058	6.1%
Bread, white	0.0074	0.00028	5.5%	Lettuce	0.0079	0.0029	5.8%	French fries	0.0078	0.0010	5.4%
Pasta	0.0064	0.00067	4.8%	Seeds	0.0071	0.0027	5.2%	Potato chips	0.0064	0.0018	4.4%
Total Cd Dose	0.13	0.0043	44%	Total Cd Dose	0.14	0.0087	46%	Total Cd Dose	0.14	0.013	41%

Ecozone 3				Ecozone 4			
Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose
Potatoes	0.033	0.0035	234%	Potatoes	0.023	0.0022	18%
Pasta, plain	0.0095	0.0016	6.8%	Pasta, plain	0.010	0.0015	8.2%
Bread, white	0.0084	0.00060	6.1%	French fries	0.0083	0.0012	6.7%
Pasta	0.0083	0.0017	6.0%	Bread, white	0.0075	0.00054	6.1%
French fries	0.0078	0.0014	5.7%	Pasta	0.0061	0.0013	5.0%
Total Cd Dose	0.14	0.0058	48%	Total Cd Dose	0.12	0.0047	44%

Table 3-4 - Top 5 traditional foods for average cadmium dose (ug/kg/d) with percentage of total Cadmium dose, calculated through the total diet study.

Province				Ecozone 1				Ecozone 2			
Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose
Moose Liver	0.0052	0.0033	3.9%	Moose Liver	0.0071	0.0066	5.1%	Moose Liver	0.011	0.011	7.8%
Moose Meat	0.0029	0.00045	2.2%	Moose Meat	0.0046	0.0011	3.3%	Moose Meat	0.0011	0.00041	0.79%
Rabbit meat	0.00016	0.00013	0.12%	Whitefish	0.00034	0.00014	0.25%	Rabbit meat	0.00073	0.00056	0.51%
Whitefish	0.00016	0.000050	0.12%	Pickerel	0.00018	0.000052	0.13%	Whitefish	0.000076	0.000061	0.05%
Pickerel	0.000086	0.000021	0.060%	Trout	0.000089	0.000042	0.060%	Pickerel	0.000063	0.000055	0.04%
Total Cd Dose	0.13	0.0043	6.3%	Total Cd Dose	0.14	0.0087	8.9%	Total Cd Dose	0.14	0.013	9.2%

Ecozone 3				Ecozone 4			
Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Cd Dose (µg/kg/d)	SE	% of Total Dose
Moose Meat	0.014	0.0025	10%	Moose Meat	0.00021	0.00014	0.17%
Caribou meat	0.00081	0.00029	0.58%	Maple Syrup	0.00010	0.000047	0.080%
Turkey meat	0.00024	0.00035	0.17%	Deer Meat	0.000075	0.000036	0.060%
Whitefish	0.00017	0.00019	0.12%	Winter squash	0.000075	0.000039	0.060%
Canada goose	0.00015	0.000060	0.11%	Perch	0.000067	0.000036	0.050%
Total Cd Dose	0.14	0.0058	11%	Total Cd Dose	0.12	0.0047	0.43%

Table 3-5a - Top 10 foods contributing to the mean cadmium dose based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

Province				Ecozone 1				Ecozone 2			
	Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)		Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)		Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)
Moose Kidney	0.067	0.0073	0.47	Moose Kidney	0.18	0.011	1.2	Moose Kidney	0.0089	0.0025	0.060
Caribou Kidney	0.019	0.0036	0.13	Caribou Kidney	0.056	0.0055	0.36	Moose Liver	0.0036	0.00044	0.22
Moose Liver	0.012	0.0012	0.61	Moose Liver	0.027	0.0015	1.7	Moose	0.0017	0.000064	6.3
Caribou Liver	0.0022	0.00035	0.13	Caribou Liver	0.0073	0.00061	0.44	Deer Liver	0.0011	0.00015	0.22
Rabbit	0.0018	0.00014	0.74	Moose	0.0027	0.000059	10	Rabbit	0.00086	0.000066	0.31
Moose	0.0017	0.000058	6.6	Beaver	0.0025	0.00012	0.77	Smelt	0.00054	0.000037	0.51
Beaver	0.0011	0.00012	0.37	Rabbit	0.0019	0.000080	0.71	Raspberries	0.00032	0.000013	4.2
Goose	0.00032	0.000013	3.6	Goose	0.00052	0.000016	5.2	Deer Kidney	0.00029	0.000036	0.0086
Raspberries	0.00016	0.0000080	2.0	White Sucker	0.00047	0.000030	1.8	Black Raspberries	0.00016	0.000011	1.8
Sum of top 10	0.11		30	Sum of top 10	0.28		54	Sum of top 10	0.018		28
Total Mean Cd Dose	0.11	0.0083	38	Total Mean Cd Dose	0.28	0.013	54	Total Mean Cd Dose	0.018	0.0025	32

Table 3-5a (con't) - Top 10 foods contributing to the mean cadmium dose based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

Ecozone 3				Ecozone 4			
	Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)		Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)
Moose Kidney	0.11	0.0077	11	Moose	0.00020	0.000012	5.6
Caribou Kidney	0.024	0.0034	2.7	Rabbit	0.00015	0.000015	0.89
Moose Liver	0.011	0.00079	3.5	Black Raspberries	0.00011	0.0000097	8.5
Rabbit	0.0053	0.00027	0.52	Deer	0.000085	0.0000038	5.5
Moose	0.0031	0.000078	3.4	Raspberries	0.0000083	0.0000065	7.9
Beaver	0.0027	0.00025	1.4	Maple Syrup	0.000063	000000034	1.6
Caribou Liver	0.0018	0.00027	13	Pickerel	0.000048	0.0000016	2.8
Goose	0.0012	0.000031	12	Perch	0.000046	0.0000021	1.8
Partridge	0.00020	0.0000077	0.91	Bearberries	0.000041	0.0000020	2.7
Sum of top 10	0.16		66	Sum of top 10	0.0010		42
Total Mean Cd Dose	0.16	0.0084	70	Total Mean Cd Dose	0.001	0.000024	43

Table 3-5b - Top 10 foods contributing to the mean cadmium dose of the upper 5th percentile of the population cadmium dose distribution based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

Province				Ecozone 1				Ecozone 2			
	Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)		Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)		Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)
Moose Kidney	1.2	0.14	7.1	Moose Kidney	0.281	0.075	16	Moose Kidney	0.18	0.049	1.1
Caribou Kidney	0.36	0.071	2.2	Caribou Kidney	0.085	0.025	4.8	Moose Liver	0.064	0.0083	3.7
Moose Liver	0.15	0.022	6.1	Moose Liver	0.022	0.0045	4.6	Deer Liver	0.021	0.0029	3.9
Caribou Liver	0.03	0.00663	1.2	Caribou Liver	0.0096	0.0034	0.57	Deer Kidney	0.0036	0.00065	0.07
Beaver	0.0055	0.0019	1.1	Moose	0.0029	0.0026	10	Black Raspberries	0.00033	0.00013	2.5
Rabbit	0.0047	0.0021	1.2	Beaver	0.0028	0.00056	0.64	Lake Whitefish	0.0002	0.000041	3.3
Moose	0.0015	0.00018	6.7	Lake Whitefish	0.0027	0.00015	6.8	Maple Syrup	0.000084	0.000012	0.58
Goose	0.00034	0.000070	3.4	Rabbit	0.0015	0.00029	0.76	Lake Trout	0.000049	0.000011	2
Raspberries	0.00016	0.000031	2	White Sucker	0.00081	0.00022	1.2	Northern Pike	0.000026	0.0000030	1.1
Black Raspberries	0.00013	0.000045	1.3	Goose	0.00073	0.00015	4.5	Rainbow Trout	0.0000093	0.0000023	0.77
Sum of top 10	1.8		46	Sum of top 10	0.4		77	Sum of top 10	0.27		20.4
Total Mean Cd	1.8	0.15	54	Total Mean Cd	0.41	0.079	82	Total Mean Cd	0.29	0.048	65.1

Table 3-5b (con't) - Top 10 foods contributing to the mean cadmium dose of the upper 5th percentile of the population cadmium dose distribution based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

Ecozone 3				Ecozone 4			
	Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)		Mean Cd Dose (µg/kg/d)	SE	Mean Intake (g/d)
Moose Kidney	1.8	0.13	11	Rabbit	0.0025	0.00027	0.89
Caribou Kidney	0.43	0.065	2.7	Moose	0.0020	0.00021	5.6
Moose Liver	0.09	0.014	3.5	Black Raspberries	0.0012	0.00018	8.5
Caribou Liver	0.012	0.0040	0.52	Raspberries	0.00085	0.00012	7.9
Rabbit	0.01	0.0027	3.4	Deer	0.00034	0.000059	5.5
Beaver	0.0091	0.0034	1.4	Maple Syrup	0.00032	0.000055	1.6
Moose	0.0036	0.00034	13	Cloudberries	0.00029	0.000080	1.8
Goose	0.0012	0.00012	12	Beaver	0.00026	0.000062	0.036
Partridge	0.00022	0.000038	0.91	Strawberries	0.00017	0.000056	0.45
Snow Goose	0.00022	0.000060	3.8	Black Bear	0.00014	0.000035	0.026
Sum of top 10	2.3		66	Sum of top 10	0		42
Total Mean Cd	2.3	0.14	70	Total Mean Cd	0.0087	0.00030	43

Table 3-6 – Mean, Standard Error (SE) and 95th percentile of cadmium intakes (µg/kg/d) from Total Diet Study with ranges for age groups, sex, and smoking status.

		n	Mean	SE	95th Percentile	Min	Max
Total Province		1429	0.13	0.0043	0.32	0.0000078	3.9
Age Groups	19-30	265	0.15	0.0084	0.35	0.0065	0.72
	31-50	611	0.13	0.0072	0.29	0.0000078	3.9
	51-70	436	0.14	0.0081	0.33	0.0022	2.3
	71+	116	0.12	0.0067	0.32	0.019	0.51
Sex	F	896	0.13	0.0063	0.31	0.0022	3.9
	M	533	0.14	0.0046	0.36	0.0000078	1.1
Smoker	No	705	0.12	0.0036	0.30	0.00020	0.64
	Yes	724	0.15	0.0078	0.34	0.0000078	3.9
Ecozone 1		359	0.14	0.0087	0.33	0.0000078	2.3
Age Groups	19-30	97	0.16	0.0157	0.35	0.0065	0.72
	31-50	159	0.12	0.0084	0.28	0.0000078	0.66
	51-70	80	0.15	0.0287	0.25	0.0022	2.3
	71+	23	0.13	0.0131	0.35	0.019	0.37
Sex	F	196	0.15	0.0149	0.27	0.0022	2.3
	M	163	0.13	0.0070	0.36	0.0000078	0.66
Smoker	No	156	0.12	0.0077	0.27	0.0022	0.64
	Yes	203	0.15	0.014	0.34	0.0000078	2.26
Ecozone 2		344	0.14	0.013	0.36	0.000015	3.94
Age Groups	19-30	47	0.13	0.016	0.48	0.0092	0.59
	31-50	149	0.15	0.026	0.31	0.000015	3.9
	51-70	122	0.14	0.0099	0.37	0.017	0.60
	71+	26	0.11	0.016	0.42	0.022	0.51
Sex	F	223	0.14	0.018	0.32	0.0062	3.9
	M	121	0.15	0.012	0.38	0.000015	1.1
Smoker	No	150	0.13	0.0076	0.32	0.00015	0.51
	Yes	194	0.16	0.021	0.39	0.000015	3.9
Ecozone 3		266	0.14	0.0058	0.33	0.013	0.52
Age Groups	19-30	60	0.18	0.015	0.36	0.016	0.38
	31-50	135	0.12	0.0065	0.29	0.013	0.36
	51-70	53	0.13	0.013	0.36	0.023	0.52
	71+	18	0.19	0.025	0.37	0.034	0.37
Sex	F	174	0.14	0.0072	0.33	0.013	0.52
	M	92	0.14	0.0095	0.34	0.016	0.36
Smoker	No	114	0.13	0.0082	0.33	0.013	0.52
	Yes	152	0.15	0.0079	0.33	0.015	0.38
Ecozone 4		460	0.12	0.0047	0.31	0.005	0.64
Age Groups	19-30	61	0.13	0.015	0.34	0.016	0.54

		n	Mean	SE	95th Percentile	Min	Max
	31-50	168	0.12	0.0072	0.31	0.0060	0.56
	51-70	181	0.13	0.0079	0.32	0.0053	0.64
	71+	49	0.12	0.0099	0.27	0.0209	0.34
Sex	F	303	0.12	0.0054	0.29	0.0053	0.64
	M	157	0.13	0.0090	0.38	0.0072	0.56
Smoker	No	285	0.12	0.0058	0.29	0.0053	0.64
	Yes	175	0.13	0.0079	0.36	0.0060	0.54

Table 3-7- Cadmium concentration in Moose and Deer meat, liver, and kidney samples with standard deviation, min, max, and average % moisture content. Data presented at µg/g wet weight.

	# replicates	# of composites	Average	SD	Min	Max	Average % moisture
Deer Meat	42	10	0.0033	0.0034	0.00050	0.0095	68.9%
Deer Kidney	4	3	3.0	5.0	0.045	8.8	75.6%
Deer Liver	14	4	0.47	0.37	0.12	0.86	70.1%
Moose Meat	83	17	0.024	0.066	0.0005	0.28	72.9%
Moose Kidney	15	8	14	8.0	0.0041	25	75.4%
Moose Liver	27	12	1.5	0.77	0.012	2.7	68.9%

3.4.1. Risk Assessment of Dietary Cadmium Exposures

Based on the total diet study, the total population was below the Joint FAO/WHO Expert Committee on Food Additives (JECFA) PTDI of 0.8µg/kg/d at the 99th percentile, indicating a low risk of cadmium toxicity. For traditional food consumers, the PTDI was exceeded at the 99th percentile level, whereas the non-traditional food consuming population was entirely below this reference dose. At the ecozone level, ecozones 2 and 1 had elevated cadmium intakes at the upper 95th and 97.5th percentiles, respectively, which exceeded the reference dose.

In the Monte Carlo simulation of cadmium exposure from annual traditional food consumptions, the 95th percentile of cadmium exposures was below the PTDI value for the province, and all ecozones but ecozone 1. The 97.5th percentile exceeded the reference dose in ecozone 3, with the 99th exceeding in the province as a whole (Figure 3-1). Given the finding from the total diet study of traditional foods contributing less to the total cadmium exposure than market foods, the annual traditional food consumption suggests an elevated risk from cadmium exposure could be expected for the upper 5th percentile of the population.

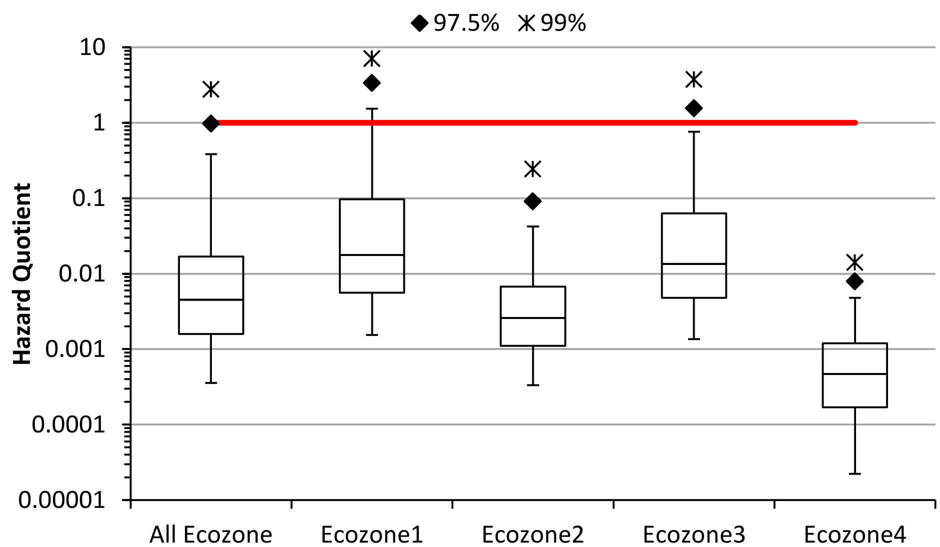


Figure 3-1 - Box plots showing the hazard quotient (exposure/reference dose of **0.8ug/kg/d**) 5th to 95th percentiles of cadmium intakes from annual consumption of traditional foods with indicators of the 96.5th and 99th percentiles.

To assess seasonal trends in cadmium exposures from traditional foods, Monte Carlo simulations were constructed (Supplementary Table B-3). Significant differences between mean seasonal cadmium exposures were observed within ecozones (Kruskal Wallis test; $p<0.0001$). In pairwise comparisons using Wilcoxon each pair testing, within ecozone seasons were all statistically different except in ecozone 2 (winter & spring; $p>0.05$), ecozone 1 (winter & summer; $p>0.05$), and for the province (winter & summer; $p>0.05$). The trends in mean cadmium intake per season, compared to the annual average are presented graphically in Supplemental Figure B-1. Distributions of cadmium intakes per season were compared to the JECFA reference dose (results not shown). At the provincial level, the reference dose was exceeded at the 96.8th percentile in the fall season, followed by the 97.1st in the winter. Ecozone 1 had the highest portion of the population exceeding the reference value in the spring season (89.6th percentile), followed by the fall (91.4th percentile). Divergent to this pattern, ecozone 3 had the highest portions of the population exceeding the reference value during fall and winter (96.3 and 95.6th percentiles, respectively). Ecozone 2 had less than 1% of the population exceeding the reference dose in any season, and the population in ecozone 4 was completely below this value.

3.5. Discussion

Dietary cadmium intakes in Aboriginal populations have been studied across Canada with varying study designs(Berti et al., 1998; Charania et al., 2014; Donaldson et al., 2010; Kuhnlein and Chan, 2000; Legrand et al., 2005). It was estimated that more than 90% of dietary cadmium exposures came from market foods in a Dene/ Metis population in the North West Territories in Canada (Kim et al., 1998), which is similar to finding from the current study where approximately 93% of the dietary cadmium was determined to be derived from market food sources. In the most recent Canadian Total Diet Study, the Canadian population has an average dietary cadmium exposure of 0.22 µg/kg/d (Health Canada, 2011), which is higher than the currently assessed average dietary cadmium intake in the First Nations population in Ontario (0.13 µg/kg/d). The total dietary cadmium intake by First Nations in Ontario was also lower than reported dietary cadmium intakes in total diet studies conducted in the United Kingdom (M. Rose et al., 2010), France (Arnich et al., 2012), Lebanon (Nasreddine et al., 2006), Hong Kong (Chen et al., 2014), and Cameroon (Gimou et al., 2014).

