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1 PCB Food Web Dynamics Quantify Nutrient and 2 Energy Flow in Aquatic Ecosystems

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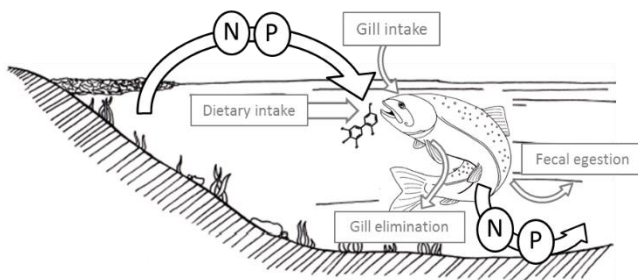
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9 KEYWORDS. Persistent Organic Pollutants as Chemical Tracers, Energy and Nutrient Flow,
10 Food webs, Lake Huron, Lake Trout.

11



12

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14 ABSTRACT. Measuring *in situ* nutrient and energy flows in spatially and temporally complex
15 aquatic ecosystems represents a major ecological challenge. Food web structure, energy and
16 nutrient budgets are difficult to measure, and it is becoming more important to quantify both
17 energy and nutrient flow to determine how food web processes and structure are being modified
18 by multiple stressors. We propose that polychlorinated biphenyl (PCB) congeners represent an
19 ideal tracer to quantify *in situ* energy and nutrient flow between trophic levels. Here, we
20 demonstrate how an understanding of PCB congener bioaccumulation dynamics provides
21 multiple direct measurements of energy and nutrient flow in aquatic food webs. To demonstrate
22 this novel approach, we quantified nitrogen (N), phosphorus (P) and caloric turnover rates for
23 Lake Huron lake trout, and reveal how these processes are regulated by both growth rate and fish
24 life history. Although minimal nutrient recycling was observed in young growing fish, slow
25 growing, older lake trout (> 5 yr) recycled an average of 482 Tonnes·yr⁻¹ of N, 45 Tonnes·yr⁻¹ of
26 P and assimilated 22 TJ yr⁻¹ of energy. Compared to total P loading rates of 590 Tonnes·yr⁻¹, the
27 recycling of primarily bioavailable nutrients by fish plays an important role regulating the
28 nutrient states of oligotrophic lakes.

29

30 **Introduction:**

31 Anthropogenic climate change, chemical pollution, nutrient loading, and habitat degradation are
32 some of the most critical factors simultaneously affecting aquatic ecosystems. These multiple
33 stressors can act synergistically resulting in a myriad of unpredictable responses causing aquatic
34 food webs to be irreparably altered. These perturbations are often enhanced by invasive species¹
35 and can lead to declining fish abundances and condition, changes in reproductive success², and
36 potentially lead to food web contractions and regime shifts³⁻⁴. While it is possible to estimate the

37 effects of environmental and anthropogenic stressors on fish ecology and physiology⁵⁻⁸, as well
38 as identify changes in resource exploitation by fish through gut contents and stable isotope
39 analyses⁹⁻¹⁰, there are currently no methods to directly measure individual-based nutrient and
40 energy flows in food webs¹¹. To achieve such measurements it is essential to be able to quantify
41 individual, *in situ* fish consumption rates.

42

43 Quantifying fish consumption rates is critical to understanding food web dynamics because fish
44 have been identified as both sinks¹² and vectors of essential nutrients and energy transport¹³. Fish
45 communities play a critical role regulating the transport and fate of nutrients in aquatic
46 ecosystems as they are an important part of the overall nutrient pool¹². Understanding nutrient
47 cycling and transport in biota is vital for predicting ecosystem responses to issues such as
48 eutrophication, species invasions and setting fisheries quotas.

49

50 The importance of quantifying fish consumption rates has long been recognized. Species-specific
51 bioenergetics models incorporating growth, metabolic and waste processes have been developed
52 for a suite of aquatic species, both juvenile and adult, starting with bluegills in 1974¹⁴. As of
53 2000, papers being published a year on bioenergetics modeling have increased rapidly¹⁵. These
54 models rely on both laboratory and field data to estimate average consumption rates of different
55 age cohorts of fish¹⁶. Validations of bioenergetics models, however, generally tend to
56 demonstrate a poor fit between model predicted and lab or field data¹⁵. Thus, efforts to complete
57 hypothesis based testing of individual model parameters will serve to improve model structure
58 and performance¹⁵. Further, bioenergetics modelling efforts tend towards population based
59 predictions of predator demand rather than assessing individual based responses to ecosystem

60 perturbations^{11,17}. Similarly, the mass-balance approach is incorporated into Ecopath, Ecosim,
61 and Ecospace model estimates of consumption and trophic interactions¹⁸. Again, however, these
62 are population-wide estimates with a Bayesian resampling approach to estimating uncertainty¹⁸
63 without tracking individual responses within populations.

64

65 Chemical tracer mass balance approaches using Mercury and Cesium^{19,20} have also provided
66 alternative methods of calculating fish consumption rates. However, these approaches are limited
67 as singular metrics of dietary consumption and do not necessarily track similar environmental
68 processes. As food webs are temporally and spatially complex²¹, there is need for a method that
69 directly quantifies fish consumption rates and provides statistical power through multiple
70 repeatable metrics while tracking fish bioenergetics responses to environmental change.

