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Contaminant concentrations in bald eagles nesting on Lake Superior, the upper Mississippi River, and the St. Croix River

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ABSTRACT

We measured concentrations of DDE, total PCBs, and mercury in bald eagle (*Haliaeetus leucocephalus*) nestlings at three locations in the upper Midwest: Lake Superior, the upper Mississippi River, and the St. Croix River, 2006–2008. We also analyzed trends in concentrations of these contaminants for eagles on the southern shore of Lake Superior, from 1989 to 2008, using the current and previously published data. Concentrations of DDE in nestling blood plasma samples were greatest on Lake Superior (geometric mean: 16.2 μg/kg, $n = 29$), whereas concentrations of total PCBs were highest in Mississippi River samples (88.6 μg/kg, $n = 51$). Mercury concentrations were highest along the upper St. Croix River (6.81 μg/g wet weight in feathers, $n = 19$). For Lake Superior, DDE concentrations declined significantly in nestling blood plasma samples from 1989 to 2008, an average of 3.0% annually. Similarly, total PCBs in Lake Superior eaglets decreased 4.0% annually from 1989 to 2008, and mercury concentrations in nestling feathers from Lake Superior nests also decreased significantly from 1991 to 2008, 2.4% per year. With the possible exception of mercury on the upper St. Croix River, mean concentrations in 2006–2008 of all three compounds were below levels associated with significant impairment of reproduction for all sites, and reproductive rates at all three sites averaged >1.2 young per occupied territory, which is greater than the rate indicative of a healthy population.

Introduction

Bald eagles (*Haliaeetus leucocephalus)* are useful biosentinels for contaminants in waterbodies, due to their trophic position as top piscivores (Elliott and Norstrom, 1998; Elliott and Harris, 2001/2002; Cesh et al., 2008). In particular, lipophilic compounds such as organochlorines, can biomagnify through the food web and reach their greatest concentrations in predators such as bald eagles, common loons (*Gavia immer*), and gulls (*Larus* spp.). Monitoring contaminant concentrations and any associated demographic impairments in such species on a regular basis can provide critical information on trends and locations of new point sources, and serve as an early warning for emerging contaminants, such as polybrominated diphenyl ethers (PBDEs).

Bald eagles are especially valuable as biosentinels, because much is known about their population status, distribution, reproductive rates, and historical heavy metal and organochlorine contaminant burdens. Concentration thresholds of some of these compounds (e.g., PCBs, DDE) are associated with impairment in eagle populations (Wiemeyer et al., 1984, 1993; Elliott and Harris, 2001/2002), and the minimum productivity that permits population sustainability (Buehler et al., 1991; Bowman et al., 1995) is known. The abundance of such data is likely unmatched for a free-ranging wild top-predator species, and is invaluable in assessing and understanding contemporary conditions in bald eagle populations.

Some historical and long-term databases date to the early 1970s, after populations of bald eagles nesting in the continental United States declined due to reproductive failure caused mainly by DDE, a metabolite of the organochlorine insecticide DDT (Wiemeyer et al., 1972; Colborn, 1991). Following the bans on most uses of DDT and PCBs in the 1970s, the North American eagle population rebounded quickly throughout most of its range. However, bald eagle populations in a few regions, including the Great Lakes shorelines (Colborn, 1991; Best et al., 1994) did not increase as rapidly, primarily because of the high concentrations of these persistent compounds in the aquatic ecosystems.

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Even as other species recovered (Weseloh et al., 1994), bald eagles of Lake Superior continued to exhibit depressed reproductive rates and elevated organochlorine burdens throughout the 1980s and 1990s. In the 1980s, eagle reproduction at the Apostle Island National Lakeshore (AINL) in southern Lake Superior averaged 20% below the rate considered indicative of a healthy population (Buehler et al., 1991; Best et al., 1994; Bowman et al., 1995) and 40% below the rate of neighboring inland eagles. Two addled eagle eggs at AINL contained a mean of 13.5 μg/g DDE and 9.4 μg/g total PCBs, which led researchers to conclude that one or both of these organochlorines was depressing eagle reproduction at that time (Kozie and Anderson, 1991). By the early 1990s, concentrations in addled eggs from Lake Superior had declined to 2.5 μg/g DDE and 9.1 μg/g total PCBs, yet reproduction was still somewhat depressed; concurrent studies of prey delivery rates illustrated that low food availability on Lake Superior was the most likely cause of depressed reproduction, although a possible contribution by PCBs could not be excluded (Dykstra et al., 1998).

Large rivers in the upper Midwest have a similar legacy of PCB and DDT contamination. Organochlorine pesticides, including DDT, were used heavily in both agricultural and urban areas, reaching rivers through contaminated groundwater and runoff from precipitation (Gilliom et al., 2006). PCBs were used in a variety of industries that were built along rivers (common locations for some industries because rivers were useful for transportation of raw materials and for easy disposal of waste, and because they provided freshwater to assist with processing).

