Local country food sources of methylmercury, selenium and omega-3 fatty acids in Nunavik, Northern Quebec

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A B S T R A C T

Country foods are central to Inuit culture and replete in selenium (Se) and long-chain omega-3 polyunsaturated fatty acids (n−3 PUFA). However, some marine country foods bioaccumulate high concentrations of methylmercury (MeHg). Se and n−3 are associated with several health benefits in Nunavik, Northern Quebec, but, recent studies show that prenatal MeHg exposure is associated with visual, cognitive and behavioral deficit later in childhood. The study objectives are to identify contemporary country food sources of MeHg, Se and long-chain n−3 PUFA in Nunavik, particularly among childbearing-age women, taking into account regional differences in consumption profiles. The contribution of different country foods to daily MeHg, Se, long-chain n−3 PUFA intake (µg/kg body weight/day) was estimated using: (i) country food consumption and blood biomarkers data from the 2004 Nunavik Health Survey (387 women, 315 men), and (ii) data on MeHg, Se, long-chain n−3 PUFA concentrations found in Nunavik wildlife species.

In the region where most traditional beluga hunting takes place in Nunavik, the prevalence of at-risk blood Hg (≥8 µg/L) in childbearing-age women was 78.4%. While most country foods presently consumed contain low MeHg, beluga meat, not a staple of the Inuit diet, is the most important contributor to MeHg: up to two-thirds of MeHg intake in the beluga-hunting region (0.66 of MeHg intake) and to about one-third in other regions. In contrast, seal liver and beluga mattaaq – beluga skin and blubber – only mildly contributed to MeHg (between 0.06 and 0.15 of MeHg intake), depending on the region. Beluga mattaaq also highly contributed to Se intake (0.30 of Se intake). Arctic char, beluga blubber and mattaaq, and seal blubber contributed to most long-chain n−3 PUFA intake.

This study highlights the importance of considering interconnections between local ecosystems and dietary habits to develop recommendations and interventions promoting country foods’ benefits, while minimizing the risk of MeHg from beluga meat, especially for childbearing-age women.

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1. Introduction

Traditional Inuit knowledge passed down through generations emphasizes the importance of wild animals and plants to Inuit diet, medicine, and culture (Cuerrier and the Elders of Kangiqsualujuaq, 2012a,b). These country foods, including fish, caribou, birds, whales, seal, seafood and berries, represent Inuit values and identity, as well as their intricate relationship with surrounding ecosystems. For Inuit communities, the hunting, harvesting and sharing of country foods is fundamental to social cohesion and cultural continuity. From an Inuit perspective, despite the increasing presence of market-imported foods in the modern Inuit diet, country foods remain essential for a healthy spirit, mind, intellect and body (ITK and ICC, 2012).

Scientific research endorses traditional Inuit knowledge regarding the health benefits of country foods. They are replete in proteins, vitamins, minerals and phytochemicals, and those of marine origin, are exceptionally rich in selenium (Se) and long-chain omega-3 polyunsaturated fatty acids (n-3 PUFA) (Blanchet et al., 2000; Kellogg et al., 2010). However, Arctic ecosystems have changed markedly over recent decades, and the safety of some country foods may be compromised since they bioaccumulate high concentrations of environmental contaminants, in particular methylmercury (MeHg) (AANDC, 2012).

Arctic populations are especially vulnerable to MeHg exposure (AMAP, 2011; Donaldson et al., 2010). While no known anthropogenic sources of mercury (Hg) are found in the North, inorganic Hg emissions, primarily now released from coal burning and artisanal and small-scale gold mining in East and Southeast Asia, accumulate at the poles following long-range atmospheric, oceanic, and river transport (UNEP, 2013). In northern aquatic ecosystems, inorganic Hg is transformed into MeHg by microbial action, and further bioaccumulated and biomagnified at the top of the aquatic food chain, primarily in predatory fish and marine mammals, which are often consumed by local populations (AMAP, 2011). In contrast to inorganic Hg, this highly toxic form of Hg is almost completely absorbed by the human intestine and can readily cross the placenta and blood–brain barriers, causing major health effects, particularly on child development (Clarkson, 2002).

Reducing MeHg exposure is a health priority in the Arctic, particularly during pregnancy (AMAP, 2011; NRBHSS, 2011). The Nunavik Child Development Study (NCDS), a mother-and-child cohort study in Nunavik, Northern Quebec, recently showed that prenatal exposure to MeHg is associated with shorter duration of pregnancy and consequently reduced fetal growth (Dallaire et al., 2013). In addition, prenatal exposure to MeHg is associated with several neurological outcomes in school-age children: poorer early process of visual information; altered attentional mechanisms modulating the early processing of sensory information; poorer memory performance; and increased risk of attention problems and Attention Deficit Hyperactivity Disorder of inattentive type (Boucher et al., 2010, 2012a,b; Ethier et al., 2012). While none of these visual, cognitive and behavioral outcomes were associated with MeHg exposure during childhood, current exposure to MeHg was associated with lower heart rate variability in school-age Inuit children (Valera et al., 2012), and to lower heart rate variability and hypertension in Nunavik adults (Valera et al., 2008; Valera et al., 2009).

Some nutrients found in country foods may counterbalance MeHg toxicity (Chapman and Chan, 2000). In Nunavik, high long-chain n-3 PUFA intake during pregnancy is associated with several benefits including increased birth weight, and child’s visual and memory functions (Boucher et al., 2011; Jacques et al., 2011; Lucas et al., 2004) and a lower prevalence of cardiovascular risk factors among adults (Ayotte et al., 2011; Dewailly et al., 2001b; Valera et al., 2009, 2011). High dietary Se intake may also contribute to offset some Hg-toxic effects among adults in Nunavik and other fish-eating populations (Ayotte et al., 2011; Fillion et al., 2013; Lemire et al., 2010b, 2011; Valera et al., 2009).

Since the 1980’s, several authors have pointed to marine mammal consumption or different parts of marine mammals as the predominant source of MeHg exposure in different regions of the Arctic (Dewailly et al., 2001a; Fontaine et al., 2008; Laird et al., 2013; Milman et al., 1994; Tian et al., 2011). Nonetheless, considering the evolution of Inuit dietary habits in Nunavik (Blanchet and Rochette, 2008), current sources of exposure need to be assessed. Furthermore, because Inuit populations are culturally diverse and live in varying socio-economic contexts and ecosystems, regional differences within the Nunavik territory need to be taken into account in assessing dietary sources of MeHg and nutrients.

