



HAL
open science

Trophic position and individual feeding habits as drivers of differential PCB bioaccumulation in fish populations

T. Masset, V. Frossard, M.E. Perga, N. Nathalie Cottin, C. Piot, S. Cachera, E. Naffrechoux

► To cite this version:

T. Masset, V. Frossard, M.E. Perga, N. Nathalie Cottin, C. Piot, et al.. Trophic position and individual feeding habits as drivers of differential PCB bioaccumulation in fish populations. *Science of the Total Environment*, 2019, 674, pp.472-481. 10.1016/j.scitotenv.2019.04.196 . hal-02165156

HAL Id: hal-02165156

<https://univ-smb.hal.science/hal-02165156v1>

Submitted on 22 Oct 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

1
2 Trophic position and individual feeding habits as drivers of differential PCB
3 bioaccumulation in fish populations.

4
5
6 Masset T.^{a*}, Frossard V.^b, Perga M.E.^c, Cottin N.^a, Piot C.^a, Cachera S.^d and Naffrechoux E.^a

7 ^a Univ. Savoie Mont-Blanc, LCME, 73000 Chambéry, France

8 ^b Univ. Savoie Mont-Blanc, INRA, CARTELE, 73000 Chambéry, France

9 ^c UNIL, IDYST, CH-1015 Lausanne, Suisse

10 ^d CISALB, 73000 Chambéry, France

11 **Abstract**

12 Despite PCBs being banned since the 1980's, some European peri-alpine lakes, and consequently their
13 fish populations, are still contaminated by these xenobiotics. We investigated the relative contribution of
14 physiological and trophic factors that could be implicated in fish PCB bioaccumulation in Lake Bourget
15 (France), one of the most contaminated in Europe, by collecting Arctic char (n = 55) and European
16 whitefish (n = 89) from 2013 to 2016. Concentrations of 7 indicator PCBs were 9 - 168 ng.g w.w⁻¹ in
17 whitefish and 90 - 701 ng.g w.w⁻¹ in Arctic char. The fish trophic positions calculated from $\delta^{15}\text{N}$ values
18 were positively correlated with PCB concentrations ($r^2 = 0.45$; $p < 0.001$). A biomagnification model
19 relying on TP and lipid content of fish was then designed, and it confirmed this result. A Bayesian mixing
20 model based on $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values was used to estimate the relative contribution of preys in the fish
21 diet, which explained a significant proportion of the biomagnification model residuals (i.e., 17%).
22 Zooplankton consumption was negatively correlated with PCB concentrations, whereas consumption of
23 chironomids enhanced the PCB burden in fish. Correction of the biomagnification model for individual
24 diets of fish increased the correlation between the predicted and measured fish PCB contents ($R^2 = 0.71$;
25 $p < 0.001$), highlighting the importance of fish feeding habits in the bioaccumulation process.

26 **Keywords:** biomagnification, Polychlorobiphenyls, peri-alpine lake, Arctic char, whitefish, Bayesian
27 mixing model

28 **1. Introduction**

29 As in numerous places around the world, some lakes in Europe are still contaminated by polychlorinated
30 biphenyls (PCBs), either from primary sources or from global atmospheric pollution¹⁻³. Lake Geneva in
31 Switzerland⁴, and Lake Como⁵ and Lake Maggiore in Italy⁶ were found to be highly contaminated with
32 PCBs. Of all the subalpine lakes, Lake Bourget is probably the most heavily polluted⁷.

33
34 Fish PCB exposure has numerous consequences: reduction of their metabolic capacities^{8,9}, induction of
35 genotoxic effects¹⁰ and introduction of intersex occurrence¹¹. Bioaccumulation of pollutants in fish can
36 occur through two distinct mechanisms. First, bioconcentration describes the accumulation process of
37 water-dissolved compounds via tegumentary and branchial absorption. Second, when the
38 thermodynamic activity of a compound in an organism exceeds its diet, bioaccumulation of the pollutant
39 can be attributed to biomagnification. In contrast to bioconcentration, biomagnification varies with the
40 contamination level of the diet. It depends on the species feeding preferences, increasing with each

41 food-web trophic level^{12,13}. Therefore, through biomagnification, predation on contaminated fish can
42 also enhance the PCB burden of the upper trophic food chain^{14–17}. Biomagnification of lipophilic
43 compounds such as PCBs has been related to three different factors: (i), the nature of the contaminant
44 itself, commonly quantified by the octanol-water coefficient (K_{ow}); (ii) the physiology of the exposed
45 individual, i.e., its lipid content (as fatter fish tend to accumulate more contaminant), body size and age
46 (as the contaminant gets diluted with individual growth), and sex (as spawning could decrease
47 concentration in females)^{18,19}; and (iii) trophic factors such as living habitat (i.e., feeding location) and
48 diet preferences^{20,21}, food chain length and therefore trophic position (TP)²².
49

50 The relative contribution of mechanisms by which contaminants bioaccumulate in fish has been
51 extensively studied but remains quite uncertain in alpine lakes, where environmental conditions (carbon
52 sources, temperature, contaminant concentrations) can greatly differ yearly and seasonally. Some
53 studies could not link PCB levels in fish to any biomagnification processes, as PCB contamination was
54 neither correlated with body size, living habitat nor TP, especially in mountain lakes. Indeed, Catalan et
55 al²³ questioned the relative importance of biomagnification in contamination of salmonids in high-
56 altitude Lake Redo and suggested that bioconcentration could be a significant mechanism for PCB
57 bioaccumulation. Perga et al²⁴ concluded that PCB contamination may be mainly due to a nontrophic
58 (i.e., bioconcentration) process in Lake Muzelle. In contrast, numerous studies have shown that a
59 contaminated diet was responsible for bioaccumulation in fish in the American Great Lakes^{15,25,26} and
60 that contamination levels increased with TP^{14,22}. As a lipophilic contaminant, differences in fat content
61 result in differences in PCB contamination within the same species¹⁴. Living habitat and particularly
62 carbon sources (benthic or planktonic) were environmental factors directly linked to discrepancies in PCB
63 contamination between individuals and between species²⁰ when PCB contamination is heterogeneous
64 between trophic habitats. Even though their work did not focus on PCBs, Kahilainen et al.²¹ went further
65 and recently showed that ecomorphological divergences in whitefish could be responsible for a large
66 part of the intraspecies variability in mercury bioaccumulation.

67 PCB contamination in our study lake (Lake Bourget), remains considerable today; water concentrations
68 exceed US-EPA guidelines for the protection of aquatic wildlife²⁷ and highly contaminated sediment.
69 Therefore, Arctic char (*Salvelinus alpinus*) fishing is prohibited by a prefectural decree, despite being the
70 most emblematic fish species of Alpine lakes²⁸. European whitefish (*Coregonus lavaretus*), the other
71 typical fish of Alpine lakes, represents 90% (91 tons/year) of the fish caught in Lake Bourget since 2014
72 (S. Cachera, personal communication 2017). These species feed from pelagic and littoral food-web and
73 are suspected to venture into the benthic zone²⁹, which make them vulnerable to PCB biomagnification,
74 possibly to different extents. For these reasons, Arctic char and whitefish were identified as the most
75 relevant species to investigate for the research hypothesis of the present study. Herein, we assumed that
76 biomagnification necessarily implies that fish PCB contamination can be related to fish physiological and
77 trophic parameters, i.e., under the biomagnification hypothesis, interspecies, interindividual and
78 interannual PCB contamination levels shall be determined by (i) physiological parameters (body size or
79 lipid content), (ii) living habitat (determined from carbon isotope composition $\delta^{13}C$), or (iii) fish TP
80 (estimated from nitrogen stable isotope composition $\delta^{15}N$). We investigated the potential importance of
81 the fish feeding habits^{30,31} (different foraging behavior leading to a differential use of carbon sources) on
82 the interindividual variability of the PCB burden. Finally, we strove to highlight how a combination of
83 those parameters could result in important PCB contamination discrepancies in fish from a single lake.

