# Historic and Contemporary Mercury Exposure and Potential Risk to Yellow-Billed Loons (Gavia adamsii) Breeding in Alaska and Canada

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# Historic and Contemporary Mercury Exposure and Potential Risk to Yellow-billed Loons (*Gavia adamsii*) Breeding in Alaska and Canada

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**Abstract.**—The Yellow-billed Loon (*Gavia adamsii*) is one of the rarest breeding birds in North America. Because of the small population size and patchy distribution, any stressor to its population is of concern. To determine risks posed by environmental mercury (Hg) loads, we captured 115 Yellow-billed Loons between 2002 and 2012 in the North American Arctic and sampled their blood and/or feather tissues and collected nine eggs. Museum samples from Yellow-billed Loons also were analyzed to examine potential changes in Hg exposure over time. An extensive database of published Hg concentrations and associated adverse effects in Common Loons (*G. immer*) is highly informative and representative for Yellow-billed Loons. Blood Hg concentrations reflect dietary uptake of methylmercury (MeHg) from breeding areas and are generally considered near background levels if less than 1.0 µg/g wet weight (ww). Feather (grown at wintering sites) and egg Hg concentrations can represent a mix of breeding and wintering dietary uptake of MeHg. Based on Common Loon studies, significant risk of reduced reproductive success generally occurs when adult Hg concentrations exceed 2.0 µg/g ww in blood, 20.0 µg/g fresh weight (fw) in flight feathers and 1.0 µg/g ww in eggs. Contemporary mercury concentrations for 176 total samples (across all study sites for 115 Yellow-billed Loons) ranged from 0.08 to 1.45 µg/g ww in blood, 3.0 to 24.9 µg/g fw in feathers and 0.21 to 1.23 µg/g ww in eggs. Mercury concentrations in blood, feather and egg tissues indicate that some individual Yellow-billed Loons in breeding populations across North America are at risk of lowered productivity resulting from Hg exposure. Most Yellow-billed Loons breeding in Alaska overwinter in marine waters of eastern Asia. Although blood Hg concentrations from most breeding loons in Alaska are within background levels, some individuals exhibit elevated feather and egg Hg concentrations, which likely indicate the uptake of MeHg originating from eastern Asia. Feather Hg concentrations tended to be highest in individuals overwintering farthest west (closer to Asia). A retrospective analysis of museum specimens ($n = 25$) found a two-fold increase in Yellow-billed Loon feather Hg concentrations from the pre-1920s (as early as 1845) to the present. The projected increase in Hg deposition (approximately four-fold by 2050) along with the uncertainty of Hg being released through the thawing of permafrost and Arctic sea ice suggest that Hg body burdens in Yellow-billed Loons may increase. These findings indicate that Hg is a current and potentially increasing environmental stressor for the Yellow-billed Loon and possibly other Nearctic-Palearctic migrant birds. *Received 19 June 2013, accepted 4 July 2013.*

**Key words.**—Alaska, Asia, Common Loon, *Gavia adamsii*, *Gavia immer*, Mercury, Yellow-billed Loon.

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The Yellow-billed Loon (*Gavia adamsii*) is one of the rarest breeding birds in North America (Earnst 2004) and the rarest of the world's five species of loons (Family Gavidae). Approximately 3,000 Yellow-billed Loon individuals occur in Alaska, the only state in the United States where they breed (Stehn *et al.* 2013). The majority of the Alaska breeding population (80%) is found on the Arctic Coastal Plain (ACP), with the remaining (20%) nesting in the northern half of the Seward Peninsula. In an average year, < 1,000 pairs nest on the ACP, where their population is patchy and unevenly distributed (Earnst *et al.* 2005). Monitoring surveys, initiated in 1985, indicate that the population appears stable (Earnst *et al.* 2005; Stehn *et al.* 2013). Because of the small population size and patchy distribution, any habitat changes or anthropogenic stressors are cause for concern. Potential population threats include expansion of the oil industry into relatively high density breeding areas (90% of the Yellow-billed Loon breeding population in the ACP is within the National Petroleum

Reserve - Alaska) (Earnst 2004). Other population threats include climate change, subsistence hunting (Schmutz 2009), commercial fishnets (bycatch; Žydelis *et al.* 2009), and contaminants such as mercury (Hg) and other pollutants along nearshore marine ecosystems where loons forage, especially in eastern Asia (Agusa *et al.* 2007) where the majority of Alaska's Yellow-billed Loon population overwinters (North 1994; Fair 2002; Schmutz *et al.* 2014). As such, the Yellow-billed Loon is being considered for listing under the Endangered Species Act (U.S. Fish and Wildlife Service 2009). The potential anthropogenic threat to Yellow-billed Loons addressed in this study is the exposure to methylmercury (MeHg) available in ecosystems.

Recent evidence suggests that Hg exposure in piscivorous birds is increasing in the North American Arctic (Braune 2007; Rigét *et al.* 2011). The origin for the increase of environmental Hg loads is related to atmospheric Hg deposition from Asian emission sources (Sunderland *et al.* 2009; Blum *et al.* 2013), and possible changes in climate patterns that are resulting in thawing of permafrost and melting of Arctic sea ice with subsequent releases of bromine and increased Hg deposition (Brooks *et al.* 2006).

