Time trends of mercury in feathers of West Greenland birds of prey during 1851–2003.

ARTICLE *in* ENVIRONMENTAL SCIENCE AND TECHNOLOGY · NOVEMBER 2006

Impact Factor: 5.33 · Source: PubMed

12 AUTHORS, INCLUDING:

Rune Dietz
Aarhus University

Frank Rigét
Aarhus University

Jon Fjeldså
University of Copenhagen

Søren Møller
Roskilde University

Time Trends of Mercury in Feathers of West Greenland Birds of Prey During 1851−2003

RUNE DIETZ,*,† FRANK F. RIGET,† DAVID BOERTMANN,† CHRISTIAN SONNE,† MORTEN T. OLSEN,† JON FJELDSÅ,‖ KNUD FALK,‡ MAJA KIRKEGAARD,† CARSTEN EGEVANG,§ GERT ASMUND,† FRANK WILLE,⊥ AND SØREN MØLLER#

Department of Arctic Environment, National Environmental Research Institute of Denmark, Frederiksborgvej 399, DK-4000 Roskilde, Denmark, Teglstrupvej 6A 1. tv, DK-2100 Copenhagen Ø, Denmark, Greenland Institute of Natural Resources, Greenland, Zoological Museum, University of Copenhagen, Universitetsparken 15, DK-2100 Copenhagen, Denmark, Thorsvej 18, 4180 Sorø, Denmark, Roskilde University Library, P.O. Box 258, DK-4000 Roskilde, Denmark

Temporal trends of mercury (Hg) in West Greenland gyrfalcons, peregrine falcons, and white-tailed eagles were determined over 150 years from 1851 to 2003. Hg was measured in the fifth primary feather. Results showed that Hg increased in the order gyrfalcon (lowest) < peregrine falcon (intermediate) < white-tailed eagle (highest). All species showed significant age accumulations, which were taken into account in the temporal trend analysis. Of eight time trend analyses (three species and three age groups of which one was missing), seven showed an increase in primary feather concentrations. Of these, four were significant at the 5% level, two were close to being significant, and one was not significant. The linear regressions of which three out of four showed significant increases were for juvenile and immature gyrfalcon and juvenile peregrine falcon, which covered only periods prior to 1960, owing to limited data from the last half-century. The two sample comparisons of Hg 10-year medians for adult peregrine falcons and juvenile and adult white-tailed eagles indicated a continued increase during recent decades. However, low levels of Hg in a few recent collections among gyrfalcons and peregrines could indicate a change in the increasing trend.

Introduction

The Arctic is assumed to act as a sink for global atmospheric mercury. Hence, mercury emission inventories, transport models, and the recent discovery of mercury depletion events from the atmosphere observed each spring in the Arctic emphasize the global nature of mercury pollution (*1*−*3*). Although mercury is a naturally occurring element, human activities worldwide have led to increased levels in the Arctic environment when compared with those of pre-industrial times (*2*). In some areas, mercury levels in the environment continue to increase, which is of particular concern as impacts are already suspected on fish, birds, and marine mammals (*4*). Recent reviews and assessments conducted under the Arctic Monitoring and Assessment Programme has demonstrated that some Arctic people may ingest sufficient mercury via their diet to negatively affect children's neural development (*5*). Current mercury emissions have decreased in Europe and North America, but these declines have been offset by increases in East Asia (*6*). Of particular concern are the rising emissions of mercury in Asia and the discovery of mercury depletion events in the Arctic (*3*). The global spread of this pollutant means that actions to reduce the problem need to take place on a global scale. Recent increases in mercury levels in some Arctic animals indicate that the threat to the Arctic ecosystems and people may be increasing (*6*, *7*). Very few data from soft tissue are available to provide information on long time trends from the Arctic (*8*). Riget and Dietz (*9*) have summarized the Greenland data, and the need for longer time series has been documented by power analysis (*8*, *10*). One approach is to look for Hg deposits in more stable tissue like feathers, hair, teeth, bone, and shell material. All these matrices are believed to be reliable indicators of the Hg body burden in biota (*11*−*14*). Our present investigation concentrates on the long-term trends in Hg concentrations in selected feathers of gyrfalcon (*Falco rusticulus*), peregrine falcon (*Falco peregrinus*), and white-tailed eagle (*Haliaeetus albicilla*) from West Greenland.

Materials and Methods

Sampling. The distal end of primary no. 5 (P5) feather (counted from inward) was sampled from all birds for mercury (Hg) analysis as these are believed to be shed and generated in Greenland during summer. However, from female gyrfalcons, the fourth (P4) primary feather was sampled to harmonize the periods of feather growth. In the falcons, molt patterns ensure that these feathers are generated during the breeding season (*15*).

Gyrfalcon. Primaries from 331 gyrfalcons collected in West Greenland were analyzed. The samples covered 244 juvenile, 13 immature, and 74 adult birds. The samples were obtained from the Zoological Museum of Copenhagen (ZMUC; 1851 to 1998), Greenland Institute of Natural Resources (GINR; 1998−2002) and two recently moulted feathers (1984 and 2003; see Table 1. for information on *N* relative to years).

Peregrine Falcon. Primaries from 110 peregrine falcons from West Greenland were analyzed. The samples covered 64 juvenile, 2 immature, and 44 adult birds. The samples were obtained from ZMUC (1859−2000), GINR (*N* = 3; 1998−2001), and recently moulted feathers from breeding birds (1981−2003, *N* = 17) from West Greenland.

