2022 SALISH SEA TOXICS MONITORING SYNTHESIS

a selection of research

PUGET SOUND ECOSYSTEM MONITORING PROGRAM

The Salish Sea
& Surrounding Basin
The Salish Sea extends from the north end of the Strait of Georgia to the south end of the Puget Sound and west to the mouth of the Strait of Juan de Fuca
Mt. Waddington
Monmouth Mountain
Desolation Sound
Squamish River
Basin Boundary
N. Gulf Islands
Vancouver Island
Strait of Georgia
Fraser River
S. Gulf Islands
Mt. Baker
Skagit River
San Juan Islands
Cape Flattery
Strait of Juan de Fuca
Puget Sound
Glacier Peak
Pacific Ocean
Olympic Peninsula
Hood Canal
Nisqually River
Mt. Rainier
N
0 50 100 km.
0 50 mi.
Data Sources:
SRTM (CGIAR-CSI), NOAA, Wa. DFW, Wa. DNR, BC Digital Atlas, ESRI
stefan freelan - 2009
http://staff.wwu.edu/stefan/SalishSea.htm

Dedication

We owe a debt of gratitude for the innovative, collaborative and effective work spearheaded by our colleague and friend Ken Zarker, who passed in 2021. Our efforts and successes to make our lives safer and healthier are rooted in his optimism and integrative approaches. He is dearly missed.

Puget Sound Ecosystem Monitoring Program (PSEMP)

This effort was organized through the Puget Sound Ecosystem Monitoring Program Toxics Workgroup. The Toxics Workgroup focuses on improving toxics-related monitoring in the region by encouraging coordination and collaboration, identifying priorities and gaps, and increasing the knowledge and understanding. The group meets bi-monthly.

More information can be found at: https://pspwa.box.com/s/cbyi59r5gpjk7yro89k0pb4g61gi3ksm

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Credits

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Disclaimer

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INTRODUCTION

The Puget Sound Ecosystem Monitoring Program (PSEMP) toxics workgroup aims to improve collaboration and information exchange among professionals working to understand the impacts of toxic contaminants in the Salish Sea. One way we achieve this is by compiling and publishing a synthesis report that includes brief updates on the monitoring, research, and management of toxic contaminants.

This is the third synthesis published, with those prior in 2016 and 2018. Together they provide a suite of snapshots of activities that document progress in our investigation, understanding, and management of anthropogenic chemicals. Our main goal is to provide a summary of toxics-related monitoring and research in a single document to 1) communicate the range of efforts currently underway, 2) provide a brief summary of findings to managers and policy makers, and 3) form a basis to develop an inventory of research needs and monitoring gaps.

SECTION 1: STATUS AND TRENDS

SECTION 2: RESEARCH

SECTION 3: MANAGEMENT

This 2022 version represents a compilation of activities of almost 50 groups in both the United States (U.S.) and Canada. We would like to note our appreciation to the contributors and applaud their dedication to their work.

The synthesis is organized into three broad categories: status and trends, research, and management. Section 1: "What is happening? (Status and trends)" includes work that examines the presence, distribution, magnitude, and change through time of contaminants in the Salish Sea ecosystem. Section 2: "What is being learned? (Research)" includes investigations that assess why changes in contaminant levels may be occurring, as well as new methods that may allow us to improve our approaches to assess contaminants. Section 3: "What is being done about it? (Management)" describes efforts that address and remediate toxic contaminants in the environment.

Our community of practice recognizes that many of the contaminants we address through our monitoring programs lead to adverse human health impacts. While at this time we are not evaluating human health directly, we do have many collaborative partners who are working in that area.

1. What is happening? | Status and trends

The status and trends of toxic contaminant levels in the ecosystems of the Salish Sea are monitored by numerous groups, including research conducted under PSEMP (Langness et al. (a,b); Weakland et al.; Carey et al.) in the U.S., and under the newly formed Pollution Tracker program in Canada (Ruberg et al.; Kim et al.). The goal of monitoring is to assess the status and trends of contaminants in the environment and document measurable changes relative to baseline, including progress towards the reduction of contaminants.

The Status and Trends Section of the Toxics Synthesis presents 19 contributions from regional monitoring groups. These contributions broadly assess: (1) the impacts to adult and juvenile Chinook salmon from both legacy and emerging contaminants, (2) the prevalence of both emerging and legacy chemicals in the Salish Sea basin using a variety of abiotic and biotic media, (3) the presence of microplastics, and (4) baseline datasets associated with large-scale cleanup efforts in the Puget Sound basin.

In recent years, the health of resident killer whales (Orcas) has become a focus. Chinook salmon are the primary prey of the Northern (NRKW) and Southern Resident Killer Whales (SRKW). In their contribution, Holbert et al. investigated the legacy contaminant profile and stable isotope ratios of returning adults from 10 priority Chinook populations within the Fraser River Basin. They found variation in contaminant profiles and concentrations among the populations, and suggested that stable isotopes ratios indicate possible differences in feeding. Kim et al. also focused on legacy contaminants possibly impacting NRKW and SRKW by measuring polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in sediments within critical habitat for resident killer whales. They found these contaminants were widespread and possibly of sufficient concentration to pose a threat to resident Orcas. Brown et al. expanded the suite of contaminants (total of 714) to include many emerging chemicals in water samples from the Fraser River Basin. Because there are no environmental thresholds for the vast majority of these contaminants, they used a risk-based approach to prioritize contaminants of concern. Urban environments and wastewater treatment plants appeared to be associated with higher concentrations of contaminants.

The potential impacts to juvenile out-migrating Chinook salmon from contaminants have also been of interest when considering the health of resident killer whales. Contaminant levels may impact the disease susceptibility of Chinook populations and affect successful return rates. This is central to the work of Carey et al., who analyzed contaminant concentrations in the tissues of juvenile Chinook from 12 large Puget Sound rivers, representing estuarine and nearshore locations. Juvenile Chinook from five of these rivers had concentrations of PCBs and PBDEs high enough to potentially impact their health and increase mortality. In the Snohomish River, where PBDE concentrations were high in juvenile Chinook, Gipe et al. present their findings from a source identification study. The authors have been measuring PBDEs in the Snohomish basin since 2019 in a variety of media. Their work shows the composition of PBDEs changes during bioaccumulation of these compounds and that there are elevated concentrations in the Snohomish estuary and Skykomish River near wastewater treatment plant outfalls that may be impacting juvenile Chinook. In the Fraser River basin, both legacy and emerging contaminants were analyzed in juvenile Chinook tissues and compared with results from water samples from the same locations (Lo et al.). Over 600 chemicals were analyzed and approximately 50% of them were detected in juvenile Chinook tissue. Very few of the chemicals have regulatory thresholds.

While legacy organic contaminants are often the focus of monitoring work looking at impacts to juvenile Chinook and other salmonids, inorganic contaminants such as copper are also being investigated. Waugh et al. present results from the Strait of

Georgia in the Metro Vancouver area where the copper binding ligand pool was sampled to ascertain the bioavailable dissolved copper over multiple seasons and locations. The results provide a snapshot of how copper speciation varies both spatially and temporally in this area of the Salish Sea.

Per- and polyfluoroalkyl substances (PFAS) have received more attention in the scientific community over the last few years. This group of chemicals are prevalent in many industrial and consumer products. Following earlier studies documenting PFAS in Washington lakes and rivers, Wong et al. present the first phase of a source identification study in the Lake Washington watershed. PFAS were detected in 224 of 226 lake, tributary, groundwater, stormwater, and bulk atmospheric deposition samples collected over multiple events. The authors highlight the prevalence of PFAS compounds that were phased out of production, but remain in the environment in multiple media. Also in the Puget Sound region, Strivens et al. analyzed four previously-collected sediment cores for 24 PFAS compounds. The long-chain, perfluorooctanesulfonic acid (PFOS) was the sole compound detected. The largest fluxes of PFOS to the sediments were documented in Central Puget Sound near Seattle.

Contaminant concentrations in mussel tissue throughout Puget Sound have been used for many years to assess nearshore inputs of legacy contaminants. In one of two contributions, Langness et al.(a) show the applicability of using transplanted mussels to assess the inputs of legacy contaminants from stormwater in the nearshore environment, both spatially and temporally. While the spatial extent of contamination by legacy contaminants (e.g., PCBs, dichlorodiphenyltrichloroethane [DDT] and PBDEs) has not changed since 2015/2016, the mean or median concentrations have decreased. In their second contribution, Langness et al.(b) present an overview of emerging chemicals in transplanted mussels deployed for three months at 55 locations. The most frequently detected contaminant of emerging concern (CEC) use groups were antibiotics, PFAS, alkylphenol ethoxylates, psychiatrics, and antidiabetics. Lanksbury et al. present a baseline dataset of emerging chemicals in fish tissues from urbanized Seattle area waterways. In rockfish and smallmouth bass tissues, 20 and 9 CECs, respectively, were detected. Also in the Seattle area, Replinger et al. present a baseline dataset of legacy contaminants in fish and shellfish from the Lower Duwamish Waterway (LDW). The samples were collected in 2018 and are being used to track reductions in contaminant concentrations following early sediment remedial actions. Chemical concentrations were found to be above the target tissue levels presented in the LDW Record of Decision.

The spatial coverage of microplastics is also being monitored. In the contribution by Sorenson, a GIS map is presented for the northern Salish Sea that highlights areas of highest potential microplastic accumulation based on six physical marine characteristics and the overlap with nine ecological parameters. The areas of highest risk include salmon-bearing stream estuaries, protected areas, and marine mammal haul-outs. The contribution by Hall presents two years of microplastics monitoring data on one major Puget Sound river, the Puyallup, in the southern Salish Sea. The results showed a higher microplastic abundance in 2018 compared to 2017 with spatially variable abundance and composition (fibers and fragments).

The spatial status of emerging and legacy contaminants is central to Pollution Tracker (pollutiontracker.org), which is the first long-term marine pollution monitoring program in Canada, established in 2015 along the British Columbia coast (Ruberg et al.). The program includes 79 sampling locations and monitors 550 analytes across 14 contaminant classes. Results from sediment samples collected from 2015-2020 highlight the diversity of contaminants present in the marine environment. Within Puget Sound, Weakland et al. present the most recent results from the long-term sediment monitoring program as part of the PSEMP. Weakland et al. analyzed 87 chemicals across 50 sampling locations. The highest concentrations of contaminants were detected near Seattle, Tacoma, Bremerton, and Olympia.

Monitoring of contaminants in sediments from historically contaminated locations in the Seattle area was the focus of three contributed papers. Eash-Loucks et al. presented continued monitoring data for a location of historical wastewater discharge (north shelf of Discovery Park) that had been sampled in the 1990s. Previous sampling documented elevated concentrations of polycyclic aromatic hydrocarbons (PAHs), which appear to be decreasing and may suggest a low rate of natural recovery. South of this study in the LDW, McGroddy et al. present baseline concentrations of PCBs, dioxins/furans, arsenic, and carcinogenic PAH (cPAH) toxic equivalents (TEQs) in sediment following early actions. Spatially-weighted sediment concentrations were established as a baseline from which to assess site-wide concentrations following the sediment remediation. Also in the LDW, Godtfredsen et al. present an overview of sediment sampling results from the initial phases of the sediment remedial design. This pre-design investigation provided data from hundreds of locations and resulted in an estimate of 14.6 acres needing remedial action.

2. What is being learned | Research

Human development in the Salish Sea region brings with it landscape changes that lead to inputs of contaminants to marine and fresh waters. These contaminants originate from both nonpoint sources (i.e., runoff from impervious surfaces, agricultural inputs, and aerosols from car exhaust and wood smoke, etc.), and point sources (i.e., oil spills, industrial and municipal wastewater discharges, etc.).

The papers in this section reflect the diversity of contaminants entering the Salish Sea and the complexity of their potential influence on Salish Sea resources. A variety of contaminants were investigated, including CECs such as pharmaceuticals and personal care products (PPCPs), N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine (6PPD-quinone), as well as familiar urban and industrial contaminants such as metals, PAHs, PBDEs, PCBs, and DDT. The 14 papers in this section are organized into groups related to: treatment of stormwater contamination by bioretention, elucidation of contaminant exposure in the Salish Sea food web, CEC distribution and risk, contaminant toxicity to Salish Sea biota, and developments in our understanding of roadway runoff and the tire wear chemical 6PPD-quinone toxicity to fish.

Preventing contaminants in stormwater from entering the Salish Sea using bioretention has been shown to be effective

in protecting resources from contaminants inputs. Research summarized in this section identifies optimal bioretention media. Knappenberger et al. used Bayesian statistical modeling in a comprehensive analysis of permitted stormwater outfall discharge data to determine the relative efficacy of mixtures of sand, compost, water treatment residuals, and shredded bark in different proportions to reduce concentrations of chemical oxygen demand (COD), nutrients, and metals. This approach facilitated identification of the optimal bioretention mixture to alleviate the effects of stormwater on surface water quality. Mitchell et al. show that two novel bioretention amendments, biochar and fungi, were equally effective as the control treatment (a Washington State standard 60% sand and 40% compost bioretention mix) at removing PAHs from highway runoff (>97% reduction for all but two samples). They found that despite continuing PAH inputs over the two years of the study, PAH concentrations in bioretention media decreased over time indicating continuous bioremediation of PAHs.

Three studies in this section advance our understanding of the spatial distribution of contaminants within the Salish Sea and pathways of exposure throughout the food web. West et al. document the transfer of PCBs from the water column to diatoms, suggesting a mechanism for entry into Puget Sound's pelagic food web from phytoplankton to primary consumers. In this study, PCBs in particulate organic matter samples, comprised primarily of diatoms, collected throughout Elliott Bay showed a declining gradient with distance from the LDW Superfund site, with the highest total PCB concentrations detected in the East Waterway (45 ng/g ww). Leidtke et al. also advance our understanding of PCBs in the food web by showing that male sandlance from representative urban and undeveloped shoreline locations in Puget Sound had higher PCB concentrations than females. They also observed the ratio of lipid-normalized PCB concentrations in eggs/female tissue is close to one, confirming maternal transfer of PCBs to early life stages. Because early life-stage sand lance are prey for a wide range of species, their PCB contamination poses increased challenges for mitigation of PCBs in the food web. Further up the food web, Wainstein et al. report increasing concentrations of PCBs, PBDEs, DDTs and PAHs in river otter scat with increasing urbanization in the Green River watershed. The highest concentrations in river otter scat occurred in the LDW Superfund Site. Concentrations of PCBs and PAHs were among the highest published for wild river otters with almost 70% of the LDW Superfund Site samples exceeding levels of concern. This study indicated otter scat is a good tool for biomonitoring of food web contaminant bioaccumulation.

Three studies in this section increase our understanding of the distribution and potential effects of a wide range of CECs. James et al. compare non-target high resolution mass spectrometry (HRMS) analysis of wet and dry season water samples collected from throughout a 100 river mile length of the Lower Columbia River in 2021. Based on a comparison to spectral libraries, 886 potential chemical matches were identified, of which 119 were validated using additional criteria. Their results indicate that organisms in the Columbia River are exposed to a wide variety of CECs, some of which bioaccumulate and are suspected to pose a risk to biota. The Columbia River CEC results may help us understand the potential CEC exposures of organisms living in the Salish Sea. Faber et al. compare CEC data from over 950 water, wastewater, and tissue chemistry samples from 20 individual studies from Puget Sound relative to biological effects thresholds for potential toxicity. Of the 240 detected contaminants, 56 had concentrations greater than effects thresholds by more than a factor of 100. These contaminants were classified as priority chemicals for further monitoring and research. Ball et al. report that juvenile Chinook salmon exposed for 10 days to secondary-treated King County wastewater effluent showed signs of increased stress, and altered endocrine function, neurological function, and metabolism accompanied by decreased lipid content and visible liver anomalies. Concentrations of three estrogenic hormones measured in effluent were similar to concentrations known to affect endocrine function, and many classes of toxicants were measured in effluent and Chinook blood plasma at concentrations potentially capable of causing adverse physiological responses, suggesting that a mixture of toxicants may be acting together to cause the observed effects.

Further advances in our understanding of contaminant toxicity to Salish Sea biota are summarized in two studies. Harding et al. document xenoestrogen exposure and endocrine disruption in English sole at 2 of 12 Puget Sound locations sampled in May of 2017 and 2019. Plasma vitellogenin, a female reproductive hormone, in male English sole co-occurred with unusual reproductive timing in females at the Seattle Waterfront and Carr Inlet, indicating xenoestrogen exposure and endocrine disruption at these two locations. Their analysis suggests that steroid hormones from sewage or other chemicals not measured are a more likely cause of the observed effects than known estrogenic bisphenols. Strivens et al. provide a means of evaluating the potential toxicity of bioavailable metals by establishing toxicological effects thresholds for protection of mussel larvae from labile copper and urchin larvae from labile zinc with the use of in situ passive samplers called Diffusive Gradients in Thin films (DGTs). Comparison of DGT passive sampler copper and zinc data collected from reference and shipyard locations in Sinclair and Dyes inlets to respective DGT copper and zinc toxicological thresholds shows that all areas had labile copper levels below its threshold, whereas 13% of labile zinc samples exceeded its threshold, suggesting zinc may be affecting urchin survival near some industrial shipyard activities.

Finally, four papers describe the tremendous progress Salish Sea researchers have recently made on increasing our understanding of contaminants in stormwater roadway runoff, particularly the tire wear derived chemical 6PPD-quinone and its impacts on coho pre-spawn mortality and early life stage coho and other salmon species. Peter et al. studied a representative small urban Puget Sound watershed during the fall, when coho pre-spawn mortality occurs. They found that a rapid first flush of contaminants, including tire-wear derived contaminants, are mobilized from urban roadways early in each storm event (even during light drizzle) that result in substantial, rapid water quality degradation. Contaminant concentrations remained high after peak flows indicating the reservoir of contaminants is not depleted by the first flush. McIntyre et al. report that contaminants from tires are responsible for acute coho spawner mortalities. They show that pre-spawn coho exposed to tire wear particulate (TWP) leachate exhibited acute mortality at

contaminant concentrations expected in roadway runoff, with the same behaviors and blood parameters impacted. Chum salmon appeared insensitive to TWP leachate similar to what has been previously observed in Chum from streams impacted by roadway runoff. Chow et al. summarize their work showing that juvenile coho exposed to roadway runoff exhibit similar changes in behavior and blood chemistry as pre-spawn adults, and also die within a few hours. This study establishes juvenile coho as a model for urban runoff mortality syndrome, which has facilitated insights into the mechanism of action summarized in Blair et al. Among other symptoms, coho deaths from 6PPD-quinone exposure include a severe hematocrit rise (increased concentration of red blood cells by volume). Blair et al. provide evidence that a breakdown of the blood-brain barrier underlies extreme hematocrit rise, which may allow passive diffusion of toxic substances from the blood into the central nervous system leading to impaired neuronal function and death.

3. What is being done? | Management

The eight papers in this section describe a suite of programs and approaches that fall on a continuum of contaminant management approaches, from effective upstream pollution prevention, to priority pollutant removal and clean-up, to permitting programs.

An earmark of this collective work is the continued acceleration of geospatial tools that can target priority pollutants, through the application of spatial modeling tools. The papers were submitted by a range of transboundary practitioners, from Federal and State/Provincial Governments, including the Washington State Governor's Office, to multiple non-profit organizations and their private sector collaborators.

These programs have been developed in response to our industrial economy, which has resulted in releases of chemicals into Puget Sound. With it came great conveniences but also legacy chemicals that persist and cause long-term harm to both people and wildlife—PCBs are an example. That said, we now have promising opportunities to use technology to change how products are made (green chemistry), and are considering redevelopment that helps protect regional aquatic species and the communities that use them.

