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Determination of mercury in the eggs of common murres (*Uria aalge*) for the seabird tissue archival and monitoring project†

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Received 23rd May 2002, Accepted 21st June 2002
First published as an Advance Article on the web 4th July 2002

An analytical method using isotope dilution cold vapor inductively coupled plasma mass spectrometry (ID-CV-ICPMS) was developed for the determination of total mercury in the eggs of seabirds. Components including error magnification, verification of method accuracy and assignment of analytical uncertainty are presented in the context of collecting mercury data for single sample aliquots. Forty-one egg samples collected from common murre (*Uria aalge*) colonies on Little Diomede and Saint George Islands in the Bering Sea and East Amatuli and Saint Lazaria Islands in the Gulf of Alaska yielded mercury mass fraction values ranging from approximately 0.010 μg g^{-1} to 0.360 μg g^{-1}. Relative expanded uncertainties for the individual determinations ranged from 1.2% to 4.4%. A one-way analysis of variance including pairwise comparisons across the colonies showed that mercury levels in eggs collected from the Gulf of Alaska colonies were significantly higher than their counterparts in the Bering Sea. Mercury data from each colony were normally distributed, suggesting a ubiquitous regional deposition of mercury and corresponding incorporation into local foodwebs.

Introduction

The transport and fate of airborne mercury (Hg) species in marine and coastal environments is an active area of research. The atmospheric chemistry responsible for the ultimate wet and dry deposition of reactive Hg species into important regional areas such as Alaska and the Arctic has been extensively studied.[1–6] Large quantities of Hg are preferentially deposited into high latitude landmasses over relatively short periods of time. This is due to a net flux of atmospheric contaminants from lower latitudes and the well-documented polar sunrise effect,[2,3] that causes rapid depletion of atmospheric Hg. Wet- or dry-deposited Hg species are then readily washed into the coastal zone in high concentrations during rain and snowmelt events. This complex combination of atmospheric Hg chemistry, meteorology, terrain and Hg chemistry in the coastal zone will control the bioavailability and accumulation of Hg into marine food webs, impacting high trophic level species such as seabirds and marine mammals[7–10] and humans.[11,12]

Seabird tissues, including eggs, have been used to monitor mercury temporally and geographically since the late 1960's.[13] Braune and coworkers have recently shown that modern analytical protocols for mercury determination can provide new insight into seabird egg contaminant trends, through reanalysis of archived samples and coupling this data with data obtained for new samples.[14] Collecting new contaminant data for sentinel species such as seabirds helps underpin the models that describe the mercury cycling processes occurring in Alaska and the Arctic and the understanding of the impact of Hg contamination on wildlife and human health.

Numerous agencies and research organizations are involved in mercury measurement. The scientific integration of these measurements requires quality assurance in terms of accuracy and sampling protocols that ensure representativeness. Thus, research institutions benefit from implementing structured and well-documented sample collection and archival protocols (to maintain specimen quality control) and benchmarking their environmental Hg determinations for quality. Structured sample collection and archival allows for confident temporal analysis of environmental contaminants. Accurate contaminant measurements yield reliable and useful scientific data for researchers studying environmental Hg cycling and provide solid risk assessment data for wildlife health managers and native subsistence populations that rely upon their local environment as a valuable food source.

This paper describes the development of an analytical method using isotope dilution cold vapor inductively coupled plasma mass spectrometry (ID-CV-ICPMS).[15] This recently developed primary technique has been shown to be highly accurate. We have uniquely applied this technique to the determination of Hg content in the eggs of common murres (*Uria aalge*). The relative total uncertainty of the measurements is estimated to be better than 5% in the range of 0.010 μg g^{-1} to 0.360 μg g^{-1} observed in these samples. Method reproducibilities at the high and low ends of this range were 0.64% and 1.25%, respectively (1*s*, $n = 4$).

The national marine analytical quality assurance program and the seabird tissue archival monitoring project

The National Institute of Standards and Technology (NIST) plays an active role in contaminant research in the marine environment through the National Marine Analytical Quality Assurance Program (NMAQAP). This program was established in 1995 through an agreement with the National Oceanic and Atmospheric Administration (NOAA). The NMAQAP, which is conducted by the NIST Chemical Science and Technology Laboratory's Analytical Chemistry Division, focuses on marine environmental quality assurance activities. These activities include the cryogenic banking of marine environmental specimens, the development of reference and control materials specific to marine matrices, the administration and coordination of interlaboratory comparison exercises, and the development and exportation of high-accuracy methods for the measurement of trace element and organic contaminants in marine samples.

