Environmental Research 158 (2017) 409–420

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

A total diet study and probabilistic assessment risk assessment of dietary mercury exposure among First Nations living on-reserve in Ontario, Canada

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ARTICLE INFO

Keywords:
Methyl mercury
First Nations
Ontario
Fish consumption
Exposure assessment

ABSTRACT

Methyl Mercury (MeHg) exposure is a global environmental health concern. Indigenous peoples around the world are susceptible to MeHg exposure from often higher fish consumption compared to general populations. The objective of this study was to estimate dietary exposure to methylmercury (MeHg) among First Nations living on-reserve in the province of Ontario, Canada. A total diet study was constructed based on a 24-h recall from the First Nations Food, Nutrition, and Environment Study (FNFNES), and measured contaminant concentrations from Health Canada for market foods, and FNFNES for traditional foods. A probabilistic assessment of annual and seasonal traditional food consumptions was conducted for 1429 adult participants. Results were compared to exposures in the general Canadian population and reference values from Health Canada for adults and women of childbearing age (ages 19–50). Results indicated traditional foods to be the primary contributor to the dietary total MeHg intake (72%). The average dietary total MeHg 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 the reference values. Mercury concentrations in participants' hair samples (n = 744) ranged from 0.03 to 13.54 μg/g, with an average of 0.64 μg/g (geometric average of 0.27 μg/g). Less than 1% of the population had a hair mercury value above the 6 μg/g level, and 1.3% of women of child bearing age had values greater than 2 μg/g. Fish species contributing to the MeHg intake included pickerel-walleye, pike, perch and trout. Only 7.9% of the population met the recommended fish consumption rate of two, 3.5 oz servings per week from the American Heart Association. Therefore, consumption of lower trophic level fish can be promoted to provide the maximum nutritional benefit with minimal risk of MeHg exposure.

1. Introduction

Mercury is a ubiquitous environmental global pollutant causing an increasing public health concern (Sheehan et al., 2014; WHO, 2010). Human activities have released large quantities of mercury to the environment, greatly enriching concentrations relative to natural levels (Depew et al., 2013; Streets et al., 2017; Trip et al., 2000; Wang et al., 2004). Mercury emissions are distributed globally though the atmosphere and deposited into ecosystems where they 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; Hsu-Kim et al., 2013). 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, 2013).

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

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http://dx.doi.org/10.1016/j.envres.2017.06.025
Received 23 April 2017; Received in revised form 19 June 2017; Accepted 21 June 2017

marine species consumption (Grandjean et al., 1997; 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, resulting 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 Canada, mercury has been a priority area of study for Aboriginal populations due to documented elevated exposures compared to the general Canadian population (Donaldson et al., 2010). 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 a national biomonitoring program (1970–1992) (Wheatley and Paradis, 1995) and 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 Canadian Arctic. The history of industrialization in this province includes seven chlor-alkali plants operating between the 1930's to 1990's 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 reserves of Grassy Narrows and Wabaseemong situated on the English-Wabigoon river system where approximately 10 metric tonnes of inorganic mercury was discharged into the river system in the 1960's, prompting consumption and sport-fishing bans on locally caught fish due to elevated levels of MeHg of up to 20 mg/Kg wet weight (Kinghorn et al., 2007). 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, (Neff et al., 2012), and biomonitoring data from community members reflecting elevated blood MeHg levels of up to 323 μg/L (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). 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 in this province (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 (Kuhnlein, 1995). 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). 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 1 cm growth portions represents a month of exposure. Hair is commonly used as an integrated exposure 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). However, both hair and blood mercury levels do not provide insights on the sources of exposure which is why comprehensive exposure characterization exercise are necessary.

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 disease 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 that cardiovascular health should be a public health priority for this population (Reading, 2015). In addition to the rising prevalence of cardiovascular disease, there has been a rise to epidemic proportions of chronic metabolic morbidity among First Nations. Diseases such as diabetes, obesity, and chronic kidney disease are 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 Indigenous 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 exposure to MeHg in First Nations peoples in Ontario 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 Indigenous populations, as well as contribute to the global call for research on mercury exposures in sensitive populations.

