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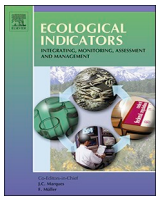

Short Note

Mercury concentrations provide an indicator of marine foraging in coastal birds

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ABSTRACT

The transfer of nutrients between marine and terrestrial systems has important ecological consequences, and animal movement is an important driver of nutrient transfer. Coastal birds forage in marine environments and breed in terrestrial habitats, and thus serve as vectors moving nutrients from the sea to the land. However, urbanization can influence the extent to which coastal birds forage in marine or terrestrial environments due to the availability of human subsidies. Establishing a reliable and straightforward indicator of marine foraging would be useful for assessing changes in the use of terrestrial vs. marine habitats in the face of urbanization and broader environmental change, and for understanding the flow of nutrients and energy between terrestrial and marine environments. Mercury (Hg) is a highly toxic heavy metal which bioaccumulates in marine food webs, and is a potential indicator of marine foraging. Methylmercury (MeHg), is present only in aquatic ecosystems and reaches elevated concentrations in the prey species of marine birds. Thus, high concentrations of MeHg would be expected for birds foraging in marine environments in comparison to those foraging on terrestrial sources. We hypothesized that the degree of marine foraging influences Hg uptake in coastal birds. To test this hypothesis, we combined GPS tracking data with measurements of Hg concentrations in the blood of herring gulls (*Larus argentatus*) along an urban gradient in the northeast United States. We examined Hg concentrations for 51 individual herring gulls tracked with GPS tags at study sites representing high, medium and low degrees of urbanization. Our results showed a strong and significant positive relationship between Hg concentrations and the proportion of herring gull foraging locations occurring in offshore waters. Hg concentrations differed significantly between herring gulls whose primary foraging habitat occurred in marine vs. terrestrial environments. Gulls in more urban colonies spent less time foraging in marine environments, and had significantly lower Hg concentrations than those at the more remote study. Our results suggest that Hg concentrations in blood can be used to reflect the extent of marine foraging for animals using both marine and terrestrial habitats. Hg concentrations could be valuable monitoring tool to assess how the use of marine foraging habitats changes through time (dietary shifts) or relative to environmental change such as urbanization.

1. Introduction

The transfer of nutrients and energy across terrestrial and aquatic boundaries can have important effects on community dynamics and ecosystem function (Polis and Strong, 1996; Huxel et al., 2002; Takimoto et al., 2002; Greig et al., 2012; Doughty et al., 2016). Animal movement greatly facilitates nutrient transfer between terrestrial and marine systems, with seabirds, sea turtles, anadromous fish, pinnipeds, and brown bears playing an important role in moving nutrients from the sea to the land (Cederholm et al., 1999; Hilderbrand et al., 1999; Anderson and Polis, 1999; Bouchard and Bjorndal, 2000; Fariña et al., 2003; Matsubayashi et al., 2015; Doughty et al., 2016). Urbanization is rapidly expanding over natural habitats and is influencing factors that drive nutrient transfer, including the movement and foraging behavior of a range of different coastal predators (Hobson and Stirling, 1997; Hebert et al., 1999; Matsubayashi et al., 2015; Deacy et al., 2017; Fuirst et al., 2018). For example, seabirds transport nutrients from marine prey to their terrestrial breeding colonies, and have marked effects on nutrient levels in proximate terrestrial ecosystems (Duda et al., 2020). In some cases, soil phosphorous concentrations have been found to be

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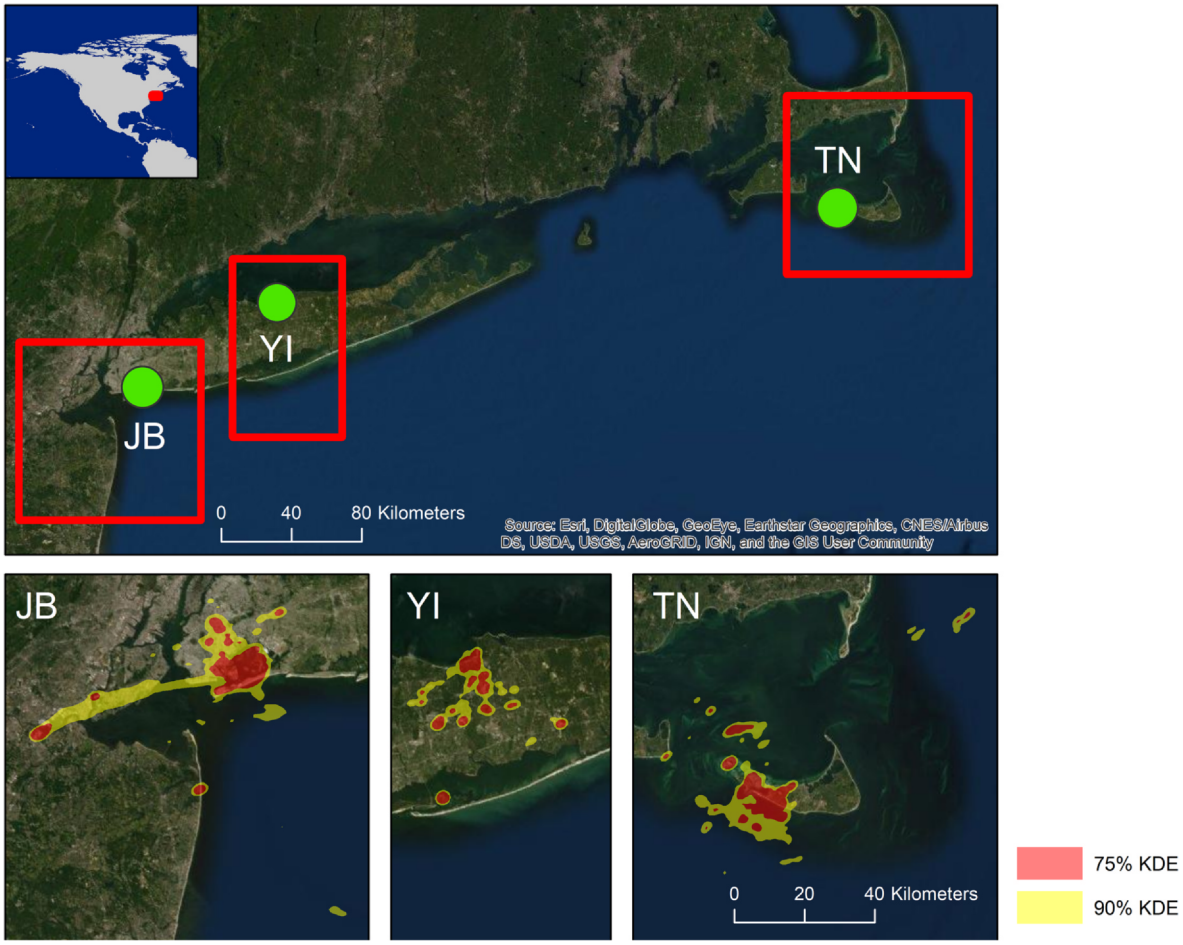

