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re: <u>Updated analysis of predicted increases in methylmercury concentrations and downstream export</u> <u>from Muskrat Falls Reservoir</u>

1 Introduction

In 2017-2018, Nalcor retained Reed Harris Environmental Ltd. and others to update or extend previous studies to predict increases in methylmercury concentrations in water and biota in Muskrat Falls Reservoir and downstream in Goose Bay and Lake Melville. An important component of the analysis was the use of field data collected since 2011, both within the reservoir area and downstream. This technical memorandum describes:

- 1. updated predictions of increases in methylmercury concentrations in Muskrat Falls Reservoir waters and fish; and
- 2. predicted methylmercury concentrations and masses exported from Muskrat Falls Reservoir.

Results of this study were used by Baird & Associates (Brunton, 2018) in a model analysis to predict increases in methylmercury concentrations in Goose Bay and Lake Melville waters. The Baird results were then used by Wood (2018) to estimate increases in methylmercury concentrations in biota in Goose Bay and Lake Melville. Finally, predicted increases in methylmercury levels in biota were used by Azimuth (2018) to develop an interim update to the human health risk assessment by Dillon (2016).

2 Methods

The primary goal of this study was to estimate increases in methylmercury concentrations and loads delivered downstream following the creation of Muskrat Falls Reservoir. Two approaches were used (Figure 1). The first approach was based on field data from an experimental reservoir study in Ontario called FLUDEX. This experiment investigated mercury and greenhouse gases intensively in newly flooded uplands and advanced our understanding of mercury in reservoirs (*e.g.* Bodaly *et al.*, 2004, Hall *et al.*, 2005). The second approach was based on a mechanistic model that predicted methylmercury concentrations in water, sediments and biota in the reservoir over time, as well as downstream export rates. The remainder of this section of the document provides additional information on each approach.

While downstream methylmercury export was the focus of the analysis, fish mercury concentrations within Muskrat Falls Reservoir were also predicted and are presented here. Predicted fish mercury concentrations in the reservoir were important in their own right and provided a line of evidence to help assess confidence in concentrations of methylmercury in water predicted by the mechanistic model.



This is because there are long-term data available for fish mercury concentrations from existing reservoirs that can be compared to model predictions, but no analogous data exist for methylmercury concentrations in water over time from full scale reservoirs, to compare directly with models.



Figure 1. Approach used to estimate downstream export of methylmercury from Muskrat Falls Reservoir

2.1 Approaches to predict increases in fish mercury concentrations in Muskrat Falls Reservoir

Mechanistic and regression models were used to predict increases in fish mercury concentrations in Muskrat Falls Reservoir. Both approaches were previously applied to Muskrat Falls Reservoir in support of the Environmental Assessment (Nalcor, 2009a; Harris *et al.*, 2010).

Mechanistic model description

ResMerc is a process-based simulation model for reservoirs and lakes, originally developed as part of FLUDEX and a companion flooded wetland experiment called ELARP (Harris *et al.*, 2009, Harris and Hutchinson, 2009). In addition to being applied previously to Muskrat Falls Reservoir to support the Environmental Assessment, ResMerc was used for the Site C project in British Columbia (Harris *et al.*, 2012). Model compartments include the water column, sediments, and a simplified food web that consists of several trophic levels (phytoplankton, zooplankton, benthos and up to four fish species) (Figure 2). Fish mercury concentrations tend to increase with age and are therefore followed in each year class (up to 20 cohorts). The model predicts concentrations, mercury pools and major fluxes for each mercury form through time.



ResMerc mercury processes include atmospheric deposition, inflows and outflows (surface and groundwater), adsorption/desorption, particulate settling, particle decomposition at the sediment/water interface and within sediments, resuspension, burial, air/water gaseous exchange, industrial point sources, in-situ transformations (*e.g.* methylation, demethylation, methylmercury photodegradation, Hg(II) reduction and Hg(0) oxidation), methylmercury uptake kinetics in plankton and partitioning in benthos, and methylmercury bioaccumulation in fish.