The objectives of the present study are to identify contemporary country food sources of MeHg, Se and long-chain n-3 PUFA in Nunavik, particularly among childbearing-age women, which take into account regional differences in country food consumption profiles and contaminant exposure and nutrient intake patterns. Strategies to balance country food risks and benefits, and develop public health recommendations and community interventions are also discussed.

2. Methods

2.1. Study population

The “Qanuippitaa? How are we?” Nunavik Inuit Health Survey is a cross-sectional study conducted in the fall of 2004 (August 27 to October 1st) among the 14 communities of Nunavik set along the eastern coast of the Hudson Bay (hereafter Eastern Hudson Bay), and the coasts of the Hudson Straight and the Ungava Bay (Fig. 1). Nunavik lies north of the 55th parallel in Quebec (Canada) and covers a third of the total surface area of the province. This study is based on a large and representative sample of the Inuit population (889 adults aged between 18 and 74 years old), which represented 10% of the Inuit population of Nunavik at that time (Statistics Canada, 2001). The study design and 2-stage stratified random sampling strategy are detailed elsewhere (Rochette and Blanchet, 2007), and the participation rate was estimated at 52% for the complete clinical examination (Valera et al., 2009). Participants were invited on-board the Amundsen research vessel for biological sample collection, and various anthropometric and physiological measurements. Questionnaires were administered to document socio-demographic data, lifestyle habits, and food consumption frequencies. Among the 889 participants, a total of 702 Inuit adults (387 women and 315 men) provided blood samples and completed the anthropometric examination and questionnaires (including general background information, lifestyles and diet), and were included in the present study. Pregnant women were excluded from this analysis (n = 26). Based on our maternal biomonitoring studies in Nunavik, childbearing-age was defined between 18 and 39 years old. Each individual who agreed to participate signed a consent form, which was read to them in their preferred language (Inuktitut, French or English). The study protocol was approved by the Ethics Committee of the Centre Hospitalier Universitaire de Québec (CHU de Québec) and Université Laval.

2.2. Blood Hg, Se and long-chain n−3 PUFA analyses

Total Hg and Se concentrations were determined in whole blood. Total Hg in whole blood is a validated biomarker of recent intake and acceptable surrogate of MeHg exposure given the elevated consumption of fish and marine mammals in this study population (Kershaw et al., 1980). Whole blood samples were collected from an antecubital vein in a 6 mL plastic vacutainer containing potassium EDTA as the anticoagulant (BD Medical), stored frozen at −80 °C and sent to the Centre de Toxicologie du Québec of the Institut National de Santé Publique du Québec for total Hg and Se determination by inductively coupled plasma mass spectrometry. The detection limit for blood Hg and Se was respectively 0.1 and 7.9 μg/L. The complete analytical method is detailed elsewhere (Valera et al., 2009). The long-chain n-3 PUFA composition of the erythrocyte phospholipids (eicosapentaenoic acid (20:5; EPA), docosapentaenoic acid (22:5; DPA) and docosahexaenoic acid...
(22:6; DHA)) was measured after membrane purification, chloroform/methanol lipid extraction, and methylation of fatty acids, followed by capillary gas–liquid chromatography using a DB-23 column (39.0 × 0.25 mm ID × 0.25 μm thickness) in a Hewlett-Packard gas chromatograph. The results are expressed as percentage of total fatty acids and the complete analytical method, described in Lucas et al. (2010c).

2.3. Data collection

Body weight (in kg) was measured without shoes. Socio-demographic characteristics and food frequencies were documented using interviewer administered questionnaires, which were available in Inuktitut, English and French. The food frequency questionnaire (FFQ) documented the frequency of consumption during the year prior to the survey for 69 food items and beverages. Foods were divided into 2 major groups. The “country foods” group refers to 25 items derived from fishing, hunting and harvesting, and were recorded for each of the 4 seasons of the year prior to the interview. This group included country fish, marine mammal, seafood, land mammal and wildfowl species found and most consumed in Nunavik, the different parts and organs of animal possibly eaten by Inuit, and the most common traditional food preparations (listed in Supplemental Table S4). Country meats are usually eaten raw, but also frozen or cooked. Inuit also traditionally prepare and eat air-dried meat made of beluga and caribou (known as nikku), dried fish (also known as pitsik), and aged meat (fermented) made of flesh, liver, intestines and blubber of walrus and/or seal (also known as igunak). Pitsik is commonly made from Arctic char, brook trout or lake whitefish. Beluga mattaaq, an Inuit delicacy, is composed of beluga skin and raw blubber. The proportion skin-to-blubber of mattaaq is quite variable depending on individual taste, but usually beluga blubber represents at least one-fifth of one mattaaq piece. Beluga mattaaq is often consumed raw, slightly aged or boiled. The blubber (fat) of beluga and seal is commonly consumed aged (i.e. fermented to liquify) as a dip and then known as misiruk, but also consumed raw or included in other traditional recipes. The “market foods” group refers to most store-bought foods imported from southern regions and consumed during the month prior to the survey (44 items, listed in Rochette and Blanchet, 2007). The FFQ specified the

Fig. 1. Map of Nunavik.
usual serving size. To do so, pre-defined serving sizes were weighted (in grams) and a corresponding food model was shown to the respondents. Analysis of the FFQ data provided estimates of consumption frequency of the different food items per day on an annual basis (average of the four seasons for the country foods) and the usual intake in grams. The daily food intake on an annual basis were calculated by multiplying the food frequency by the usual intake for each food (in g) and expressed as g/day on an annual basis. Energy intake (in kilocalories) from country and market foods was estimated using the method described in Rochette and Blanchet (2007).

The FFQ developed for the present study took into account the methodological problems identified in the 1992 Santé-Québec survey in Nunavik (Santé Québec, 1995). Consultations and a field test with a consortium of Inuit representatives, experts in nutrition, translators, and researchers were also conducted in 2004 prior to the study to verify the accuracy of food items included in the FFQ and the absence of inconsistency in results, to apply corrections when needed, and to verify and validate the language translations and consistency (Rochette and Blanchet, 2007).

