



Elevated mercury levels in a wintering population of common eiders (*Somateria mollissima*) in the northeastern United States



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ABSTRACT

In North America and Europe, sea ducks are important indicators of ecological health and inshore marine pollution. To explore spatial variation in mercury accumulation in common eiders in the northeastern United States, we compared concentrations of total mercury in common eider blood at several New England locations between 1998 and 2013. Eider food items (mollusks) were collected and analyzed to determine if mercury concentrations in eider blood were indicative of local mercury bioavailability. Eiders from Plum Island Sound, MA had a significantly higher mean blood mercury concentration (0.83 µg/g) than those in other locations. Mean mercury levels in this population were also nearly three times higher than any blood mercury concentrations reported for common eiders in published literature. We observed consistent patterns in eider blood mercury and blue mussel mercury concentrations between sites, suggesting a tentative predictive quality between the two species.

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1. Introduction

Mercury (Hg) deposition into the environment through anthropogenic activities continues to be an important environmental issue across the globe (Pirrone and Mason, 2009). Pollutant levels tend to be higher in aquatic environments due to direct run-off or input from rivers, point-source pollution, and atmospheric deposition (Chen et al., 2008; Goodale et al., 2008). Similarly, species that forage in aquatic environments are at higher risk of increased levels of contamination because of the potential for rapid movement of contaminants in aquatic food chains and the accumulation of pollutants in the bottom sediments of intertidal and shallow marine environments (Burger and Gochfeld, 2009). Following deposition, inorganic Hg may be converted to its more bioavailable, and toxic form, methylmercury (MeHg). Throughout the food chain MeHg increases through a bioaccumulation factor of approximately 10 million, reaching toxic concentrations at the highest trophic levels (Driscoll et al., 2007). Exposure to high concentrations of Hg may have detrimental effects on wildlife, possibly leading to population declines. In different bird species these

effects can include wing area asymmetry, lethargy, and reduced productivity (common loons; Evers et al., 2008), erratic chick behavior and lowered response to maternal calls (mallards; Heinz, 1979), and reduced abdominal fat accompanied by increased levels of parasitic intestinal worms (common eiders; Wayland et al., 2001b).

The life history and foraging strategies of many species of waterfowl expose them to potential Hg poisoning. Mercury exposure in waterfowl is a multi-step process that involves direct uptake through ingestion, transport in blood and subsequent accumulation in internal tissues such as liver, kidneys, and muscle. Redistribution to plumage occurs during feather growth and elimination occurs through deposition to eggs and excreta (Monteiro and Furness, 1995). For long-lived species such as sea ducks which feed at higher trophic levels, biomagnification of Hg presents a greater risk of neurological, behavioral, and reproductive impacts (Burgess and Meyer, 2008; Evers et al., 2005; Evers et al., 2008).

In North America and Europe, sea duck species have become recognized as important indicators of ecological health and inshore marine pollution (Mallory et al., 2010; Goodale et al., 2008; Savinov et al., 2003). Declines in several of these species have led to increased research on the effects of environmental contaminants on their populations and physiology (Provencher et al.,

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2014; Wayland et al., 2008a; Fisk et al., 2005; Barjaktarovic et al., 2002). Trace elements in sea ducks have typically been examined in liver or kidney samples; however non-lethal sampling methods using feather and blood samples have recently been used with increased frequency (Bond and Diamond, 2009; Burger and Gochfeld, 2009; Wayland et al., 2007). Blood is best used to indicate short-term exposure and recent dietary uptake (Evers et al., 2008; Monteiro and Furness, 2001; Kahle and Becker, 1998). As a result of intensified sampling efforts and an improved understanding of Hg contamination dynamics in aquatic ecosystems, waterfowl species have become increasingly important bioindicators of both freshwater and marine ecosystem health. Species of waterfowl in the northeastern U.S. at highest risk to Hg availability include piscivores and molluscivores (Evers et al., 2005). Common eider (*Somateria mollissima*; hereafter eider), a common sea duck in the northeastern U.S., feed heavily on mollusks. Where available, blue mussels (*Mytilus edulis*; hereafter mussels) represent a large percentage of an eider's diet (Goudie et al., 2000). Blue mussels are suspension feeders that filter large volumes of water and readily accumulate toxins and environmental contaminants in their tissues. This accumulation represents the portion of contaminants in the ecosystem that is biologically available to higher trophic level species (Leblanc et al., 2009). The blue mussel's sedentary behavior and ability to accumulate contaminants from the water column have led to its wide use as an indicator of aquatic ecosystem health (Julshamn and Grahl-Nielsen, 1996; Airas et al., 2004; Garron et al., 2005; Burger and Gochfeld, 2006). While there are currently no known studies that examine correlations between blood and prey item Hg in free-living eiders, understanding this dietary preference suggests that Hg levels in eider blood should be correlative with Hg levels found in blue mussels collected at the same location.

Common eiders are the largest ducks in the Northern Hemisphere and inhabit marine environments throughout all stages of their life cycle. Eiders nest in large colonies on marine islands and congregate in large numbers in inshore waters in arctic and subarctic coastal habitats during the non-breeding season (Goudie et al., 2000). The life history characteristics and long lifespan of eiders, which may exceed 20 years (Klimkiewicz and Fletcher, 1989), make eiders a sentinel species for contaminant monitoring in coastal and inshore marine habitats.

