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Review

The distribution and trends of persistent organic pollutants and mercury in marine mammals from Canada's Eastern Arctic

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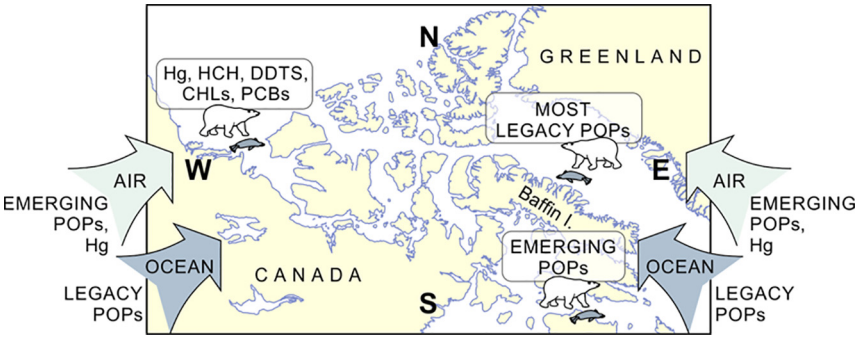
HIGHLIGHTS

- Mercury and legacy HCH in marine mammals are highest in the Western Arctic.
- Most legacy POPs in marine mammals are highest in the East and the Beaufort Sea.
- More recent POPs are highest in seals and polar bears at lower latitudes.
- Food web structure is shaping spatial trends of mercury and some legacy POPs.
- Water is driving legacy POPs whereas atmospheric deposition is driving recent POPs.

GRAPHICAL ABSTRACT



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ABSTRACT

Arctic contaminant research in the marine environment has focused on organohalogen compounds and mercury mainly because they are bioaccumulative, persistent and toxic. This review summarizes and discusses the patterns and trends of persistent organic pollutants (POPs) and mercury in ringed seals (*Pusa hispida*) and polar bears (*Ursus maritimus*) in the Eastern Canadian Arctic relative to the rest of the Canadian Arctic. The review provides explanations for these trends and looks at the implications of climate-related changes on contaminants in these marine mammals in a region that has been reviewed little. Presently, the highest levels of total mercury (THg) and the legacy pesticide HCH in ringed seals and polar bears are found in the Western Canadian Arctic relative to other locations. Whereas, highest levels of some legacy contaminants, including ∑PCBs, PCB 153, ∑DDTs, *p,p′*-DDE, ∑CHLs, ClBz are found in the east (i.e., Ungava Bay and Labrador) and in the Beaufort Sea relative to other locations. The highest levels of recent contaminants, including PBDEs and PFOS are found at lower latitudes. Feeding ecology (e.g., feeding at a higher trophic position) is shaping the elevated levels of THg and some legacy contaminants in the west compared to the east. Spatial and temporal trends for POPs and THg are underpinned by historical loadings of surface ocean reservoirs including the Western Arctic Ocean and the North Atlantic Ocean. Trends set up by the distribution of water masses across the Canadian Arctic Archipelago are then acted upon locally by on-going atmospheric deposition, which is the dominant contributor for more recent contaminants. Warming and continued decline in sea ice are likely to result in further shifts in food web structure, which are likely to increase contaminant burdens in marine mammals. Monitoring of seawater and a range of trophic levels would provide a better basis to inform communities about contaminants in traditionally

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harvested foods, allow us to understand the causes of contaminant trends in marine ecosystems, and to track environmental response to source controls instituted under international conventions.

