Sea Otters, Mercury, and Monitoring Climate Change

Duffy LK*[1], Hirons AC[2], Schaaf JM[3], McRoy CP[4], Murray MS[5] and Muelken MV[6]

[1]Department of Chemistry and Biochemistry, University of Alaska Fairbanks, Fairbanks, AK, USA

[2]Halmos College of Arts and Sciences, Nova Southeastern University, Fort Lauderdale, FL, USA

[3]National Park Service, Anchorage, AK, USA

[4]International Arctic Research Center, University of Alaska Fairbanks, Fairbanks, AK, USA

[5]Arctic Institute of North America, University of Calgary, Calgary, AB, Canada

[6]Kassiska Division of Health Sciences, Idaho State University, Pocatello, ID, USA

***Corresponding author:** Lawrence K Duffy, Department of Chemistry and Biochemistry, University of Alaska Fairbanks, Fairbanks, AK, USA, Email: lkduffy@alaska.edu

Abstract

An increase in mobility of heavy metals, like mercury (Hg), has the potential to be one of the climate change related impacts on Arctic and sub-Arctic ecosystems. Sea level rise and flooding events in high latitude coastal ecosystems could increase the bioavailability of contaminants such as mercury. Mercury concentrations have been used as an indicator of past exposure to heavy metals in ancient Pacific cod and here we report on concentrations in archeologically recovered sea otter bones (Enhydra lutris). Methods utilizing stable isotope ratios can be used to reconstruct ancient food webs and help identify prey which may have bioaccumulated high concentrations of mercury. Modern sea otters have δ^{13}C, δ^{15}N, and mercury values corresponding largely to a benthic diet. Conversely, if higher δ^{15}N and mercury levels were found in ancient sea otter bones located in a coastal ecosystem, these increases may be associated with rising sea level following the last glacial maximum. These data place present day and projected climate change related perturbations, like sea level rise, in a historical context.

Keywords: Sea Otters; Enhydra Lutris; Mercury; Sea Level; Climate Change; Stable Isotopes

Introduction

Pollution problems involving the release of mercury (Hg) into the food web have been given extensive coverage by the media, especially when associated with the mining and energy industries. The health effects of the release of Hg into water or air as a pollutant has led to the creation of legislation to regulate heavy metals. Hg in the environment is a major concern for the Polar Regions because of its tendency to biomagnify in the marine food web [1-4]. This is a particular problem in Arctic and subarctic regions where people are heavily dependent upon marine resources for subsistence. Rapid climate warming and increased mineral and energy development in the north can alter the complex fluxes in the polar Hg fate and transport system [5]. One approach to studying change in Hg dynamics is increasing site specific studies that provide an historical timeline of Hg in the marine system. Sampling of keystone species such as sea otters (*Enhydra lutris*) will allow regional monitoring of the impact of Hg contamination in a changing climate [6-11] (Figure 1).
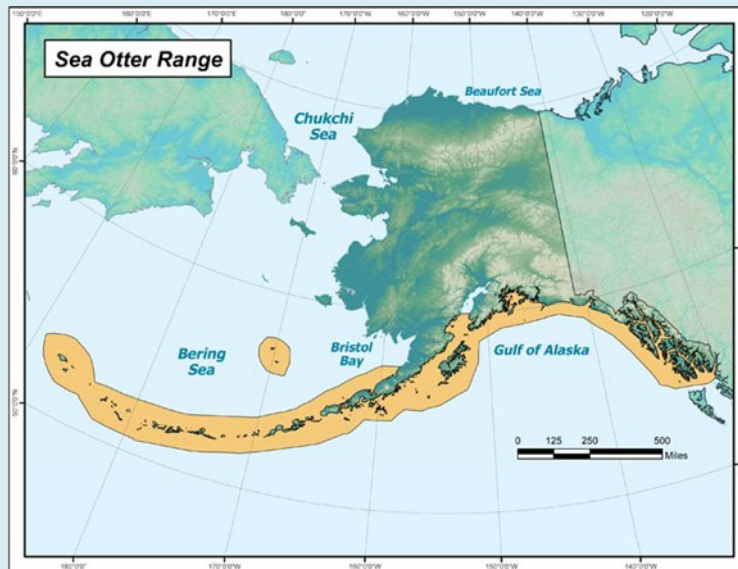
Figure 1: Sea otter range in Alaska. This is the range of northern sea otters (*E. lutris kenyoni*). Sea otters range from western Canada to the Bering Sea. Local oceanographic and climate conditions influence community sea otter ecology [12]. Sea otters are a keystone species in ecosystem structure and used as indicators in food webs.

