

Historical patterns in mercury exposure for North American songbirds

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Abstract

Methylmercury (MeHg) is a global environmental contaminant that poses significant risks to the health of humans, wildlife, and ecosystems. Assessing MeHg exposure in biota across the landscape and over time is vital for monitoring MeHg pollution and gauging the effectiveness of regulations intended to reduce new mercury (Hg) releases. We used MeHg concentrations measured in museum specimen feathers (collected between 1869 and 2014) and total Hg concentrations (as a proxy for MeHg) of feathers sampled from wild birds (collected between 2008 and 2017) to investigate temporal patterns in exposure over approximately 150 years for North American songbirds. For individual species, we found greater concentrations for samples collected post-2000 compared to those collected during historic times (pre-1900) for six of seven songbird species. Mean feather concentrations measured in samples collected post-2000 ranged between 1.9 and 17 times (mean 6.6) greater than historic specimens. The proportion of individual songbirds with feather concentrations that exceeded modeled toxicity benchmarks increased in samples collected after 1940. Only 2% of individual songbirds collected prior to 1940 had feather concentrations greater than 2.4 $\mu\text{g/g}$ (a toxicity benchmark related to a 10% decrease in nest success) compared to 35% of individuals collected post-1940. Many species included in this study have a vulnerable or near-threatened conservation status, suggesting recovery actions are needed to address mercury pollution.

Keywords mercury, temporal trend, feathers, songbirds, museum specimens

Introduction

Methylmercury (MeHg) is a deleterious environmental contaminant produced from inorganic mercury (Hg) in aquatic environments (Gilmour et al. 2013). Human activities have substantially perturbed the global biogeochemical Hg cycle, enriching concentrations in surface environments (Amos et al. 2013, 2015). Efforts to reduce anthropogenic Hg emissions, such as the global agreement (Minamata Convention on Mercury) administered by the United Nations Environmental Programme (UNEP) are essential for protecting the health of humans, wildlife, and ecosystems (UNEP 2013a). Understanding how anthropogenic releases of Hg are related to MeHg exposures in biota is vital for gauging the effectiveness of global regulations (Evers et al. 2016; Gustin et al. 2016). A better understanding of MeHg exposure in bioindicator species across the landscape and over time can help focus regulatory and conservation efforts on regions and communities at greatest risk.

Methylmercury poses a significant health threat to wildlife (UNEP 2013b) and negative effects have been determined for multiple bird species (Seewagen 2010; Scheuhammer et al. 2011; Ackerman et al. 2016; Evers 2018). Recent research indicates that MeHg exposure in songbirds is associated with reduced reproductive success (Brasso and Cristol 2008; Hallinger and Cristol 2011; Jackson et al. 2011), disruption of the endocrine system (Wada et al. 2009), suppressed immune function (Hawley et al. 2009), changes in song (Hallinger et al. 2010; McKay and Maher 2012), and decreased flight performance (Carlson et al. 2014; Ma et al. 2018). During migration, risks may increase for songbirds because tissue catabolism during flight can result in a surge in blood MeHg concentrations during this already strenuous period (Seewagen et al. 2016). Many species of North American songbirds have experienced population

declines over the past approximately 50 years (Sauer et al. 2013), and MeHg exposure may play a role.

Previous studies have used birds as bioindicator species to better understand environmental MeHg risks across large spatial gradients and temporal scales (Vo et al. 2011; Keller et al. 2014; Bond et al. 2015; Jackson et al. 2015; Ackerman et al. 2016). Feathers are frequently used as a biomarker for MeHg exposure because they can be sampled easily and non-invasively. During molt, MeHg from recent exposure and accumulated MeHg remobilized from other body tissues is deposited into growing feathers (Honda et al. 1986; Braune 1987; Agusa et al. 2005). Methylmercury is incorporated with feather keratin and forms strong bonds with disulfides (Crewther et al. 1965). This allows MeHg concentrations within feathers to remain stable over time without special storage conditions (Appelquist et al. 1984). Additionally, most total Hg (THg) within feathers is present as MeHg (Online Resource Table 1). Archival feather samples from museum specimens can thus provide valuable insights into temporal changes in MeHg exposures of different bird populations (Evers et al. 2014; Bond et al. 2015; DuBay and Fuldner 2017; Movalli et al. 2017)

Prior studies that used museum specimens have investigated temporal trends in marine and freshwater bird species and information on temporal changes in MeHg exposure for terrestrial North American birds is lacking (Montiero and Furness 1997; Frederick et al. 2004; Vo et al. 2011; Head et al. 2011; Evers et al. 2014; Bond et al. 2015; Carravieri et al. 2016). Within the terrestrial environment, songbirds foraging within wetter habitats have been shown to have higher MeHg exposure (Jackson et al. 2015), and therefore may be at greater risk for associated adverse effects. Wetland habitats contain geochemical conditions conducive to the activity of

methylating microbes and thus can produce large quantities of MeHg that subsequently biomagnifies through the food web (Benoit et al. 2003).

The main objective of this study was to investigate temporal patterns in MeHg exposure of northeastern North American songbirds. To do this, we used feather samples from wild birds and museum specimens collected between the 1860s and present. We selected songbird species that forage and breed in wetter habitats such as marshes, swamps, bogs, and wet forests. Many of the selected songbird species are of conservation concern. We examined how MeHg exposures changed over time by species and discuss plausible links to changes in environmental contamination over the same time periods.

Materials and methods

Sample selection

We obtained flank feathers of seven songbird species from the Harvard Museum of Comparative Zoology ($n = 332$). Species included: olive-sided flycatcher (*Contopus cooperi*), red-eyed vireo (*Vireo olivaceus*), wood thrush (*Hylocichla mustelina*), saltmarsh sparrow (*Ammodramus caudacutus*), rusty blackbird (*Euphagus carolinus*), northern waterthrush (*Parkesia noveboracensis*), and palm warbler (*Setophaga palmarum*). In order to expand the timeline of samples for the olive-sided flycatcher, we also obtained flank feathers of specimens from the University of Alaska Museum Bird Collection for this species ($n = 13$). Selected specimens were collected between 1869 and 2014, with the majority collected prior to 1930 (87%). Most specimens selected were collected within northeastern North America during the breeding season (Table 1). Olive-sided flycatchers molt body feathers on the wintering grounds (Altman and Sallabanks 2012), therefore, we sampled breeding and wintering specimens for this species from collection locations across North and Central America (Table 1). We could not

select samples based on age or sex since these were unknown for many of the specimens in the Harvard Museum collection. A limitation of this study is thus potential differences in feather MeHg concentrations related to sex and age.

We also obtained flank feathers from the Harvard Museum of Comparative Zoology for yellow-bellied flycatcher (*Empidonax flaviventris*, $n = 67$) and boreal chickadee (*Parus hudsonicus*, $n = 69$), however, samples collected after 1930 were not available for these species. Additionally, flank feathers were obtained for a small number of northern waterthrush ($n = 30$) and wood thrush ($n = 2$) specimens sampled on their wintering grounds. The feather MeHg concentrations for these specimens were not included in analyses investigating temporal patterns in exposure and are provided in Online Resource Table 2 as a resource for future studies.

Feather THg concentrations for wild bird samples were selected from Biodiversity Research Institute's Hg database and included a total of 461 samples from seven songbird species. Selected feather samples were collected between 2008 and 2017, with the majority collected from 2015 to 2017 (90%). We aimed for these selected samples to be comparable with museum samples regarding collection locations, season, and feather type analyzed for THg concentrations (Table 1). Similar to the museum specimens, samples were not selected based on age or sex and we included birds likely to have occupied breeding habitats in northeastern North America (Table 1). We excluded any individuals sampled at study sites with a known source of Hg contamination. However, all of these samples were associated with other studies investigating MeHg exposure in songbirds. Lack of data for randomly selected sampling locations that would allow spatial extrapolation represents another limitation of this study.

