

Applying Trophic Tracers to Study Contaminant Levels and Trends in Arctic Marine Wildlife and Food Webs: A Review

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Abstract—Arctic biota are exposed to a complex suite of anthropogenically-sourced contaminants including mercury (Hg) and persistent organic pollutants (POPs). Levels of these contaminants are determined by source emissions and pathways as well biological and ecological factors. As the use of chemical tracers in trophic ecology matures and gains recognition, these tracers have increasingly been applied to study trophic influences on contaminant levels and trends in biota. The aim of this article is to review information on trophic tracer applications to contaminant studies of individuals, populations and species in and across arctic food webs. Stable isotope (SI) ratios of carbon ($\delta^{13}\text{C}$) and particularly nitrogen ($\delta^{15}\text{N}$) have been used to explain primary production source (e.g., benthic vs. pelagic) and trophic position, respectively. These SI tracers have also been used to explain variation in contaminant levels in arctic invertebrates, fish, mammals and overall food webs. Fatty acid (FA) tracers have been used only recently and in few studies to interpret Hg and POP levels in relation to diet. These studies collectively demonstrate the utility and limitations of SI and FA tracers and highlight the influence of trophic relationships, food webs and their dynamics on contaminant levels and food web bioaccumulation.

Keywords: trophic tracers, stable isotopes (SI), fatty acids (FA), persistent organic pollutants, trace metals, mercury, trends, review

INTRODUCTION

Arctic environments and ecosystems are contaminated by numerous anthropogenically-sourced pollutants. Elevated concentrations of, in particular, mercury (Hg) and persistent organic pollutants (POPs) in upper trophic level wildlife represent a health concern for certain populations of marine mammals, polar bears (*Ursus maritimus*), Greenland sharks (*Somniosus microcephalus*) and seabirds (Letcher *et al.*, 2010). There are numerous other recent-use and current-use chemicals that have been detected in arctic biota, including the current-use pesticide endosulfan and consumer product chemicals like polyfluorinated compounds (PFCs) and brominated flame retardants (BFRs) (Letcher *et al.*, 2010).

Contaminant levels and patterns in biota vary spatiotemporally due to differences in emissions and long-range transport pathways, as well as biological factors including diet, trophic position, habitat use, body condition and size, biotransformation capacity, sex, age, and reproductive status (Borgå *et al.*, 2004). Trophic factors like diet and food web structure substantially determine wildlife POP concentrations and may be especially critical to investigate in light of reported climate-related changes to arctic food webs (e.g., Grebmeier *et al.*, 2006). Yet, such factors have been difficult to quantitatively assess.

It is not practical to directly measure prey consumption in individual free-ranging biota at spatial and temporal scales relevant to interpreting contaminant trends. Fecal and stomach contents have been examined to assess diets (e.g., Pagh and Hersteinsson, 2008). However, stomachs are often empty and scat collection is not practical in arctic marine environments. Moreover, both techniques only provide a snapshot of diet and suffer bias from differential digestibility of prey items preventing quantitative diet assessment. Instead, stable isotope (SI) ratios and fatty acids (FAs) have been used as chemical trophic tracers. These biochemical techniques can overcome aforementioned biases and can provide complimentary information when used in combination, such as an integrated signal of diet over various time scales (depending on tracer, tissue and species) which may offer different insights into contaminant accumulation, biomagnification and trends. Trophic tracers can also give information on overall food web structure (Cabana and Rasmussen, 1994; France, 1995; Iverson *et al.*, 2004) and have been applied to clarify arctic diets and food webs (Hobson and Welch, 1992; Thiemann *et al.*, 2008).

Nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) SI have become widespread to infer trophic level and foraging strategies. Since $\delta^{15}\text{N}$ enrichment occurs with increasing trophic level through a food web, it has been used as an estimate of trophic position (Post, 2002). Because $\delta^{13}\text{C}$ enrichment is minimal through food webs, it has instead been used as an indicator of primary production sources, providing information on feeding patterns, such as freshwater versus marine and pelagic versus benthic feeding (France, 1995). As many higher trophic level organisms do not produce sufficient amounts of specific FAs and must obtain them from diet, FAs are also used as trophic tracers (Budge *et al.*, 2006). The profile of FAs of carbon chain length ≥ 14 in a predator reflects that of its prey, with some predictable and correctable differences due to biosynthesis and metabolism. Predator and prey SI and FA data may also be used to generate more powerful quantitative diet composition estimates by frequentist and Bayesian mixing models (Iverson *et al.*, 2004; Moore and Semmens, 2008).

