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## Article

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# PCB Concentrations in Lake Trout (*Salvelinus namaycush*) Are Correlated to Habitat Use and Lake Characteristics

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This study considers the importance of lake trout habitat as a factor determining persistent organochlorine (OC) concentration. Lake trout is a stenothermal, cold water species and sensitive to hypoxia. Thus, factors such as lake depth, thermal stratification, and phosphorus enrichment may determine not only which lakes can support lake trout but may also influence among-lake variability in lake trout population characteristics including bioaccumulation of OCs. A survey of 23 lakes spanning much of the natural latitudinal distribution of lake trout

provided a range of lake trout habitat to test the hypothesis that lake trout with greater access to littoral habitat for feeding will have lower concentrations of OCs than lake trout that are more restricted to pelagic habitat. Using the  $\delta^{13}\text{C}$  stable isotope signature in lake trout as an indicator of influence of benthic littoral feeding, we found a negative correlation between lipid-corrected  $\delta^{13}\text{C}$  and  $\Sigma\text{PCB}$  concentrations supporting the hypothesis that increasing access to littoral habitat results in lower OCs in lake trout. The prominence of mixotrophic phytoplankton in lakes with more contaminated lake trout indicated the pelagic microbial food web may exacerbate the biomagnification of OCs when lake trout are restricted to pelagic feeding. A model that predicted  $\Sigma\text{PCB}$  in lake trout based on lake area and latitude (used as proximate variables for proportion of littoral versus pelagic habitat and accessibility to littoral habitat respectively) explained 73% of the variability in  $\Sigma\text{PCBs}$  in lake trout in the 23 lakes surveyed.

## Introduction

Lake trout, the top predator in many oligotrophic northern lakes can accumulate high concentrations of persistent organochlorine compounds (OCs), and this presents a serious health risk when consumed on a regular basis as is often the case in remote communities in northern Canada. In 1998–2001 a large survey of lakes ranging from northern Alberta to the northeastern US was conducted with the goal of increasing our ability to predict the risk of OC contaminant exposure to human consumers. This survey rendered information on PCB concentrations and other contaminants (1) over a wide range of lakes and also collected auxiliary information on some limnological characteristics as well as stable isotope composition of the fish and associated biota. We have used these data to test hypotheses about ecosystem controls on PCB bioaccumulation by lake trout.

Much progress has been made in the understanding of the biomagnification of OCs in piscivores. Studies have clearly demonstrated that biomagnification increases with food chain length and organism fat content (2, 3), and this is understood in terms of the hydrophobic, recalcitrant nature of the contaminants that are retained in prey lipids and are efficiently transferred through the food web. Food web biomagnification predicts that organisms occupying higher trophic positions, both within a given food web and across systems, will achieve higher contaminant concentrations assuming that similar system-wide contaminant loadings are present. Other studies have demonstrated that biomagnification can decrease with trophic enrichment of the water body (4, 5), and this has been attributed to enhanced phytoplankton growth which more effectively transports dissolved contaminants from the water column to sediments via algae sinking (4) and more efficient gross growth efficiencies of fish inhabiting these systems which consume less food per unit of body mass as a result of greater food abundance and/or food quality (6, 7). This trophic dilution hypothesis therefore predicts decreasing contaminant concentrations in more productive systems or in species exhibiting higher growth efficiencies.

Both food web biomagnification and trophic dilution hypotheses have been used to explain interlake differences in contaminant accumulation in lower and higher trophic level species. Berglund et al. (8) found a negative relationship between lake trophic status and OC concentrations in phytoplankton. They concluded that nutrient stressed phytoplankton had higher lipid content, and this explained the

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higher OC concentrations in oligotrophic phytoplankton. The predominance of the microbial food web in oligotrophic water bodies has also been suggested as an important contributor to food web biomagnification. For example, in oligotrophic lakes such as Lake Superior where the plankton includes a high proportion of very small bacteria, it has been suggested that micrograzers including protozoans and mixotrophic phytoplankton create an extra trophic level in the food web (9, 10). Alternatively, other studies have implicated trophic dilution as a major factor affecting food web bioaccumulation in productive systems. Taylor et al. (11) reported that net plankton in low nutrient lakes had higher concentrations of OCs compared to nutrient-enriched waterbodies, a phenomenon they attributed to biomass dilution. Likewise Swackhamer and Skoglund (12) demonstrated that growth dilution depresses chemical accumulation in phytoplankton under high growth conditions in laboratory cultures.

