Environmental Science & Technology

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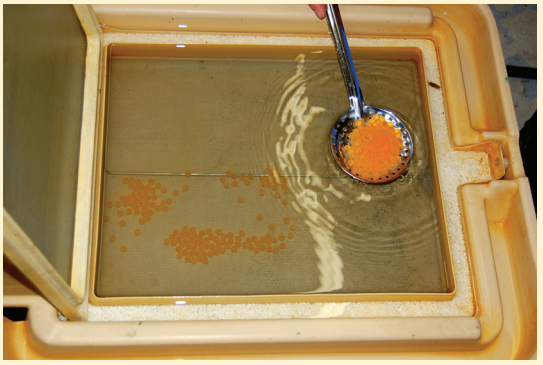
Bioamplification and the Selective Depletion of Persistent Organic Pollutants in Chinook Salmon Larvae

Jennifer M. Daley,*[,†] Todd A. Leadley,[†] Trevor E. Pitcher,[‡] and Ken G. Drouillard[†]

[†]Great Lakes Institute for Environmental Research, and [‡]Department of Biological Sciences, University of Windsor, Windsor, Ontario, Canada

Supporting Information

ABSTRACT: The maternal provisioning of yolk to eggs transfers significant quantities of persistent organic pollutants (POPs). As yolk utilization progresses via metabolic activity, there is a potential to realize further increases in POP concentrations if yolk lipids are depleted at a faster rate than POPs, a condition referred to as bioamplification. This study investigated the bioamplification of POPs in Chinook salmon (*Oncorhynchus tshawytscha*) eggs and larvae. Chinook eggs were sampled from the Credit River, ON, Canada, and brought to an aquaculture facility where they were fertilized, incubated, and maintained posthatch until maternally derived lipid reserves became depleted (approximately 168 days). The loss of chemicals having an octanol−water partition coefficient (log K_{OW}) greater than 5.8 was slow to negligible from days 0−135. However, during the increase in water temperatures in early spring, K_{OW}-dependent elimination of POPs was observed. Bioamplification was maximized for the highest log K_{OW} POPs, with an approximate 5-fold increase in lipid equivalents concentrations in 168 day old larvae as compared to newly fertilized eggs. This study demonstrates that later yolk-sac Chinook larvae (before exogenous feeding) are exposed to higher lipid equivalents POP concentrations than predicted by maternal deposition, which could lead to underestimates in the toxicity of critical life stages.



INTRODUCTION

Salmon are an important economic resource as they support both commercial and sport fisheries as well as form the bulk of aquaculture fish production in Canada.[1−3] Persistent organic pollutants (POPs) including dioxins and polychlorinated biphenyls (PCBs) have been demonstrated to elicit toxic effects on salmon, including growth inhibition, immune dysfunction, and reproductive impairments.[2−5] Toxicity studies have shown that the early life stages of fish (egg and larval stages) demonstrate greater toxicological sensitivity to POPs than do the juvenile and adult life stages.[6−8] Furthermore, life stage specific toxicity studies on salmonids have shown that the yolk sac and early fry stages appear to be a critical period in the life history of salmonids where toxicity-induced mortality is highest.[8−10] This has been interpreted to be due to the yolk sac acting as a "toxicant sink" for hydrophobic compounds, where consequently the fry receives a greater dose of POPs near the end of yolk resorption.[8,10−12]

Chinook salmon (*Oncorhynchus tshawytscha*) have a complex life history that often involves the exposure to POP compounds by bioconcentration and biomagnification processes in the adult life stages that integrate wide spatial gradients of contamination, due to its andromonous life history and its upper trophic level piscivorous feeding ecology. Pacific salmon such as Chinook and sockeye salmon (*Oncorhynchus nerka*), which are semelparous species that return to their native spawning rivers and streams and die postreproduction, have been shown to be significant biological vectors of both nutrients as well as POPs to streams and tributaries.[13−15] This transfer of contaminants from ocean/lake habitats to rivers/streams through decomposing somatic tissues may be mirrored in reproductive products via maternal transfer of POPs to eggs and their subsequent transfer to embryo and larvae.[16,17]