71

72 This study proposes the use of persistent organic pollutants (POPs) as metrics to quantify
73 nutrient and energy flow within aquatic food webs. POPs, such as polychlorinated biphenyls
74 (PCBs), are globally ubiquitous pollutants that span a wide range of chemical hydrophobicities²²
75 and many congeners are highly resistant to environmental and biological degradation^{23,24}. These
76 chemical properties regulate their bioaccumulation in fish²², with the fraction of accumulation
77 from dietary versus aqueous sources ranging from 70% for less hydrophobic congeners ($\log K_{ow}$
78 ≤ 6.5) to 100% for increasingly hydrophobic congeners²⁵. Furthermore, elimination rates of the
79 more hydrophobic congeners ($\log K_{ow} > 6.5$) are very low resulting in long chemical half-lives
80 with respect to the life span of fish²⁶. Therefore, the body burden of super-hydrophobic
81 congeners (those with $\log K_{ow} > 6.5$) represents a proxy of the total amount of food a fish has
82 consumed in its lifetime. Thus, the kinetics of PCB congeners in aquatic consumers have the

83 potential to provide an *in-situ* repeatable method to quantify the total mass of food consumed
84 over the duration of a fish's life. Therefore persistent hydrophobic chemicals can be used as
85 multiple independent markers to directly measure nutrient and energy flow through the upper
86 trophic levels of food webs.

87

88 This study investigates (1) the use of PCB bioaccumulation dynamics to develop quantitative
89 measurements of both nutrient uptake and loss as well as the energy consumed over a fish's
90 lifespan as compared with previous model prediction methods, and, (2) quantify the relative
91 importance of fish for nutrient recycling in aquatic systems.

92

93 **Experimental:**

94 *Sample Processing*

95 Lake trout (*Salvelinus namayacush*; n = 195), rainbow smelt (*Osmerus mordax*; n = 34), round
96 goby (*Neogobius melanostomus*; n = 27), alewife (*Alosa pseudoharengus*; n = 8), and whitefish
97 species (*Coregonus artedi* and *Coregonus hoyi*; n = 54) were collected from the Canadian waters
98 of the Main Basin, Georgian Bay, and North Channel regions of Lake Huron throughout the
99 summers of 2010, 2011, and 2012. Fish were collected by overnight gill nets set by the Upper
100 Great Lakes Unit of the Ontario Ministry of Natural Resources. At each site a total of 18 nets
101 were set, and each net consisted of 15m panel of (32mm) mesh and a 25 m panel of 38 mm mesh
102 followed by 50m panels of 51, 64, 76, 89, 102, 114 and 127 mm meshes. Length and weight
103 measurements were taken, sex was determined, and otoliths and gut tracts removed, then samples

104 were placed on dry ice. Frozen samples were transported back to the Great Lakes Institute for
105 Environmental Research (GLIER) and stored at -25 °C until processing.
106
107 Sample processing included homogenization of whole fish samples and the measurement of
108 whole body lipid contents. Moisture contents were obtained by drying approximately 0.5 g of
109 homogenate for 24 hours. Individual PCB congener concentrations and lipid contents were
110 determined using the microextraction method described by Daley et al.²⁷ In brief, 0.5 g of whole
111 body homogenate was ground with 15 g of sodium sulfate using a glass mortar and pestle, and
112 then wet packed into a glass chromatography column containing 15 mL of a 50 : 50
113 hexane(Hex):Dichloromethane(DCM) (v/v) extraction mixture, along with 35 ng of a PCB 34
114 extraction performance recovery standard. After solvent elution, an additional 15 mL of
115 Hex:DCM was added to extract the homogenate. Sample extracts were then evaporated under
116 vacuum to ~2 mL, and then diluted to 10 mL with hexane in a volumetric flask. Neutral lipid
117 content was determined gravimetrically using 1 mL of this solution²⁸. Six grams of Florisil
118 topped with approximately 1 g of sodium sulfate was then used for sample clean up with 50 mL
119 hexane wash. The final extract was evaporated under vacuum to <1 mL and brought to a final
120 volume of 1 mL with iso-octane for analysis by gas chromatography-electron capture detector
121 (GC-ECD)²⁹. All samples were analyzed for the following PCB congeners (IUPAC #): 18/19,
122 31/28, 33, 52, 49, 44, 74, 70, 95, 101, 99, 87, 110, 151/82, 149, 118, 153, 105/132, 138, 158,
123 187, 183, 128, 177, 156/171, 180, 191, 170, 199, 195/208, 194, 205, 206, and 209.
124
125 A method blank and an in house reference tissue homogenate of Detroit River carp were
126 extracted simultaneously as quality assurance for every set of 6 sample extractions. All sum PCB

127 concentrations quantified in the reference tissue were in compliance with the Great Lakes
128 Institute for Environmental Research organic analytical laboratory's quality assurance guidelines
129 (mean \pm 2 standard deviations (SD)). Recoveries of the internal standard averaged 89% \pm 1%
130 (Standard Error; n = 195), and sample concentrations were not recovery corrected.

131

132 Gut content analyses were performed using the Ontario Ministry of Natural Resources protocol.
133 Contents of the gut were removed and food items were identified by using calcified structures
134 such as otoliths that are generally resistant to digestion. Prey items were then enumerated and
135 species proportions calculated using the number of each prey item obtained.

