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Changes in mercury exposure of marine birds breeding in the Gulf of Maine, 2008–2013

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ABSTRACT

Mercury is a potent contaminant that can disrupt an organism's behavior and physiology, ultimately affecting reproductive success. Over the last 100 years, environmental deposition of anthropogenic sourced mercury has increased globally, particularly in the U.S. Northeast region. Marine birds are considered effective bioindicators of ecosystem health, including persistent marine contaminants. Goodale et al. (2008) found that mercury exposure exceeded adverse effects levels in some marine bird species breeding across the Gulf of Maine. We reexamined mercury contamination in four species identified as effective bioindicators. Compared with the previous sampling effort, inshore-feeding species showed significant increases in mercury exposure, while one pelagic-feeding species remained stable. This suggests that a major shift may have occurred in methylmercury availability in inshore waters of the Gulf of Maine. Understanding environmental mercury trends in the Gulf of Maine, and its significance to marine birds and other taxa will require a dedicated, standardized, long-term monitoring scheme.

1. Introduction

Mercury is a potent environmental contaminant and an issue of great concern globally (UNEP, 2013). In the form of methylmercury, it accumulates in wildlife species and has serious neurological impacts that can disrupt an organism's behavior and physiology (Hawley et al., 2009; Moore et al., 2014; Kobiela et al., 2015), ultimately affecting reproductive success (Evers et al., 2008a; Jackson et al., 2011; Provencher et al., 2016). Globally, anthropogenic-sourced mercury increased enormously over the industrial period of the last 100 years or so (Schuster et al., 2002). Although there has been a downturn in atmospheric emissions in the last 20 years (Zhang et al., 2016), mercury deposition is still particularly pervasive in certain regions, such as the Arctic (Kirk et al., 2012), and the northeastern United States (Evers and Clair, 2005; Evers et al., 2007).

Oceans are particularly at risk to mercury contamination, due to the methylating actions of sulfate-reducing bacteria that thrive in marine surface waters (Fitzgerald et al., 2007), and the concentration of mercury in surface marine waters may have increased by two to three times (UNEP, 2013; Lamborg et al., 2014) over the last 100 years due to anthropogenic emissions. Once available in the ecosystem, methylmercury increases at each trophic level (known as biomagnification; Lavoie et al., 2013) and can become concentrated within individuals over time (known as bioaccumulation). Marine ecosystems are particularly sensitive to the effects of biomagnification and bioaccumulation as they are often highly structured systems with multiple trophic levels that increase methylmercury levels, particularly for the many long-lived, top predators (UNEP, 2002).

Because they are widespread, visible, relatively easily-accessible, well-studied, and represent a range of trophic levels, marine birds are considered useful and effective bioindicators of marine ecosystem health worldwide (Furness and Camphuysen, 1997; Burger and Gochfeld, 2004), including persistent marine contaminants such as mercury (Monteiro and Furness, 1995; Mallory and Braune, 2012; Provencher et al., 2014). The Gulf of Maine Seabird Contaminant Assessment Network (GOMSCAN) examined mercury contamination in a broad suite of seabird species ($n = 17$) breeding at islands across the Gulf of Maine ($n = 35$), and presented baseline data for the region for 2001–2006 (Goodale et al., 2008). The GOMSCAN data indicated that mercury exposure clearly exceeded adverse effects levels in individuals of some seabird species. Goodale et al. (2008) also made recommendations on future sampling methods, including (1) identification of effective bioindicator species to sample across multiple food webs, (2) determination of suitable sample sizes to detect change, and (3) an appropriate monitoring timeline to assess long-term temporal trends.

