Marine Pollution Bulletin 117 (2017) 178–183

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Geographic and temporal patterns of variation in total mercury concentrations in blood of harlequin ducks and blue mussels from Alaska

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ARTICLE INFO

Article history:
Received 20 September 2016
Received in revised form 30 January 2017
Accepted 31 January 2017
Available online 3 February 2017

Keywords:
Harlequin duck
Histrionicus histrionicus
Alaska
Mercury
Blue mussel

ABSTRACT

We compared total mercury (Hg) concentrations in whole blood of harlequin ducks (*Histrionicus histrionicus*) sampled within and among two geographically distinct locations and across three years in southwest Alaska. Blue mussels were collected to assess correlation between Hg concentrations in locally available forage and birds. Mercury concentrations in harlequin duck blood were significantly higher at Unalaska Island (0.31 ± 0.19 mean ± SD, μg/g blood) than Kodiak Island (0.04 ± 0.02 mean ± SD, μg/g blood). We found no evidence for annual variation in blood Hg concentration between years at Unalaska Island. However, blood Hg concentration did vary among specific sampling locations (i.e., bays) at Unalaska Island. Findings from this study demonstrate harlequin ducks are exposed to environmental sources of Hg, and whole blood Hg concentrations are associated with their local food source.

1. Introduction

Mercury (Hg) is a pervasive and persistent environmental contaminant, found both in marine and freshwater environments. Mercury becomes available to most marine fish and wildlife through atmospheric deposition, point source pollution, or transport from rivers and estuaries emptying into the ocean (Burger and Gochfeld, 2009a; Scheuhammer et al., 2012; Wolfe et al., 2007). Mercury is of particular concern in marine environments, due to its ability to rapidly move up food chains (Ackerman et al. 2016; Burger and Gochfeld, 2009a; Chen et al., 2008; Driscoll et al., 2013). Concentrations of methylmercury (MeHg), the organic and highly toxic form of Hg, are bio-magnified through marine and freshwater food webs and can reach levels harmful to birds (Jackson et al., 2011; Evers et al., 2008). For long-lived species such as sea ducks, the biomagnification of Hg renders greater risk of behavioral and reproductive impacts (Burgess and Meyer, 2008; Evers et al., 2005; Evers et al., 2008). Potential adverse effect concentrations of Hg for many bird species, including sea ducks, are still largely unknown (Franson, 2015). Exposure to dietary Hg can be highly variable among species of birds due to their specific prey selection, foraging strategies (Burger and Gochfeld, 2009b; Dietz et al., 2013; Evers et al., 2005), and the proximity of wintering, molting, and breeding locations to contaminated areas (Cristol et al., 2012). Identifying baseline and threshold contaminant concentrations in wildlife is valuable in establishing regional toxicological benchmarks (Mallory et al., 2010).

Studies of contaminants, including Hg in sea ducks, have primarily focused on residues found in the internal organs of harvested birds (Franson et al., 1995; Henny et al., 1995; Wayland et al., 2001; Braune and Malone, 2006), eggs (Akearok et al., 2010; Goodale et al., 2009; Rave et al., 2014; Zicus et al., 1988), and feathers (Burger and Gochfeld, 2009a,b). The use of blood to interpret contaminant concentrations in sea ducks has become more common in recent years (Meattey et al., 2014; Heard et al., 2008; Franson et al., 2004; Wayland et al., 2008; Grand et al., 2002; Wilson et al., 2004; Franson et al., 2000, Hollmén et al., 1998; Wayland et al., 2001; Wayland et al., 2003). Contaminants in harlequin ducks (*Histrionicus histrionicus*) have been rarely studied (Ackerman et al. 2016; Franson, 2015; Heard

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http://dx.doi.org/10.1016/j.marpolbul.2017.01.084
0025-326X/

et al., 2008), and the Sea Duck Joint Venture has identified the specific need to determine contaminant concentrations in wintering harlequin ducks (Sea Duck Joint Venture Management Board, 2008).

Previous studies have found whole blood is the most appropriate tissue to determine recent intake of contaminants through daily foraging and can be sampled non-lethally; Hg content in the whole blood of birds often reflects an individual's Hg exposure through diet over the past several days (Evers et al., 2005). Furthermore, Hg in the blood of birds is primarily MeHg (>95%; Fournier et al., 2002; Rimmer et al., 2005; Edmonds et al., 2012). Blood sampling allows non-lethal capture options to conduct follow up studies (e.g., behavioral, survival, and reproductive investigations).