OVERVIEW OF TROPHIC TRACER USE IN ARCTIC MARINE CONTAMINANT STUDIES

Trophic tracers have been used to study POP levels in invertebrates, fish, birds, mammals and overall food webs. This paper presents an overview of recent, relevant publications that have employed trophic tracers in arctic marine biota and food webs to study contaminants.

Marine invertebrates

The influence of SI on Hg, POP and BFR levels has been studied in Canadian and Norwegian Arctic marine zooplankton. Fisk *et al.* (2003) examined lipid content, habitat use, $\delta^{15}\text{N}$ -derived trophic level and $\delta^{13}\text{C}$ -derived carbon source influences on legacy POP concentrations in 14 eastern Canadian Arctic zooplankton species. Although the overall variation explained was low (r^2 : 0.25–0.56), POP concentrations generally increased with increasing lipid content, higher trophic level and more benthic carbon source. However, Hallanger *et al.* (2011a) did not find that $\delta^{15}\text{N}$ -derived trophic position described legacy POP and BFR concentrations in seven Norwegian Arctic zooplankton species. Contaminant concentrations were better explained by feeding strategy (herbivore, omnivore, predator).

Fish

In three Aleutian Islands marine fish species, hepatic POP levels were correlated with muscle $\delta^{15}\text{N}$, and to a lesser extent, $\delta^{13}\text{C}$ (Miles *et al.*, 2009). However, the variance explained by SI ratios was small. POP levels were better explained by proximity to military installations, implying the importance of point sources in this region. Fisk *et al.* (2002) found that $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ and fork length were not correlated with POP concentrations in eastern Canadian Arctic Greenland sharks. The authors concluded that elevated POP concentrations in the sharks relative to other species with similar SI values implied that shark trophic levels were higher than indicated by their SI values. However, McMeans *et al.* (2010) found a relationship between total Hg (THg) and $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in Greenland Shark and 14 teleost species sampled near Iceland.

Seabirds

Braune *et al.* (2002) analyzed eggs in four Canadian Arctic seabird species collected at several breeding colonies for $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, THg, selenium and POPs. Between-colony differences in contaminant concentrations were generally supported by SI, suggesting differences in trophic positions and/or feeding behaviours, such as terrestrial or freshwater versus marine feeding. In seven northern Baffin Bay seabird species, liver and fat POP concentrations were correlated with $\delta^{15}\text{N}$ -derived trophic positions (r^2 : 0.17–0.47) (Buckman *et al.*, 2004). However, POP levels were sometimes lower than expected based on $\delta^{15}\text{N}$ due to biotransformation. Akearok *et al.* (2010) found that $\delta^{15}\text{N}$ explained variation in Canadian High Arctic marine duck Hg levels. In four Barents Sea seabird species, $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, migratory pattern, age and contaminant metabolism explained liver POP concentrations. In three Aleutian Islands seabird species collected across a latitudinal gradient, POPs and Hg were correlated with $\delta^{15}\text{N}$ (r^2 : 0.24–0.48), but were also related to distance from local and point sources (Ricca *et al.*, 2008).

Marine mammals

In seven Greenland, Denmark and Antarctica seal species, fur Hg concentrations were highly correlated with $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$, although $\delta^{13}\text{C}$ relationships may have been related to regional food web baseline $\delta^{13}\text{C}$ variation (Aubail *et al.*, 2011). Blubber PFC- $\delta^{15}\text{N}$ and PFC- $\delta^{13}\text{C}$ relationships were found for some Canadian Arctic and Subarctic ringed seal (*Pusa hispida*) populations, and $\delta^{13}\text{C}$ values were used to adjust PFC concentrations to better understand spatial differences in seal PFC levels (Butt *et al.*, 2008). Elevated $\delta^{15}\text{N}$ in Eastern Hudson Bay walrus (*Odobenus rosmarus*) suggested that unexpectedly high POP concentrations were likely due to ringed seal consumption (Muir *et al.*, 1995). High variation in contaminant levels in Svalbard walrus was also suggested to be due to more contaminated individuals occasionally consuming ringed seals based on inner blubber layer FA signatures between differentially contaminated individuals (Wolkers *et al.*, 2006). Loseto *et al.* (2008) described a relationship between FA- and SI-derived feeding behaviour and liver and muscle Hg concentrations in Beaufort Sea beluga whales (*Delphinapterus leucas*). This study showed that of length, age, FA and SI values, blubber FA signatures and liver $\delta^{15}\text{N}$ values best described liver and muscle Hg concentrations, respectively, suggesting that different diets within the same beluga population were responsible for differences in Hg contamination. Bentzen *et al.* (2008) found that age, sex, lipid content, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values significantly determined POP concentrations in male and female southern Beaufort Sea polar bears. Levels of Hg in this subpopulation were negatively correlated with hair and blood $\delta^{13}\text{C}$, suggesting higher Hg levels were associated with consumption of more pelagic prey items (Cardona-Marek *et al.*, 2009). Both SI and FA signatures were combined to create a composite “Diet-Index” for polar bears sampled across the circumpolar north, which explained a significant amount of explained variation (10–21%) in the levels of POPs and BFRs (McKinney *et al.*, 2011). These results demonstrated that both regional diet and regional contamination differences influence contaminant spatial trends in polar bears. Profiles of SI, FA and POPs from North Pacific killer whales (*Orcinus orca*) sampled near Alaska, Nicaragua and California together provided a weight of evidence delineating three ecotypes based on habitat use and diets (Krahn *et al.*, 2007).

