Murre Eggs (*Uria aalge* and *Uria lomvia*) as Indicators of Mercury Contamination in the Alaskan Marine Environment

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Sixty common murre (*Uria aalge*) and 27 thick-billed murre (*Uria lomvia*) eggs collected by the Seabird Tissue Archival and Monitoring Project (STAMP) in 1999−2001 from two Gulf of Alaska and three Bering Sea nesting colonies were analyzed for total mercury (Hg) using isotope dilution cold vapor inductively coupled mass spectrometry. Hg concentrations (wet mass) ranged from 0.011 μg/g to 0.357 μg/g (relative standard deviation = 76%), while conspecifics from the same colonies and years had an average relative standard deviation of 33%. Hg levels in eggs from the Gulf of Alaska (0.166 μg/g ± 0.011 μg/g) were significantly higher ($p < 0.0001$) than in the Bering Sea (0.047 μg/g ± 0.004 μg/g). Within the Bering Sea, Hg was significantly higher ($p = 0.0007$) in eggs from Little Diomede Island near the arctic than at the two more southern colonies. Although thick-billed and common murres are ecologically similar, there were significant species differences in egg Hg concentrations within each region ($p < 0.0001$). In the Bering Sea, eggs from thick-billed murres had higher Hg concentrations than eggs from common murres, while in the Gulf of Alaska, common murre eggs had higher concentrations than those of thick-billed murres. A separate one-way analysis of variance on the only time−trend data currently available for a colony (St. Lazaria Island in the Gulf of Alaska) found significantly lower Hg concentrations in common murre eggs collected in 2001 compared to 1999 ($p = 0.017$). Results from this study indicate that murre eggs may be effective monitoring units for detecting geographic, species, and temporal patterns of Hg contamination in marine food webs. The relatively small intracolony variation in egg Hg levels and the ability to consistently obtain adequate sample sizes both within and among colonies over a large geographic range means that monitoring efforts using murre eggs will have suitable statistical power for detecting environmental patterns of Hg contamination. The potential influences of trophic effects, physical transport patterns, and biogeochemical processes on these monitoring efforts are discussed, and future plans to investigate the sources of the observed variability are presented.

Introduction

The cycling of mercury (Hg) in the environment and the potential impacts of this toxicant on humans and wildlife have received considerable attention in the literature. Despite reductions in Hg emissions in North America and Western Europe in recent decades, population and economic growth in developing regions may be increasing global Hg emissions (*1*). There is evidence suggesting Hg contamination has increased in some North American arctic species since the 1980s (*1*). However, recent temporal trends vary depending on species and region, and declining trends have been reported from other arctic regions (*1*). Despite relatively modest local sources of pollution, high latitude regions often contain surprisingly high levels of contaminants such as Hg that can be transported globally. The transport and deposition of Hg in the arctic during atmospheric Hg depletion episodes that occur during polar sunrise suggest that this region may serve as a global sink for Hg (*2*−*4*). Recent evidence also suggests that Hg deposited from the atmosphere and incorporated into the snowpack may be transformed into methylmercury (MeHg) during snowmelt, thus making it available for incorporation into the food web during the most biologically active season (*5*). Although the understanding of large-scale patterns of Hg transport and cycling continues to improve, there are few examples of these patterns being reflected in the Hg distribution in biota. This is due to variability created by local physicochemical characteristics that affect transport and bioavailability and inconsistencies in the life history and food web characteristics of organisms selected for monitoring. Thus far, there has been some success using bird eggs to detect geographic patterns of Hg in biota. A geographic comparison on loon eggs showed an east−west gradient across North America consistent with Hg atmospheric deposition patterns (*6*). Also, a study on atmospheric Hg deposition found that areas in the Canadian arctic with favorable atmospheric conditions for Hg deposition also had relatively high Hg levels in gull eggs compared to other areas (*4*, *7*). The same authors also found that Hg levels in eggs from thick-billed murres and northern fulmars increased significantly between 1975 and 1998, suggesting that seabird eggs may also be useful for detecting long-term temporal trends (*8*).

Aquatic environments are particularly sensitive to Hg contamination because aquatic physicochemical conditions can facilitate the conversion of inorganic Hg (IoHg) into more bioavailable MeHg, which accumulates very efficiently in top predators (particularly piscivorous species) (*9*). The life histories and ecological niches occupied by many aquatic birds predisposes them to accumulating Hg, and the toxicology and merits/limitations of using feathers and eggs for monitoring Hg have been studied extensively. In 1998, the International Arctic Monitoring and Assessment Programme (AMAP) identified eggs from alcids (seabirds belonging to the family Alcidae) as key tissues for long-term circumpolar monitoring of contaminants by arctic nations (*10*). Common

