

Mercury contamination in Peregrine Falcons (*Falco peregrinus*) in coastal Washington, 2001–2016

Joseph G. Barnes,^{1*} Daniel E. Varland,² Tracy L. Fleming,³ Joseph B. Buchanan,⁴ and Shawn L. Gerstenberger⁵

ABSTRACT—Mercury (Hg) is a naturally occurring widespread and persistent contaminant globally, and its organic form is highly toxic to living organisms and is known to impact humans and wildlife. Our primary goal was to use feathers to establish a contemporary baseline of total Hg contamination levels in Peregrine Falcons (*Falco peregrinus*) that occur on the outer coast of Washington. We document concentrations of total Hg in feathers of 151 peregrines primarily captured on beaches from 2001 to 2016. Peregrines were captured throughout the year, with breeding and natal areas of most individuals undetermined. The bulk of our samples consisted of fourth secondary (s4) feathers, but we include fourth primary and undertail covert feathers for comparison. All s4 feather samples contained detectable concentrations of total Hg (range = 0.7–69.83 µg/g), with mean concentrations in hatch-year (HY) feathers (mean = 6.05 µg/g) significantly lower than in second-year (mean = 22.55 µg/g) and after-second-year (mean = 24.48 µg/g) feathers. We captured 23 individuals more than once to track total Hg concentrations over time (up to 12 years between first and last capture), detecting an increasing trend through their third year before stabilizing in subsequent years. All individuals first captured while in HY plumage and later recaptured ($n = 20$) exhibited an increased concentration of total Hg in later years (mean maximum difference over time = 25.39 µg/g). Our 16-year study illustrates widespread contamination of total Hg in peregrines captured in coastal Washington, with evidence of bioaccumulation within individuals and between age classes. Encouragingly, peregrines in HY plumage sampled during the final third of our study period exhibited a significantly lower mean total Hg concentration than the first two-thirds of our study. We detected greater total Hg concentrations in coastal Washington peregrines than in nearly all known published studies involving peregrines of various subspecies in North America and Europe, although additional research is needed to establish toxic effects levels in this species. *Received 30 July 2017. Accepted 22 August 2018.*

Key words: contaminant testing, *Falco peregrinus*, feathers, mercury, migration, Peregrine Falcon, Washington.

Contaminación por mercurio en halcones peregrinos (*Falco peregrinus*) en la costa de Washington, 2001–2016

RESUMEN (Spanish)—El mercurio (Hg) es un contaminante persistente global que se encuentra ampliamente distribuido de manera natural. Su forma orgánica es altamente tóxica a organismos vivos con impactos conocidos en humanos y vida silvestre. Nuestra meta principal fue utilizar plumas para establecer la línea base contemporánea de los niveles de Hg total en halcones peregrinos (*Falco peregrinus*) que se encuentran en la costa exterior de Washington. Documentamos las concentraciones de Hg total en plumas de 151 halcones capturados principalmente en playas de 2001 a 2016. Los peregrinos fueron capturados a lo largo del año y sus áreas natales y de anidación son indeterminadas. Gran parte de nuestras muestras de plumas consistió de cuartas secundarias (s4), aunque incluimos la cuarta primaria y infracoberteras caudales para comparación. Todas las plumas s4 contenían concentraciones detectables de Hg total (rango = 0.7–69.83 µg/g), con concentraciones medias en plumas de aves del primer año (media = 6.05 µg/g) significativamente más bajas que en plumas de individuos del segundo año (media = 22.55 µg/g) y de después del segundo año (media = 24.48 µg/g). Capturamos 23 individuos más de una vez para registrar concentraciones de Hg total a lo largo del tiempo (hasta 12 años entre la primera y última capturas), detectando una tendencia incremental hacia el tercer año antes de estabilizarse en años subsecuentes. Todos los individuos que fueron capturados en plumajes del primer año y capturados posteriormente ($n = 20$) incrementaron sus concentraciones de Hg total en años posteriores (media de la diferencia máxima al paso del tiempo = 25.39 µg/g). Nuestro estudio de 16 años ilustra la contaminación ampliamente distribuida de Hg total en *Falco peregrinus* capturados en la costa de Washington, con evidencia de bioacumulación individual y entre clases de edad. Es estimulante que aquéllos en plumaje del primer año muestreados durante el tercio final de nuestro periodo de estudio tenían medias de Hg total menores que las de los primeros dos tercios de nuestro estudio. Detectamos mayores concentraciones de Hg total en halcones costeros de Washington que en casi la totalidad de los estudios publicados conocidos que involucran a varias subspecies de este halcón en Norteamérica y Europa, aunque se requiere de investigaciones adicionales para establecer los niveles tóxicos y efectos en esta especie.

Palabras clave: mercurio, migración, plumas, pruebas de contaminación.

Mercury (Hg) is a naturally occurring, globally distributed element (Tchounwou et al. 2003) that

can pose a risk to human and wildlife health (Wolfe et al. 1998, Mergler et al. 2007). It persists in the environment, which contributes to dramatically increasing global concentrations largely driven by anthropogenic activities since the industrial revolution in the mid-1800s (Swain et al. 1992, AMAP 2011). Atmospheric deposition has been the major mode of Hg distribution and is a substantial driver of elevated global Hg levels,

¹ Nevada Department of Wildlife, Las Vegas, NV, USA

² Coastal Raptors, Hoquiam, WA, USA

³ Battle Ground, WA, USA

⁴ Olympia, WA, USA

⁵ Department of Environmental and Occupational Health, University of Nevada, Las Vegas, NV, USA

* Corresponding author: jrbarnes@ndow.org

specifically in oceanic, high latitude, and other areas downwind of intensive carbon-based industry (Sunderland et al. 2009, AMAP 2011). Recent research has indicated increasing Hg contamination in the northern Pacific Ocean, with modeling suggesting a basin-wide doubling of 1995 Hg levels by 2050 (Sunderland et al. 2009). Research has detected elevated Hg levels in wildlife samples from marine areas in western Washington (Calambokidis et al. 1984, Custer and Myers 1990, Henny et al. 1991, Paulson et al. 2010, Noel et al. 2011), the Aleutian Islands and southeastern Alaska (Ackerman et al. 2016), the Columbia River Estuary between Washington and Oregon (Anthony et al. 1993), the northern Pacific Ocean (Burger and Gochfeld 2009), and in Arctic Alaska (Perkins et al. 2016, Saalfeld et al. 2016). Furthermore, Herring et al. (2018) found Hg exposure in Common Ravens (*Corvus corax*) and Turkey Vultures (*Cathartes aura*) was 17–27 times greater in coastal areas of Washington and Oregon than in inland areas.

