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INFLUENCE OF LAKE CHARACTERISTICS ON THE BIOMAGNIFICATION OF PERSISTENT ORGANIC POLLUTANTS IN LAKE TROUT FOOD WEBS

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Abstract—The biomagnification of polychlorinated biphenyls (PCBs) and major organochlorine pesticides (OCPs) was studied using lake trout (*Salvelinus namaycush*) and other food web organisms collected from 17 lakes in Canada and the northeastern United States between 1998 and 2001. Whole lake trout (n = 357) concentrations of the sum (Σ) of 57 PCB congeners ranged between 1.67 and 2,890 ng/g wet weight (median 61.5 ng/g wet wt). Slimy sculpin had the highest mean concentrations of ΣPCB of all forage fish (32–73 ng/g wet wt). Positive relationships between log (lipid wt) concentrations of PCB congener 153, PCB congener 52, *p,p′*-dichlorodiphenyldichloroethylene, hexachlorobenzene, *cis*-chlordane, *trans*-nonachlor, or dieldrin and trophic level (determined using stable nitrogen isotope ratios) were found for most of the 17 food webs, indicating biomagnification of these PCBs and OCPs. The *p,p′*-dichlorodiphenyldichloroethylene had the highest trophic magnification factors (TMFs) of the 14 individual compounds studied, averaging 4.0 ± 1.8 across the 17 lakes, followed by *trans*-nonachlor (3.6 ± 1.5) and PCB congener 153 (3.4 ± 1.2). Average TMFs for 14 individual PCBs or OCPs were significantly correlated with log octanol–water partition coefficient, implying that the rate of accumulation along the food web is dependent on hydrophobicity and recalcitrance. Significant correlations ($p < 0.05$) were found between TMFs of ΣPCBs, hexachlorobenzene, α-hexachlorocyclohexane, and lindane and lake area, latitude, and longitude, but not for 11 other PCBs or OCPs. Overall, the results of the present study show that biomagnification of PCBs and most OCPs, as measured by TMFs, is only weakly influenced by such factors as latitude and longitude. Exceptions are hexachlorocyclohexane isomers and hexachlorobenzene, which had generally greater TMFs in northern lakes, possibly due to lower rates of elimination and biotransformation in the food web.

Keywords—Persistent organic pollutants Lake characteristics Lake trout food webs Biomagnification

INTRODUCTION

Lake to lake variability in concentrations of polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) in lake trout and other top predator fish has been widely observed [1,2] and continues to be difficult to fully explain. Food web length and structure, as well as lipid content, have been shown to be important factors influencing the concentrations of PCBs in lake trout (*Salvelinus namaycush*) food webs [1–3]. Higher concentrations of PCBs were observed in lake trout from lakes with the longest underlying food webs and in fattier organisms [1,4], but these factors do not account for all the among-lake differences in these contaminants.

A more recent approach in biomagnification studies has been to use stable isotopes of carbon and nitrogen to understand food web structure and, in turn, advance our understanding of the accumulation of persistent pollutants through aquatic food webs. Stable nitrogen isotope ratios (^{15}N:^{14}N) provide a continuous measure of trophic positioning of the organisms within a food web because of the predictable fractionation (3–4‰) of the lighter to heavier isotope for this element from prey to predator [5]. In contrast, ratios of the heavier-to-lighter carbon isotopes (^{13}C:^{12}C) are used to assess the importance of different energy sources to organisms because there is little (0–1‰) fractionation as this energy is moved up the food web [6]. Several studies have used $\delta^{15}N$ to quantify the trophic positioning of individual species, food web length, and the biomagnification of PCBs and other persistent pollutants in the Great Lakes [7,8] and subarctic lakes [2,3] because of the significant relationship between the concentration of these pollutants and the $\delta^{15}N$ of biota. However, these studies focused on one or a few systems and did not assess trophic transfer of PCBs or OCPs over a broader scale, despite the potential to use these stable isotope and pollutant data to contrast lakes that differ in their biological or chemical characteristics [9].

Most studies of among-lake variability of PCBs or OCPs in fish have emphasized the influence of food chain length and lipids; less attention has been paid to trophic status of lakes. A significant negative relationship between plankton PCB levels and plankton biomass was reported by Taylor et al. [10] in lakes of southern Ontario, Canada. Berglund et al. [11] found that the total PCBs in phytoplankton and microzooplankton were negatively correlated with lake trophic state (as indicated by total P) and that this was explained by lower lipid content of the plankton in eutrophic lakes. They found no

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differences among lakes in PCB concentrations in phytoplankton and microzooplankton when normalized to lipid content.

The objectives of the present study were to determine the biomagnification of PCBs and OCPs in lake trout food webs across a range of lakes that varied in their physical, biological, and chemical characteristics. A companion study examined the geographic trends, and relationships with lake characteristics, of PCBs or OCPs in lake trout from 24 lakes across Canada and the northern United States [12].

MATERIALS AND METHODS

Biotic and abiotic sample collections

Lake trout and food web organisms were collected from 17 lakes across Canada and in the northeastern United States during the period of 1998 to 2001 (Table 1). These lakes ranged in surface area from 11.2 to 82,170 km^2 and were oligotrophic or mesotrophic. Lake trout and forage fish species were collected in each lake using gill nets, seining, minnow traps, and angling. Collections of forage fish varied from lake to lake (1–5 species/lake) because of inherent differences in the fish communities and included longnose dace (*Rhinichthys cataractae*), lake herring (*Coregonus artedii*), alewife (*Alosa pseudoharengus*), rainbow smelt (*Osmerus mordax*), lake whitefish (*Coregonus clupeaformis*), yellow perch (*Perca flavescens*), white (*Catostomus commersoni*) and longnose (*C. catostomus*) sucker, slimy sculpin (*Cottus cognatus*), shiner (emerald, *Notropis atherinoides;* mimic, *N. volucellus;* spottail, *N. hudsonius;* common, *N. cornutus*), ninespine stickleback (*Pungitius pungitius*), and trout-perch (*Percopsis omiscomaycus*; see Table 1 for sample numbers and lakes). Bulk zooplankton tows (>110 μm) and benthic invertebrates were collected in 17 and 13 of these lakes, respectively; the zooplankton predators *Mysis relicta* and *Diporeia hoyi* were also collected when present. Benthic invertebrates were collected in the nearshore areas of the lakes and included snails (Gastropoda), amphipods (Amphipoda), mussels (Unionidae), caddisflies (Trichoptera), damselflies (Zygoptera), and mayflies (Ephemeroptera). Invertebrates were typically collected qualitatively during one field trip to the lakes, and this varied in the time of year from one system to another. Benthos was sorted to major taxa in the field, voucher specimens were preserved for later identification, and the remaining samples and bulk zooplankton were frozen in solvent-rinsed jars or Whirl-Pak® (Nasco, Fort Atkinson, WI, USA) polyethylene bags. Sampling in Lake Superior is described in detail in Muir et al. [13].

Phytoplankton biomass and taxonomic composition were determined for 14 of these lakes by collecting standardized water column samples at the same time as the fish and invertebrate collections. Surface water samples (2 L) were collected from the upper mixed layer, and subsamples were preserved with Lugol's iodine solution for further microscopic analysis. Water was also subsampled for particulate carbon, nitrogen, phosphorus, chlorophyll *a,* and silicon by filtration through glass fiber filters (nominal pore size 0.75 μm). Filters for carbon and nitrogen were ashed. Polycarbonate filters were used for Si. Sample and blank filters were dessicated in the dark and then frozen. The filtrate was analyzed for basic water chemistry parameters (nitrogen, phosphorus, dissolved organic carbon [DOC], silica, conductivity, alkalinity, and pH).

