

Bioaccumulation and Transport of Contaminants: Migrating Sockeye Salmon As Vectors of Mercury

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Biological transport by migratory animals is increasingly recognized as important to the long-range dispersal of toxic contaminants. Mercury (Hg) contamination is a widespread environmental concern with serious health implications for humans and wildlife. Due to their unique life history, anadromous salmon may act as important vectors for this contaminant, transferring Hg between marine and freshwater ecosystems. Previous analyses have considered contaminant transport by salmon to be unidirectional. These studies have evaluated Hg import to freshwater by spawning adults, but have not quantitatively assessed export through the migration of juveniles to the ocean. To determine the total Hg burden to freshwater systems by sockeye salmon, we reconstructed the net transport of Hg to the Wood River System in Bristol Bay, Alaska accounting for fluxes in (via adults) and out (via juveniles) of the system. Hg concentrations were higher in juvenile than adult salmon. Hg export from freshwater systems by salmon ranged from 3 to 30% of total import. Proportional export by smolts may be higher for populations under heavy exploitation with strong density dependence in juvenile recruitment. Full consideration of contaminant loading by migratory species requires attention to the relative contaminant flux at all life history stages and the effects of density dependent growth and survival.

Introduction

The release of persistent and bioaccumulating contaminants to the environment is an issue of long-standing concern (1). Environmental contaminants are increasingly found in remote and otherwise pristine environments, raising questions as to how these materials are transported at the global scale. Recently the potential for migratory species to concentrate and transport toxic substances across ecosystem boundaries has received considerable attention (2). Biological vectors, through migration, may become the predominant pathway for contaminant introduction to recipient systems. This phenomenon is particularly relevant to contaminants that biomagnify through food webs and may be accumulated in one system and subsequently deposited in another as the

result of a particular species' life history. Deposition by biovectors introduces contaminants directly to recipient food webs at high trophic levels in readily available and concentrated forms. Species that are both anadromous and semelparous, characterized by mortality following migration between marine and freshwater ecosystems, may be especially effective in driving such transfers.

There is an extensive body of research on the role of salmon in transporting nutrients from the ocean to freshwaters (3, 4). More recently, it has been recognized that spawning salmon may also input significant quantities of contaminants (5), leading to elevated concentrations in recipient systems and resident consumers (6, 7). As a result of an anadromous and semelparous life history, salmon accumulate more than 95% of their biomass in the marine environment (3) and deposit this biomass in their natal freshwater systems where they spawn and die. Contaminants acquired in the marine environment are also deposited at spawning grounds (6). Recent studies have found that anadromous salmon may be the primary route of entry for polychlorinated biphenyls and persistent organic pollutants in Pacific watersheds (8) and the main source of contamination to resident fish and foraging bears (9). Zhang et al. (10) estimate that sockeye salmon transport 1 kg/year of methylmercury (MeHg) to Bristol Bay Alaska watersheds.

Mercury (Hg) is a persistent bioaccumulative environmental contaminant that is harmful to human and ecosystem health (11). In aquatic ecosystems, Hg is biogeochemically transformed to MeHg, a highly toxic and persistent form (12) that readily accumulates and magnifies in aquatic communities (13). Hg is released into the environment as a result of human activities and occurs naturally as a result of volcanic eruptions and weathering of soils. Atmospheric transport is suggested as the key mechanism responsible for its distribution to aquatic systems (14).

In addition to physical processes, anadromous salmon provide an important pathway for Hg transport between the ocean and coastal watersheds. Biological transport of anthropogenic contaminants is of particular concern in high latitude environments that have few direct sources of contamination and are subject to large-scale seasonal migrations (15). Contaminants transported by salmon to freshwater systems may affect the contaminant burden of resident fish and other consumers in these watersheds.

