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# Mercury and other trace elements in a pelagic Arctic marine food web (Northwater Polynya, Baffin Bay)

Linda M. Campbell<sup>a</sup>, Ross J. Norstrom<sup>b</sup>, Keith A. Hobson<sup>c</sup>, Derek C.G. Muir<sup>a</sup>, Sean Backus<sup>a</sup>, Aaron T. Fisk<sup>d,\*</sup>

<sup>a</sup>National Water Research Institute, Environment Canada, 867 Lakeshore Road, Burlington, Ontario, Canada, L7R-4A6 <sup>b</sup>Chemistry Department, Carleton University, Ottawa, Ontario, Canada K1A 0H3

<sup>c</sup>Prairie and Northern Wildlife Research Centre, Canada; Canadian Wildlife Service, Saskatoon, SK, Canada, S7N 0X4 <sup>d</sup>Warnell School of Forest Resources, University of Georgia, Athens, Georgia, 30602-2152 USA

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#### Abstract

Total mercury (THg), methylmercury (MeHg) and 22 other trace elements were measured in ice algae, three species of zooplankton, mixed zooplankton samples, Arctic cod (Boreogadus saida), ringed seals (Phoca hispida) and eight species of seabirds to examine the trophodynamics of these metals in an Arctic marine food web. All samples were collected in 1998 in the Northwater Polynya (NOW) located between Ellesmere Island and Greenland in Baffin Bay. THg and MeHg were found to biomagnify through the NOW food web, based on significant positive relationships between log THg and log MeHg concentrations vs.  $\delta^{15}$ N in muscle and liver. The slope of these relationships for muscle THg and MeHg concentrations (slope=0.197 and 0.223, respectively) were similar to those reported for other aquatic food webs. The food web behavior of THg and  $\delta^{15}$ N appears constant, regardless of trophic state (eutrophic vs. oligotrophic), latitude (Arctic vs. tropical) or salinity (marine vs. freshwater) of the ecosystem. Rb in both liver and muscle tissue and Zn in muscle tissue were also found to biomagnify through this food web, although at a rate that is approximately 25% of that of THg. A number of elements (Cd, Pb and Ni in muscle tissue and Cd and Li in seabird liver tissue) were found to decrease trophically through the food web, as indicated by significantly negative relationships with tissue-specific  $\delta^{15}$ N. A diverse group of metals (Ag, Ba, La, Li, Sb, Sr, U and V) were found to have higher concentrations in zooplankton than seabirds or marine mammals due to bioconcentration from seawater. The remaining metals (As, Co, Cu, Ga, Mn, Mo and Se in muscle tissue) showed no relationship with trophic position, as indicated by  $\delta^{15}$ N values, although As in liver tissue showed significant biomagnification in the seabird portion of the food web.

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\* Corresponding author. Tel.: +1 706 542 1477. *E-mail address:* afisk@forestry.uga.edu (A.T. Fisk).

# 1. Introduction

The occurrence of heavy metals in Arctic marine food webs is of concern despite limited anthropogenic activity in this region. Mercury (Hg) and other metals have been detected in various biota and environmental samples across this region, and there has been concern about sources and concentrations in sample food items frequently consumed by humans and wildlife (Dietz et al., 1996; Fisk et al., 2003). In fact, concentrations of many metals in Arctic biota are similar to concentrations measured in temperate regions (Muir et al., 1999). While Hg and other metals are found naturally in the environment, the origin of elevated concentrations observed in biota from across the Arctic have been attributed to anthropogenic sources in southern regions (Muir et al., 1999). Of particular concern are lead (Pb), cadmium (Cd) and Hg compounds, which have reached concentrations in many Arctic biota that exceed Health Canada and World Health Organization guidelines for human consumption (Muir et al., 1999). Pb in Arctic environments has been declining, partially due to increased global limits on alkyl lead additives in gasoline (Jensen et al., 1997). Cd in seal livers and other dietary items is of concern since it has been found that traditional hunters and their families can potentially ingest Cd above Health Canada guidelines of 450 mg Cd/week (Jensen et al., 1997; Johansen et al., 2004). Despite their importance in the Arctic, the fate and dynamics of metals and other elements in Arctic marine systems remain largely unknown.

Polynyas are persistent regions of open water surrounded by sea ice and are a vital component of Arctic marine ecology (Stirling, 1980). Polynyas are also significant from a socio-economic perspective, since the Inuit from Canada and Greenland hunt traditionally important animals such as ringed seals *Phoca hispida* that congregate in polynyas. The Northwater Polynya (NOW) is the largest (50,000 km<sup>2</sup>) and most productive polynya in the Canadian Arctic (Fig. 1). As such, it supports large populations of seabirds and mammals in northern Baffin Bay.



Fig. 1. Approximate geographical position of the Northwater Polynya between Greenland and Ellesmere Island in Baffin Bay in May/June. Baffin Island and Lancaster Sound are also indicated.

The dynamics of carbon and anthropogenic persistent contaminants in the NOW polynya food web have previously been characterized using stable nitrogen  $(\delta^{15}N)$  and carbon  $(\delta^{13}C)$  isotopes (Fisk et al., 2001; Hobson et al., 2002). Stable isotopes have increasingly been used in aquatic ecosystems to evaluate food web structure and energy pathways. Stable nitrogen isotope ratios ( $\delta^{15}$ N) increases 2‰ to 4‰ with each trophic level, providing a means to assess trophic level (Peterson and Fry, 1987). In contrast,  $\delta^{13}$ C values often demonstrate lower trophic fractionation (0.8% to 1%), and are useful for evaluating sources of primary production within a food web (Peterson and Fry, 1987). Stable isotopes can also be used to estimate the rate of biomagnification of a chemical across the entire food web. The slope of contaminant levels against trophic levels as determined by  $\delta^{15}$ N, often called trophic magnification factors (TMFs) of contaminants, have been applied in Arctic food webs to determine the rate of biomagnification through the food web (Atwell et al., 1998; Borgå et al., 2004).

The objective of this research was to assess the trophic transfer of 22 elements and methylmercury in the NOW marine food web by determining metal and stable isotopes of nitrogen and carbon values in a wide range of species.

# 2. Methods

#### 2.1. Sample collection

Samples were collected during May 1998 from the CCGC Pierre Radisson in the Northwater Polynya located in northern Baffin Bay (Fig. 1). Zooplankton were collected from vertical tows from bottom to surface using a 1-m diameter zooplankton net with 520-µm mesh. After collection, zooplankton were either unsorted ("mixed") or sorted by species, including the amphipod Themisto libellula, the calanid copepod Calanus hyperboreus and the mysid, Mysis oculata (Table 1). Arctic cod Boreogadus saida (Table 1) were opportunistically sampled at one location using hand-held nets. Seabirds (Table 1) were collected by shotgun from a Zodiac boat, measured and sexed, and dissected for liver and muscle samples. Ringed seal P. hispida (age 8 to 10 years only) liver and muscle samples

Table 1

List of biota from the NOW marine food web analysed for metals, along with assigned codes used in subsequent tables

Organism	Code	Common name	Taxonomic name	TL
Primary	IALG	Ice algae	~	1.3
Invertebrate	CHYP	Copepod	Calanus	2.0
			hyperboreus	
Invertebrate	ZOOP	Mixed	~	2.2
		zooplankton <sup>a</sup>		
Invertebrate	TLIB	Amphipod	Themisto libellula	2.5
Invertebrate	MOCC	Mysid	Mysis oculata	2.7
Fish	ACOD	Arctic cod	Boreogadus saida	3.7
Sea bird	DOVE	Dovekie	Alle alle	3.2
Sea bird	BLKI	Black-legged	Rissa tridactyla	3.8
		kittiwake		
Sea bird	BLGU	Black guillemot	Ceppus grylle	3.9
Sea bird	TBMU	Thick-billed	Uria lomvia	3.9
		murre		
Sea bird	IVGU	Ivory gull	Pagophilia eburnea	4.0
Sea bird	NOFU	Northern fulmar	Fulmarus glacialis	4.1
Sea bird	GLGU	Glaucous gull	Larus hyperboreus	4.7
Sea bird	THGU	Thayer's gull	Larus thayeri	4.9
Mammal	RSEA	Ringed Seal	Phoca hispida	4.6

The common and taxonomic names are indicated. Mean estimated trophic levels (TL) were estimated from  $\delta^{15}N$  values of muscle samples using Eqs. (2) and (3) (see text). The mean muscle  $\delta^{15}N$  values are listed in Table 2, along with those for liver samples. <sup>a</sup> Mixed zooplankton were predominantly *Calanus* copepods.

(Table 1) were obtained from Inuit hunters from Ausuittuq (Grise Fijord) in Ellesmere Island and Qânâq in north western Greenland. All samples were placed in Whirl-Pak bags, cryo-vials or aluminium foil and frozen until analysed for stable isotopes or elements.

