TEMPORAL CHANGES OF FISH MERCURY CONCENTRATIONS IN MINING-AFFECTED PINCHI LAKE, BC.

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ABSTRACT

The Pinchi Mine produced metallic mercury from 1940 to 1944 (historical) and from 1968 to 1975 (modern operation). From 2010 – 2012, the mine underwent decommissioning and reclamation to ensure that terrestrial areas affected by the mine do not pose unacceptable risks to ecological resources. Historical operations included placement of roasted ores (calcines) in the lake opposite the old mill. This resulted in highly elevated mercury concentrations in nearshore sediments. This source, as well as broad aerial deposition of elemental mercury during the roasting process in both operations, increased the mercury load to sediments throughout the lake. Prior to 2000, there were limited data on mercury in fish from Pinchi Lake. Detailed fish mercury studies were conducted in 2000, 2006 and 2011, focusing on mercury-size relationships of lake trout (*Salvelinus namaycush*), whitefish (*Coregonus* sp.) and rainbow trout (*Oncorhynchus mykiss*). While fish mercury concentrations have declined by nearly an order of magnitude from their peak in 1974, the rate of decrease in mercury concentrations has slowed since 2000. The focus of temporal comparisons is based on length-standardized fish size, because of the positive correlation between mercury concentration and size. Overall, mercury concentration in fish have declined from their peaks in the late 1970s (>5 ppm), but remain elevated in Pinchi relative to nearby Stuart and Tezzeron lakes in lake trout (0.53 ppm), whitefish (0.25 ppm) and rainbow trout (0.18 ppm). Given the longevity and size of lake trout and slow burial rate of historically contaminated sediments, the timeframe for recovery of the lake to 'regional' fish mercury concentrations is unknown. Risk assessments have evaluated potential implications of fish consumption on local wildlife species (eagle, grebe, otter), as well as on human health. Continued monitoring of fish mercury concentrations is part of the risk management plan for the site.

Key Words: Pinchi Mine, Mercury, Fish, Temporal Trends

INTRODUCTION AND BACKGROUND

This paper is one of two papers prepared in 2014 on closure activities at the Pinchi Mine. The other paper is entitled "A Risk Assessment / Risk Management Perspective on Mercury Contaminated Sediments in Mining Affected Pinchi Lake, BC, by G.S. Mann, R.F. Baker and P.J. Allard. (Mann et al. 2014).

Pinchi Mine is located in a heavily wooded wildland setting in central British Columbia (BC) on the north shore of Pinchi Lake, northwest of Fort St. James in the headwaters of the Fraser River system (**Figure 1**). Pinchi Lake is 25 km long and relatively narrow (1.2 km to 3.7 km) with a surface area of 55 km^2. The Ocock and Tsilcoh Rivers are the only named tributary streams entering Pinchi Lake. Pinchi Creek, situated at the southwest end of the lake is the only outflow from Pinchi Lake, discharging to Stuart Lake, and ultimately, the Fraser River.