Unfortunately, in spite of these advances in understanding the factors that influence the biomagnification of contaminants, attempts to predict variability in PCB concentration in lake trout using a single factor such as food chain length or lake trophy have not been successful (1). Although lake trout are top predators, they are also omnivorous and will feed on different prey items depending not only on their ontogenetic development but also on environmental conditions in the water body (13).

An important component rarely considered in contaminant bioaccumulation studies is the habitat volume available to lake trout and how this can vary over seasonal cycles and between systems that differ in trophic status. Among-system differences in habitat volume can alter the composition of fish's diet (proportion of littoral versus pelagic organisms) and subsequently influence chemical exposures via diet and food web transfer. In addition to food web effects, alterations in habitat volume can also have consequences to fish growth that can, in turn, modulate growth dilution effects. Lake trout metabolism is negatively affected at dissolved oxygen (DO) concentrations less than 7 mg/L (14), and trout will avoid habitat where DO falls below that concentration. Thus, during the cooler months of the year when lakes are not thermally stratified, trout move freely throughout the well oxygenated water column and can feed in both the shallow littoral zone (the nearshore part of a lake shallow enough to support benthic algal photosynthesis) as well as in the deep pelagic zone (the open water part of a lake). When lakes warm in the summer, trout are generally restricted to deep, cool, well oxygenated, hypolimnetic waters (the stratum of water below the thermocline).

Kidd et al. (15) have shown that fish feeding in littoral communities tend to have lower PCBs than those feeding pelagically, likely a result of trophic dilution experienced in the generally much more productive littoral portion of the food web. At the southern extent of the range of lake trout where summers are warmer and longer, lake trout will be restricted for longer periods to feeding only in deep, pelagic waters and have reduced access to productive littoral areas. Under nutrient poor conditions, it is thus predicted that lake trout which have strong diet connectivity with littoral habitats will exhibit lower bioaccumulation of OCs, and this effect will manifest itself as a latitudinal gradient. However if the lake is even moderately nutrient enriched (i.e., total phosphorus (TP) > 10 µg/L), decomposition of sedimenting algal-derived organic matter can reduce hypolimnetic oxygen concentration below that suitable for lake trout (14). If the hypolimnion is not well oxygenated as a result of nutrient enrichment and/or there is deeper-than-normal thermal stratification as a result of warmer temperatures, lake trout preferred habitat will be reduced and lake trout metabolic activity and feeding behavior will be affected, causing a reduction of gross growth efficiencies of fish (14). In this

case, trophic enrichment could lead to lower growth efficiencies of cold water species such as lake trout and enhance concentrations of OCs in top piscivores. Thus, the feeding opportunities for lake trout in water bodies that differ in the quality and extent of favorable lake trout habitat may be a factor confounding the relationships among bioaccumulation of OCs, food chain length, and lake trophic status.

The survey data span much of the entire natural latitudinal range of lake trout and includes waterbodies spanning a wide range of lake area and depth as well as a range of trophic enrichment. Lake trout feeding behavior is expected to be variable across such a wide range of conditions. Thus, the survey data can be used not only to test existing hypotheses regarding OC concentrations, food chain length, and trophic enrichment but also to develop and test the hypothesis that variability in lake trout habitat volume will affect OC bioaccumulation. We hypothesize that lake trout with access to a wide range of habitat for feeding, including both littoral and pelagic zones, will have lower concentrations of OCs than lake trout that are restricted to a pelagic food web. However, under conditions of trophic enrichment, the above pattern may be attenuated or reversed. The data from this large survey are used to test these hypotheses; in particular we use the stable isotope of carbon ( $\delta^{13}\text{C}$ ) as a metric of littoral feeding as did Kidd et al. (15). Benthic algae grow in a boundary layer that restricts  $\text{CO}_2$  diffusion to photosynthesizing cells while phytoplankton grow as individual cells or colonies that generally have a negligible boundary layer. Consequently benthic algal isotopic fractionation is reduced compared to pelagic phytoplankton, resulting in a different stable isotope signature that can be used to trace benthic and pelagic primary productivity supporting higher trophic levels (16). We also used stable isotopes of nitrogen ( $\delta^{15}\text{N}$ ) to determine the length of the food chain supporting lake trout. Other lake characteristics including area, depth, TP, and phytoplankton composition were examined to explore the food chain length, trophic enrichment, and feeding behavior hypotheses.

