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Patterns of Hg Bioaccumulation and Transfer in Aquatic Food Webs Across Multi-lake Studies in the Northeast US

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Abstract. The northeastern USA receives some of the highest levels of atmospheric mercury deposition of any region in North America. Moreover, fish from many lakes in this region carry Hg burdens that present health risks to both human and wildlife consumers. The overarching goal of this study was to identify the attributes of lakes in this region that are most likely associated with high Hg burdens in fish. To accomplish this, we compared data collected in four separate multi-lake studies. Correlations among Hg in fish (4 studies) or in zooplankton and fish (2 studies) and numerous chemical, physical, land use, and ecological variables were compared across more than 150 lakes. The analysis produced three general findings. First, the most important predictors of Hg burdens in fish were similar among datasets. As found in past studies, key chemical covariates (e.g., pH, acid neutralizing capacity, and SO_4) were negatively correlated with Hg bioaccumulation in the biota. However, negative correlations with several parameters that have not been previously identified (e.g., human land use variables and zooplankton density) were also found to be equally important predictors. Second, certain predictors were unique to individual datasets and differences in lake population characteristics, sampling protocols, and fish species in each study likely explained some of the contrasting results that we found in the analyses. Third, lakes with high rates of Hg bioaccumulation and trophic transfer have low pH and low productivity with relatively undisturbed watersheds suggesting that atmospheric deposition of Hg is the dominant or sole source of input. This study highlights several fundamental complexities when comparing datasets over different environmental conditions but also underscores the utility of such comparisons for revealing key drivers of Hg trophic transfer among different types of lakes.

Keywords: mercury; bioaccumulation; aquatic food web; plankton; fish

Introduction

Environmental Hg levels have increased substantially during the industrial age and are now present at levels that adversely affect humans and wildlife. Transcontinental and global transport of Hg, to even the most remote regions, results in significant Hg contamination. Current atmospheric models of Hg transport show that the highest North American deposition rates occur in the northeastern US as well as the lower Great Lakes, Ohio Valley and scattered areas in the south (NESCAUM, 2003). As a result, Hg levels in fish from a significant

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proportion of the lakes in the Northeast region exceed levels of concern for piscivorous mammals (Whittier et al., 2002, Evers et al., 2003, Kamman et al., 2005). Accordingly, there is intense interest in investigating the impact of Hg on biota in lakes and watersheds of this region.

One approach often undertaken to identify regional patterns in Hg and other environmental stressors has been to conduct multi-lake surveys. These studies have revealed important relationships between Hg bioaccumulation, lake characteristics, local deposition patterns, and watershed attributes (Watras et al., 1995; Watras et al., 1998; Rose et al., 1999; Kamman et al., 2005). Earlier studies focused on physico-chemical variables and fish in largely forested lake systems (Hakanson et al., 1988; Suns and Hitchin, 1990; Simonin et al., 1994). Later studies included food web characteristics and measurements of Hg in lower trophic levels (Cabana et al., 1994; Back and Watras, 1995; Stemberger and Chen, 1998; Chen et al., 2000). In studies of lakes across a wide range of nutrient conditions, variation in plankton abundance affects Hg bioaccumulation in plankton and fish (e.g., bloom and density dilution; Pickhardt et al., 2002; Kamman et al., 2004; Chen and Folt, in press). Land use characteristics have also been included in more recent regional surveys and prove to be useful in identifying lake types with higher levels of Hg bioaccumulation.

The extent to which different regional studies can be compared directly, or their inferences generalized across the entire region is often limited. This arises because studies are usually conducted for a particular purpose, or agency. As such, they commonly involve different sets of indicators, scales and regions of interest, lake types and numbers, and target species, selected to meet a specific set of criteria. Regional Hg studies have spanned different ranges and types of lakes and fish sampled. One of the greatest inconsistencies among studies has been the selection of fish species, age classes, and/or body tissues (filet or whole body) for Hg analysis (Wong et al., 1993; Driscoll et al., 1995; Kamman et al., 2005). Hence, factors that best explain variation in Hg in fish and plankton have differed by study system.