In national total diet studies, cadmium concentrations are typically higher in organ meats and shellfish, as well as cereal grains (rice and wheat), potatoes, and leafy vegetables (Muñoz et al., 2005; Nasreddine et al., 2006; M Rose et al., 2010; Ysart et al., 2000). Sunflower seeds, peanuts, flaxseed, and linseed accumulate cadmium from the soil in a manner similar to that of tobacco (Reeves and Vanderpool, 1997), whereas cadmium concentration in shellfish is impacted by oceanographic inputs and sediment (Lekhi et al., 2008). In national total diet studies, the proportion of the cadmium intake from food groups varies due to different consumption patterns. For instance, the Chilean total diet study indicated fish and shellfish consumption to be the leading contributor at 43% of the cadmium dose (Muñoz et al., 2005), whereas cereal and breads were among the top contributors in the United Kingdom (21%), Hong Kong (21%), Cameroon (54%), and Lebanon (36%), and Spain (38%) (Chen et al., 2014; Gimou et al., 2014; Llobet et al., 2003; Nasreddine et al., 2006; M Rose et al., 2010). Since the province of Ontario is abundant in fresh water, there was low reported consumption of shellfish in the current study population, which limited the contribution of this food group to the dietary cadmium intake. The top traditional foods contributing to the cadmium intake were dominated by kidney and liver of terrestrial land mammals; however, the percentage of their contribution to the total diet was less than 4% for the province, and up to 7.8% in ecozone 2.

Previous studies of dietary cadmium intakes in Aboriginal populations have focused on key traditional food items to evaluate the risk from its consumption. This includes a study of snowshoe hare consumption in a First Nations community near a copper smelter in Eastern Canadian, which concluded a low cadmium exposure risk, as well as limited evidence of a concentration gradient from the smelter (Bordeleau et al., 2016). Other, more extensive studies of cadmium in traditional food items include the consumption of large terrestrial game such as Caribou, Moose, and Deer. In general, cadmium content is higher in the kidneys than the liver of these mammals, as the distribution follows similar patterns as in humans with creation of metallothionein complexes (Charania et al., 2014; Crete et al., 1987; Crête et al., 1989; Elkin and

Bethke, 1995; Gamberg and Scheuhammer, 1994). A similar trend was observed with the aggregate samples analyzed through the FNFNES where the cadmium concentration in the kidney was higher than in liver and muscle/meat samples of these mammals (Table 3-7). Caribou have been extensively documents in terms of cadmium content due to their importance as a key-stone species in Arctic food systems and ecological health, as well as the relative simplicity of cadmium intake from consuming lichen, which accumulate atmospheric metal deposition (Gamberg et al., 2005). Caribou liver and kidney samples were not provided through the FNFNES, and therefore cadmium concentrations were represented by values from moose liver and kidney, respectively. Given that moose tend to have higher cadmium concentrations than caribou from the same region (Larter et al., 2016), this is a conservative assumption that would over, rather than under-estimate the cadmium exposures from this food source. Although a relationship between the age of caribou and moose and the cadmium concentrations in the liver and kidneys has been established (Archibald and Kosatsky, 1991; Crête et al., 1989; Gamberg and Scheuhammer, 1994; Larter and Nagy, 2000; Robillard et al., 2002), the age of the animal from which samples were provided through the FNFNES was not collected to facilitate comparison; however the range of results is presented in Table 3-7.

Compared to cadmium concentration in moose kidneys reported by other studies, the mean concentration of samples from FNFNES were lower than the average (26.6µg/g w/wt) reported for Alaskan moose (O'Hara et al., 2001), however they were within the range of averages (0.7-51.4µg/g w/wt) reported in Ontario moose sampled from the Algonquin area (Glooschenko et al., 1988), Nova Scotia (3-143 µg/g w/wt) (Frank et al., 2004), and Alaskan moose kidney cortex (0.1-65.7µg/g w/wt) (Arnold, 2006). The mean cadmium concentration in liver samples from the FNFNES (1.5µg/g w/wt) was lower than the average cadmium content found in Alaskan moose (3.13μg/g w/wt) observed by O'Hara et al. (2001), but within the range reported by Arnold et al. (2006) in Alaska. The range of cadmium in liver samples was also within the ranges reported from moose from the Canadian provinces of Nova Scotia (Frank et al., 2004) and Ontario (Glooschenko et al., 1988), however the maximum observed through the FNFNES was less than the maximum concentration reported in these studies. Previous studies have observed differences in the order of 4-5 times between the cadmium content of the liver and kidney of moose and deer (Crete et al., 1987), whereas, the difference in moose samples was 9 times, and deer 13 times based on FNFNES samples. However, this observation should be regarded with uncertainty, as kidney and liver samples provided for analysis in FNFNES were analyzed as composites and it cannot be confirmed whether they originated from the same mammal.

Previous assessments of the cadmium risk from consuming organ meats of moose concluded the cadmium intake to be elevated enough to recommend limiting the consumption (Crête et al., 1989). However, in more recent assessments of consumption of organ meats by Aboriginal populations, it was concluded that given the importance of traditional food for the health of Aboriginal populations, especially since a collectively dietary shift to a more market based diet with corresponding rises in chronic diseases is being observed, the consumption of offal from large game poses a moderate risk considering the low frequency of consumption (Archibald and Kosatsky, 1991). A study of Frist Nations in Northern Quebec found no significant relationship between the consumption of organ meats and blood cadmium levels, which further supports that the restriction of the consumption of these traditional foods is unwarranted (Charania et al., 2014). Moreover, it has been proposed that the high iron content in Caribou kidneys could contribute to the lower absorption of cadmium (approximately 2%) as iron deficiency is a factor that increases cadmium absorption, which suggests that the actual risk may be lower than computed estimates based on the general absorption assumption of 10% (Chan et al., 2001). In the presentation of FNFNES results to participating First Nation communities, a hunter noted the traditional cultural practice of consuming the organ meat immediately after a moose is killed which highlights the challenge of balancing culturally appropriate advice with health advisories (Chan et al., 2014).

Although the dietary exposure level for cadmium set by JECFA was lowered in 2010, the total dietary cadmium intake in the First Nations population assessed through this study remains largely below this threshold and lower than the dietary exposure level in the general Canadian population. This is in contrast to the results of the FNBI, where blood and urine cadmium concentrations were elevated in comparison to the general Canadian population. Previous biomonitoring studies analyzing cadmium exposures in Inuit adults have indicated similar trends compared to the general Canadian population with respect to smoking being the primary contributor to the cadmium body burden (Fontaine et al., 2008). Likewise, in a study of Cree First Nations communities in northern Quebec, no significant correlation was observed between traditional food consumption and blood cadmium concentrations, whereas a significant correlation was observed between blood cadmium and self-reported smoking (Charania et al., 2014). Given that blood cadmium is more reflective of recent exposures, a chronic cadmium dose from dietary sources would be better reflected in comparison to urinary cadmium levels and should be included in future study designs.

The smoking rate in the general Canadian population is 14.6% (Reid et al., 2015), whereas in First Nations adult population assessed through the FNBI, a rate of 64.7% was reported (Assembly of First Nations, 2013), and a rate of 62% was noted in the Ontario First

Nations Regional Health Survey (MacMillan et al., 2003). The First Nations population in Ontario assessed through the FNFNES had a slightly lower reported self-reported smoking rate of 51% (Chan et al., 2014). These high prevalence rates in First Nations are consistent with reports from other Canadian Aboriginal groups such as Cree First Nations of northern Quebec (67.8% in males, 70.2% in females between 15-39 years of age), the Inuit at 77% (Fontaine et al., 2008), and Dene/ Metis at 82% (Kim et al., 1998). Detailed characterization of the cadmium intake from smoking was beyond the scope of the FNFNES and the present study, as the FNFNES did not assess characterize second-hand smoke exposure, or other smoke exposures. However, the cadmium intakes from the total diet were contrasted to observe smokers having statistically significantly higher total dietary cadmium intakes than non-smokers at the provincial aggregate level. Although this trend remained when evaluating the total dietary cadmium intakes at the ecozone level, the finding was not statistically significant. It is unclear based on solely dietary patterns what could explain the elevated dietary cadmium intake for smokers, but a trend between smoking and organ meat consumption was also observed by Charania *et al.* (2014).

In the upper 5^{th} percentile of cadmium exposures from annual traditional food consumption for the provincial population, the average amount of liver and kidney meats consumed from moose and caribou was a cumulative 17g/d (range of 0 – 85g/d), contributing an average exposure dose of 1.7µg/kg/d (range of 0-31µg/kg/d) (97% of the daily cadmium exposure from traditional foods in this subpopulation). In contrast to this value, if a person was to smoke 20 cigarettes per day, with an average cadmium content of 1.7µg per cigarette, of which, 0.14-0.19µg is inhaled as found by Elinder et al., (1983) the cadmium exposure from smoking would be an estimated 2.8-3.8µg/d, irrespective of body weight. Therefore, if a heavy daily smoker was also a consuming an average of approximately 1 serving of organ meats per week, they would be exceeded the PTDI 5.6 - 6.9 times, however, the cadmium exposure from dietary sources would be below the guidance value, and this exceedance would be driven by cadmium exposures from smoking.

The PTDI was based on the detection of β2-microglobulin (β2MG), an early and sensitive biomarker of acute, yet reversible, kidney injury (EFSA, 2009). In Aboriginal populations in Canada, there is a higher prevalence of chronic kidney disease compared to the general population, which makes this endpoint particularly relevant for population health (Dyck, 2001; Gao et al., 2007; Samuel et al., 2014; Yeates and Tonelli, 2006). This is true for adults, and also children as a study found Aboriginal children under the age of 22 to have 2.18 times higher odds of end stage renal disease characterized by glomerulonephritis than non-Aboriginals of the same age group (Samuel et al., 2014). Diabetes has been cited as the leading cause of the higher prevalence of end stage renal disease in approximately 60% of cases in Aboriginal populations, with the remaining attributed to glomerulonephritis (Dyck, 2001). The prevalence of sever chronic kidney disease is significantly higher in First Nations populations than non-First Nations populations (Gao et al., 2007) which makes this a critical population health concern. The contributing role cadmium exposures have in the development of chronic kidney disease in First Nations populations is unknown.

3.6. Conclusion

This study comprehensively quantifies the dietary cadmium exposure in First Nations people living on reserve in Ontario for the first time in a total diet context as well as more detailed characterization of annual traditional food consumption patterns. Results indicated market foods to be the primary contributor to the dietary cadmium intake, with similar exposure magnitudes between reginal ecozones. The average dietary cadmium exposure in the First

Nations population in Ontario is less than the general Canadian population, and below dietary exposure criteria. However, when traditional foods are consumed, there is an elevation in the cadmium exposure that brings these individuals to an exposure with similar magnitude to the general Canadian population. The top traditional foods contributing to the cadmium exposure are the kidney and liver of large terrestrial game. This study observed a trend of smokers having higher dietary cadmium exposures than non-smokers. It is more urgent for heavy consumers of kidney and liver of large terrestrial game (>100 g per week) to stop smoking as their risk for decreasing kidney and bone health was elevated.

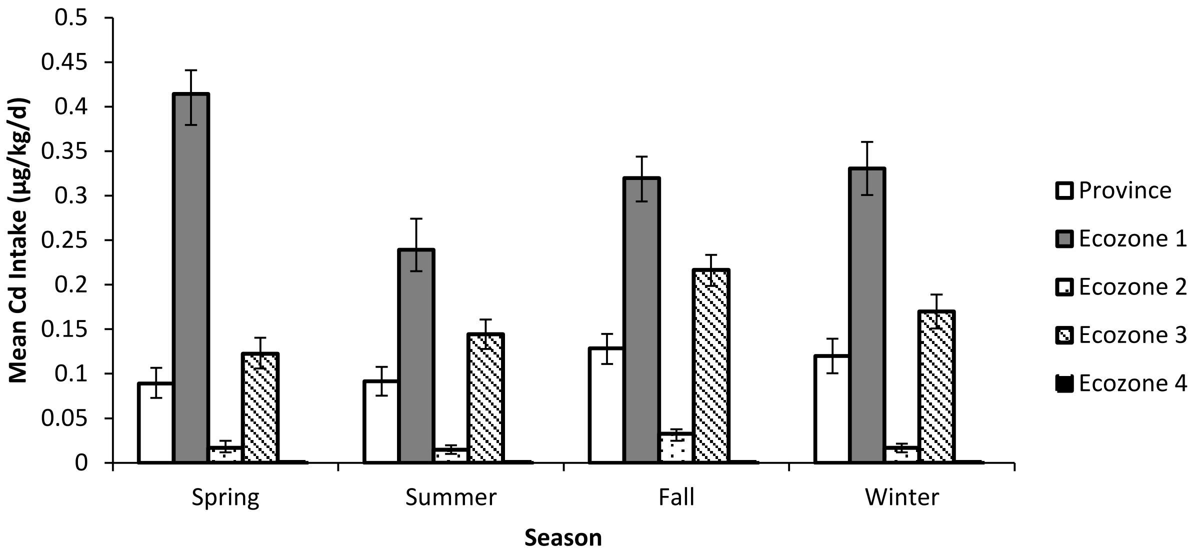
Appendix B- Supplemental Tables

Supplementary Table B-1 - Summary of mean cadmium dose (µg/kg/d) with standard error (SE) for different dietary components, with percent contribution to mean cadmium intake based on 24-hour recall Total Diet Study.

		Market Food			Traditional Food			Water			Total	
	n	Mean (µg/kg/d)	SE	%	Mean (µg/kg/d)	SE	%	Mean (µg/kg/d)	SE	%	Mean (µg/kg/d)	SE
Province	1429	0.13	0.0028	93%	0.0088	0.0033	6.6%	1.2E-05	1.3E-06	0.0093%	0.13	0.0043
Ecozone 1	359	0.13	0.0059	91%	0.012	0.0066	9.0%	6.7E-06	9.9E-07	0.0049%	0.14	0.0087
Ecozone 2	344	0.13	0.0056	91%	0.013	0.011	9.3%	2.8E-05	4.5E-06	0.019%	0.14	0.013
Ecozone 3	266	0.12	0.0054	89%	0.016	0.0026	11%	1.5E-05	3.1E-06	0.011%	0.14	0.0058
Ecozone 4	460	0.12	0.0046	100%	0.00057	0.00016	0.46%	9.3E-06	2.2E-06	0.0076%	0.12	0.0047

Supplementary Table B-2 - Summary of dietary cadmium exposure (µg/kg/d) from Market food sources in non-traditional food consumers based on 24-hour recall.

	n	Mean Cd Dose (µg/kg/d)	Standard Error	50th	90th	95th	97.5th	99th
Province	1239	0.13	0.0030	0.10	0.24	0.31	0.38	0.53
Ecozone 1	289	0.13	0.0069	0.10	0.22	0.31	0.41	0.66
Ecozone 2	318	0.13	0.0060	0.11	0.26	0.34	0.46	0.58
Ecozone 3	204	0.12	0.0062	0.095	0.22	0.27	0.33	0.37
Ecozone 4	428	0.12	0.0047	0.10	0.24	0.31	0.38	0.53



Supplemental Figure B-1 - Mean seasonal Cadmium intakes (ug/kg/d) with 95th% confidence intervals

Supplementary Table B-3 - Summary seasonal Cadmium intake (µg/kg/d) based on FFQ data in a Monte Carlo simulation (n=10,000)

Region	Season	50th	75th	90th	95th	97.5th	99th	Range (min - max)	Mean	SE
Province	Fall	0.0029	0.013	0.12	0.48	1.2	2.6	0-31	0.13	0.0089
	Spring	0.0017	0.0062	0.031	0.15	0.59	2.1	0-31	0.089	0.0083
	Summer	0.0022	0.010	0.04	0.22	0.67	2.0	0-31	0.092	0.0082
	Winter	0.0014	0.010	0.07	0.34	0.94	2.6	0-39	0.12	0.010
Ecozone 1	Fall	0.012	0.078	0.63	1.5	3.0	6.0	0-51	0.32	0.014
	Spring	0.0037	0.060	0.84	2.1	4.2	7.9	0-52	0.41	0.018
	Summer	0.0073	0.032	0.34	1.0	2.3	5.3	0-51	0.24	0.012
	Winter	0.007	0.063	0.60	1.5	3.1	6.4	0-52	0.33	0.015
Ecozone 2	Fall	0.0016	0.0051	0.021	0.051	0.12	0.43	0-22	0.033	0.0040
	Spring	0.00090	0.0037	0.015	0.031	0.06	0.15	0-16	0.017	0.0025
	Summer	0.0012	0.0027	0.0070	0.016	0.04	0.13	0-16	0.015	0.0025
	Winter	0.00093	0.0036	0.015	0.031	0.06	0.15	0-16	0.017	0.0025
Ecozone 3	Fall	0.011	0.061	0.45	1.0	1.8	3.6	0-25	0.22	0.0092
	Spring	0.0041	0.015	0.084	0.33	0.99	2.7	0-25	0.12	0.0084
	Summer	0.0056	0.022	0.18	0.57	1.1	2.9	0-25	0.14	0.0085
	Winter	0.0042	0.026	0.22	0.69	1.4	3.4	0-27	0.17	0.0097
Ecozone 4	Fall	0.00019	0.00071	0.002	0.0038	0.0069	0.014	0-0.12	0.00098	3.5E-05
	Spring	0.00018	0.00067	0.0021	0.0041	0.0076	0.013	0-0.12	0.00096	3.2E-05
	Summer	0.00037	0.00096	0.0023	0.0038	0.0063	0.011	0-0.057	0.0010	2.4E-05
	Winter	0.00015	0.00063	0.002	0.0041	0.0083	0.018	0-0.14	0.0011	4.6E-05

Supplementary Table B-4 - Top 20 foods contributing to the mean cadmium dose for the province based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented as well as % contribution to the variance in the dose for intake and contaminant concentration.

