Environmental Science & Technology

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American Dippers Indicate Contaminant Biotransport by Pacific Salmon

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ABSTRACT: Migrating salmon can increase productivity in Pacific Northwestern streams and lakes through the deposition of nutrients from their decomposing carcasses after spawning. Several studies also report simultaneous biotransport of persistent organic pollutants that have contaminated lake food webs, although no similar effect has been shown conclusively in rivers. We tested the prediction that salmon enhance contaminants in river food webs using the American dipper (*Cinclus mexicanus*), an aquatic songbird and a recognized indicator of stream quality. Over 3 years, we analyzed 29 dipper eggs and aquatic invertebrate samples from 14 different rivers in 10 catchments in southern British Columbia, Canada to assess whether variations in autumn spawning density of Pacific salmon were reflected in dipper egg contamination or stable carbon and nitrogen isotopes. δ^{13}C isotope signatures, but not δ^{15}N, in aquatic invertebrates and dipper eggs increased among catchments in proportion to the average density of spawning salmon. Concentrations of brominated flame retardants (PBDEs), dichlorodiphenyltrichloroethane metabolites (DDTs), and chlordane compounds were related in part to the δ^{13}C measure of salmon density, but mercury, chlorobenzenes, and polychlorinated biphenyls (PCBs) were explained better by dipper trophic level. We conclude that spawning Pacific salmon result in the increased availability of salmon fry as dipper prey and salmon are a significant source of PBDEs, DDTs, and chlordanes to river ecosystems. However, contrary to lake studies, postspawn concentrations of legacy PCBs in river birds, even in salmon-rich rivers, were not significantly higher than would be expected from atmospheric deposition alone. We recommend using δ^{13}C isotopes to trace salmon-derived lipids which may persist over winter particularly in rivers, and are potentially a better reflection of lipophilic contaminant transfer.

INTRODUCTION

Annual runs of Pacific salmon provide an important source of energy and nutrients to aquatic and terrestrial organisms.[1,2] Salmon acquire up to 95% of their biomass in marine ecosystems but return to spawn and die in lakes and streams. Large amounts of marine-derived nutrients are then deposited to freshwater ecosystems via salmon roe and decaying carcasses.[3,4] A relatively large body of literature now supports the assertion that marine-derived nutrients from decaying carcasses, eggs, and ultimately emergent salmon fry affect primary, secondary, and tertiary levels in freshwater and associated terrestrial ecosystems.[5] This nutrient-rich resource provides a positive feedback to freshwater and riparian ecosystems by increasing productivity, growth, survival, and fecundity in organisms as diverse as riparian plants,[6,7] aquatic invertebrates,[8−10] and subsequent generations of juvenile salmonids.[11,12] Recent evidence suggests riparian birds are also directly and indirectly influenced by salmon spawning. Increased bird density,[13,14] species diversity,[15] reproductive success,[16] and survival,[17] have all been related to the salmon subsidy.

Salmon can also act as important vectors for contaminant transfer to remote lakes.[18−20] If such effects were large and widespread, they might offset some of the benefits of enhanced productivity, for example through toxicity from accumulated pollutants. To date, the majority of work quantifying transfer of contaminant loads from salmon to freshwater ecosystems is confined to lakes occupied by sockeye salmon (*Onchorhynchus nerka*).[18,19,21] The role of multiple salmon species in contributing contaminants to other freshwater ecosystems, such as rivers and streams, is poorly understood. Large differences exist between river and lake systems in physical structure, residence

Received: August 11, 2011
Revised: November 10, 2011
Accepted: December 6, 2011
Published: December 6, 2011

   dx.doi.org/10.1021/es2028058 | Environ. Sci. Technol. 2012, 46, 1153−1162

time, energetics, major primary producers, and factors affecting primary production.[22−24] Variations in physical features of individual streams including discharge, water temperature, sediment size, and salmon density can all influence the ecological effect of salmon which may also be important for contaminant flux or fate.[25,26] Therefore, it is possible that the transport and retention of salmon-derived contaminants in rivers may differ substantially from lakes.

Stable isotope tracers have revealed quantitatively that the proportion of marine-derived carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) in various stream biota is related directly to the local density of spawning salmon since the heavier isotopes (^{15}N and ^{13}C) are highly enriched in the marine environment relative to freshwater or terrestrial biomes. As a consequence, salmon tissues produced largely in the marine environment are also enriched.[4,27] Stable nitrogen isotopes ($\delta^{15}N$) can also provide an indication of consumer trophic position.[28] Additionally, when coupled with measurements of contaminants, isotopic measurements can provide insight into linkages between nutrient and contaminant sources.[29] Earlier research revealed that eggs of obligate aquatic passerines, the Eurasian (*Cinclus cinclus*) and American dippers (*Cinclus mexicanus*), are effective monitors of stream pollution given that they are recognized indicators of stream productivity, top predators in stream ecosystems, acquire nutrients for egg-laying within a confined territory, and a direct relationship exists between diet using stable isotope ratios and contaminants in eggs.[30−32] However, in contrast to lake ecosystems with varying densities of salmon, preliminary indications were that salmon might not be a larger contributor of some contaminants than atmospheric deposition.[33,34]

Here, we test the prediction that salmon import both nutrients and contaminants into rivers of the Pacific Northwest. We hypothesized that if salmon are the main contributor of non-point source pollution to rivers, then American dippers, an established indicator species, would have an enriched $\delta^{13}C$ and $\delta^{15}N$ signature from salmon and proportionately higher contaminant concentrations (e.g., polychlorinated biphenyls (PCBs), organochlorine pesticides (OCs), brominated flame retardants (PBDEs), and mercury (Hg)) in their eggs.

EXPERIMENTAL SECTION

Study Area. We initially selected 2 catchments in 2006 with salmon (Chilliwack, Silverhope) and 2 without salmon (Skagit and Similkameen) in southern British Columbia, Canada. Sampling was expanded in 2008 and 2009 to cover a broad geographic area in southern British Columbia with suitable dipper habitats on fast-flowing rocky rivers. We surveyed 28 different river catchments (n = 16 with salmon, n = 12 without salmon) across southern British Columbia to identify dipper breeding territories (Figure 1). Data on salmon presence, salmon species, and annual escapement (numbers of spawning salmon) were derived from the Department of Fisheries and Oceans (DFO) online database Mapster v.2.2 (http://www.canbcdw.pac.dfo-mpo.gc.ca/ows/imf.jsp?site=mapster) which uses geographic information systems (GIS) to report fish distributions, habitat, and escapement overlaid on basic land cover maps for British Columbia (Figure 1). Average annual escapement was calculated as the cumulative total of all species spawning on a given river averaged over a 20 year period (1989−2009) divided by available spawning habitat (total river length). River catchments with salmon contained up to 6 anadromous species (*Onchorhynchus tshawytscha*, *O. keta*, *O. kisutch*, *O. gorbuscha*, *O. nerka*, and *O. mykiss*) with populations known to spawn naturally from August to January or are enhanced by hatcheries. Catchments without salmon have been devoid of anadromous fish for decades because of natural or man-made barriers or historic changes in land use downstream that have prevented salmon migration. Additional information on stream length, stream order,[35] and magnitude was used to describe the relative size, position, and topography of the collection sites within the stream network which can have a profound influence on stable isotope values.[36,37] Stream data were obtained from the British Columbia Watershed Altas, a georeferenced database in a 1:50000 map interpreted using ArcGIS Desktop v.10 software (ESRI).

River Surveys and Sampling. Pairs of breeding dippers and nests were located between April and June, approximately 6−8 months after the peak period of Pacific salmon spawning. Most dipper nests were inaccessible because of their height (e.g., highway bridges) or locations (e.g., on cliff walls over fast flowing water), but where possible, a single random egg from a clutch of 4−5 eggs was taken from accessible nests during incubation (n = 29 from 14 rivers in 10 catchments) (Table 1). Whole eggs were weighed and measured (length and width), then refrigerated for up to 4 weeks. Egg contents were transferred into glass jars chemically cleaned with acetone and hexane, then stored frozen at −40 °C prior to stable isotope and contaminant analysis. Wherever eggs were collected, we sampled benthic aquatic invertebrates using a 3-min kick sample spread over different areas and river habitats (riffles and margins) to reflect the breadth of available prey. Samples were live-sorted to order in the field and sealed in polyethylene bags filled with streamwater and kept on ice until processing at the end of each day. Orders Ephemeroptera, Plecoptera, Trichoptera, and Diptera were obtained in all rivers. All invertebrate samples were subsequently rinsed 3× with distilled water to remove debris and drained prior to freezing at −20 °C. Invertebrates were later identified and counted to family but were pooled to order for stable isotope analysis.