Pollution prevention

Preventing pollution before it reaches the environment and humans is the most effective and high-return investment to reduce the adverse effects of contaminant inputs to the Salish Sea.

Smith et al. made its first set of recommendations for taking preventive action on a subset of hazardous chemicals in 2022. The Safer Products for Washington law promotes transparency and reduces hazardous chemicals in consumer products. The criteria for "safer" aligns with existing chemical hazard assessment methods, such as Green Screen® for safer chemicals and the EPA's Safer Chemical Ingredients List Master Criteria. This program advances understanding of hazardous chemicals in consumer products, accelerates the use of safer alternatives, promotes green chemistry training for scientists, connects businesses with technical assistance, and connects contaminant science with the lived experiences of people through consumer products. Since 2019, the program identified safer, feasible, and available alternatives to PFAS, phthalates, organohalogen flame retardants, bisphenols, alkylphenol ethoxylates, and PCBs. Follow-up restrictive regulations and bans will accelerate prevention efforts overall.

Pollution removal and clean-up

The following papers address the use of GIS technology to identify priority stormwater retrofit action areas, the use of community engagement to increase the use of green stormwater infrastructure and using moss as a way to identify metal concentrations in watersheds.

Proper removal and cleanup of contaminant sources is critical to reducing releases and exposures. Stormwater is one of the major pathways by which chemicals released into the environment reach aquatic habitats of the Salish Sea. Thus, stormwater treatment is considered the next line of defense for reducing the adverse impacts of contaminants on both aquatic organisms and humans.

The Stormwater Heatmap described in Howe et al. identifies hotspots of pollution generation and runoff throughout the Puget Sound watershed. It is an interactive mapping tool, report generator, and data repository that quantitatively visualizes hotspots of pollution and runoff generation. The tool merges local monitoring data, statistical modeling, and cloud computing technologies and improves the ability to establish priorities for stormwater infrastructure investments. Howe et al. used a high resolution landcover dataset; conducted continuous hydrology simulations for 32 hydrologic response units (HRUs) that combine landcover, soils and slope; and used pollution statistics to predict pollution concentration across the Puget Sound watershed. The tool enables users to visualize and aggregate stormwater pollution loading data at several spatial resolutions that support local, watershed, and regional-scale planning.

Korwel et al. developed a Salmon Friendly green stormwater retrofit program for Water Resource Inventory Areas (WRIAs) 8 & 9, one of the most populous regions in the Salish Sea, including the Seattle metropolitan area. This targeted, voluntary non-point source pollution approach focuses on private landowners. The Mid Sound Fisheries Enhancement Group committed to installing 30 Best Management Practices in rain gardens and cisterns near Chinook salmon streams. A gap is measuring the types and quantities of pollution being imported to, and exported from, particular best management practices (BMPs). New and novel data collection methods will need to be identified to calculate treatment of heavy metals and other pollutants from Salmon Friendly projects.

Metals are of high concern in urban areas. A study by Messager et al. reveals that 27% of the Puget Sound watershed exceeds background levels for metal indicators (copper, zinc, and a synthetic index of pollution). Metals are particularly problematic due to their persistence, bioavailability, and toxicity. Pathways include atmospheric deposition, roadway runoff, and emissions and wear from vehicles of all types, including brakes, tire wear, and tailpipe emissions. Messager et al. demonstrated novel scalable methods to monitor and predict urban metal pollution at high resolution (<5 m) across large areas to guide pollution

reduction and stormwater management. By leveraging vehicle traffic characteristics with biological monitoring data from moss on tree trunks, identifying metal emission hotspots in densely urbanized areas is now possible, revealing that 50% of total pollution in the Puget Sound watershed is deposited over only 3.3% of the land area for copper and zinc.

Pollution permitting and management

The health of the Southern Resident Killer Whale population embodies a complex set of relationships between toxics, salmon habitat and health, green infrastructure, and regulatory controls.

Occasionally, there is misalignment between regulatory and policy structures across the British Columbia and the Washington State border. Barford et al. report on the weaknesses of regulatory controls for the cruise industry in Canada. In the Salish Sea, Puget Sound is protected by a no-discharge zone, which limits fecal coliforms to 14 Colony Forming Units (CFUs). Once ships enter Canada, they are able to discharge 250 CFU/100 mL within 12 nautical miles of shore.

As described in Barford et al., sewage and graywater from ships contain personal care products and pharmaceuticals, CECs that pose risks to Chinook salmon and SRKW. Over 90% of the volume of discharges from cruise ships is scrubber wash water that rinses the exhaust created by burning high sulfur fuel with sea water. This contributes to ocean acidification. One partial solution is aligning transboundary regulatory frameworks and enforcement and requiring low sulfur fuel use.

On a counter note, Hilborn et al. report the release of a new tool to close data gaps and create a comprehensive geospatial database of contaminant contributions from multiple sectors/activities and ambient monitoring loads. The Pollutants Affecting Whales and their Prey Inventory (PAWPIT), is part of Canada's Recovery Strategy and Action Plan under the Species at Risk Act for the NRKW and SRKW. PAWPIT enables identification and prioritization of key contaminants and their sources. The tool also identifies contaminant hot-spots and locations of exceedances of environmental quality guidelines, and enables a comparison of releases of different contaminants across locations, which helps identify data gaps and uncharacterized sources.

Current use pesticides were analyzed across three Canadian provinces by Blukacz-Richards et al. to assess urban and agricultural runoff affecting the Southern Resident Killer Whale, St. Lawrence Estuary Beluga whales and the Threatened Northern Resident Killer Whale. The analysis revealed glyphosphate had the highest yields across all sites. The Fraser Basin in B.C. analysis revealed the highest diazinon exceedances of the ECCC's Environmental Quality Guidelines. Atrazine and chlorpyrifos were also found in amounts exceeding the guidelines. The yields, exceedances of the Guidelines, and contaminant runoff estimates will be used to identify contaminant hot spots to inform potential recovery actions.

In a related action, Galuska et al. report on the contaminant-related priorities of the Southern Resident Orca Task Force (Task Force) Report in Washington State, which identifies 49 recommendations within 6 threat areas. Reducing contamination sources and pathways is a core tenet of the overall strategy. The Washington State Recreation and Conservation Office efforts, which include a ban on PCBs in state-purchased products, seeks to identify, prioritize, and take action on protecting SRKWs and their prey, improve enforcement efforts, prevent regulatory rollbacks, and explore setting minimum standards for local stormwater funding.

KEY MESSAGES

- **Some contaminants impact the health and safe consumption of fish and shellfish, particularly benthic species in urbanized areas, and pelagic fish from throughout Puget Sound. Persistent legacy contaminants, such as PCBs, are still widespread and cycle through the pelagic food web. PBDEs have been declining but are still affecting juvenile salmon in the Snohomish and Puyallup basins.**
- **A number of regional monitoring groups are providing important exploratory and baseline data on legacy and emerging contaminants potentially impacting Orca and their main food source, Chinook salmon.**
- **Although Puget Sound nearshore sediments are relatively clean, mussel tissue monitoring shows higher contamination in urban centers. Temporal trends of legacy contaminants in mussel tissues show an overall decrease in concentrations from 2017/18 to 2019/20.**
- **Contaminants in roadway and stormwater runoff negatively affect the health of salmon and forage fish. More is being learned about the toxic effects and treatment of tire wear particles and 6PPD-quinone. For example, bioretention using soil-based media is a highly effective means of protecting biota from contaminant exposure and impacts.**
- **Laboratory studies indicate that wastewater treatment plant effluent could cause adverse effects in fish where it enters the Salish Sea.**
- **CECs, including pharmaceuticals and personal care products, have been detected in Salish Sea waters, and some like PFAS are being found widely in freshwater. Some of the CECs occur at levels high enough to harm fish and wildlife.**
- **Pollution prevention efforts and cleanup, particularly those that include local community engagement and clear connections with human health, are effective strategies that can reduce contaminant exposures. Upstream strategies are also more cost effective and signal to manufacturers and processors to find alternatives.**

SECTION 1: STATUS AND TRENDS

Thai Do and Nina Mass collect intertidal sediment in the Lower Duwamish Waterway. Photo: Suzanne Replinger

Contaminant and Stable Isotope Profiles in Priority Chinook Salmon (*Oncorhynchus tshawytscha*) Stocks Consumed by Northern and Southern Resident Orca (*Orcinus orca*) Populations

Fisheries and Oceans Canada / Pêches et Océans Canada

SFU SIMON FRASER UNIVERSITY ENGAGING THE WORLD

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- **Contaminant profiles and feeding ecology varied significantly among priority Chinook salmon populations consumed by resident killer whales.**
- **Stable isotope values ($\delta^{15}N$, $\delta^{13}C$) varied among Chinook populations and influenced the contaminant concentrations and patterns that differed among stocks.**

The critically endangered Southern Resident killer whale (SRKW) (*Orcinus orca*) population faces significant threats including reduced abundance and quality of their primary prey (Chinook salmon, *Oncorhynchus tshawytscha*) and high levels of endocrine disrupting contaminants (Heise, 2008; CWR, 2022). Even though SRKW and the sympatric Northern Resident killer whales (NRKW) have overlapping habitat ranges and similar diets, NRKW have lower contaminant burdens and have experienced increased population growth (Ross et al., 2000; Rayne et al., 2004; Towers, 2015). Polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), organochlorine pesticides, hexabromocyclododecane, alkylphenols, and chlorinated paraffins have been identified as contaminants of concern for SRKW and Chinook salmon (ECCC, 2020). Studies have reported adverse health effects from contaminants in transient killer whales and NRKWs, and contaminant exposure modeling has predicted protracted health risks for both resident killer whale populations (Brown et al., 2014; Pearce, 2018; Noël & Brown, 2020).

Despite Chinook salmon from the Fraser River watershed in British Columbia, Canada comprising up to 90% of the SRKW diet during the summer months (Hanson et al., 2010; Hanson et al., 2021), little information exists on contaminants in these priority stocks, or in Chinook stocks from other Canadian rivers. The objectives of this study were to characterize a broad range of contaminant classes in ten priority Chinook salmon populations consumed by SRKW and NRKW, investigate factors affecting Chinook contaminant profiles (i.e., stable isotopes, marine migration), and to evaluate contaminant patterns in Chinook to characterize local versus global sources.

Adult Chinook salmon heads were collected and head muscle was subsampled during 2018 and 2019 in the Fraser River estuary, southwest coast and east coast of Vancouver Island, and Haida Gwaii in partnership with the Albion Test Fishery, Pacheedaht First Nation and the University of British Columbia. Based on SRKW and NRKW diet studies (Ford et al., 2010; Hanson et al., 2010 & 2021), individuals (n = 83) from ten priority Chinook salmon populations were selected: Cowichan River (East Coast Vancouver Island (ECVI)), West Coast Vancouver Island (WCVI), Puget Sound, Columbia River, Skeena River, North Mainland British Columbia (NM BC), Upper Fraser, Middle Fraser, South Thompson River, and Harrison River (Lower Fraser). These individuals were analyzed for 317 analytes across 7 contaminant classes and for stable isotopes ($\delta^{13}C$ and $\delta^{15}N$).

The highest average contaminant concentrations in all samples (n = 83) ranked as: ∑PCB > ∑ Dichlorodiphenyltrichloroethane (DDT) > ∑PBDE > ∑Chlordane > ∑Alkylphenols > Hexachlorobenzene. Stable isotope values and contaminant concentrations varied significantly across Chinook populations (Figure 1A & 1B). Chinook from the Cowichan River and Harrison populations had enriched $\delta^{13}C$ values reflecting their coastal shelf habitat use (COSEWIC, 2018), while Upper and Mid Fraser Chinook showed more depleted $\delta^{13}C$ compared to other stocks reflecting their use of offshore open ocean food webs (COSEWIC, 2018) (Figure 1A). Average Cowichan Chinook $\delta^{15}N$ values were greater than seven other Chinook populations. Resident Chinook stocks (Cowichan and Harrison) had higher average contaminant concentrations than other stocks (Figure 1B). Stable isotope values varied among the ten Chinook populations and influenced contaminant concentrations and patterns.

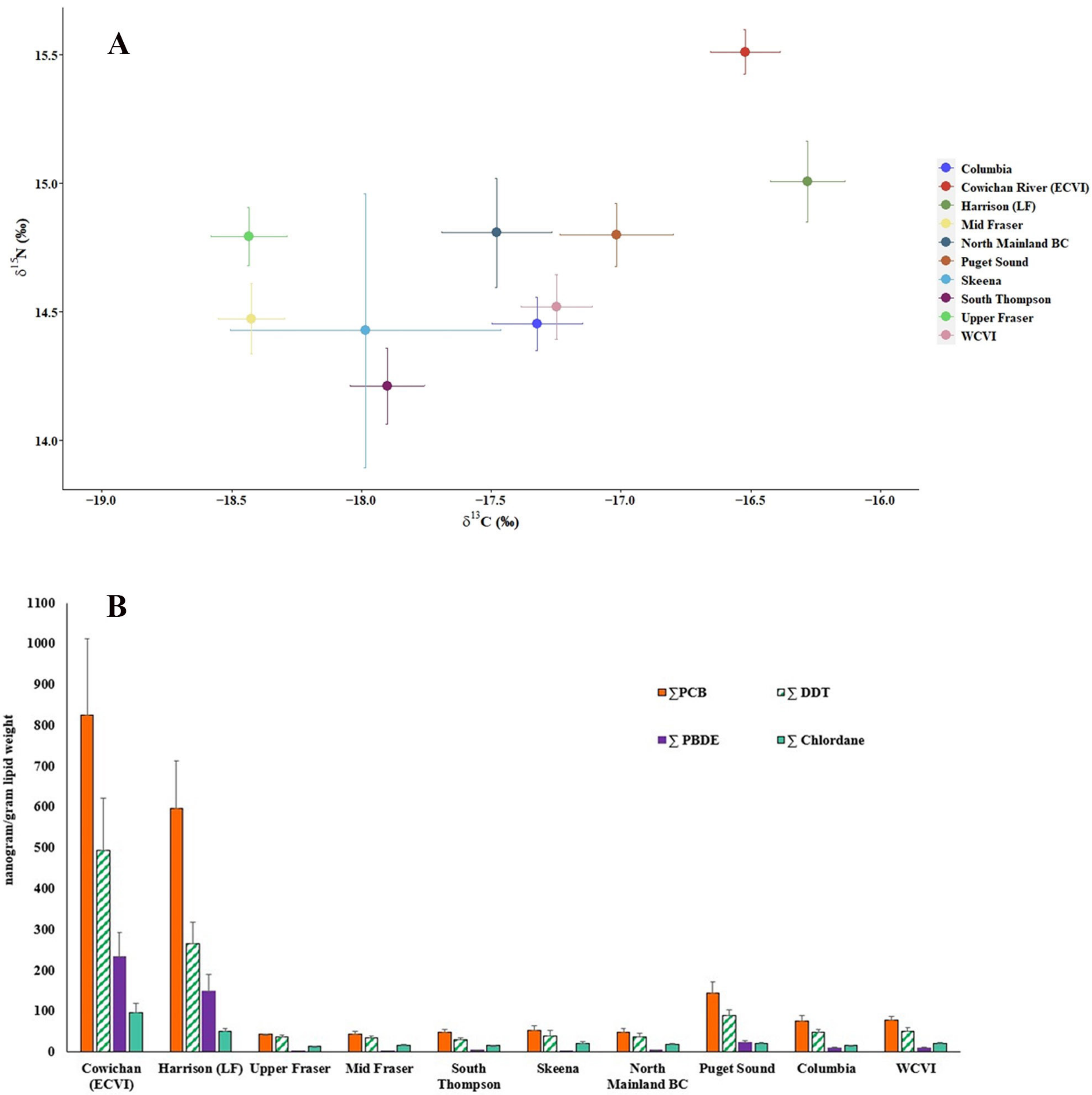

Figure 1. (A) $\delta^{13}C$ and $\delta^{15}N$ values (mean ± SE) varied among the ten priority Chinook stocks consumed by NRKW and SRKW (ANOVA; $\delta^{13}C$ $F_{9,102} = 18.1$, $p < 0.001$; $\delta^{15}N$ $F_{9,102} = 6.17$, $p < 0.001$). (B) Top contaminants (mean + standard error nanograms/gram lipid weight) detected with significant differences among Chinook stocks: $\sum_{159}$PCB ($X^2_{(9)} = 49.6$, $p < 0.001$), $\sum_{40}$PBDE ($X^2_{(9)} = 52.3$, $p < 0.001$), $\sum_{6}$DDT ($X^2_{(9)} = 48.6$, $p < 0.001$), and $\sum_{5}$Chlordane ($X^2_{(9)} = 45.1$, $p < 0.001$).

Risk Assessment of Sediment Polychlorinated Biphenyls and Polybrominated Diphenyl Ethers in Resident Killer Whale Habitat along the Coast of British Columbia, Canada

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- **Current levels of PCBs and PBDEs in sediment from coastal British Columbia pose a threat to the health of northern and southern resident killer whale populations.**

The northern and southern resident killer whale (Orcinus orca) populations are listed as threatened and endangered in Canada, respectively, with persistent, bioaccumulative contaminants, such as polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), being a threat to their recovery.

PCBs and PBDEs have been identified as primary contaminants of concern for these whales (Baird, 2001; Krahn et al., 2007; Ross et al., 2000; Ross, 2006). Toxicological effects of PCBs and PBDEs include endocrine disruption, immunotoxicity, and reproductive and developmental impairments (Bergman et al., 1992; Brouwer et al., 1989; De Swart et al., 1994; Helle et al., 1976; Mos et al., 2006; Reijnders, 1986; Ross, 2006). Contaminant levels in southern resident killer whales exceeded marine mammal immune and endocrine effects threshold concentrations for PCBs as well as the thyroid hormone disruption threshold for PBDEs (Noël & Brown, 2021).