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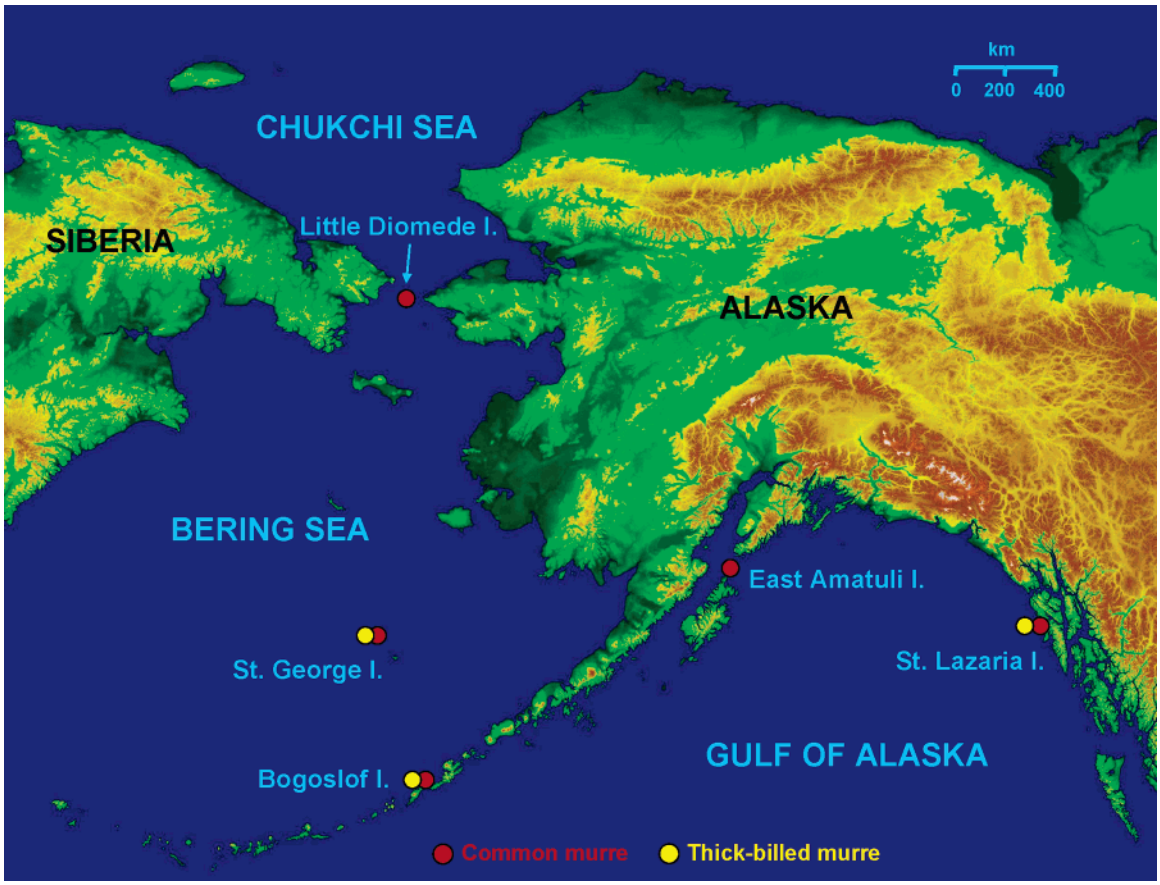

FIGURE 1. Map of the study area showing the five murre (*Uria* spp.) colonies where eggs were collected.

murres (*Uria aalge*) and thick-billed murres (*Uria lomvia*) are primarily piscivorous, deep-diving alcids with life history characteristics that make them good candidates for biomonitoring of contaminants in marine food webs. They feed near the top of their marine food web, the adults and their eggs are abundant and logistically practical to collect, and they have a circumpolar distribution. Murre eggs are also harvested for human consumption in many regions of the arctic.

When using bird eggs for monitoring geographic and temporal trends in contaminants, the influence of long-term bioaccumulation in the female on the egg contaminant level and what portion of this body burden may come from distant and sometimes poorly known over-wintering grounds should be considered. A study on herring gulls in the Great Lakes showed that in some cases the winter severity (and presumably the concomitant differences in migratory patterns) was related to the organochlorine concentrations in eggs the following spring (*11*). It is likely that Hg mobilized from tissue storage compartments contributes some fraction of the Hg deposited in developing eggs; however, laboratory studies of maternal transfer in chickens showed that egg Hg concentrations are good indicators of maternal dietary exposure during egg formation (*12*−*14*). Field studies of loons have also demonstrated good agreement between Hg concentrations in eggs and blood from females in the same territory (6), and concentrations in blood closely reflected Hg concentrations in local prey (*15*, *16*). This means that seasonal Hg intake during over-wintering and age-dependent bioaccumulation in females are minor contributors to egg Hg concentrations. Murres typically over-winter in the open ocean where variability from point-sources does not exist and they forage on their breeding grounds for several weeks prior to nesting (*17*, *18*), further reducing the potential influence that distant wintering grounds may have on contaminant levels in their eggs. Murres also lay a single egg (*18*), eliminating potential variability in egg Hg burdens due to laying order that has been observed for other species (*6*, *19*−*20*).

Data reported here are from the Seabird Tissue Archival and Monitoring Project (STAMP), a collaborative U.S. Fish and Wildlife Service (USFWS), U.S. Geological Survey (USGS), Bureau of Indian Affairs (BIA), and National Institute of Standards and Technology (NIST) study designed to provide long-term monitoring of contaminant trends in Alaskan marine environments using seabirds. This study reports the geographic, temporal, and species trends in egg Hg levels from the first three sampling years at murre colonies in the Bering Sea and Gulf of Alaska. It also discusses the potential mechanisms that may drive these trends and the implications for biomonitoring using seabird eggs.

Methods

Sample Collection and Processing. A total of 60 common murre and 27 thick-billed murre eggs were collected by USFWS personnel and subsistence harvesters at colonies in the Gulf of Alaska (St. Lazaria and East Amatuli Islands) and Bering Sea (St. George, Little Diomede, and Bogoslof Islands) from 1999 to 2001 (Figure 1). Because murres may lay a replacement egg if their single first-laid egg is lost (*18*), egg collection was timed to target first-laid eggs. Egg contents were frozen and cryogenically homogenized using procedures described by Zeisler et al. (*21*). Portions of the cryohomogenates not used for analysis are archived in the NIST Marine Environmental Specimen Bank (Hollings Marine Laboratory, Charleston, SC) for use during future studies.

