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Mercury concentrations in multiple tissues of Kittlitz's murrelets (*Brachyramphus brevirostris*)

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ABSTRACT

Mercury (Hg) is a non-essential, toxic metal that is distributed worldwide. Mercury biomagnifies in food webs and can threaten the health of top predators such as seabirds. The Kittlitz's murrelet (*Brachyramphus brevirostris*) is a seabird endemic to Alaska and the Russian Far East and is a species of conservation concern in the region. We determined Hg concentrations in eggshells, guano, blood, and feathers of Kittlitz's murrelets sampled from four locations in Alaska. Mercury concentrations in eggshells, guano, and blood were low compared to other seabird species. Mean Hg concentrations of breast feathers from Adak Island and Glacier Bay were significantly greater than those from Agattu Island or Icy Bay. Two Kittlitz's murrelets at Glacier Bay and one Kittlitz's murrelet at Adak Island had Hg concentrations above those associated with impaired reproduction in other bird species, and may merit further investigation as a potential threat to individuals and populations.

1. Introduction

The Kittlitz's murrelet (*Brachyramphus brevirostris*) is an endemic seabird found in Alaska and the Russian Far East and is listed as Near Threatened by the International Union for the Conservation of Nature and Natural Resources (IUCN) Red List owing to an apparent population decline (BirdLife International, 2017) and a small global population size of approximately 34,000 individuals (USFWS, 2013). Causes for the apparent population decline are uncertain, in part because many information gaps exist, including exposure to environmental contaminants.

Mercury (Hg) is a non-essential metal that is distributed worldwide, largely due to anthropogenic activities such as coal burning and waste incineration (Pacyna and Pacyna, 2002), as well as natural processes such as volcanic and local geologic activities (Mason, 2009). Once deposited in aquatic habitats, inorganic Hg is converted by bacteria to the more toxic and bioavailable form, methylmercury (MeHg), which has potential to bioaccumulate in individuals and biomagnify through food webs (Watras et al., 1998).

The extended atmospheric lifetime of inorganic Hg facilitates long-range transport by atmospheric and ocean currents to regions far from the source of emission (Holmes et al., 2010; Driscoll et al., 2013). Long-range transport of Hg is a concern in Alaska due to the accelerated development in coal-burning industries throughout the world and the prediction of warmer temperatures in Arctic regions related to global warming that could enhance the bioavailability of Hg (AMAP, 2002). Furthermore, recent research has identified potential avian Hg contamination hotspot regions in Alaska, including the Aleutian Archipelago and southeastern Alaska (Ackerman et al., 2016).

Due to their piscivorous diet, Kittlitz's murrelets may be at elevated risk to Hg exposure (Burger and Gochfeld, 1997; Evers et al., 2004; Ackerman et al., 2016). High concentrations of Hg in avian tissues have been associated with physiological, behavioral, and reproductive effects (Scheuhammer et al., 2007; Ackerman et al., 2016). Low reproductive success has been documented for Kittlitz's murrelets throughout its Alaskan breeding range (Kaler et al., 2009; Lawonn, 2012; Kissling et al., 2015; Kissling and Lewis, 2016), and has largely been attributed to nestling mortality, presumably due to exposure and starvation; however, the ultimate cause of nestling deaths remains poorly understood (Kaler et al., 2009; Lawonn, 2012). The apparent population decline of Kittlitz's murrelets in the core of their breeding range, and the range-wide low rate of reproductive success merits study of contaminants in tissues to help fill an important knowledge gap.