136

137 *Consumption Estimates*

138 One of the major assumptions of the PCB approach proposed in this paper is that
139 biotransformation and depuration mechanisms do not contribute significantly to the whole body
140 elimination of the super hydrophobic congeners being used as food web tracers. All
141 metabolizable congeners were removed from the analyses to ensure no biotransformation was
142 occurring³⁴. A review of literature estimates of elimination rates of PCB congeners by fish
143 demonstrated that congeners with a logK_{OW} greater than 6.5 did not demonstrate significant
144 elimination (see Fig. 1.³⁵⁻⁴⁰). Furthermore, the non-significant elimination rates for these
145 congeners resulted in a t₉₅, or time to steady state, vastly exceeding the life expectancy of the
146 organism. Therefore, the total body burden of any one of these super-hydrophobic congeners
147 represents an individual's lifetime of food consumption.

148 Amounts of nitrogen and phosphorus recycled were calculated using the concentrations of PCB
149 congeners in the whole fish sample. First, concentration data were converted to mass values. The
150 amount of each PCB congener ingested was then calculated according to equation (1).

$$151 \quad (1) \text{ Amount } PCB_i \text{ ingested [ng]} = m_{PCB_i} \times f_{PCB_i} \times (E_{d,PCB_i})^{-1}$$

152 where PCB_i is the chosen PCB congener, m_{PCB_i} is the mass of PCB_i in the consumer (in ng), E_{d,PCB_i}
153 PCB_i is the chemical assimilation efficiency for PCB_i in food (prey), and f_{PCB_i} is the fraction of
154 PCB_i mass which is accumulated through dietary uptake as opposed to gill intake (Table S1).

155 Using the results from the gut content analyses (Figure S2), the number of smelt, round goby and
156 other fish consumed were calculated according to equation (2)

$$157 \quad (2) \text{ number of fish consumed} = \left(\frac{\text{Amount of } PCB_i \text{ ingested}}{PCB_{i,prey}} \right) \times p_{prey}$$

158 where amount of PCB_i ingested is calculated using equation (1), $PCB_{i,prey}$ is the average mass
159 (ng) of PCB_i in the prey species (smelt, round goby, or other), and p_{prey} is the proportion of diet
160 made up of that prey species (where proportion of smelt, round goby, and other fish consumed
161 by lake trout were, respectively, 0.88, 0.075, and 0.045 for the Main Basin of Lake Huron, 0.77,
162 0.1, and 0.13 for the Georgian Bay, and 0.98, 0.015, and 0.005 for the North Channel).

163 Consumption estimates were then compared to consumption estimates presented in Stewart et
164 al.¹⁶ and Pazzia et al.²⁰ by extrapolating their estimates to include 13 year old fish using the
165 equations provided in their work.

166

167 *Nitrogen and Phosphorus Recycling Estimates*

168 Determination of nitrogen content was conducted in the Chemical Tracers laboratory at GLIER
169 using a continuous-flow isotope ratio mass spectrometer (Finnigan MAT Deltaplus; Thermo

170 Finnigan, San Jose, California) on freeze dried homogenates (Labconco co., Kansas City Missouri)
171 which had been ground using mortar and pestle, and then lipid extracted using chloroform-
172 methanol²⁹.

173

174 Total mg of nitrogen was subsequently calculated from dry weight (g) according to equations 3
175 and 4.

176 (3) $Dry\ weight[g] (dw) = ww - ww \left(\frac{\% moisture}{100} \right)$

177 (4) $N [mg] = \left(\left(\frac{\% N}{100} \right) \times dw \right) \times 1000$

178 where ww is the wet weight of the fish. The amount of phosphorus was then estimated using the
179 P:N ratio of 1:10.6 for wild caught fish³⁰. Total calories in each diet item were calculated using
180 fish energy densities³¹.

181

182 Using the calculated mass of N, the total amount of N ingested by each lake trout was
183 determined according to equation 5.

184 (5) $N\ ingested [mg] = \sum N_{prey} \times number\ of\ fish\ consumed_{prey}$

185 where N_{prey} is the average amount of nitrogen (mg) in the prey fish calculated using equation (4),
186 and the number of fish consumed_{prey} by a lake trout was estimated using equation (2). This was
187 then summed across all prey species. Both P and calories ingested were calculated in a similar
188 manner, where $P (mg)_{prey}$ was estimated using a P:N ratio of 1:10.6³⁰, and calories in prey fish
189 were calculated using energy densities². Finally, mass of N recycled was calculated according to
190 equation 6.

191 (6) $N\ recycled [mg] = N_{ingested} - N_{lake\ trout}$

192 where $N_{\text{lake trout}}$ (mg) was calculated using equation (4), and N_{ingested} (mg) was calculated using
193 equation (5). The mass of P recycled was calculated in a similar manner.

194

195 *Data analysis*

196 Growth rates of Lake Huron lake trout were calculated using the von Bertalanffy (VBL) growth
197 rate model comparing total length and age (equation 7)

198

$$199 \quad (7) \quad L_t = L_\infty \left(1 - e^{-k(t-t_0)}\right)$$

200

201 where t is lake trout age (yr), L_t is the total length (cm) of the fish at time t , L_∞ is the asymptotic
202 length (cm), and k the growth coefficient (yr^{-1}). The model was calculated using a value of $t_0 = 0$,
203 for the theoretical age at a total length of 0, an assumption validated for whitefish³², another
204 salmonid, and used for lake trout³³. Calculations of the VBL growth models were done using the
205 non-linear regression module of SYSTAT (SYSTAT 11). Multiple iterations were done to
206 achieve optimal model fit. The square of the correlation coefficients between observed and
207 predicted values were used to calculate the coefficient of determination (r^2) values for the VBL
208 growth models. Individual growth rates ($\% \cdot \text{year}^{-1}$) were obtained by equation 8.

