

Future Impacts of Hydroelectric Power Development on Methylmercury Exposures of Canadian Indigenous Communities

Ryan S. D. Calder,^{*,†,‡} Amina T. Schartup,^{†,‡} Miling Li,^{†,‡} Amelia P. Valberg,[†] Prentiss H. Balcom,[‡] and Elsie M. Sunderland^{†,‡}

[†]Department of Environmental Health, Harvard T. H. Chan School of Public Health, Boston Massachusetts 02215, United States

[‡]Harvard John A. Paulson School of Engineering and Applied Sciences, Cambridge Massachusetts 02138, United States

S Supporting Information

ABSTRACT: Developing Canadian hydroelectric resources is a key component of North American plans for meeting future energy demands. Microbial production of the bioaccumulative neurotoxin methylmercury (MeHg) is stimulated in newly flooded soils by degradation of labile organic carbon and associated changes in geochemical conditions. We find all 22 Canadian hydroelectric facilities being considered for near-term development are located within 100 km of indigenous communities. For a facility in Labrador, Canada (Muskrat Falls) with planned completion in 2017, we probabilistically modeled peak MeHg enrichment relative to measured baseline conditions in the river to be impounded, downstream estuary, locally harvested fish, birds and seals, and three Inuit communities. Results show a projected 10-fold increase in riverine MeHg levels and a 2.6-fold increase in estuarine surface waters. MeHg concentrations in locally caught species increase 1.3 to 10-fold depending on time spent foraging in different environments. Mean Inuit MeHg exposure is forecasted to double following flooding and over half of the women of childbearing age and young children in the most northern community are projected to exceed the U.S. EPA's reference dose. Equal or greater aqueous MeHg concentrations relative to Muskrat Falls are forecasted for 11 sites across Canada, suggesting the need for mitigation measures prior to flooding.



INTRODUCTION

Hydroelectric power accounts for 16.2% of global electricity generation and plans to greatly expand capacity are underway as countries seek to develop carbon neutral energy sources.^{1,2} In Canada, 59% of the electricity supply is from hydroelectric power and expansion is a key component of meeting international agreements on carbon dioxide (CO₂) reductions.³ Enhanced releases of CO₂, methane (CH₄), and methylmercury (MeHg) that are sustained for one to three decades following flooding are widely acknowledged.^{1,4,5} Impacts of CO₂ and CH₄ releases are global but MeHg is a neurotoxin that bioaccumulates in food webs and adversely affects individuals who rely on local ecosystems for food.⁶ Previous studies show reservoir characteristics can be used to project MeHg levels in water^{7,8} and fish⁹ following flooding but a prospective analysis of risks to human health from hydroelectric power expansion is lacking.

Traditional diets of indigenous people in the Arctic and Subarctic are rich in fish, birds, seal, and whale that provide many nutritional and cultural benefits^{10,11} but also biomagnify environmental contaminants.^{12,13} Negative impacts of MeHg exposure on neurodevelopment are well-established and widely used as the basis for regulatory thresholds.¹⁴ In northern indigenous populations, increased MeHg exposure has been

significantly associated with cardiovascular risk factors for adults such as increased resting heart rate and heart rate variability,^{15,16} as well as increased incidence of attention deficit/hyperactivity disorder (ADHD) among children with high prenatal exposures.¹⁷ Acute MeHg toxicity is associated with widespread neurological abnormalities, paresthesia and ataxia.¹⁸ In Canadian indigenous communities previously impacted by hydroelectric flooding,^{19,20} measured MeHg exposures have surpassed the lowest observed effects levels for acute MeHg toxicity.¹⁸

Inorganic mercury (Hg) is a natural component of soils and has been enriched globally by anthropogenic sources.^{21,22} MeHg is the only Hg species that biomagnifies in aquatic food webs.²³ Previously, we simulated flooding using soil cores from a planned hydroelectric reservoir in Labrador, Canada and found a 14-fold MeHg enrichment in overlying water within 3 days that was increasing exponentially at the end of the five-day experimental period.²⁴ These results suggest enhanced MeHg availability to fish, birds and seals occurs almost immediately

Received: September 1, 2016

Revised: October 21, 2016

Accepted: October 21, 2016

after reservoir flooding.²⁴ Similarly, whole ecosystem experiments in Northern Ontario show MeHg production peaks within the first 1–3 years following impoundment.^{4,7} Elevated MeHg levels in previously flooded reservoirs have gradually declined back to baseline over several decades.²⁵

Here we quantify expected increases in MeHg exposures for three Inuit communities in Labrador, Canada surrounding a hydroelectric facility to be flooded in 2016–2017 (Muskrat Falls). Our analysis considers: (a) potential MeHg enrichment in the flooded reservoir, (b) MeHg accumulation in the downstream environment (an estuary known as Lake Melville), (c) MeHg biomagnification in country foods, and (d) shifts in MeHg exposures for Inuit individuals. We use information from the Muskrat Falls site to forecast MeHg concentrations for planned hydroelectric reservoir expansion areas across Canada and discuss potential impacts on human health and mitigation strategies.

MATERIALS AND METHODS

Data from nine sites across three ecosystems were used to derive a relationship between soil organic carbon and peak methylmercury (MeHg) content of flooded soils.^{7,26–28} We excluded data from sites inundated more than three decades prior to MeHg measurements because MeHg production diminishes over time and smaller increases are observed in periodically flooded environments.^{6,24,29} For data from the Experimental Lakes Area (ELA) of Canada (boreal inceptisol soils), we used the highest MeHg concentrations following flooding for each site.⁷ Soil organic matter was converted to organic carbon using a conversion factor of 0.58, where needed.³⁰

Methods used to calculate peak MeHg fluxes from flooded soils into overlying reservoir waters for the Muskrat Falls, Labrador site are shown in [Supporting Information \(SI\) Table S1](#). Satellite data were used to derive the organic carbon content (%) of the upper 30 cm of soil in each planned reservoir.^{31,32} Post-flooding peak water column MeHg concentrations were simulated probabilistically using the distributions described in [SI Table S2](#), including (1) the 90th percentile solids diameter, (2) the sediment-water partition coefficient (K_d , L kg⁻¹) for MeHg, and (3) the MeHg fraction photochemically degraded during downstream transport.

We repeated this analysis for hydroelectric power development sites currently in the planning phase or under construction across Canada. All planned reservoirs are within 100 km of indigenous population reserves, settlements or communities, which is the approximate distance of treaty negotiated Inuit hunting and fishing territory from the Muskrat Falls facility. For all facilities, we modeled peak water column MeHg concentrations expected following flooding based on site-specific data for water discharge, flooded area, reservoir soil organic carbon, and the Muskrat Falls diffusive boundary layer estimate ([SI Table S3](#)).

For the Muskrat Falls site, downstream impacts of peak reservoir MeHg concentrations on the Lake Melville estuary were quantified using the model developed by Schartup et al.²⁴ ([SI Figure S1](#)). The estuary is permanently stratified and our previous work shows biological productivity is concentrated in the low-salinity surface layer (upper 10 m), which is the focus of this analysis. The estuarine model is based on extensive field measurements collected between 2012 and 2014 ([SI Table S4](#)). It is externally forced with probabilistically modeled freshwater MeHg inputs from the impounded river (Churchill River) from

this work, and previously characterized atmospheric deposition, and tidal inputs.²⁴ Depth-specific tidal inflows and outflows to the Lake Melville estuary are based on buoy measurements and detailed hydrodynamic modeling.³³ The annual mean flux of seawater from the subsurface to the surface layer (2.83×10^8 m³ d⁻¹) was calculated from the hydraulic budget for each vertical layer. We updated redox reactions for inorganic Hg species following the parametrization by Soerensen et al.³⁴

Baseline MeHg concentrations in locally harvested foods from the Lake Melville region were derived with the assistance of a community-led harvesting program ([SI Table S5](#)). Local foods were selected for MeHg analysis in 2014–2015 after consulting the Community Research Advisory Committee, North West River. Fish MeHg concentrations often exhibit a relationship with length.²⁵ For this study, we separated juvenile and adult size ranges and retained those most frequently consumed by Inuit community members. All fish and shellfish samples were analyzed for total Hg/MeHg and stable isotopes of carbon, nitrogen and Hg ([SI Table S6](#)).³⁵ Locally consumed seal (*Phoca hispida hispida*) muscle, liver and kidney were obtained from Inuit hunters in the spring of 2015 and analyzed for total Hg and MeHg at Environment Canada in Burlington, Ontario (see the [Supporting Information](#) for details). Data for other birds and wildlife were obtained from Environment Canada and literature values, where applicable.

Site-specific bioaccumulation factors (BAFs) for 65 locally harvested foods including fish, birds, eggs and seal ([SI Table S5](#)) were used to link modeled MeHg increases in the Churchill River and Lake Melville estuary following flooding to changes in locally harvested food concentrations ([SI Table S7](#)). This analysis assumes steady state biological MeHg concentrations with peak MeHg fluxes from the reservoir. Data from previously flooded environments indicates up to ten years are required for biota to reach maximum MeHg levels.^{29,36}

We calculated BAFs from measured MeHg concentrations in each locally consumed species and annual mean concentrations measured in the river, estuary and outer marine regions (i.e., $BAF = \text{MeHg biota/water MeHg}$). Exposure to aqueous MeHg for each species was calculated from the fraction of their lifespan spent in each environment (i.e., the sum product of aqueous MeHg concentration multiplied by the lifespan in each region). We estimated the predominant habitat/foraging regions of each species using $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, $\Delta^{199}\text{Hg}$, and $\delta^{202}\text{Hg}$ as tracers,³⁵ and literature information on their habitat preferences. We accounted for uncertainty in the time spent in each foraging region using uniform distributions that envelope the likely ranges for each species ([SI Table S2](#)) and probabilistically simulating MeHg increases. At previously flooded hydroelectric reservoirs, some typically herbivorous fish have been observed to eat fish stunned or killed by passage through hydroelectric turbines, effectively raising their trophic level and magnifying MeHg concentrations.³⁷ We do not include such potential effects in our enrichment calculations.

Hair samples were used as biomarkers of MeHg exposure for individuals in three Inuit communities (Happy Valley–Goose Bay, North West River, and Rigolet) downstream from the Muskrat Falls development area ([SI Figure S2](#)). Samples were obtained from the occipital region of the scalp with the assistance of 26 Inuit research assistants. Participants were recruited by the Nunatsiavut Government using membership rolls, which is limited to persons with demonstrated Inuit identity/ancestry. Samples were collected in both the June/July 2014 and September/October 2014 to account for any seasonal

variability in MeHg intake and ensure overlap with the peak harvest season for seals in the spring. 656 hair samples were analyzed across these two periods, representing 571 unique Inuit individuals and 19% of the total Inuit population in the region (SI Table S8). Total Hg was analyzed in the two-centimeter proximal end of hair using thermal decomposition, amalgamation, and atomic absorption spectrophotometry (EPA method 7473) with a Nippon MA-3000 or Milestone DMA-80 at Harvard University. Most of the Hg in hair is present as MeHg (>90%) and potential demethylation in the hair follicle means that total Hg is the best indicator of internal MeHg exposure.³⁸ At least one method blank and one certified hair reference materials (GBW-07601 and ERM-DB001) were tested every 10 samples and all recoveries were within certified ranges. Precision, calculated by replicate analysis of the duplicate hair samples (RSD) was better than 8.6%.

Food frequency questionnaire (FFQ) data using overlapping 24-h, 1-month and 3-month recall periods were collected in March/April 2014 concurrently with hair sampling in June/July and September/October 2014. The final FFQ survey population included 38% of Inuit individuals in the region (SI Table S9) and 1145 unique individuals. The survey included information on height, weight, sex, age. Focus group sessions were conducted with Community Research Advisory Committees to ensure comprehensiveness of country foods listed, local names and preparation methods. Interviews were conducted in-person with the use of visual aids for identification of fish meal sizes and species. Research protocols, consent procedures and the survey instrument were reviewed and approved by the Harvard Office of Human Research Administration, the Newfoundland and Labrador Health Research Ethics Authority, and the Nunatsiavut Government Research Advisory Committee prior to recruitment.

Three-month FFQ recall data from September 2014 (highest-enrollment sampling period) and the one-compartment pharmacokinetic model developed by the U.S. Environmental Protection Agency^{39,40} were used to probabilistically model baseline MeHg exposures in the three Inuit communities prior to flooding. We chose the 3-month survey period because it most closely matches the exposure period recorded by hair samples. Variability in pharmacokinetic parameters for MeHg in the human body was probabilistically simulated following the methods outlined in Li et al.⁴¹ We scaled individual fish servings to match the total meal number reported over the recall period because recall data on species-specific fish consumption tends to overestimate total consumption.^{41–43} Lognormal or gamma distributions were developed from measured MeHg concentrations in country foods (SI Table S6) and used in probabilistic exposure simulations.⁴⁴ MeHg variability in store-bought foods (SI Table S10) was simulated following Carrington and Bolger.⁴⁴

Modeled MeHg exposures were scaled by the ratio between measured and modeled hair Hg to ensure agreement with actual exposure levels. For individuals who did not provide hair samples, we adjusted modeled exposures by the median of these correction factors (mean = 0.96). Gender and age from 2011 census data were used to match the demographic distribution of the Inuit population in each of the three communities.^{45,46} Shifts in exposure resulting from flooding of the Muskrat Falls reservoir were propagated from probabilistically simulated increases in MeHg concentrations in country foods in each individual's diet.

RESULTS AND DISCUSSION

Methylmercury Increases in Flooded Reservoirs. We find a strong linear relationship across multiple ecosystems between MeHg concentrations in soils inundated within approximately three decades and their organic carbon content (Figure 1). This relationship is consistent with site-specific

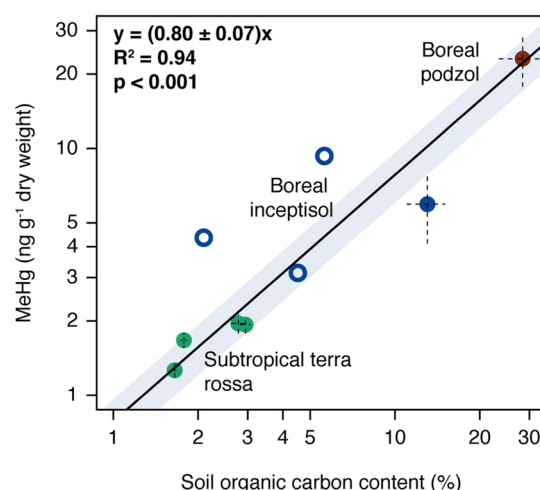


Figure 1. Relationship between soil organic carbon content and MeHg concentrations (ng g⁻¹ dry weight) of flooded soils. Each data point represents an individual sampling location. Hatched lines indicate standard errors around the mean. Soil cores are from the Wujiangu reservoir, China (subtropical terra rossa),²⁸ the Experimental Lakes Area (ELA, boreal incepsisol) in Northern Ontario, Canada,^{7,26} and La Grande-2 (Robert Bourassa) Reservoir in Quebec, Canada.²⁷ ELA data indicate the site-wide peak in MeHg (1–2 years postflood) except for the filled circle, which represents 9-years post flooding.

results from prior work.^{6,7,26} Labile organic carbon stimulates the activity of methylating microbes by providing substrate for respiration.⁴ Oxygen consumed during organic carbon degradation creates optimal geochemical conditions for anaerobic microbes (mainly sulfate reducers in flooded soils),^{4,6} thereby increasing MeHg production.

