

Report prepared for Wrangell-St. Elias National Park and Preserve

Mercury in Fishes from Wrangell-St. Elias National Park and Preserve, Alaska

Open-File Report 2014–1145

U.S. Department of the Interior
U.S. Geological Survey

Cover: Photograph of Mount Drum, Wrangell-St. Elias National Park and Preserve, 2007.
Photograph taken from the Tok Cutoff by J. Willacker, U.S. Geological Survey.

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By Brandon M. Kowalski, James J. Willacker, Jr., Christian E. Zimmerman, and Collin A. Eagles-Smith

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**U.S. Department of the Interior
U.S. Geological Survey**

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Conversion Factors and Datum

SI to Inch/Pound

Multiply	By	To obtain
Length		
millimeter (mm)	0.03937	inch (in.)
meter (m)	3.281	foot (ft)
kilometer (km)	0.6214	mile (mi)
Area		
acre	0.0015625	square mile (mi ²)
Mass		
gram (g)	0.03527	ounce, avoirdupois (oz)

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:

$$^{\circ}\text{F} = (1.8 \times ^{\circ}\text{C}) + 32.$$

Datum

Vertical coordinate information is referenced to the North American Vertical Datum of 1988 (NAVD 88).

Elevation, as used in this report, refers to distance above the vertical datum.

Abbreviations and Acronyms

AIC	Akaike's Information Criterion
AK-DHSS	Alaska Department of Health and Social Services
ANCOVA	Analysis of Covariance
dw	dry weight
EPA	U.S. Environmental Protection Agency
Hg	mercury
LOER	lowest-observed-effects-residue
MeHg	methylmercury
n	sample size
NOER	no-observed-effects-residue
R²	coefficient of determination
RSD	percent relative standard deviation
SE	standard error
THg	total mercury
WRST	Wrangell-St. Elias National Park and Preserve
ww	wet weight
yr	year or years

Mercury in Fishes from Wrangell-St. Elias National Park and Preserve, Alaska

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Abstract

In this study, mercury (Hg) concentrations were examined in fishes from Wrangell-St. Elias National Park and Preserve, Alaska, the largest and one of the most remote units in the national park system. The goals of the study were to (1) examine the distribution of Hg in select lakes of Wrangell-St. Elias National Park and Preserve; (2) evaluate the differences in Hg concentrations among fish species and with fish age and size; and (3) assess the potential ecological risks of Hg to park fishes, wildlife, and human consumers by comparing Hg concentrations to a series of risk benchmarks. Total Hg concentrations ranged from 17.9 to 616.4 nanograms per gram wet weight (ng/g ww), with a mean (\pm standard error) of 180.0 ± 17.9 across the 83 individuals sampled. Without accounting for the effects of size, Hg concentrations varied by a factor of 10.9 across sites and species. After accounting for the effects of size, Hg concentrations were even more variable, differing by a factor of as much as 13.2 within a single species sampled from two lakes. Such inter-site variation suggests that site characteristics play an important role in determining fish Hg concentrations and that more intensive sampling may be necessary to adequately characterize Hg contamination in the park. Size-normalized Hg concentrations also differed among three species sampled from Tanada Lake, and Hg concentrations were strongly correlated with age. Furthermore, potential risks to park fish, wildlife, and human users were variable across lakes and species. Although no fish from two of the lakes studied (Grizzly Lake and Summit Lake) had Hg concentrations exceeding any of the benchmarks used, concentrations in Copper Lake and Tanada Lake exceeded conservative benchmarks for bird (90 ng/g ww in whole-body) and human (150 ng/g ww in muscle) consumption. In Tanada Lake, concentrations in most fishes also exceeded benchmarks for risk to moderate- and low-sensitivity avian consumers (180 and 270 ng/g ww in whole-body, respectively), as well as the concentration at which Alaska State guidelines suggest at-risk groups limit fish consumption to 3 meals per week (320 ng/g). However, the relationship between Hg concentrations and fish size in Tanada Lake suggests that consumption of smaller-sized fishes could reduce Hg exposure in human consumers.

Introduction

Contamination of aquatic ecosystems with mercury (Hg) is a widespread threat, affecting fish and wildlife, and posing risks for humans that consume fish (Scheuhammer and others, 2007). Atmospheric deposition of anthropogenic Hg emitted from fossil fuel combustion is among the primary pathways into aquatic ecosystems, although there also are many natural sources (for example, volcanoes and forest fires) that contribute to global background of Hg contamination (Driscoll and others, 2013). Within aquatic ecosystems, natural processes convert inorganic Hg to methylmercury (MeHg), a form of Hg that biomagnifies in aquatic food webs (Lavoie and others, 2013) and is highly toxic to fish and wildlife (Evers and others, 2007; Scheuhammer and others, 2007; Schwindt and others, 2008).

The widespread distribution of atmospheric Hg places even remote environments at risk for Hg contamination. In particular, national parks in the United States often are remote and among the most protected public lands, yet are affected by anthropogenic Hg (Landers and others, 2008; Eagles-Smith and others, 2014). As a result, it is important to understand the extent and environmental consequences of Hg contamination in national parks. Moreover, Hg contamination has important implications for consumption of fish by subsistence users and park visitors who fish regularly in contaminated waters.

Wrangell-St. Elias National Park and Preserve (WRST) is an Alaska National Interest Lands Conservation Act national park, and as such, it provides access to subsistence harvest for many rural residents. A recent study of Hg in fish from 21 western national parks documented that some of the highest Hg concentrations were measured in fish from WRST (Eagles-Smith and others, 2014), with geometric mean values exceeding health criteria for the protection of fish-eating birds, mammals, and humans. However, it is unclear whether elevated Hg concentrations in fish from WRST are due to biological characteristics of the fish sampled (that is, age, size, species, trophic status, etc.) compared to local factors that enhance the availability of Hg within these food webs. Therefore, in order to better understand the distribution of Hg in WRST and the associated risk to aquatic communities and park visitors, Hg concentrations were measured in several fish species from four lakes in WRST, three of which were sampled previously by Eagles-Smith and others (2014). The primary objectives were to:

1. Determine the distribution of Hg concentrations in fishes among four lakes in WRST;
2. Evaluate the differences in fish Hg concentrations among fish species, and with fish age and size; and
3. Assess toxicological risk within the park by comparing Hg concentrations in fishes to a series of eight previously developed toxicological benchmarks for fish, fish-eating birds, and humans.

Methods

Study Sites and Sample Collection

Wrangell-St. Elias National Park and Preserve is the largest national park in the United States, spanning about 13.2 million acres and bounded on the south by the Gulf of Alaska coast (fig. 1). The park was established in 1980 and is managed to protect ecological integrity and heritage resources of south-central Alaska, while providing public use in a wild setting (Hood and others, 2006). The climate in the park is variable and extreme. Winter low temperatures can reach -40°C and summer high temperatures can reach 33°C (Sousanes, 2012). Wrangell-St. Elias National Park and Preserve contains more than 96,000 acres of lakes and ponds, and more than 20,000 km of flowing waters. The park is divided into six major watersheds—the Bremner River, Chitina River, Copper River, Nabesna River, and White River drainages—and coastal streams (Weeks, 2003). The Copper River is the major river in the region, and the Bremner and Chitina Rivers flow westward into it and ultimately to the Gulf of Alaska. In contrast, the Nabesna and White Rivers are part of the Yukon River watershed, which drains into the Bering Sea.



Figure 1. Schematic of four study lakes in Wrangell-St. Elias National Park and Preserve, Alaska.