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 DOI: 10.1039/b205018h

The Seabird Tissue Archival and Monitoring Project (STAMP) is conducted jointly by the US Geological Survey's Biological Resources Division (USGS/BRD), US Fish and Wildlife Service's Alaska Maritime National Wildlife Refuge (USFWS/AMNWR) and NIST. The purpose of STAMP is to document long-term trends in environmental quality through the collection of tissues (including eggs) of Alaskan colonial seabirds, archive these specimens under conditions that ensure chemical stability during long-term storage (decades), and analyze aliquots of these specimens for anthropogenic contaminants. Temporal and spatial monitoring data for anthropogenic contaminants, including persistent organic pollutants and mercury, can be used to assess the impact contaminants have on Alaskan wildlife and human health. Samples are processed under stringent protocols and maintained in conditions that are suitable for long-term archival. Retrospective studies are afforded through an infrastructure of analytical support, cryogenic storage, database and cleanroom facilities. The sample collection plan for STAMP includes collecting specimens from several seabird colonies over tens of islands dispersed throughout AMNWR, which covers some 18,000 square kilometers in total. Targeted bird species for STAMP include the common murre (*Uria aalge*), thick-billed murre (*Uria lomvia*), black guillemot (*Cepphus grylle*), fork tail storm petrel (*Oceanodroma furcata*) and black-legged kittiwake (*Rissa tridactyla*).

In this study, 41 eggs were cryogenically homogenized using a method described by Zeisler and coworkers[16] and aliquots of these 41 homogenates were analyzed for Hg content. Additionally, the samples have been analyzed for organochlorine pesticides and polychlorinated biphenyls (PCBs).[17] The eggs were collected from four colonies depicted geographically in Fig. 1 and represent the entire 1999 STAMP collection of common murre eggs cryogenically archived in the National Biomonitoring Specimen Bank at NIST Charleston. Nine and 11 samples respectively, are from Little Diomede and Saint George Islands in the Bering Sea, and 10 and 11 samples, respectively, are from Saint Lazaria and East Amatuli Islands in the Gulf of Alaska. The murre eggs from the four colonies were collected by USFWS personnel. Alaskan Natives periodically harvest murre eggs for consumption and local villagers actually assisted USFWS with the egg collection on Little Diomede Island.

Experimental

Reagents

High-purity nitric acid was purchased from Fisher Scientific (Suwanee, GA). Tin chloride, hydrochloric acid and potassium dichromate were purchased from JT Baker (Phillipsburg, NJ). For the isotope dilution experiments, a 100 ng g^{-1} mercury spike solution in 10% (mass fraction) HNO_3 was created from 98.11% ^{201}Hg enriched solid mercury(II) oxide obtained from Oak Ridge National Laboratory (Oak Ridge, TN). NIST Standard Reference Material SRM 3133 Mercury Spectrometric Solution obtained from NIST (Gaithersburg, MD) was used as a calibrant. All sample and standard solutions were diluted with high quality water obtained from a Millipore (Bedford, MA) deionization station capable of producing 18 MΩ cm resistivity water.

Sample preparation and spike calibration

Samples were run in 12 analytical batches of six samples, each of which consisted of four eggs, one control sample and one method blank. The sample dissolution procedures used multiple iterations of microwave digestion. For each digestion batch, approximately 0.9 g sample aliquots of homogenized egg tissue were digested along with one approximately 0.6 g control sample aliquot of SRM 2976 Mussel Tissue (Trace Elements and Methylmercury) and a procedural blank. The certified value for total Hg in SRM 2976 is 0.0610 ± 0.0036 μg g^{-1}. Each analytical sample was spiked with a known quantity of isotopically enriched mercury (^{201}Hg enriched isotopic spike) prior to addition of the 5 mL nitric acid decomposition medium. The amount of spike delivered to the blank vessels was reduced to curtail errors due to overspiking. All samples were digested in a Perkin-Elmer (Shelton, CT) Multiwave® microwave oven at the highest possible temperatures (up to 300 °C) and pressures (up to 8 MPa) in order to equilibrate the spiked mercury with the natural mercury present in the samples. The resulting digests were vented and diluted with high purity water to a total volume of approximately 40 mL and non-quantitatively transferred into 60 mL polyethylene bottles. Further sample dilutions ranging from 1 : 3 to 1 : 40 were required prior to analysis.