While specific adverse effects of Hg on the Yellow-billed Loon population in North America have not yet been measured, they are well established for the closely related Common Loon (*G. immer*). The two species are considered a super-species (American Ornithologists' Union 1998) and they share similar plumages, vocalizations, behaviors, and ecology (Sjölander and Ågren 1976; North 1994; Evers *et al.* 2010). Yellow-billed Loon weights range from 3.7 to 6.4 kg (this study), and Common Loon weights range from 3.5 to 7.6 kg (Gray *et al.* 2014). Because of these strong ecological and morphometric similarities between species, the robust literature on the exposure and effects of Hg on the Common Loon can be used to make relevant inferences about Hg exposure to Yellow-billed Loons (Evers *et al.* 1998, 2008b; Burgess and Meyer 2008; Schoch *et al.* 2014).

Specific Hg concentrations in blood, feather and egg tissues of Common Loons can now be directly related to levels of reduced fledging success in breeding populations in Maine and New Hampshire (Evers *et al.* 2008b), Wisconsin and New Brunswick (Burgess and Meyer 2008), and New York (Schoch *et al.* 2014). These studies found close agreement among tissue Hg concentrations and subsequent effects, with levels causing significant reproductive concern generally above 2.0 µg/g wet weight (ww) in the blood, 1.0 µg/g ww in the egg and 20.0 µg/g fresh weight (fw) in the feather. Hg effect concentrations based on these studies are believed to be physiologically relevant for use with the similar-sized and closely related Yellow-billed Loon as the pharmacokinetics and overall ecology are comparable.

We examined current and historic tissues from Yellow-billed Loons while on their breeding territories in Alaska to assess levels of exposure to Hg and to compare these values with effect concentrations in Common Loons. Our specific objectives were to: 1) document current patterns of Hg exposure to Yellow-billed Loons through examination of multiple tissue types that portray different time periods of exposure; 2) infer the magnitude of possible reproductive effects of Hg exposure on Yellow-billed Loons; and 3) examine evidence for change in Yellow-billed Loon exposure to Hg over the past 170 years. The contemporary and retrospective assessment conducted here is timely because environmental Hg loads have increased over the past century and further increases are expected over the next several decades (Sunderland *et al.* 2009).

## METHODS

### Study Area

The primary study area is in Alaska within the ACP, roughly 100 km southeast of Barrow, interior from the Beaufort Sea coast, and 60 km southwest of Teshekpuk Lake (Fig. 1). Here exists an extensive system of variable-sized fish-bearing lakes and several major drainage rivers. This study area is within the high-density region for breeding Yellow-billed Loons