The objectives of this study were to (1) compile blood Hg data from past and recent sampling efforts in an attempt to identify spatial variations in Hg accumulation in eiders at inshore marine habitats throughout the northeastern United States, and (2) collect prey items from capture locations to determine if Hg concentrations in eider blood are indicative of Hg bioavailability in the immediate environment. Obtaining this information will provide a more comprehensive understanding of Hg spatial distribution in sea ducks and enable future studies designed to determine the effects of elevated Hg concentrations.

2. Materials and methods

2.1. Study areas

Capture efforts took place in near-shore waters of Maine (ME; 1998–2011), Massachusetts (MA; 2009–13), and Rhode Island (RI; 2010–11), USA (Fig. 1). Eider captures in Maine spanned the state from Ogunquit in the south to Sagadahoc and Penobscot Bays in the mid-coast area, with the majority of captures taking place at Pine Point near the coastal edge of Scarborough Marsh. Other captures occurred amongst the several small, coastal islands within Casco Bay. Captures in Massachusetts took place just inside the mouth of Plum Island Sound at Ipswich Bluffs. Using a data set of

biotic Hg concentrations, Evers et al. (2007) identified the surrounding area as a biological Hg hotspot for the northeast region. We suspected that blood Hg levels in eiders sampled in the vicinity of this location would be higher than those caught elsewhere in the study area. Additional captures in Massachusetts took place near Calf Island in Boston Harbor. Capture of eiders in Rhode Island took place on the Sakonnet River, Middletown, Easton's Beach, Newport, and Beavertail Point, Jamestown, all three of which are in the eastern portion of the state. Additional eiders were captured at Quonochontaug Breachway, Charlestown along the western portion of the south coast.

2.2. Collection of tissue samples

Bird sampling efforts occurred opportunistically between the months of December and May during all sampling years. With the exception of birds from Boston Harbor, MA all birds were captured during wintering or staging periods. Birds in Boston Harbor were captured during the early stages of nesting, but none were taken off nest. Non-lethal capture methods were used for the sampling of all birds. Eiders were captured using floating mist nets as described by Brodeur et al. (2008). Blood was drawn using a 23 or 25-gauge needle to puncture either the cutaneous ulnar vein in the wing or the tarsal vein and collected in heparinized capillary tubes. No more than 1% of the bird's body weight in blood was collected and the physical condition of each bird was briefly evaluated prior to sampling to check for capture-related injuries or signs of poor health. The tubes were sealed on both ends with Critocaps® or Critoseal and placed in a labeled 10 cc plastic vacutainer. All samples were frozen at -20°C within six hours of collection. Prior to release all birds were banded, weighed. Standard morphometrics including tarsus length and width, bill length, width, and depth, and resting wing chord were also taken. Birds were aged and sexed based on plumage. Sixty-six females and eighty-one males were analyzed, of which 88% (130/147) were determined to be second-year or older. Mussels and periwinkle snails (*Littorina littorea*; hereafter snails) were collected by hand from rocks in shallow intertidal zones near feeding and capture areas in Pine Point, ME, and Ipswich Bluff, MA in 2010. Prior to analysis of prey items, standard morphometrics were taken and soft tissue was extracted and frozen at -20°C . In mussels, shell length, width, and depth were measured along with fresh whole weight. Only a fresh whole weight was recorded for snails. Soft tissue of both mussels and snails was extracted by carefully prying open the shell with a stainless steel micro spatula and scraping contents into a clean plastic container. Soft tissue weight was recorded after extraction. Subsets of similarly-sized prey specimens were analyzed for each of these locations. Blue mussels collected in both locations ranged from 18 to 45 mm in shell length with a combined average of 33 mm. This size class was chosen based on available mussels in the collection area but is supported by a study in Germany in which 80% of blue mussels consumed by eiders were between 30 and 55 mm in length (Nehls and Ketzenberg, 2002). A total of 147 eider blood samples were collected and analyzed, along with 38 mussels and 20 snails.

2.3. Determining mercury concentrations in eider and prey tissue

Samples collected in 2007 were analyzed at the Battelle Marine Sciences Laboratory in Sequim, Washington, USA, whereas samples collected in 2009 were shipped to the Utah Veterinary Diagnostic Laboratory in Logan, Utah, USA for analysis. All subsequent blood samples, along with prey item tissues, were analyzed at the Wildlife Mercury Research Lab at BRI Headquarters in Gorham, Maine, USA. Whole blood samples sent to the Utah State Veterinary Diagnostic Laboratory (UVDL, Logan, UT) were analyzed for total Hg