Despite differences between regional studies, there is still great value in systematically comparing datasets to identify lake types in which Hg bioaccumulation in fish is of greatest concern for the management of lakes and their resources. Therefore, the goal of our study was to identify common attributes of lakes in the northeastern region that are most likely to predict high Hg burdens in fish by concurrently analyzing 4 northeastern lake datasets for which physical, chemical, land use, and ecological variables were available. Within each dataset, we correlated Hg in fish (all 4 studies) or in zooplankton and fish (2 of the 4 studies) with chemical, physical, land use, and ecological variables that had been related to Hg bioaccumulation in past studies. Since physical and chemical variables have been the most commonly identified in the past, our goal was to further characterize the lake types by identifying the land use and ecological attributes predictive of increased Hg bioaccumulation. In comparing the four datasets, we asked three general questions:

(1) Which variables correlate with Hg in fish or zooplankton in all data sets?
(2) Which variables are unique to particular data sets, and why?
(3) Are there particular lake types (e.g., oligotrophic, urbanized, etc.) that appear most at risk from Hg bioaccumulation?

Methods

This study evaluates datasets from 4 field surveys all derived in part from the USEPA EMAP (Environmental Monitoring and Assessment Program) regional programs (described in Larsen et al., 1991; Allen et al., 1999). The EMAP survey includes a large number variables reflecting broad ecological conditions, landscape and watershed land use features, and chemical and physical characteristics including heavy metal and organic toxin concentrations in fish tissues. Lakes in the EMAP survey were selected based on their surface area using a stratified probability design. This sampling design ensures that lake types are sampled in proportion to their presence in the region (Larsen et al., 1991; Allen et al., 1999). The datasets in this study included: the NIEHS (National Institute of Environmental Health Sciences) survey (Chen et al., 2000); the USEPA (Regional

Environmental Monitoring and Assessment Program)-sponsored VT/NH REMAP survey (Kamman et al., 2003); the USEPA Environmental Monitoring and Assessment Program (EMAP-SW) survey (Whittier, 2002); and the USEPA-sponsored ME-REMAP survey (DiFranco et al., 1995).

Description of datasets

The four datasets varied in the spatial extent of their geographic coverage, in the years that the surveys took place and in the ranges of physical, chemical, and land use characteristics and biological communities sampled (Figure 1, Table 1). The NIEHS dataset was collected and analyzed by investigators at Dartmouth in 1995–1996 and contained 20 lakes in ME, NH, VT, CN, and NY. The lakes were partially selected from the EMAP-SW 1992–1993 dataset with the addition of several other lakes of particular interest. Lakes were selected that had a modal chain length of 5 (i.e., 5 predator–prey links) but differed in other key structural features of the pelagic web (see Stemberger and Chen, 1998). The NIEHS dataset contained the greatest number of measurements for Hg concentrations in mixed fish species (data from EMAP-SW 1992–1993, see method below) and Hg in large (>202 μm) and small zooplankton (45–202 μm) net fractions measured in each lake.

The REMAP-VT/NH dataset was collected and analyzed in 1998–2000 by a team of investigators led by the Vermont Department of Environmental Conservation, and contained 103 lakes in NH and VT. Hg was measured in yellow perch (<10 cm whole body, >10 cm fillets), >201 μm zooplankton, water, and sediment (Kamman et al., 2003, 2004). Thirty-six lakes were complete for all biological variables and hence, were included in this analysis. The 105 EMAP lakes analyzed in this study were sampled during the summers of 1992–1993 for Hg in fish (mixed species), zooplankton abundances, and for water chemistry, fish species and watershed land use variables (Stemberger and Chen, 1998; Whittier et al., 2002). Generally, 3–5 whole fish of a single species were homogenized for a composite tissue sample (Yeardley et al., 1998). Top trophic species (largemouth bass, smallmouth bass, rainbow trout, yellow perch, pumpkinseed sunfish, etc.) were preferred for analysis, but sizes and species varied in the composite. The REMAP-ME dataset contained 125 lakes that were

