Published in final edited form as:

Environ Toxicol Chem. 2012 November ; 31(11): 2647–2652. doi:10.1002/etc.1990.

MERCURY CONCENTRATIONS OF A RESIDENT FRESHWATER FORAGE FISH AT ADAK ISLAND, ALEUTIAN ARCHIPELAGO, ALASKA

Leah A. Kenney[†], Frank A. von Hippel^{†,*}, James J. Willacker^{†,‡}, and Todd M. O'Hara[‡]

[†]Department of Biological Sciences, and Environment and Natural Resources Institute, University of Alaska Anchorage, Anchorage, Alaska, USA

[‡]Department of Biology and Wildlife, University of Alaska Fairbanks, Fairbanks, Alaska, USA

Abstract

The Aleutian Archipelago is an isolated arc of over 300 volcanic islands stretching 1,600 km across the interface of the Bering Sea and North Pacific Ocean. Although remote, some Aleutian Islands were heavily impacted by military activities from World War II until recently and were exposed to anthropogenic contaminants, including mercury (Hg). Mercury is also delivered to these islands via global atmospheric transport, prevailing ocean currents, and biotransport by migratory species. Mercury contamination of freshwater ecosystems is poorly understood in this region. Total Hg (THg) concentrations were measured in threespine stickleback fish (*Gasterosteus aculeatus*) collected from eight lakes at Adak Island, an island in the center of the archipelago with a long military history. Mean THg concentrations for fish whole-body homogenates for all lakes ranged from 0.314 to 0.560 mg/kg dry weight. Stickleback collected from seabird-associated lakes had significantly higher concentrations of THg compared to non-seabird lakes, including all military lakes. The $\delta^{13}\text{C}$ stable isotope ratios of stickleback collected from seabird lakes suggest an input of marine-derived nutrients and/or marine-derived Hg.

Keywords

Biotransport; Carbon isotopes; Marine-derived mercury; Threespine stickleback; Formerly used defense sites

INTRODUCTION

Mercury (Hg) is a ubiquitous environmental pollutant that is known to bioaccumulate and biomagnify in food webs [1]. Mercury enters ecosystems through natural sources such as volcanism, as well as a variety of anthropogenic activities, including mining and the burning of coal. Once deposited, inorganic Hg is converted by bacteria into the more toxic methylmercury (MeHg). Nearly all Hg found in biota is incorporated in the form of MeHg [2]. Methylmercury in wildlife and humans can impair or suppress the nervous,

cardiovascular, and endocrine systems [3], decrease reproductive success [4], and disrupt development [5]. Elevated levels of Hg have been found in arctic ecosystems despite the paucity of local anthropogenic sources [6].

The Aleutian Archipelago (hereafter, Aleutians) is an isolated arc of over 300 volcanic islands stretching 1,600 km that geographically separates the North Pacific Ocean from the Bering Sea. Many Aleutian Islands were impacted by military activities during World War II and continuing until the end of the Cold War in the 1990s, and were exposed to a variety of anthropogenic contaminants including Hg [7]. Global distillation and the biotransport of contaminants by migratory species enhance delivery of pollutants from lower latitudes into this remote region [6].

Mercury contamination of marine and migratory species has been documented throughout the Aleutians [8–11], but we know of only one study that has examined Hg contamination in a freshwater fish (C. Jeitner, 2010, Master's thesis, Rutgers University, New Brunswick, NJ, USA). Several studies documented Hg and organochlorine compounds in marine mammals and birds collected at islands with a history of military activity [8,9,12], whereas other studies found that concentrations of contaminants in biota increased westward along the chain regardless of military history [10,11,13]. The general trend toward increased levels of contaminants in the western Aleutians has been attributed to the dynamic weather systems that build in this region and pull storms and contaminants from Asia eastward along the archipelago [6].

Military activities and long-range transport have been implicated as important contributors of Hg contamination in the Aleutians, but the biotransport of Hg by seabirds to freshwater systems has not been studied. The Aleutian Archipelago provides an essential breeding habitat for over 10 million seabirds [14] that utilize both freshwater and marine ecosystems during the breeding season [15,16]. Seabirds are top predators of marine food webs that forage at sea and return to colony sites where they deposit marine-derived nutrients and contaminants in guano and tissues [15,16]. In addition to time spent at colony sites, seabirds spend considerable time resting and bathing on inland freshwater lakes and ponds. Consequently, seabirds have the potential to biotransport contaminants on a large scale (from wintering to breeding grounds) and on a local scale (daily movements between marine and freshwater environments). Given that Hg both bioaccumulates and biomagnifies, seabirds have the potential to deliver large quantities of marine-derived MeHg to freshwater systems.