Fish eggs are considered to achieve equilibrium with somatic tissues at the time of egg formation resulting in similar lipid normalized POP concentrations between eggs and maternal tissues.[6,17] Furthermore, fall spawning fish (as exemplified by many populations of Chinook salmon) are required to provide significant maternal resources to eggs to enable overwintering incubation and survival of larvae during a prolonged critical period, which reflects the transition from endogenous (use of yolk reserves) to exogenous feeding.[6,18,19] This increased transfer of maternal resources to the egg results in the transfer of a greater POPs mass to eggs and larvae. The matter is further complicated by fish migrations during spawning, exemplified in dramatic fashion by salmon where some populations are known to swim distances of over 3000 km (range: 1 to >3200 km[20]) and utilize as much as 95% of their somatic lipid contents to

Received: November 9, 2011
Revised: January 9, 2012
Accepted: January 11, 2012
Published: January 11, 2012

support the bioenergetic costs of spawning and reproduction.[4] Under conditions where an organism loses body weight and chemical partitioning capacity faster than it can eliminate contaminants, lipid normalized POP concentrations become enriched. This process was referred to as solvent depletion by Macdonald et al.[21] and was later coined bioamplification.[22,23] deBruyn et al.[4] found that migrating female sockeye tissues exhibited bioamplification factors, characterized as the ratio of lipid normalized tissue concentrations in post- relative to prespawning individuals, of 3.4−5.6 for POP compounds following migration. They also reported bioamplified lipid normalized POP concentrations in the roe of fish post migration. Kelly et al.[3] observed similar trends between eggs and muscle of spawning Pacific salmon and further demonstrated how migration distance of a given population influences the magnitude of POPs bioamplification. Although enrichment of POP concentrations in gonads was found to occur to a lesser extent (1.9−2.5) than measured in somatic tissues, the above studies highlight how migration and bioenergetic costs serve to increase POPs exposure in eggs and larvae.

Our laboratory recently demonstrated that POP residues can be further bioamplified within the eggs of yellow perch (*Perca flavascens*) as a result of yolk utilization by fish embryos during incubation.[22] This process, should it occur in incubating Chinook salmon eggs, would be in addition to POPs bioamplification in eggs occurring from maternal spawning activities described above. It is predicted that bioamplification in fish species such as Chinook salmon, with larger eggs, greater yolk provisioning, and longer incubation periods, will experience higher POP bioamplification than observed for yellow perch. The objectives of this study were to examine whether bioamplification of POP residues occurs in incubating eggs and/or larvae of Chinook salmon and to place this into the context of potential POPs exposures contributed by bioamplification during parental spawning, egg incubation, and in larvae, up to the critical period when larvae rely primarily on the yolk-sac to satisfy bioenergetic costs.

MATERIALS AND METHODS

Experimental Section. Chinook salmon eggs were collected from a single spawning female in the Credit River, Mississauga, ON, Canada (43°34′39.24″ N, 79°42′8.19″ W). The female would have achieved its exposures to POP compounds within Lake Ontario, known for relatively high concentrations of POP compounds including PCBs and mirex.[24] The eggs were brought back to an aquaculture facility (Great Lakes Institute for Environmental Research) in Windsor, ON, Canada, and manually fertilized using the combined milt from three males. The eggs were then incubated in Heath trays using a flow through system receiving filtered water from the Detroit River at in situ temperatures. Water temperatures were recorded daily throughout the experiment. Replicate eggs (n = 10 at each time point) were collected postfertilization at day 1 and at a later egg developmental stage on day 25. The majority of eggs hatched between days 30−34. Similar to natural, under the ice conditions, larvae were not provisioned with food during the experimental trials. Larval Chinook salmon (n = 10 at each time point) were collected on days 35, 61, 91, 105, 134, 155, and when larvae had fully absorbed their yolk on day 168 corresponding to the onset of ice-out and spring temperature increases. Following sampling, the eggs and larvae were kept frozen at −20 °C until chemical and proximate analysis.