Sample storage and processing

All samples were shipped frozen to Burlington (ON, Canada) and stored at −20°C until processed. Fish length and weight were obtained either by individual collaborators or at the laboratory of Department of Fisheries and Oceans in Burlington. Prior to processing fish for PCBs and OCPs, aging structures were removed (data not shown) and skinless dorsal muscle samples were taken for stable isotope analysis. Subsamples of invertebrates were taken, removed from their cases or shells (if present), and then processed as described later for stable isotope analyses. Whole fish and invertebrate samples were then homogenized by the Burlington laboratory. Homogenates were stored (−20°C) at the Department of Fisheries and Oceans Great Lakes fish tissue archive. All samples, except those from Lake Superior, were sent to the Great Lakes Institute for Environmental Research in Windsor (ON, Canada) for analysis of PCBs and OCPs. Samples from Lake Superior were analyzed by the National Laboratory for Environmental Testing in Burlington [14].

Dorsal muscle and foot tissue from individual fish and mussels, respectively, and pooled snails (shells removed), caddisflies (cases removed), mayflies, amphipods, damselflies, *Mysis*, *Diporeia*, and bulk zooplankton were dried in an oven at 60°C, ground to a fine powder, and then analyzed as described later for stable isotopes of nitrogen and carbon ($\delta^{15}N$ and $\delta^{13}C$).

Laboratory analyses

Polychlorinated biphenyl congeners and major OCP groups—DDT, chlordanes (CHLs), chlorobenzenes, hexachlorocyclohexanes (α-HCH and γ-HCH or lindane), and α-endosulfan were the main groups of persistent organochlorines analyzed in the present study. Method details, including quality assurance steps, are provided in Guildford et al. [12] and Muir et al. [13]. In brief, homogenized samples were mixed with sodium sulfate and Soxhlet or pressurized fluid extracted. Lipids were removed by gel permeation chromatography, and the extracts were fractionated on a Florisil® (U.S. Silica, Berkeley Springs, WV, USA) column to separate PCBs, hexachlorobenzene (HCB), and *p,p′*-dichlorodiphenyldichloroethylene (*p,p′*-DDE) from other OCPs. Fractions were then evaporated to a small volume and transferred quantitatively to vials for analysis. Extracts were analyzed for 57 PCB congeners (includes co-eluters), chlorobenzenes, and three OCP groups by capillary gas chromatography with electron capture detection. Extractable lipid (%) was determined gravimetrically on each sample on a fraction of the tissue extract, and moisture (%) was determined by drying a subsample to constant weight.

Fish muscle and invertebrate samples (1.0–1.5 mg dry wt) were analyzed at the Environment Canada stable isotopes laboratory in Saskatoon, Saskatchewan, for $\delta^{15}N$ and $\delta^{13}C$ using a Micromass Optima (Waters, Milford, MA, USA) continuous-flow isotope-ratio mass spectrometer directly coupled to a Carlo Erba NA1500 elemental analyzer (Elemental Microanalysis, Okehampton, UK). Samples were standardized against atmospheric nitrogen or Canyon Diablo PeeDee Belemnite (National Institute of Standards and Technology, Gaithersburg, MD, USA) using the equation $\delta^{15}N$ or $\delta^{13}C$ (‰) $= [(R_{sample} - R_{reference})/(R_{reference})] \cdot 1{,}000$, where $R = {}^{15}N{:}{}^{14}N$ or ${}^{13}C{:}{}^{12}C$. Replicate samples had a precision of ±0.05‰. Samples were not pre-extracted to remove lipids.

Water was analyzed using standard methods of the National Laboratory for Environmental Testing [14]. Total P and DOC data for Lake Champlain and Seneca Lake were obtained from recent reports [15,16]. Phytoplankton was analyzed according to standard inverted microscope methods as outline by Findlay and Kling (www.eman-rese.ca/eman/ecotools/protocols/freshwater/phytoplankton/intro.html). Small-celled organisms were measured and enumerated at ×938 magnification, and large, less numerous cells were analyzed at ×234 magnification using phase contrast inverted microscopy. Biomass was calculated from mean cell size using a geometric shape closely approximating that of the organism and a density of 1.

Statistical analysis

Statistical analyses were carried out using Systat® (Systat Software, Point Richmond, CA, USA). Concentrations less than method detection limits were assigned a value of half the method detection limit for calculation of means and trophic magnification factors (TMFs). All PCB and OCP data were converted to lipid weight for regression analyses and for calculating TMFs. All PCB or OCP concentration data, as well as results for lake water chemistry and phytoplankton composition, were examined for their fit to a normal distribution (Shapiro-Wilk test). The log10 (concentration data) or square root transformation (% composition data) were used to generate normally distributed data for simple correlation and regression analysis.

Trophic magnification factor calculations

For the food webs, TMFs were determined based on the relationship between log PCB or OCP (lipid wt) concentration and $\delta^{15}N$ [7,8]. Converting $\delta^{15}N$ to trophic level (TL) was performed according to the relationship TL = 2 + ($\delta^{15}N_{consumer} - \delta^{15}N_{zooplankton}$)/3.4, where 2 is the assumed TL of zooplankton and 3.4 is the trophic enrichment factor constant [9]. We used zooplankton to standardize the base of the food web because it was the only invertebrate collected in all 17 lakes. Trophic magnification factors were calculated based on the antilog value of the regression slope between log PCB/OCP concentrations (on a lipid wt basis) and TL. Analysis of covariance used the model PCB or OCP = TL + Lake + Lake·TL to test for differences of the slopes among lakes of the TMF relationships. Mean TMFs of individual PCBs or OCPs for northern and southern lakes were compared using one-tailed *t* tests.

RESULTS

PCBs or OCPs in lake trout, forage fish, and invertebrates

Lake trout represented the largest pool of organisms collected in the present study ($n = 357$), and they generally had the highest concentrations of PCBs and OCPs. Concentrations of the sum (Σ) of 57 PCB congeners detected in individual lake trout (whole fish) ranged between 1.67 and 2,890 ng/g wet weight (median 61.5 ng/g wet wt) (*Supporting Information*, Table S1; http://dx.doi.org/10.1897/08-071.S1). Results for PCBs and OCPs in lake trout are discussed in more detail in Guildford et al. [12]. Mean $\delta^{15}N$ of lake trout ranged from 9.9 (Opeongo) to 17.6‰ (Seneca) and was typically higher in lakes with high $\delta^{15}N$ of the primary consumers, zooplankton. With two exceptions (Seneca and Champlain), the lake trout had mean $\delta^{15}N$ that was higher than that of any other fish species sampled from that system; mean $\delta^{15}N$ of trout was, on average, 3.2‰ higher than that of the next highest species in the food web. For Seneca and Champlain lakes, mean $\delta^{15}N$ of lake trout was similar to that of some of the forage fish collected.

The number of fish species other than lake trout analyzed for PCBs and OCPs ($n = 235$) differed from one lake to another and reflected the different fish assemblages in the 17 systems studied. Sculpin had the highest concentrations of ΣPCB of all forage fish, with mean concentrations ranging from 32 ng/g wet weight in Lake Simcoe to 73 ng/g wet weight in Lake Champlain. Relatively high mean ΣPCB concentrations were also found in rainbow smelt (range from 5.9 ng/g wet wt in Eva to 55 ng/g wet wt in Champlain). Lake herring were obtained from Lake Superior, six lakes in northwestern Ontario, and in Saskatchewan. Mean ΣPCB concentrations in lake herring ranged from 2.0 ng/g wet weight in Cold Lake to 63 ng/g wet weight in Lake Superior (Table S1, http://dx.doi.org/10.1897/08-071.S1). Spottail shiners generally had lower concentrations of ΣPCB and OCP than those in other species, with mean ΣPCB ranging from 0.05 ng/g wet weight (Cold Lake) to 14 ng/g wet weight (Sandybeach Lake). Mean $\delta^{15}N$ values of these forage fish varied from lake to lake but were generally lower (see exceptions given earlier) than those of lake trout and higher than those of the invertebrates. For example, the most commonly collected forage fish, the shiner, had $\delta^{15}N$ that ranged from 5.9‰ in Grist Lake to 8.8‰ in Reindeer Lake; in these same two lakes, lake trout had mean $\delta^{15}N$ of 12.1 and 11.8‰, respectively, and zooplankton had mean $\delta^{15}N$ of 4.1 and 3.2‰, respectively.