Stable Isotope Sample Preparation and Analysis. Invertebrate samples were freeze-dried for 24 h before grinding to fine powder. Egg contents were homogenized using a Retch Ball Mill to minimize sample loss during homogenization and a 1-mL subsample was freeze-dried for 24 h. Approximately 1 mg of each dried homogenate was weighed and sealed in 4 × 6 mm tin capsules (Costech Analytical Technologies, Inc., Valencia, CA). Stable isotopes were determined at the University of California, Davis Stable Isotope Facility using a Europa Hydra 20/20 continuous flow isotope mass spectrometer (CFIRMS). Sample isotope ratios of $^{13}C/^{12}C$ and $^{15}N/^{14}N$ were expressed using standard delta notation ($\delta^{13}C$ and $\delta^{15}N$) in parts per mil (‰). Replicate laboratory reference standards were run every 10 samples and analytical measurement errors (SD) were ±0.03 ‰ for $\delta^{13}C$ and ±0.2 ‰ for $\delta^{15}N$.

Contaminant Analysis. Egg contaminants were analyzed at the National Wildlife Research Centre (NWRC) in Ottawa, Canada (2006 samples) and the Great Lakes Institute of Environmental Research (GLIER) in Windsor, Canada (2008−2009 samples) and included 49 PCB congeners, 16 PBDE congeners, total Hg, and organochlorine (OC) compounds including ∑chlorobenzenes (tetra, penta, hexa-chlorobenzene), ∑hexachlorocyclohexanes (α-, β-, γ-HCH), ∑chlordane-related compounds (oxychlordane, trans-chlordane, cis-chlordane, trans-nonachlor, cis-nonachlor and heptachlor epoxide), and ∑DDTs (p,p'-DDT, p,p'-DDE, and p,p'-DDD), mirex and photomirex, oxychlorostyrene and dieldrin. The 2 laboratories used

dx.doi.org/10.1021/es2028058 | Environ. Sci. Technol. 2012, 46, 1153−1162

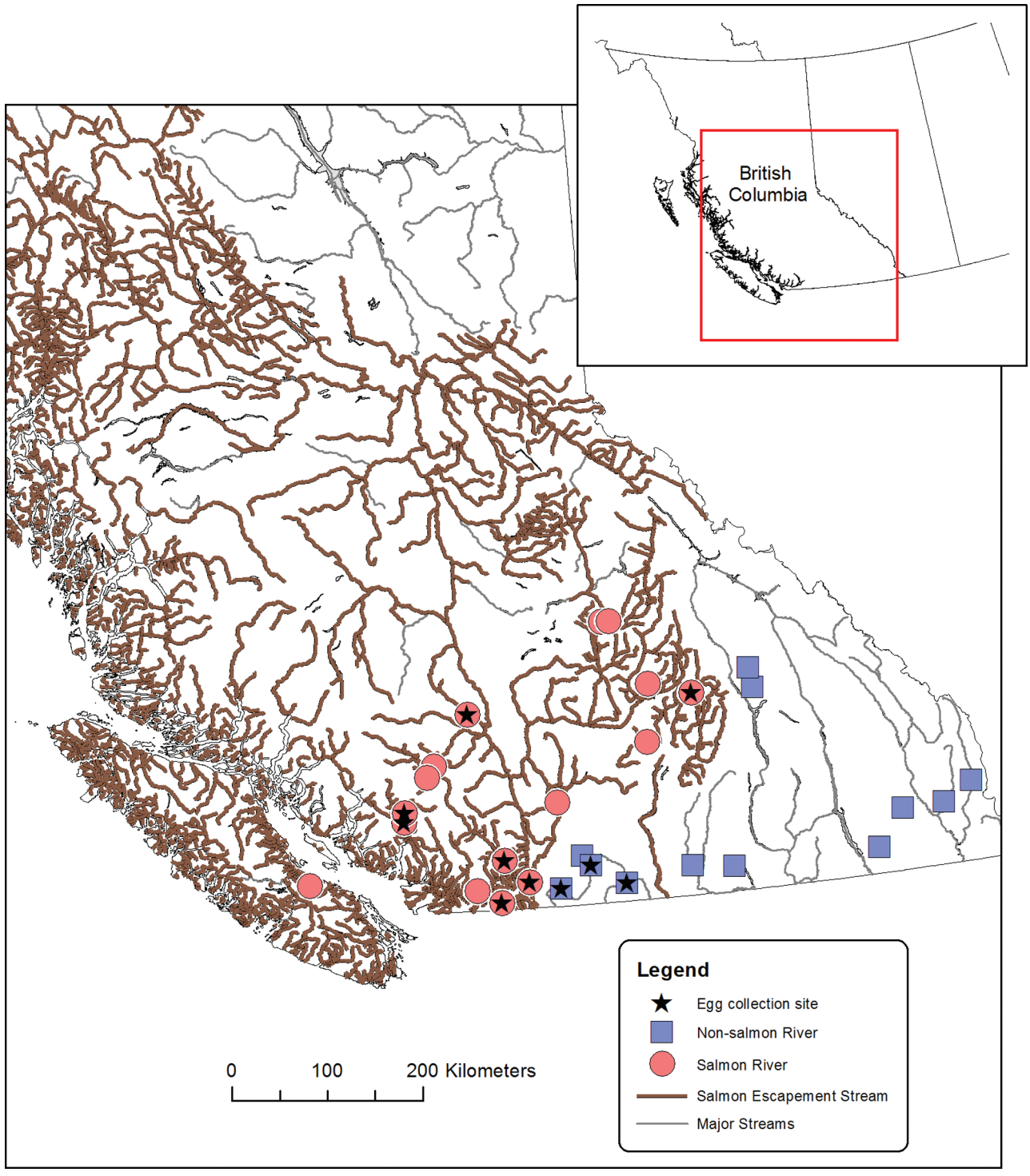

Figure 1. Map of study area in southern British Columbia, Canada showing locations of 16 salmon (circles) and 12 non-salmon (squares) rivers surveyed for dippers in addition to the 10 catchments where dipper eggs were collected (stars). Darker lines indicate streams where Department of Fisheries and Oceans (DFO) have identified salmon runs exist (salmon escapement stream).

comparable analytical methods and identical protocols including shared Double Crested Cormorant egg-pool reference material, internal spiked samples, routine solvent blanks and duplicate runs of selected samples for quality control.[32] Purified sample extracts (lipid extracted and removal of biogenic materials) were analyzed for OCs, PCBs, and PBDEs using a capillary gas chromatograph (Agilent 6890N), coupled with a mass selective detector (Agilent 5973N) operated in selected ion monitoring mode. Each extract was injected 3 times for (1) quantification of OCs; (2) determination of PCB congeners; and (3) determination of selected PBDEs. Dry masses were analyzed for mercury using an AMA-254 Mercury Analyzer and recoveries of reference material were within acceptable limits ranging from 83.8 to 110.1%. Egg contaminant concentrations are presented in ng/g on a wet weight basis except mercury data are reported in ng/g dry weight. Where appropriate for data analysis and comparison, data were lipid normalized using residuals of the contaminant concentration against percent lipid[38] and log

dx.doi.org/10.1021/es2028058 | *Environ. Sci. Technol.* 2012, 46, 1153–1162

transformed. The nominal detection limit for all compounds was 0.1 ng/g wet weight.

