

Environmental context and contaminant biotransport by Pacific salmon interact to mediate the
bioaccumulation of contaminants by stream-resident fish

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Summary

1. The extent to which environmental context mediates the uptake of biotransported contaminants by stream-resident organisms is not understood. For example, there is no clear understanding of the extent to which contaminant type, instream characteristics, or resident fish identity interact to influence the uptake of contaminants deposited by Pacific salmon (*Oncorhynchus* spp.) during their spawning runs.
2. To address this uncertainty, we sampled four stream-resident fish species from 13 watersheds of the Laurentian Great Lakes in locations with and without salmon across a gradient of instream and watershed characteristics. We determined the polychlorinated biphenyl (PCB) and mercury (Hg) concentration along with the stable isotope ratio of C and N for each stream-resident fish.
3. We found that stream-resident fish PCB concentrations were 24-fold higher in reaches with salmon and were positively related to $\delta^{15}\text{N}$. In contrast, stream-resident fish Hg concentrations were similar or lower in reaches with salmon and either exhibited a negative or no relationship with $\delta^{15}\text{N}$.
4. Based upon AIC_c , stream-resident fish exhibited species-specific PCB concentrations that were positively related to salmon PCB flux. Hg burdens exhibited an interaction between fish length and salmon Hg flux – as salmon Hg inputs increased, Hg levels decreased with increasing resident fish length. We found no support for models that included the mediating influence of instream or watershed factors. Salmon eggs are enriched in PCBs but have very low Hg concentrations, so our results may be driven by the consumption of salmon eggs by stream-resident fish.

5. *Synthesis and applications.* Our results highlight that contaminants bioaccumulate differently depending on contaminant type, species identity, and the trophic pathway to contamination. Consequently, consideration of the recipient food web and route of exposure is critical to understanding the fate of biotransported contaminants in ecosystems. The transfer of contaminants by migratory organisms represents an understudied stressor in ecology. Effective management of biotransported contaminants will require the delineation of “hot-spots” of biotransport and implementation of best management practices in those watersheds that receive contaminants from spawning salmon.

Introduction

Animal migrations are important ecological phenomena responsible for the transport of nutrients and energy across ecosystem boundaries (Bauer & Hoyer, 2014). These predictable pulses of material influence the structure and function of reciprocal habitats (Polis et al., 2004), and mediate ecosystem stability and community biodiversity (Bauer & Hoyer, 2014). In addition, migratory animals are also a dispersal agent by which contaminants are relocated to different ecosystems (Blais et al., 2007). Pacific salmon (*Oncorhynchus* spp.) provide a compelling example of organisms controlling the flow of material to recipient ecosystems (Schindler et al., 2003). Salmon are effective as both a resource subsidy and contaminant biovector because they exhibit a semelparous and anadromous life-history strategy. Consequently, salmon deposit vast quantities of nutrient-rich but potentially contaminated carcass and egg material in locations where they spawn and die (Gerig et al., 2016). This material is consumed by a variety of organisms, including stream-resident fish (Janetski et al., 2009). However, as with all pulsed nutrient subsidies, the availability of salmon varies considerably across space and time due to environmental heterogeneity.

Freshwater ecosystems exhibit extensive spatial variation in abiotic and biotic characteristics. This variability, or *environmental context*, can directly control community structure and ecosystem function (Poff & Ward, 1990). In addition, environmental context strongly influences how contaminants bioaccumulate in freshwater food webs (Clements et al., 2012). Bioaccumulation varies in aquatic food webs as a function of three potentially interacting mechanisms. First, contaminant inputs to a watershed vary by landscape and instream characteristics, thereby influencing the quantity of contaminants available for bioaccumulation. Second, the rate of bioaccumulation can differ as a function of organism-specific diet,

physiology, or behavior. Last, differences in food web structure and food chain length can mediate the magnitude of bioaccumulation (Cabana et al., 1994, Lavoie et al., 2013). The rate of bioaccumulation among food webs can be assessed directly by relating the stable isotope ratio of nitrogen to the contaminant of interest thereby establishing how context mediates bioaccumulation (Clements et al., 2012). However, it remains unclear whether environmental context is a critical driver of salmon-mediated contaminant biotransport to stream-resident organisms such as fish.

The Laurentian Great Lakes offer a unique setting to study the role of context on salmon-mediated contaminant biotransport. Pacific salmon were introduced to the Great Lakes in the 1960s to control invasive prey fish, rehabilitate predator populations, and establish a recreational fishery (Crawford 2001, Dettmers et al., 2012). Since then, salmon have established naturally reproducing populations (Kerns et al., 2016), while being intensively managed by natural resource agencies (Tsehaye et al., 2014). As a result, managers are concerned about factors such as population instability and contaminant accumulation that might compromise the value of the fishery (Murphy et al., 2012). Contaminants are of particular concern in the Great Lakes due to the extensive legacy of industrial pollution that includes numerous Areas of Concern (Murphy et al., 2012). Hence, migratory species, such as salmon, represent a link for the transfer of lake-derived contaminants accumulated within muscle and gametic tissue to streams of the Great Lakes. While transfer of salmon-derived persistent organic pollutants (POPs) to stream biota has been documented in the Great Lakes (e.g., Gerig et al., 2016, Janetski et al., 2012), the significance of salmon transfer of mercury (Hg), or how physical, chemical and biological attributes of environmental context mediate the transfer and uptake of biotransported contaminants is unknown.

In this study, we evaluated whether environmental context mediates the transfer and uptake of salmon-derived contaminants to stream-resident fish across watersheds of the upper Laurentian Great Lakes. We first quantified PCB and Hg concentrations in tissue and eggs of Pacific salmon spawners, the putative source of contaminants. Second, we related stream-resident fish $\delta^{15}\text{N}$ to PCB and Hg concentrations to compare rates of contaminant bioaccumulation between locations with and without salmon. Last, we used an information theoretic approach to assess the mediating influence of various components of the environmental context on the relationship between salmon contaminant flux and stream-resident fish contaminant concentrations. We hypothesized, first, that salmon would accumulate high concentrations of PCBs and Hg in their muscle-tissue and eggs. Second, we predicted a positive relationship between $\delta^{15}\text{N}$ and contaminant concentration reflecting salmon as a significant source of both PCB and Hg. Last, we expected that specific attributes of the environmental context, which modify background watershed contaminant levels, enhance retention of salmon carcasses and eggs, and influence stream resident diet or behavior would control the uptake and bioaccumulation of biotransported contaminants by resident fish.

Materials and methods

Study sites

We sampled Pacific salmon and stream-resident fish during fall of 2013 and 2014 from 13 watersheds in the Lake Michigan and Lake Huron drainage basins of the Upper Great Lakes (Fig. 1, Table 1). Within each watershed, we selected one 300-m stream reach accessible (salmon reach) and one 300-m reach inaccessible (reference reach) to Pacific salmon (cf. Janetski et al., 2012). Reference reaches were located in adjacent streams within the same watershed or

upstream of dams within the same stream that served as salmon barriers. Reference sites were intended to control for non-salmon pollutant inputs. Sites represented a gradient of salmon spawner densities, stream characteristics, and landscape conditions representative of streams throughout the study area (Table S1). Each tributary accessible to salmon received a fall run of Chinook (*Oncorhynchus tshawytscha*) and coho (*Oncorhynchus kisutch*) salmon. Chinook salmon were the predominant spawners in our study streams. We sampled stream-resident fish species representative of the cold-water fish community, including brook trout (*Salvelinus fontinalis*), brown trout (*Salmo trutta*), mottled sculpin (*Cottus bairdii*), and rainbow trout (*Oncorhynchus mykiss*), although they were not always present at each sampling location (Table 1). All fish were collected using standard fisheries techniques including backpack electrofishing (cf. Pepino et al., 2012). Salmon were collected during the peak of the run in early-mid October and resident fish were collected 45-60 days after the spawning run peaked.