2. Method

2.1. Ethics

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

2.2. Traditional food samples & analysis

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). A total of 18 First Nation communities from the province of Ontario, were selected to participate based on a

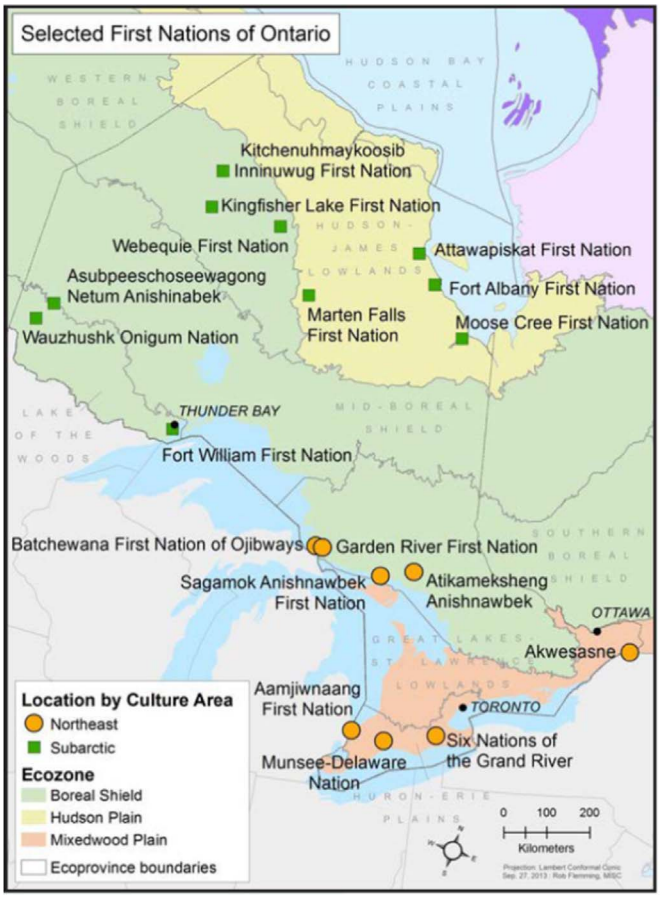

Fig. 1. Map of participating First Nations communities and four ecozones in Ontario (Chan et al., 2014).

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 (Ecological Stratification Working Group, 1995): the Boreal Shield, the Hudson Plains, and the Mixedwood Plains; and two cultural areas (Sturtevant, 1978): 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) (Fig. 1). 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-identified as being a First Nation person living on-reserve in Ontario; and whose birthday was next. A total of 1429 individuals participated (Chan et al., 2014).

All participating individuals completed a household interview which included the following sections: a 24-h dietary recall; traditional food frequency questionnaire (FFQ); socio/health/lifestyle questionnaire; and food security questionnaire. For information on survey data collection, refer to Chan et al. (2014).

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

2.2.1. *Exposure characterization*

Two complementary methods were applied to assess dietary MeHg exposure in the study population. The first was a Total Diet Study to determine MeHg exposures from all sources of food in the diet (market foods and traditional foods). The second method was a probabilistic exposure model exploring MeHg exposures from traditional food consumption reported throughout the year using a food frequency questionnaire. The probabilistic exposure modelling provided more detailed insight on the leading dietary contributors to MeHg population exposures and seasonal trends.