Unlike atmospheric transport, which facilitates not only introduction but also removal of contaminants, biovector transport mechanisms may lack a viable loss route, leading to amplification in the recipient system (16, 2). This assumption has dominated existing analyses of biologically mediated contaminant transport. To date, studies via salmon assume the predominant transfer to be unidirectional, whereby the return of spawning adults represents a one-way movement of contaminants from marine to freshwater systems. This ignores the export of contaminants from freshwater systems by juveniles migrating to the ocean as smolts. Recent analyses of nutrient transport by salmon have demonstrated that smolts export a considerable proportion of nutrients imported by adults (17, 18). Depending on the relative Hg load in adults and smolts and the strength of density dependence (the degree to which juvenile abundance is a function of the number of spawning adults), smolts may also export a significant portion of Hg.

We characterized the net flux of Hg in the Wood River system as a case study to illustrate the bidirectional role of salmon as vectors for contaminant transport. Using 15 years of corresponding brood year data for returning adults

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and outmigrating smolts, we estimated the transport of Hg between freshwater and marine environments and evaluated the potential for density dependent effects to influence the mass balance transport of Hg by salmon. We also contrast Hg concentrations in anadromous salmon to concentrations in several species of fishes resident to this system.

Experimental Section

Study Location. Research was conducted in the Wood River system in the Bristol Bay region of southwest Alaska (Map, Supporting Information). The system has an annual mean return of 2.9 million sockeye (*Oncorhynchus nerka*) salmon and a mean annual escapement (returning adult salmon that have escaped the fishery and entered freshwater) of 1.2 million (19). The total watershed area for the Wood River is 3590 km². Development is minimal within the watershed, however the region has a number of abandoned cinnabar (HgS) mines, indicating relatively enriched geological deposits of Hg (Supporting Information).

Sample Collection and Analyses of Hg. From 2001 to 2008, muscle samples were collected from adult sockeye salmon returning to the Wood River (June–July) to determine the contaminant load in fish entering the system. Adults were collected at entry to freshwater or within 2–3 weeks of entry (indicated by ocean coloration, i.e., silver). In 2007 and 2008 whole bodies of outmigrating smolts were collected periodically during the out-migrating smolt run (June–July) to determine contaminant loads in fish leaving the system. Muscle samples from resident fish were taken opportunistically throughout the study period. Whole body samples were homogenized and muscle samples were trimmed to remove skin, fat, and connective tissue. Moisture content was estimated via weight measurements before and after freeze-drying.

Sockeye salmon samples were analyzed at the University of Washington Environmental Health Laboratory. Muscle and whole body samples were acid-digested (1:1 ratio of HNO₃, H₂O) via the Mars Xpress Microwave Digestion System. Total Hg (THg) was determined by inductively coupled plasma mass spectrometry (ICP-MS). National Institute of Standards and Technology reference materials (SRM 1946 - Lake Superior Fish Tissue) were analyzed throughout sample analysis, measuring 107.3% of the certified value of 0.433 µg/g. Percent recovery (SR efficiency) of laboratory spike recovery control samples was 96% in adults and 95% in smolts. Mean relative percent difference (RPD) in replicate samples was 2.6 for adults and 5.6 for smolts.

Samples of fishes resident to the freshwater system were analyzed through the University of Alberta Biogeochemistry Laboratory. Subsamples of fish tissue (250 mg) were digested in 60-mL Teflon bombs using 7 mL of 7:3 (v/v) HNO₃/H₂SO₄. Bombs were heated in a vented oven for 2 h at 125 °C. Samples were allowed to cool and 19 mL of Milli-Q water and 1 mL of BrCl were added. Bombs were closed and heated overnight at 60 °C. A 0.2 mL subsample of digest was then analyzed using SnCl₂ reduction, purge and trap, and cold vapor atomic fluorescence spectrometry (CVAFS). Recovery of National Research Council of Canada certified reference material (DORM-2) was 99.4% of the average certified concentration of 4.47 µg/g. Sample and blank spikes recoveries were >95% and mean RPD was 4.8.

Comparison of THg in Whole Body and Muscle Samples.

