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Total mercury concentrations in anadromous Northern Dolly Varden from the northwestern Canadian Arctic: A historical baseline study

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HIGHLIGHTS

- THg were measured in Dolly Varden from the Yukon and Northwest Territories.
- Length-adjusted THg concentrations were not related to latitude or longitude.
- Among-population variation in THg was best explained by fork-length.
- Length-adjusted THg concentrations were related to age, $\delta^{15}N$, and $\delta^{13}C$.
- Mean THg were below Health Canada's consumption guideline for commercial fish.

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ABSTRACT

Previous research has documented the significance of total mercury (THg) as a northern contaminant in general and of fish in particular. While much research has been devoted to documenting both spatial and temporal changes in THg in consumed fish, little effort has been directed at understanding patterns of THg in Dolly Varden (*Salvelinus malma*), a prized subsistence species throughout the western North American Arctic. Here we report historical THg concentrations for anadromous Dolly Varden from 10 populations in the Yukon and Northwest Territories sampled across a range of latitudes (67–69°N) and longitudes (136–141°W) between the years 1988–91. Unadjusted mean THg concentrations ranged from 15 to 254 ng/g wet weight. Length-adjusted THg concentrations were significantly different among sites, but were not related to latitude or longitude. Within and among populations, THg was significantly related to fork-length, age, $\delta^{15}N$, and $\delta^{13}C$, with the variation in THg found among populations being best explained by size. The data serve as an important baseline against which future changes in THg levels in this important subsistence fishery may be compared to determine the significance of any observed trends.

1. Introduction

Total mercury (THg) pollution from natural and anthropogenic sources has caused concern for the health of Arctic aquatic ecosystems (Braune et al., 1999), with long range atmospheric transport being the main pathway of exposure (Schroeder et al., 1998). After entering the aquatic ecosystems, mercury is methylated, resulting in toxic methylmercury (MeHg), which bioaccumulates and biomagnifies up the food chain (Booth and Zeller, 2005). Thus, fish consumption is a major exposure route of Hg for humans (Gupta et al., 2005). In the western Canadian Arctic, contamination levels in Dolly Varden (*Salvelinus malma*), a riverine charr, are of particular interest as this species is fished for subsistence purposes by the Inuvialuit and Gwich'in communities (Gallagher et al., 2011).

Dolly Varden (DVCH) occur in two distinct forms, southern and northern (Reist et al., 2002), with the northern form being distributed north of the Alaska Peninsula in the Bering, Chukchi, and Beaufort sea drainages east to the Mackenzie River (Reist et al., 2002). While there are many detailed THg studies of other subsistence fish species from the Canadian Arctic, THg studies of DVCH are limited. For example, other subsistence fish species for which detailed THg studies have been completed include: Arctic charr (*Salvelinus alpinus*) (Muir et al., 2005; Gantner et al., 2010a,b; Gantner et al., 2012; van der Velden et al., 2013a,b), northern pike (*Esox lucius*) (Evans et al., 2005), broad whitefish (*Coregonus nasus*) (Snowshoe and Stephenson, 2000; Evans and Lockhart, 2001; Evans et al., 2005), walleye (*Sander vitreus*) (Evans and Lockhart, 2001; Evans et al., 2005), burbot (*Lota lota*) (Snowshoe and Stephenson, 2000; Carrie et al., 2010), lake whitefish

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(*Coregonus clupeaformis*) (Bodaly et al., 1984), brook charr (*Salvelinus fontinalis*) (Braune et al., 1999), longnose sucker (*Catostomus catostomus*) (Lockhart et al., 2005), Arctic grayling (*Thymallus arcticus*) (Lockhart et al., 2005), inconnu (*Stenodus leucichthys*) (Snowshoe and Stephenson, 2000; Lockhart et al., 2005), cisco (*Coregonus artedi*) (Lockhart et al., 2005), and lake charr (*Salvelinus namaycush*) (Power et al., 2002; Evans et al., 2005). In contrast, Dolly Varden THg concentrations have been studied only incidentally, typically being reported as a single value in a list of studied fish (e.g., Deniseger et al., 1990; Lockhart et al., 2005).

Given the paucity of published mercury data on Dolly Varden, a key aim of this study was to create a spatially explicit baseline for the species to address the lack of information on mercury concentrations in DVCH and to facilitate establishing the significance of future rates of change in THg concentrations in DVCH that may be associated with industrial development and/or larger scale ecological changes (e.g., climate change). Using archival muscle tissue samples, the study further sought to explicitly test the following hypotheses concerning THg levels in Dolly Varden: [i] as has been noted for other species of northern fish (Muir et al., 2005; Gantner et al., 2010a), there will be significant among- population level differences in THg; but, [ii] as has also been noted for other studied northern fish species (Gantner et al., 2010a), there will be no significant latitudinal or longitudinal trends in the spatial differences; [iii] measured THg levels within- and among-populations will be positively correlated with the size and age of tested fish (Bache et al., 1971; Grieb et al., 1990; Jewett et al., 2003); and [iv] measured differences in THg levels within and among populations will be correlated with carbon sources and trophic level, with THg being, respectively, negatively and positively correlated with $\delta^{13}C$ and $\delta^{15}N$ stable isotope measures (Power et al., 2002; Gantner et al., 2010a,b).