Fig. 1. Location of study colonies in Jamaica Bay (JB) in New York City, Young's Island (YI) on Long Island, and Tuckernuck Island (TN) in Massachusetts representing high, medium, and low degrees of urbanization, respectively. The lower panel shows Kernel Density Estimates (KDEs) of herring gull tracks at each of the study colonies.

more than twice that of islands where seabirds were absent (Anderson and Polis, 1999; Mulder et al., 2011). In urban environments, coastal birds forage in easily accessible and predictable human habitats, and have shown an increased reliance on human subsidies over the past 100 years (Auman et al., 2011; Hobson et al., 2015; Osterback et al., 2015; Fuirst et al., 2018). Decreases in the use of marine prey due to increased urbanization would have important consequences on nutrient cycling, and the implications for energy and nutrient flux between marine and terrestrial ecosystems merits further attention. For example, plants have lower phosphorous content than meat/ fish (D'Alessandro et al., 2015), and corn is a major component of human diets and manufactured foods, which often make up human refuse. Thus birds feeding on human refuse would be expected to transfer less phosphorous to breeding colonies than those feeding on fish in natural habitats. Quantifying the effects of urbanization on diet is challenging as it requires comparisons of food types in both urban and non-urban environments, but is a necessary first step in understanding implications for nutrient cycling. Reliable indicators of marine foraging will be useful for assessing changes in the use of terrestrial vs. marine habitats in the face of environmental change and urbanization, and for understanding the flow of nutrients and energy between terrestrial and marine environments.

Mercury (Hg) is a global pollutant that is emitted into the environment primarily through the burning of fossil fuels (Joensuu, 1971; Hutton and Symon, 1986; Jackson, 1997; Pacyna et al., 2010; Outridge et al., 2018). It is fixed from its inorganic forms into its bioaccumulative form, monomethylmercury (MeHg), by various anaerobic unicellular organisms (Furutani and Rudd, 1980; Morel et al., 1998; Gilmour et al., 2013). MeHg is ubiquitous in the marine realm and upon its entry at the bottom of the pelagic food web, it biomagnifies, reaching high concentrations in upper trophic level species (Gilmour et al., 1992; Hall et al., 1997; Bargagli et al., 1998; Jæger et al., 2009; Lehnherr, 2014; Ruus et al., 2015). Seabirds are broadly used to monitor pollutants such as Hg in marine systems since they are top predators, and as long-lived and wide-ranging species, they integrate pollutant levels over broad areas and timescales (Furness, 1993; Furness and Camphuysen, 1997; Burger and Gochfeld, 2004; Elliott and Elliott, 2013). In addition to transporting nutrients, seabirds can act as vectors transporting toxic metals from the sea to land (Headley, 1996; Liu et al., 2006; Mallory et al., 2015). Various seabird species occurring in different regions of the world and using diverse foraging strategies show Hg levels that are elevated above background levels (e.g., Thompson et al., 1992; Burger and Gochfeld, 2000; Braune et al., 2001). Diet type directly influences Hg concentrations in marine predators, including birds (Stewart et al., 1997; Monteiro et al., 1998; Thompson et al., 1998a, 1998b; Bond and Diamond, 2009), given that Hg is entirely diet derived. Moreover, differences in isotopic composition of Hg that accumulated in bird eggs reflects the degree of aquatic vs. terrestrial diet of the parent (Hebert, 2019).

Here, we argue for the application of Hg as a tracer of the habitat on a fine scale. This application is based on the well demonstrated enrichment of MeHg in aquatic organisms, which is negligible in terrestrial organisms due to very low levels of methylation of Hg in terrestrial environments (Rudd, 1995). Thus, significantly higher concentrations

of MeHg are expected in pelagic marine consumers in comparison to those foraging on terrestrial sources, particularly for piscivorous birds. We assess the hypothesis that concentrations of total Hg in blood positively correlate with the degree of foraging in marine habitats, using herring gulls (*Larus argentatus*) as a case study. Herring gulls present a compelling study model for examining how Hg relates to predator foraging behavior since they are abundant and easily studied, show highly plastic foraging behavior, forage in both marine and terrestrial environments, and have a broad geographic distribution allowing for spatial comparisons. Previous studies have demonstrated that gulls show considerable variability in their use of urban vs. marine environments, with gulls in close proximity to urban areas predominantly foraging in human habitats and gulls in less urbanized areas foraging in marine habitats (Fuirst et al., 2018). We predict that individual gulls foraging in marine environments will have higher Hg concentrations than those foraging in human habitats. Further, we predict that these trends will be evident at the colony level, with gulls in more urban colonies foraging less frequently in marine habitats and showing lower Hg concentrations.

2. Methods

2.1. Study area

We examined herring gull foraging behavior in relation to blood concentrations of Hg at three study colonies that differed in their degree of urbanization, as reflected by population densities, and in the use of human habitats by foraging gulls (Fuirst et al., 2018; Fig. 1). Following Fuirst et al. (2018), the study site in Jamaica Bay (JB) within New York City represented a high degree of urbanization, while the study sites on Youngs Island in Stony Brook Harbor on Long Island, New York (YI) and on Tuckernuck Island (TN) off the coast of Nantucket in Massachusetts represented medium and low degrees of urbanization, respectively.

2.2. GPS tracking

We sought to compare quantifiable metrics of foraging behavior with mercury concentrations in gulls, and analyzed GPS tracks to assess herring gull use of marine vs. terrestrial foraging areas. Assessing GPS tracks is advantageous for examining foraging behavior in that it allows for direct and continuous examination of a bird's position and allows multiple trips to be observed for a specific individual over consecutive days. Other techniques are limiting; for example, the use of regurgitates or pellets to examine prey species are biased towards recent meals, are labor intensive to collect and require regularly disturbing the colony. Further, analyses of regurgitates may underestimate small, soft prey items (Ramos et al., 2009), and it can be difficult to assign pellets to specific individuals from which blood samples were taken for Hg analyses (described below).