In this study, we compare total Hg (hereafter Hg) concentrations in whole blood from harlequin ducks sampled from two marine sites in Alaska (Fig. 1) from 2005 to 2008. Harlequin ducks were sampled during wing molt (August 2005) at Kodiak Island and during winter (February 2006 and 2008) at Unalaska Island. In addition, blue mussels (*Mytilus edulis*) were collected at Unalaska Island in 2008 because mollusks are an important winter forage component for harlequin ducks (Goudie and Ankney, 1986; Robertson and Goudie, 1999) and have been shown to bioaccumulate Hg (Garron et al., 2005; Burger and Gochfeld, 2006; Meattey et al., 2014), thus contributing to the Hg exposure of their consumers. We selected the blue mussel as an indicator of Hg in mollusks because they could be readily collected from all of the sampling locations and previous studies have found similar Hg concentrations of Hg among species of bivalve mollusks collected at the same location (Szkoda et al., 2015).

2. Materials and methods

2.1. Collection of harlequin duck blood samples

Harlequin ducks were live-trapped and whole blood was collected from individuals at two sites in Alaska (Fig. 1), from 2005 to 2008. In August 2005, flightless molting harlequin ducks were herded into a swim-in net trap and sampled by Kodiak Island National Wildlife Refuge biologists among three bays (Uyak Bay, Terror/Viekoda Bay, Bluefox Bay) at Kodiak Island (~57°20′N, 153°50′W). In February 2006 and 2008, wintering harlequin ducks were trapped using a floating mist net set up (Brodeur et al., 2008) and sampled by U.S. Geological Survey biologists from four bays (Chernofski Bay, Skan Bay, Portage Bay, Humpback Bay) at Unalaska Island (~53°41′N, 167°9′W) (Fig. 1). Harlequin ducks

