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Stable sulfur isotopes identify habitat-specific foraging and mercury exposure in a highly mobile fish community

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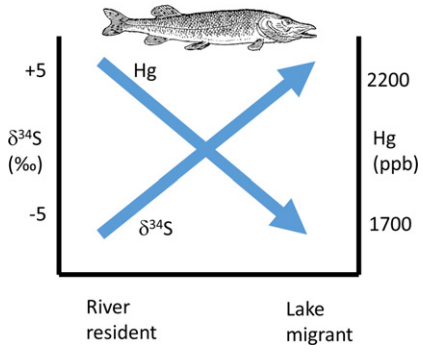
HIGHLIGHTS

- Fish experience differential contaminant exposure pathways in delta ecosystems.
- Sulfur, carbon, and nitrogen isotopes differentiate resident from migrant fishes.
- Food chain of migrant fishes has greater Hg trophic magnification than resident fish.
- Large resident fish have higher Hg concentrations than large migrant fish.

GRAPHICAL ABSTRACT



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ABSTRACT

Tracking the uptake and transfer of toxic chemicals, such as mercury (Hg), in aquatic systems is challenging when many top predators are highly mobile and may therefore be exposed to chemicals in areas other than their location of capture, confounding interpretation of bioaccumulation trends. Here we show how the application of a less commonly used ecological tracer, stable sulfur isotope ratios ($^{34}S/^{32}S$, or $\delta^{34}S$), in a large river-delta-lake complex in northern Canada allows differentiation of resident from migrant fishes, beyond what was possible with more conventional $^{13}C/^{12}C$ and $^{15}N/^{14}N$ measurements. Though all large fishes ($n = 105$) were captured in the river, the majority (76%) had $\delta^{34}S$ values that were indicative of the fish having been reared in the lake. These migrant fishes were connected to a food chain with greater Hg trophic magnification relative to the resident fish of the river and delta. Yet, despite a shallower overall trophic magnification slope, large river-resident fish had higher Hg concentrations owing to a greater biomagnification of Hg between small and large fishes. These findings reveal how S isotopes can trace fish feeding habitats in large freshwater systems and better account for fish movement in complex landscapes with differential exposure pathways and conditions.

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1. Introduction

Despite some restrictions on emissions, anthropogenic loading of contaminants to the environment remains high (Malaj et al., 2014; Zhang et al., 2016), and identifying the source and fate of these contaminants in aquatic food webs is critical in developing regulations to limit exposure for fish-eating humans and wildlife. Certain compounds, including methyl mercury (MeHg) and several organic chemicals, biomagnify in food webs to potentially toxic concentrations in top predators (Lavoie et al., 2013; Walters et al., 2016). Some of these same top predators have wider home ranges than smaller, lower trophic level organisms (McCann et al., 2005) and their movement can confound interpretation of biomagnification trends because organisms may have fed and been exposed to chemicals at a location far from their point and time of capture (Fisk et al., 2001; Jardine et al., 2006; Kim et al., 2016).

Stable isotope analyses (SIA) can provide insight into patterns of foraging and resource use, migration, and trophic relationships of large mobile predators in aquatic systems (Fry and Chumchal, 2011; McCarthy and Waldron, 2000). Food webs in fluvial ecosystems are characterised by strong longitudinal gradients in environmental conditions, heterogeneity at multiple spatial scales, and substantial temporal variation (Finlay and Kendall, 2007). However, stable isotopes are well suited to studying ecological interactions and processes in river food webs. Stable isotopes of $\delta^{13}C$ and $\delta^{15}N$ can trace the source of productivity fueling food webs (resource type, habitat, or in some cases, the specific taxa), the origin of organisms, and the trophic position of consumers (Finlay and Kendall, 2007). $\delta^{15}N$ techniques allow estimation of trophic position (Jepsen and Winemiller, 2002), due to a consistent increase in ^{15}N of consumers that is related to trophic level in food webs (Post, 2002; Vander Zanden et al., 1997). Carbon transfer along food webs is more conservative and can help trace the flow of carbon from food sources or habitats with different ^{13}C values.

When $\delta^{13}C$ fails to differentiate among sources or habitats (France, 1995; Jardine et al., 2008), other tracers can assist in resolving differences. Often applied in estuarine systems (Peterson et al., 1986), $\delta^{34}S$ can be useful in differentiating among different food sources because trophic fractionation of sulfur isotopes is minimal (McCutchan et al., 2003) and the range in $\delta^{34}S$ of potential diet items tends to be much greater than the range of $\delta^{13}C$ or $\delta^{15}N$ (Peterson and Fry, 1987). Biogeochemical differences in sediments versus the water column that lead to fractionation of $\delta^{34}S$ in source pools (Croisetiére et al., 2009; Ponton and Hare, 2015) suggest possible broad-scale differences within and among freshwater habitats, making it a potentially valuable tool in hydrologically and ecologically complex aquatic systems such as inland and coastal deltas. Yet $\delta^{34}S$ has rarely been used in freshwater systems despite early indications it could assist in identifying origins of mobile fishes (Hesslein et al., 1991; McCarthy et al., 1997)

