

Mercury in Air, Water and Biota in Kejimikujik National Park, Nova Scotia, Canada

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Abstract. Mercury in the environment (Hg) has re-emerged as an environmental issue in light of recent findings of elevated mercury levels in the tissues and blood of fish, mammals and aquatic birds in many areas of eastern North America including the lakes and streams of Kejimikujik National Park. The Atlantic provinces lie downwind of North American anthropogenic pollution emission source regions and various atmospheric studies have shown that pollutants can travel long distances and be deposited far from initial emission sources. Anthropogenic input of mercury to regional ecosystems via long range transport of airborne pollutants is a concern in this region. Studies of atmospheric total gaseous mercury (TGM) in the Atlantic region have found that TGM concentrations in ambient air at Kejimikujik National Park in 1996 ranged from 0.51 to 2.9 ng/m³ with an annual median concentration of 1.52 ng/m³. TGM levels were similar at two other regional sites, St. Andrews, N.B. (median 1.50 ng/m³) and Acadia National Park, Maine (median 1.73 ng/m³). Total mercury in precipitation for a 6 month period in 1996 in Kejimikujik averaged 11.4 ng/L compared to 9.1 ng/L in St. Andrews. Mercury deposition via wet precipitation was estimated to be 8.4 ug/m²/yr in the St. Andrews area and 10.5 ug/m²/yr in Kejimikujik. Mercury in biota at Kejimikujik National Park has been measured at levels which have been shown elsewhere to cause detrimental effects to wildlife and, in the case of some sportfish, at levels (0.35 - 2.0 ug/g wet wt.) which exceed safe consumption guidelines.

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1. Introduction

Environmental mercury has been a concern for a number of years due to the ability of mercury to both bioaccumulate in selected tissues of organisms and biomagnify progressively higher up the trophic food chain. Recent developments in technologies directed at improving the precision and accuracy of mercury measurements in the environment as well as renewed concerns regarding mercury levels in biota have led to a resurgence in research into the environmental fate and effects of mercury.

The sources and pathways of chemicals released into the environment can be varied and complex. Mercury is such a chemical because of its wide use in industry, its abundance in the natural environment and its ability to deposit and revolatilize, thus continually recycling in the

environment. Nevertheless, it is important to understand the dynamics of mercury emissions, transport, chemical transformation, deposition and movement in the environment as well as the effects of mercury on biota in order to gain an understanding of the relationship between naturally occurring and anthropogenic mercury and thus, the feasibility and efficacy of controls on releases of anthropogenic mercury into the environment.

There are a variety of methods which can be applied to improve the understanding of environmental cycling of mercury. One method which has been used successfully in acidification and toxics related research is the mass balance approach (eg Diamond *et al.*, 1993). It is proposed that the mass balance approach be implemented to understand mercury dynamics in selected watersheds in Kejimikujik National Park. This paper presents a summary of the preliminary results of recent studies in Kejimikujik National Park aimed at defining the mercury issue regionally and to be used in the development of a watershed mass balance approach to understanding mercury in regional ecosystems.

2. Materials and Methods

2.1 Ambient Air

Total gaseous mercury (TGM) in air was measured with a continuous automated mercury vapor analyzer (TEKRAN TM) using dual cartridge gold traps followed by thermal desorption of adsorbed mercury and analysis by Cold Vapor Atomic Fluorescence Spectrophotometry (CVAFS) at $\lambda = 253.7$ nm. Ambient air was continuously sampled at a rate of 1.5 L/min for 5 min through a heated sampling line and Hg trapped onto the gold cartridges. The TGM analyzer underwent automated calibrations every 25 hr. using an internal Hg permeation source in addition to quarterly calibrations done manually using an external temperature controlled Hg perm source. Various quality assurance (QA) and quality control (QC) checks were performed regularly including injection testing for line loss, zero air flow checks for line source and flow meter/controller calibrations.

TGM measurements at Kejimikujik National Park (44°26'10" 65°12'20") and St. Andrews (45°05'12" 67°04'30") were based on 12 months of data. Acadia National Park (44°22'26" 68°15'38") TGM measurements began in July 1996 thus results for this site represent only 6 months of measurement.

2.2 Precipitation

Precipitation samples were collected using an Aerochem Metrics Model 301 Automatic Sensing Wet/Dry Precipitation collector with TeflonTM coated lid supports and gasket pads to prevent evaporative sample loss. The sampler was equipped with a 128 cm sampling orifice and a borosilicate glass sampling train leading to a 1 litre borosilicate glass bottle housed in a temperature controlled enclosure. Precipitation amounts were measured using a Belfort Model B-5-780 recording rain gauge. Belfort precipitation amounts were compared with an adjacent standard fixed mount rain gauge.