We obtained GPS tracks and blood samples from a total of 51 herring gulls from late April to late June in 2016, 2017 and 2018 during incubation (22 at JB, 15 at YI and 14 at TN, respectively). We tracked gulls using iGotU (Mobile Action Technology, Taiwan) and CatTrack Gen2 GPS tags (Catnip Technologies, Hong Kong). While the majority of tags were programmed to recorded spatial locations at a 2 min sampling interval, 4 tags recorded at a 30 s interval and were resampled to a 2 min interval prior to analyses. GPS tags were sealed in heat-shrink tubing and attached to 3–4 central tail feathers with medical grade adhesive tape (Tesa Tape Inc., Charlotte NC). Birds were captured twice: once during tag deployment and once during tag recovery.

2.3. Blood sampling

Blood samples (2 mL) were obtained from the brachial or medial metatarsal veins using a 25 gauge syringe upon recapture of individual gulls so as to reflect the period when the bird was tracked. We examined Hg concentrations in whole blood, which reflects Hg incorporated from the diet over short time scales during blood formation (Anderson et al., 2009). Blood samples were stored in a −20 freezer within 12 h of collection and transferred to a −80 freezer for long-term storage (maximum of 10 days later for TN samples).

Animal capture, handling and sampling procedures were reviewed and approved by the Institutional Animal Care and Use Committee (IACUC) at Stony Brook University (IACUC #: 2016-2247 -Bi −4.19.19-R1) and were approved by relevant state and federal agencies (Federal Bird Banding Permit #22795, New York State Department of Environmental Conservation Permit 2035). Access to the Jamaica Bay Wildlife Refuge and Young's Island were approved by the National Park Service (Permit: GATE-2017-SCI-0020) and the New York State Department of Environmental Conservation (TRP:2018–001).

2.4. Movement analyses

We identified foraging locations along herring gull GPS tracks by identifying regions of Area-Restricted Search (ARS) using First Passage Time (FPT) and identifying the habitat type of ARS locations as in Fuirst et al. (2018). Briefly, we identified the scale of ARS by locating the peak in the variance of log(FPT), and assessed FPT at this scale (Fauchald and Tveraa, 2003). FPT data were subsampled to reduce autocorrelation, and the upper quartile of values from resulting FPT data were used to represent foraging points as in Suryan et al. (2006). We then used satellite imagery in ArcGIS (ESRI World Imagery May 2017) to identify the habitat type where ARS points occurred for each foraging trip, with habitats designated as terrestrial or marine. A small number of points (approximately 3% of all foraging locations) occurred in estuarine habitats, and these points were classified as marine. For each gull, the proportion of marine habitat use was assessed as the mean proportion of ARS points occurring in marine habitats across all foraging trips. We identified the primary habitat type used, defined as the habitat type in which the majority of ARS points occurred.

To ensure that habitat use in marine vs. terrestrial environments was adequately represented by GPS tracks, we assessed individual variability in habitat preference for birds tracked for at least 10 foraging trips (Table 1). We then assessed the mean proportion of marine foraging after each trip and assessed how many trips were required for this mean to be within 5% of the mean for all trips combined. We found that on average it took 5.2 foraging trips for the mean proportion of marine foraging to be within 5% of the overall mean for all trips required, and therefore used only data from birds tracked for at least 6 foraging trips for further analyses.

2.5. Mercury analyses

Total Hg analysis was conducted by direct mercury analyzer MA 3000 (Nippon), through sample pyrolysis, amalgamation of Hg with gold, and detection by atomic absorption based on the method USEPA 7473. Concentrations of THg were determined based on 5-point standard curve prepared from external Hg standards (Wako) suspended in a 1 mg L^{-1} solution of L-Cysteine for stabilization. Standards were used when reached the room temperature. Moreover, certified reference material TORT-2 (average mass was 24 mg), was also analyzed for quality assurance every tenth sample. Blood samples were thawed shortly prior to analysis and kept on ice. Blood samples were pipetted into purged ceramic sample boats and their weights were recorded. Masses of wet blood used for THg analysis ranged from 17.3 to 110.6 mg with a median of 99.3 mg. To determine the analytical error on sample analyses, every tenth sample was triplicated or duplicated depending on available volume of blood sample. Therefore, THg concentrations generated through this analysis were on wet weight basis. Select samples were analyzed in duplicate generating errors of < 2%. The analysis of the certified reference material TORT-2 resulted in a range of concentrations from 0. 295–0.311 mg kg^{-1} (certified THg

Table 1
Summary of tag deployments for herring gulls. Prop. Marine and Prop. Terrestrial represent the proportion of foraging locations occurring in each habitat type, respectively.

Tag ID	Date of deployment	Duration of tag deployment (days)	No. foraging trips	Prop. Aquatic	Prop. Terrestrial
JB100a	2016-05-17	13.04	36	0.24	0.76
JB102a	2016-05-18	3.28	5	0.32	0.68
JB104b	2017-05-19	3.76	7	0.00	1.00
JB106a	2016-05-17	5.58	8	0.21	0.79
JB109b	2017-05-16	7.13	12	0.02	0.98
JB113a	2016-05-17	5.16	9	0.07	0.93
JB205a	2017-05-10	5.31	12	0.10	0.90
JB205b	2017-05-19	14.03	44	0.41	0.59
JB206a	2017-05-10	3.22	4	0.56	0.44
JB209a	2017-05-09	9.61	14	0.12	0.88
JB212a	2017-05-18	0.96	1	0.00	1.00
JB214a	2018-05-17	3.65	6	0.14	0.86
JB217a	2018-05-17	3.70	9	0.15	0.85
JB222a	2017-05-05	4.12	7	0.35	0.65
JB227b	2018-05-17	3.94	11	0.58	0.42
JB230c	2018-05-17	4.00	13	0.61	0.39
JB237b	2018-05-17	4.27	12	0.00	1.00
JB239b	2018-05-17	3.34	3	0.17	0.83
JB240b	2018-05-17	3.96	9	0.04	0.96
JB302a	2018-05-15	6.05	7	0.04	0.96
JB304a	2018-05-14	5.83	7	0.33	0.67
JB305a	2018-05-15	14.21	27	0.44	0.56
TN109a	2016-05-26	3.76	4	0.88	0.12
TN115a	2016-05-26	3.96	6	0.80	0.20
TN117a	2016-05-26	2.62	6	1.00	0.00
TN208a	2017-06-01	1.06	1	1.00	0.00
TN210b	2017-05-28	5.01	8	0.52	0.48
TN230b	2017-06-12	4.36	8	0.94	0.06
TN231b	2017-06-17	4.48	16	0.70	0.30
TN233b	2017-05-30	3.85	6	0.51	0.49
TN234a	2017-05-29	4.27	5	0.50	0.50
TN235a	2017-05-29	2.99	4	0.37	0.63
TN237c	2018-05-31	0.92	2	0.97	0.03
TN239c	2018-05-31	0.84	2	0.73	0.27
TN240b	2017-06-14	2.03	2	0.56	0.44
TN302b	2018-05-27	5.30	8	0.85	0.15
YI103b	2017-05-15	5.54	7	0.00	1.00
YI117a	2017-05-15	5.66	5	0.00	1.00
YI206b	2017-05-21	11.31	16	0.22	0.78
YI210a	2017-05-15	5.75	7	0.00	1.00
YI211a	2017-05-11	4.15	10	0.41	0.59
YI227b	2017-05-25	0.91	4	0.00	1.00
YI228a	2017-05-11	4.12	6	0.20	0.80
YI229a	2017-05-21	4.12	2	0.08	0.92
YI232a	2017-05-11	4.04	7	0.00	1.00
YI236a	2017-05-11	3.96	8	0.00	1.00
YI237a	2017-05-18	4.51	8	0.13	0.88
YI239a	2017-05-11	4.18	3	0.00	1.00
YI301a	2018-05-08	7.12	15	0.04	0.96
YI306a	2018-05-18	4.90	11	0.01	0.99
YI307a	2018-05-18	3.89	5	0.00	1.00