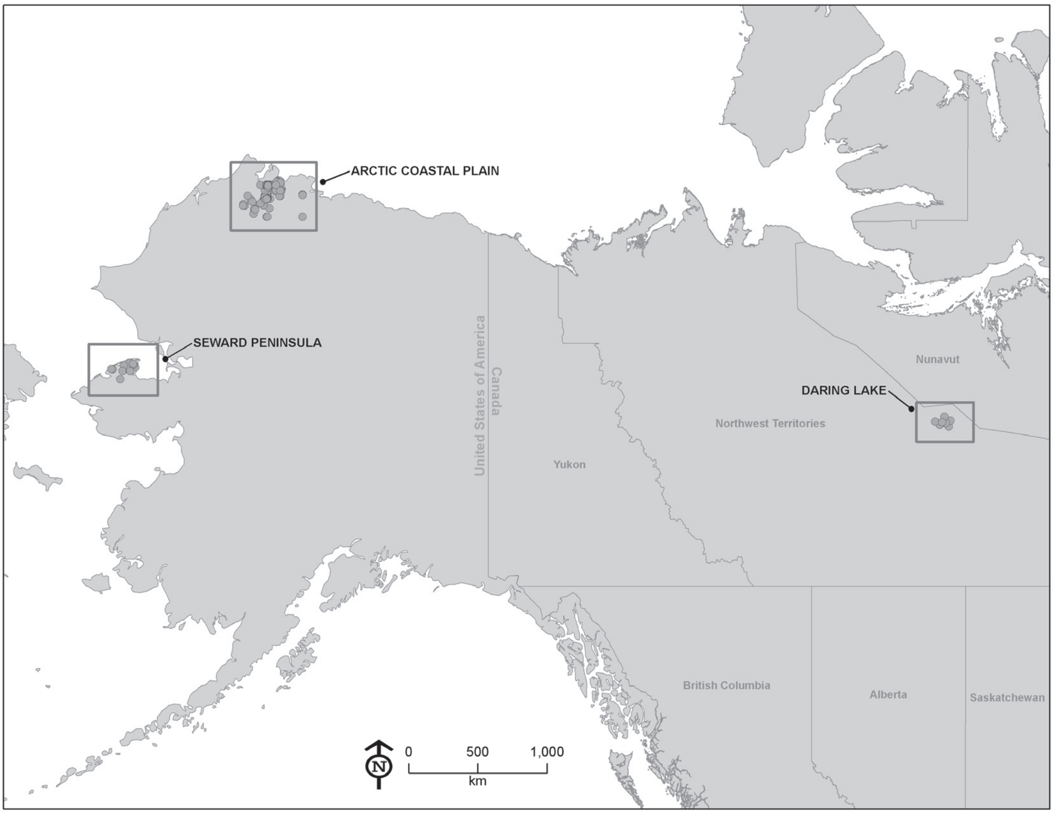

**Figure 1. Study sites for sample collection in Yellow-billed Loons, 2002 to 2012.**

(Earnst *et al.* (2005). The landscape is composed of a continuous permafrost environment, with a shallow active layer under tundra plant communities (< 1 m) and somewhat deeper active layers (taliks) underneath lakes. Unlike boreal forest areas, such as those within interior Alaska, where discontinuities in permafrost allow deep subsurface flow and connectivity, the shallow active layers of the ACP and the impermeability of the permafrost trap labile water near or at the surface. Thus, despite the ACP receiving relatively minimal annual precipitation (15-25 cm/year), it is replete with many small and large lakes (> 100,000), as well as highly saturated soils in many plant communities (Walker *et al.* 2005).

Our secondary, less intensive study sites were southwest and southeast of the ACP (Fig. 1). We sampled Yellow-billed Loons on the northern part of the Seward Peninsula, Alaska, which is similar to the ACP but with a slightly deeper active layer (for further description, see Schmutz *et al.* 2014). In Canada, our study site was the Daring Lake region of the Northwest Territories (65° 50' N and 111° 38' W), approximately 300 km northeast of Yellowknife. This area, just north of tree line, consisted of multiple oligotrophic lakes with irregular shorelines and coves that harbor multiple breeding pairs of Yellow-billed Loons. The lakes are larger and deeper than those in Alaska and surrounded by Arctic tundra, but with prevalent rock outcroppings.

### Contemporary Capture and Field Tissue Sampling

We evaluated Hg exposure in Yellow-billed Loons using blood, feathers, and eggs from 2002 to 2012, but not all tissue types were collected at every site (Table 1) or in every year. To locate individuals for capture and sampling purposes, we conducted aerial surveys using fixed-wing and rotary-wing aircraft as well as ground surveys. We captured Yellow-billed Loons during the mid to late incubation period using two methods: 1) nest trapping; and 2) off-nest trapping. For nest trapping, we used an approximately 1-m diameter spring-loaded aluminum bow-net. To prevent accidental breakage, we replaced Yellow-billed Loon eggs with wooden dummy eggs during the capture process. We captured approximately 15% of the Yellow-billed Loons while off nest by luring them into mist nets with decoys and call playbacks. Once captured, we recorded body and bill measurements, attached bands to their legs (one or two color bands on each leg along with a U.S. Geological Survey aluminum band), and collected blood and feather samples following protocols of Evers *et al.* (1998, 2008b). We collected partially-incubated eggs in the ACP (one

**Table 1. Mercury (Hg) concentrations in blood, feather, and eggs from 115 Yellow-billed Loons sampled ($n$ = 176) on breeding territories in Alaska, USA, and Northwest Territories, Canada, 2002-2012.**

| | Blood Hg (µg/g ww) | | | Feather Hg (µg/g fw) | | | Egg Hg (µg/g ww) | | |
|---|---|---|---|---|---|---|---|---|---|
| Region | *n* | Mean (SE) | Range | *n* | Mean (SE) | Range | *n* | Mean (SE) | Range |
| Arctic Coastal Plain, Alaska | 58 | 0.38 (0.03) | 0.08-1.45 | 77 | 8.18 (0.57) | 3.01-24.92 | 9 | 0.49 (0.13) | 0.21-1.23 |
| Daring Lake, Northwest Territories | 13 | 0.61 (0.05) | 0.30-0.96 | 13 | 6.22 (0.36) | 4.09-8.12 | — | — | — |
| Seward Peninsula, Alaska | — | — | — | 6 | 7.37 (1.31) | 3.62-11.09 | — | — | — |

egg per nest), wrapped them in aluminum foil that was cleansed with acetone, and kept them cool until they could be frozen.

### Field Tissue Analyses

We followed analytical protocols previously outlined for blood and feathers (Evers *et al.* 1998) and for eggs (Evers *et al.* 2003). Blood and feather samples for 2007-2012 were analyzed for total Hg concentrations at the Biodiversity Research Institute Wildlife Mercury Research Lab (Gorham, Maine). Samples included secondary feathers (5 cm tips) and whole blood. We placed samples into nickel boats that were then weighed and analyzed for total Hg concentration using a thermal decomposition technique with an automated direct Hg analyzer via the U.S. Environmental Protection Agency Method 7473 (U.S. Environmental Protection Agency 2007). Before and after every set of 30 samples, we included one sample each of two standard reference materials (Dorm-3 and Dolt-4), two methods blanks, and one sample blank. After every 20 samples, a duplicate was analyzed. Mean percent recoveries for total Hg of standard reference materials were within acceptable levels (U.S. Environmental Protection Agency Method 7473). Blood and egg samples from 2002-2003 were analyzed for total Hg at the Research Triangle Institute (Research Triangle Park, North Carolina) using standard and comparable analytical procedures with cold vapor atomic adsorption. We report Hg results in µg/g for all tissues and reported as ww for blood, fw for feathers, and dry weight (dw) converted to ww for eggs.