209

$$210 \quad (8) \quad \text{Growth rate } (\% \cdot \text{year}^{-1}) = \left(\frac{w}{w_t}\right) \times \ln\left(\frac{w_t}{w_{t+1}}\right) \times 100$$

211 where w is the measured weight of the individual, w_t the von Bertalanffy predicted weight at age
212 t , and w_{t+1} the von Bertalanffy predicted weight at age $t+1$.

213

214 **Results & Discussion:**

215 A unique aspect of using PCBs as markers of individual consumption rates is their potential to
216 offer multiple, repeatable metrics for calculating consumption dynamics in individual fish. To
217 demonstrate the power of the PCB approach, we used an ANOVA to compare congener-specific
218 consumption estimates for the 15 most abundant congeners and found no significant differences
219 among the 9 congeners with $\log K_{ow} > 6.5$ ($p > 0.15$). This lack of significant differences
220 demonstrates that the measured consumption rates are not chemical dependent, hence any of
221 these highly hydrophobic congeners represent tracers of prey consumption. For our purposes, we
222 chose PCB 153, a highly recalcitrant and ubiquitous PCB congener that is commonly monitored
223 in trophic magnification studies^{41,42}, as well as other contaminant ecology studies^{43,44}. Moreover,
224 the similar variability among the congeners further demonstrates that each congener is tracking
225 common bioaccumulation processes (Figure 2). This correspondence in individual variability and
226 bioaccumulation rates among congeners confirms that super-hydrophobic PCB congeners offer
227 multiple, repeatable metrics for calculating consumption dynamics in fish.

228

229 Quantifying the consumption rates of top predator in aquatic ecosystems is essential for
230 understanding pollutant and nutrient trophodynamics and multiple approaches have been
231 developed to generate such consumption estimates. For instance, bioenergetics studies have used
232 sub-models of physiological characteristics such as metabolism, growth, excretion, and
233 reproduction to develop consumption estimates for age cohorts within a population^{14,16,45}, while
234 more empirical approaches have used Mercury (Hg) and Cesium-137 (¹³⁷Cs) dynamics^{19,20}.
235 Figure 3 compares fish consumption estimates using the PCB 153 method presented here with
236 estimates modeled by Stewart et al.¹⁶ for Lake Michigan lake trout, and by ¹³⁷Cs estimates

237 observed by Pazzia et al.²⁰ for Lake Ontario lake trout. Calculations of maximum consumption
238 (C_{\max}) provided in Stewart et al.¹⁶ are also included in Figure 3 using temperature and weight
239 data obtained from the present study. As observed in Figure 3, the PCB model developed in the
240 current study provides consumption estimates below those of C_{\max} , and similar to those obtained
241 or estimated from the other studies or models. The PCB method, however, provides direct
242 measurement of individual-based consumption, using concentrations observed in individual fish.
243 As Figure 3 demonstrates, there is considerable individual variability in consumption within a
244 population and the PCB method offers greater resolution as to the causes of these individual-
245 level differences.

246

247 The PCB method not only provides a way of calculating individual consumption estimates, but
248 provides a foundation for estimating nitrogen and phosphorus recycling by individual organisms.
249 Estimates of the lifetime nitrogen (N) and phosphorus (P) recycled by lake trout (*Salvelinus*
250 *namaycush*) based on PCB 153 accumulation dynamics revealed a power relationship, where the
251 mass of N and P recycled by fish increased with age (Figure 4). As the fish reached the
252 maximum asymptotic length predicted by the von Bertalanffy (VBL) growth model, the mass of
253 recycled nutrients increased exponentially. Specifically, as growth rates slow to this asymptote,
254 the majority of consumed prey energy and nutrients will be turned over via metabolic respiration
255 rather than assimilated into new somatic growth. Therefore these older individuals become
256 increasingly important sources of nutrient recycling. The relationship between nutrient recycling
257 and fish age and size is further resolved by examining fish growth rates. Individual growth rates
258 ($\% \cdot \text{year}^{-1}$) as a function of fish age (Figure 4, d), indicates that lake trout ≥ 5 years of age had
259 individual growth rates below $50\% \text{ yr}^{-1}$. At this time in their life history, these upper age cohorts

260 become nutrient sources through recycling rather than net nutrient sinks. The importance of older
261 fish regarding nutrient recycling increases exponentially as their growth rates continue to
262 decrease with age.

263

264 In one year, a single 13 year old lake trout from Lake Huron will have consumed 65 MJ of
265 energy and will have recycled 1441 g of N and 136 g of P. If fish population age structure
266 estimates are made using the yearly stocking levels of 4.3×10^6 yearlings with 40%
267 mortalities/year⁴⁶ and assuming a simple exponential decay model, then over a one year period,
268 in total, the lake trout population between 5 and 13 years of age (estimated at 1.7×10^6
269 individuals; capped at 13 as that is the oldest fish captured in the study) will have recycled 482
270 Tonnes of N, 45 Tonnes of P, and have acquired 22 TJ of energy. This compares to zebra
271 (*Dreissena polymorpha*) and quagga (*Dreissena bugensis*) mussels that are estimated to divert up
272 to 20 Tonnes of P in Lake Huron's Saginaw Bay region and are also associated with the near-
273 shore shunt in Great Lakes ecosystems⁴⁷. Total annual P loads to Lake Huron are estimated to be
274 590 Tonnes and the results of our study indicate that lake trout can recycle up to 7.6% of this
275 total load. However, it must be emphasized that much of the phosphorus recycled by fish will be
276 in bioavailable form and thus capable of directly supporting a significant proportion of primary
277 production in the lake⁴⁸. Furthermore, nutrient recycling by other fish species will increase this
278 estimate.

