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Resident Fishes Display Elevated Organic Pollutants in Salmon Spawning Streams of the Great Lakes

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Supporting Information

ABSTRACT: Pacific salmon (*Oncorhynchus* spp.) can transport bioaccumulated organic pollutants to stream ecosystems where they spawn and die. We quantified PCBs, DDE, and PBDEs in resident fishes from 13 Great Lakes tributaries to assess biotransport of pollutants associated with introduced Pacific salmon. Resident fishes sampled from salmon spawning reaches had higher mean pollutant concentrations than those from upstream reaches lacking salmon (93.5 and 4.1 $\mu g \cdot kg^{-1}$ [PCB], 24.0 and 3.1 $\mu g \cdot kg^{-1}$ [DDE], 8.5 and 1.0 $\mu g \cdot kg^{-1}$ [PBDE], respectively), but differences varied substantially among lake basins. In Lake Michigan tributaries, PCB concentrations in resident fishes from salmon reaches were over four times higher than those from salmon reaches in Lake Huron and over 30 times higher than those from Lake Superior. Moreover, resident fish pollutant concentrations were better explained by pollutant inputs from salmon ($\mu g \cdot m^{-2}$; R^2 = 0.76 [PCB], 0.64 [DDE], 0.64 [PBDE]) than by land development/agriculture, watershed area, resident fish species, body length, or lipid content. These results suggest that pollutant dispersal to stream ecosystems via biotransport is an often overlooked consequence of salmon stocking and historical food web contamination in the Great Lakes. Our findings have implications for Great Lakes management, including dam removal and wildlife conservation.

INTRODUCTION

Persistent organic pollutants (POPs) are distributed globally through a number of ecological processes, including biological transport.[1,2] During migrations from one ecosystem to another, organisms can distribute large quantities of pollutants to new environments.[3,4] Unlike atmospheric transport, which facilitates both the introduction and removal of POPs, biological transport often lacks a viable loss route, resulting in accumulation in recipient ecosystems.[1,5] Migratory Pacific salmon (*Oncorhynchus* spp.) can be particularly effective agents of biological transport because they bioaccumulate POPs while feeding in the ocean and then transfer these pollutants to freshwater ecosystems (i.e., streams and lakes) where they spawn and die.[3,6]

The role of Pacific salmon in transporting nutrients through watersheds has been frequently highlighted,[7] and salmon can similarly transport POPs to organisms residing in spawning streams.[6] This ability to disperse large quantities of material during spawning runs makes Pacific salmon introductions a potentially important mechanism of pollutant dispersal in non-native ranges. A prime example is the Laurentian Great Lakes, a historically polluted region where millions of non-native Pacific salmon are stocked annually.[8] Since their introduction over 50 years ago, salmon from the Great Lakes have exhibited among the highest POP concentrations ever recorded for Pacific salmon.[9,10] Although environmental concentrations of some POPs (e.g., PCBs [polychlorinated biphenyls] and DDE [1,1-dichloro-2,2-bis[p-chlorophenyl] ethylene]) have declined due to bans in the United States and later internationally under the Stockholm Convention,[9,11] concentrations in fish remain high enough to warrant sport fish consumption advisories[12] and to elicit concern over effects on piscivorous wildlife.[13] Although instances of biological transport have been reported,[14,15] the relationship between salmon inputs and POP levels in stream biota has not been established, and the implications of biological transport for resource management in the Great Lakes remain unclear.

Discharge of pollutants into Great Lakes tributaries is a clear driver of lake pollutant levels,[16] but rarely have reverse (lake to stream) flows been considered in pollutant management or abatement. Research suggests that migratory fishes may funnel

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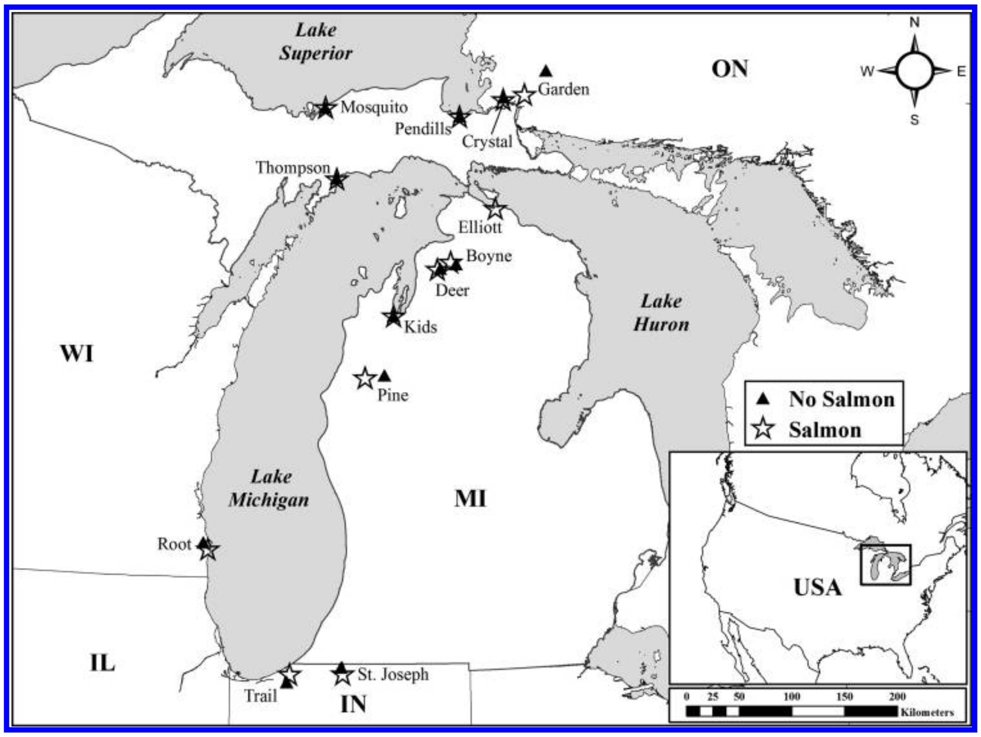