Our objectives were to: 1) quantify Hg concentrations in multiple

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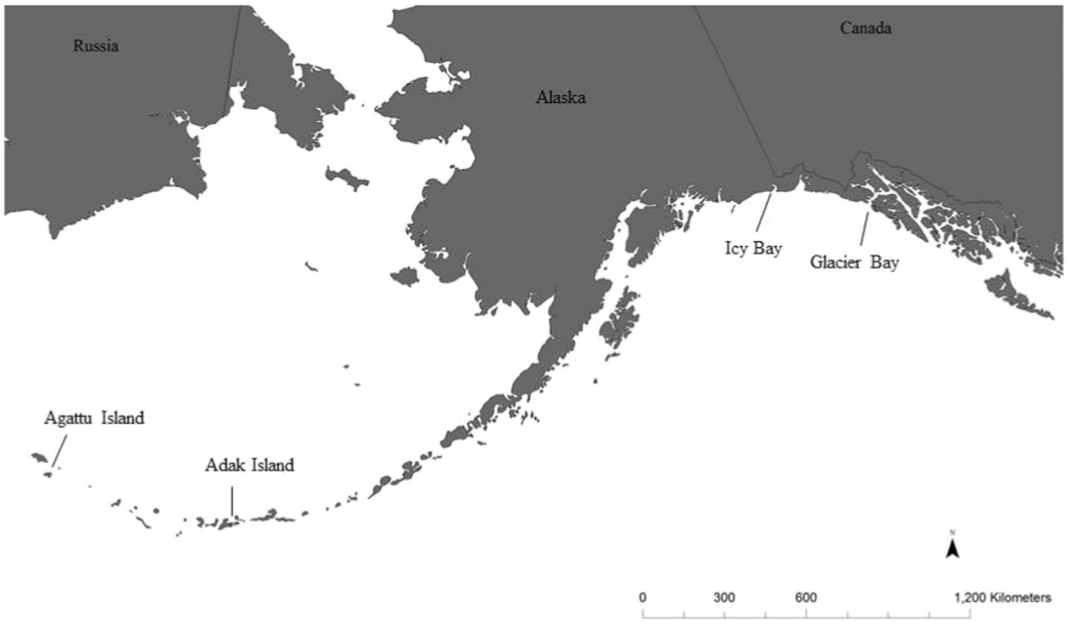

Fig. 1. Map showing sampling locations of mercury concentrations of Kittlitz's murrelets tissues in Alaska, U.S.A.

tissues of Kittlitz's murrelets; 2) compare Hg concentrations in Kittlitz's murrelets feathers collected from four regions within Alaska; and 3) compare Hg concentrations in Kittlitz's murrelet tissues to published Hg threshold levels developed for other marine birds.

2. Methods

2.1. Study sites

Sample tissues were collected from 2008 to 2011 at four areas across the breeding range of Kittlitz's murrelet in Alaska. All tissue samples were collected during the breeding season (May–August) from two areas in southeast Alaska (Glacier Bay and Icy Bay) and two areas in the Aleutian Islands (Adak Island and Agattu Island; Fig. 1).

2.2. Tissue sampling

Eggshell fragments were collected from Kittlitz's murrelet nest scrapes (n = 20) after eggs hatched at Agattu Island in 2008 to 2011. Kittlitz's murrelets lay a single egg, and infrequently lay a replacement egg if the first is depredated (Kaler et al., 2008). Eggshells were collected from the nest scrape using stainless steel forceps, air dried, and placed in sterile plastic bags.

During the 24–40-day nestling period, nestlings defecate along the edge of the nest, resulting in a fecal ring surrounding the perimeter of the nest scrape (Kaler et al., 2009; Kenney and Kaler, 2013). Fecal materials were collected during the nestling period at Adak Island in 2011 (n = 5), using stainless steel forceps, air dried, and stored in sterile plastic bags in a − 20 °C freezer until analytical processing.

Whole blood samples (n = 102) were obtained from adult Kittlitz's murrelets captured on the water at Icy Bay, May and June 2008 and 2009, using spot lights and salmon dip nets (Kissling et al., 2015). Following capture, < 2 ml of blood was sampled from the ulnar vein, using a heparinized syringe and vacutainer. All blood samples were frozen immediately.

Similar to blood sample collection, we collected breast feathers from adult Kittlitz's murrelets captured in Icy Bay, May and June 2008 and 2009 (n = 234), and in Glacier Bay, May 2011 (n = 21; Kissling et al., 2015). Kittlitz's murrelets molt twice annually; they undergo a partial molt of only body feathers during the spring (pre-alternate molt) and a full molt of body and flight feathers during the fall (pre-basic molt; Sealy, 1977; Day, 1999; Pyle, 2009). Breast feathers grown in the spring are dark-tipped (hereafter, alternate) and can be differentiated from breast feathers presumably grown only in the fall, which lack pigmentation and are all white in color (hereafter, basic; Hatch, 2011).