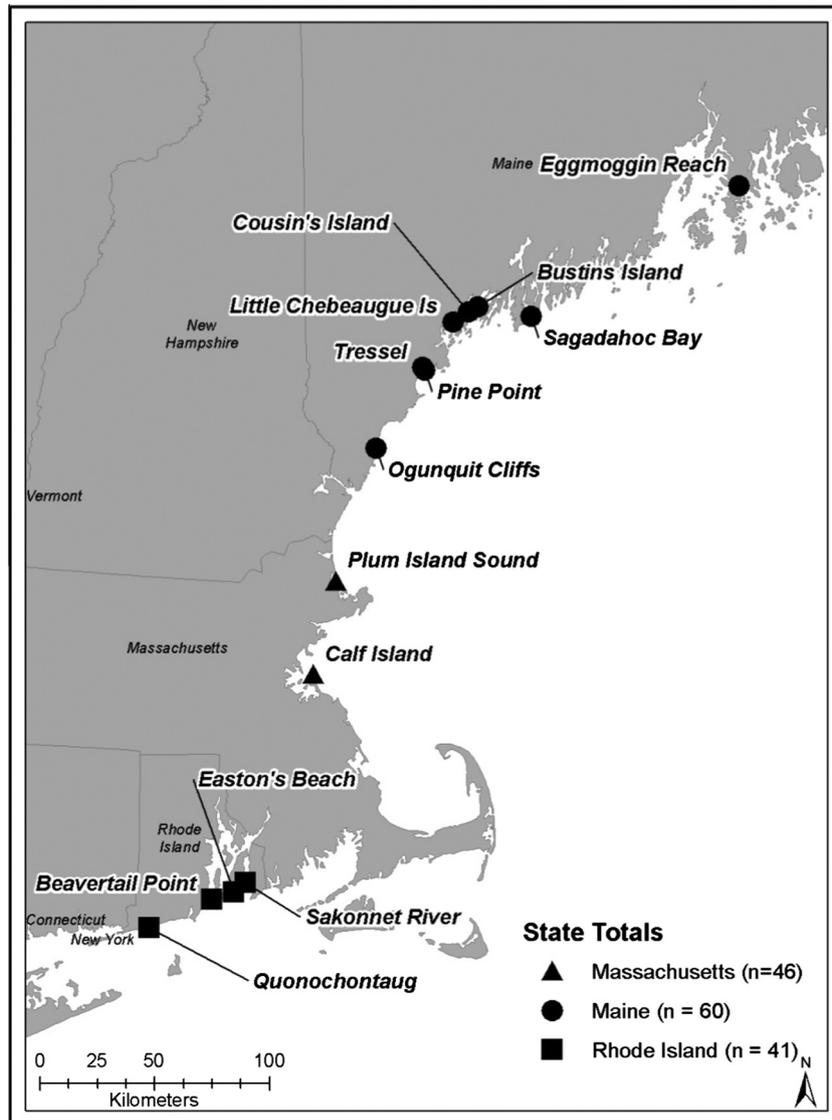


Fig. 1. Locations and sample size of blood samples collected from common eiders in 3 northeastern states between 1998 and 2013.

using validated protocols with argon plasma mass spectrometry. To quantify Hg content, whole blood analyses were performed using nitric acid digested samples. The whole blood samples were digested (1:1) in trace mineral grade nitric acid (Fisher Scientific, Pittsburgh, Pennsylvania 15275, USA) in sealed 10 ml Oak Ridge screw-cap Teflon digestion tubes (Nalge Nunc International, Rochester, New York 14625, USA) on a heat block for 2 h at 90 °C. The digest was then 1:10 with 18.2 mohm water in a 15 ml polypropylene trace metal free tube (ELKAY, Mansfield, Massachusetts 01801, USA). This provided 5% nitric acid matrices for analysis, which was matrix matched for all standard curve and quality control samples. Mineral content analysis was performed using an ELAN 6000 inductively coupled plasma mass spectrometer (ICP-MS) (Perkin Elmer, Shelton, Connecticut 06484, USA). To prevent any Hg carryover between samples a flush solution composed of 5% nitric acid with 10 ppm gold added was used between samples. Sequential 1:10 dilutions of samples exceeding the Hg standard curve were made and re-analyzed, using 5% nitric acid. Standard curves and quality control (QC) samples were analyzed every 5 samples. National Institute of Standards and Technology (NIST) standards were also analyzed to verify accuracy of the analytical results. QC analyses were considered acceptable if $\pm 10\%$ of the known Hg concentration, but were typically less than $\pm 5\%$.

Blood samples sent to the Battelle Marine Sciences Laboratory were analyzed following USEPA Method 1631e (nitric/sulfuric digestion followed by SnCl_2 reduction and purge and trap Cold Vapor Atomic Fluorescence Spectroscopy). All other tissue samples were analyzed for total Hg at the Wildlife Research Mercury Lab at BRI Headquarters using a thermal decomposition technique with a direct Hg analyzer (DMA 80, Milestone Incorporated) following the USEPA Method 7473 (US EPA, 2007). All results from each lab were within 10% of reference samples. Mussel and snail samples were dried whole in a Labconco Freezone 4.5 bench top freeze drier prior to analysis.

Eider blood Hg concentrations are reported in micrograms per gram ($\mu\text{g/g}$) and on a wet weight (ww) basis. Mercury concentrations in the soft tissue of prey items were reported and graphically presented in $\mu\text{g/g}$, dry weight. For comparison with other literature in the discussion, these values were converted to wet weight using methods described in Stickel et al. (1973).

2.4. Statistical analysis

Results throughout this study are reported as arithmetic means. All statistical analyses were performed on log-transformed Hg concentrations using JMP v.9.0 statistical software (SAS Institute Inc.,

2010). A one-way analysis of variance (ANOVA) was used to test hypotheses. When the ANOVA determined significant difference, Student's *t*-tests were used to compare paired data sets. Non-parametric Wilcoxon and/or Steel–Dwass tests were used to compare multiple groups (i.e. sites within a state). Results of statistical tests were considered significant at $p < 0.05$. Back-transformed data are presented in tables and figures. Generalized linear models and simple linear regressions were used to determine if age, sex, and body weight influenced eider Hg concentrations. Sampling year and location were also considered as factors. Within-year, temporal changes in body weight and Hg concentrations were not considered due to short sampling periods at each site. In this same way, we tested whether size parameters such as shell length and soft tissue weight affected prey item Hg concentrations. We used an analysis of covariance (ANCOVA) to examine the relationship between Hg concentrations in eiders and mussels in Maine and Massachusetts and determine if one value was predictive of the other at these sites.