The precipitation collection, analysis and QA/QC were carried out according to protocols developed by the National Atmospheric Deposition Program (NADP)/Mercury Deposition Network (MDN). Descriptions of instrumentation, sampling, analytical and QA/QC protocols are contained in NADP (1996a, 1996b). Precipitation sampling began in Kejimikujik in July 1996. The data presented for Kejimikujik National Park and other MDN sites represents only the July - Dec 1996 (6 month) period for purposes of comparison.

Annual Hg deposition at Kejimikujik and St. Andrews were estimated by factoring net precipitation and deposition amounts measured over the 6 month sampling period to the site specific 25 yr. climate normal precipitation depths.

2.3 Water -

Water samples were collected in the summer of 1996 for analysis of mercury concentrations. This was a single event sampling of 31 lakes in Kejimikujik National Park included single samples taken for most lakes but replicate samples taken from a subset of lakes for verification of spatial representativeness.

All water samples submitted for mercury analysis were analyzed by Cold Vapor Atomic Fluorescence Spectrometry (CVAFS). Water samples were collected in 500 ml Teflon bottles. Samples were oxidized through the combined action of bromine monochloride and UV irradiation for 24 hr. Prior to analysis, each sample was pre-reduced with hydroxylamine. An aliquot of 250 ml was then transferred to a bubbling apparatus containing stannous chloride and the free mercury volatilized was purged through a gold coated sand trap. The trap was connected to the CVAFS detector which measured the thermally desorbed mercury. Peak area was used to calculate concentrations. The result represented all mercury species that could be liberated from the sample matrix and reduced to Hg⁰.

2.4 Human Consumption and Mercury in Biota

Fish were collected in the field by various methods including electro-seining, gill netting, trapping and angling. Whole fish were stored in Zip-Lock™ bags, packed in ice, then frozen. Samples for total mercury content were analyzed by digestion and by Cold Vapor Atomic Absorption Spectroscopy (CVAAS).

Two fish sampling protocols were used in this study. The two different protocols were used to provide better representation of wildlife versus human consumption patterns. Humans tend to consume only muscle tissue (fillets) so human consumption Hg data is based on analysis of fish fillets only. Loons and other piscivorous birds and wildlife generally consume whole fish and these data are based on whole fish samples and analysis.

2.4.1. Human Consumption:

Muscle tissue (fillets) from brook trout (*Salvelinus fontinalis*) and white perch (*Morone americana*) were sampled separately with emphasis on collecting larger fish which are more representative of sizes angled for human consumption. Fish with a fork length greater than 20 cm were collected from various water bodies for Hg analysis.

2.4.2 Wildlife Consumption:

Composite samples of whole yellow perch (*Perca flavescens*) were sampled for total and methyl-Hg analysis (methyl Hg analyses are not presented here). Several criterion were used to determine where fish were sampled. The lake selection criterion were as follows: loons (*Gavia immer*) must be present on that lake, lakes should show a range in representative water chemistry the most important of which is based on organic content (e.g. clear vs. colored), geology (representative and unique), historical land use (old mine tailings) and lakes with current related collaborative studies (behavioral, nesting success brood rearing).

A total of 22 lakes in Kejimikujik were selected for fish sampling. Nine composite samples were collected with 3 fish per composite, each composite representing 1 of 3 size classes (age) where fish < 10 cm ~1-2 yr. old, 10.1-14.9 cm 3-4 yr. and > 15 cm ~ 15+ yr. of age for a total of 27 fish covering the 3 - 10 cm size class intervals within the size range normally consumed by loons. Kejimikujik L. was sampled similarly but in 3 locations due to its size and heterogeneity.

2.4.3 Common Loons

Adult loons were captured using night light techniques. Blood samples were collected and analyzed for total mercury by CVAFS.

3. Results and Discussion

3.1 Ambient Air -

The atmosphere has an important role in mercury transport and deposition (Fitzgerald *et al.*, 1991; Lindberg *et al.*, 1991). Mercury inputs to ecosystems can result from direct deposition via precipitation or through air-water (Schroeder *et al.*, 1992) and air-soil gas exchange processes (Xiao *et al.*, 1991; Lindberg *et al.*, 1992; Schroeder, 1995; Poissant *et al.*, 1996).