Mercury concentrations in breast feathers are derived from circulating blood Hg during the time of active feather growth (Braune and Gaskin, 1987; Monteiro and Furness, 2001), and represents a dynamic equilibrium between both dietary Hg exposure and remobilization of Hg stored in tissues. Breast feathers grown in the spring presumably represent dietary Hg exposure during the non-breeding season, and breast feathers molted during the fall should reflect dietary Hg exposure during the breeding season. We attempted to collect both alternate and basic breast feathers from each captured murrelet and separate them prior to analytical processing; however, in some cases it was not possible and either the bird did not initiate the alternate molt at the time of capture in May (i.e., no dark-tipped breast feathers present for collection) or the two types of breast feather were not separated prior to laboratory processing (see Statistical analysis). For example, in 2008, alternate and basic breast feathers were combined and not differentiated, but in 2009 and 2011, these feather types were analyzed separately. In addition, during 2011, we also collected 2–3 cm of the fifth secondary feather from adult murrelets captured on the water in Icy Bay and Glacier Bay (n = 157, n = 19, respectively).

At Agattu Island, adult breast feathers were collected during 2008–2011 from nest scrapes during the incubation period (n = 29), and from nestlings during the nestling period (n = 8; Kaler et al., 2014). At Adak Island, adult breast feathers (n = 7), and one secondary feather, were collected during the incubation period from nest scrapes in 2011. Kittlitz's murrelets alternate incubation duties approximately every 24–72 h (Day, 1999; Kaler *unpublished data*); therefore, we assume feather samples from Agattu and Adak islands represent a composite sample from both adults from a single nest site. Feathers were stored in paper envelopes until laboratory processing.

2.3. Laboratory analysis

Using gold-amalgamation atomic absorption spectroscopy with a Direct Mercury Analyzer (DMA-80 Milestone), we analyzed Kittlitz's murrelet tissues for total Hg. Eggshells, guano, and feathers, collected from Kittlitz's murrelets at Agattu Island, Adak Island, and Glacier Bay, were analyzed at the U.S. Geological Survey, Contaminant Ecology

Table 1
Mercury concentrations (mean ± standard deviation) and sample size (n) in tissues of Kittlitz's murrelets collected at four locations in Alaska, U.S.A. All values are reported as μg/g dry weight, except for blood which is reported as μg/g wet weight.

Location	Mixed Breast feather	Secondary feather	Blood	Eggshell	Guano
Agattu Island (2008–2011)	2.06 ± 1.28 (37)			0.016 ± 0.004 (20)	
Adak Island (2011)	5.15 ± 2.51 (7)	37.18 (1)			0.006 ± 0.003 (5)
Icy Bay (2008–2009)	1.21 ± 0.22 (46)	2.01 ± 0.68 (157)	0.260 ± 0.076 (71)[a] 0.425 ± 0.031 (31)[b]		
Glacier Bay (2011)	9.31 ± 1.28 (21)	2.87 ± 1.95 (19)			

[a] 2008 collection.
[b] 2009 collection.

Research Lab, Corvallis, OR. Adult blood samples and breast feathers from Icy Bay were analyzed at the Utah Veterinary Diagnostic Laboratory for samples collected in 2008 and at Biodiversity Research Institute, Gorham, Maine, for samples collected in 2009. Analytical equipment was calibrated using certified standard solutions prior to analysis, and accuracy and precision were evaluated within each analytical batch through the inclusion of certified reference materials (either dogfish muscle tissue [DORM-3] or dogfish liver [DOLT-4] from the National Research Council of Canada), calibration verifications (liquid standards), duplicates, and blanks. Recoveries for feathers, eggshells, and guano averaged 98.3 ± 3.2% and 104.2 ± 2.3% for calibration checks and certified reference materials, respectively. Absolute relative percent difference for all duplicates averaged 12.0 ± 10.8%. Recoveries for blood samples averaged 98.2 ± 4.50% and 102.3 ± 2.5% for calibration checks and certified reference materials, respectively.

Total Hg was used as a proxy for MeHg for in feathers and blood (Fournier et al., 2002; Rimmer et al., 2005; Bond and Diamond, 2009), however the proportion of total Hg comprised of MeHg in eggshells and guano is unknown. Mercury in eggshells, guano, and feathers are reported as dry weight (dw) and blood Hg concentrations are reported as wet weight (ww).