concentration in TORT-2: 0.26 ± 0.06 mg kg^{-1} dry weight; mean ± standard deviation).

2.6. Statistical analyses

For each herring gull tracked, we assessed the proportion of foraging locations occurring in terrestrial vs. marine environments and the proportion of foraging trips that used these habitat types. To examine how Hg concentrations varied with the use of marine habitats, we pooled data from all three study sites in order to compare Hg concentrations in gulls using a range of habitat types. To assess the hypothesis that individual gulls foraging in marine environments show higher Hg concentrations, we performed a linear regression to examine how Hg concentrations were affected by the proportion of foraging

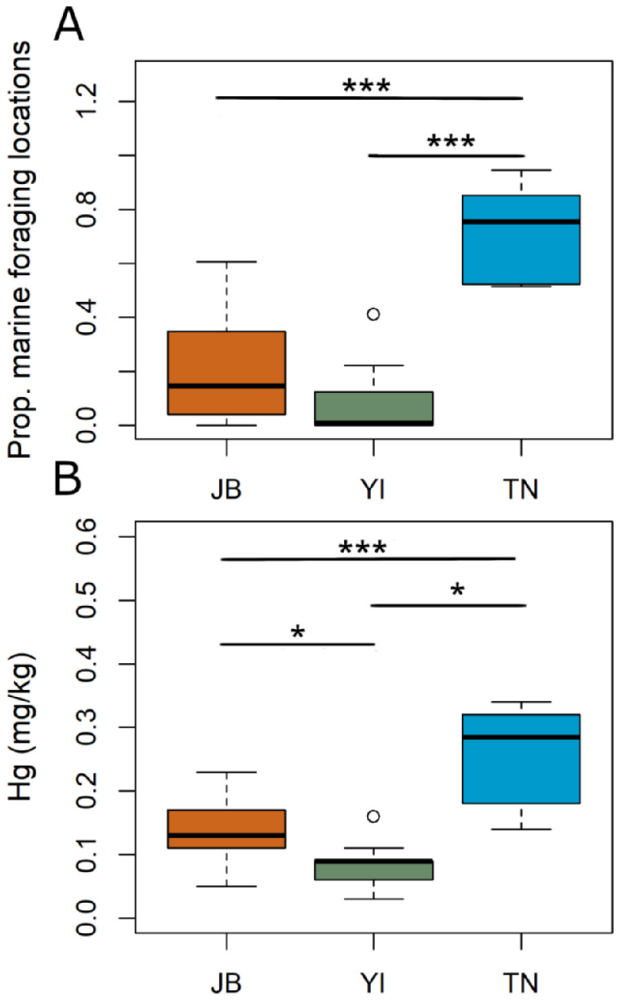

Fig. 2. Colony means of the use of marine environments (A) and blood mercury concentrations (B) in herring gulls. Asterisks reflect significant differences with a Bonferroni correction (* reflects p values < 0.05, *** reflects p values < 0.0001).

locations occurring in marine habitats, using the mean proportion across all foraging trips for all individuals. We assessed differences in Hg concentrations relative to the primary habitat type used by each gull using a Wilcoxon test. Lastly, we examined colony-level differences in both the use of marine environments and Hg concentrations in gulls using Kruskal-Wallis tests with post-hoc Dunn's tests with a Bonferroni correction. All statistical analyses were performed in program R statistical software (v. 3.3.2, R Core Team, 2016).

3. Results

GPS tag deployments ranged from 0.84 to 14.21 days (mean of 5.2 days; Table 1), and individual gulls typically took 2 to 3 foraging trips per day. Gulls at all three study colonies foraged in both marine and terrestrial habitats, though in variable proportions. Gulls at JB and YI predominantly foraged in terrestrial environments (e.g. public parks, landfills, urban centers, and grasslands), while gulls at TN predominantly foraged in offshore waters (Figs. 1 and 2). Gulls at all colonies showed a high level of individual specialization, and low variability in their habitat use of marine vs. terrestrial habitats between trips (Supplementary Fig. 1). The proportion of birds foraging exclusively in terrestrial environments (i.e., no marine foraging locations observed during any foraging trips) was 11%, 40% and 0% for JB, YI and TN, respectively, for birds tracked for at least 6 foraging trips. In contrast, 14% of birds at the TN colony foraged only in marine habitats, while no

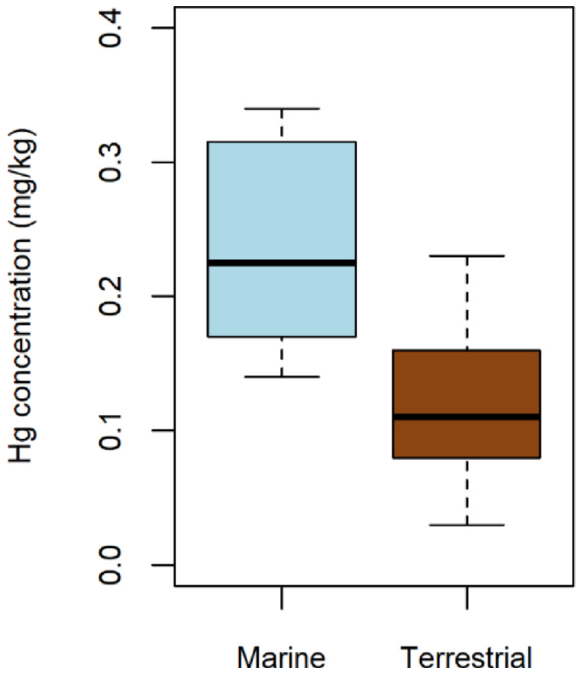

Fig. 3. Hg concentration (mg/kg) in whole blood of herring gulls by primary habitat type. Gulls whose primary foraging habitat occurred in marine vs. terrestrial differed significantly in their blood Hg concentrations (Wilcoxon test, $p = 6.90 \times 10^{-4}$).