This study focuses on fish collected from four lakes in WRST (fig. 1; table 1). Tanada Lake is a large (2,718 acres), deep (53 m) lake in the Copper River headwaters at an elevation of 880 m. Copper Lake, also is in the Copper River headwaters, 3 km west of Tanada Lake. Copper Lake has a surface area of 1,803 acres, with a maximum depth of 72 m, and is at an elevation of 886 m. Both Tanada lake and Copper lake contain burbot (*Lota lota*), lake trout (*Salvelinus namaycush*), Arctic grayling (*Thymallus arcticus*), round whitefish (*Prosopium cylindraceum*), longnose sucker (*Catostomus catostomus*), and sockeye salmon (*Oncorhynchus nerka*), which provide subsistence and recreational fishing opportunities (Weeks, 2003; Schwanke and McCormick, 2010; Kukkonen and Zimpelman, 2012). Copper Lake also has non-anadromous kokanee (*Oncorhynchus nerka*). Much less information is available for Grizzly and Summit Lakes. Grizzly Lake is at an elevation of 1,110 m within the Nabesna River drainage, and has a surface area of 252 acres and a maximum depth of 10 m. Grizzly Lake is known to contain burbot and Arctic grayling. Summit Lake is about 130 km southwest of Tanada Lake at an elevation of 860 m in the Chitna River drainage of the Chugach Mountains, and contains introduced rainbow trout (*Oncorhynchus mykiss*).

National Park Service and Alaska Department of Fish and Game personnel collected fish from each of these four lakes in 2013. Fish were sampled using hoop nets, fyke nets, and hook and line. Four species of fish—Arctic grayling, burbot, lake trout, and rainbow trout—were collected. However, not all species were collected at all sites (table 1). After collection, fish were euthanized, placed on ice, measured (total length), wrapped in aluminum foil, individually labeled, frozen, and shipped to the laboratory for Hg analysis. After arrival at the laboratory, fish were stored at -20° C until dissection and processing.

Table 1. Site locations and sample information for sites sampled in Wrangell-St. Elias National Park and Preserve, Alaska.

[SR: Size regression used—Y, yes; N, no]

Site and species	Coordinates			Number	Standard length (millimeters)		SR
	Latitude	Longitude	Elevation (meters)		Mean	Range	
Copper Lake lake trout	62.425	-143.555	886	15	378	297-454	N
Grizzly Lake burbot	62.222	-143.365	1110	15	563	505-615	N
Summit Lake rainbow trout	61.315	-144.191	860	15	309	200-388	N
Tanada Lake Arctic grayling	62.425	-143.371	880	10	301	250-355	Y
burbot				13	483	345-565	Y
lake trout				15	413	330-470	Y

Sample Preparation

In the laboratory, we thawed fish at room temperature and then measured standard length (SL) to the nearest millimeter on a fish board, and mass to the nearest gram with a digital hand-held fish scale. Equations for converting standard length to total length for each species are provided in appendix 1. We excised 5–10 g of skinless axial muscle just anterior to the dorsal fin using acid-rinsed stainless-steel scalpels. After removal, muscle tissue was rinsed with deionized water, blotted dry with a lint-free wipe, and weighed to the nearest 0.0001 g. The muscle tissue was then placed onto a clean aluminum weigh pan and dried in a convection oven at 50° C until a constant mass was achieved (typically 48 hours). Once dried, samples were stored in a desiccator until they cooled to room temperature. Finally, the dried muscle samples were weighed to the nearest 0.0001 g and ground to a fine powder using a ceramic mortar and pestle. Each mortar and pestle was rinsed with deionized water, dilute HCl, and deionized water again between samples to avoid cross contamination

Otolith Removal and Preparation

Left and right sagittal otoliths were removed from each fish in order to estimate fish age. The shape and size of the otoliths varied among species; therefore, we used one of three otolith preparation methods depending on species. Rainbow trout otoliths were mounted flat, with the sulcus facing up on a clear glass microscope slide using cyanoacrylate adhesive. Excess otolith material was removed using 400- and (or) 600- μm grit wet/dry sandpaper followed by polishing with a sequence of progressively finer lapping paper ranging from 30 μm to 1 μm . Burbot otoliths were prepared using a “break and burn” method (Panfili and others, 2002; Committee of Age-Reading Experts, 2006) because they are very large and were difficult to polish using whole-otolith mounting methods. Briefly, burbot otoliths first were scored in the center using a scalpel, cracked in half, burned using a butane lighter, and polished using the sequential sanding-polishing methods described above. Finally, lake trout and Arctic grayling otoliths were mounted in thermoplastic cement (Crystalbond™ 509, Ted Pella, Inc.), sectioned, and polished following the methods outlined in Black and others (2013) at the U.S. Geological Survey Alaska Science Center. In all cases, polishing was considered complete when the growth increments were clearly visible from the margins to the nucleus, and age was estimated by counting the sequence of transparent and opaque annuli along the plane from the nucleus to the margin. Age estimates were determined independently by two individual readers, and any discrepancies were resolved using a third independent estimate.

Mercury Determination

Total Hg (THg) concentrations were determined in dried axial muscle following U.S. Environmental Protection Agency (EPA) method 7473 (2000) on a Milestone tri-cell DMA-80 Direct Mercury Analyzer® (Milestone Inc., Monroe, Connecticut). Total Hg concentration is an accepted proxy for MeHg concentrations because nearly all Hg in fish muscle is in the MeHg form (Hall and others, 1997). Quality-assurance measures included analysis of three certified reference materials (DORM4, TORT2, or DOLT4; National Research Council of Canada, Ottawa, Canada), two system and method blanks, and two duplicates per batch of 30 samples. Recoveries (\pm standard error [SE]) averaged 97.5 \pm 2.4 percent (n=11) and 101.5 \pm 2.8 percent (n=10) for certified reference materials and calibration checks, respectively. Absolute relative percent differences (RSD) for all duplicates averaged 2.3 \pm 0.7 percent (n=11).

Statistical Analyses

We conducted all statistical analyses of Hg concentrations on a dry weight (dw) basis in order to control for variability in tissue moisture. However, we also calculated wet weight (ww) concentrations using individual-based moisture content and present the data in wet weight to facilitate comparisons with other studies. Moisture content in the fish muscle ranged from 65.1 to 81.4 percent with a mean (\pm SE) of 76.6 ± 0.4 percent.

Because fish size is often correlated with THg concentrations (Wiener and Spry, 1996), we first evaluated site- and species-specific length compared to THg relationships. Where appropriate, we size-normalized THg concentrations of each fish to facilitate comparisons across sites and species. To make this determination, we evaluated the strength of each relationship based on their coefficient of determination (R^2) and Akaike's Information Criterion (AIC) for size compared to THg concentration (\log_{10} -transformed) regression models from each species-site combination. No size normalization was conducted for models with R^2 less than 0.2. For regression models with R^2 greater than 0.2, AIC values were compared between the regression model and a null model. If the size by THg model had an AIC value of 2 units or lower (indicating a meaningful difference in model fit; Burnham and Anderson, 2010), size adjustments were made. We determined the sizes at which THg concentrations were normalized based on the size distribution of fish in each sample and the comparisons being made (see below). Mercury concentrations were size-normalized to median fish lengths by extracting the site- and species-specific regression model coefficients, and adding the individual residuals to the normalized predicted value.

Because we could not directly compare samples across all species and sites due to differences in species collected from each lake, we used a tiered statistical design to assess differences in THg concentrations among species within a lake, as well as among lakes for select species. For the within-lake assessment, Analysis of Covariance (ANCOVA) was used to compare size-normalized (350 mm SL) THg concentrations in Arctic grayling, burbot, and lake trout from Tanada Lake. In order to account for the effects of variable moisture content in the fish while facilitating intuitive interpretation, we converted the normalized THg concentrations (derived using dry weight data) to wet weight concentrations using percent moisture values for each fish and then included percent moisture in the ANCOVA model as a covariate. Thus, statistical results are presented on a wet weight basis, but adjusted for variable moisture content.

Because burbot and lake trout were both collected from more than one lake, ANCOVA was used to examine inter-lake variation in THg concentrations of burbot (from Grizzly and Tanada Lakes) and lake trout (from Copper and Tanada Lakes). Neither burbot from Grizzly Lake nor lake trout from Copper Lake had significant relationships between standard length and THg concentration that necessitated size adjustments of THg concentrations as described above. Conversely, THg increased with increasing standard length in both species from Tanada Lake. Therefore, burbot and lake trout from Tanada Lake were normalized to the median length of the species in Grizzly Lake (burbot, 550 mm SL) and Copper Lake (lake trout, 378 mm SL). Variation in moisture contents of individuals were accounted for by including percent moisture as a covariate in the ANCOVA model as described above.