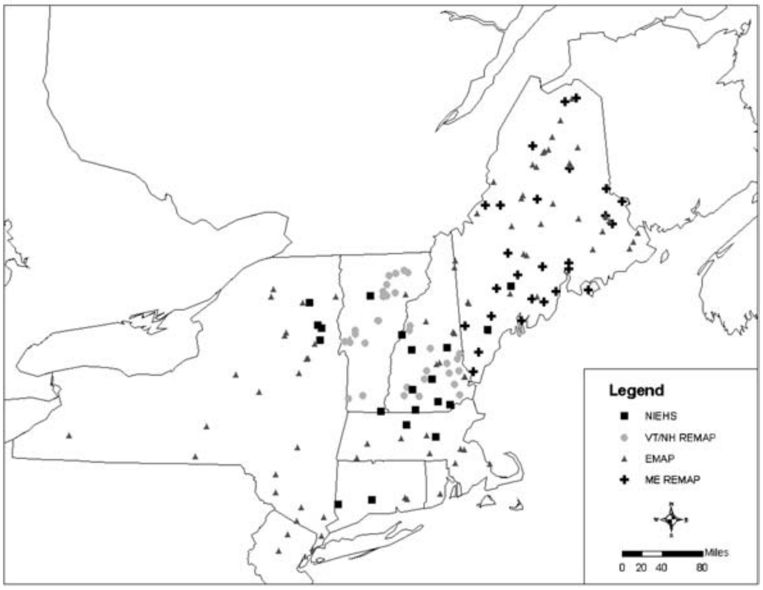

Figure 1. Geographic distribution of lakes in each dataset. 5 lakes in the NIEHS and EMAP datasets are the same and shown as NIEHS lakes. XX lakes in the ME-REMAP and EMAP lakes are the same and are shown as ME REMAP lakes.

Table 1. Ranges of selected characteristics for the lakes sampled in each sampling program. Yellow perch (YP), zooplankton Hg concentration (zoop)

Dataset	# lakes	Hg in BIOTA	Basin Area (ha)	Lake Area (ha)	Total P (µg/l)	pH	Cond.	DOC mg C/L	% Urban	Chl a µg/l	Fish Hg µg/g WW
NIEHS NE US	20	Mix fish sp. zoop	1–799	3–902	3.0–22	6.0–7.9	27–615	2.9–12.6	0–80	0.7–13. 7	0.017–1.372
REMAP VT-NH	36	YP zoop	44–110,592	10–669	3–27	5.2–8.0	11.2–400	1.9–6.5	0–11. 6*	NA	0–0.7
EMAP NE US	105	Mix Fish sp.	0.1–2645.8	6–3305.8	0–176	4.4–9.1	NA	2.0–18.2	0–94.05	0. 3–191.9	0.009–1.949
ME **REMAP**	26	Mix Fish sp.	1–1974	17–5834	5–28	6.3–7.3	25–75	1–18.4	NA	1.3–10.7	0.02- 2.50

*% commercial/industrial and transportation.

collected by the Maine Department of Environmental Protection in 1993 and 1994. However, only 26 of these lakes were sampled for zooplankton and were included in our analysis. Fish species were targeted for collection based on trophic level and categorized as a predator and an omnivore (DiFranco et al., 1995). In an effort to obtain specimens from different lakes that were of comparable age, target fish sizes (lengths) were chosen based on available length/age relationships, legal length limit, "desirability" as game species, and likelihood of capture. In all cases, larger fish near the top of the target size range were preferred. One to five fish of a single species were combined for analysis and analyzed as whole fish (omnivores and predators) or filets (predators only). From each of the four datasets, a group of chemical, physical, land use, and ecological variables were selected for analysis based upon their potential relationships to Hg bioaccumulation as suggested in past studies.

Contrasting ranges of datasets

Although all the lakes were in the northeast region, each dataset differed in its range of lake attributes (Table 1). The NIEHS dataset contained the most ecological variables including Hg concentrations in all trophic levels. The VT/NH REMAP dataset contained the smallest range of lakes sizes with the largest range of watershed areas and the narrowest dissolved organic carbon (DOC) concentrations across lakes. The NIEHS and VT/NH REMAP surveys were the only datasets containing aqueous and zooplankton Hg concentrations. The Hg concentrations in fish were much lower in the VT/NH REMAP dataset than in the other three (Table 1). This may have been due to the overall lower trophic level of the yellow perch resulting in lower Hg concentrations than for the composite samples from the other datasets. Zooplankton Hg concentrations were also much lower in the VT/NH-REMAP dataset than in the NIEHS dataset: NIEHS (> 202 µm zooplankton: 0.028–7.477 µg/g) and VT/NH-REMAP (> 201 µm zooplankton: 0.016–0.335 µg/g). In general, the EMAP dataset was the largest and most representative of the region in regard to number of lakes, geographic extent of the study area, and ranges and quantity of lake and watershed variables. In particular, ranges of values in the EMAP dataset that reflect human disturbance (nutrients, chlorophyll, urbanized shoreline) were much broader than in the other three datasets. The ME-REMAP lakes spanned the narrowest pH range (6.3–7.3) and had a very low range in conductivity (25–75 µS/cm) reflective of the generally low concentrations of cations and pristine quality of many lakes in the state of Maine. This dataset did not include land use variables except for population density (Table 2).