2.4. Statistical analysis

We assessed all data for normality using Shapiro-Wilk's test (Shapiro and Wilk, 1965), and log-transformed Hg concentrations when it improved normality. We tested for differences among years in eggshell and blood Hg concentrations, and among sites in secondary feather Hg concentrations using a general linear model (GLM), with Tukey's Honest Significant Differences (HSD) post hoc test for multiple comparisons.

Breast feathers represented two feather generations (see Methods). We first examined differences among sites in basic breast feather Hg using a general linear mixed model (GLMM) with site as a fixed factor and sampling year as a random factor, and comparing this to a GLMM with only an intercept and the random factor with an analysis of variance (ANOVA). (See Table 2.)

Some samples from Agattu Island (n = 37) included both feather generations (hereafter mixed breast feather). To compare these with other sites, we first tested for differences in Hg concentrations in these two feather generations using paired samples from Icy Bay in 2009 (n = 46) and Glacier Bay in 2011 (n = 21) in a GLMM. Because we found significant differences in Hg between feather generations (see Results), we generated random samples of resampling (without replacement) either alternate or basic feathers from Icy Bay (2009) and Glacier Bay (2011) to compare with mixed samples from Adak Island (all feathers pooled together because of small sample size) and Agattu Island. We resampled 15 feathers from each population (Icy Bay and Glacier Bay), calculated the mean, and repeated this 15 times. We chose n = 15, as this closely matched the sample sizes of mixed breast feathers. Using resampled data and mixed breast feather samples, we examined differences among sites using a GLMM with year as a random factor, and compared this to an intercept-only model as detailed above.

We also examined the relationship between blood and feather Hg in 62 individuals from Icy Bay in 2008 and 2009 from which both tissues were sampled. We used a general linear model to test whether blood Hg differed with feather Hg, and feather generation. Analyses were conducted in R 3.2.4 using the package lme4 (Bates et al., 2014; R Core Team, 2016). Values are presented as the mean ± SD, and range.

3. Results

Eggshell Hg concentrations were log-transformed (W = 0.919, p = 0.09), and there were no differences among years ($F_{2,17}$ = 0.101, p = 0.91). Blood Hg concentrations were normally distributed (W = 0.990, p = 0.73), and concentrations were significantly higher in 2009 than 2008 ($F_{1,85}$ = 41.47, p < 0.001). Log-transforming secondary feather Hg concentrations improved, but did not achieve normality (W = 0.854, p < 0.001). Secondary feather Hg concentrations differed among sites ($F_{2,174}$ = 42.82, p < 0.001, Tukey's HSD, all p < 0.001) with the highest concentrations at Glacier Bay (2.87 ± 1.95 μg/g, range: 1.5–10.12 μg/g, n = 19), and the lowest at Icy Bay (2.01 ± 0.68 μg/g, range: 0.82–5.17 μg/g, n = 157); the one sample from Adak Island had the highest Hg concentration (37.17 μg/g; Table 1).

Log-transformed basic breast feather Hg concentrations were not normally distributed (W = 0.879, p = 0.001). The GLMM including site, fit the data significantly better than an intercept-only model (χ^2_2 = 13.54, p = 0.001). Basic breast feather Hg was higher at Adak Island (5.80 ± 2.00 μg/g, range: 3.60–8.25 μg/g, n = 6) and Glacier Bay (14.87 ± 19.59 μg/g, range: 1.51–57.85 μg/g, n = 8) than at Icy Bay (0.50 ± 0.20 μg/g, range: 0.21–0.96 μg/g, n = 23; Tukey's HSD, p < 0.001).

Similarly, log-transforming improved, but did not achieve normality in alternate breast feather Hg (W = 0.915, p < 0.001). The GLMM with site differences was a better fit to the data than an intercept-only model (χ^2_2 = 94.60, p < 0.001). Alternate breast feather Hg concentrations were higher at Glacier Bay (4.89 ± 2.90 μg/g, range: 1.14–9.96 μg/g, n = 13) than Icy Bay (1.25 ± 0.63 μg/g, range: 0.39–6.01 μg/g, n = 23; Tukey's HSD, p < 0.01). Basic and alternate breast feathers had significantly different Hg concentrations ($F_{1,63}$ = 30.28, p < 0.001) that also differed between sites ($F_{1,63}$ = 101.96, p < 0.001), and the interaction between breast feather generation and site was significant ($F_{1,63}$ = 28.86, p < 0.001), with basic breast feather Hg higher than alternate breast feather Hg in Icy Bay, and the reverse in Glacier Bay.