### Museum Tissue Analyses

To gain inference into historical Hg levels, we obtained secondary covert feathers from Yellow-billed Loon specimens at the Harvard University Museum of Comparative Zoology ($n$ = 19) and the University of Michigan Museum of Zoology ($n$ = 6). Museum specimens originated from the ACP, Alaska ($n$ = 17), Northwest Territories ($n$ = 6), and the Seward Peninsula, Alaska ($n$ = 2) between 1845 and 1949. We analyzed these specimens at the University of Michigan for organic Hg since inorganic Hg has sometimes been used as a preservative in museum specimens and more than 90% of total Hg in the feather exists in an organic form (Head *et al.* 2011). Feathers were washed to remove any surface contamination and homogenized whole with a grinder using a stainless steel vial and ball pestle. We then digested the ground feathers and extracted organic Hg using the method described by Head *et al.* (2011) and Nam and Basu (2011). Every 10 samples included a standard reference material (Dolt-4, Dogfish Liver Certified Reference Material for Trace Metals) and a method blank.

### Statistical Analysis

*Spatial patterns of mercury concentrations.* Blood and feather Hg values were log-transformed to normalize data and reduce heteroscedasticity. Normality was checked with the Shapiro-Wilk test and homogeneity of variance was checked with the Bartlett test. Using the nonparametric Spearman's rank correlation test, correlations between normally distributed blood Hg and non-normally distributed feather Hg concentrations were determined for individuals where both sample types were collected. One of our analytical goals was to evaluate the effects of sex and body mass on Hg concentration. However, since males are consistently heavier than females, sex and body mass are confounded if one uses the raw data in analysis. Thus, we transformed body mass data into normalized z-scores by region and sex. We then statistically examined the effects of sampling year, sampling region, sex, and body mass on blood and feather Hg concentrations using a general linear model framework. We distinguished among competing models of suites of covariates by ranking the relative fit of each model with the Akaike Information Criterion adjusted for small sample size ($\text{AIC}_c$). The model with the lowest $\text{AIC}_c$ and those having $\Delta\text{AIC}_c < 2$ had the most statistical support, those between 4 and 7 had considerably less support, and those > 10 had virtually no support (Burnham and Anderson 2002). Additional insight to the relative amount of statistical support for a given model was provided by each model's Akaike weight. Statistical analyses were performed in Microsoft EXCEL and JMP SAS (SAS Institute, Inc. 2008).

Male and female Yellow-billed Loons are monomorphic and were sexed using HINTZ and CHD primers, following the molecular genetic techniques outlined in Guzzetti *et al.* (2008). However, DNA blood samples for a subset of individuals ($n = 33$) were not collected. To include these samples where sex data were unavailable, we used a logistic regression function to predict the sex of Yellow-billed Loons based on morphometric measurements of individuals of known sex. Morphometric measurements included diagonal tarsus (mm), tarsus width (mm), tarsus breadth (mm), toe length (mm), culmen length (mm), culmen width (mm), culmen depth (mm), and body mass (g). Analyses were performed in Program R (R Development Core Team 2012). Diagonal tarsus ($P = 0.02$) and body mass ($P = 0.02$) were significant predictors of sex and so individuals were included in the Hg analysis if their combination of diagonal tarsus and mass measurements resulted in a 90% or higher probability of correct sex assignment. Specifically, individuals weighing less than 5,145 g had a 90% probability of being female, and individuals weighing more than 5,924 g had a 90% probability of being male (for Alaska individuals only). This function allowed the inclusion of an additional 21 Yellow-billed Loons in the blood and feather Hg analyses.

To evaluate the relationship between Hg in secondary flight feathers and wintering area, we used the most westerly location (degree of longitude) estimate (Douglas *et al.* 2012) from 48 Yellow-billed Loons marked with satellite transmitters (Schmutz *et al.* 2014). We used a quantile regression approach (Cade *et al.* 1999), PROC QUANTREG (SAS Institute Inc. 2008), which is appropriate for situations where limiting factors may exert their influence near the tails of the dependent variable's distribution and thus are not easily detected by ordinary least squares regression, which focuses on the mean response. We evaluated the relationship of wintering longitude to Hg concentration by examining estimates from the 50, 55, 60, 65, 70, 75, 80, 85, 90, and 95% quantiles (Fig. 2).