Figure 1. Map of study sites in the upper Great Lakes region. Resident fishes were sampled from stream reaches with (empty stars) and without (black triangles) salmon spawners.

POPs from lake to stream environments in the Great Lakes basin, as resident fishes in stream reaches accessible from the lakes can contain pollutant concentrations many times higher than those above barriers.[14,17] Pollutant transport by migratory fishes has been discussed qualitatively as a potential negative consequence of dam removal,[18] but no tools exist for predicting upstream biotransport of POPs after dam removal. An evaluation of biological transport relative to other factors that can affect POP levels is critical for assessing the influence of biotransport on POP levels in aquatic biota and for reasonably anticipating impacts.

We tested the prediction that POP concentrations in stream-resident fishes are positively related to the abundance of Pacific salmon spawners in 13 Great Lakes tributary streams. First, we quantified POP concentrations in spawners (the agents of biological transport) and compared concentrations among salmon species from tributaries to three Great Lakes (Michigan, Huron, and Superior). Second, we compared POP concentrations in stream-resident fishes between stream reaches with and without spawning salmon, as well as among lake basins and among resident fish species. Third, we compared POP concentrations in sediments collected from stream reaches with and without spawning salmon. Fourth, we evaluated potential factors affecting POP concentrations of resident fishes and sediments, including POP inputs from spawning salmon, development/agricultural densities, watershed area, resident fish species, body length, and lipid content.

■ MATERIALS AND METHODS

Study Design and Fish Sampling. We sampled fish from tributaries to Lake Michigan (n = 8), Lake Huron (n = 3), and Lake Superior (n = 2) (Figure 1; Supporting Information (SI) S1). Each stream receives annual spawning runs of chinook (*Oncorhynchus tshawytscha*), coho (*O. kisutch*), and/or pink (*O. gorbuscha*) salmon and has a resident (assumed nonmigratory) fish community. We limited our scope to fall-run Pacific salmon rather than other anadomous species (e.g., native suckers [*Catostomus* spp.], rainbow trout [*O. mykiss*], and brown trout [*Salmo trutta*]) because salmon are semelparous (i.e., die after a single spawning bout) and deposit all POPs accumulated in their bodies to streams after they spawn. Salmon were obtained by electrofishing from nine streams in 2008 and 2009 and three streams in 2010. Salmon sampling occurred during the spawning run, typically late September through October. To approximate the relative abundance of salmon spawners and estimate POP inputs from salmon, salmon biomass (kg·m^{-2}) was estimated in each stream from visual counts of live salmon and carcasses (averaged over 2 years) within the wetted channel at the peak of the salmon run.[19]

Stream-resident fishes were sampled by electrofishing of reaches (100−500 m in length) with and without salmon (i.e., below and above, respectively, an impassable dam or waterfall). Upstream of standing water created by artificial dams, nonsalmon study reaches were selected that had conditions as similar as possible to downstream reaches. The inclusion of reaches lacking salmon was intended to control for watershed influences on resident fish POPs unrelated to salmon, such as atmospheric deposition and point sources of pollution. Thus, differences between reaches were assumed associated with salmon. Resident fish species taken from salmon and nonsalmon reaches included brook trout (*Salvelinus fontinalis*), mottled sculpin (*Cottus bairdii*), blacknose dace (*Rhinichthys atratulus*), creek chub (*Semotilus atromaculatus*), and/or white sucker (*Catostomus commersoni*) (S1). Sample sizes depended on availability and ranged from n = 1 to n = 13 individuals per reach.

We also sampled stream sediments from reaches with and without salmon at six sites in 2008 to compare the benthic reservoir of POPs across streams (S1). We used a 20-cm tubular gravity corer to obtain sediments from shallow (<30 cm water depth) backwater habitats where silt and organic matter had accumulated. All cores from one reach ($n = 3$) were homogenized and analyzed as a composite to generate a single concentration for each reach, normalized for organic content.[20]

Pollutant Extraction and Analysis. Whole fish were prepared for analysis as previously described (see S2 for details).[21,22] When only small individuals were available, such as was often the case for mottled sculpin, blacknose dace, and small brook trout, composites of two or more individuals were used to obtain sufficient mass (>20 g) for analysis. Identification and quantification of 62 polychlorinated biphenyl (PCB) and 9 polybrominated biphenyl ether (PBDE) congeners, as well as 1,1-dichloro-2,2-bis(p-chlorophenyl) ethylene (DDE) was accomplished by gas chromatography/ mass spectrometry in negative chemical ionization mode.[21,22] Results are reported in $\mu g \cdot kg^{-1}$ wet weight (w.w.). Values were not adjusted for surrogate recovery. Quality control samples (blanks, matrix spikes, matrix spiked duplicates, and standard reference material) were analyzed to ensure precision and accuracy.[23]