Indigenous lands are located within 100 km of all potential hydroelectric sites across Canada planned for near-term development (Figure 2). Modeled sediment-to-water MeHg fluxes across reservoirs range from 11–977 ng m⁻² day⁻¹. When normalized to soil organic carbon content, modeled fluxes (19–52 ng m⁻² day⁻¹) are consistent with those calculated from peak water column MeHg concentrations for a whole-ecosystem flooding experiment in the Experimental Lakes Area (ELA), Canada (24–115 ng m⁻² day⁻¹). For the Muskrat Falls reservoir, the expected mean flux (664 ng m⁻² day⁻¹) is within the range reported for other natural systems (2–830 ng m⁻² day⁻¹).⁴⁷

Across Canada, MeHg concentrations in hydroelectric reservoirs following flooding range from negligible for generating stations and run of the river facilities to greater than 0.5 ng L⁻¹. Forecasted MeHg concentrations in reservoir water for the Muskrat Falls site (0.19 ng L⁻¹) are moderate compared to other facilities across Canada due to its relatively smaller planned flooded area (41 km²). Highest forecasted concentrations are for a planned facility in Quebec with a relatively large flooded area (144 km²) (SI Table S3). Ten of the planned sites across Canada are expected to have postflooding MeHg concentrations lower than Muskrat Falls,

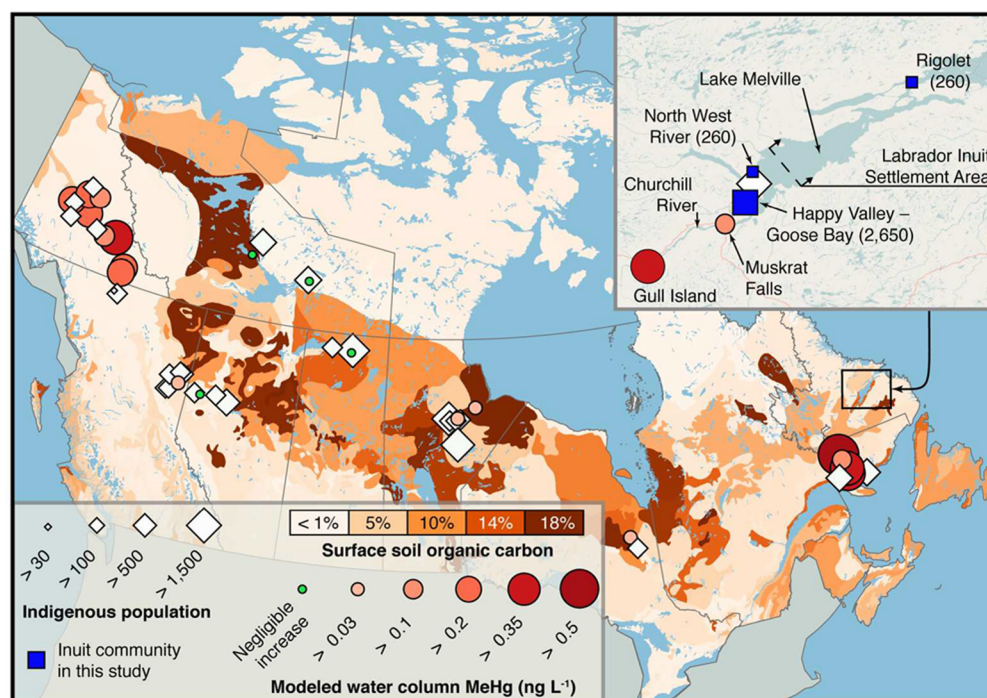


Figure 2. Planned locations for hydroelectric power expansion in Canada and indigenous populations with reserves or communities within 100 km of development regions (SI Table S3). Inset map shows the Muskrat Falls facility in Labrador and the three Inuit communities studied in this work. Reservoir MeHg concentrations are modeled for each site using the relationship shown in Figure 1 and site specific data on soil organic carbon content (upper 30 cm) of flooded reservoirs derived from satellite data, and the sediment-water flux parametrization shown in SI Table S1.

and 11 are expected to be higher (SI Figure S3). The four sites with highest projected MeHg concentrations ($>0.35 \text{ ng L}^{-1}$) have relatively large flooded areas ($85\text{--}144 \text{ km}^2$). Cumulatively, sites with projected MeHg concentrations higher than Muskrat Falls account for greater than 50% of the proposed new energy generation (SI Figure S3).

After flooding of the Muskrat Falls reservoir, the annual flow-weighted mean MeHg concentration in the Churchill River is projected to increase approximately 10-fold from a measured baseline value of $17.5 \pm 11.5 \text{ pg L}^{-1}$ (SI Table S4) to an expected mean of 180 pg L^{-1} . The fifth and 95th percentile scenarios represent 5.5 to 17-fold enrichment ($90\text{--}300 \text{ pg L}^{-1}$) relative to baseline concentrations (Figure 3). These changes represent substantial increases in MeHg concentrations in the freshwater environment that will be magnified in local food webs.

Impacts on the Downstream Environment. Few studies have considered the downstream impacts of enhanced MeHg concentrations in hydroelectric reservoirs. Kasper et al.⁴⁸ noted elevated fish MeHg concentrations up to 250 km downstream of the impoundment. However, the Muskrat Falls environmental impact assessment posited there would be no impact on a large fjord (Lake Melville) approximately 40 km downstream that contains treaty-negotiated hunting and fishing territory for Labrador Inuit, due to potential dilution throughout the water column.⁴⁹ By contrast, our previous research indicates the estuary is permanently stratified and freshwater inputs from the Churchill River are concentrated in the upper 10 m of the water column with limited mixing.²⁴ This concentrates riverine inputs within a relatively small volume of the estuary (the photic zone) that is most important for biological productivity, facilitating uptake at the base of estuarine food webs.²⁴

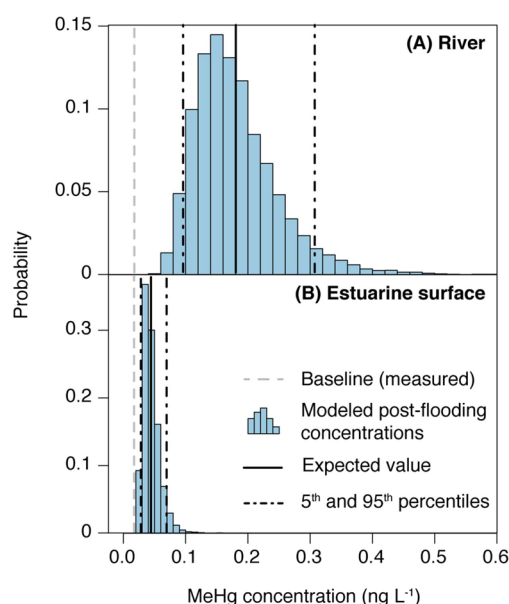


Figure 3. Probabilistically modeled scenarios for MeHg increases in downstream river and estuary of the Muskrat Falls hydroelectric facility. Photochemical MeHg demethylation is assumed to occur continuously down the reach of the Churchill River into Lake Melville thus the river concentration reflects the average of reservoir concentrations and downstream inputs to Lake Melville.

Modeling conducted here indicates expected mean MeHg concentrations in Lake Melville surface waters will increase 2.6-fold following flooding of the Muskrat Falls reservoir from 17 pg L^{-1} to a peak level of 44 pg L^{-1} (Figure 3). The fifth percentile scenario suggests a lower bound increase of 1.6-fold (28 pg L^{-1}) and the 95th percentile scenario represents a 4-fold

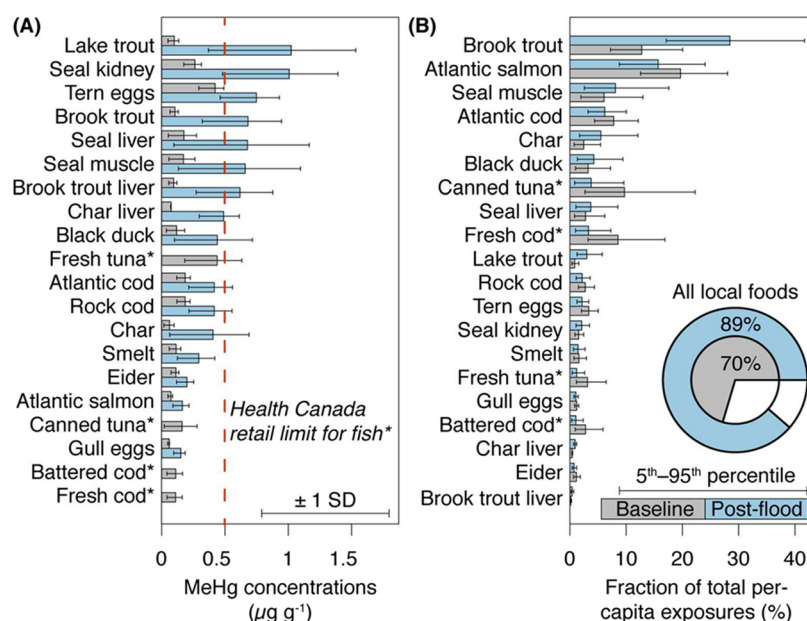


Figure 4. Top 20 MeHg exposure sources for Inuit downstream of the Muskrat Falls hydroelectric reservoir before (measured in 2014) and after flooding (modeled peak concentration) planned for 2016–2017 (SI Table S1). Commercial species unaffected by local conditions are denoted by “*”. Panel (A) shows MeHg concentrations in country foods relative to Health Canada retail limits for fish other than tuna ($0.5 \mu\text{g g}^{-1}$).⁶⁴ Error bars indicate ± 1 standard deviation for baseline and postflooding (simulated) mean. Panel (B) shows relative changes in per-capita exposures based on the expected mean exposures from probabilistic simulations. Error bars indicate 5th–95th percentiles simulated for each species. Pie charts show population-wide MeHg exposure from country foods before (measured) and after (modeled) flooding, where white space corresponds to MeHg exposure from commercial foods. A complete list of MeHg concentrations in aquatic foods are available in SI Tables S6 and S11.

increase (69 pg L^{-1}). These results suggest substantial increases in MeHg concentrations in the downstream estuary will result from flooding of the Muskrat Falls reservoir, contrasting the results of the initial Environmental Impact Assessment.⁴⁹

Methylmercury Increases in Biota. Impacts of enhanced aqueous MeHg concentrations in the river and estuary surrounding the Muskrat Falls site depend on the extent of bioaccumulation in local food webs. Site-specific BAFs for fish, birds, eggs and seal range from 10^6 to 10^8 (SI Table S7). Highest baseline MeHg concentrations are found in loon eggs, tern eggs, seal liver, and porpoise (literature value). Only porpoise presently exceeds the $0.5 \mu\text{g MeHg g}^{-1}$ Canadian retail limit⁵⁰ for most fish (Figure 4, SI Table S6).

Modeled MeHg concentrations in the top 20 local foods contributing to Inuit MeHg exposure after flooding range from 1.3 to 10 times measured baseline concentrations (Figure 4). This is consistent with two- to 9-fold increases in fish MeHg concentrations previously reported for other Canadian reservoirs.^{36,51} Variable impacts of flooding across species downstream of Muskrat Falls mainly reflects differences in foraging activity (i.e., time spent in the river, estuary and outer marine regions, SI Table S7). For example, brook trout are highly enriched in MeHg following flooding due to the large fraction of their lifespan spent in the freshwater environment (SI Table S11).

After flooding, expected mean MeHg concentrations in lake trout, seal, tern eggs, brook trout and char liver are all projected to be at or above the Canadian retail limit for MeHg (Figure 4A). Black duck, Atlantic cod and rock cod also exceed this level under the 95th percentile environmental increase scenario. After flooding, almost 90% of population-wide MeHg exposure is projected to be from locally caught foods (Figure 4B). Increasing MeHg burdens of traditional country foods consumed by Inuit will elevate their MeHg exposures and

may adversely affect local wildlife that are sensitive to high levels of MeHg exposure.⁵²

Inuit Exposures and Risks. Measured hair Hg concentrations in 474 individuals from the three Inuit communities downstream of Muskrat Falls show over 90% of baseline (ca. 2014) MeHg exposures are below regulatory guidelines for MeHg in the U.S. and Canada (Figure S4). Highest exposures are found in the most northern community of Rigolet, where 24% of individuals presently exceed the U.S. Environmental Protection Agency’s (U.S. EPA) Reference Dose for MeHg (RfD, $0.1 \mu\text{g kg}^{-1} \text{ body weight day}^{-1}$), and 3% are above Health Canada’s (HC) provisional tolerable daily intake (pTDI, $0.20\text{--}0.47 \mu\text{g kg}^{-1} \text{ body weight day}^{-1}$). Mean exposure levels in Rigolet in 2014 were similar to those reported in the 2007–2008 Inuit Health Survey for other communities along the Labrador coastline.⁵³ All three Inuit communities downstream of Muskrat Falls have higher MeHg exposure levels than the general Canadian population due to greater consumption of aquatic foods.⁵⁴

Following flooding of the Muskrat Falls reservoir, median MeHg exposures are expected to at least double for the majority of the downstream Inuit population (Figure 5A). Projected increases are greatest in the community of Rigolet, where the median exposure increase is projected to be almost three times baseline values. Disproportionate increases in MeHg exposures occur for individuals who are already the most highly exposed and consume the greatest quantities of country foods. For example, mean MeHg intake increases from 0.15 to $0.50 \mu\text{g kg}^{-1} \text{ day}^{-1}$ for individuals at the 90th percentile of postflooding exposures and this demographic accounts for nearly 60% of the total additional MeHg intake ($\mu\text{g day}^{-1}$) following flooding.

Average MeHg exposure levels for women of childbearing age^{16–49} and young children (age <12) in the community of

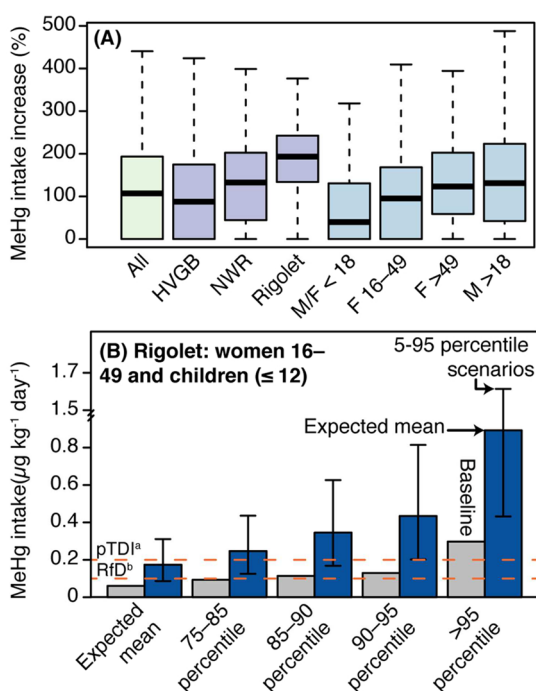


Figure 5. (a) Health Canada (HC) provisional tolerable daily intake (pTDI); (b) U.S. EPA reference dose (RfD) for MeHg. Modeled changes in Inuit MeHg exposures following flooding of the Muskrat Falls reservoir. Panel (A) shows exposure increases relative to measured baseline intake in 2014. Error bars indicate the 5th-95th percentile scenarios for MeHg increases in the flooded reservoir and biota based on probabilistic simulations. Panel (B) shows baseline and postflooding MeHg intake in women of childbearing age¹⁶⁻⁴⁹ and children (age <12) in the community of Rigolet. HVGB = Happy Valley–Goose Bay, NWR = North West River.