Using wildlife as a biomonitor of environmental contaminants can be a useful tool to identify areas of concern and track spatial and temporal contaminant trends (Evers et al. 2005, Bond et al. 2015, Gustin et al. 2016). Birds have a history of use as indicators of Hg contamination (Solonen and Lodenius 1990, Hahn et al. 1993, Burger and Gochfeld 2004), particularly because Hg is sequestered in feathers during feather growth (Appelquist et al. 1984), the concentration of which is correlated with dietary Hg consumption (Fimreite and Karstad 1971, Lewis and Furness 1991, Spalding et al. 2000), and it persists over time in stable disulfide bonds (Berg et al. 1966). Raptors and some piscivorous birds are top trophic level predators and, as such, are vulnerable to persistent environmental contaminants due to biomagnification. Consequently, such species are strong indicators of environmental pollution (Evers et al. 1998, Burger and Gochfeld 2004, Lodenius and Solonen 2013). Mercury contamination is typically greatest in aquatic systems (AMAP 2011), with highest levels found in birds from marine systems, followed by freshwater and terrestrial systems (Ackerman et al. 2016). Common Loons (*Gavia immer*; Evers et al. 1998), marine birds (Burger and Gochfeld 2004, Bond and Diamond 2009), wading birds (Frederick et al. 2004), Bald Eagles (*Haliaeetus leucocephalus*;

Anthony et al. 1993, Bowerman et al. 2002), and Osprey (*Pandion haliaetus*; DesGranges et al. 1998) have all been used to assess environmental Hg contamination. These species, and the bulk of avian contaminant studies thus far, tend largely to be limited to, or focused on, aquatic systems and often exhibit limited geographical distribution.

Peregrine Falcons (*Falco peregrinus*) can be useful biomonitors because they are distributed globally, nest across various aquatic and terrestrial habitats (e.g., marine systems, freshwater rivers and lakes, wetlands, arctic tundra, temperate forests, and open desert), have high breeding site fidelity, are long-lived, and have a diverse diet consisting largely of birds (White et al. 2013). Any spatial or temporal assessment of Hg exposure in peregrines should consider that many of their populations are migratory, and they often feed on birds with highly variable migration patterns with exposure risk of Hg throughout their life cycle. Previously, contaminant studies focusing on peregrines have looked at organochlorine pollutants and other chemical pesticides (Newton et al. 1989, Peakall et al. 1990, Franke et al. 2010), petroleum exposure (Seegar et al. 2015), and Hg (Lindberg and Mearns 1982, Dietz et al. 2006, Barnes and Gerstenberger 2015), among other trace metals and environmental contaminants (Ambrose et al. 2000). Mercury concentrations in feathers of peregrines are correlated with concentrations found in their prey (Lindberg and Odsjö 1983, Barnes and Gerstenberger 2015), although the toxic effect levels that may impair their reproduction, health, and survivorship are undetermined.

Our primary goal was to use feathers to establish a contemporary baseline of total Hg contamination levels in peregrines that occur on the Washington coast. In addition, we assessed changes in total Hg contamination over time within the sampled population and within recaptured individuals between years. We also sought to assess differences in total Hg by age class and sex, and between feather types.

Methods

Study area

Our study area consisted of 3 beaches on the outer coast of western Washington, USA (Fig. 1): Ocean Shores (23.5 km; 46°56'N, 124°10'W), Grayland (11.3 km; 46°45'N, 124°06'W), and

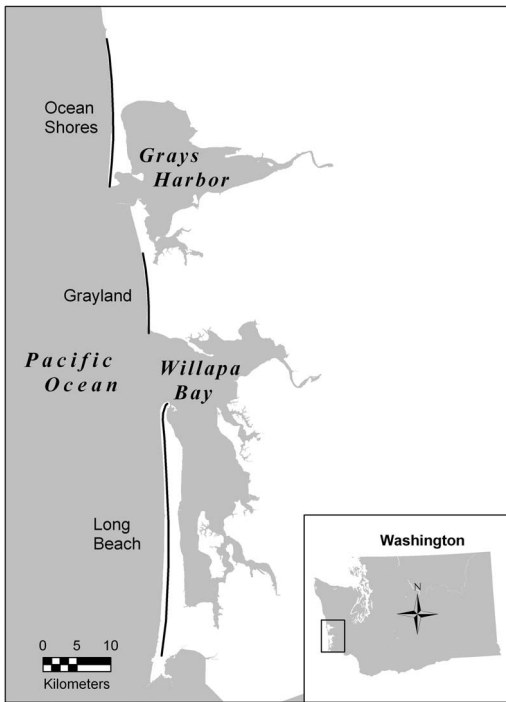


Figure 1. Location of the 3 study area beaches and Grays Harbor where we collected feather samples from Peregrine Falcons between 11 March 2001 and 27 April 2016 on the southern coast of Washington, USA. Segments of the beaches we covered in survey efforts are indicated by black lines.

Long Beach (39.9 km; 46°18'N, 124°04'W). These intertidal beaches contained unvegetated fine-grained sand and were bounded to the east by sand dunes stabilized primarily by European beach grass (*Ammophila arenaria*; Buchanan et al. 2001, Varland et al. 2012). In addition, we collected feather samples from peregrines captured on, or adjacent to, Grays Harbor.

Feather collection

We collected feathers from peregrines trapped during 735 raptor surveys between 11 March 2001 and 27 April 2016. Surveys generally began within 1 h of sunrise, lasted 2–4 h, and occurred in all seasons but with increased frequency from September to May (Varland et al. 2012). We captured an additional 7 peregrines at Grays Harbor (Fig. 1), 3 of which were breeding adults, 2 were nestlings, and 2 were of unknown origin with hatch-year (HY) and second-year (SY) plumage. We captured

peregrines primarily using harnessed Rock Pigeons (*Columba livia*) as lures (Bloom et al. 2007), with techniques further described in Varland et al. (2012).

Upon capture, we banded each peregrine and identified sex using wing chord, tail length, and culmen measurements (White et al. 2002). We assigned peregrines to age classes based on plumage (Pyle 2008), such that the plumage of HY peregrines was entirely juvenal feathers, while SY peregrines exhibited a mix of juvenal and second basic feathers, and after-second-year (ASY) peregrines exhibited the definitive basic plumage. At times, we classified peregrines as after-hatch-year (AHY) for pooled age class comparisons. To classify individuals to subspecies, we used sources of information identified in Varland et al. (2012), primarily using plumage characteristics. Some peregrines exhibited intermediate plumage and morphometric measurements; we did not identify these individuals to subspecies.

When possible, we recaptured previously banded peregrines to construct a time series of total Hg contamination by individual based primarily on sampled fourth secondary flight feathers (s4). Other research indicated minimal differences in total Hg concentrations in HY peregrines among feathers grown concurrently as nestlings (Barnes and Gerstenberger 2015). For this reason, we include 3 individual peregrines in which we initially sampled an alternate feather type on their first capture as HY birds but collected s4 feathers in future years. We first trapped 21 peregrines in HY or SY plumage, so all subsequent feather samples from these individuals were of known age.

