

## Special Feature: Methylmercury

### Original Contribution

# Marine Foraging Birds As Bioindicators of Mercury in the Gulf of Maine

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**Abstract:** From existing databases, we compiled and evaluated 604 total mercury (Hg) levels in the eggs and blood of 17 species of marine foraging birds from 35 Gulf of Maine islands to provide baseline data and to determine the best tissue, age class, and species for future biomonitoring. While mean Hg levels in most species did not exceed adverse effects thresholds, levels in some individual eggs did; for all species arithmetic mean egg Hg levels ranged from 0.04 to 0.62 ( $\mu\text{g/g}$ , wet weight). Piscivorous birds had higher Hg levels than invertivores. Leach's storm-petrel (*Oceanodroma leucorhoa*), razorbill (*Alca torda*), and black guillemot (*Cephus grylle*) adult blood and egg Hg levels were higher than other species. Our results indicate that adult blood is preferable to chick blood for detecting long-term temporal trends because adult levels are higher and not confounded by metabolic effects. However, since we found that eggs and adult blood are comparable indicators of methylmercury bioavailability, we determined that eggs are the preferred tissue for long-term Hg monitoring because the relative ease in collecting eggs ensures consistent and robust datasets. We suggest specific sampling methods, and based on our results demonstrate that common eider (*Somateria mollissima*), Leach's storm-petrel, double-crested cormorant, and black guillemot are the most effective bioindicators of Hg of the Gulf of Maine.

**Keywords:** mercury, seabirds, waterbirds, Gulf of Maine, bioindicators

## INTRODUCTION

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Although mercury (Hg) is a naturally occurring element (Nriagu and Pacyna 1988; Nriagu 1989), anthropogenic mercury levels in the North Atlantic have increased over the last 100 years (Slemr and Langer 1992; Asmund and Nielsen 2000; Mason and Sheu 2002) and in Maine have increased since 1970 (Perry et al. 2005). This increase is attributed generally to anthropogenic input (Lockhart et al. 1998; Mason and Sheu 2002). The historical increase has been reflected in tissues from seabirds in the North Atlantic (Appelquist et al. 1985; Thompson et al. 1992, 1998; Monteiro and Furness 1997), Canadian Arctic (Braune 2007), and within the Gulf of Maine watershed (Evers et al. 1998, 2005). This increase in global Hg levels since the 1900s is of concern because Hg is a persistent toxic heavy metal that bioaccumulates, is biomagnified in wildlife, and has negative neurological and reproductive impacts (Scheuhammer et al. 2007; Wolfe et al. 2007).

Studies on seabirds in many parts of the world have found Hg levels thought to be elevated above background levels, specifically in Antarctica (Norheim et al. 1982), North America (Pearce et al. 1979; Braune et al. 2001), Europe (Furness et al. 1995), Russia (Stout et al. 2002), Asia (Kim et al. 1996), and the North Pacific (Burger and Gochfeld 2000). Moreover, researchers have found these elevated Hg levels in species with diverse foraging strategies (Elliott et al. 1992; Thompson et al. 1992, Burger and Gochfeld 2000).

Birds are used frequently as bioindicators to evaluate where and to what extent Hg is bioavailable (Scheuhammer 1987; Wolfe et al. 1998, 2007; Evers et al. 2005; Scheuhammer et al. 2007). Past studies in eastern Canada found differences in Hg levels in the eggs of several seabird species (Pearce et al. 1979). While there has been a significant effort to characterize Hg levels in seabirds in North America, no studies have sampled marine foraging birds broadly and concurrently at multiple sites in the Gulf of Maine—a region that has been identified to have some of the highest Hg levels in North America (Evers and Clair 2005). Therefore, bird researchers in the Gulf of Maine formed the Gulf of Maine Seabird Contaminant Assessment Network (GOMSCAN) to share existing waterbird Hg data. GOMSCAN is led by BioDiversity Research Institute and is composed of the Canadian Wildlife Service, Kent Island Bowdoin Scientific Station, Maine Coastal Islands National Wildlife Refuge, Maine Department of Inland Fisheries and Wildlife, National Audubon Society, Shoals Marine Laboratory of

Cornell University, University of New Brunswick, University of New Hampshire, and U.S. Fish and Wildlife Service. GOMSCAN's goals are to identify species, locations, and trophic levels where Hg is concentrating, and to refine sampling methods for future contaminant studies.

This article presents findings of an initial collaborative screening effort and methods for future coordinated sampling. The main goals of this study were to determine the relationship and patterns of Hg levels in waterbirds within the Gulf of Maine, to evaluate blood and eggs as indicators of methylmercury (MeHg) bioavailability, and to identify species that are the most effective bioindicators of Hg availability in this marine system. We used the following criteria to evaluate if a species was suitable as a bioindicator: Are the birds abundant and widespread in Gulf of Maine, do they represent specific foraging guilds, and/or do they have the potential for Hg levels above estimated effects thresholds. We focused on a 6-year time period (2001–2006) and did not attempt to assess temporal trends in Hg levels.