SRM 3133 Mercury Spectrometric Solution was used to calibrate the isotopic spike solution prior to its use. First, an

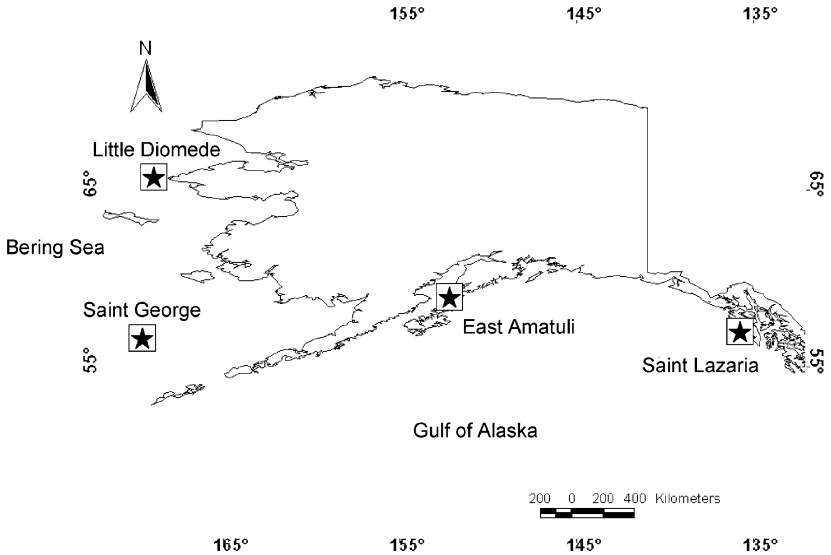

Fig. 1 Outline map of Alaska depicting the egg sample collection sites for common murre colonies at Saint George, Little Diomede, East Amatuli and Saint Lazaria Islands.

approximately 100 ng g^{-1} ^{201}Hg isotopic spike solution was prepared using 3% (mass fraction) HNO_3 and 0.5% (mass fraction) $K_2Cr_2O_7$ as the diluent to reduce loss of elemental mercury. Two approximately 100 ng g^{-1} natural mercury solutions were quantitatively prepared from SRM 3133 in 3% (mass fraction) HNO_3 only. The natural and enriched Hg solutions were then mixed (by mass) to obtain four spike calibration solutions having a target $^{201}Hg/^{202}Hg$ ratio of 2 : 1. Thus, the mean spike concentration obtained from the four spike calibration mixes was used as the working concentration for the spike solution. The spike solution was periodically re-calibrated with freshly prepared SRM 3133 as even the $K_2Cr_2O_7$ stabilizer could not indefinitely postpone the liberation of elemental Hg from the spike solution. The variability in the spike calibration (expressed as the percent relative standard deviation of the working spike concentration $\pm$ 1*s*) as calculated from four spike calibration mixes for $n = 5$ separate spike calibration experiments was 0.22 $\pm$ 0.03%.

ID-CV-ICPMS measurements

The mercury reduction chemistry and sample introduction system has been described previously[15] and is only briefly summarized here. The mercury in each sample solution was reduced to elemental Hg using a reductant solution of 10% (mass fraction) $SnCl_2$ in 7% (mass fraction) HCl in water. A gas–liquid separator was used to strip the Hg from the sample digest solutions using a stream of Ar gas (approximately 250 mL min^{-1}). Delivery of Hg^0 to the mass spectrometer was achieved by plumbing the gas output of the gas–liquid separator into the ICPMS injector line. A mass flow controller (AALBORG Model GFC 171, Greenwich, CT), controlled with LabView™ software and National Instruments (Austin, TX) data acquisition hardware, regulated gas flow through the gas–liquid separator.