2.2.1.1. 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-h recall survey, the intake of recalled food items (in grams) was multiplied by the concentration of mercury in that food item (μg/g) based on a database of contaminant concentrations to determine the dietary mercury intake. Total mercury intakes were summed per participant and divided by the individual's body weight to derive an individual's mercury expsoure. Total mercury is not routinely included in the assessment of Canadian market foods through the Canadian Total Diet Study. Therefore to facilitate the total dietary assessment for this contaminant, mercury concentrations from 1998 to 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 total 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 (0.004 μg/g), and water (0.005 μg/g). Dietary mercury intakes have been reported as using survey weights to account for factors such as design weight (the inverse of the selection probability) and adjustment factors (non-response rate).

2.2.1.2. Hair analysis. A total of 744 participants provided hair samples for mercury analysis. Samples were taken as a 5 mm 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 with the analysis conducted on the first three segments. Each 1 cm 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.

2.2.1.3. Traditional food probabilistic exposure assessment. A probabilistic approach was used to estimate Hg 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 the 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). Mercury intake for each iteration $_j$ ($\mu g \cdot kg^{-1} \cdot d^{-1}$) was modeled based on the sum of the product of the consumption of each food $_i$ ($g \cdot d^{-1}$) by mercury concentration $_i$ ($\mu g \cdot g^{-1}$), divided by body weight $_j$ (Eq. (1)).

$$\boldsymbol{HgIntake_j(\mu g/kg/d)} = \sum_{i=1}^{69} \frac{[\boldsymbol{food_i(g/d)}]\boldsymbol{x}[\boldsymbol{Hg}]_i(\mu g/g)}{\boldsymbol{BodyWeight_j(kg)}} \quad (1)$$

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-h 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 at the regional level, 47 in Ecozone 1, 63 in Ecozone 2, 41 in Ecozone 3, and 55 in Ecozone 4. Consumption data were parameterized using the custom distribution function in Crystal Ball as the sample data were representative of the provincial and ecozone populations of First Nations in Ontario. Input distributions of Hg concentrations in each traditional food item was represented through FNFNES traditional food composite analysis fitted to lognormal distributions described by the average, the standard deviation derived as an assumed coefficient of variation of 100%, and bounded by LOD/2 and three standard deviations. Body weight data were obtained through the FNFNES and included 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.

2.2.1.4. Risk assessment of dietary intake. Dietary intakes of total Hg were contrasted to the Health Canada provisional tolerable daily intake (PTDI) for MeHg of 0.47 μg/kg bw/d for adults (Environment Canada and Health Canada, 2010), and 0.2 μg/kg bw/d for women of childbearing age (ages 19–50) published by Health Canada (Health Canada Mercury Issues Task Group, 2004). 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. Reporting of results will be presented as MeHg for simplicity. 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 compared to 6 μg/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 compared to the increased risk level of 2 μg/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 mercury 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.

2.2.1.5. 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 $p < 0.05$.

3. Results

3.1. Total diet study

The average mercury exposure from the total diet of First Nations living on reserve in Ontario was 0.039 μg/kg/d. At the ecozone level, mean exposures ranged from 0.018–0.064 μg/kg/d, with the total dietary mercury being significantly different between ecozones 3 and 4 (Wilcoxon each pair test; $p = 0.0148$) (Table 1). The total mercury exposure was significantly higher among First Nation adults reporting traditional food consumption compared to adults not reporting it in the region and at each ecozone (Kruskal Wallis test; $p < 0.0001$).

A summary of total dietary mercury exposures by population

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

		n	Mean	SE	50th	90th	95th	97.5th	99th
Total Population	Ontario	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 Food Consumers	Ontario	1239	0.011	0.00098	0.0056	0.013	0.019	0.057	0.19
	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	Ontario	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

Table 2
Summary of dietary Mercury (MeHg) (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
Ontario		1429	0.039	0.0049	0.0061	0.12	0.89
Age Group	19–30	265	0.023	0.0065	0.0061	0.017	0.73
	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
	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

demographic is presented in Table 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 +) 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 aged 51 + , however this finding was not statistically significant except in ecozone 3 ($p = 0.002$). At the 95th percentiles of mercury exposures in the female population, women of childbearing age had lower exposures than women 51 + years of age.