Statistical Analysis. We used several statistical analyses to establish (1) differences in POP concentrations among salmon species and lakes, (2) differences in resident fish POP concentrations between salmon and nonsalmon reaches and among species and lakes, (3) differences in sediment POP concentrations between salmon and nonsalmon reaches, and (4) the relationship between POP concentrations in resident fishes and sediments with various explanatory factors (watershed area, land use, resident fish species, body length, lipid content, and POP inputs from salmon).

First, we used two-way analysis of variance (ANOVA) to compare salmon species and their eggs (collected from $n = 12$ streams), where species (chinook, coho, pink, as well as chinook eggs and coho eggs) and lake basin (Michigan, Huron, Superior) were factors, and salmon tissue POP concentration (PCB, PBDE, or DDE) was the response variable.[24] Pink salmon eggs were not available for collection. A significant species-by-lake interaction was interpreted to indicate that POP concentrations for a certain salmon species varied among lake basins.

Second, we used multiway ANOVA to test for differences ($n = 13$ streams) in resident fish POP concentrations, where species (e.g., brook trout, mottled sculpin, and blacknose dace), reach (salmon or no salmon), and lake basin (Michigan, Huron, and Superior) were factors, and resident fish POP concentration (PCB, PBDE, or DDE) was the response variable.[24] A significant reach-by-lake interaction was interpreted to indicate that differences between salmon and nonsalmon reaches varied by lake basin, suggesting that the magnitude of POP transport by salmon to resident fishes also varied among basins.

Third, we used paired t-tests to compare sediment POP concentrations (PCB, PBDE, or DDE) between salmon and nonsalmon reaches ($n = 6$ streams).[24] We used a paired t-test because we were interested in determining whether the mean difference between salmon and nonsalmon reaches was different from zero, and low sample sizes ($n = 1$ per stream reach) precluded the use of alternative tests. Pairs consisted of salmon and nonsalmon reaches within the same stream.

Data were tested for conformity to ANOVA assumptions using normal probability plots, residual plots, and Shapiro–Wilk normality tests ($\alpha < 0.05$). Data were $\log_{10}(x + 1)$ transformed when necessary to conform to ANOVA assumptions. An alpha level of 0.05 was used for all tests. Statistical analyses were performed using R v2.13.2.[25]

Fourth, we used an information-theoretic approach[26] to compare the strength of support for various hypotheses, or models, about the relationship of certain biological and landscape factors with resident fish POPs ($n = 10$ steams) and sediment POPs ($n = 6$ streams). Models were chosen to assess the influence of POP inputs from salmon relative to other variables known to influence environmental levels of POPs. The information-theoretic approach offers the advantage over traditional model evaluation (e.g., stepwise regression) of allowing for simultaneous comparison of several competing models.[27] We conducted separate analyses for stream-resident fishes and for sediments. Hypotheses were compared using linear regression models.[26] We first calculated Akaike's information criterion corrected for small sample bias (AIC_c) to evaluate the fit of each model, with the lowest AIC_c score indicating the best-fitting model. Next, we calculated the AIC difference (Δ_i), which is AIC_c for candidate model i minus the minimum AIC_c among all candidate models. Models with low Δ_i are most plausible given the data set. Finally, we calculated Akaike weight (w_i), or the strength of evidence for each model, to select the best-fit model.

We evaluated six models of the observed POP concentrations of resident fishes: (1) watershed area, (2) land use (% total watershed area with >50% cover by constructed materials for PCBs and PBDEs; % total watershed area covered by agriculture for DDE), (3) resident fish species (e.g., brook trout), (4) resident fish total body length (cm; mean length of individual fish for composite samples), (5) resident fish species and body length, (6) lipid content (% wet mass), (7) POP inputs from salmon ($\mu g \cdot m^{-2}$), and (8) all variables combined. Watershed area was chosen because larger catchments entrain more POPs settling from the atmosphere than small catchments, and atmospheric deposition is a significant component of the POP cycle in the Great Lakes region.[28] Land use was selected as an indicator of anthropogenic influence, which can impact POPs in biota.[29] Species, length, and lipid content were chosen because these variables have been found to be associated with the magnification and retention of POPs in fish (refs 30, 31, but see 32). Inputs from salmon were included to evaluate the importance of POP influx via spawning runs. Salmon inputs ($\mu g \cdot m^{-2}$) were estimated for each stream by multiplying salmon biomass ($kg \cdot m^{-2}$) by the average POP concentration ($\mu g \cdot kg^{-1}$ w.w.) of salmon collected from that stream. Land use data (% agriculture and % developed) were determined using GIS information available through the National Land Cover 2006 Database (S3).[33] Watershed area was determined from data made available by the Michigan Department of Technology, Management, and Budget Center for Geographical Information and the Wisconsin Department of Natural Resources.[34,35]