## Materials and Methods

**Study Area.** Twenty three lakes ranging from as far north as Lake Athabasca in western Canada (58.5°N, 111.0°W) to Cayuga Lake in the northeastern U.S (42.8°N, 77.6°W) were sampled during the years 1998 to 2001 (Table 1 and Supporting Information Table S1). The lakes ranged in surface area from 11 to 82 170 km<sup>2</sup> and had mean depths that ranged from 5 to 148 m (Table S1). They also extended from urban- and industrial-influenced catchments in the southern part of the study area to remote areas where anthropogenic impacts are minimal except possibly from regional and global atmospheric effects.

**Field Sampling Procedures.** Most of the lakes were sampled once during the middle of the open water season (at the end of June or early July). Exceptions are four lakes in northwestern Ontario (Sandybeach, Thunder, Paguchi, Eva) and Lake Simcoe and Opeongo Lake in southern Ontario (Table S1, Supporting Information) that were sampled 3 times in the open water season and Lake Superior which was sampled in May and in July 1998 (17). Cayuga and Seneca Lakes and Lake Champlain in the northeastern US were sampled only once in May. Further details on sampling are provided in Houde et al. (1) and in Supporting Information. Lake trout were obtained by gill netting programs. Bulk zooplankton ( $n = 2-5/\text{lake}$ ) were collected using vertical water column tows of a 153 µm net (1). Whole fish were then homogenized by the Department of Fisheries and Oceans (DFO) Burlington, Ontario laboratory, and homogenates were stored at -20 °C. All samples, except for those from Lake Superior and Siskiwit Lake, were sent to the Great Lakes Institute for Environmental Research (GLIER) for analysis of

**TABLE 1. Location, Area, Number (n) of Individual Trout or Zooplankton Net Hauls, Arithmetic Means of Weight, Length, Total PCB ( $\Sigma$ PCB) Expressed per Lipid Weight (lw) and Wet Weight (ww), and  $\delta^{15}\text{N}$  (‰),  $\delta^{13}\text{C}$  (‰), and % Lipid for Lake Trout and Zooplankton (where available) in Each Lake<sup>a</sup>**