Rigolet exceed the U.S. EPA's RfD (Figure 5B) and are within 15% of Health Canada's provisional tolerable daily intake (pTDI) level.⁵⁵ This demographic is most sensitive to the neurodevelopmental impacts of MeHg exposure.⁵⁶ Beyond the 75th percentile of this population, all individuals are above both regulatory guidelines for MeHg (Figure 5B). Grandjean and Budtz-Jorgensen⁵⁷ found imprecision in the biomonitoring data used to formulate the U.S. EPA's RfD led to an overestimate of 50% and proposed that a revised RfD of $0.05 \mu\text{g kg}^{-1} \text{ day}^{-1}$ ($0.58 \mu\text{g Hg g}^{-1} \text{ hair}$) would be more appropriate. In Rigolet, 77% of individuals exceed this level (Figure S5). Exposures are lower in the other two communities due to more limited consumption of country foods (Figure S6). Across the three communities, 41% of the total population and 28% of women of childbearing age (Figure S5) exceed the level proposed by Grandjean and Budtz-Jorgensen.⁵⁷

Regulatory thresholds such as a RfD imply the existence of a safe level of chronic exposure. However, when formulating the RfD, the U.S. EPA itself acknowledged that “no evidence of a threshold arose for methylmercury-related neurotoxicity”.³⁹ Recent data from prospective birth cohorts support this conclusion.⁵⁸⁻⁶⁰ For adults, the Health Canada pTDI is the least conservative across international regulatory agencies ($0.47 \mu\text{g kg}^{-1} \text{ day}^{-1}$). Therefore, all consumers of local foods are likely to face decreased net health benefits as a result of increased MeHg in local foods.

Pan Canada Implications. Modeled reservoir MeHg levels at 11 of the proposed 21 hydroelectric sites across Canada are comparable or greater than the Muskrat Falls reservoir (Figure

2). The communities of Happy Valley-Goose Bay and Northwest River consume fewer country foods than typical of most indigenous populations in Canada,⁵³ suggesting potentially greater exposures of other indigenous communities with moderate and high projected reservoir MeHg levels.

Country foods are known to confer a wide-range of nutritional and social health benefits to indigenous communities and nutritious alternative food choices are limited in the Canadian North.^{10,11} Past studies suggest reducing or avoiding consumption of country foods may also result in substantial adverse impacts on individual health.^{61,62} Reducing environmental MeHg concentrations associated with hydroelectric flooding should thus be prioritized as a mitigation measure. For example, soil organic carbon content could be used as a screening criterion for site selection or reservoirs could be designed to minimize flooded area. Mailman et al.⁶³ review a number of other interventions, such as the removal of organic carbon from the planned reservoir regions prior to flooding.

■ ASSOCIATED CONTENT

● Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b04447.

Details on reservoir model parameters and algorithms, characteristics of planned hydroelectric power facilities across Canada, probabilistic exposure assessment methods, dietary survey data and MeHg measurements in water column and biota are provided (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*Phone: (617) 637-6484; fax: (617) 495-4551; e-mail: ry.calder@mail.harvard.edu.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was supported by the U.S. National Science Foundation (OCE 1260464 and 1130549), the Canadian Northern Contaminants Program, ArcticNet Inc., Tides Canada Oak Arctic Marine Fund Program, and the Nunatsiavut Government (NG). We thank research assistants employed by the NG for collection of dietary survey data, and Marina Biasutti-Brown, Tom Sheldon, Rodd Laing, and Trevor Bell for assistance with sample collection and human health survey work. We thank Nil Basu (McGill U.), Jane Kirk and Derek Muir (Env. Canada) for assistance with seal sample shipping and analysis and Neil Burgess (Env. Canada) for information on egg and bird Hg data and foraging behavior.

■ REFERENCES

- (1) Barros, N.; Cole, J. J.; Tranvik, L. J.; Prairie, Y. T.; Bastviken, D.; Huszar, V. L. M.; del Giorgio, P.; Roland, F. Carbon emission from hydroelectric reservoirs linked to reservoir age and latitude. *Nat. Geosci.* **2011**, 4 (9), 593–596.
- (2) International Energy Agency (IEA). *Renewable Energy Essentials: Hydropower*, 2010.
- (3) Natural Resources Canada (NRCAN). About Renewable Energy. <http://www.nrcan.gc.ca/energy/electricity-infrastructure/about-electricity/7359> (accessed February 28, 2016).
- (4) St. Louis, V. L.; Rudd, J. W.; Kelly, C. A.; Bodaly, R.; Paterson, M. J.; Beaty, K. G.; Hesslein, R. H.; Heyes, A.; Majewski, A. R. The Rise

and fall of mercury methylation in an experimental reservoir. *Environ. Sci. Technol.* **2004**, *38* (5), 1348–1358.

(5) Rudd, J.; Harris, R.; Kelly, C.; Hecky, R. Are hydroelectric reservoirs significant sources of greenhouse gases? *Ambio*. **1993**, *22* (4), 246–248.

(6) Rosenberg, D. M.; Berkes, F.; Bodaly, R.; Hecky, R.; Kelly, C.; Rudd, J. W. Large-scale impacts of hydroelectric development. *Environ. Rev.* **1997**, *5* (1), 27–54.

(7) Hall, B. D.; St. Louis, V. L.; Rolfhus, K. R.; Bodaly, R. A.; Beaty, K. G.; Paterson, M. J.; Cherewyk, K. A. P. Impacts of reservoir creation on the biogeochemical cycling of methyl mercury and total mercury in boreal upland forests. *Ecosystems* **2005**, *8* (3), 248–266.

(8) Rolfhus, K.; Hurley, J.; Bodaly, R.; Perrine, G. Production and retention of methylmercury in inundated boreal forest soils. *Environ. Sci. Technol.* **2015**, *49* (6), 3482–3489.

(9) Johnston, T. A.; Bodaly, R.; Mathias, J. Predicting fish mercury levels from physical characteristics of boreal reservoirs. *Can. J. Fish. Aquat. Sci.* **1991**, *48* (8), 1468–1475.

(10) Kuhnlein, H.; Receveur, O. Local cultural animal food contributes high levels of nutrients for arctic Canadian indigenous adults and children. *J. Nutr.* **2007**, *137* (4), 1110–1114.

(11) Receveur, O.; Boulay, M.; Kuhnlein, H. V. Decreasing traditional food use affects diet quality for adult Dene/Métis in 16 communities of the Canadian Northwest Territories. *J. Nutr.* **1997**, *127* (11), 2179–2186.

(12) Donaldson, S.; Van Oostdam, J.; Tikhonov, C.; Feeley, M.; Armstrong, B.; Ayotte, P.; Boucher, O.; Bowers, W.; Chan, L.; Dallaire, F.; Dallaire, R.; Dewailly, E.; Edwards, J.; Egeland, G.; Fontaine, J.; Furgal, C.; Leech, T.; Loring, E.; Muckle, G.; Nancarrow, T.; Pereg, D.; Plusquellec, P.; Potyrala, M.; Receveur, O.; Shearer, R. Environmental contaminants and human health in the Canadian Arctic. *Sci. Total Environ.* **2010**, *408* (22), S165–S234.

(13) Stow, J.; Krümmel, E.; Leech, T.; Donaldson, S., What is the impact of mercury contamination on human health in the Arctic? In *AMAP Assessment 2011: Mercury in the Arctic*; Arctic Monitoring and Assessment Programme (AMAP): Oslo, Norway, 2011.

(14) National Research Council (NRC). *Toxicological Effects of Methylmercury*; National Academies Press: Washington, DC, 2000.

(15) Valera, B.; Dewailly, E.; Poirier, P. Impact of mercury exposure on blood pressure and cardiac autonomic activity among Cree adults (James Bay, Quebec, Canada). *Environ. Res.* **2011**, *111*, 1265–1270.

(16) Valera, B.; Dewailly, E.; Poirier, P. Association between methylmercury and cardiovascular risk factors in a native population of Quebec (Canada): A retrospective evaluation. *Environ. Res.* **2013**, *120*, 102–108.

(17) Boucher, O.; Jacobson, S.; Plusquellec, P.; Dewailly, E.; Ayotte, P.; Forget-Dubois, N.; Jacobson, J.; L, J.; Muckle, G. Prenatal methylmercury, postnatal lead exposure, and evidence of attention deficit/hyperactivity disorder among Inuit children in Arctic Quebec. *Environ. Health Perspect.* **2012**, *120* (10), 1456–1461.

(18) Clarkson, T. W.; Amin-Zaki, L.; Al-Tikriti, S. An outbreak of methylmercury poisoning due to consumption of contaminated grain. *Federation proceedings* **1976**, *35* (12), 2395–2399.

(19) Methylmercury Study Group. *McGill Methylmercury Study*. 1980.

(20) Dumont, C.; Girard, M.; Bellavance, F. O.; Noël, F. Mercury levels in the Cree population of James Bay, Quebec, from 1988 to 1993/94. *Can. Med. Assoc. J.* **1998**, *158* (11), 1439–1445.

(21) Amos, H. M.; Jacob, D. J.; Streets, D. G.; Sunderland, E. M. Legacy impacts of all-time anthropogenic emissions on the global mercury cycle. *Global Biogeochem. Cy.* **2013**, *27* (2), 410–421.

(22) Smith-Downey, N.; Sunderland, E.; Jacob, D. Anthropogenic impacts on global storage and emissions of mercury from terrestrial soils: Insights from a new global model. *J. Geophys. Res.* **2010**, *115*, G03008.

(23) Lavoie, R. A.; Jardine, T. D.; Chumchal, M. M.; Kidd, K. A.; Campbell, L. M. Biomagnification of mercury in aquatic food webs: a worldwide meta-analysis. *Environ. Sci. Technol.* **2013**, *47* (23), 13385–13394.

(24) Schartup, A. T.; Balcom, P. H.; Soerensen, A. L.; Gosnell, K. J.; Calder, R. S. D.; Mason, R. P.; Sunderland, E. M. Freshwater discharges drive high levels of methylmercury in Arctic marine biota. *Proc. Natl. Acad. Sci. U. S. A.* **2015**, *112* (38), 11789–94.

(25) Anderson, M. Duration and extent of elevated mercury levels in downstream fish following reservoir creation. *River Syst.* **2011**, *19* (3), 167–176.

(26) Rolfhus, K. R.; Hurley, J. P.; Bodaly, R. A.; Perrine, G. Production and retention of methylmercury in inundated boreal forest soils. *Environ. Sci. Technol.* **2015**, *49* (6), 3482–3489.

(27) Mucci, A.; Montgomery, S.; Lucotte, M.; Plourde, Y.; Pichet, P.; Tra, H. V. Mercury remobilization from flooded soils in a hydroelectric reservoir of northern Quebec, La Grande-2: results of a soil resuspension experiment. *Can. J. Fish. Aquat. Sci.* **1995**, *52* (11), 2507–2517.

(28) Meng, B.; Feng, X.; Qiu, G.; Li, Z.; Yao, H.; Shang, L.; Yan, H. The impacts of organic matter on the distribution and methylation of mercury in a hydroelectric reservoir in Wujiang River, Southwest China. *Environ. Toxicol. Chem.* **2016**, *35* (1), 191–199.

(29) Schetagne, R.; Verdon, R. Post-impoundment evolution of fish mercury levels at the La Grande Complex, Québec, Canada (from 1978 to 1996). In *Mercury in the Biogeochemical Cycle*; Lucotte, M. et al., Eds.; Springer, 1999; pp 235–258.

(30) Qian, Y.; Follett, R. Carbon Dynamics and Sequestration in Urban Turfgrass Ecosystems. In *Carbon Sequestration in Urban Ecosystems*; Lal, R.; Augustin, B., Eds.; Springer, 2012, 161–172.

(31) European Soil Data Centre (ESDAC). *Global Soil Organic Carbon Estimates*, 2015.

(32) Périé, C.; Ouimet, R. Organic carbon, organic matter and bulk density relationships in boreal forest soils. *Can. J. Soil Sci.* **2008**, *88* (3), 315–325.

(33) Lu, Z.; DeYoung, B.; Banton, S. *Analysis of Physical Oceanographic Data from Lake Melville, Labrador, September 2012 - July 2013*; Memorial University of Newfoundland: St. John's, Newfoundland, 2014.

(34) Soerensen, A. L.; Jacob, D. J.; Schartup, A. T.; Fisher, J. A.; Lehnher, I.; Louis, V. L., St; Heimburger, L. E.; Sonke, J. E.; Krabbenhoft, D. P.; Sunderland, E. M. A mass budget for mercury and methylmercury in the Arctic Ocean. *Global Biogeochem. Cy.* **2016**, *30* (4), S60–S75.

(35) Li, M.; Schartup, A. T.; Valberg, A. P.; Ewald, J. D.; Krabbenhoft, D. P.; Yin, R.; Balcom, P. H.; Sunderland, E. M. Environmental Origins of Methylmercury Accumulated in Subarctic Estuarine Fish Indicated by Mercury Stable Isotopes. *Environ. Sci. Technol.* **2016**. DOI: [10.1021/acs.est.6b03206](https://doi.org/10.1021/acs.est.6b03206).

(36) Verdon, R.; Brouard, D.; Demers, C.; Lalumière, R.; Laperle, M.; Schetagne, R. Mercury evolution (1978–1988) in fishes of the La Grande hydroelectric complex, Quebec, Canada. *Water, Air, Soil Pollut.* **1991**, *56*, 405–417.

(37) Brouard, D.; Doyon, J.-F.; Schetagne, R. Amplification of Mercury Concentrations in Lake Whitefish (*Coregonus clupeaformis*) Downstream from the La Grande 2 Reservoir, James Bay, Quebec. In *Mercury Pollution Integration and Synthesis*; Watras, C. J.; Huckabee, J. W., Eds.; CRC Press, 1994.

(38) Berglund, M.; Lind, B.; Bjornberg, K. A.; Palm, B.; Einarsson, O.; Vahter, M. Inter- individual variations of human mercury exposure biomarkers: a cross-sectional assessment. *Environ. Health* **2005**, *4*, 20.

(39) United States Environmental Protection Agency (US EPA). Methylmercury (MeHg); CASRN 22967-92-6. In *Integrated Risk Information System (IRIS)*, 2002.

(40) World Health Organization (WHO). *Methylmercury*, 1990.

(41) Li, M.; von Stackelberg, K.; Rheinberger, C. M.; Hammitt, J. K.; Krabbenhoft, D. P.; Yin, R.; Sunderland, E. M. Insights from mercury stable isotopes into factors affecting the internal body burden of methylmercury in frequent fish consumers. *Elementa*. **2016**, *4*, 000103.

(42) Lincoln, R. A.; Shine, J. P.; Chesney, E. J.; Vorhees, D. J.; Grandjean, P.; Senn, D. B. Fish consumption and mercury exposure among Louisiana recreational anglers. *Environ. Health Perspect.* **2011**, *119* (2), 245–251.