Traditional foods accounted for 72% of the average mercury exposure in the province, despite only accounting for 1.8% of the average caloric intake. However, between ecozones, the contribution of traditional foods to the total dietary mercury exposure varied between 23% in ecozone 4–88% 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 3. Canned fish was the leading contributor for market food sources. Table 4 presents the top 5 traditional foods contributing to the mean mercury exposure from the total diet. Pickerel-walleye, trout, and perch were among the top traditional foods, all of which are predatory fish. Table 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 0.5 μg/g, with the exception of pike which had an average mercury concentration of 0.63 ± 0.81 μg/g (0.30 ± 0.28 μg/g MeHg).

3.2. Assessment of annual traditional food consumption

MeHg exposure estimates from annual traditional food consumption are presented in Table 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 5). For MeHg, all seasons were statistically different 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 observed in each ecozone (Supplementary Table).

The top 10 traditional foods contributing to the total MeHg doses for First Nations adults living on-reserve in Ontario are presented in Table 7, with the top 10 traditional foods for the upper 5th percent of the population are presented in Table 8. The major foods contributing to MeHg intakes were pickerel-walleye, northern pike, and trout.

3.3. Hair mercury

A total of 744 participants in the FNFNES provided hair samples for mercury analysis. Hair mercury concentrations ranged from 0.03 to 13.54 μg/g, with an average of 0.64 μg/g (geometric average of 0.27 μg/g) as calculated with population weights applied. Less than 1% of the total First Nations adult population had a hair mercury value above the 6 μg/g level which is associated with increased risk for adults established by Health Canada (Environment Canada & Health Canada, 2010). In women of childbearing age, 1.3% had hair mercury values above the increased risk level of 2 μg/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 7% of the women of childbearing age in this region exceeding the 2 μg/g increased risk level.

There was a significant association between hair mercury concentration and estimated total dietary mercury from the 24-h 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 ($\rho = 0.23$; $p = 0.0043$). Fig. 2 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-h recall. In contrast, traditional food consumers had a significant positive correlation between total dietary mercury and hair mercury.

Percentiles of seasonal mercury intakes were plotted against percentiles of hair mercury (Fig. 3a) 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. Fig. 3b illustrates percentiles of mercury intake from the total diet, market food sources, and traditional food sources from the 24-h recall compared to hair mercury percentiles. The total dietary

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

Ontario				Ecozone 1				Ecozone 2			
Food[a]	Mean MeHg Dose (μg/kg/d)	SE	% of Total Dose	Food	Mean MeHg Dose (μg/kg/d)	SE	% of Total Dose	Food	Mean MeHg 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 MeHg Dose	0.039	0.0049		Total MeHg Dose	0.064	0.013		Total MeHg Dose	0.026	0.0094	

Ecozone 3				Ecozone 4			
Food	Mean MeHg Dose (μg/kg/d)	SE	% of Total Dose	Food	Mean MeHg 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 MeHg Dose	0.018	0.0040		Total MeHg Dose	0.020	0.0034	

[a] Types of fish was differentiated as market food versus traditional food based on coding in the 24-h recall which included terms such as “Fast Food” or fish products identified with a brand.

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 ($p = 0.0056$), suggesting that the total dietary mercury explains approximately only 4% of the hair mercury values ($p < 0.003$), likely because hair mercury values were highly variable across Ontario and within each ecozone.