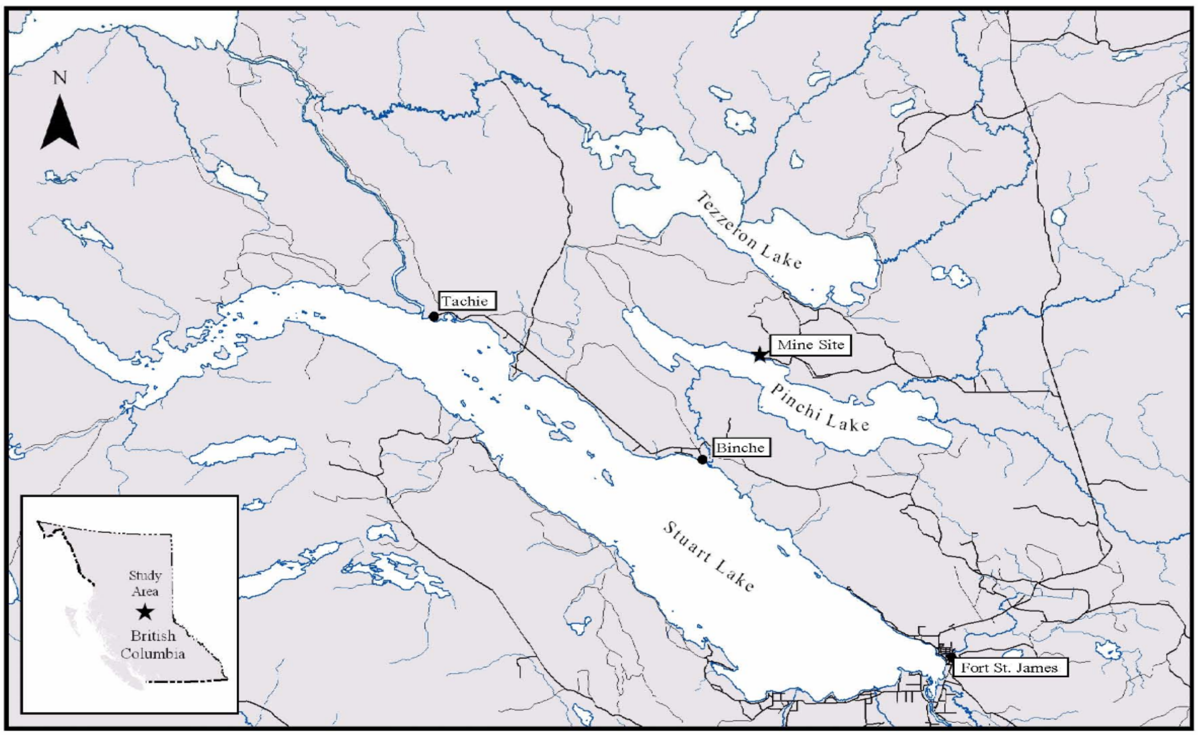

Figure 1. Map of Pinchi Lake region

The Pinchi fault region, located in central BC, is naturally enriched in mercury because of the abundance of cinnabar mineralization in bedrock along several portions of the fault (Plouffe 1995). Predecessor companies (now Teck Metals Ltd.) produced metallic mercury from the Pinchi Mercury Mine from 1940 to 1944 (historic operation) and from 1968 to 1975 (modern operation). During the historic operation, cinnabar ore was roasted without concentration and both the waste rock and the residue from roasting (calcine) were stockpiled or pushed into the lake opposite the old mill. During the modern operation, using a new mill, ore was concentrated and then processed to recover metallic mercury. Residues from this operation were slurried to a large on-site Tailings Impoundment Area. During both operations there was loss of elemental, atmospheric mercury to the environment during the roasting process.

Prior to remediation, the lake foreshore and small on-site ponds that gathered waste products from the mill had already been capped with clean fill and vegetated. Further details on mine operations can be found in Donald and Unger (2013) and in the Mine Closure Plan (Marsland Environmental Associates, 2009). After mining ceased in 1975, the Pinchi Mine was placed under long-term care and maintenance and has been the subject of numerous aquatic and terrestrial investigations. Aquatic studies focused on mercury and methylmercury in surface water, plankton, and sediment (EVS et al. 1999), sediment toxicity and benthic community health (Baker and Mann 2002) and fish (Baker et al. 2001; Azimuth 2008; Azimuth 2013).

Mercury has no known biological function and is a known toxicant to wildlife and to humans. Unlike other metals, mercury is accumulated in the tissues of animals at concentrations higher than in the environment (bioaccumulation) and these concentrations become progressively higher moving up the food chain from invertebrates, to peak in fish and fish-eating animals (biomagnification). Methylmercury, the dominant form of mercury that is present in fish (95%; Bloom 1992), is highest in species that consume other fish (e.g., lake trout, burbot) compared to those fish that eat primarily plankton (e.g., lake whitefish, kokanee). Methylmercury is acquired almost exclusively via dietary means (Hall et al. 1997) and exposure via other pathways such as through water, is very small.

Mercury is very persistent in the environment and can represent a potential human health risk in the event of long-term exposure to elevated concentrations. The process of methylation of inorganic mercury occurs principally in aquatic environments, in sediments of wetlands, marshes and lakes. In Pinchi Lake, the greater abundance of inorganic mercury has resulted in greater methylmercury production and bioaccumulation by fish. While fish is a very healthy and nutritious food source, consuming too many fish of certain species can pose a potential risk. However, as with any substance, the ‘dose’ determines the level of exposure. A human health risk assessment (Wilson 2010) did not identify health risks from consumption of most Pinchi Lake fish. The only exception was for large lake trout and burbot, where frequent and routine consumption would potentially pose a risk.

OBJECTIVES

Fish is the major source of methylmercury exposure to humans as well as to fish-eating wildlife such as mink, otter, eagles and other predators. The amount of mercury in the top predator fish, such as lake trout or burbot, is very much dependent on mercury concentrations in their diet, especially other fish species like whitefish, suckers, or kokanee. Consequently, the study design for fish mercury in more recent investigations conducted by Azimuth (2008, 2013) has focused on measuring meristics (length, weight, age), other life history information (growth, diet, maturity, gender) and stable isotopes, to better understand food web relationships from a wide range of fish species, from the top of the food chain to the bottom.

Key objectives of the 2006 (Azimuth 2008) and 2011 (Azimuth 2013) studies were as follows:

1. Characterize basic limnology, water and sediment quality of Pinchi Lake and place into context relative to two nearby lakes: Stuart and Tezzeron lakes.
2. Characterize key components of the Pinchi Lake aquatic ecosystem (phytoplankton, zooplankton, benthos and fish).
3. Determine mercury accumulation patterns and food web relationships using stable isotopes across fish species in Pinchi Lake and place into context with Stuart and Tezzeron lakes.
4. Determine historical changes and temporal trends in fish tissue mercury concentrations in Pinchi Lake.