River deltas provide important habitat and migration corridors for fish and are vital resources for human communities, including those in northern Canada (Gummer et al., 2000; Wolfe et al., 2007). As depositional areas, deltas receive high sediment and nutrient loads, which creates considerable habitat diversity and high productivity (Leconte et al., 2001; Tripp et al., 1981). However, their depositional nature also makes them vulnerable to contamination from a variety of upstream anthropogenic activities (Brock et al., 2010; Green et al., 2016; Leconte et al., 2001). For example, upstream developments such as mining, oil sands, pulp and paper, and forestry within the Slave River Basin (Gummer et al., 2000; MRBB, 2003) have manifested as community concerns regarding, among other things, fish safety for consumption (Mantyka-Pringle et al., in review, Ohiozebau et al., 2016, Pembina Institute 2012, Wolfe et al., 2007). Of particular concern are concentrations of MeHg in various consumable fish species. In aquatic food webs, diet has been identified as the primary route of exposure for MeHg and other persistent contaminants (Hall et al., 1997), and both trophic (vertical) and habitat (horizontal) food web structure can influence MeHg concentrations in fishes (Power et al., 2002). Thus, SIA can be a useful tool for understanding the trophic influences on concentration and bioaccumulation of contaminants in aquatic food webs (Kidd et al., 2012) such as river delta ecosystems.

In this study, $\delta^{34}S$ in combination with more-commonly employed stable isotopes, $\delta^{13}C$ and $\delta^{15}N$, were used to characterise habitat use and related risk of contaminant exposure for various fish species in a complex river-delta-lake ecosystem, the Slave River and Delta, Northwest Territories (NT), Canada. First, $\delta^{13}C$ and $\delta^{34}S$ were used to classify fish sampled at two river locations as resident (remain in the river year round) or migrant (migrate from nearby Great Slave Lake). $\delta^{15}N$ was then used to identify biomagnification trends of Hg in the food web of migrant and resident fish. Finally, we consider how these food web interactions and spatial distribution of fish and prey lead to differential risk of Hg exposure for fish species in the Slave River Delta (SRD) region.

2. Methods

2.1. Study site

The SRD is the terminus of the Peace-Athabasca-Slave River corridor and is located along the southern shore of Great Slave Lake, approximately 13 km north-east of Fort Resolution, NWT (Fig. 1). The SRD is a large wetland complex consisting of forests, swamps, marshes, fens, bogs, shallow lakes, and numerous river channels (Brock et al., 2010). The Jean River which originates 14.5 km upstream of the active delta flowing north, and Nagle Channel which originates 3.5 km upstream flowing south, mark the current outer boundaries of the SRD. These channels are relatively stable, with few signs of erosion or extensive flooding (Tripp et al., 1981). The active Delta, which occurs downstream of a major bifurcation of the Slave River into secondary distributaries known as Resdelta Channel, Middle Channel, Old Steamboat Channel, and East Channel, covers an area of approximately 400 km^2 (English et al., 1997) (Fig. 1). Resdelta Channel is the largest of these secondary channels, accounting for >80% of flow, with depths ranging from 12 m to 32 m (Tripp et al., 1981). Other main channels range from 5 m to 12 m deep with minor channels ranging from 1 to 2 m (Tallman, 1996). Approximately 300 km upstream of the SRD, near the town of Fort Smith are the Rapids of the Drowned, one of four sets of large rapids that act as a barrier to upstream movement of fish (Boag and Westworth and Associates Ltd., 1993; Howland et al., 2000). Near Fort Smith the Slave River is turbid with fast flows and steep river banks, and habitats are relatively homogeneous compared to the more diverse channels of the Delta (Little, 1997).

Twenty-four species of fish are known to either migrate through or live in the SRD which is an important feeding, spawning and nursery area for many of these species (Tallman, 1996; Tripp et al., 1981). The SRD has been recognized as an important migration corridor between Great Slave Lake and the lower Slave River for lake whitefish (*Coregonus clupeaformis*), inconnu (*Stenodus leucichthys*), longnose sucker (*Catostomus catostomus*), and walleye (*Sander vitreus*), and is a critical spawning area for large numbers of small bodied ciscos (*Coregonus artedi*) and burbot (*Lota lota*) (Tripp et al., 1981). It is also known as an important rearing area for young-of-the-year burbot and northern pike (*Esox lucius*) (Tripp et al., 1981). Our interest was in characterising the use of three possible habitats by mobile fishes: 1) nearshore Great Slave Lake, close to the mouth of the Slave Delta and dominated by littoral production, 2) offshore Great Slave Lake in deep waters dominated by pelagic production, and 3) the Slave River.

2.2. Sample collection

2.2.1. Collection and processing of food resources

To characterise isotope ratios at the base of the food web, we sampled lower-trophic level organisms from four main locations in August of 2014 and 2015 (Fig. 1), focusing on invertebrates. Macroinvertebrates

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