White-Tailed Eagle. Primaries from 113 samples from white-tailed eagles from West Greenland were analyzed. The samples covered 35 juvenile, 33 immature, and 45 adult birds, obtained from ZMUC (1884−1996), GINR (1985−2003), NERI (1997−2000), and recently moulted feathers (1980−2003; *N* = 8) from West Greenland (Table 1).

Sex and Age Determination. Age of the birds was estimated according to Forsman (*16*). Sex was determined from the size of the birds (females being considerably larger than males, particularly peregrine falcon and white-tailed eagle), if the sex was not stated on the label. Feathers collected

* Corresponding author phone: +45 46301938; fax: +4546301914; e-mail: rdi@dmu.dk.
† National Environmental Research Institute of Denmark.
‖ University of Copenhagen.
‡ Teglstrupvej 6A 1. tv.
§ Greenland Institute of Natural Resources.
⊥ Thorsvej 18.
Roskilde University Library

10.1021/es0609856 CCC: $33.50
Published on Web 08/29/2006

TABLE 1. Number of Samples (*N*), Median, Mean, and Standard Deviation (SD) for Hg Analyses of West Greenland Gyrfalcon, Peregrine Falcon, and White-Tailed Eagle Fifth Primaries Within the 10 Year Intervals Given in Figures 1−3[a]

	juvenile		immature		adult	
year	median (*N*)	mean ± SD	median (*N*)	mean ± SD	median (*N*)	mean ± SD
			Greenland gyrfalcon			
1845−1854	0.38 (1.00)	0.38	.	.	.	.
1855−1864	3.36 (1)	3.36	.	.	.	.
1875−1884	0.43 (13)	0.53 ± 0.32	0.57 (1)	0.57	9.51 (2)	9.51 ± 7.22
1885−1894	0.61 (40)	1.45 ± 2.15	4.38 (2)	4.38 ± 0.72	6.39 (10)	6.75 ± 6.09
1895−1904	0.61 (73)	1.03 ± 1.12	2.65 (4)	3.19 ± 1.61	3.93 (28)	4.56 ± 4.17
1905−1914	0.55 (35)	1.30 ± 1.51	5.16 (4)	4.79 ± 3.77	5.37 (6)	6.63 ± 4.85
1915−1924	0.69 (49)	1.51 ± 1.95	10.8 (1)	10.8	7.73 (18)	10.4 ± 9.12
1925−1934	0.69 (20)	1.35 ± 1.33	15.0 (1)	15.0	0.97 (1)	0.97
1935−1944	2.77 (1)	2.77	.	.	.	.
1945−1954	0.23 (1)	0.23	.	.	.	.
1955−1964	1.00 (4)	2.20 ± 3.03	.	.	10.1 (2)	10.1 ± 9.20
1965−1974	0.17 (1)	0.17	.	.	.	.
1985−1994	0.51 (2)	0.51 ± 0.31	.	.	0.39 (5)	8.37 ± 17.0
1995−2004	0.24 (3)	0.57 ± 0.60	.	.	1.88 (2)	1.88 ± 1.24
N	244		13		74	
			Peregrine falcon			
1855−1864	1.03 (2)	1.03 ± 0.17	.	.	10.8 (1)	10.8
1885−1894	1.71 (3)	2.01 ± 1.00	.	.	5.06 (4)	8.14 ± 8.39
1895−1904	1.43 (15)	1.78 ± 1.04	.	.	4.14 (5)	4.43 ± 1.68
1905−1914	1.99 (14)	2.84 ± 2.85	2.79 (1)	2.79	5.47 (4)	4.75 ± 2.48
1915−1924	1.77 (20)	1.84 ± 0.58	7.45 (1)	7.45	4.45 (11)	4.90 ± 1.71
1925−1934	2.39 (4)	2.57 ± 0.90	.	.	4.25 (4)	5.94 ± 4.53
1975−1984	6.50 (2)	6.5 ± 3.97	.	.	5.31 (3)	6.50 ± 2.19
1985−1994	0.78 (1)	0.78	.	.	10.4 (6)	9.47 ± 2.57
1995−2004	0.65 (3)	0.66 ± 0.21	.	.	6.08 (6)	6.11 ± 1.49
N	64		2		44	
			White-tailed eagle			
1875−1884	1.90 (4)	2.64 ± 1.84	4.17 (1)	4.17	4.56 (1)	4.56
1885−1894	2.03 (1)	2.03	5.10 (2)	5.10 ± 4.22	4.21 (2)	4.21 ± 1.61
1895−1904	1.07 (1)	1.07	3.30 (4)	3.29 ± 1.22	5.12 (3)	5.15 ± 1.45
1905−1914	1.22 (5)	1.22 ± 0.39	2.00 (7)	2.29 ± 1.15	7.49 (6)	6.60 ± 2.67
1915−1924	1.62 (9)	2.02 ± 0.89	2.20 (9)	5.14 ± 5.78	4.03 (12)	5.34 ± 3.96
1925−1934	1.50 (1)	1.50	1.85 (3)	2.79 ± 1.63	9.02 (2)	9.02 ± 3.05
1935−1944	5.57 (1)	5.57	.	.	5.70 (1)	5.70
1945−1954	.	.	14.7 (1)	14.7	.	.
1965−1974	.	.	3.88 (2)	3.88 ± 3.27	.	.
1975−1984	1.33 (1)	1.33	.	.	6.21 (2)	6.21 ± 0.59
1985−1994	2.88 (6)	5.61 ± 5.44	2.90 (1)	2.90	10.2 (6)	11.9 ± 7.53
1995−2004	2.79 (6)	3.02 ± 1.14	1.68 (3)	5.22 ± 6.28	11.0 (10)	10.8 ± 4.52
N	35		33		45	
total *N*	342		48		163	

[a] Periods (.) indicate that data are missing.

at the nest site are believed to originate from adult female birds that moulted the same year.