Table 2
Tukey's post hoc comparisons of mixed feather mercury as a function of collection location, based on a general linear mixed model (see Methods). Significant p-values in bold.

Location	Agattu Island	Glacier Bay	Icy Bay
Adak Island	**0.008**	0.43	< **0.001**
Agattu Island		**0.022**	0.90
Glacier Bay			< **0.001**

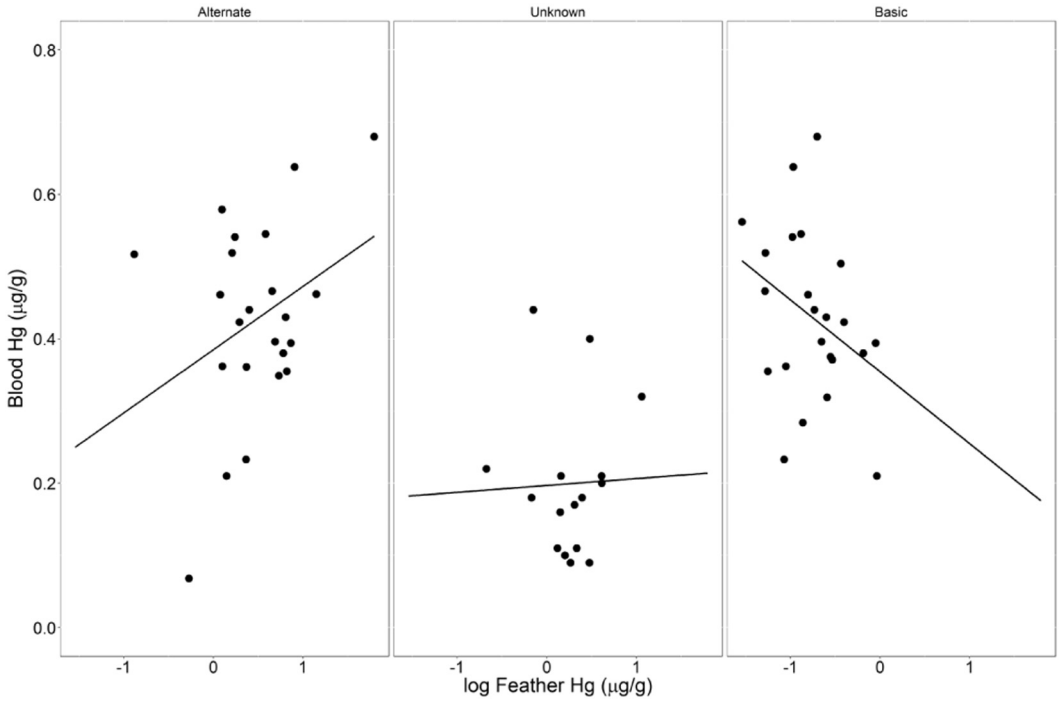

Fig. 2. Total mercury (Hg) concentrations in blood versus Hg concentrations of alternate breast feathers, unknown breast feathers, and basic breast feathers of Kittlitz's murrelets, from Alaska, U.S.A.

There were significant differences in mixed breast feather Hg among sites compared to an intercept-only model ($\chi^2_2 = 26.38$, $p = 0.001$), with two groupings: Hg was higher at Glacier Bay (9.31 ± 1.28 µg/g, range: 6.49–10.57 µg/g) and Adak Island (5.15 ± 2.51 µg/g, range: 1.26–8.25 µg/g; Tukey's HSD, $p = 0.10$) than at Icy Bay (1.21 ± 0.22 µg/g, range: 0.86–1.61 µg/g) and Agattu Island (2.06 ± 1.28 µg/g, range: 0.14–6.09 µg/g; Tukey's HSD, $p = 0.21$; Table 1); all Tukey's HSD values between these two groups with ($p < 0.001$).

Blood Hg concentrations were related significantly to breast feather Hg concentration × feather generation interaction ($F_{2,\ 58} = 12.95$, $p < 0.001$), indicating a different relationship with each generation. The relationship was positive for alternate feathers, absent for feathers of unknown generation, and negative for basic feathers (Fig. 2).