- (43) Dong, Z.; Jim, R. C.; Hatley, E. L.; Backus, A. S.; Shine, J. P.; Spengler, J. D.; Schaider, L. A. A longitudinal study of mercury exposure associated with consumption of freshwater fish from a reservoir in rural south central USA. *Environ. Res.* **2015**, *136*, 155–62.
- (44) Carrington, C. D.; Bolger, M. P. An exposure assessment for methylmercury from seafood for consumers in the United States. *Risk Anal* **2002**, *22* (4), 689–699.
- (45) Statistics Canada. 2011 National Household Survey. 2013.
- (46) Statistics Canada. 2011 Census. 2012.
- (47) Gill, G. A.; Bloom, N. S.; Cappellino, S.; Driscoll, C. T.; Dobbs, C.; McShea, L.; Mason, R.; Rudd, J. W. Sediment-water fluxes of mercury in Lavaca Bay, Texas. *Environ. Sci. Technol.* **1999**, *33* (5), 663–669.
- (48) Kasper, D.; Forsberg, B. R.; Amaral, J. O. H.; Leitão, R. P.; Py-Daniel, S. S.; Bastos, W. R.; Malm, O. Reservoir Stratification Affects Methylmercury Levels in River Water, Plankton, and Fish Downstream from Balbina Hydroelectric Dam, Amazonas, Brazil. *Environ. Sci. Technol.* **2014**, *48* (2), 1032–1040.
- (49) Nalcor Energy. *Biophysical Assessment*, 2009.
- (50) Health Canada. *Health Canada's Maximum Levels for Chemical Contaminants in Foods*. <http://www.hc-sc.gc.ca/fn-an/securit/chem-chim/contaminants-guidelines-directives-eng.php> (accessed August 16, 2016).
- (51) Harris, R.; Hutchinson, D. *Assessment of the Potential for Increased Mercury Concentrations*. 2008.
- (52) Depew, D. C.; Basu, N.; Burgess, N. M.; Campbell, L. M.; Devlin, E. W.; Drewnick, P. E.; Hammerschmidt, C. R.; Murphy, C. A.; Sandheinrich, M. B.; Wiener, J. G. Toxicity of dietary methylmercury to fish: derivation of ecologically meaningful threshold concentrations. *Environ. Toxicol. Chem.* **2012**, *31* (7), 1536–1547.
- (53) Chan, H. M. L. *Contaminant Assessment in Nunatsiavut*. 2011.
- (54) Lye, E.; Legrand, M.; Clarke, J.; Probert, A. Blood total mercury concentrations in the Canadian population: Canadian health measures survey cycle 1, 2007–2009. *Can. J. Public Health.* **2013**, *104* (3), E246–E251.
- (55) Health Canada. *Mercury: Your Health and the Environment: A Resource Tool*, 2004.
- (56) Mahaffey, K. R.; Sunderland, E. M.; Chan, H. M.; Choi, A. L.; Grandjean, P.; Mariën, K.; Oken, E.; Sakamoto, M.; Schoeny, R.; Weihe, P.; Yan, C.-H.; Yasutake, A. Balancing the benefits of n-3 polyunsaturated fatty acids and the risks of methylmercury exposure from fish consumption. *Nutr. Rev.* **2011**, *69* (9), 493–508.
- (57) Grandjean, P.; Budtz-Jorgensen, E. Total imprecision of exposure biomarkers: implications for calculating exposure limits. *Am. J. Ind. Med.* **2007**, *50* (10), 712–719.
- (58) Rice, G. E.; Hammitt, J. K.; Evans, J. S. A probabilistic characterization of the health benefits of reducing methyl mercury intake in the United States. *Environ. Sci. Technol.* **2010**, *44* (13), 5216–5224.
- (59) Karagas, M.; Choi, A. L.; Oken, E.; Horvat, M.; Schoeny, R.; Kamai, E.; Grandjean, P.; Korrick, S. Evidence on the human health effects of low level methylmercury exposure. *Environ. Health Perspect.* **2012**, *120* (6), 799–806.
- (60) Roman, H. A.; Walsh, T. L.; Coull, B. A.; Dewailly, E.; Guallar, E.; Hattis, D.; Marien, K.; Schwartz, J.; Stern, A. H.; Virtanen, J. K.; Rice, G. Evaluation of the cardiovascular effects of methylmercury exposures: current evidence supports development of a dose-response function for regulatory benefits analysis. *Environ. Health Perspect.* **2011**, *119* (5), 607–14.
- (61) Wheatley, B.; Paradis, S. Balancing human exposure, risk and reality: questions raised by the Canadian aboriginal methylmercury program. *Neurotoxicology.* **1996**, *17* (1), 241–249.
- (62) Furgal, C.; Powell, S.; Myers, H. Digesting the message about contaminants and country foods in the Canadian North: a review and recommendations for future research and action. *Arctic* **2005**, *58*, 103–114.
- (63) Mailman, M.; Stepnuk, L.; Cicek, N.; Bodaly, R. A. Strategies to lower methyl mercury concentrations in hydroelectric reservoirs and lakes: A review. *Sci. Total Environ.* **2006**, *368* (1), 224–235.
- (64) Health Canada. *Updating the Existing Risk Management Strategy for Mercury in Retail Fish*, 2007.

Supporting Information

Future Impacts of Hydroelectric Power Development on Methylmercury Exposures of Canadian Indigenous Communities

Ryan S.D. Calder^{a,b*}, Amina T. Schartup^{a,b}, Miling Li^{a,b}, Amelia P. Valberg^a, Prentiss Balcom^b, and Elsie M. Sunderland^{a,b}

^a Department of Environmental Health, Harvard T.H. Chan School of Public Health, Boston MA, 02215, USA

^b Harvard John A. Paulson School of Engineering and Applied Sciences, Cambridge MA, 02138, USA

*Corresponding author: 29 Oxford St., Cambridge MA, 02138, United States. T: (617) 637-6484; F: (617) 495-4551. ry.calder@mail.harvard.edu

Contents

Supplemental Information on Seal Mercury Analyses	10
---	----

Tables

Table S1. Methods used to calculate transport of MeHg from sediment into the water column	2
Table S2. Distribution of parameters for MeHg flux from sediments to water column	3
Table S3. Characteristics of planned hydroelectric power projects across Canada	4
Table S4. Measured methylmercury concentrations in the Churchill River between 2012-2015	6
Table S5. Community-based monitoring of fish species from the Lake Melville region	7
Table S6a. MeHg concentrations in aquatic species harvested from the Lake Melville region	8
Table S6b.	9
Table S7a. MeHg bioaccumulation factors, concentrations and lifetime habitat fractions	11
Table S7b.	12
Table S8. Hair mercury analyses of Inuit individuals	14
Table S9. Food frequency questionnaire (FFQ) data collected from Inuit	15
Table S10. MeHg concentrations in aquatic foods harvested outside the Lake Melville region	16
Table S11a. Modeled MeHg concentrations in country foods after flooding	18
Table S11b.	19

Figures

Figure S1. Schematic of model for mercury cycling the Lake Melville estuary	5
Figure S2. Map of the Labrador Inuit Settlement Area	13
Figure S3. Number of proposed hydroelectric sites with reservoir MeHg concentrations	17
Figure S4. Measured concentrations of total Hg in hair from three Inuit communities	20
Figure S5. Baseline and post-flooding MeHg intake relative to intake levels	21
Figure S6. Fraction of population exceeding exposure thresholds at baseline	22

Table S1. Methods used to calculate sediment-water exchange of MeHg.

J (ng m ⁻² s ⁻¹)	Flux of MeHg into the water column based on the mass transfer formulation of Steinberger and Hondzo (1)	$J = \frac{C_{pw} - C_w}{\delta_d} D$
D (m ² s ⁻¹)	Molecular diffusivity for MeHg (2)	2x10 ⁻¹⁰ (macromolecular organic complexes)
dC_w/dt (ng L ⁻¹ s ⁻¹)	Rate of change of MeHg concentration in the water column determined by flux from flooded soil and outflow from river	$\frac{1}{V} \left(\frac{C_{pw} - C_w}{\delta_d} D A_f - Q C_w + Q C_{wb} \right) 10^3 \frac{L}{m^3}$
C_w (ng L ⁻¹)	Steady state ($dC_w/dt = 0$) concentrations of MeHg in reservoir water	$\frac{C_{pw} \cdot D \cdot A_f}{D \cdot A_f + \delta_d \cdot Q} + C_{wb}$
C_{pw} (ng L ⁻¹)	Concentration of MeHg in interstitial waters	Derived from empirical relationship in Figure 1 and K_d
C_{wb} (ng L ⁻¹)	Pre-impoundment riverine MeHg (3)	0.0175
$\log K_d$ [L kg ⁻¹]	Sediment-water partition coefficient based on measurements (3)	2.93±0.16
A_f (m ²)	Land area flooded	Table S2
Q (m ³ s ⁻¹)	River flow	Table S2
δ_d (m)	Thickness of the diffusive sublayer controlled by turbulent action based on Peterson (4)	$\nu^{1-\frac{1}{n}} \cdot \frac{1}{\sqrt{\tau/\rho}} \cdot D^{\frac{1}{n}} \cdot c^{-\frac{1}{n}}$
ν (m ² s ⁻¹)	Kinematic viscosity of water	1.3x10 ⁻⁶
ρ (kg m ⁻³)	Density of water	10 ³
C (unitless)	Coefficient	0.000463
n (unitless)	Coefficient	3.38
τ (N m ⁻²)	Post-impoundment shear stress at the sediment-water interface based on Wilcock (5)	$\left[U \cdot \kappa / \ln \left(\frac{h}{e \cdot a \cdot \frac{d_{90}}{30}} \right) \right]^2 \rho$
U (m s ⁻¹)	Average current velocity based on Muskrat Falls facility (6)	0.1
κ (unitless)	von Karman constant	0.41
d_{90} (mm)	90 th percentile solids diameter based on the predominant soil type in the Muskrat Falls reservoir area (7, 8)	0.2
a (unitless)	Constant	2.85
h (m)	Height of the channel based on Muskrat Falls facility	16.8
e (unitless)	Base of the natural logarithm	2.718

Table S2. Distributions of uncertain parameters used to simulate MeHg enrichment in water and biota in flooded reservoirs. Table S1 contains the complete parameterization for sediment-to-water fluxes of MeHg.

Parameter	Distribution
90 th percentile solids diameter in reservoir (d_{90} , mm) ^a	Triangular: min = 0.005, max = 1, mode = 0.2
Sediment-water partition coefficient ($\log K_d$, L kg ⁻¹) ^b	Normal: $\mu = 2.96$, $\sigma = 2.54$
Degradation of MeHg during downstream transport to estuary (fraction lost) ^c	Uniform: min = 0.3, max = 0.5
Fraction of excess riverine MeHg demethylatable in Lake Melville ^d	Uniform: min = 0, max = 1
Estuarine fraction of lifespan for key marine species ^e	Uniform: min = 0, max = 0.5
Estuarine fraction of lifespan for key bird species ^f	Uniform: min = 0.5, max = 1
Riverine fraction of lifespan for seals ^g	Uniform: min = 0, max = 0.25

^a Mode based on the dominant soil type (podzol) in the Muskrat Falls region (7); minimum and maximum values represent ranges across a variety of soil types (8).

^b Probability distribution for site-wide mean derived from measurements (5).

^c Maximum degradation is based on upper limit suggested by Schartup et al. (3); minimum is based on degradation rate measured by Jonsson et al. (9).

^d MeHg complexed to terrestrial organic ligands may be resistant to degradation (9).

^e Fraction of MeHg obtained from the estuarine environment during foraging and/or spawning is uncertain for Atlantic cod, Atlantic salmon, and rock cod.

^f Seabirds (eider, tern, guillemot and gull) are found in both the marine and estuarine environments. Some birds consumed by Inuit may spend their entire life history foraging in the estuary (maximum) or in outer marine areas (minimum).

^g Inuit hunters report seasonal seal foraging in the freshwater environment.

Table S3. Characteristics of planned hydroelectric power projects across Canada.

Hydroelectric Project (River, Province/Territory)	Flow (m ³ s ⁻¹)	Flood area (km ²)	Post-flood MeHg (ng L ⁻¹)	Capacity (MW)	Indigenous populations within 100 km ^a
False Canyon (Liard, YT) ^b	151	160	0.24	58	Liard
Middle Canyon (Liard, YT) ^b	160	90	0.21	38	Liard, Dease
Detour Canyon (Pelly, YT) ^b	257	135	0.22	65	Selkirk, Little Salmon
Granite Canyon (Pelly, YT) ^b	362	170	0.21	254	
Hoole Canyon (Pelly, YT) ^b	97	25	0.13	13	Ross River
Slate Rapids (Pelly, YT) ^b	53	136	0.35	42	
Fraser Falls (Stewart, YT) ^b	359	570	0.29	300	Nacho Nyak Dun, Selkirk
Two Mile Canyon (Stewart, YT) ^b	166	105	0.18	53	Nacho Nyak Dun
La Martre (La Martre, NT) ^c	31	0	•	13	Whati
Lutselk'e (Snowdrift, NT) ^c	42	0	•	1	Lutsel K'e Dene
Site C (Peace, BC) ^d	1251	53	0.04	1100	West Moberly, Saulteau, Doig River, Halfway River, Blueberry River
Amisk (Peace, AB) ^e	1600	8	•	330	Duncan's, Horse Lake, Peavine Metis
Tazi Twé (Fond du Lac, SK) ^f	304	0	•	50	Black Lake, Fond du Lac, Fox Lake, War Lake, York Factory, Tataskweyak, Bunibonibee
Keeyask (Nelson, MB) ^g	3100	45	0.06	695	Fox Lake
Conawapa (Nelson, MB) ^h	3100	5	0.04	500	Taykwa Tagamou
New Post Creek (Abitibi, ON) ⁱ	42	2	0.04	25	Quebec Innu (Ekuanitshit, Nutashkuan)
Romaine 1 (La Romaine, QC) ^j	291	12	0.35	270	
Romaine 2 (La Romaine, QC) ^j	291	85	0.38	640	
Romaine 3 (La Romaine, QC) ^j	291	37	0.20	395	
Romaine 4 (La Romaine, QC) ^j	291	144	0.55	245	Labrador Inuit, Innu and Metis
Muskrat Falls (Churchill, NL) ^k	1829	41	0.19	824	
Gull Island (Churchill, NL) ^k	1829	85	0.37	2250	

• Negligible increase from baseline.

^a First Nations unless otherwise specified. Locations on Figure 2 are centroids of traditional lands (10, 11). First Nations populations are those living on their respective reserves and uncaded lands (12).

^b Comparative feasibility assessment ongoing (13).

^c Under review (14).

^d Construction began in 2015 and will continue through 2024 (15, 16).

^e Permitting process ongoing. Peavine settlement is 169 km from project but traditional lands review is ongoing (17).

^f Permitting process ongoing (18).

^g Construction began in 2014 and will continue through 2021 (19, 20).

^h Planning activities suspended pending results of resources planning review (21).

ⁱ Construction began in 2015 and will continue through 2018 (22).

^j Construction began in 2009 and will continue through 2017 (Romaine 3) – 2020 (Romaine 4). Construction complete on Romaine 1 and 2. Nutashkuan (132 km from Romaine 1) and Ekuanitshit and are the indigenous communities found to use the land impacted by the development (23).

^k Construction of Muskrat Falls began in 2013 and will continue through 2017 (24). A construction timetable for Gull Island has not been released. Labrador Metis (NunatuKavut) is not plotted on Figure 2 because it does not have a recognized land claim.