Laboratory Processing. Individual eggs or larvae were analyzed to determine neutral lipid content, moisture content, and POP concentrations. POPs and lipids were analyzed using a micro extraction technique described in Daley et al.[22,23] using PCB 30 as a recovery standard. One milliliter of sample was removed for the gravimetric determination of neutral lipids using a microbalance.[25] The remaining extract was concentrated to 2 mL, and sample cleanup was performed by florisil chromatography as described by Lazar et al.[26] In the present study, the first (50 mL hexane; ACP, Montreal, Quebec, Canada) and second fractions (50 mL; hexane/dichloromethane 85/15 v/v; Fisher Scientific, Fair Lawn, NJ) were collected. After florisil chromatography, extracts were concentrated to 1 mL by rotary-evaporator. Samples were analyzed for individual POPs by gas chromatography electron capture detection (GC-ECD). For each batch of six samples, a reference homogenate, method blank, an external PCB standard (Quebec Ministry of Environment Congener Mix; AccuStandard, New Haven, CT), external certified organochlorine pesticide standard (AccuStandard, New Haven, CT), and PCB 30 recovery standard were analyzed. Only PCB congeners (IUPAC no.) 18/17, 31/28, 44, 49, 52, 70, 99, 101, 110, 118, 138, 149, 151/82, 153, 158, 170, 177, 180, 183, 187, 194, and 201, as well as QCB, HCB, OCS, and Mirex, were detected with sufficient frequency to be included in the data analysis. Recoveries of individual PCB congeners in the in-house reference tissue extracted with each batch of samples were within 2 standard deviations from the mean laboratory database value derived from laboratory control charts from the Great Lakes Institute for Environmental Research accredited organic analytical laboratory (Canadian Association for Environmental Analytical Laboratories Accreditation and ISO17025 certified). The recovery of the PCB 30 standard was 87 ± 5.0% across samples.

Data Analysis. Persistent organic pollutant concentrations were expressed on a lipid equivalents basis.[27] The lipid equivalents concentration for each POP congener was calculated as follows:

$$C_{org(lipidequiv)} = C_{org(wet)}/(X_{lipid} + 0.05 \cdot X_{LDP}) \quad (1)$$

where $C_{org(lipid\ equiv)}$ is the POP concentration normalized for lipid equivalents (ng/g lipid equivalents), $C_{org(wet)}$ is the wet weight POP congener concentration (ng/g) in the pooled sample, and X_{lipid} and X_{LDP} are the fractions (unitless) of neutral lipids and lean dry protein (LDP), respectively, in the sample. The lean dry protein was determined by subtracting the total lipid weight from the total dry weight of each sample and dividing by the wet sample weight. The constant 0.05 indicates that lean dry protein has a partitioning capacity that is equivalent to 5% of lipids.[27] Percent lipids and lipid equivalents POP concentrations in the salmon eggs and posthatched larvae at different time intervals were compared using a t test or an analysis of variance (ANOVA) and Tukey's post hoc test where applicable.

POP mass balance estimates were derived by multiplying the wet weight POP concentration (ng/g wet wt) by the mass (g) of individual eggs and larvae at the time of sampling. The bioamplification factor (BAmF) is the lipid equivalents

concentration ratio between two stages of an organism's life cycle.

$$\text{BaMF} = C_{\text{org}(t)}/C_{\text{org}(t-1)} \quad (2)$$

where C_{org} is the concentration in the organism (ng/g lipid equivalents). Bioamplification factors in eggs and larvae were calculated by expressing the ratio of POP lipid equivalents concentrations in the 168 d larvae to the mean fresh (newly fertilized) egg value. Bioamplification factors were contrasted against chemical hydrophobicity (log K_{OW}) values to examine the relationship with chemical physical properties. Log K_{OW} values for PCBs were obtained from Hawker and Connell.[28] The OCs were obtained from Mackay et al.[29]

Unless otherwise stated, all data presented in the text and figures are reported as mean ± standard deviation (SD).

RESULTS AND DISCUSSION

There was no change in lipid content (t test, $p = 0.58$) or total lipid mass per egg (t test, $p = 0.52$) during the egg incubation period. This trend of no lipid loss in the embryo has also been observed with Eurasian perch and turbot and may indicate that these species rely primarily on amino acids and/or carbohydrates rather than lipids to support bioenergetic demands during egg incubation.[30,31]

Alternatively, a significant decreasing linear trend in lipid contents was observed in posthatched Chinook larvae (r^2 = 0.94; $p < 0.0001$). The mean lipid contents (±SD) for individual Chinook salmon eggs and larvae across time are presented in Figure 1 and demonstrate linear lipid decreases in