Mercury Analysis. Analyses were performed at the Department of Arctic Environment laboratory (DAE). The outer part of the feather was cut by stainless steel scissors and put into a polyethylene plastic bag. The feather samples were rinsed in a standard detergent (RBS 35), containing a sodium hydroxide/sodium hypochlorite solution to clean the feathers and prevent possible external contamination. Approximately 0.5 g of tissue was transferred to the tarred Teflon liner of a Berghof stainless steel bomb. After the addition of 3 mL of 65% HNO_3 (Merck Suprapur), the bombs were closed and incubated for 12 h at 120−150 °C. Following a cooling period, the digests were transferred quantitatively to 50 mL screw-cap polyethylene bottles and adjusted to ca. 25 g weight using metal-free, deionized water (Millipore). Approximately 8% HNO_3 was used for all further dilutions. Mercury analyses were performed using hydride generation and the amalgam technique, as described by Nielsen and Dietz (*17*). The detection limit was 0.005 μg/g (mg/kg). All data are presented on a dry weight basis. Analytical quality was checked by repeating analyses and by the frequent analysis of various reference materials, especially TORT-1 (lobster hepatopancreas), a certified reference material supplied by the National Research Council of Canada (Marine Analytical Chemistry Standards Program), and the dried tuna laboratory reference material from the National Food Agency of Denmark. The DAE laboratory participates in the international intercomparison exercises conducted by the International Council for the Exploration of the Sea (ICES), EEC (QUASIMEME), National Research Council, Canada, and the Department of Fisheries and Oceans, Winnipeg, Canada (*18*, *19*).

Statistical Analyses. Analyses of variance and *t*-test of log-transformed Hg concentrations were used when testing for possible differences in mean concentrations between species, regions, or age groups (SAS Version 8e). The log-transformation was done in order to approximate data to the normal distribution and variance homoscedasticity. Tukey's *post hoc* test was used to test differences between group means. Prior to the time trend analyses, a rough estimate on species and age differences was generated from the entire dataset, as the age and species sampling did not prove to be unbalanced over time in a way that could confound with these comparisons. As differences in both variables (species and age groups) were detected, the following time trend analyses were conducted separately for

TABLE 2. Results of Weighted (with Number of Individuals) Linear Regression of Log-Transformed Median Hg Concentrations of Gyrfalcon, Peregrine Falcon, and White-Tailed Eagle from West Greenland in 10-Year Periods[a]

species	age group	period	test type	slope/ trend	*P*	*N*
gyrfalcon	juvenile	1880–1960	linear regression of log medians	0.0088	0.003	7
	immature	1880–1935	linear regression of log values	0.045	0.020	13[#]
	adult	1880–1960	linear regression of log medians	0.0044	0.50	6
peregrine falcon	juvenile	1860–1930	linear regression of log medians	0.011	0.012	6
	adult	1890–1930 vs 1980–2000	two period comparison of medians (*t*-test)	increase	0.075	5 vs 3
white-tailed eagle	juvenile	1880–1920 vs 1990–2000	two period comparison of medians (*t*-test)	increase	0.016	3 vs 2
	immature	1890–1930 vs 1970–2000	two period comparison of medians (*t*-test)	no	0.93	5 vs 2
	adult	1880–1920 vs 1980–2000	two period comparison of medians (*t*-test)	increase	0.11	5 vs 3

[a] *N* = Number of 10-year periods except for numbers marked with #, which represents N for single values. Trend abbreviations used: increase, an increasing trend was seen (see *P* for significance level); no, no trend was detected between the two calculated means.

each of the three species (gyrfalcons, peregrine falcons, and white-tailed eagles) and three age groups (juvenile, immature, and adult). Based on the number of samples and years covered, either log-linear regressions or comparison of means were chosen for the time trend analysis among the different species and age groups. The median Hg concentrations in 10-year periods were calculated to generate data points for the time trend analyses and to harmonize for differences caused by variance in numbers and variability in food selection. Medians based on only one Hg determination were omitted in the further analyses. A log-linear regression analysis was applied to test for temporal trends in cases (combinations of species and age groups) where the data gap was no longer than 40 years. In the case of immature Gyrfalcon, the log-linear regression analyses was performed on individual Hg concentrations as too few data exist to calculate medians in 10-year periods. Student's *t*-test of log-transformed Hg concentrations was used when testing for possible differences in mean concentrations between two well-defined time periods (SAS Version 8e).

Results

Species Comparison. A graphical comparison was employed to evaluate the general levels among the three species analyzed. This analysis revealed levels in the following order: gyrfalcon (lowest) < peregrine < white-tailed eagle (highest). A significant species difference for juvenile birds (Tukey *post hoc* test) showed that white-tailed eagles and peregrines had significantly higher Hg levels than the gyrfalcons. No significant difference could be detected between juvenile white-tailed eagles and peregrines, and neither among immature birds of all three species, which could be due to the relatively small sample size. An ANOVA showed a significant species difference on the log normalized concentrations of the adult birds as well. The Tukey *post hoc* test showed that the adult white-tailed eagles contained significantly higher levels of Hg than the adult gyrfalcons ($P < 5\%$). However, no significant difference could be detected between adult white-tailed eagles and peregrines or between peregrines and gyrfalcons. The following analyses were performed separately for the three species.