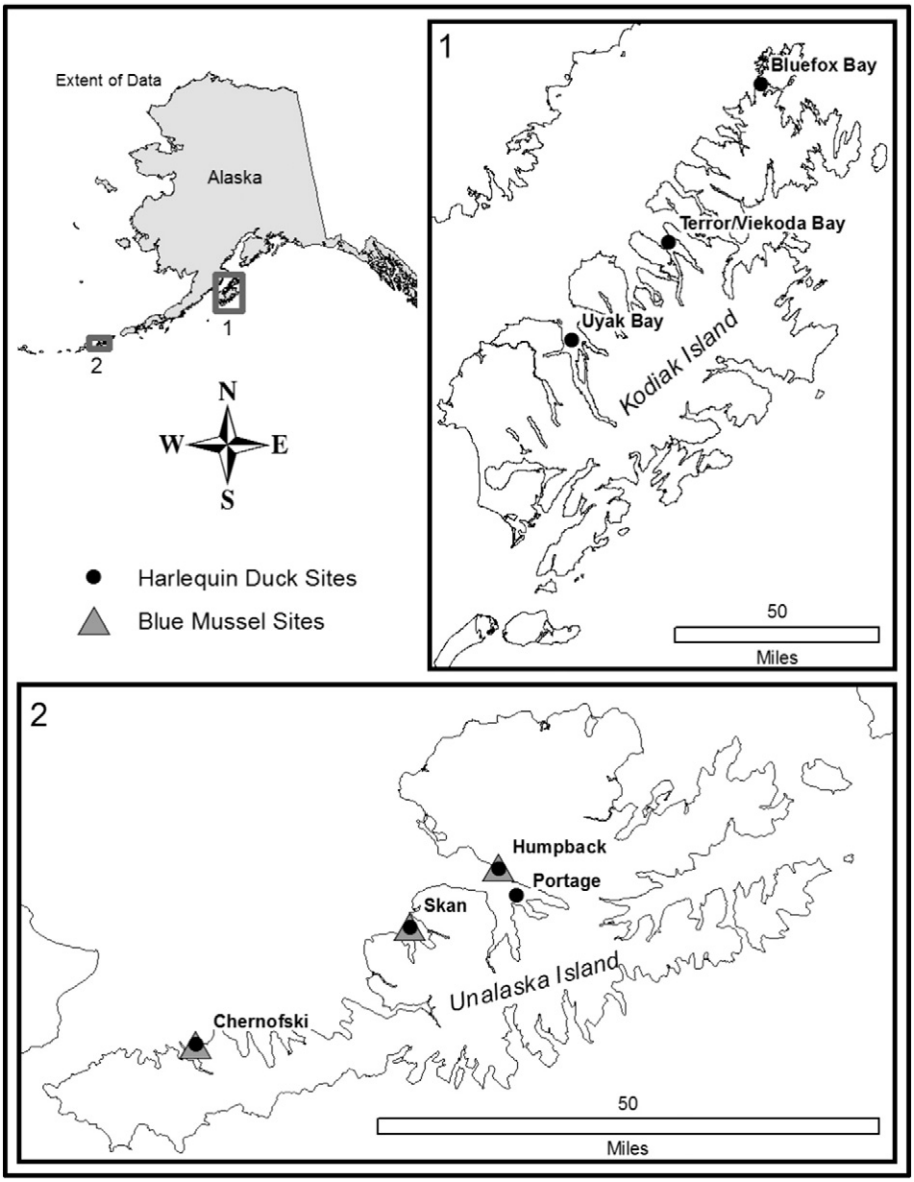

Fig. 1. Harlequin duck sampling and blue mussel collection sites in Alaska.

sampled at Kodiak Island and Unalaska Island were aged by two differing methods. At the Unalaska Island site, harlequins were aged by plumage characteristics (Robertson and Goudie, 1999) while harlequins from Kodiak Island were aged by bursal measurements (Mather and Esler, 1999). Gender at each location was determined based on plumage (Robertson and Goudie, 1999). We therefore separated harlequins into four age groups; Kodiak Island: >2 years and >1 < 2 years and Unalaska Island: >1 year or <1 year. Between 0.2 and 0.5 mL of whole blood was drawn by jugular venipuncture from each bird and stored in lithium heparinized sterile plastic vials. Vials were placed in coolers containing ice while in the field and transferred to freezers as soon as possible for archive. The samples were stored in freezers (−25 °C) prior to Hg analysis (Varian-Ramos et al., 2011).

2.2. Collection of blue mussels

Blue mussels were collected by hand from intertidal rock ledges at three of the Unalaska Island bays in which harlequin ducks were also sampled in 2008; we selected blue mussels of varying size (11–38 mm). Mussels were collected opportunistically and were not collected from Portage Bay or Kodiak Island sites. Mussels were placed in a ziplock bag and frozen (−25 °C) until laboratory processing. In the laboratory, the total shell length (0.01 mm) and the whole mussel weight (0.01 g) was measured. The soft tissue was then removed from the shell, and placed in a sterile glass jar, frozen then freeze-dried using a Labconco © Benchtop Freeze Dry System (Labconco, Inc., Kansas City, MO).

2.3. Hg analysis

Whole blood from harlequin ducks and soft tissue of blue mussels were analyzed for total Hg using multiple laboratories. All analytical methods included either cold vapor atomic absorption spectrometry (CVAAS) (USEPA, 1991), inductively coupled plasma mass spectrometry (ICP-MS), or thermal decomposition spectrophotometry (US EPA, 2007). Blood samples were analyzed at Texas A&M University, Trace Element Research Laboratory (College Station, Texas), Savannah River Ecology Laboratory, University of Georgia (Aiken, South Carolina), and Biodiversity Research Institute Wildlife Mercury Laboratory (Portland, Maine), by thermal decomposition spectrophotometry, with an automated Direct Mercury Analyzer (DMA-80, Milestone, Inc., Shelton, CT) following the United States Environmental Protection Agency, USEPA, method 7473 (US EPA, 2007). Blood samples sent to the University of Connecticut, Center for Environmental Science and Engineering (Storrs, Connecticut) were analyzed using USEPA method 245.6 (US EPA, 1991) (CVAAS, Perkin Elmer Flow Injection Mercury System, Milford, CT). Blood samples submitted to the Utah State Veterinary Diagnostic Laboratory (Logan, Utah) were analyzed with an ELAN 6000 inductively coupled argon plasma mass spectrometry (ICP-MS, Perkin Elmer, Shelton, CT) using a modification of USEPA method 3050 (US EPA, 1989) and analyzed for Hg content as previously described (Heard et al., 2008). Soft tissues of blue mussels were analyzed for Hg at Biodiversity Research Institute Wildlife Mercury Laboratory (Portland, Maine), by thermal decomposition spectrophotometry, with a Direct Mercury Analyzer (DMA-80, Milestone, Inc., Shelton, CT) using the USEPA method 7473 (US EPA, 2007).