84

85 2. Material and Methods

86 **2.1 Study site:**

87 Lake Bourget is located on the northwest edge of the French Alps. It is the largest natural lake in France,
88 with a surface of 44 km², a length of 18 km and a width of between 1.6 and 3.5 km, for a mean depth of
89 85 m (maximum: 145 m). Lake Bourget is a meso-oligotrophic lake with a mean temperature of 14.7°C
90 and 4.5 to 10.1 mgO₂.L⁻¹ of dissolved oxygen at 140 and 2 meters depth, respectively. Its pH is 8.3 and
91 the conductivity is 299 μs.cm⁻¹ for the period 2015-2017^{32,33}. PCB inputs are mainly due to a point source
92 located upstream on the Tillet River, which was contaminated from 1930 to 1987 by an electrical
93 transformer factory using Pyralène™. Despite the cessation of the industrial activity and the recent clean-
94 up of the downstream section of the river from 2012 to 2014, Lake Bourget is still contaminated. Even
95 though contamination decreased in the last decades, superficial sediments remain polluted with a mean
96 indicator PCB concentration (sum of PCB 28, 52, 101, 118, 153, 138, 180) of 12.1 ± 6.4 ng.g⁻¹ d.w, with
97 yet heterogeneous contamination of the lake, reaching 113 ng.g⁻¹ d.w at the Tillet River outlet (Fig. S1).
98 Consequently, the lake water still exceeds the USEPA recommended maximum concentration of 170
99 pg.L⁻¹ for the protection of aquatic wildlife²⁷, with a mean dissolved Σ7PCBi concentration of 322 ± 102
100 pg.L⁻¹.

101 **2.2 Processing:**

102 2.2.1 Sampling and preparation of organisms

103 European whitefish (n = 89) and Arctic char (n = 55) were collected from 2013 to 2016 using net fishing
104 (fish physiological characteristics can be found in Tab S1). The two species are suspected to be mobile in
105 the water body, and exact locations were not reported. Diverse stomach content analyses (i.e.,
106 volumetric and qualitative estimations) punctually performed on fishes obtained from recreational and
107 professional fishers revealed that the two fish species shared common prey in the lake, such as aquatic
108 insects and zooplankton, with some piscivory also detected for Arctic char (data not shown).
109 Consequently, similar resources were considered as possibly supporting whitefish and Arctic char in Lake
110 Bourget: littoral (i.e., -2 m to -5 m in depth) and deep (i.e., -20 m and -40 m in depth) chironomids larvae,
111 zooplankton (i.e., composite samples of *Daphnia* sp. and *Bythotrophes* sp.) plus young-of-the-year (yoy)
112 perch for Arctic char. Yoy perch (5-7.5 cm) were caught in the pelagic compartment at approximately 15
113 m in depth. Chironomids (n = 48) were collected at two depths (2-5 m and 20-40 m) in September 2015
114 using an Ekman grab and sieved using a 500-μm net. Zooplankton (n = 14) were sampled alongside with
115 a 50-μm net at the depth range of 0-20 m.

116 All samples were transferred to the laboratory in a cooling box and immediately processed or stored at -
117 18°C until further analysis. Organism processing has already been described previously³⁴. Briefly, fish
118 were dissected and filets ground using Ultraturax (3000 rpm). The same process was applied to
119 chironomids with the whole organism. Five g of filet (1 g of chironomids) were spiked with an internal
120 standard (PCB 116-d5), and an appropriate amount of anhydrous sodium sulfate (Na₂SO₄) was added for
121 water removal. PCB extraction was performed in heptane/acetone mixture (75:25 v/v) for 1 h at 45°C in
122 an ultrasonic bath. The solvent mixture was removed and the extraction procedure repeated once.
123 Extracts were then purified on an SPE cartridge loaded with silica (previously acidified with H₂SO₄ to
124 ensure lipid elimination of the extract) using a heptane/dichloromethane mixture, concentrated with
125 TurboVap II and resuspended in 500 μL of heptane. The lipid content in the fish filets were determined
126 for every sample. After the PCB extraction procedure above-mentioned and before the purification step,
127 5 mL of the extract was transferred in a pre-weighted vial and evaporated under a gentle stream of
128 nitrogen until steady weight. The mass collected in the vial corresponded to lipids and was ultimately
129 used for calculation of lipid content in the fish filets.

130 2.2.2 Chemicals

131 Heptane, acetone, dichloromethane and methanol were HPLC quality and purchased from Carlo Erba.
132 Anhydrous sodium sulfate was purchased from Fisher Chemical (analytical reagent grade). Silica gel 60
133 (70-230 mesh) for column chromatography was from Merck. Copper was activated with 10 mL of sulfuric
134 acid 1N (Chimie-Plus) and then rinsed three times with Milli-Q water to neutral pH and, subsequently,
135 with acetone for water removal. Standard mixture of indicator PCB congeners (28, 52, 101, 118, 138,
136 153, 180) at 100 µg.mL⁻¹ in iso-octane and internal standard PCB 116-d5 at 100 µg.mL⁻¹ in iso-octane
137 were purchased from Cluzeau Info Lab (France).

138 2.2.3 PCB analysis

139 All PCB solutions were analyzed using a Gas-Chromatography Clarus 580 from Perkin-Elmer with a
140 nonpolar fused silica capillary column Optima® HP-5-MS accent 30 m x 0.25 mm from Machery-Nagel®,
141 coupled with a Pulsed Discharge Detector in photoionization mode (Vici®). The samples were injected in
142 split/splitless mode at 300°C. Helium was used as the carrier gas (1.0 mL.min⁻¹). After injection, the oven
143 temperature was set at 60°C for 1 min, and it was increased gradationally at 10°C/min to 240°C and at
144 5°C/min to 320°C and kept steady for 5 min. Quantification was ensured for the 7 indicator PCB
145 congeners (PCB 28, 52, 101, 118, 153, 138, 180), because they represent a vast majority of the
146 compounds found in Pyralene® mixtures commercialized in France³⁵. Calibration curves were established
147 with authentic standards, and PCB 116-d5 was used as the internal standard (Fig. S2). Extraction yields
148 for each PCB_i are presented in Tab. S2 but were not used for result correction; normalization to PCB116-
149 d5 values spiked before extraction accounted for extraction correction. Detection limits were
150 determined as the mean response + three times the standard deviation of procedural blanks for each
151 PCB_i (Tab. S3). These concentrations were subtracted from the samples concentrations to avoid false
152 positives.

153 2.2.4 Stable isotope analysis

154 After sampling and dissection, fish filets, chironomids and zooplankton were dried at 50°C, ground into
155 fine powder using a mortar and pestle and kept in tin capsules away from humidity prior to isotopic
156 analysis. Samples in tin capsules were sent to SINLAB (NB, Canada). One mg was analyzed on a Finnigan
157 Delta Plus mass spectrometer coupled with a ConFlo II NC2500 Elemental Analyzer for δ¹³C and δ¹⁵N
158 quantification. Samples were calibrated against 7 plant and animal laboratory standards covering a wide
159 range of δ¹³C and δ¹⁵N values (i.e., from -35.08‰ to -13.25‰ and 0.02‰ to 16.14‰, respectively).
160 Three replicates for each sample were run. The precision values of the C and N isotope analyses were
161 <0.2‰.

162 2.2.5 Statistical analysis

163 All statistical tests were performed using the OriginPro 8 SR0 software and R 3.5.0 software. All data
164 were tested for normality using the Shapiro test and, if appropriate, for homoscedasticity using the
165 Bartlett test. When appropriate, data were log-transformed to ensure normality and homoscedasticity.
166 Differences in fish PCB concentrations and fish size, lipid content, δ¹⁵N and δ¹³C between year and season
167 were tested using an ANOVA or the Kruskal-Wallis test. Post hoc comparisons were performed using
168 Tukey's honest significant difference (HSD) or Dunnett tests. The relationship between fish PCB_i
169 concentrations and the measured parameters (size, lipid content, δ¹⁵N and δ¹³C) were tested using
170 Pearson correlations for normal variables or Spearman correlations otherwise.