Following decreased use in the 1960s, total DDT (DDT, DDE and DDD) declined rapidly in stream-bed sediments and stream biota across the nation, including those in the upper Mississippi River basin (Schmitt, 2002; Gilliom et al., 2006). These declines slowed by the mid 1980s, however, and DDT and DDE continue to be the most frequently detected organochlorine compounds in fish tissue and bed sediments (Gilliom et al., 2006). In the late 1970s and early 1980s, PCBs and DDE were elevated in great blue heron (*Ardea herodias*) eggs and nestlings along the upper Mississippi River (Ohlendorf et al., 1979; Nosek and Faber, 1984). Ohlendorf et al. (1979) sampled young from four heron colonies along the upper Mississippi River and found mean wet-weight concentrations highest in a colony near south St. Paul, MN (6.43 ppm total PCBs and 1.31 ppm DDE) and lowest levels in young from a colony near Royalton, MN about 100 km upriver (0.22 ppm total PCBs and 0.37 ppm DDE). These same colonies and six others were sampled in 1993 and embryo liver concentrations averaged 3.04 ppm total PCBs and 1.31 ppm DDE (Custer et al., 1997). Between 1975 and 1978, 15 heron eggs were collected from a colony near Fountain City Bay, WI in the upper Mississippi River for organochlorine analysis (Nosek and Faber, 1984); total PCB concentrations ranged from 0.44 to 37.2 ppm wet weight (mean = 14.1 ppm) and average DDE concentrations were 1.41 ppm, with all samples <5.72 ppm. Geometric mean levels of DDE in white sucker (*Catostomus commersoni*) and common carp (*Cyprinus carpio*) tissue changed little between 1986 and 1995 at five sites on the upper Mississippi River (Schmitt, 2002). By contrast, PCB levels in water and stream-bed sediments declined from 1985 to 1995 (Anderson and Perry, 1999), although PCBs in walleye (*Sander vitreus*) and common carp were still among the highest in the upper Mississippi River (Lee and Anderson, 1998), where PCBs continue to be the cause of fish consumption advisories (Minnesota Department of Health, 2010). McNellis et al. (2000) found the highest levels of PCBs in fish tissues immediately downstream from urban areas such as the Twin Cities on the Mississippi River and Hudson on the St. Croix River, and concluded that PCB levels in fish varied by land use in the corridor, with forested sites being lowest.

In addition to organochlorines, mercury (Hg) is also a contaminant of concern in the Great Lakes region. Elevated concentrations of mercury have been detected in many species, including loons (Fevold et al., 2003), bald eagles (Scheuhammer et al., 2008), and many prey- and game-fish (Wiener et al., 2006). Much of that mercury can be traced to anthropogenic and atmospheric sources of inorganic mercury (Rada et al., 1989; Watras et al., 1994; Wiener and Sandheinrich, 2010). Inorganic mercury from coal combustion, waste incineration, and other sources is deposited on the landscape, where it is methylated by anaerobic bacteria found in wetlands and lakes, creating methylmercury, the most toxic, bioaccumulative form of mercury. In the St. Croix River basin, Christensen et al. (2006) found that streams draining forest/wetland watersheds had significantly higher fish-tissue mercury levels than streams draining agricultural or strictly forested watersheds. In the Mississippi River, mercury concentrations in walleye fillets have declined in the some reaches of the river, but not in others (Wiener and Sandheinrich, 2010). Mercury contamination has resulted in fish consumption advisories for many lakes and streams in the upper Mississippi and St. Croix River watersheds (Minnesota Department of Health, 2010; Wisconsin Department of Natural Resources, 2010).

In light of this history, and because the bald eagle serves as a biosentinel species, the Wisconsin Department of Natural Resources (WDNR) monitored the productivity and contaminant burdens of bald eagles on the Wisconsin south shore of Lake Superior from 1989 through 2001. In 2006 the National Park Service (NPS) and WDNR began a cooperative program to monitor bald eagle productivity and contaminant burdens on the Apostle Islands National Lakeshore, the St. Croix National Scenic Riverway, and the Mississippi National River and Recreation Area. Currently, the two agencies cooperatively monitor these areas as reference sites for a suite of environmental contaminants, including PCBs, DDT (and metabolites DDE and DDD), and Hg using bald eagles as sentinel species (Route et al., 2009).

These study areas span a gradient of human land-use practices and represent distinctly different freshwater ecosystems. As such they illustrate the different ways contaminants enter, bioaccumulate, and flush from aquatic systems. Lake Superior is deep, cold, relatively unproductive, has a large surface area to absorb contaminants from global sources, and a long water residence time. By contrast, the Mississippi and St. Croix Rivers start as small, clear, swift-flowing streams, with minimal human influences, but progressively become slower, more productive, sediment-laden rivers that drain large agricultural regions and flow through urban and suburban areas (Minneapolis–St. Paul, MN).

We here compare and contrast concentrations of DDE, total PCBs, and mercury in bald eagle nestlings at these three locations in the upper Midwest. We also report trends in concentrations of these contaminants for eagles on the southern shore of Lake Superior, from 1989 to 2008.

Methods

Study areas

Lake Superior nests were located within 8 km of the Lake Superior shore in Wisconsin or on the Apostle Islands in Lake Superior in Wisconsin (including, in part, the Apostle Islands National Lakeshore; Fig. 1). St. Croix River nests were located within the St. Croix National Scenic Riverway in Wisconsin and Minnesota, and Mississippi River nests were located within the Mississippi National River and Recreation Area in Wisconsin and Minnesota (Fig. 1).