We collected undertail covert feathers from 2001 to 2006 and s4 feathers from 2003 to 2016, standardizing collection of s4 feathers in 2003 because of the predictable timing of replacement during the molt cycle (Pyle 2008). Fourth secondary feathers are usually among the first 2 secondary feathers replaced each year (Pyle 2008), and breeding adult peregrines generally begin molt on territory during the nesting season (White et al. 2002). Furthermore, s4 was the feather recommended by the US Fish and Wildlife Service for collection for contaminant testing in the post-delisting monitoring plan (USFWS 2003). In autumn 2015, we began collecting fourth primary

flight feathers (p4) in addition to s4 feathers to assess differences between the 2 feathers and to facilitate comparison with other recent North American studies (Barnes and Gerstenberger 2015, Barnes et al. forthcoming). We used the North American feather numbering scheme where primaries are numbered distally (innermost to outermost) and secondaries are numbered proximally (outermost to innermost; Pyle 2008). We sampled undertail coverts by cutting near the base of the feather vane, excluding the calamus, using clean stainless steel scissors. When sampling s4 and p4 feathers, we removed the distal 1.5–2.0 cm portion of each feather. Adhering to a set length of standardized feathers allowed us to account for variable growth rates in various feathers (Bortolotti 2010). All feather samples were stored in paper envelopes at room temperature prior to analysis.

Feather analysis

We assessed total Hg in feathers using an AMA 254 atomic absorption spectrometer (method detection limit = 2.5 ng/g [ppb]; Leco Corporation, St. Joseph, MI, USA) at the Environmental and Occupational Health Laboratory at the University of Nevada, Las Vegas. Results are presented as total Hg on a fresh weight (fw) basis in $\mu\text{g/g}$ (equivalent to ppm). We cut feather samples to fit in a small nickel weigh boat for analysis. Analyzed portions of feather samples averaged 7.1 mg for undertail coverts ($n = 33$), 5.8 mg for s4 feathers ($n = 172$), and 7.0 mg for p4 feathers ($n = 9$). Samples underwent thermal decomposition (750 °C for 320 s), and were carried by ultrapure O₂ to a gold-plated amalgamator to determine total Hg using a 253.65 nm wavelength. We ran quality control tests every 10 samples using a method blank and 1 or 2 samples of standard certified reference material (CRM; CRM 2711; National Institute of Standards and Technology, Gaithersburg, MD, USA) to verify calibration. Accepted CRM recoveries ranged from 100% to 111% of the certified values of total Hg (mean = 106.2%, SE = 2.8, $n = 26$).

Statistical analysis

We first looked at total Hg concentrations in different feathers (undertail coverts vs. s4, s4 vs. p4) from the same bird in the same year. The significance of the relationships between feather

types was assessed using a major axis regression, while the ordinary least squares line equation was reported because it best describes the relationship (Legendre and Legendre 1998). We used a mixed-effects model with age class as a fixed effect and a random bird effect to assess differences in mean total Hg concentrations in s4 feathers, while pairwise differences were based on a Tukey post hoc test. In a separate analysis, we pooled individuals into HY and AHY categories to test for a possible interaction between sex and feather age class for total Hg concentration in s4 feathers. We employed a linear model with sex, feather age, and their interaction as fixed effects, using Tukey post hoc tests to explore differences in means.

An assessment of Hg contaminant levels in AHY individuals is complicated by seasonal movement, potential dietary shifts during the nonbreeding season, and possibly by accumulated body burden from preceding years, whereas Hg in feathers of HY birds reflect short-term, site-specific exposure of individuals during the nestling stage at the natal area (Evers et al. 2005). Thus, we focused our assessment of trends in total Hg concentration in s4 feathers across years solely on HY peregrines. To assess variation in concentration of total Hg in HY s4 feathers by year, we used a linear model (Pinheiro et al. 2016) with “year” as a random effect. We used a log likelihood ratio test to assess significance. To look at trends in mean total Hg concentrations in s4 feathers of HY peregrines over time, we ran an analysis of variance with a Bonferroni post hoc test for significance based on early (2003–2007), mid (2008–2011), and late (2012–2015) periods of similar length during our study. To assess change in total Hg concentrations in s4 feathers of peregrines captured in consecutive years, we used pairwise *t*-tests based on untransformed total Hg concentrations from individuals sampled at HY and SY, and at SY and third year (TY) age classes. For individuals recaptured several times over a period of years (i.e., maximum 12 years), we used the median value recorded for the age class for each individual when called for in categorical tests.

Unless otherwise noted, we log transformed total Hg concentrations prior to analysis to meet model assumptions of normal distribution and homogeneity of variance. We back-transformed reported values, report arithmetic means (SE), and consider results significant at $P \leq 0.05$. All

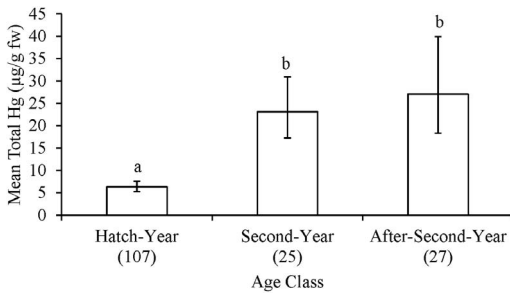


Figure 2. Concentrations of mean total mercury (Hg, µg/g fresh weight) by age class, as detected in fourth secondary flight feathers collected from Peregrine Falcons trapped in coastal Washington, USA, 2001–2016. Back-transformed mean total Hg concentrations are shown with 95% confidence intervals. Sample size is indicated in parentheses for each group. Different letters above bars indicate significant differences among means based on Tukey post hoc tests.

analyses were done in R3.3.0 (R Development Core Team 2016) using the *nlme* package (Pinheiro et al. 2016), the *predictmeans* package (Dongwen et al. 2014), or the *lmodel2* package (Legendre 2014).

Results

We collected feather samples from 151 peregrines in coastal Washington from March 2001 to April 2016. By subspecies, these individuals included 123 *F. p. pealei*, 6 *F. p. tundrius*, 4 *F. p. anatum*, and 18 of uncertain subspecies. In total, we analyzed feathers from 46 males, 103 females, and 2 peregrines of undetermined gender. Four individuals we captured were banded as nestlings by other researchers; 3 in the San Juan Islands, Washington, and 1 on Kiis Gwaii (Langara Island), British Columbia. We sampled feathers from 23 individuals that we captured more than once (range = 2–6 captures, mean = 2.9 captures/individual), spanning up to 12 years from first to last capture.