Although multiple laboratories and methods were used, all analyses included quality assurance data. All the laboratory methods used in this study have provided highly comparable results when measuring total Hg concentrations in avian blood (Cristol et al., 2012; Meattey et al., 2014; Perkins et al., 2016). For all samples analyzed, each laboratory generally included 2 analytical blanks, 1 sample replicate, 2 spiked samples, and 2 standard reference materials (SRM, National Research Council Canada). Reference materials were measured for every set of 20–30 samples and included fish protein (DORM-2), dogfish liver (DOLT-2), fish protein (DOLT-3), dogfish muscle (DORM-3) and dogfish liver (DOLT-4). The Utah State Veterinary Diagnostic Laboratory used an in-house developed SRM (bovine whole blood spiked with 500 ppb). Quality assurance data were considered acceptable if within 20% of the known Hg concentration, and were within 0.8–12%. All samples were above method detection levels and all laboratories met USEPA quality assurance standards (US EPA, 2007) and Hg results are considered comparable (Table 1). Blood results were reported as total Hg, in parts per million (μg/g) wet weight (ww), and blue mussel results were reported and displayed graphically as total Hg parts per million (μg/g) dry weight (dw). For comparison purposes in the discussion, blue mussel wet weight Hg values were calculated by using 89.8% moisture content based on findings reported in Franson et al. (1995).

2.4. Data analysis

Results are reported as arithmetic means. All statistical analyses were performed using log-transformed Hg concentrations using JMP v.9.0 statistical software (SAS Institute Inc., 2010). A non-parametric Steel-Dwass test was used to compare Hg concentrations of individual sampling bays within each location. A two-way analysis of variance (ANOVA) was performed to compare Hg concentrations among sampling bays between years. Bays not containing significant differences between years were pooled in further analyses. A two-way analysis of variance (ANOVA) was used to compare Hg concentrations between gender and age class of harlequin ducks at each location. Results of statistical tests were considered significant at $p < 0.05$. Back-transformed data are presented in tables and figures.

3. Results

3.1. Harlequin duck blood Hg

We collected blood samples from 33 harlequin ducks from Kodiak Island in 2005 and 27 and 55 harlequin ducks from Unalaska Island in 2006 and 2008, respectively. Mercury was detected in all 115 blood samples and ranged from 0.01 to 0.92 μg/g (parts per million, ppm),

Table 1
Quality assurance data for total mercury analysis of harlequin duck whole blood at 5 laboratories: Trace Elements Research Laboratory at Texas A&M University, Biodiversity Research Institute Wildlife Mercury Laboratory, Savannah River Ecology Laboratory at the University of Georgia, Center for Environmental Science and Engineering at the University of Connecticut, and Utah State University Veterinary Diagnostic Laboratory. We report method detection limits (μg/g, ww), percent recovery from standard reference materials (DORM-2, DORM-3, DOLT-2, DOLT-3, DOLT-4, and bovine blood) and percent recovery of added mercury detected in spiked samples.

	Texas A&M University	BRI Wildlife Hg Lab	Savannah River Ecology Lab	University of Connecticut	Utah State University
Method detection limit	0.001	0.001	0.001	0.0001	0.0001
DORM-2	103.1			106.9	
DORM-3		100.8			
DOLT-2			108–112		
DOLT-3	110.0			101.2	
DOLT-4		105.8			
Bovine					95–108
Sample spike	112.0			96.5	92–107
Analytical method	DMA-80	DMA-80	DMA-80	CVAAS	ICP-MS