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Based on these recommendations, we examined the trend in mercury contamination at a range of trophic levels through the collection and analysis of marine bird egg and blood tissues from across the Gulf of Maine. Two species provide information on the inshore, benthic community – the Common Eider (*Somateria mollissima*), which forages coastally on largely sessile organisms, such as mollusks (Goudie et al., 2000), and the Black Guillemot (*Cepphus grylle*), which forages on small demersal fishes, such as rock eels (Butler and Buckley, 2002). Two species reflect the trophic extremes of the offshore marine community – the Leach's Storm-Petrel (*Oceanodroma leucorhoa*), which forages far offshore on surface plankton (Huntington et al., 1996), and the Double-crested Cormorant (*Phalacrocorax auritus*), which forages more inshore on large mid-water pelagic fishes that they catch at depth (Hatch and Weseloh, 1999). Specifically, we used data from this study (2013), and data for the same four focal species from the previous study (Goodale et al., 2008) to establish the first indication of the general trends in their mercury exposure, and examine what that may mean for the broader marine community in the Gulf of Maine.

2. Study area and methods

2.1. Study area

The Gulf of Maine is an international water body, terrestrially bounded and shared by three US states (Maine, New Hampshire, Massachusetts) and two Canadian provinces (New Brunswick, Nova Scotia). It is one of the most productive marine ecosystems in the world, with a rich blend of ecological, economic, recreational, and environmental resources (Sherman and Skjoldal, 2002). Marine birds nest on hundreds of islands in the Gulf of Maine, only a handful of which are managed specifically for these species (MDIFW, 1993).

2.2. Sample collection

We collected egg or blood samples from four focal avian species from 8 island colonies spaced geographically across the Gulf of Maine (Fig. 1). In May–June of 2013, we collected 122 samples. Eggs were collected from nests of the Common Eider ($n = 32$) and Double-crested Cormorant ($n = 36$). Each egg was placed in a sealable polyethylene bag, and labeled on site. Blood samples were collected from adult Leach's Storm-Petrels ($n = 31$) and Black Guillemots ($n = 23$). Leach's Storm-Petrels were captured in a mist net as they visited their colony at night, while Black Guillemots were 'grubbed' from their nests in rock crevices. Blood sampling involved the puncture of the brachial vein of birds with a fine needle and collection of blood in 1–3 capillary tubes ($< 1.0\ cm^3$). The ends of each capillary tube were sealed with critocaps, and tubes were placed in labeled vacutainers for storage and transportation. Egg and blood samples were initially kept chilled, then frozen as soon after collection as possible, usually within 24–48 h, and stored at the Biodiversity Research Institute (BRI).

2.3. Mercury determination

All samples were later analyzed for total mercury concentrations at BRIs Wildlife Toxicology Laboratory, using a Direct Mercury Analyzer (DMA-80, Milestone Inc., Shelton, CT), and following EPA method 7473 (United States Environmental Protection Agency, 2007). During processing, we collected egg morphometrics (length, breadth, total egg weight, egg content weight, and volume), determined embryo development stage, and placed the contents in clean, labeled jars, before freeze-drying them. Eggs were homogenized prior to analysis, and egg mercury was measured as dry weight and converted to wet weight through percent moisture that we measured, using:

$$wet\ weight = (dry\ weight \times (100 - \%moisture))/100$$

2.4. Statistical analysis

To quantify the differences in mercury exposure among the four study species over space and time we constructed a general linear mixed model. This model used mercury levels as a response variable (log-transformed) then tissue type, species, study year, and site. Our mercury data were right-skewed and the log-transformation was successful in normalizing the distributions and made the liner mixed model framework appropriate. For 'tissue type', we only used samples of adult blood or eggs. Juvenile blood was collected for 28 individuals in the previous study, but, given the low mercury levels of the tissue and the inconsistent sampling across species, sites, and time periods, we removed these data from the analysis. 'Species' was a categorical variable consisting of the four study species. 'Study year' was a categorical variable assigning data from Goodale et al. (2008) as '2008' and data from the current study as '2013'. And 'site' was a random variable consisting of the 22 sites which represents undocumented differences between each of the breeding colonies, like spatial autocorrelation or unmeasured environmental covariates. Lastly, we included an interaction between species and study year to determine the trend in mercury levels over time by species. Tukey comparisons of least squared means were used to determine if there are any differences in groups within a categorical variable in a post-hoc test. Model fit was evaluated using an ANOVA comparison with the null model, r^2, and a visual assessment of the residuals. All statistical analyses were performed using JMP v.9.03 (SAS Institute Inc., Cary, NC).