lake	lat.	long.	area, Km <sup>2</sup>	n	weight, g	length, mm	lake trout					zooplankton				
							$\Sigma\text{PCB} \pm \text{SD}$ (ng g lw <sup>-1</sup> )	$\Sigma\text{PCB}$ (ng g ww <sup>-1</sup> )	$\delta^{15}\text{N}$ , ‰	$\delta^{13}\text{C}$ , ‰	lipid, %	n	$\Sigma\text{PCB}$ (ng g ww <sup>-1</sup> )	$\delta^{15}\text{N}$ , ‰	$\delta^{13}\text{C}$ , ‰	FCL, ‰
Athabasca	58.5	111.0	7900	19	4560	797	628 ± 127	63	12.0	-27.2	11.4	4	1.3	2.7	-31.8	9.3
Wollaston	58.0	103.0	2062	20	1622	537	162 ± 16	16	12.8	-26.7	10.3	3	3.7	5.4	-31.4	7.4
Namur	57.5	111.5	42	18	2396	625	100 ± 20	9	13.4	-23.1	9.8	2	2.9	7.2	-30.2	6.2
Reindeer	57.0	102.0	5569	20	1817	574	668 ± 228	35	11.8	-27.3	6.9	3	6.6	3.2	-31.0	8.6
La Ronge	55.3	110.0	25	18	3416	654	555 ± 139	35	13.9	-26.4	5.8	3	2.4	6.1	-29.8	7.9
Grist	55.3	105.0	1178	18	2614	614	225 ± 63	15	12.1	-28.4	7.9	6	3.3	4.1	-31.3	8.0
Cold	54.5	110.0	373	16	4714	733	785 ± 233	75	12.9	-26.3	11.5	2	4.8	3.3	-27.3	9.6
Kingsmere	54.0	106.0	47	13	2521	652	278 ± 63	18	14.1	-24.8	7.7	3	5.6	6.7	-27.6	7.4
Harp	55.0	61.8	125	5	3622	700	713 ± 129	41	9.5	-23.6	5.7	na	na	na	na	na
Snegamook	54.8	61.5	133	5	1911	588	642 ± 185	20	12.0	-21.5	3.6	na	na	na	na	na
Seal	54.3	61.3	125	5	2192	374	571 ± 283	21	11.0	-21.2	5.9	na	na	na	na	na
Shabogamo	53.2	66.5	207	5	2105	616	732 ± 152	26	11.1	-22.4	4.1	na	na	na	na	na
Sandybeach	49.8	92.3	38	15	3066	657	1350 ± 649	132	11.9	-29.4	14.4	3	0.1	3.2	-31.6	8.7
Thunder	49.8	92.6	11	15	1902	591	758 ± 78	59	11.8	-28.1	8.5	3	0.1	4.2	-31.0	7.6
Paguchi	49.5	91.5	25	14	2877	680	1036 ± 262	58	12.0	-25.6	7.1	3	0.1	3.4	-29.7	8.6
Eva	48.6	91.2	17	15	2852	637	861 ± 307	89	10.4	-26.8	13.9	3	2.0	1.9	-30.3	8.5
Siskiwit	48.0	89.0	17	20	2015	574	2928 ± 494	124	10.5	-25.2	4.9	na	2.2	-28.9	8.3 <sup>b</sup>	
Superior	47.0	90.5	82170	75	2370	646	5772 ± 614	587	9.6	-24.9	11.5	7	15.0	4.8	-30.4	4.9
Opeongo	45.6	78.3	59	20	1504	469	746 ± 87	67	9.9	-26.9	10.9	3	0.5	2.9	-28.6	7.0
Simcoe	44.5	73.5	1127	15	1927	530	1220 ± 153	214	16.2	-27.5	18.1	4	10.0	9.2	-29.9	7.0
Champlain	44.5	79.5	725	21	4518	422	4519 ± 612	356	17.1	-24.8	7.7	5	2.2	5.2	-35.0	11.9
Cayuga	42.8	76.6	172	10	496	328	2893 ± 406	99	17.8	-23.8	4.0	5	4.3	16.5	-30.2	2.0
Seneca	42.8	77.0	175	7	1834	552	2708 ± 269	263	17.5	-23.8	9.9	5	3.9	12.0	-31.3	5.6

<sup>a</sup> Food chain length (FCL) calculated as the difference between the  $\delta^{15}\text{N}$  of lake trout and zooplankton for each lake. <sup>b</sup> The zooplankton from Siskiwit Lake were sampled in 2005 (D. Muir, unpublished data).

PCBs. Samples from Lake Superior and Siskiwit Lake were analyzed by the National Laboratory for Environmental Testing (NLET), Burlington, Ontario.

**Analysis of PCBs.** PCBs were measured on whole fish homogenates using the method of Norstrom and Won (18) with minor modifications. Further details on extraction and cleanup steps are provided in Supporting Information. Extracts were analyzed for 57 PCB congeners (includes coeluters) by capillary gas chromatography with electron capture detection. Both laboratories analyzed the same suite of PCB congeners. The analysis included the use of blanks every six samples spiked with TCMX and PCB-209. Blanks had uniformly low or nondetectable PCBs. Method detection limits based on S/N (sample to noise ratio) = 3 ranged from about 0.01 ng g<sup>-1</sup> to 0.05 ng g<sup>-1</sup> for individual congeners in 5 g (wet weight) samples. Both laboratories analyzed a standard reference material (cod liver 1588a) from the National Institute of Standards and Technology (NIST, Gaithersburg MD) after every 20 samples. PCB congeners were generally within ± 25% of certified values.