Within individual fish, tissues vary in relative Hg concentration (20). Despite this variation, the ratio of muscle to whole body concentration has been shown to be consistent ($R^2 = 0.82$) in fishes across species and trophic groups (21), with Hg in muscle 30% greater than the corresponding whole-body homogenate (22). To standardize measurements on muscle tissues, we converted concentrations to whole-body

burdens according to the relationship in Peterson et al. ((23); eq 1).

$$\log 10(\text{THg}_{\text{Whole body}}) = -0.2712 + 0.9005 \cdot \log 10(\text{THg}_{\text{Muscle Biopsy}}) \quad (1)$$

Numerous studies have demonstrated that greater than 90% of Hg in fish is in the form of MeHg (24–26). THg and MeHg concentrations are highly correlated ($R^2 = 0.93$) (27) and the relative proportion in salmon is not age dependent (28). Our analyses measured THg, which provides a more accurate measure of total Hg flux and facilitates analyses of larger sample sizes.

Reconstruction of Net Flux of Hg via Salmon. We reconstructed gross import and export of Hg using smolt migration and adult return (escapement) data from the Alaska Department of Fish and Game (ADFG) and University of Washington Alaska Salmon Program. Estimates of the number of spawning adult sockeye salmon and their migration timing were based on hourly sampling throughout the run via visual counting towers, which enumerated fish that had transited the fishery and entered the Wood River system (29). The age composition and average mass-at-age of adults were estimated through weight measurements and analyses of scale annuli from daily representative in-river sampling of the run (30). Estimates for the number and timing of the outmigration of sockeye salmon smolts were determined through hydroacoustic surveys conducted at the outflow of the Wood River system (31). The age composition and average mass-at-age of smolts were estimated through weight measurements and scale annuli taken from daily fyke net samples conducted throughout the duration of the smolt migration (31, 32). The vast majority (>90%) of smolts in the Wood River rear for one year (rather than two) in freshwater (31). All smolts included in analyses of mercury concentration spent one year in freshwater.

Import and export of Hg by salmon was calculated on a brood year basis, such that each year of outmigrating smolts corresponds to the population of adult spawners that produced it (Supporting Information).

For each brood year t , we calculated gross import of Hg, as:

$$\text{import}_t = \sum_{\text{age}} (\text{escapement}_{\text{age},t} \cdot \overline{\text{mass}}_{\text{age},t} \cdot \text{THg}_{\text{adult}}) \quad (2)$$

where import is the sum, across ages, of the product of the age-specific escapement of spawning adults in year t , the average mass of that age class in year t , and the estimated percent wet weight content of THg in returning adult sockeye.

We calculated gross export of Hg by smolts produced from brood year t as:

$$\text{export}_t = \sum_{\text{age}} (\text{smolt migration}_{\text{age},t} \cdot \overline{\text{mass}}_{\text{age},t} \cdot \text{THg}_{\text{smolt}}) \quad (3)$$

where export is the sum, across ages, of the product of the age-specific outmigration of juveniles in year t , the average mass of that age class in year t , and the estimated percent wet weight content of THg in sockeye smolts.

Results and Discussion

THg Concentrations in Adults and Smolts. Mean whole body values for THg in adult sockeye salmon entering the Wood River system were 0.0242 ± 0.0081 µg/g (SD) wet weight ($n = 59$). Mean whole body concentrations for sockeye salmon smolts exiting the Wood River system were slightly higher than adults (0.0277 ± 0.0081 µg/g, $n = 80$). Our range of THg

values for adults ($0.01\text{--}0.05\text{ }\mu\text{g/g}$ ww) was similar to values ($0.02\text{--}0.06\text{ }\mu\text{g/g}$ ww) reported in other studies of adult sockeye salmon in this region of Alaska (10, 33) and 10 times lower than the EPA consumption advisory of $0.29\text{ }\mu\text{g/g}$ (11).

We assume Hg in adult salmon is entirely of marine origin due to the fact that sockeye salmon accrue more than 95% of their lifetime growth at sea (34). We assume Hg in juvenile salmon is entirely of freshwater origin due to minimal maternal transfer of Hg from spawning females to their eggs and rapid growth and turnover of tissue in juvenile salmonids prior to smoltification. Hg concentrations in sockeye salmon eggs have been shown to be a small percentage (<10%) of that in corresponding adult muscle tissue (26, 10).