Stable isotopes of carbon ($\delta^{13}\text{C}$) have been used elsewhere to relate seabird activities with Hg contamination and trace the sources and pathways of Hg in aquatic environments [17,18]. The ratio of carbon isotopes changes minimally as carbon moves between a food source and its consumer, thus allowing for the identification of the source of carbon [19,20]. Although differences in the $\delta^{13}\text{C}$ of primary producers can cause variation in the $\delta^{13}\text{C}$ of consumers within and between systems, marine-derived carbon is typically more enriched than freshwater carbon and has been widely used to track marine inputs into freshwater systems [21–23].

The threespine stickleback (*Gasterosteus aculeatus*) is a ubiquitous fish that is commonly used as a model for toxicology studies [24,25] and is the most widely distributed resident freshwater fish in the Aleutians (L. Kenney, 2011, Master's thesis, University of Alaska Anchorage, Anchorage, AK, USA). Stickleback are generalist consumers of both benthic and limnetic animal prey [26]. Stickleback are an important food resource for predatory fish including coho salmon (*Oncorhynchus kisutch*) and Dolly Varden (*Salvelinus malma*) [27], as well as for waterbirds such as red-throated loons (*Gavia stellata*), red-breasted mergansers (*Mergus serrator*), and Aleutian terns (*Onychoprion aleuticus*; <http://bna.birds.cornell.edu/BNA>).

Previous studies of Hg contamination in the Aleutians focused on top predators that are usually long-lived and marine species. Potential biases related to dietary changes over multiple life stages and migration behavior of top predators make attributing sources of Hg difficult. In contrast, resident-fresh-water stickleback (hereafter, stickleback) have the potential to reflect local Hg exposure, given their short life span (typically two years) and small home ranges. Our objective is to examine potential sources of Hg contamination by comparing Hg levels in stickleback resident in lakes that vary in seabird use and past military activity, but are unlikely to vary in exposure to atmospheric deposition. We compared Hg concentrations of stickleback at Adak Island that were collected from lakes within watersheds with known military disposal sites, from lakes with active seabird use, and from a lake that is neither adjacent to a military site nor regularly used by seabirds. Stable isotope ratios of carbon were used to trace marine inputs at lakes and determine if these inputs were associated with increased Hg concentrations in stickleback.

MATERIALS AND METHODS

Study area

Adak Island (51.87°N, 176.63°W) is located in the center of the Aleutian Archipelago and is part of the Alaska Maritime National Wildlife Refuge (AMNWR; Fig. 1). The island covers an area of 75,112 ha (~39 × 45 km) and is composed of rugged mountainous terrain surrounded by broad rolling lowlands. The lowland habitat is characterized by many lakes and streams and two tidal lagoons.

The Aleutian Islands were established as a National Wildlife Refuge in 1913, and Adak was added to the National Wildlife Refuge System in 1940. From 1942 to 1997, the United States military occupied the northern portion of the island, where the present study takes place. Owing to contamination from military activities, Adak was placed on the U.S. Environmental Protection Agency's (U.S. EPA) National Priorities List in 1994 (http://www.atsdr.cdc.gov/HAV/pha/adak/Ada_p3.html). Adak's population has fluctuated in size from tens of thousands of military personnel during World War II to a current population estimate of approximately 200 people. Land ownership of Adak is shared by the AMNWR and the Aleut Corporation.

All lakes sampled in the present study are oligotrophic and broadly similar in morphometry and water chemistry (Table 1). The estimated surface area of lakes ranges from 3 to 20 hectares and altitudes range from near sea level to approximately 72 m above sea level. Lake

bottoms are comprised of bedrock-cobble or finer sediments. Aquatic vegetation is sparse and dominated by *Hippurus vulgaris* and *Ranunculus trichophyllus*. Stickleback and Dolly Varden are the most common fish in lakes sampled. The weather is typical of a northern maritime climate with average summer temperatures of 8.8°C at sea level and an annual average of 4.8°C. Mean annual rainfall is approximately 166 cm and annual snowfall averages 250 cm.