Sample analysis

Inorganic Hg (mercuric chloride, HgCl_2) has commonly been used to preserve museum specimens and previous studies reported contamination of feather samples from museum collections (Head et al. 2011; Bond et al. 2015; Strekopytov et al. 2017). We investigated inorganic Hg contamination for 98 songbird specimens from the Harvard Museum of Comparative Zoology collected between 1861 and 1962. We found widespread inorganic Hg contamination of feathers as indicated by the low percentage of MeHg that comprised the THg concentrations in the feather samples (mean: $11.22 \pm 18.94\%$, range: 0.009-87.93%). Therefore, all museum specimen feathers were analyzed for MeHg concentrations at either Acadia University or Harvard University (Table 1). A small number of feather samples collected from wild birds were also analyzed for MeHg at Acadia University (northern waterthrush, $n = 2$; rusty blackbird, $n = 21$). Most feather samples collected post-2000 were analyzed for THg concentrations at Biodiversity Research Institute's Wildlife Mercury Research Laboratory. Since the majority of THg within feathers is in the form of MeHg for feathers from birds sampled in the wild (post-1989) and museum specimen samples (post-1936) not contaminated with HgCl_2 (Online Resource Table 1, mean across species = 101%), feather THg and MeHg concentrations are comparable. Additionally, while sample analysis for the current study was conducted using differing methods at multiple laboratories, quality assurance data for all analyses show that the data were of high quality across laboratories. Feather cleaning methods prior to analysis also differed among laboratories and therefore may have resulted in differences related to external surface Hg contamination. However, excluding HgCl_2 contamination of museum specimens, external Hg contamination should be minimal compared to internal Hg sources (Goede and DeBruin 1986).

Methylmercury

At Acadia University, feather samples were analyzed for MeHg concentrations using purge and trap, gas chromatography separation, and cold vapor atomic fluorescence spectroscopy detection following U.S. EPA Method 1630 (U.S. EPA 1998) with a Brooks Rand Model III analyzer, as detailed elsewhere (Edmonds et al. 2010, 2012). Prior to analysis, samples were washed in triplicate with trace grade acetone and Milli-Q deionized water to remove external debris. Samples were subsequently dried, and a subsample of 2-5 mg was weighed and put into 2 ml polypropylene screw top vials. Samples were digested using 1 ml of 25% KOH/MeOH by agitation for 1 h followed by heating for 1 h at 95 °C. A sample aliquot of 20 µl was transferred to a reaction bubbler and pH adjusted by the addition of sodium acetate buffer. Samples were ethylated with sodium tetraethylborate (NaBEt₄) and purged with argon for analysis. Quality control methods included analytical sample replication, internal standards, method blanks, and certified reference material (CRM, DOLT-4 fish liver; National Research Council of Canada). Average recovery for the CRM was $97.5 \pm 8.3\%$ ($n = 26$). Analytical precision, calculated as mean percent relative standard deviation (RSD), was 20.4% ($n = 8$) for method duplicates and 11.4% ($n = 6$) for analytical triplicates.

At Harvard University, museum specimen feathers analyzed for MeHg concentrations were cleaned prior to analysis using a surfactant (0.5% Triton X-100 solution) and sonication, followed by rinsing with ultrapure water three times. Feathers were freeze-dried overnight to remove moisture. Methylmercury analysis followed a modified U.S. EPA Method 1630 (U.S. EPA 1998) procedure previously reported (Li et al. 2016). Entire feather samples (1-3 feathers) were weighed, spiked with enriched Me²⁰¹Hg (2 ng/ml), and digested overnight at 70 °C using a 5N HNO₃ solution. Samples were neutralized with 8 N KOH, buffered using a 2 M acetate

buffer, ethylated with sodium tetraethylborate (NaBEt₄) and purged with argon for analysis. Analysis for MeHg was carried out using purge and trap and gas chromatography separation with a Tekran 2700 MeHg autoanalyzer coupled to a Thermo iCAP-Q ICP-MS. Quality control methods included CRMs (DOLT-5 fish liver, DORM-4 fish protein, and TORT-3 lobster hepatopancreas, National Research Council of Canada), reagent blanks, analytical duplicates, and ongoing precision and recovery (OPR) standards analyzed every 10 samples. Average daily recovery for the CRMs was $104.1 \pm 11.6\%$ ($n = 10$) for DOLT-5, $97.6 \pm 10.7\%$ ($n = 8$) for DORM-4, and $89.6 \pm 5.1\%$ ($n = 21$) for TORT-3. Analytical precision, calculated as RSD for CRMs and relative percent difference for analytical duplicates was $6.2 \pm 4.2\%$ and $9.7 \pm 6.9\%$ ($n = 32$), respectively. Accuracy and precision of OPRs were $97.8 \pm 10.9\%$ ($n = 38$) and $4.0 \pm 3.3\%$, respectively.

Total mercury

At Biodiversity Research Institute's Wildlife Mercury Research Laboratory (WMRL), feather samples were wiped clean of visible debris and the entire feather was cut in half, placed into a nickel boat, and weighed for analysis. All samples were analyzed using a thermal decomposition and atomic absorption spectrophotometry technique with a direct Hg analyzer (DMA 80, Milestone Incorporated) using US EPA Method 7473 (U.S. EPA 2007). Quality control methods, including the use of CRMs, two method blanks, and one sample blank were analyzed before and after every set of 30 samples. Six different CRMs were used over the course of the analyses (DOLT-4 and DOLT-5 fish liver and DORM-3 and DORM-4 fish protein, National Research Council of Canada; BCR-463 and CE-464 tuna fish tissue, The European Commission Institute for Reference Materials and Measurements). Average recoveries for the CRMs were 96.0% (DOLT-4), 99.2% (DOLT-5), 100.6% (DORM-3), 98.7% (DORM-4), 99.6%

(BCR-463), and 98.1% (CE-464). Analytical precision, calculated as RSD, for CRMs was within 10%.

Statistical analyses

Methylmercury and THg concentrations of feather samples are reported in micrograms per gram ($\mu\text{g/g}$) and referred to as Hg concentrations from this point forward for ease of discussion and comparisons. We report the mean and standard deviation of feather Hg concentrations by species and decade collected. Feather Hg concentrations were \log_{10} -transformed prior to all analyses to better meet assumptions of normality. We assessed differences in feather Hg concentrations among species for samples collected post-2000 using analysis of variance (ANOVA) followed by Tukey's HSD post hoc tests.

We examined the relationship between collection year and feather Hg concentrations for each individual species. Due to limitations in the availability of museum specimens, samples sizes were variable across collection years and samples were limited after the 1930s for some species. Therefore, statistical analyses were tailored to data availability for individual species. Northern waterthrush and olive-sided flycatchers had the most comprehensive datasets, therefore, we fit a linear regression and a generalized additive model (GAM), respectively, to decadal means to examine changes in feather Hg concentrations over time. Feather Hg concentrations for wood thrush, saltmarsh sparrow, and rusty blackbird were compared among collection years grouped by century (pre-1900, 1900 to 2000, and post-2000) using ANOVA tests followed by Tukey's HSD post hoc tests. Palm warbler feather Hg concentrations were compared between samples collected pre-1900 and post-2000 using a *t*-test. Similarly, red-eyed vireo feather Hg concentrations were compared between samples collected pre-1912 (only two samples were collected after 1899) and post-2000 using a *t*-test.

To understand the relationship between flank and tail feather Hg concentrations, we used separate linear regressions for four species (red-eyed vireo, wood thrush, saltmarsh sparrow, and palm warbler) that had both flank and tail feather Hg concentrations available within Biodiversity Research Institute's Hg database ($n \geq 15$ individuals per species, after hatch year only). All statistical analyses were conducted in R (Version 3.4.4).

Mercury risk benchmarks

We investigated how temporal patterns in MeHg exposure for the sampled songbirds were related to risks of adverse effects. Risk benchmarks based on feather THg concentrations are limited, though Jackson et al. (2011) modeled the association of nest success and body feather Hg concentrations for Carolina wren (*Thryothorus ludovicianus*). They determined that Carolina wren body feather THg concentrations of 2.4 and 6.2 $\mu\text{g/g}$ were associated with a 10% and 50% reduction in nest success, respectively. Therefore, we grouped all individual songbirds into body feather Hg risk categories: 1) $<2.4 \mu\text{g/g}$, low risk; 2) 2.4 to $<6.2 \mu\text{g/g}$, moderate risk; 3) $>6.2 \mu\text{g/g}$, high risk. We determined the proportion of songbirds with feather Hg concentrations that corresponded to each risk category for each species and separated them by collection year groupings (pre-1900, 1900-1999, post-2000).