Numerous $^{201}Hg/^{202}Hg$ isotope ratio pairs were collected for all calibration and analytical samples using a Thermo Elemental PQ3 ICPMS operating in the time resolved analysis mode. The ICP power was maintained at 1350 W forward power and Ar gas flow rates were 13.5 L min^{-1}, 0.85 L min^{-1} and 0.81 L min^{-1} for the coolant, auxiliary and injector lines, respectively. Each analytical run produced a temporal profile that consisted of a 240 s data collection window. Measurements during the first 20 s to 40 s of the profile when a 5% (mass fraction) HNO_3 wash solution (and not a sample) was present in the gas–liquid separator established the reference baselines. A typical isotope–time profile is depicted in Fig. 2 for a spiked egg sample from East Amatuli Island. The mean of eight baseline-corrected 10 s integration windows was used to establish the measured $^{201}Hg/^{202}Hg$ isotope ratio for all spike calibration, blank, control and unknown samples. All data were corrected for mass discrimination and detector dead time using the method outlined by Vanhaecke and coworkers.[18]

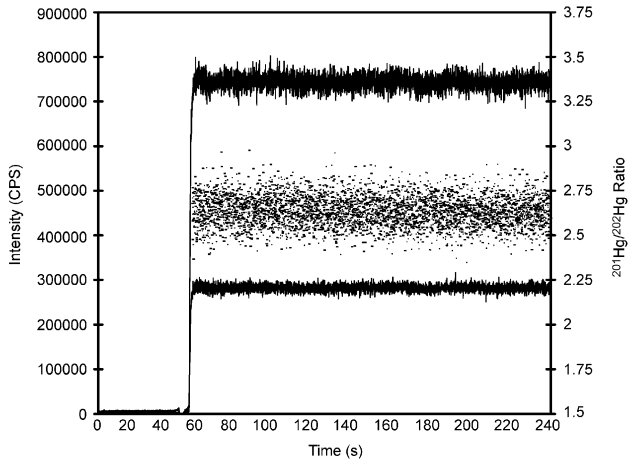

Fig. 2 Transient profiles for ^{201}Hg (top trace) and ^{202}Hg (bottom trace) and overlay of corresponding $^{201}Hg/^{202}Hg$ isotope ratio pairs for a ^{201}Hg spiked common murre egg sample.

Results and discussion

Isotope dilution method

Isotope dilution techniques such as ID-CV-ICPMS for Hg determinations are often used at NIST for high-accuracy measurements needed for the certification of analytes in NIST Standard Reference Materials. Analytical uncertainties are robustly determined and the precision of the technique allows heterogeneity of reference material batches to be assessed. However, many laboratories view isotope dilution methods unsuited for routine environmental measurements that typically feature large sample numbers and wide-ranging differences in analyte concentration. For example, the natural variation of Hg mass fraction in the 41 common murre eggs studied here ranges from approximately 0.010 μg g^{-1} to 0.360 μg g^{-1}, making it challenging to design an optimal ID experiment for each "unique" sample. A discussion follows that considers the use of ID-CV-ICPMS under such conditions.

Error magnification and ICPMS isotope ratio measurements

The ID-CV-ICPMS experiments involve measuring $^{201}Hg/^{202}Hg$ isotopic ratios of spiked samples. The uncertainty in ratio measurement is propagated non-linearly in the calculated concentration; the sensitivity factor is called the "error magnification factor." Ideally, a sample should be spiked with the amount of ^{201}Hg that minimizes the error magnifaction factor. For the ^{201}Hg spike used here, the error magnification factor is plotted against isotope dilution $^{201}Hg/^{202}Hg$ ratios in Fig. 3. The error magnification blows up if the sample is either "underspiked" or "overspiked".[19] The effective dynamic range for spiking and ratio measurement (where the error magnification factor is between 1.2 and 2) extends over roughly a range from 0.8 to 50. Practically, there are other considerations that favor ratio measurements of 1.00. Fig. 4 shows the measured $^{201}Hg/^{202}Hg$ ratios collected for all batches of unknown, blank and control samples in the form of a radar plot. The experiment was designed so that most of the samples were spiked at a $^{201}Hg/^{202}Hg$ ratio between 1.5 and 10, coinciding with the lowest part of the error magnification curve in Fig. 3. Allowing for some variability in the isotope dilution ratios greatly improves the ability to automate the spiking process, using the same quantity of spike aliquot for each sample, with the exception of the blank, where the size of the aliquot is reduced to minimize the error magnification factor.

