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Assessing element-specific patterns of bioaccumulation across New England lakes

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ABSTRACT

Little is known about differences among trace elements in patterns of bioaccumulation in freshwater food webs. Our goal was to identify patterns in bioaccumulation of different elements that are large and consistent enough to discern despite variation across lakes. We measured methylmercury (MeHg) and trace element (As, Cd, Hg, Pb, and Zn) concentrations in food web components of seven New England lakes on 3–5 dates per lake, and contrasted patterns of bioaccumulation across lakes, metals and seasons. In each lake, trace element concentrations were compared across trophic levels, including three size fractions of zooplankton, planktivorous fish, and piscivorous fish. The trophic position of each food web component was estimated from N isotope analysis. Trace element concentrations varied widely among taxa, lakes and sampling dates. Yet, we identified four consistent patterns of bioaccumulation that were consistent across lakes: (1) MeHg concentration increased (i.e., was biomagnified) and Pb concentration decreased (i.e., was biodiminished) with increased trophic position. (2) Zinc concentration (as with MeHg) was higher in fish than in zooplankton, but overall variation in Zn concentration (unlike MeHg) was low. (3) Arsenic and Cd concentrations (as with Pb) were lower in fish than in zooplankton, but (unlike Pb) were not significantly correlated with trophic position within zooplankton or fish groups. (4) Average summer concentrations of As, Pb, Hg, and MeHg in zooplankton significantly predicted their concentrations in either planktivorous or piscivorous fish. Our secondary goal was to review sampling approaches in forty-five published studies to determine the extent to which current sampling programs facilitate cross-lake and cross-study comparisons of bioaccumulation. We found that studies include different components of the food web and sample too infrequently to enable strong cross-lake and cross-study comparisons. We discuss sampling strategies that would improve our capacity to identify consistent patterns of bioaccumulation and drivers of elevated trace element concentrations under naturally high levels of variability.

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1. Introduction

Accumulation of toxic elements in lake food webs is an ongoing concern for human and wildlife health. At high levels, toxic elements can cause mortality or disease and suppress wildlife populations (Hinck et al., 2009). Even at low levels, effects of toxic elements together with other stressors can result in significant individual and population impacts on fish and other aquatic life (Chen et al., 2004; Folt et al., 1999). Terrestrial wildlife that consume contaminated aquatic organisms are also at risk of reduced reproductive success, hormonal changes, and motor skill impairment (Hinck et al., 2009; Scheuhammer et al., 2007). Human consumption of fish from thousands of inland lakes in the US and worldwide is restricted or subject to regulatory advisories because of concerns about exposure to elevated levels of toxic trace

elements and other contaminants (US EPA, 2007). The overall goal of this study and our related work (Chen et al., 2005; Chen et al., 2000; Karimi and Folt, 2006; Ward et al., 2010b) is to characterize the strongest drivers and most consistent patterns of trace element accumulation in fish and aquatic food webs under natural conditions. This knowledge will improve our ability to identify sites and conditions that are most likely to produce fish with elevated concentrations of trace elements and provide a quantitative basis for bioaccumulation models used in risk assessment.

Pollution, associated with urbanization, agriculture, mining, or localized atmospheric deposition can lead to elevated and variable concentrations of toxic trace elements in lake organisms (Chen and Folt, 2000; Driscoll et al., 2007; Renshaw et al., 2006; Suchanek, 2008). While this variation in inputs can account for differences between lakes, even lakes with similar inputs of trace element pollution can produce fish with widely different trace element concentrations, owing to variation in biological, geochemical, and environmental factors that affect trace element uptake and accumulation (Chen et al., 2009; Folt et al., 2002; Pickhardt et al., 2002; Pickhardt et al., 2005; Stemberger and Chen, 1998). Measuring trace element concentrations in organisms consumed by fish and throughout aquatic food webs has proven useful for identifying factors that drive such variation, especially

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for MeHg in fish (Chen et al., 2005; Chen et al., 2000; Garcia et al., 2007; Hall et al., 1997; Orihel et al., 2007; Ward et al., 2010b). Yet, food-web accumulation patterns of trace elements other than MeHg are not as well understood, with inconsistent results reported for some elements across studies (e.g. Barwick and Maher, 2003; Chen and Folt, 2000).

Our study involved sampling trace elements from biota across a range of lake types and watershed land uses. We excluded lakes receiving heavy point-source contamination to minimize confounding effects of large variations in trace element inputs or direct toxic effects of trace elements on organisms. In each lake, we measured a suite of trace elements across a trophic gradient of zooplankton and fish in order to 1) compare patterns of bioaccumulation within lakes for different elements; 2) measure the correlation in the concentrations of different elements between zooplankton and fish; and 3) characterize variation in zooplankton trace element concentrations associated with sampling effort (i.e. number and timing of sample dates) and technique (i.e. mesh size).

We assessed the importance of differences in sampling effort and technique to the interpretation of zooplankton trace element data by evaluating the sampling designs and results in forty-five published studies. Our goal was to illustrate how sampling can influence the interpretation of pattern and process (Folt et al., 1998) and offer suggestions for sampling designs that will facilitate broader cross-lake and cross-study comparisons. This is particularly important as national and international monitoring programs for assessing ecological risk due to MeHg exposure are being proposed and designed (Evers et al., 2008).