Ocean Wise's Pollution Tracker is a coast-wide, integrated marine pollution monitoring program in British Columbia (BC), Canada, documenting the levels of hundreds of contaminants in mussels and nearshore ocean sediments in partnership with First Nations,

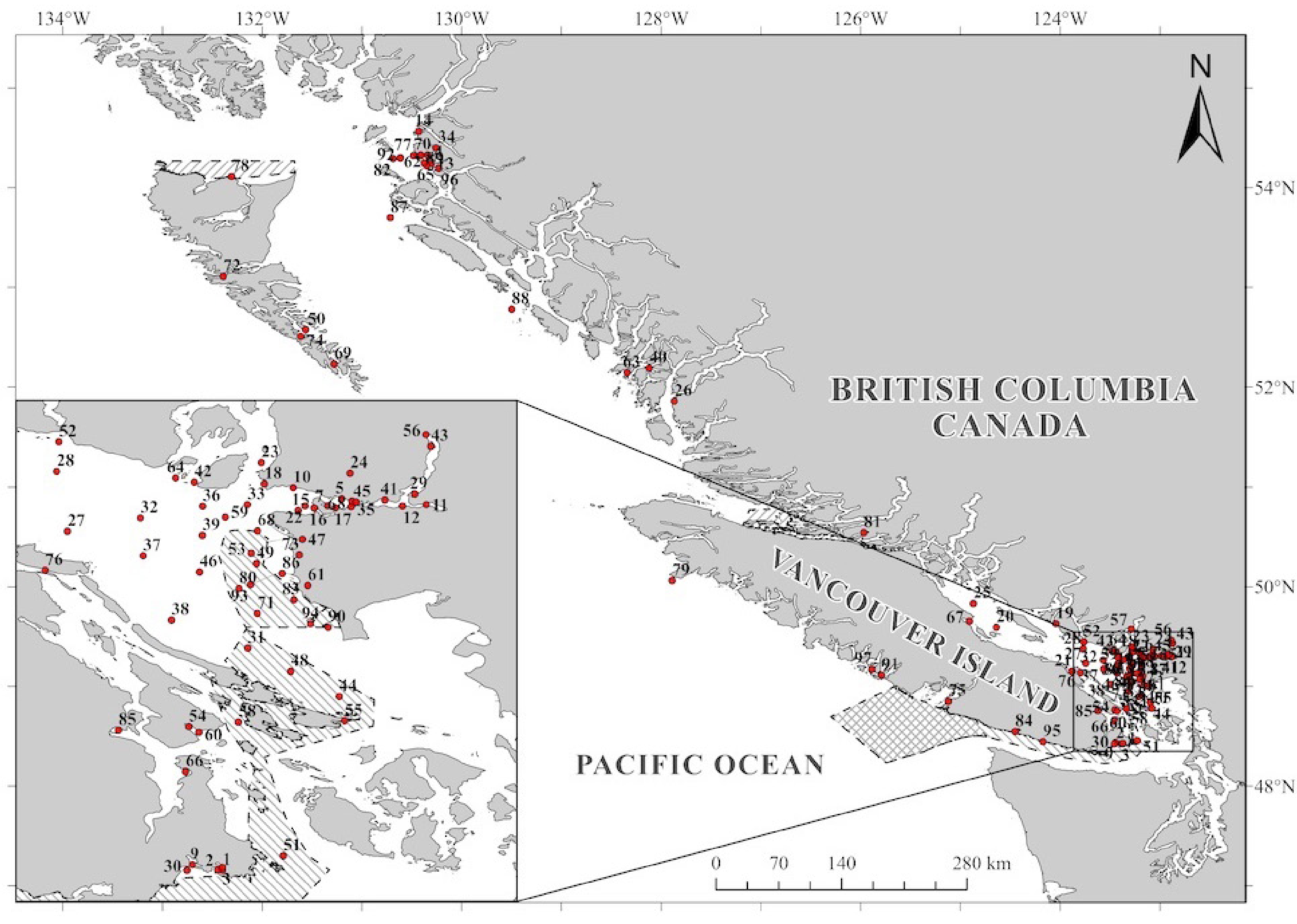

Figure 1. Subtidal surface sediments collected from 97 sites along the coast of British Columbia, Canada.

community groups, port authorities, industry, and government agencies (Full list of partners). In the present study, we compiled Pollution Tracker sediment PCB and PBDE concentration data collected between 2018 and 2020 from 97 sites along the entire British Columbia coast to assess resident killer whale habitat quality (Figure 1). Geostatistical analysis was used to identify distributions of PCBs and PBDEs in and around resident killer whale critical habitat and to compare levels in sediments to British Columbia's Ministry of Environment and Climate Change Strategy Working Sediment Quality Guidelines (PCBs 0.0037 µg/kg dw and PBDEs 1 µg/kg dw), considered protective of killer whales (Alava et al., 2012, 2016) (Figure 2).

Total PCB concentrations exceeded the Working Sediment Quality Guidelines at all sites, and total PBDEs exceeded the guidelines at 34% of sites, with 100% PCB exceedances (18/18 sites) and 10% PBDE exceedances (2/18 sites) located within killer whale critical habitat. Total PCB concentrations were lower within (0.56 ± 0.14 µg/kg dw of 18 sites) compared to outside (16.26 ± 5.17 µg/kg dw of 79 sites) critical habitat (Mann–Whitney U test, $p = 0.001$), and average concentrations within and outside critical habitat exceeded the guideline (~150- and 4,000-fold higher, respectively). Total PBDE concentrations were similar within (0.52 ± 0.19 µg/kg dw) and outside (4.13 ± 2.15 µg/kg dw) critical habitat (Mann–Whitney U test, $p = 0.122$), and only the average concentration outside critical habitat exceeded the guideline.

For further insight into killer whale habitat quality, Kernel interpolation with barrier was used to estimate levels in sediment in and around northern and southern resident killer whale critical habitat (Figure 2 A,B) and to generate probability maps by comparing those levels to the guidelines (Figure 2 C,D). The probability Kernel interpolation with barrier maps showed that all of the interpolated areas for PCBs and parts of interpolated areas for PBDEs had > 0.5 probability of exceeding guidelines, including all and most of northern and southern resident killer whale critical habitat, respectively. Our study suggests that PCBs and PBDEs remain widely distributed in the marine environment at concentrations that are harmful to the health of killer whales, and therefore, have the potential to constrain their recovery.

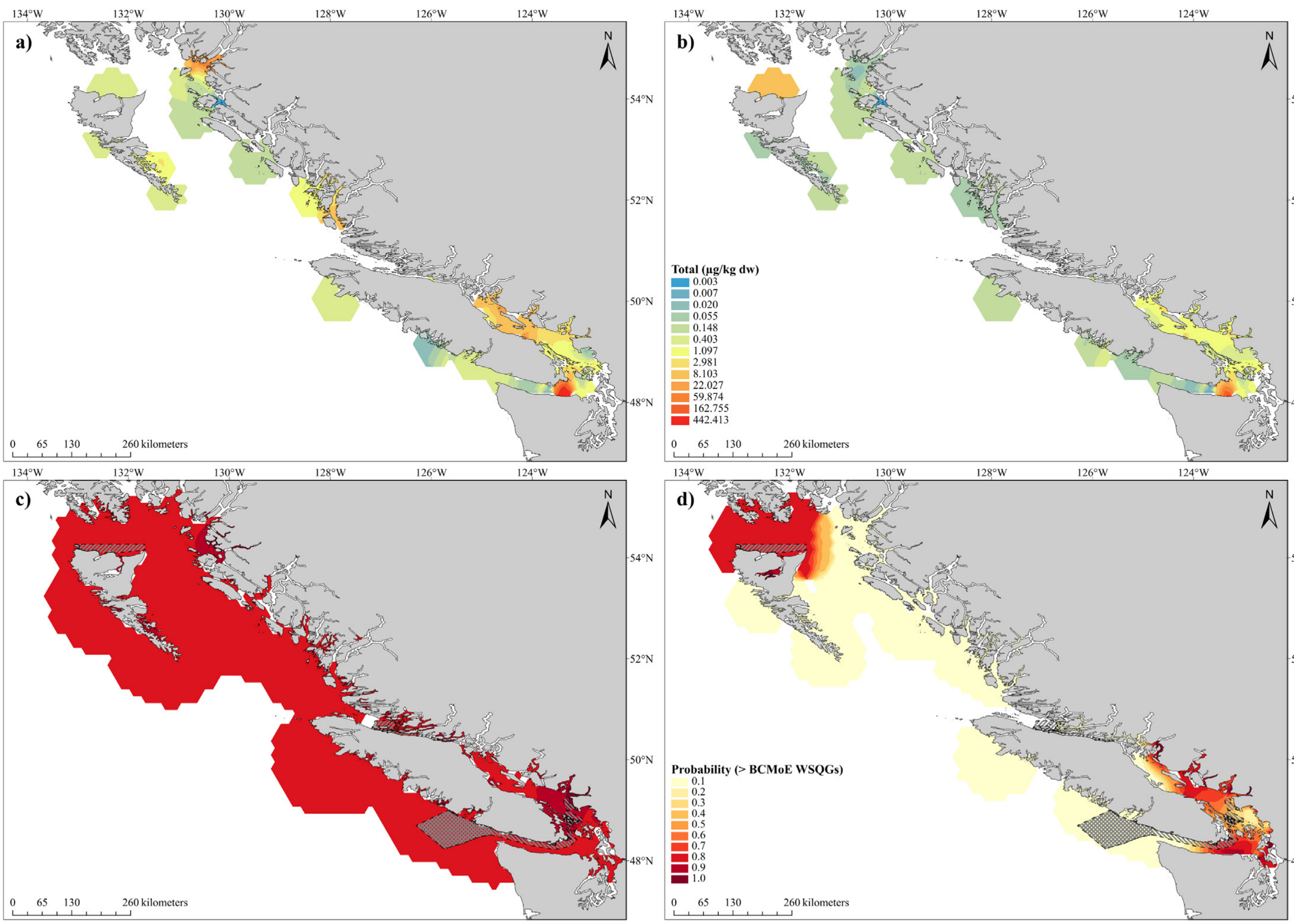

Figure 2. Contour maps of total concentration for (A) PCBs and (B) PBDEs. Contour maps for probability of exceeding established sediment-derived thresholds for killer whale risk are depicted for (C) PCBs and (D) PBDEs.

Prioritizing Contaminants of Concern in the Fraser River Watershed: a Risk-based Evaluation for Outmigrating Juveniles and Returning Adult Chinook Salmon

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- **Proximity to urban developments and wastewater treatment plants explained higher concentrations of some analytes.**
- **Higher pesticide concentrations during peak Chinook salmon occupancy raise concerns about impacts.**

The Fraser River watershed is home to 54 unique spawning populations of salmon, including 19 Chinook (Oncorhynchus tshawytscha) populations (Birtwell et al. 1988). Fraser Chinook provide 80-90% of the food source consumed by Southern Resident killer whales (SRKW, Orcinus orca) in the spring and summer (Hanson et al., 2010; Hanson et al., 2021). Over 94% (15/16) of Fraser Chinook populations are at risk (COSEWIC 2018). Extensive forestry, agricultural, industrial and urban activities take place in the Fraser Valley that expose early life stages of emigrating salmon and returning adult salmon to a mix of legacy and emerging contaminants. Many of these contaminants can elicit adverse health effects in vertebrates, including endocrine disruption and reproductive effects. However, there is limited information on the nature of contaminants discharged into British Columbia's salmon habitat and their associated effects, hampering solution-oriented opportunities for natural resource managers and stakeholders. The objective of our work is to conduct a risk-based evaluation using a combination of water quality guidelines, toxicity quotients and exposure activity ratios to prioritize contaminants for long-term monitoring and to identify chemicals suspected of posing a potential risk to outmigrating juveniles and returning adult Chinook salmon.

Surface water samples were collected monthly (2018-2020) from seven urban and semi-urban sites in the Fraser River watershed, and one site in the Serpentine River, a lower discharge river that flows directly into SRKW critical habitat. Samples were analyzed for 714 contaminants in order to prioritize contaminants of concern to outmigrating juveniles and retuning adult salmon, especially Chinook salmon. Measured chemical concentrations were compared to water quality guidelines for the protection of aquatic life and chemical-specific biological activities determined in high-throughput (ToxCast) in vitro assays. Preliminary results revealed that 26% (182/714) of the analytes were detected at all sites, and were dominated by Polychlorinated biphenyls (PCBs), metals, and polybrominated diphenyl ethers (PBDEs). Only 12% of the analytes measured had water quality guidelines and 18% and 30% of the water samples exceeded water quality guidelines for organics and metals, respectively. Pharmaceuticals and personal care products decreased with distance from wastewater treatment plants and urban areas. Pesticide concentrations were highest during maximum occupancy by Chinook of the Serpentine river, which raises concerns about the potential impacts. This study is the first step toward a comprehensive risk-based evaluation for contaminants of concern to salmon in the Fraser River. Results will support the Government of Canada's Whales Initiative in its quest to identify contaminants of greatest concern to Chinook salmon and to guide SRKW recovery efforts.

Water sampling in the lower Fraser River estuary. Photo: Tanya Brown

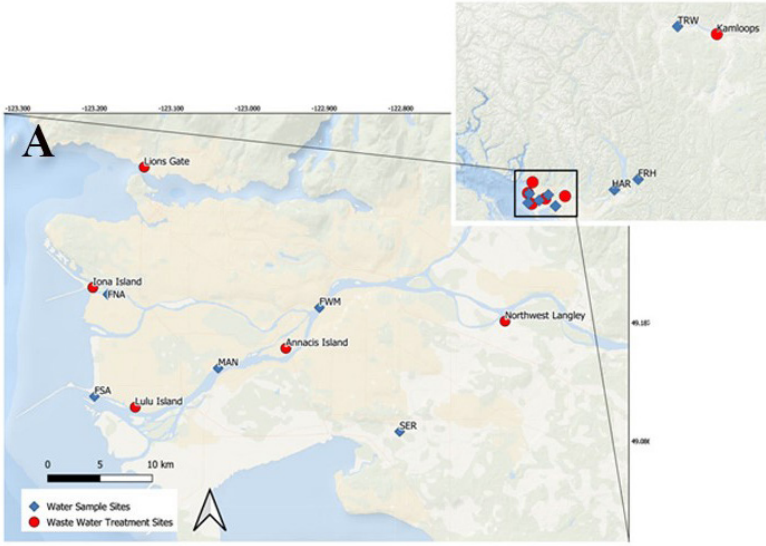

Figure 1 (A) Water sampling locations in the Fraser River.

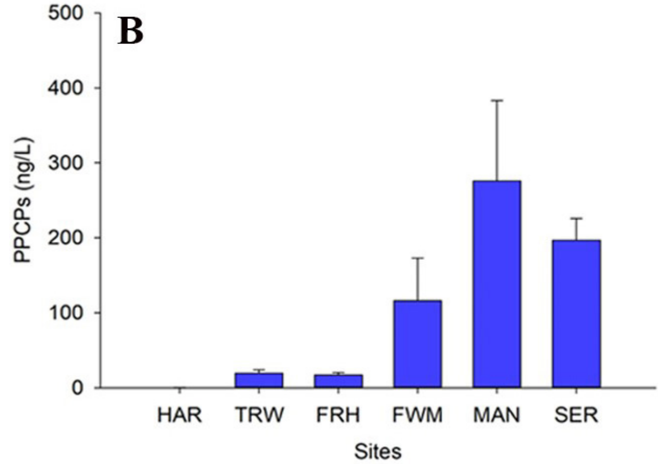

Figure 1 (B) Pharmaceuticals and personal care products (PPCP) decreased with distance from wastewater treatment plants and urban areas.

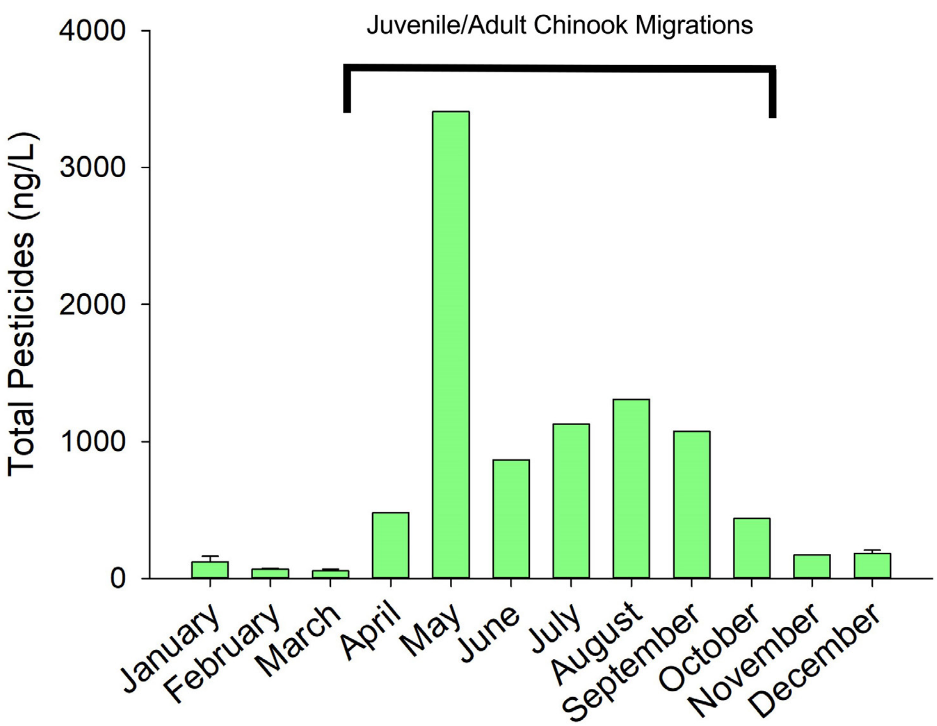

Figure 2. Pesticide concentrations appear to be highest during maximum occupancy of Chinook in the Serpentine River.

Juvenile Chinook Salmon from Puget Sound Accumulate Harmful Levels of Toxic Contaminants During their Seaward Migration

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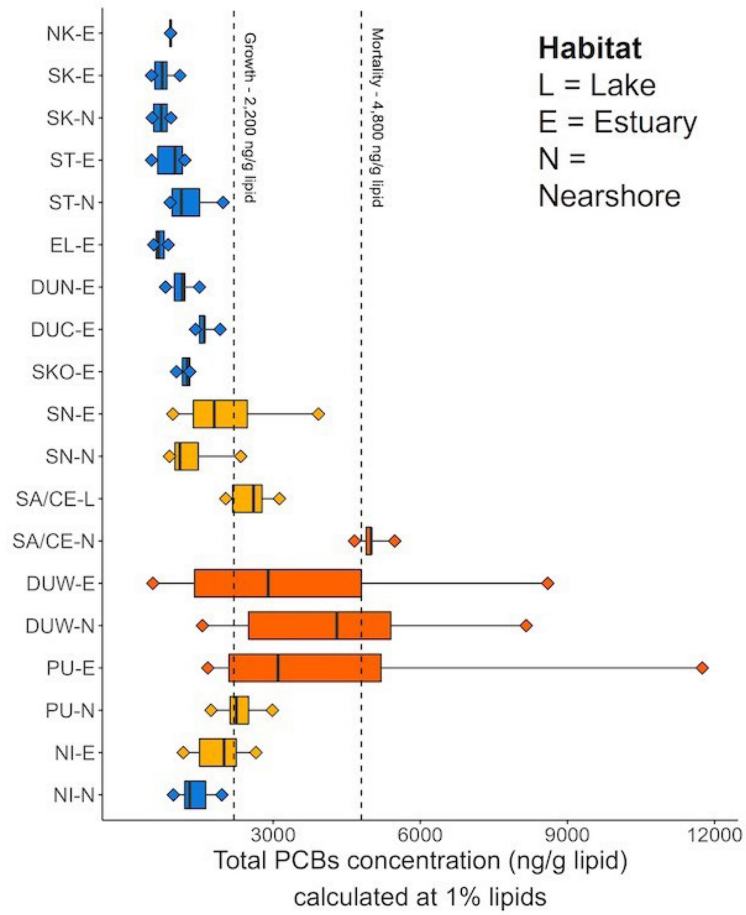
- **Seaward migrating juvenile Chinook salmon from five Puget Sound river systems are being exposed to harmful levels of contaminants.**
- **Levels of PCBs and PBDEs in juvenile Chinook salmon from five river systems are high enough to potentially impact their health and increase mortality.**

Toxic contaminant exposure is a contributing factor in the decline of Puget Sound Chinook salmon (*Oncorhynchus tshawytscha*), which are listed as threatened under the Endangered Species Act (ESA). During their seaward migration, juvenile Chinook salmon rear and feed for extended periods in estuary and nearshore habitats, exposing them to contaminants that accumulate disproportionately there (O'Neill et al. 2015, 2020; Sloan et al. 2010; Meador et al. 2010, 2016; Johnson et al. 2013). Exposure to contaminants can lead to reduced growth, a weakened immune response, reproductive impairment, and may ultimately reduce their survival (Berninger and Tillitt 2019; Arkoosh et al. 2010, 2018; Meador et al. 2002, 2006. 2014). The intent of this study is to document the extent that Puget Sound Chinook salmon are adversely impacted by contaminant exposure during their seaward migration, based on published critical body residues (CBRs) (Meador et al. 2011).

Juvenile Chinook salmon were collected from the estuary and adjacent nearshore habitat of 12 major river systems in 2013, 2016 and 2018. Collectively, these sampling locations provide estuarine and nearshore habitat for all 22 ESA-listed Puget Sound Chinook salmon populations. For this study, 1,627 individual salmon were collected and composited into 363 whole-body samples before being analyzed for persistent organic pollutants such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and other organochlorine pesticides (Sloan et al. 2014).