Results

Feather Hg concentrations in this study varied by more than two orders of magnitude, with concentrations ranging from 0.052 to 9.5 $\mu\text{g/g}$ for samples collected pre-1900, from 0.035 to 6.1 $\mu\text{g/g}$ for samples collected from 1900 to 1999, and from 0.13 to 31 $\mu\text{g/g}$ for samples collected post-2000. Mean feather Hg concentrations varied across decades and differed among species (Table 2). For samples collected post-2000, rusty blackbirds ($8.7 \pm 7.3 \mu\text{g/g}$) and northern waterthrush ($5.9 \pm 5.3 \mu\text{g/g}$) had the greatest mean feather Hg concentrations, while olive-sided

flycatcher ($1.1 \pm 0.82 \mu\text{g/g}$) and red-eyed vireo ($0.46 \pm 0.21 \mu\text{g/g}$) had the lowest mean feather Hg concentrations ($F_{6,462} = 113, P < 0.001$, Fig. 1).

We found greater feather Hg concentrations for samples collected post-2000 compared to historic samples (collected pre-1900) for six out of the seven species examined (Table 3). In addition to having the greatest feather Hg concentrations for recently collected samples, rusty blackbirds showed the greatest increase in feather Hg concentrations over time (Table 3, Fig. 2). The mean feather Hg concentration for this species was 17 times greater for samples collected from 2008 to 2015 than those collected from 1871 to 1894, though no difference was found between the pre-1900 and the 1900 to 1928 collection period groupings ($F_{2,55} = 77, P < 0.001$). Palm warbler showed the second greatest increase in mean feather Hg concentrations, samples collected post-2000 were 8.9 times greater than historic samples ($t = 15, df = 27, P < 0.001$, Fig. 2). For northern waterthrush, we found a strong, increasing relationship for decadal mean Hg concentrations ($F_{1,8} = 73, P < 0.001, r^2 = 0.90$, Fig. 3). This species also had a large (6.1 times) increase in mean feather Hg concentrations between samples collected post-2000 and historic samples. Mean feather Hg concentrations for saltmarsh sparrow increased across all three collection year groups ($F_{2,206} = 73, P < 0.001$), with a 3.3 times greater mean concentration found for samples collected post-2000 compared to those collected during the pre-1900 period (Fig. 2). For olive-sided flycatcher, the trend in decadal mean feather Hg concentrations initially decreased until ca. 1900, which marked the lowest mean Hg concentration (Fig. 3). Mean feather Hg concentrations then increased from ca. 1900 through ca. 1950 and remained relatively steady into the 21st century (Fig. 3). Similarly, for wood thrush we saw no difference in mean feather Hg concentrations for samples collected after 1952, though we did observe a 1.9 times greater mean Hg concentration for samples collected from 2015 to 2016 compared to those collected

pre-1900 ($F_{2,44} = 9.4$, $P < 0.001$, Fig. 2). Red-eyed vireo was the only species for which we found no increase in Hg concentrations ($t = -0.078$, $df = 21$, $P = 0.94$). This species had the lowest mean post-2000 feather Hg concentration (0.46 ± 0.21 $\mu\text{g/g}$) and one of the highest feather Hg concentrations seen for specimens collected prior to 1900 (7.4 $\mu\text{g/g}$, collection year 1891).

The availability of archived museum feathers for Hg analysis and those from current day studies often differ by feather type, which can limit studies interested in assessing MeHg exposure in birds over time. We compared tail and flank feathers and found positive linear relationships between feather Hg concentrations for red-eye vireo ($P < 0.001$, $r^2 = 0.58$, $n = 102$) and saltmarsh sparrow ($P < 0.001$, $r^2 = 0.58$, $n = 75$), with a slightly weaker relationship found for palm warbler ($P = 0.002$, $r^2 = 0.46$, $n = 18$, Fig. 4). No relationship was observed between flank and tail feather Hg concentrations for wood thrush ($P = 0.27$, $n = 15$).

Discussion

Humans have been releasing mercury to the environment for centuries with the greatest increases in environmental concentrations occurring after the 1850s (Amos et al. 2013; Streets et al. 2017). The results of the current study indicate that MeHg exposure has increased for multiple species of North American songbirds over this same period. Mean feather Hg concentrations for samples collected post-2000 were between 1.9 and 17 times (mean 6.6) greater than historic samples (pre-1900) for six songbird species, with post-2000 sample concentrations >5 times greater for 3 species (Table 3).

Previous studies have also found increasing MeHg exposure over time for birds foraging in the marine environment (Montiero and Furness 1997; Vo et al. 2011; Evers et al. 2014; Bond et al. 2015; Carravieri et al. 2016), freshwater systems (Frederick et al. 2004), and for birds of prey (Dietz et al. 2006). Montiero and Furness (1997) found mean feather Hg concentrations

were 1.7 to 5.0 times greater for samples collected post-1990 compared to pre-1931 for five species of seabirds from the northeast Atlantic. Bond et al. (2015) found a much greater increase in feather Hg concentrations for an Arctic species, the ivory gull (*Pagophila eburnea*), with a 45 times greater feather Hg concentration found for the most recent collected sample (2004) compared to the earliest collected sample (1880). While increasing temporal patterns in MeHg exposure were not found for all species previously investigated (Head et al. 2011; Fournier et al. 2016), the results of the current study combined with previous research strongly indicate that birds are being exposed to increasing levels of MeHg over time.

In addition to historical pollution, shifts in foraging ecology and trophic position can lead to changes in Hg concentrations of biomarker tissues of birds. Foraging at a higher trophic position can result in greater MeHg exposure (Rimmer et al. 2010; Lavoie et al. 2013). Research on thick-billed murre (*Uria lomvia*) sampled from 1993 to 2013 found that temporal changes in prey consumption influenced changes in Hg biomarker tissue concentrations (Braune et al. 2014). Therefore, some studies using museum specimens to investigate temporal trends in Hg have included $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ stable isotope analysis to better understand temporal changes in foraging and trophic dynamics (Vo et al. 2011; Bond et al. 2015) We were unable to analyze $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ stable isotopes for the current study and, therefore, did not investigate whether temporal changes in prey consumption may have influenced feather Hg concentrations for the songbird species included in this study. Songbird feather Hg concentrations may also be influenced by changes in foraging habits prior to molt. Wood thrush, for example, switch from eating invertebrates to eating more fruit during late summer and fall when they are likely to be molting (Evans et al. 2011). Additionally, a recent study suggests that numerous songbird species may change locations between breeding and molting and that these molt-migrations may be

heterogeneous and influenced by a variety of factors (Pyle et al. 2018). We were unable to account for how these changes in location or foraging habits may have influenced feather Hg concentrations over time.

The current study allowed for the comparison of temporal patterns in MeHg exposure for bird species occurring in wet terrestrial habitats, including boreal forested wetlands, forested riverine systems, and coastal marsh, in addition to upland forests (Table 2). Previous research found greater Hg concentrations for songbirds occupying wet habitats compared to upland habitats (Jackson et al. 2015). We found the greatest increase in feather Hg concentrations for the rusty blackbird, a species that breeds in wet habitats. This species also had the highest mean feather Hg concentration for samples collected post-2000 across all species (Table 2). Previous studies have also found high MeHg exposure for rusty blackbirds during the breeding season in northeastern North America (Edmonds et al. 2010, 2012). Many of its life history traits may predispose this species to increased MeHg exposure. During the breeding season, it forages at ground level on terrestrial and aquatic invertebrates within northern boreal forested wetlands (Avery 2013) where MeHg is more available (Edmonds et al. 2012). However, the results of the current study indicate that this species was not always exposed to high levels of MeHg since feather Hg concentrations for rusty blackbird samples collected before 1930 were not elevated. Rusty blackbird populations have suffered population declines since the mid-1800s. However, the rate of decline has increased with an approximately 90% decline observed since 1980 (Greenberg and Droege 1999; Greenberg and Matsuoka 2010). While the loss of wooded wetland habitat on both the breeding and wintering grounds has likely played a large role in this decline (Greenberg and Droege 1999; Greenberg and Matsuoka 2010), MeHg exposure may also be a factor. The increasing feather Hg concentrations seen in the current study, combined with

the high rate of population decline, indicates that more research is needed to better understand the extent of adverse effects from MeHg exposure on rusty blackbirds.

