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Trophic calculations reveal the mechanism of population-level variation in mercury concentrations between marine ecosystems: Case studies of two polar seabirds

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ABSTRACT

The incorporation of quantitative trophic level analysis in ecotoxicological studies provides explanatory power to identify the factors, trophic or environmental, driving population-level variation in mercury exposure at large geographic scales. In the Antarctic marine ecosystem, mercury concentrations and stable isotope values in Adélie penguins (*Pygoscelis adeliae*) were compared between the Antarctic Peninsula and the Ross Sea. Correcting tissue δ^{15} N values for baseline δ^{15} N values revealed population-level differences in trophic position which contributes to differences in mercury. Data from Thick-billed murres (*Uria lomvia*) were synthesized from published values from Baffin Bay and Svalbard to demonstrate the utility of baseline δ^{15} N values in identifying differences in environmental mercury exposure independent of diet. Here, we demonstrate the importance of calculating population-specific trophic level data to uncover the source of variation in mercury concentrations between geographically distinct populations of marine predators.

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1. Introduction

Due to their wide geographic distribution and elevated trophic position, marine predators such as seabirds and marine mammals can serve as effective biomonitors of mercury availability in marine food webs at local, regional, and global scales (Burger and Gochfeld, 2004; Aguilar et al., 2002; Aubail et al., 2011). However, geographically distinct populations could be exposed to different concentrations of mercury based on diet, habitat-specific environmental factors lending to increased bioavailability of mercury, or proximity to local point sources of mercury pollution (Evers et al., 2007; Scheuhammer et al., 2007; Aubail et al., 2011; Pouilly et al., 2013). Despite a lack of point sources of mercury pollution in many ocean ecosystems, variation in geologic and oceanographic processes could affect the distribution and bioavailability of mercury setting the stage for geographic variation in the risk of exposure to mercury in marine biota (Sunderland and Mason, 2007; Cossa et al., 2011; Point et al., 2011). Spatial heterogeneity in oceanographic processes can also lead to geographic variation in prey availability driving dietary and thus trophic level differences among populations (Gaston and Bradstreet, 1993; Bradshaw et al., 2000) that could also result in differential dietary mercury exposure.

Due to the large geographic ranges of many marine predators, studies on contaminants are typically snapshots of exposure within a specific portion of the species range. For example, Blévin et al. (2013) analyzed mercury concentrations in chicks of 21 species of seabirds from the Kerguelen archipelago. While the mercury concentrations reported in their study are an appropriate representation mercury exposure at this sub-Antarctic archipelago, it is important to note that several of the species examined have large breeding ranges that extend into southern South America, the Antarctic Peninsula, and other sub-Antarctic islands. As oceanographic conditions and trophic relationships in distinct regions of the ocean differ, rates of mercury deposition and trophic transport also have the potential to vary between regions. Supporting the above prediction, Blévin et al. (2013) compiled published accounts and found a wide range of geographic variation in the feather mercury concentrations of seabird chicks. These findings suggest caution is warranted when attempting to use mercury concentrations from a single-site in species with large breeding ranges to derive predictions of species-level toxicological risks. High mercury concentrations in one population do not imply similar exposure across the species' range nor should values from a single population be used as an estimate of species-level exposure (Evers et al., 1998, 2007; Bond and Lavers, 2011). This same reasoning also applies to migratory species spending portions of the year in geographically distinct regions or those that experience dietary shifts between wintering and breeding habitats (Evers et al., 1998; Winder and Emslie, 2011).