Figure 2. Representation of mercury cycling and bioaccumulation in ResMerc

Methylmercury concentrations in fish are predicted using a bioenergetics approach described by Harris and Bodaly (1998). Methylmercury fluxes are expanded from individual fish to entire fish populations by computing the fluxes for individual fish and then multiplying by the number of fish in each age class.

While many factors affect fish mercury concentrations in natural lakes, one process takes on special importance in new reservoirs: decomposition. Flooding stimulates decomposition and more activity by microbes that convert inorganic mercury into methylmercury. Sediments are divided into a maximum of 5 zones in the model, based on terrain type and elevations set by the user. These zones can include littoral and profundal zones in the original lake, flooded uplands and flooded wetlands. Each sediment zone has two vertical sediments layers with thicknesses defined by the user. Sediments below the 2nd layer are treated as a boundary condition. Each sediment layer has its own initial conditions, characteristics and inputs. Additional information on ResMerc is available in the model user guide (Harris and Hutchinson 2009) and a report describing the model development (Harris *et al.*, 2009).



The steps involved in the application of ResMerc to Muskrat Falls Reservoir were as follows:

- The model was calibrated to Robert Bourassa Reservoir in Quebec to estimate the methylmercury loads required from flood zones to support observed increases in fish mercury concentrations. Robert Bourassa reservoir had some of the highest reported mercury concentrations in Canadian Reservoirs, exceeding 3 µg/g in 700 mm northern pike (Schetagne *et al.*, 2003). In lieu of having information characterizing the flood zone at Robert Bourassa Reservoir, it was assumed that the flood zone conditions were the same as estimated during a field survey of Muskrat Falls Reservoir (AMEC Foster Wheeler, 2017).
- 2. The model calibration from Robert Bourassa Reservoir was applied to Notigi Reservoir, Manitoba, comparing predicted and observed fish mercury concentrations.
- 3. The model was next calibrated to pre-flood conditions in Muskrat Falls Reservoir.
- 4. Simulations were carried out to predict mercury concentrations in Muskrat Falls Reservoir after flooding, using the areal flood zone methylmercury loading rates from the Robert Bourassa simulation.

Regression Model

A regression model (Harris *et al.*, 2015) was also used to predict peak increases in mercury concentrations in northern pike in Muskrat Falls Reservoir. The model is derived from a simplified mass balance expression for methylmercury sources and sinks in reservoirs and predicts peak fish mercury concentrations on the basis of three site conditions: flooded area, total area, and mean annual flow. This model does not predict how concentrations change with time. The version of the model applied to Muskrat Falls Reservoir predicts the increase in fish mercury concentration, which is then added to the baseline concentration.

The sites used to develop the regression model had data for peak concentrations but typically did not measure pre-flood concentrations on a site-by-site basis. Fish mercury concentrations in the vicinity of the Muskrat Falls Reservoir site are low, *e.g.* 0.26 μ g/g in 700 mm northern pike (additional information is presented below) and possibly outside the range of values bounded by the model development data. To help address this issue, the regression model was tested for a range of assumptions regarding baseline concentrations (0.25 μ g/g at all sites or 0.55-0.59 μ g/g based on regional data for 12 reservoirs), flooded areas that contribute to methylmercury supply, and whether to allow the regression intercept to float or be forced through the origin. The equation for the base case model for 700 mm northern pike was as follows:

Increase in fish Hg (
$$\mu$$
g/g) = 0.322 * (A_f/(Q + 0.09* A_t) + 0.202 (1)

Where:

A_f = flooded area (km²) A_t = Total reservoir area (km²) Q = mean annual flow (km³/yr)



The overall peak concentration was then calculated as the increase plus the baseline concentration of $0.26 \,\mu$ g/g.

2.2 Approaches to predict increases in methylmercury concentrations exported downstream from Muskrat Falls Reservoir

Two approaches were used to estimate the increase in methylmercury concentrations and loads exported downstream as a result of flooding at Muskrat Falls Reservoir. The first approach was to use estimates from the ResMerc model application to the reservoir. Daily predictions of methylmercury concentrations and loads exported from the reservoir were generated by the model, for 30 years postflood. The first 5 years after flooding were predicted to have the highest concentrations and export rates and were used in downstream model simulations by Baird (Brunton, 2018).