The sea otter range spans the Gulf of Alaska to the western edge of the Aleutian Islands (Figure 1). Human use of this area is continuous from the early Holocene to the present as is human exploitation of otters [13-17]. From the late prehistoric period to the present, the Gulf has been home to Alutiiq, Athabascan, Tlingit, Asian and European people [18,19] with evidence for intra-and interregional interaction extending well into the early/mid-Holocene [20,21]. Faunal remains recovered and analyzed from coastal archeological sites indicate a focus on marine resources from at least 4000 BP in Prince William Sound (PWS) [22], 6500 BP in the Kodiak Archipelago, and from 7500 BP on the Pacific side of the Alaska Peninsula [23] and in the southeastern Gulf of Alaska [12,24]. Consumption of marine species couples the ecosystem with the human food system and to changes in contaminant bioavailability as the climate changes.

Elemental Hg in the air or sediments can be converted into the Hg (II) ion which can be methylated by anaerobic bacteria to produce the cation, CH_3Hg^+. Elemental Hg can be found in all environmental spheres, but bioavailability of Hg has been shown to vary with the chemical species. Lipophilic organometallic species of heavy metals (such as methyl mercury) tend to persist and concentrate in higher trophic level organisms. This persistence in the food web results in biomagnification of Hg in organisms living in niches at higher trophic levels, including predatory fish, birds and mammals [25-28]. THg levels (ng/g) observed in modern food systems may be linked to industrial anthropogenic Hg outputs [29]. Sea otters are a keystone species that feeds at mid-trophic

levels, consuming mainly sea urchins, shellfish and forage fish [21]. Archeologically recovered animal remains can provide useful data for assessing trends in the presence of contaminants in the food system [12] and also serve as proxies for assessing human exposure as such remains are the likely by-product of human consumption practices. Hg in Arctic and sub-arctic food webs is a constant issue of concern for subsistence harvesters and regulators [30]. Local legacy mining and global transport are major sources [31,32] of heavy metal pollutants like Hg. The Arctic is now the "canary in the coal mine" for climate change with temperatures predicted to increase at twice the average global rate. Rainfall, flooding events and erosion can redistribute heavy metals, including Hg, into aquatic and marine ecosystems [33-36]. Climate change-related threats can be pan-arctic in nature, including increased coastal erosion from sea level rise, increased precipitation, and diminished sea ice which influences the severity of storm-related coastal flooding and erosion from increasing storm strength [37,38]. Stable isotope ratios and mercury analysis provide insight into the relationships among prehistoric subsistence activities, marine ecosystem structure and function, and the amount of mercury at various trophic levels. We hypothesize that if environmental change allows marine vertebrates to forage at higher trophic levels, this will be recorded by incremental increases in amounts of mercury accumulated. In other words, as there is an increase in food web length, there is increased access to mercury, with all its potential impacts on human and marine system health. The hypothesis that sea level rise and flooding increases bioavailability of Hg in

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the marine, and eventually the human food system, could be tested by determining Hg concentrations in keystone species such as sea otters. In this study, we show that Hg in ng/g can be detected in ancient sea otter bones. This suggests that paleontological bone samples can be used to predict the impact of sea level rise on Hg mobility in the food web [11,12].

Materials and Methods

The sea otter material analyzed included modern sample from the Alexander Archipelago and the Prince William Sound areas and ancient samples recovered from the Mink Island archeological site (XMK-030) located in the Shelikof Strait, between the Alaska Peninsula and Kodiak Island. The Mink Island site remains were retrieved from various strata [12,23].

Mercury Analysis

Total Hg (THg) analysis was performed by Frontier GeoSciences (Seattle, Washington) following established procedures. Briefly, sub-sampled tissues were homogenized and approximately 1 g of tissue was digested in 7:3 HNO_3/H_2SO_4 and oxidized with 10% BrCl in 12N HCL. The sample was reduced to Hg^0 with $SnCl_2$ and purged with N_2 onto gold coated quartz sand traps followed by dual thermal desorption to a Cold Vapor Atomic Fluorescence Spectrometer (Tekran Model-2500 CVAFS Hg Detector) with argon gas carrier. The minimum detection limit for total Hg was 0.001 µg/g. Tissue concentrations are expressed as ng/g wet weight (ww) unless otherwise noted. Certified dogfish tissue (DORM-2) was used as a standard and the percent recovery was greater than 80%. A check standard and blank was run after every 10 samples. Matrix Spike Recovery Duplicates varied by 10%.