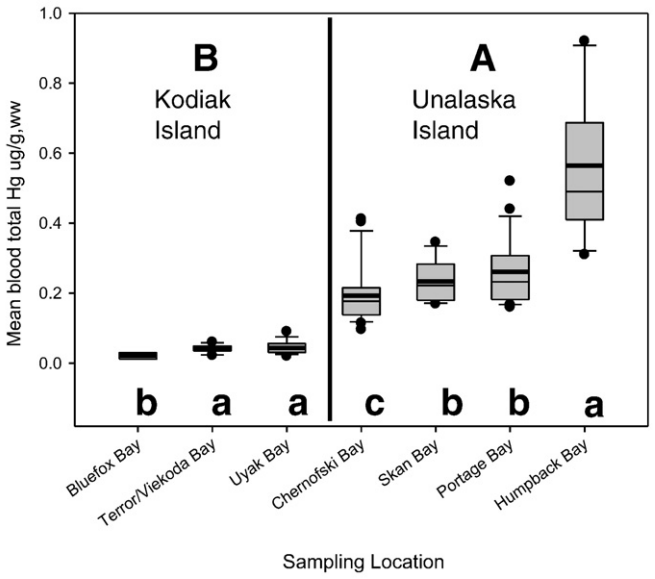

Fig. 2. Mean whole blood Hg (μg/g, ww) in harlequin ducks among bays at Kodiak Island and Unalaska Island, Alaska. *Uppercase letters represent significant differences between Unalaska Island and Kodiak Island based on Steel-Dwass multiple comparison procedure. Lowercase letters represent significant differences among sampling bays at Kodiak Island and Unalaska Island based on Steel-Dwass multiple comparison procedure. The bolded line represents the mean Hg concentration. The middle line represents the median value; the lower and upper lines represent the 25th and 75th percentiles, respectively. The lower and upper "whiskers" represent the 10th and 90th percentiles, and the black dots represent outliers beyond the 10th and 90th percentiles.

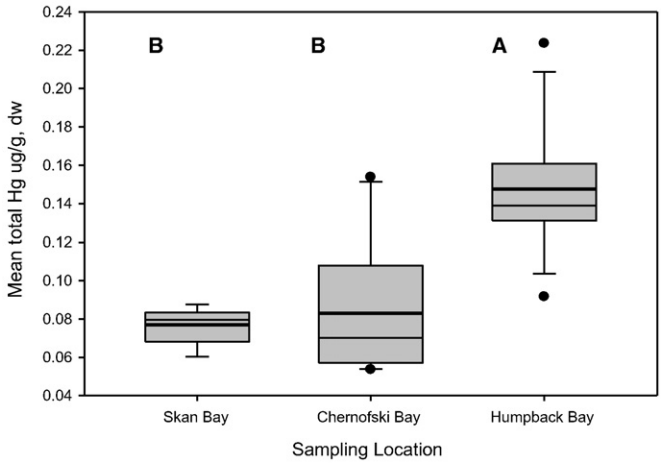

Fig. 3. Mean soft tissue Hg concentrations (μg/g ± SD dry weight) in blue mussels from Unalaska Island, Alaska. *Letters represent significant differences among sampling locations based on Steel-Dwass multiple comparison procedure. The bolded line represents the mean Hg level. The middle line represents the median value; the lower and upper lines represent the 25th and 75th percentiles, respectively. The lower and upper "whiskers" represent the 10th and 90th percentiles, and the black dots represent outliers beyond the 10th and 90th percentiles.

wet weight (ww) (Fig. 2). We found marginally significant differences in Hg concentrations between years among bays sampled at Unalaska Island ($F_{7,74} = 2.833$, $p = 0.04$). Difference in Hg concentrations between years was observed for Chernofski Bay only, but the magnitude of the difference in least square means was minimal (0.26) and was similar to Portage Bay (0.24). We therefore pooled data across years for each bay in further statistical analyses (Fig. 2).

Harlequin ducks sampled at Kodiak Island consisted of 28 males (>2 years = 11, >1 < 2 years = 17) and five females (>2 years = 1, >-1 < 2 years = 4) and for Unalaska Island, 51 males (>1 year = 44, <1 year = 7) and 31 females (>1 year = 29, <1 year = 2). We conducted post hoc analyses to evaluate potential differences in blood Hg concentrations among gender and between age classes of harlequin ducks at Kodiak Island and Unalaska Island. Overall, gender and age class did not influence Hg concentrations in harlequin ducks at both Kodiak Island ($F_{1,29} = 0.05$, $p = 0.82$) and Unalaska Island ($F_{1,78} = 0.11$, $p = 0.74$) and therefore, harlequins from each gender and age classes were combined for further statistical analyses.

Overall, Hg concentrations in the blood of harlequin ducks were significantly higher from Unalaska Island than from Kodiak Island ($F_{1,113} = 368.698$, $p < 0.0001$). Mercury concentrations in harlequin duck blood collected from Kodiak Island ranged from 0.01 to 0.09 μg/g (ww), with a mean of 0.04 ± 0.02 μg/g (ww) (Fig. 2). Mercury concentrations in harlequin duck blood collected from Unalaska Island ranged from 0.10 to 0.92 μg/g (ww) with a mean of 0.31 ± 0.19 μg/g (ww) (Fig. 2). We also found that Hg concentrations varied significantly among specific bays within each of the two sites ($F_{6,108} = 156.613$, $p < 0.0001$) (Fig. 2).