Total gaseous mercury (TGM) in ambient air was measured at 3 sites in the Atlantic region including Kejimikujik National Park. Table 1 provides a statistical summary of TGM measurements conducted at the 3 sites. All of the sites showed similar TGM concentrations in ambient air during 1996 with median concentrations in the 1.5 to 2.0 ng/m³ range. However, annual mean TGM concentrations were significantly different ($p < 0.001$) between the sites. Acadia National Park showed the highest mean TGM concentration at 1.79 ng/m³ (median 1.73 ng/m³). Minimum values were also similar at all 3 sites, never going below 0.5 ng/m³. Maximum TGM values did not exceed 3 ng/m³ at Kejimikujik but Acadia National Park reached close to 4 ng/m³. Maximum TGM at St. Andrews and Kejimikujik were slightly lower at 3.36 and 2.87 ng/m³ respectively.

Median TGM concentrations exhibited little variation on a diurnal or seasonal basis at the St. Andrews or Acadia sites. Kejimikujik TGM behaved somewhat differently from the other 2 sites (Figure 1).

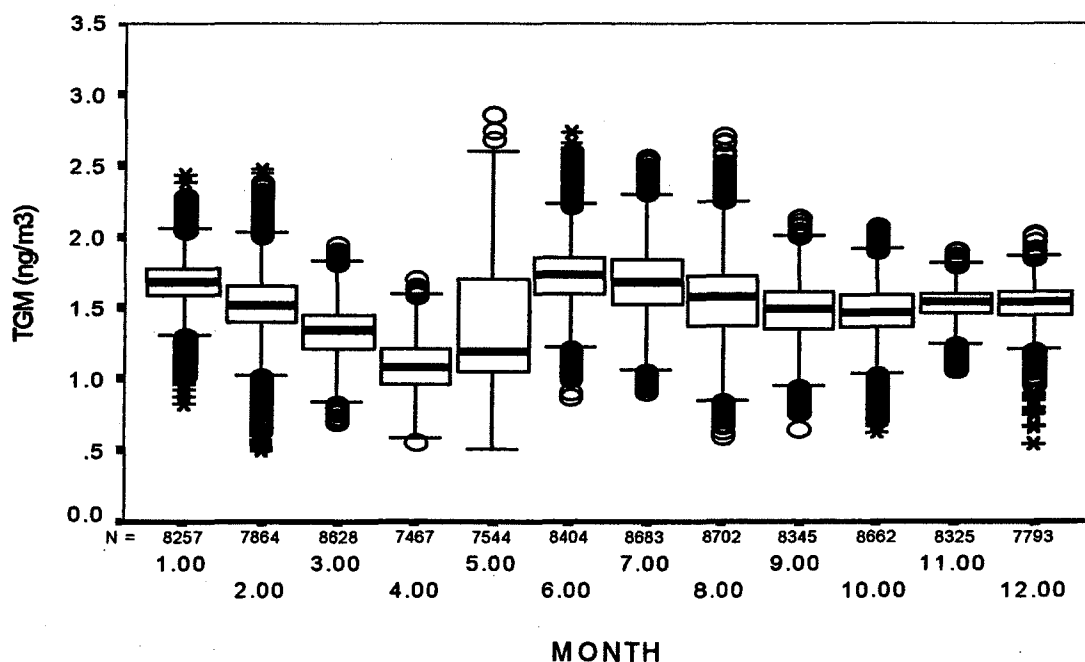


Figure 1: Monthly variability in total gaseous mercury concentrations in ambient air at Kejimikujik National Park in 1996 (circles represent near outliers and asterisks represent far outliers).

Table 1 : Statistical summary of annual total gaseous Hg in ambient air at 3 East Coast sites in 1996 (data for Acadia National Park represents the July-Dec 1996 period only).

Kejimikujik National Park (Nova Scotia) -

	N		Mean	Median	Std. Dev	Var	Min	Max	Percentiles			
	Valid	Missing							25	75	95	99
TGM (ng/m ³)	98674	6446	1.49	1.52	.27	.07	.51	2.87	1.33	1.67	1.90	2.09

St. Andrews (New Brunswick) -

	N		Mean	Median	Std. Dev	Var.	Min	Max	Percentiles			
	Valid	Missing							25	75	95	99
TGM ng/m ³	80534	7053	1.56	1.50	.31	.10	.50	3.36	1.31	1.84	2.07	2.21

Acadia National Park (Maine) (July-Dec only) -

	N		Mean	Median	Std. Dev	Var	Min	Max	Percentiles			
	Valid	Missing							25	75	95	99
TGM (ng/m ³)	37063	3544	1.79	1.73	.24	.06	.53	3.97	1.64	1.89	2.23	3

TGM in Kejimikujik showed a marked seasonal cycle with the highest median values found in the summer months. Fall median TGM concentrations were relatively stable but declined markedly in the colder winter months. The greatest variation in monthly concentrations occurred in May when TGM concentration ranged from 0.5 to 2.9 ng/m³. The seasonal variability in Kejimikujik is partially related to a strong diurnal TGM cycle present on warm sunny days during the summer months. It is uncertain whether this diurnal cycle is related to local volatilization or boundary layer dynamics. Given the relatively long atmospheric lifetime of elemental mercury in the atmosphere, limited reactivity and relatively uniform vertical distribution (Beauchamp *et al.* 1996) it appears that boundary dynamics may not account for the variability.