Field collections

Eight lakes were sampled for stickleback from June 28 to July 9, 2010. Thirty stickleback at each lake were collected for a total of 240 individuals. Lakes were chosen based on proximity (i.e., within the watershed) to known military disposal sites (hereafter, military lakes, $n = 5$), lack of association to military disposal sites (lakes with no known military disposal sites within that watershed, hereafter, nonmilitary lakes, $n = 1$), or common use by seabirds (hereafter, seabird lakes, which are also nonmilitary lakes, $n = 2$, Fig. 1).

Military lakes were selected based on Navy documentation of disposal sites (e.g., landfills, metals dumps, or disposal of unknown materials) that were within the associated watershed [7]. Bird observations were made daily during the collection period and opportunistically from May to September during 2002 and from 2006 to 2010 by AMNWR personnel and the authors. Seabird use at each lake was based on observations of birds resting, bathing, and/or foraging during the breeding season. Based on the frequency of birds present, two lakes were classified as seabird lakes (seabirds present > 80% of observation days) and all other study lakes were classified as non-seabird lakes (seabirds present < 20% of observation days). Approximate numbers of birds present during the fish collection period (June 28–July 9, 2010) on both seabird lakes were recorded before traps were set to avoid flushing birds.

Stickleback were trapped using unbaited 0.6 cm mesh minnow traps set at 0.5 to 2.0 m depth for 24 h. Stickleback were euthanized using an overdose of MS-222 anesthetic, rinsed in lake water, blotted dry with a clean paper towel, placed in Ziploc bags as one continuous layer, and stored on ice packs in the field (< 2 h). Fish were transferred to a –20°C freezer where they were stored for 1 week, then transferred to a –80°C freezer until laboratory analysis.

Temperature, pH, salinity, and conductivity were measured with a YSI Professional Plus meter at 0.5 to 1.0 m depth at each trap location and the mean for all measurements was reported for each lake.

Sample preparation

Stickleback were removed from the –80°C freezer and allowed to partially thaw before processing. Each fish received a unique identification number. Standard length (SL, anterior tip of premaxilla to posterior border of hypural plate) was measured with digital calipers (± 0.01 mm). Stomach contents were removed with stainless steel forceps and discarded, and the stomach was placed back into the fish. Additionally, *Schistocephalus solidus*, a common stickleback tapeworm, were removed from the body cavity of the fish and discarded. Each fish was placed in a uniquely labeled sterile Whirl Pak and freeze-dried for 48 to 72 h.

Whole fish were cut using stainless steel scissors to mince bony elements and skin, and then homogenized to a fine powder by hand using a ceramic mortar and pestle. All utensils were thoroughly cleaned with lab-grade soap and deionized water and sterilized with 70% ethanol between sample preparations. Homogenates were stored in sterile Whirl Paks until processing.

Mercury analysis

Total Hg (THg) concentrations were analyzed as a surrogate for MeHg concentrations in stickleback tissues, given that almost all Hg in fish is in the form of MeHg [28]. Total Hg concentrations for approximately 25 ± 4 mg of whole-fish homogenate were determined using a Milestone Direct Mercury Analyzer (DMA-80, Milestone) at the Wildlife Toxicology Laboratory, University of Alaska, Fairbanks. Quality assurance and quality control followed standard laboratory procedures, which included method blanks, method blank spikes, sample spikes, sample duplicates, and standard reference materials, run with each batch of 20 to 30 samples. The standard reference materials consisted of dried fish protein homogenate (DORM-3, THg = 0.382 ± 0.060 mg/kg dry wt) obtained from the National Research Council of Canada. All samples were initially analyzed in duplicate with a coefficient of variation (CV) <10%. Five samples had CVs >10%; therefore, triplicates ($n = 1$) or quadruplicates ($n = 4$) were run, giving these five samples a mean CV of 10.6%. The overall mean CV on all samples was 4.1%. Recovery rates for method blank spikes, sample spikes, and standard reference materials ranged from 93.6 to 101.7%.

Mercury data are presented on a dry-weight basis. To facilitate comparisons with other studies in which only wet-weight values were reported, percentage of moisture was calculated for a subset of samples ($n = 70$). Following the removal of stomach contents and parasites, whole-body wet weights were measured ± 0.001 g. Fish were then freeze-dried for 52 to 65 h and dry weights were obtained to the nearest ± 0.001 g.