3.2. Blue mussel Hg

Mercury was detected in all 34 blue mussels analyzed. Mercury ranged from 0.05–0.22 μg/g, dry weight (dw), with a mean Hg concentration of 0.11 ± 0.04 μg/g (dw). Mercury content in blue mussels displayed a similar trend to harlequin duck blood Hg, with significant differences in concentrations among bays ($F_{2,31} = 25.283$, $p < 0.0001$) (Fig. 3). The pattern of variation in Hg concentration in mussels was similar to patterns observed in harlequin duck blood Hg concentrations; Chernofski Bay and Skan Bay contained the lowest mean mussel Hg concentrations, 0.08 ± 0.03 and 0.08 ± 0.01 μg/g, (dw), respectively, while Humpback Bay contained the highest mean Hg concentrations (0.15 ± 0.03 μg/g, dw).

4. Discussion

The Hg concentrations were considerably higher at Unalaska Island compared to Kodiak Island. The potential sources of Hg in both locations are unknown. We found substantial variation in the concentration of Hg in harlequin duck blood within and between Unalaska Island and Kodiak Island sampling locations. However, we found little annual differences in Hg concentration within a specific subset of sampling locations. Furthermore, the pattern in blood Hg concentrations in harlequin ducks were similar to the pattern in blue mussel Hg tissue concentrations among sampling bays at Unalaska Island and generally fits with broad scale sediment sampling at Unalaska Island and near Kodiak Island (Meador et al., 1998). Thus, we conclude that blood Hg concentrations represent a valid indicator of local environmental Hg contaminations in this marine system. Given the patterns of variation in blood Hg concentration among specific sampling bays, we caution against extrapolating our results to broader, geographic areas. In fact, we caution that our estimates may not be representative of the broader Kodiak Island and Unalaska Island areas. Because Hg blood concentration varied among bays, the appropriate estimate for the larger area would be an average of the blood concentration from each bay weighted by the proportion of the harlequin duck population that occurred in each bay. Survey data of molting harlequin ducks were collected at Kodiak Island in August 2005 (Zwiefelhofer, 2005). Unfortunately, we do not have survey data for Unalaska Island that can be used to compare estimates for the total population size and distribution among all bays. Our current estimate of the harlequin duck population at Unalaska Island and Kodiak Island areas assumes equal distribution among bays which is unlikely.

The presence of Hg and other contaminants in wildlife is well documented in the Aleutian Chain of Alaska (Ackerman et al. 2016; Anthony et al., 2007; Rocque and Winker, 2004; Burger and Gochfeld, 2006; Burger et al., 2007; Ricca et al., 2008; Burger and Gochfeld, 2009a; Burger and Gochfeld, 2009b). Meador et al. (1998) sampled sediments

Table 2
Mean Hg concentrations (μg/g, dw ± SD and μg/g, ww ± SD) in blue mussels from Unalaska Island, 2008.

Bay	n	Hg (dw)	Range	Hg (ww)[a]	Range
Humpback Bay	15	0.15 ± 0.03	0.09–0.22	0.015 ± 0.003	0.009–0.023
Skan Bay	9	0.08 ± 0.01	0.06–0.09	0.008 ± 0.001	0.006–0.009
Chernofski Bay	10	0.08 ± 0.03	0.05–0.15	0.008 ± 0.003	0.005–0.016
Location totals	34	0.11 ± 0.04	0.05–0.22	0.011 ± 0.005	0.005–0.023

[a] Calculated wet weight values using percent moisture value of 89.9% (Franson et al., 1995).

and found higher levels of Hg at Unalaska Island than at 10 other sites in Alaska. Identifying the source, however, is extremely difficult. Potential sources include natural inputs from prevalent volcanic activity in the Aleutians (Ricca et al., 2008), historical military activity (Anthony et al., 2007; Burger and Gochfeld, 2006; Ricca et al., 2008), and atmospheric and oceanic pollution from increasing industrial activity in southern Asia (Anthony et al., 2007; Rocque and Winker, 2004; Driscoll et al., 2013) and Russia (Fisher et al., 2012). Nonetheless, all the birds we sampled appeared healthy, upon visual inspection. The concentrations of Hg we found were lower than Heard et al. (2008) reported for wintering harlequin ducks (0.82 ± 0.32, μg/g, ww) sampled in Prince William Sound, Alaska in 2005. A western North America bird Hg synthesis study reported a mean blood Hg concentration of 0.18 (μg/g, ww) in harlequin ducks (Ackerman et al. 2016). Thus, the concentrations of Hg we report are not considered to be unusually high and we do not believe that there are likely negative population consequences for the exposure we documented.