2. Methods

2.1. Study sites

We sampled seven lakes over the summer and fall of 2001 and 2002. In 2001, we sampled four lakes (Gregg Lake, Hillsborough County, NH; Island Pond, Cheshire County, NH; Turkey Pond, Merrimack County, NH; and Tewksbury Pond, Grafton County, NH), with sampling conducted in three bouts (24 June–4 July, 27 August–4 September, and 8 October–18 October) at each lake. In 2002, we sampled three lakes (Echo Lake, Rutland County, VT; Horseshoe Pond, Rockingham County, NH; and Post Pond, Grafton County, NH), with sampling conducted in five bouts (19–27 June, 9–11 July, 29–31 July, 23–27 August, 12–18 September) at each lake. All of the lakes were thermally stratified during sampling, except for the last sampling date in 2001 which occurred after fall mixing. Physical and biological characteristics of the study lakes are summarized in Table 1.

2.2. Field methods

All samples were taken from a fiberglass rowboat using trace element clean techniques as described in our earlier work (Chen and Folt, 2000; Chen et al., 2000). Briefly, zooplankton were collected with vertical tows of a plankton net in the deepest portion of each lake. Separate tows were collected with nets of different mesh size

for each of three zooplankton size fractions (cone net with 202 μm mesh, cone net with 100 μm mesh, Wisconsin net with 45 μm mesh). Small size fractions were subsequently sieved through the next larger mesh to remove larger organisms. On each sample date, we collected three replicates samples for each zooplankton size fraction. For each replicate of each size fraction, multiple tows were collected and composited to obtain sufficient biomass, then divided with a plankton splitter to provide separate aliquots for trace element analysis, isotope analysis, and biomass measurement. Trace element samples were filtered onto acid-cleaned Teflon filters in the field, along with a field blank filtered with deionized water. Zooplankton biomass and isotope samples were filtered onto pre-weighed glass filters. Zooplankton sampling gear was acid cleaned and rinsed in deionized water between all sample days. Fish were collected by gill and hoop nets from all seven study lakes on a single sample date per lake. A subset of the fish captured at each lake, representing the full range of sizes encountered for all of the abundant species in samples, was euthanized and placed in acid cleaned bags. All zooplankton and fish trace element samples were kept frozen until subsequent laboratory processing.

Particulate samples (0.45 μm – 45 μm) and filtered water samples were also collected for particulate trace elements, isotopes, and biomass, and dissolved elements analyses on all zooplankton sampling occasions. Particulate samples were taken by filtering 30–60 mL of water that had passed through the 45 μm filter onto a 0.45 μm glass (isotopes, biomass) or Teflon (elements) filter (3 replicates per lake per date). A field blank was collected by filtering a deionized water sample in the field using identical methods. Many of the particulate and water samples were below method detection limits (measured concentrations in samples were less than or similar to those in field blanks), so only the isotope data for particulates were used in the analyses presented here.

2.3. Laboratory methods

Sample preparation for trace element analyses followed methods detailed in (Chen and Folt, 2000; Chen et al., 2000). Zooplankton samples on Teflon filters were digested with an aqua regia solution (2:1 concentrated nitric acid and hydrochloric acid) heated to 70 °C for 8–10 h. After digestion, Teflon filters were removed and the sample was centrifuged. Trace element samples were analyzed by the Dartmouth Trace Element Analysis core facility by inductively-coupled plasma mass spectrometry (ICPMS). Zooplankton MeHg concentrations were measured using distillation, ethylation, purge and trap, and cold vapor atomic fluorescence spectrophotometry at Flett Research, Ltd. or isotope dilution gas chromatography-ICPMS at the Dartmouth trace element analysis core facility. The fish were homogenized whole in a food processor and replicate 1-g subsamples were digested in nitric acid and analyzed for trace elements as above. Quality control was ensured through the analysis of two standard reference materials (NIST 2976 mussel tissue and CRC DORM-2 dogfish muscle percent recovery \pm SE: As 100 \pm 2, Cd 109 \pm 2, Hg 113 \pm 8,

Table 1

Characteristics of the seven study lakes. Chlorophyll a (Chl), total phosphorous (TP), dissolved organic carbon (DOC), and specific conductivity (Sp Cond) were sampled at the location of zooplankton sampling and processed as in Chen and Folt (2005). Land cover data includes the percent area as developed (Dev), agricultural (Ag), forest, and wetland for the shoreline within 200 m of the lake and is from (Homer et al., 2004).

Lake	Chl (ug/L)	TP (ug/L)	DOC (mg/L)	Sp Cond ($\mu\text{S}/\text{cm}$)	pH	Avg depth (m)	Max Depth (m)	Lake Area (ha)	Dev	Ag	Forest	Wetland
Echo	4.1	5.5	5.8	179	8	7.1	13	21.5	7%	8%	72%	12%
Gregg	3.7	7	5.6	25	5.7	6.1	10.7	81.3	2%	2%	90%	7%
Horseshoe	11	19.6	6.7	392	6.1	2.4	7	17.8	59%	21%	10%	10%
Island	5.3	9.8	6.8	20	5.2	1.5	4	12.9	5%	24%	62%	9%
Post	5.5	4.6	4.3	82	6.6	7	11.6	46.1	13%	7%	67%	13%
Tewksbury	1.3	3.3	7.3	64	5.7	7.6	15.2	19	11%	5%	63%	21%
Turkey	3.8	12.8	9	174	6.2	0.9	4.6	66.4	0%	3%	52%	45%

Pb 112 ± 2 , Zn 97 ± 2) and two analytical blanks with every processing batch of 40 samples. Measured sample values were not corrected for recovery.

We did not conduct mercury speciation on fish samples. For data analysis, all Hg in fish was assumed to be MeHg (Bloom, 1992), so total Hg measurements were used for both Hg and MeHg in data analysis for fish samples, whereas MeHg and total Hg measurements in invertebrates were distinct (see Wyn et al., 2009 for a discussion of this approach). The assumption that all Hg in fish is in the form of MeHg is common, but may not be valid for low trophic-level fish (Lepak et al., 2009). This could lead to some bias in our analysis, but the consistent patterns of MeHg accumulation we observed from zooplankton to low trophic level fish and from low trophic level fish to higher trophic level fish (see results) suggests that this was not the case.