## 2.2. Element analysis

Element samples were analysed at the National Laboratory for Environmental Testing (NLET) at National Water Research Institute in Burlington, Ontario (National Laboratory for Environmental Testing, 2003). Total mercury in liver and muscle tissue (NLET Method 02-2802) was analysed by cold vapour atomic absorption spectrometry (CVAAS), while methylmercury in muscle tissue was determined by GC–atomic emission detection (AED). Liver tissue was not analysed for MeHg. Analyses of 22 elements in liver and muscle samples (NLET Method 02-2705), were analysed by inductively coupled plasma-Sector Field spectrometry

(ICP-SFMS), with 20 elements analysed at low resolution (Ag, Ba, Cd, Co, Cr, Cu, Ga, La, Li, Mn, Mo, Ni, Pb, Rb, Sb, Sr, Tl, U, V and Zn) and the rest at high resolution (As and Se). Instrumental detection limits for most elements is 0.001  $\mu$ g/g, except for Sr (0.05 µg/g), Pt (0.01 µg/g), Pd (0.1 µg/g), Li  $(0.1 \ \mu g/g)$  and Sb  $(0.01 \ \mu g/g)$ . All elements except MeHg were analysed with wet tissue, and therefore are expressed as µg/g wet weight. MeHg concentrations were calculated using freeze-dried muscle samples, so to bring MeHg values in line with other element values, these were converted to wet weight values assuming 80% water for fish and invertebrates and 70% water for birds and mammals (unpublished data, Canadian Wildlife Services, Canada Centre for Inland Waters, Burlington, ON). NLET is certified by the Canadian Environmental Analytical Laboratory program of the Canadian Standards Association and participates in the QA program for the Northern Contaminants Program, with good results (Stokker et al., 2003).

## 2.3. Stable isotope analysis

Samples were analysed for  $\delta^{15}N$  and  $\delta^{13}C$  at the Department of Soil Science, University of Saskatchewan, Saskatoon, Saskatchewan. Samples were washed in distilled water, freeze-dried, powdered and treated with a 2:1 chloroform-methanol solution to remove lipids. Samples were then dried under a fume hood. Zooplankton and ice algae were soaked in a 0.1 N HCl to remove carbonates and allowed to dry without rinsing. One milligram homogenized samples were loaded into tin cups and combusting at 1800 °C in a Robo-Prep elemental analyzer. Resultant CO2 and N2 gases were then analysed using an interfaced 'Europa 20:20' continuous-flow isotope ratio mass spectrometer (CFIRMS), with every five samples separated by two laboratory standards Stable isotope abundances (albumen). were expressed in the  $\delta$  notation as the deviation from standards in parts per thousand (‰) according to the following equation:

$$\delta \mathbf{X} = \left( \left( R_{\text{sample}} / R_{\text{standard}} \right) - 1 \right) * 1000 \tag{1}$$

Where X is <sup>13</sup>C or <sup>15</sup>N and *R* is the corresponding ratio  ${}^{13}C/{}^{12}C$  or  ${}^{15}N/{}^{14}N$ . The  $R_{\text{standard}}$  values were

based on the PeeDee Belemnite for <sup>13</sup>C and atmospheric N<sub>2</sub> for <sup>15</sup>N. Replicate measurements of internal laboratory standards indicate measurement errors of  $\pm 0.1\%$  and  $\pm 0.3\%$  for  $\delta^{13}$ C and  $\delta^{15}$ N measurements, respectively.

## 2.4. Determination of relative trophic level

Relative trophic levels were determined using equations derived from the model reported in Hobson et al. (2002). Trophic level was determined relative to the copepod *C. hyperboreus* which we assumed occupied trophic level (TL) 2 as a primary herbivore (although it is known to be an omnivore during low productivity periods in the winter), and their overall  $\delta^{15}$ N value from all samples collected in 1998 and 1999 was  $7.7 \pm 0.1\%$  (Fisk et al., 2001). For each individual zooplankton, fish and ringed seal sample (whole or muscle), the following equation was used:

$$TL_{consumer} = 2 + \left(\delta^{15}N_{consumer} - 7.7\right)/3.8$$
 (2)

For sea birds, rearing studies suggest that diet-tissue isotopic enrichment factor is +2.4% and so we adopted the TL model developed in Hobson et al. (2002):

$$TL_{bird} = 3 + (\delta^{15}N_{bird} - 10.1)/3.8$$
(3)

Data analyses were done using SYSTAT version 10.0 (SPSS Inc, Chicago IL, USA). As a pelagic counterpart to benthic ice algae, the published data for POM (Hobson et al., 2002) were included in the food web analyses (Fig. 2). As Se and Hg are known to be antagonistic in many vertebrates, Se and THg wet weight concentrations were converted to molar values (atomic weights 78.96 and 200.59 g/mol, respectively) to calculate molar ratios. Sex differences in metal accumulation were not observed for most elements in all seabird species and ringed seals. The exceptions include As, Cd, Se and Sr (two-group *t*-test; p < 0.1) in black-legged kittiwake liver tissue as well as As and Cd in northern fulmar liver tissue. In general, both sexes of most species in this study exhibited similar bioaccumulation and biodilution patterns, therefore both sexes were pooled. Metals were log-transformed to normalize

the distribution, and simple linear regressions were performed using metal concentrations in muscle and whole animal tissue vs. trophic level (as indicated by  $\delta^{15}$ N values) as a possible tool to estimate metal levels in the food web (Ricker, 1973). Muscle tissue concentrations were used in order to enable comparisons with other biomagnification studies which usually examined muscle tissue from vertebrates and whole invertebrates. Metals were regressed against  $\delta^{13}$ C values in seabird samples to assess tissue-specific and possible diet-specific variation within the seabird portion of the food web. However,  $\delta^{13}$ C values were not included in metal trophodynamics for the whole food web because  $\delta^{15}N$ and  $\delta^{13}C$  of all sampled biota are correlated  $(p \le 0.01)$ . This correlation can be explained by the smaller trophic fractionation of  $\delta^{13}$ C ( $\approx 1\%$  to 2‰) that happens in unison with  $\delta^{15}N$  fractionation (3.2-3.6‰). Hobson et al. (2002) gives a detailed review of the NOW Polynya food web (with more species) and they also examine the interrelationships between  $\delta^{13}$ C and  $\delta^{15}$ N in depth, based on data for muscle tissue and whole organisms. Element values below detection limits were not included in the regressions, although the total numbers of data points above detection limit are included to indicate the degree of "censoring" (Helsel, 1990). Correlations of element concentrations in liver vs. muscle samples for each ringed seal and bird species were Bonferroni-corrected to provide protection for multiple tests.

## 3. Results

## 3.1. Food web structure

The food web structure of the North Water Polynya, based on nitrogen and carbon stable isotope values was consistent with that reported by Hobson et al. (2002), which included a much larger sample set and more species. As expected, a continuum of increasing  $\delta^{15}$ N values was observed from ice algae to zooplankton to Arctic cod (*B. saida*) to seabirds and ringed seals (*P. hispida*) (Fig. 2). Seabirds occupied a trophic gradient from the dovekie (*Alle alle*), which feed on zooplankton and fish (Karnovsky and Hunt, 2002), having a trophic level of about 3.2 to the glaucous gull (*Larus hyperboreus*), which feeds on fish, carrion and seabird chicks (Godfrey, 1986), having a trophic level of approximately 4.7 (Table 1).

## 3.2. Distribution of elements in the food web

Zinc (Zn) and copper (Cu), both essential nutrients, had the highest concentrations of all elements found in all biota, ranging from 2.9  $\mu$ g/g Zn and 0.8  $\mu$ g/g Cu in ice algae to 68.3  $\mu$ g/g Zn in northern



Fig. 2. The relationship between  $\delta^{15}$ N indicating trophic position and  $\delta^{13}$ C indicating dietary carbon source for 16 species from Northwater Polynya. The code for each species are as defined in Table 1. The POM data were obtained from Hobson et al. (2002).