This study determined juvenile Chinook salmon are being exposed to contaminant levels high enough to potentially cause health effects, with fish migrating through five of the 12 studied river estuaries predicted to be impacted. Concentrations of PCBs and PBDEs were highest and sometimes above CBRs in juvenile Chinook salmon migrating through the estuaries of the Snohomish (PCBs and PBDEs), Sammamish/Cedar (PCBs), Green/Duwamish (PCBs and PBDEs), Puyallup/White (PCBs and PBDEs) and Nisqually (PCBs) rivers. Juvenile Chinook salmon in the adjacent nearshore habitat of the Snohomish (i.e., Possession Sound), Sammamish/Cedar (i.e., Shilshole/Salmon Bay area), Green/Duwamish (i.e., Elliott Bay), and Puyallup/White (i.e., Commencement Bay) Rivers also had PCB and PBDE concentrations above CBRs. Overall, juvenile Chinook salmon sampled from these five river systems exceeded PCB CBRs for reduced growth (57%) and mortality (19%) and PBDE CBRs for increased disease susceptibility (17%) and altered thyroid function (1.6%; Berninger and Tillitt 2019; Arkoosh et al. 2010, 2018). Furthermore, health impacts to salmon are likely greater than indicated by these described individual CBRs because of combined effects from exposure to other contaminants including chemicals of emerging concern and per- and polyfluoroalkyl substances.

Results from this work are being used to connect science to action, guide remediation efforts, and track the effectiveness of recovery actions to improve the health of Puget Sound Chinook salmon. At a minimum, these results are "red flags" for the health of the juvenile Chinook salmon migrating through these river systems, indicating further actions, such as those outlined in the Toxics in Fish Implementation Strategies, and more are needed to protect them and the broader ecosystem they support. Reductions in juvenile Chinook salmon survival caused by contaminant exposure may lower abundance of returning adult Chinook salmon to Puget Sound, limiting recreational, commercial and subsistence fisheries in addition to reducing the food supply of ESA-endangered Southern Resident killer whales.



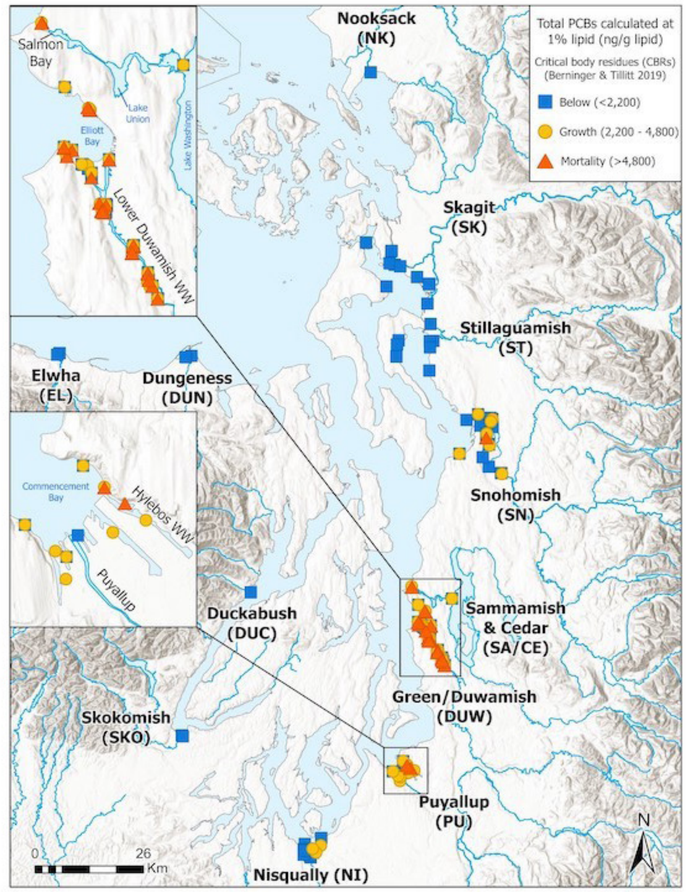


Figure 1 (A) Concentrations of Total PCBs (ng/g lipid calculated at 1% lipids) measured in juvenile Chinook salmon collected from the estuaries (E), a lake (L), and nearshore (N) marine habitats of 13 Puget Sound River systems (see map for river names) in 2013, 2016, and 2018. In the box plot (left) the median concentration for each sampling location is indicated by a solid horizontal line within the box plot. Box ends are 25th and 75th percentiles and diamonds are 5th (lower) and 95th (upper) percentiles. For each sampling location, the box color is determined by whether the upper diamond is above the critical body residue (CBR) for growth (yellow) or mortality (orange). All sampling locations with Total PCBs below the growth CBR are colored blue.

Figure 1 (B) Additionally, a map (right) shows the collection location of each juvenile Chinook salmon sample, and the points are colored based on whether the Total PCB concentrations are above the CBRs for growth (yellow circles), mortality (orange triangles) as well as the samples below the CBRs (blue squares). Two map insets show the areas where the juvenile Chinook salmon are most impacted by PCBs, the Green/Duwamish (top) and Puyallup/White (bottom) Rivers' estuaries and nearshore habitats.

Assessing Sources of Polybrominated Diphenyl Ether (PBDE) Flame Retardants Impacting Juvenile Chinook Salmon in the Snohomish River Watershed

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- **A multi-year source assessment of PBDEs in the Snohomish watershed identified elevated concentrations in the Snohomish estuary and Skykomish River near WWTP outfalls.**
- **PBDEs were found throughout the Snohomish River system in water, sediments, biofilms, and invertebrates.**

A survey by Washington Department of Fish and Wildlife (WDFW) conducted across five major river systems and four marine basins in the Puget Sound region found approximately 30% of juvenile Chinook salmon sampled contained levels of toxic contaminants high enough to produce sublethal effects (O'Neill et al., 2015). A follow up survey within the Snohomish River found juvenile Chinook of natural origin contained levels of polybrominated diphenyl ether (PBDEs) flame retardants at concentrations above fish health thresholds (O'Neill et al., 2020). associated with nearby wastewater treatment outfalls within the rivers. Although multiple wastewater treatment outfalls discharge to the Snohomish River, elevated PBDE concentrations were restricted to the lower mainstem and were not present in sloughs and the upper mainstem.

PBDEs were found throughout the river environment and accumulated at higher concentrations in sediments, biofilms, and biota relative to water. Figure 1 describes concentrations

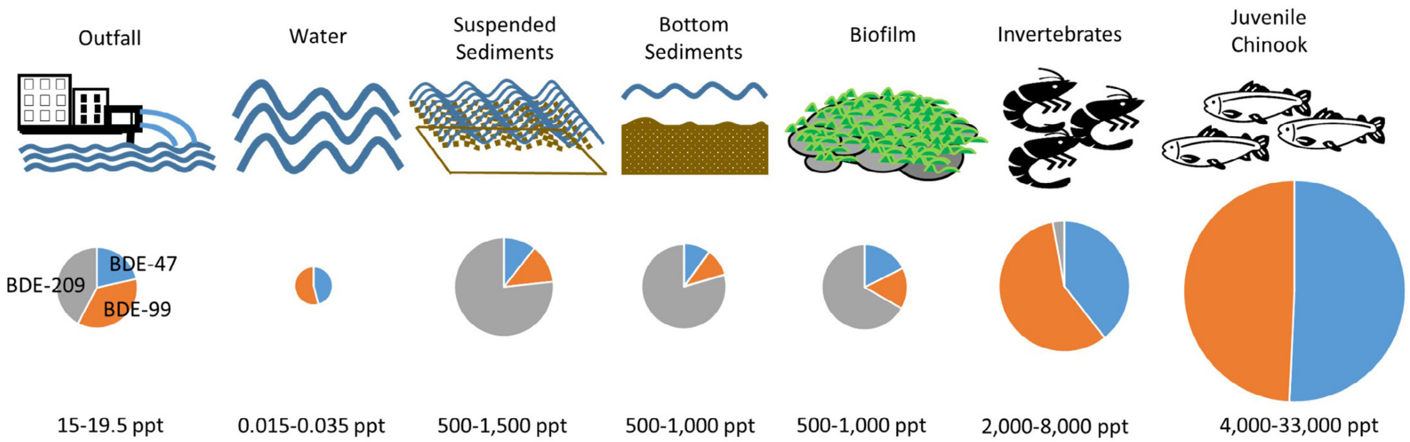

Figure 1. Concentrations of PBDE congeners BDE-209 (Gray), BDE-99 (Orange), and BDE-47 (Blue) in wastewater treatment plant effluent, ambient water, suspended sediments, bottom sediments, biofilms, invertebrates, and juvenile chinook from the lower Snohomish mainstem. ppt= part per trillion. Preliminary wastewater treatment plant outfall concentrations provided by City of Everett Public Works. Juvenile Chinook PBDE tissue concentrations provided by WDFW.

Juvenile Chinook salmon accumulate toxicants from rivers and estuaries in urban and developing environments that receive stormwater and wastewater discharges. Tissue concentrations of toxics above the fish health threshold may adversely affect salmon health, reducing their survival and limiting the abundance of returning adult Chinook to Puget Sound.[1] The decline of Chinook populations within the Puget Sound reduces the food supply available to Southern Resident killer whales and impacts tribal, commercial, and recreational harvests.

Beginning in 2019, the Washington Department of Ecology, in partnership with WDFW, started a source assessment of PBDEs in the Snohomish River watershed. The goals of this work were to 1) identify sources of PBDEs in the river system and 2) determine the route of PBDE exposure from source to juvenile Chinook. To identify sources of PBDEs and areas of elevated water concentrations, passive water samplers (semi-permeable membrane devices) were deployed throughout the Snohomish, Skykomish, and Snoqualmie rivers during high and low flow events from 2019 to 2022. Additionally, sediment (suspended and benthic), biofilms, and invertebrate samples were collected to determine partitioning and uptake of PBDEs throughout the environment and in juvenile Chinook prey.

We identified increased PBDE concentrations in the lower Snohomish River mainstem and at sites near the cities of Monroe and Sultan along the Skykomish River, all of which were

1 *Fish health threshold for PBDEs (sum of PBDE-47 and PBDE-99) in juvenile Chinook is ≥9.8 ng/g ww and ≤40 ng/g ww. (Chen et al. 2018, supplemental material).*

Retrieval of semi-permeable membrane device (SPMD) from Union Slough in the Snohomish estuary. SPMDs were deployed for approximately 30 days at sites across the Snohomish, Skykomish, Snoqualmie Rivers during low in high flow events from 2019 to 2022. Photo: Will Hobbs

of PBDEs in wastewater treatment effluent, ambient water, sediments, biofilms, and invertebrates in the lower Snohomish mainstem. PBDE compositions were different among the media sampled, with varying ratios of PBDE congeners, reflecting the chemical partitioning of PBDEs and possible metabolism of the compounds by organisms. These data suggest juvenile Chinook in part accumulate PBDEs through their prey, which feed on and live in biofilms and sediments rich in PBDEs.

Our findings provide information to Snohomish watershed stakeholders about the sources of PBDEs and inform recommendations to mediate the impacts of PBDEs on out-migrating juvenile Chinook salmon.

A Preliminary Ranking of Contaminants of Concern in Juvenile Harrison Chinook and their Habitat in the Fraser River, British Columbia

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- **A small number of environmental quality guidelines exist for detected analytes therefore multiple methods will be used to characterize risk.**
- **Contaminant profiles in tissue and water differed, confirming measurements in multiple matrices are important in assessing health risk to Chinook.**

The Fraser River is the largest river in British Columbia and was once considered the world's most productive salmon river (Gray and Touminen, 1999). This river is home to 19 Chinook populations (*Oncorhynchus tshawyscha*), of which 94% of the assessed populations are classified as being at risk (COSEWIC, 2018, Environment and Climate Change Canada, 2021). The Harrison River Chinook stock, which numerically dominate the lower Fraser River stocks, spend 30-50 days in the Fraser River estuary, where they feed and grow prior to entering the marine environment (Chalifour et al., 2020). The lower Fraser River and estuary is heavily impacted by a number of anthropogenic activities including, but not limited to, forestry, mining, pulp and paper, wood preservation, chemical manufacturing, urban and agricultural runoff and wastewater (sewage) treatment (Gray and Touminen, 1999). The objective of our work is to conduct a comprehensive assessment of anthropogenic contaminants in Chinook salmon and their habitat in the lower Fraser River and evaluate the risks posed to the health of this salmonid.

Samples of juvenile Chinook, water and sediment were collected and analyzed for over 600 contaminants of concern, including various pesticides, polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), flame retardants, personal and pharmaceutical care products, and metals. Whole juvenile Chinook salmon composites were sampled during their outmigration (April-June 2020, March-June 2021), water was collected monthly (May-June 2020, March-June 2021) and sediment was sampled annually. Sampling sites consisted of three impacted sites in the lower Fraser River and one reference site up-river in a semi-urbanized area with lower anthropogenic impact.

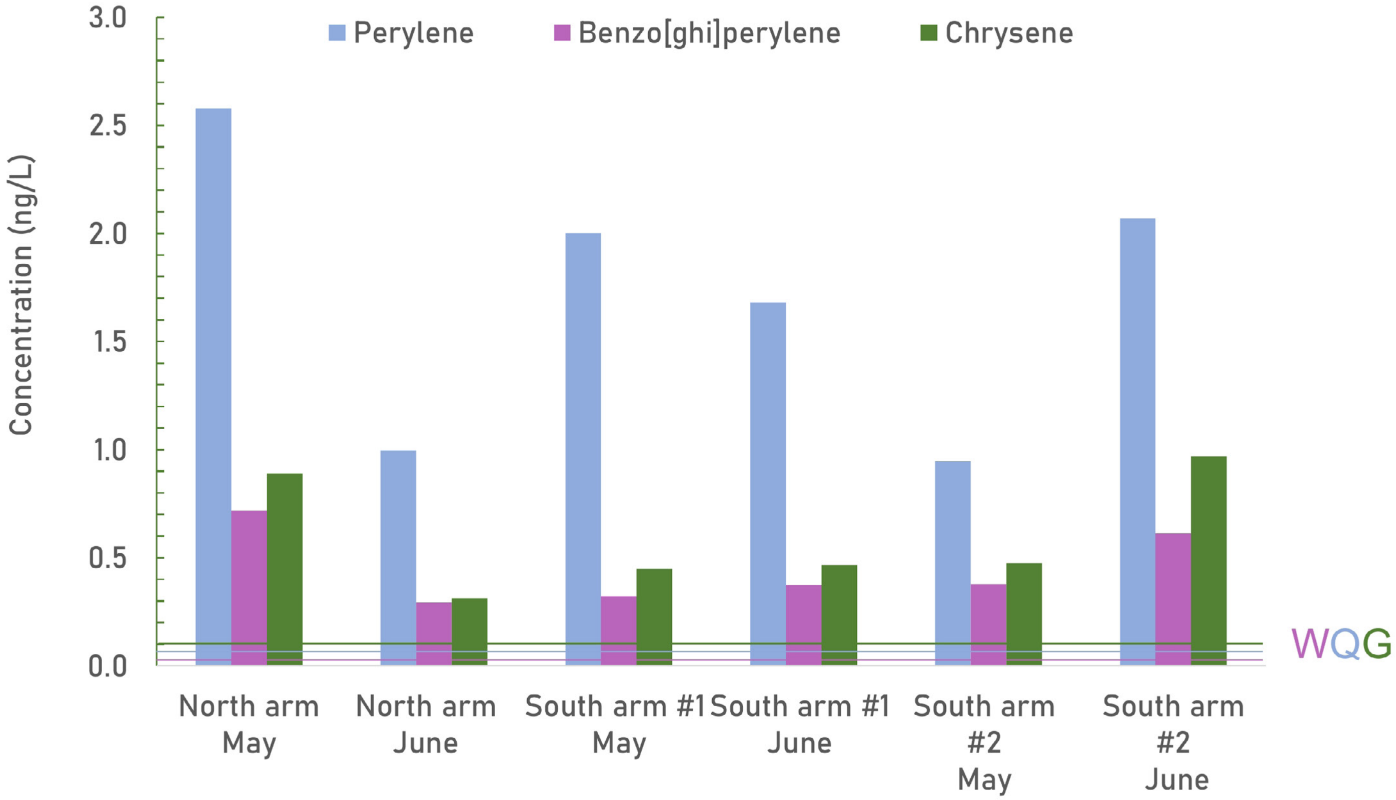

Figure 1. Measured water concentration of perylene (blue), benzo[ghi]perylene (purple) and chrysene (green) at three sites in the lower Fraser River.

Beach seining in the lower Fraser River estuary to collect juvenile Chinooks samples. Photo: Hasnah Nathani

Preliminary findings of our work focused on organic contaminants in Chinook tissue and water collected in 2020 at the three impacted sites (North arm, South arm #1 and South arm #2). Of the contaminants measured, between 46 and 50% of analytes (total of 612 assessed) were detected in Chinook tissue. The analyte with the highest concentration across all sites was 4-Nonylphenol. Also, in the list of the five highest concentrations of analytes, the antibiotic Virginiamycin (used in agriculture) was detected in both south arm sites but not the north arm. The antibiotic was not detected in any of the water samples. In the water samples, approximately 30% of analytes (total of 590 assessed) were detected. Of the 71 relevant Canadian water quality guidelines, five of them were exceeded, including two PCB and three polycyclic aromatic hydrocarbon WQGs.

The preliminary results suggest that the number of available environmental quality guidelines is insufficient to assess the risk of contaminants to Chinook and their habitat in the Fraser River estuary. We will continue to characterize and rank contaminant risk by using a combination of Exposure Activity Ratios and risk and toxicity quotients. In the future, we will also assess the health of juvenile Chinook in the Fraser River estuary using multi-omics techniques as well as traditional apical measurements. Results from this research will be used to inform Chinook conservation, recovery, and management efforts, especially as they relate to Fraser River Chinook and Southern Resident killer whales (*Orcinus orca*).

Hierarchical Clustering Captures Seasonal and Spatial Trends in Salish Sea Dissolved Copper Speciation

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- **99% of seawater dissolved copper is bound by a heterogeneous pool of organic ligands, buffering copper toxicity.**
- **We've defined distinct clusters of copper speciation parameters between seasons and regions of the Salish Sea.**

While copper is a metabolically essential micronutrient to marine biota, copper can also become toxic at relatively low concentrations (Brand, 1986; Ransberry et al., 2015). Presumably, more than 99% of dissolved copper (dCu) in the marine environment is complexed by a heterogeneous pool of natural organic ligands, which form stable, less bioavailable organic complexes that reduce bioavailable copper(2+) concentrations and buffer against copper toxicity (van den Berg et al., 1987). Cu binding ligands may be biologically derived from phytoplankton exudates (Moffett & Brand, 1997; Laglera et al., 2003; Dupont et al., 2004; Kim et al., 2005) or be of terrestrial origin, such as humic substances in river water (Kogut & Voelker, 2001; Voelker & Kogut, 2001) and biological macromolecules carried by municipal wastewater (Sedlak et al., 1997). Competitive ligand exchange – adsorptive cathodic stripping voltammetry measures ligand concentrations (L) and weighted average binding strengths ($K^{cond}_{CuL,\,Cu^{2+}}$) of the ambient heterogeneous copper binding ligand pool within the coastal environment. In collaboration with Metro Vancouver's Georgia Strait Ambient Monitoring Program, we attempted to understand the seasonal and spatial speciation of dCu in the Strait of Georgia (SoG), by measuring depth profiles of L and $\log K^{cond}_{CuL_i,\,Cu^{2+}}$ at four locations in the Salish Sea (i.e., northern SoG, southern SoG, Haro Strait, and Juan de Fuca Strait), in August 2018, and during four seasons (i.e., September 2017, December 2017, April 2018, and June 2018) within the southern SoG. We performed hierarchical clustering on this dataset to define distinct patterns of dCu speciation between seasons and regions.