Data Analysis. Average salmon escapement densities ranged from 0 to ~8000 salmon/river km, and were $\log_{10}$ transformed (salmon spawner density + 1) prior to any further analysis to normalize the data and homogenize variances. We applied lipid corrections to dipper egg and invertebrate $\delta^{13}C$ data using C:N isotope ratios based on published relationships for aquatic organisms:[39]

$$^{13}C_{\text{lipid normalized}} = \delta^{13}C_{\text{untreated}} - 3.32 + 0.99 \times \text{C:N}$$

Egg trophic level (TL) was calculated using a baseline correction equation after Jardine et al.:[29]

$$TL = (\delta^{15}N_{egg} - \delta^{15}N_{Ephemeroptera})/\Delta^{15}N_{egg} + 2$$

where $\delta^{15}N_{Ephemeroptera}$ = baseline mean values of ephemeropteran larvae (dominantly grazers from families Baetidae and Heptageniidae) obtained from the same location as the egg samples and $\Delta^{15}N_{egg}$ = 3.4 ‰ for diet-tissue discrimination estimate of eggs derived from previous studies.[40]

Linear regressions were applied to assess the relationship between isotopic ratios in invertebrates and dipper eggs among sites. We report reduced major axis regression (RMA) slopes as a direct measure of local invertebrate contributions to dipper diet.[37] Analysis of covariance (ANCOVA) assessed the effect of salmon density ($\log_{10}$ + 1) on stable isotope ratios in invertebrates or dipper eggs with stream order as a covariate to control for potential variation, particularly in $\delta^{13}C$, associated with longitudinal changes downstream in the watershed.[36,37]

Differences in contaminant concentrations between salmon and nonsalmon rivers were assessed by nonparametric Wilcoxon tests. For several contaminant classes (chlordanes, chlorobenzenes, hexachlorocyclohexanes, mirex, and dieldrin), the data set included a large proportion of nondetectable values (10−50%), known as left-censored data. For each of those contaminant data sets, we applied nonparametric maximum likelihood methods (survival analysis) after data "flipping" to compare contaminant levels between salmon and nonsalmon rivers.[41,42] Nonparametric methods were preferred for this analysis as the sample sizes were uneven, variances were unequal, and the data set was small (29 samples).[42] We then used stepwise regression models to assess the factors that were influencing contaminant concentrations in dipper eggs. The initial models incorporated factors: $\delta^{13}C$ invertebrates and $\delta^{15}N$ invertebrates as measure of salmon influence, egg TL, stream order, stream magnitude, and the sampling year. We controlled for any potential year or confounding lab effect on the results by including these as random effects in all models. Selection of terms for deletion or inclusion in the final model was based on forward selection and Akaike's information criterion (AIC). All data were analyzed using JMP v.7 (SAS Institute).

Given that the PBDEs appeared to be the primary set of contaminants influenced by salmon and that PCBs are traditionally associated with salmon in freshwaters, we ran a multivariate ordination (CANOCO v.4.56, Biometris, The Netherlands) on the major PBDE and PCB congener patterns to identify sites most affected. We only included 21 PCB and PBDE congeners that were detected in the majority (>65%) of the samples to remove the variation caused by the many nondetectable values. Because we were largely interested in patterns, contaminant data were standardized by subtracting the mean and dividing by the standard deviation to remove the effect of concentration and log transformed throughout.[43] Using an initial detrended correspondence analysis (DCA), first axis gradients were relatively short (<2.0 SD units) and we proceeded with principal components analysis (PCA).[43] PCA scores were then regressed against salmon density and egg TL to determine if the salmon were influencing the PCBs and PBDEs congener patterns in dipper eggs.

RESULTS

Influence of Salmon on Stable Isotope Signatures. Isotopic signatures in invertebrates were strongly related to dipper eggs with the effect greater for $\delta^{13}C$ (r^2 = 0.72, p < 0.0001) than $\delta^{15}N$ (r^2 = 0.4, p = 0.0002) (Figure 2). Slopes of

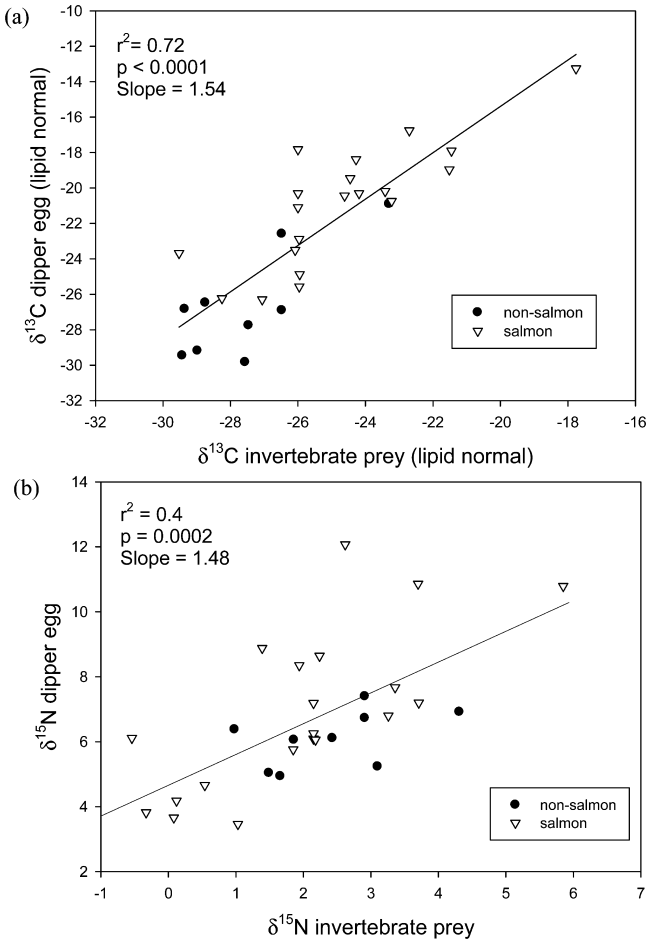

Figure 2. Relationships between mean (a) $\delta^{13}C$ and (b) $\delta^{15}N$ isotope signatures in aquatic invertebrate prey and American dipper eggs showing all samples collected from salmon (open triangle) and nonsalmon (solid circle) rivers. R^2 values and slopes of the lines are derived from reduced major axis regressions.

the regressions were 1.5 ($\delta^{13}C$) and 1.4 ($\delta^{15}N$) which exceeded 1, indicating dipper diets were likely derived exclusively from local invertebrate sources. $\delta^{13}C$ isotope signatures in invertebrates and dipper eggs were not directly affected by stream order, length, or magnitude—likely because most of our study streams chosen were similar mid-order rivers—but stream order was retained in subsequent analyses to reduce the variability in the models. Invertebrate $\delta^{13}C$ was more enriched in salmon