Lower concentrations PCBs and OCPs were found in invertebrates than in fish from the 17 lakes. *Mysis* (Champlain, Simcoe, Superior) generally had the highest concentrations of ΣPCB, ΣDDT, and ΣCHL of any invertebrate taxa analyzed, followed by *Diporeia* (Superior) (Table S2, http://dx.doi.org/10.1897/08-071.S1). Zooplankton from all 17 lakes had ΣPCB concentrations that ranged from 0.06 ± 0.03 ng/g wet weight in Thunder Lake to 15 ± 7.0 ng/g wet weight in Lake Superior. Zooplankton in Lake Superior also had the highest mean ΣCHL (3.9 ng/g wet wt) and ΣHCH (1.9 ng/g wet wt) when compared to all other lakes sampled. In contrast, mean ΣDDT in plankton was highest in Lakes Simcoe (10.3 ng/g wet wt) and Seneca (3.9 ng/g wet wt), two lakes with agricultural land within their catchments. A variety of other invertebrates (Gastropoda, Trichoptera, Ephemeroptera) were analyzed, mainly from lakes in northwestern Ontario (Eva, Paguchi, Sandybeach, and Thunder). All had low (generally <1 ng/g wet wt) concentrations of ΣPCB and major OCPs (Table S2, http://dx.doi.org/10.1897/08-071.S1). Zooplankton had mean $\delta^{15}N$ that ranged from 1.9 (Eva) to 9.2‰ (Simcoe), but the majority were less than 5‰. In four lakes, both zooplankton and longer-lived mussels were collected, and their mean $\delta^{15}N$ values were similar (1.9 and 2.0‰ in Eva; 3.3 and 3.4‰ in Paguchi; 3.2 and 2.3‰ in Sandybeach; 4.2 and 2.9‰ in Thunder, respectively). In Cold and Grist lakes, the primary consumers, Gastropoda, also had $\delta^{15}N$ values that were similar to those measured in bulk plankton (means within 1.7‰); in Kingsmere Lake, however, the Gastropods had a mean $\delta^{15}N$ signature that was 3.9‰ lower than that of plankton.

PCB or OCP concentrations in relation to trophic level

Significant positive relationships between log chlorinated biphenyl (CB) congener 153, CB101, CB138, CB180, *p,p′*-DDE, or *trans*-nonachlor (lipid wt) concentrations and TL were found for all 17 food webs analyzed ($r^2 = 0.12 - 0.92$; $p < 0.05$). Relationships between log CB153 and TL of food web organisms are shown for all lakes in Figure 1. Log-transformed concentrations of CB52, CB99, HCB, and *cis*-CHL had significant relationships with TL in 15 or more lakes. The TMFs for these compounds were significantly greater than 1 (slope of regression line was significantly greater than 0), indicating biomagnification of these PCBs and OCPs in these freshwater food webs (Table 2). In contrast, log concentrations

Table 1. Location and selected biological characteristics of the 17 lakes in North America in which food webs were sampled between 1998 and 2001[a]

Lake	Province/ state[b]	Approx. latitude	Approx. longitude	Location	Forage fish[c]	Benthic and pelagic invertebrate[d]
Cold	AB	54.5	110.0	Northern	LH, LS, Sh, LT	Ben, zoo
Namur	AB	57.5	111.5	Northern	WF, LT, WS	Ben, zoo
Athabasca	AB/SK	58.5	111.0	Northern	WF, WS, Sh, LT, LS	Ben, zoo
Grist	AB	55.3	110.0	Northern	LH, WF, LT, Sh, ST	Ben, zoo
La Ronge	SK	55.3	105.0	Northern	LH, YP, LT, WS	Ben, zoo
Reindeer	SK	57.0	102.0	Northern	LH, YP, Sh, LT	Ben, zoo
Wollaston	SK	58.0	103.0	Northern	LS, LT	Ben, zoo
Kingsmere	SK	54.0	106.0	Northern	LT, Sh	Ben, zoo
Paguchi	ON	49.5	91.5	NW Ontario	Sh, LH, YP, LT	Ben, zoo, phyto
Eva	ON	48.6	91.2	NW Ontario	S, Sh, D, LT, S	Ben, zoo, phyto
Sandybeach	ON	49.8	92.3	NW Ontario	S, Sh, WS, LT	Ben, zoo, phyto
Thunder	ON	49.8	92.6	NW Ontario	LT, YP	Ben, zoo, phyto
Simcoe	ON	44.5	79.5	Southern	S, Sc, WF, LT	Zoo, phyto
Opeongo	ON	45.6	78.3	Southern	WF, LH, LT	Zoo, phyto
Seneca	NY	42.8	77.0	Southern	AL, Sc, LT, S	Zoo, phyto
Champlain	VT/NY	44.5	73.5	Southern	Sc, TP, S, LT	Zoo, *Mysis*, phyto
Superior[g]	ON/MI	47.0	90.5	Southern	LH, Sc, S, LT	Zoo, *Mysis*, phyto

[a] For additional characteristics, see Muir et al. [12].
[b] Vermont, New York, and Michigan (VT, NY, and MI) are in the United States. All others are in Canada.
[c] Lake trout were sampled in all lakes. AL = alewife; D = longnose dace; LH = lake herring; LS = longnose sucker; LT = lake trout; P = pike; S = rainbow smelt; Sc = slimy sculpin; Sh = shiner; ST = ninespine stickleback; TP = trout-perch; WF = lake whitefish; WS = white sucker; YP = yellow perch.
[d] Ben = benthic organisms; phyto = >1–<100 μm; zoo = net zooplankton tow (>110 mm).
[e] Trophic status of the lakes is based on total P concentrations. Meso = mesotrophic, 11 to 20 μg/L total P; oligo = oligotrophic, ≤11 μg/L total P (unfiltered).
[f] — = no data available.
[g] Apostle Islands area in the western arm of Lake Superior. Fish and zooplankton collection in 1998; phytoplankton samples collected in 2002.