3. Results

3.1. Mercury in eider blood

Blood samples from 147 individual eiders, representing 14 capture locations within three different states, were analyzed for total Hg. We found the lowest individual Hg concentration in Sagadahoc Bay, ME (0.013 $\mu\text{g/g}$) and the highest in Plum Island Sound, MA (1.75 $\mu\text{g/g}$) (Table 1). In Maine and Rhode Island, no significant differences were determined among sites ($p = 0.2477$ and 0.2480 , respectively). Statistical tests were therefore run combining all sites within those states.

Mercury levels in Plum Island Sound were significantly higher than those at Calf Island, Boston Harbor, MA ($p = 0.0003$) and therefore each site is presented separately. Mean blood Hg in individuals captured in Plum Island Sound was 0.83 $\mu\text{g/g}$ and ranged from 0.14 to 1.75 $\mu\text{g/g}$ ($n = 26$). No significant difference in Hg concentrations was reported based on age or sex ($p = 0.0901$ and 0.9179 , respectively). Blood Hg concentrations at Calf Island, Boston Harbor, MA ranged from 0.14 to 0.85 with a mean of 0.36 $\mu\text{g/g}$ ($n = 20$). No significant difference in Hg concentrations was reported based on age or sex ($p = 0.7746$ and 0.7657 , respectively).

Between the years 1998 and 2011, 60 eiders were sampled at various locations along the Maine coast. The mean blood Hg concentration in Maine was 0.21 $\mu\text{g/g}$ and ranged from 0.013 to 0.59 $\mu\text{g/g}$. Neither age nor sex significantly influenced results ($p = 0.2725$ and 0.3327 , respectively).

In 2010 and 2011, a combined 41 eiders were captured and sampled in areas of coastal Rhode Island. The mean blood Hg concentration in Rhode Island was 0.24 $\mu\text{g/g}$ and ranged from 0.08 to 0.85 $\mu\text{g/g}$. There was no significant difference in Hg concentrations based on age or sex ($p = 0.0830$ and 0.1992 , respectively).

Mercury values in eiders sampled in Plum Island Sound, MA were significantly higher than those in Boston Harbor, Maine or Rhode Island ($p = 0.0003$, $p < 0.0001$ and $p < 0.0001$, respectively) (Fig. 2).

Body weights were highest in Maine, followed by Massachusetts and Rhode Island. There was no significant difference between Maine and Massachusetts weights ($p = 0.0644$) but both were significantly heavier than those in Rhode Island ($p < 0.0001$ and $p = 0.0003$, respectively). Across all sites, body weight was not a significant predictor of Hg concentration ($R^2 = 0.0004$; $p = 0.8152$).

From the ANCOVA, we found a significant relationship between eider blood and blue mussel tissue in Pine Point, ME and Ipswich Bluff, MA. At each site, eider blood Hg concentrations were consistently 34 times higher than blue mussels ($R^2 = 0.90$; $p < 0.0001$).

3.2. Mercury in prey items

Mussels and snails collected in Massachusetts contained significantly greater concentrations of Hg than those collected in Maine ($p < 0.0001$ and $p = 0.0030$, respectively). Concentrations of Hg in mussels from Massachusetts had a mean value of 0.13 $\mu\text{g/g}$ and ranged from 0.07 to 0.17 $\mu\text{g/g}$. In comparison, mussels sampled in Maine had a mean Hg concentration of 0.07 $\mu\text{g/g}$ and ranged from 0.03 to 0.09 $\mu\text{g/g}$ (Fig. 3). Mean Hg concentrations in snails from Massachusetts in 2010 ranged from 0.06 to 0.16 $\mu\text{g/g}$ with a mean value of 0.11 $\mu\text{g/g}$. Mean Hg levels in snails sampled in Maine ranged from 0.05 to 0.08 $\mu\text{g/g}$ with a mean Hg level of 0.06 $\mu\text{g/g}$ (Fig. 4). For comparison with other literature, Hg values in mussels and snails from this study were converted to wet weight using methods described in Stickel et al. (1973). We use 89.8%-moisture for conversion per findings reported in Franson

Table 1
Summary statistics of blood Hg concentrations ($\mu\text{g/g}$, wet weight) in common eiders sampled throughout the study period.