Environmental site characterization

A suite of biological, chemical, and physical variables was measured during stream habitat surveys conducted in September 2014 (Table S1). To determine watershed characteristics, a Geographic Information System (GIS) was used to measure watershed land cover (% Forested, % Wetland, % Developed) and watershed area (km², Table S1) from the National Land Cover Database 2011 using the HUC-12 watershed delineation (Homer et al., 2011). Additional watershed attributes were extracted from state geodatabases (e.g., Michigan Geographic Data Library, www.mcgi.state.mi.us/mgdl/). Instream habitat was assessed using both instream and GIS-based metrics (Table S1). For instream measurements, we established 30 lateral transects within each 300-m sampling reach. At each transect, we haphazardly measured five substrate

particles and assessed the total volume of large wood (m^3/m^2) that intersected transect (Lamberti & Gregory, 2006). In addition, physicochemical variables were measured at three locations within each reach using a multi-parameter probe (YSI, Yellow Springs, OH). At one location per reach, we collected filtered water samples ($n=3$) to determine dissolved nutrient concentrations. Stream gradient (m/km) was derived from a digital elevation model related to stream flow path. Water temperature was estimated from a regional groundwater model for mean July stream temperatures in Michigan (Wang et al., 2011). To assess biological context, we measured attributes related to both the salmon run and individual stream-resident fish. For salmon, we estimated salmon spawner density by counting all live and dead salmon in the wetted stream channel within each 300-m sampling reach accessible to salmon (cf. Chaloner et al., 2004). We multiplied counts of live and dead salmon by the mean salmon weight and mean salmon contaminant load in a given stream to estimate the total flux of contaminants delivered by salmon to the stream (cf. Janetski et al., 2012). For stream-resident fish, biometric data were measured, including species identity, length, and weight. Fish condition was determined from length and weight measurements.

Analytical chemistry

To determine stream nutrient status, water samples were filtered through a Whatman GF/F ($0.7 \mu\text{m}$) into polyethylene bottles, and stored frozen at $-20 \text{ }^\circ\text{C}$ for later analysis of soluble reactive phosphorus (SRP), nitrate (NO_3^- -N), and ammonium (NH_4^+ -N) (cf. Tiegs et al., 2008). A Lachat QC8500 Flow Injection Autoanalyzer (Lachat Instruments, Loveland, CO) was used to determine SRP, NO_3^- -N, and NH_4^+ -N concentrations. DOC samples were filtered through a Whatman GF/F ($0.7 \mu\text{m}$) into amber glass bottles, acid stabilized with hydrochloric acid, and

later measured on a Shimadzu TOC-5000 (Shimadzu Corp., Tokyo, Japan) at the Center for Environmental Science and Technology (CEST) (University of Notre Dame, Notre Dame, IN).

Polychlorinated biphenyl (PCB) concentrations of homogenized salmon (whole body, no gametes), salmon eggs, and stream-resident fish samples were determined using EPA method 1668 (USEPA Method 1668). Individual PCB congeners were quantified using an Agilent 6890 gas chromatograph (Agilent Technologies, Santa Clara, CA) equipped with a micro-electron capture detector. Instrumental conditions were previously described (Stapanian et al., 2013). This method determined a total of 89 PCB congeners. Total PCB concentration was calculated by summing the concentration of all 89 congeners and reported as ng/g wet weight. The instrument was calibrated with individual congener standards at 5 concentration levels (beginning at 0.10 ng/g) from AccuStandard (New Haven, CT). The West Coast Fish Studies standard (AccuStandard) was analyzed for calibration verification. Method blanks were run at a frequency of 1 per 20 samples, and the mean concentration (\pm SE) was 0.41 (\pm 0.08 ng/g) ng/g. Matrix spikes and matrix-spiked duplicates were also performed at a 5% frequency, with mean recovery and mean relative percentage difference equal to 81% (\pm 5%) and 11% (\pm 3%), respectively. Surrogate recoveries averaged $86 \pm 9\%$. Detection limits for the individual congeners were set at 3 times the baseline noise (\approx 0.01 ng/g). Detection limits were verified by analyzing a low-level standard at 0.025 ng/g, which yielded a signal to noise ratio between 12 and 15. See Table S2 for a list of PCB congeners measured.

Mercury concentrations of homogenized salmon (whole body, no gametes), salmon eggs, and stream-resident fish samples were determined using a Direct Mercury Analyzer 80 (DMA-80, Milestone S.r.l., Sorisole, Italy), located at CEST. Prior to analysis, all samples were freeze-dried, homogenized into a fine powder, and stored at -20°C . For analysis, 0.02 g of homogenized

sample was weighed into ashed nickel boats, placed into the DMA-80, and analyzed via fixed wavelength atomic absorption spectrophotometry (cf. Gerig et al., 2017). The DMA-80 was calibrated using standard reference materials (National Research Council of Canada, DORM-4, Ottawa, ON). Dry weight Hg concentration was converted to wet weight concentrations using the percent water content of each homogenized sample and expressed as ng/g wet weight. Quality control measures, including blanks, matrix spikes, matrix spiked duplicates, and standards were analyzed to ensure precision and accuracy of analyses. Percent recovery from DORM-4 standard was $100.6 \pm 6.8\%$ and the detection limit was 0.2 ng/g.

Carbon and nitrogen stable isotope and C:N ratios were measured for homogenized samples of salmon and stream-resident fish using an Elemental Analyzer (Costech, Valencia, CA) coupled to a Delta Plus Isotope Ratio Mass Spectrometer (Thermo Scientific, Waltham, MA) located at CEST. All samples were prepared for isotope analysis in the same manner as the Hg analyses. Isotope ratios were corrected within each individual run using a 3-point standard curve developed from known isotope standards (EA Consumables, Pennsauken, NJ). Standards used to develop the standard curve included wheat flour, sorghum, and protein. Stable isotope ratios of N ($\delta^{15}\text{N}$) and C ($\delta^{13}\text{C}$) were expressed as:

$$\delta^{15}\text{N} \text{ or } \delta^{13}\text{C} = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 1000 \quad (\text{Equation 1})$$

where R is the ratio of ^{15}N to ^{14}N or ^{13}C to ^{12}C . Tissue C:N data was determined using an acetanilide standard (cf. Chaloner et al., 2002). Data were deemed acceptable if the standard deviation of acetanilide standards during the run was <0.2 per mil (‰). The standard deviations for acetanilide standards were 0.14 and 0.09‰ for N and C, respectively. All $\delta^{13}\text{C}$ values were lipid-corrected using individual C:N ratios (cf. Post et al., 2007).

Statistical analyses

Two-way analysis of variance (ANOVA, $\alpha = 0.05$, Zar, 2010) was used to analyze salmon contaminant concentrations. The main factors were (1) contaminant type (PCB or Hg), and (2) tissue type (whole fish or eggs). We interpreted a significant interaction as evidence that contaminant accumulation was dependent on the contaminant and tissue type combination under consideration. Two-way ANOVA was also used to determine whether stream-resident fish $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ differed between the factors of location (salmon present or absent) and stream-resident species identity (brook trout, brown trout, mottled sculpin, or rainbow trout). Analysis of covariance (ANCOVA) was used to assess whether the relationship between stream-resident fish concentrations and $\delta^{15}\text{N}$ ratio was influenced by the factors of location (salmon present or absent), species identity (brook trout, brown trout, mottled sculpin, or rainbow trout), or contaminant type (PCB or Hg) (cf. Clements et al., 2012). An interaction between salmon presence, species identity, and contaminant type was used as the justification to conduct individual ANCOVAs for each species-contaminant combination. Assumptions of ANOVA and ANCOVA were assessed visually using plots of residuals (Zar, 2010), and all data was log-transformed prior to statistical analysis due to heteroscedasticity. All statistical analyses were performed using the R software platform (<https://cran.r-project.org/>).