Food webs

A collection of papers studying the Northwater Polynya food web determined $\delta^{15}\text{N}$ -derived trophic level in ice algae, zooplankton, fish, ringed seals and seabirds to investigate food web pathways and biomagnification of POPs and trace elements (Fisk *et al.*, 2001; Moisey *et al.*, 2001; Campbell *et al.*, 2005). Persistent POPs and Hg strongly biomagnified in the food web, whereas other trace elements and less persistent POPs showed either low or minimal concentration change or biodilution. However, biomagnification varied by species in relation to energy requirements and biotransformation. Similar results for POPs and/or Hg have been reported in Barents Sea, southern Beaufort-Chukchi Sea and western

Greenland food webs (Hop *et al.*, 2002; Hoekstra *et al.*, 2003; Rigét *et al.*, 2007). In a western Canadian Arctic food web study, $\delta^{15}\text{N}$ -PFC and $\delta^{15}\text{N}$ -BFR relationships were examined to infer biomagnification (Tomy *et al.*, 2009). The C_8 – C_{11} PFCAs appeared to biomagnify, whereas polybrominated diphenyl ether BFRs generally did not. Seasonality of POP biomagnification was recently reported in a Svalbard food web, with the highest magnification factors in July and lowest in May for most POPs (Hallanger *et al.*, 2011b).

Temporal trends

Trophic tracers have been used to investigate possible temporal changes or fluctuations in diet and food web structure in relation to contaminant time trends. Braune *et al.* (2007) examined concentrations of various POPs and BFRs and $\delta^{15}\text{N}$ in archived ivory gull (*Pagophila eburnea*) eggs from 1976, 1987 and 2004. The authors did not find a consistent directional change in $\delta^{15}\text{N}$, and thus concluded that contaminant time trends were not affected by temporal changes in trophic position. In a 1994–2008 study of Hg and POPs, arctic char (*Salvelinus alpinus*) from a southwest Greenland lake were also assessed in two years for $\delta^{15}\text{N}$ (Rigét *et al.*, 2010). The $\delta^{15}\text{N}$ values were similar in both years suggesting that trophic position had only a minor influence on contaminant trends.

Recently, a small number of studies have also used chemical tracers to directly measure climate change-associated trophic changes that may impact contaminant concentrations in arctic biota. Trends in total Hg (THg) concentrations between 1973–2007 in western Canadian Arctic ringed seals were correlated not with year but with length of ice-free season (Gaden *et al.*, 2009). In years of both long (5 months) and short (2 months) ice-free seasons, seals showed elevated THg concentrations. Although SI results were not conclusive, the findings may have been due to differences in summer prey composition under differing environmental conditions. Recent (1991–2007) feeding changes in western Hudson Bay polar bears were suggested by adipose $\delta^{13}\text{C}$ and FA changes, and resulted in more rapid rates of increase or slower rates of decline of tissue POP concentrations than if dietary patterns had not changed (McKinney *et al.*, 2009). A significant proportion of the between-year trophic tracer variation (46–84%) was explained by differences in timing of the annual Hudson Bay sea ice breakup. These empirical studies support predications that climate change will influence contaminant levels, pathways and fates within arctic environments and ecosystems (Macdonald, 2005). Stable isotopes and fatty acids will be important tools in addressing the interrelated, combined effects of climate change and persistent contaminants on arctic species and food webs.

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