171 **2.3 Modeling:**

172 2.3.1 Resource contributions

173
174 Bayesian mixing models (BMM) based on a Markov chain Monte Carlo simulation were used to assess
175 prey contributions to the diet of the two species at an individual level. BMM offer the opportunity to
176 account for the uncertainty (i.e., standard deviations) of the different data involved in the computation
177 (i.e., isotope composition of prey, trophic fractionation, elemental concentration) and provide robust
178 and accurate estimates of prey contributions to fish diets. The different resources considered as possibly
179 supporting whitefish and Arctic char in Lake Bourget were littoral (n = 16) and deep (n = 26) chironomids,
180 zooplankton (n = 14), plus yoy perch (n = 15) for Arctic char. $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ isotopic values were used for
181 computations. The trophic fractionations retained for carbon and nitrogen were those widely reported in
182 the literature^{36,37}: $\Delta\delta^{13}\text{C} = 0.8 \pm 1$ sd and $\Delta\delta^{15}\text{N} = 3.4 \pm 1$ sd. The computation of the Bayesian mixing
183 models were performed using the R package *simmr*³⁸ and considered 1.10^4 iterations, four Markov
184 chains and a burn-in period of 10^3 . Seasonal variability may bias prey contributions to fish diets,
185 especially for short-lived prey. To account for this possible bias, BMM were also performed (data not
186 shown) by increasing the standard deviation of zooplankton by 2 and 4 (i.e., to mimic possible seasonal
187 variability of their isotopic values) which led to minor (i.e., a few percent) and nonsignificant (i.e.,
188 Kruskal-Wallis tests, $p > 0.4$) changes in zooplankton contributions to the fish diets. Consequently, the
189 prey contributions estimated from the BMM were poorly affected by changes in the spread of their
190 isotopic values that would be expected if prey sampling would account for different seasons.
191 Salmonid muscle can exhibit high lipid contents that are typically ^{13}C -depleted and may alter the $\delta^{13}\text{C}$
192 values of their dorsal muscle³⁹. Consequently, $\delta^{13}\text{C}$ values for both whitefish and Arctic char were
193 corrected according to Post et al.³⁹, considering the sample C/N ratio to estimate lipid content in the
194 sample used for isotopic analysis following equation 1:

$$195 \delta^{13}\text{C}_{corrected} = \delta^{13}\text{C}_{measured} - 3.32 + 0.99 \times \text{CN}_{ratio} \quad (1)$$

198 2.3.2 Trophic position

199
200 The estimations of the trophic position at an individual level were performed based on the $\delta^{15}\text{N}$ values of
201 the different preys weighted by their respective contributions to individual fish diets estimated by the
202 BMM, as suggested by Post⁴⁰. This approach enabled estimates of the trophic position of each whitefish
203 (TP_{WFind}) and Arctic char (TP_{ACind}) individual according to equations 2 and 3, which generalized the initial
204 formula of Vander Zanden³⁷ for multiple baselines:

$$205 \text{TP}_{\text{WFind}} = [\delta^{15}\text{N}_{\text{WFind}} - (\alpha_{\text{zoo}} \times \delta^{15}\text{N}_{\text{zoo}} + \alpha_{\text{invdeep}} \times \delta^{15}\text{N}_{\text{invdeep}} + \alpha_{\text{invlitto}} \times \delta^{15}\text{N}_{\text{invlitto}})] /$$
$$206 3.4 + \overline{\text{TP}}_{\text{resources}} \quad (2)$$

$$207 \text{TP}_{\text{ACind}} = [\delta^{15}\text{N}_{\text{ACind}} - (\alpha_{\text{zoo}} \times \delta^{15}\text{N}_{\text{zoo}} + \alpha_{\text{invdeep}} \times \delta^{15}\text{N}_{\text{invdeep}} + \alpha_{\text{invlitto}} \times \delta^{15}\text{N}_{\text{invlitto}} +$$
$$208 \alpha_{\text{perch}} \times \delta^{15}\text{N}_{\text{perch}})] / 3.4 + \overline{\text{TP}}_{\text{resources}} \quad (3)$$

209
210 with $\delta^{15}\text{N}_{\text{WFind}}$ or $\delta^{15}\text{N}_{\text{ACind}}$ as the $\delta^{15}\text{N}$ values of individual fish, and $\alpha \times \delta^{15}\text{N}$ as the product of the
211 proportions of the different resources to their $\delta^{15}\text{N}$ values. The mean trophic position of resources (i.e.,
212 $\overline{\text{TP}}_{\text{resources}}$) of whitefish was 2, because zooplankton and invertebrate are typical primary consumers.
213 $\overline{\text{TP}}_{\text{resources}}$ of Arctic char was 2.125, because yoy perch is considered to have a trophic position of 2.5.
214 At a population level, the mean trophic positions for the two fish species were compared to those
215 estimated from a Bayesian framework recently developed by Quezada-Romegialli et al.⁴¹ to ensure the
216 robustness of the results and support their trophic differences. This Bayesian approach considers the
217 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of both whitefish and Arctic char, as well as those of their putative prey. The trophic
218
219

220 fractionations of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ consisted of a vector of 188 simulated trophic fractionations (i.e., equal
 221 length of the total number of isotope data for the two fish species) following a normal distribution with
 222 the same mean and standard deviation as the BMM. Bayesian trophic positions were computed using
 223 the R package *tRophicposition*⁴¹ considering 2.10^4 iterations and four Markov chains. In the model
 224 current implementation, consideration of more than two different baselines was not possible, while the
 225 two species feed at least on three or four different main resources. To be consistent in the computation
 226 of the trophic position for the two species, the two different baselines considered were zooplankton
 227 (i.e., pelagic resource) and deep invertebrates (i.e., benthic resource), since these resources had the
 228 highest contributions according to the BMM and accounted for the main differences in the $\delta^{15}\text{N}$ values of
 229 the resources.

231 2.3.3 Biomagnification model

232
 233 Total PCB concentrations in fish muscle used in our model were estimated as twice the measured Σ7PCBi
 234 in our samples⁴². As we expect that fish physiology, feeding habits and bioaccumulation mechanisms
 235 should not differ among years and remain similar overtime, one single biomagnification model was built
 236 for the period 2013-2016. The biomagnification model was built considering TP, measured lipid contents
 237 and their putative interactions from every individual fish of both species together as follows:

$$238 \log_{10} PCB_{estimated} = \alpha \times TP + \beta \times lipid\ content + \gamma \times (TP \times lipid\ content) \quad (4)$$

239 with α , β and γ as the regression coefficients estimated by the model and TP estimated from equations
 240 2 and 3.

241 To examine individual data more precisely, the residuals were extracted (i.e., equation 5) and could help
 242 highlight the different processes of contamination. Specifically, negative residuals would indicate lower
 243 contamination than expected, whereas positive residuals would indicate higher contamination than
 244 expected.

$$245 Residuals = \log_{10} PCB_{obs} - \log_{10} PCB_{estimated} \quad (5)$$

246 To identify the origin of these different processes (i.e., under and over contamination), residuals were
 247 modeled as a function of the different common resource contributions shared by the two species
 248 previously estimated using the BMM (i.e., equation 6), which could highlight the influence of prey type
 249 on the variability of fish contamination levels.

$$250 Residuals = f(a \times \alpha_{zoo} + b \times \alpha_{invdeep} + c \times \alpha_{invlitto} + \varepsilon) \quad (6)$$

251 The difference in the proportions of preys in the diet of the different individuals could reflect specific
 252 feeding habits (i.e., FH). Consequently, equation 6 could also be formulated as follows:

$$253 Residuals = f(FH + \varepsilon) \quad (7)$$

254 Following these different steps, equation 8 provided a general formulation of the PCB concentration in
 255 the different fish muscles at an individual level accounting for the generic biomagnification (i.e.,
 256 $\log_{10} PCB_{estimated}$) refined by a correction considering the individual feeding habits of the fish (i.e., FH):

$$257 \log_{10} PCB_{obs} = \log_{10} PCB_{estimated} + FH + \varepsilon \quad (8)$$

268

269

270 3. Results and discussion

271

272 **3.1 Temporal, interspecies and interindividual variability in PCB contamination:**

273 Arctic char was significantly more contaminated than whitefish (Fig. 1), both when wet weight and lipid-
274 normalized concentrations were considered (t-test, $p < 0.0001$ for both tests). In light of the relationship
275 between lipid content and PCB_i contamination ($r^2 = 0.52$; $p < 0.001$) and of the important discrepancies
276 regarding lipid contents between both species (i.e., mean lipid composition of 3.2% for whitefish and
277 8.9% for Arctic char, Tab. S1), only lipid-normalized PCB_i concentrations in fish were considered
278 thereafter. Interspecies and interindividual PCB_i profiles were consistent between years, with a major
279 contribution (i.e., 80%) of heavy PCB_i (Fig. S3).

280 Differences were observed between years for both species (ANOVA, $F_{3,51} p < 0.05$ for Arctic char and $F_{2,76}$
281 $p < 0.05$ for whitefish (Fig. 1)). Combining our data with data from 2008 from the French PCB follow-up
282 program⁴³ emphasized the important decline of lake fish contamination due to the clean-up of the Tillet
283 River between September 2012 and December 2013. Subsequently, a steady level of contamination was
284 observed for both species.

285 Within both species, the interindividual variability of the PCB_i concentrations was large (standard
286 deviation of normalized PCB_i concentration was 54% and 52% for whitefish and Arctic char, respectively).
287 Those results encouraged us to determine the relative importance of trophic and physiological
288 parameters potentially implicated in such interindividual variability.