To assess whether uptake of salmon-derived contaminants by stream-resident fish was influenced by environmental context, we used a generalized linear mixed modeling (GLMM) approach coupled with AIC_c model selection (cf. Johnson & Olmstad, 2004, Bolker et al., 2009). For each model, stream-resident fish contaminant concentration was considered the response variable and predictor variables included instream variables (substrate size, coarse wood debris volume, stream gradient, mean water temperature, DOC concentration, nutrient concentrations

[NO₃⁻N, NH₄⁺-N, SRP], pH), watershed characteristics (percent forested, percent wetland, percent urban, watershed area), resident fish biological variables (species identity, length, condition, δ¹⁵N, δ¹³C), and salmon run characteristics (contaminant flux). In all models, watershed was treated as a random effect to account for non-independence of fish sampled within the same watershed (cf. Swain et al., 2013). Stream-resident fish contaminant concentrations were log-transformed prior to analysis (cf. Bolker et al., 2009). We evaluated the relative support for GLMMs using an automated model selection procedure that solved all permutations of a global model (model fit with all predictor variables, Barton, 2012). To avoid overparameterization, we restricted our analysis by including a maximum of 4 predictor variables per model and only tested the interaction between salmon contaminant flux and individual variables related to instream, watershed, and biological characteristics (cf. Swain et al., 2013). Model performance and uncertainty were assessed using AIC_c, which ranks models based upon the principle of parsimony (Burnham & Anderson, 2002). The lower the AIC_c score for a given model, the better the trade-off in complexity and more optimal the fit between the model and data. Distinguishing between high-ranking models based on AIC_c values alone can be difficult, so we calculated model-averaged parameter estimates and relative variable importance (sum of AIC_c weights from all models containing a variable of interest) for fixed effects from models with a Δ AIC_c of less than 10 (Bolker et al., 2009). Last, we calculated a pseudo-R² for all models with Δ AIC_c less than 10; this metric represents the variance explained by fixed and random effects, and provides complimentary inference to the AIC analysis (Barton, 2012). Model selection was performed using the MuMIn package in the R software platform (Barton, 2012; <https://cran.r-project.org/>).

Results

Contaminant source: salmon isotope and contaminant patterns

Pacific salmon eggs and tissue had elevated $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ relative to stream-resident fish, irrespective of location (ANOVA, $\delta^{15}\text{N}$ $p < 0.001$, $\delta^{13}\text{C}$ $p < 0.001$, Table 2). Contaminant accumulation in Pacific salmon exhibited a strong interaction between contaminant and tissue type (ANOVA, $p < 0.001$, Fig. 2). Salmon eggs had 1.2-fold higher PCB concentrations relative to whole body concentrations. In contrast, Hg concentrations in salmon eggs were 15-fold lower than were whole body Hg concentrations. Overall, our results suggest that contaminant accumulation in salmon is dependent on the contaminant and tissue type considered.

Contaminant recipient: stream-resident fish isotope and contaminant patterns

The isotopic composition of stream-resident fish varied among species and between locations with and without salmon. Stream-resident fish in locations accessible to salmon exhibited higher isotope values than conspecifics in locations without salmon (ANOVA, $\delta^{15}\text{N}$ $p < 0.001$, $\delta^{13}\text{C}$ $p < 0.001$, Fig. 3). On average, brook trout, rainbow trout, and brown trout exhibited isotopic enrichment of 0.7‰, 1.8‰, and 1.8‰, respectively, for $\delta^{15}\text{N}$, and 1.5‰, 1.8‰, and 1.8‰, respectively, for $\delta^{13}\text{C}$, in locations with salmon compared to those without salmon. In contrast, mottled sculpin exhibited smaller increases of 0.2‰ for $\delta^{15}\text{N}$ and 0.5‰ for $\delta^{13}\text{C}$ in locations having salmon spawners.

The relationship between stream-resident fish contaminant concentration and $\delta^{15}\text{N}$ ratio was dependent upon salmon presence, species identity, and contaminant type (ANCOVA, $p < 0.001$, $R^2 = 0.77$, Fig. 4). As predicted, stream-resident fish PCB concentration was positively related to $\delta^{15}\text{N}$ (Fig. 4, Table 2). However, only brown trout exhibited a significant positive interaction between $\delta^{15}\text{N}$ and salmon presence (Fig. 4, Table 2). Across species, mean stream-

resident fish PCB concentrations in locations with salmon were 205.4 ± 23.4 ng/g (mean \pm SE) compared to 7.4 ± 0.5 ng/g in locations without salmon. The magnitude of this effect differed strongly among species (Table 2). Brown trout were 57-fold, rainbow trout were 29-fold, brook trout were 18-fold, and mottled sculpin were 8-fold more contaminated with PCBs in locations with salmon runs (Fig. 4).

Contrary to PCBs, we found no consistent relationship between Hg concentration and $\delta^{15}\text{N}$, either among locations or among stream-resident fish species (Fig. 4, Table 2). Brown trout again exhibited a significant interaction between $\delta^{15}\text{N}$ and salmon presence. In locations with salmon, Hg concentrations in brown trout decreased as $\delta^{15}\text{N}$ increased, in locations without salmon, brown trout Hg concentrations increased with increasing $\delta^{15}\text{N}$ (Fig. 4, Table 2). Brook trout Hg concentrations exhibited a similar, albeit non-significant, pattern to brown trout (Fig. 4). We found no evidence that salmon spawners increased stream-resident fish Hg concentrations. Across species, stream-resident fish exhibited similar or lower Hg concentrations in locations with salmon (70.8 ± 3.54 ng/g, mean \pm SE) compared to locations without salmon (83.1 ± 4.1 ng/g, Fig. 3).

Contaminant biotransport in relation to environmental context

Specific biological characteristics appeared to modulate the uptake and incorporation of salmon-derived contaminants by stream-resident fish. Stream-resident PCB concentrations were best explained by the interaction between species identity and salmon-derived PCB flux, and $\delta^{13}\text{C}$ (Table 3, Table S3). This model received a high AIC_c w_i (0.58) and explained a large proportion of variation (pseudo- $R^2=0.85$) in stream-resident fish PCB concentrations (Table 3). Furthermore, PCB concentrations of all stream-resident fish species exhibited a positive,

saturating relationship with salmon PCB flux into the system (Fig. 5). Stream-resident fish PCB concentrations were also positively related to $\delta^{13}\text{C}$, which better explained patterns than $\delta^{15}\text{N}$ (Table 3). The second ranked model also received a high $\text{AIC}_c w_i$, and was similar to the top model except that the instream volume of large wood was included as a covariate rather than $\delta^{13}\text{C}$ (Table 3). Overall, every model with an AIC_c less than 10 included the species identity-salmon PCB flux interaction. Physical or chemical variables including watershed area, stream temperature, percent forested land cover, and stream substrate size, were selected in 4 of 6 models with an $\text{AIC}_c < 10$ (Table 4). However, each of these models had low $\text{AIC}_c w_i$, low variable importance (0.02-0.04), and did not improve the proportion of variance explained by the model (Table S3), suggesting that their inclusion in the model did not substantially increase the model's explanatory power.

Factors that influenced Hg concentrations were distinct from those that affected PCB concentrations in stream-resident fish. The Hg concentration of stream-resident fish was best explained by the interaction between fish length and salmon-derived Hg flux, species identity, and the random effect of location (Table 3). The top Hg model received a high $\text{AIC}_c w_i$, but explained a much smaller proportion of variance when compared to the top PCB model (Table 3, Table S3; pseudo $R^2 = 0.32$). In contrast to the PCB model, salmon-mediated Hg flux did not directly influence stream-resident fish Hg concentrations (Table 3, Fig 5). The interaction between stream-resident fish length and salmon-derived Hg flux indicated that at low Hg fluxes, stream-resident fish Hg concentrations increased with increasing fish length (Fig. 6). However, as the flux of Hg supplied by salmon increased, Hg concentration in stream-resident fish decreased with increasing fish length. Other models that included biological, chemical, or

physical covariates received low $AIC_c w_i$ and were not considered for additional inference (Table 3).