Variation in THg as a Function of Ontogenetic Growth and Condition. Bioaccumulation is related to time of exposure, ontogenetic growth, and accumulation kinetics. Since Hg has high retention and gradual elimination rates, Hg levels will remain constant relative to sudden shifts in mass; thus Hg concentration will vary as a function of body condition or relative energy density (35–37). Previous studies have attributed seasonal and interannual variability in fish Hg concentrations to fluctuations in fish growth and body condition (38). Faster growth rates yield proportionally less accrual of Hg per unit growth. Hg concentrations therefore decrease with increased growth efficiency (39, 40). Conversely, starvation leads to elevated concentrations (35, 41). As fish catabolize muscle tissue to access energy reserves, Hg is redistributed within remaining tissues, increasing the relative concentration (35).

This relationship between Hg concentration and fish condition has relevance to our comparison of THg concentrations in adult returns and outmigrating smolts. We noted significant differences in THg concentration as a function of sampling date (Figure 1). Adult concentrations increased through the migration season whereas smolt concentrations decreased. Since smolts experience higher growth rates in summer months (32), THg concentrations are expected to decrease as a result of rapid summer growth (42). In contrast, returning adult sockeye salmon are in a state of physiological stress and starvation once they enter fresh water. Thus THg concentrations will increase as a function of freshwater residence.

We sought to determine mean THg concentrations for fish at the stage corresponding to our estimates of biomass exiting and entering the system. Biomass estimates were calculated from daily sampling, which accounts for growth over the season. To account for decreasing THg concentration in smolts related to increasing growth, we fit a linear regression to THg values as a function of calendar date to estimate THg concentrations at the midpoint of the migration. The estimated value for the peak smolt outmigration date of June 26 is $0.0295\text{ }\mu\text{g/g}$ THg (Supporting Information). In the case of adults, we assume that fish enter the system in robust condition and lose energy density only after freshwater entry. We therefore set our baseline value for returning adult sockeye salmon to the mean THg concentration of adults sampled at entry into freshwater ($0.0248 \pm 0.0067\text{ }\mu\text{g/g}$, $n = 30$). We used these standardized THg values (adults = $0.0248\text{ }\mu\text{g/g}$; smolts = $0.0295\text{ }\mu\text{g/g}$) in our analyses of net Hg flux.

Net Hg Import to the Wood River System by Salmon.

We estimate that adult salmon imported 80 ± 37 (SD) grams and smolts exported 8 ± 4 g for a net annual import of 71 ± 37 g of Hg to the Wood River system, given a mean annual escapement of $1,287,000 \pm 672,000$ (1973–1987) (Figure 2). The percentage of parental Hg that smolts exported was variable across years (3–30%), driven primarily by the number of smolts produced relative to the number of spawning adults ($R^2 = 0.86$). In many years a substantial portion of the contaminant load imported by adults is exported in subsequent years by outmigrating smolts. At lower densities of