4. Discussion

Overall, Hg concentrations in Kittlitz's murrelet eggshells, guano, and blood were low compared to other seabirds. Mean Hg concentrations of mixed breast feathers from Adak Island and Glacier Bay were significantly greater than those from Agattu Island or Icy Bay and one individual from Adak Island (37.17 µg/g) and two individuals from Glacier Bay (29.78 µg/g and 57.85 µg/g) had feather Hg concentrations greater than the potential toxic threshold of 20.0 µg/g dw that has been suggested for other avian species (Bond et al., 2015).

Other studies have focused on the use of eggshells as biomonitoring tools for assessing Hg contamination in seabirds (Burger, 1994; Morera et al., 1997; Sanpera et al., 2000; Aliakbari et al., 2011; Xu et al., 2011; Brasso et al., 2012), and guano (Liu et al., 2006; Chen et al., 2012; Signa et al., 2013). Based on comparison to these other studies, Hg concentrations in Kittlitz's murrelet eggshells are relatively low (0.016 µg/g; Table 1). For example, mean (+ SD) Hg concentrations in eggshells of Audouin's gull (*Ichthyaetus audouinii*) were 0.22 (0.11) µg/g dw (Morera et al., 1997) or ranged from 0.13–0.18 µg/g dw and were not correlated with reproductive impairment (Sanpera et al., 2000). Similarly, higher Hg concentrations in guano have been reported for other seabirds including mean Hg concentrations in red-footed booby (*Sula sula*; 0.107 µg/g dw; Liu et al., 2006) and seasonal differences in yellow-legged gull (*Larus michahellis*) guano Hg concentrations ranged from 0.80–1.02 µg/g dw (Signa et al., 2013), compared to concentrations reported in our study (0.006 µg/g dw; Table 1).

Kittlitz's murrelet eggshells and guano are easy to collect at the nest scrape and both tissues could be useful non-invasive biomonitoring tools for evaluating local sources of Hg contamination, as well as other contaminants (Evenset et al., 2007; Dauwe et al., 2000; Yin et al., 2008; Chen et al., 2012; Joshi et al., 2013). However, in order to facilitate their use as contaminant biomonitoring tools, detailed studies are needed to determine how eggshells and guano reflect sources of Hg exposure and potential impacts to the health of individual birds. For example, eggshells are primarily composed of calcium carbonate with little accumulation potential for Hg unless membrane tissues remain in the egg. Thus, it's possible that eggshells are more reflective of surface contamination than of egg contents. If egg membranes contribute Hg to eggshells, then accounting for the amount of membrane remaining in the egg may be important in order to standardize measurements. Similarly, guano represents a combination of undigested food, urine, and metabolic wastes. Mercury in guano likely reflects some combination of unassimilated Hg from prey, as well as remobilized tissue-bound Hg associated with waste. Controlled studies and paired tissue analyses are needed to determine what Hg in these tissues represents.

Mercury in blood reflects recent dietary exposure generally within 2–3 months (Stickel et al., 1977). Although mean blood Hg concentrations were higher at Icy Bay in 2009 (0.425 ± 0.031 µg/g) compared to 2008 (0.260 ± 0.076 µg/g), these mean values are below the suggested toxic threshold of 1.0–3.0 µg/g ww established for other piscivorous birds (Burger and Gochfeld, 1997; Evers et al., 2004; Evers et al., 2008; Ackerman et al., 2016). Furthermore, Ackerman et al. (2016) identified a "lower risk" category of avian blood Hg concentrations between 0.2 and 1.0 µg/g ww. All Kittlitz's murrelet's sampled had blood Hg concentrations below the upper limit of this category (range: 0.036–0.680 µg/g ww, $n = 87$). These results suggest that during the breeding season Kittlitz's murrelets at Icy Bay, are exposed to low concentrations of dietary Hg, and the values reported in this study likely do not represent a toxicological risk to individuals.