Discussion

This study used both a persistent organic pollutant and a heavy metal to understand the influence of salmon-mediated contaminant biotransport on the bioaccumulation of stream-resident fish. Stream-resident fish from locations with salmon had higher PCB concentrations (Gregory-Eaves et al., 2007, Janetski et al., 2012) and exhibited isotopic enrichment (Schlundt & Hershey, 1995, Chaloner et al., 2002). Moreover, stream-resident fish PCB concentrations were strongly linked to the PCB flux delivered by spawning salmon, although this relationship was variable among species and individuals. PCB concentrations of resident fish from our study were dramatically higher compared to fish from studies in the native range of salmon (cf. Gregory-Eaves et al., 2007, this study) but similar to a previous study from the Great Lakes (Janetski et al., 2012). In contrast, salmon-mediated transport of Hg was associated with decreases in Hg concentrations of stream-resident fish. Our result corresponds with a study from Alaska where resident fish had lower Hg concentrations in locations with salmon relative to control sites (Baker et al., 2009, Cyr et al., 2016). In addition, contaminant bioaccumulation in stream-resident fish was mediated by environmental context related to a few select biological attributes of stream-resident fish and characteristics of the salmon run. Overall, our findings suggest the outcome of salmon-mediated contaminant biotransport is dependent on the class of contaminant considered and species-specific attributes which regulate contaminant uptake by resident fish in recipient ecosystems where salmon spawn.

Context dependent contaminant biotransport

The conceptual model proposed by Clements et al. (2012) provides a useful construct to consider how bioaccumulation of biotransported contaminants differs from the traditional model of food web bioaccumulation. Our PCB results conform to the Clements et al. (2012) model, with PCB concentrations being positively related to $\delta^{15}\text{N}$ and the magnitude of this effect being mediated by the flux of PCBs supplied by salmon suggesting context dependence. However, counter to the model, the relationship between $\delta^{15}\text{N}$ and Hg was inconsistent between species and locations. In the presence of salmon, Hg concentrations either decreased with increasing $\delta^{15}\text{N}$ (e.g., brown trout, rainbow trout), or exhibited no relationship (e.g. brook trout, mottled sculpin), while all stream fish from locations without salmon exhibited a positive relationship between $\delta^{15}\text{N}$ and Hg. These Hg results differ from Clements et al. (2012) model and a global meta-analysis (Lavoie et al., 2013), which found a ubiquitous positive relationship between $\delta^{15}\text{N}$ and Hg in aquatic systems, across both biomes (e.g., arctic, temperate, tropical) and ecosystems (e.g., streams, lakes, estuaries). The counterintuitive pattern of bioaccumulation in salmon streams may be a function of differences in the chemistry of organic contaminants and heavy metals, and individual variation in diet.

We observed differential accumulation of PCBs and Hg between different types of salmon tissue. Salmon eggs had high concentrations of PCBs but low Hg concentrations. In contrast, salmon whole body samples have lower PCB and much higher Hg concentrations relative to eggs. PCBs are highly lipophilic and accumulate readily into lipid-rich tissues, including salmon eggs (Blais et al., 2007, Walters et al., 2016). In contrast, Hg accumulates by forming a strong bond with cysteine in muscle tissue (Kuwabara et al., 2007), which is largely absent from salmon eggs leading to low Hg concentrations in salmon eggs (Zhang et al., 2001).

As such, we hypothesize that consumption of salmon eggs over salmon carcass tissue could lead to a tradeoff in the bioaccumulation of PCBs and Hg in resident fish. Previous studies have demonstrated that stream-resident fish in the Great Lakes readily consume salmon eggs with considerable variation among individuals within the same stream (Ivan et al., 2011, Johnson et al., 2016). As a consequence, egg consumption was a key factor in predicting the PCB concentrations of trout subject to salmon runs (Merna 1986). In contrast, consumption of eggs may mediate a reduction in Hg concentrations in resident fish. Cyr et al., (2016), observed that increased egg consumption was negatively correlated to resident fish Hg concentrations in a study from SE Alaska. Prior studies have hypothesized that consumption of carcass tissue is a primary route for energy acquisition (Collins et al., 2016) and contaminant exposure to stream-resident fish (Gerig et al. 2017). However, if resident fish were consuming large quantities of carcass material, then we would observe a concomitant increase in both PCB and Hg concentrations due to the high PCB and Hg concentrations in salmon tissue (Christensen et al., 2005, Gerig et al., 2017). Overall, our findings suggest that differential accumulation of contaminants between salmon tissue types coupled with a diet selection could be a primary determinant of the magnitude of bioaccumulation of biotransported contaminants by stream-resident fish.

Biological context overshadows physical or chemical variables

Biological variables overwhelmed the influence of instream or watershed characteristics on stream-resident fish bioaccumulation. As a result, the influence of watershed or instream characteristics, which typically mediate the bioavailability, and bioaccumulation of contaminants were marginal compared with the salmon effect (King et al., 2004, Jardine et al., 2012). This was

particularly evident for PCBs and may reflect the interaction between low environmental concentrations of PCBs relative to the large flux of PCBs supplied by salmon during spawning. The stronger influence of biological over physicochemical factors on resident fish also parallels other studies of salmon spawners where salmon run size was the most important variable controlling the stable isotope ratios of resident fish (Reisinger et al., 2013, Swain et al., 2013).

Stream-resident fish exhibited a logarithmic relationship between salmon PCB flux and stream-resident fish PCB burden with streams that received larger salmon runs being more contaminated with PCBs. However, resident fish species-identity was a strong mediating factor influencing the magnitude of the salmon effect. Variation in resident fish contaminant concentration could arise from variation in diet, habitat use, and physiology (McGill et al., 2017). Contaminant bioaccumulation in fish is strongly controlled by diet, as diet is the chief route of contaminant exposure in higher-level vertebrates (Madenjian et al., 2016). Stream-dwelling salmonids exhibit considerable dietary plasticity and adaptive ration size that allows them to exploit rare, but high magnitude resource pulses including salmon material (Armstrong et al., 2013, Jaecks et al., 2014) thereby increasing their PCB exposure (Janetski et al., 2011). The high level of variation observed in individual fish PCB concentrations, particularly at low salmon PCB fluxes, supports the idea of diet variability.

Differences between brook, brown, and rainbow trout PCB concentrations may also reflect the variation in spatial distribution and habitat use. For instance, brown trout had 2-fold higher PCB concentrations when compared to brook trout. Our sampling revealed that brown trout were spatially segregated from brook trout (cf. Fausch & White, 1981), generally occupying habitats lower in watersheds with 4-fold larger salmon runs than areas higher in

watershed. Consequently, brown trout have a higher probability of interacting with spawning salmon, presumably enabling brown trout to become more contaminated than brook trout.

The only non-salmonid sampled in this study, mottled sculpin, exhibited the weakest relationship between PCB concentration and salmon PCB flux. Freshwater sculpin are small benthic fish with a relatively large gape that forage on salmon material when available (Swain et al., 2013). However, sculpin may lack the dietary plasticity or gut capacity of stream salmonids, which limits their ability to gorge on resource pulses provided by salmon (Armstrong & Schindler, 2013). In addition, as a result of physiological differences, sculpin may have higher elimination rates of PCBs than salmonids (Stapleton et al., 2001). A related freshwater species, deepwater sculpin (*Myoxocephalus thompsonii*) can metabolize PCBs, thereby reducing their contaminant burden (Stapleton et al., 2001). Last, the benthic orientation of sculpin may make them susceptible to displacement during redd construction when salmon spawn, reducing their access to salmon material (Moore et al., 2004).