				Contribution to Dose Variance	
	Mean Intake (g/d)	Mean Cd Dose (ug/kg/d)	Mean SE	Intake (g/d)	Concentration (µg/g)
Moose Kidney	0.47	0.067	0.0073	24%	*
Caribou Kidney	0.13	0.019	0.0036	4.6%	*
Moose Liver	0.61	0.012	0.0012	20%	*
Caribou Liver	0.13	0.0022	0.00035	3.0%	*
Rabbit	0.74	0.0018	0.00014	9.5%	*
Moose	6.6	0.0017	0.000058	12%	1.2%
Beaver	0.37	0.0011	0.00012	5.2%	*
Goose	3.6	0.00032	0.000013	1.9%	*
Raspberries	2.0	0.00016	8.0E-06	0.4%	0.2%
Lake Whitefish	2.5	0.00014	6.8E-06	0.4%	*
Pickerel	5.6	0.00013	3.8E-06	*	*
White Sucker	0.46	0.00012	0.000015	0.9%	*
Grouse	0.31	0.00011	7.5E-06	0.5%	*
Smelt	0.13	0.00011	0.000011	0.3%	*
Black Raspberries	1.1	0.00010	8.3E-06	0.3%	*
Partridge	0.39	0.000094	5.2E-06	0.3%	*
Deer	1.6	0.000057	2.6E-06	*	*
Caribou	1.0	0.000049	5.0E-06	*	*
Sturgeon	0.52	0.000043	2.3E-06	0.1%	*
Northern Pike	1.7	0.000041	2.0E-06	*	*
Sum of top 20	30	0.11			
Total Mean Cd Dose	38	0.11		85%	

*<0.1%

Supplementary Table B-5 - Top 20 foods contributing to the mean cadmium dose in the upper 5th percentile of the province based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (µg/kg/d)	Mean SE
Moose Kidney	7.1	1.2	0.14
Caribou Kidney	2.2	0.36	0.071
Moose Liver	6.1	0.15	0.022
Caribou Liver	1.2	0.030	0.0066
Beaver	1.1	0.0055	0.0019
Rabbit	1.2	0.0047	0.0021
Moose	6.7	0.0015	0.00018
Goose	3.4	0.00034	0.000070
Raspberries	2.0	0.00016	0.000031
Black Raspberries	1.3	0.00013	0.000045
Smelt	0.12	0.00012	0.000046
Partridge	0.36	0.00012	0.000033
Lake Whitefish	2.4	0.00012	0.000024
Grouse	0.32	0.00010	0.000025
Pickerel	4.4	0.000091	9.0E-06
White Sucker	0.44	0.000086	0.000035
Deer	1.8	0.000078	0.000014
Caribou	1.5	0.000066	0.000030
Northern Pike	2.1	0.000050	6.9E-06
Sturgeon	0.46	0.000041	8.7E-06
Sum of top 20 foods	46	1.8	
Total Mean Cd	54	1.8	0.15

Supplementary Table B-6 - Top 20 foods contributing to the mean cadmium dose for the Ecozone 1 based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (ug/kg/d)	Mean SE	Contribution to Dose Variance Intake (g/d)	Contribution to Dose Variance Concentration (µg/g)
Moose Kidney	1.2	0.18	0.011	40%	*
Caribou Kidney	0.36	0.056	0.0055	9.6%	*
Moose Liver	1.7	0.027	0.0015	23%	0.4%
Caribou Liver	0.44	0.0073	0.00061	4.8%	*
Moose	10	0.0027	0.000059	3.4%	0.9%
Beaver	0.77	0.0025	0.00012	3.9%	0.2%
Rabbit	0.71	0.0019	0.000080	3.4%	0.1%
Goose	5.2	0.00052	0.000016	0.4%	*
White Sucker	1.8	0.00047	0.000030	0.6%	*
Lake Whitefish	6.6	0.00035	0.000012	*	*
Pickerel	14	0.00033	7.5E-06	0.1%	*
Grouse	0.86	0.00031	0.000015	0.3%	*
Raspberries	2.3	0.00018	5.4E-06	0.2%	*
Partridge	0.54	0.00014	7.2E-06	*	*
Sturgeon	1.2	0.00011	5.4E-06	*	*
Northern Pike	4.0	0.000095	4.1E-06	*	*
Red Sucker	0.34	0.000089	6.8E-06	0.2%	*
Black Raspberries	0.71	0.000074	3.7E-06	*	*
Goldeneye	0.077	0.000069	6.3E-06	*	*
Caribou	1.3	0.000060	4.3E-06	*	*
Sum of top 20	54	0.28			
Total Mean Cd Dose	54	0.28	0.013	91%	

*<0.1%

Supplementary Table B-7 - Top 20 foods contributing to the mean cadmium dose in the upper 5th percentile of Ecozone 1 based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (µg/kg/d)	Mean SE
Moose Kidney	16	0.28	0.075
Caribou Kidney	4.8	0.085	0.025
Moose Liver	4.6	0.022	0.0045
Caribou Liver	0.57	0.0096	0.0034
Moose	10	0.0029	0.00026
Beaver	0.64	0.0028	0.00056
Lake Whitefish	6.8	0.0027	0.00015
Rabbit	0.76	0.0015	0.00029
White Sucker	1.2	0.00081	0.00022
Goose	4.5	0.00073	0.00015
Pickerel	14	0.00066	0.000074
Grouse	0.76	0.00031	0.000074
Sturgeon	1.0	0.00023	0.000055
Northern Pike	4.4	0.00020	0.000034
Raspberries	2.4	0.00016	0.000023
Partridge	0.67	0.00014	0.000030
Lake Trout	1.8	0.00012	0.000025
Black Raspberries	0.81	0.000062	0.000011
Caribou	0.77	0.000061	0.000015
Red Sucker	0.42	0.000043	0.000015
Sum of top 20 foods	77	0.4	
Total Mean Cd	82	0.41	0.079

Supplementary Table B-8 - Top 20 foods contributing to the mean cadmium dose for the Ecozone 2 based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (ug/kg/d)	Mean SE	Contribution to Dose Variance	
				Intake (g/d)	Concentration (µg/g)
Moose Kidney	0.06	0.0089	0.0025	4.6%	*
Moose Liver	0.22	0.0036	0.00044	12%	*
Moose	6.3	0.0017	0.000064	24%	2.0%
Deer Liver	0.22	0.0011	0.00015	4.3%	*
Rabbit	0.31	0.00086	0.000066	12%	0.1%
Smelt	0.51	0.00054	0.000037	7.4%	0.8%
Raspberries	4.2	0.00032	0.000013	2.7%	0.3%
Beer Kidney	0.0086	0.00029	0.000036	3.2%	*
Black Raspberries	1.8	0.00016	0.000011	1.3%	0.1%
Lake Whitefish	2.8	0.00014	5.5E-06	1.5%	*
Deer	2.6	0.000097	3.5E-06	0.9%	0.1%
Partridge	0.40	0.000094	4.2E-06	0.8%	0.1%
Beaver	0.029	0.000092	0.000012	1.8%	*
Pickerel	3.2	0.000074	2.7E-06	0.4%	*
Cloudberries	0.69	0.000074	5.4E-06	0.8%	*
Maple Syrup	0.56	0.000064	2.7E-06	0.7%	0.1%
Grouse	0.18	0.000061	4.6E-06	0.8%	*
Lake Trout	2.0	0.000052	2.7E-06	0.5%	*
Northern Pike	1.2	0.000029	8.7E-07	0.2%	*
Perch	0.52	0.000023	1.3E-06	*	*
Sum of top 20	28	0.018			
Total Mean Cd Dose	32	0.018	0.0025	84%	

*<0.1%

Supplementary Table B-9 - Top 20 foods contributing to the mean cadmium dose in the upper 5th percentile of Ecozone 2 based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (μg/kg/d)	Mean SE
Moose Kidney	1.1	0.18	0.049
Moose Liver	3.7	0.064	0.0083
Deer Liver	3.9	0.021	0.0029
Deer Kidney	0.070	0.0036	0.00065
Black Raspberries	2.5	0.00033	0.00013
Lake Whitefish	3.3	0.00020	0.000041
Maple Syrup	0.58	0.000084	0.000012
Lake Trout	2.0	0.000049	0.000011
Northern Pike	1.1	0.000026	3.0E-06
Rainbow Trout	0.77	9.3E-06	2.3E-06
King Chinook Salmon	0.20	8.0E-06	2.1E-06
Brook Trout	0.63	7.5E-06	1.9E-06
Lowbush Cranberries	0.094	7.1E-06	2.1E-06
Round Whitefish	0.022	7.0E-06	6.1E-06
White Sucker	0.017	5.2E-06	2.6E-06
White Bass	0.091	4.2E-06	1.4E-06
Blue Huckleberries	0.038	2.6E-06	1.3E-06
Largemouth Bass	0.23	2.4E-06	4.2E-07
Hickory Nuts	0.026	1.6E-06	6.1E-07
Splake Trout	0.024	2.6E-07	1.1E-07
Sum of top 20 foods	20	0.27	
Total Mean Cd	65	0.29	0.048

Supplementary Table B-10 - Top 20 foods contributing to the mean cadmium dose for the Ecozone 3 based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (ug/kg/d)	Mean SE	Contribution to Dose Variance Intake (g/d)	Contribution to Dose Variance Concentration (µg/g)
Moose Kidney	11	0.11	0.0077	47%	2.1%
Caribou Kidney	2.7	0.024	0.0034	6.8%	*
Moose Liver	3.5	0.011	0.00079	14%	*
Rabbit	0.52	0.0053	0.00027	9.0%	0.3%
Moose	3.4	0.0031	0.000078	5.0%	*
Beaver	1.4	0.0027	0.00025	3.3%	0.2%
Caribou Liver	13	0.0018	0.00027	1.6%	*
Goose	12	0.0012	0.000031	1.3%	0.2%
Partridge	0.91	0.00020	7.7E-06	0.4%	*
Caribou	3.8	0.00014	9.0E-06	*	*
Snow Goose	3.0	0.00013	7.0E-06	*	*
Gooseberries	0.31	0.00013	0.000011	*	*
Pickerel	3.9	0.00010	2.6E-06	*	*
Grouse	1.6	0.000096	5.9E-06	*	*
Sturgeon	0.29	0.000087	3.9E-06	*	*
Round Whitefish	1.1	0.000085	6.1E-06	*	*
Mallard	0.83	0.000084	2.8E-06	*	*
Northern Pike	0.77	0.000057	2.2E-06	*	*
Raspberries	0.38	0.000048	1.9E-06	*	*
Black Raspberries	2.2	0.000047	2.2E-06	*	*
Sum of top 20	66	0.16			
Total Mean Cd Dose	70	0.16		91%	

*<0.1%

Supplementary Table B-11 - Top 20 foods contributing to the mean cadmium dose in the upper 5th percentile of Ecozone 3 based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (µg/kg/d)	Mean SE
Moose Kidney	11	1.8	0.13
Caribou Kidney	2.7	0.43	0.065
Moose Liver	3.5	0.090	0.014
Caribou Liver	0.52	0.012	0.0040
Rabbit	3.4	0.010	0.0027
Beaver	1.4	0.0091	0.0034
Moose	13	0.0036	0.00034
Goose	12	0.0012	0.00012
Partridge	0.91	0.00022	0.000038
Snow Goose	3.8	0.00022	0.000060
Caribou	3.0	0.00015	0.000031
Grouse	0.31	0.00012	0.000034
Pickerel	3.9	0.000094	0.000011
Mallard	1.6	0.000087	0.000013
Gooseberries	0.29	0.000085	0.000023
Sturgeon	1.1	0.000084	0.000013
Round Whitefish	0.83	0.000070	0.000016
Raspberries	0.77	0.000057	8.8E-06
Black raspberries	0.38	0.000055	0.000015
Northern Pike	2.2	0.000052	6.4E-06
Sum of top 20 foods	66	2.3	
Total Mean Cd	70	2.3	0.14

Supplementary Table B-12 - Top 20 foods contributing to the mean cadmium dose for the Ecozone 4 based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (ug/kg/d)	Mean SE	Contribution to Variance: Intake (g/d)	Contribution to Variance: Concentration (µg/g)
Moose	5.6	0.00020	0.000012	19%	0.4%
Rabbit	0.89	0.00015	0.000015	8.7%	*
Black Raspberries	8.5	0.00011	9.7E-06	8.4%	*
Deer	5.5	0.000085	3.8E-06	6.1%	0.7%
Raspberries	7.9	0.000083	6.5E-06	6.5%	0.3%
Maple Syrup	1.6	0.000063	3.4E-06	5.8%	0.3%
Pickerel	2.8	0.000048	1.6E-06	3.3%	0.5%
Perch	1.8	0.000046	2.1E-06	4.0%	0.2%
Bearberries	2.7	0.000041	2.0E-06	2.1%	*
Cloudberries	1.8	0.000035	4.3E-06	3.0%	*
Beaver	0.036	0.000029	3.6E-06	3.4%	*
Smelt	0.068	0.000018	2.1E-06	2.5%	*
Black Bear	0.026	0.000018	2.3E-06	2.6%	*
White Bass	0.59	0.000015	9.2E-07	1.2%	*
Strawberries	0.45	0.000014	3.0E-06	1.1%	*
King Chinook Salmon	0.37	0.000012	1.3E-06	0.8%	*
Sturgeon	0.16	0.000011	8.9E-07	1.3%	*
Acorns	0.24	5.6E-06	5.9E-07	0.2%	*
Smallmouth Bass	0.41	4.4E-06	3.3E-07	0.2%	*
Hickory Nuts	0.10	4.3E-06	6.2E-07	0.4%	*
Sum of top 20	42	0.0010			
Total Mean Cd Dose	43	0.0010		83%	

*<0.1%

Supplementary Table B-13 - Top 20 foods contributing to the mean cadmium dose in the upper 5th percentile of Ecozone 4 based on annual traditional food consumption reported through the Food Frequency Questionnaire, and simulated using Monte Carlo Simulation. Mean intake (g/d) is presented

	Mean Intake (g/d)	Mean Cd Dose (µg/kg/d)	Mean SE
Rabbit	0.89	0.0025	0.00027
Moose	5.6	0.0020	0.00021
Black Raspberries	8.5	0.0012	0.00018
Raspberries	7.9	0.00085	0.00012
Deer	5.5	0.00034	0.000059
Maple Syrup	1.6	0.00032	0.000055
Cloudberries	1.8	0.00029	0.000080
Beaver	0.036	0.00026	0.000062
Strawberries	0.45	0.00017	0.000056
Black Bear	0.026	0.00014	0.000035
Smelt	0.068	0.00014	0.000035
Perch	1.8	0.00012	0.000027
Bearberries	2.7	0.00011	0.000025
Pickerel	2.8	0.000078	0.000011
White Bass	0.59	0.000038	9.9E-06
King Chinook Salmon	0.37	0.000031	0.000017
Sturgeon	0.16	0.000018	7.4E-06
Lake Trout	0.24	8.1E-06	4.3E-06
Hickory Nuts	0.10	6.3E-06	4.5E-06
Smallmouth Bass	0.41	5.1E-06	1.4E-06
Sum of top 20 foods	42	0.0	
Total Mean Cd	43	0.0087	0.00030

4. Risk Assessment of Dietary Mercury Exposure among First Nations People in Ontario, Canada - a Total Diet Study and Probabilistic Assessment

4.1. Abstract

Mercury exposure is a public health priority as methyl mercury can have significant adverse effects on neurodevelopment. The objective of this study was to estimate dietary exposure to mercury and methyl mercury among First Nations living on-reserve in the province of Ontario, Canada. A total diet study was constructed based on a 24-hour recall and contaminant concentrations from Health Canada and the First Nations Food, Nutrition, and Environment Study (FNFNES). A probabilistic assessment of annual and seasonal traditional food consumptions was conducted. Results were compared to exposures in the general Canadian population and reference values from Health Canada and FAO/WHO for adults and women of childbearing age. Results indicated traditional foods to be the primary contributor to the dietary mercury intake (72%). The average dietary mercury exposure in the First Nations population in Ontario (0.039 µg/kg/d) was 1.6 times higher than the general Canadian population; however, the majority (97.8%) of the population was below dietary exposure criteria. Fish species contributing to the MeHg intake included pickerel, pike, perch and trout. Only 7.9% of the population meet the recommended fish consumption rate of two, 3.5oz servings per week from the American Heart Association. The consumption of lower tropic level fish should be promoted to provide the maximum nutritional benefit while reducing the mercury exposure risk.

Key Words: Mercury, Methyl Mercury, First Nations, Ontario, Fish Consumption, Exposure Assessment

4.2. Introduction

Mercury is a ubiquitous environmental global pollutant causing an increasing public health concern (Sheehan et al., 2014; WHO, 2010b). Although mercury is a naturally occurring element entering the environment through the weathering of mercury-containing rocks, volcanic eruptions, and geothermal activity, anthropogenic activities are the main routes in which it is emitted to the environment, including activities such as non-ferrous industrial processes, smelting, and the combustion of coal and municipal waste (Depew et al., 2013; Trip et al., 2000; Wang et al., 2004). Currently, natural sources contribute 10% of annual emissions, anthropogenic sources an estimated 30%, and the remaining 60% of emissions arise from the re-emission of previously released mercury residing in surface soils and oceans (UNEP, 2013a). Mercury emissions are a global concern as the chemical and physical properties result in its cycling for long ranges and deposition into soils and water bodies wherein it may be converted by microbes to the more toxic and bioaccumulative methyl mercury (MeHg) (Driscoll et al., 2013). The degree to which inorganic mercury is methylated and accumulates in food systems depends on multiple biotic and abiotic factors such as pH, water temperature, and the presence of microorganisms (Driscoll et al., 2013; UNEP, 2013a). Recognizing the adverse effects caused by mercury, especially MeHg on neurodevelopment of fetuses and children, the Minamata Convention was signed in 2013 as a binding framework with the objective of "protect[ing] the health and the environment from anthropogenic emissions and releases of mercury and mercury compounds" through reducing intentional mercury uses and emissions (UNEP, 2013b).

Humans are primarily exposed to MeHg through their diet, particularly through the consumption of fish, and in some populations, marine mammals (ATSDR, 1999; Ha et al., 2016). Exposures in terms of biomonitoring levels as well as dietary intakes have been monitored for decades in high risk population characterized by elevated fish and marine species consumption (Ha et al., 2016). One of the most prominent epidemiological findings of adverse health effects was in Minamata, Japan where fish, a main dietary staple, accumulated high levels of MeHg after an industrial release of mercury into a local water body, to result in high population exposures and increased prevalence of adverse effects (Harada, 1995). The neurotoxic manifestations of MeHg exposure are most sensitive in fetuses and young children, as MeHg is able to cross the placenta as well as the blood-brain barrier to result in behavioural changes and reduced cognitive and motor ability (Ha et al., 2016; Sheehan et al., 2014). In adults, the neurological effects include, but are not limited to, motor disturbances such as ataxia (awkward gait, difficulty swallowing and articulating words), tremors, and sensory dysfunction such as impaired vision (ATSDR, 1999; Ha et al., 2016).

In Canada, mercury has been a priority area of study for Aboriginal populations due to documented elevated exposures compared to the general Canadian population. In the Arctic, studies have characterized exposures in Inuit populations to be higher than southern dwellers in similar age and sex groupings (Chételat et al., 2015; Curren et al., 2014; Donaldson et al., 2010; Van Oostdam et al., 2005). Among First Nations populations, mercury has been assessed in smaller scale studies and remains a contaminant of concern, particularly in the province of Ontario, where point source industrial emissions play a greater role in mercury exposures than in the non-industrialized Arctic. The history of industrialization in this province includes 7 chlor-alkali plants operating between the 1930's to 1990's in the cities/ towns of Cornwall, Dryden, Thunderbay, Marathon, Hamilton, and Sarnia which utilized mercury in their processing (Paine, 1994). Waste discharges, particularly waste effluent from these facilities contributed to the local mercury contamination. The mercury discharges from the Dryden facility were particularly impactful on the First Nations reservations of Grassy Narrows and Wabaseemong situated on the English-Wabigoon river system. Between 1962-1970 approximately 10 metric tonnes of inorganic mercury was discharged into the river system, prompting consumption and sport-fishing bans on locally caught fish due to elevated levels of MeHg (Kinghorn et al., 2007). In this case, concentrations in top predatory species were within the ranges reported in species sampled from well-known, highly contaminated water systems such as Minamata Bay in Japan, St. Clair River in Ontario, and Lake St. Claire in Ontario (Neff et al., 2012), while biomonitoring data from community members reflecting elevated blood MeHg levels (Wheatley et al., 1997). Since the 1970's, mercury concentrations in regional fish have declined in the areas where this legacy point-source pollution occurred (Kinghorn et al., 2007; Neff et al., 2012; Weis, 2004), as have concentrations in biomonitoring data (Wheatley and Paradis, 1996, 1995).