dx.doi.org/10.1021/es2028058 | Environ. Sci. Technol. 2012, 46, 1153−1162

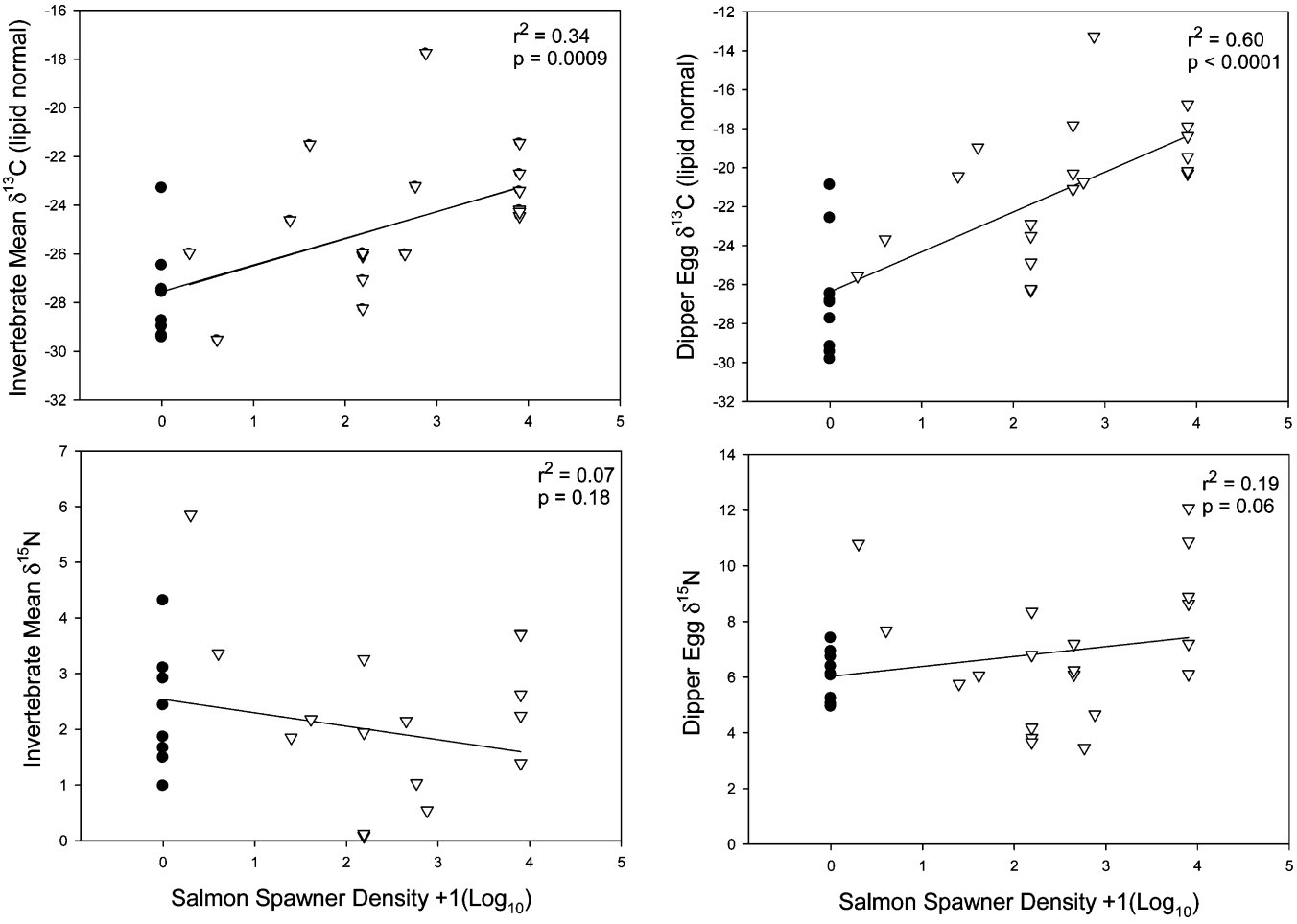

Figure 3. Relationship between $\delta^{13}C$ (top graphs) and $\delta^{15}N$ (bottom graphs) values in aquatic invertebrates, dipper eggs, and salmon spawner density in rivers of British Columbia. Open triangles represent sites with salmon, and solid circles are sites without salmon. Regression lines, r^2 and p values represent results of an ANCOVA with spawner density (main effect) and stream order (covariate).

rivers than those without salmon (t_{27} = 2.9, p = 0.008), while salmon spawner density was an effective predictor of $\delta^{13}C$ enrichment in both aquatic invertebrates (r^2 = 0.54, p < 0.0001) and dipper eggs (r^2 = 0.72, p < 0.0001) even after controlling for any potential confounding influence of longitudinal effects (Figure 3). In contrast, $\delta^{15}N$ in invertebrates (r^2 = 0.07, p = 0.18) and dipper eggs (r^2 = 0.07, p = 0.06) were apparently unrelated to salmon abundance (Figure 3). These relationships were not influenced by salmon altering lipid content (and subsequently $\delta^{13}C$) of eggs as we used lipid normalized carbon isotope values and found no correlation between salmon density and lipid content of dipper eggs measured directly (r = 0.06, p = 0.7) or using C:N ratios as a proxy (r = 0.19, p = 0.3).

Influence of Salmon on Dipper Egg Contaminants. Of the contaminants measured, only ∑chlordanes were in significantly greater concentration in dipper eggs on salmon rivers while ∑PBDEs and ∑DDTs showed a similar trend that was not formally significant (Table 1, Figure 4). Salmon-derived nutrients, expressed as invertebrate $\delta^{13}C_{\text{lipid normalized}}$, were related to egg contaminants for ∑PBDEs (r = 0.41, p = 0.03), ∑DDTs (r = 0.52, p = 0.004) and ∑chlordanes (r = 0.39, p = 0.03), while other OCs, ∑PCBs, and Hg showed no relationships with salmon in any analysis (Figure 5).

Stepwise regression confirmed that salmon did not influence the concentrations of ∑PCBs, Hg, and ∑chlorobenzenes in dipper eggs (Table 2) but were instead predicted better by trophic level or sampling year. This would be consistent with dippers feeding on higher trophic level fish or predatory macroinvertebrates and thereby increasing their contaminant exposure. In support, dipper egg trophic level estimated using baseline corrected $\delta^{15}N$ increased significantly with salmon density (Figure 6) suggesting that increased availability of salmon or other fish prey during egg laying could influence foraging behavior and ultimately contaminant concentrations in dippers. Stepwise regression also confirmed that ∑DDTs, ∑PBDEs, and ∑chlordanes were apparently influenced by salmon even after controlling for other environmental variables.

The major PBDE and PCB congeners in dipper eggs were BDE 99, 47, and 100 and PCB 153, 138, 180, and 118. In the principal components analysis, 89.9% of the variance in PCB and PBDE congener concentrations was explained by the first two axes. BDE congeners 47, 99, 100, 153, 154 were positively related to salmon abundance with samples from the high salmon density Cheakamus and Chilliwack Rivers most closely associated with those congeners. In contrast, the majority of PCB congeners and the lighter BDE 17 and 28 congeners were most prevalent in the Similkameen and Bridge Rivers which had no salmon or extremely low salmon density. Salmon density and egg TL were both significantly related to PCA axis 2 (whole model $F_{2,26}$ = 7.3, p = 0.003; salmon density: p = 0.0008, egg TL: p = 0.05). Further, axis 2 sample scores were generally higher for eggs collected on salmon rivers (t_{27} = 2.62, p = 0.01). This implies that salmon are the source of the majority of the

dx.doi.org/10.1021/es2028058 | Environ. Sci. Technol. 2012, 46, 1153–1162

Table 1. Summary Characteristics of the Ten River Catchments Sampled in Southern British Columbia Including the Stream Length, Order, and Magnitude (Ranges for Multiple Streams Sampled in the Same Catchment) and the Average Density of Spawning Salmon (Number of Salmon Spawners/km)[a]

catchment	Ashnola	Similkameen	Skagit	Shuswap	Bridge	Silverhope	Squamish	Harrison	Cheakamus	Chilliwack
salmon density (no./stream km)	0	0	0	1–40	3	154	447	581	759	7981
stream length (km)	55	197	57	21–48	59	41	89	8.6	68	64
stream order	6	5	4–5	3–4	6	5	3–5	3	4	4–5
stream magnitude	378–600	119–144	60–231	6–49	926	116–150	5–160	5	35	107–340
no. dipper eggs	3	3	3	3	1	5	3	1	1	6
% lipids	4.9 ± 1.5	6.0 ± 1.2	5.7 ± 0.4	5.3 ± 1.6	7.6	4.9 ± 1.0	6.3 ± 0.5	4.2	4.3	5.2 ± 1.9
Hg (μg/g dw)	0.1 ± 0.05	0.2 ± 0.02	0.1 ± 0.02	0.1 ± 0.1	0.1	0.1 ± 0.01	0.1 ± 0.02	0.1	0.1	0.2 ± 0.1
total PCB	4.5 ± 0.9	66.4 ± 31.5	13.4 ± 7.2	9.2 ± 5.1	52.5	12.6 ± 7.1	13.0 ± 3.2	12.0	46.1	34.3 ± 28.6
total PBDEs	2.6 ± 2.7	7.1 ± 2.4	19.4 ± 11.9	4.6 ± 3.5	3.8	13.8 ± 10.6	9.2 ± 1.4	8.3	354.6	27.2 ± 12.2
hexa-CB	2.7 ± 0.3	1.5 ± 0.2	2 ± 0.6	3.1 ± 1.1	5.0	1.7 ± 1.5	4.5 ± 1.0	3.5	4.1	1.1 ± 1.8
penta-CB	0.2 ± 0.03	0.2 ± 0.03	0.3 ± 0.15	0.3 ± 0.1	0.4	0.1 ± 0.1	0.4 ± 0.1	0.2	0.4	0.1 ± 0.1
t-nonachlor	0.3 ± 0.02	0.4 ± 0.03	0.4 ± 0.1	0.7 ± 0.5	1.2	0.5 ± 0.4	1.2 ± 0.2	0.7	1.4	0.5 ± 0.5
c-nonachlor	0.1 ± 0.01	0.05 ± 0.08	0.0	0.2 ± 0.2	0.6	0.2 ± 0.1	0.4 ± 0.1	0.1	0.5	0.2 ± 0.2
oxy-chlordane	0.2 ± 0.05	0.0	0.0	0.3 ± 0.1	1.0	0.0	0.9 ± 0.2	0.5	1.3	0.0
heptachlor epox	0.03 ± 0.03	0.0	0.0	0.1 ± 0.01	0.5	0.0	0.2 ± 0.01	0.3	0.3	0.0
p,p′-DDE	32.9 ± 36.7	7.2 ± 1.5	6.8 ± 1.6	28.2 ± 14.9	9.5	11.0 ± 8.2	12.2 ± 2.5	13.6	49.4	25.5 ± 25.2
p,p′-DDD	0.1 ± 0.02	0.0	0.1 ± 0.17	0.2 ± 0.2	0.2	0.1 ± 0.2	0.1 ± 0.01	0.1	0.1	0.1 ± 0.2
p,p′-DDT	0.3 ± 0.02	0.0	0.0	0.4 ± 0.2	0.2	0.0	0.3 ± 0.1	0.2	0.4	0.0
dieldrin	0.03 ± 0.01	0.0	0.0	0.0	0.3	0.0	0.1 ± 0.01	0.2	0.1	0.0
mirex	0.2 ± 0.05	0.0	0.0	0.3 ± 0.2	0.4	0.0	0.3 ± 0.1	0.3	0.6	0.0

[a]Lipid content, sample sizes, and arithmetic mean contaminant concentrations (ng/g ww) in American dipper eggs collected from the sites are reported.