The second approach was to use data from the FLUDEX experiment as the basis for an estimate. FLUDEX was an upland flooding experiment carried out from 1999-2003 at the Experimental Lakes Area in Ontario (Bodaly *et al.*, 2004, Hall *et al.*, 2005). Three small reservoirs were created, with different carbon pools in the flood zone (per m²). Each year the experimental reservoirs were flooded at approximately the beginning of June and drained in mid to late September. Methylmercury generation and greenhouse gases were studied intensively. Methylmercury concentrations were measured at the inflow and outflow from each reservoir approximately every two weeks during the flood seasons. This information was used to generate net methylmercury loads and export occurring due to flooding as water passed through the reservoirs. Net loads for the 1st three years of the experiment were published (Bodaly *et al.*, 2004, Hall *et al.*, 2005) and loads for years 4 and 5 were obtained from Britt Hall (Hall, 2018). These loading estimates, per m² of flood zone, were scaled up for Muskrat Falls to estimate the downstream methylmercury loads associated with flooding. The FLUDEX site with the highest methylmercury net loads (medium carbon site) was used in the Muskrat Falls Reservoir analysis.

Because flooding occurred from June to September each year during FLUDEX, it was necessary to estimate methylmercury loads for the remainder of the year. One extreme approach would be to assume that no methylmercury would be produced and exported from September through June. That would be unrealistically low. Another option would be to assume that the methylmercury loads produced from June-September would be maintained all year. This would likely be an overestimate, because methylation is temperature dependent. A decision was made to use the average of these two options, effectively using half of the June-September daily average rate for the September to June period. This approach resulted in more than half of the annual estimated methylmercury export occurring from September-June each year.

The two estimates of reservoir methylmercury export were used by Baird (Brunton, 2018) in the downstream modelling analysis.



3 Site Characteristics for Muskrat Falls, Robert Bourassa and Notigi Reservoirs

Muskrat Falls Reservoir

Muskrat Falls Reservoir will have a maximum depth of approximately 27 m at full impoundment, and a mean annual water residence time of approximately 10 days, based a mean annual flow of 1781 m³/s (average for 2006-2015, Water Survey of Canada, 2017). Based on monitoring at station N1 located at the upstream end of the reservoir from December 2016 - December 2017, river water temperatures ranged from -1 to 18 C, the mean pH was 7.4 and the mean dissolved organic carbon was 4.6 mg/L (derived from Nalcor, 2018). The water column is predicted to remain well mixed and oxygenated after reservoir creation (Nalcor, 2009b).

The total area of the reservoir will be 101.5 km². The amount of flooded terrain is 43.9 km² (Table 1). Within the flooded area, 6.9 km² are gravel bars and 6.6 km² are riparian areas with very low carbon content. It was assumed that the flooded area that effectively contributed to elevated methylmercury supply should exclude the gravel bars, and possibly exclude flooded riparian areas. For the purpose of ResMerc and FLUDEX based analyses we conservatively assumed that riparian areas would contribute to methylmercury supply, and the relevant flooded area was 37 km². The regression model was applied using scenarios including and excluding flooded riparian areas.

ELC type	Area (km ²)	% of Reservoir area	% of Flooded Area
Black Spruce / Feathermoss Forest	8.59	8.5	19.6
Fir - White Spruce Forest	8.14	8.0	18.6
Black Spruce / Lichen Woodland	0.91	0.9	2.1
Hardwood Forest	2.20	2.2	5.0
Mixedwood Forest	6.96	6.9	15.9
Spruce Fir / Feathermoss Forest	1.16	1.1	2.6
Bl. Spruce/Sphagnum Woodland	0.20	0.2	0.5
Unvegetated	0.04	0.04	0.1
Wetland	2.18	2.2	5.0
Riparian	6.56	6.5	15.0
Gravel Bar	6.92	6.8	15.8
All flooded forest	28.18	27.8	64.2
All flooded forest + wetland	30.38	29.9	69.2
Total flooded terrain	43.91	43.3	100.0
Total flooded terrain minus gravel bar	36.98	36.4	84.2
Total flooded terrain minus gravel bar and riparian	30.42	30.0	69.3
Water	57.59	56.7	
Total	101.51	100.0	