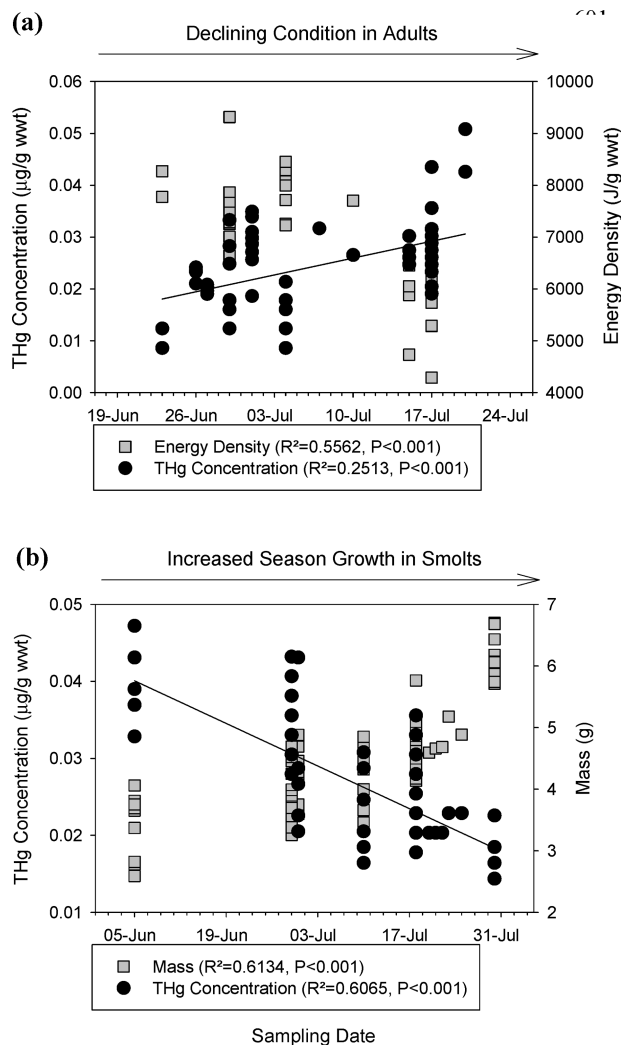


FIGURE 1. (a) THg concentration and energy density in adults as a function of calendar date of sampling. There is a distinct increase in THg concentration over time and a corresponding decrease in energy density (Supporting Information). Many adults displayed here were sampled after entry to the Wood River system, therefore sampling date is not an accurate measure of date of freshwater entry. (b) THg concentration and mass in smolts as a function of calendar date. Smolt mass increased over the season due to improved growth opportunities while THg concentrations decreased due to growth dilution. Due to allometric growth at this life stage (smolts grow long relative to their weight), mass rather than energy density or condition factor is used as a metric to demonstrate growth dilution.

adult spawners (low escapements) smolts export a greater portion of this contaminant burden. On average smolts exported 12% of the Hg brought into the system by their parental spawning class.

No significant differences in adult sockeye salmon Hg concentrations have been recorded among several of the major river systems in southwestern Alaska (10). However, extending our results from the Wood River system to other Bristol Bay watersheds is complicated by site variability in natal systems (geology and terrestrial Hg available to juveniles), differences in the life history of sockeye in each system (juvenile growth rates and age at smoltification), and differences in sockeye productivity in each system. The relative export of Hg by smolts is also influenced by the density dependent dynamics characteristic of each watershed. Where exploitation reduces adult returns, smolts will

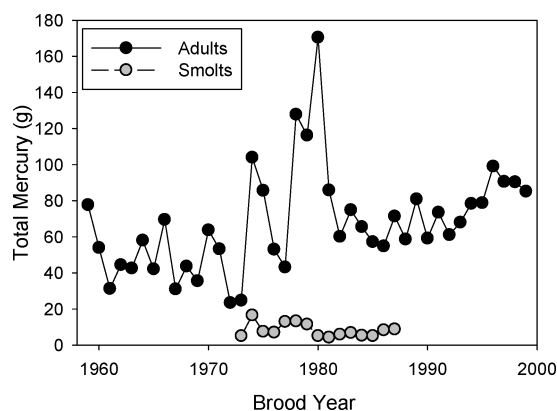


FIGURE 2. Import and export of Hg by spawning adults and outmigrating smolts.

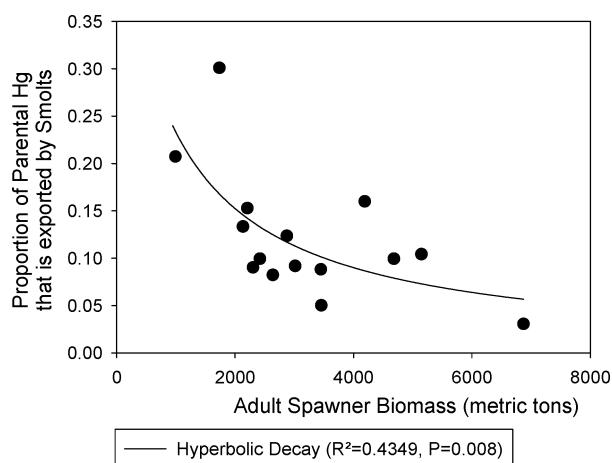


FIGURE 3. Proportion of parental Hg (Hg imported by adult spawners) that is exported by their progeny (smolts) as a function of adult spawner biomass.

demonstrate greater relative export in systems with high per capita production and strong density dependent relationships.