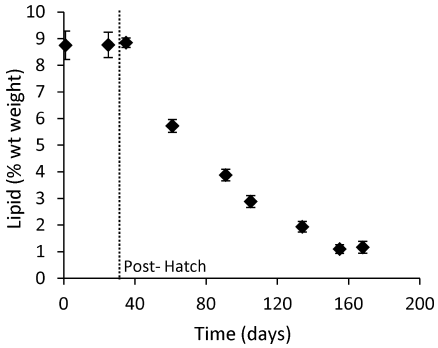


Figure 1. Mean ± standard deviation lipid contents (%) of Chinook eggs and larvae samples over time. Time refers to incubation date relative to newly collected fertilized eggs. Vertical line indicates the day of hatching (n = 6 observations per time point).

larvae through time throughout the experiment. Throughout the egg and larval period, lipid contents of eggs decreased from 8.6 ± 0.21% (range: 7.7–9.0%) in fresh eggs to 1.2 ± 0.21% (range: 0.9–1.5) in 168 d old larvae with 168 d larvae containing only 14% of the total lipid mass originally deposited to the fresh egg. These declines in lipid content are consistent with what has been reported for overwintering larval and juvenile populations of salmon.[32−34] For example, juvenile Chinook salmon from the Yakima River were reported to exhibit lipid declines from approximately 7–8% in early fall to under 2% in early spring.[32]

The experimental design that utilized eggs from a single female to reduce potential interegg variation of maternally deposited POPs, as well as the ability to sample individual eggs/larvae for chemical analysis without pooling, enabled precise mass balances to be performed to test for chemical elimination during egg incubation and in larvae following hatching. Figure 2 presents the change in chemical mass within

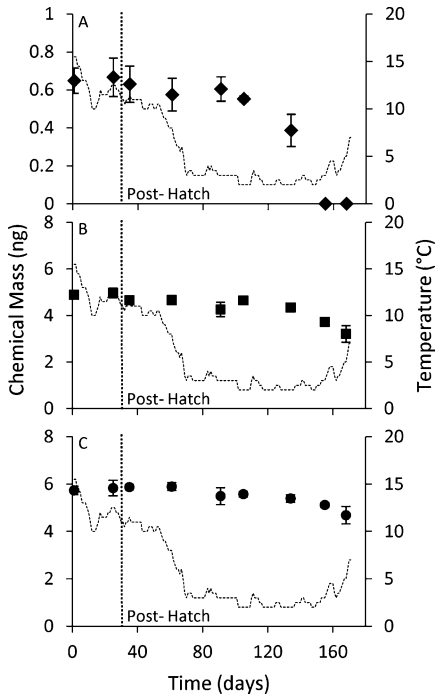


Figure 2. Mean ± standard deviation of chemical mass of selected polychlorinated biphenyl (PCB) congeners in individual Chinook eggs and larvae over time. PCB 18/17 designated by "◆", PCB 99 designated by "■", and PCB 180 designated by "●". Vertical line indicates the day of hatching (n = 6 observations per time point). The secondary axis represented by hatching dots is the temperature profile over the experimental duration.

eggs/larvae over time for three select POP compounds, a representative from the low log K_{OW} (below 6), moderate log K_{OW} (6–7), and high log K_{OW} (above 7) groups: PCB congeners 18/17 (log K_{OW} = 5.24), 99 (log K_{OW} = 6.39), and 180 (log K_{OW} = 7.36). For all chemicals of study, there were no significant changes ($p > 0.05$; ANOVA) in chemical mass between eggs over the period of fertilization to hatch-out.

Following hatch-out, mass balance estimates indicate that POP depuration from Chinook larvae occurred for several compounds, but different trends occurred for chemicals with different hydrophobicities. For the compounds in the low log K_{OW} group, 50–100% of chemical mass was eliminated by day 168 as compared to fresh eggs, whereas for the moderate range log K_{OW} compounds, 19–62% was eliminated, and only 11–24% of chemical was eliminated for those compounds with the highest log K_{OW}'s (Supporting Information 2).