Historic mercury biomonitoring data in some First Nations populations has been collected since the 1970's, and on aggregate has shown a decline in exposures (Wheatley and Paradis, 1995). In 2011, the First Nation Biomonitoring Initiative (FNBI) found blood mercury levels in First Nations on a national average to be similar to the general Canadian population reported in Cycle 1 of the Canadian Health Measures Survey (CHMS), however a high amount of variability between the communities participating in the study was noted (Assembly of First Nations, 2013). Collection of biomonitoring data has varied in methodology from blood analysis which represents a shorter-term exposure history, to hair samples in which 1cm growth portions represents a month of exposure. Hair is commonly used as an indicator because of its non-invasive nature, however, there is high variability across populations in respect to the representativeness of this measure for oral dietary exposures, such as fish consumption (Canuel et al., 2006; Liberda et al., 2014).

Given the abundant access to fresh water in Ontario from the Great Lakes water system, fish have historically comprised a large portion of traditional foods consumed by First Nations (Wheatley and Wheatley, 2000). Although traditional foods, like all foods, can be a vector for environmental pollutants, they represent an important source of essential and beneficial nutrients. This is especially true for fish which are an important source of dietary omega-3 fatty acids, an essential nutrient for brain and cardiovascular development and health (Ha et al., 2016; Hu et al., 2016; Sheehan et al., 2014). The prevalence of cardiovascular heart disease is higher in First Nations populations than in the general Canadian population, which highlights the importance of promoting fish consumption in public health initiatives (Anand et al., 2001; MacMillan et al., 2003; Reading, 2015; Yeates et al., 2015). Results from the Ontario First Nations Regional Health Survey observed a two-fold increase in self-reported heart problems between First Nation populations and the general provincial population (9.3% vs. 4.7%, respectively) (MacMillan et al., 2003). Other studies have found an increase in the prevalence of hospitalizations for ischemic heart disease in First Nation populations, while the rate in the general Canadian population has remained stable, or even declined (Shah et al., 2000), which suggest cardiovascular health must be a public health priority for this population (Reading, 2015). Cardiovascular disease is one health outcome that has seen a rise in its prevalence over the past few decades in Aboriginal populations, but other diseases such as diabetes, obesity, and chronic kidney disease are also significantly more prevalent in these populations than the general population (Dyck et al., 2010; Gao et al., 2007; MacMillan et al., 2003). Diet is a key contributing factor to all of these conditions; and as indigenous populations globally are in a dietary transition away from traditional foods, market foods of poorer nutritional quality are more frequently consumed in place of traditional foods (Egeland et al., 2011; Kuhnlein et al., 2004; Kuhnlein and Receveur, 1996; Laberge Gaudin et al., 2015; Schuster et al., 2011). The quality of the diet of First Nations is substantially better on days when traditional foods are consumed, as there are significantly lower intakes of saturated fats, sugars, and sodium than on days when only market foods are consumed (Chan et al., 2014). Furthermore, traditional foods have additional benefits for Aboriginal populations as they represent cultural and social ties which contribute to overall health and wellbeing (Kuhnlein, 1995; Laberge Gaudin et al., 2014). The majority of First Nations surveyed through the First Nations Food, Nutrition and Environment Study (FNFNES) indicated they would like more traditional foods in their diet; however the barriers to this included lack of time, transportation, and equipment/resources, as well as external factors such as the presence of industry (Chan et al., 2014, 2012, 2011)

Given the history of MeHg exposure in the First Nations population of Ontario, the assessment of dietary intakes continues to be a priority for determining risk management strategies. The objectives of this study were to quantify the mercury and MeHg exposure to Ontario First Nations peoples from the total diet, identify the key contributing food items, assess exposure risk to sensitive subpopulations (women of child bearing age) and compare dietary exposure to biomonitoring results of hair mercury concentrations. This study will contribute to the characterization of mercury exposures in Canadian Aboriginal populations, as well as contribute to the global call for research on mercury exposures in sensitive populations.

4.3. Method

Details on ethics, the First Nations Food, Nutrition and Environment Study (FNFNES), and probabilistic risk assessment methodology are presented in Chapter 2 of this thesis.

4.3.1. Traditional Food Samples & Analysis

The methodology for the collection of traditional food samples has been provided in Chapter 2 of this thesis. Mercury content was analyzed from homogenized composite samples digested in an open vessel using a combination of nitric acid and hydrogen peroxide based on EPA 200.3/6020A. Inductively coupled plasma mass spectrometry (ICP/MS) was employed to quantify mercury concentrations with a limit of detection of 0.004μg/g. Recovery of certified reference material ranged between 70-130%.

Not all composite samples were analyzed for MeHg content. MeHg concentrations were determined with a procedure using the methodology published by Liang, Bloom, and Horvat (1994) and US EPA Method 1630. Tissue samples were digested with methanol and potassium hydroxide, and content quantified using Cold Vapour Atomic Fluorescence Spectroscopy. Out of the 69 food items included in the provincial assessment, a total of 24 items had no MeHg data, and 5 (fish species and organ meats) food items included in the simulation used surrogate MeHg data. Items without MeHg data mainly plants, roots, shoots, and teas which are unlikely to contain methylated mercury.

4.3.2. Total Diet Study

Total diet studies are conducted to assess the intakes of key nutrients and contaminants in a population (EFSA et al., 2011). Using data from the 24-hour recall survey, recalled food items intake as weight/day were multiplied by with by the concentration of mercury of that food item in the database to determine the dietary mercury intake. Total mercury is not routinely included in the assessment of Canadian market foods through the Canadian Total Diet Study. Therefore to facilitate the conduction of a total dietary assessment for this contaminant, mercury concentrations from 1998-2000 were used as the most recent assessment of mercury in market foods (Dabeka et al., 2003). These concentrations were used under the assumption that no temporal variation existed between this assessment and the collection of traditional foods in 2010-2011. Concentrations of mercury in traditional food items were obtained through samples collected through the FNFNES, which were analyzed in uncooked, raw states. Water consumption was included in the calculation of mercury intakes as per harmonized total diet study guidance (EFSA et al., 2011), with concentrations in tap water represented by community-specific values measured by FNFNEs. Contaminant concentrations values below the limit of detection were represented by an upper-bound approach to provide conservative estimate as the limit of detection varied between traditional foods (0.004μg/g), and water (0.005μg/g). Dietary mercury intakes have been reported as weighted values based survey weights accounting for factors such as design weight (the inverse of the selection probability) and adjustment factors (non-response rate).

4.3.3. Hair Analysis

A total of 744 participants provided hair samples for mercury analysis. Samples were taken as a 5mm bundle of hair from the occipital region of the scalp. The hair bundle (full length, as cut from the scalp) was placed in a polyethylene bag and fastened to the bag with staples near the scalp end of the hair bundle. For samples collected in 2011, hair samples were analyzed in the CALA accredited Health Canada FNIHB Laboratory in Ottawa, Ontario. For samples collected in 2012, hair samples were analyzed in the SCC accredited Health Canada Regions and Programs Bureau Québec Region Laboratory in Longueuil, Québec using the same equipment and procedures as the Ottawa laboratory. For analysis, hair bundles were cut into three 1 cm segments, starting from the scalp end. Each 1cm segment was assumed to represent one month of hair growth and mercury exposure. Total mercury was analyzed by first being chemically treated to release ionic mercury species which are further selectively reduced to elemental mercury, followed by analysis using Cold Vapor Atomic Fluorescence

Spectrophotometer (CVAFS). The limit of quantification was 0.06 ppm (or μg/g) for total mercury. Any unused hair for the analysis was returned to participants as per cultural protocol.

4.3.4. Risk Assessment of Dietary Intake

Dietary intakes of MeHg were contrasted to the Health Canada provisional tolerable daily intake (PTDI) of 0.47μg/kg/d for adults (Environment Canada and Health Canada, 2010), and 0.23 μg/kg/d for women of childbearing age published by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (JECFA, 2007). Dietary exposures were contrasted to this reference dose to generate hazard quotients (exposure divided by reference dose), with hazard quotients greater than a value of one indicating increased population risk. Exposure estimates from the total diet study were limited to total mercury. It was assumed the MeHg exposure would be equivalent to the total mercury exposure, and were therefore compared to MeHg reference values. This is a conservative approach which would over, rather than underestimate risk, as methyl mercury is usually estimated to be 90% of the total mercury observed in fish (ATSDR, 1999).

Hair mercury values for the study population were contrasted to 6μg/g, the level which is associated with increased risk for adults established by Health Canada (Environment Canada & Health Canada, 2010). In women of childbearing age, hair mercury values were contrasted to the increased risk level of 2μg/g established by Health Canada (Legrand et al., 2010, 2005). Dietary mercury intakes from the total diet study were paired with each participant's hair mercury results and correlation analysis was conducted using Spearman's Rho correlation coefficient. Percentiles of hair mercury were contrasted to percentiles of seasonal MeHg dietary estimates from traditional food consumption (generated from FFQ data and simulated in a Monte Carlo assessment). Linear regression models were fitted to the data to observe the slope of the regression line between the distributions of the two data sets as a measure of the explained variability.

4.3.5. Statistical Analysis

JMP statistical software (version 12.1.0) was used to obtain summary statistics. Output distributions of exposures were not characterized by normal distributions, and therefore non-parametric statistics were applied to test differences. Differences between ecozones were assessed using Kruskal-Wallis test, and when significance was observed, Wilcoxon each pair test was applied to assess the significant of ecozone comparisons. Significance was considered as $p<0.05$.

4.4. Results

4.4.1. Total Diet Study

The average mercury exposure from the total diet in the province was 0.039µg/kg/d. At the ecozone level exposures ranged from 0.18 - 0.64µg/kg/d, with the total dietary mercury being significantly different between ecozones 3 and 4 (Wilcoxon each pair test; $p=0.0148$) (Table 4-1). The total mercury exposure was significantly higher in the population reporting traditional food consumption compared to the population not reporting it for the province and at each ecozone (Kruskal Wallis test; $p<0.0001$).

Table 4-1 - Summary of mercury (Hg) exposure (µg/kg/d) from the total diet for the total population, as well as traditional food consuming and non-consuming populations. Results based on 24-hour recall.

		n	Mean	SE	50th	90th	95th	97.5th	99th
Total Population	Province	1429	0.039	0.0049	0.0061	0.018	0.12	0.39	0.89
	Ecozone 1	359	0.064	0.013	0.0062	0.10	0.58	0.91	1.7
	Ecozone 2	344	0.026	0.0094	0.0059	0.015	0.036	0.17	0.56
	Ecozone 3	266	0.018	0.0040	0.0066	0.016	0.023	0.14	0.32
	Ecozone 4	460	0.020	0.0034	0.0057	0.016	0.088	0.24	0.46
Non-Traditional	Province	1239	0.011	0.00098	0.0056	0.013	0.019	0.057	0.19

		n	Mean	SE	50th	90th	95th	97.5th	99th
Food Consumers	Ecozone 1	289	0.007	0.00082	0.0055	0.012	0.015	0.020	0.042
	Ecozone 2	318	0.008	0.00078	0.0056	0.014	0.019	0.037	0.066
	Ecozone 3	204	0.011	0.0026	0.0057	0.011	0.015	0.049	0.20
	Ecozone 4	428	0.016	0.0024	0.0056	0.014	0.059	0.15	0.35
Traditional Food Consumers	Province	190	0.20	0.031	0.013	0.69	1.0	1.7	2.2
	Ecozone 1	70	0.36	0.062	0.066	0.98	1.7	2.1	2.2
	Ecozone 2	26	0.26	0.12	0.012	0.88	2.3	2.7	2.7
	Ecozone 3	62	0.039	0.017	0.012	0.062	0.14	0.58	1.1
	Ecozone 4	32	0.12	0.050	0.0072	0.46	0.86	1.4	1.4

A summary of total dietary mercury exposures by population demographic is presented in Table 4-2. An increasing trend between age and total dietary mercury was observed for the province (Kruskal-Wallis; $p=0.0179$), with significant difference between the oldest age group (71+ year olds) and all other age groups (Wilcoxon each pair; $p<0.014$). Males had significantly higher total dietary mercury exposure estimates than females for the province ($p=0.0002$), and ecozone 1 ($p=0.0211$) and ecozone 3 ($p=0.0012$). Women of childbearing age had lower average mercury exposures than compared to women not of childbearing age, however this finding was not statistically significant except in ecozone 3 ($p=0.002$). At the 95^{th} percentiles of mercury exposures in the female population, women of childbearing age had lower exposures than those who were not, with the this population having as low as 2% of the mercury dose of the non-sensitive population in ecozone 1, to 55% of the dose of non-childbearing age women in ecozone 4.

Table 4-2 - Summary of dietary Mercury (Hg) (µg/kg/d) from the total diet, exposures by age group, sex, and women of child-bearing age are presented.

		n	Mean	SE	50th	95th	99th
Province		1429	0.039	0.0049	0.0061	0.12	0.89
	19-30	265	0.023	0.0065	0.0061	0.017	0.73
Age Group	31-50	611	0.026	0.0046	0.0059	0.046	0.85
	51-70	436	0.043	0.010	0.0060	0.17	1.2

		n	Mean	SE	50th	95th	99th
	71+	116	0.10	0.027	0.0077	0.92	1.9
Sex	Female	896	0.026	0.004	0.0058	0.059	0.58
	Male	533	0.055	0.010	0.0067	0.35	1.1
Women of Child-Bearing Age	No	335	0.038	0.0083	0.0060	0.20	0.96
	Yes	561	0.018	0.0034	0.0057	0.024	0.48
Ecozone 1		359	0.064	0.013	0.0062	0.58	1.67
Age Group	19-30	97	0.043	0.017	0.0061	0.40	1.08
	31-50	159	0.051	0.014	0.0062	0.47	0.91
	51-70	80	0.10	0.043	0.0060	0.65	2.2
	71+	23	0.30	0.100	0.1035	1.8	1.9
Sex	Female	196	0.056	0.016	0.0059	0.34	1.5
	Male	163	0.10	0.024	0.0069	0.85	2.15
Women of Child-Bearing Age	No	57	0.080	0.034	0.0064	1.0	1.9
	Yes	139	0.024	0.0093	0.0056	0.020	0.89
Ecozone 2		344	0.026	0.0094	0.0059	0.036	0.56
Age Group	19-30	47	0.0080	0.0014	0.0071	0.021	0.06
	31-50	149	0.010	0.0015	0.0059	0.022	0.15
	51-70	122	0.047	0.023	0.0060	0.15	2.2
	71+	26	0.077	0.061	0.0053	1.1	1.6
Sex	Female	223	0.015	0.0035	0.0058	0.042	0.37
	Male	121	0.051	0.026	0.0062	0.026	2.42
Women of Child-Bearing Age	No	93	0.016	0.0056	0.0056	0.088	0.58
	Yes	130	0.012	0.0022	0.0060	0.026	0.16
Ecozone 3		266	0.018	0.0040	0.0066	0.023	0.32
Age Group	19-30	60	0.0060	0.00050	0.0060	0.014	0.02
	31-50	135	0.012	0.0023	0.0064	0.020	0.18
	51-70	53	0.026	0.010	0.0089	0.13	0.49
	71+	18	0.069	0.058	0.0086	1.1	1.1
Sex	Female	174	0.012	0.0021	0.0061	0.030	0.21
	Male	92	0.027	0.013	0.0078	0.034	1.1
Women of Child-Bearing Age	No	38	0.020	0.0081	0.0085	0.089	0.24
	Yes	136	0.015	0.0026	0.0055	0.018	0.18
Ecozone 4		460	0.020	0.0034	0.0057	0.088	0.46
Age Group	19-30	61	0.018	0.0070	0.0060	0.14	0.37
	31-50	168	0.026	0.0095	0.0056	0.079	0.77
	51-70	181	0.020	0.0045	0.0055	0.091	0.40
	71+	49	0.027	0.012	0.0059	0.15	0.56
Sex	Female	303	0.022	0.0055	0.0055	0.079	0.37
	Male	157	0.026	0.0065	0.0060	0.14	0.53
Women of Child-Bearing Age	No	147	0.019	0.0048	0.0055	0.11	0.37
	Yes	156	0.018	0.0065	0.0055	0.061	0.89

Traditional foods accounted for 72% of the average mercury exposure in the province (-Supplemental Tables

Supplementary Table C-1), despite only accounting for 1.8% of the average caloric intake (*results shown in Chapter 2*). However, between ecozones, the contribution of traditional foods to the total dietary mercury exposure varied between 23% in ecozone 4 to 88% in ecozone 1 (corresponding range of traditional food contribution to caloric intake 0.6% (ecozone 1) to 5.3% (ecozone3)).

The top 5 market foods contributing to the average mercury exposure with the percentage contribution to the mean dose are presented in Table 4-3. Canned fish was the leading contributor for market food sources. Table 4-4 presents the top 5 traditional foods contributing to the mean mercury exposure from the total diet. Pickerel, trout, and perch were among the top traditional foods, all of which are predatory fish. Table 4-6 presents the mercury and MeHg concentrations in most frequently consumed fish species. For traditional foods, nearly all were below Health Canada guidance concentrations of 0.5µg/g, with the exception of pike which had an average mercury concentration of 0.63±0.81µg/g (0.30±0.28µg/g MeHg).

4.4.2. Assessment of Annual Traditional Food Consumption

Mercury and MeHg exposure estimates from annual traditional food consumption are presented in Table 4-5. Significant differences were observed between all ecozones for MeHg exposures in the total and women of childbearing age populations, and all except ecozones 2 and 3 for mercury exposures (Table 4-5). Seasonal mercury and MeHg exposures were simulated with results presented in

Supplementary Table C-2 and Supplementary Table C-3 For mercury, all seasons were statistically difference within each ecozone, with the exception of Spring/Summer in the province, and summer/fall in ecozone 1 ($p>0.05$). For MeHg, all seasons were statistically difference within each ecozone, with the exception of spring/fall in the province, and fall/winter in ecozone 4 ($p>0.05$). For MeHg, exposures peaked in the summer season, with the exposure being at least twice that of any other season in the province, a trend which was observed in each ecozone (Supplementary Table C-3).

The top 10 traditional foods contributing to the mercury and MeHg doses for the province are presented in Table 4-7, with the top 10 traditional foods for the upper 5th percent of the population presented in Table 4-8. The major foods contributing to mercury and MeHg intakes were pickerel, northern pike, and trout. Top traditional foods contributing to the mean mercury and MeHg intakes for each ecozone are presented in

Supplementary Table C-5 to C-8, while top foods for the upper 5th percentile of ecozone population exposures are presented in Supplementary Table C-9 to C12.