279

280 Although the impact of fish as a phosphorus sink has been previously documented¹², their
281 importance as nutrient recycling sources has been highly disputed. It is recognized that
282 anadromous species, such as Pacific salmon (*Oncorhynchus* spp), are important sources of

283 nutrients in coastal freshwater communities, acting as biovectors, transporting nutrients
284 accumulated in the marine environment to coastal freshwater systems^{49,50}. Studies of larger,
285 piscivorous fish have noted their importance as potential nutrient sources^{51,52} whilst others
286 examining smaller, forage fish argue to the contrary⁵³. Previous nutrient studies relied on
287 bioenergetics modeling and population density and growth estimates to estimate the relative
288 importance of fish on nutrient recycling in aquatic ecosystems^{14,17}. The PCB method developed
289 in the current study provides a novel approach to quantify *in situ* the magnitude of nutrient
290 recycling achieved by fish using multiple, repeatable metrics. In this capacity, top predator
291 species such as lake trout can act as off-shore vectors of these limiting nutrients thereby reducing
292 the impact of the near shore shunt phenomenon as associated with dreissenid mussel
293 establishment⁴⁷.

294

295 The life history of other large piscivores, however, can prevent these top predators from acting as
296 off-shore nutrient shunts. For instance, Lake Huron is also stocked with Pacific salmonids
297 (*Onchorhynchus* spp) which, in contrast to lake trout, grow more rapidly, have shorter life spans
298 and migrate to tributaries and streams at maturity where they spawn and die⁵⁴⁻⁵⁶. This specific
299 reproductive ecology has garnered Pacific salmonids much attention as sources of nutrients and
300 contaminants in spawning tributaries⁵⁷⁻⁶¹. Pacific salmonid spawning migrations generally occur
301 only when individual growth rates decline below 50% per year. These older salmon provide
302 limited contributions to offshore nutrient recycling with the mature senescent individuals
303 exporting a significant mass of nutrients out of the lake⁶¹. These observations highlight the
304 importance of piscivore life history with respect to nutrient transportation and export in aquatic
305 ecosystems.

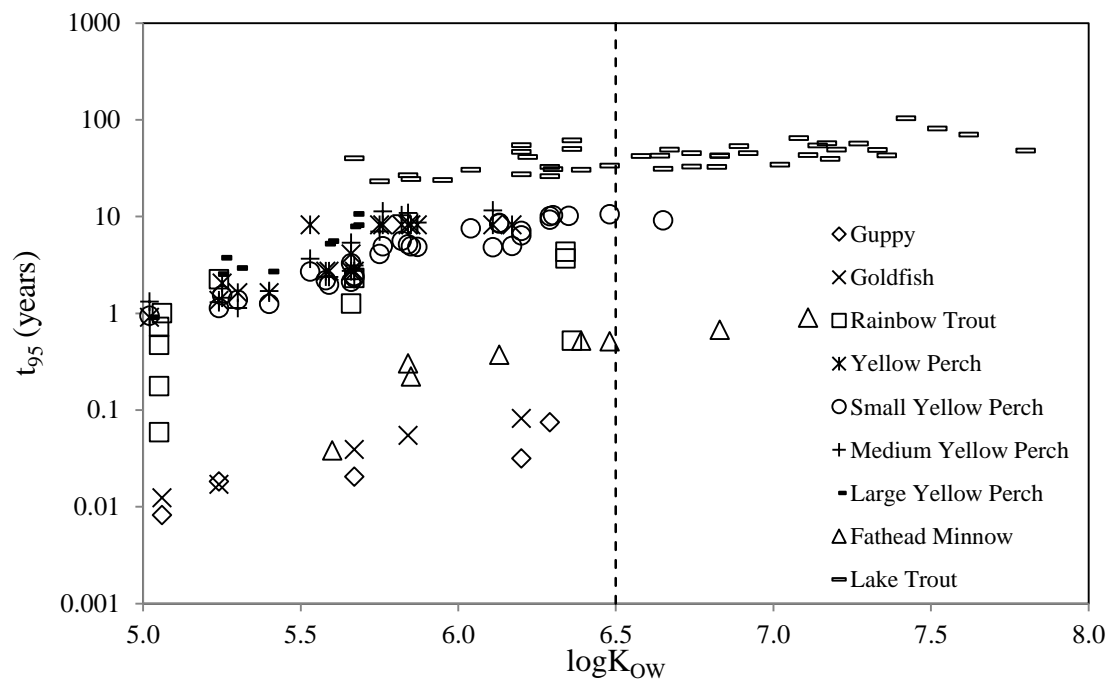
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307 While multiple factors have been identified as contributing to the regime shift in Lake Huron⁶²,
308 there is general agreement the lake is showing definite signs of ‘oligotrophication’. The results of
309 this study indicate that older lake trout can play an important role in the nutrient recycling in
310 oligotrophic lakes, systems to which they have well-adapted life histories. The stocking of fish
311 like salmon, however, is predicted to contribute significantly to a decline in bioavailable
312 nutrients, especially in the pelagic compartment. In Lake Huron, however, Pacific salmonid
313 abundances have declined dramatically following the collapse of alewife stocks⁴⁶ and older (> 5
314 yrs) lake trout have become the predominant off-shore salmonid predator population in the
315 lake⁶³. Given the differing life-spans, growth rates and reproductive strategies of lake trout
316 relative to stocked Pacific salmonids in the Great Lakes⁵⁴, the results of this study demonstrate
317 that lake trout provide a critical ecosystem service by effectively recycling offshore nutrients to
318 enhance food web stability and productivity in highly oligotrophic ecosystems.