Analytical Methods. Total Hg (THg) concentrations (based on wet mass) in egg contents were determined using isotope

dilution cold vapor inductively coupled plasma mass spectrometry (ID-CV-ICPMS). This analytical method has been previously described in detail (*22*) and is only briefly summarized here. Isotopically enriched ^{201}Hg spike solution was prepared and calibrated using NIST Standard Reference Material (SRM) 3133 Hg Spectrometric Solution. The spike was then added quantitatively to a mass of sample (0.4−1.0 g) to yield an isotopic ratio ($^{201}Hg/^{202}Hg$) that minimized random error propagation. Samples were then digested and equilibrated in a Perkin-Elmer (Shelton, CT) Multiwave microwave oven at the highest possible temperatures (up to 300 °C) and pressures (up to 8 MPa) using quartz microwave decomposition vessels and high-purity nitric acid (Fisher Scientific, Suwanee, Georgia). The digestant was mixed with a $SnCl_2$ and HCl reductant solution in a gas−liquid separator, allowing cold vapor transfer of the resulting Hg^0 in a stream of argon to the inductively coupled plasma mass spectrometer (ICPMS) injector. A VG Elemental Plasma Quad 3 ICPMS (Windsford, Cheshire, U.K.) using typical power and gas flow was used in time resolved analysis mode for measurement of isotope ratios.

Samples were analyzed for THg in 19 analytical batches from May 5, 2001, to July 24, 2004. Each batch contained up to eight egg samples, one NIST SRM 2976 Mussel Tissue (trace elements and methylmercury) used as an external control, and one procedural blank. The mean and standard deviation of the 19 measured values of SRM 2976 was 0.0610 μg/g ± 0.0021 μg/g compared to the certified value of 0.0610 μg/g ± 0.0036 μg/g. Reproducibility was previously determined by analyzing 4 aliquots of homogenized egg from one individual (repeated for two individuals) within the same analytical batch, yielding relative standard deviation (RSD) values of 1.25% and 0.64% (*22*).

MeHg was measured in 3 eggs using GC-ICPMS and reported by Davis et al. (*23*). This subsample of 3 eggs came from different colonies that cover the entire range of concentrations observed.

Statistics. All statistics were performed using commercially available software (SAS Institute's JMP 3.26, Cary, NC). Conformity with the assumptions of ANOVA was confirmed using a Shapiro-Wilk test for normality and the O'Brien, Brown-Forsythe, Levene, and Bartlett tests for homoscedasticity. Hg concentrations for the species and geographic comparisons were natural log transformed to meet these assumptions and analyzed using a two-way factorial analysis of variance (ANOVA). Comparisons between individual cells in the species−region interaction term were performed using a Tukey-Kramer test. Individual colonies within the Bering Sea were coded by island and analyzed using a Welch ANOVA (allowing for heteroscadasticity) to explore geographic variability within this region, and a Tukey-Kramer HSD test was used for pairwise comparisons. Temporal variation was analyzed using a separate one-way ANOVA on the only multiyear collections for the same species and location that are currently available (common murres from St. Lazaria Island, Gulf of Alaska). The experiment-wide α was adjusted for multiple tests according to the Dunn-Sidak method, giving an adjusted α of 0.017 at the 95% confidence level.

Results

THg concentrations (wet mass) in eggs across all colonies, species, and years ranged from 0.011 μg/g to 0.357 μg/g with an overall RSD of 76% (Figure 2), while conspecifics from the same colonies and years had an average RSD of 33% (Table 1). The fraction of the total Hg present as MeHg ranged from 84.5% to 88% (*23*).

Geographic Trend. The model comparing geographic and species differences was highly significant ($r^2 = 0.72$, $F = 70.17$, df = 3, $p < 0.0001$), with the regional effect explaining more

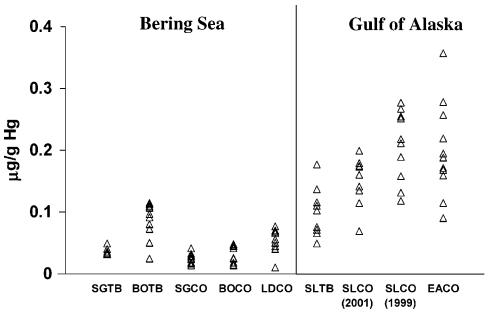

FIGURE 2. Total Hg concentrations (wet mass) for murre eggs for each collection event are reported. The first two letters of the four letter code indicate location (BO = Bogoslof, LD = Little Diomede, SG = St. George, EA = East Amatuli, SL = St. Lazaria) and the second two letters indicate species (CO = common murre, TB = thick-billed murre).

of the variation in egg Hg levels than either the species or species−region interaction terms. Hg concentrations in murre eggs from the Gulf of Alaska (0.166 μg/g ± 0.011 μg/g, mean ± 1 SE) were significantly higher ($F = 101.30$, df = 1, $p < 0.0001$) than those from the Bering Sea (0.047 μg/g ± 0.004 μg/g) (Figure 3). This difference transcended any species differences, with both common and thick-billed murres in the Gulf of Alaska having significantly higher Hg ($p < 0.05$) than either of these species in the Bering Sea (Figure 3). Although not included in the statistical analyses, visual inspection of the data revealed that the variability between islands within each region was small relative to between-region differences (Figure 2). This suggests the mechanism driving the majority of the geographic variability may operate at a relatively large spatial scale. Comparison of egg Hg levels from individual colonies within the Bering Sea showed there were also significant geographic differences within this region ($F = 7.18$, df = 2, $p = 0.0073$). A pairwise comparison showed that egg Hg burdens on Little Diomede Island in the Bering Strait were significantly higher ($p < 0.05$) than either of the two colonies in the southern Bering Sea (Figure 4).

Species and Temporal Comparison. The species effect was not significant ($F = 0.001$, df = 1, $p = 0.92$) when compared across all regions; however, the highly significant interaction term between species and region ($F = 29.18$, df = 1, $p < 0.0001$) indicates a closer inspection of the species comparison is required. Separate comparisons of these species within each region revealed that in the Bering Sea thick-billed murre eggs had significantly higher Hg levels than common murre eggs, and that in the Gulf of Alaska common murre eggs had significantly higher Hg levels than thick-billed murre eggs (Figure 3). In addition, pairwise comparisons among all combinations of species−region categories yielded significant differences. The Hg burden in common murre eggs collected from St. Lazaria Island in 1999 were compared to those collected in 2001 to investigate temporal variability. Hg concentrations in eggs from 1999 were significantly higher ($r^2 = 0.28$, $F = 6.99$, df = 18, $p = 0.0165$) than in 2001 (Figure 5).