Little is known about Kittlitz's murrelet molting or wintering areas, which makes understanding sources and patterns of Hg exposure difficult. Recent data on post-breeding movements of 35 Kittlitz's murrelets in the Aleutian Islands and south-central Alaska (Piatt, *unpublished data*) and southeastern Alaska (Kissling, *unpublished data*), demonstrated similar migration routes westward to the Bering Sea and then northward to the Chukchi Sea. Based on this information, we hypothesized that alternate breast feathers would have similar concentrations of Hg at all sampled sites in the present study, reflecting a similar overwintering area and presumably diet. Contrary to our prediction, we found that alternate breast feathers from Kittlitz's murrelets at Glacier Bay had significantly higher Hg concentrations compared to alternate breast feathers sampled from Icy Bay and Adak Island. Furthermore, we found that within a study site, these patterns varied. At Icy Bay, basic breast feathers had higher Hg concentrations than alternate breast feathers, and at Glacier Bay, the opposite pattern was observed with alternate breast feathers having higher Hg concentrations than basic breast feathers.

Our results indicate a positive relationship between alternate breast feathers and blood, and a negative relationship between basic breast feathers and blood of Kittlitz's murrelets (Fig. 2). Although the kinetics of Hg in feathers is complex and not fully understood, the significant positive relationship between alternate breast feathers and blood suggests that alternate breast feathers may be used as a non-invasive biomonitoring tissue for better understanding Hg exposure during the breeding season. Understanding specific molt patterns, migration routes, and overwintering areas of Kittlitz's murrelets will further elucidate patterns and potential sources of Hg concentrations in feathers observed in the present study.

Mean Hg concentrations for mixed breast feathers of Kittlitz's murrelets sampled at Icy Bay (1.21 ± 0.22 μg/g), Glacier Bay (9.31 ± 1.28 μg/g), Adak Island (5.15 ± 2.51 μg/g), and Agattu Island (2.06 ± 1.28 μg/g) are below the adverse effects threshold of 20.0 μg/g dw that has been suggested for avian species (Burger and Gochfeld, 1997; Bond et al., 2015). However, feathers sampled from two Kittlitz's murrelets at Glacier Bay (29.78 μg/g and 57.85 μg/g) and one Kittlitz's murrelet at Adak Island (37.17 μg/g) exceed the suggested toxic threshold of 20.0 μg/g dw. These data suggest that a small proportion of Kittlitz's murrelet individuals may be suffering from deleterious effects due to Hg exposure. Due to the sensitive conservation status of this species, potentially deleterious levels of Hg exposure in even a small proportion of the population, compounded with other conservation threats, warrants further study.

Although Hg concentrations in some feathers from our study are near the maximum reported levels for wild seabirds, laboratory studies that examined lethal doses of Hg in liver and brain, found that with the addition of selenium, the symptoms of Hg toxicity were greatly reduced (Hoffman and Heinz, 1998). Field studies have documented similar patterns (Kim et al., 1996; Burger, 1997; Burger et al., 2007; Ikemoto et al., 2004; Scheuhammer et al., 2007; McHuron et al., 2014). The interpretation of risk and potential health effects related to high Hg concentrations in Kittlitz's murrelets feathers should be considered in conjunction with selenium concentrations, in addition to physiological traits of Kittlitz's murrelets that may ameliorate adverse effects of Hg exposure.

5. Conclusion

Substantial variability exists among bird species in their sensitivity to Hg, and no information exists for Kittlitz's murrelet Hg thresholds for any tissue. Based on the feather Hg effect threshold of 20.0 μg/g dw, established for other seabirds, our results suggest that at least one Kittlitz's murrelet at Adak Island and two individuals from Glacier Bay may be exposed to high levels of Hg at some point during their annual cycle. Mercury concentration in blood of Kittlitz's murrelets from Icy Bay, feathers and eggshells from Agattu Island, and guano from Adak Island do not appear to represent high exposure rates, but more information on the physiological effects of Hg on Kittlitz's murrelets is needed. In addition to the apparent population declines in the core of their breeding range, and low reproductive success in the Aleutians (Kaler et al., 2009; Kaler *unpublished data*), northern Alaska (Kissling and Lewis, 2016), southcentral Alaska (Lawonn, 2012), and southeastern Alaska (Kissling et al., 2015), the effects of potentially high Hg exposure at Adak Island and Glacier Bay could further exacerbate Kittlitz's murrelets tolerance to other environmental stressors.

Author contributions

L.A.K and R.S.A.K conceived and designed the study. L.A.K, R.S.A.K., and M.L.K. collected samples. C.A.E. performed mercury analyses. A.L.B, L.A.K, and C.A.E. performed statistical analyses. L.A.K and R.S.A.K drafted the manuscript. A.L.B., C.A.E, and M.L.K contributed to manuscript development and preparation.

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