birds at the JB or YI colonies foraged exclusively in marine habitats. All birds at TN used offshore habitats for foraging. The proportion of foraging locations in marine habitats differed significantly between colonies (Kruskal-Wallis test, $p = 4.32 \times 10^{-4}$), with gulls at TN foraging significantly more often in marine habitats than those at JB or YI (Dunn's post hoc tests with Bonferroni correction, $p = 5.5 \times 10^{-3}$ and 2.8×10^{-4}, respectively; Fig. 2). Similarly, blood Hg concentrations differed significantly between the three herring gull colonies (Kruskal-Wallis test, $p = 4.30 \times 10^{-4}$), with TN showing significantly higher Hg concentrations than JB or YI and YI showing significantly higher Hg concentrations than JB (Dunn's post hoc test with Bonferroni correction, $p = 3.8 \times 10^{-2}$ for TN vs. JB; 2.7×10^{-4} for TN vs. YI and 3.8×10^{-2} for YI vs. JB, respectively; Fig. 2).

Hg concentrations differed significantly between herring gulls whose primary foraging habitat occurred in marine vs. terrestrial environments (Wilcoxon test, $p = 6.90 \times 10^{-4}$; Fig. 3). On an individual level, blood Hg concentrations showed a high degree of variability in herring gulls, ranging from 0.02 mg/kg to 0.43 mg/kg. We found a strong and significant positive relationship between Hg blood concentrations and the proportion of herring gull foraging locations occurring in offshore waters for individual gulls (linear regression, $R^2 = 0.453$, $p = 1.08 \times 10^{-5}$; Fig. 4).

4. Discussion

Our hypothesis that differences in foraging behavior influence diet and ultimately Hg uptake in coastal birds was upheld, and our results suggest that Hg could serve as an indicator of marine foraging in coastal predators. We quantified individual variability in Hg concentrations relative to movement patterns in gulls, and showed that higher Hg concentrations reflect increased time spent foraging in marine environments. Moreover, gulls in more urban colonies spent less time foraging in marine environments, and had significantly lower Hg concentrations than those at the more remote study colony. Our results suggest that Hg concentrations in blood can be used to reflect the extent of marine prey in the diet for animals that forage in both marine and terrestrial habitats. By reflecting the degree to which birds forage in the water vs. on land, Hg concentrations in the tissues of coastal birds could serve as a valuable component of studies investigating the movement of energy and nutrients between marine and terrestrial systems (Peterson et al., 2017). Monitoring Hg concentrations could provide a useful tool to assess how the use of marine foraging habitats changes through time (dietary shifts) or relative to environmental change such as urbanization (English et al., 2020).

We examined Hg concentrations in blood and compared these concentrations with foraging patterns evaluated over 5.9 days, on average. Different avian tissues reflect Hg exposure over different temporal scales, and it is therefore important to consider biomonitoring objectives when selecting avian tissues to sample (Evers et al., 2005). The organic form of Hg, MeHg, which seabirds assimilate from their diet, can cycle in the blood stream for a few weeks to months before being eliminated through the feathers and excreta, as well via the eggs in females (Braune and Gaskin, 1987). Demethylation of MeHg in the liver has been hypothesized in birds. MeHg detoxification may occur via its binding with selenides which results in the formation of HgSe, an inert nontoxic compound (Thompson and Furness, 1989; Kim et al., 1996; Arai et al., 2004). Thus, while tissues such as liver and feathers reflect long-term Hg exposure, blood reflects dietary uptake over shorter time scales. Lab-based studies of seabirds have demonstrated that Hg exposure through the diet is reflected in blood Hg concentrations within days, with a half-life of approximately 30–60 days (Bearhop et al., 2000; Monteiro and Furness, 2001). We therefore argue that a study of blood chemistry, including Hg concentrations, is particularly useful for studies of foraging patterns, both because Hg concentrations in blood reflect short-term exposure, and because blood can be obtained without lethal research. Studies of Hg in the liver, kidney or muscle of birds rely on lethal research to obtain specimens.

While blood is the best tissue for reflecting Hg exposure over short time scales (Evers et al., 2005; Ackerman et al., 2008), we observed a wide range of Hg concentrations for birds that did not show any foraging locations within marine habitats (Fig. 4). This discrepancy suggests that Hg blood chemistry might be impacted over longer time periods than was represented in the GPS tracks. Birds that displayed higher Hg concentrations in their blood may have foraged in marine environments shortly before they were tagged. While we found strong relationships between the proportion of foraging locations in marine habitats and blood Hg concentrations, longer tracking periods would be preferable for studies examining changes in marine foraging patterns, and could result in stronger relationships between habitat use and blood Hg concentrations.

Our study focused on sites within New York and Massachusetts in regions that have not been impacted by point source Hg pollution, and we are therefore confident that our results were not impacted by site-specific pollution. However, it is important to note that studies seeking to use Hg as an indicator of marine foraging should eliminate sites of known historical Hg pollution such as sites of industrial and chemical manufacturing. Spatial and temporal patterns of Hg contamination could influence Hg concentrations in marine prey, and it is therefore important to design studies with consideration of these factors. For example, point sources of Hg contamination have resulted in elevated Hg concentrations in forage fish in coastal systems (Gehrke et al., 2011). In New York, Hg concentrations are elevated in the Hudson River watershed. Concentrations of Hg in fish in the Hudson River have declined significantly since 1970, likely due a decrease in mercury released into the Hudson River watershed by factories, along with a decline in atmospheric deposition of Hg transported over a long range from the Midwest (Levinton and Pochron, 2008). These examples demonstrate that both the foraging location as well as its timing can greatly impact Hg concentrations in the tissues of aquatic predators.