Table 2

Means and standard deviations for selected metals ( $\mu$ g/g ww), stable nitrogen and carbon isotope values (‰) for whole (W), liver (L) and muscle (M) samples

Code	Tissu	ıe	n d	<sup>15</sup> N	$\delta^{13}C$	А	s	Cd	Cu	Mn
IALG	W		3	$5.13 \pm 0.13$	$-17.75 \pm 0.1$	10 (	$0.20 \pm 0.01$	$0.21 \pm .02$	$0.84 \pm 0.82$	$0.52 \pm .003$
CHYP	W		3	$7.83 \pm 0.01$	$-21.04 \pm 0.0$	05 1	$1.42 \pm 0.42$	$1.62 \pm 0.79$	$1.55\pm0.57$	$0.29 \pm 0.07$
ZOOP	W		4	$8.47 \pm 0.07$	$-21.09 \pm 0.0$	)4 1	$1.13 \pm 0.00$	$0.89 \pm 0.46$	$1.60\pm0.46$	$0.43\pm0.37$
TLIB	W		3	$9.69 \pm 0.05$	$-20.41 \pm 0.0$	07 (	$5.53 \pm 1.69$	$7.15 \pm 2.27$	$3.79 \pm 1.48$	$0.46 \pm 0.14$
MOCC	W		3 1	$0.40 \pm 0.08$	$-20.36 \pm 0.0$	)3 2	$2.65 \pm 0.16$	$1.17\pm0.24$	$3.83\pm0.55$	$1.14 \pm 0.32$
ACOD	W		1 1	3.88	-19.39	9	9.47	0.06	1.96	1.21
	L		2 1	4.14–14.18	-19.33 to -	19.23	11.3-40.9	1.2-1.3	1.00-9.43	0.76-0.91
DOVE	Μ		10 1	$0.85 \pm 0.43$	$-20.13 \pm 0.2$	23 (	$0.48 \pm 0.10$	$0.41\pm0.19$	$6.97 \pm 0.56$	$0.65\pm0.07$
	L		9 1	$2.01 \pm 0.27$	$-20.85 \pm 0.2$	27 1	$1.51 \pm 0.37$	$5.78 \pm 1.19$	$8.18 \pm 1.70$	$3.32\pm0.31$
BLKI	Μ		10 1	$3.32 \pm 0.40$	$-19.21 \pm 0.3$	32 3	$3.02 \pm 2.19$	$0.46 \pm 0.24$	$5.75 \pm 0.53$	$0.52\pm0.08$
	L		10 1	$5.20 \pm 0.79$	$-20.74 \pm 0.2$	29 8	$8.83 \pm 6.96$	$8.62\pm5.66$	$6.05\pm0.91$	$3.52\pm0.67$
BLGU	М		10 1	$3.69 \pm 0.11$	$-19.31 \pm 0.0$	07 1	$1.57 \pm 0.36$	$0.42 \pm 0.15$	$6.90\pm0.55$	$0.61\pm0.09$
	L		10 1	$6.45 \pm 0.46$	$-20.74 \pm 0.3$	35 (	$5.46 \pm 2.42$	$6.82 \pm 1.66$	$8.07 \pm 1.46$	$2.67 \pm 0.42$
TBMU	М		10 1	$3.50 \pm 0.59$	$-19.71 \pm 0.2$	29 3	$3.29 \pm 1.09$	$0.54 \pm 0.26$	$8.09 \pm 3.35$	$0.49 \pm 0.05$
	L		10 1	$5.16 \pm 0.37$	$-20.63 \pm 0.1$	15 12	$2.84 \pm 3.19$	$12.84 \pm 3.19$	$8.55 \pm 1.74$	$3.28 \pm 0.30$
IVGU	М		5	$14.1 \pm 0.64$	$19.32 \pm 0.2$	.9 2	$2.18 \pm 1.45$	$0.19 \pm 0.12$	$7.43 \pm 1.18$	$0.50 \pm 0.12$
	L		2 1	6.83-17.32	-20.63 to -	20.52	4.19-12.20	2.17-7.72	4.17-4.54	2.05-2.36
NOFU	М		10 1.	$4.21 \pm 0.48$	$-19.54 \pm 0.3$	30 2	$2.72 \pm 1.50$	$1.17 \pm 1.16$	$5.33 \pm 0.40$	$0.68 \pm 1.32$
	L		10 1	$5.34 \pm 0.47$	$-20.83 \pm 0.1$	10 0	$5.94 \pm 6.97$	21.84 + 13.22	$6.24 \pm 0.97$	$4.29 \pm 0.55$
GLGU	М		10 1	$6.52 \pm 0.73$	$-18.46 \pm 0.3$	34	$1.86 \pm 1.20$	$0.21 \pm 0.16$	$5.17 \pm 0.38$	$0.54 \pm 0.05$
	L		9 1	$7.81 \pm 1.02$	$-19.97 \pm 0.5$	58	7.567.62	4.80 + 3.72	$5.67 \pm 0.96$	$3.72 \pm 0.87$
THGU	М		1 1	7.2	-18.56	4	5.37	0.08	5.33	0.49
	L		1 1	8.4	-19.78	19	9.6	1.77	7.31	4.31
RSEA	М		9 1	$7.43 \pm 0.28$	$-18.04 \pm 0.3$	38 (	$0.30 \pm 0.15$	$0.07 \pm 0.06$	$1.29 \pm 0.31$	$0.19 \pm 0.09$
	L		9 n	a	na	1	$1.44 \pm 0.62$	$6.16 \pm 4.98$	$6.15\pm2.35$	$2.79\pm0.52$
Code	Tissue	п	Мо	Ni	Pb	Rb	Se	Sr	Zn	THg
IALG	W	3	$0.02 \pm 0.02$	$0.11 \pm 0.01$	$0.06 \pm 0.06$	$0.23 \pm 0.02$	$0.15 \pm 0.06$	$3.46 \pm 0.21$	$2.93 \pm 0.03$	0.003*
CHVP	W	3	$0.02 \pm 0.02$	$5 133 \pm 0.15$	$0.00 \pm 0.00$	$0.23 \pm 0.02$ $0.34 \pm 0.07$	$1.87 \pm 0.00$	$7.05 \pm 1.65$	$2.93 \pm 0.05$ 17.73 ± 0.65	0.005 + 0.017
ZOOP	W	4	$0.03 \pm 0.00$	$0.80 \pm 0.13$	$1.05 \pm 1.10$	$0.34 \pm 0.07$ $0.24 \pm 0.05$	$1.07 \pm 0.09$ $1.70 \pm 1.15$	$5.86 \pm 1.05$	$17.75 \pm 0.05$ $14.30 \pm 4.41$	$0.025 \pm 0.017$ $0.006 \pm 0.002$
TUB	W	3	$0.03 \pm 0.01$	$0.00 \pm 0.22$	$1.05 \pm 1.17$	$0.24 \pm 0.03$ $0.73 \pm 0.13$	$1.79 \pm 1.19$ $4.47 \pm 0.89$	$140.67 \pm 15.31$	$14.30 \pm 4.41$ $14.83 \pm 3.14$	$0.000 \pm 0.002$
MOCC	W	3	$0.07 \pm 0.01$	$1.20 \pm 0.58$	$0.23 \pm 0.02$ 1 53 ± 2 35	$0.73 \pm 0.13$ 0.53 ± 0.08	$4.71 \pm 1.12$	$74.20 \pm 21.58$	$14.03 \pm 3.14$ $14.47 \pm 1.76$	$0.020 \pm 0.009$
ACOD	W	1	0.08 ± 0.05	nd	0.13	0.39 1 0.08	1.04	173	57.8	0.04
neob	T	2	0.08_0.10	0.12_0.26	0.05_0.06	0.28_0.47	1.09_1.54	0.52_0.98	16.6-18.2	0.015_0.002
DOVE	M	10	0.03 - 0.10 0.04 + 0.01	0.12 - 0.20 0.04 + 0.01	$0.09 \pm 0.22$	$1.73 \pm 0.17$	1.09 - 1.04 1.98 + 0.61	0.32 - 0.98 $0.13 \pm 0.09$	10.0-10.2 $11.17 \pm 1.08$	0.013 - 0.002 0.08 + 0.02
DOVL	I	9	$0.04 \pm 0.01$ $0.70 \pm 0.08$	$0.09 \pm 0.06$	$0.09 \pm 0.22$ $0.27 \pm 0.61$	$1.75 \pm 0.17$ $1.95 \pm 0.29$	$3.85 \pm 0.83$	$0.13 \pm 0.07$ $0.12 \pm 0.07$	$32.27 \pm 4.82$	$0.00 \pm 0.02$ $0.27 \pm 0.08$
BLKI	M	10	$0.04 \pm 0.00$	$0.09 \pm 0.00$ $0.02 \pm 0.01$	$0.27 \pm 0.01$ $0.02 \pm 0.01$	$1.99 \pm 0.29$ $1.39 \pm 0.08$	$5.09 \pm 0.09$ $5.79 \pm 1.49$	$0.12 \pm 0.07$ $0.07 \pm 0.04$	$16.03 \pm 1.02$	$0.27 \pm 0.00$ $0.30 \pm 0.09$
BLIU	L	10	$0.67 \pm 0.00$ $0.62 \pm 0.07$	$0.02 \pm 0.01$ $0.07 \pm 0.04$	$0.02 \pm 0.01$ $0.02 \pm 0.01$	$1.89 \pm 0.00$ $1.89 \pm 0.27$	$11.22 \pm 4.50$	$0.58 \pm 0.76$	$35.55 \pm 8.33$	$1.05 \pm 0.03$
BI GU	M	10	$0.02 \pm 0.07$ $0.03 \pm 0.01$	$0.07 \pm 0.04$ $0.04 \pm 0.03$	$0.02 \pm 0.01$ $0.04 \pm 0.02$	$1.09 \pm 0.27$ $1.70 \pm 0.22$	$146 \pm 0.25$	$0.09 \pm 0.04$	$13.35 \pm 1.69$	$0.34 \pm 0.10$
DLGC	I	10	$0.03 \pm 0.01$ $0.71 \pm 0.14$	$0.04 \pm 0.03$	$0.04 \pm 0.02$ $0.03 \pm 0.02$	$1.70 \pm 0.22$ $1.96 \pm 0.28$	$4.76 \pm 0.29$	$0.05 \pm 0.04$ $0.25 \pm 0.12$	$15.55 \pm 1.07$ 35 80 + 4 17	$1.17 \pm 0.10$
TRMU	M	10	$0.12 \pm 0.08$	$0.09 \pm 0.02$ $0.29 \pm 0.29$	$0.05 \pm 0.02$ $0.06 \pm 0.05$	$1.90 \pm 0.20$ $1.73 \pm 0.16$	$1.85 \pm 0.93$	$0.23 \pm 0.12$ $0.23 \pm 0.25$	$15.00 \pm 1.17$	$0.33 \pm 0.09$
TDIVIC	I	10	$0.12 \pm 0.00$ $0.82 \pm 0.14$	$0.25 \pm 0.25$ $0.15 \pm 0.08$	$0.00 \pm 0.03$ $0.05 \pm 0.02$	$1.73 \pm 0.10$ $1.91 \pm 0.22$	$3.12 \pm 0.95$	$0.23 \pm 0.23$ $0.29 \pm 0.12$	$46.94 \pm 3.99$	$1.17 \pm 0.03$
WGU	M	5	$0.02 \pm 0.14$ $0.03 \pm 0.01$	$0.15 \pm 0.00$ $0.05 \pm 0.02$	$0.03 \pm 0.02$ $0.12 \pm 0.20$	$1.91 \pm 0.22$ 2.04 ± 0.30	$2.12 \pm 0.05$ 2.19 ± 0.54	$0.29 \pm 0.12$ $0.10 \pm 0.07$	$16.08 \pm 1.02$	$0.21 \pm 0.06$
1,00	I	2	$0.05 \pm 0.01$	0.01_0.01	$0.12 \pm 0.20$ 0.01 - 0.01	1 97_1 98	4 58_5 44	0.22_0.25	2450-3280	$0.21 \pm 0.00$ $0.80 \pm 0.18$
NOFU	M	10	0.03 + 0.01	$0.03 \pm 0.01$	$0.01 \pm 0.01$	$2.19 \pm 0.40$	$4.09 \pm 1.32$	$0.22 \ 0.23$ $0.14 \pm 0.16$	$20.56 \pm 2.79$	$0.39 \pm 0.10$
11010	T	10	$0.54 \pm 0.07$	$0.03 \pm 0.01$ $0.03 \pm 0.02$	$0.01 \pm 0.01$	$2.19 \pm 0.10$ 2 74 + 0 40	$10.03 \pm 2.51$	$0.25 \pm 0.12$	$63.21 \pm 15.03$	$3.41 \pm 1.62$
GLGU	M	10	$0.04 \pm 0.07$	$0.05 \pm 0.02$ $0.05 \pm 0.04$	$0.02 \pm 0.02$ $0.04 \pm 0.05$	$2.74 \pm 0.40$ $2.23 \pm 0.40$	$151 \pm 0.26$	$0.23 \pm 0.12$ $0.09 \pm 0.06$	2329 + 251	$0.80 \pm 0.27$
3200	L	9	$0.82 \pm 0.00$	$0.03 \pm 0.04$ $0.14 \pm 0.10$	$0.02 \pm 0.03$	$2.23 \pm 0.40$ 2 43 + 0 53	$3.80 \pm 0.57$	$0.05 \pm 0.00$ $0.26 \pm 0.17$	$40.36 \pm 7.58$	$2.00 \pm 0.27$ $2.92 \pm 1.06$
THGU	M	1	0.02 - 0.15	0.02	nd	1.65	1 79	0.02	26.7	0.48
11100	L	1	0.86	0.02	0.07	1.85	4 74	0.15	44 3	1 94
RSEA	M	9	0.01 + 0.01	0.05 + 0.03	$0.01 \pm 0.01$	$1.66 \pm 0.31$	$0.44 \pm 0.07$	$0.06 \pm 0.02$	$20.60 \pm 5.01$	$0.68 \pm 0.29$
10001	L	9	$0.67 \pm 0.01$	$0.05 \pm 0.03$ $0.05 \pm 0.03$	$0.01 \pm 0.01$	$2.14 \pm 0.38$	$10.19 \pm 6.72$	$0.10 \pm 0.02$ $0.10 \pm 0.05$	$43.73 \pm 6.06$	22.51 + 16.87
		~					/ _			