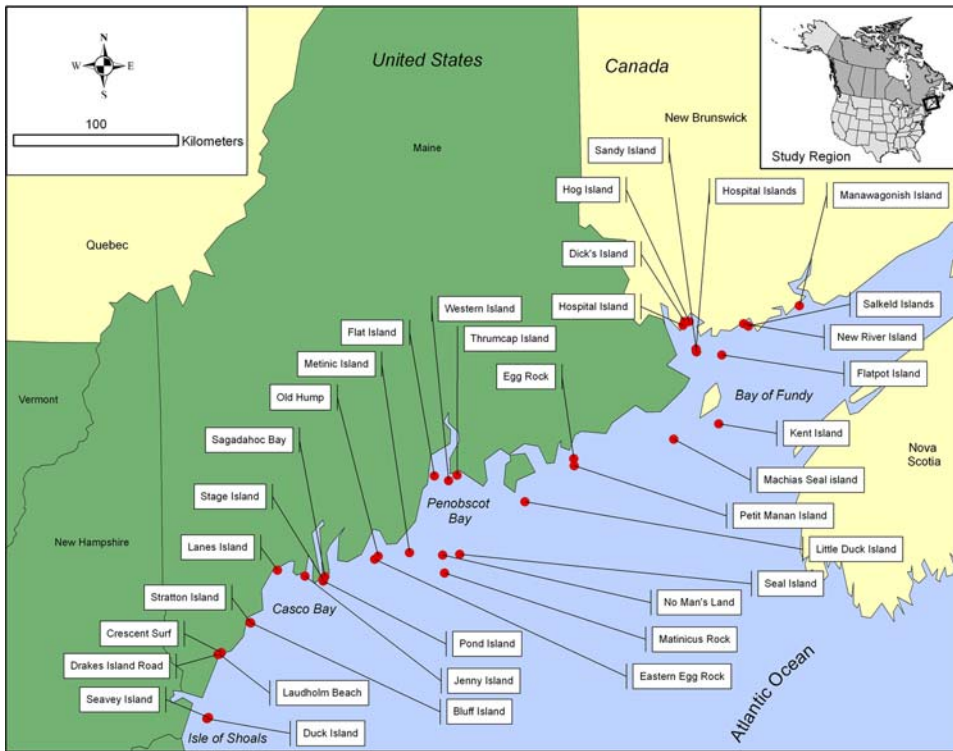
## METHODS

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From 2001 to 2006 (plus two sites in 1998), GOMSCAN members collected data on Hg levels in individual eggs, egg composites, and blood through multiple concurrent studies of 17 species of aquatic birds breeding on 35 sites in the Gulf of Maine (Figure 1, Appendix A). Viable and non-viable bird eggs were collected and placed in polyethylene bags (15% of the samples were analyzed as composites). During processing, we collected standard egg morphometrics (length, breadth, total egg weight, egg content weight, and volume), determined embryo development, placed the contents into labeled, chemically clean jars, and froze the samples (see detailed methods in Evers et al. 2003).

Juvenile (nest-bound chicks, young of year) and adult birds were captured at their breeding colonies and blood taken. Blood was collected by venipuncture of the coetaneous ulnar (wing) vein. Generally, less than 1.0 cc of blood was collected because most laboratories require only 0.25 cc of blood for Hg analysis. Blood was placed in labeled vials or tubes and frozen. All necessary state and federal permits were in place prior to field collections.

The data utilized in this compilation were generated at multiple laboratories over a period of several years. Differences in sample preparation and analytical methods were considered insignificant, although they were not



**Figure 1.** Sampling locations of the Gulf of Maine study area ( $n = 35$  islands).

quantified and split samples were not submitted to all labs to determine inter-lab differences. The quality assurance and quality control procedures—including standard reference material (SRM), blanks, duplicates, and spikes—that each laboratory used did meet federal standards set for the U.S. and Canada (e.g., recoveries of all SRMs were within established certified ranges). Mercury analysis was either by cold vapor atomic absorption spectroscopy (CVAAS) or direct mercury analyzer. Eggs were homogenized, lyophilized, and powdered. Only total Hg was analyzed because  $> 90\%$  of egg (Scheuhammer et al. 2001; Bond and Diamond 2008) and  $95\%$  of blood (Fournier et al. 2002; Rimmer et al. 2005) Hg levels are MeHg. Therefore, we used total Hg levels in these tissues to measure the availability of MeHg. Egg Hg was analyzed as dry weight and converted to wet weight using  $[(\text{dry weight} \times (100 - \% \text{ moisture})) / 100]$ . We standardized our Hg data to wet weight because it was the common measure in the multiple databases we used for this study. Each laboratory determined percent moisture during the freeze-drying process. Many eggs cracked during the freezing process in the field; therefore, we were able to measure egg volume on only a subset of the eggs. Using this subset, we calculated egg fresh mass (total egg mass/egg volume) and found no significant difference from measured egg mass, demonstrating insignificant loss of moisture (ANOVA,  $F_{1,134} = 0.14$ ,  $P = 0.71$ ). Hg in blood was measured as wet weight.

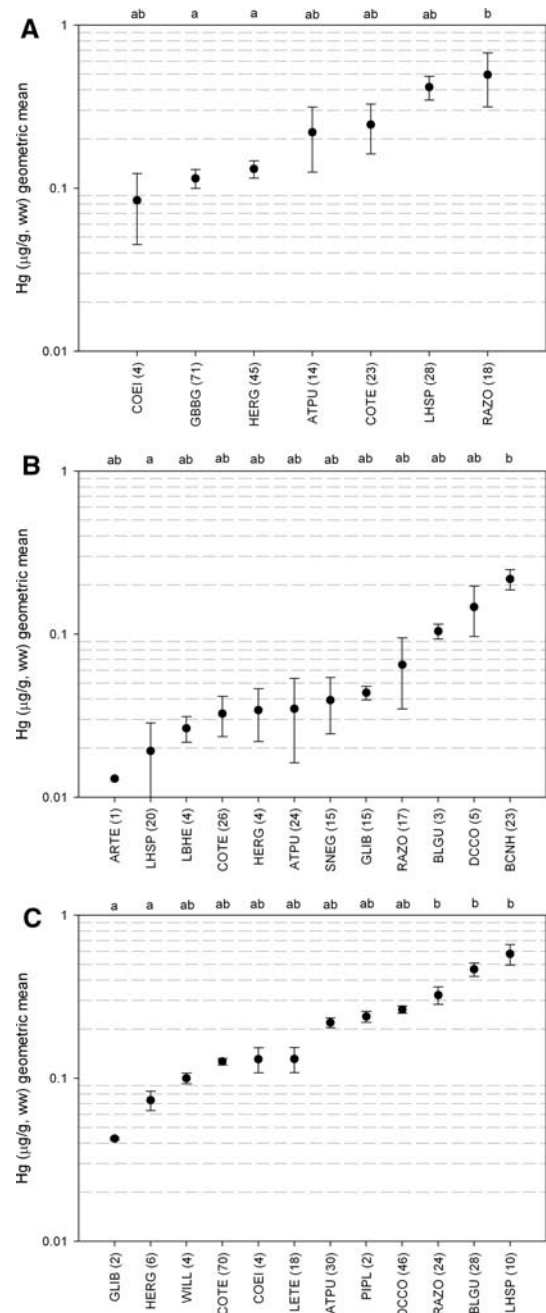
We performed statistics with JMP (SAS Institute Inc. 2001). Each egg composite (two or more eggs homogenized) was treated as a sample size of one. We  $\log_{10}$  transformed the data to improve normality and homoscedasticity. Breaking the Hg data down by bird species and age into 30 groups, the log-transformed Hg data were normally distributed for  $76\%$  of the groups as determined using one-sample Kolmogorov–Smirnov tests (SYSTAT 2007). We tested for differences among species in tissue Hg concentrations using analysis of variance (ANOVA), followed by Tukey–Kramer HSD paired comparisons. We pooled data from the same location and species across years (2001–2006). We were not able to test for interaction between island and species because all species were not present at each sampling site. For species sampled at more than four colonies, we tested for Hg differences among islands. We compared the variance of  $\log_{10}$ -transformed Hg data for individual eggs within double-crested cormorant (*Phalacrocorax auritus*) clutches and among composite samples of five eggs taken from different cormorant nests within several nesting colonies, to assess the relative importance of within-clutch and within-island variation in egg Hg concentrations. To determine the influence of foraging strategy and trophic status on tissue Hg levels among species, we grouped waterbirds into three foraging categories based upon documented diets (Appendix B).