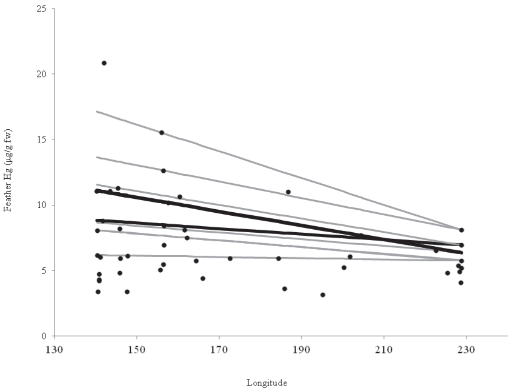

**Figure 2. Feather mercury (Hg) (µg/g, fw) concentrations in Yellow-billed Loons compared longitudinally to overwintering territories (smaller or more westerly longitudes are along the Asian coast, while larger or more easterly longitudes are along the North American coast). Longitudes east of the international dateline were recast as 180° plus the number of degrees east of the dateline. Darkened prediction lines reflect slopes whose confidence intervals did not overlap zero.**

*Long-term patterns of feather mercury concentrations.* To compare Hg concentrations in museum secondary coverts with secondary flight feathers collected in the field since 2007, it was necessary to determine the correlation in Hg between secondary coverts and secondary flight feathers. Log-transformed Hg concentrations found in secondary coverts and secondary fight feathers collected from the same individual were plotted in a linear regression to determine the Hg relationship between the two feather types. The regression calculated from this analysis was then used to create post hoc estimates of secondary flight feather Hg concentrations for museum specimens where only secondary coverts were available for Hg analysis. Based on available data, sample collection periods were divided into three groups: Pre-1920, 1930-1950, and Post-2000. Differences in Yellow-billed Loon secondary flight feather Hg concentrations among the three periods were examined with analysis of variance (ANOVA) and pairwise comparisons were tested with Tukey's honestly significant difference (HSD) test.

## Results

### Contemporary Patterns of Mercury Exposure

We sampled 115 Yellow-billed Loons or 176 tissues (10 for blood only, 47 for feathers only, 9 for blood and egg only, and 49 for both blood and feathers) in the three study areas during the summers of 2002 to 2003 and 2007 to 2012 (Table 1). Most samples originated on the ACP.

Overall, blood Hg concentrations ranged from 0.08 to 1.45 µg/g ww, feather Hg concentrations ranged from 3.01 to 24.92 µg/g fw, and egg Hg concentrations ranged from 0.21 to 1.23 µg/g ww (Table 1). Blood and feather Hg were positively correlated in individuals where both sample types were available (Spearman rank correlation coefficient $\rho = 0.45$, $P = 0.001$, $n = 49$). Body mass ranged from 3,700-6,450 g among all sampling regions (Table 2). Male Yellow-billed Loons in the ACP were significantly larger than females, averaging 17% greater body mass ($t_{71} = 14.26$, $P < 0.001$), and males in the Daring Lake region averaged 22% greater body mass than females ($t_{12} = 6.41$, $P < 0.001$). Small sample size precluded comparison of male and female body masses in the Seward Peninsula region.

### Spatial Patterns of Mercury Concentrations

We had no blood data from Seward Peninsula and only 1 year of blood data from Daring Lake in Canada. Thus, we conducted two analyses of variation in blood Hg: one examining year, sex, and body mass effects for loons sampled in the ACP over 6 years and one examining regional variation (ACP vs. Daring Lake) in one year (2010). For the time series of data from the ACP, blood Hg concentrations were highest in 2002 and 2003 ($\bar{x} = 0.69 \pm 0.15$ µg/g ww) and lowest in 2012 ($\bar{x} = 0.32 \pm 0.05$ µg/g ww). While blood Hg tended to be higher in males than females (Table 2), much of the variation in the data remained unexplained ($r^2 = 0.24$; Table 3). In 2010 (our lone sampling year for Daring Lake), blood Hg concentrations were greater at Daring Lake ($\bar{x} = 0.61 \pm 0.13$ µg/g ww) compared to the ACP ($\bar{x} = 0.38 \pm 0.03$ µg/g ww; $r^2 = 0.56$; Table 3). As with the previous analysis, the sex covariate fit substantially better than normalized body mass, suggesting that greater blood Hg in males is a function of ecological attributes other than body size or mass. Blood Hg concentrations were significantly higher in males ($\bar{x} = 0.56 \pm 0.08$ µg/g ww) compared to females ($\bar{x} = 0.41 \pm 0.05$ µg/g ww). The top supported model for feather Hg included no covariates. Thus, no differences in feather Hg concentrations

**Table 2. Body mass (g) of Yellow-billed Loons sampled in Alaska, USA, and Northwest Territories, Canada, 2002-2012.**

| | Body Mass (g) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | Female | | | Male | | | Unknown | | |
| Region | *n* | Mean (SE) | Range | *n* | Mean (SE) | Range | *n* | Mean (SE) | Range |
| Arctic Coastal Plain, Alaska | 49 | 4,945 (40) | 4,032-5,450 | 24 | 5,893 (55) | 5,300-6,450 | 29 | 5,369 (113) | 4,325-6,360 |
| Daring Lake, Northwest Territories | 8 | 4,150 (131) | 3,700-4,900 | 6 | 5,183 (60) | 4,900-5,300 | — | — | — |
| Seward Peninsula, Alaska | 4 | 5,203 (214) | 4,740-5,760 | 2 | 5,855 (205) | 5,650-6,060 | 4 | 5,272 (390) | 4,862-6,442 |