Glass fiber zooplankton filters and subsamples of homogenized tissue from each whole fish sample were dried to constant mass at 60 °C. Zooplankton biomass was calculated as the difference of the dried mass and the pre-weighed filter mass. A small section of each zooplankton isotope glass fiber filter or approximately 1 mg of homogenized fish sample was packaged in a tin capsule for stable carbon and nitrogen stable isotope analysis. Isotope analysis was conducted by continuous flow isotope ratio mass spectrometry at the UC Davis Stable Isotope Facility.

2.4. Data analysis

Our primary analysis of trace element bioaccumulation was a comparison of the lake-specific relationship between trace element concentration and trophic position across different lakes and elements. We calculated trophic position for each zooplankton and fish sample as the difference between the sample $\delta^{15}\text{N}$ and the lake-specific mean $\delta^{15}\text{N}$ of particulate samples divided by 3.4 (Post, 2002). Using simple linear regression, we calculated the slope of the trophic position-trace element concentration relationship for each lake using three datasets: just zooplankton (trophic gradient across three size fractions on each date), just fish (trophic gradient across species), and combined zooplankton and fish. Each individual fish and the mean of each zooplankton size fraction on a given date were considered independent observations. This approach allowed us to determine whether the overall trophic position-element concentration slope was driven by variation within zooplankton or fish groups, or by differences between zooplankton and fish.

We also tested whether zooplankton trace element concentrations predicted concentrations in fish. We only conducted this analysis for elements for which we found significant differences in zooplankton concentrations across lakes (see Section 3.3); Cd and Zn were excluded because our analysis found that concentrations of Cd and Zn in zooplankton did not differ across lakes. The species composition of fish samples varied across lakes, and no fish species were present in all lakes. Most element concentrations varied across fish species, often associated with differences in trophic position, so averaging element data across different species compositions for across-lake comparisons could confound analyses. We took two approaches to evaluate correlations between zooplankton and fish concentrations. First, we tested correlations for the specific species of planktivorous (pumpkinseed sunfish *Lepomis gibbosus*; 6 lakes) and piscivorous (chain pickerel *Esox niger*; 5 lakes) fish that were found across the most lakes. Second, we tested correlations using mean fish element concentrations across all fish species at each lake adjusted for differences in mean trophic position by using an analysis of covariance of fish element concentrations on lake and trophic position.

Finally, we assessed variation in zooplankton element concentrations associated with sampling strategy using hierarchical models. We used restricted maximum likelihood to estimate variance in zooplankton element concentrations associated with lake, sample date,

and size fraction. Sampling dates differed across lakes and years, so we included date as a nominal factor nested within lake. Lake and date were considered random factors and size fraction was considered fixed. The full model included two interactions: size fraction x lake, and size fraction x sample date. Due to the nesting structure, other interactions were not estimable.

All analyses of trace element concentrations were conducted on \log_{10} -transformed data as this yielded approximately normal residuals and homogeneous variance. We used JMP 5.01 (SAS Institute Inc, 2002) and the R statistical package (R Development Core Team, 2010) for statistical analyses.

3. Results

Across all lakes, we analyzed trace element concentrations and stable isotope ratios in 243 zooplankton samples (81 of each size fraction) and 128 fish of 11 different species. Our standardized sampling approach yielded comparable zooplankton and fish samples from all lakes (Tables 2, 3), with samples from all lakes spanning trophic positions from small zooplankton (median trophic position (TP) = 0.56) to piscivorous fish (adult chain pickerel, largemouth bass *Micropterus salmoides*, and smallmouth bass *M. dolomieu*; median TP = 2.6–3.4).

3.1. Trace element accumulation through the food web

Although concentrations of most trace elements ranged widely across lakes (Tables 2, 3), each trace element displayed a characteristic pattern of variation across taxa and trophic position that was consistent across lakes (Fig. 1). As and Cd concentrations were always much higher in zooplankton than fish, driving a negative slope with trophic position when the two groups were combined (Figs. 1, 2). However, As and Cd slopes were not consistently negative within trophic groups (Figs. 1, 2), so differences in accumulation between zooplankton and fish drive the strong overall negative relationship, not biodiminishing concentrations with increasing trophic position. Total Hg concentrations were not consistently different between zooplankton and fish. Total Hg had a weak tendency toward biodiminishing concentrations in zooplankton, but biomagnified consistently in fish (Figs. 1, 2). MeHg consistently biomagnified both within and across zooplankton and fish analyses in all lakes (Figs. 1, 2; analysis for MeHg in fish is the same as for total Hg). In contrast, Pb concentrations consistently biodiminished both within and across zooplankton and fish (Figs. 1, 2). Zn concentrations were slightly higher in fish than zooplankton (Figs. 1, 2) but were overall much less variable across lakes and taxa than all other trace elements.

3.2. Correlation of zooplankton and fish

Zooplankton trace element concentrations predicted concentrations in planktivorous or piscivorous fish for all trace elements whose mean concentrations in zooplankton varied consistently across lakes (As, Hg, MeHg, and Pb; see section 3.3). In analyses of fish of a single species, zooplankton MeHg and Pb concentrations predicted Hg and Pb concentrations in planktivorous pumpkinseed sunfish and mean As, Hg, and MeHg concentrations in zooplankton predicted As and Hg concentrations in piscivorous chain pickerel (Table 4).