2.4. Concentrations of mercury, selenium and long-chain n-3 PUFA in Nunavik country foods

Data reports for Hg and Se concentrations in several country foods analyzed at the Nunavik Research Center can be found at the Arctic Science and Technology Information System website (www.aina.ucalgary.ca/astis) and compiled in Supplemental Tables S1 and S2. These samples were collected in different regions of Nunavik between 1996 and 2013. Hg and Se concentrations are reported with their geometric means (GM) or age-adjusted GM (when available) in raw samples in μg/g (wet weight), with the exception of beluga niku samples, which were prepared from archived beluga meat samples that were air-dried using the traditional Inuit method. When no data was available from the Nunavik Research Center for a specific country food, we used mean Hg or Se concentrations (in wet weight) from other studies that included samples collected in 1990 from the Great Whales and Nottaway–Broadback–Rupert areas in Québec’s province (Blanchet et al., 2000; Dewailly et al., 1996) or in 1998–2000 and 2005 in Greenland (Johansen et al., 2004, 2009). For EPA, DPA and DHA concentrations (in mg/g), data were compiled from two studies in Nunavik (Dewailly et al., 1996; Lucas et al., 2010a) and elsewhere in the Arctic and Canada (CINE, 2013; Hassan et al., 2012; Health Canada, 2012) (Supplemental Table S3).

As mentioned in the Introduction section, the chemical forms of Hg found in marine foods will influence intestinal Hg absorption and toxic effects (Clarkson, 2002). Contrary to fish in which most of Hg is MeHg (Bloom, 1992), a very large proportion of Hg found in the organs of marine mammals and seabirds has been shown to be inorganic Hg or inorganic Hg–Se complexes, which are much less absorbed than MeHg by the human intestine (Clarkson, 2002; Ikemoto et al., 2004; Lemes et al., 2011; Wagemann et al., 1998). Unpublished data from Nunavik Research Center show that the percentage of MeHg varies between 97.6 and 100% in anadromous Arctic char (sea-run subspecies) flesh samples. The % of MeHg (age-adjusted GM) measured in beluga samples is 80% in skin, 65% in meat, 12% in liver and 8% in kidney for a beluga of 16.5 years of age. In ringed seal, the % of MeHg (age-adjusted GM) is 80% in skin, 65% in meat, 12% in liver and 8% in kidney (Table S2, Nunavik Research Centre, unpublished data). Total Hg concentrations reported in marine mammal blubber are consistently low, and no data was found on Hg speciation in blubber.

Since the objective of the present study is to estimate MeHg intake, total Hg concentrations were used as a surrogate for most country foods (MeHg concentrations not available for many species), with the exception of marine mammal offals. Indeed, although marine mammal offals can present very high total Hg concentrations, their effective concentrations in MeHg are below 26%. In addition, recent data show that the gastro-intestinal bioavailability of total Hg in seal liver, measured using in vitro model simulating the physiological conditions of the human gastro-intestinal tract, is on average less than 25%, supporting the hypothesis that overall absorption of Hg in marine mammal offals is low (Laird, personal communication). Therefore, the estimation of MeHg intake from different marine mammal offals was based on average MeHg instead of total Hg concentrations in those organs (Nunavik Research Centre, see above). For seal liver, the most frequently consumed offal, a conservative proportion of 25% of total Hg as MeHg instead of 11% was used (based on B. Laird findings) to avoid underestimation of MeHg intake from this particular food. Additional estimations using total Hg in marine mammal offals were also conducted.

Reference categories for MeHg concentrations in country foods were based on Canadian reference values in fish (Health Canada, 2007): low (<0.20 μg/g), low-medium (0.20–0.50 μg/g), high (0.51–1.00 μg/g) and very high in MeHg (>1.00 μg/g). For Se, reference categories were established according to the literature (Lemire et al., 2010a; Rayman, 2008): low source (<0.20 μg/g), good source (0.20–0.50 μg/g), very good source (0.51–1.00 μg/g) and exceptional source (>1.00 μg/g). Reference categories for long-chain n-3 PUFA in food were estimated based on health recommendations for EPA and DHA intake and cardiovascular risk factors (Gebauer et al., 2006; Kris-Etherton et al., 2002; Lichtenstein et al., 2006; Mozaffarian and Rimm, 2006): low source (<2.5 mg/g), good source (2.5–5 mg/g), very good source (5–10 mg/g) and exceptional source (>10 mg/g).

2.5. Country food methylmercury, selenium and long-chain n-3 PUFA intakes

The contribution of the different country foods to the daily MeHg, Se and long-chain n-3 PUFA intakes (in μg/kg bw/day or mg/kg bw/day) was calculated by multiplying average Hg, Se and long-chain n-3 PUFA (sum of EPA, DPA and DHA) concentrations of each country food by their respective daily intake adjusted for participant’s body weight (in kg) to minimize inter-individual variability (expressed as g/kg body weight/day on an annual basis; hereafter μg/kg bw/day). This weight-adjusted approach was favored to compare estimated MeHg intakes to the most recent Canadian guidance values for MeHg exposure. As detailed in Legrand et al. (2010), Health Canada has set the provisional Tolerable Daily Intake (pTDI) at 0.20 μg/kg bw/day for childbearing-age women and children, corresponding to a concentration of 8 μg/L of total Hg in whole blood, the provisional blood guidance value for MeHg exposure. For men above 18 years old and women of 40 years of age and older, the provisional blood total Hg guidance value in Canada is 20 μg/L, and the corresponding pTDI is 0.47 μg/kg bw/day (Legrand et al., 2010). In the United States, no distinction is made for at-risk sub-populations; the U.S. Environmental Protection Agency (U.S. EPA) established the Reference Dose at 0.1 μg/kg bw/day for all peoples (U.S. EPA, 2001) while the Centers for Disease Control and Prevention (CDC) uses a Minimal Risk Level of 0.3 μg/kg bw/day (ATSDR, 2011).

2.6. Statistical analysis

The normality of the distribution of each variable was verified and appropriate transformations applied to normalize distributions if needed (Log10(x) or −1/(x) for blood biomarkers and Log10(x + 1) for food intakes). Descriptive analyses were used to illustrate the distribution of blood Hg, Se and long-chain n-3 PUFA biomarkers, anthropometric measures, food intakes, and Hg, Se and long-chain n-3 PUFA food intakes, for the entire study population and stratified by region and gender groups when appropriate. Considering the right-skewed distribution of most of blood biomarkers and food intake variables, geometric mean (GM) values were provided. In this case, because
of the logarithm properties, the contribution of each food item to the total intakes of country foods, MeHg, Se and long-chain n−3 PUFA were calculated as described in Supplemental material section S1. Multiple regression models were used to examine the associations between blood biomarkers or food intakes (as continuous variables) and region, age and gender (continuous or categorical variables). Pearson correlations (r) were used to evaluate the correlations between blood biomarkers, estimated total intakes and blood predicted values (continuous variables with normalized distribution).