Note



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Stable isotope analysis has become a popular tool in ecotoxicological investigations due to a general correlation between stable nitrogen isotopes (δ^{15} N) and trophic level (Jardine et al., 2006). Within a given ecosystem, $\delta^{15}N$ can be used to distinguish among trophic levels as δ^{15} N concentrations tend to be enriched by 3–4‰ between a consumer and its prey (Post, 2002). This relationship has allowed researchers to track biomagnification of mercury within marine and aquatic food webs (e.g. Atwell et al., 1998; Chasar et al., 2009) and identify trophic differences among sympatric species which explain patterns of mercury exposure (e.g. Blévin et al., 2013; Pouilly et al., 2013). However, while consumer $\delta^{15}N$ values can aid in establishing trophic relationships within a given food web, marine ecosystem baseline $\delta^{15}N$ values vary through time and space with factors such as primary productivity, latitude, and ocean frontal region preventing direct comparison of isotope values among geographically distinct food webs (Post, 2002; McMahon et al., 2013). Though the merit of integrating trophic ecology and contaminant dynamics has been documented across a variety of taxa (fish, seabirds, and marine mammals), too often δ^{15} N values are compared among geographically distinct populations without correcting for differences in baseline $\delta^{15}N$ (Braune et al., 2002; Geisz et al., 2008; Aubail et al., 2010; Aubail et al., 2011; Vo et al., 2011; Brasso et al., 2012; Zhang and Wang, 2012; Sluis et al., 2013). For example, Aubail et al. (2010) concluded that elevated mercury concentrations in ringed seals (Phoca hispida) on the western coast of Greenland were the result of higher environmental bioavailability in this region as δ^{15} N values were lower in the west coast population relative to the east coast. However, it is possible that differences in $\delta^{15}N$ between these geographically isolated populations simply reflect disparities in baseline $\delta^{15}N$ values between food webs (Post, 2002; Popp et al., 2007; Choy et al., 2012), not inherently implying elevated trophic position. On the other hand many biomonitoring efforts report only tissue mercury concentrations leaving crucial dietary and trophic interactions unresolved altogether, offering little more than speculation in terms of explaining geographic variation in contaminant concentrations (Riget et al., 2004; Day et al., 2006; Bond and Lavers, 2011: Ferris and Essington, 2011).

A growing number of studies are taking a whole food web approach to stable isotope ecology and δ^{15} N values for primary producers and low-trophic consumers in marine ecosystems are increasingly available (Davenport and Bax, 2002; Campbell et al., 2005; Ciancio et al., 2008; Jæger et al., 2009; Stowasser et al., 2012; Pinkerton et al., 2013; Pouilly et al., 2013). Though limited in the literature, this growing source of information can now allow

researchers to pair ecosystem-specific baseline and consumer δ^{15} N values to calculate population-specific trophic level data for use in ecotoxicological studies across large geographic scales (e.g. Day et al., 2012). The use of this approach enables researchers to move beyond speculation and use a hypothesis testing framework to examine the root cause of geographic differences in mercury exposure. For example, when differences in trophic level and mercury concentrations are mirrored between populations, it provides support for the hypothesis that differences in trophic position between populations contributes to their different exposure to mercury. On the other hand, when trophic levels are similar, but mercury concentrations differ between populations (or vice versa), it lends support to the hypothesis that differences in the bioavailability of mercury between ecosystems contributed to the observed differences between populations.

The purpose of the present study was to demonstrate the utility of pairing baseline and consumer δ^{15} N values to identify the trophic and environmental sources of population-level variation in mercury exposure. Tissue samples were collected in the field and literature derived data were synthesized from two wide-ranging circumpolar seabirds the Adélie penguin (*Pygoscelis adeliae*) and Thick-billed murre (*Uria lomvia*), respectively. Here, we demonstrate how determining the mechanism of population-level variation in mercury exposure can allow researchers to identify specific populations that may be at risk to elevated exposure to mercury as well as pin point potential geographic "hot spots" of mercury availability.

2. Methods

2.1. Adélie penguin

Secondary down was collected from 3–4 week old Adélie penguin chicks at Admiralty Bay, King George Island in the Antarctic Peninsula ($62^{\circ}10'$ S, $58^{\circ}27'$ W; n = 20) and at Cape Crozier, Ross Island in the southern Ross Sea ($77^{\circ}31'$ S, $169^{\circ}24'$ E, n = 20) during the 2009/2010 austral summer (December 2009–January 2010; Fig. 1). Secondary down serves as an effective biomonitoring unit as the lifetime exposure to a given contaminant is limited to several weeks following the loss of natal down, individuals are of known age, the foraging range of the adult birds is constrained during the chick-rearing period, and any difference in adult dietary specialization is averaged through bi-parental care (Janssens et al., 2002; Blévin et al., 2013).