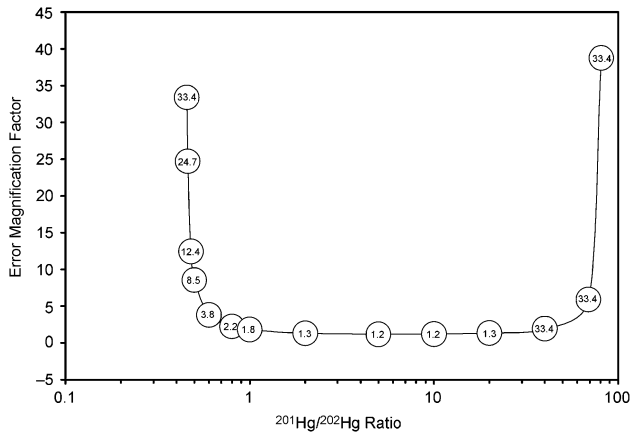

Fig. 3 Error magnification factor as a function of $^{201}Hg/^{202}Hg$ isotope dilution ratio.

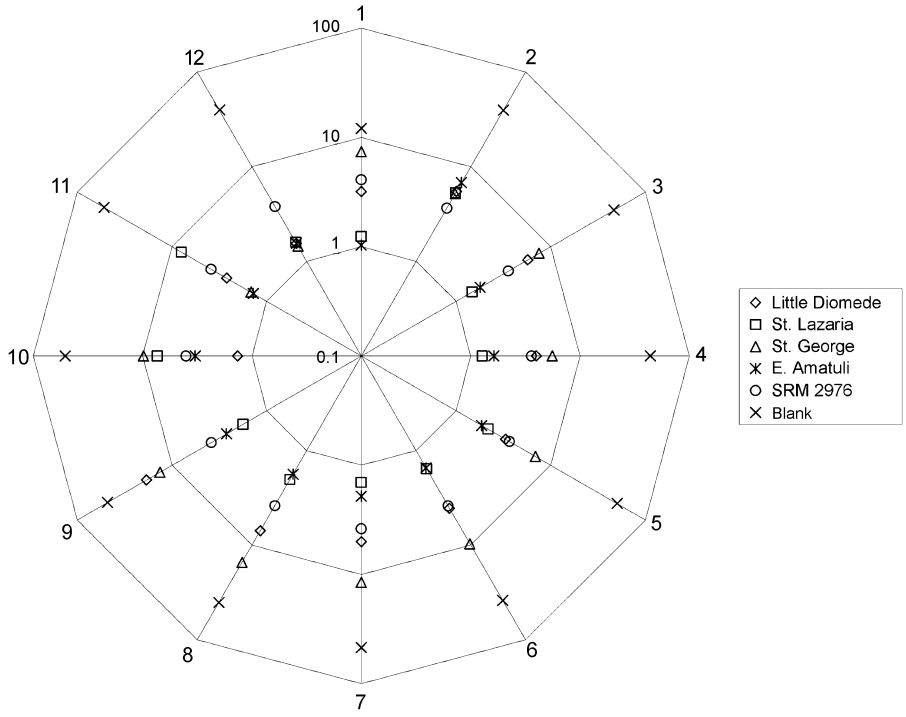

Fig. 4 Radar plot showing all measured $^{201}Hg/^{202}Hg$ isotope dilution ratios (log axis) for each sample batch.

Nonetheless, the blank samples remain overspiked indicating that there is only a small amount of contamination introduced from the analytical method. Blank corrections are of the order of approximately 1–4% for most of the egg samples, with a typical blank having an absolute value of approximately 0.3 ng. For some of the lowest Hg content eggs collected from Little Diomede and Saint George Islands, the typical uncertainty of the blank correction (approximately 0.17 ng g^{-1}) is on a par with the uncertainty obtained from the ratio measurement, whereas for the higher Hg content eggs, the uncertainty due to the blank correction is nearly an order of magnitude lower than the ratio measurement uncertainty.