This paper focuses on the last objective, how fish mercury concentrations in Pinchi Lake have changed over time. While there has been a significant reduction in mercury concentrations in Pinchi Lake fish since active mining ceased, concentrations remain elevated relative to nearby lakes; this paper speculates on the time course to recovery and puts fish mercury concentrations from Pinchi Lake in perspective other lakes with Tezzeron and Stuart lakes and other lakes in Canada (DePew et al. 2013).

METHODS

During fish mercury surveys conducted since 2000, mercury concentrations have been measured in up to 10 fish species from Pinchi, Stuart and Tezzeron lakes, including lake trout (*Salvelinus namaycush*), lake whitefish (*Coregonus clupeaformis*), mountain whitefish (*Prosopium williamsoni*), rainbow trout (*Oncorhynchus mykiss*), burbot (ling; *Lota lota*) and five other species. Prior to this, surveys were opportunistic and did not target specific species or investigate size-mercury relationships. In each of the 2000, 2006 and 2011 fish surveys (Azimuth 2013), fish were captured using short-set gill nets or by angling with a focus on three key species: lake trout a top level predator and a targeted sport fishing species; lake whitefish, benthic feeders and key food chain species; and rainbow trout, an insectivore and sport species. Tissue samples were harvested non-destructively from live lake and rainbow trout using biopsy tools (Baker et al. 2004) or destructively from whitefish and analysed for total mercury concentration by ALS Laboratories, Burnaby.

There is a well-known positive relationship between increasing mercury concentration and fish length (or weight or age) (Bodaly et al. 1984; Somers and Jackson 1993), as larger, older fish tend to have higher mercury concentrations than smaller, younger fish. This is partly due to differences in diet and the length of time of exposure. This positive relationship is typically seen for strongly carnivorous species (e.g., bull trout, lake trout, walleye) and sometimes for whitefish but seldom for suckers, forage fish or fish that consume terrestrial insects such as rainbow trout. Consequently, it is necessary to collect tissue from target species over a wide size range to determine the species-specific, size-mercury relationship. To accurately represent mercury concentrations across the size range typically observed, up to 30 – 35 fish are required, stratified (5 – 7 fish) within several discrete size intervals (e.g., 201 – 300 mm; 301 – 400 mm). Using linear regression on log-transformed data, a 'size-adjusted' mercury concentration is derived for lake trout (550 mm), lake whitefish (350 mm) and rainbow trout (300 mm) at a standardized length, specific to each species. The standardized size approximates the mean length of fish typically targeted by fishermen and allows for comparisons of mercury concentrations over time or between waterbodies for the same species that are unbiased by differences in fish size of the population sampled. Appropriate statistical methods are used (e.g., analysis of covariance, Tukey's test) to determine whether significant changes in mercury concentration have occurred over time, within or between lakes.

RESULTS AND DISCUSSION

The earliest documented collection of fish from Pinchi Lake for mercury analysis occurred in 1969 (Peterson et al. 1970), shortly after the Pinchi Mine resumed operations. Several further surveys were conducted but did not target specific species, use standardized methods, or attempt to determine size-mercury relationships or changes in fish mercury concentrations over time in a systematic way. In 2000, the first dedicated study of fish mercury concentrations in key fish species of Pinchi Lake using size-standardized techniques and analytical procedures was conducted (Baker et al. 2001). Following is a brief chronology of fish mercury investigations of Pinchi Lake.

- BC Fish and Wildlife (Peterson et al. 1970) surveyed heavy metals in sportfish from BC lakes in 1969/70, including several from the Pinchi Fault region. Lake trout from Pinchi Lake stood out, with elevated muscle tissue mercury concentrations, (> 4 mg/kg or ppm wet weight). These fish were collected three decades after cessation of historic operations and shortly after commencement of the modern operation.
- BC Fish and Wildlife (Reid and Morley 1975) followed up this study in 1974, several years later. The arithmetic mean mercury concentration of 11 lake trout was 5.2 ppm, with a concentration of >8 ppm in a large fish.
- BC Ministry of Environment (Martin et al. 1995) collected 16 lake trout in 1986, a decade after the termination of modern operations. They reported a substantial decrease in mean lake trout mercury tissue concentration (1.1 ppm).
- EVS (1996) collected 10 lake trout in 1995, approximately two decades after termination of mine operations. The arithmetic mean concentration was 0.96 ppm. The trend towards decreased mercury concentrations in lake trout that was observed between 1974 and 1986 was not detected in 1995.
- Baker et al. (2001) undertook a survey of fish mercury of six regional lakes in 2000 (Pinchi, Stuart, Tezzeron, Trembleur, Tchentlo, Francois) using biopsy techniques and a size-standardized approach. The objective was to understand spatial trends in fish mercury concentrations, explore potential "recovery concentrations" and to put the influence of Pinchi Fault geology into perspective. Pinchi Lake fish mercury concentrations were

elevated (~2-3x) relative to other Pinchi Fault lakes, which were not significantly more elevated than fish from Francois Lake, a reference lake.