Fig. 1. (a) Study sites for Adélie penguins at King George Island (62°10′ S, 58°27′ W) in the Antarctic Peninsula and Cape Crozier, Ross Island (77°31′S, 169°24′E) in the Ross Sea; (b) Study sites for Thick-billed murres in Kongsfjorden, Svalbard (79°N, 12–13°E; Jæger et al., 2009) and Baffin Bay (~76°–79°N, 70°–80°W; Campbell et al., 2005).

Approximately 10 mg of down from each individual was rinsed in an alternating series of acetone and deionized water baths, dried under a fume hood for 24 h and analyzed for total mercury via atomic absorption spectrophotometry on a Tri-Cell Direct Mercury Analyzer (DMA-80) at the University of North Carolina Wilmington (Wilmington, NC, USA). Because nearly all mercury in feathers is present in the form of methylmercury, total mercury concentration was used as a proxy for this highly bioavailable form (Evers et al., 2005; Bond and Diamond, 2009). Each set of 20 samples analyzed was preceded and followed by two method blanks, a sample blank, and two samples each of standard reference material (DORM-3 and DOLT-4). Mercury concentrations in chick down are reported as parts per million (ppm) fresh weight (fw). Mean percent recoveries for standard reference materials were 99.3 ± 2.1% (DORM-3) and $98.6 \pm 2.6\%$ (DOLT-4) with relative significant differences in mercurv concentration <2.6%.

Prior to stable isotope analysis, down was cleaned using a 2:1 chloroform:methanol rinse and cut into small fragments with stainless steel scissors. Approximately 0.5 mg of down was loaded into tin cups, flash-combusted (Costech ECS4010), and analyzed for δ^{15} N through an interfaced Thermo Delta V Plus continuous-flow stable isotope ratio mass spectrometer (CFIRMS). Raw δ values were normalized on a two-point scale using depleted and enriched glutamic acid standard reference materials (USGS-40 and USGS-41). Sample precision based on duplicate standard and sample materials was 0.2‰. Stable isotope abundances are expressed using a δ notation in per-milliliter units (‰) based on the following equation:

 $\delta X = [(R_{\text{sample}}/R_{\text{standard}}) - 1] * 1000$

where X is ¹⁵N, and R is the corresponding ratio of ¹⁵N:¹⁴N and $R_{\text{stan-dard}}$ value are based on atmospheric N₂ for ¹⁵N.

2.2. Thick billed-murre

We analyzed published data on mercury concentrations and δ^{15} N values in muscle tissue of adult Thick billed-murres from the Norwegian and Canadian sectors of the Arctic (Fig. 1). These included individuals (n = 10) collected by [æger et al. (2009) from Kongsfjorden, Svalbard in the Norwegian Arctic (79°N, 12-13°E) in the arctic summers of 2005 and 2006 and individuals (n = 10)collected by Campbell et al. (2005) from the Northwater Polynya in northern Baffin Bay (~76°N to 79°N and 70°W to 80°W) in the Arctic summer of 1999. Analytical methods were similar between the two studies with total mercury concentration determined via cold vapor atomic absorption spectrometry, and $\delta^{15}N$ values determined using CFIRMS (for complete analytical details see: Campbell et al., 2005; Jæger et al., 2009). We used total mercury concentration data from these two studies as proxy for bioavailable methylmercury because nearly all mercury in seabird muscle tissues is present in the form of methylmercury (Thompson et al., 1991; Campbell et al., 2005).