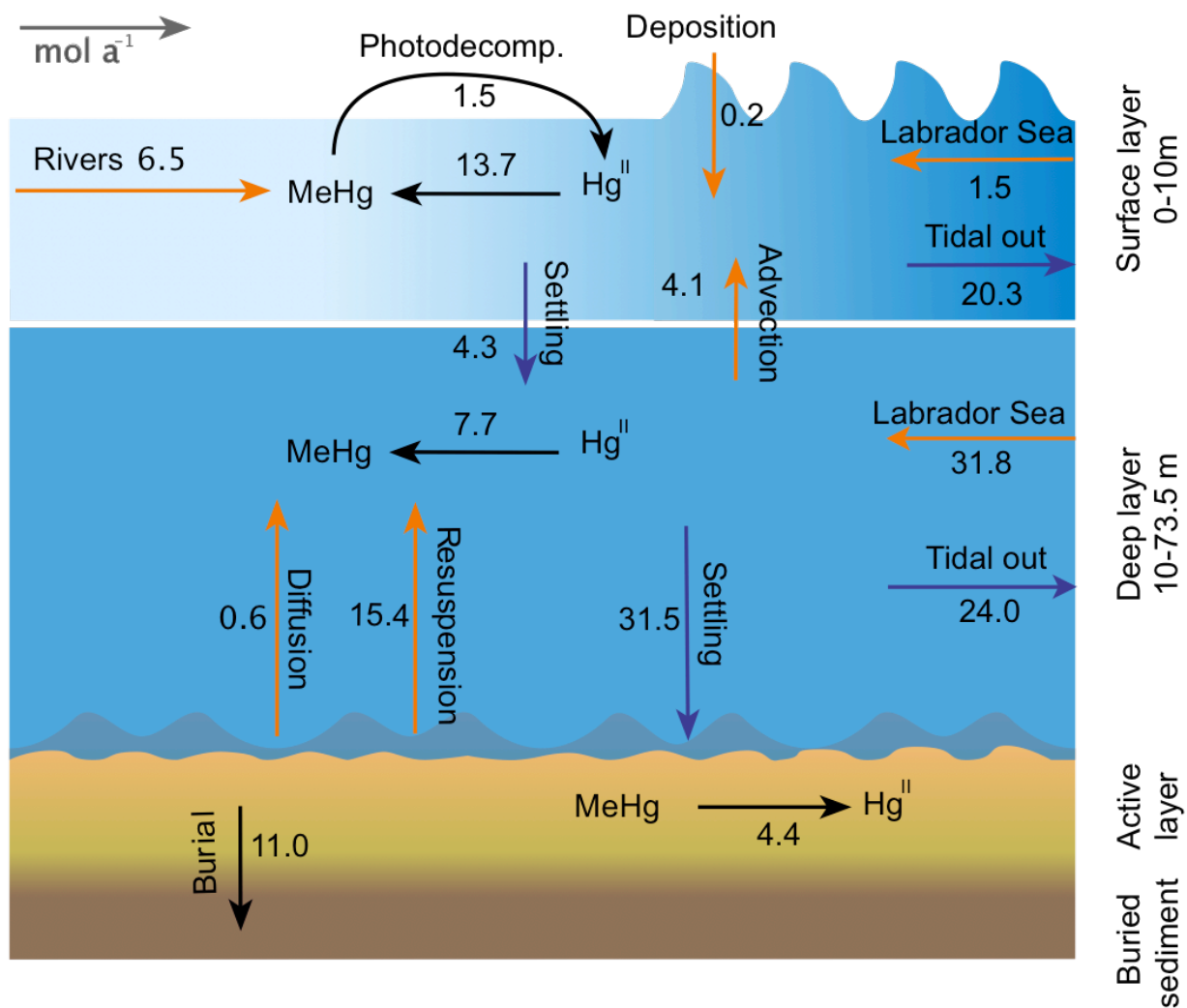


Figure S1. Schematic of model for mercury cycling the Lake Melville estuary Labrador adapted from Schartup et al. (3) for this analysis. Hydrodynamic data used to calculate mixing are from Lu et al. (25).

Table S4. Measured MeHg concentrations in the Churchill River between 2012-2015. Analytical procedures are described in Schartup et al. (3).

Season	Month - Year	Churchill River discharge (m ³ day ⁻¹)	MeHg (pg L ⁻¹)	<i>n</i>	Weighted mean (pg L ⁻¹)	Weighted SD (pg L ⁻¹)
Winter					26.53	1.66
	Dec	1.56E+08	27.49 ^a	—		
	Jan-15	1.56E+08	27.49	1		
	Feb-15	1.57E+08	24.62	1		
Spring					26.36	12.76
	Mar-15	1.47E+08	23.21	1		
	Apr-14	1.35E+08	11.83	1		
	May-14	2.31E+08	36.91	1		
Summer					4.99	1.15
	Jun-13/14	2.03E+08	5.91	2		
	Jul-14	1.45E+08	5.01	1		
	Aug-14	1.36E+08	3.61	1		
Fall					11.22	—
	Sep-12/14	1.32E+08	11.20	2		
	Oct	1.45E+08	11.20 ^a	—		
	Nov	1.53E+08	11.20 ^a	—		
Annual					17.94	11.46

^a No data were available for this month so MeHg concentrations are based on a month with similar water discharges.

Table S5. Community-based monitoring of fish species from the Lake Melville region between 2014-2015. Analytical methods for total Hg and MeHg analysis are provided in Li et al. (26)

Sample	Location	Date	<i>n</i>	Sampled By
Smelt	Churchill River	September 2014	7	Inuit residents of North West River and Rigolet
Brook Trout	Lake Melville		20	Inuit residents of North West River and Rigolet
Lake Trout	Churchill River	June-July 2014	13	Field Research Coordinator
Stickleback	Churchill River and Lake Melville	July-Sept 2014	30	Field Research Coordinator
Salmon	Lake Melville (Rigolet area)	July 2014	3	Rigolet fishers
Long Nose Sucker	Lake Melville (between NWR/Rigolet)	July-Aug 2014	20	Inuit fishers, North West River and Rigolet
Whitefish	Lake Melville (between NWR/Rigolet)	July-Aug 2014	20	Inuit fishers, North West River and Rigolet
Flatfish	Lake Melville (between NWR/Rigolet)	July-Aug 2014	20	Inuit fishers, North West River and Rigolet
Pike	Churchill River	July-Aug 2014 August 2015	13	Inuit fishers (HVGB)
Arctic Char	20 miles East of Rigolet	August 2015	10	Inuit fisher (Rigolet)
Atlantic Cod	St. Lewis Bay	September 2014	5	Labrador fisher
Mussels	Rigolet and NWR areas	June 2015	10	Inuit hunter
Misc. river fish	Churchill River above Muskrat Falls	August 2015	10	Inuit fishers

Table S6a. MeHg concentrations in aquatic species harvested from the Lake Melville region. Fish and bird concentrations are for fillets/muscle unless noted.

Species	MeHg ($\mu\text{g g}^{-1}$) Mean \pm SD	<i>n</i>	Data Source
Seal (<i>Phoca hispida</i>)			
<1 year (80%) ^a			
Muscle	0.11 \pm 0.09	34	This study
Liver	0.13 \pm 0.16	50	This study
Kidney	0.24 \pm 0.12	14	This study ^b
Seal 1-4 years (10%) ^a			
Muscle	0.21 \pm 0.17	18	This study, Brown et al. (27)
Liver	0.28 \pm 0.29	n/a	Mean of age classes < 1 year
Kidney	0.31 \pm 0.15	n/a	and > 4 years.
Seal > 4 years (10%) ^a			
Muscle	0.39 \pm 0.51	68	This study, Brown et al. (27)
Liver	0.43 \pm 0.37	3	This study
Kidney	0.38 \pm 0.17	3	This study ^b
Atlantic Salmon (<i>Salmo salar</i>)			
Fillet	0.07 \pm 0.02	12	Li et al. (26)
Roe	0.01 \pm 0.004	n/a	This study ^c
Liver	0.09 \pm 0.02	n/a	This study ^d
Atlantic cod (<i>Gadus morhua</i>)	0.19 \pm 0.06	5	Li et al. (26)
Arctic char (<i>Salvelinus alpinus</i>)			
Fillet	0.06 \pm 0.04	4	Li et al. (26)
Roe	0.01	n/a	This study ^c
Liver	0.08	n/a	This study ^d
Sculpin (<i>Myoxocephalus scorpius</i>)			
Fillet	0.23 \pm 0.09	10	Li et al. (26)
Liver	0.11 \pm 0.11	10	This study ^e
Brook trout (<i>Salvelinus fontinalis</i>)			
Fillet	0.10 \pm 0.03	48	Li et al. (26)
Liver	0.10 \pm 0.03	18	This study ^f
Roe	0.05 \pm 0.02	17	This study
Ouananiche (<i>Salmo salar m. sebago</i>)	0.15 \pm 0.11	18	Jacques Whitford Environment Ltd (28)
Lake trout (<i>Salvelinus namaycush</i>)	0.99 \pm 0.46	28	Jacques Whitford Environment Ltd (28)

^a Fraction of total seal harvest in each age class estimated by Inuit seal hunters in 2015.

^b Fraction of total Hg as methylmercury in kidney estimated as 26% from Northern Quebec ringed seals; moisture content estimated as 29% (29).

^c Estimated from salmon fillet:roe ratio (30).

^d Estimated from salmon fillet:liver ratio (30).

^e Estimated as 50% MeHg as a fraction of total Hg from literature values (31).

^f Estimated 62% MeHg as a fraction of total Hg based on salmon liver (30).

^d Estimated from salmon fillet:liver ratio (30).

^e Based on 44% MeHg as a fraction of total Hg as for molluscs (32).

^f Converted from dry weight using moisture content from gull samples.

Table S6b. MeHg concentrations in aquatic species harvested from the Lake Melville region. Fish and bird concentrations are for fillets/muscle unless noted.

Species	MeHg ($\mu\text{g g}^{-1}$) Mean \pm SD	<i>n</i>	Data Source
Flatfish (<i>Pleuronectoide sp.</i>)	0.07 \pm 0.04	20	Li et al. (26)
Capelin (<i>Mallotus villosus</i>)			
Fillet	0.02 \pm 0.002	6	Li et al. (26)
Roe	0.002 ^a		
Rainbow smelt (<i>Osmerus mordax</i>)	0.11 \pm 0.05	18	Li et al. (26)
Mussels (<i>Mytilus edulis</i>)	0.004 \pm 0.0005	6	Li et al. (26)
Porpoise (<i>Phocoena phocoena</i>)			
Muscle	0.60 \pm 0.06 ^b	20	Das et al. (33) (Atl. Norway)
Liver	1.22 \pm 0.87 ^{b,c}	21	Das et al. (33) (Atl. Norway)
Rock cod (<i>Gadus ogac</i>)			
Fillet	0.19 \pm 0.06		Assumed equal to cod
Liver	0.23 ^d		
Green sea urchin (<i>Strongylocentrotus droebachiensis</i>)	0.04	8	Noël et al. (34)
Periwinkle (<i>Littorina littorea</i>)	0.04	40	Noël et al. (34)
Clams (<i>Arctica islandica</i>)	0.01 \pm 0.01	15	US FDA (35)
Scallops (<i>Amusium laurenti</i>)	0.01 ^e	200	Karimi et al. (36)
Gull (<i>Rissa tridactyla</i>)			
Muscle	0.23 \pm 0.27	7	Lavoie et al. (37)
Eggs	0.06 \pm 0.01	20	Lavoie et al. (38)
Tern (<i>Sterna paradisaea</i>)			
Muscle	0.23 \pm 0.25 ^f	12	Clayden et al. (39)
Eggs	0.42 \pm 0.25 ^f	17	Clayden et al. (39)
Guillemot (<i>Cepphus grylle</i>)			
Muscle	0.27 \pm 0.07	3	Braune et al. (40) (Nfld.)
Eggs	0.21 \pm 0.01	20	Lavoie et al. (38)
Black duck (<i>Anas rubripes</i>)			
Muscle	0.11 \pm 0.08	12	Braune et al. (40) (Nfld. + Labrador)
Eggs	0.03 \pm 0.003		Schwarzbach and Adelsbach (41) – mallards, CA.
Eider (<i>Somateria mollissima</i>)			
Muscle	0.11 \pm 0.03	8	Braune et al. (40) (Nfld. + Labrador)
Loon (<i>Gavia immer</i>)			
Eggs	0.90 \pm 1.88	29	Evers et al. (42) (Maritimes)
Sandpiper (<i>Calidris pusilla</i>)			
Muscle	0.07 \pm 0.01	19	Burger et al. (2014)

^a Estimated from salmon fillet:roe ratio (30).

^b Converted from dry weight using moisture content from seal.

^c Based on 29% MeHg as a fraction of total Hg (43).

^d Estimated from salmon fillet:liver ratio (30).

^e Based on 44% MeHg as a fraction of total Hg as for molluscs (32).

^f Converted from dry weight using moisture content from gull samples.

Supplemental Information on Seal Mercury Analyses

MeHg concentrations in seal liver and muscle were measured at the Environment Canada laboratory in Burlington, Ontario. Samples were freeze dried and homogenized, then digested with 5N HNO₃ solution at 55 °C overnight. Digested samples were buffered with acetate and ethylated using sodium tetraethylborate (NaTEB). Ethylated MeHg was purged onto a Tenax packed column, separated by gas chromatography, and detected by cold vapor atomic fluorescence spectroscopy using a Brooks Rand MERX automated MeHg analyzer following established methods (44, 45). The average recovery for the DOLT 5 Certified Reference Material (CRM) included in each digestion cycle was 96.8±5.6% (SD; $n=8$). Precision, estimated by replicate analysis of duplicate samples was on average 6% ($n=6$).

Table S7a. Bioaccumulation factors (BAFs) between aquatic MeHg concentrations and measured concentrations in biota and the estimated fraction of lifespan for each species spent in the freshwater environment (River), Lake Melville (Estuary) and outer marine regions (Marine).

Species	log BAF	River	Estuary	Marine	References
Arctic char		0.5	0.5	0	Dunbar (46), Bradbury et al. (47) ^{a,b}
Muscle	6.6				
Liver	6.6				
Roe	5.6				
Atlantic cod	7.7	0	0–0.50	0–0.50	Li et al. (26) ^{c,d}
Atlantic salmon		0	0–0.50	0–0.50	Li et al. (26) ^{c,d}
Muscle	7.3				
Liver	7.4				
Roe	6.4				
Brook trout		0.5	0.5	0	Backus (48), Pilgrim et al. (49) ^{a,e}
Muscle	6.8				
Liver	6.7				
Roe	6.5				
Capelin		0	0.25	0.75	Li et al. (26) ^c
Muscle	6.0				
Roe	5.1				
Clams	5.8	0	1	0	Harvest location ^f
Black duck		0.5	0.5	0	Longcore et al. (50) ^g
Muscle	6.8				
Eggs	6.2				
Eider		0	0.5–1	0.5–1	BirdLife International (51) ^{d,g}
Muscle	6.9				
Flatfish	6.6	0	1	0	Armstrong and Starr (52) ^a
Green sea urchin	6.4	0	1	0	Harvest location ^f
Guillemot		0	0.5–1	0.5–1	Butler et al. (53) ^d
Muscle	7.4				
Eggs	7.2				
Gull		0	0.5–1	0.5–1	Baird et al. (54) ^g
Muscle	7.3				
Eggs	6.7				

^a Stable Hg isotopes suggest mixed habitat (26).

^b Time spent in open ocean is short (several weeks per year) (46, 47).

^c Habitat is predominantly offshore and fish migrate into the estuary to feed and/or spawn.

^d Habitats modeled probabilistically (see Table 2). Reported BAF is expected value.

^e Habitat is predominantly freshwater. Radiotelemetry monitoring in the Churchill River revealed short (90% < 10 km) seasonal displacements (55).

^f Sessile and low-motility species are based on predominant fishing location.

^g Increased MeHg following flooding is scaled by time spent in region (0.5) for migratory species.

Table S7b. Bioaccumulation factors (BAFs = MeHg biota/aqueous MeHg) and the estimated fraction of lifespan for each species spent in the freshwater environment (river), Lake Melville (estuary) and outer marine regions (marine).

