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Mercury in the northeastern Chukchi Sea: Distribution patterns in seawater and sediments and biomagnification in the benthic food web

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ABSTRACT

Mercury contamination in the atmosphere, snow and marine mammals of the Arctic has been a continuing environmental concern and the focus of many investigations. Much less is known about the distribution of Hg in seawater, sediments and organisms from lower trophic levels in the Arctic, especially the Chukchi Sea. The onset of sea-ice retreat, severe coastal erosion, enhanced primary productivity and offshore energy development in the northeastern Chukchi Sea (NECS) signal changes to a system for which we have limited data for potentially toxic chemicals. To help us better understand and explain any future changes, we present here a combined data set for Hg in seawater, sediments and the following organisms from the NECS: amphipods (*Ampelisca macrocephala*), clams (*Astarte borealis*), snow crabs (*Chionoecetes opilio*), arctic cod (*Boreogadus saida*) and whelks (*Buccinum* spp. and *Neptunea heros*). Concentrations of total dissolved Hg (THg_d) averaged ($\pm$ standard deviation) 2.8 ± 1.4 pM in the NECS, ~2 times greater than values of 1.5 ± 0.5 pM for the Bering Strait. Overall, consistently lower concentrations of THg_d were found at depths with markedly higher concentrations of chlorophyll *a*. Concentrations of total Hg (THg) in sediments from the NECS averaged 31 ± 10 ng g^{-1}, correlated well with silt+clay, Al and TOC, and showed a long-term record consistent with the natural, background environment. Very localized occurrences of sediment with elevated THg concentrations were identified near two exploratory drilling sites where drilling mud and formation cuttings were discharged in 1989. Concentrations of sediment monomethylmercury (MMHg) averaged 0.15 ± 0.07 ng g^{-1} and accounted for only 0.43 ± 0.17% of the sediment THg. The lowest average value ($\pm$ standard error) for THg in biota was found for *A. borealis* at 44 ± 4 ng g^{-1} dry weight (d. wt.) with 33% of the THg present as MMHg. The highest average values for THg were identified for the whelks *N. heros* (195 ± 29 ng g^{-1}, d. wt) and *Buccinum* spp. (269 ± 54 ng g^{-1}, d. wt) with 95% of the THg present as MMHg in *N. heros*, the highest trophic level organism in this study as determined using data for $\delta^{15}N$. Biomagnification of MMHg was observed in this benthic food web with the following relationship: $\log[MMHg] = 0.19[\delta^{15}N] - 0.84$ ($R^2 = 0.80$, $p = 0.02$).

1. Introduction

The Chukchi Sea, the only passage between the Pacific and Arctic oceans, is of great ecological, political and economic importance. At present, the Arctic, including the Chukchi Sea, is experiencing rapid climate change and is susceptible to the effects of sea-ice retreat, severe coastal erosion and enhanced primary productivity (Grebmeier et al., 2010; Holland et al., 2006). Increased offshore energy development and trans-Arctic shipping have further heightened concern for anthropogenic impacts in the Chukchi Sea. Several persistent pollutants that biomagnify, including mercury (Hg), are of particular interest in the Arctic because of past instances of neurological damage and adverse effects on the excretory, immune and reproductive systems of marine mammals and humans (Jaeger et al., 2009). We present here the first comprehensive data set for Hg in seawater, sediment and benthic biota from the northeastern Chukchi Sea (NECS) to document and explain the present-day distribution of Hg and to help establish a point of reference for tracking any future changes.

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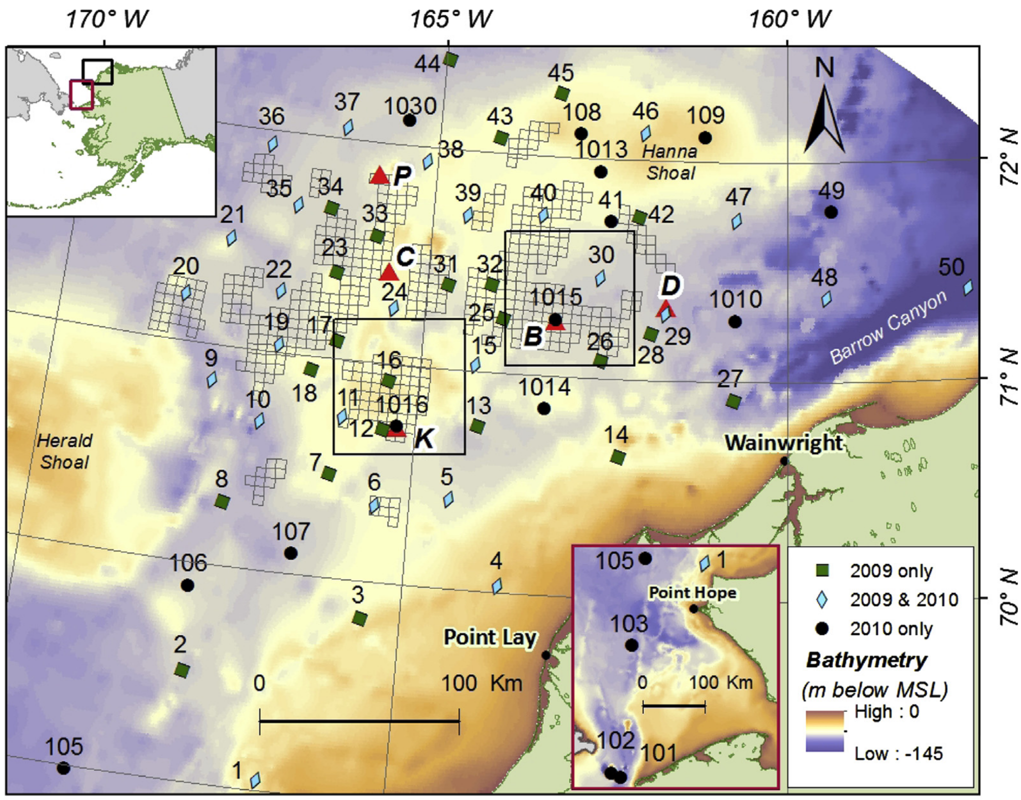