## RESULTS

Geometric mean Hg levels in the blood of adult waterbirds differed among seven species measured (Figure 2A; ANOVA,  $F_{6,196} = 17.63$ ,  $P < 0.0001$ ). Blood Hg concentrations in adult razorbills were greater than those in great black-backed (*Larus marinus*) and herring gulls (*L. argentatus*) (Tukey HSD,  $P < 0.05$ ). Arithmetic mean Hg levels in tissues of each species are listed in Appendix B. Juvenile waterbirds also differed in their geometric mean Hg levels in blood (Figure 2B; ANOVA,  $F_{11,144} = 15.57$ ,  $P < 0.0001$ ). Blood Hg concentrations in juvenile black-crowned night-herons (*Nycticorax nycticorax*) were greater than those in Leach's storm-petrels (*Oceanodroma leucorhoa*) (Tukey HSD,  $P < 0.05$ ). In paired data sets, Hg levels in adult blood were significantly higher than juvenile blood (Table 1; all  $P < 0.0001$ ). The ratio of geometric mean blood Hg levels in adults versus juveniles of Leach's storm-petrel, herring gull, common tern (*Sterna hirundo*), razorbill (*Alca torda*), and Atlantic puffin (*Fratercula arctica*) ranged from 3.8 to 21.61, averaging overall 7.8:1 in the five species (Table 1).

Geometric mean egg Hg levels differed among the 12 species measured (Figure 2C; ANOVA,  $F_{11,232} = 37.08$ ,  $P < 0.0001$ ). Egg Hg concentrations were greater in Leach's storm-petrel, black guillemot (*Cepphus grylle*), and razorbill compared to herring gull and glossy ibis (*Plegadis falcinellus*) (Tukey HSD,  $P < 0.05$ ). Egg Hg levels were not different among islands for common terns (ANOVA,  $F_{5,64} = 0.909$ ,  $P = 0.48$ ) nor for composite samples of double-crested cormorant eggs (ANOVA,  $F_{6,39} = 2.09$ ,  $P = 0.07$ ). However, black guillemot egg Hg levels were different among islands (ANOVA  $F_{4,23} = 17.24$ ,  $P < 0.0001$ ). Geometric mean egg Hg levels were greater in guillemot eggs from Western Island than those from the other four islands sampled (Tukey HSD,  $P < 0.05$ ). There were low within-clutch (Table 2) and within-island differences in Hg levels of cormorant eggs (Table 3). In cormorant clutches where we analyzed each egg, clutches with higher mean Hg levels also had a greater Hg range and standard deviation, despite using  $\log_{10}$ -transformed Hg data. Based on limited data, this relationship was positive and significant for a polynomial curve fit (2nd degree polynomial,  $r^2 = 0.99$ ,  $df = 5$ ;  $P = 0.01$ ).

Geometric mean Hg concentrations in eggs and adult blood differed among waterbirds classified by foraging



**Figure 2.** Geometric mean Hg levels ( $\mu\text{g/g}$ , ww)  $\pm$  SE in adult blood (A), juvenile blood (B), and eggs (C). Sample sizes in parentheses. Means not sharing a common letter are significantly different ( $P < 0.05$ ). ARTE, Arctic tern; ATPU, Atlantic puffin; BCNH, black-crowned night-heron; BLGU, black guillemot; COEI, common eider; COTE, common tern; DCCO, double-crested cormorant; GLIB, glossy ibis; GBBG, great black-backed gull; HERG, herring gull; LBHE, little blue heron; LETE, least tern; LHSP, Leach's storm-petrel; PIPL, piping plover; RAZO, razorbill; SNEG, snowy egret; and WILL, willet.