We also found a large increase in mean feather Hg concentrations between historic (1888-1894) and post-2000 (2015-2017) samples for palm warbler (Fig. 2). This species may also exhibit life history traits that predispose it to increased MeHg exposure, as during the breeding season this species tends to forage on invertebrates both on the ground and aerially in bogs and wet coniferous forests (Wilson Jr. 2013), where MeHg is likely to be more available. It is important to note that the majority of historic samples were collected in Michigan and Western Ontario and consist of the western palm warbler subspecies (*S. p. palmarum*), while samples collected post-2000 were from Central to Eastern New York and consist of the yellow palm warbler subspecies (*S. p. hypochrysea*, Wilson Jr. 2013). Regional differences in MeHg contamination on the breeding grounds as well as possible differences in habitat use, foraging, and migratory patterns between subspecies may play a role in the increase documented in feather Hg concentrations for this species. Regardless, the elevated feather Hg concentrations found for samples collected post-2000 in the current study indicate that some eastern North America populations of palm warblers are likely at risk from adverse effects of MeHg exposure.

For northern waterthrush, we observed a linear increase in feather Hg concentrations over the approximately 150-year sampling period (Table 2, Fig. 3). This species inhabits forested riverine systems during the breeding season, foraging on invertebrates within this wet habitat (Whitaker and Eaton 2014). A recent study, which sampled northern waterthrush during southward migration, also indicated high MeHg exposure for this species (Seewagen 2013). While northern waterthrush populations are thought to be relatively stable (IUCN 2017), the

increasing temporal trend observed for this species indicates that it may be at increasing risk for adverse effects from MeHg exposure.

Feather Hg concentrations also increased across collection years for the saltmarsh sparrow (Table 2, Fig. 2), a species that forages on invertebrates within coastal marsh habitats (Greenlaw and Rising 1994). A previous study also indicated high Hg concentrations for saltmarsh sparrows sampled at numerous sites across northeastern North America during the breeding season (Lane et al. 2011). These high concentrations, as well as the increasing temporal trend in MeHg exposure, are of particular concern for saltmarsh sparrow due to its conservation status as vulnerable (IUCN 2017). In addition, saltmarsh sparrow face the threat of sea level rise due to their coastal habitat.

For olive-sided flycatchers, mean feather Hg concentrations increased during the first half of 20th century and then remained relatively steady after 1950 (Fig. 3). During the breeding season, this species also uses boreal forested wetlands where MeHg is more available (Edmonds et al. 2012). However, since olive-sided flycatchers molt on the wintering grounds in Mexico and Central America (Altman and Sallabanks 2012), their feather Hg concentrations are more likely to reflect MeHg exposure from this region than from its northern breeding grounds. Olive-sided flycatchers are also considered to be a species of high conservation concern (ADFG 2015; EC 2016; IUCN 2017) and are indicated to be experiencing sizeable populations declines at ~3.5% per year (Sauer et al. 2013). The current study suggests MeHg exposure has increased for olive-sided flycatchers, though to a lesser extent than for some other species. However, due to its declining populations, additional studies may be warranted to monitor MeHg risk for this species in the future.

We found increasing feather Hg concentrations for wood thrush, an upland forest species that forages predominantly on the ground (Evans et al. 2011). While the observed increase was less than that found for songbird species occurring in wetter habitats, these results indicate that some songbird species inhabiting upland forests during the breeding season are also at risk of increasing MeHg exposure.

For red-eyed vireo, another upland forest species, we found no difference between feather Hg concentrations for samples collected from 1879-1912 and those collected from 2015-2017. This species also had the lowest mean feather Hg concentration for recent samples (2010-2017), with a mean concentration between 2.5 and 18.8 times lower than that of the other species in this study (Table 2). Red-eyed vireo forage predominantly in the upper canopy (Cimprich et al. 2000), where MeHg may be less available within upland forests.

This study aimed to better understand temporal changes in available MeHg among terrestrial systems of eastern North America using songbirds as bioindicator species. Feathers are a commonly used MeHg biomarker tissue and they have previously provided an important resource for investigating temporal patterns in MeHg exposure in birds (Montiero and Furness 1997; Frederick et al. 2004; Dietz et al. 2006; Vo et al. 2011; Evers et al. 2014; Bond et al. 2015; Carravieri et al. 2016). However, the analysis of museum specimens for feather MeHg concentrations has some limitations. For instance, while feather THg concentrations have been shown to be stable over time and under a variety of conditions (exposure to UV, heat, freezing, and weathering, Appelquist et al. 1984), the long-term stability of MeHg concentrations in archived museum specimen feathers is unknown. Further, while research shows that the majority of THg within feathers is in the form of MeHg for museum specimen samples not contaminated

with HgCl₂ collected after 1936 (Head et al. 2011), the proportion of MeHg in feathers from museum specimens collected prior to the 1930s is unknown.

Additionally, the availability of feathers from specific feather tracts may differ among museum specimen samples and those collected during current day research, which may limit studies using museum specimens. A recent study found high intra-individual variation in feather THg concentrations for songbirds in the thrush and sparrow families (Low et al. 2019, this issue). Therefore, we investigated the relationship between flank and tail feather THg concentrations for 4 songbird species to better understand differences in THg concentrations among songbird feather tracks. We found a positive relationship overall between the two feather types (Fig. 4). However, the relationship differed among species, and no relationship was observed for wood thrush (Fig. 4). These limitations should be considered when comparing feather MeHg concentrations from museum specimens with THg concentrations from feathers collected during current day studies.

The increasing temporal trends in feather Hg concentrations found for songbirds in the current study also indicate that these songbirds are at an increased risk from the adverse effects of MeHg exposure. However, caution should be taken when using feather Hg concentrations to determine adverse risk for songbirds. While body feather THg concentrations related to reduced nest success for the Carolina wren (Jackson et al. 2011) provide the best toxicity benchmarks for comparison, benchmarks for other songbird species are unknown. Additionally, a recent study indicates that feather Hg concentrations may be poor indicators of Hg concentrations of internal tissues for some songbird species (Low et al. 2019, this issue). Nonetheless, using the benchmarks provided in Jackson et al. (2011), the current study found 19% of all songbirds sampled had feather Hg concentrations related to moderate (15%) or high (4%) risk categories

for adverse effects of MeHg exposure. We found that only 2% of individual songbirds collected between 1860 and 1940 had feather Hg concentrations related to moderate or high risk categories, while 34% of individuals collected after 1940 fell into these categories (Fig. 5). A recent study found a 36% decrease in blackpoll warblers (*Setophaga striata*) returning to the breeding grounds in the spring with tail feather THg concentrations $> 1.43 \mu\text{g/g}$ and a 16% decrease in spring returns for American redstarts (*Setophaga ruticilla*) with tail feather THg concentrations $> 1.95 \mu\text{g/g}$ (Ma et al. 2018a). While many other factors may play a role in these reduced return rates, the Ma et al. (2018a) study indicates that feather Hg concentrations lower than $2.4 \mu\text{g/g}$ might be related to negative health effects for migratory songbirds. Many North American songbird species are suffering population declines (Sauer et al. 2013), and several of the species included in the current study are considered to be of high conservation concern (IUCN 2017). Though our data cannot directly address causation, the combined pattern of increasing MeHg exposure and declining songbird populations is consistent with the notion that MeHg may be one of many mechanisms influencing declines in migratory birds, which is clearly cause for concern.