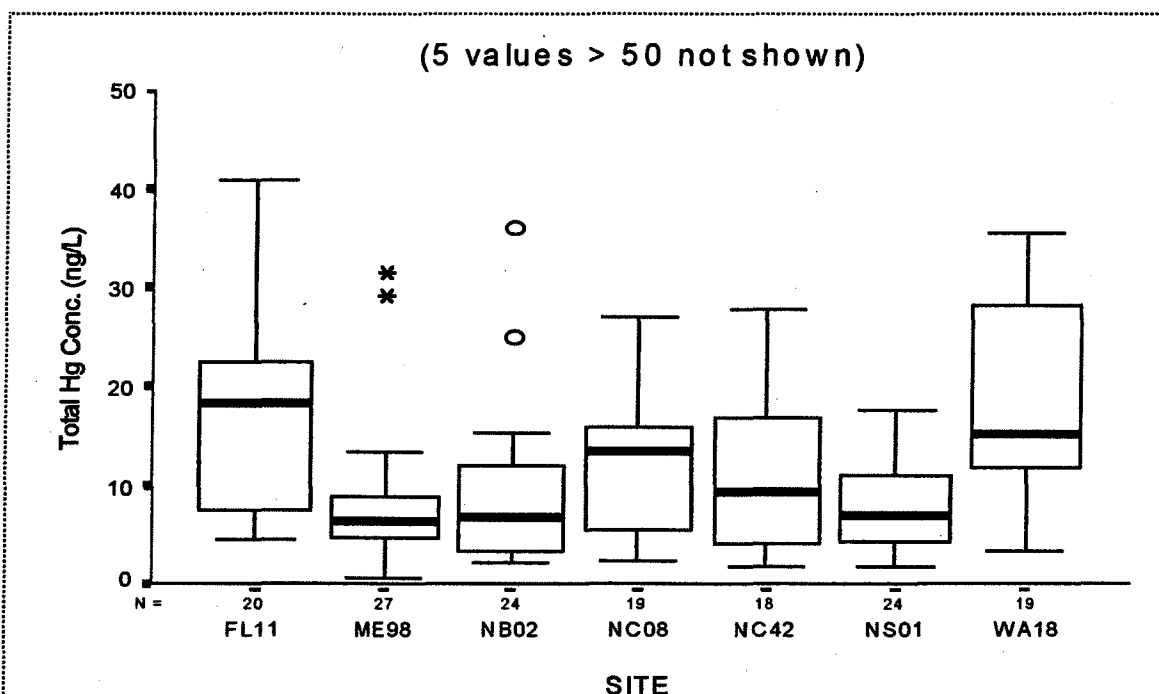
TGM concentrations at Kejimikujik and the other two locations were similar to levels found at other North American sites (Schroeder, 1994; Gill *et al.*, 1995; Burke *et al.* 1995; Poissant *et al.*, 1995) and globally (Iverfeldt *et al.* 1995).

3.2 Precipitation -

3.2.1. Hg in precipitation:

Mercury input to ecosystems through the wet deposition pathway can be substantial depending on the concentration of mercury in precipitation and the amount of precipitation occurring on an annual basis in a particular region. Mercury concentration in weekly bulk samples from Kejimikujik National Park were collected and analyzed as part of the Canada/US Mercury

Deposition Network (MDN) and are compared with samples collected at other North American sites in Figure 2 .



NOTE: FL11= Florida Everglades, FL; ME98 = Acadia National Park, ME; NB02 = St. Andrews, NB; NC08 = Waccamaw State Park, NC; NC42 = Pettigrew State Park, NC; NS01 = Kejimikujik National Park, NS; WA18 = Sand Point, Seattle, WA (US data courtesy of NADP/MDN).

Figure 2 : Total mercury concentration in weekly bulk precipitation samples collected at several North American sites in the period July-Dec 1996 (circles represent near outliers and asterisks represent far outliers)

The median concentration of total mercury in Kejimikujik precipitation was amongst the lowest of the 7 MDN sites during the July-Dec 1996 sampling period (median = 7.0 ng/l) and were comparable to concentrations in precipitation measured at the Maine (median = 6.5 ng/l) and the St. Andrews sites (median = 6.85 ng/l). Precipitation mercury concentrations tended to be greater and more variable at the other south eastern US and the west coast sites.