2.3. Trophic position calculations

Tissue δ^{15} N values were converted into relative trophic positions using a modification of the model described by Hobson et al. (1994, 2002):

$TL_{consumer} = 2 + (\delta^{15}N_{consumer} - \delta^{15}N_{primary\ consumer})/\Delta_N\ {}_{foodweb}$

This model uses a consumer's $\delta^{15}N$ value to estimate its trophic position relative to the mean $\delta^{15}N$ value of a primary consumer (assumed trophic position of 2) and the mean ¹⁵N food web trophic discrimination per trophic transfer (Δ_N). However, the avian tissues used in our case studies differed (down feathers and muscle) and $\delta^{15}N$ values in seabird tissues are dependent on tissue-specific

discrimination factors (Bond and Jones, 2009; Polito et al., 2009). Therefore, we modified the above formula to standardize calculations between tissues by adding an additional term to account for tissue-specific dietary isotopic discrimination factors as proposed by Hobson and Bond (2012):

$TL_{bird} = 3 + (\delta^{15}N_{bird} - \Delta_{N \text{ avian tissue}} - \delta^{15}N_{primary \text{ consumer}})/\Delta_{N \text{ food web}}$

Using the above formula we incorporated Adélie penguin and Thick-billed murre isotopic values for $\delta^{15}N_{consumer}$ and food webspecific primary consumer isotopic values ($\delta^{15}N_{primary consumer}$) into separate models. For models with Adélie penguins we used published discrimination factors ($\Delta_{N \text{ avian tissue}}$) for feathers (+3.5%; Polito et al., 2011) and mean δ^{15} N values for salps (*Salpa thompson*i) from the Antarctic Peninsula (2.7%; Stowasser et al., 2012) and Ross Sea (3.9%; Pinkerton et al., 2013). We used individual $\delta^{15}N$ values to calculate mean ± SD TL_{bird} for Adélie penguins. For models with Thick-billed murres we used published discrimination factors ($\Delta_{N \text{ avian tissue}}$) for muscle (+2.4‰; Mizutani et al., 1991) and mean δ^{15} N values from copepods (*Calanus hyperboreus*) collected around Svalbard (7.5%; Søreide et al., 2006) and Baffin Bay (7.9%; Hobson et al., 2002). As individual δ^{15} N values were not available for Thick-billed murres we used published mean ± SD δ^{15} N values to calculate mean ± SD TL_{bird} for this species. For all models we assumed a mean $\Delta_{N \text{ food web}}$ value of 3.4‰ as this value is robust across multiple food webs (Deniro and Epstein, 1981; Minagawa and Wada, 1984; Post, 2002; Søreide et al., 2006).

2.4. Statistical analysis

For each species, we used two sample *t*-tests to identify significant differences in total mercury concentrations, $\delta^{15}N$ values, and calculated trophic levels. Prior to analysis data were examined for normality and equal variance; mercury concentrations were log-transformed in order to create a data set with a normal distribution. All tests were two-tailed, significance was assumed at the 0.05 level, and means are presented ±SD. Statistical calculations were performed using SAS (version 9.1, SAS Institute 1999).

3. Results

Mercury concentrations in Adélie penguin chicks differed significantly between regions with mercury concentrations in the Ross Sea nearly five times higher than in the Antarctic Peninsula (Table 1). Chick δ^{15} N values also differed significantly between regions; δ^{15} N values in the Ross Sea were 2.6 ‰ higher than those in the Antarctic Peninsula. This difference in δ^{15} N between regions translated to a significant difference in trophic position (Table 1, Fig. 2). Adélie penguin chicks in the Ross Sea consumed a diet nearly one-half trophic level higher (0.4) than chicks in the Antarctic Peninsula.

Mercury concentrations in Thick-billed murres differed significantly between regions; mercury concentrations in Baffin Bay were approximately three times higher than in Svalbard (Table 1). Adult muscle $\delta^{15}N$ values also differed significantly between regions; $\delta^{15}N$ values in Baffin Bay were 0.8‰ higher than those in Svalbard. However, in this case the significant difference in $\delta^{15}N$ did not translate to a difference in trophic position between populations in Baffin Bay and Svalbard (Table 1; Fig. 2).