4.1. Blue mussel Hg

Blue mussels are a typical food item for harlequin ducks on most of the species' wintering areas (Robertson and Goudie, 1999) and were abundant at the Unalaska Island trapping sites (Flint pers. comm.). Sedentary bivalves, such as the blue mussel are frequently used as an aquatic indicator species of contaminants, including Hg (Airas et al. 2004; Garron et al., 2005; Burger and Gochfeld, 2006).

Mercury concentrations in the soft tissues of blue mussels collected from Skan and Chernofski Bays were lower than those reported in previous studies from sites focused on the Aleutian chain of Alaska (range of means: 0.01–0.02, μg/g, ww) (Burger and Gochfeld, 2006). Blue mussels collected from Humpback Bay in 2008 contained similar Hg concentrations from those collected at Adak Island, AK (Burger and Gochfeld, 2006) and a historically polluted site in Norway (0.01 and 0.03, μg/g, ww) (Airas et al. 2004). A study in New Brunswick, Canada reported a mean Hg concentration of 1.40 (μg/g, ww) in blue mussels collected near a known point-source Hg polluted chemical plant (Garron et al., 2005).

Mercury concentrations in harlequin duck blood were greater than an order of magnitude higher than Hg measured in blue mussels. Patterns of Hg concentrations among sampling bays at Unalaska Island were similar between harlequin duck blood and blue mussels (Table 2). Blue mussels and harlequin ducks from Humpback Bay had significantly higher Hg concentrations than from the other bays, suggesting that Humpback Bay and its biota have greater exposure to Hg.

4.2. Blue mussel size and mercury concentrations

Post hoc analyses were performed to evaluate whether size (e.g., whole weight, shell length, content weight) of blue mussels influenced Hg concentrations in their soft tissue, and therefore influencing mean Hg concentrations and comparisons of mussel Hg among sampling bays. Previous literature provides conflicting findings with regards to correlations between blue mussel size and corresponding Hg concentrations. Some studies have found larger blue mussels, using shell length measurements, correlated with increasing Hg concentrations in their soft tissue (Burger and Gochfeld, 2006) and this same pattern has been shown in other mollusks (Saavedra et al., 2004). However, other studies report no correlation between shell length and soft tissue Hg concentrations in blue mussels (Anderson et al., 1996). In our study we found no correlation between blue mussel size and Hg concentrations. Thus, the variation in Hg concentration in blue mussels among bays we sampled is not a result of variation in mussel size class.

5. Conclusion

Our data demonstrate that harlequin duck blood Hg concentration tends to reflect local environmental availability. The fact that blood Hg concentration varied among bays within larger study areas indicates that extrapolation of results at larger geographic scales warrants caution. Future studies seeking to develop regional estimates of Hg exposure need to account for variation within areas and adjust estimates based on proportional distributions of birds. We found that blood Hg concentrations were relatively consistent over time, suggesting that sources of Hg are consistent, however it is unclear at this time how climate change will influence Hg availability in the marine ecosystem. Results determined age and gender does not influence Hg concentrations in the whole blood of harlequin ducks. We determined harlequin ducks are able to accumulate locally available environmental Hg and at varying concentrations. However, it does not appear that Hg exposure at our study sites is of a magnitude to cause population level effects.

Acknowledgements

We thank the crews involved with field collections. We also thank the captains and crews of the *M/V Kittiwake* and *R/V Ursa Major II*, for safe passage through bays of Unalaska Island and Kodiak Island. Jon Atwood and Iain Stenhouse provided helpful reviews on drafts of this manuscript and Kim Trust (U.S. Fish and Wildlife Service) facilitated access to archived samples. Kevin Regan assisted in laboratory analyses. Evan Adams provided assistance with statistical analyses. This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors. All procedures were approved by the U.S. Geological Survey Alaska Science Center Animal Care and Use Committee, and were authorized under U.S. Fish and Wildlife Service and State of Alaska permits. Any use of trade, product, or company names is solely for descriptive purposes and does not imply endorsement by the U.S. government.

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