Discussion

Geographic Trends. Describing spatial patterns of contaminants on local, regional, and global scales is a fundamental part of measuring sources and sinks of toxicants, assessing environmental risks, and understanding the abiotic and biotic processes that drive these patterns. The Hg levels reported here are comparable to those reported from other regions of the world (Table 1). Within this data set there is a distinct trend of higher Hg contamination in murres from colonies in the Gulf of Alaska compared to the Bering Sea. This geographic trend is consistent with that found in Pacific

TABLE 1. Mean Total Hg Concentrations (Wet Mass) in Common (*U. aalge*) and Thick-billed (*U. lomvia*) Murre Eggs from Colonies Worldwide

species	location	year	Hg (μg/g)	RSD (%)	N	ref
U. aalge	Alaska (Little Diomede)	1999	0.053	38.1	9	22
U. aalge	Alaska (St. George)	1999	0.026	31.6	11	22
U. aalge	Alaska (Bogoslof)	2000	0.031	46.3	9	current
U. aalge	Alaska (East Amatuli)	1999	0.200	38.0	11	22
U. aalge	Alaska (St. Lazaria)	1999	0.207	27.5	10	22
U. aalge	Alaska (St. Lazaria)	2001	0.150	25.1	10	current
U. aalge	California (Farallones)	1994	0.165	37.3	12	24
U. aalge	California (Farallones)	1993	0.196	n/a	15	25
U. aalge	Russia (Kola)	1993	0.08	12.5	5	26
U. aalge	Norway (Finmark)	1993	0.10	40	5	26
U. aalge	Norway (E. Finmark)	1983	0.12	33.3	10	27
U. aalge	Norway (W. Finmark)	1983	0.11	54.5	9	27
U. aalge	Norway (Nordland)	1983	0.13	30.1	7	27
U. aalge	Norway (Lofoten)	1983	0.08	12.5	8	27
U. aalge	Norway (northern)	1972	0.07	n/a	30	28
U. lomvia	Alaska (St. George)	2000	0.037	16	7	current
U. lomvia	Alaska (Bogoslof)	2000	0.086	35	10	current
U. lomvia	Alaska (St. Lazaria)	2001	0.104	38	10	current
U. lomvia	Canada (Coats I.)	1998	0.176	n/a	15	7
U. lomvia	Canada (Prince Leopold)	1998	0.332	n/a	15	7
U. lomvia	Canada (Coburg I.)	1993	0.423	n/a	15	7
U. lomvia	Canada (Digges I.)	1993	0.238	n/a	15	7
U. lomvia	Canada (Coats I.)	1993	0.237	n/a	15	7
U. lomvia	Canada (Prince Leopold)	1993	0.290	n/a	15	7
U. lomvia	Canada (Prince Leopold)	1988	0.258	n/a	9	8
U. lomvia	Canada (Prince Leopold)	1987	0.269	n/a	9	8
U. lomvia	Canada (Prince Leopold)	1977	0.150	n/a	9	8
U. lomvia	Canada (Prince Leopold)	1976	0.236	n/a	9	8
U. lomvia	Canada (Prince Leopold)	1975	0.188	n/a	9	8

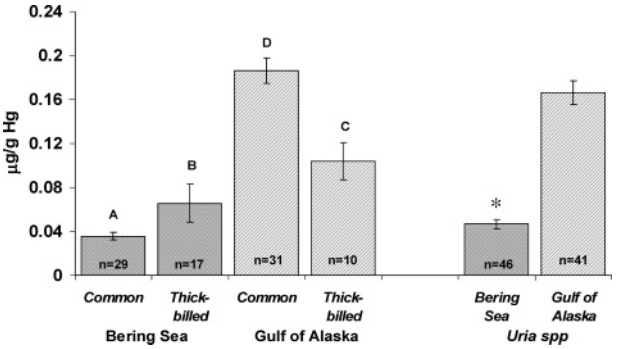

FIGURE 3. Results of a two-way factorial ANOVA comparing geographic and species differences in total Hg concentrations in common (*U. aalge*) and thick-billed (*U. lomvia*) murre egg from colonies in the Gulf of Alaska and Bering Sea. Eggs from the Gulf of Alaska had significantly higher Hg than eggs from the Bering Sea ($p < 0.0001$). There were also significant differences between species within each region, with higher levels of Hg in thick-billed murres in the Bering Sea, and lower levels of Hg in thick-billed murres in the Gulf of Alaska ($p < 0.05$). Error bars represent $\pm$ 1 standard error, the asterisk (*) indicates a significant difference between regions, and the different letter codes (A, B, C, D) indicate a significant difference between pairwise comparisons of each species−region combination.

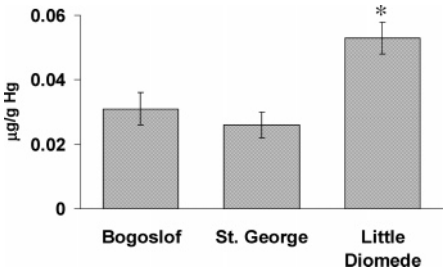

FIGURE 4. Total Hg concentrations (wet mass) in common murre eggs collected at insular colonies in the Bering Sea. A one-way ANOVA indicated egg Hg levels were significantly higher ($p = 0.0073$) at Little Diomede Island in the Bering Strait than at either Bogoslof Island near the Aleutian Islands or St. George Island in the Pribilof Islands. Error bars represent $\pm$ 1 standard error, and the asterisk (*) indicates a significant difference from the other colonies.