RESULTS AND DISCUSSION

Pollutant Concentrations in Salmon and Resident Fishes. Pollutant concentrations varied among salmon species and eggs (ANOVA, PCB $F_{4,86} = 79.5$, $P < 0.0001$; DDE $F_{4,86} = 68.7$, $P < 0.0001$; PBDE $F_{4,86} = 33.1$, $P < 0.0001$; S4). The highest concentrations were observed in chinook salmon (PCB [mean $\mu g \cdot kg^{-1}$ wet weight (SE)] = 390.0 (28.0), DDE = 93.0 (6.5), PBDE = 36.2 (2.0), $n = 38$), followed by coho salmon (PCB = 84.7 (16.5), DDE = 20.4 (4.2), PBDE = 12.3 (1.7), $n = 28$) and pink salmon (PCB = 22.2 (1.6), DDE = 5.3 (0.8),

Table 1. Mean Contaminant Levels in Salmon Spawners and Stream-Resident Fishes Sampled from Reaches Without (No Salmon) and With Salmon (Salmon) in Each Stream[a]

			salmon contaminant levels ($\mu g \cdot kg^{-1}$ wet weight)			mean resident fish contaminant levels ($\mu g \cdot kg^{-1}$ wet weight)					
						PCB		DDE		PBDE	
lake basin	stream	salmon biomass ($kg \cdot m^{-2}$)	PCB	DDE	PBDE	no salmon	salmon	no salmon	salmon	no salmon	salmon
Michigan	Thompson Creek, MI	0.785	328	90.8	33.0	6.0	97.9	2.3	18.5	1.1	7.4
	Pine Creek, MI	0.305	394	107	37.3	4.0	245	1.4	65.8	0.8	19.1
	Root River, WI[b]	0.199	444	81.9	34.6	8.8	126	14.2	35.7	1.8	9.8
	Kids Creek, MI	0.160	456	113	54.1	4.0	31.4	6.6	14.4	1.0	4.0
	Deer Creek, MI	0.008	199	41.3	23.6	2.3	8.7	1.8	2.5	1.6	2.3
	Trail Creek, IN[b]	-	324	51.3	22.0	6.8	198	18.2	46.3	0.4	21.1
	St. Joseph River, IN[b]	-	159	30.5	16.2	11.1	15.2	5.9	3.5	2.4	2.2
	Boyne River, MI	-	-	-	-	2.7	160	1.2	19.1	1.0	13.9
	mean	**0.291**	**328**	**76.2**	**30.3**	**4.8**	**139**	**4.3**	**35.7**	**1.2**	**11.9**
Huron	Elliott Creek, MI	0.080	165	48.2	18.5	-	47.3	-	14.7	-	5.2
	Garden River, ON	0.042	180	47.9	20.8	2.5	31.0	0.9	6.0	0.7	7.4
	Crystal Creek, ON	0.003	51.5	14.7	10.6	3.3	18.4	1.0	3.1	1.0	3.8
	mean	**0.041**	**154**	**42.9**	**18.4**	**2.9**	**33.8**	**1.0**	**9.0**	**0.8**	**4.9**
Superior	Pendills Creek, MI	0.006	46.0	13.5	13.0	2.0	4.4	0.7	1.0	0.4	0.9
	Mosquito River, MI	0.002	36.9	9.2	8.4	3.9	3.8	1.1	0.6	1.1	1.1
	mean	**0.004**	**42.4**	**11.7**	**11.1**	**3.0**	**4.3**	**0.9**	**0.9**	**0.8**	**1.0**

[a]Values represent means of all individuals of all fish species collected and analyzed in 2008 and 2009 (salmon = chinook and/or coho; resident fishes = brook trout, mottled sculpin, blacknose dace, creek chub, and/or white sucker). Salmon biomass is the mean of 2008 and 2009. Hyphens (-) indicate instances where data were not collected. [b]Sampled only in 2010.

PBDE = 4.3 (0.4), n = 4). Pollutant concentrations in salmon eggs were 20−84% higher on average than in adults of the same species (chinook egg PCB = 480.7 (43.1), DDE = 133.2 (11.8), PBDE = 50.8 (4.4), n = 16; coho egg PCB = 101.9 (23.4), DDE = 30.0 (9.6), PBDE = 22.6 (4.5), n = 12; pink eggs were not available).

Lake Michigan salmon had the highest POP concentrations (except for PBDE), followed by Lake Huron, and then Lake Superior (Table 1; ANOVA, PCB $F_{2,86}$ = 19.7, P < 0.0001; DDE $F_{2,86}$ = 12.4, P < 0.0001; PBDE $F_{2,86}$ = 0.4, P = 0.44). Differences in POP concentrations among the salmon species may at least partly explain the variation among lake basins. Chinook salmon, the species with the highest average POP concentration, predominated in the Lake Michigan tributaries that we sampled, while coho salmon were dominant in Lake Superior, and both species were present in Lake Huron. Moreover, Lake Michigan chinook had POP concentrations 2−3 times higher than coho (Lake Michigan chinook PCB [mean $\mu g \cdot kg^{-1}$ (SE)] = 395.7 (30.4), PBDE = 35.7 (2.1), DDE = 92.1 (7.0), n = 33; Lake Michigan coho PCB = 142.3 (30.9), PBDE = 15.5 (3.4), DDE = 32.5 (8.5), n = 12). The observed variation in POP concentration among salmon species could be explained by life history differences; chinook attain a much larger size and typically live longer than coho, and size and life span are positively associated with tissue POP levels.[31] Lake basin also appears to influence salmon POPs since two significant interactions (ANOVA, PCB Species × Lake $F_{5,86}$ = 3.19, P = 0.01; DDE Species × Lake $F_{5,86}$ = 3.14, P = 0.01) indicate that species differences vary by lake basin. Thus, POP concentrations in salmon appear linked to both lake basin and species. These results suggest that the biotransport of POPs to streams via salmon may be influenced by species- and basin-specific factors.