Toxicological Risk Estimation

In addition to comparing THg concentrations among lakes and species, we also evaluated the potential ecological risk of Hg exposure to fish, fish-eating birds, and humans by comparing THg concentrations to a series of eight previously developed toxicological benchmarks. Mercury concentrations in muscle were converted to whole-body concentrations (Peterson and others, 2005) in order to assess the toxicological risks of Hg to wildlife and fish, whereas muscle Hg concentrations were used to assess the toxicological risks to humans.

To assess potential toxicological effects of Hg to fish, we compared estimates of whole-body THg concentrations to a no-observed-effects-residue (NOER) of 200 ng/g ww (Beckvar and others, 2005) and a lowest-observed-effects-residue (LOER) of 300 ng/g ww (Sandheinrich and others, 2011). Tissue THg concentrations less than the NOER benchmark are not commonly associated with altered behavioral, development, growth, or reproduction of fishes, whereas THg concentrations in fishes greater than the LOER benchmark have been consistently associated with sublethal effects of Hg, including changes in reproductive health (Sandheinrich and others, 2011). For fish-eating birds, whole-body THg concentrations were compared to three toxicological benchmarks representing risks to highly sensitive, moderately sensitive, and relatively insensitive species. Because robust estimates of dietary thresholds exist for only a few species, we used the threshold derived for a well-studied, moderately sensitive species, the common loon (*Gavia immer*; Scheuhammer and others, 2007; Kenow and others, 2008; Mitro and others, 2008; Scheuhammer and others, 2008; Heinz and others, 2009; Kenow and others, 2011), as a baseline. A review of field and laboratory studies on Hg toxicity in common loons found that Hg concentrations greater than 180 ng/g ww in prey fish were associated with significant reductions in reproductive success of wild loons (Depew and others, 2012). We used this concentration as the benchmark for risk to moderately sensitive avian consumers, and evaluated the likely range of risk to more-or-less-sensitive avian consumers by scaling this threshold by ± 50 percent. Thus, risk to avian consumers was assessed by comparing Hg concentrations in whole-body fish to 90 (high sensitivity), 180 (moderate sensitivity), and 270 ng/g ww (low sensitivity) benchmarks. For human health risk, muscle THg concentrations were compared to three fish consumption guidelines recommended by the Alaska Department of Health and Social Services (AK-DHSS) to protect women who are or can become pregnant, nursing mothers, and young children (aged 12 years or younger; Verbrugge, 2007). These guidelines (150, 320, and 640 ng/g ww in fish muscle) correspond to the concentrations at which AK-DHSS recommends at-risk groups limit fish consumption to 4, 3, and 1 meal per week, respectively, and aim to balance the risks and health benefits of fish consumption.

Comparisons were made to these benchmarks using two approaches. First, we compared raw (that is, not size-normalized), whole-body (fish and avian benchmarks) or muscle (human benchmarks) THg concentrations to each of the eight benchmarks and enumerated the proportion of individuals of each species in each lake that exceeded the benchmarks. This approach provides a coarse overview of risk for each species and site, but does not account for the effects of fish size on risk in those populations with size-THg relationships. Therefore, the second approach assessed the proportion of individuals that would exceed each benchmark over the range of standard lengths observed in the sample. To accomplish this comparison, site- and species-specific length-THg relationships were used to model risk at 1-mm size increments over the entire range of lengths observed in the sample. Effectively, all fish in a sample were normalized to the size of the smallest fish, then in 1-mm increments until the size of the largest fish in a sample was reached. At each of these size increments, the proportion of fish exceeding each benchmark was enumerated. Uncertainty in these size-specific risk estimates was preserved by comparing the bounds of a 95-percent prediction interval for each fitted value to the benchmarks. Thus, the resulting graphics show the estimated proportion of fish in a population that would exceed a threshold at a given size and can be used to identify the size of fish at which some threshold of acceptable risk is exceeded.

Results

We analyzed THg concentrations in a total of 83 fish from the four study lakes in 2013—15 lake trout from Copper Lake, 15 burbot from Grizzly Lake, and 15 rainbow trout from Summit Lake, as well as 15 lake trout, 10 Arctic grayling, and 13 burbot from Tanada Lake. Total Hg concentrations ranged from 17.9 to 616.4 ng/g ww across all sites and species, with a mean (\pm SE) THg concentration of 180.0 ± 17.9 ng/g ww. Site- and species-specific mean THg concentrations were highest in burbot (382.5 ± 40.3 ng/g ww) and lake trout from Tanada Lake (371.7 ± 26.8 ng/g ww), followed by lake trout from Copper Lake (144.7 ± 6.7 ng/g ww), Arctic grayling from Tanada Lake (108.6 ± 9.2 ng/g ww), burbot from Grizzly Lake (40.6 ± 4.4 ng/g ww), and rainbow trout from Summit Lake (35.2 ± 3.3 ng/g ww; fig. 2). However, THg concentrations were correlated with fish size in some populations (fig. 3), necessitating size-adjustment of THg concentrations in order to make accurate comparisons. Similarly, some, but not all, populations had significant positive relationships between age and THg concentration (fig. 4). Age generally was a better predictor of fish THg concentrations than length (table 2). Total Hg concentrations in Tanada Lake Arctic grayling ($R^2=0.55$, $P=0.0042$) and lake trout ($R^2=0.72$, $P<0.0001$) were correlated with age, whereas burbot from Tanada ($R^2 = 0.37$, $P = 0.0230$) and Grizzly Lakes ($R^2=0.31$, $P=0.0247$) had weaker correlations. There was no relationship between age and THg concentration in lake trout from Copper Lake ($R^2=-0.01$, $P=0.4354$) or rainbow trout from Summit Lake ($R^2=-0.05$, $P=0.5449$). Length by age plots indicated that fish size in several populations approached an asymptote of maximum size in the oldest individuals (fig. 5).

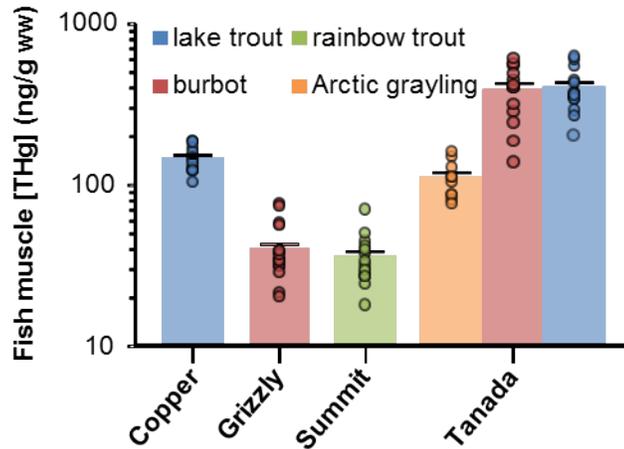


Figure 2. Arithmetic-mean total mercury concentrations (THg, in nanograms per gram wet weight [ng/g ww]) \pm 1 standard error in fish species collected from four lakes in Wrangell-St. Elias National Park and Preserve, Alaska.

Table 2. Comparison of coefficient of determination (R^2) and P-values between mercury-age and mercury-length regressions from fish sampled in Wrangell-St. Elias National Park and Preserve, Alaska.