319

320 Most importantly, this study demonstrates that PCB congeners can be used to quantify *in situ*
321 nutrient and energy flow in aquatic systems. Through the use of this metric, we have been able to
322 demonstrate the importance of fish growth rates and life history on the recycling of essential
323 limiting nutrients in oligotrophic lakes. The ubiquitous nature of PCBs implies that this
324 technique is applicable to aquatic systems across the globe. Moreover, due to the presence of
325 different congeners, PCBs provide repeatable, independent metrics for measuring nutrient and
326 energy flow which will allow us to quantify the effects of multiple environmental and
327 anthropogenic stressors across different aquatic ecosystems.

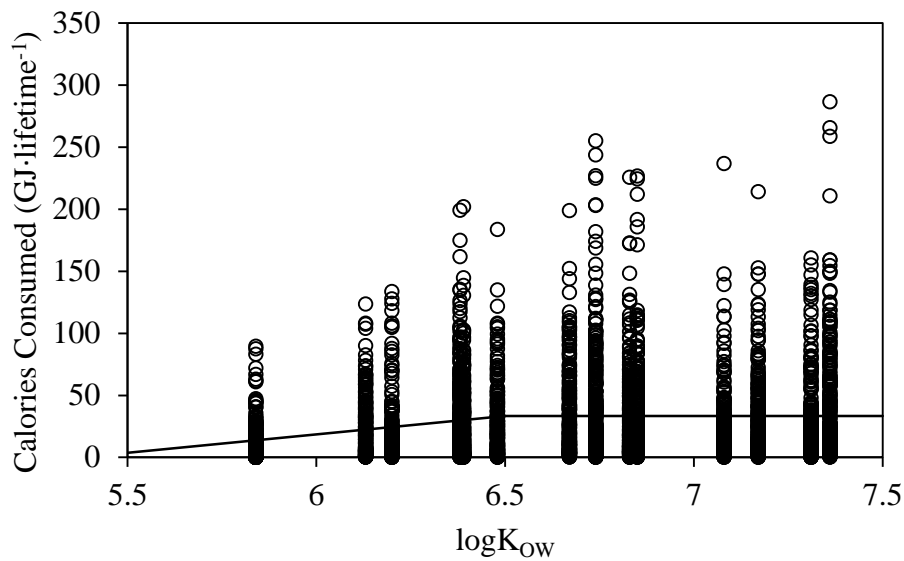
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330

331 **Figure 1:** Literature t_{95} estimates across a range of $\log K_{OW}$ s. Only those congeners depicting
 332 significant elimination are shown³⁵⁻⁴⁰. The dashed line represents the cut off at a $\log K_{OW}$ of 6.5,
 333 after which very few studies revealed significant elimination of congeners and these congeners
 334 were only in small fish. All estimates presented here are measured t_{95} estimates, with the
 335 exception of lake trout which were modeled; hence the elimination rates may not have been
 336 significant. However, the elimination rate estimates demonstrate that it takes at least a hundred
 337 years for congeners with higher $\log K_{OW}$ s to reach steady state.