3.4. Risk assessment

Dietary mercury intakes from the total diet were largely below the PTDI of 0.47 $\mu g/kg_{bw}/d$ for MeHg for the adult population (assuming that total mercury exposure was equivalent to MeHg exposure). At the regional or provincial level, this reference dose was exceeded at the 97.8th percentile, while the ecozones exceeded this value at the following percentiles: 94.6th (ecozone 1), 98.6th (ecozone 2), 99.5th (ecozone 3), and 99.2nd (ecozone 4). Among the exceedance in the province, 94% reported traditional food consumption. As shown in Table 1, traditional food consumers had higher total dietary mercury levels than those who did not report traditional food consumption on the 24-h recall prior to being interviewed. In the subpopulation of

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

Ontario				Ecozone 1				Ecozone 2			
Food	Mean MeHg Dose (μg/kg/d)	SE	% of Total Dose	Food	Mean MeHg Dose (μg/kg/d)	SE	% of Total Dose	Food	Mean MeHg Dose (μg/kg/d)	SE	% of Total Dose
Pickerel-walleye	0.012	0.0030	32%	Pickerel-walleye	0.025	0.0075	40%	Pickerel-walleye	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 MeHg Dose	0.039	0.0049		Total MeHg Dose	0.064	0.013		Total MeHg Dose	0.026	0.0094	

Ecozone 3				Ecozone 4			
Food	Mean MeHg Dose (μg/kg/d)	SE	% of Total Dose	Food	Mean MeHg 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-walleye	0.0014	0.00099	6.8%
Moose Meat	0.0015	0.00027	8.4%	Deer Meat	0.000040	0.000020	0.18%
Pickerel-walleye	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 MeHg Dose	0.018	0.0040		Total MeHg Dose	0.020	0.0034	

Table 5
Summary of total MeHgdoses (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
Total Mercury (MeHg)	Ontario	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) **WCBA**	Ontario	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 6
Concentrations of total mercury (Hg) and methyl mercury (MeHg) in top consumed traditional food items. Average with standard deviation (SD) and range is presented.

			Total Hg		MeHg	
	n (composites)	n (replicates)	Average (SD)	Range (min-max)	Average (SD)	Range (min-max)
Pickerel-walleye	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

traditional food consumers (n = 190), the reference dose for adults (0.47 μg/kg_{bw}/d) was exceeded at the 85th percentile. In the sub-population of women of childbearing age, the total dietary mercury exposure was compared to the sensitive reference dose of 0.2 μg/kg_{bw}/d. At the provincial level, this reference dose was exceeded at the 89.7th percentile, while at the ecozone level only ecozones 1 and 3 exceeded this value (65.8th and 92.8th percentiles respectively) (Fig. 4).

Comparing the results from the assessment of annual traditional food consumption to the reference values demonstrated similar trends to the results of the 24-h recall. Fig. 3 shows the population distribution of the hazard quotient (risk) for MeHg exposures (based on the reference dose of 0.47 μg/kg_{bw}/d). The First Nations adult population at the provincial level (or regional level) 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. Fig. 5 shows the MeHg hazard quotient (risk) distribution the women of childbearing age subpopulation. Similar trends were observed in this population as in the general adult population, despite a comparison to a lower reference dose (0.2 μg/kg_{bw}/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.

4. Discussion

First Nations living on-reserve in Ontario have elevated mercury exposures compared to the general Canadian population; however the population is largely below the dietary reference suggesting low population risk. 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 = 0.022 μg/kg/d) as published by Dabeka et al. (2003). At the ecozone

Table 7
Summary of mean consumption (g/d) and total 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 First Nations adult population and Women of Child-Bearing Age (WCBA) sub population.

Total MeHg				MeHg in **WCBA**			
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-walleye	5.6	0.021	0.00066	Pickerel-walleye	3.0	0.0071	0.00020
Northern Pike	1.7	0.012	0.00064	Northern Pike	0.84	0.0029	0.00016
Lake Trout	1.0	0.003	0.00022	Lake Trout	0.90	0.0013	0.00011
Lake Whitefish	2.5	0.0024	0.00012	Lake Whitefish	1.5	0.00063	0.000033
Sturgeon	0.52	0.0014	0.000071	Smallmouth Bass	0.15	0.00048	0.000032
Smallmouth Bass	0.27	0.0013	0.00012	Sturgeon	0.20	0.00030	0.000015
Perch	0.50	0.0010	0.000062	Perch	0.34	0.00023	0.000024
Trout	0.21	0.00087	0.000069	King Chinook Salmon	0.15	0.00020	0.000026
White Sucker	0.46	0.00039	0.000050	Partridge	0.29	0.00010	0.0000060
Brook Trout	0.26	0.00035	0.000032	Mallard	0.41	0.000072	0.0000071
Sum of top 10	13	0.044		Sum of Top 10	7.8	0.013	
Total	38	0.047	0.00097	Total	27	0.014	0.00029