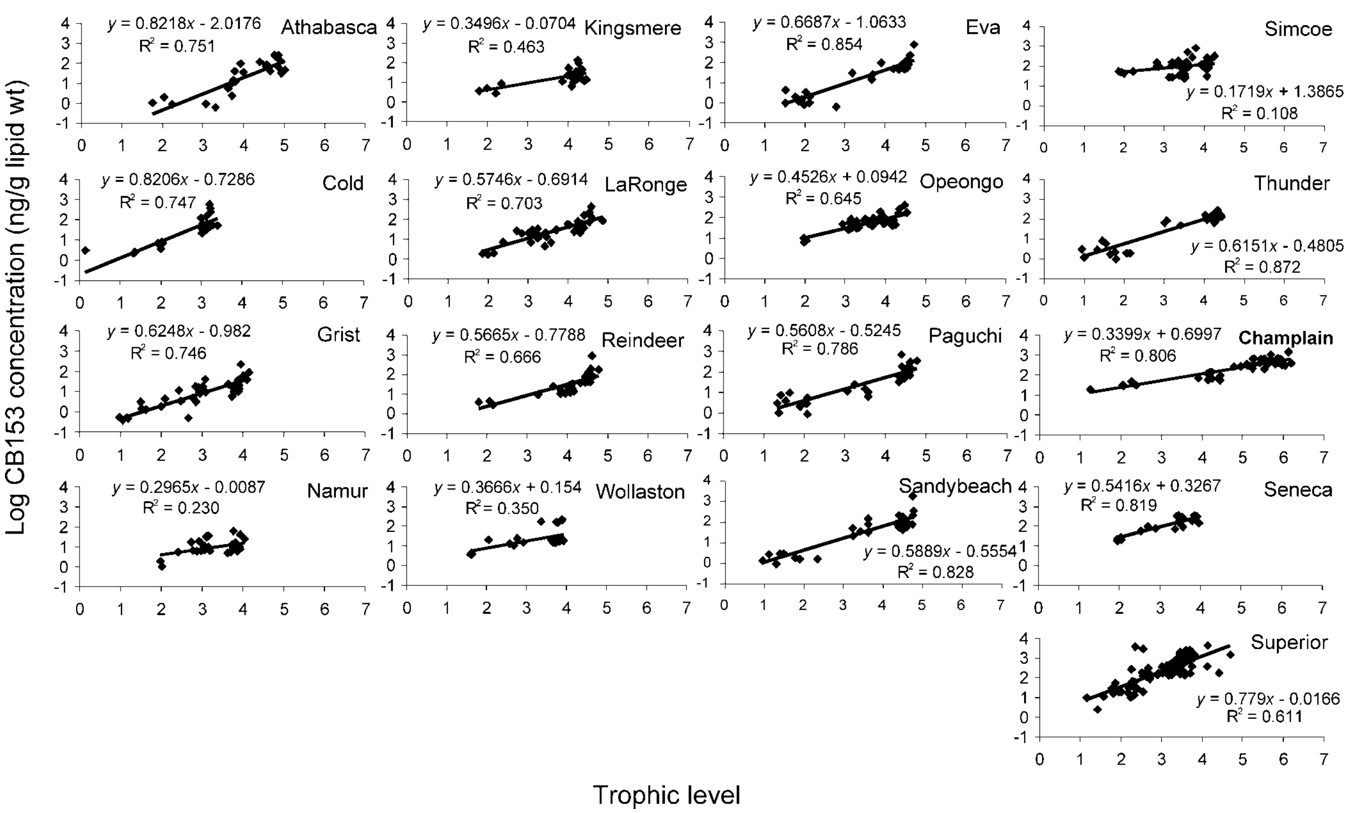

Fig. 1. Relationships between the concentrations of log chlorinated biphenyl (CB) congener 153 (ng/g lipid wt) and trophic level (calculated using $\delta^{15}N$; see *Materials and Methods* section) of organisms in 17 lake trout food webs by location (all in North America). All relationships were statistically significant ($p < 0.05$; see Table 2).

Table 1. Extended

Phytoplankton composition (%)						Trophic status[e]
Chlorophyta	Cyanophyta	Chrysophyceae	Diatomeae	Cryptophyceae	Dinophyta	
7.4	50.2	11.1	11.8	12.7	6.8	Meso
—[f]		—	—	—	—	Meso
28.4	24.1	0.4	20.3	26.9	0.0	Meso
—		—	—	—	—	Oligo
3.9	3.7	59.7	29.3	2.5	1.0	Oligo
8.7	9.1	64.2	16.3	1.1	0.6	Meso
13.4	2.0	59.2	21.8	0.7	2.9	Meso
0.5	2.4	11.1	85.4	0.1	0.1	Meso
4.4	17.9	31.8	30.8	7.7	7.6	Meso
2.4	8.9	19.2	55.5	6.4	7.5	Oligo
4.5	0.9	49.2	12.4	25.8	7.3	Meso
2.6	18.9	20.8	32.5	20.6	4.8	Oligo
15.8	10.7	25.4	27.6	15.3	5.2	Meso
17.6	26.5	28.8	11.5	10.7	4.7	Oligo
16.4	0.0	5.6	0.3	65.8	11.9	Oligo
—		—	—	—	—	Meso
5.9	4.1	29.3	24.7	20.6	10.9	Oligo

of α-HCH, lindane, and endosulfan had nonsignificant relationships ($p > 0.05$) with TL for eight or more lakes. Significant negative biomagnification (TMF < 1) was observed for lindane (five lakes), dieldrin (three lakes), α-HCH (two lakes), HCB (one lake), and *cis*-CHL (one lake). However, in most cases, the negative slope of the regression line was not statistically significant ($p > 0.05$) (Table 2). The compound *p,p'*-DDE had the highest TMFs of the 10 compounds, averaging 4.0 ± 1.8 across the 17 lakes, followed by *trans*-nonachlor (3.6 ± 1.7) and CB153 (3.4 ± 1.2). For endosulfan, TMFs were calculated only for 4 lakes, because in the other 13 lakes concentrations in more than 50% of the samples were

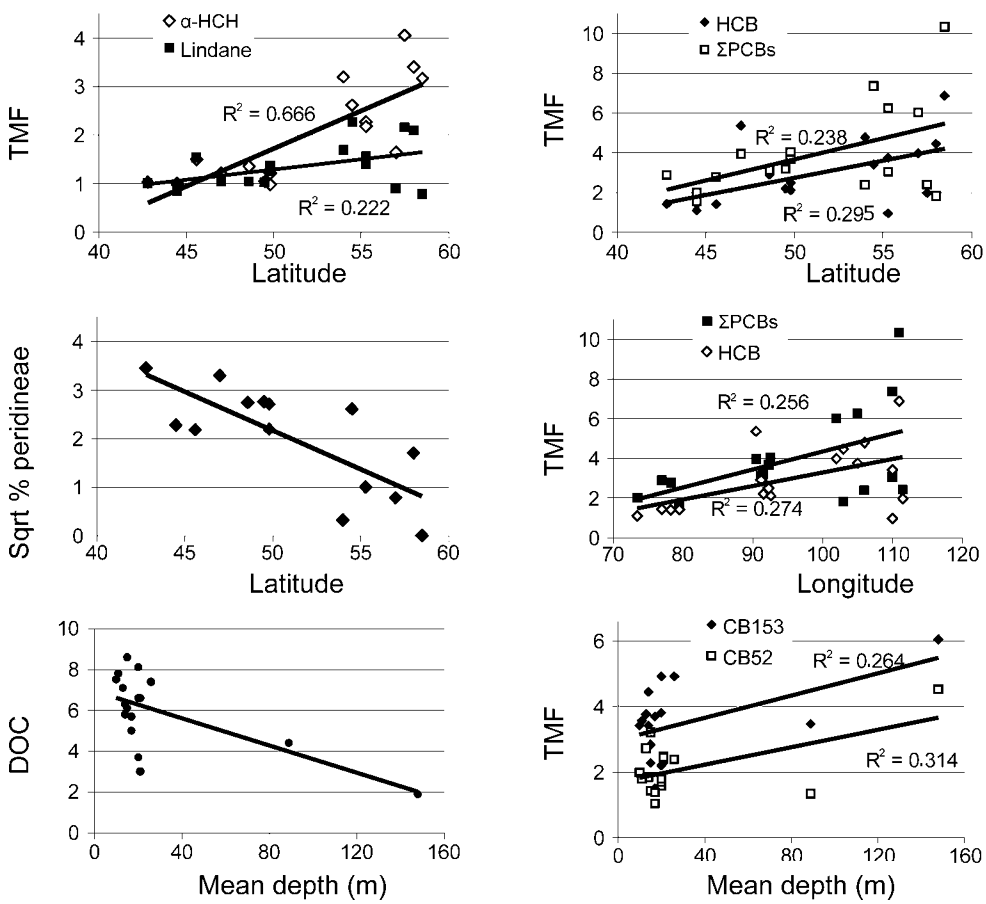

Fig. 2. Relationships between trophic magnification factor (TMF) of selected polychlorinated biphenyls (PCBs) and organochlorine pesticides and latitude, longitude, percentage of Dinophyta, and mean depth. Also shown to illustrate the covariance of water chemistry and phytoplankton composition are percentage of Dinophyta versus latitude and dissolved organic carbon (DOC) versus mean depth. All have statistically significant correlation coefficients ($p < 0.05$). CB = chlorinated biphenyl; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane.