Table 1.	Muskrat Falls Reservoir flood zone characterization.	Data from AMEC Foster Wheeler	. 2018a
TUNIC II			, 20100



Approximately 5% of the flood zone is wetland, representing ~2% of the overall reservoir area (Table 1). Carbon pools were estimated for the upland flood zone based on the following:

- Humic layer: Field survey by AMEC Foster Wheeler (2017)
- Litter: Literature review by AMEC Foster Wheeler (2018b)
- Foliage: FLUDEX experiment data from Hall *et al.* (2005)

It was assumed that foliage would represent a labile pool of carbon affecting methylation rates while other above-ground vegetation would not contribute to elevated methylmercury supply.

ResMerc has two sediment layers. The top layer was set up with a 2 cm thickness and included carbon from foliage and litter. The lower layer represented the humic layer that averaged about 8 cm in depth (Figure 3).



Figure 3. Carbon pools in model soil layers for Muskrat Falls Reservoir flood zone.

The estimated baseline mercury concentration in a 700 mm northern pike in the reservoir area was 0.26 μ g/g (Figure 4), derived from McCarthy (2017). The estimated baseline concentrations for 400 mm longnose suckers and lake whitefish were 0.17 and 0.12 μ g/g respectively.

The food web related to bioaccumulation by northern pike was set up for ResMerc as shown in Figure 5. Macroinvertebrates are an important component at the base of the northern pike food web in the freshwater system. It was assumed for based case simulations that most of the methylmercury in macroinvertebrates is derived from methylmercury in the water column. Alternative scenarios were also simulated where macroinvertebrates had a greater connection to the pool of methylmercury in sediments.





Figure 4. Observed mercury concentrations in northern pike from River Section 2 in Lower Churchill River. Data from 2012-2016, n=52. Data from McCarthy (2017)



Figure 5. Major food web compartments and links used in simulations for northern pike. Information from J. McCarthy, unpublished.



Robert Bourassa Reservoir

Robert Bourassa Reservoir was developed as part of the La Grande complex in Quebec. Reservoir filling was completed in December 1979. Flooding increased the water surface area from 205 km² to 2,835 km² (Schetagne *et al.*, 2003). The flood zone represented 92% of the total reservoir. With a mean annual flow of 3,374 m³/s, the estimated mean hydraulic residence time was 7 months. Mercury levels in 700 northern pike reached 3.3 μ g/g 11 years after flooding and then declined towards background levels (Hydro Québec, 2013; Schetagne *et al.*, 2003). Upstream reservoirs in the system, including La Grande 3 immediately upstream, which began flooding in 1981, may also have influenced fish mercury levels in Robert Bourassa Reservoir.

Notigi Reservoir

Notigi Reservoir, Manitoba was created when water was diverted south from the Churchill River through the Burntwood/Nelson River system to boost the water supply to several generating stations on the Nelson River. Reservoir filling was completed in December 1976. Flooding increased the water surface area from 198 km² to 785 km² from the South Bay diversion channel to Notigi Dam (Manitoba Hydro, 2006a). The flood zone represented 75% of the total reservoir. With a mean annual outflow of 764 m³/s from Notigi dam (1978-2005, estimated by R. Harris from Manitoba Hydro 2006b), the estimated mean hydraulic residence time was 110 days. Mercury levels in 550 mm northern pike rose to approximately 2 µg/g within 5-7 years, and then declined towards background levels (Bodaly, 2005).