Due to limited space, each taxon is identified by a code which is defined in Table 1.

\* Only one sample of three samples had measurable THg levels.

fulmar (Fulmarus glacialis) liver and 9.0 µg/g Cu in thick-billed murre (Uria lomvia) liver (Table 2). Other essential nutrients, manganese (Mn), magnesium (Mg), nickel (Ni), molybdenum (Mo) and selenium (Se) are also consistently found above detection limits in all biota (Table 2). Non-essential elements, including arsenic (As), cadmium (Cd), lead (Pb), strontium (Sr) and mercury (THg) were also detected in all species (Table 2). Many metals were found at less than 0.01  $\mu$ g/g (or were below detection limits) in seals, fish and birds, but were found at higher concentrations in zooplankton, notably, the non-essential elements, silver (Ag), barium (Ba), lanthanum (La), lithium (Li), antimony (Sb), uranium (U), palladium (Pd), and Sr (Table 3). Two nonessential elements, thallium (Tl) and platinum (Pt) concentrations were typically below detection limits, with only 9 Tl 'hits' measured just above detection limit (0.002 to 0.004  $\mu$ g/g, all from thick-billed murre liver samples). Therefore Tl and Pt were excluded from trophodynamics analyses.

## 3.3. Trophic transfer of metals

In the NOW food web, log concentrations of THg, MeHg, Zn and rubidium (Rb) in muscle and whole samples were found to significantly increase ( $p \le 0.001$ ) with trophic level (Table 4, Fig. 3a). The slopes for those elements were 0.223 for log-transformed MeHg, a highly bioaccumulative compound, 0.197 for log THg, 0.073 for log Rb to 0.042 for log Zn (Table 4).

Significant negative slopes  $(p \le 0.001)$  were observed for log-transformed Cd (-0.093), log Pb (-0.123) (Table 4, Fig. 3b). Regressions for log Ni were not significant  $(p=0.170, r_{adj}^2=0.087)$ , but untransformed Ni regressed against  $\delta^{15}$ N were significant, with a slope of -0.089 for Ni  $(p \le 0.001$ ; Table 4). This suggests that there is consistent biodilution of those elements in muscle tissue, with a slower rate for Ni, through the food web.

As mentioned previously, certain elements were found to be highly concentrated in zooplankton relative to vertebrates (Table 3). The disparity in the concentrations between zooplankton and vertebrates resulted in significant negative regressions for logtransformed Ag, Ba, Li, Sr, V and Sb (slopes -0.039 to -0.222,  $p \le 0.001$ ), despite the lack of

elected 1	nean (	± SU) element	concentrations (µg	/g ww) in zooplant	kton and ice algae sai	mples				
ode	и	Ag	Ba	La	Li	Sb	U	Λ	Pd	Sr
ALG	3	≤0.001	$0.271\pm0.030$	$0.009\pm0.002$	$0.064 \pm 0.011$	≤0.001	$0.305\pm0.039$	$0.056\pm0.003$	$0.010\pm0.000$	$3.46 \pm 0.21$
<b>HYP</b>	б	$0.011\pm0.001$	$0.131\pm0.057$	$0.005\pm0.002$	$89.600 \pm 115.650$	$0.003 \pm 0.002$	$0.751\pm0.150$	$0.112\pm0.020$	$0.023\pm0.006$	$7.05\pm1.65$
OOP	2	$0.011\pm0.005$	$0.346\pm0.416$	$0.011 \pm 0.016$	$79.820 \pm 12.848$	$0.011\pm0.016$	$0.265\pm0.078$	$0.101\pm0.058$	$0.016\pm0.005$	$5.86 \pm 1.19$
LIB	9	$0.087 \pm 0.025$	$1.102\pm0.237$	$0.019\pm0.007$	$2.003\pm0.993$	$0.009 \pm 0.993$	$0.054\pm0.044$	$0.119\pm0.050$	$0.370 \pm 0.039$	$140.67 \pm 15.31$
10CC	б	$0.104\pm0.019$	$0.960\pm0.245$	$0.015\pm0.004$	$4.363 \pm 2.313$	$0.024\pm0.028$	$0.042\pm0.014$	$0.139\pm0.052$	$0.170 \pm 0.044$	$74.20 \pm 21.58$
ach taxe	n is	identified hv a	code which is de	efined in Table 1.	Excent for Sr (Tab	ole 2). these elem	nents are found a	t < 0.001  mg/s	w or less in ver	tehrate samples.