The St. Croix National Scenic Riverway was subdivided into two study areas, upstream from the town of St. Croix Falls (upper St. Croix River and Namekagon River; Fig. 1), and downstream of St. Croix Falls (lower St. Croix River), based on ecological, hydrological, and land-use differences previously documented; for the upper St. Croix River, anthropogenic land use along the river ranges from 2 to 32%, whereas such land use is >60% along the lower St. Croix (Wan et al., 2007). In

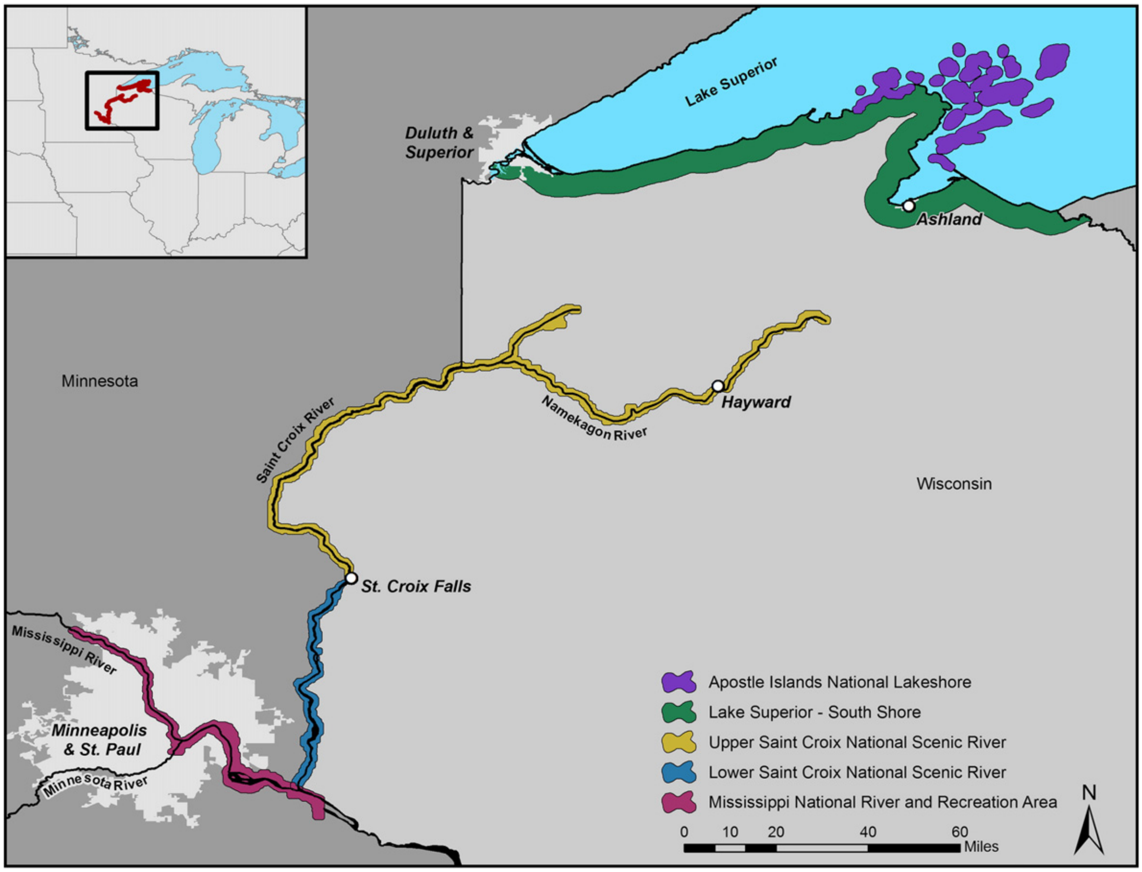

Fig. 1. Map of the study areas in northern Wisconsin and Minnesota. The Apostle Islands National Lakeshore (and associated islands) and the Lake Superior–South Shore together make up the Lake Superior study area.

addition, land along the lower river has a different groundwater source than the upper river (Wan et al., 2007).

For the trend analysis only, the Lake Superior nests were subdivided into those located on an island (Apostle Islands nests) and those along the shore of the mainland (Lake Superior shoreline); the Apostle Islands have a slightly different prey base (Warnke et al., 2002; Dykstra, 1995) and lower air temperatures than mainland shoreline nest sites, and eagles on the Apostle Islands have historically exhibited lower productivity and greater organochlorine burdens than those along the mainland shore (Dykstra, 1995).

Contaminant concentrations at Lake Superior, the Mississippi River, and the upper and the lower St. Croix River

Between 2006 and 2008, blood and breast feather samples were collected for contaminant analysis from 29 bald eagle nestlings at 21 nesting territories at Lake Superior, at 15 nesting territories ($n=19$ samples) located in the upper St. Croix National Scenic Riverway, at 10 nesting territories ($n=14$ samples) in the lower St. Croix National Scenic Riverway, and at 32 territories ($n=51$ samples) within the Mississippi National River and Recreation Area.

Between 1991 and 2002, additional feather samples were collected for analysis of mercury concentrations from 93 bald eagle nestlings at 27 territories located within 8 km of the Lake Superior shore in Wisconsin.

Nestlings were aged 5–10 weeks (based on length of the eighth primary [Bortolotti, 1984]) at the time of the sample collections. Two to four breast feathers were collected from each nestling and stored dry for mercury testing. For organochlorines, approximately 10 mL of blood was drawn from the brachial vein using sterile plastic or glass syringes previously washed with hexane and acetone. Blood was transferred to heparinized vacutainers, stored on wet ice until the end of the day, and separated by centrifuging in the evening, or stored in a refrigerator and then centrifuged <48 h after collection. Plasma was drawn off, transferred to another sterile vial, and frozen upright at approximately −20 °C. At the end of the field season, all feathers and frozen plasma samples were either hand-delivered or shipped on dry ice to the Wisconsin State Laboratory of Hygiene, Madison, Wisconsin.