Influence of Density Dependent Population Dynamics on Hg Transport by Salmon. The strength of density dependent relationships determines how many smolts are produced per adult across a range of spawning densities. Increased numbers of spawning salmon will increase the net import of Hg to freshwater systems, not only through increased amounts of Hg deposited in freshwaters, but also through an increase in the mass of adults entering the system relative to the mass of smolts exiting the system. Due to density dependent constraints on habitat and food availability, there is a carrying capacity for smolt production. At high adult spawning densities, the progeny of that brood year may experience reduced juvenile survival (43), slower growth rates (19), or both. Thus the biomass of smolts exiting the freshwater system will asymptote at some threshold capacity independent of how many adults return to spawn. We found the proportion of Hg imported by adults that was subsequently exported by their progeny declined as a function of spawner biomass in a relationship that approximates a hyperbolic decay (Figure 3). Hg export by smolts will have a greater relative impact on the net flux of Hg at low spawning densities. At high spawning densities, Hg imported by adults will overwhelm smolt export due both to the relative difference in biomass at each stage and capacity thresholds to smolt production in lake rearing environments.

To explore the potential effects of density-dependent juvenile growth and survival on the net balance of Hg transport by sockeye salmon, we modeled Hg flux in the

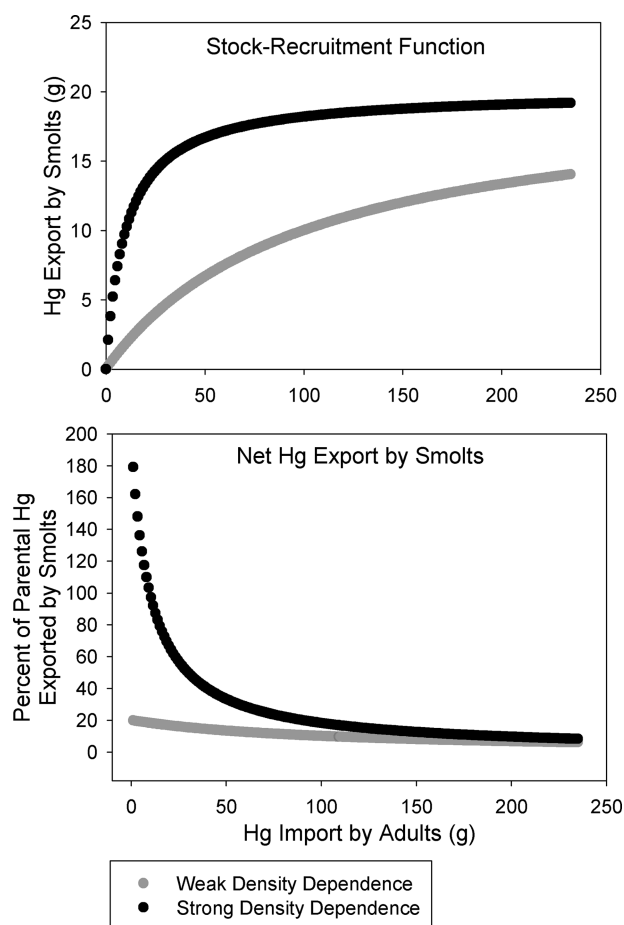


FIGURE 4. Impact of the strength of density dependence on Hg export by smolts. The top graph displays stock-recruitment curves according to the Beverton–Holt model, which suggests that production of juveniles will decrease at higher adult spawning densities and approach an asymptotic capacity for the system. The bottom graph displays the percentage of parental Hg exported by juveniles as a function of Hg import by adults. In both scenarios $a = 20$ g Hg. Weak density dependence is approximated as $b = 100$ g Hg. Strong density dependence is approximated as $b = 10$ g Hg.

Wood River system through the Beverton–Holt (44) stock–recruitment relationship.

$$\text{export}_t = \frac{a \cdot \text{import}_t}{b + \text{import}_t} \quad (4)$$

where the export of Hg in brood year t is a function of the import of Hg for that brood year, a is the maximum export of Hg by smolts, and b is the Hg import needed to produce average Hg export equal to 50% of maximum Hg export ($a/2$).