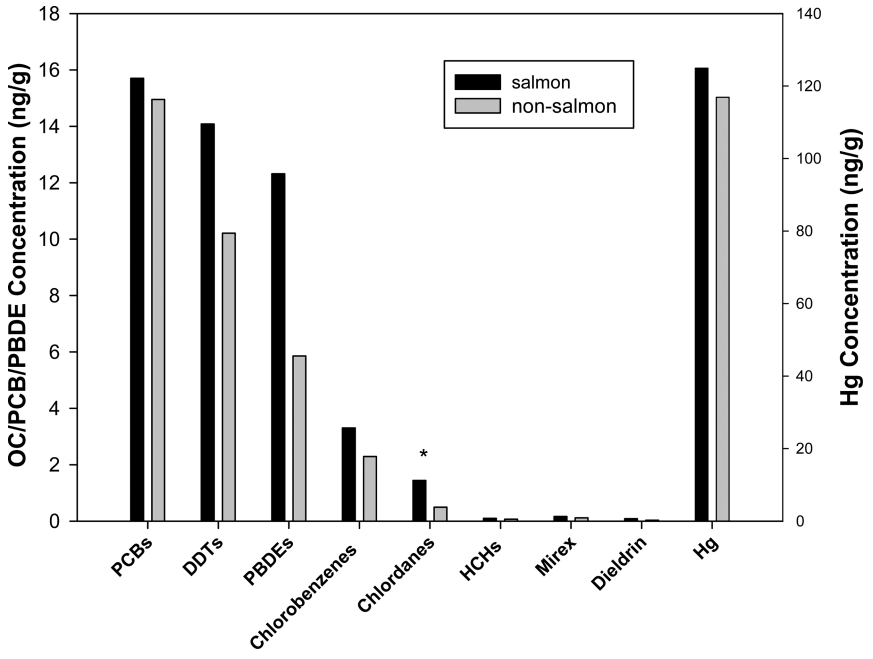

Figure 4. Geometric mean concentrations of total PCBs, total PBDEs, organochlorines (total chlorobenzenes, total chlordanes, total HCHs, mirex, and dieldrin), and mercury (Hg) contaminants in American dipper eggs collected from rivers with and without salmon in southern British Columbia, Canada. Significant differences (*) between salmon and non-salmon groups are based on non-parametric maximum likelihood methods to account for the large numbers of nondetectable values (see methods for details).

PBDE congeners (BDE 47, 99, 100, 153, 154) while atmospheric sources were the main contributor of PCBs and the lighter PBDE 17 and 28.

DISCUSSION

Stable Isotopes As Tracers of Salmon Influence. Based on our findings over a relatively large geographic area and wide range of salmon densities, we confirm that isotopic changes in $\delta^{13}C$ are indicative of salmon in stream biota and are reflected in dipper eggs in proportion to the density of salmon. Although the use of stable isotopes as a tracer of salmon influence is now popular, the mechanisms are poorly described. In quantifying salmon inputs using stable isotopes, users must account for two separate pathways by which salmon contribute nutrients to

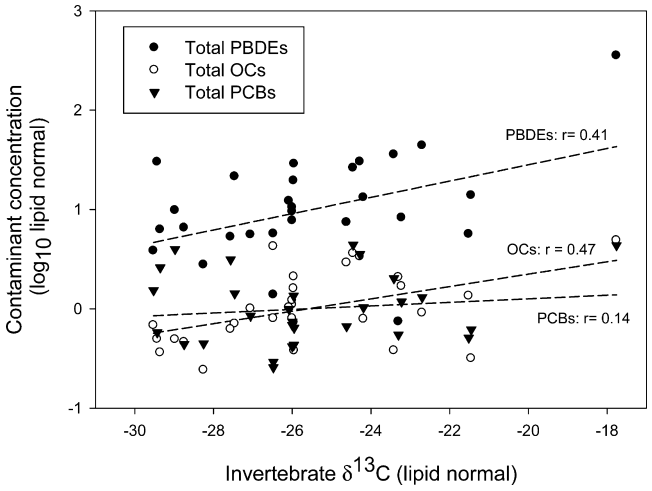

Figure 5. Correlations between mean $\delta^{13}C$ of invertebrate samples collected at dipper breeding sites (an indication of salmon density) and major classes of organic contaminants (total PBDEs, total organochlorines (OCs), and total PCBs). PBDEs (r = 0.41, p = 0.03) and total OCs (r = 0.47, p = 0.009) were significantly positively related to increasing salmon influence, while, for comparison, PCBs were not significantly related to the salmon subsidy (r = 0.14, p = 0.47).

Table 2. Results of Stepwise Multiple Regression Models Using Forward Selection to Identify Factors Influencing Contaminant Levels in Dipper Eggs[a]

contaminant	model variables	AIC	r^2	significance p (effect tests)
Hg[b]	**egg TL**	−109.6	0.22	**0.01**
∑chlorobenzenes[b]	**egg TL**	−36.47	0.40	**0.03**
	year			**0.04**
∑chlordanes[c]	**$\delta^{13}C$ invertebrate**	−33.06	0.47	**0.05**
	egg TL			0.27
	stream order			0.41
	year			0.09
∑DDT[c]	**$\delta^{13}C$ invertebrate**	−64.39	0.33	**0.008**
	year			0.1
∑PCBs[c]	$\delta^{13}C$ invertebrate	−62.32	0.24	0.21
	year			**0.01**
∑PBDEs[b]	**$\delta^{13}C$ invertebrate**	−48.27	0.53	**0.005**
	$\delta^{15}N$ invertebrate			**0.03**
	stream order			0.21

[a]All models had the following variables entered: $\delta^{13}C$ and $\delta^{15}N$ of aquatic invertebrates as measure of salmon influence, dipper egg trophic level (TL), stream order, and the year when the samples were collected. Table shows variables which passed the selection, model AIC scores, regression coefficients (r^2) and significance (p-values) of the variables tested. [b]$\log_{10}$. [c]$\log_{10}$ and lipid normalized using residuals of concentration regressed against % lipid; see methods.

freshwater and terrestrial ecosystems: (1) through direct consumption of tissues by consumers and (2) through indirect recycling of products through decomposition, leaching, and elimination.[44,45] Here we found no relationship between post spawn invertebrate or dipper egg $\delta^{15}N$ and salmon spawner density. Similarly, Holtgrieve et al.[46] also did not find any relationship between salmon and river consumer $\delta^{15}N$ in spring ($\delta^{13}C$ not reported). If consumers are feeding on salmon fry or eggs, tissue $\delta^{15}N$ will increase but this is better assessed using trophic level estimates.[29] In the case of dippers, we determined birds produced eggs at a higher trophic level by feeding on the

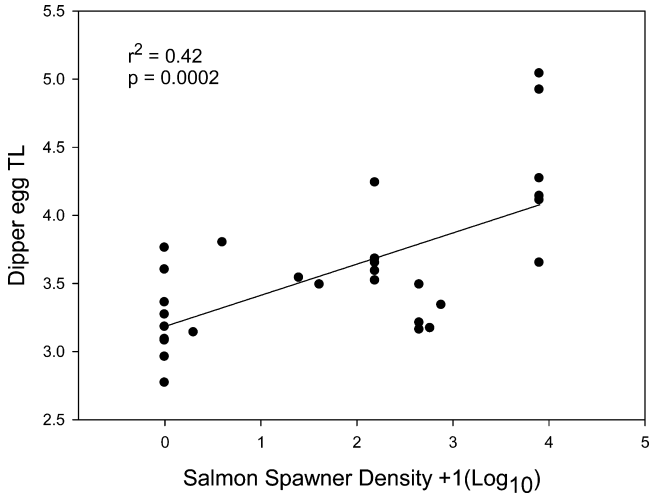

Figure 6. Relationship between the trophic level (TL) of American dipper eggs and increasing salmon spawner density among rivers of southern British Columbia.

salmon subsidy when it was available. This was true even though there were other freshwater fish species (e.g., trout) present on many rivers where salmon were absent, suggesting salmon are important prey particularly during breeding.[31,32]

Our results certainly do not negate the large numbers of studies which have detected a strong increase in $\delta^{15}N$ in aquatic invertebrates and other consumers in freshwater food webs—rather it illustrates a temporal inconsistency in persistence of the isotopic signal following salmon spawning. Since the retention time of recycled nutrients from salmon in rivers is poorly known, as well as their flux relative to other catchment sources, we cannot assume that $\delta^{15}N$ will indicate salmon influence in contaminant studies after the period when bulk salmon carcasses are no longer present. Dissolved stream nutrients, ammonium (NH_4^+), and soluble reactive phosphorus (SRP) concentrations in streamwater increase several fold in the presence of spawning salmon, but rapidly decline within weeks after carcasses degrade.[45,47] High discharge rates of rivers will increase the rate of loss compared to lakes and we evaluated isotopes in biota several months postspawn when salmon carcasses were no longer available to consumers. Biogeochemical and microbial processes in freshwater ecosystems through nitrification and denitrification will further influence the rate of loss and absolute concentrations of $\delta^{15}N$.[44] Long-term studies are needed to quantify the spiraling length and retention time of these nutrients among catchments with and without salmon.