Contaminant biotransport models revised

The process of contaminant biotransport is defined by several steps including: (1) the contaminant is bioaccumulated by a migratory organism; (2) the contaminant is transported across an ecosystem boundary; and (3) the contaminant is deposited into the recipient ecosystem (Blais et al., 2007, Kallenborn & Blais, 2015). We propose that this model should be modified to include an additional step focused on factors which influence the uptake and subsequent bioaccumulation of contaminants by resident organisms in the recipient ecosystem (Fig. 7). Specifically, a hierarchy of factors determines the effect that migratory animals will have on resident organisms, either magnifying or modulating bioaccumulation. First, the abundance and

contaminant load of the migratory animal determines the flux of pollutants deposited and available for uptake by resident organisms (Janetski et al., 2012, this study). Second, the contaminant legacy of the recipient ecosystem determines the susceptibility to biotransport impacts; ecosystems that are less contaminated may have greater risk of contaminant biotransport. Third, the trophic pathway by which energy and contaminants move within the recipient ecosystem food web influences the overall magnitude of contaminant bioaccumulation by resident organisms (Cyr et al., 2016, this study); direct consumption of biotransported material magnifies impacts. Last, individual traits including species identity, diet, physiology, depuration rates, spatial distribution, and habitat use that vary among resident organisms will interact with flux of contaminants supplied by salmon, legacy contamination, and the trophic pathway to determine the magnitude of bioaccumulation of biotransported contaminants (Clements et al., 2012, Michelutti et al., 2010, this study). Refinement of the individual factors that contribute to this broader conceptual model provides insight into the consequences of the bulk movement of material and associated contaminants carried by migratory organisms.

The migratory organism transporting the contaminants also impacts the magnitude of contaminant biotransport (Michelutti et al., 2010). Blais et al. (2007) suggest that iteroparous fish, such as steelhead, or Atlantic salmon (*Salmo salar*), represent a diminished risk for contaminant biotransport because they only deliver gametic tissues along with excretory products to recipient ecosystems as compared with semelparous species, such as Pacific salmon, that also deliver carcasses. Arguably, because the flux of material is relatively small with iteroparous species, the corresponding impact on resident fish contaminant burdens is diminished. However, our results suggest that egg consumption may disproportionately drive PCB accumulation by stream resident-fish, and therefore migratory species that survive

reproduction may also pose a risk to resident organisms. For example, over 50 fish species exhibit a migratory life history and use tributaries for spawning in the Great Lakes (Lane et al., 1996). These species differ markedly in traits related to fecundity, spawning mode, run timing, energy density, and mobility, which may all interact to influence the flux of material to recipient ecosystems (Childress et al., 2015, Janetski et al., 2012). Knowing the risk imparted by different migratory fish runs is important to managing inputs of biotransported contaminants.

Management of biotransported contaminants will likely require different mitigation strategies than those already used to manage environmental contamination (cf. Qi et al., 2014). At present, contaminant biotransport by salmon is not monitored by state or federal management agencies, in part because biotransported contaminants often defy the conventional paradigm of pollution flowing from upstream to downstream. However, literature on managing non-point sources of nutrients may be relevant to reducing contaminant biotransport by migratory fish. For instance, non-point sources can be effectively managed when threshold levels are defined, “hot-spots” are identified, and best management practices are implemented (Carpenter et al., 1998). This approach translates well to the challenges of contaminant biotransport, where managers could (1) set threshold contaminant concentrations for streams receiving migratory fish; (2) identify locations with large salmon runs and high resident fish contaminant burdens for potential advisories; (3) use adaptive stocking strategies to minimize contaminant transfer to sites of conservation concern; and (4) implement seasonal removable barriers that limit the upstream flux of contaminants by spawning salmon. Our research therefore facilitates a greater awareness of the need for management of contaminant biotransport by state and federal regulatory agencies.

The delivery of resource subsidies has been widely recognized as critical to ecosystem structure and function (Bauer & Hoye, 2014, Polis et al., 2004). Our study demonstrates that

ecosystem linkages created by spawning salmon have significant consequences for resident fishes in tributaries. Previous work salmon on migrations has emphasized the translocation of nutrients and energy (e.g., Schindler et al., 2003, Janetski et al., 2009). By contrast, the movement of contaminants across ecosystem boundaries, called the ‘dark side of ecosystem resource subsidies’ (after Walters et al., 2008, Kraus et al., 2015), has been understudied relative to subsidy effects (Blais et al., 2007, Walters et al., 2008). Overall, our results highlight that contaminant transport by a migratory organism can have large consequences for recipient ecosystems with the effects being mediated by the contaminant being transported, the stream-resident fish being considered, and the pathway of trophic transfer.

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277 Table 1. Site characteristics of watersheds sampled to evaluate the effect of Pacific salmon contaminant biotransport on stream-
 278 resident fish. Sample size refers to the total number of stream-resident fish sampled. BKT=brook trout, BNT= brown trout,
 279 RBT=rainbow trout, MTS=mottled sculpin. The order of resident species present reflects abundance in each watershed.

Watershed	Basin	Watershed Area (km ²)	Salmon Biomass (kg/m ²)	PCB Flux (ng/m ²)	Hg Flux (ng/m ²)	Resident Species	Sample Size
Albany	Huron	103.26	0.15	5.74	8.09	BKT, MTS	6
Betsie	Michigan	118.73	0.18	100.34	40.79	BNT, RBT, MTS	16
Black	Michigan	86.64	0.02	7.63	5.81	RBT, MTS, BND	4
Boardman	Michigan	40.53	0.15	53.11	38.56	BNT, MTS	15
Boyne	Michigan	138.69	0.40	139.18	68.02	BNT, MTS	14
Carp	Huron	58.82	0.01	2.06	1.27	BKT, RBT, MTS, BND	7
Crow	Michigan	89.99	0.08	34.64	19.03	BKT, MTS, BND	7
Elliot	Huron	156.76	0.00	0.00	0.00	BKT, MTS, BND	4
Kalamazoo	Michigan	90.22	0.15	54.03	36.60	BNT, MTS, BKT	14
Manistee	Michigan	139.24	0.63	291.17	136.43	BKT, MTS, BNT	21
Muskegon	Michigan	147.75	0.02	8.29	6.48	BKT, MTS	19
Thompson	Michigan	48.10	0.21	82.07	41.43	BNT, MTS, RBT	9
White	Michigan	72.22	0.00	0.00	0.00	BKT, BNT	14

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282 Table 2. Results of ANCOVA assessing if salmon presence influences the relationship between
 283 stream-resident fish contaminant concentration and $\delta^{15}\text{N}$ ($\alpha=0.05$). Significant relationships
 284 shown in bold.
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Effect	PCB			Hg		
	df	F	P	df	F	P
Brook Trout						
$\delta^{15}\text{N}$	1, 48	4.3	0.040	1, 48	0.2	0.650
Salmon Presence	1, 48	51.0	0.001	1, 48	0.3	0.570
$\delta^{15}\text{N}$ *Salmon Presence	1, 48	0.1	0.810	1, 48	1.9	0.164
Brown Trout						
$\delta^{15}\text{N}$	1, 35	562.2	0.001	1, 35	0.3	0.622
Salmon Presence	1, 35	405.0	0.001	1, 35	18.0	0.001
$\delta^{15}\text{N}$ *Salmon Presence	1, 35	5.7	0.022	1, 35	31.3	0.001
Mottled Sculpin						
$\delta^{15}\text{N}$	1, 37	9.2	0.004	1, 37	4.5	0.041
Salmon Presence	1, 37	48.0	0.001	1, 37	4.8	0.035
$\delta^{15}\text{N}$ *Salmon Presence	1, 37	0.3	0.587	1, 37	0.8	0.360
Rainbow Trout						
$\delta^{15}\text{N}$	1, 14	43.0	0.001	1, 14	0.9	0.542
Salmon Presence	1, 14	6.8	0.020	1, 14	42.5	0.001
$\delta^{15}\text{N}$ *Salmon Presence	1, 14	0.3	0.890	1, 14	0.1	0.940