Table 4-3 - Top 5 market foods contributing to the Mercury (Hg) exposure (µg/kg/d) for the total diet with mean, standard error (SE), and percentage contribution to the total dose. N=1429

Province				Ecozone 1				Ecozone 2			
Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose
Fish, canned	0.0020	0.00052	5.1%	Poultry, chicken & turkey	0.0014	0.00013	2.2%	Fish, canned	0.0020	0.00066	7.4%
Fish, fresh water	0.0015	0.00059	3.9%	Shellfish	0.00088	0.00070	1.4%	Poultry, chicken & turkey	0.0013	0.00014	4.8%
Poultry, chicken & turkey	0.0013	0.000067	3.4%	Beef, ground	0.00050	0.000060	0.78%	Fish, fresh water	0.00084	0.00066	3.2%
Shellfish	0.00068	0.00029	1.8%	Eggs	0.00040	0.00004	0.62%	Beef, ground	0.00064	0.000090	2.4%
Fish, marine	0.00061	0.00022	1.6%	Fish, marine	0.00038	0.00030	0.59%	Coffee	0.00039	0.000020	1.5%
Total Hg Dose	0.039	0.0049		Total Hg Dose	0.064	0.013		Total Hg Dose	0.026	0.0094	

Ecozone 3				Ecozone 4			
Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose
Fish, canned	0.0035	0.0020	19%	Fish, fresh water	0.0039	0.0017	19%
Poultry, chicken & turkey	0.00087	0.00010	4.9%	Fish, canned	0.0038	0.0013	19%
Eggs	0.00069	0.00006	3.9%	Poultry, chicken & turkey	0.0013	0.00013	6.2%
Beef, ground	0.00060	0.00010	3.4%	Fish, marine	0.0012	0.00057	5.7%
Rice	0.00038	0.000060	2.1%	Shellfish	0.00083	0.00056	4.1%
Total Hg Dose	0.018	0.0040		Total Hg Dose	0.020	0.0034	

Table 4-4 - Top 5 Traditional foods contributing to the mean Mercury (Hg) exposure (µg/kg/d) for the total diet with mean, standard error (SE), and percentage contribution to the total dose. N=1429

Province				Ecozone 1				Ecozone 2			
Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose
Pickerel	0.012	0.0030	32%	Pickerel	0.025	0.0075	40%	Pickerel	0.0091	0.0079	34%
Trout	0.0099	0.0036	26%	Trout	0.023	0.011	36%	Trout	0.0048	0.0047	18%
Whitefish	0.0028	0.00086	7.2%	Whitefish	0.0060	0.0024	9.3%	Whitefish	0.0013	0.0011	5.0%
Perch	0.0012	0.00063	3.3%	Sturgeon	0.0019	0.0023	2.9%	Perch	0.00081	0.00097	3.1%
Sturgeon	0.00074	0.00071	1.9%	Moose Meat	0.00048	0.00011	0.74%	Pike	0.00053	0.00094	2.0%
Total Hg Dose	0.039	0.0049		Total Hg Dose	0.064	0.013		Total Hg Dose	0.026	0.0094	

Ecozone 3				Ecozone 4			
Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose	Food	Mean Hg Dose (µg/kg/d)	SE	% of Total Dose
Whitefish	0.0029	0.0033	16%	Perch	0.0033	0.0018	16%
Caribou meat	0.0022	0.00082	12%	Pickerel	0.0014	0.00099	6.8%
Moose Meat	0.0015	0.00027	8.4%	Deer Meat	0.000040	0.000020	0.18%
Pickerel	0.0014	0.00112	8.0%	Moose Meat	0.000020	0.000010	0.11%
Canada goose	0.00020	0.000080	1.1%	Winter squash	0.000010	0.000010	0.070%
Total Hg Dose	0.018	0.0040		Total Hg Dose	0.020	0.0034	

Table 4-5 - Summary of mercury (Hg) and MeHg (MeHg) doses (µg/kg/d) from annually consumed traditional foods (µg/kg/d) based on FFQ data presented from a Monte Carlo simulation with 10,000 iterations.

		Mean	SE	50th	75th	90th	95th	97.5th	99th
Mercury (Hg)	Province	0.047	0.00097	0.018	0.049	0.11	0.18	0.27	0.44
	Ecozone 1	0.10	0.0018	0.049	0.12	0.24	0.38	0.56	0.90
	Ecozone 2	0.036	0.00062	0.018	0.042	0.084	0.13	0.19	0.28
	Ecozone 3	0.042	0.00090	0.017	0.042	0.096	0.16	0.25	0.38
	Ecozone 4	0.015	0.00037	0.0036	0.014	0.039	0.071	0.11	0.18
MeHg (MeHg)	Province	0.026	0.00056	0.0092	0.025	0.062	0.099	0.15	0.25
	Ecozone 1	0.058	0.00100	0.027	0.063	0.132	0.21	0.31	0.48
	Ecozone 2	0.019	0.00036	0.0093	0.021	0.042	0.063	0.09	0.14
	Ecozone 3	0.022	0.00045	0.0093	0.023	0.051	0.085	0.13	0.21
	Ecozone 4	0.0083	0.00022	0.0018	0.0071	0.021	0.037	0.06	0.10
MeHg (MeHg) **WCBA**	Province	0.014	0.00029	0.0049	0.014	0.035	0.056	0.087	0.14
	Ecozone 1	0.031	0.00051	0.014	0.035	0.076	0.12	0.17	0.24
	Ecozone 2	0.010	0.00019	0.0049	0.012	0.025	0.038	0.054	0.083
	Ecozone 3	0.0087	0.00018	0.0035	0.0087	0.02	0.033	0.053	0.084
	Ecozone 4	0.0066	0.00019	0.0013	0.0052	0.016	0.03	0.048	0.08

Table 4-6 - Concentrations of mercury (Hg) and methyl mercury (MeHg) in top consumed traditional food items. Average with standard deviation (SD) and range is presented.

			Hg		MeHg	
	n (composites)	n (replicates)	Average (SD)	Range (min-max)	Average (SD)	Range (min-max)
Pickerel	19	55	0.34 (0.23)	0.082 - 0.98	0.21 (0.34)	0.041 - 1.3
Whitefish	10	32	0.086 (0.048)	0.018 - 0.15	0.039 (0.022)	0.015 - 0.075
Perch	6	10	0.21 (0.074)	0.11 - 0.30	0.087 (0.059)	0.027 - 0.15
Pike	9	33	0.63 (0.81)	0.15 - 2.8	0.30 (0.28)	0.076 - 0.69
Smallmouth bass	4	7	0.45 (0.28)	0.075 - 0.67	0.29 (0.035)	0.26 - 0.31
Lake trout	7	20	0.27 (0.15)	0.063 - 0.53	0.14 (0.14)	0.019 - 0.29

Table 4-7 – Summary of mean consumption (g/d) and mercury (Hg) and MeHg (MeHg) exposure (µg/kg/d) of top 10 traditional foods for the province based on annual traditional food consumption for the total population and Women of Child-Bearing Age (WCBA) sub population.

Hg				MeHg				MeHg in **WCBA**			
Food	Mean Consumption (g/d)	Mean Hg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE
Pickerel	5.6	0.021	0.00066	Pickerel	5.6	0.013	0.00043	Pickerel	3.0	0.0071	0.00020
Northern Pike	1.7	0.012	0.00064	Northern Pike	1.7	0.0059	0.00032	Northern Pike	0.84	0.0029	0.00016
Lake Trout	1.0	0.003	0.00022	Lake Trout	1.0	0.0016	0.00011	Lake Trout	0.90	0.0013	0.00011
Lake Whitefish	2.5	0.0024	0.00012	Lake Whitefish	2.5	0.0011	0.000052	Lake Whitefish	1.5	0.00063	0.000033
Sturgeon	0.52	0.0014	0.000071	Smallmouth Bass	0.27	0.00085	0.000079	Smallmouth Bass	0.15	0.00048	0.000032
Smallmouth Bass	0.27	0.0013	0.00012	Sturgeon	0.52	0.00076	0.000049	Sturgeon	0.20	0.00030	0.000015
Perch	0.50	0.0010	0.000062	Perch	0.50	0.00035	0.000030	Perch	0.34	0.00023	0.000024
Trout	0.21	0.00087	0.000069	King Chinook Salmon	0.21	0.00029	0.000033	King Chinook Salmon	0.15	0.00020	0.000026
White Sucker	0.46	0.00039	0.000050	Brook Trout	0.26	0.00018	0.000016	Partridge	0.29	0.00010	0.0000060
Brook Trout	0.26	0.00035	0.000032	Trout	0.21	0.00016	0.000012	Mallard	0.41	0.000072	0.0000071
Sum of top 10	13	0.044		Sum of top 10	13	0.024		Sum of Top 10	7.8	0.013	
Total	38	0.047	0.00097	Total	38	0.026	0.00056	Total	27	0.014	0.00029

Table 4-8 – Summary of upper 5th percentile mean consumption (g/d) and mercury (Hg) and MeHg (MeHg) exposure (μg/kg/d) of top 10 traditional foods for the province based on annual traditional food consumption for the total population and Women of Child-Bearing Age.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (μg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (μg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (μg/kg/d)	SE
Pickerel	30	0.18	0.010	Pickerel	32	0.12	0.0065	Pickerel	14	0.056	0.0028
Northern Pike	14	0.13	0.011	Northern Pike	13	0.064	0.0054	Northern Pike	5.9	0.031	0.0026
Lake Trout	5	0.023	0.0038	Lake Trout	5.0	0.011	0.0018	Lake Trout	7.4	0.015	0.0020
Smallmouth Bass	1.2	0.0080	0.0019	Smallmouth Bass	1.1	0.0051	0.0014	Lake Whitefish	3.5	0.0023	0.00045
Lake Whitefish	5.1	0.0078	0.0015	Lake Whitefish	4.0	0.0027	0.00056	Smallmouth Bass	0.31	0.0017	0.00042
Sturgeon	0.55	0.0021	0.00062	Sturgeon	0.79	0.002	0.00073	King Chinook Salmon	0.54	0.0013	0.00041
Trout	0.30	0.0019	0.00062	King Chinook Salmon	0.47	0.0011	0.00041	Perch	0.55	0.00086	0.00031
Perch	0.69	0.0015	0.00034	Perch	0.63	0.00084	0.00034	Sturgeon	0.18	0.00031	0.000074
White Sucker	0.85	0.0015	0.00075	Teal	0.31	0.00046	0.00021	Trout	0.069	0.00011	0.000044
Brook Trout	0.65	0.00092	0.00028	Trout	0.17	0.00026	0.00010	Mallard	0.45	0.000086	0.000028
Sum of top 20	58	0.36		Sum of top 20	57	0.21		Sum of Top 10	33	0.11	
Total	83	0.36	0.011	Total	84	0.21	0.0065	Total	54	0.11	0.0029

4.4.3. Hair Mercury

A total of 744 participants in the FNFNES provided hair samples for mercury analysis. Hair mercury concentrations ranged from 0.03 to 13.54µg/g, with an average of 0.64µg/g (geometric average of 0.27µg/g) as calculated with population weights applied. Less than 1% of the population had a hair mercury value above the 6µg/g level which is associated with increased risk for adults established by Health Canada (Environment Canada & Health Canada, 2010). In women of childbearing age, approximately 18% had hair mercury values above the increased risk level of 2µg/g established by Health Canada (Legrand et al., 2010, 2005). The majority of the exceedances for women of childbearing age were observed in ecozone 1 with 47% of the women of childbearing age in this region exceeding the 2µg/g increased risk level. Ecozone 4 neither exceeded the increased risk level in the general adult population nor in the women of childbearing age subpopulation.

There was a significant association between hair mercury concentration and estimated total dietary mercury from the 24hour recall (spearman's rho correlation coefficient = 0.12, p=0.001). At the ecozone level, only ecozone 3 had a significant correlation between hair mercury and total dietary mercury (ρ=0.23; p=0.0043). Figure 4-1 presents the correlation between the total dietary mercury estimate with hair mercury concentrations of the participants, with a breakdown based on non-traditional food consumers (n=639), and traditional food consumers (n=105). There was no significant association between dietary Hg exposure and hair Hg concentrations among the non-traditional food consumers, based on the grouping from the 24-hour recall. In contrast, traditional food consumers had a significant positive correlation between total dietary mercury and hair mercury. A total of 12% of the provincial population had dietary Hg exposure above the PTDI of 0.23µg/kg/day but below the hair guidance level of 6µg/g.

Percentiles of seasonal mercury intakes were plotted against percentiles of hair mercury (Figure 4-2a) to show that the variability in the traditional food dietary estimate closely represents the variability observed in hair samples. The slope of the regression lines between seasonal MeHg exposure quantiles and hair mercury were all significant ($p<0.0001$) and positive. The strongest relationship was for summer (0.06; $p<0.0001$), which suggests that only 6% of the hair mercury variation in the population can be explained by summer traditional food consumption. Figure 4-2b illustrates percentiles of mercury intake from the total diet, market food sources, and traditional food sources from the 24-hour recall compared to hair mercury percentiles. The total dietary mercury intakes versus hair mercury values also showed a positive relationship. The slope of the regression line for the total dietary mercury intake was 0.041 ($p=0.0056$) which suggest that the total dietary mercury explains approximately only 4% of the hair mercury values ($p<0.003$).

Hair mercury values were highly variable across the province and within each ecozone.

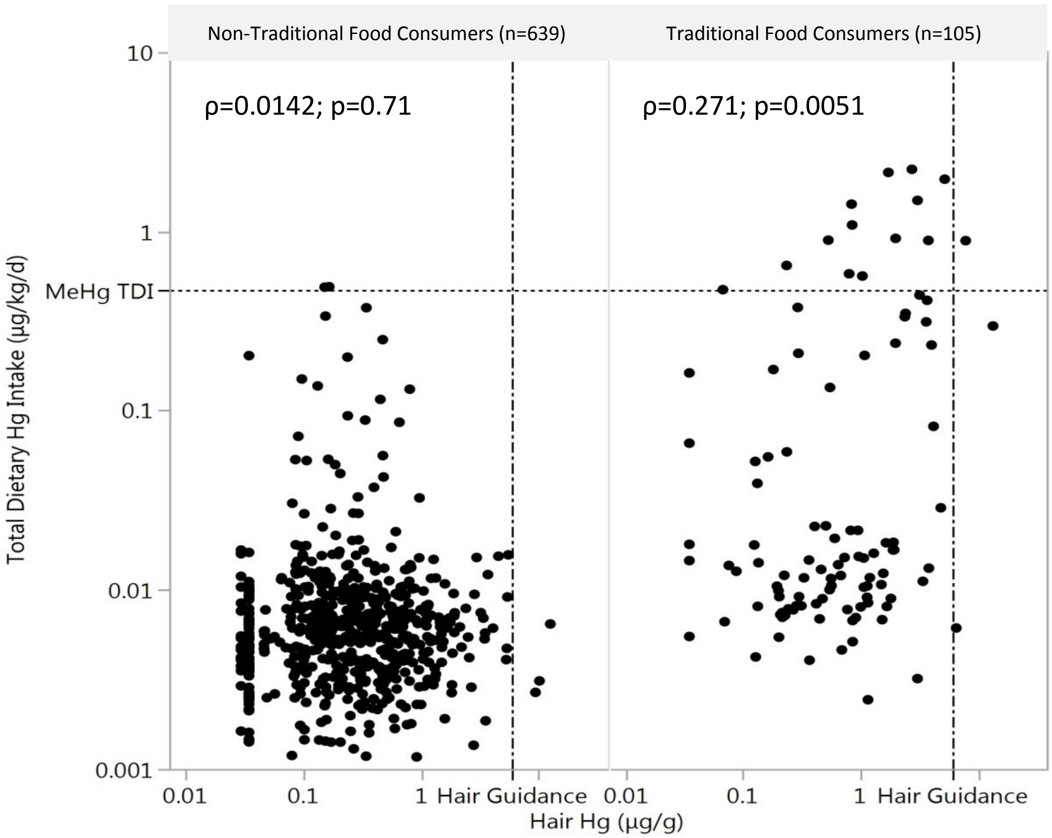


Figure 4-1 Hair Hg (µ/g) versus total dietary Hg (µg/kg/d) from 24-hour recall data in traditional food consumers (n=105) and non-traditional food consumers (n=639)

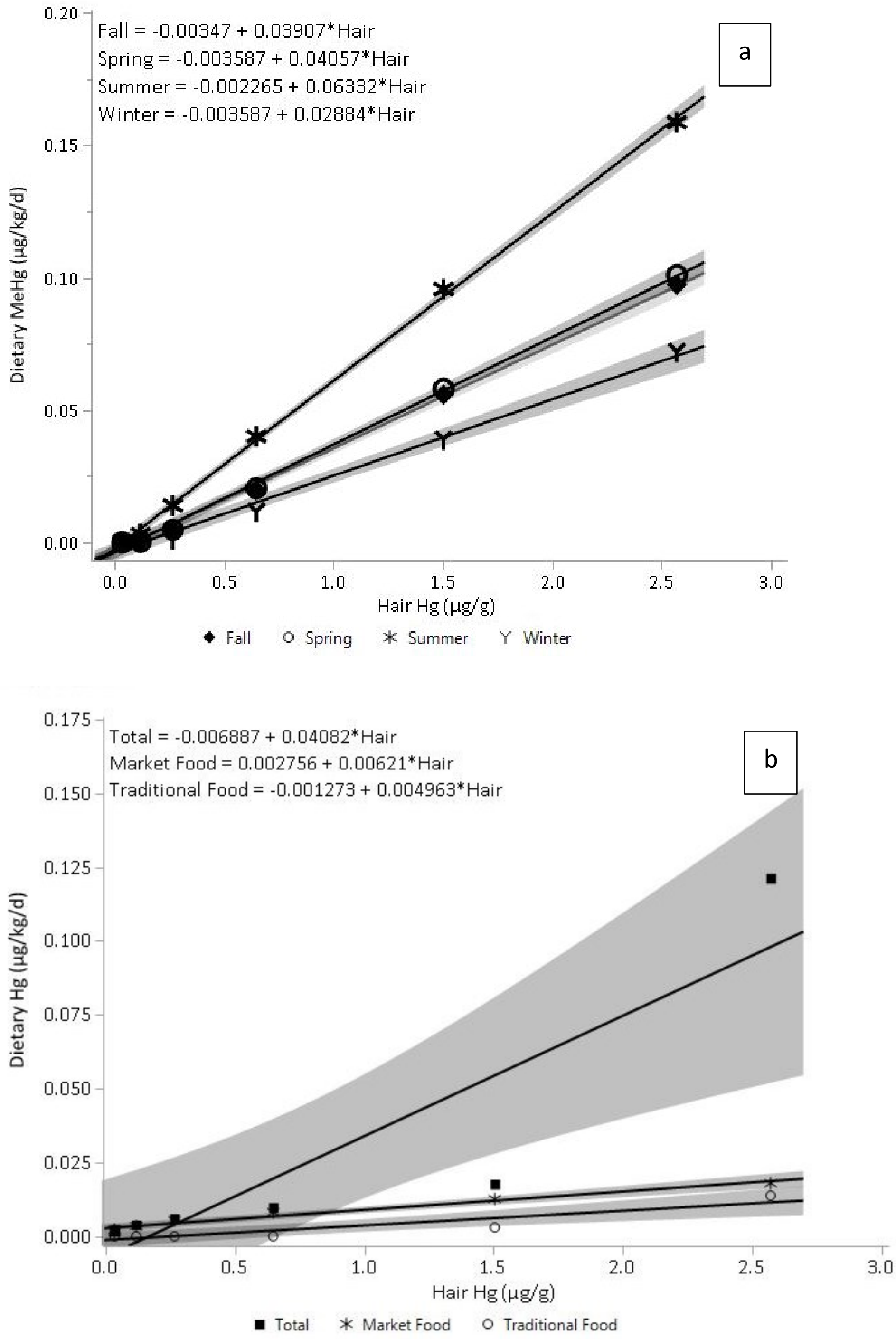


Figure 4-2 - (a) Plot of quantiles (5th-95th) of seasonal Hg Intake (µg/kg/d) versus quantiles of hair Hg (µg/g); (b) Plot of quantiles (5th-95th) of total dietary, traditional food, and market food Hg Intake (µg/kg/d) from Total Diet study versus quantiles of hair

4.4.4. Risk Assessment

Dietary mercury intakes from the total diet were largely below the PTDI of 0.47μg/kg/d for MeHg for the adult population (assuming that mercury exposure was equivalent to MeHg exposure). For the province, this reference dose was exceeded at the 97.8th percentile, while the ecozones exceeded this value at the percentiles: 94.6th (ecozone 1), 98.6th (ecozone 2), 99.5th (ecozone 3), and 99.2nd (ecozone 4). Among the exceedances in the province, 94% reported traditional food consumption. As shown in Table 4-1, traditional food consumers had higher total dietary mercury levels than those who did not report traditional food consumption in the past 24-hours. In the subpopulation of traditional food consumers (n=190), the reference dose for adults (0.47μg/kg/d) was exceeded at the 85th percentile. In the subpopulation of women of childbearing age, the total dietary mercury exposure was compared to the sensitive reference dose of 0.23μg/kg/d. At the provincial level, this reference dose was exceeded at the 98.7th percentile, while at the ecozone level only ecozones 1 and 3 exceeded this value (97.4th and 98th percentiles respectively).