Other studies have confirmed significant enrichment in $\delta^{13}C$ and its utility as a tracer of salmon in freshwater food webs.[8,21,27] Here we found that the ^{13}C isotope was a better indicator of salmon influence than the ^{15}N isotope in the spring postspawn. Since carbon isotopes are often closely associated with lipids,[48] this isotope may better reflect salmon-derived lipids and contaminants being transferred and retained in consumers such as the dipper.[44] Despite their limited use in contaminant studies, carbon isotopes have consistently been valued for their applications in estimating diet sources and establishing baseline trophic position.[28,49] Indeed, fatty acid profiles of salmon consumers change in response to the availability of marine-derived lipids which have been shown to persist over winter providing a long-term reserve.[50] In contrast, the salmon-derived nitrogen subsidy through recycling does not appear to persist in streams to the same extent as carbon.

dx.doi.org/10.1021/es2028058 | Environ. Sci. Technol. 2012, 46, 1153−1162

Therefore, researchers need to consider the timing of the incorporation of the isotopes relative to the peak salmon spawning. If this is not accounted for, one might underestimate the contribution of salmon to consumers using $\delta^{15}N$. Therefore, we recommend using $\delta^{13}C$ isotopes to trace salmon-derived lipids which may persist for a longer period, particularly in rivers, and are a better reflection of lipophilic contaminant transfer.

Biotransport of Contaminants from Salmon. Despite considerable evidence on the positive impacts of salmon on freshwater ecosystems, the contribution and impact of biotransport of contaminants from spawning salmon to various biota is less well-known. Salmon bioaccumulate contaminants from their natural diet and from the abiotic environment through water and sediment.[51,52] When large numbers of migrating salmon spawn and die in freshwaters, large influxes of contaminants such as PCBs[53] and mercury[54] have been detected. For example, large sockeye salmon runs in Alaska were found to increase sediment PCBs by 10× amounting to 6000 ng PCB/m^2/yr.[19] Despite this fact, only a handful of studies have examined the contribution of salmon-derived contaminants to freshwaters.[19−21,54,55] Of the published studies, most are limited to lakes or have evaluated contaminant deposition during or immediately following autumn spawning. Still fewer have documented comparative contaminant profiles in predators between salmon and non-salmon habitats which can be significant.[21,31,56] The debate about whether contaminant biotransport from salmon negates their benefit to consumers and freshwater ecosystems remains an important issue considering several studies have reported salmon accumulate levels of organic contaminants that are of concern to human and ecosystem health.[51,52,57−59]

Given the evidence from lakes and other predator studies we expected a much stronger influence of salmon on the contaminant levels of dippers. Concentrations of legacy organochlorines, PCBs, and Hg detected in this study were generally low by comparison to those reported for other aquatic birds in the region.[60,61] Although we observed relationships between $\delta^{13}C$ and PBDEs, DDTs, and chlordanes that could be attributed in part to spawning salmon, we found no effect for other contaminants particularly PCBs which have been strongly associated with salmon.[18,19,21] However, the presence of salmon fry and other fish can increase the trophic level from which eggs are produced, thereby increasing Hg, PCB, and chlorobenzene concentrations.

Whereas other studies have reported spawning salmon play a role in significantly enhancing the concentrations of legacy persistent organic pollutants such as PCBs, Hg, and OC pesticides to receiving ecosystems and food webs, we did not observe those large increases. Major factors in the incongruity of PCB and Hg results may be (1) sampling bias, (2) the biogeochemical and hydrological differences between lakes and rivers, and (3) the timing of sampling. Sampling bias cannot explain the differences observed, as we sampled a relatively large assortment of rivers with the range of salmon densities comparable to those in lakes where effects have been detected.[21] Unlike lakes where internal nutrient recycling typically contributes to elevated nutrients and contaminants, river ecosystems sampled 6−8 months post spawn when carcasses are not present do not retain the nutrient and contaminant subsidy likely because of higher flows and lower sedimentation. In other wildlife studies reporting higher contaminant levels associated with salmon, the primary pathway for increased accumulation is the direct consumption of salmon carcasses[56] or high availability of salmon fry and eggs.[31] Dippers have been shown to be largely "income breeders"[62] producing eggs from the resources available at the time of laying without using body reserves.[32] This implies the salmon contribution is a transient component to legacy pollutants in river catchments in comparison to atmospheric inputs or trophic level effects.

In contrast to many of the legacy pollutants, PBDEs were the most closely associated with salmon. Of particular interest was the high density Cheakamus River, where PBDE concentrations were 10−100× higher than other catchments (355 ng/g ww or 8247 ng/g lipid wt). Congeners 99, 47, and 100 dominated dipper egg samples similar to that reported for other aquatic birds[63] and for salmon worldwide.[52,58] Recent studies have shown the same PBDE congeners are present in relatively high concentrations in out-migrating wild juvenile Chinook salmon in the Pacific Northwest which ranged from 160 to 2800 ng/g lipid weight.[59] PBDEs are now considered a ubiquitous contaminant originating from industrial waste, sewage, and PBDE-containing products such as polyurethane foam[52,64] and during the past few decades have been steadily increasing in North American birds,[63,65,66] fish,[67] and marine mammals.[68] The use of PBDEs has largely been phased out of new products from markets in North America and other countries. However, many jurisdictions have legislated for addition of flame retardant chemicals to many commercial products. As a result, attention has shifted recently to the potential release of the replacement chemicals, many of which are brominated compounds.[69] Continued surveillance is thus warranted to assess persistence and bioaccumulation in food chains such as that of the dippers discussed here.

Despite the fact PBDEs, DDTs, and chlordanes were associated with salmon, at present there is no clear evidence that populations of dippers are being negatively affected. Based on an assessment of dipper presence or absence on the 28 survey streams in this study, breeding pairs of dippers were uniformly present in the appropriate habitats (fast-flowing rivers with boulders and cliffs or high ledges for nesting) regardless of salmon. In a detailed 7-year study of a dipper population in the Chilliwack River catchment of B.C.—a site with large salmon runs—birds which resided on the river year round consumed more salmon and had higher annual and lifetime reproductive rates but lower annual survival than those birds which migrated to high elevation tributaries where salmon were scarce.[16,70] American dippers in Alaska also produced larger brood sizes and heavier nestlings on stream reaches with salmon than those without.[71] Levels of legacy persistent organic pollutants in dippers generally remain low relative to other bird species measured worldwide. However, salmon appear to be an important contributor of PBDEs to freshwater ecosystems and the consumers within them. We recommend further research to evaluate whether salmon limit or enhance avian reproduction and survival in light of the potential sublethal toxic effects from contaminants such as PBDEs.

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ACKNOWLEDGMENTS

Annette Potvin, René McKibbin, and Courtney Albert helped with field work and collecting samples and Sandi Lee prepared the egg samples for analysis. Analytical support was provided by David Harris' lab at the Stable Isotope Facility at the University of California-Davis, the Great Lakes Institute of Environmental Research, and Environment Canada's National Wildlife Research

Centre. Funding was provided by Environment Canada (J.E.E.), Natural Science and Engineering Research Council of Canada (C.A.M.), and the Royal Society (S.J.O. and C.A.M.). We thank 3 anonymous referees for their insightful and helpful comments which greatly improved the manuscript.

REFERENCES

(1) Kline, T. C.; Goering, J. J.; Mathisen, O. A.; Poe, P. H.; Parker, P. L.; Scalan, R. S. Recycling of elements transported upstream by runs of Pacific salmon 0.2. delta-N-15 and delta-C-13: Evidence in the Kvichak River watershed, Bristol Bay, southwestern Alaska. *Can. J Fish Aquat. Sci.* **1993**, *50*, 2350−2365.