3.2.2. Hg deposition via precipitation:

Precipitation sampling began in July 1996 at Kejimikujik and St. Andrews thus measured concentration and deposition data is available for only 6 months and, as such, it is currently not possible to calculate the exact deposition which occurred in 1996. However, by using climate statistics (Environment Canada 1993) it is possible to approximate the amount of mercury deposited via wet deposition at these 2 sites. Kejimikujik normally receives 1397 mm/yr. and St. Andrews on average receives 1199 mm/yr. of precipitation based on 25 year climate statistics (1966-1990). By adjusting precipitation amount (depth) and Hg deposition measured over the current 6 month period at these 2 sites to the climatological average precipitation amount received over an entire year, it was estimated that Kejimikujik would receive approximately 10.5 ug/m² of mercury annually and St. Andrews 8.4 ug/m² through wet deposition. These deposition estimates are consistent with measurements in the northeastern US (Hoyer *et al.* 1995) and within the range of deposition values observed in Europe (eg. Steinnes and Anderson, 1991).

3.3 Lake Water -

Water samples were collected from 31 lakes in Kejimikujik National Park in the summer of 1996 for analysis of mercury concentrations. Samples were analyzed for total and total dissolved Hg, the results of which are shown in Table 2. Total Hg in the surface waters of 23 Kejimikujik lakes ranged from 1 ng/l to almost 10 ng/l. On average, two-thirds of the mercury was in the dissolved fraction with the remainder in the particulate fraction.

Kejimikujik lakes are comprised of oligotrophic, shallow (mean depth < 4 m), well mixed lakes, only a few of which show weak thermal stratification during the summer. The major contrast between lake types is related to organic content (water color) (Kerekes *et al.*, 1986). Lakes with high concentrations of dissolved organic carbon (DOC) tend to have higher mercury content than clear water lakes and it is likely that much of the mercury is bound in the organic fraction. If this is the case, the concentration of Hg may vary substantially year to year based on highly variable DOC content (Beauchamp and Kerekes, 1989).

Table 2: Total, total dissolved, total particulate, percent dissolved and percent particulate mercury in 31 lakes in Kejimikujik National Park (Hg in ng/l).

	N		Mean		Median	Std. Deviation	Variance	Minimum	Maximum
	Valid	Missing							
				Std. Error					
Total Hg	23	8	4.22	.43	4.65	2.05	4.18	1.04	9.57
Total Diss.	30	1	3.44	.35	3.87	1.93	3.72	.48	8.12
Part. Hg	21	10	.98	.14	.96	.63	.39	.00	2.40
% Diss. Hg	21	10	71.67	4.82	80.00	22.08	487.73	29.00	100.00
% Part. Hg	21	10	28.33	4.82	20.00	22.08	487.73	.00	71.00

Additional data on Hg concentrations in water, sediment, plankton and macrophytes collected from 3 Kejimikujik lakes is available in Freedman *et al.*, (1989).

3.4 Human Consumption and Mercury in Biota

3.4.1 Human Consumption

Mercury levels in fish are a concern from two perspectives; a) the effects of fish consumption on human health and; b) similar effects on wildlife. Sport fishing is a popular and widespread activity throughout Nova Scotia. Degradation in the quality of recreational fishery could have health implications in addition to indirect socio-economic effects through changes in ecotourism.

Recent collection and analysis of mercury in sport fish sampled at selected locations throughout the Atlantic region have shown elevated Hg concentrations in tissues of fish taken from some locations and this has raised concerns regarding the health effects of human consumption of these fish. Two of the species which are frequently angled and consumed by visitors to Kejimikujik National Park are brook trout and white perch.

Sampling of these 2 species was carried out in Kejimikujik lakes in 1996. Large brook trout (fork length > 20 cm) were collected from several lakes according to the protocols set out previously. Figure 3 illustrates the distribution of mercury in the fillets of 81 brook trout relative to fork length.