Mercury exposure estimates (average and 95th percentiles) from traditional food sources in the total diet were higher than exposures predicted from annual (Table 4-5) and seasonal traditional food consumption (

Supplementary Table C-2) for the province. However, this observation is only applicable to the population which reported traditional food consumption through the 24-hour recall. When the results from the TDS are averaged for the total population, the average mercury exposure from traditional food sources becomes less than that predicted for annual traditional food consumption. The exception to this is ecozone 4, in which the average dietary mercury from traditional foods (0.020µg/kg/d) calculated from the total diet was greater than the average mercury exposure from annual traditional food consumption (0.015µg/kg/d).

Comparing the results from the assessment of annual traditional food consumption to the reference values demonstrated similar trends to the results of the 24-hour recall. Figure 4-3 shows the population distribution of the hazard quotient for MeHg exposures (based on the reference dose of 0.47µg/kg/d). The provincial population is below the reference dose (HQ<1) at the 99th percentile of the exposure distribution. Only ecozone 1 exceeds the reference dose at the 99th percentile, with a hazard quotient of 1.03. Figure 4-4 shows the MeHg (MeHg) hazard quotient distribution the women of childbearing age sub-population. Similar trends were observed in this population as with the general adult population, despite a comparison to a lower reference dose (0.23µg/kg/d). Ecozone 1 was the only sub-region where the reference dose was exceeded at the 99th percentile, with a hazard quotient of 1.04.

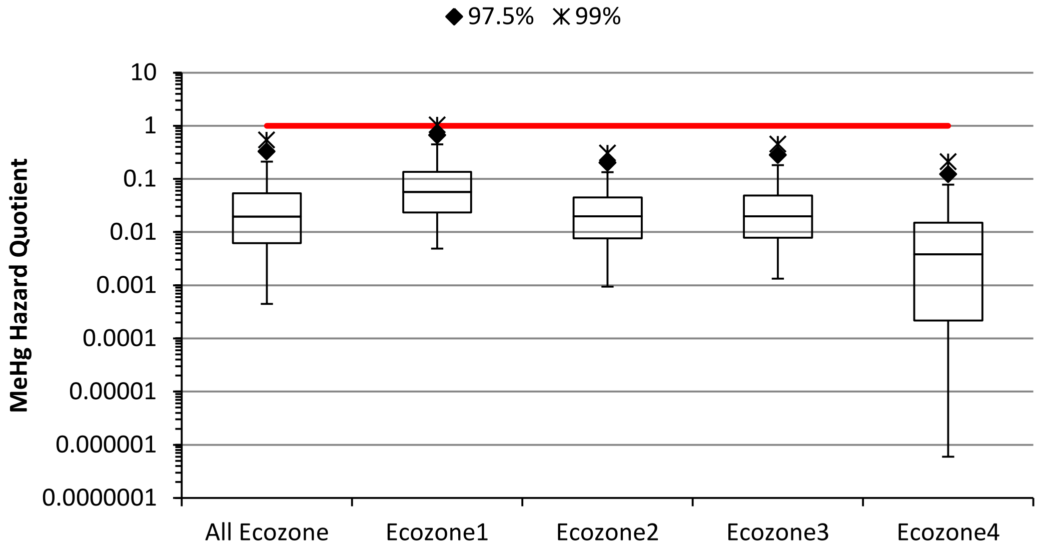


Figure 4-3 – Distribution of MeHg hazard quotients from 5th-95th percentile of the total adult population with indicators for 97.5 and 99th percentile points (hazard quotient based on a reference dose of 0.47µg/kg/d). Hazard quotient of 1, which indicated increased risk, has been indicated to facilitate comparison. Data from annual traditional food consumption as reported in the FFQ and simulated with Monte Carlo simulation (n=10,000 iterations).

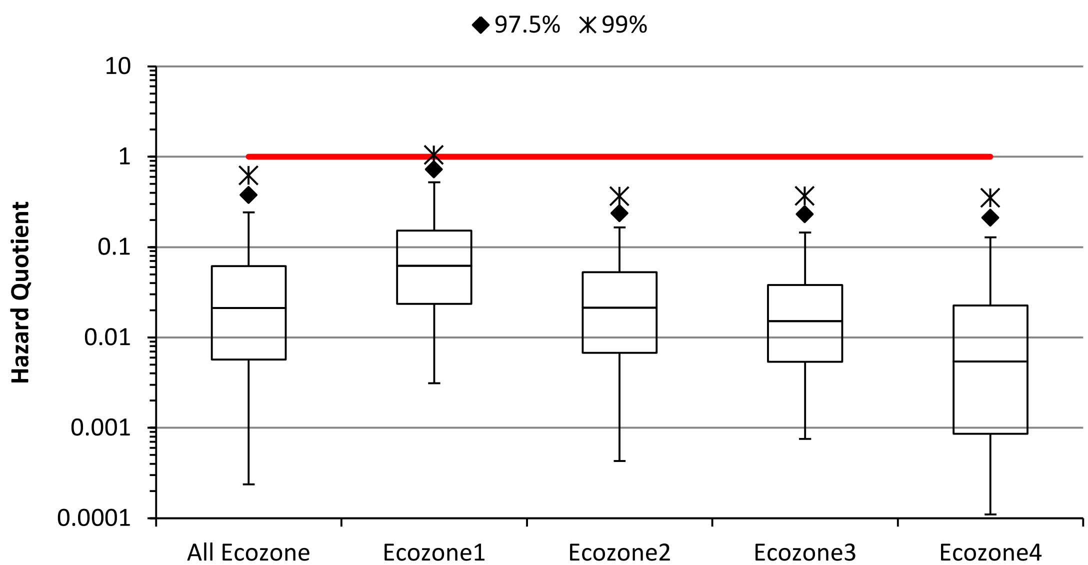


Figure 4-4 - Distribution of MeHg hazard quotients from 5th-95th percentile of the <u>women of childbearing age</u> population with indicators for 97.5 and 99th percentile points (hazard quotient based on a reference dose of 0.23µg/kg/d). Hazard quotient of 1, which indicated increased risk, has been indicated to facilitate comparison. Data from annual traditional food consumption as reported in the FFQ and simulated with Monte Carlo simulation (n=10,000 iterations).

4.5. Discussion

In comparison to dietary mercury exposures in the general Canadian population, our results indicate that First Nations in Ontario have elevated mercury exposures. Average mercury intakes from the total diet among First Nations across the province (mean= 0.039 μg/kg/d) were 1.6 times higher than those of the general Canadian population (mean=0.022μg/kg/d) as published by Dabeka et al. (2003). At the ecozone level, ecozone 1 and 2 were elevated compared to the general Canadian population (2.9 and 1.2 times higher, respectively). Direct comparison of age and sex specific exposure values is difficult as the grouping for age ranges was different between our study and previous study from Health Canada (Dabeka et al. 2003). However, the exposure estimate (0.023μg/kg/d) for the youngest group (19-30 year olds in our study) was within the range of the estimate for the Canadian general population (0.019-0.030μg/kg/d) at the 20-39 year-old age group. In contrast, our results for the two older age groups 51-70 and 71+ year olds were 2.3 and 5.3 times higher than the comparable age group of the Canadian population (65+ year olds). These results likely reflect the findings that older First Nations peoples consumed more traditional food as we found that traditional food contributed the majority of dietary mercury exposures, despite accounting for a small percentage of the caloric intake. In the provincial population reporting traditional food consumption, the average total dietary mercury exposure was nine times higher than the Canadian average. At the ecozone level, mercury exposures were the highest among traditional food consumers in ecozone 1 (16 times higher the Canadian average) and in ecozone 2 (12 times higher). Despite this elevated exposure, the population is largely below the dietary reference suggesting low population risk.

Results from the total dietary assessment conducted in this study are similar to those observed in two First Nations population in the Bay of Fundy on the east coast of Canada where total dietary mercury intakes were estimated to be an average of 0.03 and 0.05µg/kg/d (Legrand et al., 2005). Comparing the mercury exposures from traditional food consumption estimated from FFQ data to Inuit populations from the Inuvialuit Settlement Region, Nunavut, and Nunatsiavut jurisdictions, results of this study are lower than the average of 7.9 µg/kg/wk reported by Laird, Goncharov, Egeland, & Chan, (2013). First Nations population have different dietary profiles from Inuit who consume more marine mammal that contribute to the mercury exposure, which explains the difference and supports the need for cultural specific risk management strategies. This observation is supported by others who have suggested the risk of mercury exposure in non-coastal northern communities is relatively less than in marine environments since diets in these areas include more herbivorous terrestrial animals (Chan and Receveur, 2000; Hansen and Gilman, 2005).

Based on the 24-hour recall for the province, an average of 16g of fish were consumed per day (112g/week); however in the population reporting fish consumption from either market or traditional foods, fish constituted an average intake of 190g per day. In the annual traditional food assessment, an average of 14g per day (98g/week) of fish was consumed (median: 27g/d; 95th percentile: 108g/d). The majority of the population is therefore below the recommended two, 3.5oz weekly servings (200g) recommended by the American Heart Association, with only 7.9% meeting the guidance based on the 24 hour recall data, and 14% based on the annual traditional food consumption. This recommendation from the American Heart Association is based on obtaining the protective cardiovascular benefits of omega-3 fatty acids (American Heart Association, 2015). High exposures to mercury from fish and marine mammal consumption have been associated with diminished cardiovascular outcomes which persist even after accounting for the nutritional benefits of fish consumption in Inuit (Hu et al., 2016; Valera et al.,

2013) as well as in First Nations (Dewailly et al., 2002; Valera et al., 2011). This suggest that fish lower in mercury should be consumed with greater frequency, especially given the elevated prevalence of cardiovascular disease in this population (Reading, 2015). Based on the finding from our study, 55% of the average fish intake is from the consumption of pickerel and pike, which contribute 75% of the mercury exposure. These two species have well documented elevated concentration of mercury due to their high trophic level as predatory fish. In the upper 5th percentile of mercury exposure from annual traditional food consumption, this trend continued, as pickerel and pike represented 76% of the grams consumed in the top 10 food items, and 86% of the mercury exposure. Lower trophic level fish such as whitefish, which are lower in mercury concentrations, accounted for 19% of the average fish consumption and 5% of the mercury exposure, while in the upper 5th percentile of exposure whitefish represented 8.8% of the grams consumed, and 2.3% of mercury exposure. To minimize the risk of mercury exposures, yet maximize the nutritional benefits of omega-3 fatty acids, the consumption of species such as whitefish can be promoted.

Although results of the seasonal exposure assessment of traditional food consumption indicated exposures in summer to be almost twice that of other seasons, the average exposures to MeHg in these seasons remain below the guidance value, reiterating the low risk to consumers. This seasonal trend was expected, and is consistent with findings from sport fish consumers assessed in Montreal, where summer and fall fish had strong associations with blood mercury concentrations (Kosatsky et al., 2000). Anglers and sport fishers in the province of Ontario have increased consumptions of fish species such as small and largemouth bass, pickerel, northern pike, and yellow perch (Cole et al., 2004). Furthermore, it has been observed in St. Lawrence sport fishers that the consumption of pike explains most of the variations in the results of mercury biomonitoring (Kosatsky et al., 2000).

Concentrations of mercury in market foods have been discussed by Dabeka et al. (2003) and have indicated that market food samples were below threshold guidance value for mercury concentrations. Among market foods, canned fish was among the top contributors to the mercury exposure for the province, and contributed up to 19% of the average mercury exposure in ecozone 3. A study of canned tuna in the United States observed a slight increasing trend in the mercury content between 1998-2003, but also noted variation in concentrations based on the type of tuna (i.e. albacore versus skipjack) (Burger and Gochfeld, 2004). In comparison, pickerel (also referred to as Walleye), northern pike, and trout were the top contributing traditional foods to the mercury and MeHg exposure for the average consumer, as well as for consumers in the upper 5^{th} percentile of mercury exposures. Mercury concentrations in these species have been presented in Table 6. Whitefish, a lower tropic level fish, had an average mercury concentration of $0.086 \pm 0.048 \mu g/g$; the lowest among the most commonly consumed fish species contributing to the mercury exposure. A study of 17 Areas of Concern in the Great Lakes region found that pickerel had mercury concentrations ranging from 0.22 - 0.66μg/g which was within the range in of FNFNES data ($0.34 \pm 0.23 \mu g/g$), and Northern Pike Hg concentrations ranged from 0.40 to 0.60μg/g, which was slightly lower than the range observed in FNFNES (Weis, 2004). A study evaluating trends over the past 15 years in mercury concentrations in Ontario fish noted an increase in mercury concentrations, and projected levels to increase if current emission and accumulation trends persist, suggesting future consumption advisors and renewed need to reevaluate exposure risks in fish consuming populations (Gandhi et al., 2015). In the 2015 guide on consuming Ontario fish from the provincial regulator, mercury concentrations in fish were the reason for as low as 11% of consumption restriction advisories issues in Lake Erie, and up to 85% of the consumption restriction advisories issues for inland water bodies not in close proximity to industry (Ministry of the Environment and Climate Change, 2015).

In the maritime region of Canada, historical inventories illustrate that many of the most significant point sources of mercury emissions in the past such as the chlor-alkali industry, paint containing mercury additives, and pharmaceuticals, have been largely phased out so that modern sources are predominantly from fossil fuel combustion and waste disposal (Sunderland and Chmura, 2000). This has also been corroborated in the St. Lawrence River in Ontario where mercury accumulation in river bed sediment cores corresponds to the industrial releases from the area, showing concentrations peaking in the 1970's when emissions become more stringently regulated (Delongchamp et al., 2009). Further analysis of mercury in the sediment of this area suggest that this historical contamination is a probable source of presently observed elevated mercury concentrations observed in mature fish collected in the vicinity, which suggests remobilized has a local scale impact for contributing to the mercury burden (Delongchamp et al., 2010). In northern Ontario, lakes affected by mercury discharges from chlor-alkali plant continue to demonstrate a mercury concentration gradient in fish species related to the distance from release source (Kinghorn et al., 2007). These findings suggest that consumption guidance should be prioritized and routinely assessed in a framework that accounts for historical mercury releases.

The hair mercury biomonitoring data presented in this study has a high variability across the population, with dietary mercury intakes only able to account for a small percentage of the variability when either assessments of total diet using a 24-hour recall or seasonal traditional food consumption using an FFQ are applied. A similar high variability in mercury biomonitoring data was observed in blood biological specimens collected and analyzed through the FNBI (Assembly of First Nations, 2013). Relating dietary MeHg exposures to hair mercury concentrations has limitations noted in Aboriginal populations studied in Canada. For instance, a study relating dietary MeHg intakes to hair mercury levels in First Nations from eastern Canada found hair Hg to be a poor reflector of dietary MeHg intakes, citing ethnicity as a potential factor that influences the relevance of kinetic conversion factors (Canuel et al., 2006). A review of hair and blood mercury values in First Nations populations in Northern Quebec, Canada, observed similar finding, as estimations of blood mercury from hair mercury samples systematically over-estimated for males, and under-estimation for females, although a relationship between fish consumption and blood mercury levels was noted (Liberda et al., 2014). It has been observed that in individuals with infrequent fish consumption or where bolus doses of MeHg occur, the variability between biomonitoring matrices such as hair and blood may be high due to difference in the retention of mercury in each medium over the duration of the exposure (Mergler et al., 2007). This could be another factor contributing to the variability observed in hair mercury concentrations observed in this study, as traditional foods, which contributed the majority of the mercury dose, were consumed at a lower frequency which could reflect a consumption pattern that is more aligned with bolus doses rather than a steady chronic exposure.

Estimating chronic dietary intakes of contaminants has many limitations and burdens. The two commonly employed methods - 24-hour dietary recalls and food frequency questionnaires (FFQs), were employed in this study and have been validated as good tools for assessing contaminant intakes, especially when coupled together (Boucher et al., 2006; Liu et al., 2013; MacIntosh et al., 1997). In a case study of MeHg dietary intakes, Tran et al. (2004)

demonstrated the suitability of these two methods in estimating long-term dietary exposures. Tran et al. (2004) also observed a similar trend to our study, in that the long-term dietary estimates based on FFQ data were lower than the estimates produced from single day methods (i.e. 24-hour recall). This is likely due to the high within individual variability in reporting dietary patterns that over-estimates intakes on a daily basis, but when considering a population that was sampled in a representative framework and assessed with population weights, this variability becomes an accurate measure of the population variability.

Limitations to the total diet study conducted in this study include the representation of mercury concentrations in market food from data collected in 1999-2000. The assumption was made that the temporal difference between the collection of market food data and traditional food data (collected and analysed in 2011-2012) would have negligible impact on the total dietary mercury exposures as concentrations in market food remain fairly stable. The 24-hour recall employed in this study did not differentiate the type of tuna, and in the Canadian market food assessment, canned fish is neither differentiated based on the source. Since canned fish falls under the scope of regulatory surveillance, and the current levels in Canada are below the guidance value, this is assumed to have a negligible impact on the dietary risk profile for First Nations.

4.6. Conclusion

This is the first comprehensive study presenting mercury exposures for the First Nations population living on reserve in the province of Ontario in a total diet assessment, and with seasonal exposure estimates from traditional food consumption. Although this study noted elevated exposures to MeHg in First Nations compared to the general Canadian population, both dietary estimates and hair mercury biomonitoring data indicates low population risk for adverse health effects. Only 7.9% of the studied population consumed the recommended two servings per week advised by the American Health Association based on the 24-hour recall survey for increasing omega-3 nutrient intakes. Consumption of lower tropic level fish, which are lower in mercury, is recommended to be promoted to meet this advice.

Appendix C- Supplemental Tables

Supplementary Table C-1 – Mean Hg dose (µg/kg/d) with standard error (SE) and percentage contribution of market and traditional foods to the total dietary Hg exposure. Data based on the Total Diet Study conducted with 24-hour recall data.