This model can also be expressed as a ratio of export to import, such that:

$$\frac{\text{export}}{\text{import}} = \frac{a}{b + \text{import}} \quad (5)$$

We fit parameters a and b to observed data by minimizing the negative log-likelihood of the model (45) using brood-year-specific export and import of Hg, with the assumption of log-normally distributed errors. Maximum export (a) for the system was estimated at 20 g Hg (roughly equivalent to a maximum recruitment of 117×10^6 smolts).

Insight into relationship between import and export can be derived through the parameters of the Beverton–Holt

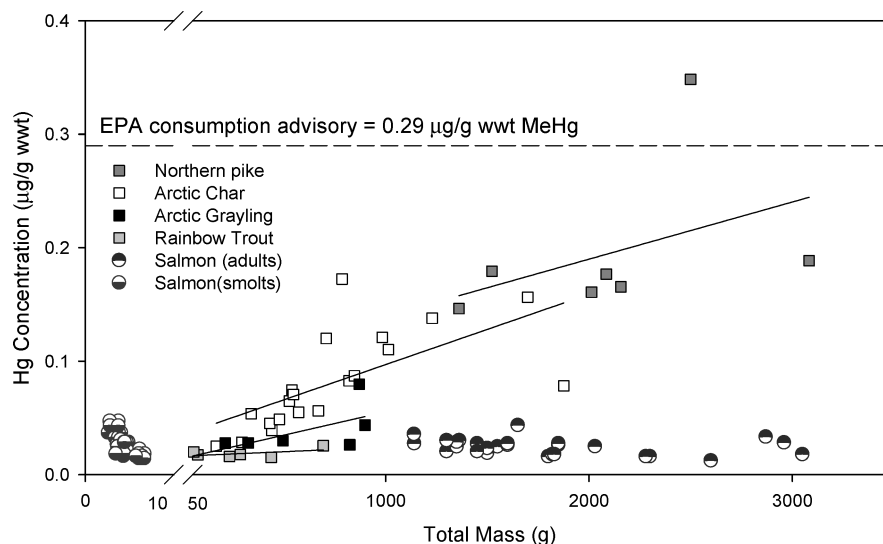


FIGURE 5. Whole body mercury concentrations of resident fish and salmon (adults and smolts) as a function of total mass. We found significant correlations in Arctic char ($R^2 = 0.4228$, $P = 0.002$) and Arctic grayling ($R^2 = 0.4552$, $P = 0.097$), though not in Northern pike ($R^2 = 0.1793$, $P = 0.344$) or Rainbow trout ($R^2 = 0.2610$, $P = 0.300$). Within species, concentrations vary with size, age, and growth rate (26, 36). Among species, Hg concentrations vary with trophic position (13), prey source (49), and habitat-specific foraging (50). Differences among resident fishes in relative THg concentration by mass are likely due to differences in physiological determinants (accumulation and elimination rates), trophic level, longevity, habitat, and diet.

recruitment function. By solving for import as a function of a and b , we are able to determine the combinations of a and b that will produce net import, net export, and a net balance in contaminant flux between marine and freshwater systems. At equilibrium, where import is set equal to export, both are equal to a minus b . According to the parameters we estimated for the Wood River system (1973–1987), the value of b (101) is greater than a (20), thus for all densities of adult spawners, smolts will export less Hg than their parents import to this system. This may not be true in all systems. Smolt export may be higher in systems with elevated local Hg or strong density dependent dynamics. Conversely, in systems with low levels of locally bioavailable Hg, the impact of smolt export may be reduced.

To demonstrate the effect of density dependent juvenile production on the net transfer of Hg between marine and freshwater systems, we assumed a constant value for a (maximum export = 20 g Hg) and applied different values for b , which alters the steepness of the curve at the origin or the relative strength of density dependence in recruitment to the population (Figure 4). In the case of weak density dependence, the target escapement of adult spawners to the Wood River system (1.1×10^6) would be expected to produce a cohort of smolts that would export 16% of the Hg imported by their parents. At the upper range of spawner densities observed in this system (6×10^6), juveniles from that brood year would export only 8%. In the case of strong density dependence, the relative difference in Hg export at high versus low spawner densities would be greater. At 1.1×10^6 adults spawners, juveniles would export 56% of the Hg imported by spawners and 12% at spawner densities of 6×10^6 .