Conflict of interest The authors declare that they have no conflict of interest

Ethical approval All applicable international, national, and/or institutional guidelines for the care and use of animals were followed

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Table 1 The collection location, sample size (*n*), and date range (mo./day) for songbirds included in the current study. Songbirds are arranged by species and collection year groupings (pre-1900, 1900-1999, post-2000) and collection location is identified by state or province code within the USA and Canada or by country. Flank feathers collected unless otherwise indicated. The laboratories (Acadia University, Harvard University, and Biodiversity Research Institute's Wildlife Mercury Research Laboratory, WMRL) where samples were analyzed for Hg concentrations are included

Species	Collection year	Collection location (<i>n</i>)	Date range	Analysis laboratory (<i>n</i>)
Olive-sided flycatcher	1869-1899	Costa Rico (3), Colombia (1), ME (12), MI (2), NB (4), NH (2), NL (5), NY (19), VT (1), WI (1)	3/8-9/15	Acadia (27) Harvard (23)
	1900-1989	AK (5), ME (1), NL (3), YK (1), Costa Rico (3), Guatemala (4), Honduras (2),	1/8-12/15	Acadia (4) Harvard (15)
	2001-2017	AK (109), NH (5)	5/8-8/23	Harvard (7) WMRL (107)
Red-eyed vireo	1879-1897	NY (19)	4/15-10/14	Harvard (19)
	1908, 1912	NY (2)	2/10, 8/20	Harvard (2)
	2015-2017	NJ (3), NY (92)	6/1-7/24	WMRL (95)
Wood thrush	1880-1897	NY (24)	5/9-9/27	Harvard (24)
	1952-1972	MA (8), NJ (1)	5/1-9/22	Harvard (9)

	2015-2016	NY (14)	6/10-7/16	WMRL (14)
	1879-1897	MA (1), ME (2), NY (30)	2/7-11/2	Harvard (33)
Saltmarsh sparrow	1900-1955	MA (22), ME (2), NH (2), NS (3)	2/27-10/25	Harvard (29)
	2010-2017	MA (19), ME (7), NY (121)	6/16-9/26	WMRL (147)
	1871-1894	MA (2), ME (3), NL (10)	5/2-8/20	Acadia (12) Harvard (3)
Rusty blackbird	1900-1928	MA (15), NH (2), NL (3), QC (1)	1/30-10/29	Acadia (3) Harvard (18)
	2008-2015	All body feathers NH (18), NY (1), VT (3)	5/18-6/30	Acadia (21) WMRL (1)
	1861-1899	ME (2), MI (2)*, NB (1), NH (2), NL (35), NY (17), QC (1)	5/19-9/30	Acadia (43) Harvard (17)
Northern waterthrush	1902-1976	MA (5), ME (3), NH (2), NJ (1), NL (12), RI (6)	6/25-9/21	Acadia (14) Harvard (15)
	2010-2016	ME (breast feather = 55, flank feathers = 4)	5/8-9/14	WMRL (59)
	1888-1894	MI (24), NL (2), ON (1)	5/17-6/8	Acadia (27)
Palm warbler	2016-2017	NY (18)	6/13-7/21	WMRL (18)

*Samples excluded for analyses of the relationship between collection year and feather Hg concentrations for individual species

Table 2 Comparisons of feather Hg concentrations (arithmetic mean, standard deviation, and sample size) from songbird museum specimens and samples collected from wild birds across collection decades. Predominate habitat type used during the breeding season is included for each species

	Olive-sided flycatcher	Red-eyed vireo	Wood thrush	Saltmarsh sparrow	Rusty blackbird	Northern waterthrush	Palm warbler
	Wet boreal forest	Upland forest	Upland forest	Coastal marsh	Wet boreal forest	Riverine forest	Bog and wet forest
1860-1869	0.50 ± 0.18 (2)	0.81 (1)	.
1870-1879	0.59 ± 0.32 (23)	1.40 (1)	.	4.44 (1)	0.31 ± 0.30 (5)	0.54 ± 0.37 (2)	.
1880-1889	0.38 ± 0.19 (14)	0.33 ± 0.08 (11)	0.91 ± 0.71 (18)	0.88 ± 1.83 (28)	.	1.01 ± 0.52 (44)	0.37 ± 0.13 (24)
1890-1899	0.27 ± 0.19 (11)	1.73 ± 2.63 (7)	0.62 ± 0.34 (6)	0.23 ± 0.05 (4)	0.62 ± 0.37 (10)	0.88 ± 0.39 (11)	0.64 ± 0.06 (3)
1900-1909	0.19 ± 0.21	0.20	.	0.72 ± 0.43	0.30 ± 0.16	.	.

	(4)	(1)		(5)	(8)		
1910-1919	0.33 ± 0.15	0.08	.	0.63 ± 0.52	0.53 ± 0.87	1.57 ± 1.07	.
	(3)	(1)		(7)	(6)	(12)	
1920-1929	0.49 ± 0.37	.	.	0.68 ± 0.36	0.50 ± 0.30	1.14 ± 0.57	.
	(2)			(5)	(7)	(2)	
1930-1939	1.06 ± 0.77	.	.	0.57 ± 0.11	.	.	.
	(2)			(4)			
1940-1949	.	.	.	0.55	.	.	.
				(1)			
1950-1959	0.71 ± 0.43	.	1.16	2.25 ± 1.90	.	2.65 ± 1.46	.
	(5)		(1)	(7)		(11)	
1960-1969	1.02	.	1.90 ± 1.12	.	.	2.04 ± 0.61	.
	(1)		(5)			(2)	
1970-1979	.	.	2.15 ± 0.35	.	.	2.43 ± 1.44	.
			(3)			(2)	
1980-1989	1.16 ± 0.31

	(2)						
1990-1999
2000-2009	0.62 ± 0.32	.	.	.	8.65	.	.
	(2)				(1)		
2010-2017	1.15 ± 0.83	0.46 ± 0.21	1.56 ± 0.72	2.96 ± 2.95	8.66 ± 7.52	5.85 ± 5.27	3.57 ± 2.19
	(112)	(95)	(14)	(147)	(21)	(59)	(18)

Table 3 Parameters for comparisons of feather Hg concentrations and collection year. Results indicate greater feather Hg concentrations over time for most songbird species sampled

Species	Years	<i>n</i>	Mean (SD)	Test	Test statistic	<i>P</i>
Olive-sided flycatcher	See Table 2			Generalized additive model (<i>k</i> = 7)	.	.
Red-eyed vireo	1879-1912	21	0.83 ± 1.60	<i>t</i> -test	-0.078	0.94
	2015-2017	95	0.46 ± 0.21			
Wood thrush	1880-1897	24	0.83 ± 0.64	ANOVA	9.4	< 0.001
	1952-1972	9	1.90 ± 0.87			
	2015-2016	14	1.56 ± 0.72			
Saltmarsh sparrow	1879-1897	33	0.91 ± 1.81	ANOVA	73.3	< 0.001
	1900-1955	29	1.03 ± 1.17			
	2010-2017	147	2.96 ± 2.95			
Rusty blackbird	1871-1894	15	0.52 ± 0.37	ANOVA	76.7	< 0.001
	1900-1928	21	0.43 ± 0.49			
	2008-2015	22	8.66 ± 7.33			
Northern waterthrush	See Table 2			Linear regression	72.9	< 0.001 <i>r</i> ² = 0.90
Palm warbler	1888-1894	27	0.40 ± 0.15	<i>t</i> -test	15.2	< 0.001
	2015-2017	18	3.57 ± 2.19			