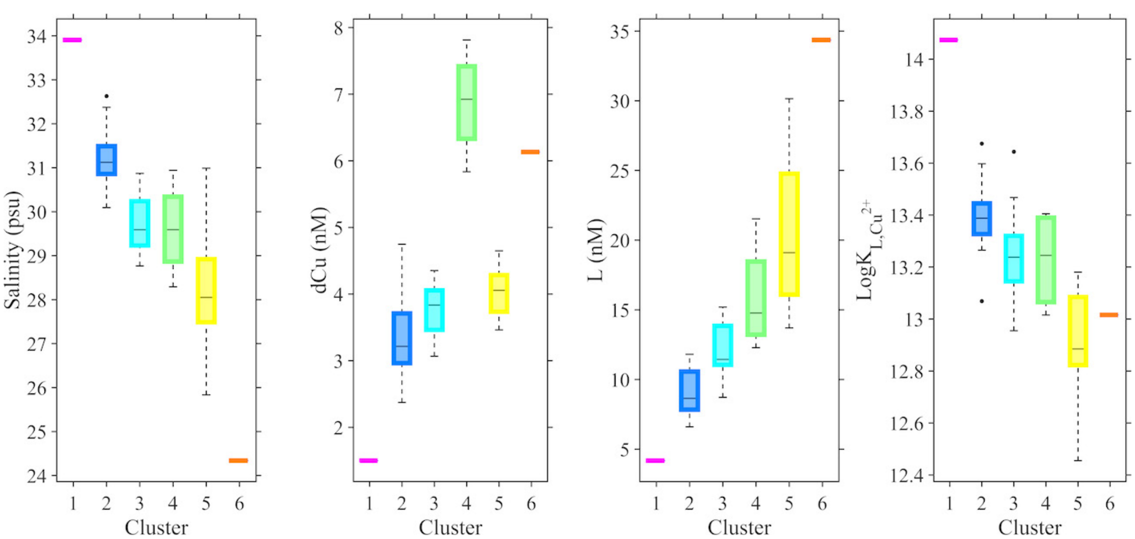

Figure 1. Boxplots of salinity, dCu concentration, L concentration, and $\log K^{cond}_{CuL,\,Cu^{2+}}$ for six clusters of seasonal and spatial SoG dCu speciation samples; 1 (n=1), 2 (n=10), 3 (n=28), 4 (n=4), 5 (n=13), and 6 (n=1). On each box, the central mark indicates the median, and the bottom and top edges of the box indicate the 25th and 75th percentiles, respectively. The dashed whiskers extend to the most extreme data points not considered outliers, and the black markers are outliers.

This analysis identified six clusters (Figure 1). Cluster 1 describes the deep Juan de Fuca Strait, which receives incoming intermediate northeast Pacific water (Pawlowicz et al., 2007), and consists of a low concentration of dCu and the strongest copper binding ligands in the SoG. Cluster 2 encompasses samples from intermediate Juan de Fuca Strait, deep Haro Strait, and deep SoG. Thus, cluster 2 describes the characteristics of the ligands which are carried to the deep SoG during summer deep water renewal (Masson, 2002). Cluster 3 describes the SoG intermediate water, which was received from Haro Strait surface water. Cluster 4 specifies spring southern SoG surface and intermediate water, where elevated dCu concentrations are evident, and aligns with regions of high particulate copper concentrations in the southern SoG, near surface and 200m depth (see Figure 22b in Flores Ruiz, 2020). Cluster 5 includes non-spring surface SoG and surface Juan de Fuca Strait, indicating input of either biologically derived ligands in sunlit depths, or the addition of terrestrial ligands from freshwater inputs. Cluster 6 is comprised of the single freshest sample, as the strongest Fraser River plume signal, signifying the highest contribution of copper binding ligands. These six clusters can be used as a baseline to understand the fate and potential toxicity of inorganic copper in the SoG in the future. For further discussion, please see Waugh et al. (2022).

Survey of PFAS in the Greater Lake Washington Watershed

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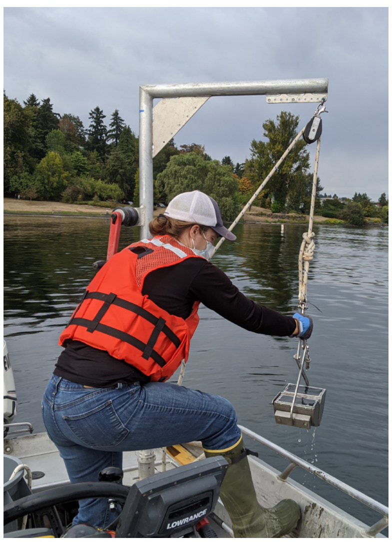
- **PFAS were detected in 224 of 226 samples, collected during multiple sampling events from the lake, tributaries, groundwater, stormwater, and bulk atmospheric deposition.**
- **Perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) were the most frequently detected of the 40 different PFAS that were analyzed.**

In 2020, the Washington State Department of Ecology initiated a field study to assess pathways and sources of per- and polyfluoroalkyl substances (PFAS) to Lake Washington. PFAS are a large group of human-made chemicals used in many industrial and consumer products, like those with water- or stain- repellant coatings, and fire-fighting foams.



Ecology staff collecting sediment sample from Lake Washington. Photo: Siana Wong

Our study follows up on previous surveys conducted in 2008 (Furl and Meredith 2010), 2016 (Mathieu and McCall 2017), and 2018 (Mathieu 2022), which documented high PFAS concentrations in urban waterbodies in Washington. The main goal of our current study is to identify and assess major pathways and sources by which PFAS enter Lake Washington, one of the urban lakes from the previous surveys.

From summer 2020 to spring 2021, we implemented the first phase of sampling. Our Phase 1 objective was to collect samples in the lake and in various pathways by which contaminants can enter the lake, including tributary inflows, stormwater, groundwater, and bulk (dry + wet) atmospheric deposition. Water, sediment, and biofilm samples were sent to a contract laboratory for analysis of 40 PFAS. Samples were also analyzed using a total oxidizable precursor (TOP) assay.

Initial results indicated that PFAS were widely present throughout the study area. PFAS were detected in 224 of 226 samples, which consisted of lake, tributary, groundwater, stormwater, and bulk atmospheric deposition samples collected during multiple sampling events. While total PFAS concentrations were fairly consistent in the lake surface water (median total PFAS concentration ~15 ng/L), concentrations in the tributaries, stormwater, and groundwater were more variable, with areas of lower and higher total PFAS concentrations that ranged from non-detect to over 100 ng/L (Figure 1). Perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) were the most frequently detected of the 40 different PFAS that were analyzed. These results suggest that, despite being largely phased out of manufacture and use in the U.S., there remain external sources of PFOS and PFOA as well as other PFAS chemicals to the lake.

The second phase of sampling began in summer 2022. During Phase 2, we will sample in areas of the watershed where higher PFAS concentrations or loads were found during Phase 1. Subsequent analyses of PFAS compositions and total concentrations, combined with knowledge about historic and current land uses, will help us prioritize and distinguish among different sources of PFAS to Lake Washington. By identifying major types of sources to the lake, we will better understand how sources can be reduced. While our current efforts focus on Lake Washington, information gained from this study will be largely applicable to other urban water bodies in Washington with PFAS contamination.

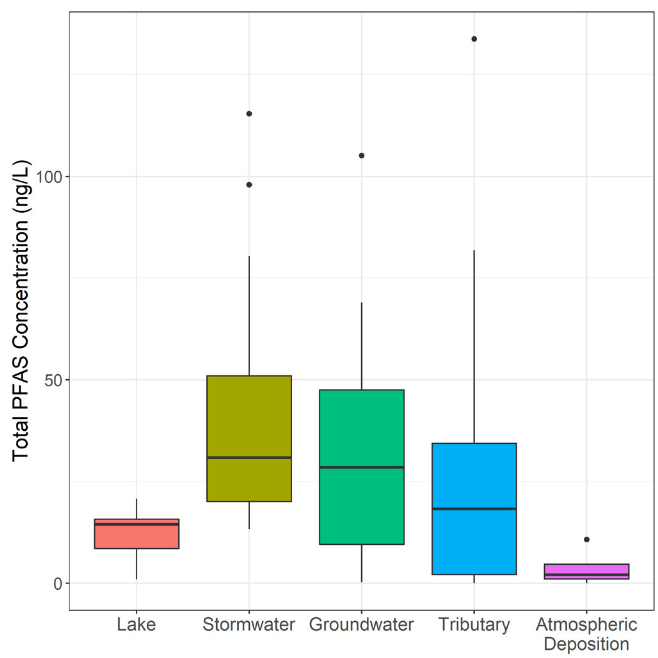


Figure 1. Box plots showing total PFAS concentrations in water samples collected from 2020-2021 in Lake Washington and contaminant pathways to the lake.

Baselines of PFOS flux to Puget Sound Marine Sediments

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- **Central Puget Sound Basin marine sediments, adjacent Seattle, experienced the largest observed PFOS flux over time.**
- **Central Hood Canal marine sediments held the greatest toxicologically relevant PFOS burden at the time of core collection.**

Poly- and perfluoroalkyl substances (PFAS), a large group of synthetic organic pollutants, have been in wide-spread use since the 1950s – due to unique properties that reduce friction, inhibit corrosion, repel liquids, and provide a thermal barrier for firefighting applications. Released PFAS are persistent in the environment, not efficiently depurated, and have negative acute and chronic effects on organisms. In marine environments, sediments can act as both a PFAS sink and a route of exposure into the base of the food-web. In addition to routine surveillance of contaminants of concern, reconstructions of historical depositions are key in providing insights into long-term ecosystem effects and allow the impacts of pollution management efforts to be quantified.

To document the flux of PFAS to Puget Sound sediments, in 2018 our team utilized four archived sediment cores, three of which were part of the seminal Puget Sound hypoxia studies detailed in Brandenberger et al. (2008), and published a Baselines paper covering the period from initial PFAS introduction through the turn of the century (Strivens et al., 2021). The cores were collected from (1) the central basin, adjacent Tacoma (core PS-1) and Seattle (core PS-4), (2) central Hood Canal (core HC-5), and (3) Carr Inlet (core CARR-1). At the quantification limits of the study, perfluorooctanesulfonic acid (PFOS) was the sole compound reported from comprehensive analyses which targeted 24 PFAS compounds.

PFOS in the marine environment has many potential origins and in a populated and militarized fjord watershed (i.e., Puget Sound) municipal wastewater treatment plant discharges and aqueous film-forming foams are significant contributors. The observed temporal trends of PFOS in Puget Sound sediments showed initial flux at centrally located stations coinciding with production history (Figure 1). At the Carr Inlet station there was a lag of ~ 20 years before detectable deposition/retention. Spatial comparisons showed that in 2005 Carr Inlet had the lowest PFOS flux at 0.206 ng/cm2/y, while PS-4 (adjacent Seattle) was experiencing the greatest flux at 0.742 ng/cm2/yr. When considering toxicological impacts, the sedimentation rate adjacent Seattle, and equivalent porosity with PS-1 (adjacent Tacoma), resulted in similar concentrations (0.723 and 0.699 ng/g [dw], respectively) in the biologically active layer (0–10 cm). Deposition characteristics resulted in HC-5 (central Hood Canal) having the greatest toxicologically relevant burden at the time of coring (0.823 ng/g [dw]), roughly double the burden of CARR-1.

Despite a phase-out of most PFOS production, initiated in the early 2000s by multiple national agencies, precursors that may transform or degrade to PFOS continue to be produced. In addition, PFOS is still imported in textiles and adequate replacements for military fire suppression systems have not been produced. The reported baselines provide spatial and temporal histories for Puget Sound's marine environment, offering insights on transport, fate, and biological impacts over time, as well as a means of interpreting effectiveness of current and future phase-out efforts.

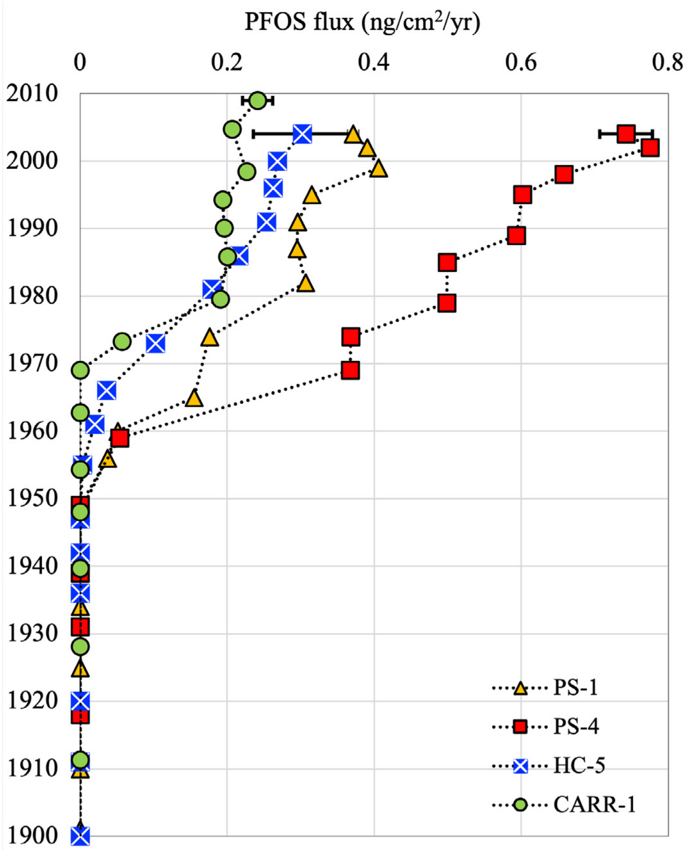

Figure 1. PFOS deposition trends in Puget Sound, WA. PS-1 is adjacent Tacoma (47° 20.830, 122° 24.580); PS-4 is adjacent Seattle (47° 36.898, 122° 26.941); HC-5 is central Hood Canal (47° 36.995, 122° 56.186); CARR-1 is central Carr Inlet (47°18.925, 122°42.902). This figure is reprinted from Strivens et al. (2021) and is in the public domain.

Low-tide at Discovery Park, Seattle, Washington. Photo: Sylvia Kantor

Occurrence and Distribution of Contaminants of Emerging Concern in the Puget Sound Nearshore Using a Marine Mussel Monitoring Program

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- **Using transplanted caged mussels requires a known starting concentration of CEC chemicals to evaluate contaminant loading and provides an opportunity to evaluate conditions in the nearshore across a variety of land-use types.**
- **Transplanted mussels from the 2019/2020 Marine Mussel Monitoring survey indicate the Puget Sound nearshore is contaminated with a wide range of CECs.**

In collaboration with the National Oceanic and Atmospheric Administration Mussel Watch Program, the Washington Department of Fish and Wildlife's Marine Mussel Monitoring program conducted a survey to assess the occurrence and distribution of contaminants of emerging concern (CECs) in Puget Sound nearshore biota. In the winter of 2019/2020, mussels (Mytilus trossulus) from a relatively uncontaminated local aquaculture source located on Whidbey Island, WA (baseline) were deployed in cages to 55 sites along the Puget Sound nearshore, with sampling locations representing a wide range of potential CEC exposures. After a three-month exposure period, a comprehensive CEC analysis targeting over 330 chemicals was performed on mussel tissue composite samples. CEC classes evaluated included pharmaceutical and personal care products (PPCPs), alkylphenols, current-use pesticides, and per- and polyfluoroalkyl substances (PFAS).

Thirty-seven unique chemicals from 19 general use chemical groups were detected at concentrations above the reporting limits in the mussels. The most frequently detected CEC use groups were antibiotics, PFAS, alkylphenol ethoxylates, psychiatrics, and antidiabetics, with detection frequencies between 82 and 96 percent (Figure 1). A closer examination of three frequently detected chemicals, Lomefloxacin (antibiotic), perfluorononanesulfonic acid (PFNS), and Sertraline (psychiatric), demonstrated they were widely detected across Puget Sound in both low and high development nearshore areas. The perfluorinated compound PFNS was detected at 52 of the 55 sampled sites, with concentrations ranging from 0.072 to 0.39 ng/g wet weight (Figure 2). Only seven of the sites had concentrations above the baseline concentration, meaning only a few sites had mussels accumulating additional loads of this contaminant. The location of these sites ranged from high development nearshore areas, such as Elliott Bay, to low development areas, such as Sequim Bay. Similarly, the psychiatric Sertraline was detected at 52 of the 55 sampled sites (detected concentrations 0.18 to 4.4 ng/g wet weight); most of these sites (43) had concentrations in mussels greater than the baseline sample concentration. Unlike PFNS and Sertraline, Lomefloxacin was not detected in the baseline sample, and 22 sites had detected concentrations ranging from 2.9 to 76 ng/g wet weight.

Mussel monitoring cage at Duwamish Head, Seattle, WA. Photo: Puget Soundkeeper Alliance

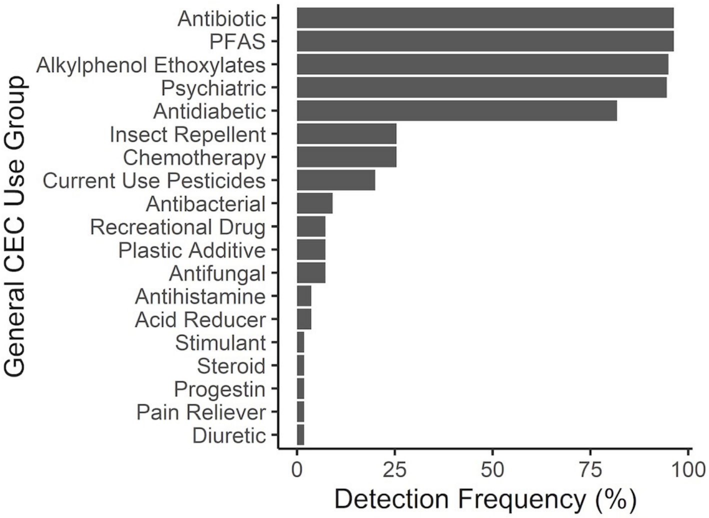

Figure 1. The detection frequency (%) of CEC use groups in mussels collected at the 2019/2020 WDFW and NOAA Mussel Monitoring sites.

This survey demonstrated the effectual use of transplanted caged mussels to monitor CECs in the Puget Sound nearshore and some of the advantages of using this approach. Sites with a wide variety of land-use types and proximity to putative CEC sources and pathways can be targeted, and it is important to know baseline concentrations in order to determine where mussels accumulate additional contaminant loads.