**Table 1.** Ratio of Hg in Adult and Juvenile Blood

Species	Geometric mean ratio (adult:juvenile)	Statistics (AVOVA)
Leach's storm-petrel	21.6:1	<i>DF</i> 1, 46; <i>F</i> = 143.00; <i>P</i> < 0.0001
Herring gull	3.8:1	<i>DF</i> 1, 47; <i>F</i> = 23.96; <i>P</i> < 0.0001
Common tern	7.5:1	<i>DF</i> 1, 47; <i>F</i> = 81.68; <i>P</i> < 0.0001
Razorbill	7.6:1	<i>DF</i> 1, 33; <i>F</i> = 67.53; <i>P</i> < 0.0001
Atlantic puffin	6.3:1	<i>DF</i> 1, 36; <i>F</i> = 44.51; <i>P</i> < 0.0001
Geometric mean of ratios	7.8:1	

**Table 2.** Within-clutch Total Hg ( $\mu\text{g/g}$ , ww) Variation of Double-crested Cormorant Eggs

Island	<i>n</i>	Geometric mean	Min	Max	SE
Bluff Island	4	0.12	0.11	0.13	0.003
Egg Rock	3	0.24	0.23	0.25	0.006
No Man's Land	3	0.30	0.25	0.33	0.024
Sugarloaf Island	4	0.24	0.23	0.26	0.008
Thrumcap Island	3	0.32	0.28	0.40	0.037

**Table 3.** Within-island Total Hg ( $\mu\text{g/g}$ , ww) Variation of Double-crested Cormorant Eggs<sup>a</sup>

Island	<i>n</i>	Geometric mean	Min	Max	SE
Duck Island	3	0.42	0.40	0.45	0.016
Egg Rock	3	0.30	0.26	0.34	0.024
No Man's Land	2	0.23	0.14	0.38	0.118
Stratton Island	3	0.35	0.31	0.40	0.025
Sugarloaf Island	3	0.36	0.32	0.42	0.032
Thrumcap Island	3	0.26	0.21	0.36	0.049

<sup>a</sup>Samples size is the number of five egg composites.

strategy (see Appendix B) (ANOVA, eggs:  $F_{2,243} = 56.52$ ,  $P < 0.0001$ ; adult blood:  $F_{2,200} = 11.07$ ,  $P < 0.0001$ ). Piscivorous birds had greater geometric mean Hg levels in eggs (geometric mean  $\pm$  SE;  $0.30 \pm 0.02$   $\mu\text{g/g}$ , ww) than species that foraged on both invertebrates and fish ( $0.14 \pm 0.02$   $\mu\text{g/g}$ , ww) or on invertebrates alone ( $0.11 \pm 0.02$   $\mu\text{g/g}$ , ww; Tukey HSD,  $P < 0.05$ ). Similarly, adult piscivorous birds had greater geometric mean Hg levels in blood ( $0.35 \pm 0.08$   $\mu\text{g/g}$ , ww) than adult birds that foraged on both invertebrates and fish ( $0.16 \pm 0.2$   $\mu\text{g/g}$ , ww) or on invertebrates alone ( $0.08 \pm 0.03$   $\mu\text{g/g}$ , ww; Tukey HSD,  $P < 0.05$ ). We did not observe any differences in juvenile blood Hg levels among foraging groups (ANOVA,  $F_{2,153} = 0.18$ ,  $P = 0.84$ ).

## DISCUSSION

### Tissue Selection

#### Blood

Our results indicate that adult blood is more useful than chick blood for detecting long-term temporal trends, because Hg levels in adults are higher and are not confounded by metabolic effects. Blood Hg levels in chicks are lower when Hg is depurated into growing feathers (Spalding et al. 2000) and, consequently, may not represent full dietary exposure. Since the specific relationship between MeHg uptake and feather molt is relatively unknown and may

vary from species to species, interpreting Hg difference is challenging and is therefore confounded by chick age.

In all species with paired data, adult blood Hg levels were significantly higher than those of juveniles (Table 1); adult/juvenile blood Hg ratios by species are valuable in predicting adult Hg levels based on juvenile's or vice versa. This trend has also been observed in herring and Franklin's gulls (*L. pipixcan*; Burger and Gochfeld 1997), common mergansers (*Mergus merganser*), tree swallows (*Tachycineta bicolor*), belted kingfishers (*Megasceryle alcyon*), common loons (*Gavia immer*; Evers et al. 2005), double-crested cormorants, snowy egrets (*Egretta thula*), and black-crowned night-herons (Henny et al. 2002). This difference is attributed to chicks' deprecating Hg into their growing feathers thereby reducing blood levels (Spalding et al. 2000; Evers et al. 2005), and to the chicks consuming smaller prey than adults—smaller fish will have bioaccumulated less Hg (Evers et al. 2005).

Leach's storm-petrel had the highest ratio with adults having 21.6 times higher Hg than chicks; this ratio is greater than that observed for other piscivorous birds (Burger and Gochfeld 1997; Burgess et al. 2005; Evers et al. 2005). Trophic position likely does not explain the difference: Hedd and Montevecchi (2006) found no trophic difference between Leach's storm-petrel adults and juveniles; and Antarctic petrel (*Thalassoica antarctica*; Hodum and Hobson 2000) and southern giant petrel (*Macronectes giganteus*; Forero et al. 2005) chicks occupied a higher trophic position than adults. Bond (2007) found no significant relationships (all  $r^2 < 0.05$ ) between  $\delta^{15}\text{N}$  and total mercury in seabird feathers, blood, and yolk, and only a weak relationship in albumen ( $r^2 = 0.25$ ) from seabirds in the Gulf of Maine region. There are at least two possible explanations for this higher ratio in storm-petrels. First, storm-petrel chicks may have different pharmacokinetics of Hg as a result of their slower development (fledging  $\pm 65$  days) compared to other species. This may allow storm-petrel chicks to deprecate a higher proportion of their ingested Hg burden to their growing feathers, since their feather growth occurs over a 60–70-day period (Huntington et al. 1996). Second, the observed difference in juvenile and adult blood Hg levels in storm-petrels may result from possible differences in prey Hg levels between the breeding and non-breeding foraging areas. Lower levels of Hg in prey fed to chicks may result in lower blood Hg levels if adults feed on different prey with higher Hg levels prior to the breeding season and bioaccumulate higher levels of Hg in their soft tissues than chicks.