Table 2. Trophic magnification factors (± standard error) of selected chlorinated biphenyls (CBs) and organochlorine pesticides in 17 lake trout food webs[a] from lakes in North America[bc]

Lake	CB153	CB52	CB138	CB101	CB180	CB99
Athabasca	4.9 ± 0.5	2.4 ± 0.7	5.5 ± 0.7	3.4 ± 0.6	6.9 ± 0.7	8.0 ± 1.3
Champlain	2.2 ± 0.2	1.6 ± 0.5	2.1 ± 0.2	1.9 ± 0.3	2.5 ± 0.2	2.6 ± 0.2
Cold	4.9 ± 0.7	1.7 ± 0.8	4.3 ± 0.7	3.3 ± 0.8	9.0 ± 0.8	5.2 ± 1.1
Eva	4.4 ± 0.4	1.8 ± 0.5	4.4 ± 0.4	4.0 ± 0.3	4.5 ± 0.4	3.7 ± 0.3
Grist	3.4 ± 0.3	2.0 ± 0.4	3.2 ± 0.4	1.4 ± 0.5	3.5 ± 0.5	2.4 ± 0.5
Kingsmere	2.3 ± 1.1	2.5 ± 0.7	2.3 ± 1.2	1.6 ± 1.0	2.5 ± 1.3	2.3 ± 1.2
La Ronge	3.8 ± 0.4	2.7 ± 0.8	3.6 ± 0.4	4.1 ± 0.6	5.9 ± 0.8	1.6 ± 1.2
Namur	2.3 ± 0.6	3.2 ± 0.6	1.9 ± 0.6	1.9 ± 0.5	5.3 ± 0.7	2.8 ± 0.8
Opeongo	2.8 ± 0.3	1.4 ± 0.5	3.1 ± 0.3	1.7 ± 0.4	5.0 ± 0.5	3.5 ± 0.3
Paguchi	3.4 ± 0.4	1.8 ± 0.5	3.7 ± 0.5	3.2 ± 0.3	3.9 ± 0.3	3.4 ± 0.4
Reindeer	3.7 ± 0.4	1.0 ± 7.8*	3.4 ± 0.4	2.6 ± 0.7	3.4 ± 0.4	4.2 ± 0.8
Sandybeach	3.8 ± 0.3	1.8 ± 0.4	3.5 ± 0.3	3.2 ± 0.3	3.4 ± 0.3	3.5 ± 0.3
Seneca	3.5 ± 0.4	1.3 ± 1.0	3.1 ± 0.4	2.3 ± 0.6	3.6 ± 0.5	3.5 ± 0.3
Simcoe	1.5 ± 0.5	1.4 ± 0.4	1.6 ± 0.5	1.5 ± 0.5	1.7 ± 0.5	1.6 ± 0.5
Superior	6.0 ± 0.5	4.5 ± 0.4	5.7 ± 0.4	3.2 ± 0.3	6.5 ± 0.5	4.8 ± 0.4
Thunder	3.6 ± 0.5	1.8 ± 0.3	4.7 ± 0.7	4.0 ± 0.6	4.9 ± 0.7	4.8 ± 0.7
Wollaston	2.3 ± 0.6	2.5 ± 0.7	1.8 ± 0.3	1.3 ± 0.5	2.7 ± 0.5	5.1 ± 0.7
Means (all)[e]	3.4 ± 1.2	2.0 ± 0.8	3.3 ± 1.2	2.6 ± 1.0	4.3 ± 1.9	3.6 ± 1.6
Northern	3.4 ± 1.2	2.3 ± 0.7	3.3 ± 1.4	2.6 ± 1.0	5.1 ± 2.4	4.2 ± 2.2
Northwest Ontario	3.7 ± 0.4	1.8 ± 0.1	3.9 ± 0.6	3.2 ± 1.0	4.1 ± 0.7	3.5 ± 0.9
Southern	3.2 ± 1.7	2.0 ± 1.4	3.1 ± 1.6	2.1 ± 0.6	3.8 ± 1.9	3.2 ± 1.2

[a] Trophic magnification factors = 10^{slope}.
[b] DDE = dichlorodiphenyldichloroethylene; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane.
[c] All trophic magnification factors based on regression slopes were statistically significant ($p < 0.05$) except those identified with * ($p > 0.05$).
[d] Dashes indicate that the trophic magnification factor was not calculated because >50% of samples had endosulfan < method detection limit.
[e] Means ± standard deviation of the trophic magnification factors for all lakes and for separate regions (northern = Athabasca, Cold, Grist, Kingsmere, La Ronge, Namur, Reindeer, Wollaston; northwest Ontario = Eva, Paguchi, Sandy Beach, Thunder; southern = Champlain, Opeongo, Simcoe, Seneca, Superior).

less than the method detection limit. Still, these TMFs were significant, with values −0.6, 1.8, and 2.2 in three of the four lakes.

When comparing the TMFs for individual PCBs and OCPs across lakes, some geographic patterns were found. The lowest TMFs were observed in the southern lakes Champlain (0.8–3.0) and Simcoe (0.82–2.17) for the PCBs and OCPs when compared to other lakes in the present study. By contrast, the food webs with the most elevated TMFs, particularly for *p,p′*-DDE, *trans*-nonachlor, and CB153, were the northern lakes and some of those in northwest Ontario, including Cold, Athabasca, Eva, Reindeer, and Superior. Average TMFs in northern lakes (Alberta or Saskatchewan) were generally higher than those in southern lakes (Ontario or New York state, including Superior; Table 2). Comparisons across regions using a *t* test (one-tailed) showed that mean TMFs for HCB, α-HCH, *trans*-nonachlor, and lindane were significantly greater ($p < 0.05$) in the northern lakes than in southern lakes (see Table 1 for lake location), while no significant differences were found for TMFs of ΣPCBs, PCB congeners, *p,p′*-DDE, dieldrin, or *cis*-CHL. For ΣDDT and CB101, TMFs were significantly higher in northwest Ontario lakes compared with southern lakes ($p < 0.05$); however, no other significant differences were found between these two regions or between northwest Ontario and northern lakes ($p > 0.05$).

TMFs and lake trophic status or phytoplankton composition

Significant correlations were found between TMFs of ΣPCBs, PCB congeners, HCB, α-HCH, or *trans*-nonachlor and lake area, latitude, longitude, and/or mean depth (Fig. 2 and Table 3). Only TMFs for HCB were significantly positively correlated ($p < 0.05$) with lake area and volume (not shown), but TMFs for several PCBs and OCPs increased significantly with latitude and/or longitude of the system. Particularly striking was the strong correlation with latitude and longitude of the TMFs for α-HCH (r^2 = 0.81 and 0.80, respectively) and lindane (r^2 = 0.47 and 0.52, respectively), indicating greater biomagnification of these OCPs in the western and northern lakes of Saskatchewan and Alberta than in those systems in the more southern and eastern regions in the present study. For CB52 and CB153, but not other PCB congeners, TMFs were significantly and positively correlated with mean depth. No significant correlations of TMFs for ΣDDT, *p,p′*-DDE, dieldrin, CB101, and CB180 were found with any lake characteristic.

Correlations between TMFs of PCBs or OCPs and water chemistry variables (DOC, total P, or phytoplankton composition [% Dinophyta]) were also found (Table 3). For *cis*-CHL, TMFs increased significantly in lakes with higher DOC concentrations. Total P concentrations were also significantly and positively correlated to TMFs for ΣPCBs, CB99, HCB, *trans*-nonachlor, and α-HCH for these 17 lakes. Significant negative correlations were found between TMFs of HCB or α-HCH and percentage of Dinophyta (square root transformed) (Table 3). However, TMFs for PCBs and OCPs were not significantly correlated with total phytoplankton biomass of lakes or the percentage of other individual algal groups (Cyanophyta, Chlorophyta, Chrysophyceae, Diatomeae; data not shown).