Concentrations of p,p'-DDE, and of 75 PCB congeners (for list of congeners, see Appendix) in nestling plasma were determined by gas chromatography, with electron capture detection and were confirmed by mass spectrometry. Methods have already been described (Bowerman, 1993; Mora et al., 1993). Detection limits were 0.2 μg/kg for DDE, and ranged from 0.1 to 6 μg/kg for PCB congeners. The sum of the PCB congener concentrations was considered to be the measure of total PCBs; individual PCB congeners with concentrations below the detection limits were assigned a value of 0 for the purposes of summing congener concentrations.

Concentrations of total Hg in feather samples (wet weight) were determined by cold-vapor atomic absorption (CVAA) spectrophotometry at the Animal Health Diagnostic Laboratory at Michigan State University or at the Wisconsin State Laboratory of Hygiene using previously described methodology (Fevold et al., 2003).

Trends in contaminant concentrations along Lake Superior

Concentrations of mercury in bald eagle nestlings along the Lake Superior shore in Wisconsin were tested for temporal trends. Concentrations of DDE and total PCBs for bald eagles along the Wisconsin shore of Lake Superior were combined with previously published (1989–2001; Dykstra et al., 2005) and unpublished data (M. Meyer, unpublished data; $n = 9$ samples from 6 territories) for analysis of temporal trends. Earlier samples were collected and analyzed using methodology similar to the above (Dykstra et al., 2005).

Reproductive rate

Reproductive rate was assessed from fixed-wing aircraft (St. Croix River and Lake Superior nests) by the Wisconsin Department of Natural Resources, or from a combination of helicopter and boat (all Mississippi River nests) by the Ramsey County Parks and Recreation Department and the National Park Service, respectively. Nests were visited during incubation and again when nestlings were 4–7 weeks old. In the first survey, the eagle pairs that were occupying territories and/or incubating eggs were counted, and in the second survey, the resulting nestlings were counted. In cases where nests were visited by a climber after the second survey, the climber's data were used to calculate productivity. An occupied territory was defined as one where eggs had been laid, or two eagles were present on the territory, or the nest had been visibly repaired (even if no adults or only one non-incubating adult was observed; Postupalsky, 1974).

Occasionally nests were missed during the first survey and found during the second survey and vice versa. Data from these incomplete surveys were excluded from productivity calculations, as inclusion of nests found after hatching may bias calculated reproductive rates upward. For a regional summary, the total number of young produced was divided by the total number of breeding attempts during 2006–2008, for nests with complete surveys in each region. Productivity rates should be considered estimates in that not all nests were completely surveyed in all years.

Statistical analysis

Concentrations of all contaminants were log-transformed before analysis because the concentration distributions were skewed to the right and bounded on the left by zero. We examined plots of residuals against fitted values from models to assess assumptions of linearity and homogeneity of variance, and normal quantile–quantile plots to assess normality of residuals. We examined standard plots of residuals from all analyses to assess whether the assumptions of the statistical models were satisfied.

Multiple samples collected from eaglets at the same territory in different years cannot be assumed to be independent, because the eaglets likely have the same parents and because characteristics of the territory do not change much from year to year. Such consistent differences among sites could be caused by differences in forage base, adult health or experience (if the site is used repeatedly by the same adults), or other factors not specified by the model. Because the most important assumption of linear regression and analysis of variance models is that observations are independent (e.g., Scheffe, 1959; Zuur et al., 2009), it is essential to account for the potential lack of independence due to multiple observations from the same territories. Mixed effects models have become the standard method to analyze statistical relationships in data sets where observations or sampling units are grouped into larger units, such as contaminant measurements within territories (Wagner et al., 2006; McMahon and Diez, 2007; Zuur et al., 2009). These models include fixed effects that describe the general response of the population, as well as random effects that describe the characteristics of individual subjects or groups within the population. For all analyses, we used mixed effects models with territory identified as a random effect to account for correlations among observations within territories. Models for comparisons of contaminant concentrations among regions included region or region and year as fixed effects; we included year in some models to ensure that regional comparisons were not confounded by year effects and differing annual sample sizes. Because only one fixed effect (region) was included in most models, we report P-values from standard hypothesis tests (no model selection was necessary). The Tukey–Kramer method was used for pairwise comparisons among regions.

Regional means of log-transformed values were estimated directly from the mixed effects models; these estimates account for the random effects in the model and thus for differing numbers of observations for each territory (see Searle, 1987). These estimated regional means are very similar to the means computed by averaging first over all observations within a territory, and then averaging over the territory means in each region. Finally, the geometric means for each region were computed by back-transforming the means of the log-transformed observations.

To examine time trends in contaminant concentrations during 1989/1991–2008 for Lake Superior eaglets, we used mixed effects models with year (trend), study area (Apostle Islands or mainland Lake Superior shoreline), and their interaction as fixed effects, and territories as random effects. We used AIC (Akaike's Information Criterion) to select among models; the model with the minimum value of AIC provides the best balance between bias and precision (Burnham and Anderson, 2002). Linear trends in log concentration correspond to exponential trends in concentration and can be expressed as % change per year. Computing was carried out using SAS PROC MIXED (Littell et al., 1996).

Results

Contaminant concentrations at Lake Superior, the Mississippi River, and the upper and lower St. Croix River

DDE

There was some evidence ($P = 0.03$) for differences among years (2006–2008) for DDE concentrations, but these did not affect the regional comparisons. For simplicity and consistency with analyses of other parameters, we discuss models with regional effects but no year effect.

DDE concentrations differed significantly among regions ($F = 27.81$, $df = 3$, 35, $P < 0.001$). Highest concentrations were observed in nestlings from Lake Superior (geometric mean 16.20 μg/kg), and lowest concentrations were found at the upper St. Croix River (2.60 μg/kg; Table 1).