**Lipid and Moisture Content.** The percentage (%) of extractable lipid was determined gravimetrically for each sample using a fraction of the tissue extract. Percent moisture was determined on a subsample of homogenized tissue by drying to constant weight.

**Stable Isotope Analysis.** Stable nitrogen and carbon isotope ratios were determined on fish muscle at the Environment Canada (Saskatoon) stable isotopes laboratory using the methods described in Houde et al. (1). Food chain length (FCL) was estimated by subtracting the mean  $\delta^{15}\text{N}$  of bulk zooplankton from the mean  $\delta^{15}\text{N}$  of the lake trout from each lake. Stable isotope data for zooplankton from Lake Siskiwit were obtained from a more recent study (Muir, D., unpublished data 2006), and these were used to calculate a FCL for Siskiwit. The stable isotope analysis also returned data on the C and N content of each sample for 19 of the lakes. These data were used to correct the  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  of the fish muscle

for lipid content using the algorithm of Post et al. (19) ( $\Delta \delta^{13}\text{C} = -3.32 + 0.99 \times \text{C:N}$ ). Lakes that did not have C and N content were Superior, Champlain, Siskiwit, and Grist.

**Water Chemistry and Phytoplankton Enumeration.** Total phosphorus (TP) was measured in unfiltered lake water samples. Water was passed through a glass fiber filter with a nominal pore size of 0.75  $\mu\text{m}$ , and these filters were extracted for chlorophyll *a* (20). Unfiltered water samples were fixed with Acid Lugols (final concentration 1%) for phytoplankton preservation. Phytoplankton species were identified and enumerated using an inverted microscope following the Utermohl method (21). Biomass of phytoplankton was calculated based on measurements of linear dimensions using appropriate volume formulas and assuming a specific gravity of 1.0.

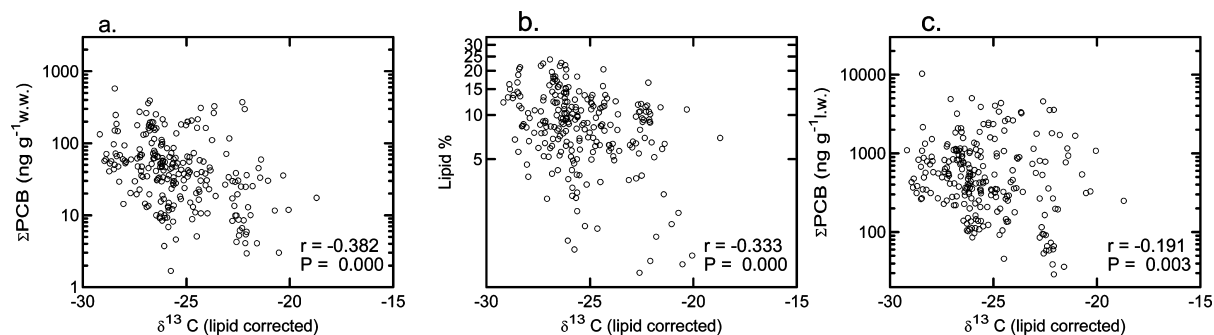
**Statistical Analysis.** Statistical analyses (Pearson's product moment correlation, simple linear and multiple linear regression) were carried out using Systat Version 9 (Systat Software, Inc., Point Richmond, CA, 2004). Variables that were not normally distributed were transformed by taking the logarithm base 10.