The water chemistry parameters, as well as percentage of Dinophyta, were, as might be expected, correlated ($p < 0.05$) with one another and with lake size, depth, latitude, and longitude (Table 4). For example, the percentage of Dinophyta was negatively correlated with latitude and longitude, while

Table 2. Extended

α-HCH	*cis*-Chlordane	Dieldrin	HCB	*p,p′*-DDE	*trans*-Nonachlor	Lindane	α-Endosulfan
3.2 ± 0.3	2.4 ± 0.6	1.8 ± 0.5	6.9 ± 0.6	4.5 ± 0.6	6.4 ± 0.8	−0.8 ± 0.6*	2.2 ± 0.9
−1.0 ± −5.7*	1.3 ± 0.3	1.1 ± 1.0*	1.1 ± 1.4*	2.1 ± 0.2	3.0 ± 0.2	−0.8 ± 0.3	—[d]
2.6 ± 0.4	2.4 ± 0.7	1.3 ± 0.9*	3.4 ± 0.5	8.8 ± 0.6	7.2 ± 0.9	2.3 ± 0.5	—
1.4 ± 0.5	2.7 ± 0.3	2.3 ± 0.4	2.9 ± 0.3	5.9 ± 0.4	4.3 ± 0.4	1.0 ± 2.0*	—
2.3 ± 0.3	2.7 ± 0.4	2.2 ± 0.4	−0.9 ± 2.6	3.3 ± 0.4	3.3 ± 0.4	1.4 ± 0.8*	—
3.2 ± 0.2	2.5 ± 0.8	1.5 ± 0.3	4.8 ± 0.3	4.6 ± 1.4	5.5 ± 1.2	1.7 ± 0.4	—
2.2 ± 0.5	3.2 ± 0.6	1.5 ± 0.7	3.7 ± 0.7	4.4 ± 0.5	3.9 ± 0.8	1.6 ± 0.8*	−0.6 ± 0.3
4.0 ± 0.8	4.4 ± 0.5	1.2 ± 4.4	2.0 ± 0.7	2.2 ± 0.7	2.5 ± 0.5	2.2 ± 0.8	—
1.5 ± 0.6	2.8 ± 0.5	2.9 ± 0.4	1.4 ± 0.3	3.6 ± 0.4	2.3 ± 0.4	1.5 ± 0.4	—
1.0 ± 2.6*	2.2 ± 0.3	1.7 ± 0.4	2.2 ± 0.2	4.0 ± 0.3	3.6 ± 0.4	1.0 ± 3.8*	—
1.6 ± 1.0	1.7 ± 0.5	1.3 ± 0.4	4.0 ± 0.4	3.7 ± 0.5	3.5 ± 0.3	−0.9 ± 0.8*	1.2 ± 1.8*
1.2 ± 0.4	2.6 ± 0.3	1.8 ± 0.3	2.5 ± 0.3	4.7 ± 0.4	4.0 ± 0.3	1.3 ± 0.5	—
1.0 ± 3.7	2.4 ± 0.4	1.1 ± 3.3*	1.4 ± 0.5*	4.8 ± 0.6	3.1 ± 0.4	1.0 ± 29*	—
1.0 ± 12	1.6 ± 0.4	−1.0 ± 4.0*	1.4 ± 0.4	1.9 ± 0.5	1.4 ± 0.5	−1.0 ± 15*	—
1.2 ± 0.4	3.3 ± 0.3	1.9 ± 0.2	5.3 ± 0.5	5.9 ± 0.5	4.0 ± 0.5	1.0 ± 1.8	1.8 ± 0.5
1.0 ± 0.1	2.8 ± 0.4	1.2 ± 0.2	2.1 ± 0.3	5.1 ± 0.8	2.9 ± 0.4	1.4 ± 0.2	—
3.4 ± 0.5	−0.8 ± 0.6*	−0.9 ± 0.9*	4.4 ± 1.0	1.4 ± 0.8*	2.7 ± 0.6	2.1 ± 0.5	—
1.9 ± 1.0	2.4 ± 0.8	1.5 ± 0.5	2.9 ± 1.7	4.0 ± 1.8	3.6 ± 1.5	1.3 ± 0.5	
2.9 ± 0.8	2.5 ± 1.1	1.4 ± 0.3	4.2 ± 1.5	4.2 ± 2.4	4.5 ± 1.8	1.6 ± 0.6	
1.4 ± 0.5	2.6 ± 0.2	1.8 ± 0.4	2.1 ± 0.7	4.6 ± 1.0	3.6 ± 0.5	1.2 ± 0.2	
1.1 ± 0.2	2.3 ± 0.8	1.6 ± 0.8	2.1 ± 1.8	3.7 ± 1.7	2.7 ± 1.0	1.1 ± 0.3	

total P was significantly positively correlated with latitude and longitude. Dissolved organic carbon was negatively correlated with lake volume and mean depth and positively correlated with total P.

DISCUSSION

Results from the present study concurred with several others on aquatic food webs and showed that concentrations of PCBs and OCPs in freshwater biota were significantly related to their trophic position (as determined using $\delta^{15}N$) and that the slopes of these relationships can be used to quantify the biomagnification of these contaminants through these food webs [2,7,8,13]. Unlike many of the prior studies that have focused on single systems or lakes within a small geographic range, we found highly significant relationships between log-transformed concentrations (lipid wt) of PCBs or OCPs and TL (calculated using $\delta^{15}N$) in the lakes we studied from New York state, USA, to northern Alberta, Canada, demonstrating that this tool can be broadly applied across systems that differ considerably in their location and characteristics. As a result, researchers can use this approach to determine whether variability in climate or physical or chemical characteristics of lakes affect the trophic transfer of these pollutants up to top predators by contrasting the TMFs derived from the slopes of the regressions across the gradients of interest. Indeed, we found that some PCBs and OCPs were biomagnified at a greater rate in larger lakes, and lakes with higher phosphorus or DOC and at greater latitudes and longitudes.

Trophic magnification of PCBs and OCPs in lake trout food webs

To date, most studies that have used $\delta^{15}N$ to determine trophic positioning of organisms, and to quantify the trophic transfer of persistent pollutants, have focused on one or a few different food webs [9,17]. Herein we sampled food web organisms from 17 lakes that all support populations of lake trout but that vary in their physical and chemical characteristics, and we showed that concentrations of several of the PCBs and OCPs were significantly related to TL within most or all of these systems. For the 12 individual compounds regressed against trophic position, relationships between lipid weight concentrations of CB153, CB138, CB101, CB180, CB99, or *trans*-nonachlor and TL were significant in all lakes. However, across lakes for these individual PCBs or OCPs, the slopes of these relationships varied significantly (TMFs for *p,p′*-DDE ranged from 1.9 to 8.8, with the interaction term in an analysis of covariance of $p < 0.001$), indicating that characteristics other than physical or chemical properties of the contaminant affect their average rate of increase per TL within these food webs. For the other PCBs or OCPs that we examined, the slopes of the lipid-adjusted concentrations versus TL were not significant ($p > 0.05$) for between one and four of the lakes. Significant relationships between chemical concentration and TL was not observed in several of the lakes (Wollaston, Simcoe, Seneca, Paguchi, Cold, Reindeer, Champlain), and this lack of a clear biomagnification process was evident regardless of chemical hydrophobicity or lake properties (location, trophic status, etc.). For example, concentrations of dieldrin were not significantly predicted by TL in 5 of the 17 lakes, and these systems ranged in their characteristics from meso- to oligotrophic, spanned a spectrum of food webs, and were located in both the southern and the northern lake groups.

