

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

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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. 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

September 1, 2016 Received: Revised: October 21, 2016 Accepted: October 21, 2016 Published: November 9, 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. 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 ($K_{\rm d}$, 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. (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 \times 10^8 m^3 $d^{-1})$ 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 stables 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 = 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 $\delta^{\hat{1}3}$ C, δ^{15} N, Δ^{199} Hg, and δ^{202} 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 twocentimeter 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.4 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

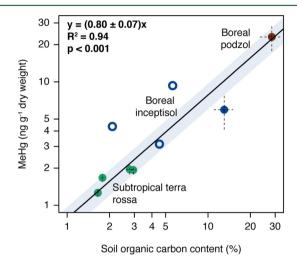


Figure 1. Relationship between soil organic carbon content and MeHg concentrations (ng g⁻¹ dry weight) of flooded soils. Each data point represents an individual sampling location. Hatched lines indicate standard errors around the mean. Soil cores are from the Wujiangu reservoir, China (subtropical terra rossa),28 the Experimental Lakes Area (ELA, boreal inceptisol) 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 $(2-830 \text{ ng m}^{-2} \text{ day}^{-1}).4$

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,

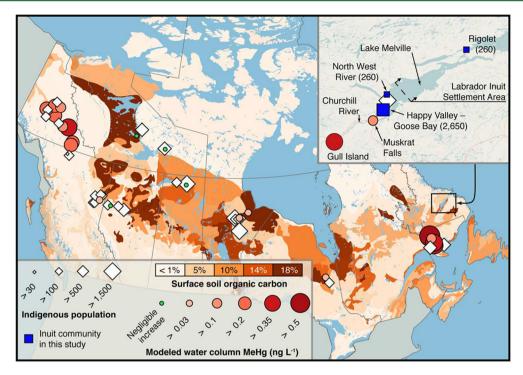


Figure 2. Planned locations for hydroelectric power expansion in Canada and indigenous populations with reserves or communities within 100 km of development regions (SI Table S3). Inset map shows the Muskrat Falls facility in Labrador and the three Inuit communities studied in this work. Reservoir MeHg 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 ng L⁻¹) have relatively large flooded areas (85–144 km²). 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 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 (90–300 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

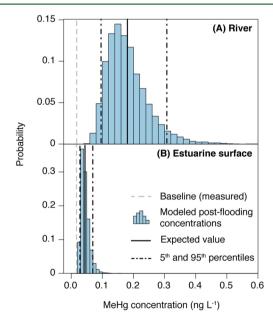


Figure 3. Probabilistically modeled scenarios for MeHg increases in downstream river and estuary of the Muskrat Falls hydroelectric facility. Photochemical MeHg demethylation is assumed to occur continuously down the reach of the Churchill River into Lake Melville thus the river concentration reflects 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 $\rm L^{-1}$ to a peak level of 44 pg $\rm L^{-1}$ (Figure 3). The fifth percentile scenario suggests a lower bound increase of 1.6-fold (28 pg $\rm L^{-1}$) and the 95th percentile scenario represents a 4-fold

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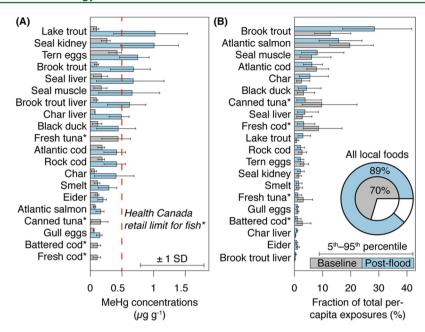


Figure 4. Top 20 MeHg exposure sources for Inuit downstream of the Muskrat Falls hydroelectric reservoir before (measured in 2014) and after flooding (modeled peak concentration) planned for 2016–2017 (SI Table S1). Commercial species unaffected by local conditions are denoted by "*". Panel (A) shows MeHg concentrations in country foods relative to Health Canada retail limits for fish other than tuna $(0.5 \,\mu g \, g^{-1})$. 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 μ g MeHg g⁻¹ Canadian retail limit ⁵⁰ 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. 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 μ g kg⁻¹ body weight day⁻¹), and 3% are above Health Canada's (HC) provisional tolerable daily intake (pTDI, 0.20- $0.47 \mu g kg^{-1}$ 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 μ g kg⁻¹ day⁻¹ for individuals at the 90th percentile of postflooding exposures and this demographic accounts for nearly 60% of the total additional MeHg intake (μ 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

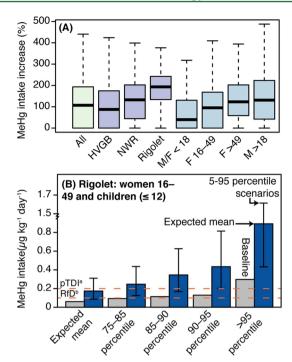


Figure 5. (a) Health Canada (HC) provisional tolerable daily intake (pTDI); (b) U.S. EPA reference dose (RfD) for MeHg. Modeled changes in Inuit MeHg exposures following flooding of the Muskrat Falls reservoir. Panel (A) shows exposure increases relative to measured baseline intake in 2014. Error bars 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 age $^{16-49}$ 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 μ g kg⁻¹ day⁻¹ (0.58 μ g Hg g⁻¹ 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". Recent data from prospective birth cohorts support this conclusion. For adults, the Health Canada pTDI is the least conservative across international regulatory agencies (0.47 μ g kg $^{-1}$ 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. Past studies suggest reducing or avoiding consumption of country foods may also result in substantial adverse impacts on individual health. 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. 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)

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Notes

The authors declare no competing financial interest.

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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), 5165-5234.
- (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,
- (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.; Lehnherr, 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), 560-575.
- (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.
- (36) Verdon, R.; Brouard, D.; Demers, C.; Lalumiere, 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/chemchim/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.; Drevnick, 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.