3.3. Components of variation in zooplankton trace element concentrations

Variation among replicate trace element samples for a given lake, date, and size fraction (residual variation) was always <25% of total variation in zooplankton trace element concentration, indicating that our sampling technique yielded reproducible results and effectively characterized concentrations for a given lake, date, and size fraction (Table 5). Despite substantial within-lake variation

Table 2

Characteristics of zooplankton samples from each study lake. Biomass and percent cladocera values are averages (standard errors) across all sample dates. Trace element concentrations are the ranges across all sample dates.

Lake	Size Fraction	Biomass	% Cladocera	As (ppb)	Cd (ppb)	Hg (ppb)	Pb (ppb)	MeHg (ppb)	Zn (ppb)
Echo	45–100 um	5.5 (1)	2.4 (0.6)	630–7200	310–4500	54–700	470–5800	3.6–18	2400–9400
	100–202 um	3.6 (0.6)	21 (10)	1100–4300	320–14000	110–520	430–5200	6–35	3700–7700
	>202 um	110 (10)	83 (5)	1100–5800	300–1900	54–430	95–1700	40–100	5100–18000
Gregg	45–100 um	9.8 (3)	9.1 (6)	430–3900	230–44000	88–1900	3100–12000	13–46	740–18000
	100–202 um	10 (3)	6.5 (3)	1100–5000	290–4900	350–4600	6200–20000	35–90	520–28000
	>202 um	30 (10)	4.9 (1)	790–4400	660–11000	390–3700	1800–7100	20–170	4100–18000
Horseshoe	45–100 um	57 (10)	39 (7)	1600–14000	230–13000	89–290	1900–15000	5.8–25	2600–6500
	100–202 um	46 (9)	50 (8)	2500–7200	460–4600	140–310	1000–16000	15–120	7100–12000
	>202 um	350 (90)	43 (4)	1100–3600	220–1000	91–310	360–1300	36–170	8100–15000
Island Pond	45–100 um	13 (6)	37 (4)	4000–20000	380–2300	1100–2000	4900–68000	46–130	4400–23000
	100–202 um	12 (5)	41 (4)	1300–6600	330–2600	800–1700	4300–29000	47–280	1300–24000
	>202 um	71 (30)	12 (7)	1300–2700	180–5200	180–1600	730–12000	76–410	1300–22000
Post Pond	45–100 um	12 (3)	23 (3)	1000–8400	100–24000	83–1300	220–9700	2.5–30	2700–5400
	100–202 um	9.8 (2)	30 (5)	1400–7000	340–5100	270–3500	780–6000	5.9–54	7100–14000
	>202 um	81 (5)	47 (4)	260–4200	160–2400	200–570	160–1000	7.3–130	2400–19000
Tewksbury	45–100 um	7.3 (0.7)	68 (7)	670–3200	100–7000	360–3000	3300–17000	6–23	1100–40000
	100–202 um	7 (0.2)	71 (9)	1400–15000	350–2300	210–550	4100–12000	7.8–30	2700–13000
	>202 um	14 (3)	32 (8)	1200–4000	440–8100	200–1200	980–4400	28–170	1800–28000
Turkey	45–100 um	15 (7)	11 (8)	1700–6700	260–8000	830–2200	9100–42000	7.5–43	2800–26000
	100–202 um	16 (10)	14 (6)	3600–6600	240–5100	330–1400	16000–51000	19–63	4800–9400
	>202 um	45 (40)	1.6 (0.1)	1400–6700	79–2700	350–5200	1000–18000	19–94	2500–25000

associated with sample date and size fraction, we detected significant across-lake variation in zooplankton concentrations of Hg, MeHg, and Pb, and marginal across-lake variation for As (Table 5). Some of the variation we identify as across-lake variation may in fact be due to differences between the two study years, but we cannot evaluate this directly because we did not sample the same lakes in the two years.

The largest source of random-effects variation for element concentrations in zooplankton was the interaction between date and size fraction within lakes (Table 5). Given the nested analysis, the substantial variation associated with the interaction of date and size fraction indicates that there is a specific pattern of temporal variation for each size class within each lake. Overall, within-lake temporal variation in zooplankton trace element concentrations was substantial, with a median 5-fold range across dates across all lakes and size fractions.

In the fixed-effect analysis of differences across zooplankton size fractions, only MeHg and Pb showed consistent trends. MeHg concentrations were significantly higher in larger size fractions, while Pb concentrations were lower. These size-related differences are consistent with our regression analyses of element concentrations and trophic position, as larger zooplankton size fractions also had significantly higher trophic position than small zooplankton (fit with same model as element concentrations; mean trophic position; large size fraction 1.26, middle size fraction 0.73, small size fraction 0.52; $F_{2,12} = 36.8$, $P < 0.0001$).

4. Discussion

Each trace element had a consistent pattern of accumulation through the food web, and patterns differed widely across elements. These element-specific bioaccumulation patterns were consistent

Table 3

Characteristics of fish samples from each study lake. Mass and trace element concentrations are the ranges across all individual fish of each species.