Population weights were incorporated into all of the statistical analyses to take into account the complex sampling design used in this study. Bootstrap procedure, a re-sampling technique for variance estimation for complex survey designs that provides gains in the precision of estimates, was also used when required as described in Valera et al. (2009). Results were defined as statistically significant at p < 0.05. Analyses were performed using JMP 9.0.0 (SAS Institute Inc., USA) and SUDANN (10.0.1).

3. Results

The study participant characteristics are shown in Table 1. High blood Hg concentrations are prevalent in this population (GM = 10.7 μg/L), and more than half of child-bearing-age women and adults of 40 years old and above had blood Hg levels exceeding Canadian blood Hg guidance values (≥8 μg/L for childbearing-age women and ≥20 μg/L for other adults). A large regional difference in MeHg exposure was also observed: residents of Hudson Strait villages presented significantly higher blood Hg than those of Eastern Hudson Bay villages, and those from Eastern Hudson Bay, had higher blood Hg than residents from the Ungava Bay villages. In the Hudson Strait region, up to 78% of childbearing-age women and 73% of adults of 40 years old and above had blood Hg levels that exceeded the Canadian guidance values. In a multiple regression model adjusted for the region of residence, blood Hg levels were significantly associated with age (standardized β = 1.25, p < 0.0001), and higher among women (adjusted GM for women = 12.2 μg/L, for men = 10.4 μg/L, p = 0.02).

Exceptionally high blood Se status (GM = 271 μg/L) and long-chain n−3 PUFA profile (sum of EPA, DPA and DHA: GM = 8.5%) of total red blood cells fatty acids, sum of EPA and DHA: GM = 6.3%) were also observed in this population (Table 1). As was observed for blood Hg, regional differences are substantial. Blood Se concentrations were significantly higher in the Hudson Strait region, followed by the Ungava Bay and the Eastern Hudson Bay regions. For long-chain n−3 PUFA, Hudson Strait and Ungava Bay presented significantly higher EPA and DPA (GM = 1.5% and GM = 2.3%, respectively) than the Eastern Hudson Bay region (GM = 1.2% and GM = 1.9%, respectively), while DHA levels were similar among the three different regions (GM = 5.0%). In a multiple regression model adjusted for the region of residence, blood Se levels were positively associated with age (β = 0.87, p < 0.0001), and higher in women, although the difference was marginally significant (adjusted GM for women = 288 μg/L, for men = 276 μg/L, p = 0.06). In a similar model adjusted for region of residence, the percentage of long-chain n−3 PUFA was also positively associated with age (β = 1.52, p < 0.0001) and significantly higher among women (adjusted GM for women = 9.1%, for men = 8.1%, p < 0.0001).

Similarly to blood biomarkers, annual country food consumption profiles display important regional and gender differences (Table 2). Compared to participants living in the Hudson Strait and the Ungava Bay regions, those from the Eastern Hudson Bay reported consuming significantly more country foods of all types except marine mammals, which were significantly more consumed in the Hudson Strait region. Wild berries and seaweeds were the only country foods equally consumed by men and women. In separate multiple regression models taking gender and the region of residence into account, the consumption of marine mammal was positively associated with age (β = 0.40, p < 0.0001), as was the consumption of fish and seafood (β = 0.37, p = 0.0001). However, this was not the case for land animals and game birds, and for wild berries and seaweed consumption (β < 0.07, p > 0.43). Market food consumption was significantly higher among men (p = 0.01), inversely associated with age (β = −0.40, p < 0.0001) but not different between regions (p = 0.38). Overall, the contribution of country foods to total food energy intake is low (14.1%), particularly in the Ungava Bay region.

Table 1

<table>
<thead>
<tr>
<th>Study population characteristics and blood biomarkers concentrations, by region of residence.</th>
</tr>
</thead>
<tbody>
<tr>
<td>All regions</td>
</tr>
<tr>
<td>Women (n, weighted %)</td>
</tr>
<tr>
<td>Age (years)</td>
</tr>
<tr>
<td>GM (range)</td>
</tr>
<tr>
<td>Weight (kg)</td>
</tr>
<tr>
<td>Whole blood Hg (μg/L)</td>
</tr>
<tr>
<td>Whole blood Hg concentrations above guidance values (weighted %)</td>
</tr>
<tr>
<td>Women 18–39 years old</td>
</tr>
<tr>
<td>Men 18–39 years old</td>
</tr>
<tr>
<td>Women ≥ 40 years old</td>
</tr>
<tr>
<td>Men ≥ 40 years old</td>
</tr>
<tr>
<td>Whole blood Se (μg/L)</td>
</tr>
<tr>
<td>Red blood cells long-chain n−3 PUFA (as % of total fatty acids)</td>
</tr>
</tbody>
</table>

Different letters represent statistically significant differences between regions.
1 GM by region are adjusted for age and gender.
2 Canadian blood guidance values: ≥8 μg/L for childbearing-age women (<40 years old) and ≥20 μg/L for other adults.
3 Sum of eicosapentaenoic acid (EPA), docosapentaenoic acid (DPA) and docosahexaenoic acid (DHA) fatty acids.
Bay villages (11.3%), not different by gender (p = 0.78), and strongly associated with age (std β = 0.76, p < 0.0001; adjusted GM for women and men < 40 y = 11.2% and GM for women and men ≥ 40 y = 19.3%).