The above observations of slow to negligible chemical elimination in post hatched larvae were unexpected, as generally with small animals chemical elimination is considered to be rapid due to their high surface area to volume ratio.[35,36] There are two hypotheses that could possibly explain the lack of chemical elimination, one being related to temperature and the other to physiological mechanisms. In the present study, the increase in the rate of chemical depuration during days 145–168 coincided with increases in water temperatures related to spring warming (see Figure 2). Water temperature directly regulates the metabolic rate of fish and has been demonstrated

to reduce depuration of POPs from larger more mature fish under cold water conditions.[37,38] For example, slow elimination rates of PCBs were observed in overwintering yellow perch where significant elimination occurred only for the least hydrophobic PCB congeners (log K_{OW} = 5.2) under cold water conditions (<10 °C) as compared to the spring period when elimination of 71 congeners was observed during a much shorter time frame.[37]

However, water temperature/metabolic relationships were not likely the only factors regulating chemical depuration rates observed in larvae from the present study. An examination of the rate of lipid reduction with time (Figure 1) indicates that larvae salmon exhibited steady declines in lipid content and total lipid mass per individual, suggesting that metabolic activity continued to proceed at a relatively high rate over much of the experimental period. Indeed, over the days 0−135 period, larvae lost 78% of their fresh egg lipid mass, representing a majority of lipid declines observed over the total study duration. Thus, while higher water temperatures have been shown to increase yolk absorption rate of larvae fish,[39,40] other factors in addition to increased metabolic rate appear to be important. Developmental studies on Atlantic salmon (*Salmo salar*) demonstrate that gill development increases rapidly by the end of yolk absorption, whereas cutaneous respiration dominates in newly hatched larvae.[41,42] Thus, elevated water temperatures as observed toward the end of the study not only correspond with increased metabolic rate and faster yolk resorption, but also with the development of gills, which can act as a more efficient method of chemical exchange.[43] Given that yolk is the major repository of maternally deposited lipids as well as POPs,[8,10,11,44] it is hypothesized that the combined onset of gill development along with final resorption of yolk sac acted in concert to increase the rate of POP elimination. Because yolk sac resorption rate and developmental cues for gill formation are related to water temperature, further studies to determine how seasonal water temperature profiles interact with these variables and influence POPs loss, and bioamplification would be useful.

As indicated from the mass balance estimates, the lipid equivalents POP concentrations did not change in eggs from fertilization to hatch-out for any of the compounds studied (see Figure 3). This differs with trends reported for yellow perch where PCBs exhibited enriched lipid normalized chemical concentrations in eggs just prior to hatching relative to that measure in fresh eggs.[22] These differences are attributed to the fact that yellow perch eggs showed declines in lipid content of eggs during the egg incubation period, but salmon did not show such declines, whereas both species showed no chemical elimination.

Post hatching, changes in lipid equivalents POP concentrations occurred for all chemicals, but different patterns of concentration trends were evident depending on chemical hydrophobicity. The lowest log K_{OW} congener presented in Figure 3, PCB18/17 (log K_{OW} = 5.24), showed a significant increase in concentration from days 60−100 (ANOVA; p < 0.0001). This was a period of rapid decreases in animal lipid content but little or no change in chemical mass. After day 100, chemical depuration for less hydrophobic compounds caused an attenuation of bioamplification trends such that by day 155, chemical loss rates exceeded lipid utilization rates, causing a drop in lipid normalized chemical concentrations to values well below those present in the fresh egg. Compounds having log K_{OW} values exceeding 6 demonstrated a different pattern of

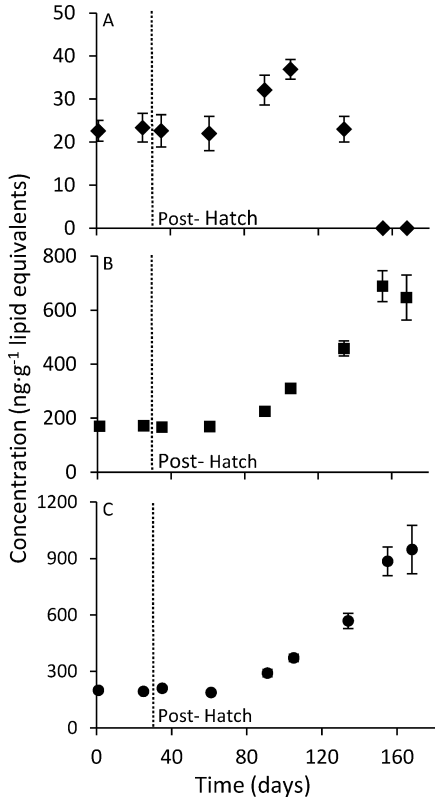


Figure 3. Mean ± standard deviation of chemical concentration (ng/g lipid equivalents) of selected polychlorinated biphenyl (PCB) congeners in Chinook eggs and larvae over time. PCB 18/17 designated by "◆", PCB 99 designated by "■", and PCB 180 designated by "●". Vertical line indicates the day of hatching (n = 6 observations per time point).