4 Results

4.1 Mechanistic model results

ResMerc was calibrated to estimate the methylmercury loads required from the flood zone to support observed mercury concentrations in northern pike and lake whitefish in Robert Bourassa Reservoir. Annual averaged methylmercury diffusion loads predicted for flooded uplands in Robert Bourassa Reservoir ranged from approximately 80-120 ng/m²/day for years 2-6 after flooding (filling occurred during the first year). The resulting modeled fish mercury concentrations matched observations well (Figure 6). These methylmercury loads from the flood zone produced peak methylmercury concentrations in water of nearly 1 ng/L (Figure 7). No water column methylmercury data were available from existing full-scale reservoirs for comparison.









Figure 7. Predicted methylmercury concentration in surface waters in Robert Bourassa Reservoir, QC.

The calibrated model was then applied to Notigi Reservoir, MB, again assuming the same flood zone characteristics and areal carbon pools in the flood zone as were estimated for Muskrat Falls Reservoir. ResMerc predictions of mercury concentrations in northern pike and lake whitefish reasonably matched observations, with a slight tendency to overpredict (Figure 8).





Figure 8. Observations and ResMerc results for mercury concentrations in northern plke (700 mm) and Lake whitefish (350 mm) in Notigi Reservoir. Observations derived from Bodaly (2005)

ResMerc was next applied to pre-flood conditions in the lower Churchill River at the Muskrat Falls site. Rate constants for mercury cycling and carbon turnover were the same as used for Robert Bourassa and Notigi Reservoirs. The simulation was "warmed up" for 100 years to allow conditions to stabilize, and results were examined for the 101st year. Simulated and concentrations reasonably matched observations of methylmercury in water (Figure 9) and fish (Figure 10). Minor adjustments were made to rate constants for fish methylmercury dynamics to improve the model fit.



Figure 9. Observed and simulated methylmercury concentrations in surface waters for pre-flood conditions at the Muskrat Falls Reservoir site. Data from Station N1 (Nalcor, 2018)





Figure 10. Observed and simulated fish mercury concentrations for pre-flood conditions at the Muskrat Falls Reservoir site. Black dots are observations. Coloured points represent predicted concentrations in different year classes. Data from 2010 to 2016 in River Section 2. Data from J. McCarthy (2017).

The model was then applied to post-flood conditions for Muskrat Falls Reservoir. Due to limitations with the model's ability to simulate the filling period, post-flood simulations started with the reservoir at full elevation (39 m asl). Predicted average annual methylmercury diffusion loads from flooded soils to overlying water ranged from approximately 80-145 ng/m²/day from flooded uplands during the first 6 years after flooding. Methylmercury concentrations were predicted to increase briefly to approximately 0.1 ng/L in surface waters of Muskrat Falls Reservoir, about 5X the baseline concentration, and the contribution from flooding briefly reached a peak of 0.07 ng/L (Figure 11). The peak export rate for methylmercury briefly reached a peak of 10 g/day (Figure 12). Peak predicted fish mercury concentrations were 0.64 μ g/g in 700 mm northern pike and 0.24 μ g/g in 400 mm lake whitefish (Figure 13). These values are roughly 2.0 – 2.5X the baseline concentrations. An alternative scenario was simulated assuming that 50% of the base of the food web derived methylmercury from sediments postflood. The peak predicted mercury concentration for 700 mm northern pike was 0.80 μ g/g, approximately 3X the baseline. Overall, peak concentrations predicted in northern pike were 2-3X the baseline concentrations for the scenarios tested.





Figure 11. Predicted methylmercury concentrations in Muskrat Falls Reservoir surface waters (and exported downstream) based on ResMerc model.



Figure 12. Predicted methylmercury export from Muskrat Falls Reservoir based on ResMerc model.





Figure 13. Predicted methylmercury concentrations in Muskrat Falls Reservoir northern pike and lake whitefish, based on the ResMerc Model base case simulation.