Within our study streams, POP concentrations varied among resident fish species (ANOVA, PCB $F_{8,119}$ = 6.0, P < 0.0001; DDE $F_{8,119}$ = 10.6, P < 0.0001; PBDE $F_{8,119}$ = 4.5, P < 0.0001). Among the resident fish species with $n \geq 5$, brook trout (PCB [mean $\mu g \cdot kg^{-1}$ ww (SE)] = 65.0 (16.0), DDE = 19.9 (4.7), PBDE = 6.2 (1.3), n = 56) and blacknose dace (PCB = 74.3 (32.7), DDE = 17.4 (8.1), PBDE = 5.5 (1.9), n = 19), both water column feeders, had higher POP concentrations than benthic feeding mottled sculpin (PCB = 24.8 (6.3), DDE = 4.8 (1.1), PBDE = 2.9 (0.6), n = 56). Our finding that POP concentrations vary among species is consistent with previous studies, which have posited that among-species variability is driven by differences in growth rate and trophic position.[31,36] However, the pattern we observed for water column versus benthic feeding fishes contrasts with patterns observed in lakes and reservoirs, where benthic species generally possess higher POP burdens than pelagic species.[37] Furthermore, high POP concentrations observed for blacknose dace were contrary to expectation. Based on species morphologies, we expected that brook trout would accumulate more POPs than blacknose dace or mottled sculpin because of their larger gape (>7 mm for our brook trout samples) and presumed superior ability to consume salmon eggs. Our finding that blacknose dace had relatively high POPs (in spite of having gape width smaller [<5 mm] than the diameter of a salmon egg [5−7 mm]) may indicate that they feed on salmon flesh or are able to nibble off small pieces of egg, but more research is needed to clarify potential uptake mechanisms (e.g., via food web pathways).

The differences in POP concentrations observed among resident fish species were relatively small (up to 4×; Table 2)

Table 2. Pollutant Levels in Stream-Resident Fishes in Reaches Without and With Salmon[a]

contrast	location	year	species	mean resident fish contaminant levels ($\mu g \cdot kg^{-1}$ wet weight)						ref
				no salmon			salmon (or GL-influenced)			
				PCB	DDE	PBDE	PCB	DDE	PBDE	
Time	Lake Michigan	1977-78	Mottled sculpin	320	100	-	1170	230	-	14
			Brown trout	390	170	-	1110	470	-	
			Rainbow trout	1660	840	-	2270	670	-	
	Lakes Michigan and Huron	1989-90	Common carp	160	60	-	6000	630	-	38
			Walleye	220	40	-	2900	330	-	
			White sucker	30	10	-	400	110	-	
			Northern pike	30	20	-	1000	150	-	
			Yellow perch	70	120	-	800	20	-	
Time / Space	Lake Michigan	2008-10	Brook trout	4.3	4.4	0.9	189	55.5	16.4	present study
			Mottled sculpin	3.7	1.7	1.2	56.8	10.6	5.3	
			Blacknose dace	4.7	2.7	0.7	303	69.3	18.4	
			Creek chub	11.3	14.6	1.8	116	35.2	8.2	
			White sucker	6.7	9.4	1.4	1230	18.6	11.8	
			Notropis spp.	13.1	23.7	2.8	160	57.2	14.0	
Space	Lake Huron	2008-10	Brook trout	3.0	0.9	0.8	55.6	17.6	6.2	
			Mottled sculpin	1.9	0.5	0.9	20.6	2.9	4.6	
			Blacknose dace	3.2	1.5	0.7	28.1	7.7	4.3	
	Lake Superior	2008-10	Brook trout	2.9	1.2	1	4.9	1.2	1.4	
			Mottled sculpin	1.8	0.3	0.3	2.1	0.4	0.5	
			Blacknose dace	4.8	0.7	0.6	9.1	1.6	1.6	
	Copper River watershed, AK	1994	Grayling	12.0	2.1	-	51.2	27.4	-	39
	Western U.S. Nat. Parks	2003-05	Salmonidae spp.	1.1	4.3	1.1	-	-	-	40[b]
	European mountains	2000-01	Salmonidae spp.	6.3	18	0.3	-	-	-	41,42
	Canadian mountains	1997-03	Salmonidae spp.	7.7	4.1	-	-	-	-	43