3

Table 4 Linear regression equations for metal concentration vs.  $\delta^{15}$ N relationships for NOW marine food web using whole invertebrates, Arctic cod and vertebrate muscle data

Object vs. $\delta^{15}$ N	п	Slope	Intercept	$r_{\rm adj}^2$	<i>p</i> -value	Notes
Log THg	92	0.197	-3.407	0.681	≤0.001	BM
Log MeHg	76	0.223	-3.882	0.742	$\le 0.001$	BM
Log Rb	94	0.073	-0.850	0.501	$\le 0.001$	BM
Log Zn	94	0.042	0.642	0.446	$\leq 0.001$	BM
Log Cd	94	-0.093	0.788	0.257	$\leq 0.001$	BD
Log Pb	88	-0.123	0.124	0.299	$\leq 0.001$	BD
Ni	81	-0.089	-0.040	0.184	$\leq 0.001$	BD
Log Ag	79	-0.084	-1.420	0.129	$\leq 0.001$	ZO
Log Ba	60	-0.145	0.363	0.373	$\leq 0.001$	ZO
Log Li	89	-0.222	1.766	0.341	$\leq 0.001$	ZO
Log Sr	93	-0.214	2.189	0.394	$\leq 0.001$	ZO
Log V	51	-0.138	0.131	0.437	$\leq 0.001$	ZO
Log Sb	85	-0.039	-2.164	0.065	0.011	ZO
Log As	94	0.011	0.020	0.000	0.507	NS
Log Co*	94	-0.030	-1.352	0.040	0.029	NS
Log Cu*	94	0.023	0.324	0.035	0.039	NS
Log Ga	94	-0.001	-1.976	0.000	0.914	NS
Log Mn	94	-0.015	-0.109	0.015	0.042	NS
Log Mo	93	-0.009	-1.313	0.009	0.385	NS
Log Se	94	-0.004	0.298	0.004	0.792	NS

Liver results are not included. Sample number (n) indicates number of samples above detection limit that were included in the regression. Total number of samples is 76 for MeHg samples and 94 for all other elements. Notes indicate whether regressions support biomagnification (BM), biodilution (BD), elevated concentrations in zooplankton only (ZO) or not significant trends (NS). Asterisks indicate metals that have significant *p*-values but are not considered biologically significant (see text for discussion).

consistent trends through the food web (Table 4). From Fig. 3c, zooplankton values were between one to three orders of magnitude higher than for the larger animals. The regression significance was due mostly to the pattern of clusters sharply offset from each other (Weisberg, 1985).

Non-significant slopes (p=0.029 to 0.792) were found for the remaining elements (Table 4), indicating that those elements do neither biomagnify nor biodilute through the food web. While Co and Cu have significant *p*-values (0.029 to 0.039), visual inspection of the plots show little change with trophic level with wide scatter present within each species (especially for dovekies, black-legged guillemots, black-legged kittiwakes and *C. hyperboreus*) and throughout the whole food web ( $r_{adj}^2=0.035$  to 0.040; Table 4), so those two elements are not considered to have the ability to consistently biomagnify in the NOW food web.

# 3.4. Seabird muscle and liver concentrations

Element concentrations were typically higher in liver tissue than in muscle tissue for seals and seabirds (Table 2). Significant and positive regressions are observed for MeHg and THg in liver and muscle tissue for all birds vs.  $\delta^{15}$ N ( $r_{adj}^2 = 0.441$  to 0.642,  $p \le 0.001$ ), with slopes (0.137 to 0.173; Table 5) lower than observed for the food web slopes (0.197 to 0.223; Table 4). As and Rb also both exhibited significant regressions with  $\delta^{15}N$  in both muscle and liver tissue  $(r_{adi}^2 = 0.117 \text{ to } 0.161, p \le 0.004; \text{ Table 5}),$ suggesting that trophic position was affecting the concentrations of those elements in seabirds, with higher trophic sea birds retaining higher concentrations than the lower trophic birds. However, As and Rb are not significantly regressed against  $\delta^{13}$ C in either liver or muscle tissue  $(r_{adj}^2 = 0.029 \text{ to } 0.000)$ , p=0.107 to 0.805). Zn in seabird muscle tissue is significantly regressed against both  $\delta^{15}N$  and  $\delta^{13}C$  $(p \le 0.001)$ , but not in liver tissue (p = 0.379) to 0.810). Cu concentrations exhibit weak significant regressions against both  $\delta^{15}N$  and  $\delta^{13}C$  in muscle and against only  $\delta^{15}$ N in liver tissue  $(r_{adj}^2=0.111$  to 0.124, p=0.002; Table 5). Both Li and Cd exhibit significant negative regressions against both  $\delta^{15}$ N and  $\delta^{13}$ C in liver and muscle tissue (Table 5).

Concentrations of elements, such as Li and Zn (Table 5), were significantly regressed against trophic position, as indicated by  $\delta^{15}$ N and with carbon source as indicated by  $\delta^{13}$ C in muscle tissue  $(r_{adj}^2=0.041 \text{ to } 0.631, p \le 0.001 \text{ to } 0.022)$ , but were not significantly regressed against with  $\delta^{15}$ N in liver tissue  $(r_{adj}^2=0.000 \text{ to } 0.013, p=0.195 \text{ to } 0.655; \text{ Table 5})$ . This suggests that while muscle concentrations of those metals were influenced by dietary patterns, these metals stored in the liver may be more affected by metabolic processes. The other elements, such as Pb, Ni, Co and Mo, did not exhibit significant relationships with either stable isotope signature in seabird liver tissue or muscle tissue (p > 0.5).

Correlations of various elements in liver against muscle can provide insights into how elements are distributed throughout the body. When all bird species were grouped together, most element concentrations in liver tissue were significantly correlated with muscle tissue, except Pb, Mn and Sr (Table 6). For individual species, correlations between liver and



Fig. 3. Mean  $\pm$  SD of log-transformed elements vs.  $\delta^{15}$ N and regressions of various metals vs.  $\delta^{15}$ N values of individual biota. The vertical dotted lines indicate which data points represent ice algae, zooplankton and vertebrates (seabirds, fish and seals). See Table 4 for regression equations. (a) shows the metals that are biomagnifying through the food web with regression lines; (b) indicates the metals that are biodiluting through the food web with regression lines; (b) indicates the metals that are biodiluting through the food web with regression lines; while (c) indicates selected metals that are higher in zooplankton than in vertebrates by at least an order of magnitude.

Object	n	Regression	Slope	Intercept	$r_{\rm adj}^{2}$	<i>p</i> -value
Bird muscle	55	Log MeHg vs. $\delta^{15}$ N	0.173	-3.066	0.642	≤0.001
		Log MeHg vs. $\delta^{13}$ C	0.517	8.477	0.473	≤0.001
Bird muscle	66	Log THg vs. $\delta^{15}$ N	0.155	-2.679	0.695	≤0.001
		Log THg vs. $\delta^{13}$ C	0.492	7.154	0.397	≤0.001
Bird liver	61	Log THg vs. $\delta^{15}$ N	0.137	-2.049	0.441	≤0.001
		Log THg vs. $\delta^{13}$ C	0.229	4.787	0.054	0.040
Bird muscle	66	Log As vs. $\delta^{15}$ N	0.084	-0.932	0.158	≤0.001
		Log As vs. $\delta^{13}$ C	0.123	2.610	0.025	0.107
Bird liver	61	Log As vs. $\delta^{15}$ N	0.091	-0.695	0.144	0.001
		Log As vs. $\delta^{13}$ C	0.031	1.369	0.000	0.805
Bird muscle	66	Log Rb vs. $\delta^{15}$ N	0.021	-0.030	0.161	≤0.001
		Log Rb vs. $\delta^{13}$ C	-0.032	0.871	0.029	0.090
Bird liver	61	Log Rb vs. $\delta^{15}$ N	0.009	0.159	0.117	0.004
		Log Rb vs. $\delta^{13}$ C	0.023	0.361	0.000	0.477
Bird muscle	66	Log Zn vs. $\delta^{15}$ N	0.053	0.474	0.614	≤0.001
		Log Zn vs. $\delta^{13}$ C	0.122	3.580	0.346	≤0.001
Bird liver	61	Log Zn vs. $\delta^{15}$ N	0.008	1.488	0.000	0.379
		Log Zn vs. $\delta^{13}$ C	0.009	1.789	0.000	0.810
Bird muscle	66	Log Cu vs. $\delta^{15}$ N	-0.022	1.092	0.124	0.002
		Log Cu vs. $\delta^{13}$ C	-0.062	-0.403	0.111	0.004
Bird liver	61	Log Cu vs. $\delta^{15}$ N	-0.021	1.159	0.117	0.004
		Log Cu vs. $\delta^{13}$ C	-0.023	0.361	0.000	0.477
Bird muscle	66	Log Cd vs. $\delta^{15}$ N	-0.070	0.517	0.101	0.005
		Log Cd vs. $\delta^{13}$ C	-0.244	-5.168	0.141	≤0.001
Bird liver	61	Log Cd vs. $\delta^{15}$ N	-0.058	1.771	0.082	0.014
		Log Cd vs. $\delta^{13}$ C	0.245	-4.174	0.083	0.014
Bird muscle	66	Log Li vs. $\delta^{15}$ N	-0.096	0.277	0.067	0.025
		Log Li vs. $\delta^{13}$ C	-0.302	-7.456	0.071	0.021
Bird liver	61	Log Li vs. $\delta^{15}$ N	-0.149	0.877	0.199	0.000
		Log Li vs. $\delta^{13}$ C	-0.419	-10.085	0.075	0.019

 Table 5

 Selected linear regression equations for elements vs. stable isotopes in seabird muscle and liver

muscle tissue concentrations were only consistently seen for gallium (Ga) for all bird species and ringed seal, while lithium (Li) in liver and muscle tissue were correlated for most bird species except blacklegged kittiwakes (Table 6). Dovekies (6 elements) and northern fulmars (7 elements) had more significant correlations of individual elements than the other species, while thick-billed murres and black guillemots had the fewest correlations, with only Ga and Li having significant correlations between liver and muscle tissue (Table 6).