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287 Table 3. AIC_c model selection table for generalized linear mixed models used to explain stream-
 288 resident fish PCB and Hg contaminant concentrations. K=number of parameters in model.
 289 Psuedo-R² represents the proportion of variance explained by fixed effects in the GLMM model.
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Contaminant	Model	K	Δ AIC	AIC w _i	psuedo-R ²
PCB	Species x PCB flux, δ ¹³ C	11	0.00	0.578	0.85
	Species x PCB flux, Large wood volume	11	1.27	0.307	0.82
	Species x PCB flux, Watershed area	11	5.53	0.036	0.83
	Species x PCB flux, Temperature	11	5.61	0.035	0.84
	Species x PCB flux, % Forested land cover	11	6.74	0.020	0.82
	Species x PCB flux, Substrate size	11	7.23	0.016	0.84
	Species x PCB flux, δ ¹⁵ N	11	10.08	0.004	0.83
	Species x PCB flux, SRP (ug/L)	11	11.47	0.002	0.82
	Species x PCB flux	10	11.47	0.002	0.82
	Species x PCB flux, Length	11	12.13	0.001	0.82
Hg	Length x Hg flux, Species	9	0.00	0.977	0.32
	Species, Length, Hg flux	8	11.04	0.004	0.25
	Species, Length, Hg flux, % Forested land cover	9	11.08	0.004	0.28
	Species, Length, Hg flux, Watershed area	9	11.60	0.003	0.35
	Species, Length	7	11.96	0.002	0.16
	Species, Length, Hg flux, δ ¹³ C	9	12.20	0.002	0.27
	Species, Length, Watershed Area	8	12.32	0.002	0.21
	Species, Length, Hg flux, % Wetland land cover	9	12.42	0.002	0.25
	Species, Length, δ ¹³ C	8	12.49	0.002	0.17
	Species, Length, Hg flux, δ ¹⁵ N	9	12.54	0.002	0.26

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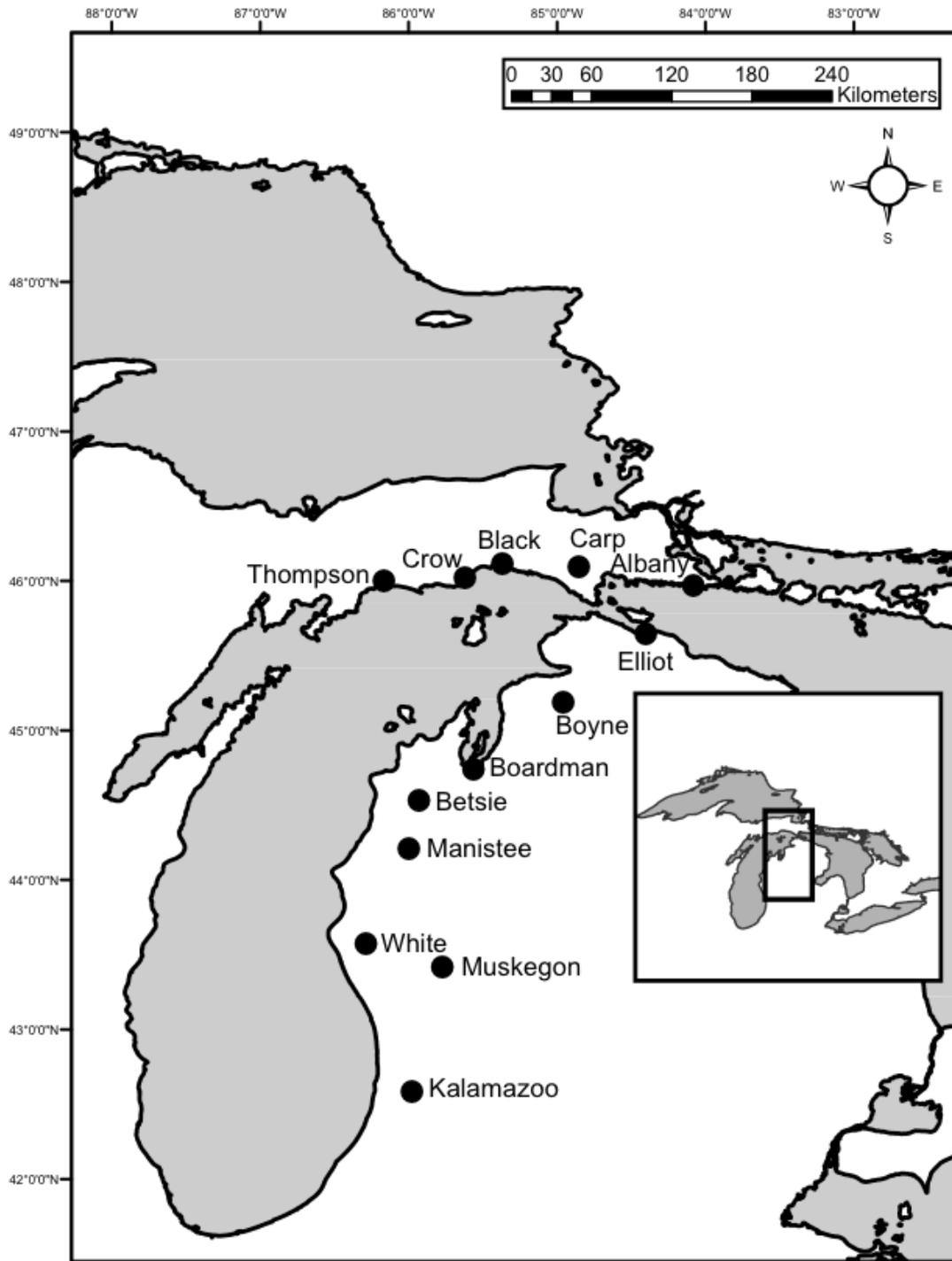


Figure 1. Location of 13 watersheds sampled to evaluate the mediating role of environmental context on salmon contaminant biotransport. See Table 1 and S1 for watershed covariates.

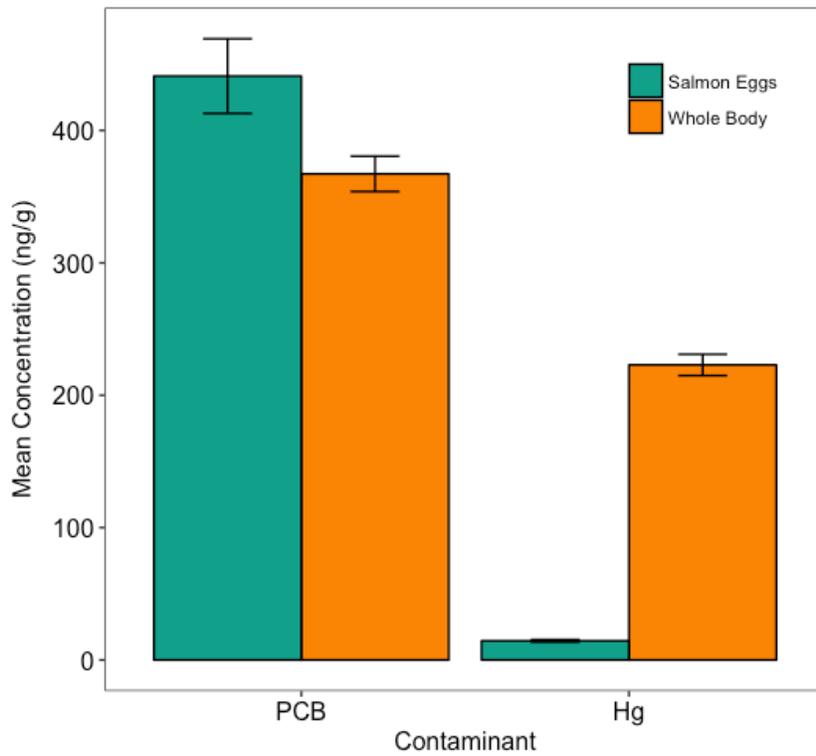


Figure 2. PCB and total Hg concentrations (ng/g wet weight) in whole body and egg samples of spawning Chinook and Coho salmon collected from tributaries of the Upper Great Lakes.