Site and species	Age		Size	
	R^2	P-value	R^2	P-value
Copper Lake lake trout	-0.01	0.4354	-0.08	0.9412
Grizzly Lake burbot	0.31	0.0247	0.11	0.1218
Summit Lake rainbow trout	-0.05	0.5449	0.04	0.2384
Tanada Lake Arctic grayling	0.55	0.0042	0.36	0.0388
burbot	0.37	0.0230	0.34	0.0216
lake trout	0.72	<0.0001	0.39	0.0077

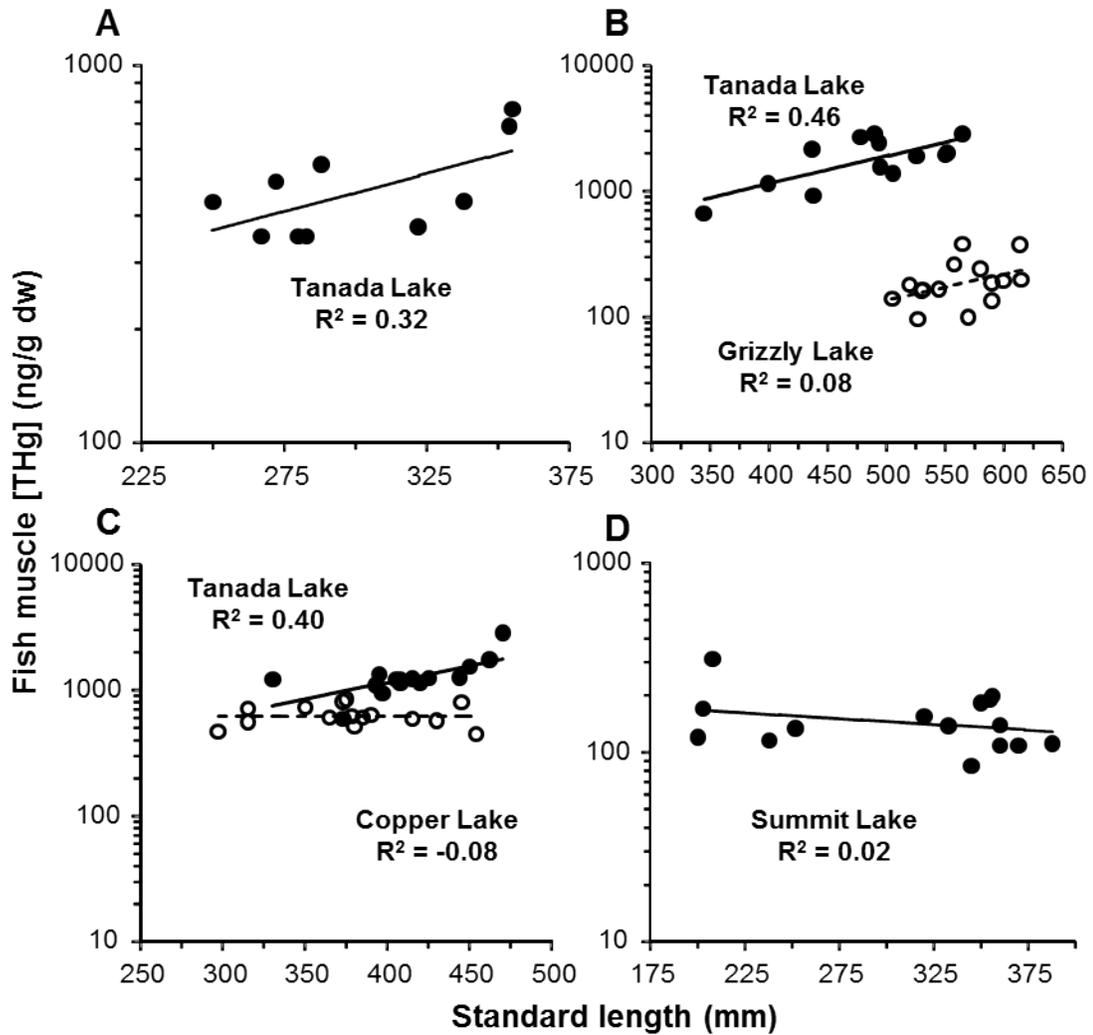


Figure 3. Linear relationships between standard length (in millimeters, [mm]) and total mercury concentrations (THg, in nanograms per gram dry weight [ng/g dw]) in fish muscle of (A) Arctic grayling, (B) burbot, (C) lake trout, and (D) rainbow trout sampled from Wrangell-St. Elias National Park and Preserve, Alaska.

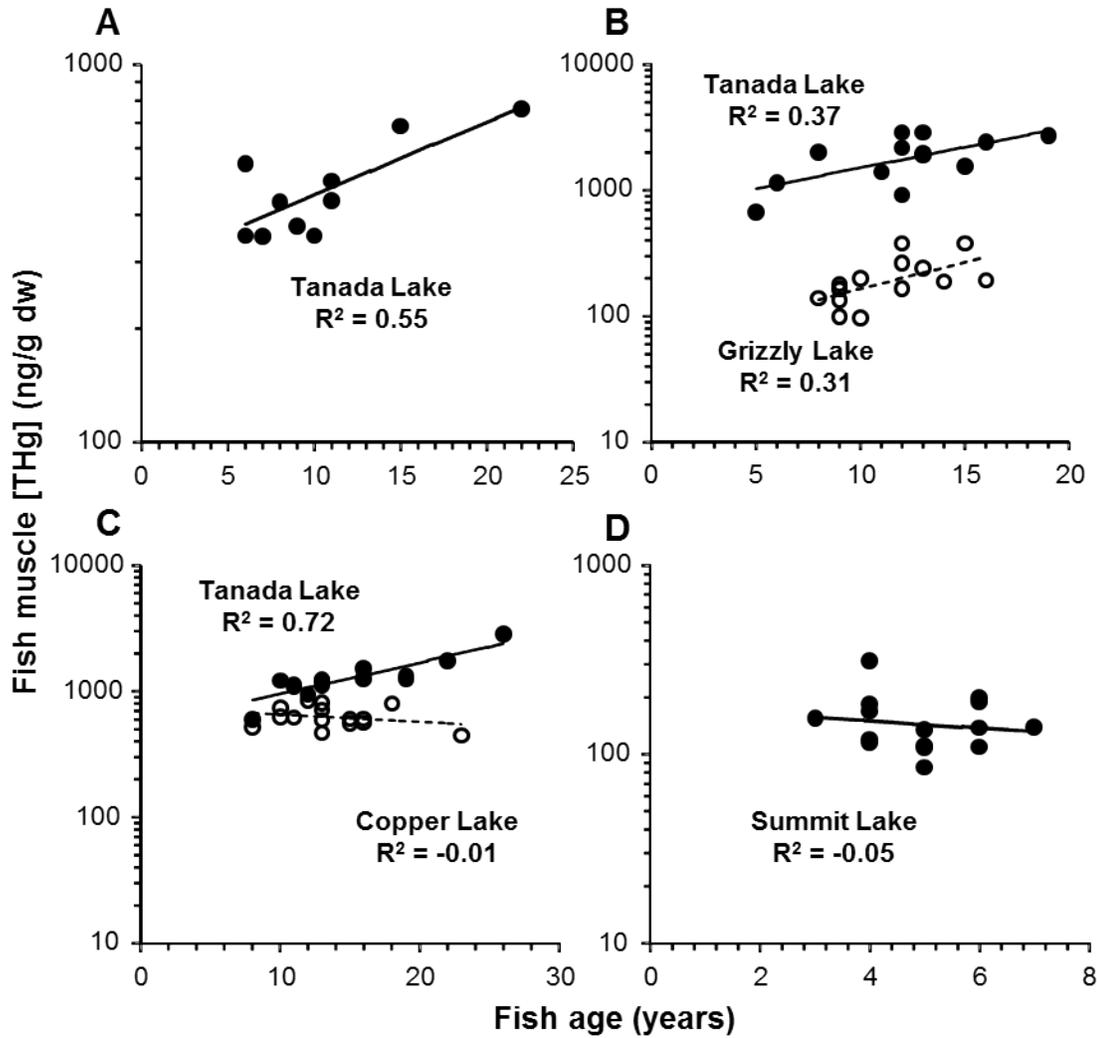


Figure 4. Linear relationships between fish age (in years) and total mercury concentrations (THg, in nanograms per gram dry weight [ng/g dw]) in fish muscle of (A) Arctic grayling, (B) burbot, (C) lake trout, and (D) rainbow trout sampled from Wrangell-St. Elias National Park and Preserve, Alaska.