4.2 Regression model results

Regression model estimates of peak increases in mercury concentration for 700 mm northern pike in Muskrat Falls Reservoir are shown in Figure 14. The y axis in the figure shows the predicted increase, which must be added to the baseline to estimate the overall peak concentration. The base case model, shown in the figure, allowed the regression intercept to float, although in reality no flooding would produce no increase. Muskrat Falls Reservoir is predicted to have a peak concentration between 0.61 and 0.64 μ g/g, about 2.4X baseline concentrations. A range of model outcomes based on different assumptions about baseline fish mercury concentrations, effective flooded area, and whether to allow the model intercept to float or be forced through the origin, is shown in the red shaded area in Figure 14. These results were very consistent with ResMerc predictions.





Figure 14. Regression model results for 700 mm northern pike. Blue dots are estimated increases in mercury concentrations in 12 Canadian reservoirs, based on observed peak concentrations and estimated baseline concentrations of 0.55-0.59 μ g/g. Blue line is model version with floating intercept. Red dots are predictions for Muskrat Falls Reservoir based on flooded area of 30-37 km². Shaded red area includes predictions for a range of assumptions related to the intercept (floating or forced through origin, flooded area (30-37 km²), and baseline concentrations (0.25 at all sites or 0.55-59 μ g/g). Predicted increase must be added to the baseline concentration to estimate the overall peak concentration. Additional information on field data for existing reservoirs available in Harris *et al.* (2015).

4.3 Methylmercury export estimates based on FLUDEX

Methylmercury export rates observed during the FLUDEX experiment are shown in Table 2. These data represent the net export rates each year, based on outflow minus inflow fluxes during the flood period from approximately June – September. The medium carbon site (#2) had the highest methylmercury export rates and was used to estimate methylmercury export for Muskrat Falls Reservoir, shown in Table 3. Results from FLUDEX were applied to Muskrat Falls Reservoir as follows:

- Areal loading rates from FLUDEX from the June -September flood season each year were extended to annual estimates by assuming that if flooding had continued each year, the daily loads would have been half the average rate during the June-September period (see earlier discussion).
- FLUDEX net export rates, per m², were multiplied by the portion of the Muskrat Falls flood zone assumed to contribute to excess methylmercury supply (37 km²), to estimate the mass of methylmercury exported each year from Muskrat Falls Reservoir. This area included about 6.5 km² of flooded riparian terrain, which may not contribute as much methylmercury (per m²) as flooded forest soils.



Year after flooding	Methylmercury export (mg/ha)		
	Reservoir 1	Reservoir 2	Reservoir 3
1	66	126	60
2	77	131	61
3	44	88	32
4	71	42	31
5	38	29	11

Table 2. Net methylmercury export (outflow minus inflow) for the three FLUDEX reservoirs from 1999-2003. Values are mg/ha for flood season each year (approximately June – September). Site 2 data wereused for Muskrat Falls Reservoir analysis. Data from Hall (2018).

	Estimated annual
Year after	methylmercury export
flooding	(kg)
1	1.08
2	1.13
3	0.76
4	0.36
5	0.25

Table 3. Estimated annual methylmercury export from Muskrat Falls Reservoir for first 5 years after flooding (excess above baseline, associated with flooding). Estimates are based on FLUDEX data, scaled up to flooded area at Muskrat Falls that contributes to excess methylmercury supply (37 km²). Annual values are based on FLUDEX Reservoir 2 data for June-September each year, plus contribution for remainder of year assuming half the average daily rate for June-September period.

Predicted average annual increases in methylmercury concentrations in water exported from the reservoir are presented in Figure 15 for the FLUDEX and ResMerc based analyses. The FLUDEX-based estimates had largely declined after 5 years. The concentrations based on the model calibration from Robert Bourassa Reservoir were higher and declined more slowly that the estimates based on FLUDEX. Both estimates are much lower than the increase in concentration in the reservoir predicted by Calder et al (2016), also shown in Figure 15. The predicted increase by Calder *et al.* (2016) was 0.16 ng/L, sustained for an undefined period long enough for fish mercury concentrations to respond in proportion. This concentration is 4.6 to 8X greater than the maximum one-year average increases predicted using FLUDEX or ResMerc based estimates, and 5-9X greater than the maximum 3 year average increases from FLUDEX and ResMerc.