## Results and Discussion

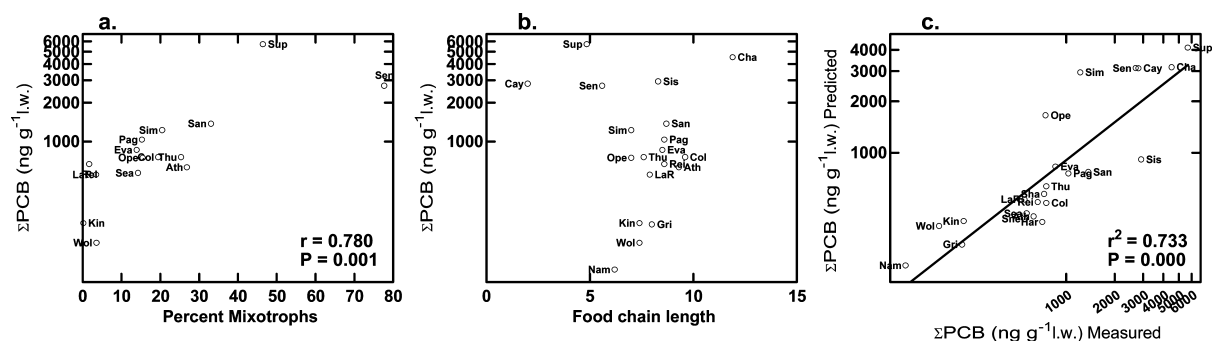
**$\Sigma\text{PCBs}$  in Lake Trout and  $\delta^{13}\text{C}$ .** We found a statistically significant negative correlation between the lipid-corrected  $\delta^{13}\text{C}$  signature and  $\Sigma\text{PCBs}$  in lake trout (both wet weight and lipid normalized; Figure 1a,b). This supports the hypothesis that lake trout supported more by carbon derived from littoral benthic algae, as indicated by their more positive  $\delta^{13}\text{C}$  values, have lower concentrations of PCBs compared to lake trout relying more upon a food web based upon pelagic phytoplankton. Percent lipid in lake trout was also negatively correlated to lipid-corrected  $\delta^{13}\text{C}$  (Figure 1c), indicating that lake trout with a pelagic-based food web were more lipid rich than lake trout with a littoral-based food web.

**$\Sigma\text{PCBs}$  in Lake Trout and the Microbial Food Web.** PCBs enter the aquatic food web mainly at the level of the smallest





**FIGURE 1.** (a)  $\Sigma\text{PCB}$  ( $\text{ng g}^{-1}$  wet wt) versus  $\delta^{13}\text{C}$  (‰) (lipid corrected) for each fish, Pearson correlation coefficient  $r = -0.382$ ,  $P = 0.0001$ . (b) Percent lipid for individual lake trout versus  $\delta^{13}\text{C}$  (‰) (lipid corrected) for each fish, Pearson correlation coefficient  $r = -0.333$ ,  $P = 0.0001$ . (c)  $\Sigma\text{PCB}$  ( $\text{ng g}^{-1}$  lipid wt) for individual lake trout versus  $\delta^{13}\text{C}$  (‰) (lipid corrected) for each fish, Pearson correlation coefficient  $r = -0.191$ ,  $P = 0.003$ . Four lakes were excluded because data were not available to correct the  $\delta^{13}\text{C}$  for lipid.



**FIGURE 2.** (a) Mean  $\Sigma\text{PCB}$  in lake trout for each lake versus percent of total phytoplankton that are mixotrophs (calculated by summing percent Cryptophyta and percent Dinophyta of total phytoplankton biomass), Pearson product moment correlation coefficient  $r = 0.780$ ,  $P = 0.001$ . (b) Mean  $\Sigma\text{PCB}$  in lake trout for each lake versus food chain length (FCL; calculated by subtracting the mean  $\delta^{15}\text{N}$  of zooplankton from the mean  $\delta^{15}\text{N}$  of lake trout for each lake), no statistically significant correlation. (c) Predicted versus measured mean  $\Sigma\text{PCB}$  in lake trout for each lake based on a multiple linear regression between  $\log_{10}\Sigma\text{PCB}$  in lake trout and  $\log_{10}$  lake area and latitude for each lake. The equation for the line of best fit was  $\log_{10}\Sigma\text{PCB} = 6.222 + 0.158 \times \log_{10}\text{area} - 0.073 \times \text{latitude}$ ,  $r^2 = 0.733$ ,  $P_{(\text{latitude})} = 0.000$ ,  $P_{(\text{area})} = 0.007$ .