#### 3.5. Mercury and selenium

The proportion of THg that is MeHg varied with each taxa. Percent MeHg ranges from 7.5% in *C. hyperboreus* to 100% in *M. oculata*, *T. libellula* and Arctic cod (Table 7). In birds and ringed seal, the

proportion of THg that was MeHg was around 80% (75% to 86% in muscle tissue).

Molar Se: Hg ratios were high in zooplankton and Arctic cod, exceeding 100. In birds, molar ratios in muscle tissue ranged from 5.3 to 67.2 and average 2.8 in ringed seal, while liver molar ratios ranged from 3.7 to 37.9 in birds and 2.2 in seals. None of the organisms sampled had molar Se:Hg ratios at 1:1. Significant positive correlation coefficients were found for molar Se and molar Hg for all individual invertebrate species, Arctic cod, both muscle and liver tissue for northern fulmars, muscle tissue of black guillemots and thick-billed murres and liver tissue of ringed seals (Table 7). In particular, logtransformed molar Se in ringed seal liver was significantly regressed against log-transformed molar Hg  $(\log \text{ molar } \text{Se} = -0.301 + 0.595 \ (\log \text{ molar } \text{Hg}),$  $r_{\rm adi}^2 = 0.960, \, p < 0.001$ ).

Table 6 Bonferroni-corrected multiple correlation coefficients of elements in liver vs. muscle tissue from ringed seals and seabirds

n	RSEA	All birds	DOVE	BLKI	BLGU	TMBU	NOFU
	9	50	10	10	10	10	10
As	0.960	0.869	0.753	0.962	0.133	0.559	0.881
Cd	-0.144	0.882	-0.317	0.586	0.022	0.497	0.928
Со	0.530	0.552	0.969	0.672	0.555	0.429	0.745
Cu	-0.189	0.599	0.011	0.745	0.638	0.782	0.166
Ga	0.946	0.957	0.986	0.997	0.993	0.989	0.984
Li	0.826	0.963	0.999	0.910	0.996	0.876	0.875
Mn	0.029	0.462	-0.284	0.804	0.326	0.500	0.415
Mo	-0.611	0.612	0.245	-0.493	-0.374	0.549	0.548
Ni	0.020	0.735	0.953	-0.532	0.387	0.726	0.239
Pb	-0.500	-0.015	-0.126	0.664	0.462	0.323	0.914
Rb	0.784	0.703	0.649	0.732	0.684	0.294	0.911
Se	0.582	0.906	0.992	0.987	0.293	0.715	0.713
Sb	0.604	0.873	0.928	0.988	0.234	0.537	0.820
Sr	-0.004	-0.067	-0.324	0.519	0.270	-0.062	-0.155
Zn	-0.470	0.743	0.020	0.271	-0.132	0.050	0.655
Hg	0.917	0.733	0.611	0.743	0.840	0.791	0.941

Each taxon is identified by a code which is defined in Table 1. Bold values are significant at p < 0.01 and Bonferroni probability < 1.

## 4. Discussion

## 4.1. Biomagnification of metals

Total mercury and MeHg were both found to biomagnify in the NOW Arctic marine food web, consistent with other studies on freshwater and marine food webs in the Arctic and sub-Arctic (Atwell et al., 1998; Power et al., 2002) and other regions (Cabana and Rasmussen, 1994; Kidd et al., 1995; Thompson et al., 1998; Bowles et al., 2001). This study found log concentration– $\delta^{15}$ N relationship slopes of 0.197 and 0.223 for THg and MeHg, respectively, providing further evidence that MeHg is the main biomagnifying agent for THg trophodynamics in food webs. These slopes are very similar to the slope of 0.20 reported for THg for an Arctic marine food web, which included many of the same species, in the adjacent Lancaster Sound (Atwell et al., 1998). In fact, the log THg (or log

Table 7

MeHg and THg concentrations (ng/g ww), molar Se:Hg ratios and proportion of THg that is MeHg in whole organism (W), liver (L) and muscle (M) tissue

Code	Tissue	п	MeHg	THg	Percent MeHg	Se:Hg (mol)	Correl.
СНҮР	W	3	$2.4 \pm 1.3$	$25.3 \pm 16.7$	$7.5 \pm 0.1$	$196.2 \pm 55.9$	0.914
ZOOP	W	4	$4.0 \pm 3.0$	$5.5 \pm 2.1$	$70.2 \pm 38.1$	$900.5 \pm 494.5$	0.058
TLIB	W	3	$28.3\pm0.8$	$20.3 \pm 9.0$	$100 \pm 0.0$	$609.7 \pm 175.4$	0.990
MOCC	W	3	$14.3 \pm 4.8$	$10.7 \pm 2.5$	$100 \pm 0.0$	$1121.8 \pm 17.6$	0.998
ACOD	W+L	3	40 (W)	$23.0 \pm 14.8$	100 (W)	$199.1 \pm 97.7$	0.668
DOVE	L	10	na	$273.6 \pm 79.6$	na	$37.9 \pm 12.6$	0.301
	Μ		$68.1\pm31.8$	$77.3\pm22.0$	$78.9 \pm 20.7$	$67.2\pm20.9$	0.381
BLKI	L	10	na	$1047.0 \pm 432.5$	na	$31.8 \pm 17.5$	-0.370
	М		$255.2\pm105.0$	$299.3 \pm 92.4$	$76.4 \pm 38.3$	$52.9 \pm 21.0$	0.213
BLGU	L	10	na	$1172.2 \pm 447.7$	na	$11.5 \pm 5.4$	-0.038
	М		$271.3\pm88.8$	$340.3\pm98.9$	$79.5 \pm 16.7$	$11.6 \pm 3.1$	0.673
TBMU	L	10	na	$1132.3 \pm 225.9$	na	$7.2 \pm 2.0$	0.265
	Μ		$200.9 \pm 65.1$	$333.1 \pm 91.0$	$77.0 \pm 30.1$	$14.1 \pm 4.9$	0.619
IVGU	L	10	na	678.0-929.0	na	14.9-17.2	na $(n \leq 2)$
	М		$205.1\pm105.8$	$213.6 \pm 61.0$	$85.8 \pm 15.5$	$26.9\pm7.5$	0.431
GLGU	L	10	na	$2923.3 \pm 1059.6$	na	$3.7 \pm 1.4$	0.034
	Μ		$621.3 \pm 334.7$	$801.0 \pm 267.0$	$74.9 \pm 23.7$	$5.3 \pm 1.9$	0.118
NOFU	L	10	na	$3411.8 \pm 1624.5$	na	$9.2 \pm 4.4$	0.525
	Μ		$327.4 \pm 156.0$	$391.4 \pm 142.5$	$81.3 \pm 18.6$	$28.5\pm8.5$	0.579
THGU	L	1	na	1940.0	na	6.2	na $(n \leq 1)$
	Μ		408.2	483.0	84.5	9.4	na $(n \leq 1)$
RSEA	L	9	na	$22506.1 \pm 16869.9$	na	$2.2 \pm 2.8$	0.982
	М		$564.6\pm245.2$	$676.4 \pm 288.8$	$78.8 \pm 14.2$	$2.8\pm3.7$	0.390

Each taxon is identified by a code which is defined in Table 1. For Arctic cod (ACOD), both whole  $(n \le 1)$  and liver  $(n \le 2)$  samples are included in Se:Hg calculations. Correlations (Correl.) of log molar Se and log molar Hg are shown for muscle and liver samples. Bold values are significant at p < 0.01.