289

290 **3.2 Physiological parameters and relationship with PCB_i contamination:**

291 For both species, no strong relationship could be drawn between the physiological fish parameters (size
292 and age) and the PCB contamination (data not shown). Fish physiology appeared to be different between
293 both species. The relatively narrow range of sizes (255 mm – 480 mm) and ages (1 year – 5 years) for the
294 target fish species sampled might be a reason behind the lack of a relationship with PCBs. A lack of
295 increasing PCB contamination with size was observed by Olsson et al. in small perch (<20 cm) in a Latvian
296 lake⁴⁴. An absence of contaminant-size relationship was also found for rainbow trout in Lake Michigan⁴⁵.
297 Metabolism of PCBs and elimination rates did not differ between Arctic char and whitefish since PCB
298 profiles and particularly metabolizable PCB (PCB 28 and PCB 52) percentages were very similar for both
299 species (Fig S3).

300

301 **3.3 Trophic plasticity of Arctic char and whitefish:**

302 Overall the two fish species had relatively similar $\delta^{13}\text{C}$ values, ranging from -34 ‰ to -31 ‰. Mean
303 annual $\delta^{13}\text{C}$ values showed significant variabilities between years (one-way ANOVA, $F_{3,51} = 5$; $p < 0.01$ and
304 $F_{2,76} = 32$; $p < 0.001$ for Arctic char and whitefish, respectively). Arctic char had a higher $\delta^{15}\text{N}$ than
305 whitefish (mean value of 15.7‰ and 13.7‰, respectively, Wilcox test $p < 0.0001$) confirming a piscivory
306 behavior resulting in a higher trophic status. The isotopic measurements for the different resources
307 considered to be consumed by whitefish and Arctic char did not overlap, although zooplankton and yoy
308 perch exhibited lower variability than the benthic invertebrates (Fig. 2). The main differences in the

309 isotopic compositions of these resources lay in their different $\delta^{15}\text{N}$ values, with pelagic resources (i.e.,
310 zooplankton and yoy perch) enriched by approximately 6 ‰ compared to benthic resources.

311 The BMMs applied at an individual level highlighted similar general patterns for the two fish species,
312 with a dominant contribution of zooplankton followed by deep invertebrate, while the littoral
313 invertebrates had a low although nonnegligible contribution (Fig. 3). For Arctic char, yoy perch appeared
314 as a nonnegligible resource, approaching the deep invertebrate contribution.

315
316 The interindividual variability in resource contribution was substantial for both species, although it was
317 higher for Arctic char than for whitefish regarding the two main resources (i.e., zooplankton and deep
318 invertebrates). The standard deviations of the zooplankton contribution were 13% and 11%, and they
319 were 31% and 13% in the deep invertebrate contribution for Arctic char and whitefish, respectively.

320
321 The TP calculated according to Post⁴⁰ was significantly higher for Arctic char than for whitefish (Wilcox
322 test, $p < 0.01$), with a mean TP of 3.21 ± 0.09 for whitefish and 3.48 ± 0.13 for Arctic char (Fig. S4 A, B).
323 Whitefish and Arctic char exhibited similar TP ranges of approximately 0.4, although one Arctic char
324 individual had an especially high TP of 4.04. No significant relationship could be identified between TP
325 and fish size (ANOVA, $p > 0.05$ for both species, Fig. S4 C, D). Bayesian inferences provided highly similar
326 results to Post's estimates for TP statistics at a population level, with mean TP of 3.28 ± 0.37 for
327 whitefish and 3.58 ± 0.48 for Arctic char (Fig. S4 E).

328

329 **3.4 Trophic parameters and relationship with PCBi contamination:**

330 $\delta^{13}\text{C}$ and TP were not correlated ($r^2 = 0.18$; $p > 0.05$ and $r^2 = 0.03$; $p > 0.05$, respectively) with PCBi
331 concentrations in whitefish for all samples considered (Fig. 4). A finer analysis for each year was
332 performed and showed the same negative results (Fig. S5 and S6). The same analyses were performed
333 for Arctic char, and both trophic parameters $\delta^{13}\text{C}$ and TP were unrelated to PCBi concentrations ($r^2 = 0$; p
334 > 0.05 and $r^2 = 0.22$; $p > 0.05$, respectively) (Fig. 4). For whitefish, advanced testing of different years did
335 not show any correlation (Fig. S5 and S6).

336 As in our study, a lack of intraspecies relationships between PCB concentrations and TP was also
337 observed in Salmonids from the Great Lakes^{13,46}. The authors hypothesized that this could be related to
338 the trophic structure and dynamic of the food web. Vander Zanden et al.¹³ found that in a short-length
339 food chain, no relationship between PCB concentrations and TP were observed in Lake Trout. However,
340 our data showed that TP and PCB concentration data were correlated ($r^2 = 0.45$; $p < 0.001$) when both
341 species were examined together, with an increase of the PCBi contamination with trophic position. This
342 followed the findings of Vander Zanden et al.¹³, for which an increasing range of TP values could also
343 enable the observation of a positive relationship between TP and PCB concentrations. The increase of
344 PCB concentrations with TP could be an indicator of the biomagnification process. The Trophic
345 Magnification Factor (TMF) is a reliable metric to assess biomagnification (i.e., $\text{TMF} > 1$) in aquatic food
346 webs⁴⁷. TMF can be determined as the regression slope of the following equation: $\log[\text{PCB}] = a\text{TP} + b$. In
347 our study, a TMF value of 1.8 confirmed biomagnification.

348 At least two parameters (lipid content and TP) seem to account for a significant part of the PCB
349 variability in fish. To clear up the different processes involved and the relative importance of
350 biomagnification in fish contamination, we built a model to which our data were compared.

351

352 **3.5 Data comparison with a biomagnification model:**

353 This model was based on the correlation described by Vander Zanden et al.¹³, taking into account TP and
354 lipid content. It enabled to estimate PCB concentrations in fish (a comparison between our model and
355 the model of Vander Zanden et al.¹³ can be found in Fig. S7). PCB concentrations of fish individuals were
356 estimated and compared with the measured PCB values (calculated as twice the measured Σ 7PCBi
357 concentration, see above). The biomagnification model using TP and the lipid content exhibited high
358 performance ($r^2_{\text{adj}} = 0.64$, Fig. 6A). Ordering the residuals of the biomagnification model enabled us to
359 highlight possible differences in contamination mechanisms (Fig. 5). A large proportion of Arctic char
360 (i.e., 64%) tended to have positive residuals, indicating that they were more contaminated than
361 predicted by the model. In contrast, 58% of whitefish exhibited the opposite patterns (i.e., less
362 contaminated than predicted).

363 The origin of the individual differences between measured contamination and modeled contamination
364 (i.e., model residuals) was investigated regarding the different resources shared by the two species,
365 which reflected their feeding habits. The fitted linear model shown in equation 7 indicated that the three
366 shared resources could explain a significant proportion of the residuals of the biomagnification model
367 ($r^2_{\text{adj}} = 0.17$, $p < 0.001$). The model estimates provided important insights in the way the different
368 resources could influence fish contamination. Both zooplankton and deep invertebrates were
369 characterized by negative estimates suggesting that an increasing contribution of these resources would
370 lead to lower contamination (PCB concentration in zooplankton was not measured in this work). In
371 contrast, littoral invertebrates were associated with positive estimates, indicating that this resource may
372 support positive residuals; that is, there were higher PCB concentrations than expected (Σ 7PCBi
373 concentration of chironomids decreased from $541 \pm 378 \text{ ng.g w.w}^{-1}$ at the outlet of the Tillet River in
374 2015 down to $41 \pm 30 \text{ ng.g w.w}^{-1}$ at -20 m to -40 m).

375 Predicted contamination of the biomagnification model associated with corrections for feeding habits
376 indicated that most Arctic char (i.e., 78%) had positive corrections, while most whitefish (i.e., 78%) had
377 negative corrections (Fig. 5). This result suggested that the feeding habits of fish may partly explain the
378 discrepancies between observed and predicted PCB concentrations. Specifically, feeding habits of fish
379 would tend to increase the predicted concentrations of the biomagnification model for Arctic char and
380 decrease those predicted for whitefish.

381 The biomagnification model provided valuable information regarding the general variables responsible
382 for PCB variability in fish muscles. Nonetheless, the complementary corrections for individual feeding
383 habits allowed even more accurate predictions ($r^2_{\text{adj}} = 0.71$), which may be especially important
384 considering the \log_{10} -scale (Fig. 6B). This was exemplified by the three highest concentrations in Arctic
385 char (i.e., boxes in Fig. 6), for which the residuals were the most penalizing on a \log_{10} -scale. For these
386 individuals, the biomagnification model alone showed a mean difference between predicted and
387 measured PCB concentrations of $637 \pm 55 \text{ ng.g}^{-1}$, while once the feeding habits correction is applied, the
388 difference decreases down to $253 \pm 83 \text{ ng.g}^{-1}$.