Almost all country foods found in Nunavik are reported to contain low MeHg concentrations (<0.2 μg/g), with the exception of beluga meat, beluga niku (traditionally air-dried meat), beluga liver, ringed seal liver and lake trout, which present in average very high MeHg concentrations (> 1.0 μg/g). Beluga and ringed seal kidneys, northern pike, walleye and marine birds present, on average, high MeHg concentrations (0.51–1.00 μg/g), while beluga skin, beluga mattaqq and ringed seal meat present MeHg concentrations in the low-medium range (0.20–0.50 μg/g) (Fig. 2a, and Supplemental Tables S1 and S2). Exceptionally high Se concentrations (> 1.0 μg/g) are found in beluga mattaqq, marine mammal offals, walrus meat, beluga niku and fish eggs (Fig. 2a, Supplemental Table S2). Nearly all other country foods present Se concentrations above 0.2 μg/g, the usual guideline for a good source of Se in other populations. Ringed seal blubber presents amazingly high EPA, DPA and DHA concentrations, higher than all other country foods consumed in Nunavik and any other marine food found on the market (Fig. 2b, Supplemental Table S3). Beluga and walrus blubbers, which contain less than half of seal blubber long-chain n−3 PUFA concentrations, are still considered exceptionally high in long-chain n−3 PUFA (>10 mg/g), together with Arctic Char (searun subspecies), Atlantic salmon and fish eggs (data only for Atlantic salmon and lake whitefish).

DPA concentration data were only found for seal, beluga and walrus blubbers, beluga niku, walrus meat and Arctic char. However, low EPA and DHA concentrations suggest very low DPA concentrations in other country foods.

Despite significant regional differences in country food consumption profiles (Table 2), the country foods most highly consumed are very similar between Nunavik regions. Indeed, as shown in Fig. 3, caribou meat, Arctic char (searun ssp.), Canada and snow geese, willow and rock ptarmigans, as well as wild berries, are important staple country foods in this population. Altogether, they contributed to up to two-thirds of the daily country food intake (proportion of the total = 0.65) on an annual basis, and this, similarly across age groups and between men and women (data not shown). A variety of other fish and bird species, different parts of marine mammals and blue mussels contributed to the remaining third, varying greatly from one region to another and reflecting regional differences presented in Table 2 (Supplemental Table S4). In the category “other fish”, fourhorn sculpin – also known as ugly fish – was the fish species most frequently reported (30%). Arctic char (landlocked subspecies), lake trout, pike and walleye fish species were also mentioned, but at a much lower frequency (between 1 and 2%). Eiders, scoters, mergansers, murre (guillemot), pintails and ducks were the species reported in the “other birds” category. Very few consumed common loon (<1%) and none reported consuming sea or herring gulls, two other marine bird species. Very few reported consuming igunak or offals, with the exception of ringed seal liver. Oysters,

Table 2

<table>
<thead>
<tr>
<th>Marine mammals (g/day)</th>
<th>Women (n = 387)</th>
<th>Men (n = 315)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fish and seafood (g/day)</td>
<td>21.5b</td>
<td>27.4a</td>
</tr>
<tr>
<td>Land animals and game birds (g/day)</td>
<td>34.4b</td>
<td>43.3a</td>
</tr>
<tr>
<td>Wild berries and seaweed (g/day)</td>
<td>4.1</td>
<td>3.5</td>
</tr>
<tr>
<td>Total country foods (g/day)</td>
<td>91.2b</td>
<td>107.7a</td>
</tr>
<tr>
<td>Total market foods (g/day)</td>
<td>629.9 b</td>
<td>727.2a</td>
</tr>
</tbody>
</table>

% of country food intake

4 Express as the geometric mean (range) of the weighted percentage of the energy (kilocalories) provided by country foods over the total of energy provided by market and country foods.

Different letters represent statistically significant differences between gender or regions.

1 Geometric mean, adjusted for age and region.
2 Geometric mean, adjusted for age and gender.
3 Combined marine mammals, fish and seafood, land animals and game birds, and wild berries and seaweed consumption.
4 Express as the geometric mean (range) of the weighted percentage of the energy (kilocalories) provided by country foods over the total of energy provided by market and country foods.
urchins, clams, scallops, bear, fox, snowshoe hare and seaweed were also not often consumed.

Beluga meat was the most important contributor of MeHg intake in this population. While not a staple of the diet, this country food alone contributed up to two-thirds of MeHg intake in villages along the Hudson Strait, and about one-third in other regions (Fig. 3, Supplemental Table S5). In Nunavik, beluga meat is often consumed as such, raw or frozen, as nikku and occasionally boiled (raw, frozen and boiled meat are labeled together as beluga meat in Fig. 3). The consumption of beluga meat and beluga nikku reported by childbearing-age women in the Hudson Strait villages provided an intake of 0.4 μg MeHg/kg bw/day, exceeding the pTDI by a factor of two (beluga meat GM = 0.097 μg MeHg/kg bw/day [proportion of the total MeHg intake = 0.17]; beluga nikku GM = 0.323 μg MeHg/kg bw/day [proportion = 0.55]). For childbearing-age women from the Eastern Hudson Bay and Ungava Bay villages, MeHg intake from beluga

Fig. 3. Average country food, MeHg, Se and long-chain n−3 PUFA daily intakes by region of residence on an annual basis*. A. Eastern Hudson Bay, B. Hudson Strait, C. Ungava Bay. *Country food items are listed in decreasing order according to their consumption within different regions.
meat and beluga *nikku* was significantly lower, but still contributed to most of MeHg intake compared to other country foods (beluga meat GM = 0.090 µg MeHg/kg bw/day [proportion = 0.36]; beluga *nikku* GM = 0.084 µg MeHg/kg bw/day [proportion = 0.48]). This was likewise the case for other age and gender groups, except for men 40 years old and above from the Eastern Hudson and Ungava Bays, for whom beluga meat, beluga *nikku* and seal liver contributed almost equally to MeHg intake (Eastern Hudson Bay: beluga meat and beluga *nikku* GM = 0.090 µg MeHg/kg bw/day [proportion = 0.21]; seal liver GM = 0.109 µg MeHg/kg bw/day [proportion = 0.25]; and Ungava Bay: beluga meat and beluga *nikku* GM = 0.036 µg MeHg/kg bw/day [proportion = 0.19]; seal liver GM = 0.043 µg MeHg/kg bw/day [proportion = 0.22], respectively).

Fig. 3 also shows the importance of other country foods, and particularly those from beluga whale, for Se and long-chain n–3 PUFA intakes. Indeed, while beluga *mattaaq* mildly contributed to MeHg intake (proportion between 0.09 and 0.13), it greatly contributed to Se intake in all regions (proportion from 0.23 to 0.45). Several other country foods moderately contributed to Se intake depending on the region. In the Hudson Strait, beluga blubber and Arctic char were equally important for long-chain n–3 PUFA intakes, while in the other regions, Arctic char was by far the most important contributor to long-chain n–3 PUFA intakes. Despite very high concentrations found in seal blubber, it only moderately contributed to long-chain n–3 PUFA intakes.