## Eggs

Our results indicate that egg Hg levels are as effective as adult blood for biomonitoring. Adult blood and egg Hg levels were within the same order of magnitude, and they both displayed species differences (Appendix A, Figure 2A, C). Although species with paired adult blood and egg data sets did not have identical Hg exposure order, piscivorous birds had significantly higher blood and egg Hg levels than birds feeding on both invertebrates and fish or on invertebrates alone. Therefore, eggs are as effective as blood in detecting difference in Hg exposure between trophic levels.

Eggs are a good tissue for long-term monitoring because they represent recent local dietary uptake (Hobson et al. 1997; Evers et al. 2003; Bond 2007). Egg nutrients are generally allocated from exogenous rather than endogenous sources (Hobson et al. 1997, 2000; Hobson 2006; Bond et al. 2007a) and most species represented in this study are documented income breeders, using exogenous nutrients for egg production (Hobson 2006; Bond 2007). In addition, eggs are relatively easy to collect, and clearly exhibit differences among species and colonies. However, power analysis and research design must take into account the varying levels of Hg within clutches as well as within colonies.

When interpreting Hg levels in eggs, it is important to consider within-clutch variation. In some studies, the first-laid egg exhibits the highest Hg levels, and the last-laid egg the lowest (Becker 1992; Evers et al. 2005). Cormorant egg results in this study show little within-clutch variation (Table 2), suggesting that one egg from a clutch can accurately characterize Hg levels of the laying female. We did find, however, that clutches with higher Hg levels also had higher Hg variation among eggs within the clutch. This indicates that analyzing each egg within a clutch will be necessary at sites where Hg levels are known to be high, such as near a known point source.

Another factor critical to understanding Hg levels is within-colony variation, which ultimately determines the number of samples needed to characterize a colony. We found little within-colony variation in Hg levels in cormorant eggs (Table 3), but, similar to the above findings, we found an increase in within-colony variation with an increase in Hg level. This indicates the importance of conducting a pilot study prior to large-scale sampling.

Several programs that monitor contaminant levels in seabird eggs use parallel data on stable carbon and nitrogen isotope ratios within the same eggs, as an aid in inter-

preparing differences in contaminant concentrations between eggs from different nests within the same colony, between different colonies, or between different years at the same colony (Jarman et al. 1996; Hebert et al. 2000; Braune et al. 2002). Using stable isotope data, it is possible to assess if differences in contaminant levels are related to differences in trophic level or source of prey, rather than general changes in contaminant levels in the marine environment.

### Species Comparisons

Mean tissue Hg levels in most waterbird species we measured were below suggested toxic thresholds of 0.6–1.3  $\mu\text{g/g}$  (ww) in eggs (Barr 1986; Thompson 1996; Evers et al. 2003), 3.0  $\mu\text{g/g}$  (ww) in adult blood (Evers et al. 2008), and levels reported for juvenile blood, which are age dependent (Kenow et al. 2007, 2008). Recently, findings on the relative sensitivity of various bird species to Hg exposure appear to vary significantly and are related to foraging guilds (Heinz et al. 2008). Therefore, while mean egg Hg levels for Leach's storm-petrel and black guillemot are generally under stated thresholds (Appendix B), evidence from Heinz et al. (2008) indicates reproductive impairment remains plausible and that toxic thresholds for these species need to be developed. Generally the adult and juvenile Hg blood levels were low and consistent with other studies (Kahle and Becker 1999; Thompson and Dowding 1999; Bearhop et al. 2000; Evers et al. 2005; Ikemoto et al. 2005). However, some adult puffins, common terns, storm-petrels, and razorbills had blood and egg Hg levels which exceeded the mean by more than three times (Appendix B). This suggests that certain individuals may have a specialized diet, which results in higher Hg levels in their blood and eggs.

Piscivores in this study had higher Hg levels than invertivores, and birds that forage on both invertebrates and fish. This relationship has been documented in other studies on marine birds (e.g., Burger 2002), waterfowl (Evers et al. 2005), wading birds (Sundlof et al. 1994), and fish (Peterson et al. 2002). This diet difference reflects trophic level differences (Hobson et al. 2002; Evers et al. 2005). Invertivores such as common eider (*Somateria mollissima*), glossy ibis, and willet (*Catoptrophorus semipalmatus*) had lower Hg levels than piscivores such as double-crested cormorant and razorbill. Piscivores also tended to have Hg levels higher than species that feed on both invertebrates and fish. For example, juvenile blood and egg Hg results show that common tern Hg levels are significantly lower than double-crested cormorant tissues.

Trophic position and foraging strategies (see Appendix B) may also explain why Hg levels in adult blood of Leach's storm-petrels and razorbills, and eggs of Leach's storm-petrels, guillemots, and razorbills tended to be higher than those of other species, explained further below.

#### *Leach's Storm-petrel*

Leach's storm-petrels consistently have some of the highest Hg levels in multi-species studies (Elliott et al. 1992; Elliott and Scheuhammer 1997; Burgess 2006; Bond 2007). The Hg levels of Leach's storm-petrel may be attributed to their mesopelagic foraging strategy. They feed 100–200 km offshore beyond the continental shelf (Huntington et al. 1996) on crustaceans and mesopelagic fish (Watanuki 1985; Hedd and Montevecchi 2006). Monteiro et al. (1996) found that mesopelagic fish have higher Hg levels than surface feeding fish; they suggested that Hg is more readily available to deep-sea fish because the methylation of inorganic Hg mostly occurs in deep water with low oxygen. Myctophids (lanternfish) are important prey for storm-petrels in British Columbia, Canada (Vermeer and Devito 1988); Pearl Island, Nova Scotia, Canada; and Middle Island, Newfoundland, Canada (Linton 1979), accounting for 55% diet mass on Green Island and Gull Island, Newfoundland (Montevecchi et al. 1992), and 77% of the mass of identified fish on Baccalieu Island, Newfoundland (Hedd and Montevecchi 2006). These mesopelagic fish become available to storm-petrels when the fish rise to the surface to feed at night (Hedd and Montevecchi 2006). Myctophids have elevated Hg levels when compared to other fish of a similar size (Monteiro et al. 1996; Lahaye et al. 2006), and have varying Hg levels in different regions in the North Atlantic (Martin et al. 2006). A fourfold increase in Hg levels in band-rumped storm-petrels (*Oceanodroma castro*) over the last 100 years further demonstrates high Hg availability in the mesopelagic zone (Thompson et al. 1998). Therefore, the Hg levels of storm-petrels represent primarily pelagic and mesopelagic zones, which are areas likely removed from point and regional Hg sources, and indicates that storm-petrels may serve as bioindicators of global Hg temporal trends.