338

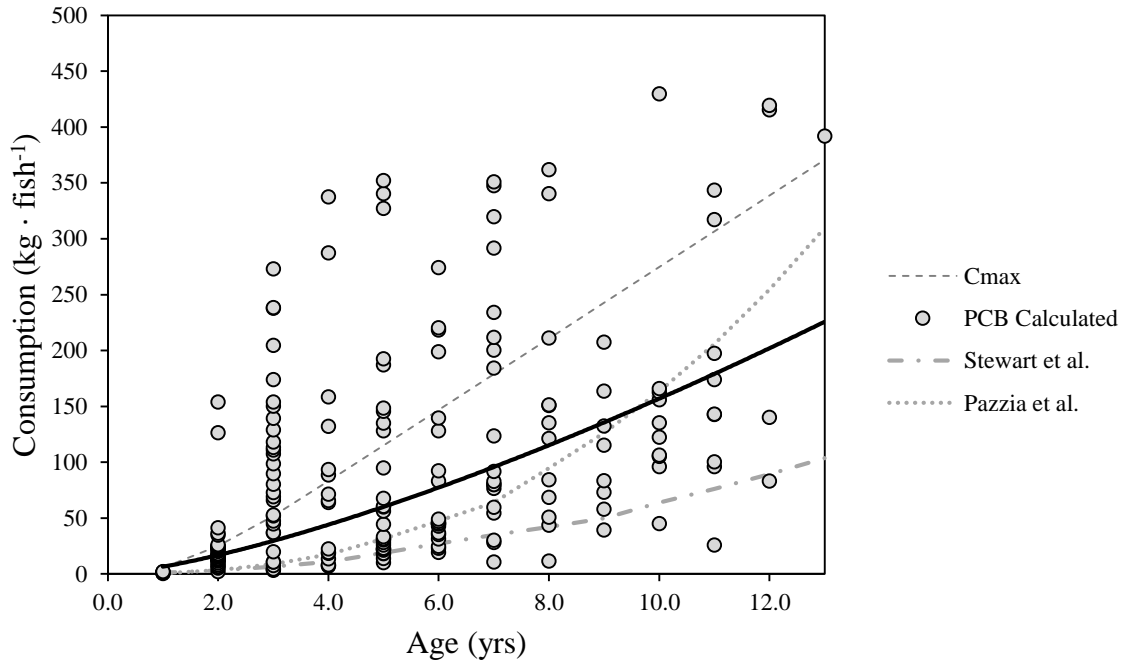


339

340 **Figure 2.** The relationship between calories consumed and logK_{ow} for the 15 most abundant
 341 PCB congeners (PCBs 52, 70, 95, 99, 101, 110, 118, 128, 138, 149, 153, 170, 177, 180, 187).

342 Piece-wise linear regressions were completed and no significant slope was found for congeners
 343 with logK_{ow}>6.5.

344



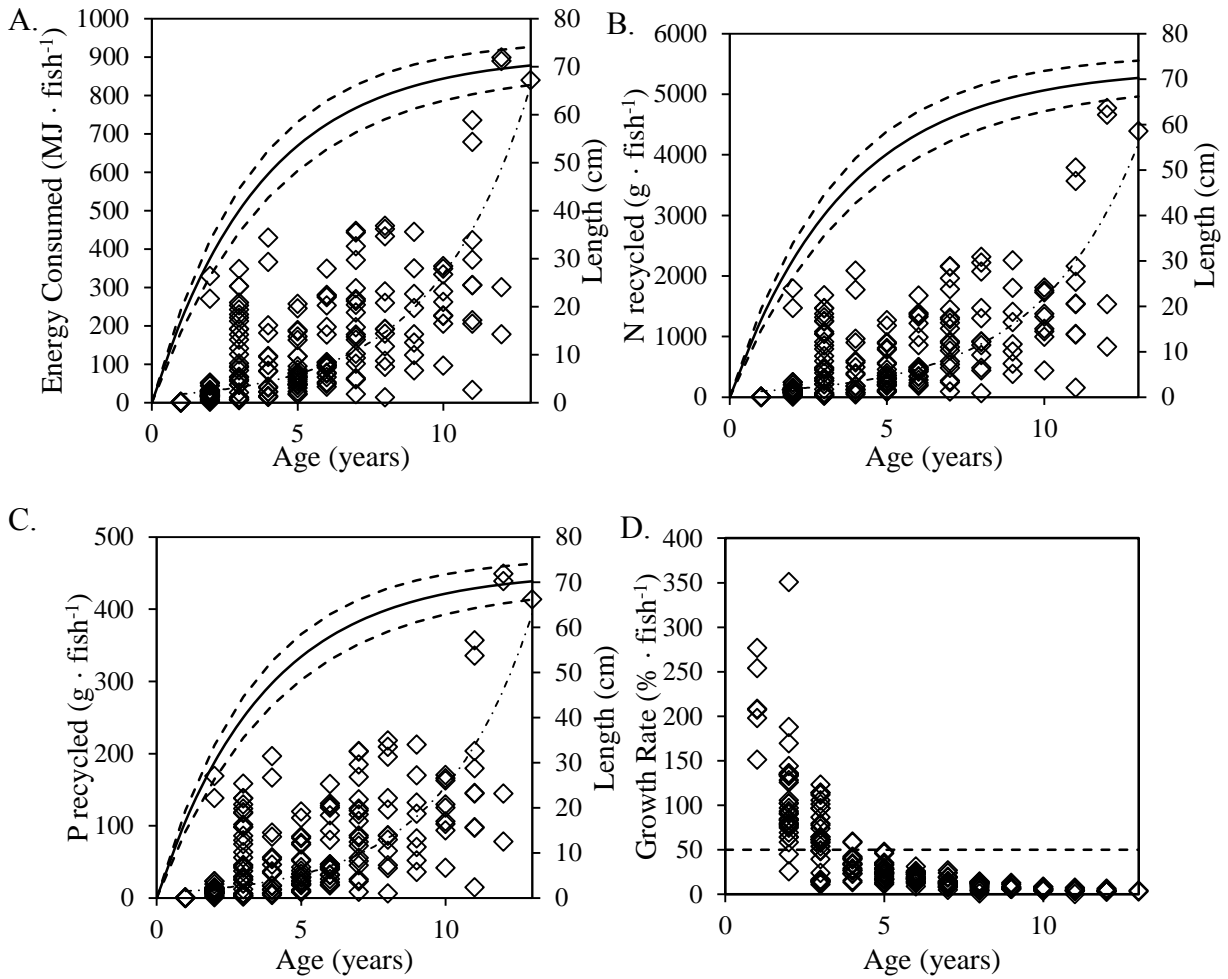
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346

347 **Figure 3:** Lifetime prey consumption estimates (kg · fish⁻¹) for lake trout calculated using PCB
 348 153 (grey circles, solid black line of best fit; $y = 6462.8x^{1.4}$ $R^2 = 0.4$), using C_{max} equation
 349 (dashed dark grey line) provided in Stewart et al. 1983¹⁶, with temperature and weight values
 350 obtained from our study, from extrapolation of bioenergetics estimates by Stewart et al. 1983¹⁶
 351 for Lake Michigan lake trout (dashed-dotted dark grey line), and from extrapolation of ¹³⁷Cs
 352 estimates for Lake Ontario lake trout by Pazzia et al. 2002²⁰ (dotted grey line).