Year	State ^a	Waterbody	Location	N ^b	Mean Hg \pm SD	Median (Min–Max)
1998	ME	Sagadahoc Bay	Sagadahoc Bay	2	0.043 \pm 0.042	0.043 (0.013–0.072)
2000	ME	Casco Bay	Bustins Island	2	0.170 \pm 0.048	0.170 (0.136–0.204)
2007	ME	Casco Bay	Little Chebeague Island	4	0.256 \pm 0.116	0.245 (0.134–0.402)
2007	ME	Southern Maine Coast	Ogunquit Cliffs	2	0.193 \pm 0.069	0.193 (0.144–0.241)
2007	ME	Penobscot Bay	Eggmoggin Reach	1	0.159	NA ^c
2007	ME	Saco Bay	Pine Point	1	0.176	NA
2010	ME	Casco Bay	Little Chebeague Island	4	0.247 \pm 0.104	0.253 (0.115–0.369)
2010	ME	Saco Bay	Pine Point	23	0.212 \pm 0.115	0.177 (0.082–0.594)
2010	ME	Scarborough Marsh	Tressel	16	0.233 \pm 0.103	0.235 (0.090–0.453)
2011	ME	Casco Bay	Cousin's Island	1	0.131	NA
2011	ME	Saco Bay	Pine Point	4	0.224 \pm 0.170	0.162 (0.097–0.474)
2009	MA	Plum Island Sound	Ipswich Bluff	12	0.965 \pm 0.474	0.991 (0.137–1.755)
2010	MA	Plum Island Sound	Ipswich Bluff	14	0.711 \pm 0.401	0.742 (0.223–1.536)
2013	MA	Boston Harbor	Calf Island	20	0.365 \pm 0.186	0.298 (0.144 – 0.848)
2010	RI	Rhode Island Coast	Easton's Beach	1	0.120	NA
2011	RI	Rhode Island Coast	Beavertail Point	14	0.203 \pm 0.118	0.161 (0.082–0.442)
2011	RI	Rhode Island Coast	Quonochontaug	25	0.266 \pm 0.148	0.256 (0.131–0.848)
2011	RI	Rhode Island Coast	Sakonnet River	1	0.220	NA

^a ME = Maine; MA = Massachusetts; RI = Rhode Island.

^b Number of samples analyzed.

^c NA, not applicable.

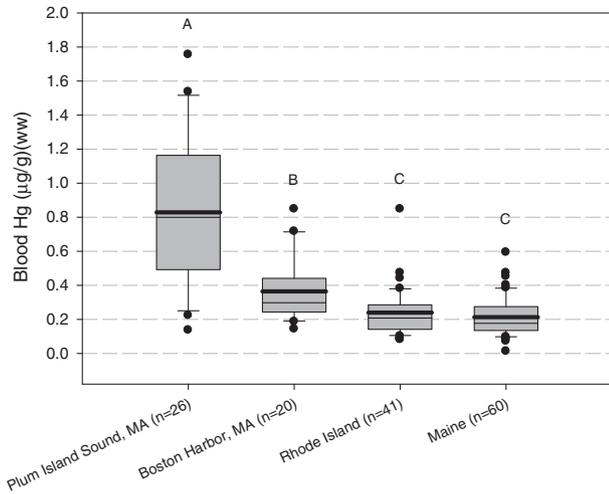


Fig. 2. Mercury concentrations (wet weight) in blood tissue of eiders sampled. Different letters indicate significant differences ($p < 0.05$). Plot depicts 25th and 75th percentiles, median (thin line), mean (thick line), and outliers. Error bars represent 10th and 90th percentiles.

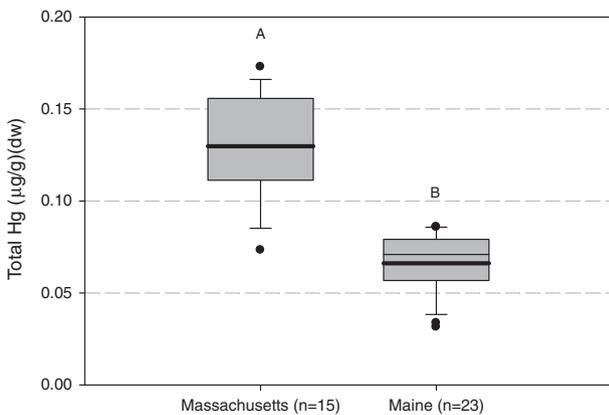


Fig. 3. Mercury concentrations (dry weight) in mussels collected at Plum Island Sound, MA and Pine Point, ME. Different letters indicate significant differences ($p < 0.05$). Plot depicts 25th and 75th percentiles, median (thin line), mean (thick line), and outliers. Error bars represent 10th and 90th percentiles.

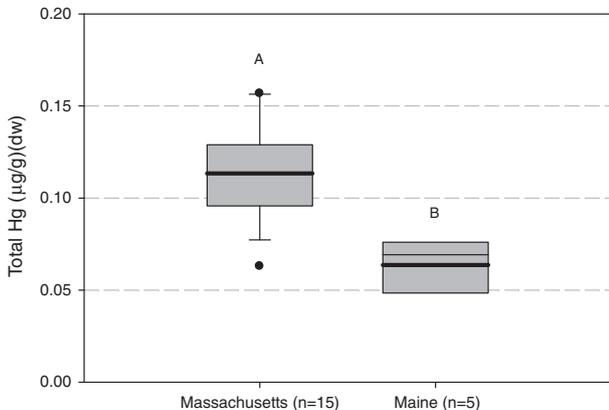


Fig. 4. Mercury concentrations (dry weight) in snails collected at Plum Island Sound, MA and Pine Point, ME. Different letters indicate significant differences ($p < 0.05$). Plot depicts 25th and 75th percentiles, median (thin line), mean (thick line), and outliers. Error bars represent 10th and 90th percentiles.

et al. (1995). Using this conversion, mussels and snails from Massachusetts had mean wet weight Hg concentrations of 0.013 and 0.011 $\mu\text{g/g}$, respectively. Mussels and snails collected in Maine both had mean wet weight Hg concentrations of 0.006 $\mu\text{g/g}$ after conversion.

