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The delivery of organic contaminants to the Arctic food web: Why sea ice matters

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HIGHLIGHTS

- Sea ice facilitates delivery of organic contaminants to the Arctic marine food web.
- Organic contaminants are concentrated in brine, under-ice seawater, and melt ponds.
- We model contaminant concentrations in melt ponds based on air concentrations.
- CUPs pose a risk of increased exposures *via* dry deposition to melt ponds.
- Dry deposition to melt ponds is important loading route of CUPs to the Arctic Ocean.

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ABSTRACT

For decades sea ice has been perceived as a physical barrier for the loading of contaminants to the Arctic Ocean. We show that sea ice, in fact, facilitates the delivery of organic contaminants to the Arctic marine food web through processes that: 1) are independent of contaminant physical–chemical properties (*e.g.* 2–3-fold increase in exposure to brine-associated biota), and 2) depend on physical–chemical properties and, therefore, differentiate between contaminants (*e.g.* atmospheric loading of contaminants to melt ponds over the summer, and their subsequent leakage to the ocean). We estimate the concentrations of legacy organochlorine pesticides (OCPs) and current-use pesticides (CUPs) in melt pond water in the Beaufort Sea, Canadian High Arctic, in 2008, at near-gas exchange equilibrium based on Henry's law constants (HLCs), air concentrations and exchange dynamics. CUPs currently present the highest risk of increased exposures through melt pond loading and drainage due to the high ratio of melt pond water to seawater concentration (Melt pond Enrichment Factor, MEF), which ranges from 2 for dacthal to 10 for endosulfan I. Melt pond contaminant enrichment can be perceived as a hypothetical 'pump' delivering contaminants from the atmosphere to the ocean under ice-covered conditions, with 2–10% of CUPs annually entering the Beaufort Sea *via* this input route compared to the standing stock in the Polar Mixed Layer of the ocean. The abovementioned processes are strongly favored in first-year ice compared to multi-year ice and, therefore, the dynamic balance between contaminant inventories and contaminant deposition to the surface ocean is being widely affected by the large-scale icescape transition taking place in the Arctic.

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1. Introduction

In the era of global warming, the Arctic is undergoing some of the most rapid changes on Earth. The areal sea ice extent toward the end of summer has decreased steadily since satellite imagery became available (Stroeve et al., 2012), partly as a consequence of the transition of

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much of the ice from multi-year ice (>3 m) to seasonal ice (1–2 m) (Barber et al., 2009). In September 2012, Arctic sea ice extent reached its lowest level on record since 1979 averaging about 3.5 million km^2 and shattering the previous minimum from 2007 by almost 20% and the long-term average from 1979–2000 by ~50% (National Snow & Ice Data Center, NSIDC, http://nsidc.org/). Also, age and thickness of Arctic sea ice decreased dramatically since 1987, particularly in the Eurasian sector (Rigor, 2005). Recent studies suggest that satellite observations may underestimate the rate of ice thinning in the Canadian Arctic (Flato and Boer, 2001; Barber et al., 2009). There are several model projections of seasonally ice-free Arctic by the end of this century (Lindsay and Zhang, 2005; Holland et al., 2006); however, Arctic sea ice cover extent continues to shrink at a much faster rate than that predicted by most models (Stroeve et al., 2007).

Legacy organochlorine pesticides (OCPs) are a group of hydrocarbon compounds containing multiple chlorine substitutions. Although no longer used, and banned under the United Nations Economic Commission for Europe (UNECE) Stockholm Convention on Persistent Organic Pollutants (POPs), these pesticides are now ubiquitous in the Arctic environment due to their semi-volatility and chemical persistence (Macdonald et al., 2000). Examples of such compounds include heptachlor epoxide, α-hexachlorocyclohexane (α-HCH), chlordanes, nonachlor and dieldrin. Large-scale application agro-chemicals, often referred to as current-use pesticides (CUPs), were first synthesized in the 1950s and 1960s and continue to be in use globally. Increasing levels of CUPs have been found in regions isolated from their use and production including the Arctic (Hoferkamp et al., 2010), even though most of them were designed to have reduced environmental persistence and low potential to undergo long-range transport (Hermanson et al., 2005). CUPs of concern in the Arctic have high production volumes and widespread current or past use in temperate regions, relatively low air–water partitioning, and potential to bioaccumulate in biota (*e.g.* chlorpyrifos, chlorothalonil, dacthal, and endosulfan) (Hoferkamp et al., 2010; Weber et al., 2010; Kurt-Karakus et al., 2011). We classify endosulfan as a CUP, because it was in global use at the time when samples for this study were collected. However, endosulfan is currently being phased out globally (placed on the UNECE Stockholm Convention list on POPs in 2011).

