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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://hal.univ-smb.fr/hal-02165156>

Submitted on 22 Oct 2021

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

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

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

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

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

^d CISALB, 73000 Chambéry, France

Abstract

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

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

1. Introduction

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

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

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

The relative contribution of mechanisms by which contaminants bioaccumulate in fish has been extensively studied but remains quite uncertain in alpine lakes, where environmental conditions (carbon sources, temperature, contaminant concentrations) can greatly differ yearly and seasonally. Some studies could not link PCB levels in fish to any biomagnification processes, as PCB contamination was neither correlated with body size, living habitat nor TP, especially in mountain lakes. Indeed, Catalan et al.²³ questioned the relative importance of biomagnification in contamination of salmonids in high-altitude Lake Redo and suggested that bioconcentration could be a significant mechanism for PCB bioaccumulation. Perga et al.²⁴ concluded that PCB contamination may be mainly due to a nontrophic (i.e., bioconcentration) process in Lake Muzelle. In contrast, numerous studies have shown that a contaminated diet was responsible for bioaccumulation in fish in the American Great Lakes^{15,25,26} and that contamination levels increased with TP^{14,22}. As a lipophilic contaminant, differences in fat content result in differences in PCB contamination within the same species¹⁴. Living habitat and particularly carbon sources (benthic or planktonic) were environmental factors directly linked to discrepancies in PCB contamination between individuals and between species²⁰ when PCB contamination is heterogeneous between trophic habitats. Even though their work did not focus on PCBs, Kahilainen et al.²¹ went further and recently showed that ecomorphological divergences in whitefish could be responsible for a large part of the intraspecies variability in mercury bioaccumulation.

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

2. Material and Methods

2.1 Study site:

Lake Bourget is located on the northwest edge of the French Alps. It is the largest natural lake in France, 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 85 m (maximum: 145 m). Lake Bourget is a meso-oligotrophic lake with a mean temperature of 14.7°C 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 the conductivity is 299 µs.cm⁻¹ for the period 2015-2017^{32,33}. PCB inputs are mainly due to a point source located upstream on the Tillet River, which was contaminated from 1930 to 1987 by an electrical transformer factory using Pyralène™. Despite the cessation of the industrial activity and the recent clean-up of the downstream section of the river from 2012 to 2014, Lake Bourget is still contaminated. Even though contamination decreased in the last decades, superficial sediments remain polluted with a mean indicator PCB concentration (sum of PCB 28, 52, 101, 118, 153, 138, 180) of 12.1 ± 6.4 ng.g⁻¹ d.w, with yet heterogeneous contamination of the lake, reaching 113 ng.g⁻¹ d.w at the Tillet River outlet (Fig. S1). Consequently, the lake water still exceeds the USEPA recommended maximum concentration of 170 pg.L⁻¹ for the protection of aquatic wildlife²⁷, with a mean dissolved Σ7PCBi concentration of 322 ± 102 pg.L⁻¹.

2.2 Processing:

2.2.1 Sampling and preparation of organisms

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

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

2.2.2 Chemicals

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

2.2.3 PCB analysis

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

2.2.4 Stable isotope analysis

After sampling and dissection, fish filets, chironomids and zooplankton were dried at 50°C, ground into fine powder using a mortar and pestle and kept in tin capsules away from humidity prior to isotopic analysis. Samples in tin capsules were sent to SINLAB (NB, Canada). One mg was analyzed on a Finnigan Delta Plus mass spectrometer coupled with a ConFlo II NC2500 Elemental Analyzer for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ quantification. Samples were calibrated against 7 plant and animal laboratory standards covering a wide range of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values (i.e., from -35.08‰ to -13.25‰ and 0.02‰ to 16.14‰, respectively). Three replicates for each sample were run. The precision values of the C and N isotope analyses were <0.2‰.

2.2.5 Statistical analysis

All statistical tests were performed using the OriginPro 8 SR0 software and R 3.5.0 software. All data were tested for normality using the Shapiro test and, if appropriate, for homoscedasticity using the Bartlett test. When appropriate, data were log-transformed to ensure normality and homoscedasticity. Differences in fish PCB concentrations and fish size, lipid content, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ between year and season were tested using an ANOVA or the Kruskal-Wallis test. Post hoc comparisons were performed using Tukey's honest significant difference (HSD) or Dunnett tests. The relationship between fish PCB_i concentrations and the measured parameters (size, lipid content, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) were tested using Pearson correlations for normal variables or Spearman correlations otherwise.

2.3 Modeling:

2.3.1 Resource contributions

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

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

2.3.2 Trophic position

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

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

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

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

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

2.3.3 Biomagnification model

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

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

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

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

$$\text{Residuals} = \log_{10} \text{PCB}_{\text{obs}} - \log_{10} \text{PCB}_{\text{estimated}} \quad (5)$$

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

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

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

$$\text{Residuals} = f(\text{FH} + \varepsilon) \quad (7)$$

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

$$\log_{10} \text{PCB}_{\text{obs}} = \log_{10} \text{PCB}_{\text{estimated}} + \text{FH} + \varepsilon \quad (8)$$

3. Results and discussion

3.1 Temporal, interspecies and interindividual variability in PCB contamination:

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

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

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

3.2 Physiological parameters and relationship with PCB_i contamination:

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

3.3 Trophic plasticity of Arctic char and whitefish:

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

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

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

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

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

3.4 Trophic parameters and relationship with PCBi contamination:

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

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

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

3.5 Data comparison with a biomagnification model:

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

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

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

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

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

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

exhibit various morphs in the lacustrine systems^{52–54}. Recently, Kahilainen et al.²¹ showed that ecomorphological divergence could be responsible for differential mercury bioaccumulation in whitefish. Moreover, fish respiratory capacity can vary between species and also between individuals^{55,56}, leading to possible difference in PCB metabolism rates. Therefore, intra and interspecies variation in PCB bioaccumulation could be partially explained by intra and interspecies physiological differences at the gills level. Hence, the diversity of behaviors and morphology potentially encountered for the same population could be partly responsible for the large span of PCB contamination observed, as different individuals could develop in high or low contamination environments in Lake Bourget.