3. Results

The fit of the general linear mixed model was good with an overall r^2 of 0.63 and visual checking of the residuals indicated a random spread that was uncorrelated with the fitted response. The variance associated with the random variable of colony site made up 31% of the total variation of the model, suggesting that there are many factors related to site that affect mercury levels that we did not explicitly test within the linear model, like spatial variance in mercury exposure.

In our general linear mixed model, we tested for the effect of tissue type, species, and year as fixed effects with the effect of year nested within species. We found that mercury concentrations in eggs were significantly higher than concentrations in adult blood across all species ($F_{1,242} = 9.17$, $p = .003$; Table 1). While not included in the model, a simple comparison of means between egg, adult blood, and juvenile blood indicates that juvenile blood has significantly lower Hg concentrations than the other two tissue types (ANOVA $F_{1,112} = 49.8$, $p < .001$; Table 2).

After we controlled for tissue type, total mercury concentrations were shown to vary considerably among species ($F_{3,31.8} = 13.5$, $p < .0001$), year ($F_{1,77.3} = 90.2$, $p < .0001$), and interaction of species and year ($F_{3,112.6} = 22.7$, $p < .0001$). Based on a Tukey test of least squared means for a post-hoc comparison, three of the four species showed significant increases in tissue mercury concentrations in 2013 when compared to 2008 (Fig. 2). All mercury concentrations presented in this section are predicted blood mercury (ww, μg/g) from the linear model (see Fig. 2). Common Eiders averaged 0.33 μg/g (95% Confidence Interval: 0.20, 0.53) in 2013 and 0.08 (0.05, 0.13) in 2008 (log-difference = 1.5 ± 0.26 μg/g, $p < .001$). Double-crested Cormorants were higher overall than eiders and averaged 0.84 μg/g (0.53, 1.35) in 2013 and 0.16 (0.10, 0.24) in 2008 (log-difference = 1.7 ± 0.13 μg/g, $p < .001$). Black Guillemots had a similar pattern to cormorants with an average of 0.65 μg/g (0.44, 0.97) in 2013 and 0.22 (0.13, 0.37) in 2008 (log-difference = 1.1 ± 0.30 μg/g Hg, $p = .01$). Only Leach's Storm Petrel showed no statistical difference between sampling occasions, 0.45 μg/g (0.30, 0.67) in 2013 and 0.47 (0.32, 0.69) in 2008. Their mean mercury concentration was the highest of all four focal species in 2008, and, despite remaining stable, was second to lowest in 2013, being surpassed by the Double-crested Cormorant and Black