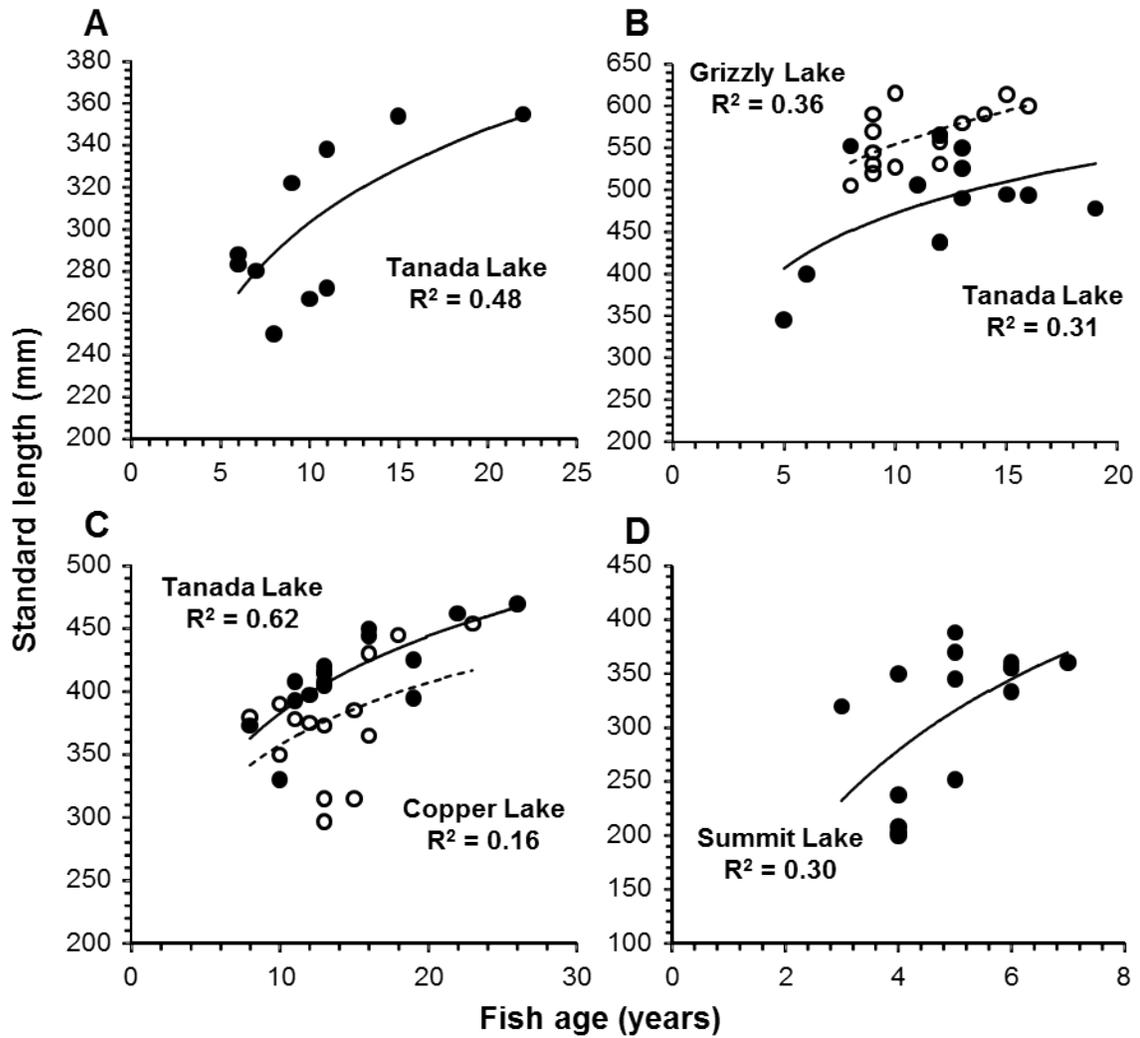


Figure 5. Logarithmic relationships between fish age (in years) and standard length (in millimeters [mm]) of (A) Arctic grayling, (B) burbot, (C) lake trout, and (D) rainbow trout sampled from Wrangle-St. Elias National Park and Preserve, Alaska.

Species Comparisons

Tanada Lake was the only site from which more than one species was collected; therefore, species comparisons were limited to Tanada Lake. After size-normalizing THg concentrations in all fish from Tanada Lake to 350 mm SL, we found significant differences in the THg concentrations among the species (fig. 6; $F_{2,37}=10.08$, $P=0.0004$). Concentrations were highest in lake trout (253.2 ± 24.6 ng/g ww), and lowest in Arctic grayling (133.2 ± 13.5 ng/g ww). Burbot THg concentrations were intermediate (184.6 ± 19.0 ng/g ww) between the other two species, but were not statistically different from either lake trout ($P=0.15$) or grayling ($P=0.08$).

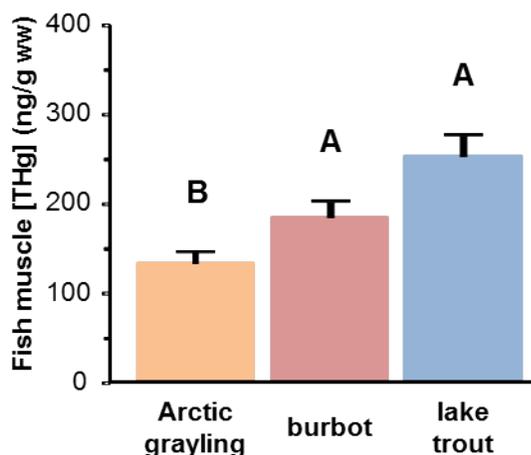


Figure 6. Back-transformed least-square mean total mercury concentrations (THg, in nanograms per gram wet weight [ng/g ww]) in muscle of fishes from Tanada Lake, Wrangell-St. Elias National Park and Preserve, Alaska. All concentrations are size-normalized to 350 millimeters standard length and error bars are standard errors estimated using the delta method.

Site Comparisons

We observed inter-lake differences in the THg concentrations of lake trout and burbot that were each sampled from two lakes (burbot, $F_{1,27}=112.82$, $P < 0.0001$; lake trout, $F_{1,29}=31.93$, $P < 0.0001$; fig. 7). Size-normalized (550 mm SL) THg concentrations in burbot were higher in Tanada Lake (535.2 ± 34.7 ng/g ww) than in Grizzly Lake (40.6 ± 31.1 ng/g ww), despite similar median ages in the two populations (Tanada Lake=12 yr; Grizzly Lake=10 yr; fig. 5). Likewise, size-normalized (378 mm SL) THg concentrations in lake trout were higher in Tanada Lake (302.9 ± 14.4 ng/g ww) than in Copper Lake (138.7 ± 25.2 ng/g ww), even though the median age of both populations was 13 yr (fig. 5).

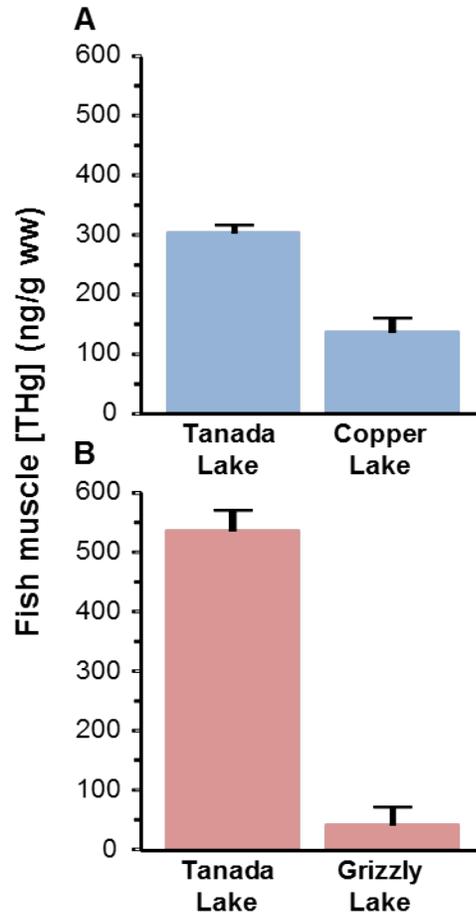


Figure 7. Back-transformed least-square mean total mercury concentrations (THg, in nanograms per gram wet weight [ng/g ww]) in muscle of (A) lake trout, and (B) burbot from lakes in Wrangell-St. Elias National Park and Preserve, Alaska. Lake trout from Tanada Lake were size-normalized to the median length of Copper Lake lake trout (378 millimeters). Burbot from Tanada Lake were size-normalized to the median length of Grizzly Lake burbot (550 millimeters). Mercury concentrations for lake trout from Copper Lake and burbot from Grizzly Lake are geometric means. Error bars are standard errors estimated using the delta method.