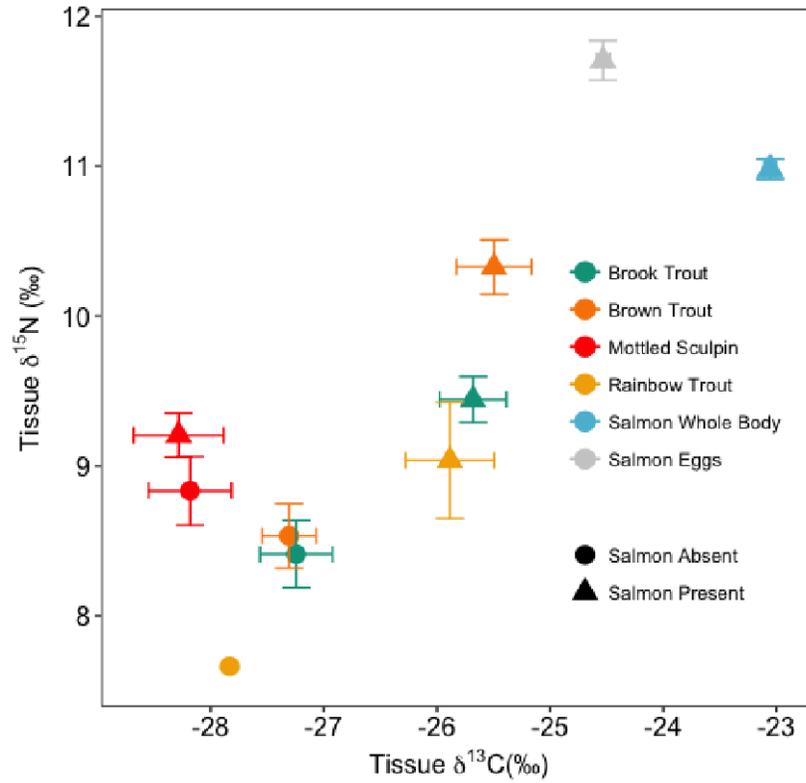


Figure 3. Stream-resident fish isotopic ratios in locations with and without Pacific salmon. Salmon whole body and egg isotopic ratios included for comparison. All values represented as mean (\pm standard error).

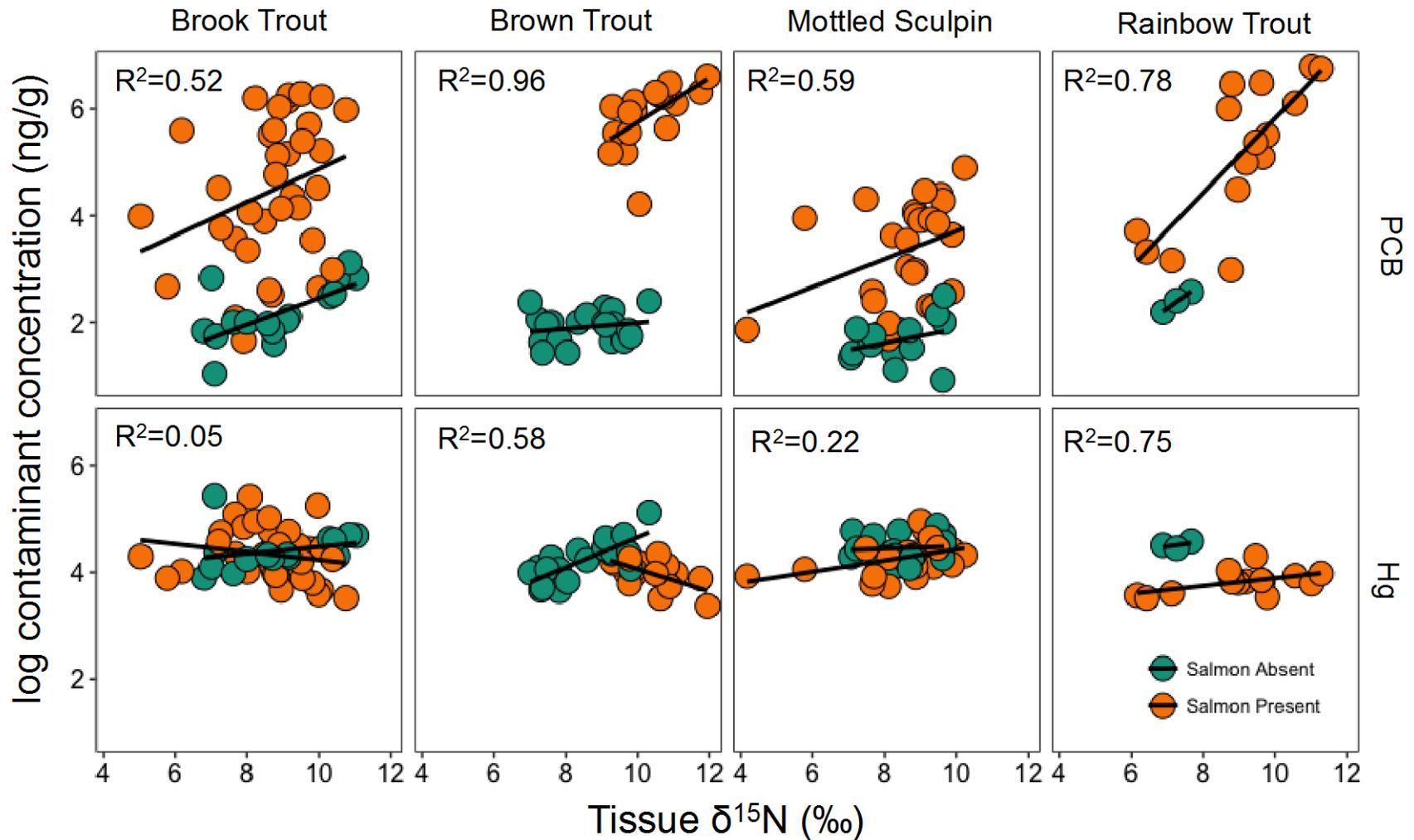


Figure 4. Relationship between contaminant concentration (PCB or Hg, ng/g wet weight) and $\delta^{15}\text{N}$ in stream-resident fish from reaches with and without salmon. Color denotes salmon presence (orange) or absence (green). Lines are least-squares regressions. See Table 3 for species and contaminant specific ANCOVA results.