All s4 feather samples contained detectable concentrations of total Hg (range = 0.7–69.83 µg/g), with mean concentrations in HY feathers significantly lower than in SY and ASY feathers ($F_{2,156} = 70.4$, $P < 0.001$; Fig. 2). Correspondingly, total Hg concentrations in s4 feathers of HY peregrines (mean = 6.05 µg/g, SE = 0.36, $n = 105$) were significantly lower than in s4 feathers of AHY individuals pooled (mean = 23.11 µg/g, SE =

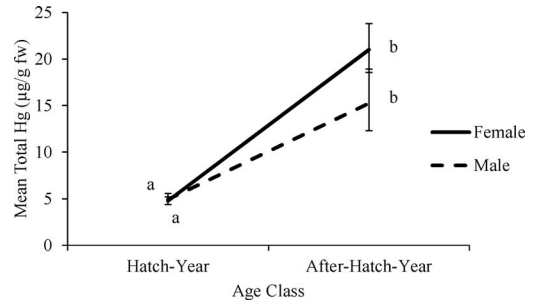


Figure 3. Concentrations of mean total mercury (Hg, µg/g fresh weight) by age class and sex, as detected in fourth secondary feathers collected from Peregrine Falcons trapped in coastal Washington, USA, 2001–2016. We compared hatch-year males ($n = 35$), hatch-year females ($n = 70$), after-hatch-year males ($n = 11$), and after-hatch-year females ($n = 33$). Back-transformed means are shown with 95% confidence intervals. Different letters indicate significant difference among means, determined using Tukey post hoc tests ($P \leq 0.05$).

1.74, $n = 44$; $t_{147} = 0.201$, $P < 0.001$). Our analysis of mean total Hg concentrations by sex did not detect differences between males and females in s4 feathers of HY or AHY peregrines ($F_{1,145} = 1.87$, $P = 0.17$; Fig. 3).

We observed significant yearly variation in total Hg concentration in HY peregrines (likelihood ratio = 9.7, $df = 1$, $P = 0.002$), but no significant trend was found over time (Fig. 4). An analysis of HY individuals pooled into early (2003–2007; mean = 7.73 µg/g, SE = 0.63, $n = 36$), mid (2008–2011; mean = 6.89 µg/g, SE = 0.59, $n = 35$), and late (2012–2015; 4.18 µg/g, SE = 0.44, $n = 36$) sampling periods indicated significant differences between periods ($F_{2,104} = 11.54$, $P < 0.001$), with significantly lower mean total Hg contamination in the late study period (Tukey post hoc test $P = 0.001$).

All individuals first captured while in HY plumage and subsequently recaptured ($n = 20$) exhibited an increased concentration of total Hg after HY (mean maximum difference over time = 25.39 µg/g, SE = 3.25, range = 5.64–59.74 µg/g). The greatest single increase between consecutive years was a change in total Hg of 39.2 µg/g (HY female captured in 2009 and recaptured in SY plumage in 2010). Of those individuals captured in their first and second year, there was a significant increase (mean = 520%) of total Hg from HY to SY in s4 feathers (mean difference between years

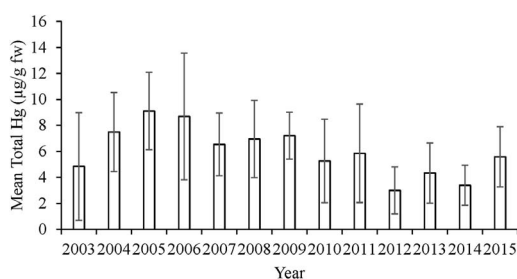


Figure 4. Concentrations of mean total mercury (Hg, $\mu\text{g/g}$ fresh weight), as detected in fourth secondary feathers collected from hatch-year Peregrine Falcons trapped in coastal Washington, USA, 2001–2016. Years indicate when feathers were grown. Back-transformed means are shown with 95% confidence intervals. The number of individuals sampled varied from 5 in 2003, 2006, and 2010, to a high of 13 in 2009.

$= 19.1 \mu\text{g/g}$, $\text{SE} = 2.63$; $t_{14} = -7.272$, $P < 0.001$). Total Hg concentrations in s4 feathers increased in 4 of 6 individuals from SY to TY, but our analysis was limited by small sample size and we did not detect a significant increase between these age classes (mean difference between years $= 10.19 \mu\text{g/g}$, $\text{SE} = 7.26$; $t_5 = -1.403$, $n = 6$, $P = 0.22$). Furthermore, we did not detect a clear trend in feather concentrations of total Hg in peregrines recaptured after their third plumage (Table 1). Indeed, in 4 individuals, highest total Hg concentrations occurred in their third or fifth plumage rather than in a following year.

When we collected more than one feather type from the same individual at the same time, we found a strong relationship between total Hg in s4 and p4 feathers ($n = 9$) and between total Hg in s4 and undertail covert feathers ($n = 12$; Fig. 5). The confidence intervals for slope and intercept were large because the comparisons were based on small sample size in both cases. Nevertheless, a strong linear relationship was found between p4 and s4 total Hg concentrations ($R^2 = 0.95$), with the 2 feather types containing nearly the same amount of total Hg (Fig. 5a). The linear relationship between undertail covert and s4 total Hg concentrations ($R^2 = 0.85$) was not as strong, with undertail coverts appearing to contain less total Hg (Fig. 5b), but we cannot exclude that the slope is 1 because the 95% CI included 1.

Discussion

Following recent research on year-round resident *F. p. anatum* in southern Nevada (Barnes and Gerstenberger 2015) and the highly migratory northern latitude *F. p. tundrius* and *F. p. anatum* (Barnes et al. forthcoming), our study further clarifies the broad-scale Hg contamination in peregrines across North America. Looking primarily at the subspecies *F. p. pealei* captured in our study area, we found total Hg contamination in all feathers across age classes and in both male and female peregrines collected from 2001 to 2016.

Table 1. Total mercury concentrations ($\mu\text{g/g}$ fresh weight) detected in fourth secondary flight feathers collected from Peregrine Falcons trapped along coastal Washington, USA, 2001–2016. Individuals are indicated by their Visual Identification Bands listed in the top row, and are those peregrines with known age recaptured after their third-year plumage. Plumage age (i.e., 1 = hatch-year, 2 = second-year, etc.) was determined by molt cycle (e.g., juvenal, second basic, definitive basic; Pyle 2008) and sequential plumage since first capture. All individuals were classified as the subspecies *Falco peregrinus pealei*, determined by plumage characteristics and measurements, and all but W/Z were female.