Toxicological Risks

Fish Risk

We first assessed risk to fishes based on the proportion of non-size-normalized, whole-body THg concentrations that exceeded the selected toxicological benchmarks. Across all sites and species, 24 percent of fish sampled (20 individuals) had whole-body THg concentrations exceeding the NOER benchmark for which Hg exposure may affect fish health (200 ng/g ww; table 3), and 6 percent (5 individuals) had concentrations exceeding the LOER benchmark (300 ng/g ww; table 3). All fish exceeding the NOER benchmark were from Tanada Lake, where 53 percent of the fish sampled (20 individuals) exceeded the benchmark. On a species specific-basis, in Tanada Lake, none of the Arctic grayling, 62 percent of the burbot (8 individuals), and 80 percent of the lake trout (12 individuals) exceeded the 200 ng/g NOER benchmark. Across all species, 13 percent of fish (5 individuals) from Tanada Lake exceeded the LOER benchmark, comprised of 23 percent of burbot (3 individuals) and 13 percent of lake trout (2 individuals), and no Arctic grayling.

Table 3. Percentage of fish from four lakes in Wrangell-St. Elias National Park and Preserve, Alaska, with raw (that is, non-size corrected) total mercury concentrations exceeding defined toxicity benchmarks for fish, avian, and human risk.

[Fish and avian benchmarks are based on wet weight whole-body concentrations whereas human benchmarks are based on wet weight concentrations in fish muscle]

Site and species	Fish toxicity			Bird toxicity			Human consumption ^c	
	NOER ^a (200 ng/g)	LOER ^b (300 ng/g)	High sensitivity (90 ng/g)	Mod. sensitivity. (180 ng/g)	Low sensitivity. (270 ng/g)	Unrestricted consumption (150 ng/g)	4 Meals per week (320 ng/g)	2 Meal per week (640 ng/g)
Across All Sites	24	6	45	25	7	41	25	--
Copper Lake								
lake trout	--	--	53	--	--	40	--	--
Grizzly Lake								
burbot	--	--	--	--	--	--	--	--
Summit Lake								
rainbow trout	--	--	--	--	--	--	--	--
Tanada Lake								
Arctic grayling	--	--	20	--	--	10	--	--
burbot	62	23	92	69	31	92	62	--
lake trout	80	13	100	80	13	100	87	--

^aNo-Observed-Effects-Residue (Beckvar and others, 2005).

^bLowest-Observed-Effects-Residue (Sandheinrich and others, 2011).

^cBased on Alaska-caught fish monthly consumption allowances for women who are or can become pregnant, nursing mothers, and young children (aged 12 years and under).

We also examined risk over the size range of fish sampled in order to account for the effects of size and age on THg concentrations. These size-specific risk models predicted that Arctic grayling would not exceed the 200 ng/g ww NOER or the 300 ng/g ww LOER benchmarks at any size sampled (fig. 8). Conversely, burbot and lake trout were predicted to first exceed the NOER toxicological benchmark at 370 and 356 mm SL, respectively, with greater than or equal to 50 percent of each population expected to exceed this threshold at 492 and 398 mm SL. One-hundred percent of burbot and lake trout with greater than 559 and 470 mm SL, respectively, were predicted to exceed the NOER (appendix 2). Burbot and lake trout first exceeded the LOER benchmark at 463 and 432 mm SL, respectively, and both plateaued at about 46 percent of the population exceeding the benchmark at 565 and 470 mm SL, respectively (fig. 8).

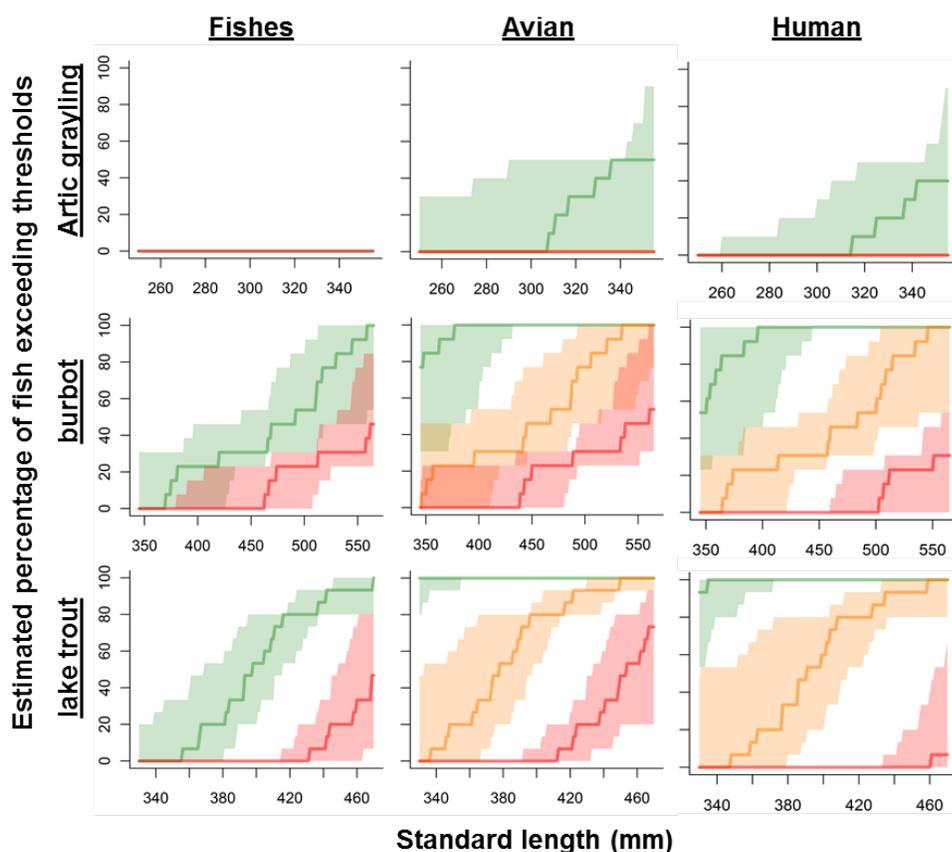


Figure 8. Modeled relationship between fish size and the percentage of Arctic grayling, burbot, and lake trout from Tanada Lake, Wrangell-St. Elias National Park and Preserve, Alaska, that exceed defined toxicological benchmarks for fishes, avian, and human consumers. Fishes: Percentage of whole-body total mercury concentrations at the sampled size range that exceed benchmarks for a generic no-observed-effects residue (green; 200 ng/g ww), and lowest-observed-effects residue (red; 300 nanograms per gram wet weight [ng/g ww]) for fish health. Avian: Percentage of whole-body total mercury concentrations at the sampled size range that exceed dietary toxicity benchmarks for piscivorous birds that can be classified as high (green; 90 ng/g ww), medium (orange; 180 ng/g ww), or low (red; 270 ng/g ww) sensitivity. Human: Percentage of muscle total mercury concentrations exceeding the Alaska Department of Health and Social Services guidelines at which they recommend women of childbearing age and children younger than 12 years of age consume no more than 4 meals per week (green; 150 ng/g ww), 3 meals per week (orange; 320 ng/g ww), or 2 meals per week (red; 640 ng/g ww).