Age Accumulation and Sex Comparisons. To investigate the temporal Hg trend, feather concentrations in the birds were tested for comparability between each age group and sex across the three species.

Gyrfalcons. The ANOVA showed a significant difference in primary Hg concentrations between age/sex groups of gyrfalcons. The Tukey *post hoc* test showed that Hg in juvenile gyrfalcons from West Greenland was significantly lower than that in all the other groups. No significant difference between any other groups could be detected.

Peregrine. In general, Hg concentrations differed significantly among age/sex groups of peregrines from West Greenland. It was clear that juveniles had the lowest Hg concentrations, while adult females had the highest concentrations and adult males had the intermediate concentrations. Hg concentrations in adult female peregrines were significantly higher (Tukey's test, $P < 0.05$) than those in adult males. A regional comparison was carried out between birds from Southwest (SW) and Central West (CW) Greenland. Of the three comparisons of juveniles, adult females, and adult males, all three exhibited highest concentrations in Southwest Greenland. However, the difference was only significant in the case of the juveniles and therefore West Greenland samples were combined in the consecutive time trend analysis.

White-Tailed Eagle. In general, Hg concentrations increased in the order juvenile, immature, and adult white-tailed eagles. Adult birds carried significantly higher levels compared to younger age groups, while no significant differences were detected between immature and juvenile birds (Tukey *post hoc* test). Southwest Greenland birds were higher in Hg concentrations than birds from CW Greenland but not significantly so. Therefore, the two datasets were merged to a common West Greenland dataset for additional analysis. Data were too few to analyze for sex differences.

Temporal Trends. *Gyrfalcons.* Most data were available for gyrfalcons. Male and female gyrfalcons of all sub-regions were pooled to obtain a sufficient sample size, as the male/female ratio did not change over time. However, the three age classes were maintained in the temporal trend analysis. Log-linear regression analyses of median Hg concentrations of 10-year periods were carried out for juveniles and adults. For immature birds all single observations were used in the log regressions, as the sample size was too small to allow for the same procedure used for the other two age groups. All age groups showed a positive slope ranging between 0.44 and 4.5% increase per year. The juvenile birds from 1880–1960 showed a highly significant positive trend of 0.88% increase per year ($P = 0.003$) (Table 2; Figure 1A). The first and especially the last median, with a two decadal gap to the previous, exerts a leverage effect on the regression line and the *p*-value strengthening the significant positive trend. The two data points ($N = 2$ and $N = 3$) from the two recent decades were in the lower end of the total dataset (medians). However, these were not included in the calculation of the regression line due to the 60 year time span back to the body of the data. Immature gyrfalcons covering only the period from 1880–1935 likewise showed a significant 4.6% yearly increase ($P = 0.020$) (Table 2; Figure 1B). The last group of adults did not show a significant trend (Table 2; Figure 1C). The standard deviation of the Hg concentrations for adults was consider-

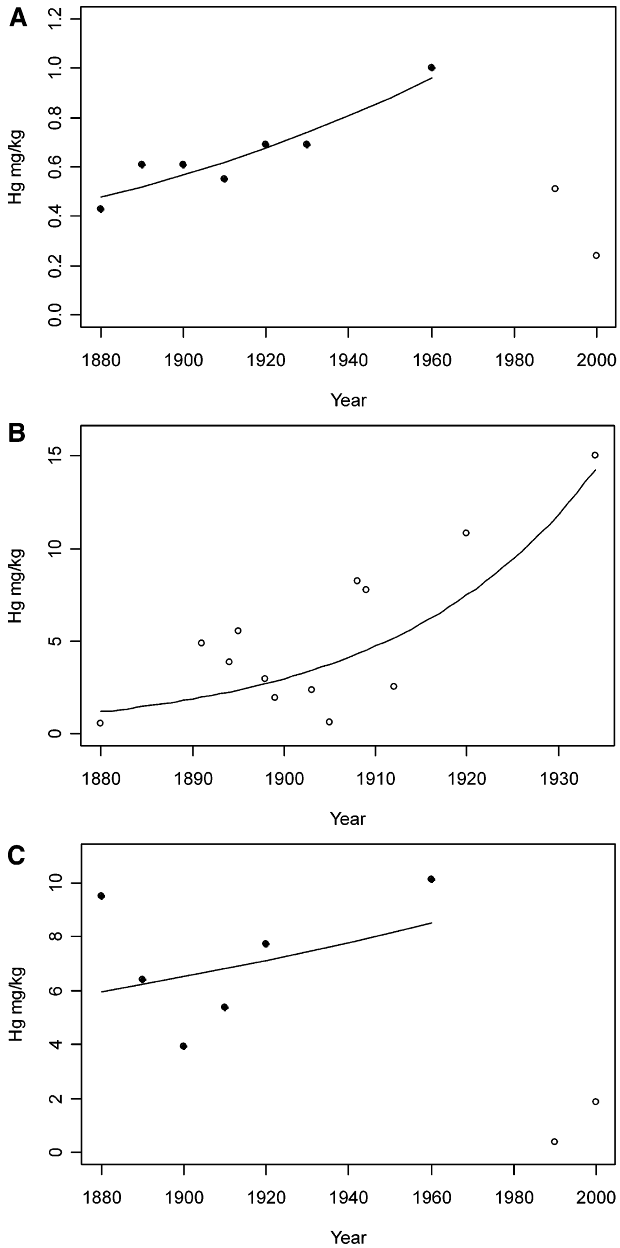

FIGURE 1. Mercury in primary feather no. 5 of gyrfalcon from West Greenland: (A) juvenile and (C) adult birds; lines are back-calculated linear regressions lines based on logarithmic medians of 10-year interval values (see Table 1); regression lines (solid) are based on black points and recent open circles not included; (B) immature birds; lines are back-calculated linear regressions lines conducted on logarithmic values of the analyzed concentrations ($n = 14$; one open circle represents one sample analysis) (see Table 1).

able, probably due to an effect of age and feeding differences between the sample periods. This variation had a pronounced impact on the outcome of the statistical analysis. In addition, this group only covered six 10-year periods between 1880 and 1960, with a significantly lower sample size compared to juveniles, which were represented by primaries generated during only the first year. As for juvenile birds, the last two decades indicated lower concentrations, and these were not included in the regressions for the same reasons as for juveniles.