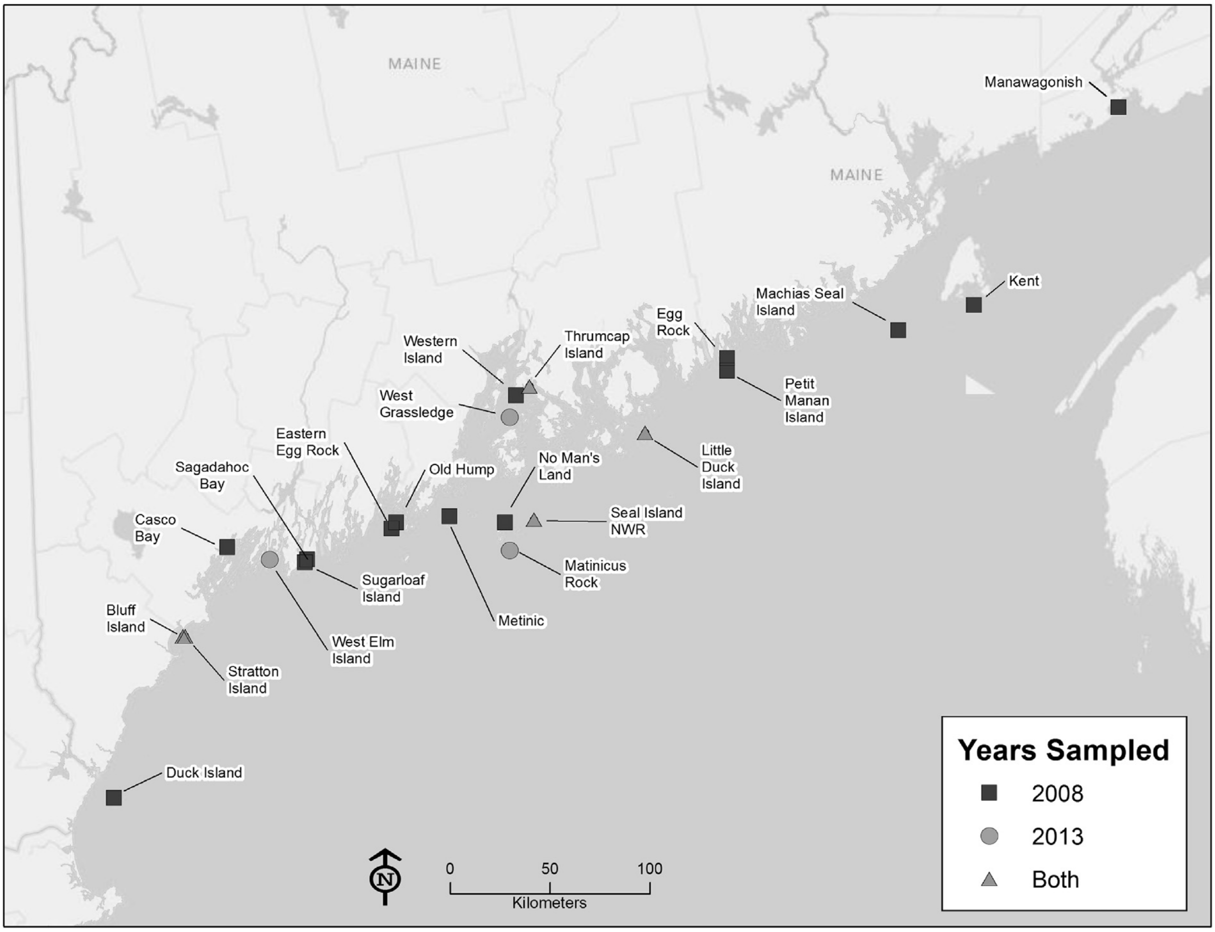

Fig. 1. Map of the sampling locations of marine bird breeding colonies in the Gulf of Maine for our focal species from the previous study (2008), this study (2013), and in both years.

Table 1
Comparison of mean mercury concentrations (μg/g, wet weight) in comparable tissue types for the four focal species between studies. Baseline samples (1998–2006) from Goodale et al. (2008).

Species	Year	Tissue type	*n*	Mean (μg/g)	StDev	Min	Max
Common Eider	1998–2005	Adult blood	4	0.109	0.078	0.025	0.204
		Egg	4	0.136	0.046	0.100	0.202
	2013	Egg	32	0.577	0.307	0.216	1.21
Leach's Storm-Petrel	2004–2006	Adult blood	28	0.540	0.366	0.034	1.994
		Egg	10	0.62	0.263	0.289	1.253
	2013	Adult blood	31	0.516	0.229	0.174	1.300
Double-crested Cormorant	2004–2005	Egg	46	0.279	0.089	0.114	0.453
	2013	Egg	36	1.41	0.819	0.403	4.39
Black Guillemot	2005–2006	Egg	28	0.521	0.231	0.163	1.010
	2013	Adult blood	23	0.916	0.490	0.413	2.571

Table 2
Mean mercury values (μg/g, wet weight) in juvenile blood sampled for three of the four focal species in the previous study (Goodale et al., 2008). No juvenile blood was sampled from Common Eiders in that study.

Species	Year	*n*	Mean (μg/g)	StDev	Min	Max
Leach's Storm-Petrel	2004–2006	20	0.030	0.041	0.005	0.196
Double-crested Cormorant	2004–2005	5	0.176	0.119	0.055	0.369
Black Guillemot	2005–2006	3	0.105	0.020	0.085	0.125

Guillemot (Fig. 2).