Lake	Common name	Latin name	N	Mass (g)	As (ppb)	Cd (ppb)	Hg (ppb)	Pb (ppb)	Zn (ppb)
Echo	Chain Pickerel	<i>Esox niger</i>	3	239–482	21–28	2.9–21	170–190	13–31	21000–35000
	Pumpkinseed	<i>Lepomis gibbosus</i>	3	33–117	110–210	7.2–11	130–220	24–36	12000–15000
Gregg	Brown Bullhead	<i>Ameiurus nebulosus</i>	2	546–588	3.8–5.2	11–13	81–99	45–56	7300–7800
	Chain Pickerel	<i>Esox niger</i>	5	174–447	6.6–19	5.5–15	400–1200	11–86	25000–43000
	Golden Shiner	<i>Notemigonus crysoleucas</i>	4	80–88	15–42	19–29	78–120	52–130	16000–22000
	Pumpkinseed	<i>Lepomis gibbosus</i>	3	39–196	29–63	29–69	180–910	110–190	13000–19000
	Smallmouth bass	<i>Micropterus dolomieu</i>	11	146–1050	8.2–35	6.2–56	520–3700	7.2–360	8400–12000
	Yellow perch	<i>Perca flavescens</i>	8	60–384	4.4–18	20–30	270–790	62–520	7000–13000
Horseshoe	Chain Pickerel	<i>Esox niger</i>	3	425–480	40–48	6–8	320–490	21–29	21000–32000
	Pumpkinseed	<i>Lepomis gibbosus</i>	7	7–93	88–180	11–60	46–160	29–130	9400–21000
Island	Brown Bullhead	<i>Ameiurus nebulosus</i>	5	89–326	14–51	17–29	94–280	84–160	9300–13000
	Chain Pickerel	<i>Esox niger</i>	10	12–714	14–95	18–41	440–1900	18–63	17000–73000
	Golden Shiner	<i>Notemigonus crysoleucas</i>	3	21–69	20–24	21–42	660–2300	100–140	12000–19000
	Largemouth bass	<i>Micropterus salmoides</i>	7	1–1100	12–62	21–76	370–3100	33–120	7400–23000
	Pumpkinseed	<i>Lepomis gibbosus</i>	8	24–79	15–66	25–67	340–1800	82–240	11000–21000
	Chain Pickerel	<i>Esox niger</i>	3	236–388	83–83	46–46	200–460	31–31	38000–38000
Post Pond	Pumpkinseed	<i>Lepomis gibbosus</i>	9	3–7	56–160	18–120	34–360	56–310	12000–23000
	Blacknose dace	<i>Rhinichthys atratulus</i>	4	1–2	89–110	10–54	150–180	7.1–23	16000–47000
	Brown Trout	<i>Salmo trutta</i>	2	140–141	250–370	7.6–20	190–230	13–150	24000–41000
	Common shiner	<i>Luxilus cornutus</i>	3	2–6	42–71	19–28	140–160	28–54	66000–86000
	White Sucker	<i>Catostomus commersonii</i>	12	71–191	35–140	4.2–17	36–170	11–68	16000–73000
Turkey	Golden Shiner	<i>Notemigonus crysoleucas</i>	3	5–68	27–98	0.09–6	100–170	60–220	15000–41000
	Pumpkinseed	<i>Lepomis gibbosus</i>	8	32–126	48–120	2–10	100–300	45–360	11000–19000
	Yellow perch	<i>Perca flavescens</i>	7	132–322	15–75	3.2–12	290–710	47–100	12000–17000