4. Discussion

In the absence of point sources of mercury contamination in the ocean, population-level differences in mercury exposure can result from disparities in dietary composition among populations or the bioavailability of mercury in geographically distinct foraging

Table 1

Tissue mercury concentrations (Hg), stable nitrogen isotope values (δ^{15} N) and calculated trophic levels of Adélie penguins from the Antarctic Peninsula (*n* = 20) and Ross Sea (*n* = 20) and Thick-billed murres from Svalbard (*n* = 10; Campbell et al., 2005) and Baffin Bay (*n* = 10; Larger et al., 2009). See text for details.

Species, tissue	Sites, comparisons	Hg (ppm)	δ ¹⁵ N (‰)	Trophic level
Adélie penguin, chick down	Antarctic Peninsula	0.11 ± 0.22	8.1 ± 0.5	3.6 ± 0.1
	Ross Sea	0.53 ± 0.08	10.7 ± 0.3	4.0 ± 0.1
	Difference	0.42	2.6	0.4
	t-test	<i>t</i> = 29.3, <i>p</i> < 0.001	<i>t</i> = 20.7, <i>p</i> < 0.001	<i>t</i> = 9.9, <i>p</i> < 0.001
Thick-billed murre, adult muscle	Svalbard	0.11 ± 0.01	12.7 ± 0.2	3.8 ± 0.1
	Baffin Bay	0.33 ± 0.09	13.5 ± 0.6	3.9 ± 0.2
	Difference	0.22	0.8	0.1
	<i>t</i> -test	t = 7.7, p < 0.001	t = 4.0, p < 0.001	t = 1.4, p = 0.174



Fig. 2. As trophic position was similar in both populations of Thick-billed murres, the higher mercury concentrations in the population in Baffin Bay relative to Svalbard indicated enhanced bioavailability of mercury in Baffin Bay (t = 7.7, p < 0.001, top; data from Campbell et al., 2005; Jæger et al., 2009). Foraging at a higher trophic position (t = 9.9, p < 0.001) explained the elevated mercury concentrations in the Adélie penguin population in the Ross Sea compared to the Antarctic Peninsula (t = 29.3, p < 0.001; bottom).

habitats. Here, we have provided two examples documenting the merit of incorporating trophic level data derived from stable isotope analysis into biomonitoring efforts to decipher between trophic and environmental sources of population-level variation in mercury. These case studies highlight how trophic level metrics that account for differences in ecosystem δ^{15} N baselines can help researchers avoid incorrect conclusions about the relative importance of trophic position as a driving factor in differences in mercury exposure between geographically distinct populations.

In the Antarctic marine food web, observed differences in Adélie penguin δ^{15} N values translated into significant differences in the trophic level between geographically distinct populations. Therefore, trophic differences between populations appear to be an important factor explaining the observed regional differences in tissue mercury concentrations. Published data on stomach contents reinforce our findings and suggest a greater reliance on fish prey by penguins in the Ross Sea relative to the Antarctic Peninsula (Volkman et al., 1980; Ainley, 2002). While these finding do not discount the possibility that differences in environmental mercury availability may exist between these two Antarctic food-webs, the higher trophic status of penguins in the Ross Sea is clearly an important driver in this population's elevated exposure to mercury.

On the contrary, the opposite trend was found when examining differences in mercury between populations of Thick-billed murres in the Arctic. As with the Adélie penguin, the δ^{15} N values of Thickbilled murres differed between regions (Baffin Bay and Svalbard). suggesting at first that trophic differences might contribute to the observed differences in tissue mercury concentration between these two regions. However, when controlling for differences in ecosystem baseline δ^{15} N values, we were able to rule out trophic level differences between populations and conclude that environmental mercury availability in Baffin Bay is likely higher than in Svalbard. Therein, more research should be focused on investigating environmental disparities between these two Arctic ecosystems to determine the source of population-level variation in mercury exposure (e.g., enhanced deposition, methylation rates, photochemical breakdown). In both cases examined here, trophic level calculations using food web specific baseline and consumer δ¹⁵N values helped to identify the most plausible mechanism driving variation in mercury between geographic distinct populations.