#### *Black Guillemots*

In contrast to the storm-petrels, guillemots serve as bioindicators of Hg in inshore benthic food webs. Guillemots in the Gulf of Maine feed their chicks primarily rock

gunnels (*Pholis gunnellus*; Butler and Buckley 2002): 68% of the diet of chicks on Kent Island, New Brunswick, Canada (Preston 1968), and 59% on Great Duck Island, Maine, U.S.A. (Hayes 1993). Rock gunnel life history indicates they may bioaccumulate Hg to elevated levels: the fish are long-lived (up to 14 years), live close to sediment in the intertidal zone, and feed on benthic polychaetes, amphipods, mollusks, and crustaceans, which accumulate contaminants (Bigelow and Schroeder 2002; Vallis et al. 2007).

The guillemots' benthic foraging may also explain why their eggs exhibited significant inter-island Hg variation, while common terns and cormorants did not. This difference may reflect prey mobility and foraging range. Common terns feed within 20 km of breeding sites (Nisbet 2002), primarily on white hake (*Urophycis tenuis*) and Atlantic herring (*Clupea harengus*) during the breeding season (Hall et al. 2000). Both of these species are schooling fish that move throughout Gulf of Maine (Scott and Scott 1988). Similarly, cormorants feed within 40 km of breeding sites (Custer and Bunck 1992) and almost exclusively on larger fish (often schooling, up to 40 cm long; Hatch and Weseloh 1999) that are generally highly mobile. In contrast, guillemots feed usually within 4 km of their nesting sites (Butler and Buckley 2002), and their primary prey, rock gunnels, tend to have low mobility during the spring and summer (Vallis et al. 2007); this suggests that Hg levels in guillemots reflect a limited area around their breeding colonies, while Hg levels in terns and cormorants reflect a much broader geographic range. These results are consistent with research on polychlorinated biphenyls (PCBs) in a contaminated site in Labrador, where the high level of PCBs in guillemots were attributed to benthic foraging, small foraging range, and limited dispersal (Kuzyk et al. 2005).

Guillemot egg Hg levels were also among the highest recorded in this study, which corroborates findings from previous studies on Petit Manan Island, Maine (Mierzykowski et al. 2005) and the Faroe Islands (Dam et al. 2004). In our study, mean guillemot egg Hg levels on Western Island (0.76 µg/g, ww) were nearly three times that of the highest levels in cormorant eggs (0.28 µg/g, ww) on nearby Thrumcap Island (6 km away). Similarly, on Eastern Egg Rock and Petit Manan, where tern and guillemot eggs were sampled, guillemot egg Hg levels were 1.8 to 3.8 times higher than terns'.

#### *Other Alcids*

Puffins and razorbills are pursuit divers, feeding mainly on local (5–20 km) schooling fish (white hake and Atlantic

herring), and marine invertebrates, particularly *Meganyctiphanes norvegica* (Crustacea: Euphausiidae; Northern krill) in the case of puffins (Diamond and Devlin 2003; Bond et al. 2007b). Despite these similarities, razorbills have consistently higher Hg than puffins in feathers (Bond 2007), eggs, and blood (this study). In general, razorbills tend to dive deeper (Piatt and Nettleship 1985) and feed on larger, and therefore older, fish (Bond et al. 2007b), which bioaccumulate contaminants (Wiener and Spry 1996) and could increase the birds' Hg exposure.

#### *Juveniles of Other Species*

In our study, blood Hg levels in juvenile black-crowned night-herons, black guillemots, and double-crested cormorants tended to be higher than other species. The higher night-heron levels could be attributed to their feeding at higher trophic levels (we observed them feeding on tern chicks at the sampling site) or in an area with high Hg availability. Cormorant chicks likely also occupy a higher trophic position as they are commonly fed large fish (up to 40 cm; Hatch and Weseloh 1999) and black guillemot levels are likely high for the reasons described above.

#### **Bioindicators**

Seabirds are used as bioindicators of persistent bioaccumulative toxins (PBTs) around the world (Pearce et al. 1989; Elliott et al. 1992; Furness and Camphuysen 1997; Cifuentes et al. 2003; Braune 2007; Wolfe et al. 2007), and specifically for Hg (Thompson et al. 1990, 1992, 1998; Monteiro and Furness 1995). Since 1972, the Canadian Wildlife Service (CWS) has analyzed Atlantic puffin, double-crested cormorant, and Leach's storm-petrel eggs for Hg in Atlantic Canada (Pearce et al. 1979; Burgess 2006). Currently, there is no such long-term monitoring in the Gulf of Maine. We used the following criteria to select bioindicators: Are the birds abundant and widespread in the Gulf of Maine, do they represent specific foraging guilds, and/or do they have the potential for Hg levels above estimated effects thresholds?

In order to create an informative dataset to monitor long-term trends of Hg and other PBTs in the Gulf of Maine, we propose using an approach modeled after the CWS protocol. From each site, our model calls for the collection of eggs, at least every 4 years (higher frequency will increase the power to detect time trends (Hebert and Weseloh 2003)), from 15 separate nests of the common



eider, Leach's storm-petrel, double-crested cormorant, and black guillemot. The sample size of 15 would detect a 15% difference between sites at a 90% confidence interval (derived from a power analysis using the mean island standard deviation, 0.11, of the  $\log_{10}$ -transformed data; SAS Institute Inc. 2001). Since there is within-clutch Hg variation, consistently collecting eggs laid in the same sequence (i.e., the first laid egg) would reduce variation within each colony. These eggs should be collected from Isle of Shoals, Casco Bay, Penobscot Bay, and Bay of Fundy—not all sites have all species. If funding limitations prevent a thorough study, sampling could be limited to storm-petrel, cormorant, and guillemot.