Isotope analysis

For each sample, approximately 0.75 ± 0.05 mg of homogenate was weighed, sealed in a tin capsule, and analyzed for stable carbon isotope ratio ($^{13}\text{C}/^{12}\text{C}$) at the University of Alaska Anchorage–Environment and Natural Resources Institute Stable Isotope Laboratory using a Finnigan MAT DeltaPlusXL Isotope Ratio Mass Spectrometer (IRMS) directly coupled to a Costech Elemental Analyzer (ESC 4010). Isotope ratios are presented in standard δ notation as parts per thousand (‰) differences between the isotope ratio of the sample and that of a standard (Vienna Pee Dee Belemnite) using the formula

$$\delta^{13}\text{C} = [(R_{\text{sample}} - R_{\text{standard}})/R_{\text{standard}}] \times 1000$$

where R is the ratio $^{13}\text{C}/^{12}\text{C}$ in the sample and standard material. Samples depleted in the heavier isotope (^{13}C) in comparison to the standard have lower delta values. Replicates of a reference material (SRM 1547: peach leaves) from the National Institute of Standards and Technology ($\delta^{13}\text{C} = -25.89 \pm 0.3\%$) and our laboratory-working standard (Cheney Lake, Alaska, threespine stickleback, $\delta^{13}\text{C} = -29.50 \pm 0.2\%$) were analyzed with every batch of 40 samples. Analytical error for carbon isotope analysis was $\pm 0.2\%$. Carbon isotope ratios were not corrected for lipid content because lipid values in threespine stickleback are

consistently low (2–3%; personal communication, J. Willacker, University of Alaska Anchorage, Anchorage, AK, USA) and thus lipid correction has a negligible impact [20].

Statistical methods

All statistics were calculated using the SPSS package v18. Data normality and variance homogeneity were assessed prior to statistical analysis using the Shapiro-Wilks test for goodness-of-fit to normal distribution at $\alpha = 0.05$ [29]. Mercury concentrations and the $\delta^{13}\text{C}$ for each lake met assumptions of normality and variance homogeneity; therefore, no data transformations were necessary. Total Hg was positively and linearly correlated with fish SL ($r = 0.35$, $n = 240$, $p < 0.001$). Thus, THg concentrations were size corrected by calculating the THg at the mean SL across all lakes ($\bar{x} = 49.3$ mm, $n = 240$), regressing SL against THg within each lake and using the residuals to adjust individual THg values by the mean SL across lakes. Variation in size-adjusted THg concentration between lakes was tested with one-way analysis of variance (ANOVA), and a Tukey's honestly significant difference (HSD) post hoc test was used to determine pair-wise comparisons between lakes.

Analysis of variance was also used to compare $\delta^{13}\text{C}$ isotopic variation of stickleback between lakes. When a significant main effect was observed, a Tukey's HSD post hoc test was used to determine pair-wise comparisons between lakes. A general linear model (GLM) was used to determine if $\delta^{13}\text{C}$ explained a significant amount of variation in THg concentrations between lakes. Spearman rank correlation was used to examine potential relationships between lake variables (lake area, temperature, and pH) and THg. All data are expressed on a dry-weight basis $\pm$ standard deviation (SD). All tests are two-tailed with a critical $\alpha = 0.05$.

RESULTS

Water quality variables and lake area were similar across all lakes and none were significantly correlated with THg (Table 1). Glaucous-winged gull (*Larus glaucescens*), Aleutian tern, and red-breasted merganser were the most commonly observed seabirds at study lakes. Mean number of birds observed on each seabird lake before traps were set (± 1 SD) was 28.2 ± 5.8 (Lake Shirley) and 13.8 ± 3.9 (Lake Ronnie). Glaucous-winged gulls were the most abundant species. The most common activities of all seabirds were resting and bathing.

Mean stickleback wet weight was 1.12 ± 0.47 g and mean moisture content of stickleback was $76.9 \pm 2.3\%$ (range = 70.0–82.1%; $n = 70$). Mean SL of stickleback was 49.3 ± 6.7 mm (range = 35–67 mm; $n = 240$). Mean size-adjusted THg concentrations for stickleback for all lakes ranged from 0.316 to 0.542 mg/kg (Table 1, Fig. 2) and differed significantly between lakes ($F_{7,232} = 18.09$, $p < 0.001$). Total Hg concentrations in stickleback were significantly higher in lakes associated with seabird use (Tukey HSD; $p < 0.001$, Fig. 2).