Mussels shell lengths were significantly higher ($p = 0.0001$) in Massachusetts (38.0 ± 2.59 mm, $n = 15$) than in Maine (29.9 ± 6.95 mm, $n = 23$). Mean soft tissue weight of mussels was also significantly higher ($p < 0.0001$) in Massachusetts (2.7 ± 0.83 g, $n = 15$) than in Maine (0.95 ± 0.70 g, $n = 23$). Soft tissue weights of snails collected in Maine were significantly higher ($p < 0.0001$) than those collected in Massachusetts. In combined mussels from both collection sites, we found shell lengths and soft tissue weights to have significant positive relationships with Hg concentrations ($p = 0.0013$ and $p < 0.0001$, respectively). However, predictive power was low to moderate in both cases ($R^2 = 0.25$ and 0.39 , respectively). In combined snails, we found a significant relationship between soft tissue weight and Hg ($p = 0.0116$) but predictive power again was low ($R^2 = 0.30$).

4. Discussion

4.1. Mercury in eider blood

Few studies have examined Hg concentrations in eider blood (Fig. 5). Those that have typically do not report elevated concentrations when compared to other seabird species sampled. Bond and Diamond (2009) found that of several seabird species breeding on Machias Seal Island, New Brunswick, Canada, eiders had lower blood Hg concentrations ($0.05 \mu\text{g/g}$, ww) than razorbills (*Alca torda*), Leach’s storm-petrels (*Oceanodroma leucorhoa*), arctic terns (*Sterna paradisaea*), Atlantic puffins (*Fratercula arctica*), common terns (*Sterna hirundo*), and common murrelets (*Uria aalge*). Wayland et al. (2001a) found a mean blood Hg concentration of $0.23 \mu\text{g/g}$ (ww) with a maximum value of $0.37 \mu\text{g/g}$ in eiders sampled in the eastern Canadian Arctic during 1997 and 1998. Of 7 adult female eiders sampled in the Gulf of Finland in 1994, Hollmén et al. (1998) found only one sample ($0.22 \mu\text{g/g}$, ww) to be above the detection limit of $<0.025 \mu\text{g/g}$. Franson et al. (2000) analyzed blood samples from 268 breeding females in Finland in 1997 and 1998 and detected Hg in less than half, with a maximum

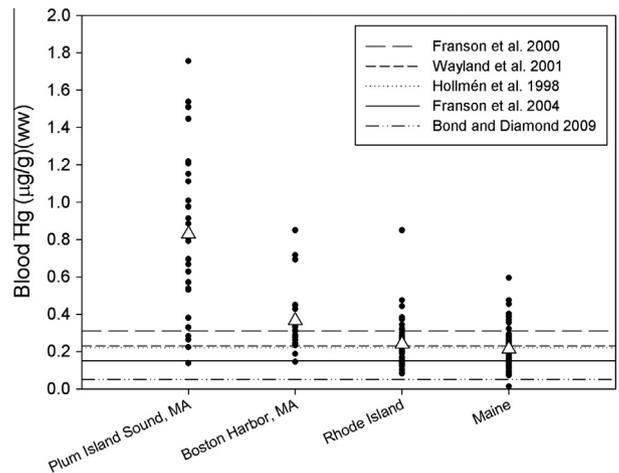


Fig. 5. Eider blood Hg levels from this study in comparison to selected published background levels. Triangles indicate mean Hg levels determined in this study. Lines represent: Franson et al. (2000) (Max: $0.31 \mu\text{g/g}$); Wayland et al. (2001a) (Mean: $0.23 \mu\text{g/g}$); Hollmén et al. (1998) (Max: $0.22 \mu\text{g/g}$); Franson et al. (2004) (Mean: $0.15 \mu\text{g/g}$); Bond and Diamond (2009) (Estimated Marginal Mean: $0.05 \mu\text{g/g}$).

concentration of 0.31 $\mu\text{g/g}$ (ww). Eiders sampled in eastern and western areas of the Beaufort Sea near Prudhoe Bay, Alaska, USA, had mean blood Hg concentrations of 0.014 and 0.017 $\mu\text{g/g}$, respectively (Franson et al., 2004; Hg values converted from $\mu\text{g/g}$ dry weight to $\mu\text{g/g}$ wet weight using methods described in Stickel et al., 1973). With the exception of birds sampled in Plum Island Sound, concentrations of total Hg were relatively low and were similar to mean values reported in common eider studies in other locations (Fig. 5): 0.05 $\mu\text{g/g}$ in New Brunswick, Canada (Bond and Diamond, 2009); 0.23 $\mu\text{g/g}$ in the Canadian arctic (Wayland et al., 2001a); <0.025–0.22 $\mu\text{g/g}$ in Finland (Hollmén et al., 1998); In Finland, a maximum concentration of 0.31 $\mu\text{g/g}$ with results from less than half of 203 samples above detection limits (Franson et al., 2000); 0.14 and 0.17 $\mu\text{g/g}$, respectively, in eastern and western Beaufort Sea locations (Franson et al., 2004). In this study, 67% (31 of 46) of birds captured in Massachusetts had blood Hg levels higher than the highest published background level (0.31 $\mu\text{g/g}$), followed by 19% (8 of 41) in Rhode Island and 15% (9 of 60) in Maine. Eiders captured in Plum Island Sound also had significantly higher blood Hg concentrations than Atlantic brant (*Branta bernicla hrota*) and white-winged scoters (*Melanitta fusca*) captured in the same location during the study period (unpublished data). The Hg concentrations we found in brant and scoters were within their respective previously reported background levels. At the time of this study, no published blood Hg data on brant species were available but concentrations from Plum Island Sound were within the range of Hg concentrations found in black brant (*Branta bernicla orientalis*) in Alaska (unpublished data). Wayland et al. (2008a) reported a mean blood Hg concentration of 0.19 $\mu\text{g/g}$ wet weight (range: 0.06–0.47) in white-winged scoters in Canada.