Other chemical markers of diet can distinguish foraging habitats, and Stable Isotope Analysis (SIA) of tissues in particular is commonly used to assess broad trends in the diet of birds and other coastal

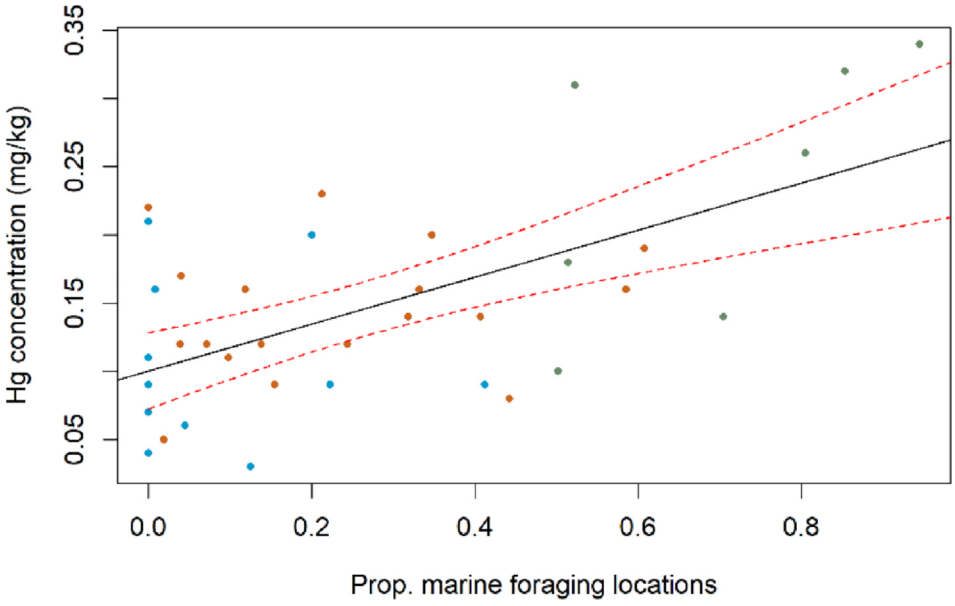

Fig. 4. Linear regression of Hg concentration (mg/kg) in whole blood of individual herring gulls on the proportion of gull foraging locations occurring in marine habitats (R^2 = 0.453, $p = 1.08 \times 10^{-5}$). Red dashed lines show the 95% confidence interval. Orange circles represent gulls foraging from Jamaica Bay, while green and blue circles represent gulls foraging from Youngs Island and Tuckernuck, respectively.

predators. Stable isotopes of hydrogen and sulfur have been used to discriminate between the use of marine and terrestrial systems in foraging predators as terrestrial foodwebs typically have lower values of δ^2H and δ^{34}S than marine foodwebs (Ramos et al., 2009; Hobson et al., 2015). However, δ^{34}S may not effectively discriminate between terrestrial and freshwater sources, and interpreting δ^2H data in terms of dietary shifts between marine and terrestrial habitats can be complex as our understanding of variability of δ^2H in marine food webs is limited (Hobson et al., 2015). Our results suggest Hg as an alternative indicator of marine foraging in coastal predators. Both SIA and analyses of Hg concentrations in animal tissues offer considerable advantages over traditional diet analyses in that they can provide rapid assessments of foraging ecology, and provide an integrated measure of diet over weeks (Hobson et al., 1994; Bond and Jones, 2009; Ramos et al., 2009). Since Hg does not greatly accumulate in terrestrial organisms due to the absence of its source in the form of bioavailable methylated Hg (MeHg), its detection in blood infers some degree of foraging on aquatic diet. Its application as an indicator of marine foraging is therefore most useful when the study organisms use both marine and terrestrial habitat and when the significance of one vs. the other is in question. In these cases, higher Hg concentrations will indicate a higher degree of marine foraging in comparison to individuals with lower blood Hg concentrations. Therefore, Hg may be a particularly useful indicator of marine foraging.

5. Conclusions

Understanding foraging patterns of coastal predators is becoming increasingly important under continued urbanization and anthropogenic change. Our results demonstrate that Hg concentrations can be used to assess differences in the foraging patterns of coastal birds at both the individual and colony levels. Hg provides a promising tool for identifying the degree of aquatic foraging, and thus for assessing changes in the use of aquatic foraging habitats through time.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

Ackerman, J.T., Eagles-Smith, C.A., Takekawa, J.Y., Bluso, J.D., Adelsbach, T.L., 2008. Mercury concentrations in blood and feathers of prebreeding Forster's terns in relation to space use of San Francisco Bay, California, USA, habitats. Environ. Toxicol. Chem.: Int. J. 27, 897–908.
Anderson, O., Phillips, R., McDonald, R., Shore, R., McGill, R., Bearhop, S., 2009. Influence of trophic position and foraging range on mercury levels within a seabird community. Mar. Ecol. Prog. Ser. 375, 277–288.
Anderson, W.B., Polis, G.A., 1999. Nutrient fluxes from water to land: seabirds affect plant nutrient status on Gulf of California islands. Oecologia 118, 324–332.
Arai, T., Ikemoto, T., Hokura, A., Terada, Y., Kunito, T., Tanabe, S., Nakai, I., 2004. Chemical forms of mercury and cadmium accumulated in marine mammals and seabirds as determined by XAFS analysis. Environ. Sci. Technol. 38, 6468–6474.
Auman, H.J., Bond, A.L., Meathrel, C.E., Richardson, A.M., 2011. Urbanization of the silver gull: evidence of anthropogenic feeding regimes from stable isotope analyses. Waterbirds 34, 70–76.
Bargagli, R., Monaci, F., Sanchez-Hernandez, J., Cateni, D., 1998. Biomagnification of mercury in an Antarctic marine coastal food web. Mar. Ecol. Prog. Ser. 169, 65–76.
Bearhop, S., Ruxton, G.D., Furness, R.W., 2000. Dynamics of mercury in blood and feathers of great skuas. Environ. Toxicol. Chem.: Int. J. 19, 1638–1643.
Bond, A.L., Diamond, A.W., 2009. Mercury concentrations in seabird tissues from Machias Seal Island, New Brunswick, Canada. Sci. Total Environ. 407, 4340–4347.
Bond, A.L., Jones, I.L., 2009. A practical introduction to stable-isotope analysis for seabird biologists: approaches, cautions and caveats. Marine Ornithol. 37, 183–188.
Bouchard, S.S., Bjorndal, K.A., 2000. Sea turtles as biological transporters of nutrients and energy from marine to terrestrial ecosystems. Ecology 81, 2305–2313.
Braune, B., Donaldson, G., Hobson, K., 2001. Contaminant residues in seabird eggs from the Canadian Arctic. Part I. Temporal trends 1975–1998. Environ. Pollut. 114, 39–54.
Braune, B.M., Gaskin, D.E., 1987. A mercury budget for the Bonaparte's gull during autumn moult. Ornis Scand. 244–250.
Burger, J., Gochfeld, M., 2000. Metal levels in feathers of 12 species of seabirds from Midway Atoll in the northern Pacific Ocean. Sci. Total Environ. 257, 37–52.
Burger, J., Gochfeld, M., 2004. Marine birds as sentinels of environmental pollution.