Species	log BAF	River	Estuary	Marine	Reference
Lake trout	6.8	1	0	0	Black et al. (56)
Loon		0.5	0.5	0	McIntyre et al. (57) ^a
Eggs	7.7				
Mussels	5.3	0	1	0	Harvest location ^b
Ouananiche	6.9	1	0	0	Bradbury et al. (47)
Periwinkles	6.4	0	1	0	Harvest location ^b
Porpoise		0	0.25	0.75	Read and Westgate (58) ^c
Muscle	8.1				
Liver	8.4				
Rainbow smelt	6.8	0	1	0	FishBase (59) ^d
Rock cod		0	0–0.50	0–0.50	Ferguson et al. (60) ^{e,f}
Muscle	7.7				
Liver	7.5				
Sandpiper	6.6	0.5	0.5	0	Gratto-Trevor et al. (61) ^a
Scallops	6.1	0	1	0	Harvest location ^b
Sculpin		0	0.25	0.75	Li et al. (26) ^c
Muscle	7.7				
Liver	7.2				
Seal		0–0.25	0.5–0.75	0.25	Sikumiut Environmental Management Ltd. (62) ^{f,g}
Muscle	7.1				
Liver	7.1				
Kidney	7.3				
Tern		0	0.5–1	0.5–1	Hatch et al. (63) ^{a,f}
Muscle	7.3				
Eggs	7.5				

^a Increased MeHg following flooding is scaled by time spent in region (0.5) for migratory species.

^b Sessile and low-motility species are based on predominant fishing location.

^c Habitat is predominantly offshore and fish migrate into the estuary to feed and/or spawn. Habitat fraction is modeled probabilistically (see Table S2). Reported BAF is expected mean.

^d Hg isotope signature in adults indicates mixed habitat (26).

^e Same $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ stable isotope signature as Atlantic cod.

^f Habitat fraction modeled probabilistically (see Table S2). Reported BAF is expected mean.

^g Pups are found in sea ice in estuarine environment.

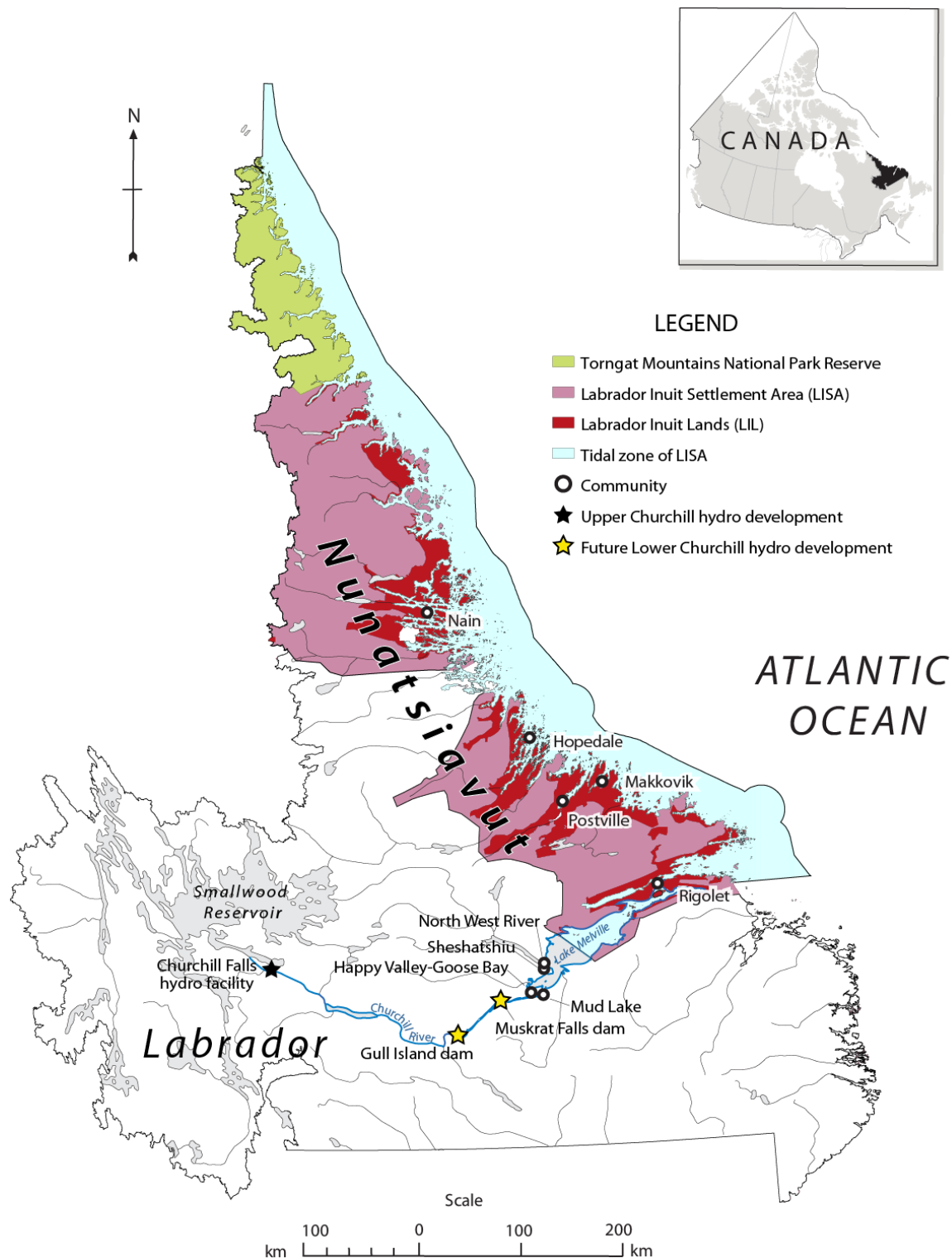


Figure S2. Map of the Labrador Inuit Settlement Area, existing and future hydroelectric developments on the Churchill River, and locations of indigenous communities. Source: Durkalec et al. (64). Reprinted with permission from Nunatsiavut Government.

Table S8. Hair mercury sampling from Inuit individuals in the communities downstream of the Muskrat Falls reservoir in June/July (spring/summer) and September/October (fall) 2014.

Demographic Group	Spring/ Summer (n)	Fall (n)	Total (n)	Unique Individuals (Percent Inuit Population ^a)
All individuals	157	499 ^b	656 ^b	571 ^b
Non-Inuit household members ^c	21	84	105	94
Inuit individuals	136	412	548	474 (19%)
<i><u>Communities</u></i>				
Happy Valley–Goose Bay ^d	96	265	361	325 (13%)
North West River	37	133	170	139 (37%)
Rigolet	24	101	125	107 (40%)
<i><u>Demographic Group</u></i> ^e				
Women of childbearing age (16-49) ^f	52	149	201	173
Children ≤ 12 years	15	29	44	40
Women of childbearing age (16-49 & children ≤ 12 in Rigolet	12	36	48	39
All male >12 years	56	174	230	200
All female >49 years	27	140	167	147

^a Hair was collected for some individuals during both sampling periods. Total Inuit population is based on the 2011 Census and National Household Survey (65, 66).

^b Including three individuals who did not report Inuit status

^c Hair samples were collected from non-Inuit individuals if they shared a residence with registered Inuit beneficiary identified by the Nunatsiavut Government.

^d Includes the nearby community of Mud Lake (n=22).

^e Combined data for all three communities.

^f As defined by the U.S. National Health and Nutrition Examination Survey (67).

Table S9. Food frequency questionnaire (FFQ) data collected from Inuit individuals from the communities downstream from the Muskrat Falls reservoir in March/April (winter), June/July (spring/summer) and September/October (fall) 2014. Dietary survey data collection overlapped with hair sampling (Table S8) in the spring and fall.

Demographic Group	Winter (<i>n</i>)	Spring/ Summer (<i>n</i>)	Fall (<i>n</i>)	Total (<i>n</i>)	Unique Individuals (Percent Inuit Population ^a)
All individuals	231	294	1054 ^b	1579 ^b	1145 ^b
Non-Inuit household ^c members	34	49	167	250	188
Inuit individuals	197	245	882	1324	952 (38%)
<i>Communities</i>					
Happy Valley-Goose Bay ^d	170	217	667	1054	745 (31%)
North West River	30	34	158	222	167 (43%)
Rigolet	31	43	229	303	233 (87%)
<i>Demographic Group^e</i>					
Women of childbearing age (16-49)	59	77	278	414	306
Children ≤12 years	55	59	166	280	179
Women of childbearing age (16-49 & children ≤ 12 in Rigolet	15	19	100	134	101
All male >12 years	74	108	387	569	406
All female > 49 years ^f	28	37	191	256	200

^a Data from some individuals are for multiple survey periods. Total Inuit population is based on the 2011 Census and National Household Survey (65, 66).

^b Total includes three individuals who did not report Inuit status.

^c Non-Inuit individuals who share a household with a registered Inuit beneficiary identified by the Nunatsiavut Government were included in the survey.

^d Includes the nearby community of Mud Lake (*n*=22).

^e Combined data for all three communities.

^f As defined by the U.S. National Health and Nutrition Examination Survey (67).

Table S10. MeHg concentrations in aquatic foods harvested outside the Lake Melville region. Commercial market categories rather than species names are listed for store-bought seafood.

Species	MeHg ($\mu\text{g g}^{-1}$) Mean \pm SD	<i>n</i>	Data Source
Minke whale (<i>Balaenoptera acutorostrata</i>) ^a	0.075 \pm 0.021	4	Riget et al. (68)
Polar bear (<i>Ursus maritimus</i>)	0.07 \pm 0.05	23	Woshner et al. (69)
Cod	0.11 \pm 0.07	115	US FDA (35)
Clams	0.01 \pm 0.002	15	US FDA (35)
Scallops	0.02 \pm 0.01 ^b	200	Karimi et al. (36)
Mussels	0.02 \pm 0.01 ^b	134	Karimi et al. (36)
Catfish	0.04 \pm 0.02 ^b	103	Karimi et al. (36)
Crab	0.06 \pm 0.03 ^b	151	Karimi et al. (36)
Haddock	0.06 \pm 0.03 ^b	78	Karimi et al. (36)
Herring	0.02 \pm 0.01 ^b	115	Karimi et al. (36)
Lobster	0.04 \pm 0.02 ^b	149	Karimi et al. (36)
Oysters (canned)	0.003 \pm 0.003 ^{b,c}	361	Karimi et al. (36)
Pollock (fish sticks)	0.02 \pm 0.01 ^b	131	Karimi et al. (36)
Brook trout	0.09 \pm 0.04 ^{b,d}	44	Karimi et al. (36)
Rainbow trout	0.03 \pm 0.02 ^b	71	Karimi et al. (36)
Sardines	0.03 \pm 0.02 ^b	246	Karimi et al. (36)
Shrimp	0.03 \pm 0.02 ^b	361	Karimi et al. (36)
Skate	0.12 \pm 0.05 ^b	13	Karimi et al. (36)
Sole	0.10 \pm 0.04 ^b	51	Karimi et al. (36)
Tilapia	0.02 \pm 0.01 ^b	114	Karimi et al. (36)
Fresh Tuna	0.44 \pm 0.25 ^d	295	US FDA (35)
Canned tuna	0.16 \pm 0.13 ^e	1002	US FDA (35)
Fresh salmon	0.04 \pm 0.02 ^b	504	Karimi et al. (36)
Canned salmon	0.04 \pm 0.04 ^f	61	Karimi et al. (36) ^e

^a Converted from dry weight using moisture content from seal muscle.

^b Standard deviation of distribution modeled following Carrington and Bolger (70).

^c Based on all market oysters.

^d Based on all unspecified freshwater.

^e Yellowfin, bigeye and albacore weighted according to relative landings reported by Sunderland (71).

^f Relative consumption of light and white canned tuna calculated from Sunderland (71).

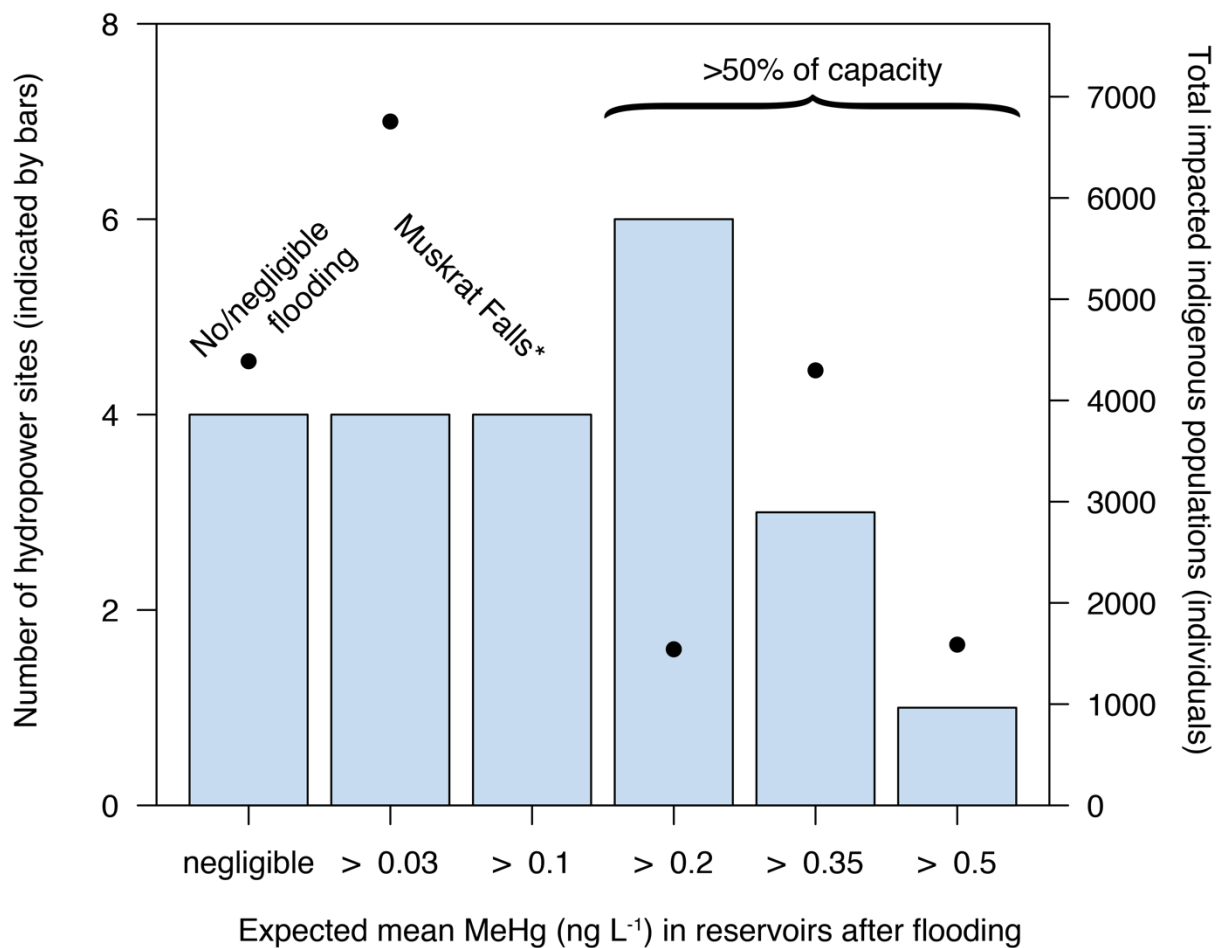


Figure S3. Number of planned hydroelectric power sites with forecasted reservoir MeHg concentrations above and below the Muskrat Falls reservoir and corresponding indigenous populations potentially impacted (circles). * Inuit population downstream from Muskrat Falls is included in the >0.35 bin because it is also potentially impacted by planned Gull Island facility.