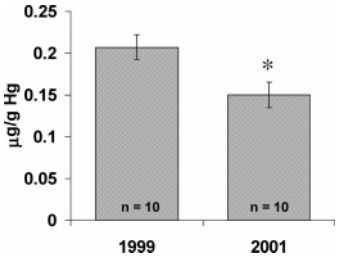

FIGURE 5. Annual variability in total Hg concentrations found in common murre eggs collected from St. Lazaria Island. Egg Hg levels were significantly lower ($p = 0.0165$) in 2001 compared to 1999, representing an approximately 20% change. Error bars represent $\pm$ 1 standard error, and the asterisk (*) indicates a significant difference.

halibut (*Hippoglossus stenolepis*) and sablefish (*Anoplopoma fimbria*) (*29*, *30*), which showed decreasing Hg concentrations from southeast Alaska, to the Gulf of Alaska, to the Bering Sea. A similar trend was also reported in persistent organic pollutants measured in these same egg samples (*31*), and Hg levels in several marine species continue to increase as one moves further south toward British Columbia, Washington, Oregon, and California (*25*, *29*, *30*, *32*). The mechanisms behind this trend are not clear, and could relate to physical transport and deposition patterns, factors related to the formation and bioaccumulation of MeHg, and trophic effects.

There are no available direct measurements on differences in contaminant transport pathways and Hg deposition rates for these regions. One clear climatic difference is the much

higher rate of precipitation in coastal Gulf of Alaska compared to the Bering Sea (*33*). While it has become increasingly clear that dry atmospheric deposition of Hg is important (*34*, *35*), it is probable that higher precipitation in the Gulf of Alaska could result in enhanced scavenging of Hg species as air masses approach the coast (*36*). Representative measurements of atmospheric Hg deposition in these regions of Alaska are currently lacking, but incorporating this type of data into the STAMP program or coordinating sampling sites with a national monitoring and deposition program in the future would greatly enhance the value of the dataset.

Local biogeochemical conditions that control the formation and bioaccumulation of MeHg are often equally as important as Hg input in determining Hg levels in biota (*37*). No quantitative assessment of these factors has currently been made, but the proximity of the Gulf of Alaska colonies to the mainland and remoteness of the Bering Sea insular colonies constitutes a substantial difference in their local environments. The influence of the erosion of natural Hg deposits (*2*), snowmelt-derived Hg (*5*), strong freshwater discharge (*33*), and estuaries that typically have favorable conditions for Hg methylation (*38*) could contribute to the higher Hg levels observed in eggs from the Gulf of Alaska. Additional work on the significance of watershed-derived MeHg and in situ MeHg production in coastal environments is needed to better understand these processes. To further investigate the influence of local ecosystem characteristics on egg Hg burdens, STAMP is currently collecting eggs in the Gulf of Alaska at more remote insular colonies and in the Bering Sea at colonies located on the mainland.

In addition to the regional trend discussed above, a geographic gradient also exists among common murres from individual colonies within the Bering Sea. Egg Hg levels in the southern colonies (Bogoslof Island in the Aleutian Islands and St. George Island in the southeastern Bering Sea) were significantly lower than at Little Diomede Island ($p < 0.05$) in the Bering Strait (Figure 4). A review of the available literature on Hg in seabirds suggests that there is not a clear geographic pattern throughout the arctic (*1*). However, in Canada there is a trend of increasing Hg in seabirds from the subarctic and low arctic to the high arctic (*7*), which is consistent with the pattern we observed here.

One possible mechanism for higher Hg in biota from these more remote arctic ecosystems relates to the specific combination of photochemical conditions during polar sunrise at high latitudes. These conditions allow gaseous elemental Hg (GEM) to be transformed into reactive gaseous Hg (RGM), which can then be rapidly deposited during atmospheric mercury depletion episodes (*3*, *4*). Little Diomede Island is located in the Bering Strait on the southern edge of the arctic where high levels of atmospheric RGM have been measured during the spring (*3*) when murres return to their breeding grounds to forage and nest. Analysis of Hg in the Alaskan snowpack also showed higher Hg in snow along the coast of the Arctic Ocean compared to the Bering Sea (*39*). Future collection and analyses of murre eggs from higher arctic colonies in the Chukchi Sea (Cape Thompson and Cape Lisburne) will help verify this trend of increasing Hg from south to north. Measuring atmospheric Hg deposition concurrently with the murre breeding season along this north−south transect will be required to corroborate the hypothesis that this trend is related to enhanced atmospheric input through mercury depletion episodes.

Additional data are also needed to determine how food web differences among colonies might influence Hg accumulation. Species diversity and food web complexity tend to decrease as one moves northward into more stressful arctic habitats, resulting in generally shorter food chains (*40*), which in turn lower the ultimate biomagnification factor for top predators (*41*). Zooplankton density may also relate to MeHg bioaccumulation in planktivorous fish (*42*), and murre prey species may vary from colony to colony (*43*). Carbon and nitrogen stable isotope analyses will be applied to archived and future samples to address the influence of these factors.

Species Differences. These data show that there are distinct differences in egg Hg burdens between common and thick-billed murres and that the observed patterns vary depending on colony. Thick-billed murres were the more highly contaminated of the two species in the Bering Sea while common murres had higher Hg levels in the Gulf of Alaska. There is currently no data to explain the mechanism behind this trend, but differences in their foraging ecology are apparently adequate to cause significant variation in the trophic transfer of Hg to these two species. Common murres tend to be more stenophagic, primarily targeting pelagic and semi-pelagic age +0 y to +1 y fish including capelin (*Mallotus villosus*), walleye pollock (*Theragra chalcogramma*), cod (Gadidae), and Pacific sand lance (*Ammodytes hexapterus*). Although thick-billed murres also prey heavily on these species, they are known to dive deeper and take more benthic fish and invertebrates, including sculpin (Cottidae), pricklebacks (Stichaeidae), small flatfish (Pleuronectidae), shrimp (e.g. Pandalidae and Crangonidae), squid (Cephalopoda), euphausiids (*Thyssanoesa* spp.), amphipods (Amphipoda), and annelids (e.g. *Nereis* spp.) (*44*−*51*). Differences in habitat utilization have also been documented in some areas, with thick-billed murres tending to forage farther from shore than common murres (*18*, *51*, *52*).