[a]The "time" contrast highlights the temporal decline in pollutant levels of various fish species in Lake Michigan since the late 1970s; the "space" contrast illustrates that pollutant levels in resident fishes from stream reaches above barriers (i.e., not influenced by the Great Lakes) are comparable to levels in other geographic regions. The term "GL-influenced" (GL = Great Lakes) is used because ref 38 compares fishes from stream reaches accessible to the Great Lakes to those from inaccessible reaches (above dams), and the presence/absence of salmon is not discussed. Common carp = *Cyprinus carpio*, walleye = *Sander vitreus*, yellow perch = *Perca flavescens*, grayling = *Thymallus thymallus*. [b]Ref 40 [PCB] = ΣPCB 74, 101, 118, 138, 153, 183, and 187.

compared with the variation observed in these same species between salmon and nonsalmon reaches (up to 61×; Table 1). Consistent with previous research, our results show that POP levels in both salmon and stream-resident fishes have declined in Great Lakes tributaries since the 1980s and 1990s, presumably due to tighter restrictions on pollutant emissions (cf. ref 9, 11). However, resident fish POP levels in stream reaches accessible to salmon remain high relative to other locations across the globe, while fish POP levels in inaccessible reaches (i.e., upstream of barriers) are comparable to regions outside of the Great Lakes basin with few or no local POP sources (Table 2). The contrast between salmon and nonsalmon reaches suggests that anadromous salmon may play an important role in POP dispersal to Great Lakes tributaries.

Pollutant Transport to Great Lakes Tributaries and Interbasin Variation. Assuming that our upstream control reaches effectively controlled for watershed influences unrelated to salmon, the presence of spawning salmon appears linked to elevated POP concentrations of stream-resident fishes. Resident fishes from stream reaches receiving spawning salmon had considerably higher POP concentrations than fishes from reaches lacking salmon (Table 1; ANOVA, PCB $F_{1,119} = 222.1$, $P < 0.0001$; DDE $F_{1,119} = 102.9$, $P < 0.0001$; PBDE $F_{1,119} = 136.0$, $P < 0.0001$; S4). Differences between reaches with and without salmon also varied among lake basins (ANOVA, PCB Reach × Lake $F_{2,119} = 21.2$, $P < 0.0001$; DDE Reach × Lake $F_{2,119} = 10.2$, $P < 0.0001$; PBDE Reach × Lake $F_{2,119} = 12.2$, $P < 0.0001$). The largest difference between salmon and nonsalmon reaches was observed in the eight Lake Michigan tributaries, where resident fishes from salmon reaches were an average of 29.0 (PCB), 8.3 (DDE), and 9.3 (PBDE) times more contaminated than fishes from nonsalmon reaches. The next highest difference was in Lake Huron (11.7 [PCB], 9.0 [DDE], and 6.1 [PBDE] times higher in salmon reaches), while Lake Superior showed only small differences between salmon and nonsalmon reaches (1.4 [PCBs], equal [DDE], and 1.3 [PBDE] times higher in salmon reaches). This pattern among the three lakes appears to be associated with salmon POP concentrations and salmon biomass (Figure 2). Although natural reproduction likely contributes to spawner biomass to some degree, biomass in the streams we sampled appeared associated with stocking rates, which are 7.1, 3.0, and 1.2 million (chinook and coho combined) for Lakes Michigan, Huron, and Superior, respectively, since 1967.[8,44]

Pollutant concentrations in stream sediments did not differ overall between reaches with and without salmon (paired t test, PCB $t_5 = 1.00$, $P = 0.36$; DDE $t_5 = 1.04$, $P = 0.35$; PBDE $t_4 = 1.03$, $P = 0.36$; S5). However, sediment PCBs and DDE in the stream with the highest abundance of salmon (Thompson Creek, Lake Michigan tributary) were respectively 41 and 19 times higher in the salmon reach (PCB [adjusted for organic content] = 270.0 $\mu g \cdot kg^{-1}$; DDE = 34.5 $\mu g \cdot kg^{-1}$) than in the nonsalmon reach (PCB = 6.5 $\mu g \cdot kg^{-1}$; DDE = 1.8 $\mu g \cdot kg^{-1}$).