**Table 3. Model selection results examining the effect of sampling year, sampling region, sex, and body mass on blood and feather mercury (Hg) concentrations in breeding Yellow-billed Loons in Alaska, USA, and Northwest Territories, Canada from 2002 to 2012. Number of estimated parameters (K), differences between model Akaike Information Criterion adjusted for small samples size ($\Delta AIC_c$) values <10, and $AIC_c$ weights ($w_i$) are shown.**

*Blood Hg across years on the ACP*

| Model | K | $\Delta AIC_c$ | $w_i$ |
|---|---|---|---|
| Year (Temporal trend) + Sex | 4 | 0 | 0.485 |
| Year (Temporal trend) | 3 | 1.153 | 0.273 |
| Early vs. Late years + Sex | 4 | 2.222 | 0.16 |
| Year (Annual variation) | 2 | 5.248 | 0.035 |
| Sex | 3 | 6.123 | 0.023 |
| Null | 4 | 6.769 | 0.016 |
| Body mass | 5 | 8.131 | 0.008 |

*Blood Hg between regions*

| Model | K | $\Delta AIC_c$ | $w_i$ |
|---|---|---|---|
| Region + Sex | 3 | 0 | 0.959 |
| Region | 2 | 6.23 | 0.041 |

*Feather Hg*

| Model | K | $\Delta AIC_c$ | $w_i$ |
|---|---|---|---|
| Null | 1 | 0 | 0.262 |
| Body mass | 2 | 0.892 | 0.168 |
| Year (Temporal trend) | 2 | 1.484 | 0.125 |
| Sex | 2 | 1.563 | 0.12 |
| Body mass + Region | 3 | 2.441 | 0.077 |
| Body mass+ Sex | 3 | 2.668 | 0.069 |
| Sex + Region | 3 | 2.836 | 0.063 |
| Region | 3 | 3.12 | 0.055 |
| Body mass+ Sex + Region | 4 | 4.151 | 0.033 |
| Year (Annual variation) | 6 | 4.449 | 0.028 |

were observed between sexes or among sampling regions (Table 3).

Estimates for the 70, 75, and 80% quantiles had confidence intervals that did not overlap zero and indicated that more westerly wintering loons (i.e., in eastern Asia) were exposed to more Hg than those wintering along the coast of North America (Fig. 2). Point estimates for the 85, 90, and 95% quantiles were even greater, but were imprecisely estimated due to sparser data.

### Long-term Trends of Mercury Exposure

Mercury concentrations in secondary coverts and secondary feathers in Yellow-billed Loons ($n$ = 15) were strongly correlated ($r^2$ = 0.97, $F_{14}$ = 410.21, $P$ < 0.001). The regression calculated from this equation [LOG Secondary Hg = -0.032575 + 1.0861684*LOG Secondary Covert Hg] (SE of m = 0.05) was used to convert 25 historical secondary covert samples collected from museum specimens to secondary feather Hg concentrations. Feather Hg concentrations in Yellow-billed Loons varied significantly among sampling periods ($F_{128}$ = 20.10, $P$ < 0.001) (Fig. 3). Pairwise comparisons between sampling periods using Tukey's HSD test indicated that feather samples collected after 2000 ($\bar{x}$ = 8.01 ± 0.45 µg/g fw; $n$ = 104) were significantly higher than those collected Pre-1920 ($\bar{x}$ = 3.81 ± 0.40 µg/g fw; $n$ = 19; $q$ = 1.96, $P$ < 0.001). The other two pairwise comparisons, involving the middle time period ($\bar{x}$ = 4.97 ± 0.76 µg/g fw; $n$ = 6) were not significant ($P$ > 0.05).

## DISCUSSION

Our results indicate current Hg concentrations for Yellow-billed Loons on their breeding range are generally below background levels (i.e., blood Hg 1.0 µg/g ww) and are consistent with earlier work