Method accuracy, reproducibility and uncertainty

A control chart for SRM 2976 (Fig. 5) verifies the method accuracy across multiple analysis batches (project duration approximately 3 months). A method reproducibility study was conducted on two egg samples to help establish an uncertainty budget for the remaining eggs that were subjected to only a single determination. Method reproducibility results expressed as percent relative standard deviation (RSD) for single homogenized egg samples from Little Diomede Island (representing a low Hg concentration sample) and East Amatuli Island (representing a high Hg concentration sample) were 1.25 and 0.64%, respectively. Each result is based on four sample aliquots taken through the analytical spiking, weighing, digestion and measurement processes. The measurement reproducibility reflects the precision of ratio measurement, sample homogeneity, and other sources of variability in the isotope dilution method, including weighing precision and sample contamination.

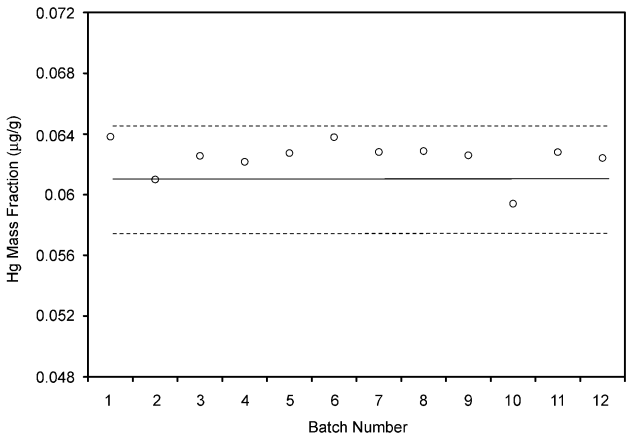

Fig. 5 Control chart for Hg mass fraction (µg g^{-1}) in SRM 2976 Mussel Tissue (Trace Elements and Methylmercury), total Hg certified value = 0.0610 ± 0.0036 µg g^{-1}.

The Hg mass fraction values and corresponding expanded uncertainties for each egg are presented in Fig. 6. The individual components of uncertainty for Hg in each egg sample were determined according to ISO guidelines.[20] Type A uncertainty components included sample measurement, spike calibration and blank correction. Type B uncertainty contributions included weighing measurements on a balance possessing 0.001 g resolution, uncertainty in concentration for the NIST SRM 3133 calibrant and instrument mass discrimination.

The Type A uncertainty contributions for each egg sample were first compiled in relative terms before conversion into absolute mass fraction terms. The reproducibility data presented in the previous paragraph were used to estimate the sample measurement repeatability for each egg sample where only a single measurement was collected ($n = 1$ measurement, 3 degrees of freedom). The RSD of 1.25% was used to estimate the measurement reproducibility for all of the eggs collected on Little Diomede and Saint George islands, *i.e.*, the eggs from the islands in the Bering Sea that possessed low relative Hg content. Similarly, an RSD of 0.64% was used to estimate the measurement reproducibility for all of the eggs collected on East Amatuli and Saint Lazaria Islands, *i.e.*, the eggs from the islands in the Gulf of Alaska that possessed a high relative Hg content. The RSD obtained from the measurement of four spike calibration mixes was used to estimate the contribution of uncertainty from the spike concentration. Finally, the standard deviation for eleven blank measurements was ratioed to each egg's Hg concentration to obtain a blank uncertainty component based on a single