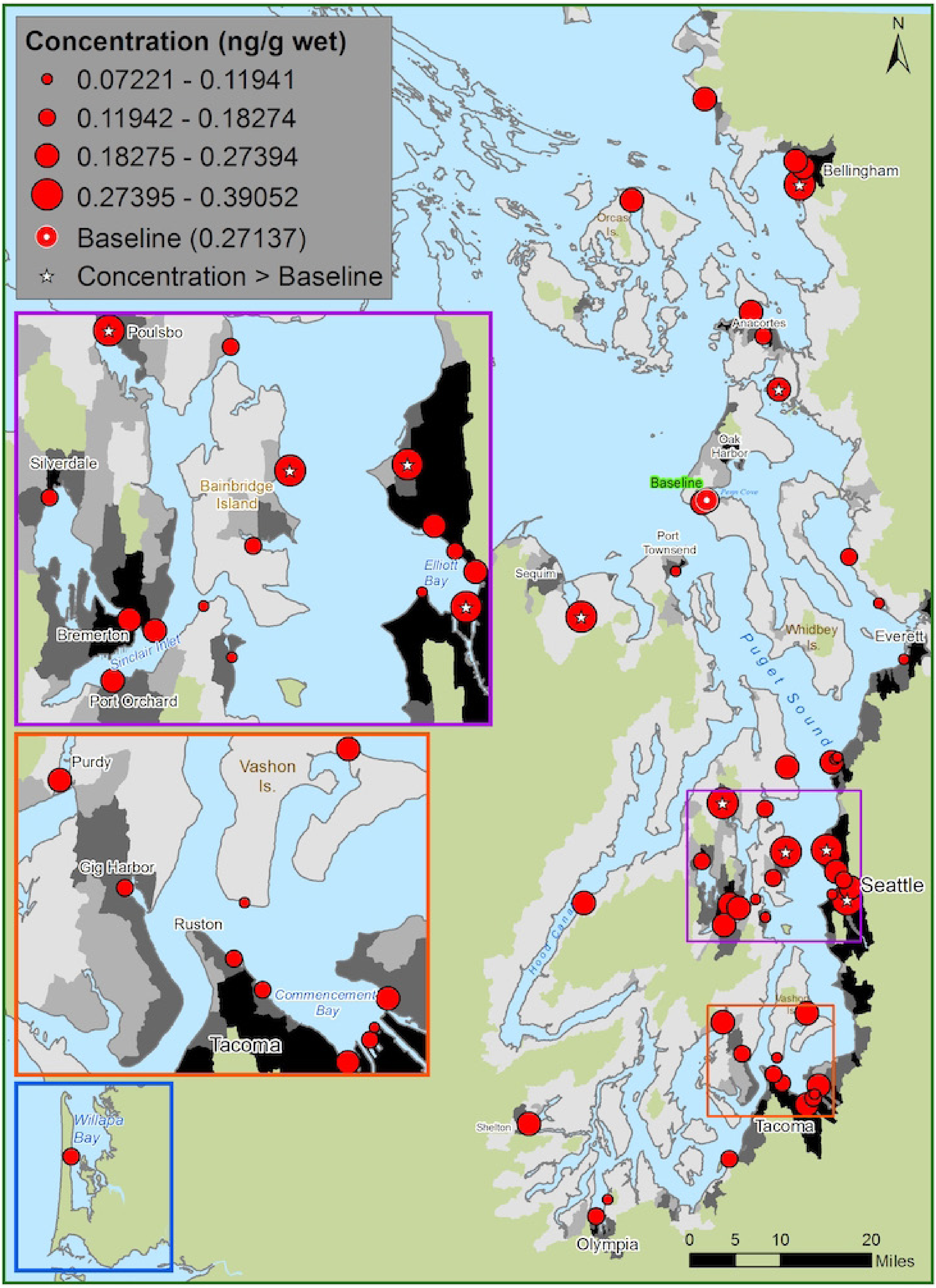

Figure 2. Map of the relative concentrations of PFNS from all the 2019/2020 WDFW and NOAA Mussel Monitoring sites. Starred sites indicate the concentration at the site was greater than the starting condition (baseline sample). Grey shading on land represents mean percent impervious surface (a proxy for development) on the adjacent shoreline watershed (darker shading = higher impervious surface).

Monitoring Contaminants in the Puget Sound Urban Nearshore Using Transplanted Mussels (*Mytilus trossulus*) to Assess the Effectiveness of Stormwater Management Actions Program

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- **Mussels in the urban nearshore of Puget Sound consistently have concentrations of PAHs, PCBs, PBDEs, and DDTs greater than those from a clean reference site.**
- **The spatial extent of PAHs, PCBs, PBDEs, and DDTs contamination in the urban nearshore has shown little to no decline in recent monitoring surveys.**
- **Central tendency concentrations of PAHs, PBDEs, and DDTs in mussels from the urban nearshore declined in the 2019/20 survey.**

Monitoring pollutants in contaminated stormwater and effects on the marine biota of Puget Sound are critical to inform best management practices and remediation efforts in this large and diverse estuary. The Stormwater Action Monitoring Status and Trends in Receiving Waters program conducts monitoring in Puget Sound to provide a regional assessment of whether collective stormwater management actions are leading to improved receiving water conditions. These Puget Sound Nearshore Monitoring studies focus on the bioaccumulation of pollutants in caged native bay mussels (*Mytilus trossulus*) to evaluate the current status and trends of nearshore conditions. In the winter of 2019/2020 the Washington Department of Fish and Wildlife's Toxics Biological Observation System team, with the help of citizen science volunteers, other agencies, tribes, and non-governmental organizations, conducted the third of a series of biennial, nearshore mussel monitoring surveys.

The 2019/2020 survey provided the first opportunity to evaluate changes in contamination of nearshore biota residing within urban growth areas of Puget Sound since the surveys conducted in 2015/2016 and 2017/2018. In each survey year, relatively uncontaminated mussels from a local aquaculture source were transplanted to over 40 monitoring locations along the Puget Sound urban shoreline, and at least one reference site with no

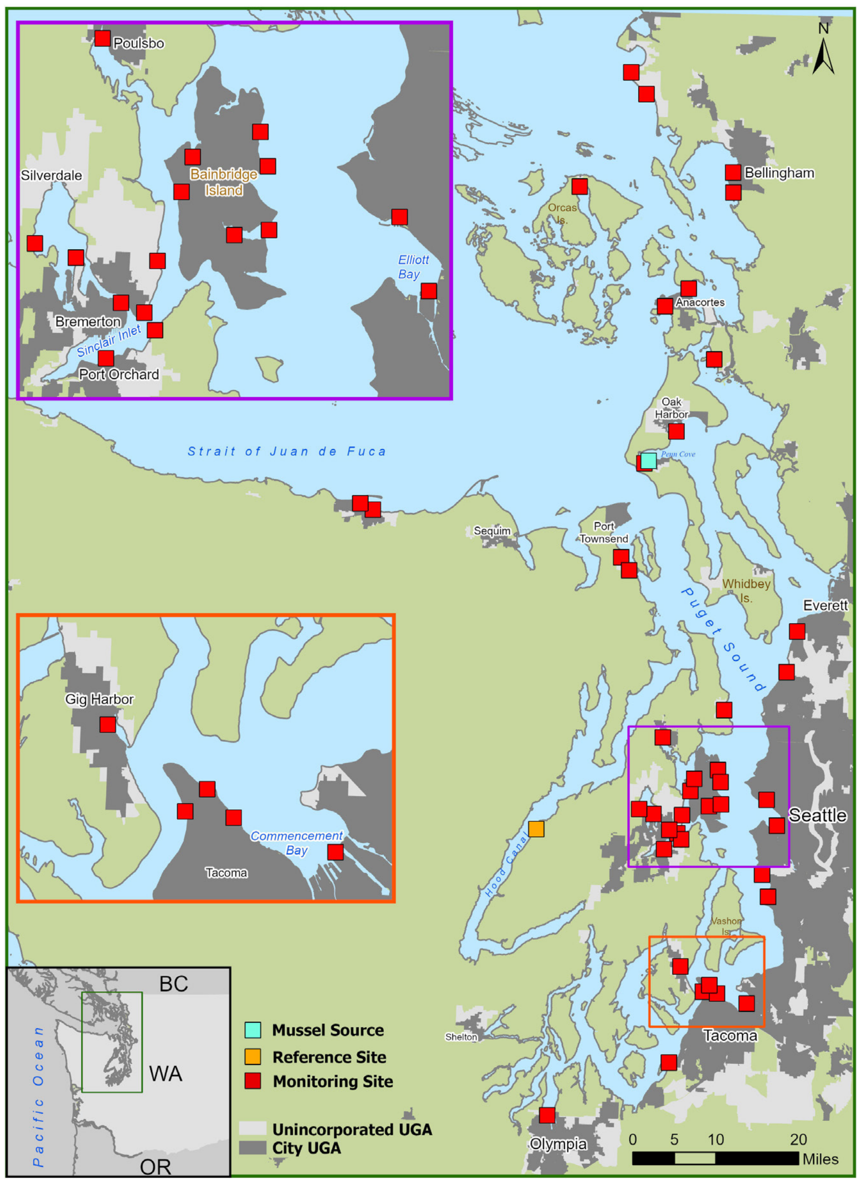

Figure 1. Stormwater Action Monitoring Nearshore Mussel Monitoring sites in the Puget Sound urban growth areas. Grey shading on land represents municipal land-use designations based on urban growth area boundaries; dark grey representing City urban growth areas and light grey representing Unincorporated urban growth areas.

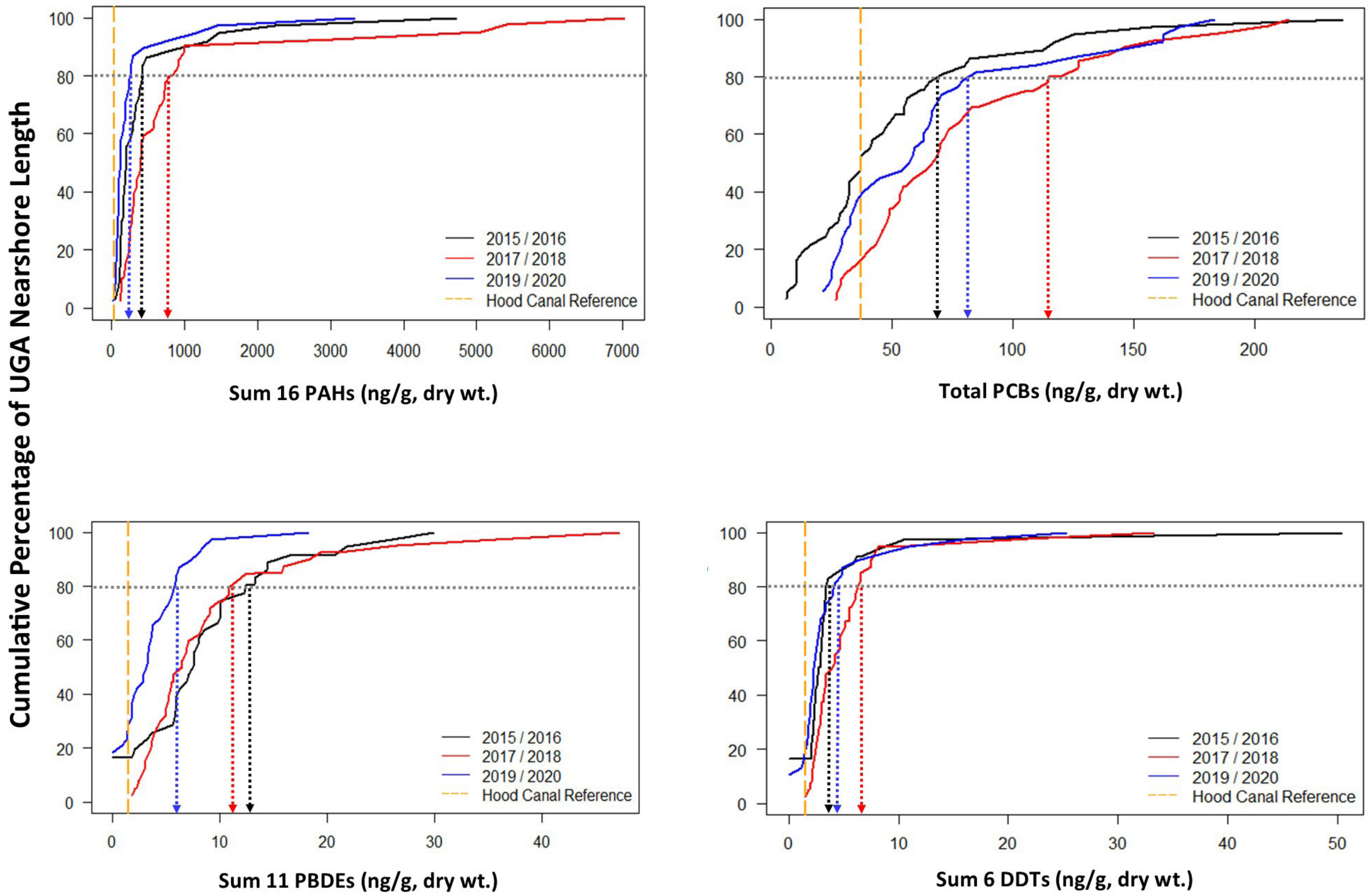

Figure 2. Cumulative frequency distribution of $\sum_{16}$PAHs, TPCBs, $\sum_{11}$PBDEs , and $\sum_{6}$DDTs concentrations in mussels from 2016, 2018, and 2020 study sites, representing the total sampled length of nearshore in Puget Sound urban growth areas. The dashed yellow line represents the Hood Canal Holly Reference site established as a regional threshold with which to compare against. Dotted lines are guides to read the plot, pointing to the concentrations observed in each survey year at 80% of the total sampled urban growth area nearshore length.

known sources of contamination to establish regional scale thresholds (Figure 1). Mussel soft tissue composites from each site were analyzed for a range of organic contaminants including polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), organochlorine pesticides (dichlorodiphenyltrichloroethane [DDTs], hexachlorocyclohexanes [HCHs], hexachlorobenzene [HCB], chlordanes, dieldrin, aldrin, mirex, and endosulfan), and metals (arsenic, cadmium, copper, lead, mercury, and zinc).

Here we report on the four most frequently detected organic contaminants (PAHs, PCBs, PBDEs, and DDTs), and characterize changes in the spatial extent of contamination in the Puget Sound urban nearshore and evaluate the temporal changes in concentrations of these key contaminants between each of the three survey years. The Σ_{16}PAHs, total PCBs (TPCBs), Σ_{11}PBDEs, and Σ_{6}DDTs concentrations in mussels collected from nearshore locations were consistently greater than those from the clean reference site established in Hood Canal, indicating elevated uptake. The spatial extent or cumulative distribution of these contaminants in mussels along the Puget Sound urban nearshore showed little to no decline during the three survey years (Figure 2). For example, 80% of the total nearshore length had TPCBs concentrations below approximately 113 ng/g in 2017/2018, below 80 ng/g in 2019/2020, and below 70 ng/g in 2015/2016 (Figure 2). Σ_{16}PAHs, Σ_{11}PBDEs, and Σ_{6}DDTs had significantly lower central tendency concentrations in mussels from the 2019/2020 survey than those in the 2015/2016 and/or 2017/2018 surveys. TPCBs data were inconclusive as there was no significant difference in mean concentration values attributable to survey year. Although additional sampling years are needed to infer conclusions regarding any significant trends in these organic contaminant concentrations, the declining Σ_{11}PBDEs concentrations but stable TPCBs concentrations were congruent with the temporal pattern in two other WDFW indicator species (English sole and Pacific herring) reported in the Toxics in Aquatic Life Vital Sign.

Contaminants of Emerging Concern in Marine and Freshwater Fish in King County

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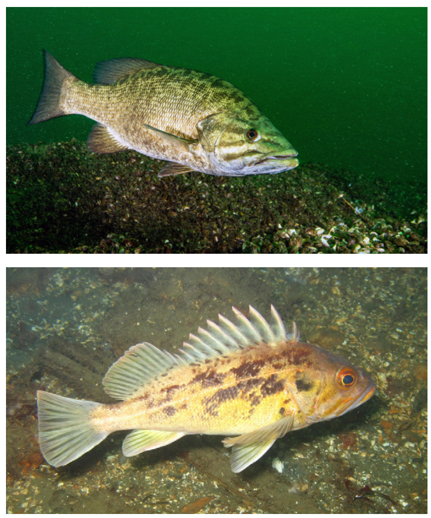
- **This study provides valuable baseline information on the recent occurrence of CECs in rockfish from Elliott Bay and smallmouth bass from Lake Union.**
- **21 chemicals detected in the rockfish or bass were listed as high priority or watch list chemicals in a recent Puget Sound CEC prioritization report.**

Human use of pharmaceuticals and personal care products leads to wastewater discharges of these chemicals to our waterways. These contaminants of emerging concern (CECs) are increasingly detected in aquatic environments and some of them may be harming fish and wildlife, but studies evaluating CECs in King County fish are limited. To expand our local knowledge about exposure of King County fish to mixtures of CECs we collected smallmouth bass from Lake Union and rockfish from Elliott Bay in 2021 and analyzed them for pharmaceuticals, personal care products, triclosan (in antibiotic soaps), and alkylphenols (active ingredients in cleaning products).



Smallmouth bass (top) and brown rockfish (bottom). Photo: Jennifer Vanderhoof

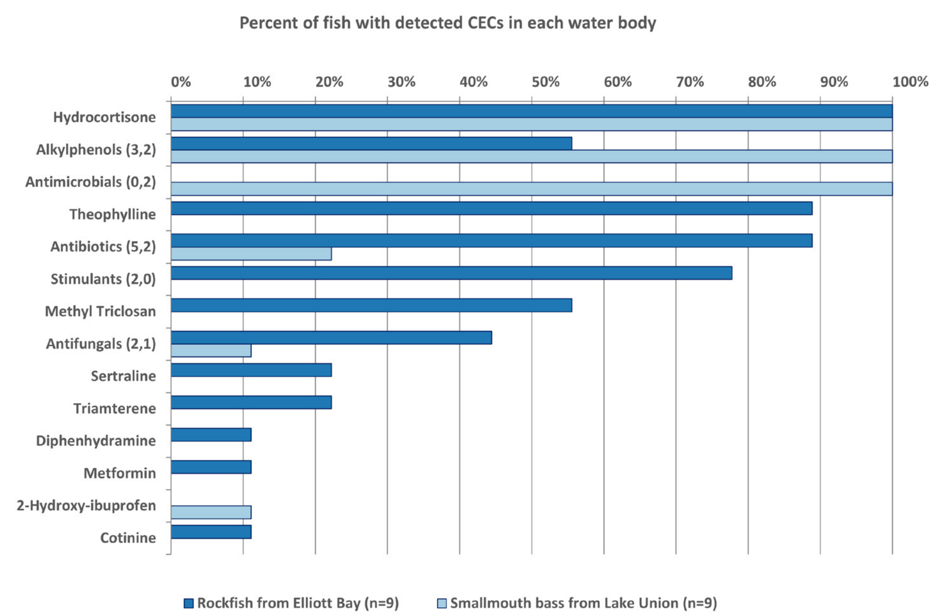


Figure 1. Percent of fish with detected CECs by individual chemical or chemical class. For chemical class, the values in parentheses show number of chemicals detected for rockfish and/or smallmouth bass, respectively.

We detected 20 CECs in rockfish and 9 in smallmouth bass. Over half the CECs we detected in rockfish have previously been detected in Puget Sound sediment, effluent, and/or water (King County, 2017; Meador et al., 2016; Long et al., 2013). Many of them also occurred in fish and/or mussels sampled in other local and national studies, indicating their widespread presence in aquatic environments (Meador et al., 2016; James et al., 2020; Krogh et al., 2017; Ramirez et al., 2009). In addition, we found multiple antibiotics and antimicrobials in the Lake Union and Elliott Bay fish, which suggests the potential for development of bacteria with antibiotic resistance genes in those water bodies. Interestingly, none of the CECs we detected in smallmouth bass were found in Lake Union water sampled in 2015 (King County, 2017). However, this may be due to a mismatch in the timing of sampling. The water samples from Lake Union were collected in the fall and winter of 2015, when discharges from wastewater, stormwater and CSOs are generally higher. In contrast the smallmouth bass were collected during a relatively dry period in mid-May to early-June of 2021.