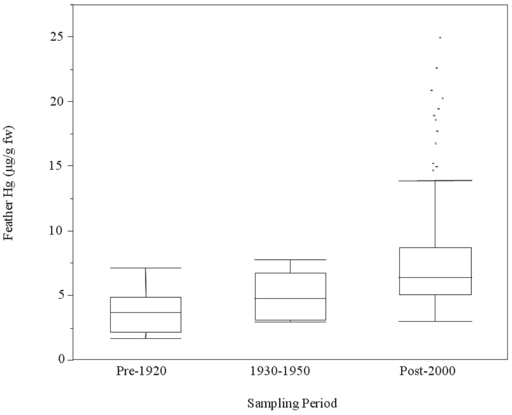

**Figure 3. Median feather mercury (Hg) (µg/g fw) concentrations in Yellow-billed Loons sampled during three time periods, Pre-1920 (*n* = 19), 1930-1950 (*n* = 6), and Post-2000 (*n* = 104, this study), across the three study areas in the Arctic Coastal Plain and Seward Peninsula, Alaska, USA, and east to the Daring Lake region, Northwest Territories, Canada. Each of these three study regions are represented in each of the three sampling periods.**

conducted on breeding Common Loons in southern, boreal Alaska (Evers *et al.* 1998). Arctic environmental Hg loads primarily originate from atmospheric deposition. Historically, sources of deposition were natural (e.g., volcanic), whereas today the origin is primarily anthropogenic through current long-range transport (e.g., release from the burning of coal for industrial purposes) or re-emissions from ocean legacy sources (Corbitt *et al.* 2011; Mason *et al.* 2012). Once Hg is deposited in Arctic ecosystems, it is then made available through *in situ* methylation within lakes and their watersheds (St. Louis *et al.* 2005; Leitch *et al.* 2007). Methylmercury is transferred through the food web and biomagnifies as it moves into higher trophic level species and is well studied in temperate ecosystems (Driscoll *et al.* 2007) and increasingly in Arctic ecosystems (Kirk *et al.* 2012). Methylation and demethylation processes are dependent on many biogeochemical factors and can vary greatly among watersheds and even within watersheds and lakes. Such landscape variability creates significant challenges for determining spatial gradients of risk to Yellow-billed Loons. Mercury input from the surrounding watershed (Gantner *et al.* 2010b) and food web length appear to be important drivers for MeHg transfer to upper trophic organisms in Arctic lakes and subsequent variability (Chételat *et al.* 2008; Gantner *et al.* 2010a).

Ultimately, Yellow-billed Loon body burdens of MeHg are dictated by their prey, and as obligate piscivores, understanding preferred fish diet and determining Hg concentrations by fish species and size class will provide an ability to predict loon Hg concentrations based on models with Common Loons. For instance, among Common Loons Evers *et al.* (2008b) found that average prey fish Hg concentrations of 0.16 µg/g ww (total Hg analyzed as whole body) relate to a 40% decline in the number of fledged chicks per territorial pair. Similarly, Depew *et al.* (2012) determined that 0.18 µg/g ww of Hg in fish (whole body analyses) was responsible for a 50% decline in overall productivity in Common Loons. Arctic lakes are known to contain fish of relevant prey size (< 25 cm; Barr 1996; Haynes *et al.* 2013) and species (e.g., cisco [*Coregonus* spp.]) that approach or exceed 0.16 µg/g ww (Power *et al.* 2002), especially for land-locked Arctic char (*Salvelinus alpinus*) (Swanson *et al.* 2011).

Preliminary findings indicate that the majority of Yellow-billed Loons breeding on the ACP winter in eastern Asia (especially offshore of northern Japan), while half of the breeding population on the Seward Peninsula winter in eastern Asia and half along the Aleutian Islands, Alaska (J. A. Schmutz, pers. commun.). The majority (60%) of Yellow-billed Loons from the Daring Lake study site overwintered in Hecate Strait, British Columbia, and the remaining individuals overwintered as far west as the south-central shoreline of the Alaska Peninsula (Schmutz *et al.* 2014). These tracking efforts indicate that individuals from distinct breeding populations do not necessarily winter together, but the majority of the Yellow-billed Loons sampled within this study, principally those from the ACP, overwinter in eastern Asia. Because the Yellow-billed Loon undergoes a full remigial molt during winter (likely between December and February), the sampling of secondary feathers from individuals on their breeding territories allows a determination of dietary uptake of MeHg in marine waters of eastern Asia.

Past studies demonstrate that elevated body burdens of contaminants (e.g., polychlorinated biphenyls) in avian piscivores are accumulated while overwintering in eastern Asia (Kunisue *et al.* 2002; Minh *et al.* 2002; Schmutz *et al.* 2009). Yellow-billed Loons follow this contaminant loading pattern. Based on winter movements of transmittered individuals, there is a significant relationship with higher feather Hg concentrations in individuals overwintering to the west along the Asian coast (Fig. 2). Locally elevated atmospheric deposition (Jaffe and Strode 2008; Pan *et al.* 2008) and watershed input of Hg to eastern Asia marine waters likely contribute to this Hg exposure pattern.

Overall, feather Hg concentrations of Yellow-billed Loons sampled from 2007 to 2012 provided the most compelling evidence of risk. Seven percent of the feather samples exceeded Hg concentrations of approximately 20.0 µg/g fw, which relate to projected lowered reproductive success in Common Loons (Evers *et al.* 2008b). However, exposure to Hg in Common Loons is greatest during the breeding season, whereas for Yellow-billed Loons it is currently greatest during the winter. Therefore, it is uncertain how MeHg loading during the winter may contribute to lowered reproductive output of Yellow-billed Loons.