EcoHealth 1, 263–274.
Cederholm, C.J., Kunze, M.D., Murota, T., Sibatani, A., 1999. Pacific salmon carcasses: essential contributions of nutrients and energy for aquatic and terrestrial ecosystems. Fisheries 24, 6–15.
D'Alessandro, C., Piccoli, G.B., Cupisti, A., 2015. The "phosphorus pyramid": a visual tool for dietary phosphate management in dialysis and CKD patients. BMC Nephrol. 16, 1–6.
Deacy, W.W., Armstrong, J.B., Leacock, W.B., Robbins, C.T., Gustine, D.D., Ward, E.J., Erlenbach, J.A., Stanford, J.A., 2017. Phenological synchronization disrupts trophic interactions between Kodiak brown bears and salmon. Proc. Natl. Acad. Sci. 114, 10432–10437.
Doughty, C.E., Roman, J., Faurby, S., Wolf, A., Haque, A., Bakker, E.S., Malhi, Y., Dunning, J.B., Svenning, J.-C., 2016. Global nutrient transport in a world of giants. Proc. Natl. Acad. Sci. 113, 868–873.
Duda, M.P., Glew, J.R., Michelutti, N., Robertson, G.J., Montevecchi, W.A., Kissinger, J.A., Eickmeyer, D.C., Blais, J.M., Smol, J.P., 2020. Long-term changes in terrestrial vegetation linked to shifts in a colonial seabird population. Ecosystems 1–14.
Elliott, J.E., Elliott, K.H., 2013. Tracking marine pollution. Science 340, 556–558.
English, M.D., Robertson, G.J., O'Driscoll, N.J., Klapstein, S.J., Peck, L.E., Mallory, M.L., 2020. Variation in isotopic niche, digestive tract morphology, and mercury concentrations in two sympatric waterfowl species wintering in Atlantic Canada. FACETS 5, 393–408.
Evers, D.C., Burgess, N.M., Champoux, L., Hoskins, B., Major, A., Goodale, W.M., Taylor, R.J., Poppenga, R., Daigle, T., 2005. Patterns and interpretation of mercury exposure in freshwater avian communities in northeastern North America. Ecotoxicology 14, 193–221.
Fariña, J., Salazar, S., Wallem, K., Witman, J., Ellis, J., 2003. Nutrient exchanges between marine and terrestrial ecosystems: the case of the Galapagos sea lion *Zalophus wollebaecki*. J. Anim. Ecol. 72, 873–887.
Fauchald, P., Tveraa, T., 2003. Using first-passage time in the analysis of area-restricted search and habitat selection. Ecology 84, 282–288.
Fuirst, M., Veit, R.R., Hahn, M., Dheilly, N., Thorne, L.H., 2018. Effects of urbanization on the foraging ecology and microbiota of the generalist seabird *Larus argentatus*. PLoS One 13.
Furness, R.W., 1993. Birds as monitors of pollutants, Birds as monitors of environmental change. Springer 86–143.
Furness, R.W., Camphuysen, K., 1997. Seabirds as monitors of the marine environment. ICES J. Mar. Sci. 54, 726–737.
Furutani, A., Rudd, J.W., 1980. Measurement of mercury methylation in lake water and sediment samples. Appl. Environ. Microbiol. 40, 770–776.
Gehrke, G.E., Blum, J.D., Slotton, D.G., Greenfield, B.K., 2011. Mercury isotopes link mercury in San Francisco Bay forage fish to surface sediments. Environ. Sci. Technol. 45, 1264–1270.
Gilmour, C.C., Henry, E.A., Mitchell, R., 1992. Sulfate stimulation of mercury methylation in freshwater sediments. Environ. Sci. Technol. 26, 2281–2287.
Gilmour, C.C., Podar, M., Bullock, A.L., Graham, A.M., Brown, S.D., Somenahally, A.C., Johs, A., Hurt Jr, R.A., Bailey, K.L., Elias, D.A., 2013. Mercury methylation by novel microorganisms from new environments. Environ. Sci. Technol. 47, 11810–11820.
Greig, H.S., Kratina, P., Thompson, P.L., Palen, W.J., Richardson, J.S., Shurin, J.B., 2012. Warming, eutrophication, and predator loss amplify subsidies between aquatic and terrestrial ecosystems. Glob. Change Biol. 18, 504–514.
Hall, B., Bodaly, R., Fudge, R., Rudd, J., Rosenberg, D., 1997. Food as the dominant pathway of methylmercury uptake by fish. Water Air Soil Pollut. 100, 13–24.
Headley, A.D., 1996. Heavy metal concentrations in peat profiles from the high Arctic. Sci. Total Environ. 177, 105–111.
Hebert, C.E., 2019. The river runs through it: the Athabasca River delivers mercury to aquatic birds breeding far downstream. PLoS One 14.
Hebert, C.E., Shutt, J.L., Hobson, K.A., Weseloh, D.C., 1999. Spatial and temporal differences in the diet of Great Lakes herring gulls (*Larus argentatus*): evidence from stable isotope analysis. Can. J. Fish. Aquat. 56, 323–338.
Hilderbrand, G.V., Hanley, T.A., Robbins, C.T., Schwartz, C.C., 1999. Role of brown bears (*Ursus arctos*) in the flow of marine nitrogen into a terrestrial ecosystem. Oecologia 121, 546–550.
Hobson, K.A., Stirling, I., 1997. Low variation in blood δ13C among Hudson Bay polar bears: implications for metabolism and tracing terrestrial foraging. Mar. Mammal Sci. 13, 359–367.
Hobson, K.A., Piatt, J.F., Pitocchelli, J., 1994. Using stable isotopes to determine seabird trophic relationships. J. Anim. Ecol. 786–798.
Hobson, K.A., Blight, L.K., Arcese, P., 2015. Human-induced long-term shifts in gull diet from marine to terrestrial sources in North America's coastal Pacific: more evidence from more isotopes (δ2H, δ34S). Environ. Sci. Technol. 49, 10834–10840.
Hutton, M., Symon, C., 1986. The quantities of cadmium, lead, mercury and arsenic entering the UK environment from human activities. Sci. Total Environ. 57, 129–150.
Huxel, G.R., McCann, K., Polis, G.A., 2002. Effects of partitioning allochthonous and autochthonous resources on food web stability. Ecol. Res. 17, 419–432.