Table S11a. Modeled MeHg concentrations in country foods after flooding of the Muskrat Falls reservoir.

Species	Post-flooding distribution of values			
	Expected mean	75 th percentile	90 th percentile	95 th percentile
Arctic char				
Muscle	0.41	0.51	0.78	1.0
Liver	0.49	0.58	0.70	0.80
Roe	0.05	0.06	0.07	0.08
Atlantic cod	0.41	0.50	0.65	0.76
Atlantic salmon				
Muscle	0.16	0.20	0.25	0.29
Liver	0.20	0.23	0.28	0.31
Roe	0.020	0.023	0.027	0.031
Black duck				
Muscle	0.44	0.55	0.83	1.1
Eggs	0.11	0.13	0.16	0.18
Brook trout				
Muscle	0.68	0.84	1.1	1.3
Liver	0.62	0.76	1.0	1.2
Roe	0.34	0.42	0.58	0.70
Capelin				
Muscle	0.04	0.05	0.06	0.07
Roe	0.01	0.01	0.01	0.01
Clams	0.03	0.03	0.04	0.04
Eider				
Muscle	0.20	0.24	0.30	0.34
Flatfish	0.17	0.22	0.32	0.40
Green sea urchin	0.10	0.12	0.14	0.16
Guillemot				
Muscle	0.68	0.82	1.0	1.2
Eggs	0.53	0.61	0.74	0.84
Gull				
Muscle	0.41	0.46	0.54	0.59
Eggs	0.15	0.18	0.21	0.24
Lake trout	1.0	1.3	1.8	2.2
Loon				
Eggs	5.6	5.7	13.3	20.9
Minke whale	0.07	0.09	0.10	0.11
Mussels	0.01	0.01	0.01	0.01
Ouananiche	1.5	1.9	3.0	3.9
Periwinkles	0.10	0.12	0.14	0.16

Table S11b. Modeled MeHg concentrations in country foods after flooding of the Muskrat Falls reservoir

Species	Post-flooding distribution of values			
	Expected mean	75 th percentile	90 th percentile	95 th percentile
Porpoise				
Muscle	1.4	1.8	2.7	3.5
Liver	2.8	3.6	5.2	6.8
Rock cod				
Muscle	0.42	0.50	0.65	0.77
Liver	0.50	0.58	0.70	0.79
Sandpiper	0.26	0.30	0.37	0.42
Scallops	0.06	0.07	0.08	0.09
Sculpin				
Muscle	0.54	0.66	0.88	1.0
Liver	0.20	0.24	0.42	0.58
Seal ^a				
Muscle	0.66	0.82	1.3	1.6
Liver	0.67	0.84	1.3	1.7
Kidney	1.0	1.2	1.6	1.9
Smelt	0.29	0.36	0.48	0.58
Tern	0.41	0.50	0.86	1.2

^a Weighted by age range (Table S6a).

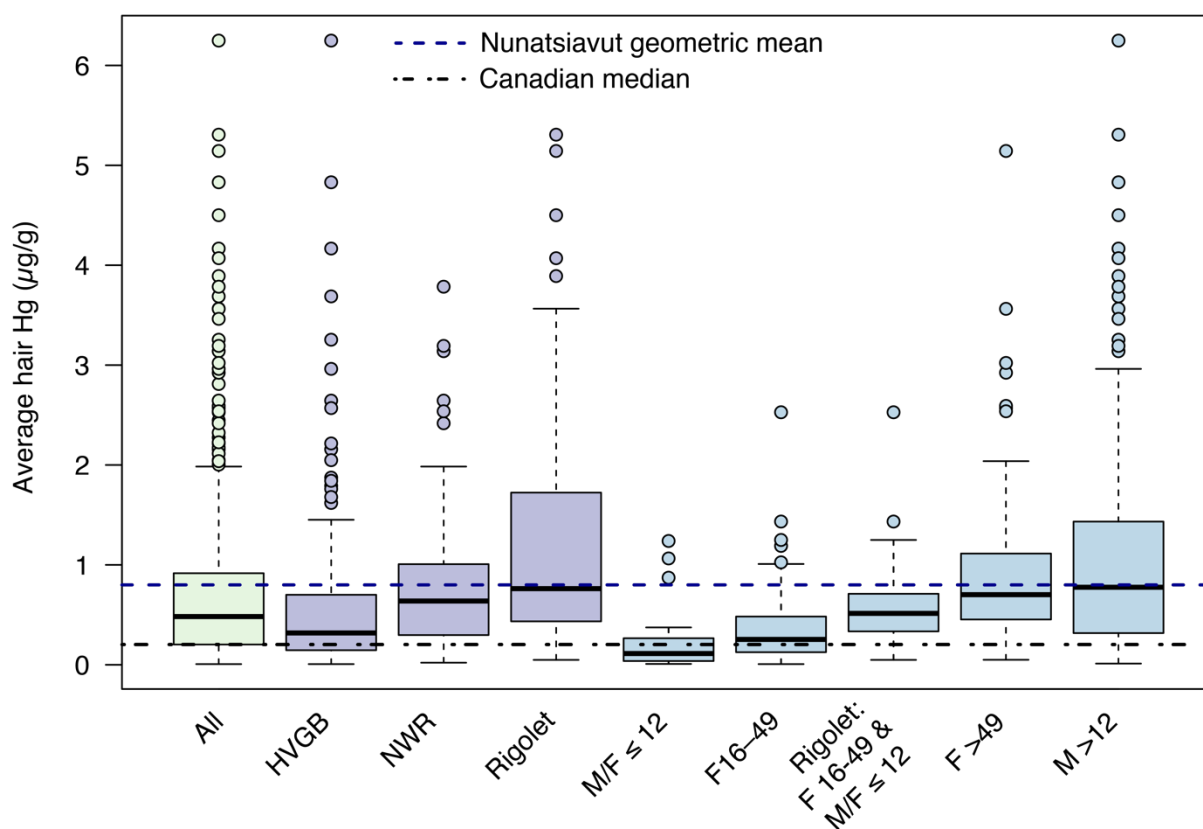


Figure S4. Measured concentrations of total Hg in hair samples from individuals in three Inuit communities downstream from the Muskrat Falls hydroelectric facility (HVGB = Happy Valley – Goose Bay; NWR = North West River) and among demographic groups (all communities together). Canadian median (6–79 years old) (72) and Nunatsiavut mean (73) are estimated using a mean blood-to-hair partition coefficient of 250 L g^{-1} (74). Most of the Hg in hair is present as MeHg (>90%) and potential demethylation in the hair follicle means that total Hg is the best indicator of internal MeHg exposure (75). At least one method blank and one certified hair reference materials (GBW-07601 and ERM-DB001) were tested every 10 samples and all recoveries were within certified ranges. Precision, calculated by replicate analysis of the duplicate hair samples (RSD) was better than 8.6%.

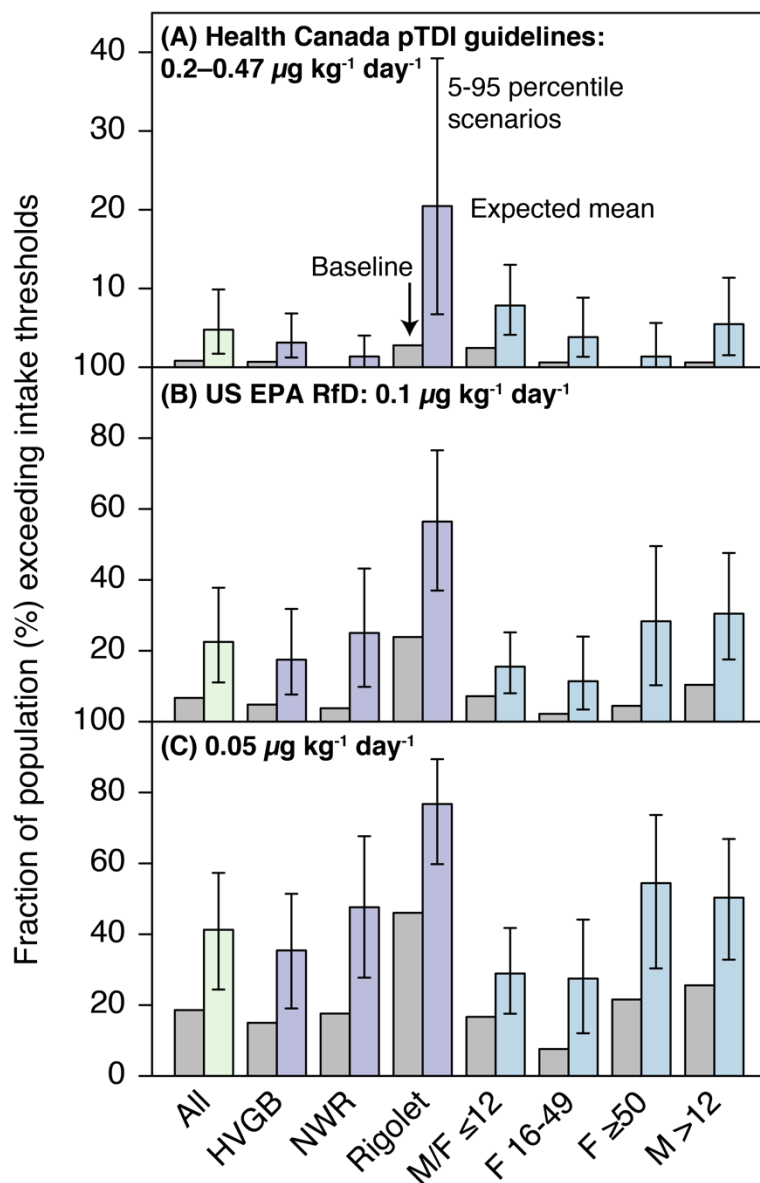


Figure S5. Fraction of population exceeding exposure thresholds in 2014 (measured) and post-flooding (modeled) by community (HVGB = Happy Valley – Goose Bay, NWR = North West River) and age/gender. Panel (A) shows the population that exceeds Health Canada provisional tolerable daily intake (pTDI) guidelines for MeHg of $0.20\ \mu\text{g kg}^{-1}\ \text{day}^{-1}$ for women of childbearing age and children 12 years and under and $0.47\ \mu\text{g kg}^{-1}\ \text{day}^{-1}$ for others (76). Panel (B) shows the population that exceeds the U.S. Environmental Protection Agency’s Reference Dose (RfD) (77), and panel (C) indicates the proportion of the population exceeding the RfD calculated based on more recent epidemiological research on neurotoxicity (78, 79).

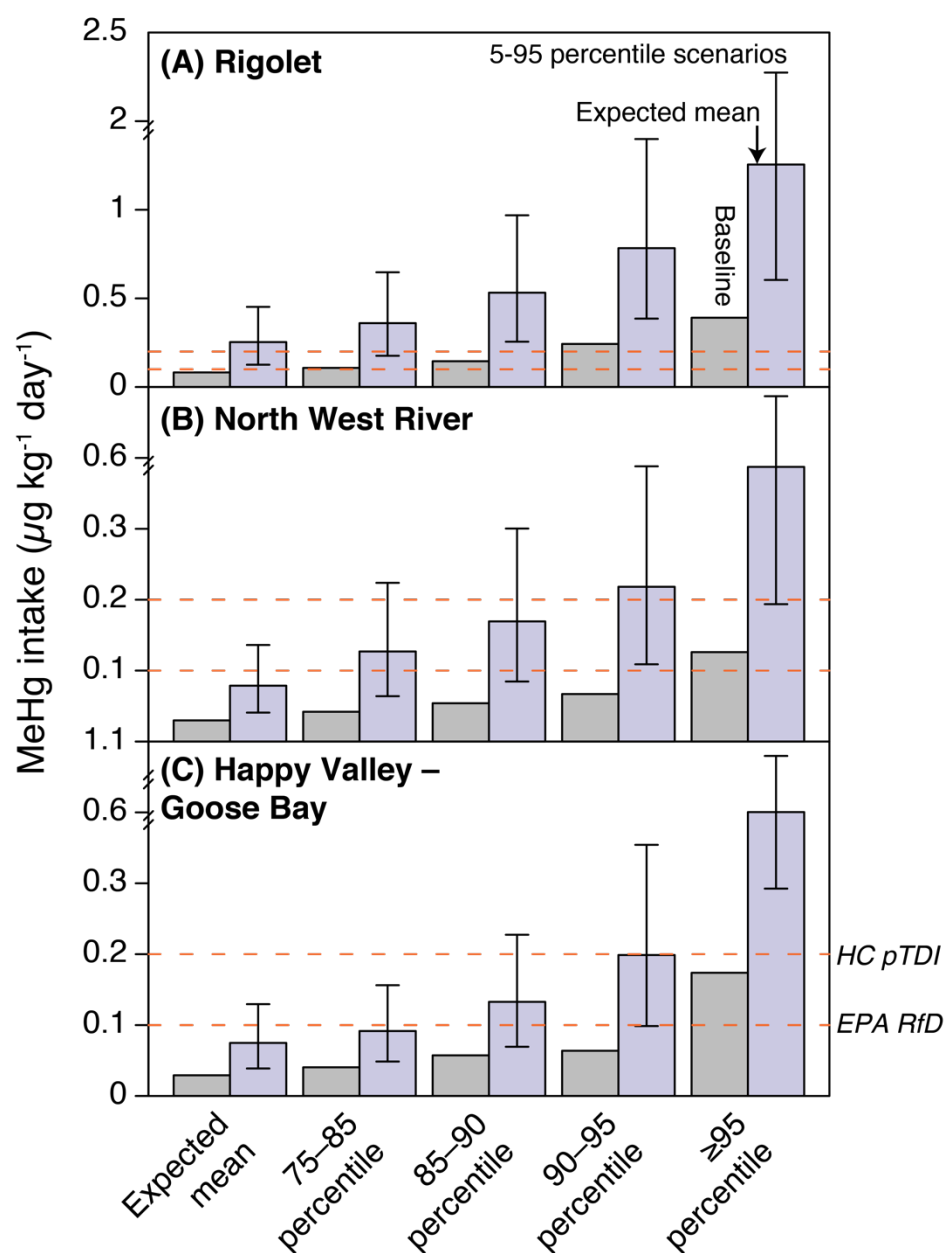


Figure S6. Baseline (measured) and post-flooding (modeled) MeHg intake relative to the Health Canada (HC) provisional tolerable daily intake (pTDI) and the U.S. EPA reference dose (RfD) for the communities of (A) Rigolet, the largest per-capita consumer of country foods, (B) North West River and (C) Happy Valley – Goose Bay