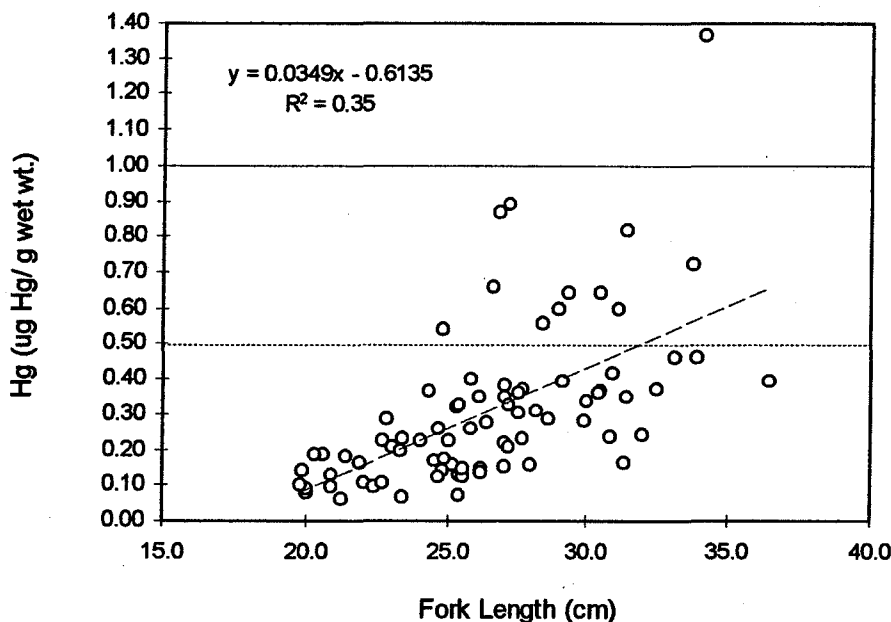


Figure 3 : Total mercury content in brook trout fillets collected from Kejimikujik lakes (horizontal dashed line at 0.50 ug/g wet wt represents the safe consumption guideline; the horizontal solid line at 1.0 ug/g wet wt represents the no consumption guideline).

There is a significant trend toward increased Hg content with increased fork length ($p < 0.01$) although there is substantial variability in the data set likely due to between lake differences. Twelve of the 81 brook trout (~15 %) exceeded the safe consumption guideline of 0.5 ug/g wet wt. and one of the sampled trout exceeded the no consumption guideline (1.0 ug/g wet wt). However, median Hg concentration in brook trout fillets (0.31 ug/g wet wt.) was below safe consumption guideline. Maximum Hg concentration was 1.37 ug/g wet wt. and the minimum level in the sample population was relatively low at 0.06 ug/g wet wt.

Similar data for white perch fillets collected from Kejimikujik lakes are illustrated in Figure 4. As was the case for brook trout, the focus of the perch sampling effort was placed on collection of larger fish to better represent size classes consumed by humans. Both brook trout and white perch showed a trend toward increased mercury concentration in muscle tissue with increased fork length although the trend in white perch was not statistically significant at $p < 0.05$.

One difference between the trout and perch data sets was that the white perch sampled tended to have substantially more mercury content in the fillets compared to a similar size range of trout. Median Hg content in white perch fillets exceeded the no consumption guideline at 1.03 ug/g wet wt., which was also 3 times the median Hg concentration measured in brook trout. Mercury content in 17 of the 30 white perch sampled (57 %) exceeded the safe consumption guideline and 11 of these (37 % of the entire sample) exceeded the no consumption guideline of 1.0 ug/g wet wt (Figure 4). These figures indicate that mercury in wild sport fish populations is a concern in the area.

Median Hg concentrations in brook trout and white perch were similar to levels found in whitefish (*Coregonus clupeaformis*) in the James Bay, La Grande Reservoir in Quebec (Brouard *et al.*, 1994) and Adirondack (NY) lakes (Simonin, 1994).

Mercury concentrations in fish vary in relation to a number of factors including length, weight, age, position in the food chain, lake chemistry and morphometry, thus the exact cause of elevated Hg levels in Kejimikujik fish remains to be determined as well as whether or not the mercury is derived from within the watersheds or from external sources such as the atmosphere or if these sources are natural or anthropogenic in nature.