Understanding the effect of density dependence in populations has useful application in the context of fisheries exploitation. Increasing harvest rates will decrease net Hg (or any environmental contaminant) transport to freshwater ecosystems, not only through reduced adult import (interception of adults), but also due to increased relative export by smolts (reduction of density dependent constraints on smolt production). This is a particularly salient issue given the high rates of exploitation of adult salmon returns in Bristol Bay (mean exploitation rate of 56%, 1955–2006).

Hg in Resident Fish and Transfer from Salmon. Nearly all resident fish sampled from the Wood River system had

higher THg concentrations than anadromous sockeye salmon: Rainbow trout (*Oncorhynchus mykiss*) $0.0186 \pm 0.0037 \mu\text{g/g}$, $n = 6$; Arctic grayling (*Thymallus arcticus*) $0.0358 \pm 0.0211 \mu\text{g/g}$, $n = 7$; Arctic char (*Salvelinus alpinus*) $0.0814 \pm 0.0421 \mu\text{g/g}$, $n = 20$; northern pike (*Esox lucius*) $0.1951 \pm 0.0690 \mu\text{g/g}$, $n = 7$ (Figure 5). Our values for THg concentration in resident fishes were similar to those reported elsewhere in the region (Supporting Information).

Elevated THg concentrations in resident freshwater fish relative to anadromous salmon are likely a function of high background Hg in the sediments of southwest Alaska (46), higher methylation rates in freshwater than marine environments (25), the increased longevity of resident fish, and slow growth dynamics in these cold and unproductive systems (47). Rainbow trout, which have the lowest THg concentration among resident fishes sampled, are unique in deriving a significant portion of their energetic budget as a direct subsidy from adult salmon and display isotopic signatures associated with marine food webs (48). In contrast, arctic char and northern pike depend nearly exclusively on resident food sources.

Impact of Salmon on Hg Loading in Southwest Alaska.

Total atmospheric deposition to the lakes in this system is estimated at roughly 2100 g Hg (Supporting Information). This suggests the amount of Hg brought to freshwater by anadromous salmonids is low relative to atmospheric and geological loading. Our analysis suggests that smolt export further reduces salmon-derived mercury inputs to lakes. The availability of Hg to freshwater biota is driven not only by volume but also methylation rates and bioaccumulation rates in freshwater food webs. Therefore, the relative effect of salmon-derived mercury transport is highly dependent on how Hg is integrated into aquatic food webs (Supporting Information). Since Hg delivered by salmon is in a concentrated and readily bioavailable form (MeHg), it may have a disproportionate impact on aquatic food webs. Alternatively, it is possible that salmon may reduce Hg concentrations in freshwater food webs through positive effects of nutrient and energy subsidies to the growth of resident consumers relative to Hg import. Without comprehensive data on how inputs from salmon are integrated into these systems relative to atmospheric and geologic watershed loading, the impact

of salmon imports on the contaminant burdens in aquatic systems is uncertain.

Regardless of the relative magnitude of Hg transport by salmon to this system, it is clear that any comprehensive analysis of contaminant loading by migratory species should consider the relative contaminant fluxes at all relevant life stages. In the case of salmon, this requires attention to both returning adults and outmigrating smolts. This also requires quantifying the dynamics of populations to understand the effect of adult densities on juvenile growth and survival and how commercial exploitation will influence net transport. Our analysis provides insight as to how incorporating juvenile emigration impacts analyses of Hg transport by salmon. While it may not be feasible to accurately measure juvenile export in many cases, it is worth recognizing that it may represent a significant source of export from freshwater to marine systems. Future analyses of biovector contaminant transport should acknowledge if not explicitly account for similar effects, given the specifics of the life history of the particular vector species.

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Supporting Information Available

Additional information on atmospheric and geological sources of Hg to this system, methodology related to biomass estimates and brood year construction, bioaccumulation in salmon and resident fishes, and health implications of fish consumption; a map of sampling areas in Bristol Bay, AK. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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