Each of these species will serve as indicators of different food webs in the Gulf of Maine. Common eiders provide an inshore, site-specific Hg signal, because they feed primarily on mollusks, crustaceans, and echinoderms (Goudie et al. 2000). Leach's storm-petrels utilize offshore pelagic and mesopelagic food webs that may reflect global Hg levels. Double-crested cormorants are higher trophic-level piscivores that represent the pelagic food web for broad coastal areas because they feed on mobile schooling fish. The black guillemot represents benthic zone near breeding colonies. Guillemots are particularly important bioindicators because benthic feeding birds have higher levels of some contaminants than other species (Braune 1987; Kuzyk et al. 2005) and Hg can concentrate and is methylated in marine sediment (Gagnon and Fisher 1997).

Our results indicate that Hg levels in adult blood and eggs provide a comparable indicator of MeHg bioavailability and are within the same order of magnitude. While both tissues represent recent dietary uptake, sampling eggs is preferred because it is consistent with CWS protocol and researchers can collect suitable sample sizes from nesting colonies over time. The relative ease of collecting eggs is critical to the success of long-term monitoring.

By selecting a diverse suite of indicator species for monitoring Hg in the Gulf of Maine region, future studies will be able to measure changes in Hg in different food webs of the marine environment accurately and across multiple trophic-levels (Evers et al. 2009). This multi-trophic approach would also augment existing programs such as MusselWatch (Kimbrough et al. 2008), and GulfWatch (Chase et al. 2001), which tend to be focused on one component of the food web. Our study provides not only the necessary, broad baseline data to which future studies can be compared, but also the important process for selecting species and tissues, thereby allowing future studies to focus on key questions regarding changes in marine Hg in this important region.

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## APPENDIX

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(See Appendix Tables)

**Appendix A.** Species, Sites, and Sample Sizes for Hg Samples from the Gulf of Maine, 1998–2006

Common name	Scientific name/ Latin name	Site/Island	Nearest town	State/ Province <sup>a</sup>	Latitude	Longitude	Year	Blood		Egg	Total
								Adult	Juvenile		
Common eider	<i>Somateria mollissima</i>	Off Lanes Island,	Yarmouth	ME	43.80	-70.12	2000	2			2
		Casco Bay									
		Old Hump, Mus- congens Bay	St. George	ME	43.88	-69.36	2000			1	1
		Off Stage Island,	Georgetown	ME	43.76	-69.76	1998	2			2
		Sagadahoc Bay									
		Stratton Island,	Old Orchard Beach	ME	43.51	-70.31	2005			3	3
Leach's storm- petrel	<i>Oceanodroma leucorhoa</i>	Casco Bay									
		Machias Seal Island	Cutler	ME	44.50	-67.10	2005–2006	13	18	7	38
		Kent Island	Grand Manan	NB	44.58	-66.76	2004			3	3
Double-crested cormorant	<i>Phalacrocorax auritus</i>	Little Duck Island	Frenchboro	ME	44.17	-68.24	2005	15	2		17
		Bluff Island	Old Orchard Beach	ME	43.51	-70.32	2004–2005		3	4	7
		Duck Island, Isle of Shoals	Kittery	ME	42.98	-70.63	2005			3	3
		Egg Rock, Pigeon Hill Bay	Milbridge	ME	44.41	-67.87	2005			6	6
		No Man's Land	Matinicus Isle	ME	43.88	-68.87	2005			5	5
		Stratton Island	Old Orchard Beach	ME	43.51	-70.31	2005			3	3
		Sugarloaf Island	Phippsburg	ME	43.75	-69.77	2004–2005		2	7	9
		Thrumcap Island	Brooksville	ME	44.32	-68.76	2004–2005			15	15
		Manawagonish Island	Saint John	NB	45.21	-66.11	2004			3	3
Snowy egret	<i>Egretta thula</i>	Stratton Island	Old Orchard Beach	ME	43.51	-70.31	2001, 2003, 2004		15		15
Little blue heron	<i>Egretta caerulea</i>	Stratton Island	Old Orchard Beach	ME	43.51	-70.31	2003–2004		4		4
Black-crowned night-heron	<i>Nycticorax nyctico- rax</i>	Stratton Island	Old Orchard Beach	ME	43.51	-70.31	2001–2004		23		23
Glossy ibis	<i>Plegadis falcinellus</i>	Stratton Island	Old Orchard Beach	ME	43.51	-70.31	2001–2004		15	2	17
Piping plover	<i>Charadrius melodus</i>	Crescent Surf	Kennebunk	ME	43.34	-70.53	2002			1	1
		Laudholm Beach	Wells	ME	43.33	-70.54	2003			1	1



## Appendix A. continued

Common name	Scientific name/ Latin name	Site/Island	Nearest town	State/ Province <sup>a</sup>	Latitude	Longitude	Year	Blood		Egg	Total
								Adult	Juvenile		
Arctic tern	<i>Sterna paradisaea</i>	Stratton Island	Old Orchard Beach	ME	43.51	-70.31	2000		1		1
Razorbill	<i>Alca torda</i>	Machias Seal Island	Cutler	ME	44.50	-67.10	2005-2006	18	17	17	52
		Matinicus Rock	Matinicus Isle	ME	43.79	-68.85	2005			7	7
Black guillemot	<i>Cepphus grylle</i>	Eastern Egg Rock	St. George	ME	43.86	-69.38	2006			4	4
		Petit Manan Island	Milbridge	ME	44.37	-67.87	2006			8	8
		Western Island	Deer Isle	ME	44.29	-68.82	2005-2006			9	9
		Seal Island	Matinicus Isle	ME	43.89	-68.74	2006			1	1
		Metinic Island	St. George	ME	43.90	-69.12	2006			6	6
Atlantic puffin	<i>Fratercula arctica</i>	Little Duck Island	Frenchboro	ME	44.17	-68.24	2005		3		3
		Machias Seal island	Cutler	ME	44.50	-67.10	2004-2006	14	18	23	55
		Matinicus Rock	Matinicus Isle	ME	43.79	-68.85	2000, 2005			4	4
		Petit Manan Island	Milbridge	ME	44.37	-67.87	2005-2006		6	3	9
Total								203	157	244	604

<sup>a</sup>ME, Maine, U.S.A.; NH, New Hampshire, U.S.A.; NB, New Brunswick, Canada.