The number of trophic levels within a given food web is a strong predictor of the potential for the biomagnification of mercury (Cabana and Rasmussen, 1994). However, there is a growing realization that the use of consumer $\delta^{15}N$ values, without baseline corrections, as a proxy of trophic position should be limited to comparisons within a given ecosystem rather than among ecosystems in avoid incorrect conclusions (Post, 2002; Popp et al., 2007; Choy et al., 2012). To this end, pairing ecosystem-specific baseline and consumer δ^{15} N values allows for the calculation of trophic level data which can be robustly compared across geographically distinct food webs. Studies examining large scale, spatial patterns of mercury that also to control for consumer trophic position using stable isotopes have been historically limited by the availability of baseline δ^{15} N values for marine ecosystems. Fortunately, a growing number of isotopic studies of the trophic structure of pelagic food webs in oceanic regions of southern South America and Patagonia

(Ciancio et al., 2008), southeastern Australia (Davenport and Bax, 2002), the southern Indian Ocean (Kaehler et al., 2000), the Arctic (Campbell et al., 2005; Jæger et al., 2009), and the Southern Ocean (Stowasser et al., 2012; Pinkerton et al., 2013) now make robust trophic comparison of across populations and large geographic regions possible. Past studies of mercury contamination which did not include stable isotope data (e.g. Riget et al., 2004; Bond and Lavers, 2011; Ferris and Essington, 2011) or those that solely relied on consumer δ^{15} N values as a proxy for trophic level to explain mercury exposure between geographically isolated populations (e.g. Aubail et al., 2010, 2011; Vo et al., 2011) could be enhanced by calculating trophic level metrics that control for inherent differences in baseline δ^{15} N values among marine ecosystems. For example, Day et al. (2012) added stable isotope data to help reinterpret previously published mercury concentrations data on Thick-billed murres and other seabirds from several regions of Alaska (Dav et al., 2006). Using regional δ^{15} N signatures derived from herbivorous zooplankton from published studies (reviewed in Point et al., 2011), Day et al. (2012) found that trophic normalized mercury concentrations in egg tissues differed across regions indicating spatial variability in mercury availability.

Similarly, in our two case studies we used food web-specific baseline δ^{15} N values to estimate trophic level and evaluate the relative importance of dietary versus environmental variation in mercury concentrations between these geographically distinct populations; a question historically left unanswered in many large-scale toxicological comparisons. Our findings provide strong evidence that differences in trophic position are driving the risk of exposure to mercury in two geographically distinct Adélie penguin populations in Antarctica. In contrast, our analysis of published data on Thick-billed murres highlighted the importance of geographic variation in the bioavailability of mercury between two geographically distinct Arctic marine food webs, a trend also observed within Alaska (Day et al., 2012). In the open ocean, most methylmercury comes from current driven transfer from coastal waters, in situ production by microbes in the water column or marine sediments, or from the upwelling of deep water (Cossa et al., 2011). In polar regions sea-ice cover can mediate atmospheric deposition and either impede (sea-ice present) or facilitate (seaice absent) the photochemical breakdown of methylmercury in surface waters (Point et al., 2011). Additional local factors such as coastal currents, bathymetry, productivity, and/or temperature are thought to also play crucial roles in the distribution and production of bioavailable mercury to marine biota (Mason and Fitzgerald, 1993; Cossa et al., 2011). The right combination of environmental factors (such as microbial activity, low pH, redox potential, and the presence of organic and inorganic complexing materials; Celo et al., 2006) can lead to "hot spots" of mercury availability. However, relationships between the above environmental factors and mercury availability are often only identifiable after ruling out trophic disparities between populations of indicator species such as seabirds (e.g. Point et al., 2011). The approaches outlined here, while not novel, are currently underutilized in the literature and a broader adoption of trophic level calculations in ecotoxicology studies using stable isotope analysis is required to facilitate large scale geographic comparisons. Monitoring mercury concentrations in geographically disparate populations throughout a species range will allow for better estimates of species-level exposure to mercury and the identification of regions, and thereby populations, at greater risk for adverse effects of elevated tissue mercury concentrations as global emissions continue to rise.

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