Avian Risk

Across all sites, 45 percent of fish sampled (37 individuals) had THg concentrations greater than the proximate benchmark for Hg effects to highly sensitive avian piscivores (90 ng/g ww), 25 percent (21 individuals) had tissue concentrations greater than the benchmark for moderate-sensitivity species (180 ng/g ww), and 7 percent (6 individuals) had concentrations greater than the benchmark for low-sensitivity species (270 ng/g ww; table 3). Whole-body THg concentrations exceeded the high-sensitivity benchmark in lake trout from Copper Lake (53 percent [8 individuals] of fish) and in all three fish species from Tanada Lake (Arctic grayling—20 percent [2 individuals]; burbot—92 percent [12 individuals]; lake trout—100 percent [15 individuals]). Only burbot and lake trout from Tanada Lake exceeded the moderate-sensitivity benchmark (69 and 80 percent [9 and 12 individuals] for burbot and lake trout, respectively) and low-sensitivity benchmark (31 and 13 percent [4 and 2 individuals], respectively; table 3).

Size-specific risk models estimated that whole-body THg concentrations exceeded the high-sensitivity benchmark for all three species in Tanada Lake (fig. 8). Arctic grayling first exceeded the high-sensitivity benchmark at 308 mm SL, with greater than or equal to 50 percent of the population expected to exceed this benchmark at 336 mm SL (appendix 2). Seventy-six percent of burbot exceeded the high-sensitivity benchmark at 345 mm SL, the lowest length observed for this species in Tanada Lake. One hundred percent of the burbot exceeded the high-sensitivity avian benchmark at 377 mm SL, and all lake trout exceeded this benchmark at the lowest observed length of 330 mm. None of the Arctic grayling were modeled to exceed the moderate- or low-sensitivity benchmarks within the size range sampled. Burbot first exceeded the moderate-sensitivity benchmark at 346 mm SL, greater than or equal to 50 percent of the population exceeded at 468 mm SL, and 100 percent at 535 mm SL. Lake trout from Tanada Lake were first modeled to exceed the moderate-sensitivity benchmark at 337 mm SL, with greater than or equal to 50 percent and 100 percent exceeding this threshold at 378 and 450 mm SL, respectively. Burbot and lake trout first exceeded the low-sensitivity benchmark at 439 and 413 mm SL, respectively. Greater than 50 percent of burbot and lake trout whole-body THg concentrations exceeded the low sensitivity threshold at 561 and 454 mm SL, respectively (appendix 2).

Human Consumption Risk

For the raw, unadjusted data across all sites, 41 percent of the fish sampled (34 individuals) had THg concentrations in their muscle that exceeded the AK-DHSS unrestricted human consumption guideline (150 ng/g ww in fish muscle), 25 percent of the fish sampled (21 individuals) exceeded the 4-meal-per-week AK-DHSS guideline (320 ng/g ww), and none of the fish had concentrations greater than the guideline at which AK-DHSS recommends consumption be limited to 1 meal per week. Forty percent of lake trout (6 individuals) from Copper Lake exceeded the unrestricted consumption guidance benchmark, although no fish from Copper Lake were determined to have muscle THg concentrations greater than the 4-meal-per-week guideline. Ten percent of Arctic grayling (1 individual) from Tanada Lake exceeded the AK-DHSS unrestricted consumption guideline, whereas none exceeded the 4-meal-per-week guideline. All lake trout and 92 percent of burbot (12 individuals) from Tanada Lake exceeded the unrestricted consumption guideline, whereas 87 percent of lake trout (13 individuals) and 62 percent of burbot (8 individuals) exceeded the 4-meal-per-week guideline.

Total mercury muscle concentrations in fish sampled from Tanada Lake were modeled across the observed size range to assess the risks of human consumption (fig. 8). Arctic grayling were first modeled to exceed the AK-DHSS unrestricted consumption guideline at 315 mm SL, but no individuals were modeled to exceed the 4- or 2-meal-per week-guidelines at any size sampled. Fifty-four and 93 percent of burbot and lake trout, respectively, exceeded the AK-DHSS unrestricted consumption guideline at the smallest sizes observed (345 and 330 mm SL, respectively; appendix 2). Models estimated that burbot and lake trout would first exceed the AK-DHSS 4-meal-per-week guideline at 365 and 348 mm SL, respectively, and that 50 percent or more fish exceeded this benchmark at 484 and 391 mm SL, respectively. One-hundred percent of burbot greater than 546 mm SL and lake trout greater than 459 mm SL were modeled to exceed the AK-DHSS 4-meal-per-week guideline. Some of the burbot greater than 503 mm SL and lake trout greater than 461 mm SL were modeled to exceed the AK-DHSS 2-meal-per-week guideline, although they were never more than 30 percent of the population (fig. 8; appendix 2).

Discussion

Results of this study indicate that Hg bioaccumulation and risk to remote aquatic ecosystems within WRST are highly variable among lakes and among species within a lake. Total Hg concentrations in individual fish ranged by an order of magnitude across the 83 individuals analyzed, with the highest concentrations exceeding 600 ng/g ww. Even within a single species, size-normalized THg concentrations differed among lakes. THg concentrations in lake trout from Tanada Lake were 2.2 times higher than lake trout from Copper Lake, and concentrations in burbot from Tanada Lake were 13.2 times higher than burbot from Grizzly Lake. Furthermore, some of the THg concentrations in fish, particularly from Tanada Lake, were high enough to be of concern for wildlife and humans that consume them. However, the relationship between THg concentrations and fish size in Tanada Lake suggests that Hg exposure in human consumers could be reduced by harvesting small-sized fishes.

Although fish THg concentrations varied substantially among sites, they generally are comparable to those measured in these species from other studies in remote northern habitats. For example, THg concentrations in lake trout and Arctic grayling averaged between 58 and 216 ng/g ww, and 54 to 112 ng/g ww, respectively, in four lakes in Arctic Alaska (Allen-Gil and others, 1997). These concentrations are similar to those that we measured in fish from Copper, Grizzly, and Summit Lakes, but are considerably less than the mean values from Tanada Lake. Similarly, mean Hg concentrations for lake trout (180 ng/g ww) and burbot (150 ng/g ww) from Great Slave Lake, Canada (Evans and others, 2013) were comparable to those that we measured in Copper and Grizzly Lakes, but are considerably less than the mean values from Tanada Lake. It is unclear why THg concentrations in fish from Tanada Lake were elevated in comparison to the other lakes, but could be related to lake or catchment characteristics that facilitate higher bioaccumulation or biomagnification of Hg in this system, or to marine-derived sources associated with the return of anadromous fish since only Tanada Lake has substantial salmon returns each year (Written Communication, Dave Sarafin, National Park Service, 5/8/2014). Many factors are known to influence these processes, including Hg inputs, MeHg production, and food web processes that regulate the flow of Hg through systems. A primary source of Hg inputs into WRST lakes likely is atmospheric deposition (Jaffe and others, 2008; Durnford and others, 2010; Phillips and others, 2011). Despite the close proximity of three of the lakes assessed in this study, the surface area of Tanada Lake (2,718 acres) is 1.5 times greater than the surface area of Copper Lake (1,830 acres) and 11 times greater than the surface area of Grizzly Lake (252 acres). Additionally, the surface area of Tanada Lake catchment basin (84,830 acres) is 3 times greater than that of Copper Lake (26,804 acres), and 47 times greater than that of Grizzly Lake (1,790 acres). Therefore, even if the

delivery rates per unit area are similar among the three lakes, the substantially larger lake and catchment size likely results in higher total atmospheric Hg loading in Tanada Lake relative to Copper Lake, and in Copper Lake relative to Grizzly Lake. Hg concentrations in fish follow these trends, suggesting that further investigation of the influence of catchment drivers is warranted. Differences in Hg concentrations among the four lakes also could reflect varying geologic conditions, such as differing bedrock composition or glacial activity in the upstream watersheds, a hypothesis that could be assessed by measuring Hg concentrations in sediment and water inputs to the lakes.