Hg, Se and long-chain n–3 PUFA blood biomarkers were highly intercorrelated (Table 3, Pearson r > 0.42, p < 0.0001), and this, particularly in the Hudson Strait region (Supplemental Table S7, blood Hg versus Se, r = 0.80, p < 0.0001; blood Hg versus long-chain n–3 PUFA, r = 0.71, p < 0.0001; blood Se versus long-chain n–3 PUFA, r = 0.59, p < 0.0001). Correlations between Hg and long-chain n–3 PUFA blood biomarkers with their respective calculated intakes were moderately high (r = 0.27, p < 0.0001, and r = 0.30, p < 0.0001), but lower for Se (r = 0.20, p < 0.0001), and this particularly in the Eastern Hudson Bay region (Supplemental Table S7, r = 0.11, p = 0.07).

MeHg intakes were recalculated using total Hg concentrations in marine mammal organs, in order to investigate the possible underestimation of Hg intake from these particular country foods, especially in comparison to beluga meat. These “worst-case scenario” analyses showed that for childbearing-age women, beluga meat still remained the most important contributor to Hg intake (beluga meat and beluga *nikku* GM = 0.147 [proportion of the total Hg intake = 0.36]; seal liver GM = 0.108 µg MeHg/kg bw/day [proportion = 0.26]), and this similarly among the three regions (data not shown). However, for men of 40 years old and above from Eastern Hudson and Ungava Bays, seal liver was now the predominant source of Hg (Eastern Hudson Bay: beluga meat and beluga *nikku* GM = 0.090 µg Hg/kg bw/day [proportion = 0.13]; seal liver GM = 0.356 µg Hg/kg bw/day [proportion = 0.51]; and Ungava Bay: Beluga meat and beluga *nikku* GM = 0.036 µg MeHg/kg bw/day [proportion = 0.12]; Seal liver GM = 0.137 µg MeHg/kg bw/day [proportion = 0.46]).

### 4. Discussion

#### 4.1. Contemporary country food sources of MeHg, Se and long-chain n–3 PUFA in Nunavik

In this study, we identified important country food sources of MeHg, Se and EPA, DPA and DHA n–3 PUFAs. Our results show that most of commonly-consumed country foods in Nunavik are quite low in MeHg, except for beluga meat, seal liver and lake trout. Beluga meat accounts for most MeHg exposure in this population. We also show that nearly all country foods are good sources of Se, and confirm that marine mammal blubber and Arctic char are exceptional sources of long-chain n–3 PUFA in Nunavik. This study highlights significant regional variations in country food consumption, MeHg exposure, and blood nutrient levels. In particular, our results indicate that residents of villages along the Hudson Strait are at particular risk of surpassing Canadian guidelines for blood Hg levels of concern.

Despite a decreasing temporal trend of 30% in MeHg exposure since 1992 (GM = 16.0 µg/L) [Dewailly et al., 2001a], MeHg exposure in Nunavik remains among the highest in the world (blood Hg GM = 10.7 µg/L), and much higher than elsewhere in Southern Canada (GM = 0.69 µg/L) [Lye et al., 2013]. Based on the MeHg blood guidance value in Canada [Legrand et al., 2010], more than half of childbearing-age women in this study had at-risk blood Hg levels (≥8 µg/L) and in villages along the Hudson Strait, over three-quarters of women had levels exceeding this threshold.

Even though it is not a staple of the Nunavik diet, beluga meat is the most important contributor of MeHg intake. Beluga meat alone contributes up to two-thirds of MeHg exposure in the Hudson Strait region, and to about one-third in other regions of Nunavik. Twice a year, beluga whales transit the Hudson Strait area to migrate from the Ungava Bay and Labrador Sea to the Hudson Bay to give birth and raise their calves [FOC, 2012]. The villages settled along the coast of the Hudson Strait are where most of the traditional beluga hunting currently takes place in Nunavik [FOC, 2012]. In contrast to beluga *mattaaq*, an Inuit delicacy readily shared by plane to other villages in Nunavik during hunting seasons, beluga meat usually remains in the villages where the whale is hunted and air-dried as *nikku* for preservation. Since MeHg displays a very high affinity for proteins, it tends to accumulate in the flesh/meat and organs of animals [Clarkson, 2002]. In the flesh/meat, MeHg represents a very high proportion of total Hg, and most of cooking processes only generally mildly increase total Hg concentrations due to moisture loss [Bloom, 1992; Morgan et al., 1997]. Unfortunately, recent data from the Nunavik Research Centre show that the traditional *nikku* process, which removes almost all water, drastically increases total Hg concentrations by 2- to 3-fold total. Consequently, since beluga is a

### Table 3

Correlation matrix for blood biomarkers, MeHg, Se and long-chain n–3 PUFA estimated intakes and predicted blood Hg concentrations.

<table>
<thead>
<tr>
<th></th>
<th>Whole blood Hg (µg/L)</th>
<th>Whole blood Se (µg/L)</th>
<th>Long-chain n–3 PUFA (% of total RBC fatty acids)</th>
<th>Total MeHg intake (µg/kg bw/day)</th>
<th>Total Se intake (µg/kg bw/day)</th>
<th>Total long-chain n–3 PUFA intake (mg/kg bw/day)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Whole blood Hg (µg/L)</td>
<td>0.70 ***</td>
<td>0.62 ***</td>
<td>0.27 ***</td>
<td>0.26 ***</td>
<td>0.26 ***</td>
<td>0.35 ***</td>
</tr>
<tr>
<td>Whole blood Se (µg/L)</td>
<td></td>
<td>0.46 ***</td>
<td>0.26</td>
<td>0.20 ***</td>
<td>0.22 ***</td>
<td>0.30 ***</td>
</tr>
<tr>
<td>Long-chain n–3 PUFA (%)</td>
<td></td>
<td></td>
<td>0.12 **</td>
<td>0.11 **</td>
<td>0.30 **</td>
<td></td>
</tr>
<tr>
<td>Total MeHg intake (µg/kg</td>
<td>0.84 **</td>
<td>0.52 **</td>
<td></td>
<td>0.72 **</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*** p < 0.0001,  ** p < 0.01
toothed whale at the top of the marine food chain, and very high concentrations of total Hg (≥ 1 μg/g) are often found in beluga meat and consistently found in beluga niku, villagers who frequently consume this country food display high blood Hg levels compared to others.