Some insight into the contribution of wintertime exposure on breeding success lies within the blood-egg relationship of both loon species. There is a strong and significant relationship between blood and eggs for Common Loons ($r^2 = 0.79$, $n = 108$; Evers *et al.* 2003). Because adult female blood Hg concentrations of Common Loons strongly correlate with prey fish on their breeding territory (Burgess and Meyer 2008), it is well understood that egg Hg concentrations primarily reflect dietary uptake of MeHg from the breeding territory. However, there is a poor blood-egg relationship for Yellow-billed Loons (based on a limited number of eggs), which indicates that individual Hg body burdens accumulated in marine waters during winter and spring have a greater contribution to eggs than observed in Common Loons. Such observation and inference is consistent with the short interval between arriving at breeding areas and egg laying for Yellow-billed Loons (Schmutz *et al.* 2014) as compared to the long interval for Common Loons (Evers *et al.* 2010). Therefore, winter and spring exposure of Hg to Yellow-billed Loons may have a direct relationship to egg Hg concentrations, which may explain why breeding Yellow-billed Loons can have elevated egg Hg concentrations (i.e., > 1.0 µg/g ww) while corresponding blood Hg concentrations are low (i.e., < 1.0 µg/g ww). Larger sample sizes are necessary to determine if this observation is representative of the ACP and other areas with breeding Yellow-billed Loons.

Feather Hg concentrations measured in Yellow-billed Loons since 1845 have increased two-fold (Fig. 3). Our finding of this increase over this time period was expected and is consistent with sediment Hg concentrations over a similar time period from multiple sites in North America, where global atmospheric deposition was three to four times higher than pre-industrial times (Swain *et al.* 1992; Kamman and Engstrom 2002; Engstrom *et al.* 2007; Drevnick *et al.* 2012).

While contemporary blood Hg concentrations in Yellow-billed Loons generally remain under background levels defined for Common Loons for North America (Evers *et al.* 1998), egg and feather Hg concentrations for some individuals are elevated. Projections of water Hg concentrations in the North Pacific Ocean indicate a four-fold increase from current levels by 2050 (Sunderland *et al.* 2009; E. M. Sunderland, pers. commun.). Such increases could be even more pronounced within nearshore marine waters of the eastern Asian coast because of additional inputs from watersheds (Mason *et al.* 2012). Therefore, winter Hg body burdens could significantly increase over current levels and, because there may be a transfer of winter dietary uptake of MeHg to eggs, reproductive success could be adversely impacted. The future overall impact to Yellow-billed Loons from projected increasing environmental Hg emissions, deposition, and re-emissions (from legacy sources) will depend on changes within Arctic ecosystems related to permafrost thawing that are linked to changing climatic patterns (Schuur *et al.* 2009).

Loons are often-used as bioindicators of environmental quality because of their high trophic position and sensitivity to contaminants (Evers 2006). It is now well established that the Common Loon is experiencing adverse reproductive effects due to elevated environmental Hg loads (Burgess and Meyer 2008; Evers *et al.* 2003, 2008b, 2011; Schoch *et al.* 2014). Because of increases in global anthropogenic Hg emissions, spatial heterogeneity within a landscape, and changes in climatic patterns that may positively influence the remobilization of legacy environmental Hg loads and methylation rates, an assessment of the risk of Hg to Yellow-billed Loons within their breeding and wintering areas is timely. Biological Hg hotspots can form within areas of particular sensitivity to MeHg availability, even though the atmospheric input of Hg is relatively low (Driscoll *et al.* 2007; Evers *et al.* 2007). A better understanding for assessing risk to loons is now available, where effects concentrations in multiple tissues (i.e., blood, feathers and eggs) and prey can be confidently related to percent reduction in overall reproductive success (Depew *et al.* 2012) across broad landscapes.

Because of the uncertain relationship between Hg concentrations in air, water and sediment with high trophic level biota, the determination of spatial gradients and temporal trends on the magnitude of MeHg food web transfer requires suitable indicator organisms. The Yellow-billed Loon (and other loon species) are good high-trophic level indicators of Hg exposure across space (breeding and wintering areas) and time (short- and long-term). Blood provides a confident approach for examining short-term Hg exposure. Feathers collected from Yellow-billed Loons captured on their breeding territories indicate MeHg dietary uptake during the mid-winter molt are useful for determining Hg exposure along the eastern Asian nearshore waters during present and past time periods. Egg Hg for Yellow-billed Loons likely reflects a mixed Hg signal from wintering areas and breeding territories. The use of these three tissues provides a robust approach for monitoring Hg in both Arctic freshwater systems and marine ecosystems of eastern Asia. Such monitoring should be integrated into national programs of the United States (Mason *et al.* 2005; Evers *et al.* 2008a; Schmeltz *et al.* 2011) and Canada (Morrison 2011) and international efforts, such as the Global Mercury Observation System (Pirrone and Mason 2009) for the global Hg treaty (United Nations Environment Programme 2013), especially in light of increasing Hg emissions, watershed runoff of MeHg, and thawing of permafrost and pack ice.

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