(2) Schindler, D. E.; Scheuerell, M. D.; Moore, J. W.; Gende, S. M.; Francis, T. B.; Palen, W. J. Pacific salmon and the ecology of coastal ecosystems. *Front. Ecol. Environ.* **2003**, *1*, 31−37.

(3) Groot, C.; Margolis, L. *Pacific Salmon Life Histories*; UBC Press: Vancouver, B.C., 1991; p 576.

(4) Naiman, R. J.; Bilby, R. E.; Schindler, D. E.; Helfield, J. M. Pacific salmon, nutrients, and the dynamics of freshwater and riparian ecosystems. *Ecosystems* **2002**, *5*, 399−417.

(5) Janetski, D. J.; Chaloner, D. T.; Tiegs, S. D.; Lamberti, G. A. Pacific salmon effects on stream ecosystems: A quantitative synthesis. *Oecologia* **2009**, *159*, 583−595.

(6) Hocking, M. D.; Reynolds, J. D. Impacts of Salmon on Riparian Plant Diversity. *Science* **2011**, *331*, 1609−1612.

(7) Bartz, K. K.; Naiman, R. J. Effects of salmon-borne nutrients on riparian soils and vegetation in southwest Alaska. *Ecosystems* **2005**, *8*, 529−545.

(8) Bilby, R. E.; Fransen, B. R.; Bisson, P. A. Incorporation of nitrogen and carbon from spawning coho salmon into the trophic system of small streams: Evidence from stable isotopes. *Can. J. Fish. Aquat. Sci.* **1996**, *53*, 164−173.

(9) Chaloner, D. T.; Martin, K. M.; Wipfli, M. S.; Ostrom, P. H.; Lamberti, G. A. Marine carbon and nitrogen in southeastern Alaska stream food webs: Evidence from artificial and natural streams. *Can. J. Fish. Aquat. Sci.* **2002**, *59*, 1257−1265.

(10) Wipfli, M. S.; Hudson, J.; Caouette, J. Influence of salmon carcasses on stream productivity: Response of biofilm and benthic macroinvertebrates in southeastern Alaska, USA. *Can. J. Fish Aquat. Sci.* **1998**, *55*, 1503−1511.

(11) Bilby, R. E.; Fransen, B. R.; Bisson, P. A.; Walter, J. K. Response of juvenile coho salmon (*Oncorhynchus kisutch*) and steelhead (*Oncorhynchus mykiss*) to the addition of salmon carcasses to two streams in southwestern Washington, USA. *Can. J. Fish Aquat. Sci.* **1998**, *55*, 1909−1918.

(12) Wipfli, M. S.; Hudson, J. P.; Caouette, J. P.; Chaloner, D. T. Marine subsidies in freshwater ecosystems: Salmon carcasses increase the growth rates of stream-resident salmonids. *Trans. Am. Fish. Soc.* **2003**, *132*, 371−381.

(13) Gende, S. M.; Willson, M. F. Passerine densities in riparian forests of southeast Alaska: Potential effects of anadromous spawning salmon. *Condor* **2001**, *103*, 624−629.

(14) Christie, K. S.; Reimchen, T. E. Presence of salmon increases passerine density on Pacific Northwest streams. *Auk* **2008**, *125*, 51−59.

(15) Field, R. D.; Reynolds, J. D. Sea to sky: Impacts of residual salmon-derived nutrients on estuarine breeding bird communities. *Proc. R. Soc. B* **2011**, *278*, 3081−3088.

(16) Morrissey, C. A. Effect of altitudinal migration within a watershed on the reproductive success of American dippers. *Can. J. Zool./Rev. Can. Zool.* **2004**, *82*, 800−807.

(17) Elliott, K. H.; Elliott, J. E.; Wilson, L. K.; Jones, I.; Stenerson, K. Density-dependence in the survival and reproduction of Bald Eagles: Linkages to Chum Salmon. *J Wildlife Manage.* **2011**, *75*, 1688−1699.

(18) Ewald, G.; Larsson, P.; Linge, H.; Okla, L.; Szarzi, N. Biotransport of organic pollutants to an inland Alaska lake by migrating sockeye salmon (*Oncorhynchus nerka*). *Arctic* **1998**, *51*, 40−47.

(19) Krummel, E. M.; Macdonald, R. W.; Kimpe, L. E.; Gregory-Eaves, I.; Demers, M. J.; Smol, J. P.; Finney, B.; Blais, J. M. Delivery of pollutants by spawning salmon - Fish dump toxic industrial compounds in Alaskan lakes on their return from the ocean. *Nature* **2003**, *425*, 255−256.

(20) O'Toole, S.; Metcalfe, C.; Craine, I.; Gross, M. Release of persistent organic contaminants from carcasses of Lake Ontario Chinook salmon (*Oncorhynchus tshawytscha*). *Environ. Pollut.* **2006**, *140*, 102−113.

(21) Gregory-Eaves, I.; Demers, M. J.; Kimpe, L.; Krummel, E. M.; MacDonald, R. W.; Finney, B. P.; Blais, J. M. Tracing salmon-derived nutrients and contaminants in freshwater food webs across a pronounced spawner density gradient. *Environ. Toxicol. Chem.* **2007**, *26*, 1100−1108.

(22) Giller, P.; Malmqvist, B. *The Biology of Streams and Rivers*; Oxford University Press, 1998.

(23) Dodds, W. K.; Whiles, M. R. *Freshwater Ecology: Concepts and Environmental Applications of Limnology*, 2nd ed.; Elsevier: San Diego, CA, 2010.

(24) Newman, M. C.; Clements, W. H. *Ecotoxicology: A Comprehensive Treatment*; CRC Press: Boca Raton, FL, 2008.

(25) Chaloner, D. T.; Lamberti, G. A.; Cak, A. D.; Blair, N. L.; Edwards, R. T. Inter-annual variation in responses of water chemistry and epilithon to Pacific salmon spawners in an Alaskan stream. *Freshwater Biol.* **2007**, *52*, 478−490.

(26) Moore, J. W.; Schindler, D. E.; Carter, J. L.; Fox, J.; Griffiths, J.; Holtgrieve, G. W. Biotic control of stream fluxes: Spawning salmon drive nutrient and matter export. *Ecology* **2007**, *88*, 1278−1291.

(27) Kline, T. C.; Goering, J. J.; Mathisen, O. A.; Poe, P. H.; Parker, P. L. Recycling of elements transported upstream by runs of Pacific salmon. I. delta-N-15 and delta-C-13 evidence in Sashin Creek, southeastern Alaska. *Can. J. Fish. Aquat. Sci.* **1990**, *47*, 136−144.

(28) Peterson, B. J.; Fry, B. Stable isotopes in ecosystem studies. *Annu. Rev. Ecol. Syst.* **1987**, *18*, 293−320.

(29) Jardine, T. D.; Kidd, K. A.; Fisk, A. T. Applications, considerations, and sources of uncertainty when using stable isotope analysis in ecotoxicology. *Environ. Sci. Technol.* **2006**, *40*, 7501−7511.

(30) Ormerod, S. J.; Tyler, S. J. Inter- and intra-annual variation in the occurrence of organochlorine pesticides, polychlorinated biphenyl congeners, and mercury in the eggs of a river passerine. *Arch. Environ. Contam. Toxicol.* **1994**, *26*, 7−12.

(31) Morrissey, C. A.; Bendell-Young, L. I.; Elliott, J. E. Linking contaminant profiles to the diet and breeding location of American dippers using stable isotopes. *J. Appl. Ecol.* **2004**, *41*, 502−512.

(32) Morrissey, C. A.; Elliott, J. E.; Ormerod, S. J. Diet shifts during egg laying: Implications for measuring contaminants in bird eggs. *Environ. Pollut.* **2010**, *158*, 447−454.

(33) Morrissey, C. A.; Bendell-Young, L. I.; Elliott, J. E. Identifying sources and biomagnification of persistent organic contaminants in biota from mountain streams of southwestern British Columbia, Canada. *Environ. Sci. Technol.* **2005**, *39*, 8090−8098.

(34) Morrissey, C. A.; Elliott, J. E.; Ormerod, S. J. Local to Continental Influences on Nutrient and Contaminant Sources to River Birds. *Environ. Sci. Technol.* **2010**, *44*, 1860−1867.

(35) Strahler, A. N. Dynamic basis of geomorphology. *Geol. Soc. Am. Bull.* **1952**, *63*, 1117−1142.

(36) Finlay, J. C. Stable-carbon-isotope ratios of river biota: Implications for energy flow in lotic food webs. *Ecology* **2001**, *82*, 1052−1064.