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

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

Acknowledgements

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

Author information

Corresponding author:

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

Supplementary material

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

References

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

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

- (37) Vander Zanden, M. J.; Cabana, G.; Rasmussen, J. B. Comparing Trophic Position of Freshwater Fish Calculated Using Stable Nitrogen Isotope Ratios ($\Delta^{15}\text{N}$) and Literature Dietary Data. *Can. J. Fish. Aquat. Sci.* **1997**, *54* (5), 1142–1158.
- (38) Parnell, A. Simmr: A Stable Isotope Mixing Model. R Package Version 0.3.1. 2016.
- (39) Post, D. M.; Layman, C. A.; Arrington, D. A.; Takimoto, G.; Quattrochi, J.; Montana, C. G. Getting to the Fat of the Matter: Models, Methods and Assumptions for Dealing with Lipids in Stable Isotope Analyses. *Oecologia* **2007**, *152* (1), 179–189.
- (40) Post, D. M. Using Stable Isotopes to Estimate Trophic Position: Models, Methods, and Assumptions. *Ecology* **2002**, *83* (3), 703–718.
- (41) Quezada-Romegialli, C.; Jackson, A. L.; Hayden, B.; Kahilainen, K. K.; Lopes, C.; Harrod, C. TRophicPosition, an r Package for the Bayesian Estimation of Trophic Position from Consumer Stable Isotope Ratios. *Methods Ecol. Evol.* **2018**, *9*, 1292–1299.
- (42) Opinion of the Scientific Panel on Contaminants in the Food Chain Related to the Presence of Non Dioxin-like Polychlorinated Biphenyls (PCB) in Feed and Food. *EFSA J.* **2005**, *3* (11).
- (43) Agence de l'eau RMC. Base de Données Micropolluants Du Programme PCB. Agence de l'eau RMC, FRANCE 2008.
- (44) Olsson, A.; Valters, K.; Burreau, S. Concentrations of Organochlorine Substances in Relation to Fish Size and Trophic Position: A Study on Perch (*Perca fluviatilis* L.). *Environ. Sci. Technol.* **2000**, *34* (23), 4878–4886.
- (45) Stow, C. A. Factors Associated with PCB Concentrations in Lake Michigan Fish. *Environ. Sci. Technol.* **1995**, *29* (2), 522–527.
- (46) Kiriluk, R. M.; Servos, M. R.; Whittle, D. M.; Cabana, G.; Rasmussen, J. B. Using Ratios of Stable Nitrogen and Carbon Isotopes to Characterize the Biomagnification of DDE, Mirex, and PCB in a Lake Ontario Pelagic Food Web. *Can. J. Fish. Aquat. Sci.* **1995**, *52* (12), 2660–2674.
- (47) Borgå, K.; Kidd, K. A.; Muir, D. C.; Berglund, O.; Conder, J. M.; Gobas, F. A.; Kucklick, J.; Malm, O.; Powell, D. E. Trophic Magnification Factors: Considerations of Ecology, Ecosystems, and Study Design. *Integr. Environ. Assess. Manag.* **2012**, *8* (1), 64–84.
- (48) Brandl, S. J.; Robbins, W. D.; Bellwood, D. R. Exploring the Nature of Ecological Specialization in a Coral Reef Fish Community: Morphology, Diet and Foraging Microhabitat Use. *Proc R Soc B* **2015**, *282* (1815), 20151147.
- (49) Guiguer, K.; Reist, J.; Power, M.; Babaluk, J. Using Stable Isotopes to Confirm the Trophic Ecology of Arctic Charr Morphotypes from Lake Hazen, Nunavut, Canada. *J. Fish Biol.* **2002**, *60* (2), 348–362.
- (50) Ulrich, K. L. *Trophic Ecology of Arctic Char (*Salvelinus Alpinus* L.) in the Cumberland Sound Region of the Canadian Arctic*; University of Manitoba (Canada), 2013.
- (51) Snorrason, S. S.; Skúlason, S.; Jonsson, B.; Malmquist, H. J.; Jónasson, P. M.; Sandlund, O. T.; Lindem, T. Trophic Specialization in Arctic Charr *Salvelinus Alpinus* (Pisces; Salmonidae): Morphological Divergence and Ontogenetic Niche Shifts. *Biol. J. Linn. Soc.* **1994**, *52* (1), 1–18.
- (52) Lu, G.; Bernatchez, L. Correlated Trophic Specialization and Genetic Divergence in Sympatric Lake Whitefish Ecotypes (*Coregonus clupeaformis*): Support for the Ecological Speciation Hypothesis. *Evolution* **1999**, *53* (5), 1491–1505.
- (53) Pothoven, S. A.; Nalepa, T. F. Feeding Ecology of Lake Whitefish in Lake Huron. *J. Gt. Lakes Res.* **2006**, *32* (3), 489–501.
- (54) Siwertsson, A.; Knudsen, R.; Adams, C. E.; Præbel, K.; Amundsen, P. Parallel and Non-parallel Morphological Divergence among Foraging Specialists in European Whitefish (*Coregonus 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 $\delta^{13}\text{C}$ (a) and TP (b) vs $\Sigma 7\text{PCBi}$ 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.