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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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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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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