389 Altogether, the lipid content and trophic position accounted for a large part of the variability in the PCB
390 contamination in fish from Lake Bourget. Our modeling, including correction for feeding habits, allowed
391 us to account for significant complementary PCB variations not accounted for by the lipid content or
392 trophic position and highlight that some fishes (mainly Arctic char) were highly contaminated due to
393 feeding on benthic littoral invertebrate, whereas less contaminated fish (mainly whitefish) fed
394 predominantly on zooplankton.

395
396 Intraspecies specialization in aquatic animals has been an emerging topic of interest the past few years³⁰.
397 Numerous studies have shown that ecological specialization could occur in fish^{31,48}. Arctic char ecology is
398 extremely difficult to encompass, because the intrapopulation variability of diverse aspects (morphology,
399 feeding strategies, life-history strategies) can be complex^{49,50,48}. Similarly, whitefish was also proved to

400 exhibit various morphs in the lacustrine systems⁵²⁻⁵⁴. Recently, Kahilainen et al.²¹ showed that
401 ecomorphological divergence could be responsible for differential mercury bioaccumulation in whitefish.
402 Moreover, fish respiratory capacity can vary between species and also between individuals^{55,56}, leading
403 to possible difference in PCB metabolism rates. Therefore, intra and interspecies variation in PCB
404 bioaccumulation could be partially explained by intra and interspecies physiological differences at the
405 gills level. Hence, the diversity of behaviors and morphology potentially encountered for the same
406 population could be partly responsible for the large span of PCB contamination observed, as different
407 individuals could develop in high or low contamination environments in Lake Bourget.

408 Altogether, our results suggested that trophic parameters have a significant effect on PCB contamination
409 variability between Arctic char and whitefish. Within species, feeding habits were shown to be
410 responsible for a substantial part of the PCB burden. This study emphasized the importance of the
411 feeding habits in the bioaccumulation process for Arctic char and whitefish in Lake Bourget.

412 Modeling or empirical studies based on the prediction of fish PCB burden according to the environmental
413 compartment PCB concentration (water, sediment) are useful tools for managers. However, it should be
414 kept in mind that considerable inter- and intraspecies variability exists that can be partly related to
415 feeding habits. Consequently, estimates can greatly differ from actual measured values, especially in the
416 most contaminated organisms, which are particularly concerning per safety guidelines. Therefore, as
417 recently stressed by Mackay et al.⁵⁷, caution should be taken when using models for regulatory purposes
418 and field monitoring accounting for inter- and intraspecies variability, and spatial variability seems to
419 remain a reliable way to assess the ecological state of lacustrine ecosystems. Field monitoring should
420 include sampling of at least 7 fish per target species, as suggested by Amiard et Al.⁵⁸, and it should be
421 performed seasonally at several locations and depths within the lake in order to account for feeding
422 habits and to prevent biases resulting from seasonal and spatial fish contamination.

423 Acknowledgements

424 The authors would like to thank ZABR CNRS and Agence de l'eau RMC, funders of the research program
425 RESTOLAC (2013-2016) as well as OLA (Observatory on LAkes), © OLA-IS, AnaEE-France, INRA Thonon-
426 les-Bains, CISALB for IS OLA Data.

428 Author information

429 Corresponding author:

430 *Tel: +33 479 758 898; email: Thibault.masset@univ-smb.fr

431

432 Supplementary material

433 Sediment sampling locations and associated congeners profile; example of chromatogram in ECD-PDID
434 mode; whitefish and arctic char physiological characteristics; extraction yields and detection limits for
435 the different matrices; mean PCB concentration and mean fish 7 indicator PCB congeners repartition;
436 trophic positions determined with frequency; plots with yearly relationships between $\delta^{13}\text{C}$ and TP with
437 PCB concentration; bayesian approaches and relationship between biomagnification model developed
438 in this study and Vander Zanden and Rasmussen (1996) model.

440 References

441

- 442 (1) Nellier, Y.-M.; Perga, M.-E.; Cottin, N.; Fanget, P.; Malet, E.; Naffrechoux, E. Mass Budget in Two
443 High Altitude Lakes Reveals Their Role as Atmospheric PCB Sinks. *Sci. Total Environ.* **2015**, *511*,
444 203–213.
- 445 (2) Carrera, G.; Fernández, P.; Grimalt, J. O.; Ventura, M.; Camarero, L.; Catalan, J.; Nickus, U.; Thies,
446 H.; Psenner, R. Atmospheric Deposition of Organochlorine Compounds to Remote High Mountain
447 Lakes of Europe. *Environ. Sci. Technol.* **2002**, *36* (12), 2581–2588.
- 448 (3) Castro-Jiménez, J.; Mariani, G.; Eisenreich, S. J.; Christoph, E. H.; Hanke, G.; Canuti, E.; Skejo, H.;
449 Umlauf, G. Atmospheric Input of POPs into Lake Maggiore (Northern Italy): PCDD/F and Dioxin-
450 like PCB Profiles and Fluxes in the Atmosphere and Aquatic System. *Halogenated Persistent Org.*
451 *Pollut. Dioxin 2005 Selected Pap. 25th Int. Symp. Halogenated Environ. Org. Pollut. POPs Held Tor.*
452 *Can. August 2005* **2008**, *73* (1, Supplement), S122–S130.
- 453 (4) Mathieu, A.; Babut, M. Contamination Des Poissons d’eau Douce Par Des Contaminants
454 Persistants: Polychlorobiphényles (PCB), Dioxines, Furanes, Mercure. Etude Des Relations Biote-
455 Sédiment Pour Les PCB. **2012**.
- 456 (5) Bettinetti, R.; Quadroni, S.; Boggio, E.; Galassi, S. Recent DDT and PCB Contamination in the
457 Sediment and Biota of the Como Bay (Lake Como, Italy). *Sci. Total Environ.* **2016**, *542*, 404–410.
- 458 (6) Binelli, A.; Ricciardi, F.; Provini, A. Present Status of POP Contamination in Lake Maggiore (Italy).
459 *Chemosphere* **2004**, *57* (1), 27–34.
- 460 (7) Naffrechoux, E.; Cottin, N.; Pignol, C.; Arnaud, F.; Jenny, J.-P.; Perga, M.-E. Historical Profiles of
461 PCB in Dated Sediment Cores Suggest Recent Lake Contamination through the “Halo Effect.”
462 *Environ. Sci. Technol.* **2015**, *49* (3), 1303–1310. <https://doi.org/10.1021/es5043996>.
- 463 (8) Jørgensen, E. H.; Vijayan, M. M.; Killie, J.-E. A.; Aluru, N.; Aas-Hansen, Ø.; Maule, A. Toxicokinetics
464 and Effects of PCBs in Arctic Fish: A Review of Studies on Arctic Charr. *J. Toxicol. Environ. Health A*
465 **2006**, *69* (1–2), 37–52.
- 466 (9) Gauthier, P. T.; Evenset, A.; Christensen, G. N.; Jørgensen, E. H.; Vijayan, M. M. Lifelong Exposure
467 to PCBs in the Remote Norwegian Arctic Disrupts the Plasma Stress Metabolome in Arctic Charr.
468 *Environ. Sci. Technol.* **2017**.
- 469 (10) Marabini, L.; Calò, R.; Fucile, S. Genotoxic Effects of Polychlorinated Biphenyls (PCB 153, 138, 101,
470 118) in a Fish Cell Line (RTG-2). *Toxicol. In Vitro* **2011**, *25* (5), 1045–1052.
- 471 (11) Hinck, J. E.; Blazer, V. S.; Schmitt, C. J.; Papoulias, D. M.; Tillitt, D. E. Widespread Occurrence of
472 Intersex in Black Basses (*Micropterus* Spp.) from US Rivers, 1995–2004. *Aquat. Toxicol.* **2009**, *95*
473 (1), 60–70.
- 474 (12) Gobas, F. A. Bioconcentration and Biomagnification in the Aquatic Environment. *Handb. Prop.*
475 *Estim. Methods Chem. Environ. Health Sci.* **2000**.
- 476 (13) Vander Zanden, M. J.; Rasmussen, J. B. A Trophic Position Model of Pelagic Food Webs: Impact on
477 Contaminant Bioaccumulation in Lake Trout. *Ecol. Monogr.* **1996**, *66* (4), 451–477.
- 478 (14) Rasmussen, J.; Rowan, D.; Lean, D.; Carey, J. Food Chain Structure in Ontario Lakes Determines
479 PCB Levels in Lake Trout (*Salvelinus Namaycush*) and Other Pelagic Fish. *Can. J. Fish. Aquat. Sci.*
480 **1990**, *47* (10), 2030–2038.
- 481 (15) Kidd, K.; Hesslein, R.; Ross, B.; Koczanski, K.; Stephens, G.; Muir, D. C. Bioaccumulation of
482 Organochlorines through a Remote Freshwater Food Web in the Canadian Arctic. *Environ. Pollut.*
483 **1998**, *102* (1), 91–103.
- 484 (16) Sørmo, E. G.; Salmer, M. P.; Jenssen, B. M.; Hop, H.; Bæk, K.; Kovacs, K. M.; Lydersen, C.; Falk-
485 Petersen, S.; Gabrielsen, G. W.; Lie, E. Biomagnification of Polybrominated Diphenyl Ether and
486 Hexabromocyclododecane Flame Retardants in the Polar Bear Food Chain in Svalbard, Norway.
487 *Environ. Toxicol. Chem.* **2006**, *25* (9), 2502–2511.