The baseline information generated from this survey will be useful in tracking CECs in fish and prioritizing future King County monitoring and water quality actions related to CECs. Based on a recent CEC prioritization effort conducted by the PSEMP Toxics Work Group (James and Sofield, 2021), three of the CECs we detected in the fish from this study are listed as high priority chemicals and 18 are listed as watch list chemicals. Considering their potential for biological effects, we recommend further monitoring of the high priority and watch list chemicals detected in King County fish from this study. We also recommend compiling a list of effects thresholds for CECs detected in all organisms from Puget Sound. This will be useful for future evaluations of effects of CECs in King County aquatic environments.

Comparison of Lower Duwamish Waterway Baseline Tissue Results with Historical Data and Target Tissue Levels

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- **Concentrations in LDW fish and shellfish tissue are above the Target Tissue Levels for the human health risk driver chemicals.**
- **Risk driver baseline tissue concentrations were less than or similar to historical data and in line with post-cleanup model predictions for PCBs.**

The Lower Duwamish Waterway (LDW), located in Seattle, Washington, is a 5-mile estuarine site that was listed as a Superfund site in 2001 and as a Washington State Model Toxics Control Act site in 2002. The Record of Decision (ROD) for the LDW was released by the Environmental Protection Agency in late 2014 and early action area sediment cleanups were completed by 2016. Baseline sampling was conducted in 2018 to serve as a foundation for assessing trends in resident fish and shellfish tissue concentrations following early action cleanups.

Processing fish and crab collected from the Lower Duwamish Waterway as part of baseline sampling in 2018. Photo: Thai Do

As part of baseline sampling in 2018, fish (English sole and shiner surfperch), crab (Dungeness and slender crab), and clam (eastern softshell) were collected throughout the LDW and analyzed for the four human health risk drivers: polychlorinated biphenyls (PCBs), dioxins/furans, carcinogenic polycyclic aromatic hydrocarbons (cPAHs), and arsenic. The baseline data provided 1) a snapshot of tissue concentrations following the completion of early remedial actions and prior to the implementation of the site-wide remedy, and 2) a point of comparison with target tissue levels (TTLs) derived for risk communication. The TTLs are primarily based on concentrations in non-urban areas of Puget Sound (ROD Table 21).

Relative to these uses, key conclusions for the risk driver chemicals (Figure 1) are as follows:

- **Total PCBs** – Early remedial actions have reduced site-wide average sediment PCB concentrations in the LDW by approximately 50%. PCB concentrations in baseline tissue samples were generally less than or similar to those in 2007 and above TTLs (which range from 0.42 µg/kg ww for clams to 12 µg/kg ww for English sole fillet tissue). The LDW food web model, which was developed for PCBs as part of the Remedial Investigation (Windward 2010), accurately predicted total PCB concentrations following early remedial actions.
- **Dioxins/furans** – No historical dioxin/furan data are available for comparison to baseline data. In general, dioxin/furan toxic equivalents (TEQs) were above the TTL for fish (0.35 ng/kg ww for English sole fillet) and below the TTL for both crabs and clams (0.53 to 2 ng/kg ww).
- **cPAHs** – Risks associated with cPAHs are driven by clam consumption. cPAH TEQs were lower than in historical data (2004) but generally higher than the TTL for clam tissue (1.5 µg/kg ww).
- **Arsenic** – Like cPAHs, risks associated with arsenic are driven by clam consumption. There were no clear temporal trends for arsenic in clam tissue. Inorganic arsenic concentrations in clam tissue were above the TTL (0.09 mg/kg ww). However, when the siphon skin tissue was removed, concentrations were close to the TTL.

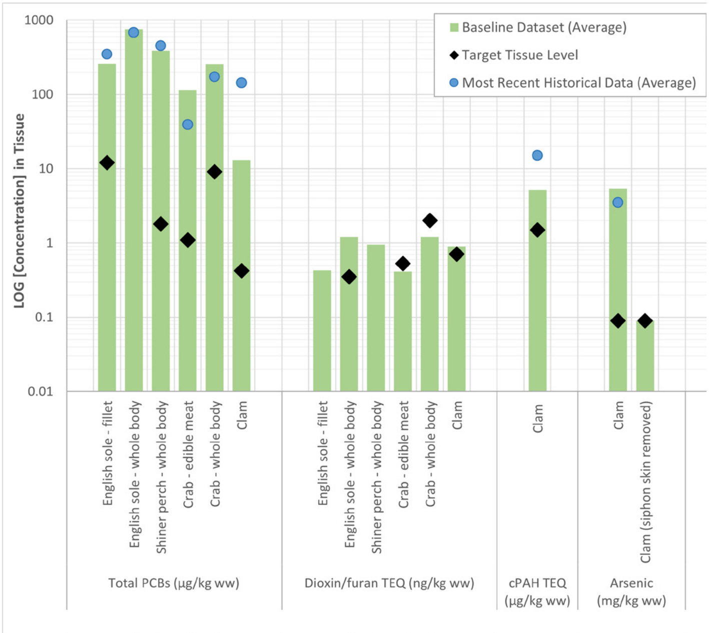

Figure 1. Comparison of baseline tissue concentrations with historical data and TTLs for the Lower Duwamish Waterway in Seattle, WA.

Predicting the Ecotoxicological Impacts of Microplastics in the Northern Salish Sea using GIS

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- **GIS is a useful tool for modelling microplastic accumulation at an ecosystem scale.**
- **The highest risk for microplastic accumulation include: salmon-bearing stream estuaries, protected areas, and marine mammal haul-outs.**

Microplastics (plastic particles less than 5mm in diameter) are of increasing concern in the marine environment due to their impacts on biota through ingestion and their ability to leach and absorb toxicants (Guzzetti et al., 2018; Koelmans et al., 2014). Trophic level interactions and resulting bioaccumulation of microplastics and associated toxicants are known to be of concern (do Sul & Costa, 2014; Cole et al., 2011). The combination of depth and exposure to wind and current patterns cause the upwelling of cold, nutrient-rich water in the northern Salish Sea; this creates a biodiversity hotspot with an estimated 37 species of mammals, 172 species of birds, 253 species of fish, and more than 3000 species of invertebrates (Gaydos & Pearson, 2011).

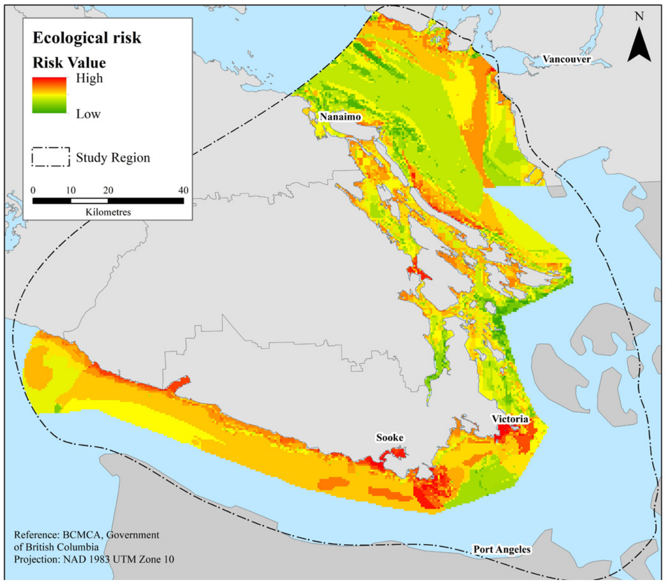
Transient orca (*Orcinus orca*) swimming in Haro Strait. Photo: Melina Sorensen

Given the known threats of microplastics to marine biota and our understanding of the marine food web relationships, the Salish Sea is an essential marine region to study to better understand, predict, and potentially mitigate microplastic accumulation. While the entire Salish Sea is important, this research is focussed on the Canadian areas of the Salish Sea, specifically the regions of the Juan de Fuca Strait, Haro Strait and Rosario Passage, the Interior Gulf Islands, and the Southern and Central Strait of Georgia as a case study.

Due to the physical properties of microplastics, such as their size, buoyancy, and ability to be transported great distances by wind and waves (Ballent et al., 2021; Zhang, 2017), conducting adequate studies at an ecosystem level is challenging. However, GIS provides tools for storing, analyzing, and displaying spatial data to address challenges in many disciplines, including ecotoxicology (Awange & Kiema, 2019; Besseling et al., 2019).

By performing weighted raster analysis using GIS, I mapped areas of the Northern Salish Sea where microplastics are likely to occur and where microplastics are likely to cause the most ecological harm. The data chosen for analysis were from two primary sources, the British Columbia Marine Conservation Analysis (BCMCA) project and the Government of British Columbia. The six physical parameters included were: (1) benthic class; (2) bathymetry; (3) high rugosity; (4) shorezone exposure; (5) benthic current; and (6) tidal current. The nine ecological parameters included were: (1) chlorophyll concentration; (2) ecologically and biologically significant areas; (3) ecological feature count; (4) fish and invertebrate feature count; (5) habitat bases estimate of salmon productivity; (6) historical and current fish distribution; (7) marine ecosections; (8) marine mammal feature count; and (9) marine plant feature count.

The areas of highest microplastic accumulation risk are off the coast of Victoria, Oak Bay, Sooke, Port Renfrew, and small regions of the Southern Gulf Islands. The areas of highest ecological risk are concentrated off Sooke, Victoria, Oak Bay, and between Salt Spring Island and Maple Bay. The locations determined to be of the highest risk



Total ecological risk value map based on ecological features and physical calculation results.

overlap with key ecological regions, including salmon-bearing stream estuaries, protected areas, and marine mammal haul-outs. By identifying vulnerable areas and where microplastics are likely to accumulate, the results could be helpful for conservation managers, fisheries management, and natural resource managers.

Comparison of Microplastics in Puget Sound via the Puyallup River from 2017-2018 Using University of Washington Tacoma Datasets

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- **Data from river stations three through seven indicated that microplastic abundance increased from 2017 to 2018.**

The pollution of our oceans is a pressing problem for the health and prosperity of marine environments. Plastic material is the most prominent constituent of pollution in the marine environment leading to a myriad of adverse consequences (Derraik, 2002). Microplastics are either manufactured or created by the breakdown of larger plastic materials. Although there is no absolute lower boundary for size, a microplastic is generally defined as being in the range of 333 microns to 5 millimeters (Arthur et al., 2009). Microplastic pollution has lasting effects on marine communities because some materials do not disintegrate easily and can even remain in the sediments. Microplastics impact marine communities via contamination of their habitats and accidental ingestion. The consumption of plastic material is harmful as it can be toxic, pose as a choking threat, or cause death (Barnes et al., 2009). Ultimately, plastic pollution transported to the ocean can have adverse consequences on humans via food sources. Plastic particles enter the human diet by the ingestion of contaminated fish and seafood (Davison & Asch, 2011).

Example of various plastics collected from a net tow through Commencement Bay, Tacoma. Photo: Keely Hall

Anthropogenic factors have played a significant role in amplifying the problem of microplastic pollution. Population density is positively correlated to the amount of microplastics in the environment (Mbedzi et al., 2020). This is especially relevant for heavily populated cities on coastlines or by rivers since these locations have direct contact with water. Additionally, rivers are significant contributors for transporting microplastics into the ocean (Xiong et al., 2019). Therefore, the Puget Sound, Washington, estuary system is an important location for this research topic due to the several rivers which can be potential sources of microplastic pollution.

This research investigated the contribution of microplastics to Puget Sound via the Puyallup River from 2017 to 2018. Datasets from UW Tacoma were analyzed to determine spatial and temporal variation of microplastic abundance across nine sampling stations. The first seven stations were river samples whereas stations eight and nine were located in Commencement Bay, Tacoma (Figure 1). Data from river stations three through seven indicated that microplastic abundance increased from 2017 to 2018 (Figure 2; P = 0.0287). Additionally, data from river stations three through seven demonstrated a positive correlation between average microplastic abundance and total population both years (2017, R2 = 0.8749; 2018, R2 = 0.6195). Although spatial results were variable, abundance increased in the Puyallup River between the two sites closest to Puget Sound. 2018 data had a larger microplastic type distribution than 2017 with 63 percent fiber and 37 percent fragment, while 2017 had only fibers. Regulations on anthropogenic related pollution are necessary as microplastics continue to accumulate in the ocean. Microplastic research is important for influencing environmental policy regarding mismanaged plastic waste and the production of microplastics. Further research with larger datasets and over longer timescales is crucial for resolving variability in the data and determining the impact plastic pollution has on marine communities.

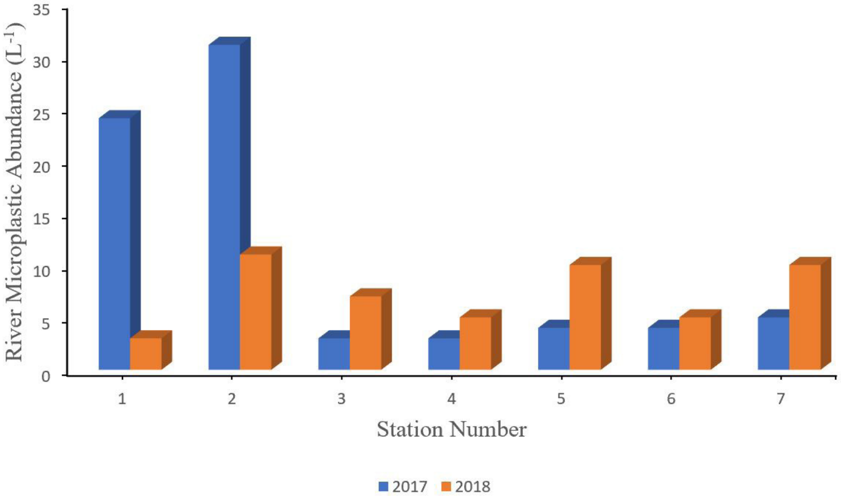

Figure 1. Microplastic abundance across all river stations, 1-7. Blue represents 2017 data and orange represents 2018 data.

Pollution Tracker, a Coast-wide Marine Pollution Monitoring Program in British Columbia, Canada

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- **79 Pollution Tracker sites have been established along the coast of British Columbia, Canada.**
- **Sediment and mussel samples are analyzed for over 550 contaminants selected from 14 contaminant classes of particular concern to ocean health.**

Ocean Wise's Pollution Tracker is a coast-wide, integrated marine pollution monitoring program in British Columbia (BC), Canada, documenting the levels of hundreds of contaminants in mussels and nearshore ocean sediments. Pollution Tracker is the first program of its kind in Canada and was designed to determine the relative state of marine pollution in coastal BC over space and time. It was launched in 2015 with a three-year sampling cycle. A total of 79 coastal locations have been established in partnership with First Nations, community groups, port authorities, industry, and government agencies (full list of partners). Samples are being analyzed for over 550 chemical contaminants and microplastics, including priority contaminants of concern for the health and recovery of the Southern Resident Killer Whale (Orcinus orca) population.

Based on results from Phase 1 (2015-2017) and Phase 2 (2018-2020), a wide variety of contaminants were detected along the coast. When available, Federal (Canadian) and Provincial (British Columbia) sediment and tissue quality guidelines protective of marine organisms were used to evaluate the quality of the marine environment. In the present chapter, we focus on results for sediment samples averaged across both phases:

- Legacy Persistent Organic Pollutants such as polychlorinated biphenyls (PCBs), dioxins and furans, and polybrominated diphenyl ethers (PBDEs) were detected at all sites. PCB levels at all sites exceeded the BC Working Sediment Quality Guideline of 0.0037 µg/kg protective of killer whales while 36% of the sites exceeded the PBDE BC Working Sediment Quality Guideline of 1 µg/kg;
- Polycyclic aromatic hydrocarbons (PAHs) were detected at all sites;
- The heavy metals cadmium, lead and mercury were detected at 98%, 100% and 92% of the sites, respectively; of those 30%, 12%, and 28% exceeded sediment quality guidelines (SQGs) protective of marine benthic invertebrates, respectively;
- The brominated flame retardants tetrabromobisphenol A and hexabromocyclododecane were detected at 3% and 36% of the sites, respectively;
- Perfluorinated compounds were detected at 21% of the sites;
- Legacy pesticides were detected at 93% of the sites while current use pesticides were detected at 54% of the sites;

Pollution Tracker is documenting the levels and trends of hundreds of contaminants in mussels and nearshore ocean sediments.

- Pharmaceutical and personal care products were detected at 45% of the sites;
- Organotins were detected at 49% of the sites;
- Alkylphenols were detected at 68% of the sites;
- Microplastics were detected in 89% of the samples (Phase 1 results only) and averaged 32.6 ± 5.3 particles per kg of sediment (Noel et al., in prep).

Overall, contaminant concentrations were highest in industrialized and urban areas; however, a small number of contaminants including alachlor, alkylphenols, cadmium, and mercury, were found at relatively high levels in more remote areas. The contaminants detected reflect a variety of sources, including local, urban, and industrial inputs to the marine environment, as well as external inputs, and oceanographic factors.

Phase 3 is currently underway, and will run until 2024, building on Phases 1 and 2 results at dedicated sites, expanding the project's spatial coverage, and continuing to add to a valuable baseline dataset that will help British Columbia better understand, manage, and prevent marine pollution.

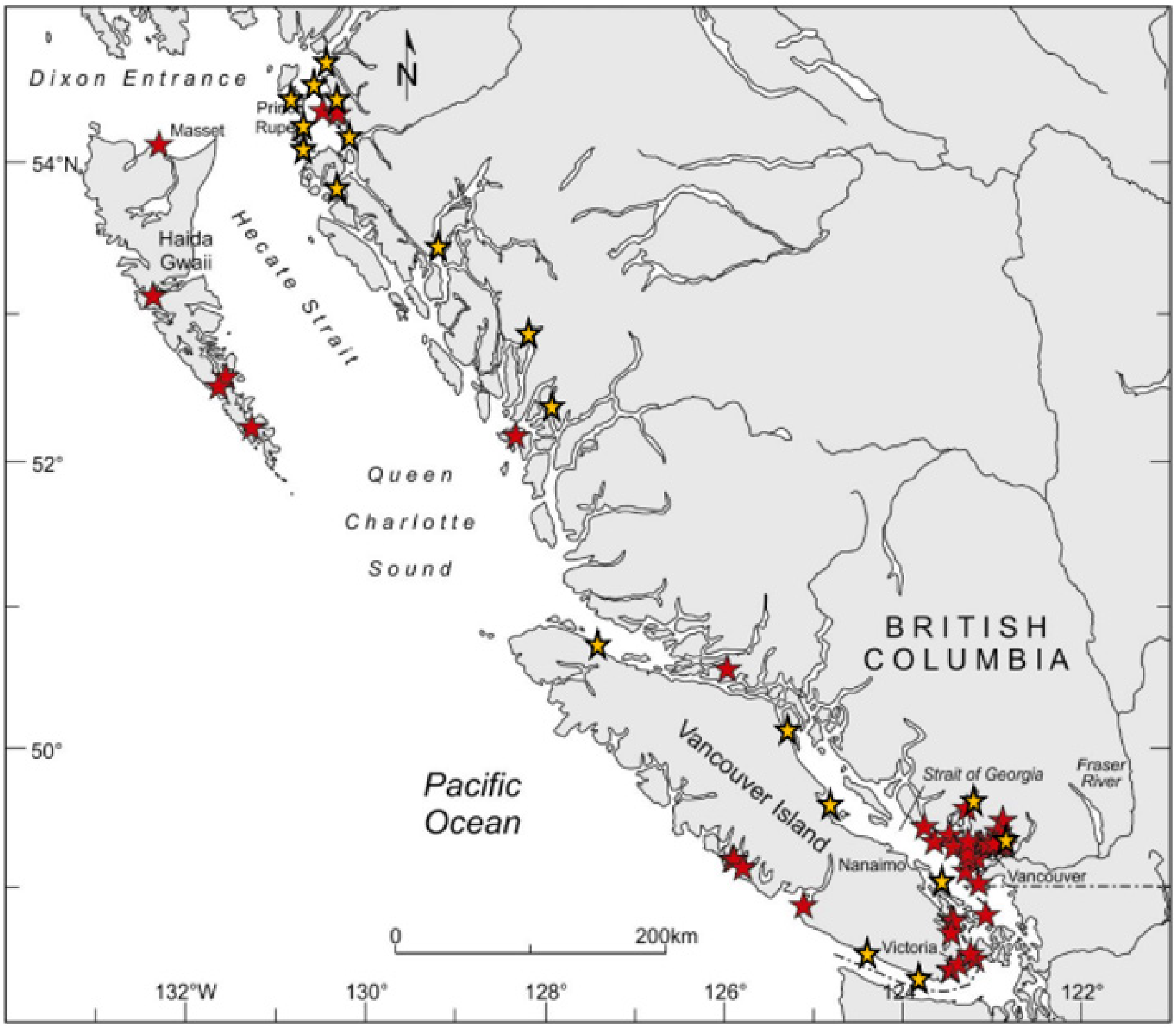

Figure 1. Pollution Tracker sites have been established all along the coast of British Columbia. Red stars indicate sites established during Phase 1 while yellow stars indicate sites that were added during Phase 2.