PCBs

Total PCB concentrations also differed significantly among regions ($F = 154.7$, $df = 3$, 35, $P < 0.001$). Highest concentrations were observed in nestlings from the Mississippi River (geometric mean 88.06 μg/kg), and lowest concentrations were found at the upper St. Croix River (2.66 μg/kg; Table 1).

Table 1
Concentrations of p,p'-DDE and total PCBs in plasma and of mercury in feathers of nestling bald eagles in northern Wisconsin, 2006–2008. Concentrations of DDE and total PCBs reported as μg/kg in plasma, wet weight; concentrations of mercury reported as μg/g in feathers, wet weight. Geometric means were estimated from analysis using mixed effects models, which accounted for multiple years of data from individual nests. For each contaminant, estimated geometric means followed by the same letter (a, b, or c) do not differ statistically. "Total PCBs" is the sum of concentrations of 75 PCB congeners.

Region	n	Estimated geometric mean DDE	Estimated geometric mean total PCBs	Estimated geometric mean mercury
Lake Superior	29	16.2 a	55.46 b	3.88 b c
Mississippi River	51	9.03 b	88.06 a	3.13 c
Lower St. Croix River	14	6.73 b	79.71 a b	4.54 b
Upper St. Croix River	19	2.60 c	2.66 c	6.81 a

Mercury

Mercury concentrations also differed significantly among regions ($F = 18.03$, $df = 3$, 34, $P < 0.001$), but the pattern differed. Highest concentrations were observed in nestlings from the upper St. Croix River (geometric mean 6.81 μg/g), and lowest concentrations were found at the Mississippi River (3.13 μg/g; Table 1).

Trends in contaminant concentrations along Lake Superior

DDE

Over the period 1989–2008, concentrations of DDE in eaglet plasma declined in both subregions of the Lake Superior study area (the Apostle Islands and the Lake Superior mainland shoreline) with concentrations slightly higher in Apostle Islands territories (Fig. 2.). The best fitting mixed effects model, as assessed using AIC, was the model including study area and trend, with no interaction between them (i.e., a difference in intercept but not in trend between the two study areas; Table 2). This model fit only slightly better than the model with a time trend only (ΔAIC = 1.0), so the evidence for a consistent difference between study areas was weaker than that for the trend over time (trend estimates were essentially identical from both models). The estimated rate of change in log concentration from this model (−0.0134) indicated that DDE concentrations have decreased at a rate of 3.0% per year over the period. DDE concentrations from Apostle Islands territories averaged 1.35 times higher than those from the Lake Superior shoreline. Geometric mean DDE concentrations for the two study areas as estimated from the model were 22.9 μg/kg for the Apostle Islands territories, and 16.9 μg/kg for Lake Superior shoreline territories.

Total PCBs

Total PCB concentrations in eaglet plasma declined during 1989–2008 in both the Apostle Islands and Lake Superior shoreline study areas (Fig. 3). As assessed using AIC, the best fitting mixed effects model included a time trend only, with no difference in either intercept or trend between the two areas. The common rate of change in log concentration of total PCBs was estimated as −0.0191 (Table 2), which corresponds to a change in total PCB concentration of 4.30% per year. The geometric mean concentration of total PCBs for the period for both study areas was 74.1 μg/kg.

Table 2
Results of mixed effects model analysis for DDE and total PCB concentrations in plasma of nestling bald eagles from the two subregions in the Lake Superior study area (the Apostle Islands and the Lake Superior shoreline), 1989–2008. Both subregions have the same slope for both DDE and total PCBs, so slope is reported only for the first subregion. For total PCBs, both subregions also have the same intercept, so intercept is also reported for the first subregion only. The *P*-value reported is for the null hypothesis that the slope differs from zero.

Contaminant	Subregion	Intercept (±SE)	Slope (±SE)	*P*	Annual change (%)
DDE	Apostle Islands	1.507 (0.079)	−0.0134 (0.0049)	0.01	−3.03
	Lake Superior shoreline	1.377 (0.068)			
Total PCBs	Apostle Islands	2.073 (0.067)	−0.0191 (0.0048)	< 0.001	−4.30
	Lake Superior shoreline				

Mercury

Concentrations of mercury in eaglet feathers during 1991–2008 were higher in Lake Superior shoreline territories than in Apostle Islands territories, but declined in both (Fig. 4). The best fitting model (lowest AIC) included study area and trend as fixed effects, with no interaction between them (a difference in intercept but not in trend). As estimated by the model, concentrations of mercury decreased at a rate of 2.44% per year (−0.0107 on the log scale), and were 1.73 times higher in mainland territories than in island territories (Table 3). Geometric mean mercury concentrations for the two study areas as estimated from the model were 3.37 μg/g for Apostle Islands territories and 5.84 μg/g for Lake Superior shoreline territories.

Reproductive rate

Reproductive rate estimates were highest at the Mississippi River and lowest at Lake Superior (Table 4). Statistical comparisons among regions were not conducted due to the incomplete nature of the survey data (not all nests were surveyed twice in all years).