Plumage age	V/V	W/X	C/4	W/Z	P/5	D/2
1	9.22	3.52	9.0	10.1	9.0	4.01
2	—	29.74	—	—	22.65	10.72
3	59.88	29.87	24.09	24.95	—	—
4	32.77	—	—	61.99	28.08	12.68
5	—	—	37.64	69.83	—	—
6	—	28.27	—	—	—	—
7	—	—	34.63	—	—	—
8	23.77	—	—	53.5	—	—
9	25.3	—	—	—	—	—
10	22.58	—	—	—	—	—
11	—	—	19.41	—	—	—
12	36.36	—	—	—	—	—

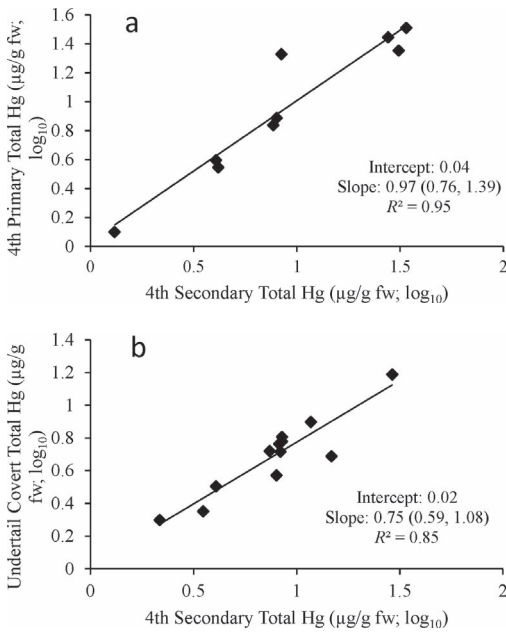


Figure 5. Relationship between concentrations of total mercury (Hg, $\mu\text{g/g}$ fresh weight; means \log_{10} transformed) detected in fourth secondary (s4) and fourth primary feathers (a; $n = 9$), and s4 and undertail covert feathers (b; $n = 12$) from Peregrine Falcons with both feather types collected from the same individual. Feather samples were collected from peregrines trapped on coastal beaches in Washington, USA, from 2001 to 2016. The relationship of the 2 different feathers from the same bird are indicated using the ordinary least squares slope and intercept, and significance is denoted by their 95% confidence intervals derived from major axis regression. Ordinary least squares trend lines are included for each comparison.

The average total Hg contaminant levels we found (HY = $6.05 \mu\text{g/g}$, ASY = $23.11 \mu\text{g/g}$) exceeded nearly all known published studies of various subspecies in North America and Europe (Table 2). However, it is important to consider which feathers were sampled because Hg body burden is reduced as growing feathers sequester Hg, so feathers replaced later in the molt cycle generally contain less Hg than those replaced earlier (Honda et al. 1986). The recent studies in southern Nevada (Barnes and Gerstenberger 2015) and the far northern latitudes of North America and Greenland (Barnes et al. forthcoming) report total Hg contamination detected in p4 feathers, and Dietz et al. (2006) assessed the fifth primary feather in western Greenland, whereas European studies in the 1970s utilized various feathers for analysis (Lindberg and Mearns 1982, Lindberg and Odsjö 1983). While the earlier European studies remain informative, we suggest limiting direct comparisons to studies analyzing feathers standardized between studies, or those with feathers replaced in molt at similar periods within the breeding and molt cycles.

The beaches of our study area are located near the southern limit of the breeding range of *F. p. pealei*, the coastal-breeding peregrine subspecies found in northwestern North America. *F. p. pealei* is known to breed from the western extent of its range in the Commander Islands of Russia, east to coastal Alaska, and southward along coastal British Columbia to Washington and, to a lesser extent, Oregon (White et al. 2013). Of those

Table 2. Mean mercury (Hg) concentrations ($\mu\text{g/g}$) detected in feathers from various studies of Peregrine Falcons. Subspecies are as reported by the authors. Hatch-year peregrines include nestlings sampled from eyries and fledged individuals in their juvenal plumage.

Location	Peregrine Falcon subspecies	Years	Hatch-year $\mu\text{g/g}$ Hg (n)	After-hatch-year $\mu\text{g/g}$ Hg (n)	Source
Western Washington	<i>pealei</i>	2001–2016	6.05 (105)	23.11 (44)	This study ^a
Southern Nevada	<i>anatum</i>	2012–2013	3.76 (24)	12.19 (25)	Barnes and Gerstenberger 2015 ^b
Northern Canada and Alaska ^c	<i>anatum</i> and <i>tundrius</i>	2009–2015	2.72 (120)	10.29 (105)	Barnes et al. forthcoming ^b
West Greenland	<i>tundrius</i>	1995–2004	0.66 (3)	6.11 (6)	Dietz et al. 2006 ^d
Northern Fennoscandia	<i>peregrinus</i>	1971–1978	8.31 (12)	17.6 (20)	Lindberg and Odsjö 1983 ^e
Southern Sweden	<i>peregrinus</i>	1971–1978	2.79 (7)	9.95 (9)	Lindberg and Odsjö 1983 ^e
Scotland	<i>peregrinus</i>	1975–1977	—	2.4 (10)	Lindberg and Mearns 1982 ^e

^a Results from fourth secondary feathers reported.
^b Results from fourth primary feathers reported.
^c Captured during autumn migration along coastal Texas and Maryland.
^d Results from fifth primary feathers reported.
^e Results from various combined feathers reported.

peregrines we identified to subspecies, 92% were *F. p. pealei*, so they were likely from natal and breeding grounds described earlier. *F. p. pealei* in the Aleutian Islands are thought largely to be year-round residents, whereas those found along mainland coastal North America include migratory individuals, with movements extending at least to Baja California (White et al. 2013). Individuals we banded were resighted by others on the Pacific Coast as far north as Seward, Alaska, and south to Newport, Oregon (Varland et al. 2012; DEV, 2018, unpubl. data). *F. p. pealei* primarily take sea birds, waterfowl, and shorebirds as prey throughout the year (White et al. 2013). As such, research indicating elevated Hg levels in the northern Pacific Ocean (Sunderland et al. 2009), in seabirds along the Aleutian Islands (Ricca et al. 2008), Bald Eagles in the Columbia River Estuary along the Washington and Oregon border (Anthony et al. 1993), and in Common Ravens and Turkey Vultures in coastal Washington and Oregon (Herring et al. 2018) all indicate likely high Hg exposure risk throughout most of the breeding range of *F. p. pealei*. Furthermore, elevated Hg concentrations were detected farther south in aquatic birds in San Francisco Bay (Eagles-Smith et al. 2009) in species that are potential peregrine prey, thereby posing a risk of Hg exposure to *F. p. pealei* on migration or wintering along coastal California (Anderson et al. 1988, Earnheart-Gold and Pyle 2001).