The field-measured TMFs of PCBs and OCPs in the 17 lakes in the present study were generally similar to those published in the literature for other freshwater food webs, although some exceptions were found. In Lake Superior, Kucklick and Baker [8] also found significant relationships between log-transformed ΣPCB and $\delta^{15}N$ of organisms. Based on their data, we calculated the TL for each organism and the TMF for the ΣPCB (lipid normalized) using amphipods as the baseline. The TMF was 3.3, which is lower than observed in the present study for Lake Superior but within the range of TMFs for the 17 lakes. Tomy et al. [18] reported TMFs for ΣPCB and *p,p′*-DDE of 6.1 and 5.7, respectively, for the Lake Ontario food web based on results from Kiriluk et al. [7], and these values

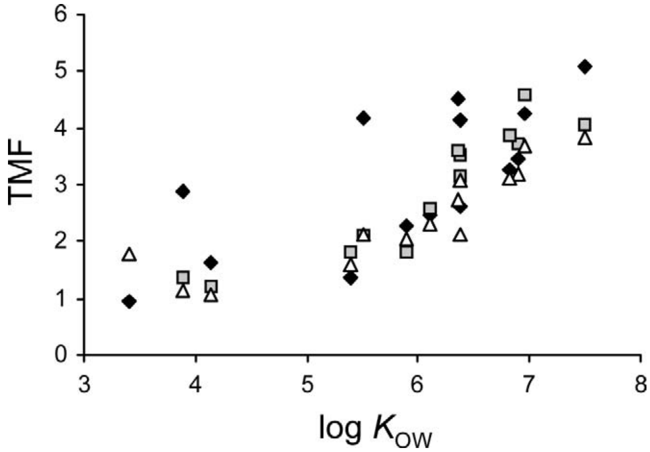

Fig. 3. Relationship of average trophic magnification factors (TMFs) in northern lakes (◆), northwestern Ontario lakes (■), and southern lakes (△) in North America with the octanol–water partition coefficient (log K_{OW}). R^2 values trophic magnification factors versus log K_{OW} for each region were highly significant ($p < 0.01$). The trophic magnification factors are given in Table 2.

were similar to what we found for Lake Superior but higher than the values for most of the other lakes in the present study.

Another advantage of calculating an average TMF for food webs is that it allows broader scale comparisons to be made. Trophic magnification factors can be compared not only across regions for freshwater food webs but also between marine waters and freshwaters or among arctic, temperate, and tropical systems. For example, Borga et al. [17] found that the TMFs for *trans*-nonachlor were similar (4.8–5.5, with or without marine mammals) across six high-latitude marine food webs in the Alaskan, Canadian, European, and Russian Arctic; one exception was a TMF of 10.4 in a Barents Sea food web. In the present study, TMFs for these freshwater food webs ranged from 1.4 to 7.2 (mean of 3.6), indicating that the average biomagnification of *trans*-nonachlor through these systems was typically lower than what has been measured in marine food webs. It is through comparisons such as these that we can obtain a broader understanding of factors affecting the biomagnification of PCBs, OCPs, and other halogenated organics in aquatic food webs.

In the present study, we used lipid rather than wet weight concentrations of PCBs or OCPs versus TL to calculate TMFs. After adjusting concentrations of these compounds for percentage of lipids, we continued to find a significant relationship between an organism's concentrations of PCBs or OCPs and its TL, indicating that both variables are important determinants of contaminant concentrations in biota. This concurs with results from previous studies. Kidd et al. [3] also found significant relationships between the slope of lipid-adjusted α-HCH, *trans*-nonachlor, CB52, CB153, or *p,p′*-DDE and $\delta^{15}N$ in a subarctic lake food web. By using lipids as the covariate, rather than calculating food web magnification factors with lipid-normalized data, the authors were able to demonstrate that lipids accounted for most of the increasing concentration with TL. However, it is important to consider that the percentage of lipids is low and variable in plankton and other invertebrates (Table S2, http://dx.doi.org/10.1897/08-071.S1) and that this may be a major source of uncertainty in the calculation of TMFs. Extraction and gravimetric determinations of the percentage of lipids in lean tissues has been shown to be quite variable among laboratories, and colorimetric methods are now recommended [19].

The TMFs calculated in the present study for individual PCBs and OCPs were significantly correlated with their oc-

Table 3. Correlation coefficients of trophic magnification factors for selected polychlorinated biphenyls (PCBs) and organochlorine pesticides versus selected lake characteristics, phytoplankton composition, and water chemistry parameters; only those with one or more significant correlations are listed[a–d]

	Latitude	Longitude	Log lake area	Mean depth	DOC	Total P unfiltered	Dinophyta[f]	Latitude + log mean depth	Latitude + log lake area
n	17	17	17	17	17	17	14		
ΣPCB[e]	0.488*	0.506*	0.394	−0.014	0.304	0.693*	−0.346	0.536*	0.592*
CB52	0.248	0.352	0.395	0.560*	−0.202	−0.078	−0.058	0.621*	0.443
CB99	0.323	0.279	0.373	0.210	−0.067	0.570*	0.003	0.534	0.466
CB101	0.040	0.126	0.001	0.065	0.256	0.085	0.120	0.045	0.045
CB138	0.049	0.172	0.236	0.393	0.051	0.251	0.109	0.346	0.237
CB153	0.107	0.244	0.354	0.514*	−0.068	0.199	0.169	0.521	0.359
CB180	0.281	0.418	0.210	0.202	0.209	0.408	0.069	0.395	0.332
ΣDDT	0.060	0.224	−0.153	0.010	0.326	0.333	0.213	0.063	0.173
p,p′-DDE	−0.027	0.192	−0.026	0.272	0.129	0.259	0.199	0.259	0.032
HCB	0.543*	0.523*	0.611*	0.278	−0.141	0.477*	−0.571*	0.784*	0.771*
cis-Chlordane	0.096	0.292	−0.248	0.189	0.493*	−0.105	0.162	0.134	0.279
trans-Nonachlor	0.414	0.544*	0.155	0.039	0.206	0.734*	−0.282	0.504	0.427
Dieldrin	−0.116	−0.035	−0.226	0.011	0.185	−0.111	0.041	0.195	0.243
α-HCH	0.816*	0.800*	0.066	−0.237	0.297	0.497*	−0.705*	0.827*	0.817*
Lindane	0.472*	0.523*	−0.242	−0.247	0.247	0.097	−0.005	0.479	0.560*

[a] The following parameters did not show significant correlations with trophic magnification factors: log lake volume ($n = 17$), phytoplankton biomass ($n = 14$), percentage of Cyanobacteria ($n = 13$), percentage of Chlorophyta ($n = 14$), percentage of Chrysophyceae ($n = 14$), square root percentage of Bacillariophyceae ($n = 14$), log C:P ratio ($n = 14$), N:P ratio ($n = 14$).
[b] Values of certain parameters were transformed to yield normally distributed data as indicated.
[c] Results for latitude + log mean depth or log lake area are *r* values for multiple regression analyses. Multiple regression used latitude and log mean depth or latitude and log lake area as an independent variable.
[d] Statistically significant relationships ($p < 0.05$) are identified by an asterisk (*).
[e] DOC = dissolved organic carbon; CB = chlorinated biphenyl; DDE = dichlorodiphenyldichloroethylene; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane.
[f] Square root transformed.