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

Total MeHg				MeHg in WCBA			
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-walleye	30	0.18	0.010	Pickerel-walleye	14	0.056	0.0028
Northern Pike	14	0.13	0.011	Northern Pike	5.9	0.031	0.0026
Lake Trout	5	0.023	0.0038	Lake Trout	7.4	0.015	0.0020
Smallmouth Bass	1.2	0.0080	0.0019	Lake Whitefish	3.5	0.0023	0.00045
Lake Whitefish	5.1	0.0078	0.0015	Smallmouth Bass	0.31	0.0017	0.00042
Sturgeon	0.55	0.0021	0.00062	King Chinook Salmon	0.54	0.0013	0.00041
Trout	0.30	0.0019	0.00062	Perch	0.55	0.00086	0.00031
Perch	0.69	0.0015	0.00034	Sturgeon	0.18	0.00031	0.000074
White Sucker	0.85	0.0015	0.00075	Trout	0.069	0.00011	0.000044
Brook Trout	0.65	0.00092	0.00028	Mallard	0.45	0.000086	0.000028
Sum of top 20	58	0.36		Sum of Top 10	33	0.11	
Total	83	0.36	0.011	Total	54	0.11	0.0029

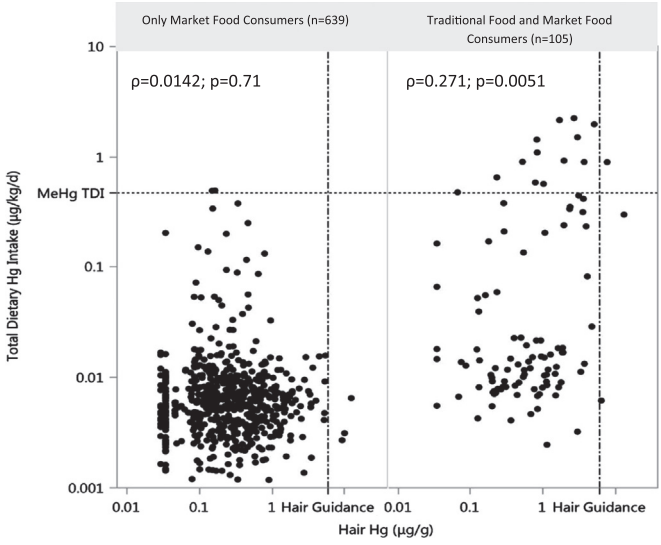

Fig. 2. Hair Hg (μ/g) versus total dietary MeHg (μg/kg/d) from 24-h recall data in consumers of traditional and market foods (n = 105) and consumers of only market foods (n = 639).