Concentrations of Hg in feathers of juvenile birds reflect short-term, site-specific exposure in proximity to nest sites, based on contamination of local prey sources at the time of feather growth. By contrast, Hg in feathers of adults reflect uptake since the previous molt and blood levels during feather growth (Evers et al. 2005). Therefore, in non-sedentary adults, the feather Hg level is an indication of Hg uptake across various locations during the breeding and nonbreeding seasons and during migration, and not solely of site-specific exposure. Total Hg concentrations we measured in HY peregrines are therefore more directly linked than our AHY peregrines to contamination from locally selected prey in natal areas along the western coast of North America, regardless of whether those prey were resident or migratory species. However, many of the avian species that peregrines prey on are migratory themselves, so while feathers of juvenile peregrines reflect site-

specific exposure based on diet, they do not indicate the source of contamination. Perkins et al. (2016) and Tsipoura et al. (2017) detected variable levels of Hg in migratory shorebirds at various migration stopover locations, illustrating variable exposure levels even sedentary predators are exposed to by preying on migratory species. Although an assessment of annual means in HY peregrines did not reveal a downward trend by year, our categorical assessment found that mean total Hg contamination in s4 feathers was significantly lower in the late sampling period (mean = 4.18 $\mu\text{g/g}$) compared to the early (mean = 7.30) and mid (mean = 6.89 $\mu\text{g/g}$) sampling periods. While this decline is encouraging, the complexity of discerning Hg exposure patterns from peregrines primarily with natal areas extending from coastal Washington to southeast Alaska preying on migratory species makes it difficult to determine exposure risk at specific locations. Nonetheless, our data may provide initial evidence of a regional coastal decline in Hg contamination in peregrines, although we are not aware of specific point-source emissions reductions to explain this decline.

We are not aware of toxicity studies specific to Hg concentrations in peregrines, and substantial species-specific differences in Hg toxicity thresholds have been documented in various bird species (Scheuhammer et al. 2007, Heinz et al. 2009). However, Eisler (1987) considered Hg concentrations in feathers as low as 5 $\mu\text{g/g}$ could be correlated with neuropathologies, with behavioral changes and reduced reproduction, whereas others suggested that feather Hg concentrations of 15 $\mu\text{g/g}$ may negatively impact reproduction in some predatory birds (Spry and Weiner 1991). By contrast, research on piscivorous raptors found no correlation between population declines in Osprey with feather Hg concentrations >40 $\mu\text{g/g}$ (DesGranges et al. 1998) or Bald Eagles at 21 $\mu\text{g/g}$ (Bowerman et al. 1994). While their research predated our investigation, Wilson et al. (2000) documented an increase of breeding pairs of peregrines from 2 to 17 during 1980–1998 in a coastal Washington study area immediately north of ours, suggesting the mean total Hg concentration we detected in AHY *F. p. pealei* (23.1 $\mu\text{g/g}$) may not yet be impacting the regional breeding population size. Similarly, Barnes and Gerstenberger (2015) detected mean total Hg concentra-

tions of 17.2 µg/g and 5.8 µg/g in feathers of AHY and HY peregrines, respectively, in an expanding local population in the southwestern United States (Barnes et al. 2015). Lindberg and Odsjö (1983) detected a mean Hg concentration of 17.6 µg/g in adult peregrine feathers in a declining population in Sweden after alkyl Hg was banned as an agricultural seed dressing in 1966; however, they were not able to distinguish between effects of Hg and chlorinated hydrocarbon pesticides. Consequently, although high concentrations of Hg in peregrines may be cause for concern, concentrations of Hg that impair individuals or populations have not been identified for this species.

Although our assessment of s4 feathers from peregrines trapped in coastal Washington from 2001 to 2016 indicates widespread total Hg contamination at levels higher than detected elsewhere, it is encouraging that Hg concentrations in HY peregrines exhibited a downward trend overall and significant decline in the last third of our study. Additional sampling is needed to determine if this resulted from an actual decline of Hg in the environment or random variation during our sampling time frame. Continued sampling, coupled with demographic monitoring of breeding territories and prey analysis, is needed to establish if thresholds exist above which Hg levels impact individuals or populations. Additionally, concentrations of contaminants in feathers are merely an indication of relative body burdens circulating in birds, so additional research to assess relationships between Hg in feathers, actual Hg dietary uptake, and Hg concentrations in blood are needed to better understand potential health and reproductive risks posed by the Hg exposure levels we detected in peregrines. We encourage research to more fully assess the utility of Peregrine Falcons as biomonitors of environmental contamination, and to ensure the conservation of this species and the avian prey assemblage it relies on into the future.

Acknowledgments

The Washington State Parks and Recreation Commission provided vehicle access to the beaches, which was essential for conducting our work. Coastal Raptors and Rayonier Inc. provided financial support. We thank those who helped in the field over the years, including M. K. Kenney, D. Moore, S. Tomlinson, D. Murnen, D. Miller, S. Miller, D. Larson, T. Rowley, and many others. Thanks also to V. Molenaar, who

designed and maintained the project database, and to C. Vanier for statistical analysis. We thank R. Keller and D. Cristol for their comments to improve this manuscript.

Literature cited

- Ackerman JT, Eagles-Smith CA, Herzog MP, Hartman CA, Peterson SH, et al. 2016. Avian mercury exposure and toxicological risk across western North America: a synthesis. *Science of the Total Environment*. 568:749–769.
- Ambrose RE, Matz A, Swem T, Bente P. 2000. Environmental contaminants in American and Arctic Peregrine Falcon eggs in Alaska, 1979–95. Fairbanks (AK): US Fish and Wildlife Service, Ecological Services. Technical Report NAES-TR-00-02.
- Anderson CM, Roseneau DG, Walton BJ, Bente PJ. 1988. New evidence of a peregrine migration on the west coast of North America. In: Cade TJ, Enderson JH, Thelander CG, White CM, editors. *Peregrine Falcon populations: their management and recovery*. Boise (ID): The Peregrine Fund; p. 507–516.
- Anthony RG, Garrett MG, Schuler CA. 1993. Environmental contaminants in Bald Eagles in the Columbia River estuary. *Journal of Wildlife Management*. 57:10–19.
- Appelquist H, Asbirk S, Drabaek I. 1984. Mercury monitoring: mercury stability in bird feathers. *Marine Pollution Bulletin*. 15:22–24.
- [AMAP] Arctic Monitoring and Assessment Programme. 2011. AMAP assessment 2011: mercury in the Arctic. Oslo (Norway): Arctic Monitoring and Assessment Programme.
- Barnes JG, Doney GE, Yates MA, Seegar WS, Gerstenberger SL. Forthcoming. A broadscale assessment of mercury contamination in Peregrine Falcons across the northern latitudes of North America. *Journal of Raptor Research*.
- Barnes JG, Gerstenberger SL. 2015. Using feathers to determine mercury contamination in Peregrine Falcons and their prey. *Journal of Raptor Research*. 49:43–58.
- Barnes JG, Haley RD, Thompson DB, Jaeger JR. 2015. Attributes of a breeding population of Peregrine Falcons associated with reservoirs on the Colorado River. *Journal of Raptor Research*. 49:269–280.
- Berg W, Johnels A, Sjostrand B, Westermark T. 1966. Mercury content in feathers of Swedish birds from the past 100 years. *Oikos*. 17:71–83.
- Bloom PH, Clark WS, Kidd JW. 2007. Capture techniques. In: Bird DM, Bildstein KL, editors. *Raptor research and management techniques*. Blaine (WA): Hancock House Publishers; p. 193–219.
- Bond AL, Diamond AW. 2009. Total and methyl mercury concentrations in seabird feathers and eggs. *Archives of Environmental Contamination and Toxicology*. 56:286–291.
- Bond AL, Hobson KA, Branfireun BA. 2015. Rapidly increasing methyl mercury in endangered Ivory Gull (*Pagophila eburnea*) feathers over a 130 year record. *Proceedings of the Royal Society B*. 282:20150032.
- Bortolotti GR. 2010. Flaws and pitfalls in the chemical analysis of feathers: bad news–good news for avian