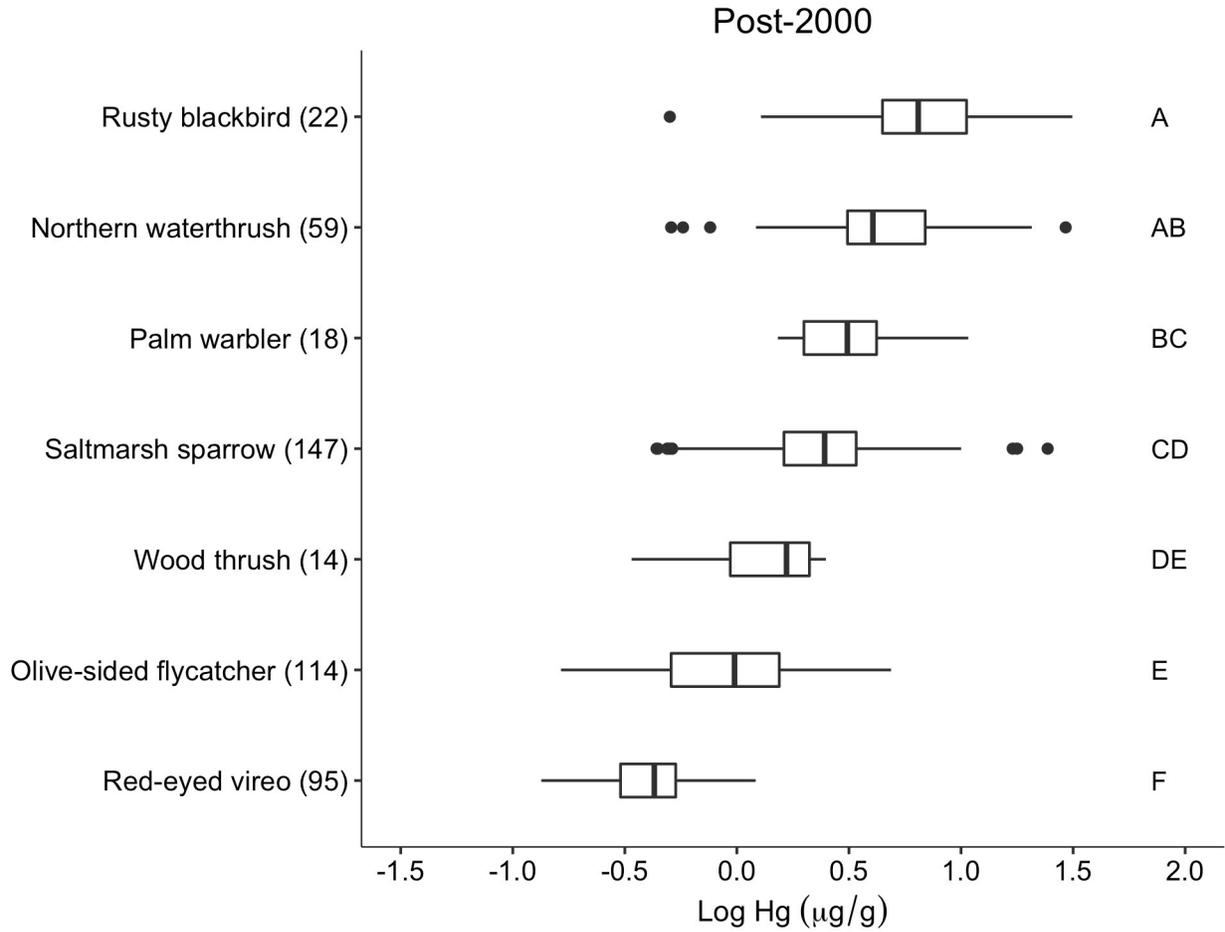


Fig. 1 Feather Hg concentrations differed among songbird species for samples collected post-2000. Box plots indicate median with the center line and the 1st and 3rd quartiles with the box outline. Species sharing the same letter are not significantly different ($P > 0.05$), sample sizes included in parentheses

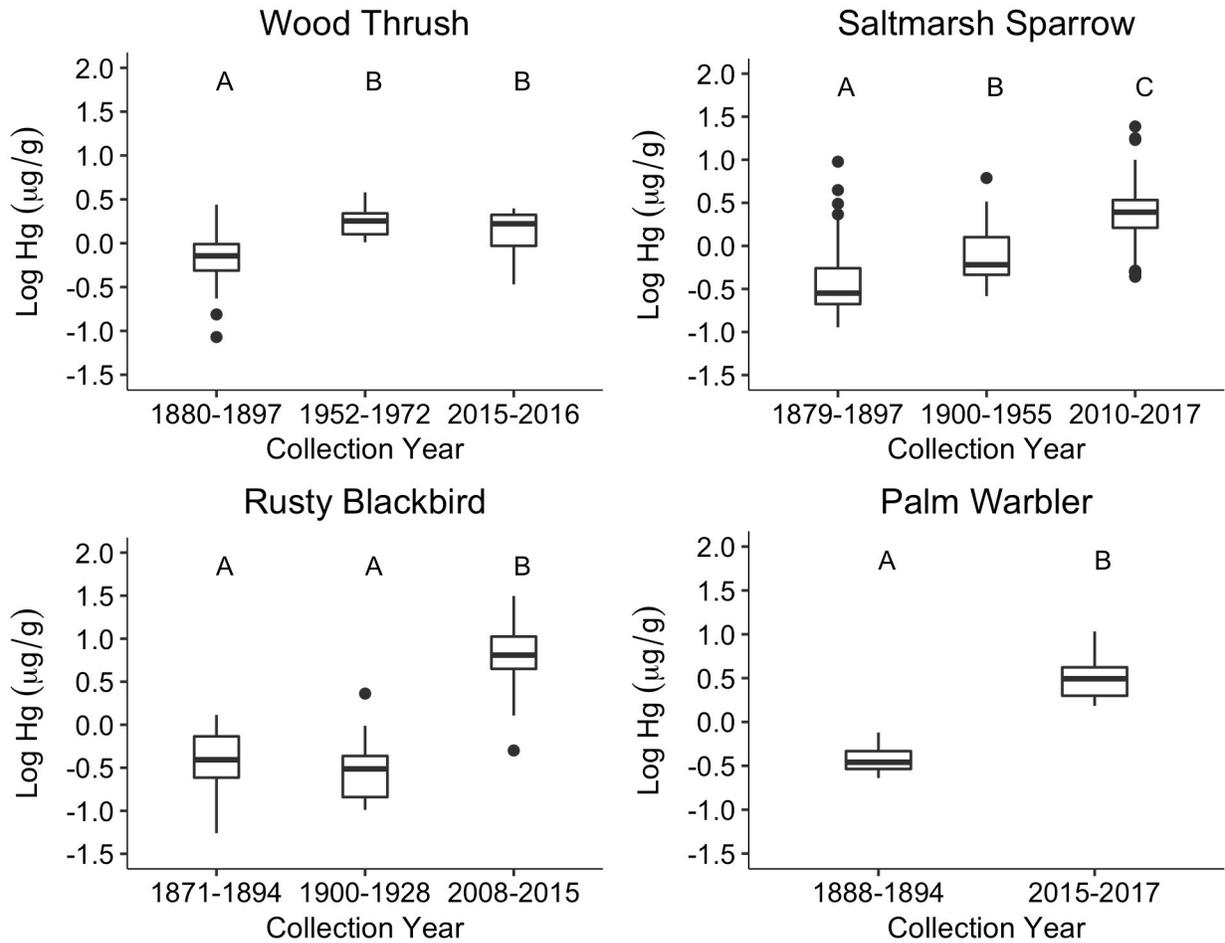


Fig. 2 Feather Hg concentrations differed among collection year groupings (pre-1900, 1900 to 2000, and post-2000) for four songbird species and greater Hg concentrations were seen for samples collected post-2000. Box plots indicate median with the center line and the 1st and 3rd quartiles with the box outline. For each species, groupings sharing the same letter are not significantly different ($P > 0.05$)