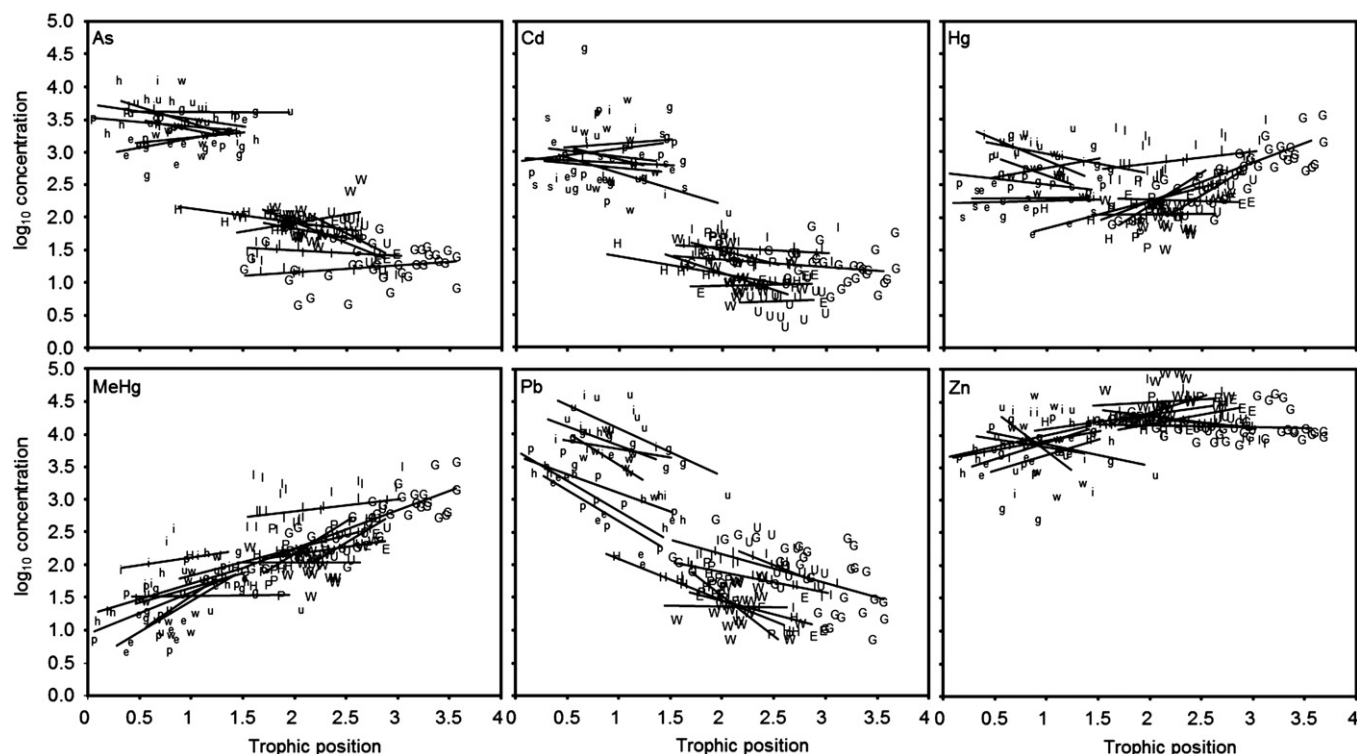


Fig. 1. Trace element concentrations (ppb dry basis) in zooplankton (small letters) and fish (capital letters) related to trophic position. Letters used as markers are the first letter in the name of the sample lake except for Tewksbury (W) and Turkey (U). Points for zooplankton are the mean of three replicates from the lake on a single sample date. Points for fish are individuals. Lines are the linear regression of \log_{10} trace element concentration on trophic position, with a separate regression for zooplankton and fish samples at each lake.

across lakes, despite substantial differences in overall concentrations of most trace elements across lakes. These results reinforce the importance of characterizing element concentrations throughout aquatic food webs and across multiple sites in order to identify factors that drive increased concentrations of toxic trace elements in fish. Standardized sampling of a large trophic gradient across multiple lakes allowed us to identify consistent patterns of bioaccumulation and drivers of elevated trace element concentrations in fish.

4.1. Trace element accumulation through the food web

Much prior work on bioaccumulation of trace elements in aquatic food webs has focused on Hg and MeHg. Our observations of consistent biomagnification of MeHg with increasing trophic position agree with other studies in marine, freshwater, lotic, and lentic systems. The slope of the relationship between trophic position and MeHg concentration within a food web is noted for being remarkably consistent across all of these systems. The overall median slope for \log_{10} MeHg concentrations across trophic position we observed in this study was 0.49 (range 0.44–0.63; equivalent to a slope of \log_{10} MeHg concentration on $\delta^{15}\text{N}$ of 0.14–0.20). The slope we observed is on the low end, but within the range of slopes reported in the literature for other systems (e.g. freshwater lakes: 0.54–1.46 (Gantner et al., 2010), 0.54–0.88 (Swanson and Kidd, 2010) 0.65–0.78 (Wyn et

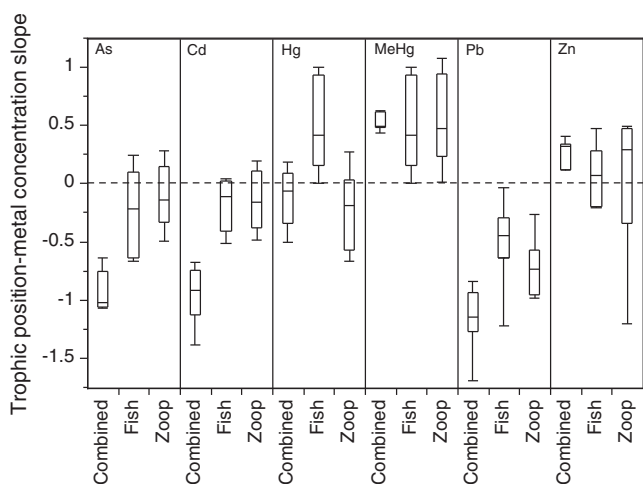


Fig. 2. Slope estimates for the relationship between \log_{10} trace element concentration and trophic position for combined analysis across all samples or for fish and zooplankton analyzed separately. Each slope estimate is from a linear regression for data from a single lake. Boxes show the median, 25th and 75th percentiles, whiskers extend to the highest and lowest estimates. Positive slopes indicate biomagnifying concentrations, negative indicate biodiminishing.

Table 4
Correlation of zooplankton and fish trace element concentrations for planktivorous pumpkinseed sunfish (N=6), piscivorous chain pickerel (N=5), and overall fish mean trace element concentrations adjusted for trophic position (N=7).

Fish variable	Element	Zooplankton–fish correlation	t	P-value
Pumpkinseed	As	0.03	0.1	0.95
	Hg	0.55	1.3	0.26
	MeHg	0.79	2.5	0.06
	Pb	0.91	4.5	0.01
Pickerel	As	0.95	5.1	0.01
	Hg	0.87	3.1	0.04
	MeHg	0.95	5.3	0.01
	Pb	0.46	0.9	0.42
Adjusted mean	As	0.42	1.0	0.36
	Hg	0.43	1.1	0.35
	MeHg	0.95	6.8	0.003
	Pb	0.77	2.7	0.05

Table 5

Components of variation in zooplankton trace element concentrations. Percentages following random effects components are the percent of random effects variation accounted for.

Metal	Total variance	R2	Random effects variance	Lake variance	Date variance	Lake x size fraction variance	Date x size fraction variance	Residual variance	Size fraction F-value*	Size fraction P-value
As	0.09	79%	0.09	0.01 (11%)	0.004 (4%)	0.004 (5%)	0.05 (59%)	0.02 (22%)	2.7	0.10
Cd	0.24	76%	0.24	0 (0%)	0.06 (25%)	0 (0%)	0.12 (51%)	0.06 (24%)	1.4	0.28
Hg	0.15	90%	0.17	0.06 (36%)	0.05 (29%)	0.01 (9%)	0.03 (17%)	0.02 (9%)	0.9	0.42
MeHg	0.21	94%	0.14	0.06 (38%)	0.02 (11%)	0.01 (8%)	0.05 (33%)	0.01 (10%)	30.6	<0.0001
Pb	0.37	94%	0.31	0.18 (58%)	0.06 (18%)	0.006 (2%)	0.05 (15%)	0.02 (7%)	45.1	<0.0001
Zn	0.28	88%	0.28	0 (0%)	0.19 (68%)	0 (0%)	0.06 (21%)	0.03 (11%)	2.3	0.15

*2 numerator degrees of freedom, 12 denominator degrees of freedom.

al., 2009); streams: 0.48–0.92 (Chasar et al., 2009); oceans: 0.76 (Campbell et al., 2005), 0.95 (Senn et al., 2010); estuaries: 0.20 C.Y. Chen, unpublished data; converted from $\delta^{15}\text{N}$ to trophic position where applicable by multiplying by 3.4).