Figure 15. Estimated increases in methylmercury concentrations in waters exported from Muskrat Falls **Reservoir.** Overall concentration = increase + baseline. Also shown is predicted increase from Calder *et al.* (2016).

5 Discussion

Two approaches were used to estimate the magnitude and timing of downstream export of methylmercury in water from Muskrat Falls Reservoir to Goose Bay and Lake Melville. The first approach used observations from FLUDEX, while the 2nd approach used the ResMerc model to backestimate flood zone methylmercury loading rates that would produce fish mercury concentrations observed from two existing reservoirs, and then applied those loading rates to predict methylmercury concentrations in water and fish in Muskrat Falls Reservoir. The use of these two approaches was influenced by the absence of measured concentrations of methylmercury in waters from full scale reservoirs. The only known datasets are from the FLUDEX upland and ELARP wetland reservoir experiments at the Experimental Lakes Area, and the FLUDEX data formed an important component of the analysis. The ResMerc model analysis provided a second means to gain insights into water column methylmercury concentrations that occur in new reservoirs.

The FLUDEX-based analysis predicted an increase of 0.02 ng/L in water exported from Muskrat Falls Reservoir (maximum one-year average). The ResMerc analysis predicted an increase of 0.035 ng/L. These estimates are within a factor of 2 of each other in magnitude, which is encouraging in the absence of being able to develop confidence limits associated with predicted increases in water column methylmercury, which would require observations of methylmercury concentrations in water from multiple reservoirs. It is also possible that the actual methylmercury loads from Robert Bourassa Reservoir, per m², were greater than occurred during FLUDEX, given that many factors affect the production of methylmercury in reservoirs. The FLUDEX site for example experienced a fire roughly 20



years prior to the experiment. Whether this reduced carbon pools relevant to methylmercury production that was available for bioaccumulation during FLUDEX is not clear. Conversely, Robert Bourassa Reservoir was downstream of other reservoirs that could have contributed to higher fish mercury levels, including La Grande 3. The ResMerc analysis did not explicitly simulate upstream reservoir contributions and allocated any methylmercury supply needed to produce observed fish mercury levels to in-situ production in Robert Bourassa Reservoir. These considerations guided the decision to use the average of the two estimates of downstream export in simulations in Goose Bay and Lake Melville by Baird (2018).

The predicted increases in water methylmercury concentrations exported from Muskrat Falls Reservoir were 4.6 to 8x lower than the 0.16 ng/L increase predicted by Calder *et al.* (2016), which predicted much higher loads of methylmercury from the flood zone than were observed from FLUDEX or predicted from the ResMerc model analysis.

The mechanistic and regression models produced very similar predictions of peak fish mercury concentrations in Muskrat Falls Reservoir, providing consistency among the various lines of evidence used in the analysis. Both models predicted that concentrations in 700 mm northern pike would increase roughly 2.5-3X from a baseline concentration of 0.26 μ g/g to a peak in the range of 0.6 to 0.8 μ g/g.

Flow dilution was predicted to be an important moderating factor in the mechanistic and regression model predictions for Muskrat Falls Reservoir. For example, the flood zone methylmercury loading rates (per m²) applied to Robert Bourassa Reservoir in ResMerc simulations produced much lower peak concentrations in water and fish when approximately the same rates were applied to Muskrat Falls Reservoir. This was related to the shorter water residence time (~10 days vs 7 months) and greater flow dilution associated with Muskrat Falls Reservoir, and the fraction of the reservoirs consisting of flooded terrain: about 92% Robert Bourassa Reservoir versus ~40% for Muskrat Falls.

Overall, methylmercury concentrations (baseline + increase) in waters exported downstream from Muskrat Falls Reservoir were predicted to peak at roughly 2-3X baseline values when averaged over time periods relevant to bioaccumulation in adult fish (*e.g.* one-year average concentration up to 0.04 to 0.055 ng/L), based on the two approaches used. The results of this analysis were used in downstream analyses by Brunton (2018) and Wood (2018) to estimate potential increases in methylmercury concentrations in water and biota in Goose Bay and Lake Melville.



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