Statistical analyses

We chose non-parametric correlations as the most direct and parsimonious analysis for comparing lake attributes across the four datasets. Spearman rank correlations were calculated using non-transformed data in each of the four datasets to identify the variables that were significantly correlated with Hg in fish and in zooplankton (JMP, SAS, 1995). A separate analysis was performed for each of the Hg variables (fish and zooplankton concentrations) to test its relationship to chemical, physical, land use, and ecological variables available within each dataset. In addition, 16 lakes in the EMAP dataset from which yellow perch were collected, were analyzed separately in order to compare results to the EMAP dataset as a whole and to the results of the VT/NH-REMAP analyses which contained only yellow perch. A Bonferroni correction was applied to account for the number of tests conducted for each analysis thus determining the most conservative assessment of significance (Miller, 1981). The four datasets were not combined and run in a single analysis because variables measured, the methods

Table 2. Variables available for analysis in each sampling program dataset. (Abbreviations: WaLa: watershed area:lake area; ANC: acid neutralizing capacity; DOC: dissolved organic carbon; YP: yellow perch)

Variable Category	Variables	NIEHS	REMAP VT-NH	EMAP	ME-REMAP
Physical	Lake area	X	X	X	X
	Watershed area	X	X	X (WaLa)	
	Depth (max.)	X	X	X	X
	Elevation			X	X
Chemical	pH	X	X	X	X
	Alkalinity		X	X (ANC)	X
	DOC	X	X	X	X
	SO4		X	X	
	Conductivity	X	X		X
	Total P	X	X	X	
	Total N	X		X	
	N:P	X			
Landuse	Population		X (911 count[1])		X
	Disturbed	X		X	
	Residential		X		
	Hay/Perm. Past,		X	X (agric.)	
	Comm/Ind/Transp.		X	X (urban)	
	Forest		X	X	
	Wetland	X		X	
	Road density	X		X	
Ecological	Linkages	X	X		
	Max. chain length	X	X		
	Chorophyll	X		X	X
	Algal biomass	X			
	Zoop. abundance	X	X	X	X
	Zoop. Biomass	X			
	Richness	X	X		X
Hg conc.	Aqueous	X	X		
	Zooplankton	X	X		
	Fish	X (mixed)	X (YP)	X (mixed)	X (mixed)

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[1] See Kamman et al., 2003.

used for sample analysis, and numbers of lakes in each dataset varied greatly.

We investigated differences between the predictors of Hg bioaccumulation in the NIEHS and VT/NH-REMAP datasets, using path analysis because these were the only two datasets for which Hg data was collected for water, zooplankton, and fish. These analyses were conducted to determine the strength of linkages between Hg concentrations in water and zooplankton compartments and Hg concentrations in fish. Within each dataset, linear regression was done for each variable in the model as dependent on the other variables that the model indicated as causal. The coefficients depicting the strength of the relationships between variables were calculated as standardized regression weights (Beta coefficients, JMP, SAS, 1995). All plausible connections between food web compartments based on past research were assembled in a path diagram for each dataset with Beta coefficients labeled to indicate the relative importance of each pathway.

Results

Significant correlates of Hg bioaccumulation

The Spearman Rank correlation analyses identified relationships between Hg concentrations in fish and chemical, physical, land use and ecological variables. We identified suites of significant variables ($p < 0.05$) some of which were common across the datasets and which contained several variables from each variable category, i.e., chemical, physical, land use, and ecological (Table 3). For example, the NIEHS, EMAP, and ME-REMAP datasets had suites of significantly correlated variables from all 4 categories whereas the VT/NH-REMAP only had chemical and land use variables. The Bonferroni correction reduced the significant variables to only one ecological variable (zooplankton density) in the NIEHS dataset, and 3 chemical (acid neutralizing capacity or ANC, pH, conductivity) and 2 land use variables (commercial/industrial/transportation and residential) in the VT/NH-REMAP dataset. The EMAP dataset included highly significant variables from all 4 categories: chemical (ANC, pH, SO_4), physical (lake area, watershed:lake area), land use (% wetland), and ecological (zooplankton density). Moreover, variables identified in the analysis of the 16 yellow perch lakes were similar to those identified by the full analysis (ANC, SO4, pH, road density, zooplankton density). In contrast, only human population density in the watershed was highly significant in the ME REMAP dataset.

Common variables

The correlation analyses identified a set of common variables across datasets that were consistent in terms of the direction of their relationship to Hg in fish (Table 4). Specifically, two physical variables (lake area and watershed:lake area) were positively related to Hg concentrations, and five chemical variables (pH, ANC, SO4, conductivity, nutrients), four human land use variables (e.g., residential, agricultural, commercial/industrial, disturbed), and one ecological variable (zooplankton density) were all negatively related to Hg concentrations in fish.