4. Discussion

The marine bird species sampled in this study utilize a variety of prey types and were specifically chosen to represent a range of trophic levels across near and offshore zones in the Gulf of Maine. Mercury concentrations increased in a statistically and ecologically significant manner in three of the four focal species, compared with the baseline levels described by Goodale et al. (2008). Species differences in mercury concentrations are likely related to trophic level differences (e.g., species that forage at upper trophic levels generally have higher mercury concentrations than those that feed at lower trophic levels), but also significantly influenced by tissue type.

4.1. Gulf of Maine seabird mercury burdens

The mean mercury concentration we found in Common Eider eggs is considerably higher than that seen by Meattey et al. (2014) from blood samples collected in the Gulf of Maine between 1998 and 2011, although the range of mercury concentrations was similar, and also higher than that seen in eggs in nearby Nova Scotia, although it is not clear exactly what years were sampled (Pratte et al., 2015). It was lower than that found in eggs of the same species breeding in the Canadian Arctic (Akearok et al., 2010; Mallory et al., 2004). As a benthic invertivore, mainly consuming bivalves, this species forages at a low trophic level, which probably limits its dietary exposure to mercury. Double-crested Cormorants have often been studied for contaminant exposure, but usually at inland sites where adults are shot or collected as part of a culling program, and their internal organ tissues are