2. Methods

2.1. Sample collection

Dorsal muscle tissue samples of anadromous Dolly Varden were obtained from an archival collection maintained by the Department of Fisheries and Oceans Canada (DFO) in Winnipeg, Manitoba (Table 1) from sample sites covering roughly half the natural geographical range of the northern form of Dolly Varden within Canada. Approximately 5 g of muscle tissue was dissected with a clean scalpel from a standard area of each fish located posterior to the dorsal fin and above the lateral line (van der Velden et al., 2013b). Obtained tissue samples were subsequently split for use in THg and stable isotope analyses. Dolly Varden exhibit variant life-history types in this area: anadromous males and females and residual (non-migratory) males in rivers with access to the sea; and isolated populations upstream of impassable falls (Morrow, 1980). This study focused only on the anadromous life-history type.

Table 1
Sites for which anadromous Dolly Varden samples were obtained for analysis. Site code, number of fish (n), geographic co-ordinates and year of capture are given below.

Site	Site code	n	Latitude	Longitude	Year of capture
Babbage River	BR	30	68°37′47″N	139°22′12″W	1991
Cache Creek	CC	30	68°31′11″N	136°13′47″W	1988
Rat River	RR	30	67°46′48″N	136°19′11″W	1988
Firth River	FR	14	68°40′12″N	140°55′11″W	1988
Ptarmigan Bay	PB	28	69°29′11″N	139°01′12″W	1988
Canoe River	CR	30	68°46′11″N	138°45′00″W	1988
Shingle Point	SP	16	68°58′48″N	137°31′12″W	1989
Pauline Cove	PC	30	69°34′47″N	138°52′12″W	1989
Thetis Bay	TB	8	69°32′59″N	139°01′48″W	1989
Big Fish River	BFR	30	68°27′00″N	136°11′60″W	1991

All fish were sampled in the period July to October from rivers located throughout the northern Yukon and Northwest Territories in 1988, 1989, and 1991 (Fig. 1) and were classified as anadromous fish on the basis of capture point (e.g., in the marine environment) or size, morphology and coloration (Reist, personal communication) (Table 1). The whole otolith method was used to determine fish age (Sandstrom et al., 1997). The outer surface of the otolith was hand-ground until the core/nucleus was visible. The otolith was then analyzed in a petri dish under a stereoscopic microscope with water or a clearing agent (e.g., oil of wintergreen) to elucidate the annuli. Sample sites were chosen because of the availability of a statistically sufficient sample size (e.g., Zar, 2010) and from as limited a range of years as possible so as to remove potential confounding temporal effects. Finally, samples were chosen so as to ensure that appropriate biological data (e.g., fork-length (mm), weight (g), age (years), gonad weight (g), maturity, and sex) for statistical testing were available.

2.2. Stable isotope analyses

Portions of all muscle samples were used for the analysis of stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) following methods described in van der Velden et al. (2013b). Briefly, the samples were dried at 50 °C for 48 h and homogenated with a Retsch MM 301 ball mill grinder (F. Kurt Retsch GmbH Co., Haan, Germany) and weighed on a Mettler Ultra micro balance (UMX2, Mettler-Toledo GmbH, Greifensee, Switzerland). A range of 0.275–0.300 mg of the homogenate was used for stable isotope analysis (SIA) completed with a Delta Plus Continuous Flow Stable Isotope Ratio Mass Spectrometer (Thermo Finnigan, Bremen, Germany) coupled to a Carlo Erba elemental analyzer (CHNS-O EA1108, Carlo Erba, Milan, Italy) at the Environmental Isotope Laboratory, University of Waterloo (Waterloo, Ontario). Machine analytical precision, respectively, for $\delta^{13}C$ and $\delta^{15}N$ was $\pm 0.1‰$ and $\pm 0.2‰$, and was determined by repeat analysis (duplicates run every $n = 11$). All resulting measurements were expressed using standard delta notation as parts per thousand differences (‰) with respect to the international reference standards, Vienna Peedee Belemnite for $\delta^{13}C$ (Coplen, 1994) and nitrogen gas in the atmosphere for $\delta^{15}N$ (Mariotti, 1983).

The comparison of organism stable isotope signatures from differing environments requires accounting for differences in the isotopic composition of primary producers from the food web within which organisms feed (Fry, 2006; Casey and Post, 2011). Selecting an appropriate baseline for marine feeding Dolly Varden is particularly challenging given variation in the factors which significantly influence baseline signatures in marine environments (e.g., temperature, depth and salinity (Casey and Post, 2011)), and the apparent movement of Dolly Varden between marine environments with differing physico-chemical characteristics (e.g., salinity, temperature, etc.). Dolly Varden enter the marine environment in spring (May–June) and move westward along the Beaufort coast. Within the marine environment, little is known about the nature and range of their movement, but they are assumed to feed in the highly productive nearshore zone (Sandstrom et al., 2009). Nevertheless, studies have reported anadromous DVCH moving over extremely large distances and outside of the coastal zone (DeCicco, 1992, 1997). Furthermore, recent reports have suggested that DVCH occur further offshore than previously thought (Cobb et al., 2008), a fact that would concur with the reported low catches of Dolly Varden in fish monitoring programs completed along the Beaufort coast (e.g., Jarvela and Thorsteinson, 1999). Given the complexity of possible Dolly Varden coastal and offshore marine movements, the lack of information about mean movement patterns for each of the studied populations, the known spatial variation in coastal baseline signatures caused by variations in depth, salinity, water temperature and water mass mixing (e.g., Jennings and Warr, 2003) and the implications for inferential error associated with incorrect baseline adjustment, no direct attempt