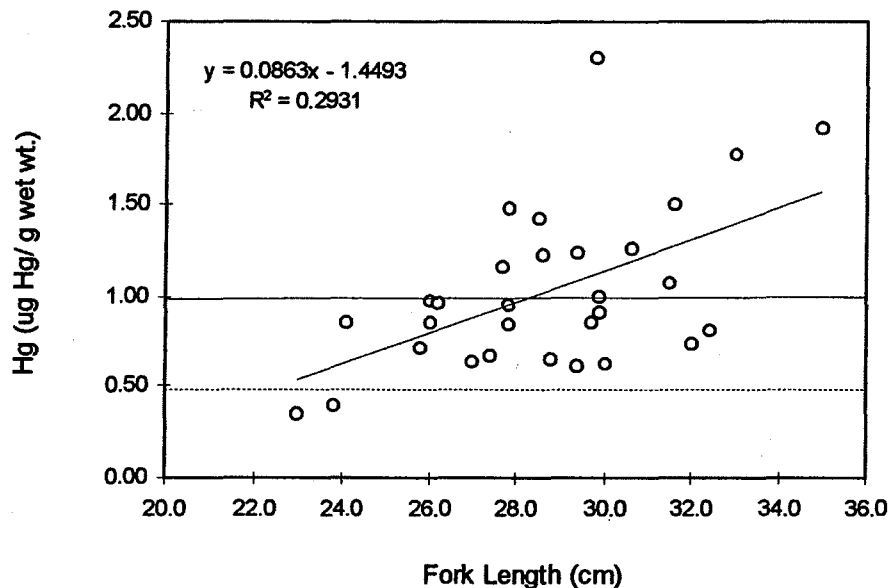


Figure 4 : Total mercury in 30 white perch fillets collected from Kejimikujik lakes (the solid line is line of best fit, horizontal dashed line at 0.50 ug/g wet wt. represents the safe consumption guideline, the horizontal solid line at 1.0 ug/g wet wt. represents the no consumption guideline).

3.4.2. Wildlife Consumption -

Fish comprise either part or the bulk of the diet for several wildlife species occurring throughout the Atlantic region including Kejimikujik National Park. As indicated earlier, many of the lakes in Nova Scotia and Kejimikujik are shallow glacial lakes. These lake waters are generally within a few degrees of mean daily temperature and as such warm water fish species are among the most abundant. One of the most widespread and abundant of these species is yellow perch which frequent shallow, near surface waters and are easily caught by piscivorous birds.

Avian and mammalian predators generally consume whole fish and data related to fillets does not give adequate representation of whole body mercury burdens thus, replicate composite samples of whole yellow perch were analyzed for mercury content based on 3 size classes (5-10, 10-15 and 15-20 cm fish).

Again, mercury content in fish tissues tends to increase with increased size class of fish (Figure 5). Median Hg content was 0.14 ug/g wet wt. in the smallest size class (5-10 cm) compared to 0.22 ug/g wet wt. in fish 10-15 cm in length and 0.37 ug/g wet wt. in the 15-20 cm length class. The entire set of 222 composite samples of whole fish had a median mercury content of 0.22 ug/g wet wt. Mercury content in whole fish are obviously lower than those of fillets only with proportionally fewer of the composite samples in any size class having mercury levels exceeding consumption guidelines (maximum composite sample Hg = 0.77 ug/g wet wt.).

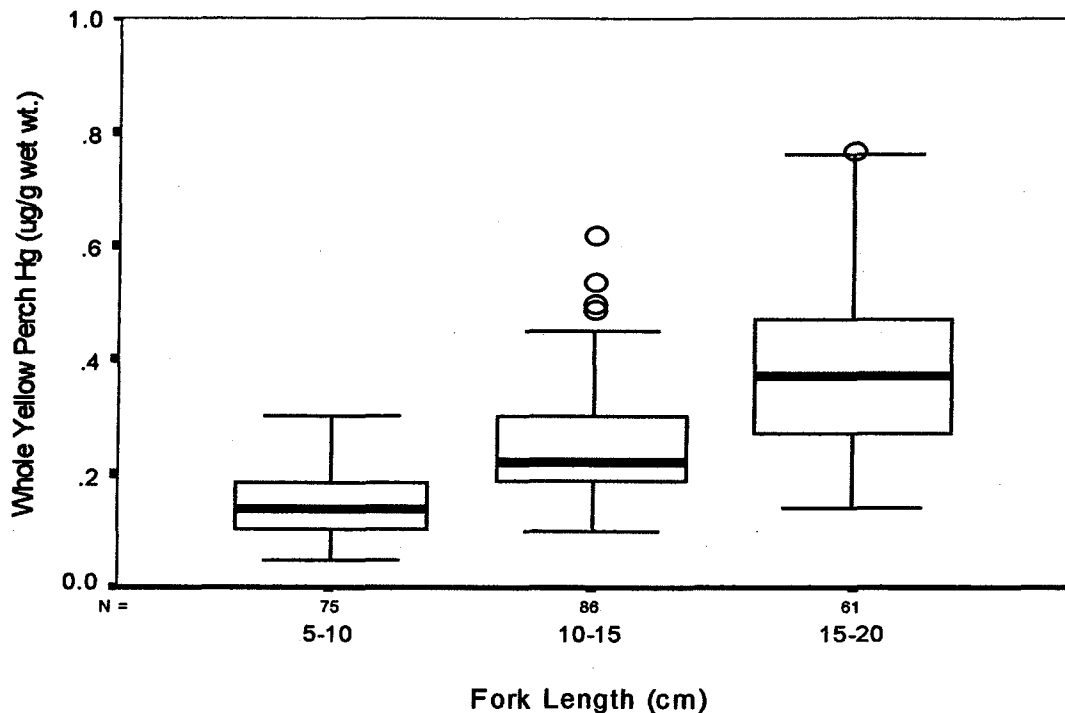


Figure 5: Total mercury content in three size classes of whole yellow perch collected from Kejimikujik lakes (N= number of samples in each size [length] class).