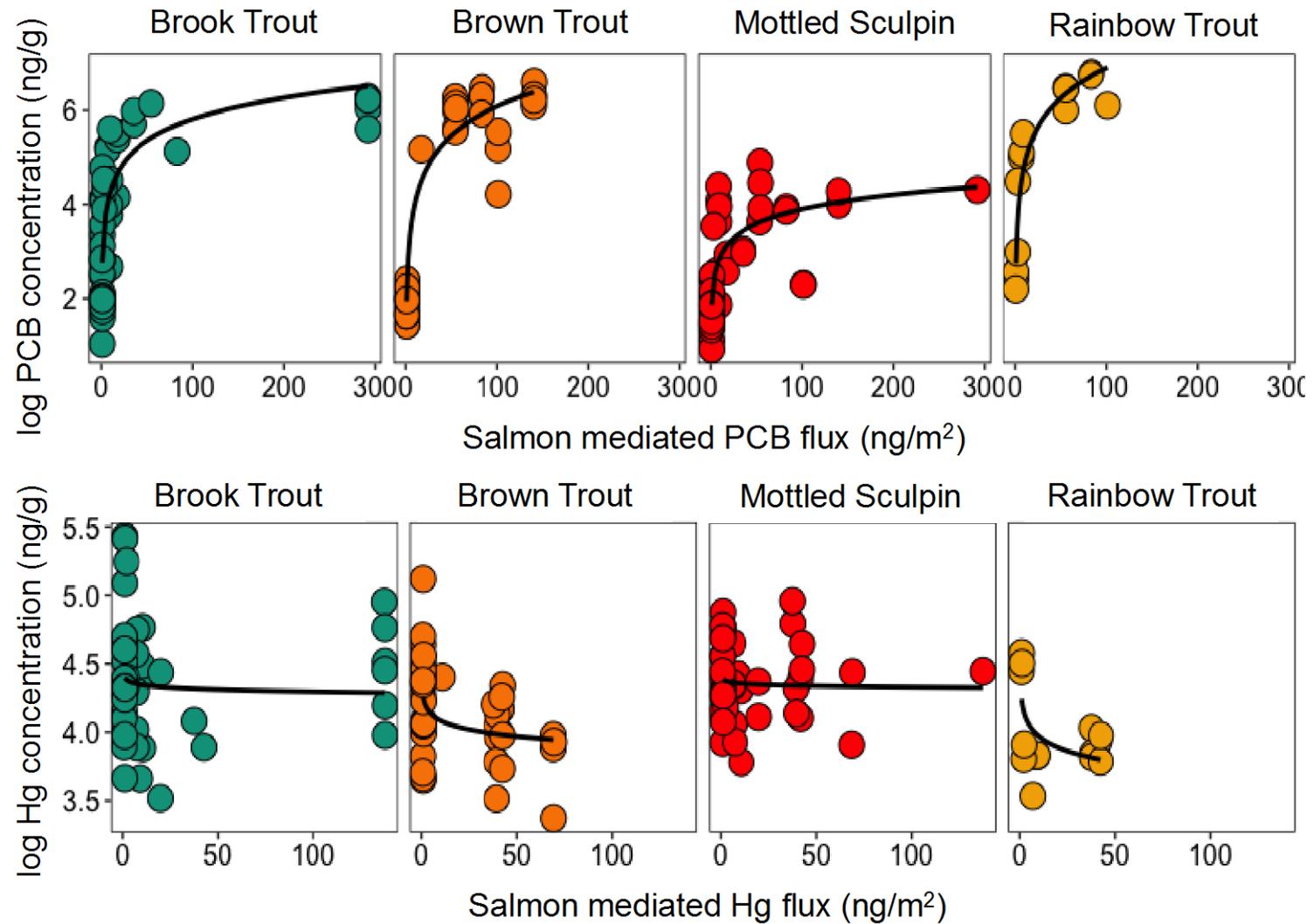


Figure 5. Relationship between log PCB concentrations (ng/g) in stream-resident fish and salmon-mediated PCB flux (ng/m²) supplied by spawning salmon. PCB flux calculated from visual abundance estimates, biometric data, and mean PCB concentration of salmon from a given watershed.

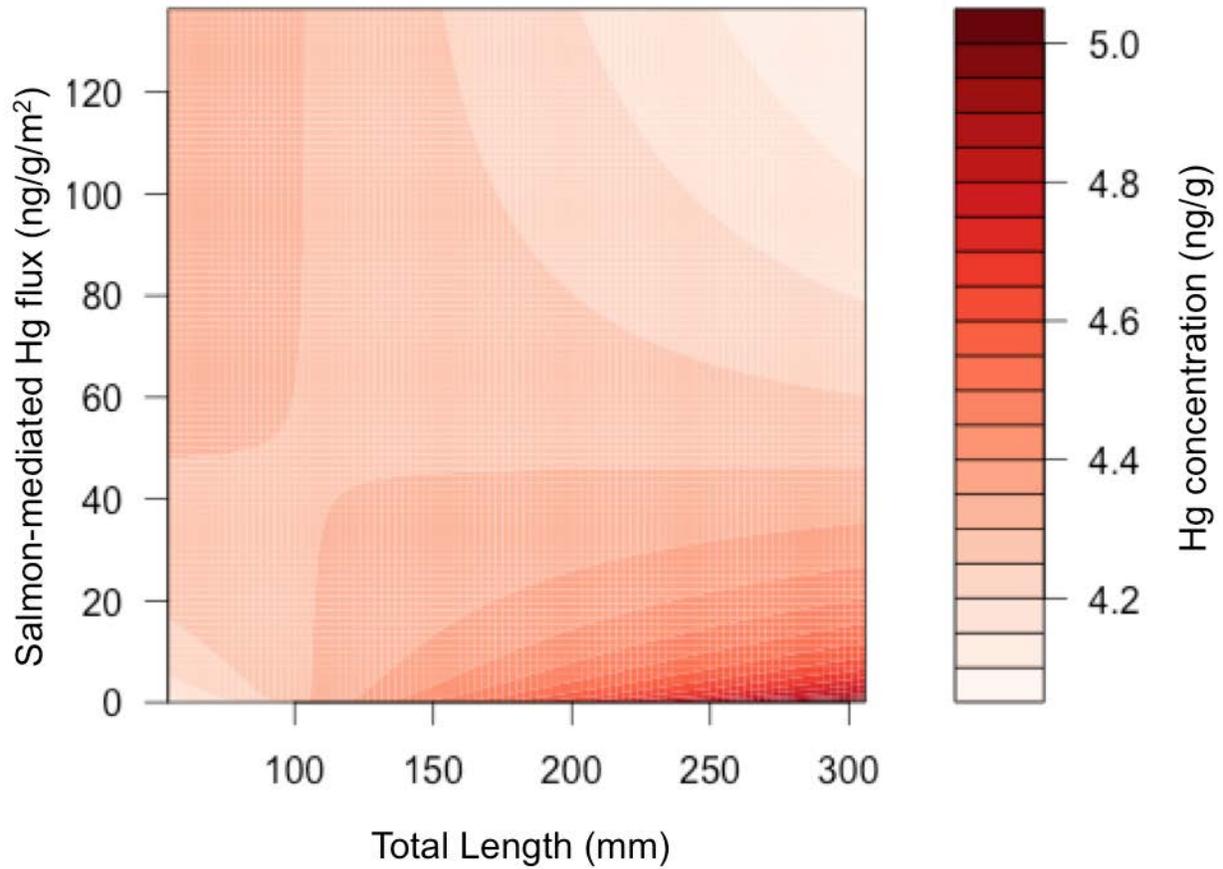


Figure 6. Interaction plot between the salmon-mediated Hg flux and stream-resident fish total length. Log of stream-resident fish Hg concentration (ng/g) is represented within the heat map. Note that stream-resident fish mercury concentrations increase with increasing fish length at low salmon-mediated Hg fluxes. At high salmon-mediated Hg fluxes, stream-resident mercury concentrations decrease with increasing fish length.

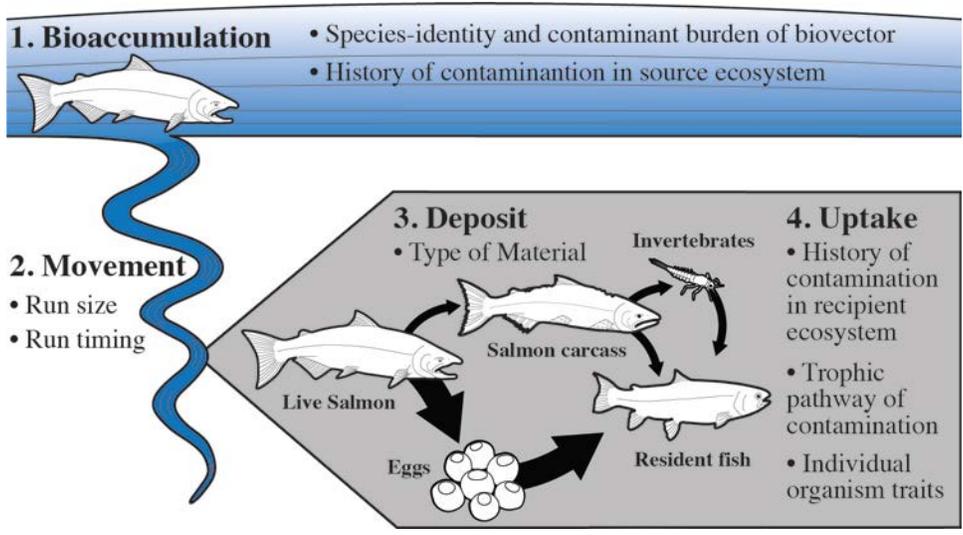


Figure 7. Conceptual model of the influence of context on contaminant biotransport by Pacific salmon. The process of contaminant biotransport is described by Blais et al. (2007) as a three-step process whereby a contaminant is bioaccumulated, moved, and deposited into a recipient ecosystem by a migratory organism. Here we revise this model and add a fourth step focused on the processes of contaminant uptake by resident organisms.