Peregrines. Median log-transformed Hg concentrations of the 10-year periods were increasing significantly ($P = 0.012$) with a 1.1% increase per year for juvenile birds over the 10-year periods from 1860–1930, where the bulk of the data were from (Table 2; Figure 2A). The first and lowest median,

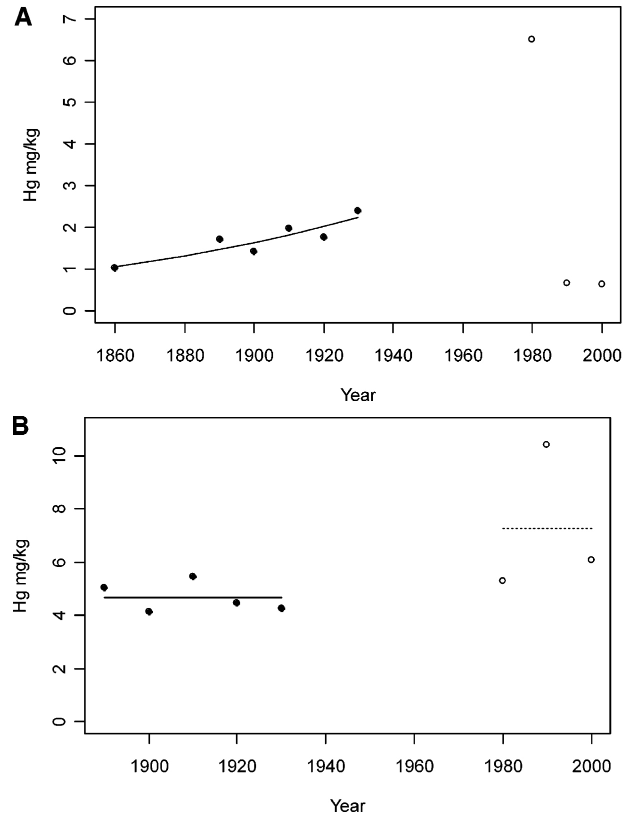

FIGURE 2. Mercury in primary feather no. 5 of peregrine falcon from West Greenland: (A) juvenile birds; lines are back-calculated linear regressions lines based on logarithmic medians of 10-year interval values (see Table 1); (B) adult birds where solid lines are averages of historic black points and recent averages (dotted lines) and open circles of 10-year interval median values.

with a two decadal gap to the consecutive value, is believed to have a strengthening effect on the significant positive trend found. The last two decades with low sample size included both the highest (1975–1984: 6.5 μg/g Hg dw, $N = 2$) and the lowest medians (1995–2004: 0.65 μg/g Hg dw, $N = 3$). The sample size of immature birds was too small ($N = 2$) for inclusion in the time trend analysis. Regarding adult peregrines, the few median points with $N > 1$ were too few to conduct linear regressions (Table 2; Figure 2B). A t-test on medians indicated a higher but not significant ($P < 0.076$) level in the 10-year period after 1980 compared to 1860–1930.

White-Tailed Eagles. All time trend analyses of white-tailed eagles were conducted as t-test comparisons of medians due to few data points and a large time gap during the period 1940–1980 among the medians. Hg concentrations in juvenile eagles showed a significant difference and twice as high Hg concentrations in the period 1990–2000 compared to the period 1880–1920. The immature with the lowest sample size ($N = 33$) showed no time trend, whereas adults covering the largest age span indicated an increase from the period 1880–1920 (mean = 5.9 mg/kg) to 1980–2000 (mean = 9.1 mg/kg), however the increase was not significant ($P = 0.11$) (Table 2; Figure 3A–C).

Discussion

Selection of Feathers. The use of feathers is a widely accepted method for monitoring Hg exposure in birds. The correlative relationship between dietary Hg intake and feather Hg concentration is known, as is the relationship between blood and feather, because both feather and blood Hg concentrations reflect Hg intake over the same period (*20–22*). To minimize variability (i.e., standardize seasonal and locality differences as shown by, for instance, Johnels et al. (*23*)) the