353

354



355

356

357

358 **Figure 4:** (A-C) Relationships between the amount of (A) energy consumed (MJ · fish) and (B)
 359 nitrogen and (C) phosphorus recycled (g · fish) by lake trout. The solid curve in panels A-C
 360 represents the von Bertalanffy growth curve for lake trout with dotted lines representing the 95%
 361 confidence intervals. The dashed line in panels A-C represent the best fit, non-linear regression
 362 ($y = 16.625e^{0.3003x}$, $R^2 = 0.42$) and N ($y = 72.339e^{0.3122x}$, $R^2 = 0.40$) and P ($y = 6.8181e^{0.3122x}$, $R^2 =$

363 0.40). Panel D provides the relationship between individual lake trout growth rate (% year⁻¹) and
364 age with the dashed line indicating a growth rate of 50% year⁻¹.

365

366 ASSOCIATED CONTENT

367 **Supporting Information.**

368 Values for congener logK_{OW}²², E_{d,PCBi}²⁶ and f_{PCBi}²⁵ used in the calculations of the amount of PCB_i
369 ingested by a fish, results from gut content analyses on Lake Trout. This material is available free
370 of charge via the Internet at <http://pubs.acs.org>.

371 AUTHOR INFORMATION

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375 **Author Contributions**

376 G.D.H., G.P., and A.M. initiated the study, while K.G.D. provided input on development of the
377 modeling component. All authors were involved in the writing of the paper, but the main
378 contributions were by A.M. All authors discussed the results and consulted on the paper.

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385 ABBREVIATIONS

386 PCB, polychlorinated biphenyl; N, nitrogen; P, phosphorus; POPs, persistent organic pollutants;
387 K_{OW} , octanol-water partition coefficient; Hex, hexane; DCM, dichloromethane; VBL, von
388 Bertalanffy Growth; dw, dry weight, m_{PCB_i} , the mass of PCB_i in the fish (in ng); E_{d, PCB_i} ,
389 the organism's chemical assimilation efficiency for food for PCB_i ; f_{PCB_i} , the fraction of PCB_i
390 mass which is accumulated through ingestion; t , lake trout age (yr); L_t , the total length (cm) of
391 the fish at time t ; L_{∞} , the asymptotic length (cm); k , the growth coefficient (yr^{-1}).

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596

597 Supporting Information: PCB Food Web Dynamics

598 Quantify Nutrient and Energy Flow in Aquatic

599 Ecosystems

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605

606 *File consists of one supporting table and one supporting figure.*

607

608 **Table S1.**

609

PCB	logK _{OW} ^{a, 21}	E _{d, PCB_i} ^{b, 25}	f _{PCB_i} ^{c, 24}
18/17	5.1	0.802	0.96
31/28	5.67	0.699	0.71
48	5.85	0.667	0.86
44	5.75	0.685	0.86
70	6.2	0.604	0.88
99	6.39	0.570	0.93
87	6.29	0.588	0.91
110	6.48	0.554	0.93
118	6.74	0.507	0.96
153	6.85	0.487	0.96
105/132	6.7	0.514	0.91
138	6.83	0.491	0.96
158	7.02	0.456	0.97
187	7.17	0.429	0.98
183	7.2	0.424	0.97
128	6.74	0.507	0.96
156/171	7.2	0.424	0.97
180	7.36	0.395	0.98
170	7.31	0.404	0.98
201	7.2	0.424	0.985
195/208	7.65	0.343	0.983
194	7.8	0.316	0.98
206	8.09	0.264	0.989
209	8.18	0.248	0.99

610

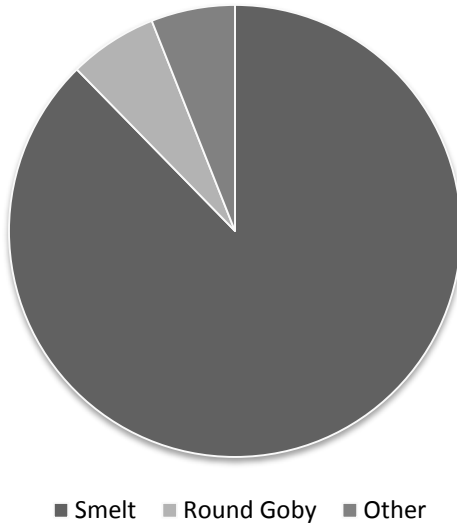
611 ^a the octanol-water partition coefficient as reported in Hawker and Connell, 1988.

612 ^b the organism's assimilation efficiency of PCB_i as reported in Liu et al. 2006.

613 ^c the fraction of PCB_i accumulated from dietary sources as reported in Arnot and Gobas 2004.

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617 **Figure S1.** Results from gut content analyses on Lake Trout from Lake Huron. Proportions are
618 based on the number of each prey item obtained.

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