- In 2006, Azimuth (2008) conducted a survey of Pinchi, Tezzeron and Stuart lakes to understand mercury dynamics and trophic relationships (based on stable isotope analysis), targeting phytoplankton, zooplankton, benthic invertebrates and fish. Similar to previous studies, tissue mercury concentrations in lake trout and other species from Pinchi Lake were elevated relative to Tezzeron and Stuart lakes. It was determined that elevated mercury tissue concentrations in Pinchi Lake are due to mine-related contamination and not to regional (i.e., Pinchi Fault background mercury) or ecological (i.e., longer food chain) factors. While lake trout continued the slow reductions observed since 1986, mercury concentrations in whitefish appeared to drop considerably. This was the first year that mercury in rainbow trout was examined. A positive size-mercury relationship for rainbow trout was established with a size-adjusted mean of 0.14 ppm for a 350 mm fish.
- A Pinchi-only fish survey was conducted in 2011 by Azimuth (2013) with a focus on mercury concentrations and stable isotopes in tissue samples from lake trout, whitefish and rainbow trout. Size-standardized mercury concentrations of lake trout were significantly lower, while mercury in lake whitefish was significantly higher than in 2006, the opposite trend as observed in 2006. Mean mercury in rainbow trout in 2011 (0.19 ppm) was slightly higher than in 2006.

This section describes our understanding of temporal changes in mercury concentrations of lake trout, lake whitefish and rainbow trout in Pinchi Lake since detailed, size-adjusted investigations of mercury concentrations began in 2000, in perspective with historic (pre-2000) studies. **Figure 2** illustrates all mercury (mg/kg ww or ppm) versus length (mm) data for lake trout (LKTR), lake (LKWH) and mountain whitefish (MNWH) and rainbow trout (RNTR) collected between 1974 and 2011. In 2006 there was a clear positive correlation between increasing fish size (length) and mercury concentration for lake trout and whitefish, as well as in rainbow trout – an atypical result for this species in most lakes. There has also been a clear reduction in mercury concentration in lake trout and lake whitefish over time, especially since 1974. There were too few data from 2006 and 2011 to make clear inferences for mountain whitefish and rainbow trout.

Although not illustrated here, mercury concentrations of these species from Pinchi Lake were significantly higher than their counterparts from Tezzeron and Stuart lakes, on a size-adjusted basis, from the most recent (2006) data (Azimuth 2008). Mercury concentration for a 550 mm lake trout from Pinchi Lake (0.91 ppm) was significantly higher ($p<0.01$) than mercury in lake trout from Stuart (0.39 ppm) and Tezzeron lakes (0.26 ppm). Mercury in 350 mm lake whitefish from Pinchi Lake (0.26 ppm) was also significantly higher than in whitefish from Stuart (0.10 ppm) and Tezzeron (0.09 ppm) lakes, although the magnitude of difference was lower. The highest mercury concentration measured in Pinchi Lake was from burbot with a concentration of 1.16 ppm for a 600 mm fish. This was much higher than for burbot from Tezzeron Lake (0.08 ppm) and Stuart Lake (0.17 ppm) on a size-adjusted basis.