Table 4. Correlations among lake characteristics, phytoplankton composition, and water chemistry parameters; only those with two or more significant correlations are shown[a]

	n	Latitude	Longitude	Log lake area	DOC	Mean depth
Log lake area	17	0.327	0.035	NA	NA	
Log volume	17	0.039	−0.009	0.980*	−0.701*	NA
DOC	17	0.362	0.418	−0.611*	NA	−0.700*
Log total P unfiltered	17	0.565*	0.554*	−0.150	0.502*	−0.058
Square root Dinophyta	14	−0.761*	−0.673*	−0.224	−0.371	0.458
Log particulate C	14	0.645*	0.724*	−0.041	0.492	−0.548*
Log particulate N	14	0.472	0.587*	0.040	0.461	−0.436
Mean depth	17	−0.386	−0.276	0.545*	−0.700*	NA

[a] Values of certain parameters were transformed as indicated to yield normally distributed data. Statistically significant relationships ($p < 0.05$) are identified by an asterisk (*). DOC = dissolved organic carbon; NA = not applicable.

tanol–water partition coefficient (log K_{OW}) values (Fig. 3), a widely used measure of bioaccumulation potential of persistent organohalogen compounds. Higher TMFs were found for the most hydrophobic compounds (CB153, CB138, CB180, CB99, and *p,p*′-DDE) compared with those of intermediate (HCB, dieldrin, and *cis*-CHL) and lower (lindane and endosulfan) hydrophobicity. Similar TMF versus log K_{OW} relationships were seen across all three regions, although some of the northern lakes had higher TMFs than either the northwest Ontario or the southern lakes for compounds at the same log K_{OW}. These results confirm that TMFs are indicative of the biomagnification potential of recalcitrant PCBs or OCPs over a wide range of chemicals and lake environments. Previous studies on a lake in the Canadian Arctic and on Lake Ontario have also shown that the slope of log-transformed PCBs or OCPs versus $\delta^{15}N$ is higher for the more lipophilic organochlorines, even after adjusting for the effects of lipid content [3,7]. The limited biomagnification of endosulfan, lindane, and to some extent, dieldrin and α-HCH found in the present study is consistent with other studies and is predicted from food web modeling. Kidd et al. [3] found a TMF of approximately 1 for α-HCH in a subarctic lake trout food web. Lindane and endosulfan have relatively short half-lives in fish in laboratory studies [20,21] and thus low predicted bioaccumulation when using the Gobas model [22] for the Lake Ontario food web.

Although we and others have shown that $\delta^{15}N$ can be used to quantify the trophic transfer of the legacy organochlorines, recent studies have shown that this approach can be applied to other emerging contaminants, such as halogenated organics, and that these compounds have biomagnification rates similar to those we obtained for PCBs, *p,p*′-DDE, and *trans*-nonachlor. For example, in the Lake Ontario food web, TMFs of 5.9 and 6.3 have been found for perfluorooctane sulfonate and hexabromocyclododecane, respectively [18,23].

Relationships of TMFs with characteristics of the lakes

One of the objectives of the present study was to examine the effects of lake trophic status, as indicated by phytoplankton composition and water chemistry parameters on TMFs. Indeed, significant positive correlations were found between TMFs of some PCBs or OCPs and lake characteristics such as total P (ΣPCB, CB99, HCB, and *trans*-nonachlor) and DOC (*cis*-CHL), and negative correlations were found between HCB or α-HCH and percentage of Dinophyta (Table 3). These results suggest that the lake trophic state has some influence over the rate of biomagnification of some of these contaminants, but it was not possible to delineate their relative influence from that of latitude and longitude. Parameters such as DOC, total P, and percentage of Dinophyta were also correlated with latitude and longitude (Table 4). Thus the positive or negative correlations of TMFs (*trans*-nonachlor, HCB, ΣPCB, CB99, and α-HCH) with total P, DOC, or percentage of Dinophyta may simply reflect the relationship ($p < 0.05$) of these water quality parameters with latitude and longitude (Table 4).

For most individual PCB congeners and OCPs (*p,p*′-DDE, *cis*-CHL, and dieldrin), TMFs were not correlated with latitude and longitude, nor with physical characteristics of the lakes, such as mean depth or surface area. Multiple linear regression analyses using latitude and (log) lake area or latitude and (log) mean depth generally did not offer much additional explanatory power compared to simple correlations (Table 3). However, positive correlations were found for CB52, *trans*-nonachlor, HCB, α-HCH, or lindane and some of these variables (Table 3). Higher TMFs for α-HCH and lindane were associated with the most western and northern lakes (northwest Ontario and northern Saskatchewan and Alberta). The results suggest that HCH isomers, which have low bioaccumulation in laboratory studies [21], biomagnify to a greater extent in colder, northern environments. Higher TMFs for ΣPCBs were also associated with these northern lakes compared to southern lakes (*t* test, $p = 0.055$), although this was not observed for major PCB congeners. Total PCB concentrations in lake trout were lower in more northerly lakes [12]; thus, the larger TMF values were not driven by elevated ΣPCBs in top predators but likely by a greater trophic transfer of these compounds up the food web.

What explains the positive correlation of TMFs for ΣPCBs, HCB, α-HCH, and lindane with latitude? Guildford et al. concluded that access to littoral sources (indicated by a heavier $\delta^{13}C$) explained differences in ΣPCB concentrations in lake trout between cold northern lakes and warmer southern lakes [12]. In the southern lakes (especially Simcoe, Seneca, and Champlain) lake trout are confined to cold pelagic waters in summer and thus feed only on the pelagic food web, while in northern lakes lake trout have year-round access to both littoral and pelagic food webs. Furthermore most of the southern lakes, except for Lake Opeongo, include agricultural activities and municipalities within their catchments. Thus, past local inputs, combined with greater atmospheric deposition (which is known to decline with latitude [24]), give rise to higher residues of PCBs and most legacy OCPs (DDT, CHL, and dieldrin) in southern lakes. The northern lakes have much lower population density within their immediate catchments, particularly Athabasca, Reindeer, and La Ronge, and are influenced mainly by atmospheric deposition. The overall effect is for invertebrates to have lower PCB and OCP residues while top predators in northern lakes, such as lake trout, with multiple feeding sources, achieve concentrations that are higher than might be predicted given low inputs. This results in a steeper relationship of TL versus lipid-adjusted concentrations. This explanation is less satisfactory for lindane and α-HCH. Concentrations of HCH isomers were generally higher in samples from Saskatchewan and Alberta lakes, particularly in invertebrates (Table S1, http://dx.doi.org/10.1897/08-071.S1), reflecting past use of technical HCH and recent lindane use in the Canadian prairies [25]). These compounds, along with endosulfan, are eliminated by fish rather rapidly due to lower hydrophobicity and due to metabolism, while the other 13 compounds are more hydrophobic and recalcitrant. One possibility is that generally lower water temperatures in the northern lakes and longer ice cover result in lower rates of volatilization, elimination, and/or biotransformation of HCH isomers within the food web; hence, they behave more like recalcitrant OCPs in these lakes.

Overall, the results of the present study showed that biomagnification of PCBs and most OCPs, as measured by TMFs, is only weakly influenced by such factors as latitude and longitude. Exceptions are HCH isomers and HCB, whose TMFs were generally greater in northern lakes, possibly due to lower rates of elimination and biotransformation in the food web. The average TMFs for 14 compounds were significantly correlated with log K_{OW}, implying that the rate of accumulation along the food web is dependent on the hydrophobicity and recalcitrance. Although correlations of TMFs with various water chemistry parameters and phytoplankton composition were found, most of these parameters correlated with latitude and longitude; therefore, the influence of water chemistry and lake trophy on TMFs could not be determined.

SUPPORTING INFORMATION

Table S1. Arithmetic mean concentrations (ng/g wet wt) of polychlorinated biphenyls (PCBs) and major organochlorine pesticides (OCPs) in fish sampled in 17 lakes across Canada and the northeastern United States.

Table S2. Arithmetic mean $\delta^{13}C$, $\delta^{15}N$, percentage of lipids, and concentrations (ng/g wet wt) of polychlorinated biphenyls (PCBs) and major organochlorine pesticides (OCPs) in invertebrates sampled in 17 lakes across Canada and the northeastern United States.

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