Seal liver is another Inuit delicacy, at times eaten raw right after the animal was hunted. Very high concentrations of total Hg are found in seal liver, often much higher than in beluga meat. Total Hg concentrations in seal liver also greatly increase with the age of the seal, and consequently a very large variation in total Hg is found between animals of different ages (2–90 μg/g). Conversely, the proportion of MeHg concentration over that of total Hg significantly decreases with the age of the seal (Wagemann et al., 1998; Kwan, personal communication). The large inorganic Hg fraction found in the liver of older seals is poorly bioavailable and/or absorbed by the human intestine (Clarkson, 2002; Laird, personal communication). In the present study, average bioavailable MeHg concentrations in seal liver (25% of total Hg; Laird, personal communication) were used to estimate MeHg intake from this particular food (instead of using total Hg as a surrogate of MeHg, as was done for other country foods). Our results showed that seal liver is not the predominant contributor to MeHg intake, and this for all age groups. Even when using total Hg instead of bioavailable MeHg concentrations, beluga meat remains the predominant source of Hg in most cases. Seal liver is not the major source of total Hg intake for childbearing-age women and other study participants, except for older men, perhaps seal hunters, from the Eastern Hudson Bay and Ungava Bay, where seal hunting is more common than beluga hunting. Other country foods most frequently consumed in Nunavik, including caribou meat, Arctic char (searun spp.), geese, ptarmigan and wild berries, are fairly low in MeHg and do not significantly contribute to MeHg intake.

Although country food consumption has declined in the last decades (Blanchet and Rochette, 2008), Se status is still exceptionally high in Nunavik (GM = 271 μg/L, range 126–3571 μg/L) in comparison to the general Canadian population (GM = 204 μg/L, 95% CI 200–208 μg/L) (Health Canada, 2010b). Almost all country foods in Nunavik are rich in Se (≥ 0.2 μg/g), particularly those of marine origin such as beluga mattaaq, which is an exceptional source of Se containing on average between 2.0 and 8.0 μg/g.

Se status in Nunavik is well above the blood Se levels of 90 μg/L required for optimal Se-enzymes activity (≥ 70 μg/L of plasma Se) (Health Canada, 2010a; Yang and Xia, 1995). A few participants (3.1%) had blood Se above the No Observable Adverse Effect Level (NOAEL) of 1000 μg/L. This guideline was set by the U.S. Environmental Protection Agency more than 10 years ago to prevent human Se intoxication (Poirier, 1994). However, more recent studies suggest that the organic forms of Se found in food may be less toxic than those found in occupational settings, nutritional supplements or drinking water (Rayman, 2012). Indeed, no evidence of selenium toxicity was found in the Brazilian Amazon, a fish-eating population with blood Se status at times surpassing 1000 μg/L (Lemire et al., 2006; Lemire et al., 2012).

In Northern and Amazonian populations highly exposed to MeHg, high Se intake may partially offset MeHg-mediated oxidative stress and/or be required to maintain optimal Se-enzyme levels. Consequently, there may be less “excess” Se and little or no Se toxicity (Khan and Wang, 2010; Khan and Wang, 2009), although this remains to be confirmed.

As was the case for Se, Inuit in Nunavik have remarkably high blood long-chain n – 3 PUFA (GM = 8.5%, range 1.7–20.2%). These levels are about four times greater than those observed in Southern Quebec populations (Dewailly et al., 2003). Marine mammal fats, and particularly seal blubber, contain exceptional content in long-chain n – 3 PUFA, especially of DPA (22:5n – 3), a DHA (22:6n – 3) precursor almost only found in seal blubber. Arctic char and beluga mattaaq also greatly contribute to long-chain n – 3 PUFA intake. In Nunavik, such a high n – 3 PUFA status has been associated with several health benefits, from healthier pregnancies and to better neurologic child development, and mental and cardiovascular health among adults (Boucher et al., 2011; Jacques et al., 2011; Lucas et al., 2004, 2010b; Valera et al., 2009). As a whole, this marine country food diet is believed to be one of the main factors that protect Inuit populations from many chronic diseases (Bjerregaard et al., 2004; Dewailly et al., 2001b).

Despite study limitations due to FFQ recall-bias, imprecisions in portion size estimation and in specific country foods MeHg concentrations, good associations were observed between blood Hg and estimated MeHg intake, suggesting that total MeHg intake was adequately estimated. The strength of this study resides in the accuracy of the data on MeHg and Se concentrations in country foods now available for the Nunavik territory. In addition, the GM and regional approach taken for the intake calculations reduced the distortions in right-skewed FFQ data distributions, and allowed a more precise estimation of the regional sources, particularly for MeHg. A good correlation was also found between estimated long-chain n – 3 PUFA intakes and the percentage of long-chain n – 3 PUFA in red blood cells, despite incomplete data for EPA, DPA and DHA concentrations in country foods consumed in Nunavik. For Se, a moderate correlation was found between estimated intake and blood Se, although the correlation in the Eastern Hudson Bay region was weak. As was the case for MeHg, important variations in Se concentrations may be found in country foods. This may also suggest that the consumption of some country foods high in Se and particularly consumed in this region may not have been evaluated in the FFQ. It is to note that other sources of Se may also be found among imported market foods in Nunavik, such as beef organs, fish, and cereals, with varying Se concentrations between 0.2 and 0.5 μg/g (Rayman, 2008), depending on the soil Se levels where the foods were raised or grown.

Lake trout, reported to be frequently consumed in villages with limited access to Arctic char or during the fall when other fish species are less available, and marine bird eggs, sometimes consumed during the nesting period, may periodically contribute to MeHg intake in some specific villages, although this has yet to be confirmed. The Nunavik Research Centre recently showed that fish eggs from various species in Nunavik are high in Se. These are also known to be high in long-chain n – 3 PUFA (CINE, 2013). For future studies in Nunavik, the FFQ will be improved to better capture regional differences in country food consumption and in these particular food items.

The chemical forms of Se and Hg found in Nunavik country foods may greatly influence Se and Hg absorption, and Se availability for Se-enzyme synthesis (Clarkson, 2002; Khan and Wang, 2010; Rayman et al., 2008). Long-chain n – 3 PUFA might also contribute to mitigate intestinal MeHg absorption (Xiao et al., 2013). Analytical and in vitro studies are on-going to elucidate the different chemical forms and bioavailability of Se and Hg in Nunavik country foods, as well as the influence of country food nutrients on Hg intestinal absorption.