lipid equivalents concentrations with time. For these compounds, the slow chemical elimination across time caused an increasing trend in concentration, with the highest concentrations occurring between days 155 and 168.

Bioamplification factors between fresh eggs and 168 d larvae are compared in Figure 4 and plotted as a function of chemical

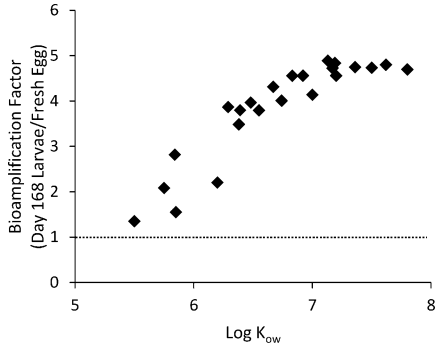


Figure 4. Persistent organic pollutant (POP) bioamplification factors comparing 168 d larvae/fresh egg as a function of chemical K_{OW}. Lower horizontal dashed line refers to the bioamplification factor of 1.

log K_{OW} for all POPs studied except for two compounds. The latter chemicals, QCB and PCB 18/17, had concentrations below detection limits at day 168, and the bioamplification factor would therefore be much less than 1 for these compounds. There was an increasing trend in larvae/egg

dx.doi.org/10.1021/es204003a | Environ. Sci. Technol. 2012, 46, 2420−2426

bioamplification factor with increasing log K_{OW} until it reached an asymptote at log K_{OW} values exceeding 7. The magnitude of bioamplification for the highest log K_{OW} POPs showed an approximate 5-fold increase in chemical lipid equivalent concentrations in larvae as compared to fresh eggs (Figure 4). This magnitude of the maximum bioamplification factor approached the degree of decrease in partitioning capacity (~6-fold) jointly contributed by lipids and lean dry protein.

As noted earlier, the trend of POPs bioamplification in Chinook salmon early life stages differed from that of yellow perch eggs and larvae. Yellow perch showed clear evidence of POPs bioamplification in the eggs, whereas posthatched perch larvae began to eliminate chemical within 2 days of hatching.[22] In Chinook salmon, there was little chemical elimination either in eggs or in larvae for over 100 days post hatching until water temperatures began to increase during spring warming. The differences between these species early life stage bioamplification trends are likely related to life history differences in reproductive strategies. In the present study, Chinook salmon represents a fall spawning, semelparous life history strategy that provisions significantly more lipids to the eggs to survive prolonged under ice periods during egg and larvae development where food resources may be limiting post hatch. Newly hatched salmon typically remain in the gravel until their yolk-sac is nearly depleted.[33] Heming[45] demonstrated at a constant water temperature of 6 °C the time to 50% emergence for Chinook salmon larvae was 192 days, which is similar to the 168 days it took for complete yolk absorption in the present study. Alternatively, yellow perch represent iteroparous spring spawners whose eggs typically hatch in 10−20 days following spawning and emerge as free-swimming larvae capable of feeding immediately following hatching.[46]