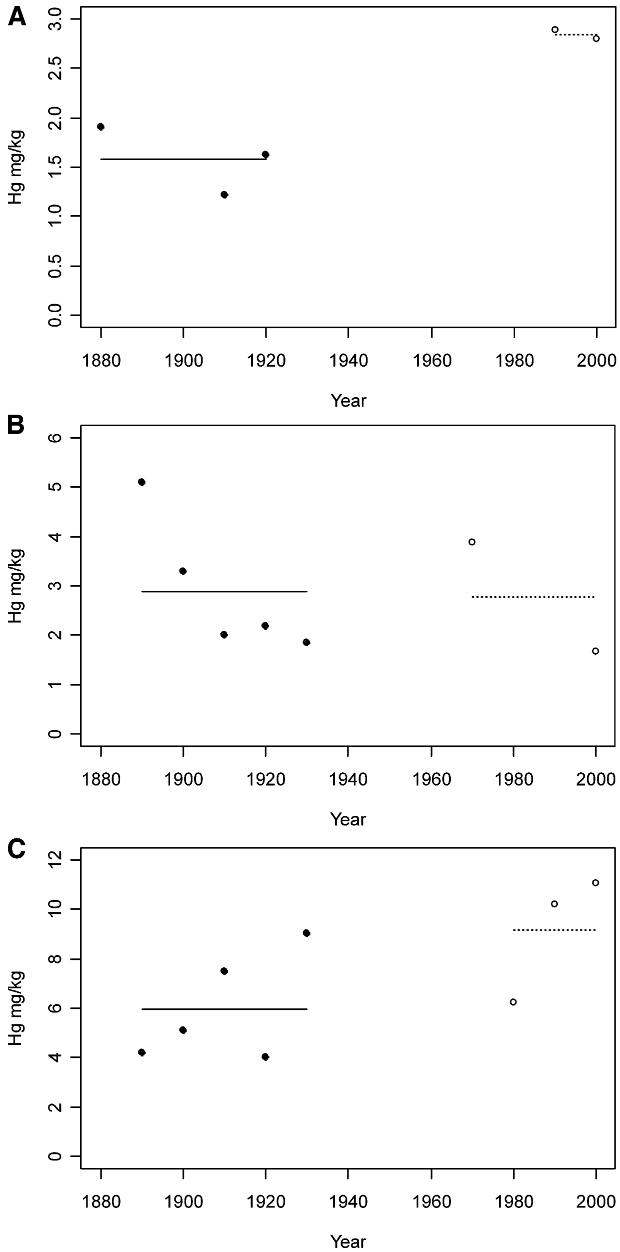

FIGURE 3. Mercury in primary feather no. 5 of white-tailed eagles from West Greenland: (A) juvenile; (B) immature; (C) adult. Lines are averages of historic (solid line and points) and recent (dotted lines and open circles) 10-year interval median values (see Table 1).

same feather was chosen from all birds. Sampling the fourth or fifth primary from the falcons ensured that the feathers were generated locally on West Greenland breeding grounds, thus reducing the influence of varying Hg exposure induced by different migration and wintering areas and habitats, as the peregrine winters in Latin America, and gyrfalcons may switch between terrestrial and marine habitats (*24*). In sparrow hawks (*Accipiter nisus*) and tawny owls (*Strix aluco*), no significant differences were found between the different parts of the feathers indicating the exact proportion of feather used for analysis may not be critical (*25*). In the present study, all samples were from the distal part of the primary and, therefore, minor differences along the length of individual primary feathers—and hence the proportion of the feather—may not affect the results.

Species Comparison. White-tailed eagles feed primarily on marine fishes (*26*), carrying the highest Hg concentrations, followed by the gyrfalcon feeding on medium size birds like ptarmigan (*Lagopus mutus*) and seabirds (*27*). Peregrines hunt in a wide range of habitats taking a wide range of birds, although in the breeding season in Greenland they mainly rely on small terrestrial passerines and an occasional ptarmigan (*28, 29*). The significantly higher Hg concentrations in juvenile white-tailed eagles compared to the juvenile gyrfalcons are linked likely to the higher proportion of marine food ingested by eagles. White-tailed eagles are 4−6 years old before they attain sexual maturity, whereas sexual maturity for the gyrfalcons and peregrines is attained after 1 year (*16*). The juvenile category of white-tailed eagles will therefore include older individuals than is the case for the falcons, resulting in higher Hg levels. The substantial variance among the adult falcons probably originates from differences in food selection and migration patterns, which may cause the lack of detectable differences. The higher averages in peregrines probably derive from Hg differences in prey at their different winter habitats. The higher concentrations—although non-significant—in adult white-tailed eagles compared to gyrfalcons and peregrines is probably linked to the longer life span and proportion of marine food items of this species.

Age and Sex Differences. All species showed significant age accumulations and this was taken into account when temporal trend analyses were conducted. Age accumulations have been documented in other studies performing this comparison (*30, 31*). Feathers are likely to reflect the food intake affecting the blood levels at the time of formation (*20−22*). However, a lifelong exposure will be mirrored in the blood concentration as this compartment is in equilibrium with the target organs (liver and kidney) which slowly excrete the accumulated mercury (*6*). The higher concentration of Hg found in the female gyrfalcons and peregrines (only significant for peregrines) compared with males are likely related to the larger size of the females, enabling them to take larger prey, including seabirds.

Temporal Trends. Seven out of eight time trend analyses showed an increase in primary feather mercury concentrations during 1850−2000. Four of these were significant while another two were close to being significant. Three of the four significant linear regressions represented periods within 1860−1960, which in turn, do not provide information from the recent decades (1960−2000). However, there were indications of a recent decline for juvenile peregrines and juvenile and adult gyrfalcons, but analyses were too few to include in any explanatory time trend models. However, the two sample comparisons of Hg 10-year medians for two of the white-tailed eagle age groups indicated a continued increase in recent decades. Further samples should be collected in the future to streghten the time trend comparisons and to solve whether Hg continue to increase in West Greenland birds of prey.