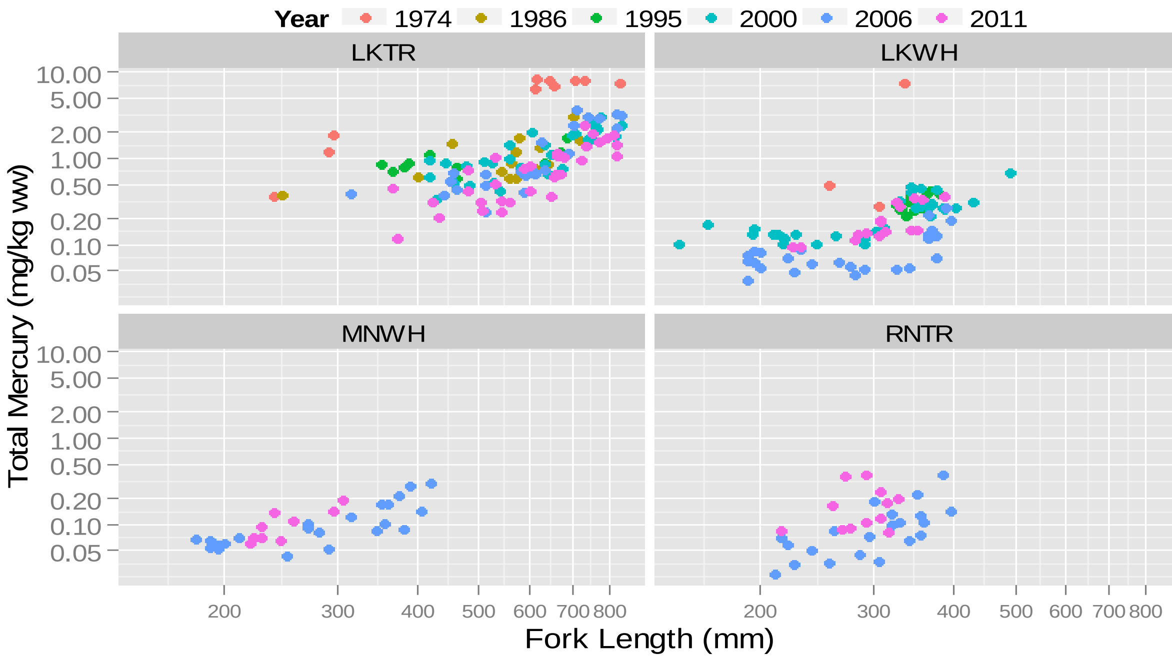

Figure 2. Log_{10} mercury (ppm) on length (mm) relationships for Pinchi Lake fish, 1974 - 2011.

Results from the most recent 2011 survey (Azimuth 2013) are illustrated in **Table 1** for the above species as well as kokanee, burbot, longnose sucker and white sucker. The relationship between increasing fish size and mercury concentration was only significant for lake trout and the whitefish species and not for rainbow trout, which was consistent with other years and lakes (Azimuth, 2008). White sucker mercury concentration also increased significantly with increasing fish size. This table also illustrates the size-adjusted mercury concentration for each species, based on the regression equations in this table. The mercury concentration for a 550 mm lake trout was 0.53 ppm, 0.25 ppm for a 350 mm lake whitefish and 0.19 ppm rainbow trout (independent of fish size).

Table 1. Fish mercury (Hg) on length, age, and weight regression results and standardized mercury concentrations for Pinchi Lake, 2011.

Species	Sample Size	R^2	p-Value	Significant Relationship	Mercury (Hg) on Length (L), Age (A), or Weight (W) Relationship	Standardized Measures[1]	
						Length or Weight	[Mercury] (mg/kg ww)
Hg-Length Relationships						(mm)	
Lake trout	32	0.65	<0.001	yes	$Log_{10}Hg = -7.65 + 2.69 (Log_{10}L)$	550	0.53
Lake whitefish	16	0.62	<0.001	yes	$Log_{10}Hg = -6.94 + 2.49 (Log_{10}L)$	350	0.25
Mountain whitefish	9	0.68	0.006	yes	$Log_{10}Hg = -7.69 + 2.80 (Log_{10}L)$	300	0.18
Rainbow trout	12	0.07	0.412	no	$Log_{10}Hg = -3.87 + 1.24 (Log_{10}L)$	350	0.19
Kokanee	3	0.89	0.216	no	$Log_{10}Hg = -4.38 + 1.58 (Log_{10}L)$	250	0.26
Burbot	2	1	IS	IS	IS	600	IS
Longnose sucker	1	0	IS	IS	IS	350	IS
White sucker	10	0.55	0.014	yes	$Log_{10}Hg = -5.45 + 1.92 (Log_{10}L)$	350	0.27

1 Using standardized length; IS = insignificant relationship

Figure 3 and **Figure 4** illustrate all mercury-length data collected since 1974 for lake trout and lake whitefish respectively, but with statistically derived log-transformed regressions for 2000 (green line), 2006 (blue line) and 2011 (pink line) based on analysis of covariance, where a common slope between years was derived. For lake trout (**Figure 3**), the magnitude of difference in the intercept of the slopes between 2000 and 2006 was small and not significant (p>0.05). However, the 2006 distribution and size-standardized mean (0.53 ppm) was significantly lower than in 2000 (0.91 ppm) and 2006 (0.81 ppm) (**Table 2**).