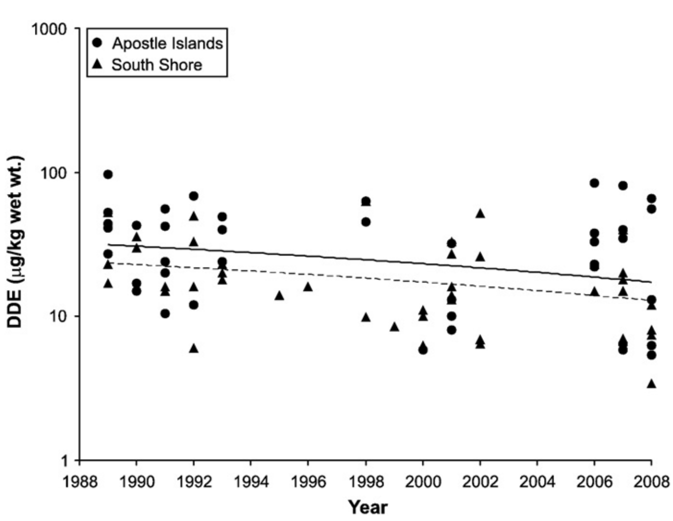

Fig. 2. DDE concentration (μg/kg wet wt) in plasma of nestling Bald Eagles from Lake Superior in Wisconsin, 1989–2008. Nests in the study area were divided into two groups: those on the Apostle Islands in Lake Superior (circles) and those along the mainland of Lake Superior (<8 km from the shoreline; triangles). Lines are the estimated trends from the mixed effects model; the upper line is the estimated trend for the Apostle Islands, lower line is the estimated trend for the mainland shoreline.

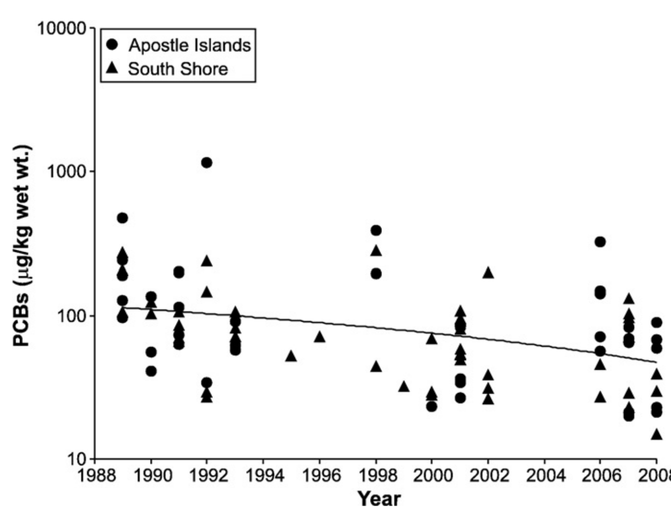

Fig. 3. Total PCB concentrations (μg/kg wet wt) in plasma of nestling bald eagles from Lake Superior in Wisconsin, 1989–2008. Nests in the study area were divided into two groups: those on the Apostle Islands in Lake Superior (circles) and those along the mainland of Lake Superior (<8 km from the shoreline; triangles). Line is the estimated trend from the mixed effects model for all territories.

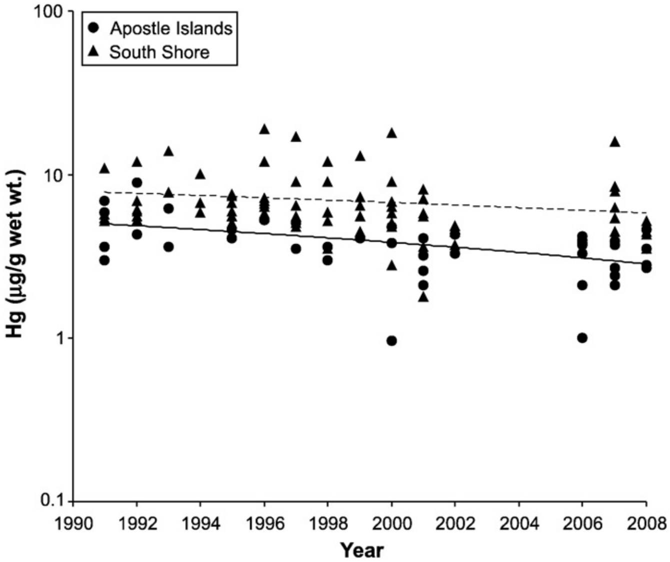

Fig. 4. Mercury concentrations (μg/g wet wt) in feathers of nestling bald eagles from Lake Superior in Wisconsin, 1991–2008. Nests in the study area were divided into two groups: those on the Apostle Islands in Lake Superior (circles) and those along the mainland of Lake Superior (<8 km from the shoreline; triangles). Lines are the estimated trends from the mixed effects model; the upper line is the estimated trend for the mainland shoreline, the lower line for the Apostle Islands.

Discussion

Contaminant concentrations

Organochlorines

Contaminant concentrations in eaglet blood generally reflect contaminant levels within the eagle territory (Olsson et al., 2000), as well as in addled eggs from the same area (Elliott and Harris, 2001/2002; Strause et al., 2007). Using the relationship between regional mean contaminant concentrations in eggs and plasma (Elliott and Norstrom, 1998; Elliott and Harris, 2001/2002), we estimated that the mean plasma concentrations of DDE, 2006–2008, in the regions we studied (16.2, 9.0, 6.7, and 2.6 μg/kg for Lake Superior, Mississippi River, lower St. Croix River, and upper St. Croix River, respectively, corresponded to egg concentrations of approximately 3.2, 2.1, 1.8, and 0.9 mg/kg). For total PCBs, mean plasma concentrations in the regions we studied (55.5, 88.1, 79.7, and 2.7 μg/kg for Lake Superior, Mississippi River, lower St. Croix River, and upper St. Croix River, respectively) corresponded to egg concentrations of approximately 7.5, 10.9, 10.0, and 0.7 mg/kg.