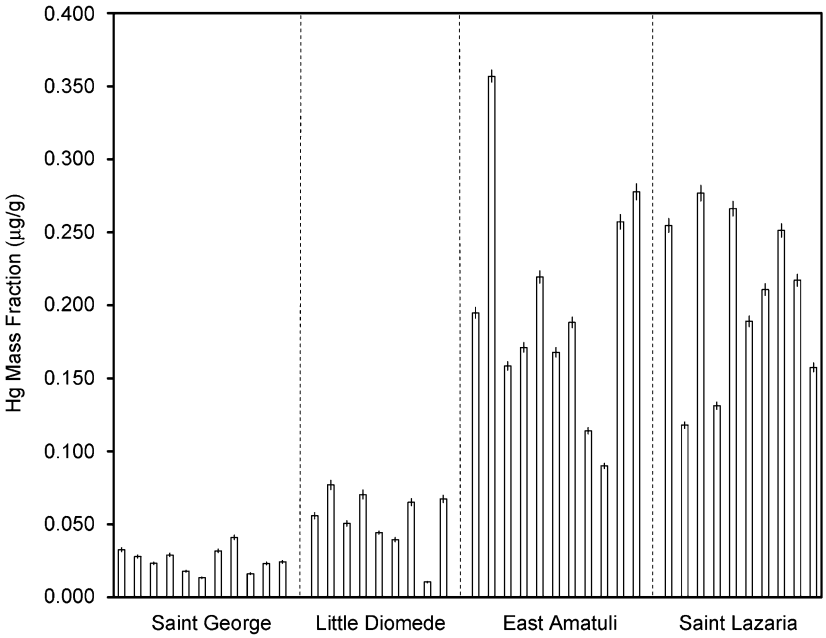

Fig. 6 Mass fraction value (μg g^{-1}) and expanded uncertainty for each individual Hg wet mass fraction determination.

measurement with 10 degrees of freedom for the blank correction.

The Type B uncertainty contributions for each egg sample were also compiled in relative terms before conversion into absolute mass fraction terms. The calibrant certification uncertainty is derived from the expanded uncertainty reported in the certificate of SRM 3133. The reported expanded uncertainty was converted to a standard uncertainty by dividing by 2 and expressed in relative terms by dividing by the concentration of Hg in the SRM. The uncertainty contribution from mass discrimination (0.34% RSD) was derived from experimental biases observed over the course of the project while collecting dead time-corrected $^{201}Hg/^{202}Hg$ isotope ratios for approximately 1 ng g^{-1} solutions of SRM 3133.

Data analysis

Mercury mass fraction data for the eggs collected from each island colony is presented in Fig. 7. The plot shows that the colony means and medians are similar, suggesting that the data are normally distributed, an atypical result for environmental contaminant data. Constructing normal plots (quantile *vs.* Hg mass fraction) for the data and applying the Shapiro-Wilk test for non-normality formally tested the normality assumption.

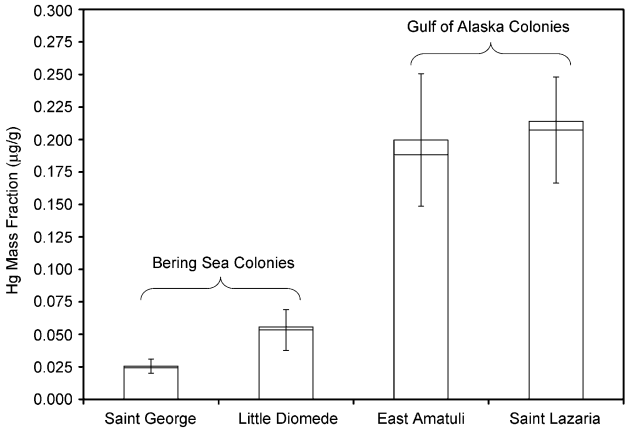

Fig. 7 Mercury mass fraction data mean ± 95% confidence interval and corresponding median plotted as a function of island location.

For example, a normal plot generated for the 11 eggs collected from Saint George Island showed a high degree of linearity ($y = 121.06x - 3.06$, $R^2 = 0.975$). This indication of data normality was also confirmed by the output of the corresponding Shapiro-Wilk test (coefficient = 0.9748 and $p = 0.934$ at 95% confidence level). Applying normality tests to the remaining colonies produced similar results. A one-way analysis of variance ($F = 38.6$, $p < 0.0001$) and pairwise comparisons across the four colonies indicate that the eggs collected from the two colonies in the Gulf of Alaska (Saint Lazaria and East Amatuli Islands) are significantly higher in Hg content than the eggs collected from the two colonies in the Bering Sea (Little Diomede and Saint George Islands).