Unique variables

Each dataset had significant variables that were unique to that group of lakes (Table 5). In some cases, the unique variables were included and analyzed in more than one dataset, but only found to be significant in one. For example, Hg levels in zooplankton were measured in both the NIEHS and VT/NH-REMAP studies, however, Hg in >202 μm zooplankton fraction was positively correlated to fish Hg levels in only the NIEHS dataset. Similarly, DOC was measured in all the datasets, but was only positively correlated with fish Hg in the VT/NH-REMAP dataset. While aqueous total Hg was measured in both the NIEHS and VT/NH-REMAP datasets, it was positively correlated with fish Hg in only the latter study. Forested shorelines with greater extent of wetlands were significantly related to higher Hg concentrations in fish only in the EMAP dataset even though similar variables were also measured in the other datasets. In the ME-REMAP dataset, human population density was positively correlated with Hg in fish and elevation was negatively correlated.

Zooplankton Hg correlates

The variables found to be significantly correlated with zooplankton Hg concentrations differed

Table 3. Significant variables identified in each program dataset by Spearman Rank correlations for Hg concentrations in fish (all variables significant at the $p < 0.05$ level). Variables in bold indicate significance at $p < .05$/number of variables tested for each dataset using a Bonferroni correction. (-) indicates negative correlations with all others positive. Asterisk in EMAP dataset indicates significant variables when tested for yellow perch only (16 of the 105 lakes). Abbreviations: Epi: epilimnetic; THg: total Hg in water; Resid: residential; Perm. Past.: permanent pasture; WHS: white sucker; BB: brown bullhead; also see Table 2.

Program	Fish Hg	Physical/Chemical Variables	Landuse/Ecological Variables
NIEHS	Mixed species	Conductivity (-)	% shoreline dist. (-)
		Lake area	**45–202** μm **dens. (-)**
		N:P ratio	**Total large zoopl. (-)**
			> 202 μm Hg conc.
REMAP VT-NH	Prey YP	**Mean conductivity (-)**	Resid. Landuse (-)
		Epi ANC (-)	Hay/perm. past. (-)
		Epi SO4 (-)	**Comm/Ind/Transp. (-)**
		Epi DOC	
		Epi THg	
		Mean pH (-)	
	YP adult fillet	**Mean conductivity (-)**	**Resid. Landuse (-)**
		Epi ANC (-)	Hay/perm. past. (-)
		Epi SO4 (-)	**Comm/Ind/Transp. (-)**
		Epi DOC	
		Epi THg	
		pH(-)	
EMAP	Mixed species	**Lake area**	Road density* (-)
		Wa:La	% Forested
		Total N (-)	**\% Wetland**
		Total P (-)	**Total zoop dens.* (-)**
		ANC* (-)	
		SO4* (-)	
		pH* (-)	
ME REMAP	Omnivore whole (WHS, BB)		Zooplankton/L (-)
	Piscivore whole (mixed)	Alkalinity (-)	Population density
		Elevation (-)	
	Piscivore fillet	PH (-)	**Population density**
		Alkalinity (-)	
		Lake area (-)	
		Elevation (-)	

Table 4. Common variables correlated with Hg concentrations in fish across lakes in more than one dataset (number of datasets in which variable is correlated/number of datasets tested)

Variable category	Common variables	Relationship to Hg in fish
Physical	Lake area (3/4)	Positive
	Watershed area (2/3)	
Chemical	pH (3/4)	Negative
	ANC (alkalinity) (3/3)	
	Conductivity (2/3)	
	SO_4 (2/2)	
	Nutrients (2/3)	
Land use	Residential, agricultural, commercial/ind./transp. road density, disturbed (3/3)	Negative
Ecological	Zooplankton abundance (3/4)	Negative

between size fractions and between datasets (Table 6). In the NIEHS dataset, high chlorophyll concentrations were negatively correlated with Hg concentrations in small zooplankton (45–202 μm) and Hg concentrations in large zooplankton were positively correlated with Hg in small zooplankton

Table 5. Significant ($p < 0.05$) variables correlated to Hg in biota unique to each dataset

Program	Hg in Biota	Variables
NIEHS	Fish	> 202 μm zooplankton Hg (+)
	Zooplankton (45–202 μm, > 202 μm)	Chlorophyll *a* (–), Zooplankton density (–)
REMAP VT-NH	Fish	Epi DOC (+)
	Zooplankton (> 201 m)	Epi THg (+) Lake depth (+)
EMAP	Fish	Watershed area (+)
		Forest (+)
		Wetland (+)
ME-REMAP	Fish	Elevation (–)
		Population density (–)