This study demonstrates that late stage Chinook salmon larvae are exposed to higher POP concentrations than predicted by maternal deposition, which could lead to underestimates in the toxicity experienced by this species if toxicity studies are confined to the maternal tissues and/or egg life stages. While the early life stages of fish are generally considered more sensitive to toxicological effects than adults,[6] some studies have demonstrated that later larval stages of salmonids experience the greatest sensitivity to maternally deposited POPs evidenced by increased mortality.[8,9] One example is 2,3,7,8-TCDD in Lake Trout (*Salvelinus namaycush*) that at concentrations of 400 pg/g generated deleterious effects on eggs but produced the greatest mortality in yolk-sac larvae.[47] Such trends would be expected given both the concentration increase in tissues and the mobilization of yolk-sac associated POPs to tissues following yolk-sac resorption. It is widely recognized that fish mortalities are increased during the critical period, representing the transition from endogenous maternally deposited yolk lipids to exogenous food resources.[8−10,48] This research suggests that POPs-based toxicity can interact with, and augment, critical-period associated mortality, potentially resulting in underestimates of the true toxic consequences of POP compounds in the environment. In comparing an estimated threshold for PCB concentrations that protects juvenile salmonids from experiencing adverse health effects of 2400 ng/g lipid,[5,49] to the sum of the 22 PCB congeners (21 600 ± 1100 ng/g lipid or 13 800 ± 1800 ng/g lipid equivalents) evaluated in this study at day 168, the concentrations in the present study were observed to be approximately 5−9 fold greater than this threshold. Although this PCB threshold is suggested for juvenile salmon, studies have demonstrated that newly hatched rainbow trout are 20−40 times more sensitive to the lethality of TCDD than 1 year old juvenile rainbow trout.[7] As such, documenting the critical tissue residues in larval fish that have completed bioamplification would be a better reference point then using the fresh egg concentration as they may have both heightened bioamplification factors and toxicological sensitivities. Further research to identify the relative sensitivity of late stage Chinook salmon larvae as compared to embryos using lipid normalized tissues concentrations is needed. Studies verifying bioamplification of POPs in lake trout fry would also be useful to extend the concentration enrichment mechanism posthatch to this more sensitive species. Notable in the present research is that the larvae bioamplification mechanism occurs only for highly hydrophobic substances (log K_{OW} > 6 and maximized for chemicals with log K_{OW} > 7) narrowing the range of chemicals that can elicit enhanced toxicity by concentration enrichment in the larvae life stage.

Finally, it is worth noting that bioamplification processes occur in Chinook salmon at multiple life stages that include both reproducing adults and larvae life stages, which can serve to increase the vulnerability of this species to hydrophobic toxics. deBruyn et al.[4] found that migrating female sockeye tissues showed bioamplification factors ranging from 3.4 to 5.6 between pre- and postmigration areas as a result of lipid utilization to satisfy migration energy costs. These bioamplification factors vary by environment and migration distance experienced by a given population.[3] Although gonads were observed to have exhibited lower bioamplification factors (1.9−2.5) as compared to somatic tissues, gonads did exhibit significant enrichment of POPs residues and are also dependent on population specific migration costs. Using the highest bioamplification factors reported by deBruyn et al.[4] of 2.5 for the postmigration adult female gonads as compared to gonads premigration and the highest bioamplification factor of 4.9 in larval Chinook salmon observed from the present study, the combined bioamplification factor could be as high as 12 in larval Chinook salmon as compared to prespawning maternal tissue concentrations. Arguably the coupling of bioamplification processes across these life stages contributes to POPs exposures that equal, and may exceed, the magnitude of biomagnification processes experienced by adult fish. On the basis of these joint observations, it is hypothesized that the sensitivity of a given salmon population to POPs toxicity can be influenced by a large number of factors including distance/duration and bioenergetics of spawning migration, maternal provisioning of yolk lipids to eggs, environmental temperature trends, and the availability of food resources to larvae during the critical period.

■ ASSOCIATED CONTENT

Supporting Information

SI 1: The mean ± standard deviations of individual POP congener concentrations (ng/g lipid equivalents) are summarized over time in Table 1. SI 2: The mean ± standard deviations of the mass of individual POP congeners (ng) for fresh eggs and 168 day larvae, the fraction of chemical eliminated, and the *p*-value for the regression of chemical mass over time are summarized in Table 2. This material is available free of charge via the Internet at http://pubs.acs.org.

AUTHOR INFORMATION

Corresponding Author

*Phone: (519) 253-3000; fax: (519) 971-3616; e-mail: daleyj@uwindsor.ca.

ACKNOWLEDGMENTS

The present study was supported by a Natural Sciences and Engineering Research Council (NSERC) grant to K.G.D. and NSERC-CGS-D scholarship to J.M.D. Additional acknowledgements are made to Andy Rubaszek from the Ringwood Fish Culture Station and to Michelle Farwell for providing sampling support for the Chinook salmon eggs.

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