**Appendix B.** Tissue Hg Levels (Arithmetic Mean  $\pm$  SD, Range, and Sample Size) from Sampling of Seabird Tissues in Gulf of Maine 1998–2006<sup>a</sup>

Species	Blood ( $\mu\text{g/g}$ , ww)		Egg ( $\mu\text{g/g}$ , ww)	
	Mean $\pm$ SD (range)	<i>n</i> (t/c) <sup>a</sup>	Mean $\pm$ SD (range)	<i>n</i> (t/c) <sup>a</sup>
Common name	Primary foraging category	Foraging habitat/diet	Adult <sup>b</sup>	Juvenile <sup>c</sup>
Common eider	Invertivore	Nearshore benthic/invertebrates, intertidal mollusks (Goudie et al. 2000)	0.11 $\pm$ 0.08 (0.03–0.20) <b>4</b>	0.14 $\pm$ 0.05 (0.10–0.20) <b>4/3</b>
Leach's storm-petrel	Invertivore and piscivore	Mesopelagic, pelagic/plankton and small nekton (Huntington et al. 1996)	0.54 $\pm$ 0.37 (0.03–1.99) <b>28</b>	0.62 $\pm$ 0.26 (0.29–1.25) <b>10/3</b>
Double-crested cormorant	Piscivore	Mid-water, benthic/fish (Hatch and Weseloh 1999)	0.18 $\pm$ 0.12 (0.06–0.37) <b>5</b>	0.28 $\pm$ 0.09 (0.11–0.45) <b>46/20</b>
Snowy egret	Invertivore and piscivore	Salt-marsh, intertidal zone/invertebrates, fish (Parsons and Master 2000)	0.07 $\pm$ 0.06 (0.02–0.20) <b>15</b>	
Little blue heron	Invertivore and piscivore	Estuary/invertebrates, fish (Rodgers and Smith 1995)	0.03 $\pm$ 0.01(0.02–0.04) <b>4</b>	
Black-crowned night-heron	Invertivore and piscivore	Water edge, marsh/invertebrates, fish, birds, small mammals, garbage (Davis 1993)	0.25 $\pm$ 0.15 (0.11–0.72) <b>23</b>	
Glossy ibis	Invertivore	Shallow water/invertebrates (Davis and Kricher 2000)	0.04 $\pm$ 0.02 (0.00–0.07) <b>15</b>	0.04 $\pm$ 0.00 (0.04–0.04) <b>2/0</b>
Piping plover	Invertivore	Shoreline/invertebrates (Haig 2004)		0.24 $\pm$ 0.03 (0.22–0.26) <b>2/0</b>
Willet	Invertivore	Mud flats, salt-marsh edge/invertebrates small fish (Lother et al. 2001)		0.10 $\pm$ 0.02 (0.09–0.12) <b>4/0</b>
Herring gull	Invertivore and piscivore	Ocean surface, intertidal/berries, invertebrates, lobster bait, fish, small mammals, garbage (Goodale 2000)	0.16 $\pm$ 0.11 (0.03–0.53) <b>45</b>	0.08 $\pm$ 0.02 (0.05–0.10) <b>6/6</b>
Great black-backed gull	Invertivore and piscivore	Ocean surface, mudflats, intertidal zone/invertebrates, fish, birds, small mammals, garbage (Good 1998)	0.16 $\pm$ 0.13 (0.02–0.73) <b>71</b>	
Least tern	Invertivore and piscivore	Shallow water, estuaries, bays/invertebrates, small fish (Thompson et al. 1997)		0.15 $\pm$ 0.10 (0.08–0.49) <b>18/0</b>
Common tern	Invertivore and piscivore	Open water/invertebrates, small fish (Nisbet 2002)	0.36 $\pm$ 0.40 (0.04–1.81) <b>23</b>	0.13 $\pm$ 0.05 (0.07–0.25) <b>70/0</b>
Arctic tern	Invertivore and piscivore	Open water/invertebrates, small fish (Hatch 2002)	0.01 (0.01–0.01) <b>1</b>	
Razorbill	Piscivore	Shallow water, nearshore/crustaceans, fish (Hipfner and Chapdelaine 2002)	0.60 $\pm$ 0.45 (0.20–2.04) <b>18</b>	0.38 $\pm$ 0.19 (0.10–0.82) <b>24/0</b>

Species	Primary foraging category	Foraging habitat/diet	Blood ( $\mu\text{g/g}$ , ww)		Egg ( $\mu\text{g/g}$ , ww)
			Adult <sup>b</sup>	Juvenile <sup>c</sup>	
Common name			Mean $\pm$ SD (range)	n	Mean $\pm$ SD (range) n (t/c)
Black guillemot	Piscivore	Shallow inshore waters, benthic/invertebrates, fish (Butler and Buckley 2002)	0.11 $\pm$ 0.02	(0.09–0.13) 3	0.52 $\pm$ 0.23 (0.16–1.01) 28/0
Atlantic puffin	Piscivore	Shallow water near breeding colonies/fish (Lowther et al. 2002)	0.29 $\pm$ 0.35	(0.01–0.43) 24	0.23 $\pm$ 0.08 (0.08–0.45) 30/3
				(0.12–1.51) 14	

<sup>a</sup>Sample size for eggs includes the total number of samples and the number that were composites (t/c).

<sup>b</sup>Individuals at least 1 year old.

<sup>c</sup>Young of the year.

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