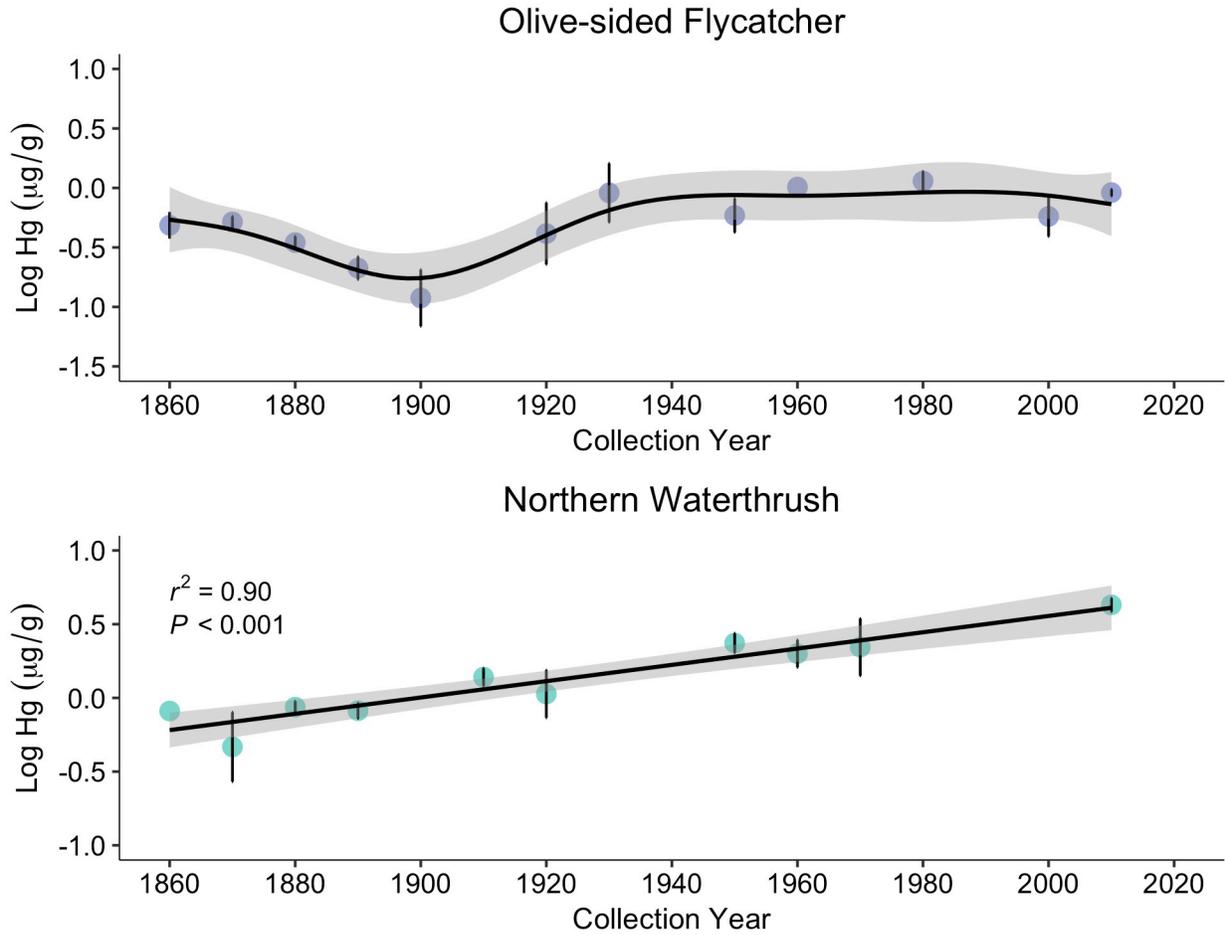


Fig. 3 Decadal mean feather Hg concentrations increased for olive-sided flycatcher from 1900 to approximately 1950, then remained stable based on a generalized additive model ($k = 7$, shaded region indicates standard error). Decadal mean concentrations increased over a ~150-year period for northern waterthrush, the coefficient of determination (r^2) and statistical significance (P -value) are provided for this linear regression (shaded region indicates standard error for the linear regression). For both species, dots and vertical lines represent decadal means and standard error

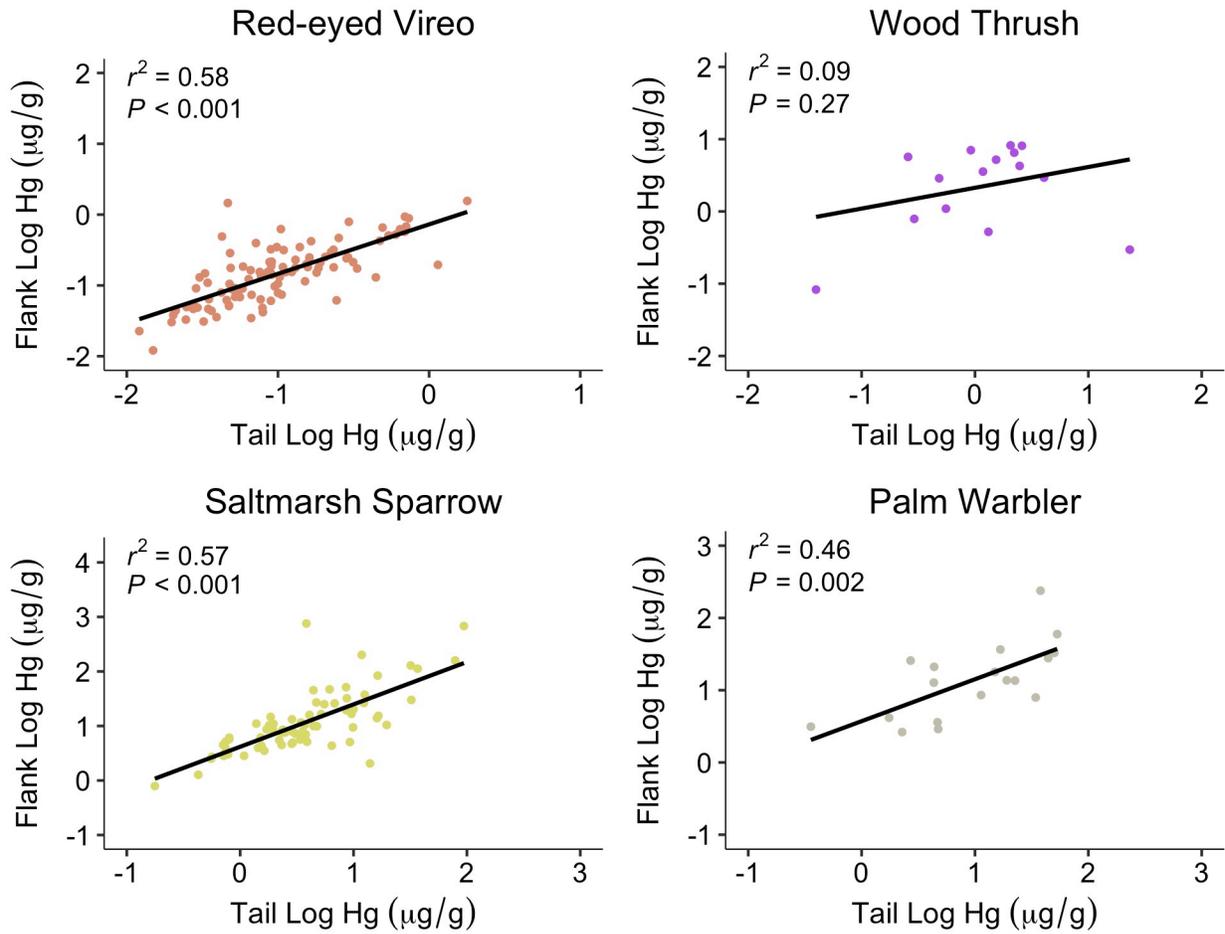


Fig. 4 Comparison of flank and tail feather Hg concentrations indicated a significant relationship for red-eyed vireo ($n = 102$), saltmarsh sparrow ($n = 75$), and palm warbler ($n = 18$), though no relationship was found for wood thrush ($n = 15$)