Table 6. Spearman rank correlations with zooplankton Hg concentrations

Program	Zooplankton Hg	Significant Variables	Rho	*p*-value
NIEHS	45–202 μm	Ave. chlorophyll	–0.5907	0.0125
	> 202 μm	45–202 μm Hg	0.5789	0.0075
		Total large zoopl.	–0.5766	0.0062
REMAP VT-NH	> 201 μm	Depth max.	0.4435	0.0067
		Mean depth	0.4378	0.0076

and negatively correlated with density of large zooplankton. In contrast, Hg concentrations in large zooplankton were positively correlated with lake depth but no other plankton variables in the VT-REMAP dataset.

Pathways for Hg bioaccumulation

A closer examination of the NIEHS and VT/NH-REMAP datasets was made using path analysis to describe the relative strengths of causal links between environmental variables and Hg in water, zooplankton, and fish (Figure 2). The path analyses suggested different potential pathways underlying the transfer of Hg to fish in each dataset. In the NIEHS dataset, for instance, Hg in large zooplankton had strong effects on fish Hg concentrations whereas dissolved aqueous Hg weakly affected zooplankton Hg concentrations and had no effect on fish Hg concentrations (Figure 2a). In contrast, zooplankton Hg concentrations in the VT/NH-REMAP dataset had minimal effect on fish Hg concentrations which were much more strongly affected by epilimnetic whole water Hg concentrations (Figure 2b). Thus, in the VT/NH-REMAP lakes, aqueous Hg concentrations appear to exert primary control over fish Hg concentrations, where phytoplankton biomass and zooplankton Hg

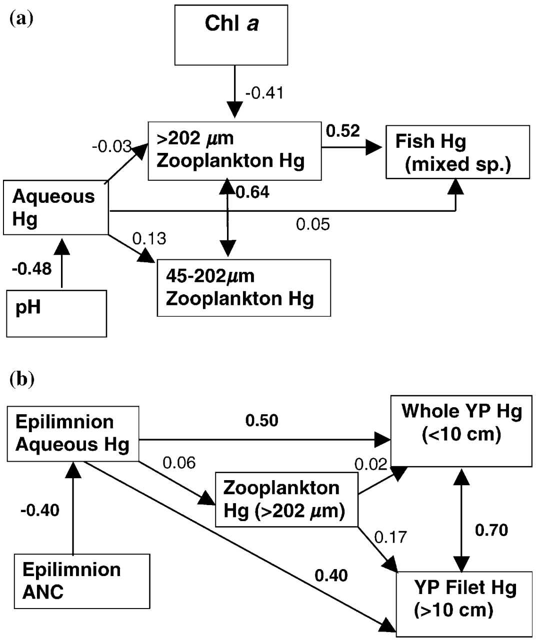

Figure 2. Path diagrams for (a) NIEHS and (b) VT/NH-REMAP datasets. The magnitude and sign of the Beta coefficients represent the relative contribution of the independent variable in the prediction of the dependent variable located at the tip of the arrows. These models assume that a 0.5 standard deviation change in the independent variable imparts a 0.5 standard deviation response in the dependent variable. Double headed arrows indicate that the relationship is correlational and not causal.

concentrations appear more influential in the NIEHS lakes.

Discussion

Comparative analysis of data from four northeast US lake surveys provided a useful method for identifying the key variables that potentially explain high rates of Hg bioaccumulation in plankton and fish. Although other studies have identified some of these variables by including a wide range of lake types we identified several potentially important factors that have not been previously recognized, particularly land use and ecological variables. In total, these variable suites describe 'lake types' most at risk to Hg bioaccumulation and biomagnification.

Four chemical variables, all covariates, (pH, ANC, DOC and SO_4) emerged as common critical predictors of Hg bioaccumulation in fish in all four data sets. This finding agrees with correlates identified in earlier multi-lake studies that are focused on more poorly buffered lakes in forested watersheds (Back and Watras, 1995; Driscoll et al., 1995; Watras et al., 1998). However, by extending the lake types surveyed, we discovered several additional common correlates with Hg bioaccumulation. These correlates fall into 2 categories, descriptors of adjacent land use (% disturbed, residential, agricultural, and commercial/industrial/transportation) and lake biota (plankton density). Although these descriptors have not been widely measured or related to Hg bioaccumulation in prior studies, our results show that they were as strong as chemical variables in identifying lakes with high Hg bioaccumulation.