Contents

1. Regional contaminant overview

Contaminants have entered the Arctic predominantly by atmospheric and oceanic long-range transport (Fig. 1) and secondarily from local sources. Long-range contaminants having the potential to produce consequential exposures in arctic biota exhibit a set of key characteristics. To transport long distances in the atmosphere or oceans, they must be volatile or semi-volatile, or partition favourably into water, but they must also be sufficiently involatile and chemically stable to deposit from the atmosphere within the Arctic once arriving there (Wania, 2003). Furthermore, to produce substantial risk at distant locations like the Arctic these contaminants must have been released in large quantities (e.g., 100 s of kt to >Mt (see, e.g., Li and Macdonald, 2005; Macdonald et al., 2000; Streets et al., 2011)), and persist in air or water for the time periods required to arrive in the Arctic or move within the Arctic (>5 days for winds, >1–10 years for ocean currents). Finally, these contaminants must readily enter biological food webs and be concentrated there to concentrations that exceed thresholds of known or potential adverse effects. Persistent, biologically accumulative, toxic (PBT) characteristics apply particularly to a wide range of organohalogen compounds (OHCs) released either intentionally (e.g., pesticides) or collaterally with industrial activities (e.g., PCBs, PBDEs and PFOS), and mercury (Hg), which has volatile and toxic forms (elemental and methyl mercury).

The OHCs comprise thousands of compounds that exhibit a wide range in physical chemical properties such as volatility and partitioning between media like air, water and particulates (Wania, 2003). Even within a single compound class (e.g., PCBs, PBDEs, DDTs, HCHs) there may be a wide range in these properties, with highly chlorinated compounds usually having higher affinity for particulates. These chemical properties ultimately decide much of the fate of OHCs: compounds with high affinity for particles like the highly chlorinated PCBs (high K_{OW}) tend to attach to particles. In the ocean some of these particles rapidly sink (days – weeks) from surface water into the deeper ocean or become buried in shallow-ocean sediments. Other OHCs, like the HCHs, tend to partition strongly from air into water, but not so much onto particles. Accordingly, the transport pathways of the OHCs into and through the Arctic Ocean are largely controlled by these physical chemical properties (Wania, 2003), and by the time scales of degradation (e.g., photolysis, hydroloyis, metabolism) and sedimentation (particle scavenging), which ultimately remove OHCs from the biosphere.

Hg can be buried in soils and sediments, which removes it from the biosphere, but unlike the OCHs, Hg cannot be destroyed. Transfer of Hg to long-term sequestration (sediments, permafrost and the deep oceans) takes centuries or longer during which the contaminant Hg has sufficient time to redistribute widely within air and ocean through volatilization, deposition and transport (e.g., see Driscoll et al., 2013). As a result of the historical loadings from human activities (e.g., mining, coal burning, industry), the ocean today is estimated to hold ~56 ± 16 kt of contaminant Hg in part manifested as an approximate tripling of the total Hg (THg) concentration in surface water above the pre-industrial, natural background (Lamborg et al., 2014).

Unlike the OHCs, Hg occurs naturally and thus anthropogenic Hg has loaded onto this natural background (Driscoll et al., 2013; Lamborg et al., 2014; Streets et al., 2011). Of great importance to fate and toxicity, Hg exhibits several forms in the environment including inorganic (elemental (Hg^0), dicationic Hg (Hg^{2+})) and organic (monomethylmercury (MMHg), dimethylmercury (DMHg)) forms (AMAP, 2011). Of these forms, MMHg presents by far the greatest concern for environmental toxicity, which implies that the inorganic forms of Hg, which dominate emissions and transport, must undergo methylation within the environment to convert them to the PBT form (Macdonald and Loseto, 2010). The potential for Hg to transfer between chemical forms presents a challenge to predicting Hg trends and toxicity within ecosystems because processes of oxidation, reduction and methylation are strongly controlled by environmental conditions like solar radiation and intensity of organic carbon processing. In this review we summarize and discuss the distribution and trends of POPs (and specifically OHCs) and mercury burdens in ringed seals and polar bears in the Eastern Canadian Arctic relative to the rest of the Canadian Arctic. Implications of climate-related changes on marine mammal contaminant levels are also discussed.

2. Contaminants in the Arctic

The risks to the Arctic from OHCs and Hg, which have long been recognized (AMAP, 1998; Barrie et al., 1992), provided much of the