The $\delta^{13}\text{C}$ values of stickleback from all lakes ranged from -29.00 to -20.92% ($\bar{x} = -26.41 \pm 2.37\%$, $n = 240$) and differed significantly between lakes ($F_{7,232} = 97.33$, $p < 0.001$, Fig. 2). Stickleback from seabird lakes were significantly more enriched in $\delta^{13}\text{C}$ (Tukey HSD; $p <$

0.001, Fig. 2) and $\delta^{13}\text{C}$ explained a significant amount of THg variation between lakes (37.3%, $F_{1,7} = 4.50$, $p = 0.035$).

DISCUSSION

Mercury sources

Identification of contaminated sites at Adak began under the Navy Assessment and Control of Installation Pollutants program in 1986. By 1992, 185 contaminated sites were identified and in 1994 Adak was placed on the U.S. EPA's National Priorities List. A wide variety of materials were documented at military disposal sites associated with lakes sampled in the present study, including Hg containing products [7]. All contaminated sites in the present study underwent remediation efforts, including testing of groundwater, surface water, sediments, and soils. In the present study, stickleback collected from nonmilitary lakes (which were also seabird lakes) had the highest concentrations of THg.

Atmospheric transport of Hg from fossil fuel combustion is the primary pathway of Hg to arctic regions [6]. Many Aleutian Islands experience the additional local source of Hg from regular volcanic activities. Inorganic Hg from volcanic emissions can enter aquatic ecosystems where, like other inorganic Hg, it is converted to the more toxic MeHg and biomagnifies through food webs. In addition to regular volcanic emissions, prominent volcanic eruptions have occurred recently in the Aleutians, including the Kasatochi Island (80 km northeast of Adak) eruption during August 2008 that deposited several millimeters of ash on Adak and the surrounding area. All Aleutian Islands are volcanic in origin, but the potential contribution of volcanic Hg at some island groups is likely more important than at others, such as the Near Islands that lack active volcanoes. To better understand distant versus local sources of Hg pollution in the Aleutians, future research should investigate differential volcanic Hg deposition by island group as well as latitudinal trends related to long-range transport from lower latitude fossil fuel combustion.

Seabird biotransport

Biological transport of contaminants and marine-derived nutrients to freshwater systems has been documented primarily in salmon [30,31]. However, recent studies have focused on the role of seabirds in the transport and release of contaminants in their guano and tissues at colony sites [15,18,32]. Although lakes in our study area were not used for colonial nesting, they were used daily for resting and bathing and represent an additional nexus for marine derived Hg and nutrients to enter freshwater ecosystems.

In the present study, stickleback collected from seabird lakes had significantly enriched $\delta^{13}\text{C}$ values and significantly higher THg concentrations compared to stickleback from non-seabird lakes. Furthermore, $\delta^{13}\text{C}$ values for other resident freshwater threespine stickleback populations range from -31.3 to -26.4% [33,34] and for anadromous stickleback range from -22.1 to -21.3% (personal communication, J. Willacker, University of Alaska Anchorage, Anchorage, AK, USA). These values are consistent with our mean values of -27.5% for stickleback from non-seabird lakes (freshwater signature) and -23.2% for stickleback from

seabird-associated lakes (marine signature). Despite our small sample sizes, these data indicate that marine-derived materials, and likely Hg, are being deposited into these lakes.

Whether seabirds are directly supplying marine-derived Hg to lakes and/or are indirectly increasing MeHg production via the delivery of marine-derived nutrients remains unclear. Recent studies documented that seabird guano contains significant amounts of MeHg [35]. Consequently, seabirds could provide a direct input of bioavailable marine-derived MeHg to these lakes. However, seabirds could be indirectly increasing bacterial methylation of Hg already present through the introduction of enhanced nutrient subsidies in their guano. Conversely, excess nutrient loadings can eutrophy lakes and thereby inhibit methylation of Hg [36]. Adak lakes are currently oligotrophic, so both MeHg loading and enhanced methylation due to nutrient loading are possible. The environmental factors that control the conversion of inorganic Hg to MeHg are complex and can differ greatly depending on specific lake variables, such as sediment organic matter, dissolved organic carbon, pH, and temperature [37]. Future monitoring should include the suite of factors that drive Hg methylation.