There are no studies covering the 20th century from the central Canadian−Greenlandic Arctic, but the few studies over decades from this time period show increases primarily in Hg in American (*Falco peregrinus anatum*) and Arctic (*F. p. tundrius*) peregrine falcon eggs from Alaska that were measured from 1988 to 1995 (*32*). This analysis showed that Hg might be increasing, at least in *F. p. anatum*. Both *F. p. tundrius* and Alaskan *F. p. anatum* are migratory and winter in the southern United States to Argentina. Therefore, contaminant trends reflected by peregrine falcons may not be exclusively indicative of the Arctic environment, but the closely similar Hg levels of adult peregrine and gyrfalcons do not suggest that this is a significant factor. Methyl-mercury in breeding plumage of black guillemot (*Cepphus grylle*) from Arctic Alaska showed an increase from an average of 581 ng/g ($N = 5$) in 1897 to 930 ng/g in ($n = 9$) 2000 (*33*). The increase over the last century was not statistically significant, but is similar to the approximately one percent increase per year seen in black guillemots from Greenland (*11*). No significant temporal trends were found for Hg concentrations from the mid-1980s to mid-1990s in livers of glaucous gulls (*Larus hyperboreus*) collected from Greenland (*9*). These time series were rather short and sample sizes were below the numbers usually needed to obtain a significant time trend. In a retrospective study, Braune et al. (*35*) analyzed eggs of thick-billed murres (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*), and black-legged kittiwakes (*Rissa tridactyla*) collected from Prince Leopold Island in Lancaster Sound, Nunavut, Canada, between 1975 and 1998. Total Hg concentrations almost doubled between 1975 and 1998 in eggs of thick-billed murres, and a 50% increase was observed in the northern fulmars. Stable isotope analyses ($\delta^{15}N$) indicated that the temporal trends observed were not the result of shifts in trophic level. Concentrations of total Hg in black-legged kittiwake eggs did not change significantly with time, probably because murres and fulmars overwinter in northern waters whereas kittiwakes overwinter at lower latitudes, where Hg concentrations have decreased due to recent reductions in Hg emissions and discharges from point sources (*34*). Concentrations in gray-headed albatrosses (*Diomedea chrysostoma*) were 113% higher in 1998 compared to 1989, which the authors interpreted as a consequence of increased mercury pollution of the southern ocean during the past decade (*35*).

Acknowledgments

This study was funded by the Danish Ministry of the Environment (Program: DANCEA "Danish Co-operation for Environment in the Arctic"). We thank Jan Bolding Kristensen from the Zoological Museum in Copenhagen for providing invaluable help and samples. We thank the Greenland Institute of Natural Resources for providing samples. Peter Aastrup is also acknowledged for providing moulted feathers. The laboratory technicians Sigga Joensen and Lene Bruun from NERI, Department of Arctic Environment, are acknowledged for their work on the heavy metal analysis.