Fig. 1. Map showing sampling stations for the COMIDA CAB Project. Exploratory drilling sites at Klondike (K), Burger (B), Diamond (D), Crackerjack (C) and Popcorn (P) are identified with red triangles. Squares outlined on the larger map show areas around the B and K drilling sites where more intensive sampling was carried out. Upper inset map shows location of study area off the northwestern coast of Alaska. Lower inset map shows stations to the south of those shown on the larger map. Small gray squares show offshore lease blocks.

With a residence time of 6–12 months, Hg^0 is transported over long distances through the atmosphere to the Chukchi Sea from various natural and anthropogenic sources, including combustion of coal at lower latitudes (Jaeger et al., 2009; Pacyna, 2005). Photochemical oxidation of Hg^0 in the atmosphere produces soluble inorganic Hg^{2+} that enters the Chukchi Sea through wet and dry deposition. In addition to atmospheric inputs, Hg also enters the Chukchi Sea with incoming water from the Bering Sea, coastal erosion and runoff from Alaskan and Russian rivers (Laurier et al., 2004; Outridge et al., 2008; Sunderland et al., 2009). Mercury not taken up by biota or adsorbed onto particles that settle to the sediment, can be carried to the Arctic Ocean relatively quickly due to a half-transit time of 1–6 months for water moving northward through the Chukchi Sea (Woodgate et al., 2005). Biological uptake of dissolved Hg by phytoplankton has been shown to be important in the Arctic (Outridge et al., 2008) and may be especially noteworthy in the NECS where the photic zone extends to the bottom of this 50-m deep, shallow sea (Hill et al., 2005). Microbial production of MMHg occurs rapidly and in the Canadian Arctic accounts for $\sim$50% of the MMHg present in the water column (Lehnherr et al., 2011).

Bottom sediments also serve as an important site of Hg methylation, a process mostly carried out by sulfate-reducing bacteria under anoxic conditions (Gilmour et al., 1992, 1998). However, MMHg accounts for $<$1% of THg in most sediments (Gagnon et al., 1996; Kannan et al., 1998). Background concentrations of THg in marine sediments have been determined by normalizing concentrations to aluminum (Al) which serves as a proxy for the natural metal-controlling variables of organic carbon, grain size and mineralogy (e.g., Trefry et al., 2003). Sediment that becomes contaminated with Hg may lead to enhanced biomagnification of Hg (Desy et al., 2000).

In the Arctic, MMHg biomagnifies by $>$50-fold from lower trophic level organisms such as zooplankton and bivalves to higher trophic level organism such as sea birds (Jaeger et al., 2009). For example, THg concentrations in edible muscle tissue from ringed seals in the Eastern Arctic averaged 1850 ng g^{-1} d. wt., relative to 20 ng g^{-1} d. wt. for zooplankton (Neff et al., 2009; Wagemann et al., 1998). Biomagnification is usually traced using concentrations of MMHg and data for stable nitrogen isotopes ($\delta^{15}N$); values for $\delta^{15}N$ increase by $\sim$3.4‰ per trophic level following well defined equations (Hobson and Welch, 1992). Even though the knowledge base for Hg in the Arctic is evolving, few data are available for the Chukchi Sea.

Results presented here were obtained during the Chukchi Sea Offshore Monitoring in Drilling Area (COMIDA): Chemical and Benthos (CAB) Project that was designed to determine the distribution of benthic biota and the chemical composition of water, sediments and biota in the NECS (Fig. 1). This region contains numerous lease areas where offshore oil and gas development may occur. The objectives of this paper are as follows: (1) establish and explain background concentrations of Hg in seawater, sediments and biota from the NECS, (2) search for any anthropogenic Hg inputs to the area, including inputs from past exploratory drilling and (3) determine the magnitude of biomagnification of MMHg in the benthic food web. These objectives need to be met if we are to effectively evaluate future changes in the distribution and impacts of Hg in the Chukchi Sea.

2. Methods

2.1. Sample collection

Sampling for the COMIDA CAB Project took place in the Bering Strait and throughout the eastern Chukchi Sea during July and