As expected, the pattern of accumulation through the food web for total Hg was more variable and less prone to biomagnification than for MeHg. For example, (Campbell et al., 2005) report a slope of 0.63 for the relationship between Hg concentration and trophic position in Hudson Bay, less than the 0.95 observed for MeHg, but still significantly positive. Yet, we found that total Hg did not biomagnify at all for zooplankton, as with other studies in which vertebrates and invertebrates were analyzed separately (Atwell et al., 1998). This pattern emphasizes the necessity of distinguishing Hg species in low trophic level organisms in order to assess MeHg accumulation potential in predators. Nonetheless, as we found in this and previous work (Chen and Folt, 2005; Chen et al., 2005; Ward et al., 2010a), total Hg in prey can still be a useful predictor of Hg concentrations in their predators when variation in the proportion of Hg as MeHg is small relative to variation in Hg concentrations. This is important, because it is much more difficult and expensive to measure MeHg than total Hg in invertebrates and other small prey.

Relative to Hg and MeHg, much less is known about patterns of accumulation of other trace elements through aquatic food webs (see review in Luoma and Rainbow, 2008). Other studies that measure Pb typically report biodiminishing concentrations with trophic position, similar to those we observed (Campbell et al., 2005; Chen and Folt, 2000). Some others suggest that As (Campbell et al., 2005; Chen and Folt, 2000; but see Barwick and Maher, 2003) and Cd (Mathews and Fisher, 2008) concentrations also biodiminish with trophic position. Yet, we found that this pattern is primarily driven by the lower concentrations of As and Cd measured in fish relative to zooplankton, with no evidence for biodiminishing concentrations within groups of fish or zooplankton. This result is similar to that reported by (Campbell et al., 2005) for Cd concentrations in fish and zooplankton and indicates that there may be key differences in physiology or exposure routes of these elements between zooplankton and fish taxa that lead to widely divergent element concentrations independent of trophic position.

As with previous work that reports Zn biomagnification (Campbell et al., 2005; Chen et al., 2000; Mathews and Fisher, 2008), we found that Zn concentrations were slightly higher in fish than zooplankton, with a weak trend toward biomagnification within groups. Overall, though, variation in Zn concentrations was much lower than for other elements, possibly due to physiological regulation of internal concentrations of this essential element (Karimi and Folt, 2006).

Variation in the partitioning of the different elements in tissues within individual organisms may explain some of the patterns of bioaccumulation that we observed. For example, relatively high concentrations of As and Cd in invertebrates relative to fish may be related to accumulation of these elements in compartments specific to invertebrates, such as surface load adsorbed to chitinous exoskeletons (Cain et al., 1992; Reinfelder and Fisher, 1994). Surficial element load would lead to high measured whole-body concentrations in invertebrates, yet not represent a pool that is readily assimilated in predators

(Reinfelder and Fisher, 1994), potentially explaining why concentrations declined from invertebrate prey to vertebrate predators without any indication of biodiminishing concentrations within groups. However, this mechanism may not be consistent with all of our observations. Previous studies have found Pb and Zn can accumulate by adsorption to exoskeletons (Cain et al., 1992; Reinfelder and Fisher, 1994), yet we found consistent patterns of accumulation of these elements along a trophic gradient in both vertebrates and invertebrates.

4.2. Correlations among element levels in zooplankton and fish

For all elements whose zooplankton trace element concentrations differed across lakes, zooplankton element levels predicted levels in planktivorous or piscivorous fish. This finding underscores the need to measure zooplankton trace element concentrations to interpret across-lake differences in element concentrations in fish; the differences in fish appear likely to arise from differences in bioaccumulation at the base of the food web (Wyn et al., 2009). Even for Pb and Cd, elements that can accumulate in fish via aqueous exposure, accumulation under typical field conditions is frequently dominated by dietary uptake (Mathews and Fisher, 2009). Interestingly, MeHg is the only analyte that showed consistent correlation between zooplankton and fish in all analyses; MeHg is unique among the elements we analyzed for accumulating almost entirely through food under a wide range of exposure conditions (Harris and Bodaly, 1998; Pickhardt and Fisher, 2007).

4.3. Evaluation of sampling designs for zooplankton trace elements in 45 published studies

Determination of trace elements in zooplankton is becoming increasingly common, particularly for Hg and MeHg. Yet, sampling techniques, sample size, and study design vary widely across studies, in ways that can hamper cross-study and cross-lake comparisons. To assess the efficacy of various designs and test for emergent patterns across lakes, we evaluated sampling approaches in 45 published studies reporting zooplankton Hg and methylmercury (MeHg) concentrations from 1990–2008 (ISI Web of Science search keywords: [mercury OR methylmercury OR Hg] AND zooplankton AND [lake OR pond OR freshwater]; the search returned 82 results, the 45 that reported concentrations in field-collected zooplankton were included in the review). We focused on two particular aspects of sampling design that our findings indicated were likely to influence estimates of trace element concentrations in zooplankton: replication and size fractionation.