Implications for wildlife and humans

Mean THg wet-weight concentrations in stickleback for all lakes sampled at Adak are below the U.S. EPA advisory levels for human consumption of freshwater fishes (0.3 mg/kg wet wt; <http://water.epa.gov/scitech/swguidance/fishshellfish/fishadvisories/index.cfm>). However, mean THg concentrations of stickleback for all lakes ($\bar{x} = 0.095$ mg/kg; range = 0.073–0.125 wet wt) exceed the advisory level for fish-eating wildlife (0.033 mg/kg wet wt) that Canadian [38] and California [39] agencies have set for whole-bodied forage fish. Levels of Hg in forage fish that exceed wildlife consumption advisories pose a direct health risk to fish-eating wildlife, especially long-lived, high-trophic level animals. In the Aleutians, sticklebacks are an important component of the diet of many wildlife species, especially waterbirds. These include species of conservation concern, such as the red-throated loon.

Due to the biomagnification of Hg through food webs, elevated levels of Hg in forage fish also pose a health risk to residents living at remote islands who rely on piscivorous fish as well as seabirds and their eggs for subsistence food. Therefore, Hg monitoring in forage fish provides important information regarding human health risks associated with consuming higher trophic level animals.

Sources of Hg contamination in this region are still poorly understood, but the present study highlights the need to assess Hg biotransport in addition to other Hg inputs to lakes. Future study at a broader geographic scale across the North Pacific coastline would help to resolve the relative importance of contaminated military sites, atmospheric deposition, biotransport, and local factors driving Hg methylation.

Acknowledgement

Invaluable logistical support was provided by the U.S. Fish and Wildlife Service-Alaska Maritime National Wildlife Refuge, especially from J. Williams, V. Byrd, and L. Spitler. We thank R. Kaler for field assistance and C. Lieske and M. Castellini for laboratory assistance. We thank R. Kaler, J. Welker, and two anonymous reviewers for

comments on earlier versions of this manuscript. Research reported in this publication was supported by the National Institute of General Medical Sciences of the National Institutes of Health under Award Number P20GM103395. The content is solely the responsibility of the authors and does not necessarily represent the official views of the National Institutes of Health. All collections during the present study were done in accordance with the University of Alaska's Animal Care and Use Committee (protocol 159870-11) and were authorized by permits from the Alaska Department of Fish and Game (SF2010-029), the U.S. Fish and Wildlife Service, and the Aleut Corporation (063010).

REFERENCES

1. U.S. Environmental Protection Agency. Integrated risk information system. Washington D.C.: Office of Water; 2010. Methylmercury (MeHg); CASRN 22967-92-6.
2. Watras CJ, Back RC, Halvorsen S, Hudson RJM, Morrison KA, Wentz SP. Bioaccumulation of mercury in pelagic freshwater food webs. *Sci Total Environ*. 1998; 219:183–208. [PubMed: 9802248]
3. Walker, CH.; Hopkin, SP.; Sibly, RM.; Peakall, DB. Principles of Ecotoxicology. London, England: Taylor and Francis; 1996.
4. Scheuhammer AM, Meyer MW, Sandheinrich MB, Murray MW. Effects of environmental methylmercury on the health of wild birds, mammals, and fish. *Ambio*. 2007; 36:12–19. [PubMed: 17408187]
5. Watanabe C, Satoh H. Evolution of our understanding of methylmercury as a health threat. *Environ Health Perspect*. 1996; 104:367–379. [PubMed: 9182044]
6. Arctic Monitoring and Assessment Programme. Arctic pollution 2002 (persistent organic pollutants, heavy metals, radioactive human health, changing pathways). Oslo, Norway: 2002.
7. U.S. Navy, U.S. Environmental Protection Agency, and Alaska Department of Environmental Conservation. Former Adak Naval Complex declaration of the record of decision, operable unit A. Poulso, WA: Engineering Field Activity; 1999.
8. Anthony RG, Miles AK, Estes JA, Isaacs FB. Productivity, diets, and environmental contaminants in nesting bald eagles from the Aleutian Archipelago. *Environ Toxicol Chem*. 1999; 18:2054–2062.
9. Burger J, Gochfeld M, Sullivan K, Irons D. Mercury, arsenic, cadmium, chromium, lead, and selenium in feathers of pigeon guillemots (*Cephus columba*) from Prince William Sound and the Aleutian Islands of Alaska. *Sci Total Environ*. 2007; 387:175–184. [PubMed: 17765292]
10. Ricca MA, Miles KA, Anthony RG. Sources of organochlorine contaminants and mercury in seabirds from the Aleutian archipelago of Alaska: Inferences from spatial and trophic variation. *Sci Total Environ*. 2008; 406:308–323. [PubMed: 18692865]
11. Rocque DA, Winker K. Biomonitoring of contaminants in birds from two trophic levels in the North Pacific. *Environ Toxicol Chem*. 2004; 23:759–766. [PubMed: 15285370]
12. Bacon CE, Jarman WM, Estes JA, Simon M, Norstrom RJ. Comparison of organochlorine contaminants among sea otter (*Enhydra lutris*) populations in California and Alaska. *Environ Toxicol Chem*. 1999; 18:452–458.
13. Anthony RG, Miles AK, Ricca MA, Estes JA. Environmental contaminants in bald eagle eggs from the Aleutian Archipelago. *Environ Toxicol Chem*. 2007; 26:1843–1855. [PubMed: 17702538]
14. Byrd GV, Renner HM, Renner M. Distribution patterns and population trends of breeding seabirds in the Aleutian Islands. *Fish Oceanogr*. 2005; 14:139–159.
15. Blais JM, Kimpe LE, McMahon D, Keatley BE, Mallory ML, Douglas MSV, Smol JP. Arctic seabirds transport marine-derived contaminants. *Science*. 2005; 309:445. [PubMed: 16020729]
16. Croll DA, Maron JL, Estes JA, Danner EM, Byrd GV. Introduced predators transform subarctic islands from grassland to tundra. *Science*. 2005; 307:1959. [PubMed: 15790855]
17. Brimble SK, Foster KL, Mallory ML, Macdonald RW, Smol JP, Blais JM. High Arctic ponds receiving biotransported nutrients from a nearby seabird colony are also subject to potentially toxic loadings of arsenic, cadmium, and zinc. *Environ Toxicol Chem*. 2009; 28:2426–2433. [PubMed: 19604032]