- chemoecology and toxicology. *Ecological Applications*. 20:1766–1774.
- Bowerman WW, Evans ED, Giesy JP, Postupalsky S. 1994. Using feathers to assess risk of mercury and selenium to Bald Eagle reproduction in the Great Lakes Region. *Environmental Contamination and Toxicology*. 27:294–298.
- Bowerman WW, Roe AS, Gilbertson MJ, Best DA, Sikarskie JG, et al. 2002. Using Bald Eagles to indicate the health of the Great Lakes environment. *Lakes and Reservoirs: Research and Management*. 7:183–187.
- Buchanan JB, Johnson DH, Greda EL, Green GA, Wahl TR, Jeffries SJ. 2001. Wildlife of coastal and marine habitats. In: Johnson DH, O'Neil TA, editors. *Wildlife habitat relationships in Oregon and Washington*. Corvallis (OR): Oregon State University Press; p. 389–422.
- Burger J, Gochfeld M. 2004. Marine birds as sentinels of environmental pollution. *EcoHealth*. 1:263–274.
- Burger J, Gochfeld M. 2009. Comparison of arsenic, cadmium, chromium, lead, manganese, mercury and selenium in feathers in Bald Eagles, and comparison with Common Eider, Glaucous-winged Gull, Pigeon Guillemot, and Tufted Puffin from the Aleutian Chain of Alaska. *Environmental Monitoring and Assessment*. 152:357–367.
- Calambokidis J, Peard J, Steiger GH, Cabbage JC, Long RL. 1984. Chemical contaminants in marine mammals from Washington state. Rockville (MD): National Oceanic and Atmospheric Administration, Pacific Office. NOAA Technical Memorandum NOS OMS 6.
- Custer TW, Myers JP. 1990. Organochlorines, mercury, and selenium in wintering shorebirds from Washington and California. *California Fish and Game*. 76:118–125.
- DesGranges JL, Rodrigue J, Tardif B, Laperle M. 1998. Mercury accumulation and biomagnifications in Ospreys in the James Bay and Hudson Bay regions of Quebec. *Archives of Environmental Contamination and Toxicology*. 35:330–341.
- Dietz R, Riget FF, Boertmann D, Sonne C, Olsen MT, et al. 2006. Time trends of mercury in feathers of West Greenland birds of prey during 1851–2003. *Environmental Science and Technology*. 40:5911–5916.
- Dongwen L, Ganesh S, Koolaard J. 2014. Predictmeans: calculate predicted means for linear models. R package version 0.99 [cited 17 Aug 2018]. <https://CRAN.R-project.org/package=predictmeans>
- Eagles-Smith CA, Ackerman JT, De La Cruz SEW, Takekawa JY. 2009. Mercury bioaccumulation and risk to three waterbird foraging guilds is influenced by foraging ecology and breeding stage. *Environmental Pollution*. 157:1993–2002.
- Earnheart-Gold S, Pyle P. 2001. Occurrence patterns of Peregrine Falcons on southeast Farallon Island, California, by subspecies, age, and sex. *Western Birds*. 32:119–126.
- Eisler R. 1987. Mercury hazards to fish, wildlife, and invertebrates: a synoptic review. Laurel (MD): US Fish and Wildlife Service. *Biological Report* 85(1.10).
- Evers DC, Burgess NM, Champoux L, Hoskins B, Major A, et al. 2005. Patterns and interpretation of mercury exposure in freshwater avian communities in north-eastern North America. *Ecotoxicology*. 14:193–221.
- Evers DC, Kaplan JD, Meyer MW, Reaman PS, Braselton WE, et al. 1998. Geographic trend in mercury measured in Common Loon feathers and blood. *Environmental Toxicology and Chemistry*. 17:173–183.
- Fimreite N, Karstad L. 1971. Effects of dietary methyl mercury on Red-tailed Hawks. *Journal of Wildlife Management*. 35:293–300.
- Franke A, Setterington M, Court G, Birkholz D. 2010. Long-term trends of persistent organochlorine pollutants, occupancy and reproductive success in Peregrine Falcons (*Falco peregrinus tundrius*) breeding near Rankin Inlet, Nunavut, Canada. *Arctic*. 63:442–450.
- Frederick PC, Hylton B, Heath JA, Spalding MG. 2004. A historical record of mercury contamination in southern Florida (USA) as inferred from avian feather tissue. *Environmental Toxicology and Chemistry*. 23:1474–1478.
- Gustin MS, Evers DC, Bank MS, Hammerschmidt CR, Pierce A, et al. 2016. Importance of integration and implementation of emerging and future mercury research into the Minamata Convention. *Environmental Science and Technology*. 50:2767–2770.
- Hahn E, Hahn K, Stoeppler M. 1993. Bird feathers as bioindicators in areas of the German environmental specimen bank—bioaccumulation of mercury in food chains and exogenous deposition of atmospheric pollution with lead and cadmium. *Science of the Total Environment*. 139/140:259–270.
- Heinz GH, Hoffman DJ, Klimstra JD, Stebbins KR, Kondrad SL, Erwin CA. 2009. Species differences in the sensitivity of avian embryos to methylmercury. *Archives of Environmental Contamination and Toxicology*. 56:129–138.
- Henny CJ, Blus LJ, Grove RA, Thompson SP. 1991. Accumulation of trace elements and organochlorines by Surf Scoters wintering in the Pacific Northwest. *Northwestern Naturalist*. 72:43–60.
- Herring G, Eagles-Smith CA, Varland DE. 2018. Mercury and lead exposure in avian scavengers from the Pacific Northwest indicate potential risks for California Condor reintroduction efforts. *Environmental Pollution*. 243:610–619.
- Honda K, Nasu T, Tatsukawa R. 1986. Seasonal changes in mercury accumulation in the Black-eared Kite, *Milvus migrans lineatus*. *Environmental Pollution*. 42:325–334.
- Legendre P. 2014. lmodel2: model II regression. R Package version 1.7-2 [cited 17 Aug 2018]. <https://cran.r-project.org/web/packages/lmodel2/index.html>
- Legendre P, Legendre L. 1998. *Numerical ecology*. 2nd edition. Developments in environmental modelling 20. Amsterdam (Netherlands): Elsevier.
- Lewis SA, Furness RW. 1991. Mercury accumulation and excretion in laboratory reared Black-headed Gull chicks. *Archives of Environmental Contamination and Toxicology*. 21:316–320.
- Lindberg P, Mearns R. 1982. Occurrence of mercury in feathers from Scottish peregrines. *Bulletin of Environmental Contamination and Toxicology*. 28:181–185.