### 4.2. Implications of study results for the broader Nunavik Public Health context

In this Inuit region, the Nunavik Nutrition and Health Committee (NNHC) is responsible for overviewing public health research conducted in Nunavik and composed of representatives of the Kativik Regional Government, Nunavik Research Centre of the Makivik Corporation, Nunavik Regional Board of Health and Social Services, Ungava Tulattavik Health Centre, Kativik School Board, Inuit Tapiriit Kanatami, Institut national de santé publique du Québec and Trent University. In 2011, a working group composed of the Director of Public Health in Nunavik, the NNHC and several authors of the present study identified the most significant results of the Nunavik Child Development Study in terms of public health for Nunavik youth. The working group also agreed to work with the Public Health Director in the development of dietary country food guidelines for the Nunavik population, with a specific focus on childbearing-age women.

All members of the NNHC were aware of a loss of confidence in country foods following inadequate or conflicting consumption...
advisories and that these had deleterious health consequences for indigenes people in the past (Donaldson et al., 2010). Several issues with respect to health effects of MeHg in children and dietary sources of exposure were taken into account: (i) Consistent with previous studies in Nunavik and elsewhere, prenatal MeHg exposure affects children’s cognitive and visual development and is associated with behavioral problems in the classroom, threatening future success in school; (ii) few adverse health outcomes have been associated with postnatal MeHg exposure in Nunavik; (iii) MeHg exposure during pregnancy is a modifiable risk factor; (v) beluga meat is the main source of exposure to MeHg and not a staple of Inuit diet; and (vi) there is no evidence for declining MeHg concentrations in beluga meat.

Elevated MeHg in certain country foods creates challenges for nutritional and public health programming. In addition to concerns about balancing MeHg risk and nutrient benefits in country foods, the working group had to consider several other priorities essential to support Inuit diet, healthy pregnancies and cultural continuity in Nunavik. First, beluga hunting and eating tradition are central to Nunavik culture, and beluga mattaqq is one of the favorite country foods across all Inuit generations. Seal liver is also highly appreciated by Inuit and believed to keep the body warm when consumed raw on the ice. Second, country food intake has also declined markedly in the last decades, by about 20% since 1992 in Nunavik (Blanchet et al., 2000; Blanchet and Rochette, 2008). As the present study shows, overall country food consumption is low (14% of total energy intake), particularly among young adults (11%). Diet across the Arctic is suboptimal. Most energy now comes from “other” food sources such as potato chips, soda pop and juices, particularly among younger generations (Blanchet and Rochette, 2008). The rapid transition towards a Western diet has translated into an important decrease in nutrients such as Se, long-chain n – 3 PUFA, iron, zinc, magnesium, calcium, fibers and vitamins A, C, D, folate, and others of the B complex, and this is combined with a drastic increase in saturated fats, sugar and salt intakes (Blanchet and Rochette, 2008; Deutch et al., 2007; Egeland et al., 2011; Gagne et al., 2012). Food insecurity is also a major challenge in the circumpolar world (Huet et al., 2012), with estimates in Nunavik ranging from 24% to 43% depending on the study (Blanchet and Rochette, 2008; Law and Harvey, 2004). Food security is defined as when all people at all times have access to sufficient, safe, nutritious food to maintain a healthy and active life (World Food Summit, 1996); when this is not the case, a situation of food insecurity arises. Consequently, increasingly poor nutrition, combined with lifestyle changes such as reduced physical activity and elevated consumption of alcohol and cigarettes, are setting the stage for a major emergence of obesity and diet-sensitive chronic diseases, particularly among Inuit youth and women (Chateau-Degat et al., 2010; Egeland et al., 2011).

In light of these considerations, the NNHC arrived at the following conclusion: “The NNHC strongly believes that country food is generally the best food for Nunavimmiut, including for pregnant women and their children. Country food consumption has significantly decreased over the last decade. Therefore, everybody must be incited to increase their consumption of country foods, the only limitation being for childbearing-age women who should limit their consumption of beluga meat. This recommendation holds until there is an evidence of a decrease in Hg content of this specific country food.” Further details can be found on the Nunavik Regional Board of Health and Social Services (NRBHSS) website (www.rssss17.gouv.qc.ca), under the NCDS section.

Moreover, in villages along the Eastern Hudson Bay, the Im\\u00a0alitsivik Health Centre, began in September 2011 a program that freely distributes Arctic char (searun spp.) to pregnant women, in order to reduce MeHg exposure and improve nutritional status among pregnant women and newborns. A study is now on-going to evaluate the efficacy of the Public Health recommendations and the Arctic char distribution program on prenatal exposure to MeHg as well as nutrient status and newborn health conditions in Nunavik. Preliminary data show that only 20% of Nunavik pregnant women are food secure, and that a quarter of pregnant women have experienced hunger (Pirkle et al., unpublished data). This highlights the urgent need for local community-adapted interventions actively promoting and facilitating the consumption of diverse local country foods and improving food security, while minimizing the risk of MeHg from beluga meat, notably among childbearing-age women.

5. Conclusion

The recommendation for childbearing-age women to limit their consumption of beluga meat in Nunavik is a short-term solution to a very complex issue. Scientists, public health and other governmental agencies agree that reducing anthropogenic Hg emissions is the only long-term solution to decrease long-range Hg transport to higher latitudes and Hg accumulation in Arctic ecosystems (Boucher et al., 2012a,b; Donaldson et al., 2010; Kirk et al., 2012; UNEP, 2013). North American emissions have decreased significantly after a peak in the 1970’s. However, global Hg emissions are increasing again due to the rapid industrial development of Asian countries and increasing artisanal gold-mining activities in low-income countries. This raises particular concerns for marine mammals and human health in the Arctic (UNEP, 2013). After several years of international negotiations, a new global legally-binding agreement, the Minamata Convention on Mercury, was signed in October 2013 to reduce global Hg emissions. However, the biochemical cycle of Hg in the Arctic is also very sensitive to global perturbations such as climate changes, which are accelerating (Stern et al., 2012). These may lead to a release of globally significant amounts of long-stored Hg and organic matter into Arctic lakes, rivers and ocean, complicating the response of Arctic ecosystems to eventual Hg global emissions reductions (UNEP, 2013).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.07.102.