Elliott and Harris (2001/2002) estimated the thresholds for significant impairment of productivity (<0.7 young per occupied territory) to be 28 μg/kg DDE and 190 μg/kg total PCBs in plasma, corresponding to 6 mg/kg DDE and 20 mg/kg total PCBs in addled eggs. In an earlier review, contaminant concentrations >3.6 mg/kg DDE or >13 mg/kg total PCBs in addled eggs were associated with bald eagle reproductive impairment (<0.7 young per occupied territory; Wiemeyer et al., 1984, 1993).

Table 3
Results of mixed effects model analysis for mercury concentrations in feathers of nestling bald eagles from the two subregions in the Lake Superior study area (the Apostle Islands and the Lake Superior shoreline), 1991–2008. Note that slope did not differ between the two areas (P = 0.19), so the results for the common-slope model are shown.

Subregion	Intercept (±SE)	Slope (±SE)	P	Annual change (%)
Apostle Islands	0.632 (±0.059)	−0.0107 (±0.004)	0.02	−2.44
Lake Superior shoreline	0.871 (±0.058)			

Table 4
Reproductive rate of bald eagles nesting at Lake Superior, the Upper Mississippi River, and the St. Croix River, 2006–2008.

Region	n of breeding attempts	n of territories	Young per occupied nest (±SE)
Lake Superior	58	35	1.2 ± 0.1
Mississippi River	65	43	1.8 ± 0.1
Upper St. Croix River	39	24	1.5 ± 0.2
Lower St. Croix River	19	13	1.6 ± 0.2

In 2006–2008, DDE and total PCB concentrations in eaglet plasma at Lake Superior, the Mississippi River and the lower and upper St. Croix River, and the calculated addled-egg-equivalent concentrations, were below the threshold levels for reproductive impairment, regardless of which threshold values were used. This suggests that neither organochlorine was depressing overall reproductive rates at any of the sites in 2006–2008. Reproductive rates at the four sites confirmed this conclusion, averaging 1.2, 1.8, 1.5, and 1.6 young per occupied territory at Lake Superior, Mississippi River, upper St. Croix River, and lower St. Croix River, respectively. All mean rates were >1.0 young per occupied territory, the rate considered indicative of a healthy, expanding population (Buehler et al., 1991; Best et al., 1994; Bowman et al., 1995).

Although overall population reproductive rates were not impaired by DDE or PCBs, some individual nestlings' contaminant concentrations exceeded threshold levels, and thus reproduction at individual nests near contaminant "hotspots" may be affected by DDE or PCBs. Concentrations at individual nests are likely affected by local prey availability and/or prey selection by the adult eagles. Eagles preying on piscivorous birds or predator fish species consume greater amounts of contaminants because of biomagnification through the food chain; blood concentrations of DDE and total PCBs have been significantly associated with trophic level in nestling bald eagles (Elliott et al., 2009). In our study area, nests on the Apostle Islands with consistently high contaminant concentrations in nestlings included one near a rocky shoal with colonies of double-crested cormorants (*Phalacrocorax auritus*) and herring gulls (*Larus argentatus*) where adult eagles regularly foraged (C. Dykstra, unpublished data).

Differences in contaminant concentrations in the water, sediment, and biota among the four study areas may be attributed to differences in hydrology, ecology, and contaminant inputs. Lake Superior, deep, oligotrophic, and with a large watershed and water turnover time of 191 years (Minnesota Seagrant, 2008), experiences primarily atmospheric deposition of contaminants (Strachan and Eisenreich, 1988; Arimoto, 1989; Stevens and Neilson, 1989); contaminant concentrations in Lake Superior and its biota represent the slow recovery from the high levels of contamination of decades past, for the most part. The rivers, with their rapid flushing, integrate the contaminant inputs of a smaller water surface area; contaminant levels in their biota may reflect upriver point sources, including contaminated sediments, which can be transported by periodic flooding. Elevated concentrations of total PCBs in the Mississippi and some lower St. Croix River eaglets (compared to the Lake Superior eaglets) are likely due to industrial development. Analyses of emergent female mayflies (*Hexagenia bilineata*) sampled in 1988 on the upper Mississippi River showed that PCB levels were greatest in the vicinity of industrialized areas (Steingraeber and Wiener, 1995). Such contamination low on the aquatic food web will be magnified in high trophic-level species such as eagles.

Mercury

Although mercury compounds have been repeatedly measured in adult and nestling feathers, the threshold concentrations that signify impaired productivity remain unknown. Some researchers have suggested that populations with mercury >20 μg/g in adult feathers should be evaluated for possible toxic effects (Welch, 1994; Evers et al., 2004; DeSorbo and Evers, 2005), although Eisler (1987) proposed that 5 μg/g in feathers might be cause for concern. Concentrations in adult and nestling feathers may be correlated (Wood et al., 1996), but they are not always so (DeSorbo and Evers, 2005). Scheuhammer et al. (2008) found that bald eagles were able to demethylate mercury to a greater degree than common loons, yet they documented significant correlations between total mercury levels in the brain and concentrations of certain brain chemical receptors in both species.