- 488 (17) Alava, J. J.; Ross, P. S.; Lachmuth, C.; Ford, J. K.; Hickie, B. E.; Gobas, F. A. Habitat-Based PCB
489 Environmental Quality Criteria for the Protection of Endangered Killer Whales (*Orcinus Orca*).
490 *Environ. Sci. Technol.* **2012**, *46* (22), 12655–12663.
- 491 (18) Johnston, T. A.; Fisk, A. T.; Whittle, D. M.; Muir, D. C. Variation in Organochlorine Bioaccumulation
492 by a Predatory Fish; Gender, Geography, and Data Analysis Methods. *Environ. Sci. Technol.* **2002**,
493 *36* (20), 4238–4244.
- 494 (19) Borgå, K.; Fisk, A. T.; Hoekstra, P. F.; Muir, D. C. Biological and Chemical Factors of Importance in
495 the Bioaccumulation and Trophic Transfer of Persistent Organochlorine Contaminants in Arctic
496 Marine Food Webs. *Environ. Toxicol. Chem.* **2004**, *23* (10), 2367–2385.
- 497 (20) Lopes, C.; Perga, M.-E.; Peretti, A.; Roger, M.-C.; Persat, H.; Babut, M. Is PCBs Concentration
498 Variability between and within Freshwater Fish Species Explained by Their Contamination
499 Pathways? *Chemosphere* **2011**, *85* (3), 502–508.
- 500 (21) Kahilainen, K. K.; Thomas, S. M.; Nystedt, E. K.; Keva, O.; Malinen, T.; Hayden, B. Ecomorphological
501 Divergence Drives Differential Mercury Bioaccumulation in Polymorphic European Whitefish
502 (*Coregonus Lavaretus*) Populations of Subarctic Lakes. *Sci. Total Environ.* **2017**, *599*, 1768–1778.
- 503 (22) Borgå, K.; Gabrielsen, G.; Skaare, J. Biomagnification of Organochlorines along a Barents Sea Food
504 Chain. *Environ. Pollut.* **2001**, *113* (2), 187–198.
- 505 (23) Catalan, J.; Ventura, M.; Vives, I.; Grimalt, J. O. The Roles of Food and Water in the
506 Bioaccumulation of Organochlorine Compounds in High Mountain Lake Fish. *Environ. Sci. Technol.*
507 **2004**, *38* (16), 4269–4275.
- 508 (24) Perga, M.-E.; Nellier, Y.-M.; Cottin, N.; Fanget, P.; Naffrechoux, E. Bioconcentration May Be
509 Favoured over Biomagnification for Fish PCB Contamination in High Altitude Lakes. *Inland Waters*
510 **2017**, *7* (1), 14–26.
- 511 (25) Oliver, B. G.; Niimi, A. J. Trophodynamic Analysis of Polychlorinated Biphenyl Congeners and
512 Other Chlorinated Hydrocarbons in the Lake Ontario Ecosystem. *Environ. Sci. Technol.* **1988**, *22*
513 (4), 388–397.
- 514 (26) Madenjian, C. P.; Elliott, R. F.; Schmidt, L. J.; Desorcie, T. J.; Hesselberg, R. J.; Quintal, R. T.;
515 Begnoche, L. J.; Bouchard, P. M.; Holey, M. E. Net Trophic Transfer Efficiency of PCBs to Lake
516 Michigan Coho Salmon from Their Prey. *Environ. Sci. Technol.* **1998**, *32* (20), 3063–3067.
- 517 (27) Agency for Toxic Substances and Disease Registry. Public Health Statement for Polychlorinated
518 Biphenyls (PCBS). November 2000.
- 519 (28) Batias, A. Le Lavaret Du Lac Du Bourget. Productivité et Alevinage. *Bull. Fr. Piscic.* **1954**, No. 173,
520 150–163.
- 521 (29) Visini, V. Salmonidés du lac du Bourget ETUDE DE LEUR REPRODUCTION. August 20, 2008.
- 522 (30) Dingemanse, N. J.; Kazem, A. J.; Réale, D.; Wright, J. Behavioural Reaction Norms: Animal
523 Personality Meets Individual Plasticity. *Trends Ecol. Evol.* **2010**, *25* (2), 81–89.
- 524 (31) Wolf, M.; Weissing, F. J. Animal Personalities: Consequences for Ecology and Evolution. *Trends*
525 *Ecol. Evol.* **2012**, *27* (8), 452–461.
- 526 (32) Jacquet, S., et Al. Suivi Environnemental Des Eaux Du Lac Du Bourget Pour l'année 2015. 2016.
- 527 (33) SOERE OLA-IS, AnaEE-France, INRA of Thonon-Les-Bains, CISALB, 09/04/2019, Developed by the
528 Eco-Informatique ORE System of the INRA.
- 529 (34) Nellier, Y.-M.; Perga, M.-E.; Cottin, N.; Fanget, P.; Malet, E.; Naffrechoux, E. Mass Budget in Two
530 High Altitude Lakes Reveals Their Role as Atmospheric PCB Sinks. *Sci. Total Environ.* **2015**, *511*,
531 203–213.
- 532 (35) Dargnat, C.; Fisson, C. Les PolyChloroBiphényles (PCB) Dans Le Bassin de La Seine et Son Estuaire.
533 *Etude Réalis. Par GIP Seine-Aval* **2010**.
- 534 (36) Minagawa, M.; Wada, E. Stepwise Enrichment of $\delta^{15}\text{N}$ along Food Chains: Further Evidence and
535 the Relation between $\Delta^{15}\text{N}$ and Animal Age. *Geochim. Cosmochim. Acta* **1984**, *48* (5), 1135–1140.