References

1. Steinberger, N.; Hondzo, M. Diffusional mass transfer at sediment-water interface. *J Environ Eng.* **1999**, *125* (2), 192-200.
2. Sunderland, E. M.; Dalziel, J.; Heyes, A.; Branfireun, B. A.; Krabbenhoft, D. P.; Gobas, F. A. Response of a macrotidal estuary to changes in anthropogenic mercury loading between 1850 and 2000. *Environ Sci Technol.* **2010**, *44* (5), 1698-1704.
3. Schartup, A. T.; Balcom, P. H.; Soerensen, A. L.; Gosnell, K. J.; Calder, R. S. D.; Mason, R. P.; Sunderland, E. M. Freshwater discharges drive high levels of methylmercury in Arctic marine biota. *P Natl Acad Sci USA*, **2015**, *112* (38), 11789-11794.
4. Peterson, E. L. Benthic shear stress and sediment condition. *Aquacult Eng.* **1999**, *21* (2), 85-111.
5. Wilcock, P. R. Estimating Local Bed Shear Stress from Velocity Observations. *Water Resour Res.* **1996**, *32* (11), 3361-3366.
6. AMEC. Lower Churchill Hydroelectric Development Freshwater Fish Habitat Compensation Plan: Muskrat Falls. **2013**.
7. Soil Landscapes of Canada Working Group (SLCWG). Soil Landscapes of Canada v. 3.2. Agriculture and Agri-Food Canada. **2011**.
8. Chan, T. P.; Govindaraju, R. S. Estimating Soil Water Retention Curve from Particle-Size Distribution Data Based on Polydisperse Sphere Systems. *Vadose Zone J.* 2004, *3* (4), 1443-1454.
9. Jonsson, S.; Skjellberg, U.; Nilsson, M. B.; Lundberg, E.; Andersson, A.; Bjorn, E. Differentiated availability of geochemical mercury pools controls methylmercury levels in estuarine sediment and biota. *Nat Commun.* **2014**, *5*, 4624.
10. Natural Resources Canada (NRCan). *Data – Canada Lands Surveys*. <http://www.nrcan.gc.ca/earth-sciences/geomatics/canada-lands-surveys/11092>. Accessed July 2, 2016.
11. Metis Settlements General Council. *Peavine Metis Settlement*. <http://www.msgc.ca/communities/peavine-metis-settlement>. Accessed July 3, 2016.
12. Aboriginal Affairs and Northern Development Canada (AANDC). *First Nation Profiles*.
13. Midgard Consulting Inc. Yukon Next Generation Hydro and Transmission Viability Study: Site Screening Inventory. **2015**.
14. Government of the Northwest Territories. *Northwest Territories Energy Report*. **2011**.
15. BC Hydro. Preliminary Construction Schedule. **2015**.
16. Klohn Crippen Berger; SNC-Lavalin. *Hatch Peace River Site C Clean Energy Project*. **2011**.
17. AHP Development Corporation. *Project Description: Amisk Hydroelectric Project: Executive Summary*. **2015**.
18. Canadian Environmental Assessment Agency. *Tazi Twé Hydroelectric Project Draft Environmental Assessment Report*. **2015**.
19. Keeyask Hydropower LP. *Keeyask Generation Project Year in Review 2014-2015*. **2015**.
20. Manitoba Hydro-Electric Board; Tataskweyak Cree Nation; War Lake First Nation; York Factory First Nation; Fox Lake Cree Nation. *Keeyask Project: Project Description*. **2009**.
21. Manitoba Hydro. *Conawapa Generating Station*. <https://www.hydro.mb.ca/projects/conawapa/index.shtml>. Accessed March 28, 2016.
22. Northern Ontario Business Staff. *Construction begins on \$300-million hydro project*. Northern Ontario Business, 2015.
23. Ministère du Développement durable, de l'Environnement et des Parcs. *Rapport d'analyse environnementale pour le projet d'aménagement du complexe hydroélectrique de la rivière Romaine sur le territoire de la municipalité régionale de comté de Minganie par Hydro-Québec*. **2009**.

24. Ernst & Young LLP. *Muskrat Falls Project: Review of Project Cost, Schedule and Related Risk Interim Report*. **2016**.
25. Lu, Z.; DeYoung, B.; Banton, S. *Analysis of physical oceanographic data from Lake Melville, Labrador, September 2012 - July 2013*; Memorial University of Newfoundland: St. John's, Newfoundland. **2014**.
26. Li, M.; Schartup, A. T.; Valberg, A. P.; Ewald, J. D.; Krabbenhoft, D. P.; Yin, R.; Balcom, P. H.; Sunderland, E. M. Environmental Origins of Methylmercury Accumulated in Subarctic Estuarine Fish Indicated by Mercury Stable Isotopes. *Environ Sci Technol*. **2016**. In press. DOI: 10.1021/acs.est.6b03206
27. Brown, T. M.; Fisk, A. T.; Wang, X.; Ferguson, S. H.; Young, B. G.; Reimer, K. J.; Muir, D. C. Mercury and cadmium in ringed seals in the Canadian Arctic: Influence of location and diet. *Sci Total Environ*. **2016**, 545-546, 503-511.
28. Jacques Whitford Environment Ltd. *Statistical Analysis of Mercury Data from Churchill Falls (Labrador) Corporation Reservoirs*. **2006**.
29. Lemire, M.; Kwan, M.; Laouan-Sidi, A. E.; Muckle, G.; Pirkle, C.; Ayotte, P.; Dewailly, E. Local country food sources of methylmercury, selenium and omega-3 fatty acids in Nunavik, Northern Quebec. *Sci Total Environ*. **2015**, 509-510, 248-259.
30. Zhang, X.; Naidu, A. S.; Kelley, J. J.; Jewett, S. C.; Dasher, D.; Duffy, L. K. Baseline concentrations of total mercury and methylmercury in salmon returning via the Bering Sea (1999-2000). *Mar Poll Bull*. **2001**, 42 (10), 993-997.
31. Harley, J.; Lieske, C.; Bhojwani, S.; Castellini, J. M.; López, J. A.; O'Hara, T. M. Mercury and methylmercury distribution in tissues of sculpins from the Bering Sea. *Polar Biol*. **2015**, 38 (9), 1535-1543.
32. Claisse, D.; Cossa, D.; Bretaudeau-Sanjuan, J.; Touchard, G.; Bombled, B. Methylmercury in Molluscs Along the French Coast. *Mar Poll Bull*. **2001**, 42 (4), 329-332.
33. Das, K.; Siebert, U.; Fontaine, M.; Jauniaux, T.; Holsbeek, L.; Bouqueneau, J. M. Ecological and pathological factors related to trace metal concentrations in harbour porpoises *Phocoena phocoena* from the North Sea and adjacent areas. *Mar Ecol Prog Ser*. **2004**, 281, 283-295.
34. Noël, L.; Testu, C.; Chafey, C.; Velge, P.; Guérin, T. Contamination levels for lead, cadmium and mercury in marine gastropods, echinoderms and tunicates. *Food Control*. **2011**, 22 (3-4), 433-437.
35. United States Food and Drug Administration (US FDA), Mercury Levels in Commercial Fish and Shellfish (1990-2010). **2014**.
36. Karimi, R.; Fitzgerald, T. P.; Fisher, N. S. A quantitative synthesis of mercury in commercial seafood and implications for exposure in the United States. *Environ Health Perspect*. **2012**, 120 (11), 1512-1519.
37. Lavoie, R. A.; Hebert, C. E.; Rail, J. F.; Braune, B. M.; Yumvihoze, E.; Hill, L. G.; Lean, D. R. Trophic structure and mercury distribution in a Gulf of St. Lawrence (Canada) food web using stable isotope analysis. *Sci Total Environ*. **2010**, 408 (22), 5529-5539.
38. Lavoie, R. A.; Champoux, L.; Rail, J. F.; Lean, D. R. Organochlorines, brominated flame retardants and mercury levels in six seabird species from the Gulf of St. Lawrence (Canada): relationships with feeding ecology, migration and molt. *Environ Pollut* **2010**, 158 (6), 2189-2199.
39. Clayden, M. G.; Arsenault, L. M.; Kidd, K. A.; O'Driscoll, N. J.; Mallory, M. L. Mercury bioaccumulation and biomagnification in a small Arctic polynya ecosystem. *Sci Total Environ* **2015**, 509-510, 206-215.

40. Braune, B. M.; Malone, B.; Burgess, N. M.; Elliott, J.; Garrity, N.; Hawkings, J.; Hines, J.; Marshall, H.; Marshall, W.; Rodrigue, J.; Wakeford, B.; Wayland, M.; Weseloh, D.; Whitehead, P. *Chemical Residues in Waterfowl and Gamebirds Harvested in Canada, 1987-95*. **1999**.
41. Schwarzbach, S.; Adelsbach, T. *Assessment of Ecological and Human Health Impacts of Mercury in the Bay-Delta Watershed*. **2003**.
42. Evers, D. C.; Burgess, N. M.; Champoux, L.; Hoskins, B.; Major, A.; Goodale, W. M.; Taylor, R. J.; Poppenga, R.; Daigle, T. Patterns and Interpretation of Mercury Exposure in Freshwater Avian Communities in Northeastern North America. *Ecotoxicology*. **2005**, *14* (1-2), 193-221.
43. Joiris, C. R.; Holsbeek, L.; Bolba, D.; Gascard, C.; Stanev, T.; Komakhidze, A.; Baumgärtner, W.; Birkun, A. Total and organic mercury in the Black Sea harbour porpoise *Phocoena phocoena relicta*. *Marine Poll Bull.* **2001**, *42* (10), 905-911.
44. Lehnher, I.; St. Louis, V. L.; Emmerton, C. A.; Barker, J. D.; Kirk, J. L., Methylmercury cycling in High Arctic wetland ponds: Sources and sinks. *Environ Sci Technol.* **2012**, *46* (19), 10514-10522.
45. Hintelmann, H.; Nguyen, H. T., Extraction of methylmercury from tissue and plant samples by acid leaching. *Anal Bioanal Chem.* **2005**, *381* (2), 360-365.
46. Dunbar, M. J., The Sea Waters surrounding the Québec-Labrador peninsula. *Cahiers de géographie du Québec*. **1965**, *10* (19), 13.
47. Bradbury, C.; Roberge, M.; Minns, C. *Life History Characteristics of Freshwater Fishes Occurring in Newfoundland and Labrador, with Major Emphasis on Lake Habitat Requirements*. Fisheries and Oceans Canada. **1999**.
48. Backus, R. The Fishes of Labrador. *Bulletin of the American Museum of Natural History*. **1957**, *113* (4).
49. Pilgrim, B. L.; Perry, R. C.; Keefe, D. G.; Perry, E. A.; Dawn Marshall, H., Microsatellite variation and genetic structure of brook trout (*Salvelinus fontinalis*) populations in Labrador and neighboring Atlantic Canada: evidence for ongoing gene flow and dual routes of post-Wisconsinan colonization. *Ecol Evol.* **2012**, *2* (5), 885-898.
50. Longcore, J. R.; McAuley, D. G.; Hepp, G. R.; Rhymer, J. M.; Poole, A.; Gill, F. American Black Duck (*Anas rubripes*). *The Birds of North America Online*. **2000**.
51. BirdLife International *Somateria mollissima*. <http://www.iucnredlist.org/details/22680405/0>. Accessed July 19, 2016.
52. Armstrong, M. P.; Starr, B. A. Reproductive Biology of the Smooth Flounder in Great Bay Estuary, New Hampshire. *T Am Fish Soc.* **1994**, *123* (1), 112-114.
53. Butler, R. G.; Buckley, D. E.; Poole, A.; Gill, F. Black Guillemot (*Cephus grylle*). *The Birds of North America Online*. **2002**.
54. Baird, P. H.; Poole, A.; Gill, F. Black-legged Kittiwake (*Rissa tridactyla*). *The Birds of North America Online*. **1994**.
55. Nalcor Energy. *Biophysical Assessment*. **2009**.
56. Black, G. A.; Dempson, J. B.; Bruce, W. J. Distribution and postglacial dispersal of freshwater fishes of Labrador. *Can J Zool.* **1986**, *64* (1), 21-31.
57. McIntyre, J. W.; Barr, J. F.; Poole, A.; Gill, F. Common Loon (*Gavia immer*). *The Birds of North America Online*. **1997**.
58. Read, A. J.; Westgate, A. J., Monitoring the movements of harbour porpoises (*Phocoena phocoena*) with satellite telemetry. *Mar Biol.* **1997**, *130* (2), 315-322.
59. FishBase. *Osmerus mordax* (Mitchill, 1814). <http://www.fishbase.org/summary/253>. Accessed July 19, 2016.

60. Ferguson, S. H.; Loseto, L. L.; Mallory, M. L. *A Little Less Arctic: Top Predators in the World's Largest Northern Inland Sea, Hudson Bay*. Springer Netherlands. **2010**.
61. Gratto-Trevor, C. L.; Poole, A.; Gill, F. Semipalmated Sandpiper (*Calidris pusilla*). *The Birds of North America Online*. **1992**.
62. Sikumiut Environmental Management Ltd. *Seal Abundance and Distribution*. **2007**.
63. Hatch, J. J.; Poole, A.; Gill, F., Arctic Tern (*Sterna paradisaea*). *The Birds of North America Online*. **2002**.
64. Durkalec, A.; Sheldon, T.; Bell, T. E., Eds. Scientific Report. **2016**.
65. Statistics Canada, 2011 National Household Survey. **2013**.
66. Statistics Canada, 2011 Census. **2012**.
67. McDowell, M. A.; Dillon, C. F.; Osterloh, J.; Bolger, P. M.; Pellizzari, E.; Fernando, R.; Montes de Oca, R.; Schober, S. E.; Sinks, T.; Jones, R. L.; Mahaffey, K. R. Hair Mercury Levels in U.S. Children and Women of Childbearing Age: Reference Range Data from NHANES 1999-2000. *Environ Health Perspect.* **2004**, *112* (11), 1165-1171.
68. Riget, F.; Moller, P.; Dietz, R.; Nielsen, T. G.; Asmund, G.; Strand, J.; Larsen, M. M.; Hobson, K. A. Transfer of mercury in the marine food web of West Greenland. *J Environ Monit.* **2007**, *9* (8), 877-883.
69. Woshner, V. M.; O'Hara, T. M.; Bratton, G. R.; Beasley, V. R. Concentrations and interactions of selected essential and non-essential elements in ringed seals and polar bears of arctic Alaska. *J Wildl Dis* **2001**, *37* (4), 711-721.
70. Carrington, C. D.; Bolger, M. P. An exposure assessment for methylmercury from seafood for consumers in the United States. *Risk Anal.* **2002**, *22* (4), 689-699.
71. Sunderland, E. M., Mercury exposure from domestic and imported estuarine and marine fish in the U.S. seafood market. *Environ Health Perspect.* **2007**, *115* (2), 235-42.
72. Lye, E.; Legrand, M.; Clarke, J.; Probert, A. Blood total mercury concentrations in the Canadian population: Canadian health measures survey cycle 1, 2007-2009. *Can J Public Health.* **2013**, *104* (3), E246-E251.
73. Chan, H. M. L. Contaminant Assessment in Nunatsiavut. **2011**.
74. World Health Organization (WHO) Methylmercury. **1990**.
75. Berglund, M.; Lind, B.; Bjornberg, K. A.; Palm, B.; Einarsson, O.; Vahter, M., Inter-individual variations of human mercury exposure biomarkers: a cross-sectional assessment. *Environ Health.* **2005**, *4*, 20.
76. Health Canada Updating the Existing Risk Management Strategy for Mercury in Retail Fish; **2007**.
77. United States Environmental Protection Agency (US EPA), Reference Dose for Methylmercury. In Federal Register, 2000; Vol. 65, pp 64702-64703.
78. Bellanger, M.; Pichery, C.; Aerts, D.; Berglund, M.; Castano, A.; Cejchanova, M.; Crettaz, P.; Davidson, F.; Esteban, M.; Fischer, M. E.; Gurzau, A. E.; Halzlova, K.; Katsonouri, A.; Knudsen, L. E.; Kolossa-Gehring, M.; Koppen, G.; Ligocka, D.; Miklavcic, A.; Reis, M. F.; Rudnai, P.; Tratnik, J. S.; Weihe, P.; Budtz-Jorgensen, E.; Grandjean, P.; Demo/Cophes, Economic benefits of methylmercury exposure control in Europe: monetary value of neurotoxicity prevention. *Environ Health* **2013**, *12*, 3.
79. Grandjean, P.; Budtz-Jorgensen, E., Total imprecision of exposure biomarkers: implications for calculating exposure limits. *Am J Ind Med.* **2007**, *50* (10), 712-719.