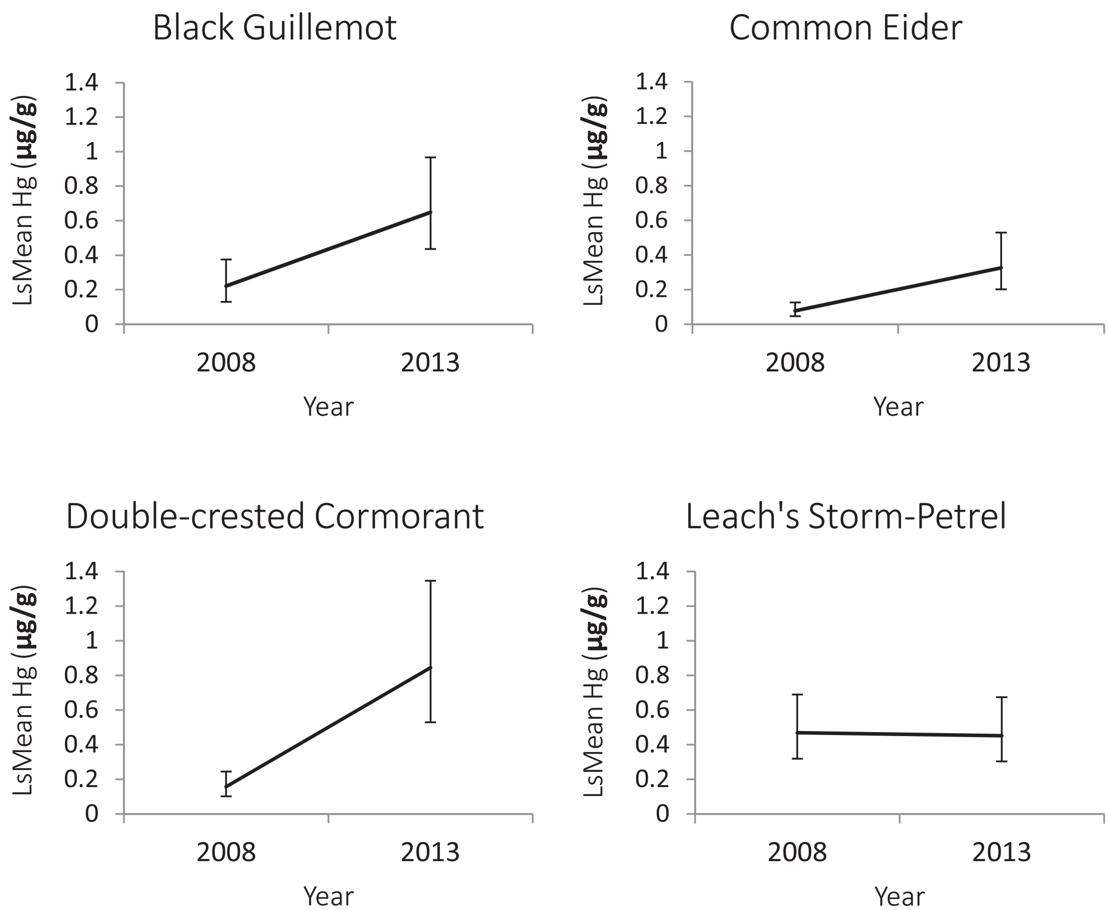

Fig. 2. The trend in general linear model predicted mean blood mercury values (µg/g ± 95% CI) of the four sentinel species between studies in 2008 and 2013. Focal species showed a significant increase over time, except the Leach's Storm-Petrel, which was high to begin with and remained stable over time.

assessed for contaminant concentrations, including mercury (Robinson et al., 2009; Robinson et al., 2011; Hall et al., 2014). It is difficult to compare results from those tissues types directly with the studies carried out to date in the coastal region of the Gulf of Maine (without inter-tissue conversions). The Black Guillemot, however, is known to be high in mercury across both its continental and global range (Akearok et al., 2010; Dam et al., 2004; Jensen et al., 1972; Goodale et al., 2008), and our results were consistent with these observations. Of the four focal bioindicator species, only Leach's Storm-Petrels remained stable. Based on other studies, this species appears to have a consistently high exposure to mercury (Goodale et al., 2008; Bond and Diamond, 2009; Pollett et al., 2016), and our results were no different. The fact that this wide-ranging pelagic species is the only one to have remained stable, while all others appear to have sharply increased, suggests that a major shift in methylmercury availability may have occurred in the inshore waters of the Gulf of Maine in the years between these studies.

4.2. Tissue type

Tissues sampled in this study varied among species and created a complex analysis when comparing results among species within and between years. The first issue is that avian tissues vary in the capacity to retain methylated mercury due to varying lipid and protein concentrations (Jackson et al., 2011; Spalding et al., 2000). While this can be potentially confounding for a study describing trends in mercury exposure over multiple tissue types, these differences can be disentangled using statistical modeling as long as there are enough samples across all tissue types. Multiple tissue samples from each individual would be ideal, as differences in acquiring different tissue types could bias our modeled comparisons of mercury among tissue types. When mercury data from various tissues of the same individual are not available, this method is a useful tool when the assumptions are met.

The second issue is that avian tissues have different metabolic half-lives and are produced at different times. This is particularly an issue in eggs where variation in mercury exposure may also relate to the amount of time individuals spent on the breeding ground prior to egg-laying. In general, blood mercury concentrations reflect exposure over the past few days, whereas eggs can indicate mercury exposure over the past few days to several months (depending on species foraging habits). Seabird eggs are generally considered good indicators of local mercury exposure (Burger and Gochfeld, 2004), since most seabirds in tropical and temperate regions spend many weeks on the breeding grounds prior to egg-laying, acquiring the resources required to produce their eggs (i.e. 'income' breeders). Seaducks, however, are considered 'capital' breeders, building up large body reserves prior to reaching their breeding areas, and can arrive with enough endogenous resources to begin egg-laying soon after arrival (Goudie et al., 2000). Common Eiders breeding in Maine colonies, however, are known to winter relatively locally, remaining in the Gulf of Maine or moving just south to the Cape Cod region (B. Allen, MDIFW, pers. comm.), suggesting they are still reliable indicators of local conditions. While we were able to correct for absolute differences in mercury concentrations between blood and eggs to obtain more reliable estimates of changes within species, we cannot control for these differences in temporal integration between tissue types.

Generally, the inshore region of the Gulf of Maine is known to be impacted by terrestrial sources of mercury, largely delivered by rivers and streams (Sunderland et al., 2012) and exposure to methylmercury could ultimately be related to (1) dietary shifts related to natural and/or anthropogenic factors, and/or (2) significantly changing climatic conditions (e.g., increasing precipitation and warmer average temperatures). How each of these potential factors play a role in the increase of methylmercury availability and ultimately increases of mercury body burdens in the chosen upper trophic level bioindicators requires further examination.