While some field studies across North America have investigated the presence of Hg in blood tissue of multiple eider species on their breeding grounds (Wilson et al., 2004; Wayland et al., 2008b, 2007, 2001a, b), none have thoroughly examined the dynamics of Hg exposure in wintering eiders. It is important to consider the entire annual cycle when examining and comparing Hg values across different studies. Throughout the year female eiders will go through several drastic fluctuations in body condition as they gain weight and increase fat stores in preparation for nesting, a period then marked by fasting and decreasing body condition (Wayland et al., 2005; Bolduc and Guillemette, 2003). These annual fluctuations may affect concentrations of Hg and other contaminants. For example, Hg concentrations may increase in certain organ tissues as body condition and organ mass decreases (Wayland et al., 2005). Conversely, concentrations of Hg may become diluted in organ tissues during pre-nesting periods when birds are gaining weight and increasing fat reserves (Fisk et al., 2005). Studies on breeding female eiders in the Canadian Arctic found that hepatic Hg concentrations were inversely correlated with body weight and organ mass, while total Hg content within the organs remained static (Wayland et al., 2005, 2003). In those studies, Hg concentrations tended to be higher in lighter birds with decreased organ mass. During winter, body composition and condition of eiders is dynamic and could possibly increase the likelihood of Hg exposure or increase its effects. A study on wintering common eiders in Greenland noted a decrease in fat reserves and body mass between late winter and early spring (Merkel et al., 2006), highlighting the importance of acquiring fat reserves during spring migration.

Captures at each site in our study took place within short periods of time (generally <2 weeks) and therefore temporal differences in body weight throughout the study period were not relevant. However, we found no relationship between blood Hg concentrations and body weight within the entire sample set. This lack of correlation is not surprising given the dynamics of Hg in the

organism. Mercury in blood represents short-term exposure and recent dietary uptake. The half-life of MeHg, which makes up >95% of total Hg in blood tissue (Fournier et al., 2002; Wayland et al., 2001a), was 40–65 days in non-molting adult Cory's shearwaters (*Calonectris diomedea*; Monteiro and Furness, 2001), 30–63 days in great skua (*Catharacta skua*; Bearhop et al., 2000), and 74 days in mallard ducks (*Anas platyrhynchos*; Heinz and Hoffman, 2004). It should be noted that birds captured in Boston Harbor for this study were in the early stages of nesting, and possibly recent arrivals to the breeding ground. Mercury concentrations in these birds may still be representative of spring staging areas occupied prior to arriving at the breeding site. Given the short time period that Hg from dietary uptake is represented in blood tissue, the relationship between blood Hg concentrations and those in prey items collected from the same locations suggests that blood Hg concentrations reported in this study are reliably indicative of Hg availability in the immediate environments where the birds were captured and to a lesser extent of Hg body burdens accumulated throughout a longer time period or in a variety of locations. While significant in this study, this relationship should be treated with caution until further studies can validate its utility across broader geographic ranges and with greater sample sizes.

Although few other blood Hg effect level threshold data exist for birds, a study by Heinz et al. (2009) examined the variability in avian species sensitivity to Hg. The authors injected a geometric progression of Hg doses ranging from 0 to 6.4 $\mu\text{g/g}$ into the eggs of 26 bird species and documented egg mortality. Species were categorized as having a low, medium, or high sensitivity to Hg, and several waterbird species appeared in each one of these categories. Eider sensitivity to Hg compared to other species of sea birds is unknown.

While adverse effect level thresholds for blood in sea ducks have not yet been determined, data from studies focused on other bird species, both aquatic and terrestrial, may be used to better understand how eiders differ from or relate to exposure levels and effects in other species. A study on common loons (*Gavia immer*) found that 3.0 $\mu\text{g/g}$ in adult blood tissue resulted in increased reproductive failure (Evers et al., 2008). A study on tree swallows (*Tachycineta bicolor*) using blood-egg pairings determined a conservative blood Hg lowest observed adverse effect level (LOAEL) at 0.63 $\mu\text{g/g}$ (Jackson, 2011). Jackson et al. (2011) also found that a blood Hg concentration of 0.70 $\mu\text{g/g}$ was associated with a 10% reduction in the reproductive success of Carolina wrens (*Thryothorus ludovicianus*). Studies have also shown that Hg at low concentrations can have sublethal effects which may impact overall health. A recent study in Spain examined the effect that low concentrations of Hg and other heavy metals had on the antioxidant system in Eurasian eagle owls (*Bubo bubo*). They found that blood Hg concentrations as low as 3 $\mu\text{g/dl}$ increased lipid peroxidation by 102% (Espín et al., 2014). While eider blood Hg values from all capture locations in this paper fall below the reported LOAEL for loons, several individuals at the Plum Island Sound, MA capture site are well above known LOAELs for swallows, wrens, and eagle owls. It is uncertain how levels of exposure and physiological sensitivity to Hg accumulation may differ among these species, but it is possible that eiders accumulating high levels of Hg within Plum Island Sound may be negatively impacted by this exposure.