Jackson, T.A., 1997. Long-range atmospheric transport of mercury to ecosystems, and the importance of anthropogenic emissions A critical review and evaluation of the published evidence. Environ. Rev. 5, 99–120.
Jæger, I., Hop, H., Gabrielsen, G.W., 2009. Biomagnification of mercury in selected species from an Arctic marine food web in Svalbard. Sci. Total Environ. 407, 4744–4751.
Joensuu, O.I., 1971. Fossil fuels as a source of mercury pollution. Science 172, 1027–1028.
Kim, E., Murakami, T., Saeki, K., Tatsukawa, R., 1996. Mercury levels and its chemical form in tissues and organs of seabirds. Arch. Environ. Contam. Toxicol. 30, 259–266.
Lehnherr, I., 2014. Methylmercury biogeochemistry: a review with special reference to Arctic aquatic ecosystems. Environ. Rev. 22, 229–243.
Levinton, J., Pochron, S., 2008. Temporal and geographic trends in mercury concentrations in muscle tissue in five species of Hudson River, USA, fish. Environ. Toxicol. Chem. 27, 1691.
Liu, X., Zhao, S., Sun, L., Yin, X., Xie, Z., Honghao, L., Wang, Y., 2006. P and trace metal contents in biomaterials, soils, sediments and plants in colony of red-footed booby (Sula sula) in the Dongdao Island of South China Sea. Chemosphere 65, 707–715.
Mallory, M.L., Mahon, L., Tomlik, M.D., White, C., Milton, G.R., Spooner, I., 2015. Colonial marine birds influence island soil chemistry through biotransport of trace elements. Water Air Soil Pollut. 226, 31.
Matsubayashi, J., Morimoto, J.O., Tayasu, I., Mano, T., Nakajima, M., Takahashi, O., Kobayashi, K., Nakamura, F., 2015. Major decline in marine and terrestrial animal consumption by brown bears (*Ursus arctos*). Sci. Rep. 5, 1–8.
Monteiro, L.R., Furness, R.W., 2001. Kinetics, dose− response, and excretion of methylmercury in free-living adult Cory's shearwaters. Environ. Sci. Technol. 35, 739–746.
Monteiro, L., Granadeiro, J., Furness, R., 1998. Relationship between mercury levels and diet in Azores seabirds. Mar. Ecol. Prog. Ser. 166, 259–265.
Morel, F.M., Kraepiel, A.M., Amyot, M., 1998. The chemical cycle and bioaccumulation of mercury. Annu. Rev. Ecol. Syst. 29, 543–566.
Mulder, C., Jones, H., Kameda, K., Palmborg, C., Schmidt, S., Ellis, J., Orrock, J., Wait, A., Wardle, D., Yang, L., 2011. Impacts of seabirds on plant and soil properties.
Osterback, A.-M.-K., Frechette, D.M., Hayes, S.A., Shaffer, S.A., Moore, J.W., 2015. Long-term shifts in anthropogenic subsidies to gulls and implications for an imperiled fish. Biol. Conserv. 191, 606–613.
Outridge, P.M., Mason, R., Wang, F., Guerrero, S., Heimburger-Boavida, L., 2018. Updated global and oceanic mercury budgets for the United Nations Global Mercury Assessment. Environ. Sci. Technol. 52, 11466–11477.
Pacyna, E.G., Pacyna, J., Sundseth, K., Munthe, J., Kindbom, K., Wilson, S., Steenhuisen, F., Maxson, P., 2010. Global emission of mercury to the atmosphere from anthropogenic sources in 2005 and projections to 2020. Atmos. Environ. 44, 2487–2499.
Peterson, S.H., Ackerman, J.T., Eagles-Smith, C.A., 2017. Mercury contamination and stable isotopes reveal variability in foraging ecology of generalist California gulls. Ecol. Ind. 74, 205–215.
Polis, G.A., Strong, D.R., 1996. Food web complexity and community dynamics. Am. Nat. 147, 813–846.
Ramos, R., Ramírez, F., Sanpera, C., Jover, L., Ruiz, X., 2009. Feeding ecology of yellow-legged gulls *Larus michahellis* in the western Mediterranean: a comparative assessment using conventional and isotopic methods. Mar. Ecol. Prog. Ser. 377, 289–297.
Rudd, J.W., 1995. Sources of methyl mercury to freshwater ecosystems: a review. Water Air Soil Pollut. 80, 697–713.
Ruus, A., Øverjordet, I.B., Braaten, H.F.V., Evenset, A., Christensen, G., Heimstad, E.S., Gabrielsen, G.W., Borgå, K., 2015. Methylmercury biomagnification in an Arctic pelagic food web. Environ. Toxicol. Chem. 34, 2636–2643.
Stewart, F., Phillips, R., Catry, P., Furness, R., 1997. Influence of species, age and diet on mercury concentrations in Shetland seabirds. Mar. Ecol. Prog. Ser. 151, 237–244.
Suryan, R.M., Sato, F., Balogh, G.R., Hyrenbach, K.D., Sievert, P.R., Ozaki, K., 2006. Foraging destinations and marine habitat use of short-tailed albatrosses: a multi-scale approach using first-passage time analysis. Deep Sea Res. Part II: Topical Stud. Oceanogr. 53, 370–386.
Takimoto, G., Iwata, T., Murakami, M., 2002. Seasonal subsidy stabilizes food web dynamics: balance in a heterogeneous landscape. Ecol. Res. 17, 433–439.
Thompson, D., Furness, R., 1989. The chemical form of mercury stored in South Atlantic seabirds. Environ. Pollut. 60, 305–317.
Thompson, D., Furness, R., Walsh, P., 1992. Historical changes in mercury concentrations in the marine ecosystem of the north and north-east Atlantic Ocean as indicated by seabird feathers. J. Appl. Ecol. 79–84.
Thompson, D., Bearhop, S., Speakman, J.R., Furness, R., 1998a. Feathers as a means of monitoring mercury in seabirds: insights from stable isotope analysis. Environ. Pollut. 101, 193–200.
Thompson, D.R., Furness, R.W., Monteiro, L.R., 1998b. Seabirds as biomonitors of mercury inputs to epipelagic and mesopelagic marine food chains. Sci. Total Environ. 213, 299–305.