Quality Control

Daily analytical runs were begun with an eight point standard curve, spanning the entire analytical range of interest, with additional standards analyzed every ten samples. The daily standard curves were calculated with the blank-corrected initial standards, and a linear regression forced through zero. Two matrix spikes, and several method blanks were co-processed and analyzed in exactly the same manner as ordinary samples. Variation of duplicate samples was less than 20%.

Stable Isotope Analysis

Stable isotopes are naturally-occurring forms of elements which characterize the shape of the food web. The $\delta^{13}C$ content established by environmental conditions (CO_2, temperature, pH and phytoplankton levels) serves as the base; trophic level is indicated by $\delta^{15}N$ content which increases $\sim3^0$/00 w/trophic level.

Collagen Extraction

Bone samples were well preserved and free of humus and tissues. Collagen was extracted following the procedure described in detail in Hirons [39]. Approximately 1g of bone was either cut as a solid piece or shaved from the mandible or the shaft of a long bone. Only cancellous bone was used for extraction due to the larger quantity of collagen it contains. The bone samples had lipids removed by a methanol/chloroform procedure [39] prior to demineralization. The bone was allowed to demineralize in 1N HCl for approximately seven days at 5°C; fresh acid was added to the samples every day. The remaining material was rinsed and then boiled in deionized water for approximately eight hours to dissolve the collagen and precipitate peptides. The solution was passed through a 0.45µ filter, and the filtrate was dried in glass shell vials at 60°C for a minimum of 48 hours.

Mass spectrometry

Subsamples of each tissue (1-1.5mg) were combusted and analyzed for stable isotope ratios with a thermos-Finnigan Delta Plus isotope ratio mass spectrometer. Replicability of standards and samples was ≤ 0.2% for both $\delta^{13}C$, $\delta^{15}N$. Stable isotope ratios were expressed in the following standard notation:

$$\delta X (0/00) = (R_{sample}/R_{standard}-1) \times 1000$$

where x is ^{13}C or ^{15}N and R_{sample} is the $^{13}C/^{12}C$ or $^{15}N/^{14}N$ respectively. $R_{standard}$ for ^{13}C is Pee Dee Belemnite; for ^{15}N it is atmospheric N_2 (air).

Results and Discussion

Generally, soft tissues in archeological sites are not preserved [10,16]; we therefore sampled bone, muscle, liver and fur from one modern otter in an effort to identify the differences in bioaccumulation among different tissues. The Hg concentration observed in the modern sea otter bone sample is different than the ancient bone samples (Table 1). The THg concentrations for sea otter bones from the Mink Island archeological site were highly variable, ranging up to 451 ng/g. The mean THg concentration for the 27 ancient bone samples was 289 ng/g. with a range of 17.8-451 ng/g (Table 2). This is a similar range to the reported values of Pacific cod bones (16 -707 ng/g) [12].

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Modern Sample	Hg (ng/g)
Muscle	58
Liver	64
Fur	186
Bone (A0137)	9.3*
Ancient Bone Sample (n=27) average	289

Table 1: Modern Sea otter mercury concentration and ancient bone average.
*Delta N was 11.55 (0/00).

Sea otters feed at an intermediate trophic level, consuming sea urchins and shell fish, supplemented with forage fish. Increased precipitation and water movement can transport Hg into the food web [31,38] . Sea level rise and precipitation would disperse THg [5,40].

The XMK-030 site is one of the most thoroughly dated in the region and is located along the western side of Shelikof Straits between the upper Alaska Peninsula and Kodiak Island, in Amalik Bay [12,23]. The sea otter remains are the by-product of human hunting and disposal activities. The prehistoric human populations that exploited these animals were capable of marine mammal hunting over a wide territory and in the open waters of the Gulf of Alaska.

Dietz and others [17,27] have discussed the diagenesis processes that are possible when working with calcified tissues from archeological sites. Since methyl Hg is bound to the protein fraction, conditions leading to rapid collagen degradation are of concern from an Hg and MeHg concentration perspective. MeHg was observed, but concentrations were below reportable levels ranged between 04-07ng/g. Diagenisis occurs in wet, alkaline soils.