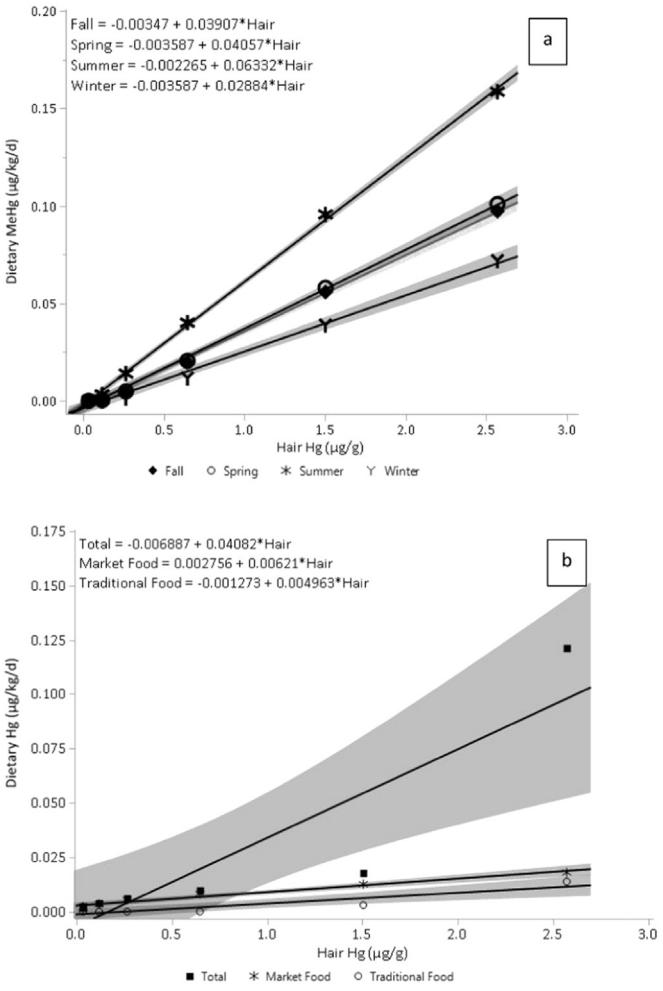

Fig. 3. (a) Plot of quantiles (5th–95th) of seasonal Hg Intake (μg/kg/d) versus quantiles of hair Hg (μg/g); (b) Plot of quantiles (5th–95th) of total dietary, traditional food, and market food MeHg Intake (μg/kg/d) from Total Diet study versus quantiles of hair.

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 (0.023 μg/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 (0.019–0.030 μg/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). Among the traditional food consumers, 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). 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 0.05 μg/kg/d (Legrand et al., 2005). Mercury exposure from traditional food in this study is also lower than the average of 7.9 μg/kg/wk reported for Inuit in the Inuvialuit Settlement Region, Nunavut, and Nunatsiavut jurisdictions (Laird et al. (2013). First Nations populations have different dietary profiles from Inuit who consume marine mammals that contribute to the mercury exposure, and different terrestrial game (i.e. caribou), which explains the difference and supports the need for regional and culturally 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

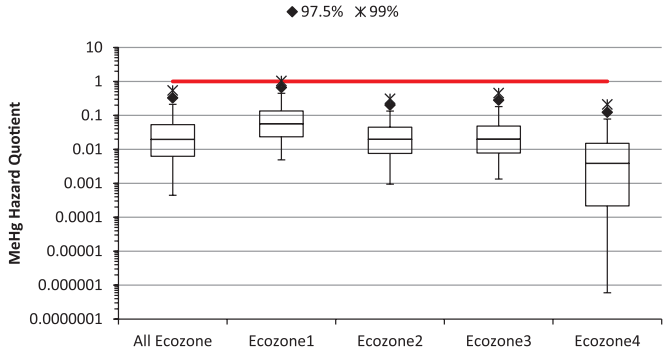

Fig. 4. Distribution of MeHg hazard quotients from 5th–95th percentile of the total adult population with indicators for 97.5 and 99th percentile points (hazard quotient based on a reference dose of 0.47 μg/kg$_{bw}$/d). Hazard quotient of 1, which indicated increased risk, has been indicated to facilitate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