- Lindberg P, Odsjö T. 1983. Mercury in feathers of Peregrine Falcon compared with total mercury content in some of its prey species in Sweden. *Environmental Pollution*. 5:297–318.
- Lodenius M, Solonen T. 2013. The use of feathers of birds of prey as indicators of metal pollution. *Ecotoxicology*. 22:1319–1334.
- Mergler D, Anderson HA, Chan LHM, Mahaffey KR, Murray M, et al. 2007. Methylmercury exposure and health effects in humans: a worldwide concern. *Ambio*. 36:3–11.
- Newton I, Bogan JA, Haas MB. 1989. Organochlorines and mercury in the eggs of British peregrines (*Falco peregrinus*). *Ibis*. 131:355–376.
- Noel M, Ross P, Jeffries S, Lance M. 2011. Toxic contaminants in harbor seal (*Phoca vitulina*) pups from Puget Sound. Lacey (WA): Washington Department of Ecology, Final Report. Publication Number 11-10-001.
- Paulson AJ, Keys ME, Scholting KL. 2010. Mercury in sediment, water, and biota of Sinclair Inlet, Puget Sound, Washington, 1989–2007. Reston (VA): US Geological Survey. Open-File Report 2009-1285.
- Peakall DB, Noble DG, Elliott JE, Somers JD, Erickson G. 1990. Environmental contaminants in Canadian Peregrine Falcons: a toxicological assessment. *Canadian Field-Naturalist*. 104:244–254.
- Perkins M, Ferguson L, Lanctot RB, Stenhouse IJ, Kendall S, et al. 2016. Mercury exposure and risk in breeding and staging Alaskan shorebirds. *Condor: Ornithological Applications*. 118:571–582.
- Pinheiro J, Bates D, DebRoy S, Sarkar D, R Core Team. 2016. nlme: linear and nonlinear mixed effects models. R package version 3.1-128 [cited 9 Aug 2018]. <http://CRAN.R-project.org/package=nlme>
- Pyle P. 2008. Identification guide to North American birds. Part II. Anatidae to Alcidae. Point Reyes Station (CA): Slate Creek Press.
- R Development Core Team. 2016. R: a language and environment for statistical computing. Vienna (Austria): R Foundation for Statistical Computing [cited 8 Aug 2018]. <https://www.R-project.org>
- Ricca MA, Miles AK, Anthony RG. 2008. Sources of organochlorine contaminants and mercury in seabirds from the Aleutian archipelago of Alaska: inferences from spatial and trophic variation. *Science of the Total Environment*. 406:308–323.
- Saalfeld DT, Matz AC, McCaffery BJ, Johnson OW, Bruner P, Lanctot RB. 2016. Inorganic and organic contaminants in Alaskan shorebird eggs. *Environmental Monitoring and Assessment*. 188:276.
- Scheuhammer AM, Meyer MW, Sandheinrich MB, Murray MW. 2007. Effects of environmental methylmercury on the health of wild birds, mammals, and fish. *Ambio*. 36:12–18.
- Seegar WS, Yates MA, Doney GE, Jenny JP, Seegar TCM, et al. 2015. Migrating Tundra Peregrine Falcons accumulate polycyclic aromatic hydrocarbons along Gulf of Mexico following Deepwater Horizon oil spill. *Ecotoxicology*. 24:1102–1111.
- Solonen T, Lodenius M. 1990. Feathers of birds of prey as indicators of mercury contamination in southern Finland. *Holarctic Ecology*. 13:229–237.
- Spalding MG, Frederick PC, McGill HC, Bouton SN, Richey LJ, et al. 2000. Histologic, neurologic, and immunologic effects of methylmercury in captive Great Egrets. *Journal of Wildlife Diseases*. 36:423–435.
- Spry DJ, Wiener JG. 1991. Metal bioavailability and toxicity to fish in low-alkalinity lakes: a critical review. *Environmental Pollution*. 71:243–304.
- Sunderland EM, Krabbenhoft DP, Moreau JW, Strode SA, Landing WM. 2009. Mercury sources, distribution, and bioavailability in the North Pacific Ocean: insights from data and models. *Global Biogeochemical Cycles* 23, GB2010. doi:10.1029/2008GB003425
- Swain EB, Engstrom DR, Brigham ME, Henning TA, Brezonik PL. 1992. Increasing rates of atmospheric mercury deposition in midcontinental North America. *Science*. 257:784–787.
- Tchounwou PB, Ayensu WK, Ninasvili N, Sutton D. 2003. Environmental exposure to mercury and its toxicopathologic implications for public health. *Environmental Toxicology*. 18:149–175.
- Tsipoura N, Burger J, Niles L, Dey A, Gochfield M, et al. 2017. Metal levels in shorebird feathers and blood during migration through Delaware Bay. *Archives of Environmental Contaminants and Toxicology*. 72:562–574.
- [USFWS] United States Fish and Wildlife Service. 2003. Monitoring plan for the American Peregrine Falcon, a species recovered under the Endangered Species Act. Portland (OR): US Fish and Wildlife Service, Divisions of Endangered Species and Migratory Birds and State Programs, Pacific Region.
- Varland DE, Buchanan JB, Fleming TL, Kenney MK, Loughin TM. 2012. Peregrine Falcons on coastal beaches of Washington: fifteen years of banding and surveys. *Journal of Raptor Research*. 46:57–74.
- White CM, Cade TJ, Enderson JH. 2013. *Peregrine Falcons of the world*. Barcelona (Spain): Lynx Edicions.
- White CM, Clum NJ, Cade TJ, Hunt WG. 2002. *Peregrine Falcon (Falco peregrinus)*. In: Poole A, Gill FB, editors. *Birds of North America*. Ithaca (NY): Cornell Lab of Ornithology. <https://doi.org/10.2173/bna.660>
- Wilson UW, McMillan A, Dobler FC. 2000. Nesting, population trend and breeding success of Peregrine Falcons on the Washington outer coast, 1980–98. *Journal of Raptor Research*. 34:67–74.
- Wolfe MF, Schwarzbach S, Sulaiman RA. 1998. Effects of mercury on wildlife: a comprehensive review. *Environmental Toxicology and Chemistry*. 17:146–160.