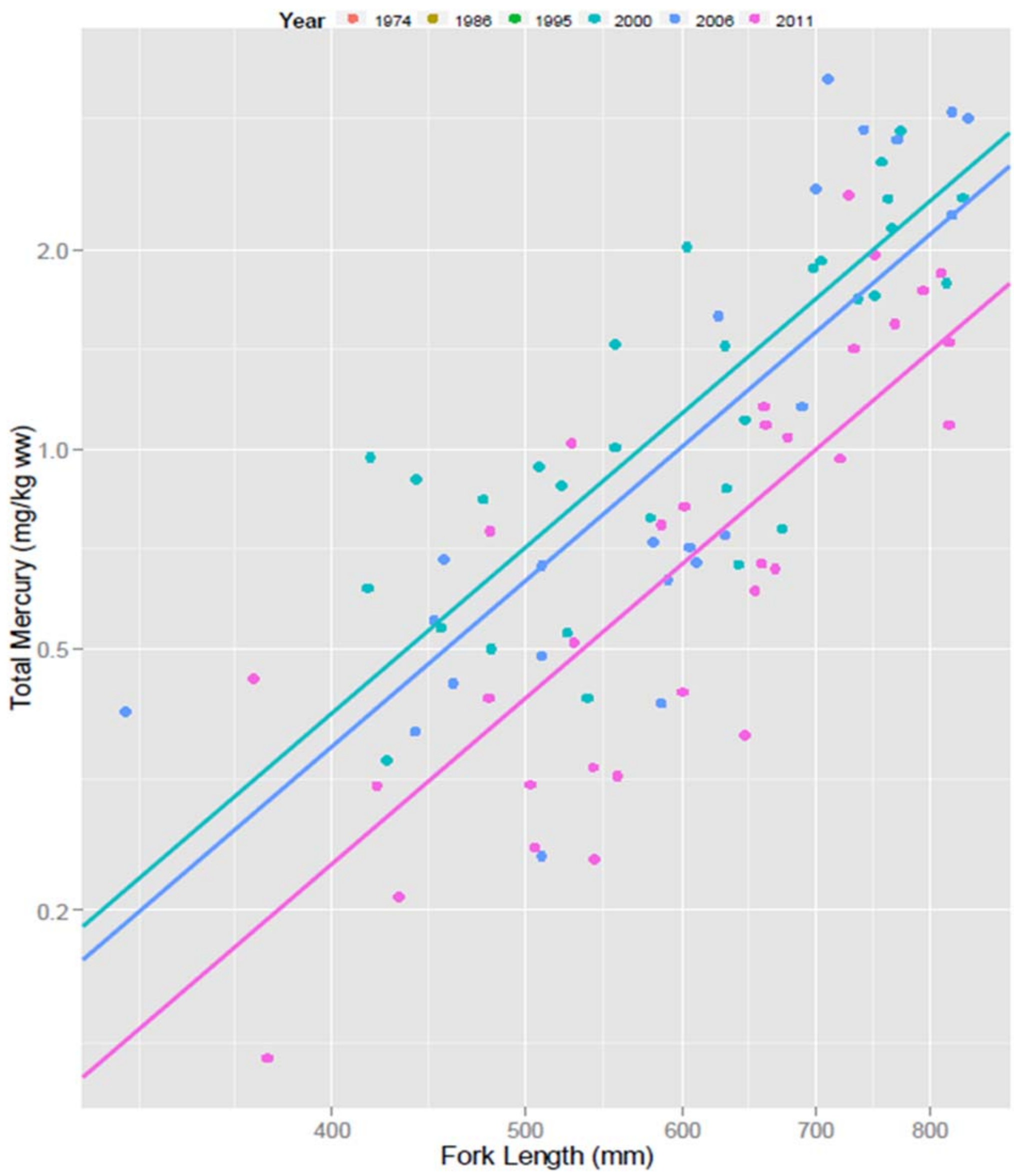

Figure 3. Lake trout ANCOVA-based (assuming equal slopes) on $\log_{10}$(mercury) vs $\log_{10}$(length) model for Pinchi Lake for 2000, 2006, and 2011.