These interesting results warrant further discussion. Normally distributed data implies that the female birds (and eggs) of a particular colony are exposed to Hg that is ubiquitously deposited and incorporated in local food webs. However, the differences in common murre egg Hg content in the Bering Sea *versus* the Gulf of Alaska imply that either the quantity or bioavailability of regionally deposited Hg is significantly different in the two regions. Much work still needs to be completed before we can definitively identify all of the factors that account for the observed data, but several factors may be influencing the Hg uptake in common murres. Differences in rates of wet and dry deposition of Hg into coastal Alaska should be considered. Mercury deposition into Alaska's terrestrial and aquatic regions will be influenced by meteorology, gas phase chemistry and terrain. The Alaskan coast along the Bering Sea features a relatively flat, rocky coastline when compared to the coastline bordering the Gulf of Alaska, which possesses a more mountainous and heavily forested topography, with higher annual precipitation.[21,22] Thus the propensity for Hg to be deposited through wet and dry scavenging events and washed into Low Arctic wetlands and the coastal zone through precipitation and snowmelt events is necessarily higher in the Gulf of Alaska region. However, the Hg must biomagnify, and more importantly, become bioavailable through Hg methylation processes before it can accumulate in seabirds. Therefore, we speculate that the Hg deposition and Hg methylation rates are greater in the coastal area that includes the Gulf of Alaska relative to the Bering Sea. An increase in Hg methylation efficiency would presumably result from greater microbial activity in a more seasonally temperate climate with higher surface air[23] and water[21] temperatures that possesses a

higher percentage of organic matter at the forest soil–surface water interface.[24] The migration patterns of both the birds and their prey must be considered as well. In the non-breeding season, common murres tend to stay north in their respective habitats,[25,26] so migration of the birds themselves is not likely to account for the differences in Hg content among the Bering Sea and Gulf of Alaska colonies. The roles that prey migration and food web effects have on contaminant uptake in Alaskan common murres need to be studied. Finally, the Alaska Current may also provide a waterborne influx of nutrients and contaminants to the Gulf of Alaska region, which may indirectly impact Hg methylation efficiencies.

Conclusions

Accurate and sensitive protocols and methods for the determination of Hg in seabird eggs have been developed and applied in a routine fashion to obtain Hg contaminant data for STAMP, a seabird contaminants monitoring program. The method of ID-CV-ICPMS was discussed in the context of its application to environmental measurements. Several benefits of the methodology include high accuracy, good method reproducibility, and the ability to estimate errors and uncertainty for a single analytical determination. The accuracy and uncertainty of critical environmental Hg measurements must be verified as this information may ultimately be used by wildlife health assessors to determine the impact of Hg contamination on colonial seabirds in the Alaska Maritime National Wildlife Refuge and by organizations concerned with identifying health issues that impact native Alaskan peoples.

We are in the process of producing total Hg, methylmercury and persistent organic pollutant data for additional STAMP colonies and more seabird species to gain a better understanding of the factors that may be influencing the preferential deposition and uptake of Hg and organic contaminants into colonial seabirds in Alaska. It still needs to be determined if the trends observed here are species-specific or universal to Alaskan colonial seabirds. The International Arctic Monitoring and Assessment Programme (AMAP) has identified alcid eggs as key matrices for contaminant monitoring in northern environments.[27] In a joint effort with our partner organizations (USGS/BRD and USFWS/AMNWR) we are combining our analytical data with information on common murre life history traits, population dynamics, breeding habits and food sources. These findings will be presented elsewhere. These studies will also help add to the current knowledge base of research regarding the fate of Hg in the northern latitudes and the Arctic environment. For example, temporal examination of the ratio of methylmercury to total mercury in seabird egg samples may provide information on Hg deposition and methylation rates across Alaska, complementing current and historical Hg data for Arctic colonial seabirds.[9,14]

Acknowledgement

The authors would like to acknowledge the STAMP project leaders, Geoff W. York (USGS/BRD Alaska Biological Science Center, Anchorage, AK) and David G. Roseneau (USFWS/AMNWR, Homer, AK). In addition, Vernon Byrd (USFWS) and Lyman Thorsteinson (USGS/BRD) were instrumental in the design and development of the project. Individuals responsible for collecting and processing eggs in Alaska include: Kristin Simak (USGS/BRD), Laura Greffenius, Dean Kildaw, Arthur B. Kettle, Leslie Slater, Alexis Paul, and Susan Hatch (all of USFWS/AMNWR), and the villagers of Little Diomede Island.

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