For decades sea ice has been perceived almost exclusively as a physical barrier for the airborne delivery of organic contaminants to the Arctic Ocean, and the presence of ice cover has been a deterrent to the flux between the seawater and air (Hargrave et al., 1997). Based on α-HCH as a process tracer, we postulate that first-year sea ice plays a profound role in the delivery of organic contaminants to Arctic food webs, increasing the exposure to biota through 1) high contaminant concentrations in the brine fraction of the sea ice itself (Pućko et al., 2010a), 2) higher concentration of contaminants in the underlying seawater during the early stages of ice formation (Pućko et al., 2013), and 3) the entry of atmospherically-deposited contaminants into surface waters mediated by melt ponds (Pućko et al., 2012). Because the efficacy of the abovementioned processes strongly depends on the ice type (first-year ice (FYI) *versus* multi-year ice (MYI)), the widespread transition from MYI to FYI in the Arctic Ocean is likewise widely altering the flux of contaminants to surface ocean. Specifically, MYI presents a weak brine engine (low content of salts in the brine fraction) and transient, relatively fresh melt ponds that refreeze in winter. In contrast, first-year ice forms widespread melt ponds containing more brine, and the water in these ponds enters the ocean as the ice rots (Barber et al., 2009) and disappears annually toward the end of summer. In this paper, we describe the role that sea ice plays in the delivery of various OCPs and CUPs to marine food webs. In particular we focus on both the spatial and temporal heterogeneity of the marine icescape. Our spatial focus examines the change from a perennial to an annual icescape; our temporal focus covers the rate of ablation and subsequent ponding mechanisms controlling contaminant exchange based on measured concentrations in the Arctic air and surface seawater, and modeled concentrations in the melt ponds.

2. Materials and methods

2.1. Field sampling

Air and surface seawater samples were collected from the Beaufort Sea during the CCGS *Amundsen* scientific cruise as part of the International Polar Year (IPY)–Circumpolar Flaw Lead (CFL) System study (Barber et al., 2010) in May–July 2008 at matching locations to spatially cover as vast an area of the region as possible (Fig. 1). 10 samples of air, and 8 samples of water were analyzed during each leg (8 and 9).

High volume air samples were collected continuously at the bow of the ship using a Gast Regenair Blower (Cole-Parmer, Vernon Hills, USA) calibrated with a Sierra 620 Mass Flow Meter (Sierra Instruments, Monterey, USA). Generally, the sampling protocol was to point the bow into the wind while on station to avoid ship contamination. The sampling train consisted of an EPM 2000 glass fiber filter (GFF; 20 × 25 cm; Whatman, Kent, UK) followed by two polyurethane foam (PUF) plugs (Tisch Environmental, Cleves, USA; 7.8 cm in diameter, 7.5 cm in length). Air volumes ranged from 500 to 1200 m^3 at a flow rate of ~0.4 $m^3 \cdot min^{-1}$. PUF plugs and GFF filters were stored at −20 °C prior to analysis.

Surface water was obtained from a continuous flow line, which ran from 7 m below the water surface to the interior of the ship. At some stations, additional water was collected *via* a submersible *in situ* SeaStar pump (AXYS Technologies, Sidney, Canada) into LDPE (Low-density Polyethylene) collapsible plastic containers. Water volumes ranged from 40 to 100 dm^3. Water samples were filtered through a 0.7 μm GFF (Whatman, Kent, UK; 14.2 cm in diameter) contained in a stainless steel filter holder (Millipore, Billerica, USA; 14.2 cm, stainless steel), and followed by a stainless steel column of Amberlite XAD-2 resin (polystyrene–divinylbenzene copolymer, 20–60 mesh, 50–75 cm^3 settled volume; Sigma-Aldrich, St. Louis, USA) at a flow rate of 100–150 $cm^3 \cdot min^{-1}$. XAD-2 columns were stored at 4 °C and GFFs at −20 °C prior to laboratory analysis.

2.2. Laboratory analysis

PUF plugs and associated GFF filters were processed as described elsewhere (Jantunen and Bidleman, 1995, 1996, 2008; Wong et al., 2011). Data reported here are for PUF plugs only; a random selection of GFFs showed no detectable amounts of CUPs or legacy OCPs. XAD-2 columns and associated GFFs were prepared and analyzed as described elsewhere (Jantunen and Bidleman, 1998; Bidleman et al., 2007; Wong et al., 2011). GFFs were analyzed for CUPs and legacy OCPs but the levels were below the limit of detection (LOD) for the method (0.5–1.0 $pg \cdot dm^{-3}$ for all the CUPs and OCPs in this study).

Quantitative analysis was done by capillary gas chromatography–electron capture negative ion mass spectrometry using an Agilent 6890 GC–5973 MSD or 5975 MSD using a DB-5 column (J&W, Agilent Technologies, Mississauga, Canada, 60 or 30 m, 0.25 mm i.d., 0.25 μm film thickness). A typical temperature program was: initial temperature 90 °C, 15 $°C \cdot min^{-1}$ to 160 °C, 1.0 $°C \cdot min^{-1}$ to 200 °C; hold for 2.0 min, 20 $°C \cdot min^{-1}$ to 270 °C; hold for 10.0 min. Samples were injected pulsed splitless (2 μL, split opened after 1.0 min). Other instrument conditions were: helium carrier gas at 1.2 $cm^3 \cdot min^{-1}$, injector temperature 250 °C, transfer line temperature 250 °C, ion source 150 °C and quadrupole 150 °C. The mass spectrometer was operated with methane at 2.2 $cm^3 \cdot min^{-1}$. The ions monitored (target/qualifying, abbreviation) were: endosulfan I (404/410, ENDO I), dacthal (332/330, DAC), chlorothalonil (266/264, CHT), chlorpyrifos (313/315, CP) *trans*- and *cis*-chlordane (410/408, TC and CC), heptachlor epoxide (316/318, HEPX), *trans*-nonachlor (444/446, TN) and dieldrin (380/382, DIEL). Mirex was used as the internal standard (404/406).

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