18. Choy ES, Gauthier M, Mallory ML, Smol JP, Douglas MSV, Lean D, Blais JM. An isotopic investigation of mercury accumulation in terrestrial food webs adjacent to an Arctic seabird colony. *Sci Total Environ*. 2010; 408:1858–1867. [PubMed: 20153017]
19. Jardine TD, Kidd KA, Fisk AT. Applications, considerations, and sources of uncertainty when using stable isotope analysis in ecotoxicology. *Environ Sci Technol*. 2006; 40:7501–7511. [PubMed: 17256487]
20. Post DM. Using stable isotopes to estimate trophic position: Models, methods, and assumptions. *Ecology*. 2002; 83:703–718.
21. Kline TC Jr, Wilson WJ, Goering JJ. Natural isotope indicators of fish migration at Prudhoe Bay, Alaska. *Can J Fish Aquat Sci*. 1998; 55:1494–1502.
22. Swanson HK, Kidd KA, Babaluk JA, Wastle RJ, Yang PP, Norman M, Halden NM, Reist JD. Anadromy in Arctic populations of lake trout (*Salvelinus namaycush*): Otolith microchemistry, stable isotopes, and comparisons with Arctic char (*Salvelinus alpinus*). *Can J Fish Aquat Sci*. 2010; 67:842–853.
23. Doucett RR, Hooper W, Power G. Identification of anadromous and nonanadromous adult brook trout and their progeny in the Tabusintac River, New Brunswick, by means of multiple-stable-isotope analysis. *Trans Am Fish Soc*. 1999; 128:278–288.
24. Katasiadaki, I. The use of the stickleback as a sentinel and model species in ecotoxicology. In: Östlund-Nilsson, S.; Mayer, I.; Huntingford, FA., editors. *Biology of the Threespined Stickleback*. Boca Raton, FL, USA: CRC; 2007. p. 319–351.
25. von Hippel, FA. *Tinbergen's Legacy in Behaviour: Sixty Years of Landmark Stickleback Papers*. Leiden, The Netherlands: Brill Academic Pub; 2010.
26. Travis, MP. PhD thesis. Stony Brook, NY, USA: Stony Brook University; 2011. The functional morphology and evolution of feeding modes in threespine stickleback.
27. Neuhold, JM.; Helm, WT. Alexandria, VA, USA: U.S. Department of Commerce, National Technical Information Service; 1971. Freshwater vertebrate and invertebrate ecology of Amchitka Island. Annual Progress Report. NTIS BMI Report #BMI-171-142.
28. Bloom NS. On the chemical form of mercury in edible fish and marine invertebrate tissue. *Can J Fish Aquat Sci*. 1992; 49:1010–1017.
29. Sokal, RR.; Rohlf, FJ. *Biometry: The Principles and Practice of Statistics in Biological Research*. 3rd ed.. New York, NY, USA: W. H. Freeman; 1995.
30. Heintz RA, Wipfli MS, Hudson JP. Identification of marine-derived lipids in juvenile coho salmon and aquatic insects through fatty acid analysis. *Trans Am Fish Soc*. 2010; 139:840–854.
31. Sarica J, Amyot M, Hare L, Doyon M, Stanfield LW. Salmon-derived mercury and nutrients in a Lake Ontario spawning stream. *Limnol Oceanogr*. 2004; 49:891–899.
32. Evenset A, Carroll J, Christensen GN, Kallenborn R, Gregor D, Gabrielsen GW. Seabird guano is an efficient conveyor of persistent organic pollutants (POPs) to Arctic lake ecosystems. *Environ Sci Technol*. 2007; 41:1173–1179. [PubMed: 17593715]
33. McIntyre JK, Beauchamp DA. Age and trophic position dominate bioaccumulation of mercury and organochlorines in the food web of Lake Washington. *Sci Total Environ*. 2007; 372:571–584. [PubMed: 17157357]
34. Power M, Klein GM, Guiguer K, Kwan MKH. Mercury accumulation in the fish community of a sub-Arctic lake in relation to trophic position and carbon sources. *J Appl Ecol*. 2002; 39:819–830.
35. Chen Q, Liu X, Xu L, Sun L, Yan H, Liu Y, Luo Y, Huang J. High levels of methylmercury in guano and ornithogenic coral sand sediments on xisha islands, South China Sea. *Arch Environ Contam Toxicol*. 2012; 63:177–188. [PubMed: 22614866]
36. Pickhardt PC, Folt CL, Chen CY, Klaue B, Blum JD. Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. *P Natl Acad Sci USA*. 2002; 99:4419–4423.
37. Munthe J, Bodaly RA, Branfireun BA, Driscoll CT, Gilmour CC, Harris R, Horvat M, Lucotte M, Malm O. Recovery of mercury-contaminated fisheries. *Ambio*. 2007; 36:33–44. [PubMed: 17408189]
38. Environment Canada. Canadian tissue residue guidelines for the protection of consumers of aquatic life: Methylmercury. In: *National Guidelines and Standards Office EQB*, Environment Canada. ,