In general, both watershed disturbance and plankton abundance were negatively correlated with fish and zooplankton Hg concentrations. With increasing human land use or disturbance in the watershed there is reduced area remaining in forest and wetland. This may result in a net decrease in mobilization of Hg from land to water because adjacent forest and wetlands are known to enhance Hg transport from watersheds to lakes (Driscoll et al., 1995; Driscoll et al., 1998; Hintlemann et al., 2002; Kramar et al., 2005). Lakes in disturbed watersheds also often receive greater nutrient and organic inputs. This results in greater lake productivity, which can be negatively correlated with Hg bioaccumulation for many reasons. For instance, Hg in plankton and fish has been shown to decline with increases in chlorophyll and zooplankton abundance as shown in Tables 5 and 6 in this study and by others (Pickhardt et al., 2002; Kamman et al., 2004). This relationship is hypothesized to arise when higher growth rates and abundances of biota result in lower mass specific Hg concentrations in plankton (i.e., algal bloom dilution – Pickhardt et al., 2002, 2004) and in fish (i.e., growth dilution – Essington et al., 2003; Kamman et al., 2004). Productive lakes also commonly have high DOC concentrations that are derived in part from the degradation of autochthonous algal material, which reduces Hg bioavailability to the biota (Watras et al., 1995; Kamman et al., 2004). Moreover, higher productivity is usually associated with higher levels of organic matter in the sediments, which binds with inorganic Hg, thus reducing the amount available for methylation (Hammerschmidt and Fitzgerald, 2004). Finally, eutrophic sediments have high sulfate reduction rates that produce sulfide which can inhibit MeHg production (Choi et al., 1994; Gilmour et al., 1998), and can serve to sequester available MeHg (DiPasquale et al., 2004).

Several other chemical (DOC, aqueous total Hg), physical (lake depth, watershed area), land cover (forest, wetland) and ecological (chlorophyll, small and large zooplankton Hg concentrations) variables emerged as unique predictors of Hg bioaccumulation in fish or plankton in individual datasets. Differences in survey design (e.g., scale/region of study, selection of dominant lake types, variables measured, sampling protocols, fish species) are very likely to have produced some of the differences in significant correlates among lake surveys that we observed. The greater number of ecological correlates (chlorophyll, small and large zooplankton Hg concentrations) for Hg bioaccumulation in zooplankton and fish in the NIEHS dataset establishes an important linkage between two lower trophic levels and Hg bioaccumulation in fish. The analysis demonstrates the importance of plankton variables to Hg bioaccumulation and suggests that they should be included more frequently in local and regional surveys. In contrast to the NIEHS dataset, the relationship of lake depth to zooplankton Hg concentrations in the

VT-REMAP dataset is potentially due to the lower abundances and larger body sizes of zooplankton in deeper lakes which tend to carry higher Hg burdens. Finally, percent of shoreline in wetlands emerged as a significant correlate with Hg bioaccumulation in the EMAP dataset that also covered the greatest range of values (0–37% versus 0–4.8% in the NIEHS dataset). Wetlands are known areas of high organic matter and extremely high rates of Hg methylation which is the main source of Hg for bioaccumulation (St. Louis et al., 1994; Heyes et al., 1998; Driscoll et al., 1998).

Differences in the significant variables that emerge across datasets may also arise from disparities among sampling methods for Hg in abiotic and biotic compartments. For instance, fish Hg levels were significantly related to Hg in large zooplankton in the NIEHS dataset but not in the VT/NH-REMAP dataset. The path analyses of the respective food webs illustrate the contrast: while Hg concentrations in fish in the NIEHS were most strongly influenced by zooplankton Hg, they were affected most directly by aqueous Hg concentrations in VT/NH-REMAP dataset. In some respects, the significant relationship between water and fish Hg concentrations is surprising given that there is no direct food web link between the two. MeHg is the form of Hg that is bioaccumulated via food consumption in the web and is the predominant species in fish (95%) but only comprises $\sim$10% of total Hg in water (Watras and Bloom, 1992; Mason et al., 1995; Hall et al., 1997; Watras et al., 1998). Nonetheless, there are 2 possible explanations for the differences between the findings in the two datasets. First, aqueous samples in the NIEHS and VT/NH-REMAP studies were not entirely equivalent. In the NIEHS survey, Hg was measured in the dissolved fraction (0.4 μm filtered) whereas unfiltered whole water samples were collected and analyzed in the VT/NH-REMAP program. Given that Hg is bioconcentrated in particulate matter, Hg in whole water samples could be greatly affected by the presence of suspended material. This bias could be most pronounced in high productivity lakes, because past studies have demonstrated declines in dissolved aqueous metal concentrations with the occurrence of algal blooms (Luoma et al., 1998; Chen and Folt, 2000). Second, some of the variation between these two surveys may be due to the temporal disjunction of fish Hg data and zooplankton and water chemistry data. In the NIEHS dataset, fish data were collected in 1992 and other variables including zooplankton and aqueous Hg in 1995–1996. In the VT/NH-REMAP, the zooplankton data were collected 1 year after the data for fish Hg and aqueous Hg concentrations.