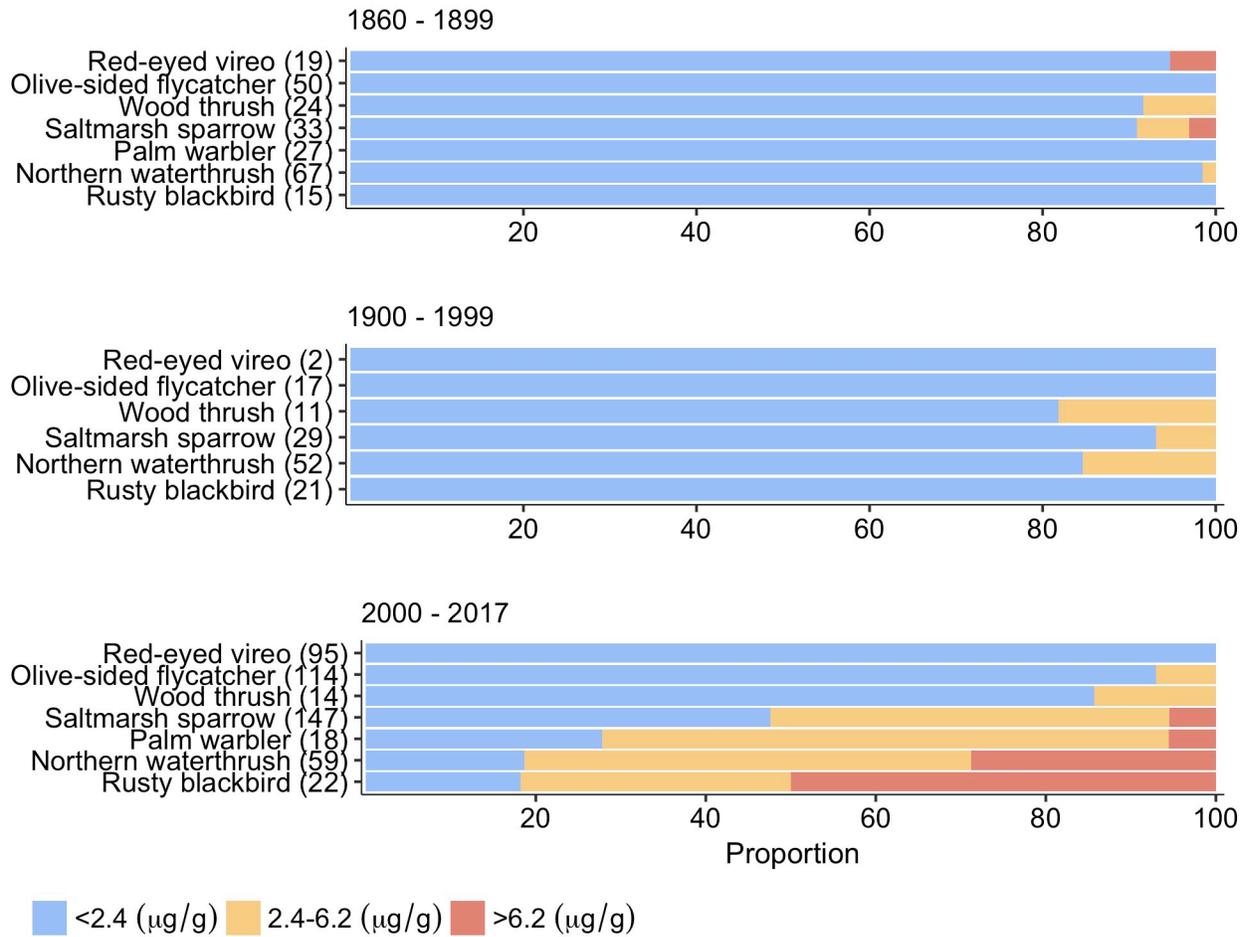


Fig. 5 Comparison of the proportion of individual songbirds for each species (sample size in parentheses) with feather Hg concentrations falling within risk categories for adverse effects of MeHg exposure. Risk categories based on body feather concentrations: 1) <2.4 µg/g, low risk; 2) 2.4 to <6.2 µg/g, moderate risk; 3) >6.2 µg/g, high risk, adapted from Jackson et al. (2011). Samples separated by collection year groupings (pre-1900, 1900-1999, post-2000) indicate increasing risk over time

Supplementary information

Historical patterns in mercury exposure for North American songbirds

Ecotoxicology

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Table 1 Summary of mean, standard deviation (SD), and sample size (*n*) for percent MeHg (Montiero and Furness 1997; Bond and Diamond 2009; Edmonds et al. 2010) and percent organic Hg (Thompson and Furness 1989; Head et al. 2011) determined for bird feathers

Species	Feather type	Mean % (SD)	<i>n</i>	Reference
5 seabird species	Ventral feather (recent samples)	101.0 (4.1*)	10	(Montiero and Furness 1997)
Herring gull	Outermost secondary or flank feather (Historic samples)	90.8 (6.2)	NA	(Head et al. 2011)
Great blue heron	Outermost secondary or flank feather (Historic samples)	90.3 (4.2)	NA	(Head et al. 2011)
Arctic tern	Breast feather	95 (15)	5	(Bond and Diamond 2009)
Common murre	Breast feather	133 (33)	5	(Bond and Diamond 2009)
Common tern	Breast feather	114 (65)	5	(Bond and Diamond 2009)
Razorbill	Breast feather	82 (35)	5	(Bond and Diamond 2009)
Atlantic puffin	Breast feather	100 (50)	5	(Bond and Diamond 2009)
Leach's storm- petrel	Breast feather	99 (40)	5	(Bond and Diamond 2009)
Rusty blackbird	1 st primary or 2 nd secondary	97 (0.3)	5	(Edmonds et al. 2010)

Wandering albatross	Body feather	95	26	(Thompson and Furness 1989)
Sooty albatross	Body feather	97	7	(Thompson and Furness 1989)
Northern fulmar	Body feather	111	15	(Thompson and Furness 1989)
Shag	Body feather	118	14	(Thompson and Furness 1989)
Great skua	Body feather	107	14	(Thompson and Furness 1989)
Arctic skua	Body feather	77	9	(Thompson and Furness 1989)
Kittiwake	Body feather	92	14	(Thompson and Furness 1989)
Razorbill	Body feather	100	16	(Thompson and Furness 1989)
Common guillemot	Body feather	113	17	(Thompson and Furness 1989)
Puffin	Body feather	98	10	(Thompson and Furness 1989)

Mean		101 (13)		
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*Standard error reported

Table 2 Comparisons of flank feather Hg concentrations (arithmetic mean, standard deviation, and sample size) by collection decade for museum specimens of yellow-bellied flycatcher and boreal chickadee and winter collected wood thrush and northern waterthrush. The collection location, sample size (*n*), and date range (mo./day) for specimens are arranged by species and collection year groupings (pre-1900, 1900-1999, post-2000) and collection location is identified by state or province code within the USA and Canada or by country. The laboratories (Acadia University, Harvard University, and Biodiversity Research Institute’s Wildlife Mercury Research Laboratory, WMRL) where samples were analyzed for mercury concentrations are included

Species	Collection year	Mean ± SD (<i>n</i>)	Collection location (<i>n</i>)	Date range	Analysis laboratory (<i>n</i>)
Yellow-bellied flycatcher	1870-1879	0.32 ± 0.23 (16)	ME (16), MI (1), MN (2), NB (7), NH (2), NL (16), NY (13), QC		
	1880-1889	0.24 ± 0.08 (29)		6/10-8/14	Acadia (58)
	1890-1899	0.58 ± 1.04 (13)	(1)		
	1900-1909	0.24 ± 0.11 (2)	ME (1), NH (2), NL (5), NS (1)	5/25-7/27	Acadia (9)
	1910-1919	0.26 ± 0.09 (6)			
	1920-1929	0.14 (1)			
Boreal chickadee	1870-1879	0.50 ± 0.74 (9)		5/16-8/12	Acadia (40)

	1880-1889	0.24 ± 0.11	ME (7), NB (8), NH (18)	(2), NL (20), NY (1),		
	1890-1899	0.25 ± 0.14	QC (2) (13)			
	1900-1909	0.10 ± 0.04	(2)			
	1910-1919	0.69 ± 0.22	NB (2), NL (22), NS (13)	(2), QC (3)	5/16-8/20	Acadia (29)
	1920-1929	0.20 ± 0.11	(14)			
Wood thrush	1952, 1958	0.97 ± 0.29	Mexico (1), Guatemala (2)	(1)	2/15, 12/4	Harvard (2)
	1860-1869	0.93 ± 0.71	Costa Rico (2), (2)	Jamaica (2), Colombia (3)	10/19- 3/17	Harvard (7)
Northern	1890-1899	0.77 ± 0.80	(5)			
waterthrush	1900-1909	0.96 ± 0.37	(9)	Colombia (2), Costa Rico (3), Cuba (2),		
	1910-1919	0.87 ± 0.16	(3)	Dominican Republic (1), Grenada (1),	9/27-5/8	Harvard (23)
	1920-1929	1.51 ± 0.06	(2)	Guatemala (5),		

1930-1939	1.87 ± 1.68	Honduras (6), Jamaica (2), Mexico (1)
1940	20.7 (1)	
195-1959	1.02 ± 0.59	(5)
1961	0.93 (1)	

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