- 536 (37) Vander Zanden, M. J.; Cabana, G.; Rasmussen, J. B. Comparing Trophic Position of Freshwater Fish
537 Calculated Using Stable Nitrogen Isotope Ratios ($\Delta^{15}\text{N}$) and Literature Dietary Data. *Can. J. Fish.*
538 *Aquat. Sci.* **1997**, *54* (5), 1142–1158.
- 539 (38) Parnell, A. Simmr: A Stable Isotope Mixing Model. R Package Version 0.3.1. 2016.
- 540 (39) Post, D. M.; Layman, C. A.; Arrington, D. A.; Takimoto, G.; Quattrochi, J.; Montana, C. G. Getting to
541 the Fat of the Matter: Models, Methods and Assumptions for Dealing with Lipids in Stable Isotope
542 Analyses. *Oecologia* **2007**, *152* (1), 179–189.
- 543 (40) Post, D. M. Using Stable Isotopes to Estimate Trophic Position: Models, Methods, and
544 Assumptions. *Ecology* **2002**, *83* (3), 703–718.
- 545 (41) Quezada-Romegialli, C.; Jackson, A. L.; Hayden, B.; Kahilainen, K. K.; Lopes, C.; Harrod, C.
546 TRrophicPosition, an r Package for the Bayesian Estimation of Trophic Position from Consumer
547 Stable Isotope Ratios. *Methods Ecol. Evol.* **2018**, *9*, 1292–1299.
- 548 (42) Opinion of the Scientific Panel on Contaminants in the Food Chain Related to the Presence of Non
549 Dioxin-like Polychlorinated Biphenyls (PCB) in Feed and Food. *EFSA J.* **2005**, *3* (11).
- 550 (43) Agence de l'eau RMC. Base de Données Micropolluants Du Programme PCB. Agence de l'eau
551 RMC, FRANCE 2008.
- 552 (44) Olsson, A.; Valters, K.; Burreau, S. Concentrations of Organochlorine Substances in Relation to Fish
553 Size and Trophic Position: A Study on Perch (*Perca fluviatilis* L.). *Environ. Sci. Technol.* **2000**, *34*
554 (23), 4878–4886.
- 555 (45) Stow, C. A. Factors Associated with PCB Concentrations in Lake Michigan Fish. *Environ. Sci.*
556 *Technol.* **1995**, *29* (2), 522–527.
- 557 (46) Kiriluk, R. M.; Servos, M. R.; Whittle, D. M.; Cabana, G.; Rasmussen, J. B. Using Ratios of Stable
558 Nitrogen and Carbon Isotopes to Characterize the Biomagnification of DDE, Mirex, and PCB in a
559 Lake Ontario Pelagic Food Web. *Can. J. Fish. Aquat. Sci.* **1995**, *52* (12), 2660–2674.
- 560 (47) Borgå, K.; Kidd, K. A.; Muir, D. C.; Berglund, O.; Conder, J. M.; Gobas, F. A.; Kucklick, J.; Malm, O.;
561 Powell, D. E. Trophic Magnification Factors: Considerations of Ecology, Ecosystems, and Study
562 Design. *Integr. Environ. Assess. Manag.* **2012**, *8* (1), 64–84.
- 563 (48) Brandl, S. J.; Robbins, W. D.; Bellwood, D. R. Exploring the Nature of Ecological Specialization in a
564 Coral Reef Fish Community: Morphology, Diet and Foraging Microhabitat Use. *Proc R Soc B* **2015**,
565 *282* (1815), 20151147.
- 566 (49) Guiguer, K.; Reist, J.; Power, M.; Babaluk, J. Using Stable Isotopes to Confirm the Trophic Ecology
567 of Arctic Charr Morphotypes from Lake Hazen, Nunavut, Canada. *J. Fish Biol.* **2002**, *60* (2), 348–
568 362.
- 569 (50) Ulrich, K. L. *Trophic Ecology of Arctic Char (*Salvelinus Alpinus* L.) in the Cumberland Sound Region*
570 *of the Canadian Arctic*; University of Manitoba (Canada), 2013.
- 571 (51) Snorrason, S. S.; Skúlason, S.; Jonsson, B.; Malmquist, H. J.; Jónasson, P. M.; Sandlund, O. T.;
572 Lindem, T. Trophic Specialization in Arctic Charr *Salvelinus Alpinus* (Pisces; Salmonidae):
573 Morphological Divergence and Ontogenetic Niche Shifts. *Biol. J. Linn. Soc.* **1994**, *52* (1), 1–18.
- 574 (52) Lu, G.; Bernatchez, L. Correlated Trophic Specialization and Genetic Divergence in Sympatric Lake
575 Whitefish Ecotypes (*Coregonus Clupeaformis*): Support for the Ecological Speciation Hypothesis.
576 *Evolution* **1999**, *53* (5), 1491–1505.
- 577 (53) Pothoven, S. A.; Nalepa, T. F. Feeding Ecology of Lake Whitefish in Lake Huron. *J. Gt. Lakes Res.*
578 **2006**, *32* (3), 489–501.
- 579 (54) Siwertsson, A.; Knudsen, R.; Adams, C. E.; Præbel, K.; Amundsen, P. Parallel and Non-parallel
580 Morphological Divergence among Foraging Specialists in European Whitefish (*Coregonus*
581 *Lavaretus*). *Ecol. Evol.* **2013**, *3* (6), 1590–1602.

- 582 (55) Jenjan, H. B.; Garduño-Paz, M.; Huntingford, F. A.; Adams, C. E. Gill Development in Sympatric
583 Morphs of Arctic Charr from Loch Awe, Scotland: A Hidden Physiological Cost of Macrobenthos
584 Feeding? *Ecol. Freshw. Fish* **2017**.
- 585 (56) Amundsen, P.-A.; Bøhn, T.; Våga, G. H. Gill Raker Morphology and Feeding Ecology of Two
586 Sympatric Morphs of European Whitefish (*Coregonus Lavaretus*); JSTOR, 2004; pp 291–300.
- 587 (57) Mackay, D.; Celsie, A. K.; Powell, D. E.; Parnis, J. M. Bioconcentration, Bioaccumulation,
588 Biomagnification and Trophic Magnification: A Modelling Perspective. *Environ. Sci. Process.*
589 *Impacts* **2018**.
- 590 (58) Amiard, J.-C. *Les Problèmes Liés à l'échantillonnage et à La Détection Des Éléments Traces En*
591 *Écotoxicologie*.; 1994.
- 592

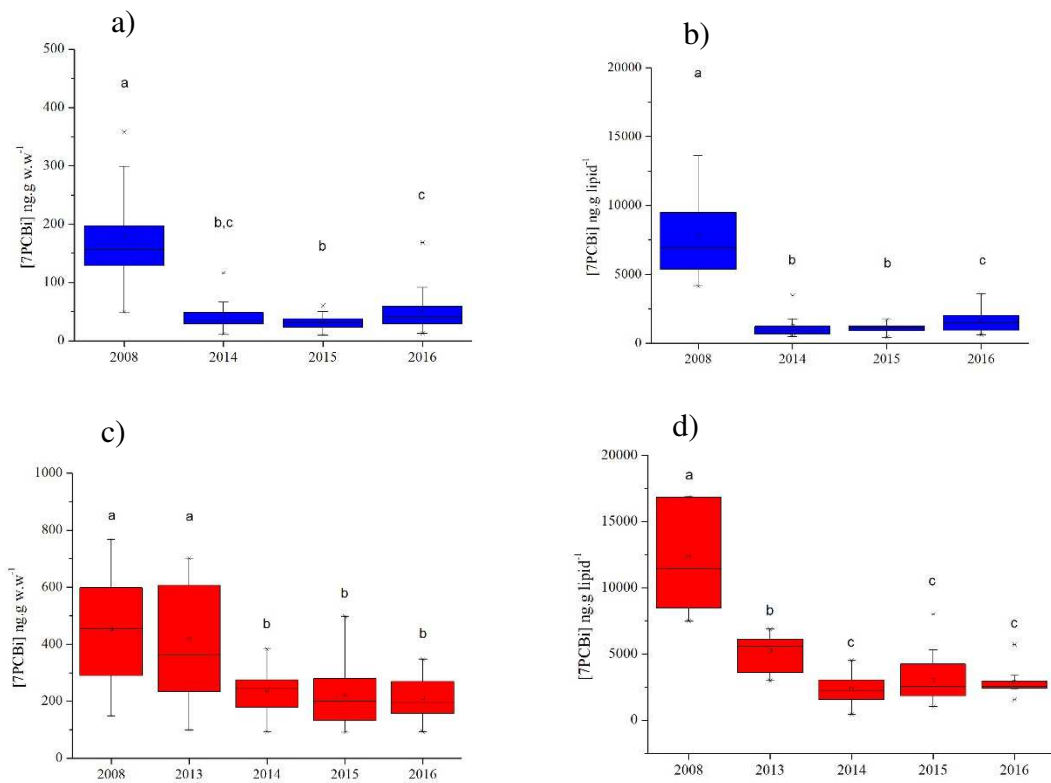


Figure 1: Boxplots of whitefish a) and b), and Arctic char c) and d) wet weight and lipid-normalized PCBi contamination. Letters are used to show statistically significant differences.

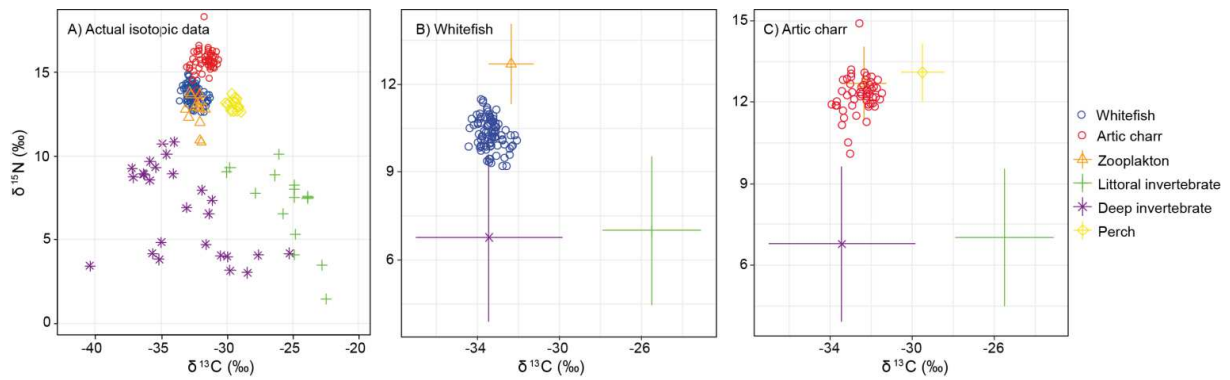


Figure 2: Isotopic space $\delta^{13}\text{C}$ - $\delta^{15}\text{N}$ showing the different isotopic measurements. A) Actual isotopic data, and isotopic values for B) whitefish and C) Arctic char corrected for isotopic fractionation, highlighting their embedding within isotopic values of putative preys (i.e., 96% and 89% of isotopic embedding for Arctic char and whitefish, respectively, when considering the standard deviation of $\pm 1\%$ for trophic fractionation). Vertical and horizontal lines in B) and C) indicate standard deviations of the isotopic values for the different resources considered in Bayesian mixing models.