The observed reversal in the relative contaminant levels between these species in the two regions suggests that the effects of these differences in foraging ecology on Hg uptake may vary depending on the particular fish and invertebrate assemblages and habitat that occur near the sampling sites. This reiterates the importance of obtaining complimentary data on murre diets, Hg content of prey, and stable isotope signatures to better understand the processes driving these patterns and improve comparability of data. While the mechanisms driving the above differences are not yet known, the current data suggests that even sympatric thick-billed and common murres have sufficient differences in Hg levels to preclude using these species interchangeably for monitoring based on availability and distribution.

Temporal Variability. One of STAMP's primary goals is to collect and analyze seabird eggs to provide baseline contaminant data for long-term temporal trends and to archive subsamples to allow future retrospective studies. Since collections for STAMP began in 1999, the initial effort has focused on investigating geographic variability and determining suitable sites for long-term monitoring, therefore multiyear data for the same species are currently available from only one colony. These preliminary data show that Hg concentrations were significantly lower in common murre eggs collected from St. Lazaria Island in 2001 compared to 1999 (Figure 5). From a statistical standpoint, this annual variability is an important factor to consider when designing a monitoring program intended to track long-term trends. It still needs to be determined what the driving forces behind the pattern detected here are to validate the interpretation. Since wet deposition is strongly correlated to precipitation (*36*), significant annual variability in Hg input based on weather patterns should be expected. The 1998 El Niño did create a warmer, wetter winter in the Gulf of Alaska (*33*), but a concomitant decrease in egg lipid content from 12.3% in 1999 to 10.5% in 2001 ($p < 0.0001$) suggests possible dietary differences may exist. Thick-billed murres may shift prey species seasonally and annually based on availability (*44*, *49*, *53*), so a shift in prey quality must be considered. A study on seabird forage fish in the northern Gulf of Alaska found high variability in the energy density among species, and a positive correlation between lipid and protein content (*54*), which

could relate to Hg levels. Understanding temporary and/or long-term shifts in the structure of Alaskan marine communities and climate will be important considerations for interpreting temporal trends in contaminants.

Monitoring Strategies. Monitoring contaminant levels in air, sediment, and water alone cannot adequately predict health risks to wildlife and humans, and these matrixes have certain biases and limitations for monitoring (*37*). Using biota for monitoring contaminants in the environment can provide researchers with single contaminant data points that integrate exposure over time, space, and multiple trophic levels. However variation also occurs in indicator species due to other inherent sources of sampling biases resulting from life-history related differences (sex, age, migration, distribution, foraging ecology, etc.) among individuals and populations. Monitoring spatial and temporal patterns of Hg using matrixes such as muscle or liver tissues that serve as long-term storage sites for Hg also make it difficult to establish the spatial and temporal scales the measured tissue concentrations are integrating to allow a valid interpretation of the data. Therefore species and matrixes for monitoring must be selected with care.

To some extent using eggs for biomonitoring is also vulnerable to these types of biases; however, these data taken together with other egg monitoring programs and experimental data suggest eggs may serve as effective Hg monitoring units that more closely reflect a discrete period of Hg exposure, are geographically distinct, and have low sampling variability relative to other matrixes. High analytical costs often preclude increasing sample sizes to compensate for large amounts of variability in the system. To maximize the detection of environmental trends, long-term monitoring programs must be designed to balance these factors and, whenever possible, employ high-accuracy techniques to reduce compounding environmental and analytical variability. The potential influence of the observed annual variability on the ability to detect long-term trends will be formally addressed through repetitive collections from representative colonies from each major oceanographic region. These data can then be incorporated into a power analysis to determine the monitoring design that provides the appropriate balance between the ability to detect trends while maintaining an economical scale of sample collection and analysis.

One of the primary goals of STAMP is to monitor the effect of changing Hg emissions on Hg levels in the environment over a decadal time scale. The limited number of local anthropogenic Hg sources in Alaska makes this an appropriate environment for tracking changes in global levels of Hg. This and concerns that polar regions may serve as a global sink for Hg makes establishing long-term monitoring programs in this region a necessity. These data demonstrate that murre eggs are sensitive indicators of geographic and temporal trends in Hg. It is also clear that understanding relationships among egg contaminant levels, murre ecology, the marine food web, atmospheric deposition, climate, and Hg cycling will be required to accurately interpret the mechanisms behind the detected trends. Initially applying a more integrative approach to validate data interpretation, and normalizing Hg measurements by covariates such as $\delta^{15}N$ or protein content in archived and future samples, will improve long-term comparability of data and maximize the impact of this research.

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Literature Cited

(1) AMAP Assessment Report: *Arctic Pollution Issues*; Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, 1998.

(2) AMAP Assessment Report: *Arctic Pollution*; Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, 2002.

(3) Skov, H.; Christensen, J. H.; Goodsite, M. E.; Heidam, N. Z.; Jensen, B.; Wahlin, P.; Geernaert, G. Fate of elemental mercury in the Arctic during atmospheric mercury depletion episodes and the load of atmospheric mercury to the Arctic. *Environ. Sci. Technol.* **2004**, *38*, 2373−2382.

(4) Lindberg, S. E.; Brooks, S.; Lin, C. J.; Scott, K. J.; Landis, M. S.; Steven, R. K.; Goodsite, M.; Richter, A. Dynamic oxidation of gaseous mercury in the Arctic troposphere at polar sunrise. *Environ. Sci. Technol.* **2002**, *36*, 1245−1256.

(5) Loseto, L. L.; Lean, D. R.; Siciliano, S. D. Snowmelt sources of methylmercury to high arctic ecosystems. *Environ. Sci. Technol.* **2004**, *38*, 3004−3010.

(6) Evers, D. C.; Taylor, K. M.; Major, A.; Taylor, R. J.; Poppenga, R. H.; Scheuhammer, A. M. Common loon eggs as indicators of methylmercury availability in North Amercia. *Ecotoxicology* **2003**, *12*, 69−81.