The geometric mean concentrations of mercury in nestling feathers we measured (3.9, 3.1, 4.5, and 6.8 μg/g wet weight at Lake Superior, Mississippi River, lower St. Croix River, and upper St. Croix River, respectively), were mostly comparable to or lower than those measured in eagle nestlings in other regions. In central Florida, nestlings had feather concentrations of 3.23 μg/g wet weight (1991–1993; Wood et al., 1996), whereas in South Carolina, concentrations of 3.06 μg/g dry weight were measured in nestling feathers (1998–1999; Jagoe et al., 2002). In Maine, concentrations averaging 6.6 and 7.8 μg/g were measured in 1991 and 1992; mercury concentrations were unrelated to productivity rates, although any such relationship may have been masked by high concentrations of DDE (Welch, 1994). Within the Great Lakes Basin, Bowerman (1993) reported nestling feather concentrations of 8.8 μg/g in the interior lower peninsula of Michigan, 8.1 μg/g in the interior upper peninsula, 8.0 μg/g at Lakes Michigan and Huron, 3.7 μg/g at Lake Erie, 20.2 μg/g at Voyageurs National Park in northern Minnesota, and 8.7 μg/g at Lake Superior, in 1985–1989, with no relationship between mercury concentrations and productivity, although, again, the analysis may have been confounded by high concentrations of organochlorines.

Although the overall means for mercury at three of the upper Midwest locations were relatively low, the cluster of nests with higher concentrations of mercury along the upper St. Croix River in northern Wisconsin (geometric mean = 6.8 μg/g) may pose some reason for concern. The upper St. Croix, a small, relatively unproductive river, drains a forested, sparsely populated watershed with few industries. This watershed, however, contains a large proportion of wetlands, which likely contribute to the high bioavailability of methylmercury, given that low pH allows bacteria to methylate elemental mercury at an accelerated rate (Fagerstrom and Jernelov, 1972; Wiener, 1987; Cope et al., 1990; Brigham et al., 2009). Christensen et al. (2006) found significantly higher median fish-tissue mercury concentrations in watersheds of the St. Croix River that drained forest/wetland areas compared to those draining agricultural/forested areas. Rivers generally exhibit lower mercury concentrations than freshwater lakes, however, primarily because of the flushing ability within riverine systems (Evers et al., 2005). The relatively high levels in the upper St. Croix River suggest that lakes and other stillwater reservoirs in this watershed may have even greater mercury burdens and should be examined for possible effects on birds and other biota using those waterbodies.

Contaminant trends in Lake Superior eaglets

Concentrations of DDE, total PCBs, and mercury in Lake Superior eaglets all declined over the last two decades, which was in agreement with our previous report for total PCBs and DDE (Dykstra et al., 2005). Concentrations of DDE declined 3.0% per year from 1989–2008, a slower decline than we reported for DDE in eaglets from 1989–2001 (5.2% per year; Dykstra et al., 2005). Similarly, concentrations of total PCBs declined at 4.3% per year from 1989–2008, slightly more slowly than for 1989–2001 (6.3% per year; Dykstra et al., 2005). The lower rates may suggest that organochlorine levels in eaglets might be nearing a plateau, although continued monitoring is needed to test this hypothesis.

Concentrations of PCBs and DDE in other species in Lake Superior have declined significantly since the 1970s (DeVault et al., 1986; Weseloh et al., 1994); however, by the 1990s, organochlorine concentrations in some Great Lakes species had stopped decreasing, having reached plateau concentrations that were still somewhat elevated compared to animals in inland regions. For example, concentrations of total PCBs and TCDD congeners in herring gull eggs collected in many Great Lakes regions had stopped declining by the mid-1980s to 1990s (Hebert et al., 1994; Stow, 1995; Pekarik and Weseloh, 1998). In Lake Michigan fishes, PCBs had reached plateau concentrations by the late 1980s (Stow et al., 1995), and DDE concentrations in Lake Ontario lake trout had stabilized in the 1980s (Borgmann and Whittle, 1991).

Point-source discharges of mercury concentrations into the Great Lakes also decreased during the last four decades, which was reflected in the decline in concentrations in Great Lakes seabirds between 1972 and 1992 (egg concentrations; Koster et al., 1996). Similarly, in inland northern Wisconsin lakes, mercury concentrations declined 4.9% annually from 1992–2000 in loons (nestling blood samples; Fevold et al., 2003), approximately 0.5% per year in walleye (1982–2005; Rasmussen et al., 2007), and 5% annually in yellow perch (*Perca flavescens*; Hrabik and Watras, 2002), with a concomitant decrease in mercury deposition (Watras et al., 2000). In contrast, mercury concentrations in eaglets at inland lakes in Maine have not decreased since blood-sampling began in 1991 (DeSorbo and Evers, 2005), and egg concentrations have not declined since the 1970s (DeSorbo and Evers, 2005). In Minnesota lakes, mercury concentrations in piscivorous fish declined until 1992, and then increased at 0.8% per year (Monson, 2009), and in southern Wisconsin lakes, there was evidence for increasing mercury concentrations in walleye (Rasmussen et al., 2007). It is unclear whether differences in trends may be attributed to differences in water chemistry, deposition patterns, or some other factor; however, the increasing concentrations in biota in at least some regions, together with the relatively high levels measured in eaglets at some nests, suggest that researchers and managers should continue to monitor mercury concentrations in eagles, at least in areas of concern.

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Appendix A

Specific PCB congeners measured in eaglets: 003, 004/010, 006, 007/009, 008/005, 015/017, 016/032, 018, 019, 022, 024/027, 025, 026, 028/031, 033, 037/042, 040, 041/071/064, 044, 045, 046, 047/048, 049, 051, 052, 053, 056/060, 063, 066, 070/076, 074, 077/110, 082, 083, 085, 087, 089, 091, 092/084, 095, 097, 099, 101, 118, 123/149, 128, 132/153/105, 135/144, 136, 137/176, 141, 146, 151, 158, 163/138, 167, 170/190, 172, 174, 177, 178, 180, 183, 185, 187/182, 193, 194, 198, 199, 201, 202/171, 203/196, 206, 207, 208/195.

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