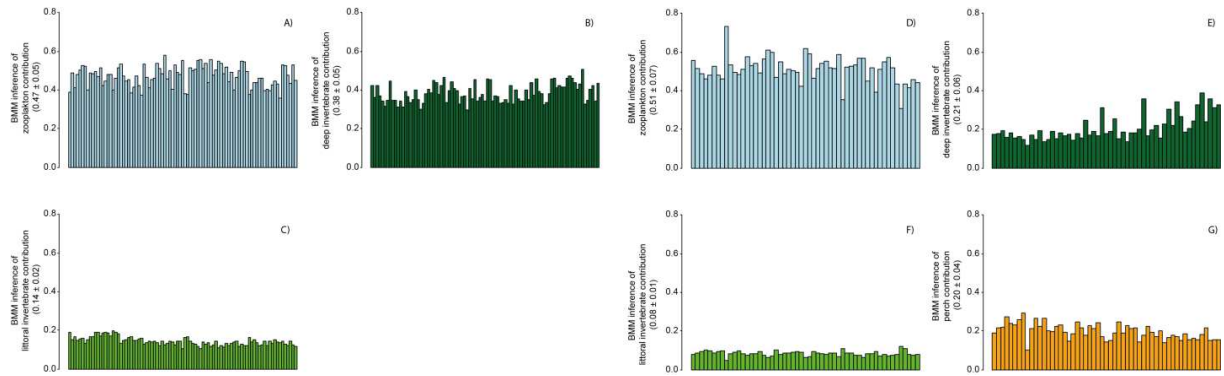


Figure 3: Barplots representing the proportions of the different resources consumed by each individual for the two fish species inferred from the BMMs. For whitefish, barplots represent A) contribution of zooplankton, B) contribution of deep invertebrates and C) contribution of littoral invertebrates. For Arctic char, barplots represent D) contribution of zooplankton, E) contribution of deep invertebrates, F) contribution of littoral invertebrates and G) contribution of yoy perch. Numbers in brackets indicate the mean \pm sd for the different resources.

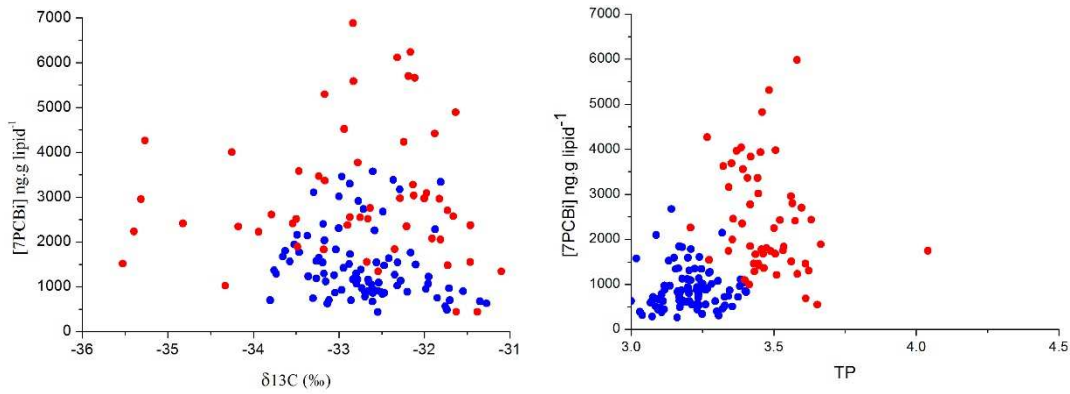


Figure 4: Plots of $\delta^{13}C$ (a) and TP (b) vs $\Sigma 7PCBi$ concentration for Arctic char (●) and whitefish (●).

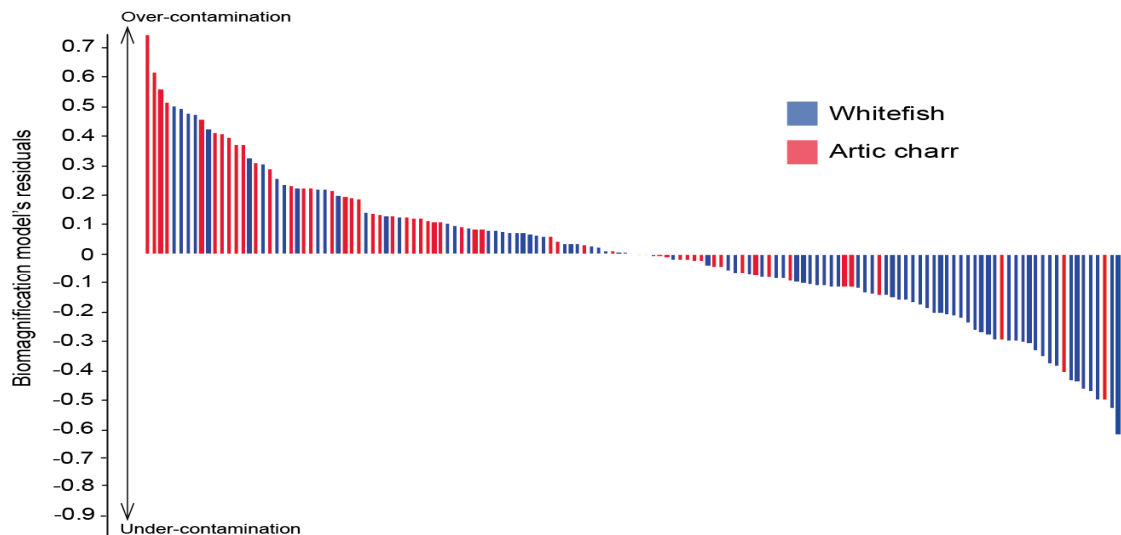


Figure 5: Individual residuals of the biomagnification model ranging from positive to negative residuals, representing a gradient of over- to undercontamination.

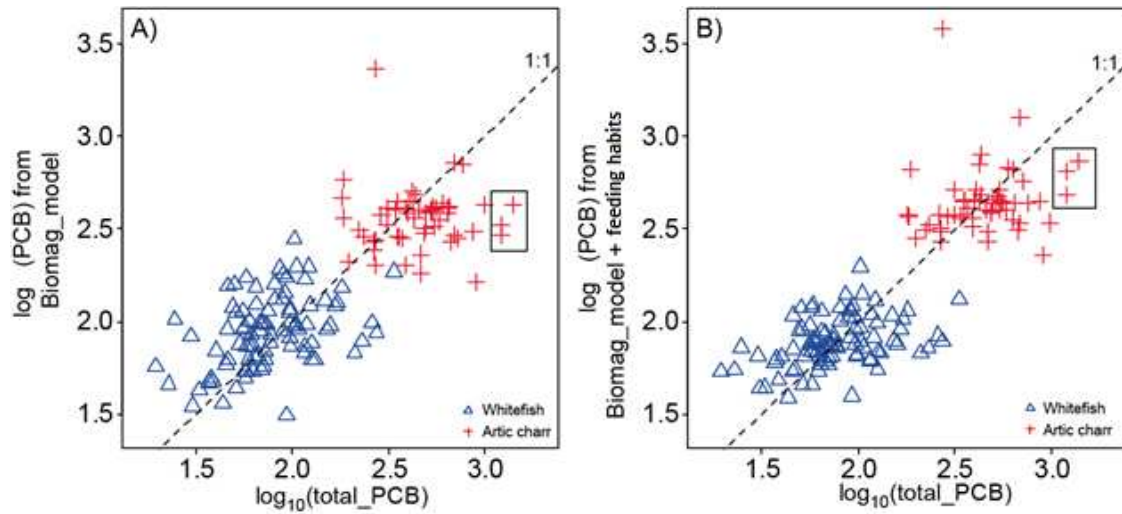


Figure 6: Biplot of A) the relationships between the predicted PCB concentrations by the biomagnification model (i.e., equation 4) and the measured total PCB concentrations and B) the relationships between the predicted PCB concentrations inferred by the biomagnification model corrected by the individual feeding habits (i.e., equation 8) and the measured total PCB concentrations. Boxes indicated the most important corrections applied to three Arctic char individuals.