editor. Ecosystem Health: Science-Based Solutions. Gatineau, Quebec, Canada: Environment Canada; 2002.

39. San Francisco Bay Regional Water Quality Control Board. Proposed basin plan amendment and staff report for revised total maximum daily load (TMDL) and proposed mercury water quality objectives. Oakland, CA, USA: 2006. p. 116

Fig. 1. Study sites at Adak Island, Aleutian Archipelago, Alaska, USA, 2010. Lakes sampled for total mercury (THg) analyses of whole-bodied threespine stickleback are labeled. Lakes within watersheds with military disposal sites are underlined and asterisks represent seabird lakes.

Fig. 2. Size-adjusted mean total mercury (THg) concentrations (± 1 standard error) and mean $\delta^{13}\text{C}$ values (± 1 standard error) of threespine stickleback from lakes at Adak Island, Aleutian Archipelago, Alaska, USA, 2010. Underlined names indicate lakes located in watersheds with military disposal sites, open circles indicate non-seabird lakes, and solid circles indicate seabird lakes.

Table 1

Mean size-adjusted total mercury (THg, dry wt) concentrations in threespine stickleback (± 1 standard deviation), and lake variables for sampled lakes at Adak Island, Aleutian Archipelago, Alaska, USA, 2010

Lake	THg (mg/kg)	Estimated surface area (ha)	Water Temp ($^{\circ}$ C)	Salinity	pH	Conductivity (μ S/cm)
Military and Non-seabird						
Jamie	0.31 (0.05)	20.5	7.5	0.05	7.92	71.1
Mitt	0.34 (0.08)	3.1	13.0	0.07	6.26	111.0
Heart	0.36 (0.04)	20.9	9.8	0.03	8.25	44.6
Leone	0.38 (0.11)	42.8	12.0	0.04	8.25	59.1
Kendall	0.41 (0.08)	10.7	9.0	0.03	7.64	58.0
Nonmilitary and Non-seabird						
Shotgun	0.40 (0.10)	4.4	7.0	0.06	7.90	91.8
Nonmilitary and Seabird						
Ronnie	0.53 (0.12)	6.0	10.0	0.06	7.83	100.0
Shirley	0.54 (0.17)	4.8	12.0	0.08	7.93	129.3