		Market Food			Traditional Food			Total	
	n	Mean	SE	%	Mean	SE	%	Mean	SE
Province	1429	0.011	0.00087	28%	0.028	0.0048	72%	0.039	0.0049
Ecozone 1	359	0.0074	0.00081	12%	0.057	0.013	88%	0.064	0.013
Ecozone 2	344	0.0097	0.00096	37%	0.017	0.0094	63%	0.026	0.0094
Ecozone 3	266	0.0096	0.0020	53%	0.0084	0.0035	47%	0.018	0.0040
Ecozone 4	460	0.016	0.0023	77%	0.0048	0.0020	23%	0.020	0.0034

Supplementary Table C-2 - Distributions of seasonal mercury exposures (µg/kg/d) from FFQ data. Monte Carlo simulation results with 10,000 iterations.

Region	Season	50th	75th	90th	95th	97.5th	99th	Range (min - max)	Mean	SE
Province	Fall	0.0089	0.038	0.10	0.17	0.30	0.45	0-1.7	0.040	0.00094
	Spring	0.010	0.041	0.109	0.18	0.28	0.50	0-2.0	0.043	0.0011
	Summer	0.030	0.080	0.18	0.28	0.43	0.72	0-3.9	0.075	0.0017
	Winter	0.000	0.020	0.070	0.13	0.21	0.38	0-1.8	0.029	0.0009
Ecozone 1	Fall	0.036	0.10	0.23	0.36	0.55	0.86	0-2.6	0.092	0.0017
	Spring	0.024	0.075	0.18	0.30	0.49	0.82	0-2.5	0.070	0.0016
	Summer	0.037	0.11	0.24	0.40	0.64	1.0	0-4.1	0.10	0.0021
	Winter	0.077	0.18	0.37	0.56	0.86	1.3	0-4.1	0.16	0.0026
Ecozone 2	Fall	0.010	0.032	0.072	0.11	0.17	0.30	0-2.1	0.030	0.00067
	Spring	0.0071	0.028	0.067	0.11	0.16	0.23	0-1.4	0.025	0.00053
	Summer	0.0080	0.029	0.069	0.11	0.17	0.30	0-1.5	0.028	0.00065
	Winter	0.027	0.062	0.12	0.17	0.24	0.33	0-1.4	0.049	0.00071
Ecozone 3	Fall	0.0090	0.036	0.11	0.19	0.29	0.45	0-1.8	0.040	0.00094
	Spring	0.0012	0.005	0.024	0.060	0.11	0.22	0-3.9	0.020	0.00089
	Summer	0.0048	0.021	0.060	0.11	0.18	0.28	0-2.3	0.030	0.00071
	Winter	0.035	0.090	0.20	0.33	0.52	0.91	0-7.4	0.092	0.0024
Ecozone 4	Fall	0.00011	0.0037	0.024	0.052	0.085	0.15	0-0.76	0.0094	0.000035
	Spring	0.000080	0.0023	0.022	0.052	0.090	0.16	0-0.76	0.0096	0.000031
	Summer	0.00021	0.010	0.037	0.072	0.12	0.20	0-1.2	0.015	0.000020
	Winter	0.0036	0.014	0.039	0.071	0.11	0.18	0-0.7	0.015	0.000046

All seasons were statistically difference within each ecozone, with the exception of Spring/Summer in the province, and summer/fall in ecozone 1 ($p>0.05$).

Supplementary Table C-3 - Distributions of seasonal MeHg exposures (µg/kg/d) from FFQ data. Monte Carlo simulation results with 10,000 iterations.

Region	Season	50th	75th	90th	95th	97.5th	99th	Range (min - max)	Mean	SE
Province	Fall	0.0046	0.020	0.056	0.097	0.15	0.26	0-0.90	0.022	0.00056
	Spring	0.0049	0.021	0.058	0.10	0.16	0.28	0-1.3	0.024	0.00064
	Summer	0.014	0.04	0.096	0.16	0.23	0.42	0-1.6	0.04	0.0009
	Winter	0.0013	0.012	0.039	0.072	0.12	0.21	0-0.82	0.016	0.00048
Ecozone 1	Fall	0.020	0.055	0.12	0.20	0.30	0.46	0-1.5	0.050	0.00091
	Spring	0.019	0.056	0.13	0.22	0.34	0.57	0-2.2	0.055	0.0012
	Summer	0.042	0.099	0.20	0.33	0.48	0.72	0-2.7	0.088	0.0015
	Winter	0.013	0.041	0.10	0.17	0.27	0.42	0-1.5	0.040	0.00085
Ecozone 2	Fall	0.005	0.017	0.038	0.062	0.098	0.16	0-1.0	0.016	0.00038
	Spring	0.0037	0.015	0.035	0.057	0.086	0.15	0-1.4	0.014	0.00039
	Summer	0.013	0.032	0.059	0.086	0.12	0.18	0-1.4	0.025	0.00041
	Winter	0.0033	0.014	0.034	0.053	0.080	0.13	0-1.4	0.013	0.00033
Ecozone 3	Fall	0.0053	0.02	0.057	0.10	0.16	0.26	0-0.92	0.022	0.00052
	Spring	0.0026	0.012	0.034	0.06	0.091	0.15	0-0.89	0.013	0.00035
	Summer	0.019	0.048	0.10	0.18	0.28	0.51	0-2.8	0.049	0.00117
	Winter	0.000	0.002	0.01	0.03	0.055	0.11	0-1.4	0.0074	0.0004
Ecozone 4	Fall	5.0E-05	0.0018	0.013	0.027	0.047	0.082	0-0.43	0.0052	0.000035
	Spring	9.0E-05	0.0054	0.020	0.038	0.062	0.11	0-0.70	0.0079	0.000031
	Summer	0.0018	0.0071	0.021	0.037	0.058	0.099	0-0.64	0.0083	0.000024
	Winter	3.0E-05	0.0011	0.011	0.026	0.049	0.084	0-0.43	0.0051	0.000046

Supplementary Table C-4 - Dietary Mercury exposure from Traditional Food consumption in 24-hour recall (µg/kg/d)

	n	Mean	SE	50th	90th	95th	97.5th	99th
Province	190	0.19	0.031	0.0078	0.69	0.98	1.7	2.2
Ecozone 1	70	0.36	0.062	0.063	0.97	1.7	2.1	2.2
Ecozone 2	26	0.26	0.12	0.0060	0.88	2.3	2.7	2.7
Ecozone 3	62	0.034	0.017	0.0064	0.057	0.14	0.58	1.0
Ecozone 4	32	0.094	0.037	0.0015	0.46	0.68	0.91	0.91

Supplementary Table C-5 - Summary table of the top 10 Traditional foods contributing to the mean mercury (Hg) and MeHg (MeHg) in Ecozone 1 and for MeHg for WCBA with standard error and the mean consumption (g/d) presented. Results based on FFQ data in a Monte Carlo simulation.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE
Pickerel	14	0.053	0.001252	Pickerel	14	0.034	0.00080	Pickerel	7.5	0.019	0.00037
Northern Pike	4.0	0.029	0.001248	Northern Pike	4.0	0.013	0.00055	Northern Pike	1.8	0.0057	0.00027
Lake Whitefish	6.6	0.0065	0.000212	Lake Trout	2.2	0.0035	0.00017	Lake Trout	2.2	0.0035	0.00021
Lake Trout	2.2	0.0062	0.000299	Lake Whitefish	6.6	0.0028	0.000087	Lake Whitefish	3.2	0.0013	0.000047
Sturgeon	1.2	0.0036	0.00017	Sturgeon	1.2	0.0019	0.000094	Sturgeon	0.46	0.00073	0.000024
White Sucker	1.8	0.0016	0.000093	Sauger	0.31	0.00066	0.000073	Burbot	0.36	0.00026	0.000020
Trout	0.31	0.0013	0.000095	Burbot	0.56	0.00042	0.000028	Grouse	0.51	0.00019	0.000015
Sauger	0.31	0.00069	0.000089	Grouse	0.86	0.00032	0.000015	Partridge	0.35	0.00013	8.9E-06
Brook Trout	0.33	0.00041	0.000037	White Sucker	1.8	0.00023	0.000015	Mallard	0.53	0.000091	9.8E-06
Burbot	0.56	0.00040	0.000026	Trout	0.31	0.00023	0.000016	Moose	8.3	0.000090	1.6E-06
Sum of top 10	31	0.10		Sum of top 10	32	0.057		Sum of Top 10	25	0.031	
Total	61	0.11	0.002	Total	61	0.058	0.0010	Total	40	0.031	0.00051

Supplementary Table C-6 - Summary table of the top 10 Traditional foods contributing to the mean mercury (Hg) and MeHg (MeHg) in Ecozone 2 and for MeHg for WCBA with standard error and the mean consumption (g/d) presented. Results based on FFQ data in a Monte Carlo simulation.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE
Pickerel	3.2	0.012	0.00045	Pickerel	3.2	0.0074	0.00029	Pickerel	1.7	0.0039	0.000098
Northern Pike	1.2	0.0084	0.00027	Northern Pike	1.2	0.0039	0.00013	Northern Pike	0.77	0.0024	0.000091
Lake Trout	2.0	0.0060	0.00028	Lake Trout	2.0	0.0031	0.00015	Lake Trout	1.1	0.0018	0.00012
Lake Whitefish	2.8	0.0026	0.000097	Lake Whitefish	2.8	0.0012	0.000045	Lake Whitefish	1.7	0.00071	0.000035
Trout	0.44	0.0018	0.00010	Rainbow Trout	0.6	0.00044	0.000030	Trout	0.34	0.00024	0.000011
Perch	0.52	0.0012	0.000069	Perch	0.52	0.00036	0.000025	Perch	0.31	0.00021	0.000020
Largemouth Bass	0.28	0.00084	0.000042	Brook Trout	0.51	0.00033	0.000021	Largemouth Bass	0.20	0.00020	0.000011
Rainbow Trout	0.60	0.00069	0.000044	King Chinook Salmon	0.24	0.00032	0.000019	Rainbow Trout	0.27	0.00019	0.000010
Brook Trout	0.51	0.00063	0.000043	Trout	0.44	0.00031	0.000014	Partridge	0.45	0.00017	7.7E-06
King Chinook Salmon	0.24	0.00033	0.000022	Largemouth Bass	0.28	0.00027	0.000014	King Chinook Salmon	0.073	0.000097	4.8E-06
Sum of top 10	12	0.034		Sum of top 10	12	0.018		Sum of Top 10	6.9	0.0099	
Total	32	0.036	6.2E-04	Total	32	0.019	0.00036	Total	25	0.010	0.00019

Supplementary Table C-7 - Summary table of the top 10 Traditional foods contributing to the mean mercury (Hg) and MeHg (MeHg) in Ecozone 3 and for MeHg for WCBA with standard error and the mean consumption (g/d) presented. Results based on FFQ data in a Monte Carlo simulation.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE
Northern Pike	2.5	0.018	0.00078	Pickerel	4.3	0.0097	0.00026	Pickerel	1.6	0.0036	0.000098
Pickerel	4.3	0.016	0.00043	Northern Pike	2.5	0.0083	0.00036	Northern Pike	0.94	0.0031	0.00014
Sturgeon	0.95	0.0026	0.00011	Sturgeon	0.95	0.0014	0.000057	Sturgeon	0.34	0.00049	0.000018
Lake Whitefish	0.80	0.00075	0.000062	Teal	0.55	0.00042	0.000025	Lake Whitefish	0.85	0.00036	0.000032
Brook Trout	0.32	0.00039	0.000041	Lake Whitefish	0.8	0.00033	0.000025	Lake Trout	0.13	0.00022	0.000022
Lake Trout	0.12	0.00037	0.000031	Round Whitefish	0.78	0.00031	0.000024	Mallard	1.0	0.00018	8.0E-06
Round Whitefish	0.78	0.00021	0.000014	Partridge	0.83	0.00029	0.000012	Goose	11	0.00016	4.0E-06
Rainbow Trout	0.19	0.00021	0.000017	Mallard	1.6	0.00028	9.8E-06	Partridge	0.36	0.00013	6.7E-06
Northern Pintail	0.54	0.00020	0.000014	Brook Trout	0.32	0.00024	0.000027	Brook Trout	0.15	0.00010	6.7E-06
Moose Kidney	0.77	0.00016	0.000011	Lake Trout	0.12	0.00020	0.000018	Moose	8.4	0.000085	1.6E-06
Sum of top 10	11	0.039		Sum of top 10	13	0.021		Sum of Top 10	25	0.0084	
Total	53	0.042	0.00090	Total	53	0.022	0.00045	Total	32	0.0087	0.00018

Supplementary Table C-8 - Summary table of the top 10 Traditional foods contributing to the mean mercury (Hg) and MeHg (MeHg) in Ecozone 4 and for MeHg for WCBA with standard error and the mean consumption (g/d) presented. Results based on FFQ data in a Monte Carlo simulation.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE
Pickerel	2.0	0.0077	0.000276	Pickerel	2.0	0.0046	0.00017	Pickerel	1.6	0.0037	0.00016
Perch	1.1	0.0026	0.000124	Smallmouth Bass	0.41	0.0013	0.00011	Perch	1.1	0.00068	0.000039
Smallmouth Bass	0.41	0.0022	0.000178	Perch	1.1	0.00071	0.000039	Smallmouth Bass	0.16	0.00050	0.000032
White Bass	0.37	0.00085	0.000055	King Chinook Salmon	0.36	0.00051	0.000051	King Chinook Salmon	0.33	0.00047	0.000047
King Chinook Salmon	0.36	0.00046	0.000045	Sturgeon	0.13	0.0002	0.000017	Lake Trout	0.18	0.00027	0.000043
Lake Trout	0.12	0.00040	0.000066	Lake Trout	0.12	0.00018	0.000027	Sturgeon	0.102	0.00016	0.000013
Sturgeon	0.13	0.00038	0.000034	Rainbow Trout	0.13	0.000092	0.000015	Corn	12	0.00013	2.3E-06
Largemouth Bass	0.1	0.00027	0.000035	Largemouth Bass	0.099	0.000089	0.000010	Rainbow Trout	0.123	0.000094	0.000013
Northern Pike	0.02	0.00017	0.000017	Northern Pike	0.024	0.000082	8.7E-06	Northern Pike	0.021	0.000070	7.3E-06
Rainbow Trout	0.13	0.00015	0.000022	Bullhead Catfish	0.043	0.000032	5.4E-06	Squash	4.7	0.00049	1.4E-06
Sum of top 10	4.7	0.015		Sum of top 10	4.4	0.0078		Sum of Top 10	20	0.0061	
Total	14	0.015	0.00037	Total	14	0.0083	0.00022	Total	36	0.0066	0.00019

Supplementary Table C-9 - Summary table of the top 10 Traditional foods contributing to the mean mercury (Hg) and MeHg (MeHg) in the **upper 5th percentile** of exposures in Ecozone 1, and for MeHg for WCBA with standard error and the mean consumption (g/d) presented. Results based on FFQ data in a Monte Carlo simulation.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (μg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (μg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (μg/kg/d)	SE
Pickerel	54	0.12	0.012	Pickerel	60	0.26	0.012	Pickerel	23	0.11	0.0047
Northern Pike	29	0.063	0.010	Northern Pike	25	0.11	0.0086	Northern Pike	10	0.051	0.0045
Lake Whitefish	7.9	0.049	0.0028	Lake Trout	6.2	0.014	0.0020	Lake Trout	12	0.035	0.0034
Lake Trout	5.7	0.0085	0.0018	Sturgeon	2.8	0.0063	0.0014	Lake Whitefish	3.8	0.0025	0.00051
Sturgeon	2	0.0071	0.0015	Lake Whitefish	7.7	0.0039	0.00034	Sturgeon	0.46	0.00083	0.00013
White Sucker	2.4	0.0021	0.00047	Sauger	0.66	0.00061	0.00018	Burbot	0.38	0.00027	0.000092
Sauger	0.52	0.0014	0.00078	Burbot	0.64	0.00040	0.000088	Round Whitefish	0.3	0.00023	0.000094
Brook Trout	0.41	0.0010	0.00028	Brook Trout	0.51	0.00038	0.00012	Grouse	0.48	0.00020	0.000079
Trout	0.38	0.00083	0.00021	White Sucker	1.7	0.00027	0.000044	Partridge	0.42	0.00017	0.000044
Moose Kidney	1.3	0.00046	0.00014	Partridge	0.59	0.00025	0.000052	Moose	8.7	0.00011	8.4E-06
Sum of top 20	120	0.25		Sum of top 20	130	0.40		Sum of Top 10	60	0.20	
Total	130	0.25	0.015	Total	140	0.40	0.011	Total	76	0.20	0.0044

Supplementary Table C-10 Summary table of the top 10 Traditional foods contributing to the mean mercury (Hg) and MeHg (MeHg) in the **upper 5th percentile** of exposures in Ecozone 2, and for MeHg for WCBA with standard error and the mean consumption (g/d) presented. Results based on FFQ data in a Monte Carlo simulation.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (μg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (μg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (μg/kg/d)	SE
Northern Pike	4.6	0.055	0.0041	Pickerel	18	0.06	0.0051	Lake Trout	12	0.024	0.0021
Lake Trout	12	0.054	0.0047	Lake Trout	12	0.027	0.0026	Smallmouth Bass	1.3	0.0056	0.00089
Lake Whitefish	4.7	0.0077	0.0012	Northern Pike	5	0.027	0.0020	Round Whitefish	7.5	0.0046	0.00063
Rainbow Trout	1.2	0.0019	0.00049	Lake Whitefish	4.3	0.0032	0.00057	Pickerel	0.86	0.0039	0.00043
Largemouth Bass	0.33	0.0015	0.00043	Rainbow Trout	1.1	0.0014	0.00040	Northern Pike	0.71	0.003	0.00055
Brook Trout	0.61	0.0013	0.00045	Perch	0.93	0.0013	0.00034	Perch	0.79	0.0011	0.00019
King Chinook Salmon	0.23	0.00048	0.00018	Brook Trout	0.79	0.00066	0.00021	Trout	0.62	0.00074	0.00014
Smallmouth Bass	0.061	0.00035	0.00014	Splake Trout	0.049	0.00052	0.00024	Partridge	0.64	0.00043	0.00011
White Bass	0.083	0.00017	0.000046	King Chinook Salmon	0.3	0.00044	0.00010	Rainbow Trout	0.349	0.00042	0.00010
Splake Trout	0.036	0.00014	0.000068	Trout	0.45	0.00043	0.000095				
Sum of top 20	26	0.12		Sum of top 20	55	0.12		Sum of Top 10	25	0.044	
Total	65	0.23	0.0068	Total	64	0.12	0.0047	Total	37	0.045	0.0020

Supplementary Table C-11 - Summary table of the top 10 Traditional foods contributing to the mean mercury (Hg) and MeHg (MeHg) in the **upper 5th percentile** of exposures in Ecozone 3, and for MeHg for WCBA with standard error and the mean consumption (g/d) presented. Results based on FFQ data in a Monte Carlo simulation.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (µg/kg/d)	SE
Northern Pike	19	0.19	0.013	Northern Pike	16	0.083	0.0059	Northern Pike	7.0	0.033	0.0023
Pickerel	17	0.11	0.0060	Pickerel	20	0.077	0.0035	Pickerel	6.7	0.027	0.0014
Sturgeon	1.6	0.0075	0.0015	Sturgeon	1.5	0.0037	0.00075	Lake Whitefish	4.2	0.003	0.00056
Lake Whitefish	1.9	0.0029	0.00086	Brook Trout	0.53	0.00094	0.00040	Lake Trout	0.47	0.0017	0.00038
Brook Trout	0.68	0.0018	0.00067	Teal	0.82	0.00091	0.00024	Sturgeon	0.46	0.001	0.00019
Trout	0.15	0.0011	0.00048	Round Whitefish	0.95	0.00075	0.00029	Mallard	1.2	0.00024	0.000044
Teal	0.64	0.00073	0.00023	Lake Whitefish	1.2	0.00072	0.00028	Goose	15	0.00022	0.000024
Caribou	3.5	0.00059	0.00022	Partridge	0.96	0.00038	0.000064	Partridge	0.47	0.00019	0.000040
Lake Trout	0.13	0.00055	0.00022	Mallard	1.9	0.00036	0.000054	Teal	0.12	0.00011	0.000031
Mallard	1.7	0.00041	0.000051	Lake Trout	0.15	0.00035	0.00011	Moose	8.5	0.000097	9.5E-06
Sum of top 20	82	0.32		Sum of top 20	80	0.17		Sum of Top 10	44	0.067	
Total	86	0.322	0.011	Total	87	0.17	0.005	Total	51	0.067	0.0019

Supplementary Table C-12 - Summary table of the top 10 Traditional foods contributing to the mean mercury (Hg) and MeHg (MeHg) in the **upper 5th percentile** of exposures in Ecozone 4, and for MeHg for WCBA with standard error and the mean consumption (g/d) presented. Results based on FFQ data in a Monte Carlo simulation.