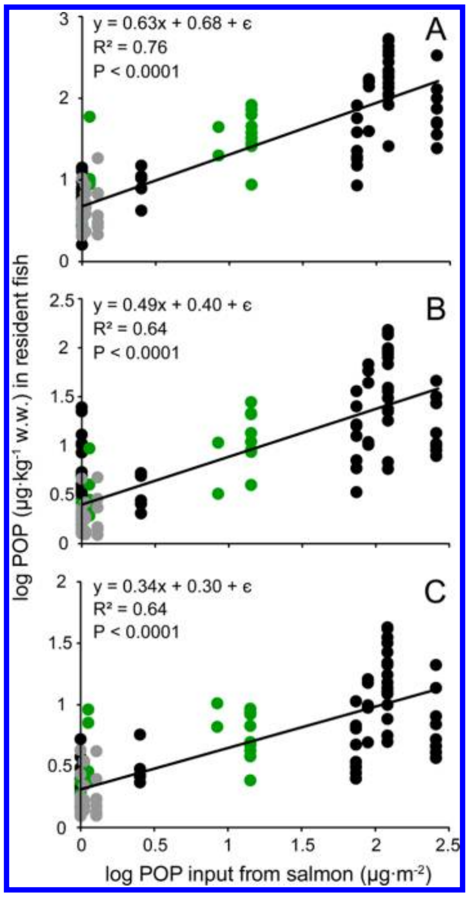

Figure 2. Resident fish PCB (A), DDE (B), and PBDE (C) concentrations as a function of pollutant inputs to streams via spawning salmon. Each point represents an individual sample (or composite sample; $\log_{10}(x + 1)$ transformed) from Lake Michigan (black), Huron (green), or Superior (gray). POP inputs from salmon ($\mu g \cdot m^{-2}$) were calculated by multiplying salmon biomass ($kg \cdot m^{-2}$) and contaminant concentration ($\mu g \cdot kg^{-1}$ wet weight). The coefficient of determination (R^2) is reported for each regression line. Regression equations indicate the slope (μ), y-intercept (β), and model error (ε) of each regression line. Standard errors of μ, β, and ε, respectively, are 0.031, 0.036, 0.33 (A); 0.032, 0.038, 0.34 (B); 0.022, 0.026, 0.24 (C).

While perhaps elevated in some instances, sediments in our salmon streams were still far less contaminated than systems directly impacted by anthropogenic discharge.[45] For instance, PCBs (corrected for % organic content) in surface sediments from the heavily industrialized Indiana Harbor and Ship Canal (Lake Michigan tributary) were several hundred times higher than the maximum concentration we observed in Thompson Creek. In our case, the absence of consistent differences in sediment POP concentrations between salmon and nonsalmon reaches contrasts with the results for resident fishes, which showed differences, and suggests that sediments in our study streams typically do not serve as substantial reservoirs for POPs delivered by salmon.

Akaike Information Criterion (AIC) results for sediment PCBs and DDE were most strongly supported by the salmon input model (Table 3). However, closer inspection of relationships suggests the sediment data analyses should be interpreted with caution, as they are largely driven by outliers (Thompson Creek salmon reach [PCB and DDE]; S5). Sediment PBDEs were best supported by the model containing watershed area, but, similar to the PCBs and DDE results, this relationship was largely driven by an outlier (Pine Creek nonsalmon reach [PBDE]; S5). We therefore consider the sediment AIC results to be inconclusive, and suggest that further research is needed to determine the influence of salmon on sediment POPs in Great Lakes tributaries.

Relationship between Spawning Salmon and Pollutants in Resident Fishes. Akaike Information Criterion results for resident fish POPs were most strongly supported by the salmon input model (Table 3). Regression analysis provided further support for an association between salmon inputs and resident fish POP concentrations ($R^2 = 0.76$ [PCB], $R^2 = 0.64$ [DDE], $R^2 = 0.64$ [PBDE]; Figure 2). Lake sediment PCBs have been found to correlate with temporal patterns of salmon spawner abundance in their native range,[3,6] and we provide evidence that POP concentrations in Great Lakes stream fishes also associate with spawner abundance. Our results suggest that, in the absence of point sources, inputs from spawning salmon can more strongly influence resident fish POPs than watershed area, land use, among-species differences, body length, and lipid-related differences. This finding does not mean that those nonsalmon factors are unimportant, but rather that under certain circumstances (such as in streams with high salmon abundance) their influence on fish POPs may be small compared to salmon inputs.

Salmon may elevate resident fish POPs via a number of mechanisms. For instance, the annual infusion of salmon material to the diets of resident fishes may lengthen the food chain and increase POP burdens by raising the trophic position of resident fishes.[cf.30] Additionally, stream-resident fish were observed consuming salmon eggs, suggesting that direct ingestion of salmon material and subsequent bioaccumulation may also be an important process, although experimentation is needed to confirm these mechanisms. Our findings also raise questions about the potential role of other anadromous fishes, such as native suckers and introduced rainbow and brown trout, in pollutant dispersal to streams in the Great Lakes basin. We suggest that POP dispersal from lakes to streams by anadromous fishes should be included in conceptual models of POP cycling in the Great Lakes (e.g., ref 28).

Implications for Natural Resource Management. The contribution of biological transport to POP burdens in piscivorous wildlife is largely unknown in the Great Lakes Basin. Our results indicated that stream fishes frequently exceeded published thresholds for adverse impacts on common consumers of salmon and stream-resident fishes. Bald eagles (*Haliaeetus leucocephalus*), for example, were observed feeding on salmon carcasses in our study area. In a risk assessment for bald eagle egg lethality, the hazard quotient of 1.0 is exceeded when the average PCB concentration of food items (dietary no observable adverse effect concentration, NOAEC) exceeds 143 $\mu g \cdot kg^{-1}$.[38] When this value is applied to our study, 94% (31/33) of chinook, 33% (4/12) of coho, and 59% (10/17) of brook trout samples (from salmon reaches) from Lake Michigan tributaries exceeded the bald eagle NOAEC for PCBs. Adverse effects associated with DDE appear to be less likely, with only 6% (2/33) of chinook salmon sampled from Lake Michigan exceeding the bald eagle NOAEC of 160 $\mu g \cdot kg^{-1}$.[38] Further research, including dietary analysis, is needed to assess the impacts of biological transport of POPs on wildlife health in Great Lakes tributaries that receive salmon migrations.