Much has been written about the importance of aligning the sampling design to the question being asked (reviewed in Folt et al., 1998). The ultimate tradeoff that determines the optimal design of sampling programs depends on the dimensions along which most variation occurs. We found significant temporal variation in all element levels of zooplankton, suggesting that considering the allocation of sampling effort among dates is essential to effective design. Replicate sampling on a single date in a lake clearly provides power for making within-lake comparisons on a single date (e.g. high spatial resolution). However, sampling over multiple locations and days is preferable if

one seeks to characterize and compare overall trace element concentrations in zooplankton across lakes and seasons (e.g. spatially extensive sampling). While sampling on a single date yields high apparent power to detect differences across lakes (Fig. 3a), the true confidence interval for the estimate of the difference between lakes based on a single sampling date is very large because the effect of lake and date are entirely confounded (Fig. 3b). Adding sampling dates, even at the loss of within-date replication, subsumes some of the temporal variation that otherwise confounds the lake-to-lake comparisons of the overall mean and improves estimates of differences between lake means, as shown by the smaller confidence interval with multiple dates (Fig. 3b). While adding dates improves the estimate of the overall lake mean and difference between lakes, identifying of the drivers of short-term temporal variation within lakes would likely require much more than the 3–5 dates we included (see Harris et al., 2007).

In keeping with our results, the data produced by studies in which samples were collected on multiple dates generally showed substantial and statistically significant temporal variation in Hg and MeHg concentrations in zooplankton (Garcia et al., 2007; Gorski et al., 1999; Harris et al., 2007; Marrugo-Negrete et al., 2008; Monson and Brezonik, 1998; Paterson and Rudd, 1998; Slotton et al., 1995; St Louis et al., 2004). Seasonal sampling effort in the studies that we reviewed varied from 1–17 sampling dates per lake per year. Although the number of sample dates was often clearly related to the goals of each study, 55% of the studies relied on a single sample date per lake per year. Such snapshot sampling

effort can be informative in large-scale surveys of tens to hundreds of lakes (Chen et al., 2005) but we conclude that, at present, zooplankton are generally being sampled too infrequently to enable strong cross-lake and cross-study comparisons of trace element bioaccumulation.

We also examined the advisability of fractionating samples by size in order to assess element levels in zooplankton across lakes. Our present study demonstrated that different size fractions of zooplankton had consistently different concentrations of MeHg and Pb. Even for the other elements the size fractions were often different for a given lake and date, but the differences were less consistent across lakes and dates (i.e., the date \times size interaction within lakes accounted for the largest proportion of the variation). Similarly, other studies that tested for differences in MeHg concentrations across zooplankton size fractions found significant differences (Back et al., 2003; Cleckner et al., 2003; Kainz et al., 2002; Masson and Tremblay, 2003; Paterson and Rudd, 1998; Plourde et al., 1997; Tremblay et al., 1998). However, our review of the literature revealed that most studies (74%) do not separate zooplankton size fractions. Moreover, the minimum mesh size of the plankton sampling devices used varies widely among the papers reviewed from 20–500 μm . Hence, we conclude that comparison of specific zooplankton trace element concentrations across studies are likely to be quite imprecise because studies include different components of the planktonic food web. In future studies, standardizing the size fractions sampled to match those in relevant completed studies would allow rigorous comparisons of trace element concentrations with earlier work.

Taken together, these comparisons highlight the importance of studies, such as this one, that sample trace element concentrations in zooplankton across multiple lakes, size fractions and dates. Explicitly accounting for seasonal variation and matching the sampling technique of completed studies when undertaking new studies will enhance the utility of lake-specific estimates of zooplankton trace element concentrations and allow for comparisons across studies. This approach will greatly improve the quantitative basis of simple bioaccumulation-based risk assessment models of trace element accumulation in lake food webs.

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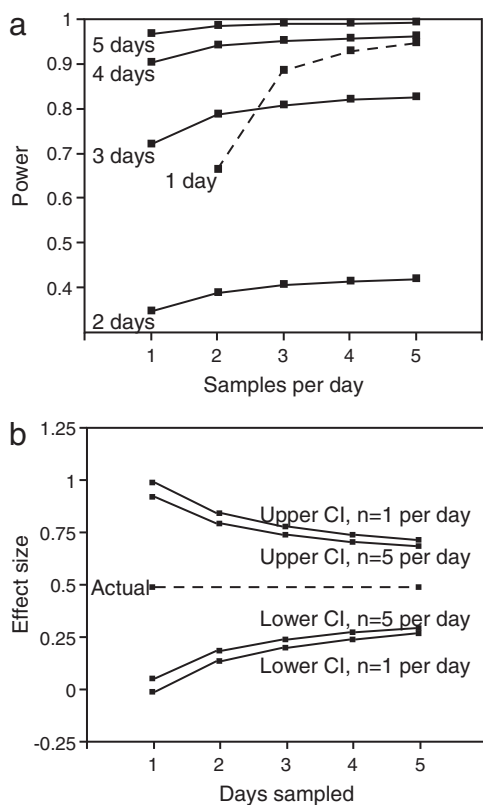


Fig. 3. (a) Power plot of the power to detect a difference in zooplankton trace element concentrations between two lakes at $P < 0.05$ for study designs sampling different numbers of sample days and samples per day. The dashed line indicates apparent power for a single sample day where the estimated difference between the two lakes is entirely confounded with temporal variation. (b) Confidence intervals (CI, 95%) for the estimate of the difference in mean zooplankton trace element concentrations over the growing season for two lakes based on sampling designs with different numbers of sample days and samples per day. Data for both plots are from 10,000 randomized simulations conducted assuming temporal and sample to sample variation as observed for Hg (see Table 1) with the analysis conducted on \log_{10} -transformed data. The actual difference in zooplankton trace element concentrations between lakes in all simulations was held at 0.5 \log_{10} units (c. 3-fold difference in means).

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