Stable isotope ratios are used to reconstruct ancient food webs and help identify sea otter prey which may have bioaccumulated high concentrations of the metal. Modern sea otters have $\delta^{13}C$, $\delta^{15}N$, and mercury values likely corresponding to a benthic diet. Higher $\delta^{15}N$ and mercury levels were found in ancient sea otter bones demonstrating upper trophic level use. An increase of $\delta15N$ may be associated with rising sea level following the glacial maximum and leading to a change in trophic levels [12]. Monitoring of mercury levels in sea otters and fish would identify trophic spacio- temporal shifts and evaluate their implications as performed in the U.S. Great Lakes [41].

During the Holocene, as today, rivers are a major component of a drainage system. Watersheds are closely linked ecosystems that are natural units of analysis and management [4], within which Hg can be mobilized and dispersed across the landscape, eventually entering the ocean [42]. Rivers are still a major component of socio-ecological systems which, in turn, have the potential to change because of chronic disturbances, i.e. the adaptive cycle. Seven percent of the human population lives below the estimated future sea level, so coastal populations will be impacted by Hg when sea levels rise [43]. Some of the damage from sea level rise can be mitigated if there is a coordinated investment in developing a framework to monitor and understand the local risks to subsistence users [43,44]. Risk assessments and assessments of individual and community health in contemporary human communities should be undertaken with sensitivity to environmental justice and the historical dimensions of the community [4,30,44]. In this study, we suggest sea otter data can add a historical dimension to monitoring climate change in the North.

Ancient Bone Sample		
Sample ID	Hg (ng/g)*	Delta N (0/00)
1219	256	13.04
1220	130	12.25
1237	26.2	12.54
1378	451	14.3
1381	48.7	11.86
1386	60.7	11.71
1415	249	15.03
1616	273	12.68
1617	18	13.71
1618	17.8	12.56
1619	47.8	12.97
1620	44.3	12.65
1621	22.9	11.94
1659	288	16.52
1660	191	14.36
1671	262	14.09
1676	252	14.36
1712	89	11.97
2130	153	14.09
2146	197	12.52
2147	354	14
2152	357	13.31
2154	199	13.73
2164	277	13.51
2169	368	15.2
2175	437	14.98
2332	329	16.07
Mean	**289**	**13.55**

Table 2: Sea Otter archeological bone samples: mercury and nitrogen stable isotope ratio.

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The sea otter inhabits coastal environments where it dives to the sea floor to forage. Today it preys mostly on marine invertebrates such as sea urchins, various mollusks and crustaceans, and some species of fish. They tend not to travel long distances and will generally remain in a home location, which can range from a few square kilometers to 40 square km. There are 3 stocks within Alaska: southeastern, south central, and southwestern Alaska [21]. Sea otters are a keystone species and offer a perspective of contamination transport often neglected in pollution monitoring of fish only eating mammals. Observations of sea otter diet show consumption of benthic invertebrates such as sea urchins and in some areas forage fish [21]. Prior to commercial hunting (1700s), the northern sea otter population was estimated to be 150,000-300,000. In 1995, the population was documented to have declined to 100,000 otters and some populations were seen to have declined by 90% in certain areas. The southwest Alaska stock is listed as threatened as of 2005 under the Endangered Species Act. The sea otter acts as a good sentinel species (i.e. bioindicator) that can inform changes in ecosystem services.

While seal level rise and flooding did increase Hg mobility following the last glacial maximum, the current situation is more complicated. Due to the increase in human ecosystem services required by human population and industrial activity, i.e. anthropogenic activities, the melting of glaciers and sea level rise, is now also influenced by human population growth [45-50].

Conclusion

Current and historical perspectives of the food web structure and its sensitivity to disturbances are relevant to our ability to predict the fate of Hg bioavailability related to sea level rise. The current movement of Hg in the ecosystem and its impact on services illustrates an increased complexity of climate change. Hg was detected in ancient sea otter bones from a paleontological site dating to the early Holocene. This study suggests the concept that anthropological induced climate change, which leads to increased precipitation and sea level rise, should be monitored for increased mercury mobility into coastal food webs. The rapid increase in heavy metal pollution is a factor that leads to higher Hg accumulation in animals, and is a more immediate result of a changing climate. Additionally, the use of stable isotopes can inform an animal's changing diet.

Acknowledgements

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