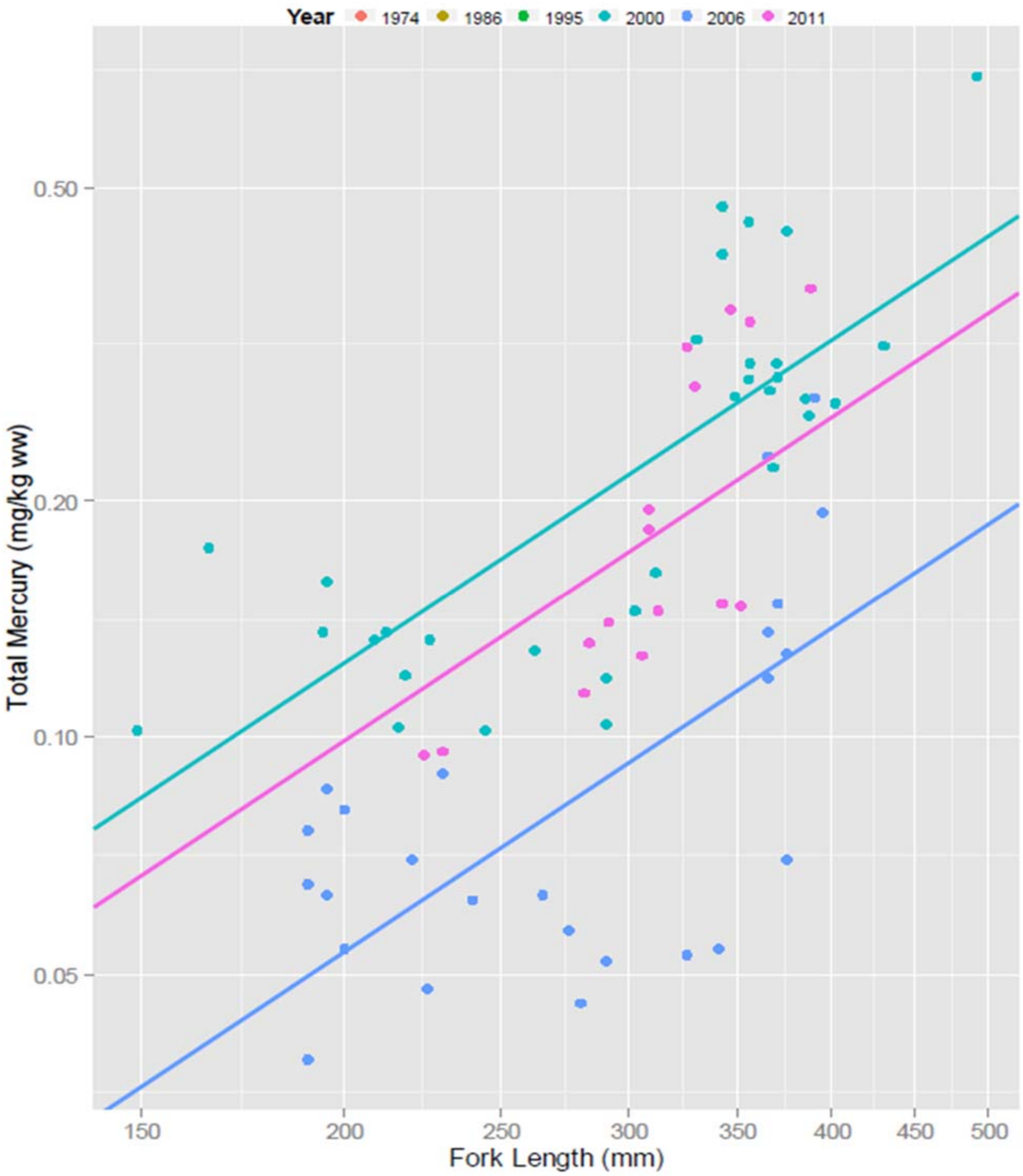

Figure 4. Lake whitefish ANCOVA-based (assuming equal slopes) on $\log_{10}$(mercury) vs $\log_{10}$(length) model for Pinchi Lake for 2000, 2006, and 2011.

The trend for lake whitefish was much more variable over time (**Table 2**). Size-standardized mean mercury concentration (350 mm) in 2000 was 0.26 ppm which was significantly higher than in 2006 (0.11 ppm) but not 2011 (0.25 ppm). The difference between 2006 and 2011 data might be related to small sample size in 2011, or other factors that might have contributed to a real increase in mercury in this species, as described below.

Table 2. Comparison of results of standardized tissue mercury (Hg) concentrations for lake trout and lake whitefish from Pinchi Lake.

Year	Sample Size	Mercury (Hg) on Length (L) Relationship $Log_{10}Hg = a + b (Log_{10}L)$	R^2	p-Value	Significant Relationship	Standardized Measures[1]	
						Length (mm)	[Mercury] (mg/kg ww)
Lake trout							
1974	9	$Log_{10}Hg = -4.25 + 1.81 (Log_{10}L)$	0.92	<0.001	yes	550	5.13
1986	16	$Log_{10}Hg = -3.31 + 1.19 (Log_{10}L)$	0.29	0.030	yes	550	0.89
1995	10	$Log_{10}Hg = -1.90 + 0.70 (Log_{10}L)$	0.37	0.064	no	550	1.04
2000	31	$Log_{10}Hg = -6.04 + 2.19 (Log_{10}L)$	0.62	<0.001	yes	550	0.91
2006	23	$Log_{10}Hg = -7.90 + 2.85 (Log_{10}L)$	0.68	<0.001	yes	550	0.81
2011	32	$Log_{10}Hg = -7.65 + 2.69 (Log_{10}L)$	0.65	<0.001	yes	550	0.53
Lake whitefish							
1986	6	$Log_{10}Hg = -7.99 + 2.99 (Log_{10}L)$	0.79	0.018	yes	350	0.41
1995	12	$Log_{10}Hg = -8.47 + 3.13 (Log_{10}L)$	0.49	0.011	yes	350	0.31
2000	32	$Log_{10}Hg = -4.09 + 1.38 (Log_{10}L)$	0.60	<0.001	yes	350	0.26
2006	25	$Log_{10}Hg = -3.89 + 1.15 (Log_{10}L)$	0.37	0.001	yes	350	0.11
2011	16	$Log_{10}Hg = -6.94 + 2.49 (Log_{10}L)$	0.62	<0.001	yes	350	0.25