The unique circumstances created by spawning migrations of stocked salmon in the Great Lakes have implications for watershed management, including dam removal. The associa-

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Table 3. Relative Strength of Support for Hypotheses (Linear Models) about the Relationship between Certain Biological and Landscape Factors and POPs (PCBs, PBDE, and DDE) in Sediments and Stream-Resident Fishes[a]

	PCB					DDE					PBDE				
model	K	SSE	AIC$_c$	Δ_i	w_i	K	SSE	AIC$_c$	Δ_i	w_i	K	SSE	AIC$_c$	Δ_i	w_i
sediment POPs[b]															
salmon inputs	3	0.92	−21.8	0.0	>0.99	3	0.59	−27.1	0.0	>0.99	3	1.23	−14.7	3.0	0.18
watershed area	3	1.80	0.1	21.8	<0.01	3	1.16	−3.5	23.6	<0.01	3	0.08	−17.8	0.0	0.81
land use	3	1.64	−8.1	13.7	<0.01	3	1.27	−10.7	16.4	<0.01	3	1.39	−9.8	8.0	0.01
SI+WA+LU	5	1.17	24.6	46.4	<0.01	5	0.15	8.3	35.4	<0.01	5	0.00	11.9	29.7	<0.01
stream-resident fish POPs															
salmon inputs	3	13.84	−288.3	0.0	>0.99	3	15.27	−275.3	0.0	>0.99	3	7.14	−375.0	0.0	>0.99
watershed area	3	54.15	−82.2	206.1	<0.01	3	37.01	−126.3	149.0	<0.01	3	17.48	−213.3	161.7	<0.01
species	3	56.77	−103.3	184.9	<0.01	3	37.08	−159.2	116.2	<0.01	3	19.55	−243.0	132.0	<0.01
body length	3	57.98	−100.6	187.7	<0.01	3	40.30	−148.3	127.1	<0.01	3	19.50	−243.3	131.7	<0.01
species+body length	4	56.18	−102.6	185.7	<0.01	4	37.16	−156.7	118.6	<0.01	4	19.57	−240.7	134.3	<0.01
lipid content	3	56.81	−103.3	185.0	<0.01	3	40.07	−149.0	126.3	<0.01	3	19.45	−243.7	131.4	<0.01
land use	3	60.42	−95.2	193.1	<0.01	3	42.41	−141.5	133.8	<0.01	3	20.54	−236.6	138.5	<0.01
SI+WA+S+BL+L+LU	8	9.38	−275.8	13.9	<0.01	8	10.27	−263.9	11.4	<0.01	8	4.81	−353.1	23.3	<0.01

[a]The number of parameters (K), error sum of squares (SSE), Akaike's information criterion corrected for small-sample bias (AIC$_c$), AIC$_c$ difference (Δ_i), and Akaike weights (w_i) are reported for each model. SI = salmon inputs, WA = watershed area, S = resident fish species, BL = resident fish total body length, L = lipid content (%), LU = land use (% developed [PCB, PBDE], % agriculture [DDE]). [b]Note that sediment results were strongly influenced by outliers; see text for details.

tion we found between resident fish POPs and inputs from spawning salmon may provide managers with a basis for anticipating upstream POP dispersal by anadromous fishes following dam removal. For example, in Lake Michigan tributaries with spawner biomass comparable to or exceeding our sites with the highest salmon abundance (i.e., ∼0.2 kg·m^{-2}), dam removal may result in significant POP dispersal to upstream biota. Although further study is needed to quantify these impacts, our research provides managers with relevant information for weighing the anticipated impacts of biotransport relative to the potential benefits (e.g., restoring natural flow regimes and improving fish passage[18,47]) and risks (e.g., species invasions and mobilization of toxic substances from reservoirs[48,49]) of dam removal.

Our findings also have relevance for determining which water bodies should be assessed for establishing fish consumption advisories. For example, a threshold of 50 μg·kg^{-1} (for fillets) is used by the Michigan Department of Community Health to establish a consumption advisory of no more than one meal per week for women of child-bearing age and children under the age of 15.[50] If we correct this threshold for our samples of whole fish (assuming the PCB ratio for whole fish to fillets for brook trout is similar to rainbow trout, which is ∼1.5:1[51]), the adjusted value of 75 μg·kg^{-1} is exceeded in 65% (11/17) of the brook trout we collected from Lake Michigan tributaries. Such findings are notable because fish assessments for consumption advisories do not routinely include stream-resident trout (Mark Tonello, Michigan Department of Natural Resources, Cadillac, MI, personal communication). Monitoring of PCB levels in trout populations may be necessary to alert consumers of potential health risks associated with consuming trout from streams receiving salmon runs.

Overall, our study suggests that biological transport of pollutants, enabled by stocking of migratory salmon and historical contamination, can be a key contributor to POP levels in resident biota in Great Lakes tributaries. This biological linkage is an important consideration for understanding patterns of pollutant distribution and dispersion, as well as for maintaining the health of rivers and streams in the Great Lakes basin.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Geographical and biological information for each sampling location (S1). Detailed description of analytical methods (S2). Land use variables for watersheds included in statistical analysis (S3). POP data for each individual fish sample (or composite sample) analyzed (S4) and sediment samples (S5). This information is available free of charge via the Internet at http://pubs.acs.org/.

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Notes

The authors declare no competing financial interest.

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