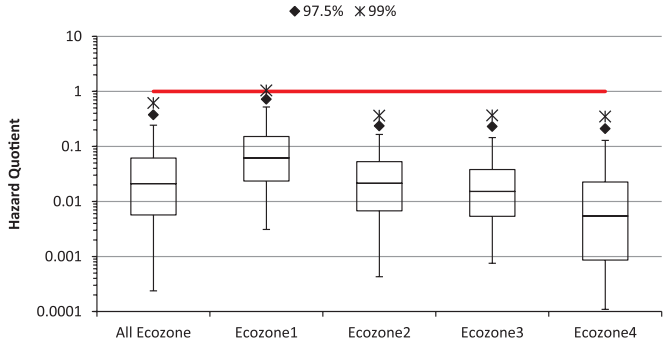

Fig. 5. Distribution of MeHg hazard quotients from 5th–95th percentile of the women of childbearing age population(19–50 y years of age) with indicators for 97.5 and 99th percentile points (hazard quotient based on a reference dose of 0.2 μg/kg$_{bw}$/d). Hazard quotient of 1, which indicated increased 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).

herbivorous terrestrial animals (Chan and Receveur, 2000; Hansen and Gilman, 2005).

Based on data from the FNFNES 24-h recall, an average of 16 g of fish were consumed per day (112 g/week) by First Nations living on-reserve in the province of Ontario; however in the subpopulation that report fish consumption from either market or traditional foods, fish constituted an average intake of 190 g per day. In the FFQ, an average of 14 g per day (98 g/week) of fish was consumed (median: 27 g/d; 95th percentile: 108 g/d). The majority of the population is therefore below the recommended two 3.5 oz weekly servings (200 g) recommended by the American Heart Association, with only 7.9% meeting the guidance based on the 24 h 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; Health Canada, 2007). High exposure 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). In this study, pickerel-walleye and pike account for 55% of fish intake and 75% of the mercury exposure. These two species have well documented elevated concentrations. This suggests 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-walleye 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-walleye 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 higher consumption of predatory fish species that are high in mercury such as small and largemouth bass, pickerel-walleye, northern pike, and yellow perch than the general population (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 (0.5 ppm). Among market foods, canned fish was among the top contributors to the mercury exposure for the First Nations living on-reserve in the province of Ontario, 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). A study in the Great Lakes region found that pickerel-walleye had mercury concentrations ranging from 0.22–0.66 μg/g which was within the range in of FNFNES data (0.34 ± 0.23 μg/g), and Northern Pike Hg concentrations ranged from 0.40 to 0.60 μg/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 the 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 (Nova Scotia, New Brunswick, Prince Edward Island), 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 before emissions became 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 remobilization has a local 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 and direct measures of mercury in local fish.

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-h 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 Indigenous 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 findings, as estimations of blood mercury from hair mercury samples systematically over-estimated mercury burdens for males, and underestimation 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 finding could explain the variability observed in hair mercury concentrations in this study, as traditional foods were consumed 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-h dietary recalls and food frequency questionnaires (FFQs), were employed in this study and have been validated as useful 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-h recall). This is likely due to the 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. In the FNFNES study design, the FFQ only captured traditional food consumption, and the 24-h recall was conducted in the fall season. Therefore trends on market food consumptions and their contribution to the variability are unknown.

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 analyzed in 2011–2012) would have negligible impact on the total dietary mercury exposures as concentrations in market food remain fairly stable. The 24-h recall employed in this study did not differentiate the type of tuna, and in the Canadian market food assessment, canned fish is not differentiated based on the source either. 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.

5. 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 indicate 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-h recall survey for increasing omega-3 nutrient intakes. Consumption of lower trophic level fish, which are lower in mercury, is to be promoted to meet this advice.

Acknowledgements

We would like to express our gratitude to all participants for their cooperation and participation in the First Nations Food, Nutrition and Environment Study (FNFNES). We thank all First Nations community members who collected food and water samples, assisted in data collection, coordinated research activities, and arranged meetings and public gathering to share information. FNFNES is funded by Health Canada and the communication of the results to the communities was also funded by the Canadian Institute of Health Research. A.K.Juric was supported by a NSERC-CREATE Research in Environmental Analytical Chemistry and Toxicology scholarship. H.M. Chan is supported by the Canada Research Chair Program. The authors have no potential conflicts of interest.

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.envres.2017.06.025.

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