SUMMARY AND CONCLUSIONS

The historical trend observed, especially for lake trout, suggests a two-phased recovery response in fish mercury concentration. The initial phase, between 1974 and 1986 would have seen a significant reduction in mine-related mercury-containing discharges to the lake. Subsequent reductions in total and methylmercury concentrations in the water column would have likely occurred rapidly and cascaded up to phytoplankton and zooplankton, as well as to any fish feeding on these organisms (e.g., lake whitefish).

The secondary phase, which continues today, is characterized by more modest reductions in tissue mercury concentrations likely due to population attrition and deposition of mercury into deeper sediments (EVS et al. 1999; Mann et al. 2014). Over the decades following mine shutdown, older, larger lake trout would have naturally died and been replaced in the population by fish exposed to progressively lower mercury during their lifetime. Concurrently, sediment coring programs (EVS et al. 1999) documented a slow burial process where deeper sediments generally act as a mercury sink. The slow reductions in mercury concentrations in biologically active surface sediments would drive lower tissue mercury concentrations in benthic organisms, which would gradually be reflected in the food chain (e.g., to juvenile lake trout and other fish relying heavily on aquatic insect larvae and ultimately to adult lake trout).

The lack of significant inputs of relatively cleaner sediments to Pinchi Lake (i.e., no large tributaries) is probably the greatest rate-limiting factor now affecting the secondary phase. However, the reduction in tissue mercury concentrations observed in smaller lake trout between 2000 and 2006 foreshadowed a future trend in the larger lake trout. The apparent drop in zooplankton mercury concentrations between 1997 and 2006 also supports this hypothesis. While these results suggest continued, but slow, reductions in the amount of mercury actively cycling through the ecosystem, verification of this hypothesis will require continued monitoring.

The 2011 results for both lake trout and lake whitefish were unexpected. For lake trout, the magnitude of the reduction in tissue concentrations relative to 2006 was greater than expected, even considering the foreshadowing we speculated about. One possible difference between the 2011 and 2006 studies was the use of non-lethal biopsy sampling procedures for total metals in 2011 (i.e., not just limited to mercury). However, this method has been shown to be accurate (Baker et al., 2004) and there was no apparent bias in the 2011 data due to sampling type. Lake whitefish on the other hand (based on a sample size of only 16 fish) showed the opposite pattern, with a reasonably large increase in tissue mercury concentrations in 2011 relative to 2006. These results suggest that the small sample size of fish did not represent the true underlying population, there are spatial differences within the lake that confound results, and/or that there was a real increase in mercury for this species, such as can be caused by logging and forest fires (Garcia and Carnignan 2000), activities that have been common in this area recently. Logging, fires and death of forests from pine beetle are known to cause soils to erode and become deposited in lakes as sediment, transporting organic material and mercury into lakes, where it is methylated. As well, readers should be cautious about over-interpreting the results of a single survey and should instead focus on broader temporal trends.

Notwithstanding the uncertainty in some data driven by small and uneven sample size distribution between years, mercury concentrations in key fish species in Pinchi Lake have been slowly declining since mining ceased in 1976. Despite the fact that mercury concentrations in fish are elevated relative to other BC lakes and reservoirs (Baker 1999), these concentrations are similar to what is observed in many hundreds of lakes elsewhere in Canada (DePew et al. 2014). This is likely why ecological effects to local fish-eating wildlife species have not been observed. For example, Weech et al. (2006) found that reproductive success and average productivity of bald eagles over the 3-year period (2000 – 2002) were 62% and 0.98 chicks/territory on Pinchi Lake compared to 64% and 1.17 chicks/territory on all other study lakes combined. While exposure to mercury was higher on Pinchi Lake (blood, feathers, eggs), birds were in excellent condition and successfully raised eaglets. An ecological risk assessment conducted by Azimuth (2009) did not predict unacceptable risks to fish-eating wildlife such as loons, grebes and otter.

In summary, given the strongly bioaccumulative nature of methylmercury, long lifespan (>30 y) and large size of lake trout, and slow deposition and burial rate of historically contaminated lake sediments (Mann et al. 2014), it is expected that recovery of the fish mercury concentrations in Pinchi Lake to ‘regional’ (i.e., lakes in close proximity to the mercury-rich Pinchi Fault) fish mercury concentrations, may take several decades, but may also remain higher than regional lakes, given possible influence of the Pinchi Fault and lake-wide elevations in mercury concentrations from historic mining operations (Mann et al. 2014).

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