MeHg) vs.  $\delta^{15}$ N slopes have been remarkably consistent between freshwater and marine ecosystems and between Arctic, temperate and tropical latitudes, with the majority of slopes falling between 0.2 and 0.3 (Bowles et al., 2001; Power et al., 2002; Campbell et al., 2003; Kidd et al., 2003). The results of these studies suggest that MeHg is biomagnified through these diverse food webs, as measured by  $\delta^{15}$ N, regardless of productivity (eutrophic vs. oligotrophic), latitude (Arctic vs. tropical) or salinity (marine vs. freshwater) of the ecosystem. Relationships between  $\delta^{15}N$  and organochlorine concentrations are not as likely to be so similar between systems because organochlorines accumulate in fatty tissues, which are influenced by different variables such as latitude and temperature (Borgå et al., 2004). In other words, it may be possible that MeHg, which is stored in protein, and  $\delta^{15}$ N, which tracks nitrogen ratios in protein, are linked by their close association with protein, and this avenue of research should be explored.

Rb and Zn were found to biomagnify in the NOW marine food web. Rb has recently been found to biomagnify consistently in other arctic and temperate aquatic food webs (Campbell et al., 2005). Zn also exhibited biomagnification properties in a Montana stream invertebrate food web (Quinn et al., 2003). The slopes of the log concentration– $\delta^{15}$ N relationships were at least one-third of those for THg and MeHg, therefore these elements biomagnify to a lesser extent relative to THg. Those findings suggest that trophic position may need to be factored in when considering Rb and Zn in Arctic marine biota. Rb is a naturally occurring alkali metal that is considered by many to be a biochemical analog for the essential element potassium K<sup>+</sup> (Peters et al., 1999). Biological uptake of Rb from food has been determined for forest and aquatic insects, particularly in tracer studies (Johnson and Reeves, 1995), and retained Rb in food items can be transferred to other trophic levels (Nyholm and Tyler, 2000). While Rb can be depurated from organisms (Johnson and Reeves, 1995; Peters et al., 1999), enough can be retained in tissues, perhaps the result of competing for K<sup>+</sup> locations on enzymes, for propagation through the food web, with a gradual accumulation with each trophic level. This closer balance between uptake and depuration is reflected in the low slope (0.073) compared to MeHg (Table 4).

Zn is an essential element with a long list of important metabolic functions, and most organisms have biochemical mechanisms to regulate the amount of Zn in their cells (Friberg et al., 1979). The discovery that Zn concentrations increased with trophic position was unexpected because it was originally hypothesized that the rate of Zn excretion would be relative to Zn ingestion, thereby ensuring a relatively constant concentration in biota throughout the food web. The fact that Zn is an essential element raises questions as to whether Zn biomagnifies or metabolic requirements vary between vertebrates and invertebrates. Given that Zn also showed biomagnification when only the seabird portion of the food web was considered suggest that it is at least partially a food web magnification phenomenon.

#### 4.2. Biodilution of metals

Biodilution of a number of elements was observed in the NOW food web. Biodilution can be defined here as a decrease in concentration of an element with increasing trophic level, i.e., a significant negative slope between log concentration and  $\delta^{15}N$  was found. Cd. Pb and Ni were all found to biodilute through the NOW marine food web (Table 4). Furthermore, the significant negative slopes of Cd and  $\delta^{15}$ N in bird liver and muscle (Table 5), further supporting biodilution of Cd in the seabird portion of the NOW food web. (No significant relationships were observed for Pb or Ni vs.  $\delta^{15}$ N in liver tissue.) Furthermore, while many metals do not show relationship with bird age, Cd have been found significantly correlated to age of thick-billed murres as well other bird species as reviewed by Donaldson et al. (1997). The birds in this study were not aged, but age may explain a portion of the variability for Cd, in addition to trophic position as indicated by  $\delta^{15}N$ values.

Cd and Pb are non-essential elements that compete for calcium (Ca<sup>2+</sup>) enzymatic locations. Pb is stored in bones, while Cd is stored mainly in liver and kidneys (Friberg et al., 1979). Ni is an essential element important for liver, hormonal and lipid regulation. These elements share a commonality in that while they have long half-lives in the body, they are poorly absorbed from food (Friberg et al., 1979). The long biological half-lives mean that biota will retain and gradually accumulate Cd, Pb and Ni in their muscle tissues throughout their lifespan, such a characteristic usually results in biomagnification. However, the poor absorption from diet and biodilution of the metal burden by body growth and cell turnover ensures reduction via trophic transfer. This is emphasized by the non-logarithmic trophic increase in Ni, which shows much slower rate of food web biodilution than either Pb and Cd. Invertebrates appear to bioconcentrate those elements which are then stored in the muscle tissues of their predators, but those elements are not efficiently transferred nor accumulated in muscle nor liver tissue through the food web.

## 4.3. Bioconcentration of metals

A diverse range of metals (Ag, Ba, La, Li, Sb, Sr, U and V) were found at higher concentrations in zooplankton relative to liver and muscle concentrations for birds, fish and seals (Table 3), resulting in significant negative food web regressions with  $\delta^{15}N$ values for many of the metals (Table 4, Fig. 3c). However, these relationships are misleading because the large 'offset' in metal concentrations between zooplankton and vertebrates creates two distinctive clusters of data. The trophodynamics of those metals have not been frequently assessed, either in the field or in the lab, but it is known that Ag (Ratte, 1999), U (Szefer, 1991), Sb (Maeda et al., 1997) and La (Tu et al., 1994; Moermond et al., 2001) are easily bioconcentrated from water by algae and zooplankton, but concentrations will decrease in higher trophic levels. Zooplankton in thick-billed murre stomachs from Barents Sea have been shown to have similar organochlorine contaminant concentrations to mixed field samples (Borgå et al., 2003), so it can be assumed that metal concentrations of zooplankton consumed by thick-billed murres and other species will have similar concentrations to field zooplankton samples. In other words, the metals listed above are easily taken up by pelagic plankton, but are not easily transferred to upper trophic biota muscle tissue. even when exposed to those levels in diets.

Strontium is a special case, as Sr is also a calcium analogue which accumulates in bones and other calcium-rich structures (Chowdhury and Blust, 2001). The high concentrations of Sr in whole Arc-

tic cod (173 µg/g) compared to Arctic cod liver  $(0.5-1.0 \text{ }\mu\text{g/g}; \text{ Table 2})$  is likely due to the importance of bone and other calcium structures as a Sr reserve (Chowdhury and Blust, 2001). Whole T. libellula and M. oculata also have elevated Sr concentrations relative to other zooplankton species, which may be associated with the larger amount of chitin borne by those species. Those concentrations are in line with those found for Antarctic krill Euphausia superba and other euphausiids (100 to 115 µg Sr/g whole body), with molting chitin accounting for most of the seasonal variation (Nicol et al., 1992). If only whole organisms are considered, then there is some evidence for trophic biomagnification of Sr from the invertebrates T. libellula and M. oculata to whole Arctic cod. Accordingly, it is recommended that any bioaccumulation study of Sr (as well as the calcium competitors, Cd and Pb as discussed above) in the environment either include whole organisms or sub-sampling the calcified structures of individual biota.

# 4.4. Metals that show a lack of trophic relationships

A lack of correlation with trophic level was observed for Se, Mn and Mo, all essential elements (Table 4). The same is argued for Cu and Co, due to the broad scatter across the food web and within species. Visual inspection of the log Cu and log Co plots indicated very little increase with trophic position as scatter was broad for all species. This is unlike Sb which has consistently low concentrations in vertebrates and wide scatter of values among invertebrates (Fig. 3c). It is noteworthy that many of the elements are essential (or non-toxic) at trace amounts and excessive amounts are usually regulated homeostatically (Friberg et al., 1979). The more toxic elements, As and Se also did not demonstrate trophodynamic behaviors in the NOW food web. Both organic and inorganic forms of As, a non-essential element, are easily absorbed from the GI tract, but are also rapidly depurated. This will minimize accumulation and transfer of As through the food web. It is interesting that while gallium (Ga), another non-essential element which is commonly used in the electronics industry, exhibited the most correlations between liver and muscle tissue in ringed seals and most seabird species (Table 7), but did not exhibit food web relationships (Table 4). While organisms will accumulate Ga proportionally throughout their bodies, Ga was not biomagnified or biodiluted through the NOW food web.