(7) Braune, B. M.; Donaldson, G. M.; Hobson, K. A. Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable isotopes for intercolony differences. *Environ. Pollut.* **2002**, *117*, 133−145.

(8) Braune, B. M.; Donaldson, G. M.; Hobson, K. A. Contaminant residues in seabird eggs from the Canadian Arctic. Part I. Temporal trends 1975−1998. *Environ. Pollut.* **2001**, *114*, 39−54.

(9) Wiener, J. G.; Krabbenhoft, D. P.; Heinz, G. H.; Scheuhammer, A. M. In *Handbook of Ecotoxicology*; Hoffman, D. J., Rattner, B. A., Burton, G. A., Jr., Cairns, J., Jr., Eds.; Lewis Publishers: New York, 2003; pp 409−463.

(10) AMAP Scientific Experts Workshop, Girdwood, Alaska, 1998.

(11) Hebert, C. E. Winter severity affects migration and contaminant accumulation in northern Great Lakes herring gulls. *Ecol. Appl.* **1998**, *8*, 669−679.

(12) Kambamamonli-Dimou, A.; Kamarianos, A.; Kilikidis, S. Transfer of methylmercury to hens' eggs after oral administration. *Bull. Envron. Contam. Toxicol.* **1991**, *46*, 128−133.

(13) March, B. E.; Soong. R.; Blinski, E.; Jonas, R. E. Effects on chickens of chronic exposure to mercury at low levels through dietary fish meal. *Poultry Sci.* **1974**, *53*, 2175−2181.

(14) Sell, J. L.; Guenter. W.; Sifri. M. Distribution of mercury among components of eggs following the administration of methylmercuric to chickens. *J. Agr. Food. Chem.* **1974**, *22*, 248−251.

(15) Burgess, N. M.; Evers, J. D.; Kaplan, J. D. Mercury levels in the blood of common loons breeding in the Maritimes and their prey. In *Mercury in Atlantic Canada: A Progress Report*; Environment Canada: New Brunswick, Canada, 1998; pp 96−100.

(16) Evers, D. C.; Lane. O. P.; De Sorbo, C.; Savoy, L. Assessing the impacts of methylmercury on piscivorous wildlife using a wildlife criterion value based on the common loon, 1998−2001. In *Report BRI2002-08 submitted to the Maine Department Environmental Protection*; BioDiversity Research Inst.: Falmouth, Maine, 2002.

(17) Hatch, S. A.; Meyers, P. M.; Mulcahy, D. M.; Douglas, D. C. Seasonal movements and pelagic habitat use of murresand puffins determined by satellite telemetry. *Condor* **2000**, *154*, 102−145.

(18) Cramp, S. Ed. *Handbook of the Birds of Europe, the Middle East, and North Africa*; Oxford University Press: New York, 1985; Vol. IV, p 170.