(37) Rasmussen, J. B. Estimating terrestrial contribution to stream invertebrates and periphyton using a gradient-based mixing model for delta 13C. *J. Anim. Ecol.* **2010**, *79*, 393−402.

(38) Hebert, C. E.; Keenleyside, K. A. To normalize or not to normalize? Fat is the question. *Environ. Toxicol. Chem.* **1995**, *14*, 801−807.

(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*, 179−189.

(40) Hobson, K. A. Reconstructing avian diets using stable-carbon and nitrogen isotope analysis of egg components - patterns of isotopic fractionation and turnover. *Condor* **1995**, *97*, 752−762.
(41) Helsel, D. R. Fabricating data: How substituting values for nondetects can ruin results, and what can be done about it. *Chemosphere* **2006**, *65*, 2434−2439.
(42) Helsel, D. R. *Nondetects and Data Analysis: Statistics for Censored Environmental Data*; John Wiley: New York, 2005.
(43) Leps, J.; Smilaur, P. *Multivariate analysis of ecological data using CANOCO*; Cambridge University Press: Cambridge, U.K., 2003.
(44) Gende, S. M.; Edwards, R. T.; Willson, M. F.; Wipfli, M. S. Pacific salmon in aquatic and terrestrial ecosystems. *Bioscience* **2002**, *52*, 917−928.
(45) Kiernan, J. D.; Harvey, B. N.; Johnson, M. L. Direct versus indirect pathways of salmon-derived nutrient incorporation in experimental lotic food webs. *Can. J. Fish. Aquat. Sci.* **2010**, *67*, 1909−1924.
(46) Holtgrieve, G. W.; Schindler, D. E.; Gowell, C. P.; Ruff, C. P.; Lisi, P. J. Stream geomorphology regulates the effects on periphyton of ecosystem engineering and nutrient enrichment by Pacific salmon. *Freshwater Biol.* **2010**, *55*, 2598−2611.
(47) Mitchell, N. L.; Lamberti, G. A. Responses in dissolved nutrients and epilithon abundance to spawning salmon in southeast Alaska streams. *Limnol. Oceanogr.* **2005**, *50*, 217−227.
(48) DeNiro, M.; Epstein, S. Mechanism of carbon isotope fractionation associated with lipid synthesis. *Science* **1977**, *197*, 261−263.
(49) Post, D. M. Using stable isotopes to estimate trophic position: Models, methods, and assumptions. *Ecology* **2002**, *83*, 703−718.
(50) Heintz, R. A.; Wipfli, M. S.; Hudson, J. P. Identification of marine-derived lipids in juvenile coho salmon and aquatic insects through fatty acid analysis. *Trans. Am. Fish. Soc.* **2010**, *139*, 840−854.
(51) Hites, R. A.; Foran, J. A.; Carpenter, D. O.; Hamilton, M. C.; Knuth, B. A.; Schwager, S. J. Global assessment of organic contaminants in farmed salmon. *Science* **2004**, *303*, 226−229.
(52) Hites, R. A.; Foran, J. A.; Schwager, S. J.; Knuth, B. A.; Hamilton, M. C.; Carpenter, D. O. Global assessment of polybrominated diphenyl ethers in farmed and wild salmon. *Environ. Sci. Technol.* **2004**, *38*, 4945−4949.
(53) Krummel, E. M.; Gregory-Eaves, I.; Macdonald, R. W.; Kimpe, L. E.; Demers, M. J.; Smol, J. P.; Finney, B.; Blais, J. M. Concentrations and fluxes of salmon-derived polychlorinated biphenyls (PCBs) in lake sediments. *Environ. Sci. Technol.* **2005**, *39*, 7020−7026.
(54) Baker, M. R.; Schindler, D. E.; Holtgrieve, G. W.; St. Louis, V. L. Bioaccumulation and Transport of Contaminants: Migrating Sockeye Salmon As Vectors of Mercury. *Environ. Sci. Technol.* **2009**, *43*, 8840−8846.
(55) Merna, J. W. Contamination of stream fishes with chlorinated hydrocarbons from eggs of Great-Lakes salmon. *Trans. Am. Fish. Soc.* **1986**, *115*, 69−74.
(56) Christensen, J. R.; Macduffee, M.; Macdonald, R. W.; Whiticar, M.; Ross, P. S. Persistent organic pollutants in British Columbia grizzly bears: Consequence of divergent diets. *Environ. Sci. Technol.* **2005**, *39*, 6952−6960.
(57) Hardell, S.; Tilander, H.; Welfinger-Smith, G.; Burger, J.; Carpenter, D. O. Levels of polychlorinated biphenyls (PCBs) and three organochlorine pesticides in fish from the Aleutian Islands of Alaska. *PLoS ONE* **2010**, *5*.
(58) Shaw, S. D.; Berger, M. L.; Brenner, D.; Carpenter, D. O.; Tao, L.; Hong, C.-S.; Kannan, K. Polybrominated diphenyl ethers (PBDEs) in farmed and wild salmon marketed in the Northeastern United States. *Chemosphere* **2008**, *71*, 1422−1431.
(59) Sloan, C. A.; Anulacion, B. F.; Bolton, J. L.; Boyd, D.; Olson, O. P.; Sol, S. Y.; Ylitalo, G. M.; Johnson, L. L. Polybrominated diphenyl ethers in outmigrant juvenile chinook salmon from the lower Columbia River and estuary and Puget Sound, Washington. *Arch. Environ. Contam. Toxicol.* **2010**, *58*, 403−414.
(60) Elliott, K. H.; Cesh, L. S.; Dooley, J. A.; Letcher, R. J.; Elliott, J. E. PCBs and DDE, but not PBDEs, increase with trophic level and marine input in nestling bald eagles. *Sci. Total Environ.* **2009**, *407*, 3867−3875.
(61) Harris, M. L.; Elliott, J. E.; Butler, R. W.; Wilson, L. K. Reproductive success and chlorinated hydrocarbon contamination of resident great blue herons (*Ardea herodias*) from coastal British Columbia, Canada, 1977 to 2000. *Environ. Pollut.* **2003**, *121*, 207−227.
(62) Drent, R. H.; Daan, S. The prudent parent - energetic adjustments in avian breeding. *Ardea* **1980**, *68*, 225−252.
(63) Chen, D.; Hale, R. C. A global review of polybrominated diphenyl ether flame retardant contamination in birds. *Environ. Int.* **2010**, *36*, 800−811.
(64) Hale, R. C.; Alaee, M.; Manchester-Neesvig, J. B.; Stapleton, H. M.; Ikonomou, M. G. Polybrominated diphenyl ether flame retardants in the North American environment. *Environ. Int.* **2003**, *29*, 771−779.
(65) Elliott, J. E.; Wilson, L. K.; Wakeford, B. Polybrominated diphenyl ether trends in eggs of marine and freshwater birds from British Columbia, Canada, 1979−2002. *Environ. Sci. Technol.* **2005**, *39*, 5584−5591.
(66) Norstrom, R. J.; Simon, M.; Moisey, J.; Wakeford, B.; Weseloh, D. V. C. Geographical distribution (2000) and temporal trends (1981−2000) of brominated diphenyl ethers in Great Lakes herring gull eggs. *Environ. Sci. Technol.* **2002**, *36*, 4783−4789.
(67) Zhu, L. Y.; Hites, R. A. Temporal trends and spatial distributions of brominated flame retardants in archived fishes from the Great Lakes. *Environ. Sci. Technol.* **2004**, *38*, 2779−2784.
(68) Lebeuf, M.; Gouteux, B.; Measures, L.; Trottier, S. Levels and temporal trends (1988−1999) of polybrominated diphenyl ethers in beluga whales (*Delphinapterus leucas*) from the St. Lawrence estuary, Canada. *Environ. Sci. Technol.* **2004**, *38*, 2971−2977.
(69) Covaci, A.; Harrad, S.; Abdallah, M. A. E.; Ali, N.; Law, R. J.; Herzke, D.; de Wit, C. A. Novel brominated flame retardants: A review of their analysis, environmental fate and behaviour. *Environ. Int.* **2011**, *37*, 532−556.
(70) Gillis, E. A.; Green, D. J.; Middleton, H. A.; Morrissey, C. A. Life history correlates of alternative migratory strategies in American Dippers. *Ecology* **2008**, *89*, 1687−1695.
(71) Obermeyer, K. E.; White, K. S.; Willson, M. F. Influence of salmon on the nesting ecology of American dippers in Southeastern Alaska. *Northwest Sci.* **2006**, *80*, 26−33.