# 4.5. Liver vs. muscle

In both birds and mammals, the liver is integral to the metabolism of nutrients and essential elements as well as for removal of non-essential elements and toxins from the bloodstream. As a result, hepatic cells can have concentrated amounts of elements relative to muscle tissue as typified by higher concentrations seen for all seabird species and ringed seal in this study (Table 1). Significant correlations of Ga between liver and muscle were consistently observed for virtually all species, while Li correlations were seen for most seabird species (Table 7). This indicates that those trace elements were proportionally distributed throughout the body tissues. However, many elements, especially Sr and Zn, did not consistently show significant correlations between liver and muscle tissue across species (Table 7), which can be attributed to the different biological functions of those tissues and the tendency of Sr to be stored in calcified structures. Hepatic tissue has higher turnover and metabolic rates than muscle tissue and accumulation of many elements will vary. This has implications for biomagnification studies that use whole invertebrates but only muscle tissue of upper trophic biota, since whole-body values may have greater significance

Table 8

Literature values for most commonly measured elements (Cd, Se, THg; converted to µg/g ww) in various organisms from the Arctic

Organism	п	Year	Location	Tissue	Cd	Se	THg	Reference
CHYP	3	1988-90	Lancaster Sound	Whole	na	na	$0.012\pm0.01$	(Atwell et al., 1998)
ACOD	9	1988–90	Lancaster Sound	Muscle	na	na	$0.04\pm0.03$	(Atwell et al., 1998)
DOVE	1	1988–90	Lancaster Sound	Muscle	na	na	0.07	(Atwell et al., 1998)
BLKI	26	1991–92	Barents Sea	Muscle	0.1 - 0.5	0.9-1.8	0.03-0.1	(Savinov et al., 2003)
		"	·· »	Liver	0.03-9.0	1.5-3.5	0.1-0.3	" "
	6	1988–90	Lancaster Sound	Muscle	na	na	$0.4 \pm 1.1$	(Atwell et al., 1998)
	8	1984–85	NW Greenland	Muscle	$0.6 \pm 1.5$	$4.4 \pm 1.2$	$0.3 \pm 1.3$	(Dietz et al., 1996)
		"	·· »	Liver	$8.2 \pm 1.5$	$14.1\pm1.4$	$1.0 \pm 1.2$	" "
BLGU	21	1992	Barents Sea	Muscle	0.07 - 0.2	0.2 - 1.1	0.02 - 0.2	(Savinov et al., 2003)
		"	«« »	Liver	0.7 - 1.3	0.2 - 2.9	0.1-0.2	66 33
	6	1988–90	Lancaster Sound	Muscle	na	na	$0.3 \pm 0.7$	(Atwell et al., 1998)
NOFU	5	1988–90	Lancaster Sound	Muscle	na	na	$0.15\pm0.23$	(Atwell et al., 1998)
TBMU	34	1991–92	Barents Sea	Muscle	0.1-0.3	0.4 - 1.6	0.03-0.1	(Savinov et al., 2003)
		cc ??	«« »	Liver	1.2-3.9	0.8 - 2.8	0.1-0.3	cc >>
	4	1988–90	Lancaster Sound	Muscle	na	na	$0.4 \pm 0.8$	(Atwell et al., 1998)
	10	1986–90	Newfoundland	Muscle	0.1 - 0.4	0.6-0.8	0.1 - 0.2	(Donaldson et al., 1997)
	12	1984–85	NW Greenland	Muscle	$0.4 \pm 2.2$	$0.6 \pm 1.5$	$0.2 \pm 1.4$	(Dietz et al., 1996)
		,	·· »	Liver	$6.7 \pm 1.4$	$1.8 \pm 1.2$	$0.7 \pm 1.3$	cc >>
IVGU	8	1984–85	NW Greenland	Muscle	$0.4 \pm 2.3$	$1.6 \pm 1.2$	$0.2 \pm 1.7$	(Dietz et al., 1996)
		"	·· »	Liver	$7.5 \pm 1.5$	$5.9 \pm 1.4$	$0.7 \pm 1.8$	" "
NOFU	9	1984–85	NW Greenland	Muscle	$0.7 \pm 2.4$	$2.7 \pm 1.4$	$0.3 \pm 1.4$	(Dietz et al., 1996)
		"	«« »	Liver	$10.1\pm1.9$	$9.5 \pm 1.3$	$2.3 \pm 1.6$	66 <b>3</b> 3
GLGU	30	1991–92	Barents Sea	Muscle	0.02-0.1	0.2 - 1.6	0.03-0.1	(Savinov et al., 2003)
		"	·· »	Liver	0.2 - 2.0	0.9-5.6	0.1 - 0.4	" "
	4	1988–90	Lancaster Sound	Muscle	na	na	$0.4 \pm 0.7$	(Atwell et al., 1998)
	8	1984–85	NW Greenland	Muscle	$0.4 \pm 1.9$	$1.1 \pm 2.2$	$0.7 \pm 0.7$	(Dietz et al., 1996)
		,	·· ··	Liver	$12.6\pm2.1$	$3.7 \pm 1.5$	$2.7\pm1.7$	«« »»
RSEA	20	1988–90	Lancaster Sound	Muscle	na	na	$0.2 \pm 0.1$	(Atwell et al., 1998)
	10-43	1984-85	NW Greenland	Muscle	$0.2 \pm 2.8$	$0.3 \pm 1.3$	$0.2\pm2.6$	(Dietz et al., 1996)
		" "	« »	Liver	$17.2\pm3.9$	$1.8\pm1.8$	$1.6\pm1.4$	66 <b>33</b>

Each taxon is identified by a code which is defined in Table 1. Lancaster Sound (northern Baffin Island) and the northwestern Greenland sites are near the NOW polynya site. TBMU from Newfoundland were determined to have migrated from eastern Arctic. Barents Sea is the portion of Arctic Ocean shared between Norway and Russia.

when considering biomagnification and trophic transfer pathways.

#### 4.6. Mercury: a special case

Mercury is unique among the elements listed here in that it is highly bioaccumulative in the methylated form, methylmercury. It is associated with neurological damage in humans and biota (World Health Organization, 1990). In zooplankton, the amount of THg that is MeHg is related to their trophic level, with the predatory zooplankton *M. oculata* and *T. libellula* having the higher proportion of MeHg compared to filter-feeding *C. hyperboreus*. In the Arctic cod, the THg burden was virtually all MeHg. This is typical for most fish worldwide (Bloom, 1992), even if recently, questions were raised about the actual bioavailability of the MeHg in fish tissue to consumers (Harris et al., 2003).

Selenium is thought to form seleno-protein complexes with Hg, and Se-related pathways such as the glutathione peroxidase enzymes may be used to mitigate the toxic effects of Hg in fish (Elia et al., 2003), birds (Hoffman, 2002) and mammals (Gailer, 2002). A review of 2510 samples from 48 marine species collected in Greenland waters (Dietz et al., 2000) indicated that several Greenland seal species (ringed seal, harp seal *Phoca geoenlandica*, hooded seal Cystophora cristata, and bearded seal Erignathus barbatus) were reported to have mean Se:Hg molar ratio values of  $10.4 \pm 35.9$  for muscle tissue and  $26.3 \pm 3.88$  for liver tissue. In the same review, the authors found that seabirds of assorted species had molar ratios of  $20.7 \pm 19.6$  for muscle tissue and  $26.3 \pm 44.1$  for liver tissue, while whole crustaceans had molar ratios of 56.4. Dietz et al. (2000) only found a few instances where molar Se:Hg ratios were at 1:1 and those were typically only found in mammals with very high THg concentrations exceeding 200 ng/g. However, because the species composition of crustaceans in our study compared to those reviewed in Dietz et al's study is very different and because we only have 1 species of seal compared to 5 in Dietz et al's study, it is difficult to draw conclusions by comparing the results in both studies. On the other hand, since Dietz's study (13 seabird species) and our study (8 species) both include a broad range of seabird species, it is interesting to note that the seabirds in this study had somewhat similar molar Se: Hg ratios of 20 for liver and 26 for muscle tissue.

## 4.7. Regional patterns

Mercury concentrations and  $\delta^{15}$ N signatures in a very similar marine food web in Lancaster Sound



Fig. 4. Comparison of mean ( $\pm$  SD) THg (circles) concentrations and  $\delta^{15}$ N values (triangles) in species from NOW (white) and Lancaster Sound (black). As the data for Lancaster Sound obtained from Atwell et al. (1998) were in dry weight values, NOW values were also converted to microgram per gram dry weight using the conversion values in Methods.

(northern Baffin Island) collected in 1988-90 (Atwell et al., 1998) were similar to our values for samples collected 10 years later (Table 8, Fig. 4). This indicates that within the region of Baffin Bay, food webs may be spatially and temporally consistent. However, variation in  $\delta^{15}$ N and  $\delta^{13}$ C values between zooplankton from different regions of the Arctic have been reported (Schell et al., 1998). The similarities of our results to those from Atwell et al. (1998) were likely because the location were geographically close and Arctic cod, marine mammals and migratory seabirds can cover wide distances which would result in the incorporation of variable isotopic signatures from across a wide region. This is reinforced by the similarities in literature values and our muscle and liver concentrations for Cd, Se and THg for most birds, black-legged kittiwakes, thick-billed including murres, ivory gulls and glaucous gulls from northwestern Greenland and Newfoundland (Table 8). Similar bird species from Barents Sea in Europe tend to have lower concentrations of Cd, Se and THg (Table 8) relative to those from the Baffin Bay (Table 8). Zn and Cu in seabird liver and muscle tissue from Barents Sea (Savinov et al., 2003), however, tend to be similar to those for the bird species from the NOW polynya.

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