(19) Morera, M.; Sanpera, C.; Crespo, S.; Jover, L.; Ruiz, X. Inter- and intra-clutch variability in heavy metals and selenium levels in Audouin's gull eggs from the Ebro Delta, Spain. *Arch. Environ. Contam. Toxicol.* **1997**, *33*, 71−75.
(20) Becker, P. H. Egg mercury levels decline with the laying sequence in Charadriformes. *Bull. Environ. Contam. Toxicol.* **1992**, *48*, 762−767.
(21) Zeisler, R.; Langland, J. K.; Harrison, S. H. Cryogenic homogenization of biological tissues. *Anal. Chem.* **1983**, *55*, 2431−2434.
(22) Christopher, S. J.; Vander Pol, S. S.; Pugh, R. S.; Day, R. D.; Becker, P. R. Determination of mercury in the eggs of common murres (*Uria aalge*) for the seabird tissue archival and monitoring project. *J. Anal. At. Spectrom.* **2002**, *17*, 780−785.
(23) Davis, W. C.; Vander Pol, S. S.; Schantz, M. M.; Long, S. E.; Day, R. D.; Christopher, S. J. An accurate and sensitive method for the determination of methylmercury in biological specimens using GC−ICP-MS with solid-phase microextraction. *J. Anal. At. Spectrom.* **2004**, *19*, 1546−1551.
(24) Jarman, W. M.; Hobson, K. A.; Sydeman, W. J.; Bacon, C. E.; McLaren, E. B. Influence of trophic position and feeding location on contaminant levels in the Gulf of the Farallones food web revealed by stable isotope analysis. *Environ. Sci. Technol.* **1996**, *30*, 654−660.
(25) Sydeman, W. J.; Jarman, W. M. Trace metals in seabirds, steller sea lion, and forage fish and zooplankton from central California. *Mar. Pollut. Bull.* **1998**, *36*, 828−832.
(26) Barrett, R. T.; Skaare, J. U.; Gabrielson, G. W. Recent changes in levels of persistent organochlorines and mercury in eggs of seabirds from the Barents Sea. *Environ. Pollut.* **1996**, *92*, 13−18.
(27) Barrett, R. T.; Skaare, J. U.; Norheim, G.; Vader, W.; Froslie, A. Persistent organochlorine and mercury in eggs of Norwegian seabirds 1983. *Environ. Pollut., Ser. A* **1985**, *39*, 79−93.
(28) Fimreite, N.; Brun, E.; Froslie, A.; Frederichsen, P.; Gundersen, N. Mercury in eggs of Norwegian seabirds. *Astarte* **1974**, *7*, 71−75.
(29) Hall, A. S.; Teeny, F. M.; Lewis, L. G.; Hardman, W. H.; Gauglitz, E. J., Jr. Mercury in fish and shellfish of the northeast Pacific. I. Pacific Halibut, *Hippoglossus stenolepis*. *Fish. Bull.* **1976**, *74*, 783−789.
(30) Hall, A. S.; Teeny, F. M.; Gauglitz, E. J., Jr. Mercury in fish and shellfish of the Northeast Pacific. II. Sablefish, *Anaplopoma fimbria*. *Fish. Bull.* **1976**, *74*, 791−797.
(31) Vander Pol, S. S.; Becker, P. R.; Kucklick, J. R.; Pugh, R. S.; Roseneau, D. G.; Simac, K. S. Persistent organic pollutants in Alaskan murre (*Uria* spp.) eggs: geographical, species, and temporal comparisons. *Environ. Sci. Technol.* **2004**, *38*, 1305−1312.
(32) Anas, R. E. Heavy metal in the northern fur seal, *Callorhinus ursinus*, and harbor seal, *Phoca vitulina richardi*. *Fish. Bull.* **1974**, *72*, 133−137.
(33) Stabeno, P. J.; Bond, N. A.; Hermann, A. J.; Kachel, N. B.; Mordy, C. W.; Overland, J. E. Meteorology and oceanography of the northern Gulf of Alaska. *Contl. Shelf Res.* **2004**, *24*, 859−897.
(34) Miller, E. K.; Vanarsdale, A.; Keeler, G. J.; Chalmers, A.; Poissant, L.; Kamman, N. C.; Brulotte, R. Estimation and mapping of wet and dry mercury deposition across northeastern North America. *Ecotoxicology* **2005**, *14*, 53−70.
(35) Grigal, D. F. Inputs and outputs of mercury from terrestrial watersheds: a review. *Environ. Rev.* **2002**, *10*, 1−39.
(36) Sakata, M.; Marumoto, K. Wet and dry deposition fluxes of mercury in Japan. *Atmos. Environ.* **2005**, *39*, 3139−3146.
(37) Mason, R. P.; Abbott, M. L.; Bodaly, R. A.; Bullock, O. R., Jr.; Driscoll, C. T.; Evers, D.; Lindberg, S. E.; Murray, M.; Swain, E. B. Monitoring the response to changing mercury deposition. *Environ. Sci. Technol.* **2005**, *39*, 14A-22A.
(38) U.S. EPA. Mercury transport and fate in watersheds. *Star Report, National Center for Environmental Research*; U.S. EPA: Washington, DC, 2000; Vol. 4, pp 1−8.
(39) Douglas, T. A.; Sturm, M. Arctic haze, mercury and the chemical composition of snow across northwestern Alaska. *Atmos. Environ.* **2004**, *38*, 805−820.
(40) Barrie, L. A.; Gregor, D.; Hargrave, B.; Lake, R.; Muir, D.; Shearer, R.; Tracey, B.; Bidleman, T. Arctic contaminants: sources, occurrence and pathways. *Sci. Total Environ.* **1992**, *122*, 1−74.
(41) Rasmussen, J. B.; Rowan, D. J.; Lean, D. R.; Carey, J. H. Food chain structure in Ontario Lakes determines PCB levels in Lake Trout (*Salvelinus namaycush*) and other pelagic fish. *Can. J. Fish. Aquat. Sci.* **1990**, *47*, 2030−2038.
(42) Chen, C. Y.; Folt, C. L. High plankton densities reduce mercury biomagnification. *Environ. Sci. Technol.* **2005**, *39*, 115−121.
(43) Gaston, A. J.; Bradstreet, M. S. Intercolony differences in the summer diet of thick-billed murres in the eastern Canadian Arctic. *Can. J. Zool.* **1993**, *71*, 1931−1840.
(44) Springer, A. M.; Roseneau, D. G.; Lloyd, D. S.; McRoy, C. P.; Murphy, E. C. Seabird responses to fluctuating prey availability in the eastern Bering Sea. *Mar. Ecol. Prog. Ser.* **1986**, *32*, 1−12.
(45) Springer, A. M.; Murphy, E. C.; Roseneau, D. G.; McRoy, P.; Cooper, B. A. The paradox of pelagic food webs in the northern Bering Sea-I. Seabird food habits. *Cont. Shelf Res.* **1987**, *7*, 895−911.
(46) Barrett, R. T.; Asheim, M.; Bakken, V. Ecological relationships between two sympatric congeneric species, common murres and thick-billed murres, *Uria aalge* and *U. lomvia*, breeding in the Barents Sea. *Can. J. Zool.* **1997**, *75*, 618−631.
(47) Roseneau, D. G.; Chance, M. F.; Chance, P. F.; Byrd, G. V. Monitoring seabird populations in areas of oil and gas development on the Alaskan continental shelf: Cape Lisburne and Cape Thompson seabird studies, 1995−1997. USGS/BRD/CR-1999−0002 Final Report. 2000.
(48) Swartz, L. G. Sea-cliff birds in *Environment of the Cape Thompson Region, Alaska, Book 2*; Willimovsky, N. J., Wolfe, J. N.; Eds.; U.S. Atomic Energy Commission, 1966; pp 611−668.
(49) Springer, A. M.; Roseneau, D. G.; Murphy, E. C.; Springer, M. I. Environmental controls of marine food webs: food habits of seabirds in the Easter Chukchi Sea. *Can. J. Fish. Aquat. Sci.* **1984**, *41*, 1202−1215.
(50) Dragoo, D. E. Food habits and productivity of kittiwakes and murres at St. George Island, Alaska. Master thesis, University of Alaska, 1991.
(51) Gaston, A. J.; Jones, I. L. The Auks. In *Bird Families of the World*, 4; Oxford University Press: New York, 1998; pp 133−160.
(52) Swartz, L. G. Distribution and movements of birds in the Bering and Chukchi seas. *Pac. Sci.* **1967**, *21*, 332−347.
(53) Carscadden, W. A.; Montevecchi, W. A.; Davoren, G. K.; Nakashima, B. S. Trophic relationships among capelin (Mallotus villosus) and seabirds in a changing ecosystem. *J. Mar. Sci.* **2002**, *59*, 1027−1033.
(54) Anthony, J. A.; Roby, D. D.; Turco, K. R. Lipid content and energy density of forage fishes from the northern Gulf of Alaska. *J. Exp. Mar. Biol. Ecol.* **2000**, *248*, 53−78.

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