Although Hg inputs play a role in determining Hg concentrations in fish, the biogeochemical factors that control MeHg production also are key drivers of Hg bioaccumulation in food webs (Ulrich and others, 2001; Benoit and others, 2003). Thus, lake and watershed characteristics, including the presence of wetlands, high-quality organic matter inputs, and low pH, facilitate Hg methylation and, thus, are often important drivers of Hg bioaccumulation in fish (Watras and others, 1998; Snodgrass and others, 2000; Greenfield and others, 2001; Wiener and others, 2006). In order to understand the role methylation plays in generating the variable THg concentrations measured in WRST fish, future research should include abiotic and low trophic-level sampling with the aim of identifying habitat, lake, and watershed characteristics that are associated with contamination in these parts of the ecosystems as well as higher trophic-level fishes.

More than 90 percent of the MeHg exposure in fish comes from dietary sources (Hall and others, 1997). Thus, differences in the feeding ecology strongly regulate the accumulation of Hg in tissues (Cabana and others, 1994; Vander Zanden and Rasmussen 1996; Power and others, 2002). The considerable variation in THg concentrations among the three species of fishes sampled from Tanada Lake suggests that variable bioavailability of MeHg is not the sole determinant of THg concentrations in these lakes. The two species with the highest THg concentrations (lake trout and burbot) are predominantly piscivorous as adults (Scott and Crossman, 1973; Mittlebach and Persson, 1998), and, therefore, occupy higher trophic positions, which results in higher levels of Hg biomagnification. In contrast, Arctic grayling tend to forage predominantly on benthic and emergent invertebrates (Armstrong, 1986). THg concentrations in invertivorous grayling were low compared to co-occurring piscivorous species in other areas of Alaska (Allen-Gil and others, 1997; Jewett and others, 2003) and throughout Canada (Lockhart and others, 2005). Although these data indicate that grayling from Tanada Lake typically are younger (median age=9.5 yr) than either burbot (12 yr) or lake trout (13 yr), THg concentrations in burbot and lake trout were consistently higher than in similarly aged grayling. Thus, it is unlikely that age alone could account for the observed variation. However, THg concentrations were more strongly correlated with age than size. This may indicate the relatively large size of the fish sampled and the asymptotic relationship between age and growth (Rao, 1958; Ricker, 1975), which could result in the decoupling of size-THg relationships once the asymptotic portion of the growth curve is reached. Because the point at which growth asymptotes varies among populations (Chen and others, 1992; Murphy and Willis, 1996), populations may have differing relationships even when age or size structures are similar (for example, lake trout from Tanada Lake compared to Copper Lake in the current study).

Although Hg exposure risk in WRST was not formally assessed, our data suggest that risk to park fish, wildlife, and humans is variable across lakes and species. Mercury concentrations in fishes from Grizzly Lake and Summit Lake did not exceed any of the guidelines used for comparisons, whereas concentrations in fishes from Copper Lake and Tanada Lake exceeded benchmarks for toxicological risk to fish, birds, and (or) humans (table 3). Despite the fact that only the most conservative benchmarks for bird (90 ng/g ww in whole body) and human (150 ng/g ww in muscle, corresponding to the AK-DHSS unrestricted consumption guideline) were exceeded in lake trout from Copper Lake, a relatively large percentage of individuals exceeded the human benchmark, suggesting that a more detailed evaluation of human consumption risk is warranted. Furthermore, THg concentrations in fishes from Tanada Lake regularly exceeded conservative benchmarks and benchmarks for risk to moderate- and low-sensitivity avian consumers (180 and 270 ng/g ww in whole body, respectively), as well as the AK-DHSS 4-meal-per-week guideline (320 ng/g ww in muscle). In particular, most of the burbot and lake trout from Tanada Lake exceeded these benchmarks (table 3). This warrants further considerations because Tanada Lake is a known site of subsistence fisheries (Weeks, 2003; Schwanke and McCormick, 2010; Kukkonen and Zimpelman, 2012). However, our analyses suggest that selectively harvesting smaller individuals is one potential way to reduce human Hg exposure from Tanada Lake. For example, the model of risk as a function of fish size in Tanada Lake predicted that no burbot less than 365 mm SL or lake trout less than 348 mm SL were expected to exceed the AK-DHSS 4-meal-per-week guideline. Conversely, all burbot and lake trout with greater than 546 and 459 mm SL, respectively, exceeded this guideline. Such insights into the size at which risk thresholds are exceeded provide a valuable basis upon which managers can communicate and mitigate exposure risk to consumers. Nonetheless, the high degree of within-park variability suggests that size thresholds generated for one population may not reflect risk in other populations. Thus, follow-up research and monitoring efforts across sites within the park will be beneficial for adequately characterizing and determining future directions for minimizing risk to park fish, wildlife, and users.

This study provides an important step towards understanding the distribution of Hg and potential risks posed to park wildlife and visitors. However, our study included only a small fraction of the aquatic resources and habitat types present within WRST. Such limited sampling is unlikely to adequately characterize Hg risk because of the substantial variation observed within other national parks (Eagles-Smith and others, 2014). Additional quantitative studies of the biogeochemical and food-web processes regulating inter-site and inter-species variation within the park will facilitate more accurate identification of the ecological risks associated with Hg. Finally, it is important that the toxicological effects of Hg on park fish and wildlife be directly assessed in order to develop risk benchmarks that reflect the species and habitats present in WRST, and, thus, more robustly portray risk to WRST ecosystems as a whole.

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Appendix 1. Equations for Converting Standard Length (SL, in millimeters) to Total Length (TL, in millimeters) for Four Species of Fish Sampled from Wrangell-St. Elias National Park and Preserve, Alaska.

Arctic grayling: $TL = 1.0312 * SL + 23.116; n = 10; R^2 = 0.9944; P < 0.0001$

burbot: $TL = 1.11 * SL + 7.6252; n = 24; R^2 = 0.9866; P < 0.0001$

lake trout: $TL = 1.1643 * SL + 18.84; n = 30; R^2 = 0.9754; P < 0.0001$

rainbow trout: $TL = 1.1386 * SL + 4.6143; n = 15; R^2 = 0.9974; P < 0.0001$

Appendix 2. Size at Which the Proportion of Modeled Individuals of Each Species from Tanada Lake Exceeded Defined Toxicity Benchmarks in Wrangell-St. Elias National Park and Preserve, Alaska.

Species	Fish Toxicity					
	NOER ^a (200 ng/g)			LOER ^b (300 ng/g)		
	0%	50%	100%	0%	50%	100%
Arctic grayling	--	--	--	--	--	--
burbot	370	492	559	463	--	--
lake trout	356	398	470	432	--	--

^a No-Observed-Effects-Residue (Beckvar and others, 2005).

^b Lowest-Observed-Effects-Residue (Sandheinrich and others, 2011).

Species	Bird Toxicity								
	High Sensitivity (90 ng/g)			Moderate Sensitivity (180 ng/g)			Low Sensitivity (270 ng/g)		
	0%	50%	100%	0%	50%	100%	0%	50%	100%
Arctic grayling	308	336	--	--	--	--	--	--	--
burbot	< 345	< 345	377	346	468	535	439	561	--
lake trout	< 330	< 330	< 330	337	378	450	413	454	--

Species	Human Toxicity								
	Unrestricted Consumption (150 ng/g)			4 Meals per Week (320 ng/g)			2 Meal per Week (640 ng/g)		
	0%	50%	100%	0%	50%	100%	0%	50%	100%
Arctic grayling	315	--	--	--	--	--	--	--	--
burbot	< 345	< 345	396	365	484	546	503	--	--
lake trout	< 330	< 330	335	348	391	459	461	--	--

-- Denotes that none of the population were modeled to exceed the benchmark at even the largest size observed.

< Denotes that 100 percent of the population were modeled to exceed the benchmark at even the smallest size observed.

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