In the Parker River watershed, within Parker River National Wildlife Refuge in Massachusetts adjacent to Plum Island Sound, elevated Hg levels have been reported in saltmarsh sparrows (*Ammodramus caudacutus*) and various species of shorebirds (Lane et al., 2011). This particular area receives freshwater input from the Parker and Ipswich rivers, both of which are likely to carry Hg-contaminated waters from inland watersheds towards the coast (Evers et al., 2007). The Ipswich River drains a large watershed that includes suburbs of Boston. While not a direct

freshwater input to Plum Island Sound, the Merrimack River, whose mouth drains to the ocean at the north end of Plum Island, flows through highly urbanized and industrialized areas of southern New Hampshire and northeastern Massachusetts (Lane et al., 2011). This is a likely reason for the elevated Hg concentrations in eiders captured in Plum Island Sound. Future studies should expand the sampling area to locations surrounding the sound to examine differences in micro-habitat Hg concentrations. Sampling birds and mussels in various locations inside the sound, on the ocean side of the island, and at the mouth of the Merrimack River could further delineate spatial variations in Hg availability in the area while potentially identifying local contaminant point sources.

4.2. Mercury in prey items

In this study, snail Hg concentrations in Massachusetts were significantly higher than in Maine (Fig. 4); however, both are slightly lower than the mean background level of 0.02 µg/g (ww) reported in a study in the Bay of Fundy, Canada (Legrand et al., 2005). Mussels collected and sampled at Plum Island Sound in Massachusetts were significantly higher in Hg than those collected in Pine Point, Maine, USA (Fig. 3). Gulfwatch, a long-term marine contaminants monitoring program focused within the Gulf of Maine, uses the blue mussel as its bioindicator species. Mercury concentrations in blue mussels at various long-term monitoring stations in the Gulf of Maine provide comparison data for mussels collected in this study. When compared to the most recent Gulfwatch data, Hg concentrations in mussels collected in Maine for this study were within or below the range of those at their monitoring sites (range: 0.086–0.399 µg/g, dw; LeBlanc et al., 2009). Comparison between the Massachusetts and Maine sites suggests that not only waterfowl species, but also their prey items, are accumulating Hg from somewhere in the Plum Island Sound environment. It should be noted that the mean soft tissue weight of mussels collected in Massachusetts was more than one and a half times that of the mean soft tissue weight of Maine mussels. Several studies have attempted to identify the relationship between mussel size and trace metal concentrations but current literature generally lacks consensus on the dynamics of this relationship. Riget et al. (2000) as well as Burger and Gochfeld (2006) found positive correlations between mussel shell length and Hg concentrations, whereas other studies (Saavedra et al., 2004; Borchardt et al., 1988; Andersen et al., 1996) found other factors, such as soft tissue weight and body condition, may be more accurate predictors of metal concentrations. We found both shell length and soft tissue weight to be significant predictors of Hg concentrations, but the correlation in both instances was weak, suggesting that Hg concentrations in mussels are affected by several interacting factors. Studies have cited that sampling season and location (Cain and Luoma, 1990), concentrations of metals in the immediate environment (Popham and D'Auria, 1983), and variations in uptake rates (Strong and Luoma, 1981) may all play a role in influencing heavy metal concentrations. Metal concentrations in mussels are dependent on a variety of environmental and physiological factors, thus comparison among studies warrants caution.

While Hg values in mussels collected at Plum Island Sound were higher than those collected in Maine, both sites are within the range of background levels reported in current literature and lower than those levels from which a point-source contaminant site was known (Garron et al., 2005). A study in New Brunswick, Canada found mean Hg levels of 1.40 µg/g (ww) at various locations surrounding a mercury cell chlor-alkali plant, which has been identified as a known point-source in the area. Mussels collected approximately 12 km away had a mean Hg concentration of 0.02 µg/g (ww) (Garron et al., 2005). Airas et al. (2004) reported mean Hg concentrations of between 0.01 and 0.03 µg/g (ww) in

mussels from Norway. Another study in Canada reported Hg concentrations in mussels ranging from 0.01 to 0.04 with a mean of 0.02 µg/g (ww) (Legrand et al., 2005).

5. Conclusions

We reported Hg concentrations in eiders and prey items from Plum Island Sound, MA that were significantly higher than those sampled in other locations throughout the study. Blood Hg concentrations in eiders from Plum Island Sound were also nearly three times higher than background blood Hg concentrations reported for common eiders in published literature. These higher levels exceed concentrations thought to be associated with sublethal effects in other wild bird species.

Consistent relationships were observed between Hg concentrations in eiders and blue mussels at sampling locations in both Plum Island Sound, MA and Pine Point, ME tentatively suggesting a predictive quality between the two. While significant in two areas in this study, this relationship should be interpreted with caution until further studies can validate its utility across broader geographic ranges and with greater sample sizes.

The results of this study underline the increased importance of determining effect level thresholds for common eiders and other sea duck species. Without such data, it is difficult to determine the relevance of, or risk associated with, elevated Hg concentrations in different sea duck habitats. We recommend more intensive sampling of eiders and associated prey items at several locations within Plum Island Sound in order to explore Hg exposure trends and spatial variation within the sound. Sampling eiders outside of the sound in nearby areas will also help determine spatial exposure patterns. In future studies, emphasis should be put on determining key biomarkers and adverse effect level thresholds for Hg in eiders and other sea ducks. Future studies may also explore trophic pathways and feeding behavior to help determine why eiders appear to be accumulating Hg more readily than other species with similar dietary preferences (i.e. scoters).

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