particles (bacteria and plankton). Oligotrophic waterbodies have proportionally more bacteria and phytoplankton that are in the picoplankton size range ( $<2 \mu\text{m}$  diameter) compared to nutrient-enriched water bodies (9). As well as being smaller in size, bacteria and phytoplankton in oligotrophic pelagic water bodies have been reported to have lower growth rates and contain higher concentrations of lipids which are deposited as a storage product in phytoplankton when nitrogen is unavailable to synthesize proteins (22). Berglund et al. (8) reported that phytoplankton in oligotrophic lakes in their study of 18 lakes in Sweden had higher concentrations of lipid and PCBs than the eutrophic lakes in their study. Picoplankton in oligotrophic lakes are too small to be efficiently grazed by typical lake zooplankton (Cladocerans and Copepods) and are instead the prey of micrograzers such as ciliates and flagellated protozoa. Typical micrograzers in oligotrophic water bodies are “mixotrophs” such as the Dinophyta and Cryptophyta which are flagellated protozoa that have photosynthetic pigments but can supplement their energy needs by phagocytosis of picoplankton. These organisms (picoplankton and micrograzers) comprise the microbial food web in oligotrophic water bodies. The microbial food web has been suggested as a potentially important pathway in the incorporation of organic contaminants into aquatic food webs both as a result of high lipid content (10) and because of the introduction of an additional trophic level into pelagic food webs (9). In typical food web studies of biomagnifications leading to lake trout, the lowest trophic level considered is usually the zooplankton so potentially important trophic transfers within the microbial food web are often missed. In our study lakes with a high

proportions of mixotrophs, lake trout had higher concentrations of  $\Sigma\text{PCBs}$  (Figure 2a). The correlation of these two variables was surprisingly strong ( $r = 0.780$ ;  $P = 0.001$ ), demonstrating the possible importance of the microbial food web to PCB bioaccumulation in lake trout.

**$\Sigma\text{PCBs}$  in Lake Trout and Food Chain Length.** Food chain length has been demonstrated to be an important factor determining the biomagnification of OCs in lake trout (2, 3); however, it is challenging to quantify the complex linkages that exist in most waterbodies especially with top predators such as lake trout that are actually omnivorous and will feed at different trophic levels, inhabit variable habitats depending on their life history stage, and have inherent within- and among-population differences (13). There is no doubt that PCBs increase with trophic transfers, but defining the basal signal and all the trophic transfers is problematic especially when littoral prey may be integrated into trout diets. In our study the food chain length estimated from the  $\delta^{15}\text{N}$  signature for zooplankton and lake trout in the survey lakes was not correlated to the  $\Sigma\text{PCB}$  concentrations in lake trout (Figure 2b) suggesting that littoral prey are also likely as important or more important than zooplankton in the food web supporting lake trout.

**$\Sigma\text{PCBs}$  in Lake Trout and Trophic Enrichment.** Several studies have demonstrated a negative relationship between lake trophic status and PCBs in different aquatic food web organisms including plankton (11), northern pike (4, 5) and profundal chironomids (23). Total phosphorus (Table S1, Supporting Information) was not correlated to  $\Sigma\text{PCBs}$  in lake trout in the survey lakes ( $r = -0.362$ ,  $P = 0.117$ ). Lake trout are a sentinel species for good water quality because of their

inability to tolerate oxygen concentrations below 7 mg L<sup>-1</sup>, and their optimum temperature for growth is 12.7 °C (14). Lakes with TP concentrations much above 10 µg L<sup>-1</sup> that experience strong summer thermal stratification are not likely to have sustainable lake trout populations because of restricted suitable habitat (14). Survey lakes with high TP concentrations were all located at higher latitudes where prolonged thermal stratification does not presently occur. Thus, although trophic enrichment may explain the variability of PCBs in some food web organisms, the negative effect of TP on DO concentration and as a result available lake trout habitat may confound any direct relationship between lake nutrient enrichment and lake trout PCBs in a geographically extensive study such as ours.