Literature Cited

(1) Schroeder, W. H.; Anlauf, K. G.; Barrie, L. Y.; Lu, A.; Schneeberger, D. R.; Berg, T. Arctic springtime depletion of mercury. *Nature* **1998**, *394*, 331−332.
(2) Nilsson, A.; Huntington, H. *Arctic Pollution*; Arctic Monitoring and Assessment Programme, AMAP, P.O. Box 8100 Dep., N-0032: Oslo, Norway, 2002; 112 pp.
(3) Schroeder, W. H.; Steffen, A.; Scott, K.; Bender, T.; Prestbo, E.; Ebinghaus, R.; Lu, J. Y.; Lindberg, S. E. Summary report: first international Arctic atmospheric mercury research workshop. *Atmos. Environ.* **2003**, *37*, 2551−2555.
(4) Derome, J.; Fairbrother, A.; Marcy, S.; Wirtz, J.; Harding, K. Biological Effects. In *AMAP Assessment Report, Heavy Metals*; Marcy, et al.; Arctic Monitoring and Assessment Programme: Oslo, Norway, 2005; Chapter 6, pp 107−127.
(5) Dewailly, E.; Weihe, P. In *Arctic Monitoring and Assessment Programme*; AMAP: Oslo, Norway, 2003; Chapter 9, p 309.
(6) Dietz, R.; Pacyna, J.; Thomas, D. J.; Asmund, G.; Gordeev, V.; Johansen, P.; Kimstach, V.; Lockhart, L.; Pfirman, S. L.; Riget, F. F.; Shaw, G.; Wagemann, R.; White, M. In *AMAP Assessment Report: Arctic Pollution Issues*; AMAP: Oslo, Norway, 1998; Chapter 7, pp 373−524.
(7) Braune, B.; Outridge, P; Wilson, S.; Bignert, A.; Riget, F. *Contaminant residues in seabird eggs from the Canadian Arctic. Part I. Temporal trends 1975−1998*; AMAP: Oslo, Norway, 2005; pp 84−106.
(8) Bignert, A.; Riget, F.; Braune, B.; Outridged, P.; Wilsone, S. Recent temporal trend monitoring of mercury in Arctic biota − how powerful are the existing data sets? *J. Environ. Monit.* **2004**, *6*, 351−355.
(9) Riget, F.; Dietz, R. Temporal trends of cadmium and mercury in Greenland marine biota. *Sci. Total. Environ.* **2000**, *245*, 49−60.
(10) Riget, F.; Dietz, R.; Cleemann, M. Evaluation of the Greenland AMAP programme 1994-1995, by the use of power analysis (illustrated by selected heavy metals and POPs). *Sci. Total. Environ.* **2000**, *245*, 249−259.
(11) Appelquist, H. I.; Drabark, I.; Asbirk, S. Variation in mercury content of guillemot feathers over 150 years. *Mar. Pollut. Bull.* **1985**, *16*, 244−248.
(12) Eide, R.; Wesenberg, G. R.; Fosse, G. R. Mercury in primary teeth in preindustrial Norway. *Scand. J. Dent. Res.* **1993**, *101*, 1−4.
(13) Outridge, P. M.; Evans, R. D.; Wagemann, R.; Stewart, R. E. A. Historical trends of heavy metals and stable lead isotopes in beluga (*Delphinapterus leucas*) and walrus (*Odobenus rosmarus rosmarus*) in the Canadian Arctic. *Sci. Total Environ.* **1997**, *203*, 209−219.
(14) Tvinnereim, H. M.; Eide, R.; Fosse, G. R. Mercury in primary teeth from contemporary Norwegian children. Poster, International Conference on Human Health Effects of Mercury Exposure, Faroe Islands, June 1997.
(15) Cramp, S. In *Handbook of the birds of Europe, the Middle East and North Africa (Birds of the Western Palearctic)*; Oxford University Press: Oxford, 1979; Vols I−IX.
(16) Forsman, D. *The Raptors of Europe and the Middle East: A Handbook of Field Identification;* Poyser, London, UK, 1999; pp 77−554.
(17) Nielsen, C. O.; Dietz, R. Distributional pattern of Zinc, Cadmium, Mercury and Selenium in livers of Hooded seal (Cystophora cristata). *Biol. Trace Elem. Res.* **1990**, *24*, 61−71.
(18) Asmund, G.; Cleemann, M. Analytical methods, quality assurance and quality control used in the Greenland AMAP programme. *Sci. Total. Environ.* **2000**, *245*, 203−219.
(19) Asmund, G.; Vorkamp, K.; Backus, S.; Comba, M. An update of analytical methods, quality assurance and quality control used in the Greenland AMAP programme. *Sci. Total. Environ.* **2004**, *331*, 233−245.
(20) Kahle, S.; Becker, P. H. Bird blood as bioindicator for mercury in the environment. *Chemosphere* **1999**, *39*, 2451−2457.
(21) Bearhop, S.; Waldron, S.; Thompson, D.; Furness, R. Bioamplification of mercury in great skua Catharacta skua chicks: The influence of trophic status as determined by stable isotope signatures of blood and feathers. *Mar. Pollut. Bull.* **2000**, *40* (2), 181−185.
(22) Spalding, M. G.; Frederick, P. C.; McGill, H. C.; Bouton, S. N.; Richey, L. J.; Schumacher, I. M.; Blackmore, S. G. M.; Harrison, J. Histologic, neurologic, and immunologic effects of methylmercury in captive great egrets. *J. Wildl. Dis.* **2000**, *36*, 423−435.
(23) Johnels, A.; Tyler, G.; Westermark, T. A History of Mercury Levels in Swedish Fauna. *Ambio* **1979**, *8* (4), 160−168.
(24) Fuller, M. R.; Seegar, W. S.; Schueck, L. S. Routes and travel rates of migrating peregrine falcons *Falco peregrinus* and Swainson's hawks *Buteo swainsoni* in the western hemisphere. *J. Avian Biol.* **1998**, *29*, 433−440.
(25) Berg, W.; Johnels, A.; Sjöstrand, B.; Westermark, T. Mercury content in feathers of Swedish birds from the past 100 years. *Oikos* **1966**, *17*, 71−83.
(26) Wille, F.; Kampp, K. Food of the white-tailed eagle *Haliaeetus albicilla* in Greenland. *Holarct. Ecol.* **1983**, *6*, 81−88.
(27) Salomonsen, F. In *Grønlands Fauna*; Muus, B., Salomonsen, F., Vibe, C. Gyldendal: 1981.
(28) Falk, K.; Møller, S.; Burnham, W. A. Foraging behaviour of thick-billed murres breeding in different sectors of the North Water polynya. *Dansk. Orn. Foren. Tidsskr.* **1986**, *80*, 113−120.
(29) Burnham, W. A.; Mattox, W. G. Biology of the Peregrine and Gyrfalcon in Greenland. *Bioscience* **1984**, *14*, 1−25.
(30) Lindberg, P.; Odsjö, T. Mercury in feathers of peregrine falcon *Falco peregrinus* in Finland. *Environ. Pollut. B.* **1983**, *5* (4), 297−318.
(31) Lindberg, P.; Odsjö, T.; Wikman, M. Mercury levels in feathers of peregrine falcon *Falco peregrinus* compared with total mercury content in some of its prey species in Sweden. *Ornis. Fennica.* **1983**, *60*, 28−30.
(32) Ambrose, R. E.; Matz, A.; Swem, T.; Bente, P. *Environmental contaminants in American and Arctic peregrine falcon eggs in Alaska, 1979-95*; U.S. Technical Report NAES-TR-00-02; Fish and Wildlife Service, Ecological Services: Fairbanks, AK, 2000; p 67.
(33) Divoky, G. University of Alaska-Fairbanks, unpublished data; cited in Braune et al., 2005.
(34) Braune, B. M.; Donaldson, G. M.; Hobson, K. A. Contaminant residues in seabird eggs from the Canadian Arctic. Part I. Temporal trends 1975−1998. *Environ. Pollut.* **2002**, *114* (1), 39−54.
(35) Becker, P. H.; González-Solís, J.; Behrends, B.; Croxall, J. Feather mercury levels in seabirds at South Georgia: influence of trophic position, sex and age. *Mar. Ecol. Prog. Ser.* **2002**, *243*, 261−269.

Received for review April 25, 2006. Revised manuscript received July 27, 2006. Accepted July 28, 2006.

ES0609856