An even greater inconsistency in methodology among the datasets in this study lies in the ages, sizes, and species of fish collected. Selection of the fish species, sizes, and tissues is crucial to understanding the pathways by which Hg is transferred from prey to consumer and yet, these variables differ across most multi-lake studies (Kamman et al., 2005). For example, the NIEHS, EMAP, and ME-REMAP programs sampled predominantly top trophic level, piscivorous species and combined them in single composites. In all 3 datasets, Hg concentrations in fish decreased with increasing zooplankton densities. Interestingly, this was also true for the subset of 16 EMAP lakes in which yellow perch alone were sampled. These results suggests that high zooplankton abundances either dilute metal concentrations or are related to increased fish growth rates and metal growth dilution (Chen and Folt, in press). This may be due to both increases in zooplankton biomass and smaller bodied zooplankton assemblages which may carry lower Hg burdens probably because of higher mass-specific respiration rates (Kainz et al., 2002). In contrast, Hg concentrations in the VT-REMAP fish were markedly lower than in the other datasets and there was no relationship observed between zooplankton variables and fish Hg as observed in the other datasets. This contrast may be due to differences in the species and age of fish sampled among programs; only yellow perch in two specific size classes (<10 cm and >10 cm) were collected in the VT/NH-REMAP program. It is well known that yellow perch feeding is highly habitat- and age-specific. Based on findings in earlier studies, the <10 cm yellow perch collected in the VT/NH-REMAP program were likely feeding on plankton and littoral invertebrates whereas the >10 cm fish were consuming other fish and invertebrates (Prout et al., 1990; Schael et al., 1991; Wu and Culver, 1992; Persson et al., 2000; Liao et al., 2002). The selection of specific age classes of yellow perch in the VT-REMAP sampling program greatly differed from the mixed species samples of the other datasets and may also have differed from yellow perch in the EMAP dataset in terms of fish age. In general, we suggest that mixed species composites of fish such as those in the NIEHS, EMAP-SW, and ME-REMAP datasets may better integrate the upper trophic level Hg signal in lakes than a single fish taxa feeding in specific habitats within the lake as seen in the VT/NH-REMAP dataset.

Our study clearly illustrates that the sampling design (e.g., range of variables in the dataset, taxonomy and age of focal species, etc.) strongly influences the interpretation of certain key predictors of Hg bioaccumulation. Some of the observed differences between common and unique predictors undoubtedly arose from the choices of variables measured in the various surveys. However, by comparing results of studies conducted over different spatial and temporal scales or for different target species, inconsistencies in the results could also be compared and underlying mechanisms could be elucidated and hypothesized (Folt et al., 1998). Taken together, the common and unique predictors can be used to derive a more thorough investigation of environmental drivers of Hg bioaccumulation across a variety of ecosystems.

In summary, we identified a suite of variables that emerge as strong predictors or indicators of Hg bioaccumulation. Some of these indicators like pH, ANC, lake area, and zooplankton abundance are already measured in many lake management programs and can be useful in identifying lakes likely to contain fish with high Hg concentrations. Other factors such as watershed vegetation cover and land use are often not quantified but can be important drivers of biotic Hg concentrations via biogeochemical processes that affect terrestrial and atmospheric Hg loading into aquatic ecosystems. Identifying such key suites of indicator variables strengthens our fundamental understanding of processes underlying Hg bioaccumulation and increases our capacity to monitor and predict patterns of Hg in biota across a variety of lake ecosystems.

Combining the results of the 4 multi-lake studies, we find that lake types associated with the greatest amount of Hg bioaccumulation are poorly buffered, low pH, low productivity lakes having forested watersheds and minimal human land use. These are general characteristics of the most remote lake ecosystems for which atmospheric transport and deposition have the greatest relative impact as sources for Hg (Miller et al., 2005). They are also lakes of high value for recreation and other human uses, and worthy of increased effort to protect and conserve.

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