3.4.3. Common Loons

Loons are top trophic feeders which frequent Kejimikujik lakes generally breeding on lakes ≥ 40 ha (Kerekes *et al.* 1994, 1996). Measurements of Hg concentration in loon blood provides a non-destructive means for acquiring an index of the Hg body burden in this species. The most important threshold for mercury effects in loons is a total mercury concentration of 0.3- 0.4 ppm in the fish upon which they feed. This appeared to be the threshold for reproductive impairment in loons in the English-Wabigoon river system in northwest Ontario in the 1970s (Barr 1986). It is evident from the whole fish data presented above, that these levels are frequently exceeded in Kejimikujik lakes.

Figure 6 illustrates blood mercury levels in adult loons sampled at various locations in the Maritimes are high compared to other North American sites and loons in Kejimikujik have blood mercury levels significantly greater than all of the other North American sites.

4. Conclusion

Mercury is ubiquitous in the global environment yet some areas when compared to others of a similar nature, show elevated concentrations in various media including biota. Data from the Kejimikujik National Park and surrounding area show that mercury is present in the air and precipitation falling on the land and water surfaces. Mercury is clearly present in water and at levels in some components of the food chain, fish and birds specifically, at levels which exceed consumption guidelines and which are known to cause behavioral and reproductive impairment. As such, there is sufficient evidence that mercury poses a real threat to ecosystem health, human health and biodiversity.

Mean mercury levels (± 1 SD) in adult common loons

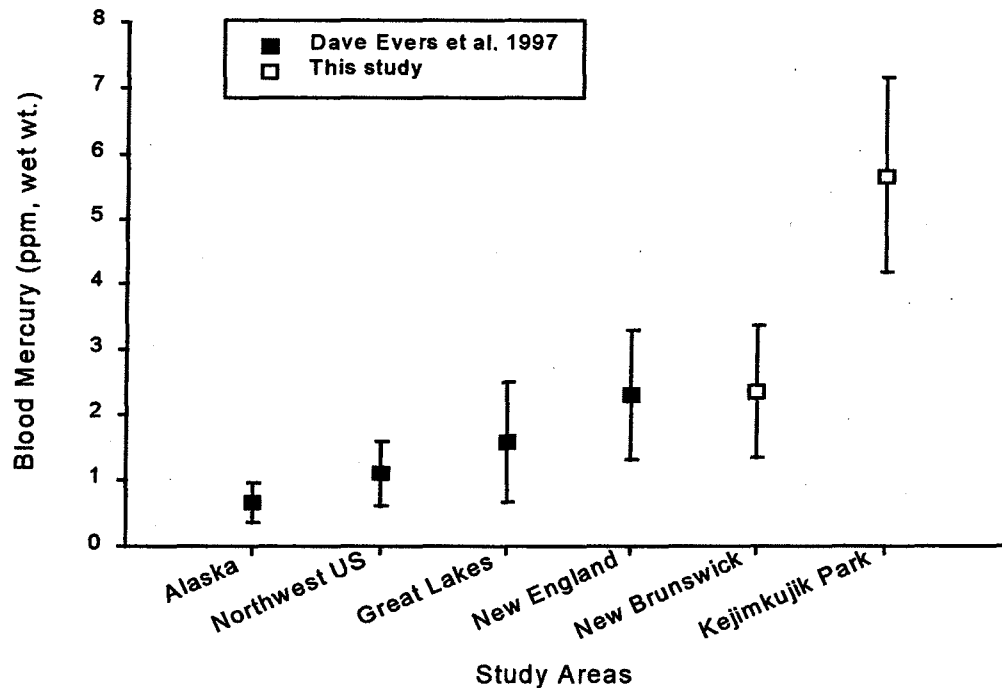


Figure 6: Blood mercury levels in adult loons in Kejimikujik National Park compared to levels found in samples collected at other North American sites (blood mercury sample sizes (n) are as follows: Alaska; 14, Northwest US; 6, Great Lakes; 141; New England; 11, Canadian sites (NB,NS); 14).

Although specific actions are being taken and further actions being proposed to reduce the anthropogenic emissions and subsequent inputs of mercury into receiving systems, the dynamics of mercury cycling in the environment in this area are not clearly understood. A more comprehensive knowledge of the role of the atmosphere in mercury input to watersheds and the dynamics of mercury within the terrestrial and aquatic components within these watersheds is necessary. Further understanding of the sources and consequences of natural versus anthropogenic mercury in ecosystems is necessary to determine the feasibility and subsequent efficacy of controls on anthropogenic emissions in various regions throughout North America.

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