Hg				MeHg				MeHg in WCBA			
Food	Mean Consumption (g/d)	Mean Hg Dose (μg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (μg/kg/d)	SE	Food	Mean Consumption (g/d)	Mean MeHg Dose (μg/kg/d)	SE
Pickerel	14	0.079	0.0041	Pickerel	14	0.048	0.0025	Pickerel	13	0.043	0.0026
Smallmouth Bass	3.7	0.025	0.0033	Smallmouth Bass	3.9	0.016	0.0021	King Chinook Salmon	2.8	0.0059	0.00086
Perch	5.9	0.020	0.0021	King Chinook Salmon	2.6	0.0057	0.00093	Perch	4.0	0.0051	0.00064
Lake Trout	1.1	0.0049	0.0012	Perch	3.6	0.004	0.00063	Lake Trout	1.95	0.0042	0.00081
White Bass	1.0	0.0042	0.00079	Lake Trout	0.65	0.0017	0.00046	Smallmouth Bass	0.67	0.0038	0.00052
King Chinook Salmon	1.4	0.0031	0.00074	Sturgeon	0.26	0.00076	0.00021	Rainbow Trout	0.314	0.00050	0.00020
Largemouth Bass	0.35	0.0016	0.00058	Rainbow Trout	0.36	0.00053	0.00022	Sturgeon	0.167	0.00049	0.00015
Sturgeon	0.19	0.0013	0.00047	Largemouth Bass	0.16	0.00019	0.000098	Northern Pike	0.03	0.00015	0.000068
Rainbow Trout	0.44	0.00072	0.00027	Northern Pike	0.034	0.00018	0.000082	Corn	12	0.00013	0.000011
Northern Pike	0.028	0.00022	0.000094	Bullhead Catfish	0.14	0.00014	0.000065				
Sum of top 20	36	0.14		Sum of top 20	30	0.077		Sum of Top 10	35	0.064	
Total	37	0.14	0.0039	Total	37	0.080	0.0026	Total	56	0.066	0.0020

5. Thesis Conclusion

This thesis assessed the dietary exposures to cadmium, lead, and mercury among adult First Nations living on-reserve in the province of Ontario, Canada. Two complementary methods were applied to assess the dietary intakes of metals in the study populations. The first was a Total Diet Study approach to quantify contaminant intakes from all dietary sources based on a 24-hour recall. The second was a detailed probabilistic exposure assessment using Monte Carlo simulations on traditional food consumption which included annual and seasonal investigations. Results of these assessments indicated that the majority of the population is at low risk of exceeding the dietary reference guidance. The average exposures of lead and mercury were higher than the general Canadian population (1.7 and 1.6 times greater, respectively), whereas average dietary cadmium was 59% lower than the Canadian population average. The upper percentiles of the population exposure distributions were closer to the guidance values, and exposures were characterized with greater detail for the contributing food items. For cadmium exposures, it was noted that smokers had elevated exposures compared to non-smokers, and given the known contribution of cigarette smoking to the cadmium body burden, future investigations should focus on the characterization of this exposure route. Women of childbearing age tended to have lower dietary exposures to mercury than the total population, as well as in contrast to women not of childbearing age and were largely below the more sensitive subpopulation reference value. This may due to the increased awareness of the risk pre-natal exposure to mercury among young women.

The methods used to assess dietary contaminants in this project have benefits in that they are harmonized with practices globally; however there are limitations with their application in the context of this study. Total diet studies are limited in that not all market foods can be quantitatively assesses as consumed by a population, therefore a certain degree of assumptions are made in these assessments. Since this study used market food contaminant concentrations from a secondary data source, consumption of foods reported in the 24-hour recall were match based on primary composition to the available database. Contaminations in Market foods are assumed to be uniform across the country. This is in contrast to the traditional foods included in the total diet study, which are sensitive to local contaminant fluxes. Furthermore, the assessment included traditional foods which were analyzed for contaminant concentrations in their raw, uncooked states due to constraints in the methodology of the FNFNES. This is assumed to have a negligible impact on the results of the study, as studies have demonstrated cooking methods to have varied, although little impacts on the total metal concentrations (Domingo, 2010).

With respect to the assessment of contaminant exposures from annual traditional food consumption, the probabilistic Monte Carlo assessments followed the 14 principles of good practice for Monte Carlo analysis published by Burnmaster and Anderson (1994) to the extent possible given the data set. See Appendix D for more detailed explanation of the simulations conformation to these principles.

The studies presented in this thesis are the first to conduct comprehensive exposure assessment of Cd, Pb, and Hg using a total diet study and probabilistic assessment approach on a provincial and regional (ecozone) scale. The results of this study are representative for the First Nations population in the province, and can be directly integrated into public health planning initiatives as the characterization of risk was presented on an aggregated ecozone level to respect the confidentiality of participating communities, while providing sufficient detail to reflect the regional dietary patterns.

Future research in the area of dietary risk assessments in First Nations communities should adopt a similar ecozone and regional approach to provide results which are representative of the population. Specific emphasis should be on the impact of lead exposures and strategies and initiatives to reduce exposures, and on assessing cigarette smoking behaviour to reduce cadmium exposures.

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Appendix D- Principles of Good Practice for Monte Carlo Analysis

The following is a detailed explanation of the probabilistic assessment models constructed as part of this thesis in comparison to the principles of good practice for Monte Carlo analysis published by Burmaster & Anderson (1994).

Exposures were derived through the application of the following formula, where the contaminant intake for each iteration $_j$ (μg•kg^{-1}•d^{-1}) was modeled based on the sum of the product of the consumption of each food $_i$ *(g•d^{-1})* multiplied by the contaminant concentration $_i$ (μg•g^{-1}), divided by body weight $_j$. The parameter values for each iteration were randomly sampled from distributions using a Monte Carlo sampling method in Oracle Crystal Ball software.

$$\boldsymbol{Contaminant\ Intake_j(\mu g/kg/d) = \sum_{i=1}^{69} \frac{[food_i(g/d)]\ x\ [contaminant]_i(\mu g/g)}{Body\ Weight_j(kg)}}$$

Percentiles of exposures where contrasted to reference values using a hazard quotient method, wherein the exposure was divided by the reference value. Reference values were obtained from published values from international agencies such as JECFA, and the national values published by Health Canada. Hazard quotients greater than a value of one indicated increased population risk, and the percentile of the population which exceeded this value was reported.

The distributions of traditional food consumption in grams per day were modelled with non-parametric methods using Crystal Ball's custom distribution function. To convert the frequency of consumption capture by the FFQ to grams consumed per day, age and sex specific portion sizes for different traditional foods types were applied to characterize the variability in the population (Chan et al., 2011). The portion sizes were not included probabilistically as distributions as it was determined that this level of complexity was unwarranted at the current stage given the large sample size and therefore characterization of population variability in consumption amounts as an input factor. Traditional foods items that were included in the simulation were limited to those with more than 5% of the population reporting consumption to limit the amount of inputs to those of relative importance.

Given the participatory study design of the FNFNES, the composition of traditional food samples collected varied across ecozones. Driven by availability and priorities set at the community level, the representation of traditional foods items analyzed for contaminants provided insufficient information at the ecozone level to conduct comprehensive exposure assessments with ecozone specific contaminant data. Furthermore, the locations of where food items were hunted or gathered were nonspecifically characterized for most samples, with some coming from ecozones other than the one the donating community was located within. Therefore, the contaminant concentrations in the assessment were an aggregate of results from across the province of Ontario. The expected impacts of this on the final assessment results are deemed to be minimal and potentially representative of modern hunting ranges from increased mobility.

Concentration of contaminants in traditional food items were characterized in the simulation by using Crystal Ball's parametric methods that fitted lognormal distributions to the parameters of the mean and standard deviation. The assumption of contaminant concentrations assuming a lognormal distribution has been followed in other dietary assessment of contaminant exposures which included traditional foods (Chan et al., 1997; Laird et al., 2013b). The standard deviation was assumed to be equal to the mean in the simulation (100% coefficient of variation). This coefficient of variation was assumed to account for the uncertainty arising from inconsistent composite sample sizes. Table 6-1 presents a summary of the coefficient of variations calculated as part of the analysis of metal concentrations which supports that this assumption reasonably represents the observed variation. Table 2 presents a summary of distribution characteristics for the input parameters in the simulation models.

Table 6-1 - Summary of coefficient of variation for traditional foods analyzed for contaminants through the FNFNES (2011-2012).

		Cd			Hg			MeHg			Pb		
	n	n[a]	Average	Range	n[a]	Average	Range	n[a]	Average	Range	n[a]	Average	Range
All Samples	**418**	**60**	**109%**	**14-310%**	**44**	**69%**	**20-139%**	**18**	**62%**	**9-157%**	**64**	**104%**	**10-347%**
Fish	101	12	135%	74-258%	15	67%	21-121%	12	73%	9-157%	11	105%	22-249%
Game	120	18	111%	22-310%	16	79%	25-133%	2	49%	44-53%	17	144%	46-347%
Mushrooms	3	0	-		0	-		0	-		0	-	
Roots/ Tea	33	4	71%	50-100%							7	62%	40-95%
Tree Foods	11	2	126%	97-155%							2	96%	84-109%
Vegetables	93	14	97%	14-165%	3	42%	33-50%				17	80%	25-176%
Water Fowl	57	10	104%	28-168%	10	65%	20-139%	4	35%	11-72%	10	105%	10-194%

a – number of composite samples with standard deviation

Table 6-2 - Summary of input distribution parameters

Contaminant Group	Distribution	Central Tendency	Truncation Min	Truncation Max	Method for inclusion of Non Detects
Metals (ug/g)	Lognormal	μ, SD	LOD/2	μ + 3(SD)	LOD/2
TF Consumption (g/d)	Custom		Defined by data		N/A
Body Weight (kg)	Custom		Defined by data		N/A

The models constructed to estimate the exposures to metal contaminants used primary source data from the FNFNES. The data was collected in a participatory research framework which integrated traditional knowledge inputs in the design and execution of the data collection, and communicated all results and data to participating communities as per the principles of ownership, control, access, and possession (OCAP).

Moderate-to-strong correlations (ρ>0.6) in input parameters in a simulation may have an effect on the tail regions of the output distributions. To investigate the presence of correlations in the traditional food consumption parameter Spearman's Rank Correlation coefficients (ρ) were computed. Table 6-2 presents traditional food pairings which had correlations greater than 0.6. Traditional foods with strong correlations ($\rho > |0.6|$), with a $p<0.0001$ were included in a model for the purpose of a sensitivity analysis. The difference between the simulation with strong correlations and the base case was a change in the total dose ranging from -18% to 12.5% in the mean doses of metals, and -19.78 to 9.76% in doses at the 95th percentile. The differences, although statistically significant, were largely a decline in the total dose with the inclusion of correlations between the frequencies of traditional foods consumed. Therefore, it was decided to exclude correlations in the simulation model to err on the conservative side of total dose estimates, and assume that all input variables were independent. Furthermore, the independence of input variables is supported by the nature of how traditional foods are not consistently available for pairings as suggested by correlating food items (e.g. goose and moose meat) which present a correlation only when annual traditional food consumption is evaluated without consideration for seasonal availability.

Table 6-3 - FFQ input variables with strong correlations (p> |0.6|) in ecozones

Ecozone	Traditional Food Item	Correlation	P
Province	Caribou Liver & Caribou Kidney	0.82	<0.0001
	Beans & Corn	0.70	<0.0001
	Moose Liver & Moose Kidney	0.69	<0.0001
	Goose & Moose Meat	0.64	<0.0001
Ecozone1	Sturgeon & Lake Whitefish	0.72	<0.0001
	Pickerel & Moose	0.60	<0.0001
	Moose Liver & Moose Kidney	0.71	<0.0001
	Caribou Liver & Caribou Kidney	0.80	<0.0001
	Goose & Whitefish	0.73	<0.0001
	Goose & Sturgeon	0.63	<0.0001
	Goose & Moose	0.65	<0.0001
Ecozone2	Fiddlehead & Dandelions	0.64	<0.0001
	Crab Apples & Mint Leaves	0.63	<0.0001
Ecozone3	Trout & Rainbow Trout	0.67	<0.0001
	Moose Liver & Moose Kidney	0.70	<0.0001
	Caribou Liver & Caribou Kidney	0.86	<0.0001
	Trout & Merganser	0.64	<0.0001
	Rainbow Trout & Merganser	0.67	<0.0001
Ecozone4	Lake Trout & Smelt	0.62	<0.0001

A one-way analysis of variance was conducted in JMP to evaluate the difference between total amounts of traditional food consumed in grams per day between ecozones. An F-ratio of 31.88 with a $p<0.001$ indicated a statistically significant difference between ecozone groups. Therefore, it was decided to conduct the exposure assessment for the total population as well as for each ecozone specifically. This would provide insight on trends that could support ecozone-specific strategies.

The number of iterations selected for simulations was 10 000 with an initial seed for the random generator of 999. To evaluate the numerical stability of simulation outputs of total doses for cadmium, mercury, methyl mercury, and lead, two simulations varying in initial starting seed values were run per iterations of 8 000 to 15 000 in increasing increments of 1 000, as well as simulations with 20 000 and 50,000 iterations. This sensitivity analysis was performed on the provincial simulation. Results of the contaminant dose outputs were averaged between the two simulations per iteration setting and compared to previous value to assess the percent change of increasing the iteration magnitude by 1000. Comparisons of the percentage change at the mean, standard error of the mean and 95th percentile estimate were made. With the addition of adding 1 000 simulations, there was a less than 5% change over the previous simulation for the mean and 95th percentiles of the total dose estimates for mercury, methyl mercury, and lead between 8 000 to 15 000 iterations, as well as comparisons between 20 000 and 50 000 iterations. The estimates for cadmium exhibited variability ranged up to 8.4% for the mean and 8.9% for the 95th percentile between the simulation with 14 000 and 15 000, and 13 000 and 14 000, respectively. Results for cadmium exhibit less stability than those of the other metal contaminants in part due to the sensitivity of the key inputs driving the cumulative dose (see supplemental tables in cadmium exposure assessment chapter to see the percentage contribution to the variance of the consumption of top food items to the total dose). Dose outputs of the simulation with 10 000 iterations was contrasted to those from simulations with 20 000 and 50 000 iterations to find a less than 5% difference in the mean and 95th percentile estimates for mercury, methyl mercury and lead; whereas cadmium continued to vary between 8 and 10% for the mean and 95th percentiles, respectively.

The standard error of the mean had an inverse relationship to the increase in iterations (Figure 6-1). This is to be expected with an increased sample size. However, given the limited differences in dose estimates for increasing the number of iterations, the precision at 10 000 iterations provides estimates of sufficient stability for the context of the present exposure assessment.

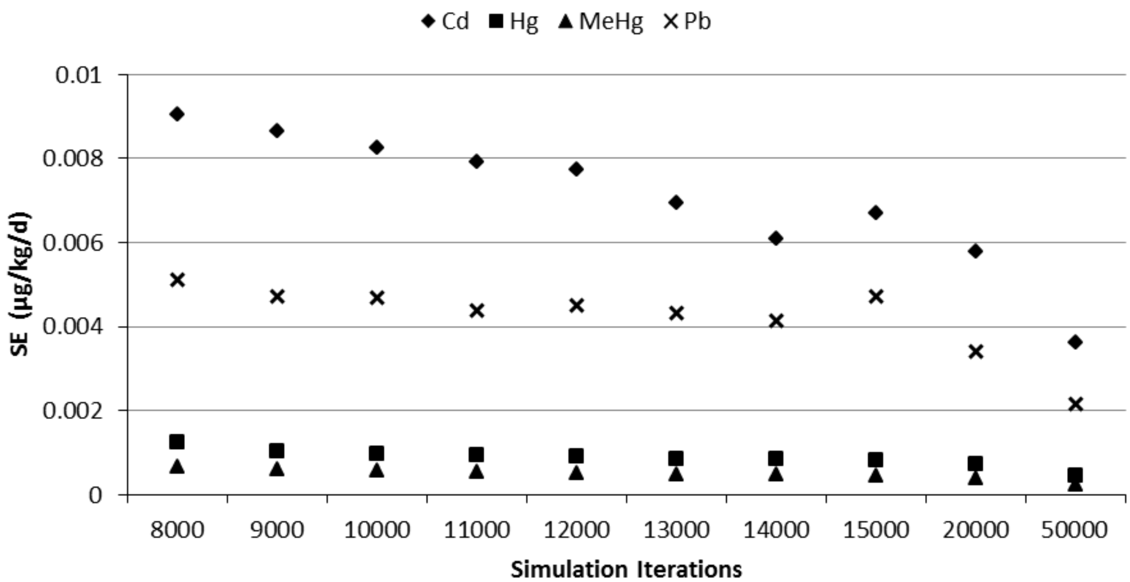

Figure 6-1 – Average Standard Error of the mean (SE) of two simulations for Cd, Hg, MeHg, and Pb at different amounts of iterations.

The random number generator in Oracle Crystal Ball uses the Multiplicative Congruential Generator method. The length of the iteration chain is 2^{31} or 2,147,483,646 before the series is repeated. The random number seed for the simulation was 999. For replicability, future simulations should begin at this seed to ensure a consistent sequence of iterations and simulation results. The formula this number generator is defined by is:

$$r \leftarrow (62089911 \cdot r) \bmod (2^{31} - 1)$$

A limitation in the consumption data is that it is based on self-reported amounts. This has the potential to introduce self-reporting bias into the data as people may either over or under report traditional food consumption. Another source of bias is the unknown collection time, geographic location, and hunting method of traditional food samples. This study was unable to differentiate seasonal trends in contaminant concentrations of hunted and harvested traditional foods. However, given that the availability of certain traditional foods is often limited to certain seasons, the samples provided by communities are likely representative of the traditional foods when consumed by the majority of the population. Further to this, traditional food samples were collected using a food drive style of collection where community members were asked to donate food samples from their personal store. This increases the representativeness of the data as the foods reflect those that would have been consumed by community members.