4.3. Dietary shift

The observed changes in mercury concentrations found in three of the four focal species could be due to increases in mercury availability in the ecosystem (e.g., Vo et al., 2011; Lavoie et al., 2015) or shifts in diet towards prey species that bioaccumulate more mercury (e.g., Arcos et al., 2002; Lepak et al., 2009). As predators, marine birds integrate contaminant loads across the ecosystem, and, thus, provide a simple method by which we can track broad-scale marine ecosystem health. However, interpretation of changes in mercury exposure over time is complicated by the potential for dietary shifts over the same time period (see Hebert et al., 2009). Stable isotope data can help distinguish these two different causes of the same pattern (e.g., Burgess et al., 2013), but those data are lacking in this study. We know that the chick dietary composition of piscivorous marine birds changes from year to year in the Gulf of Maine, with Atlantic herring (*Clupea harengus*) and sand lance (*Ammodytes dubius*) being highly available to seabirds in some years and much less so in others (Kress et al., 2016). So, if adults are forced to forage for themselves in a similar manner, year to year diet switching due to the availability of particular prey species could play a significant role in blood mercury levels. Likewise, in the benthivores, Common Eiders are traditionally heavily reliant on Blue Mussels (*Mytilus edulis*), which have declined by > 60% in the Gulf of Maine since the 1970s (Sorte et al., 2016). This change in the availability of their primary prey may have caused a major shift in their diet (towards other benthic invertebrates, such as crabs, urchins, etc.), but such a change would likely have taken place over a much longer timeframe than that measured here.

4.4. Climate change

While global mercury emissions are currently thought to be decreasing (Zhang et al., 2016), there is substantial variance in mercury bioavailability among ecosystems and habitats. Although mercury is broadly increasing across the world's oceans, concentrations in the surface waters of the North Atlantic Ocean are thought to be decreasing (UNEP, 2013). The Gulf of Maine, however, may contradict that general Atlantic trend for several reasons, potentially exacerbating an already rapidly worsening situation. An observed, extreme rate of increase in mean sea surface temperature in the region (Pershing et al., 2015) may be driving greater mercury availability, since methylation increases with temperature in cold ocean waters (Belkin, 2009; Booth and Zeller, 2005; St. Pierre et al., 2014), as well as long-term trends in foundation species and shifts in community composition (Sorte et al., 2016). Furthermore, the Gulf of Maine may also be particularly sensitive to ocean acidification (a decrease in pH caused by the absorption of atmospheric CO_2; Gledhill et al., 2015). Mercury is of particular concern in acidic environments because low pH also increases the rate of methylation and boosts the bioavailability of mercury to wildlife (Kelly et al., 2003).

5. Conclusion

Understanding trends in mercury contamination in the Gulf of Maine ecosystem, and its current and future significance to marine birds and other taxa, including human health, will require a dedicated, standardized, long-term monitoring scheme (see Evers et al., 2008b, 2016). While eggs are relatively easy to collect and are consistent with past (Mierzykowski et al., 2005) or current (Burgess et al., 2013) monitoring efforts, more consideration should be given to the interpretation of mercury concentrations in different tissue types and the specific objectives of the study. Additional considerations for monitoring mercury in marine wildlife should include: 1) developing a reasonable annual sampling scheme for birds that minimize breeding colony disturbance, 2) determining where capital breeders, like sea ducks, spend time prior to breeding, if eggs are used as a sampling type, and 3) incorporating stable isotope analyses into monitoring efforts to help disentangle changes in mercury exposure due to changes in prey mercury versus changes in foraging trophic position. Despite the interpretive limitations of our data, there appears to be clear and sufficient evidence of elevated and increasing mercury concentrations in these focal marine bird species breeding in the Gulf of Maine, particularly in the inshore region. We believe this pattern and trend warrants considerably greater research attention, as part of a focused and coordinated regional and global monitoring effort (see Evers et al., 2016).

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