**A Model for Predicting Lake Trout PCB Concentration Based on Proximate Variables for Optimal Lake Trout Habitat: Lake Area and Latitude.** The correlations observed in this large survey may be relevant to understanding the variability of PCBs in lake trout. The negative correlation between ΣPCBs and δ<sup>13</sup>C in lake trout as well as the positive correlation between ΣPCBs in lake trout and % of lake phytoplankton that are mixotrophs both support the hypothesis that lake trout with access to both littoral and pelagic zones will have lower PCBs than lake trout that are restricted to a pelagic food web. Lake area can be a good predictor of littoral habitat. On average, the larger the lake, the greater the proportion of deeper water and therefore the smaller the proportion of the total lake area that is shallow enough to support benthic algal photosynthesis taken as defining the limit of the littoral (24). However, lake area alone was not directly correlated to lake trout ΣPCB concentration because the littoral zones of some of the survey lakes are not suitable trout habitat because of their warm summer epilimnetic temperatures. Thus, lakes in this study that are located at the southern latitudes would be expected to have proportionally less preferred trout feeding habitat than lakes of a similar size located at the northern latitudes where summer thermal stratification is much less pronounced and of shorter duration. Strong and prolonged, summer thermal stratification will reduce preferred lake trout habitat to the deepest areas of the lake. Lake latitude in this large survey provides a proximate estimate of a wide range of epilimnetic temperatures and can be used in combination with lake area to construct a simple statistical model to predict littoral habitat suitable for lake trout feeding. A multiple linear regression combining lake area(log10) and latitude predicted well ΣPCB in lake trout in all the survey lakes (Figure 2c).

**Mechanisms Underlying the Feeding Behavior Hypothesis.** Lake trout are omnivores and will feed opportunistically at different trophic levels (13), and this confounds efforts to relate ΣPCB concentrations with food chain length based on zooplankton only. One explanation for the correlations between PCB concentrations in lake trout and the % mixotrophs observed in the survey lakes is that the pelagic microbial food web exacerbates the biomagnification of PCBs in pelagic oligotrophic lakes as proposed by Hudson et al. (10). Another explanation is that lake trout that can readily access littoral as well as pelagic food webs may have higher growth rates compared to lake trout in lakes where a pelagic diet dominates. Low growth rates were correlated with higher ΣPCBs in a variety of trout in a study of eight lakes in the Canadian rocky mountains (25). Similarly, Trudel and Rasmussen (7) suggest that there is a general negative relationship between growth rates and Hg contamination in fish. Lake trout inhabiting the lakes in the southern range of this survey are at the southernmost limit of their natural distribution, and species at their distribution limit are, by definition, likely to be growing at less-than-optimum rates. Some of the lakes that had high concentrations of ΣPCBs in lake trout are located in the southeastern part of Canada and

the northeastern USA in close proximity to major population centers in comparison to the high latitude survey lakes and, as a result, may have been influenced by local industrial sources of PCBs (e.g. (26)). However when the ΣPCBs in zooplankton from all the lakes are compared (Table 1), only Lake Simcoe located 100 km north of the city of Toronto exhibited PCB concentrations outside the range seen in lakes that are remote from population centers. Highest zooplankton PCB concentrations over all our survey lakes were measured in zooplankton taken from Lake Superior which has no known significant point source PCB inputs (27).

The data in this survey were collected with the goal of increasing our ability to predict the risk of OC contaminant exposure to human consumers. The survey spanned a broad range of lake size and latitude which combined to provide lakes with different proportions of accessible littoral and pelagic zones. Area and latitude may provide simple surrogates to estimate this proportion and the availability of littoral feeding habitat. These simple metrics were surprisingly good predictors of ΣPCBs in lake trout in the survey lakes. To further substantiate the underlying mechanism for this relationship and determine if the model has utility for lakes outside of those in this survey will require a research design that specifically includes direct estimates of littoral and pelagic feeding habitats available for lake trout. Until such further studies are performed, the lake area and latitude are two readily available, simple metrics that may improve our ability to predict ΣPCBs in lake trout in areas where contaminant data may be lacking.

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## Supporting Information Available

A map showing the lake locations (Figure S1), and a table giving more specific details about water sampling and analysis as well information about fish sample storage and handling and extraction and cleanup steps. The information is available free of charge via the Internet at <http://pubs.acs.org>.

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