Spatial Distribution of Chemicals in Puget Sound Sediments

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- **The highest concentrations of pollutants were found in the industrial harbors of Puget Sound near Seattle, Tacoma, Bremerton, and Olympia.**
- **Total organic carbon and finer-grained sediments tended to be higher in the same locations as higher concentrations of pollutants.**

The Washington State Department of Ecology (Ecology) conducts long-term status-and-trends monitoring of marine sediments as part of the Puget Sound Ecosystem Monitoring Program (PSEMP). Ecology analyzed for 87 chemicals in the top 2 to 3 cm of sediment at 50 locations throughout Puget Sound during 2016-2021.

Several of the individual chemicals measured were not present at concentrations above the reporting limit of the analytical methods, particularly those in the polybrominated diphenylether (PBDE), polychlorinated biphenyl (PCB), and phthalate chemical classes. Metals and polycyclic aromatic hydrocarbons (PAHs) were most often detected.

Marine sediment monitoring staff collect sediments from the bottom of Commencement Bay. Photo: Tim Zornes

The challenge is to indicate the spatial distribution of complex mixtures of potentially toxic chemicals when the concentrations vary by orders of magnitude and most do not have associated WA State Sediment Quality Standards (Ecology, 2013). In order to make use of information on all of the chemicals, we ordered and ranked each of the 87 chemicals from low to high concentration over the entire station network. Then for each station, we calculated the average rank of chemicals, focusing only on those detected at 10% or more of the stations (59 of the 87 chemicals). Finally, we mapped the station ranks to visualize areas of Puget Sound where higher concentrations of chemicals were found (Figure 1). The station ranks of concentrations were well-correlated with the numbers of detected chemicals (Pearson $r = 0.841$).

When chemicals were detected, they tended to be detected in clusters, with higher average ranks found in the industrial harbors of Puget Sound near the cities of Seattle, Tacoma, Bremerton, and Olympia. Proximity to industrial activities is only one of multiple possible drivers. Average ranks were positively correlated with total organic carbon content and fine-grained sediments (Pearson $r = 0.713$ and 0.718, respectively). High total organic carbon content and fine-grained sediments are generally thought to be associated with low-energy, depositional environments such as terminal inlets; these were often the same locations where high average ranks occurred.

In general, the majority of Puget Sound did not have concentrations of chemical contaminants above state standards, though a few chemicals did not meet state standards at a handful of locations. These results were consistent with previous Puget Sound-wide sediment chemistry assessments (Weakland et al., 2018; Partridge et al., 2018).

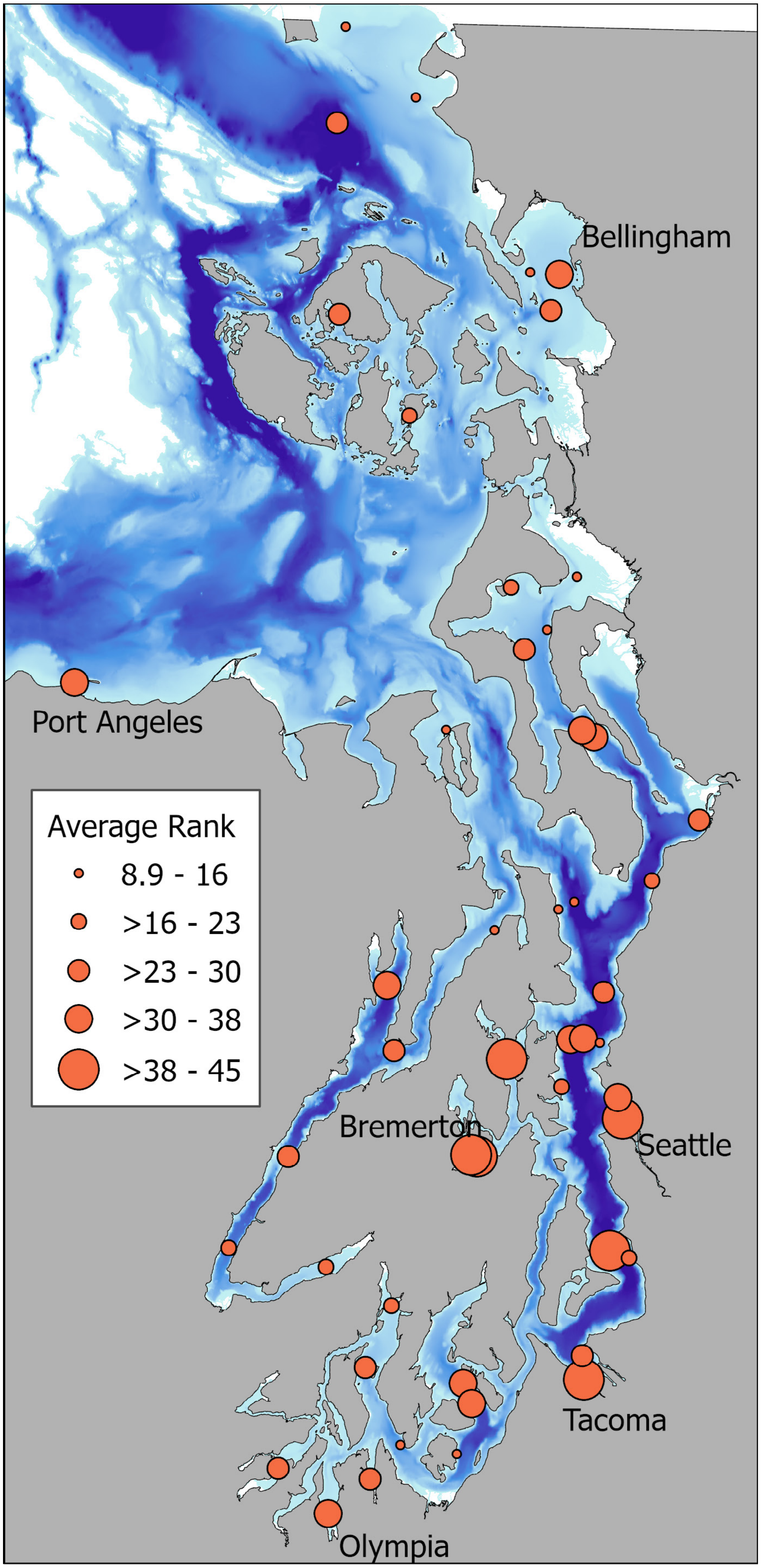

Figure 1. Average rank of stations for 59 pollutants in Puget Sound surface sediments. Higher average ranks indicate higher concentrations of potentially toxic chemicals.

Revisiting a Site of Historic Sediment Contamination in Seattle, 25 Years Later

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Websites: Final report https://green2.kingcounty.gov/ScienceLibrary/Document.aspx?ArticleID=677; sampling and analysis plan: https://green2.kingcounty.gov/ScienceLibrary/Document.aspx?ArticleID=603

- **Widespread persistent PAH contamination has been documented along West Point's north shelf, with highest concentrations located near the decommissioned North Trunk Outfall.**
- **Compared to 1996 results, PAH concentrations along West Point's north shelf have only slightly decreased, suggesting low rates of natural recovery.**

In February 2017, King County's West Point Wastewater Treatment Plant, located in Discovery Park, Seattle, Washington, experienced equipment failure related to record rainfall and flow into the plant. The result was flooding of the plant and damage to mechanical and electrical systems. To avoid further damage, the plant discharged 244 million gallons of untreated stormwater and sewage through the Emergency Bypass Outfall (EBO). The potential effect of the discharges on sediments near the EBO was initially assessed with a sediment deposition modeling effort because known historical contamination in the area would make it difficult to attribute any contamination to the release.

Data collected from the north shelf of Discovery Park in the 1990s, before the treatment plant was last upgraded and the EBO was operational, indicated that sediment along the shelf had elevated concentrations of polycyclic aromatic hydrocarbons (PAHs). Contamination was found near where the plant's EBO would be built. King County's Wastewater Treatment Division wanted to better understand the status and spatial extent of sediment contamination along the shelf and further assess any potential effect of the EBO discharges on sediments.

In December of 2019, we collected sediments along the north shelf off Discovery Park for analysis of metals, polychlorinated biphenyls (PCBs), PAHs, and other semivolatile organic compounds (SVOCs). We sampled a total of 33 locations along the shelf (including near the EBO) from the western tip of West Point to east of a decommissioned pipe known as the North Trunk Outfall, which discharged untreated sewage from 1913 to 1966. A total of 22 chemicals had concentrations above the marine sediment quality standards; the number of exceedances and high concentrations of PAHs (especially high-molecular weight PAHs [HPAHs]) was most striking (Figure 1). The patterns seen in multiple chemical groups along the shelf indicate that the highest concentrations occurred near the North Trunk Outfall. PAH contamination appears to be widespread along the shelf (Figure 1).

We also looked at how concentrations of PAHs have changed over time near the EBO between 1996 (before it was operational) and 2019 (after the recent overflows), based on a comparison of concentrations from sites within 250 ft of the EBO. While the overflow event and other occasional discharges from the outfall do not appear to have resulted in increased PAH concentrations in the area, there does appear to be persistent contamination along West Point's north shelf. Concentrations have decreased slightly over time, but our study indicates low rates of natural recovery in the study area.

Compositing sediment from multiple grab samples at a station. Photo: Wendy Eash-Loucks

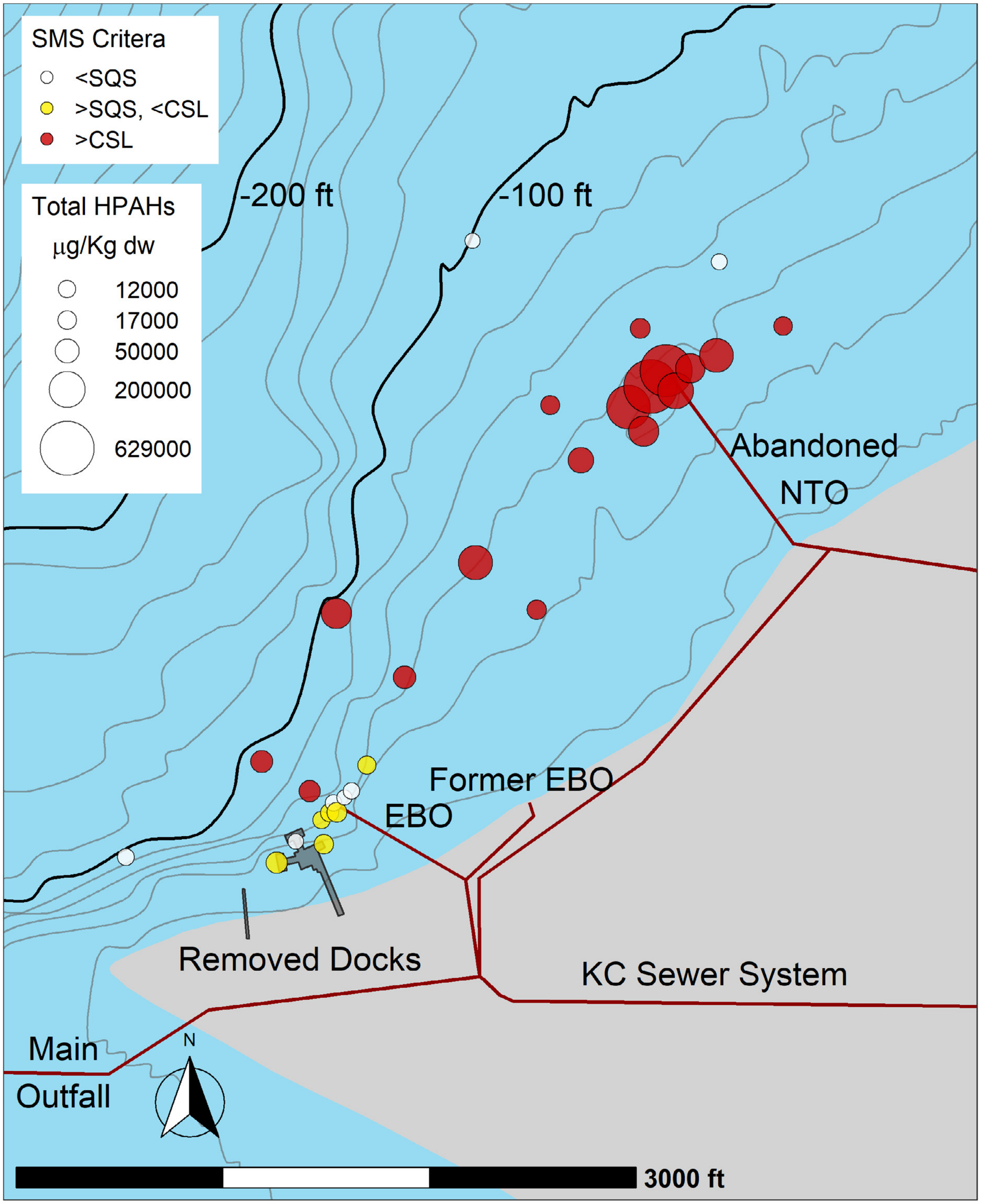

Figure 1. Total HPAH concentrations (µg/kg dry weight) measured at each West Point shelf station. Symbol size is based on concentration on a continuous scale. Color is based on a comparison to Sediment Management Standards criteria. EBO = Emergency Bypass Outfall; NTO = North Trunk Outfall; SQS = sediment quality standards; CSL = cleanup screening level.

Lower Duwamish Waterway Baseline Sediment Results Following the Completion of Early Action Area Cleanups Compared to Historical Data and Sediment Transport Model Predictions

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- **Composite sampling was efficient and cost-effective in establishing LDW baseline conditions to assess trends in long-term monitoring post-remedy.**
- **Following early actions, source control, and natural recovery, significant, site-wide reductions in human health risk driver spatially weighted average concentrations were observed.**

The Lower Duwamish Waterway (LDW) is a 5-mile, 441-acre estuarine waterway that was listed as a Superfund site in 2001. The Record of Decision (ROD) for the LDW was released by the Environmental Protection Agency in 2014 and early action area (EAA) sediment cleanups were completed by 2016. Source control actions throughout the site and its drainage basins have been ongoing since the LDW was listed. Baseline sampling was conducted in 2018 (prior to the full sediment remedy) to serve as a foundation for assessing trends in site-wide sediment concentrations following early action cleanups.

Surface sediment composite samples (0–10 cm) were collected in 2018 to establish baseline spatially weighted average concentrations (SWACs) for the following human health risk driver chemicals: polychlorinated biphenyls (PCBs) (Figure 1), carcinogenic polycyclic aromatic hydrocarbons (cPAHs), dioxins/furans, and arsenic. The statistically derived sampling plan described the creation and analysis of composite samples from 24 equally sized areas with 7 grab samples each. The SWAC was calculated as the average of the composite results.

A hydrodynamic model (Windward and QEA 2008), a sediment transport model (QEA 2008), and a bed composition model (AECOM 2012) were used to predict sediment dynamics and chemical concentrations in LDW surface sediment following sediment remedial actions. The baseline SWACs were compared to the predicted sediment SWACs immediately after early action area remediation (year 0) and five years after early action area remediation (year 5).

The 2018 baseline data confirmed significant reductions in the site-wide SWACs of LDW human health risk drivers following early action area remediation (Figure 2). The baseline SWACs for PCBs and arsenic were consistent with predicted reductions following early actions, source control, and natural recovery, whereas the SWAC reductions for cPAHs and dioxins/furans were greater than predicted.

The baseline sampling design will be used for future monitoring to assess changes in the site-wide sediment concentrations following the river-wide sediment remedy outlined in ROD.

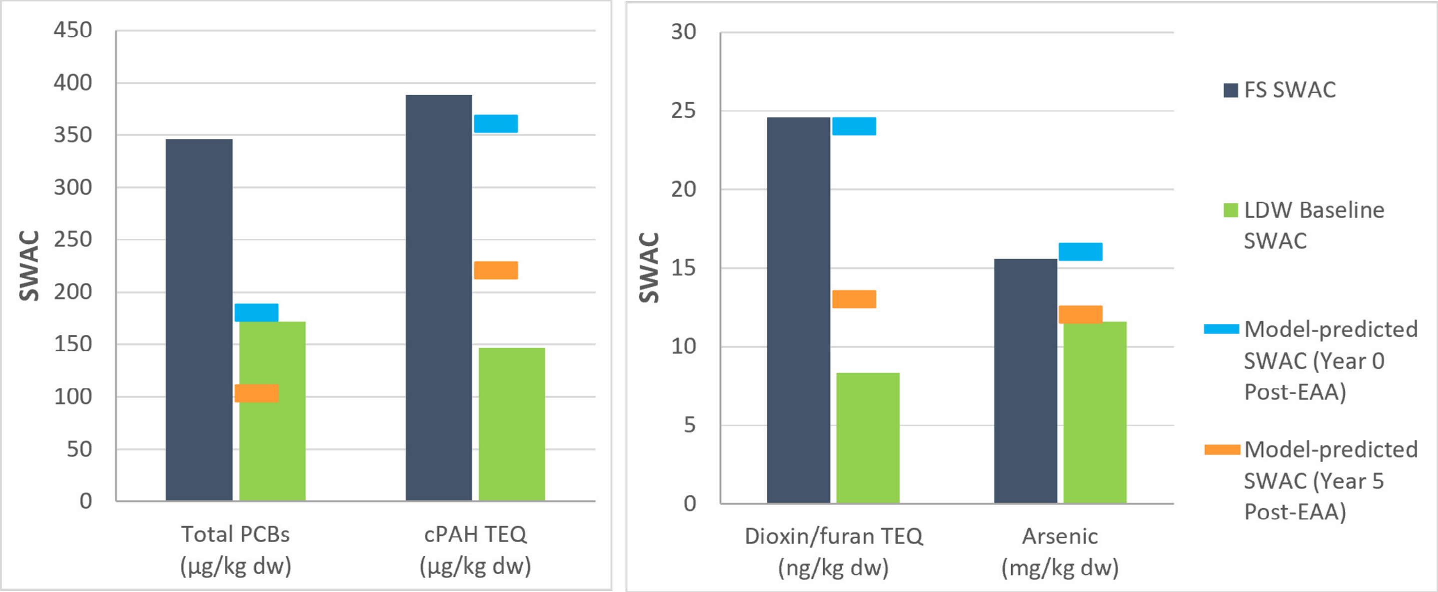

Figure 1. Baseline surface sediment SWAC (2018) relative to Feasibility Study SWAC (2012) prior to completion of EAA cleanups and post-early action model predictions.

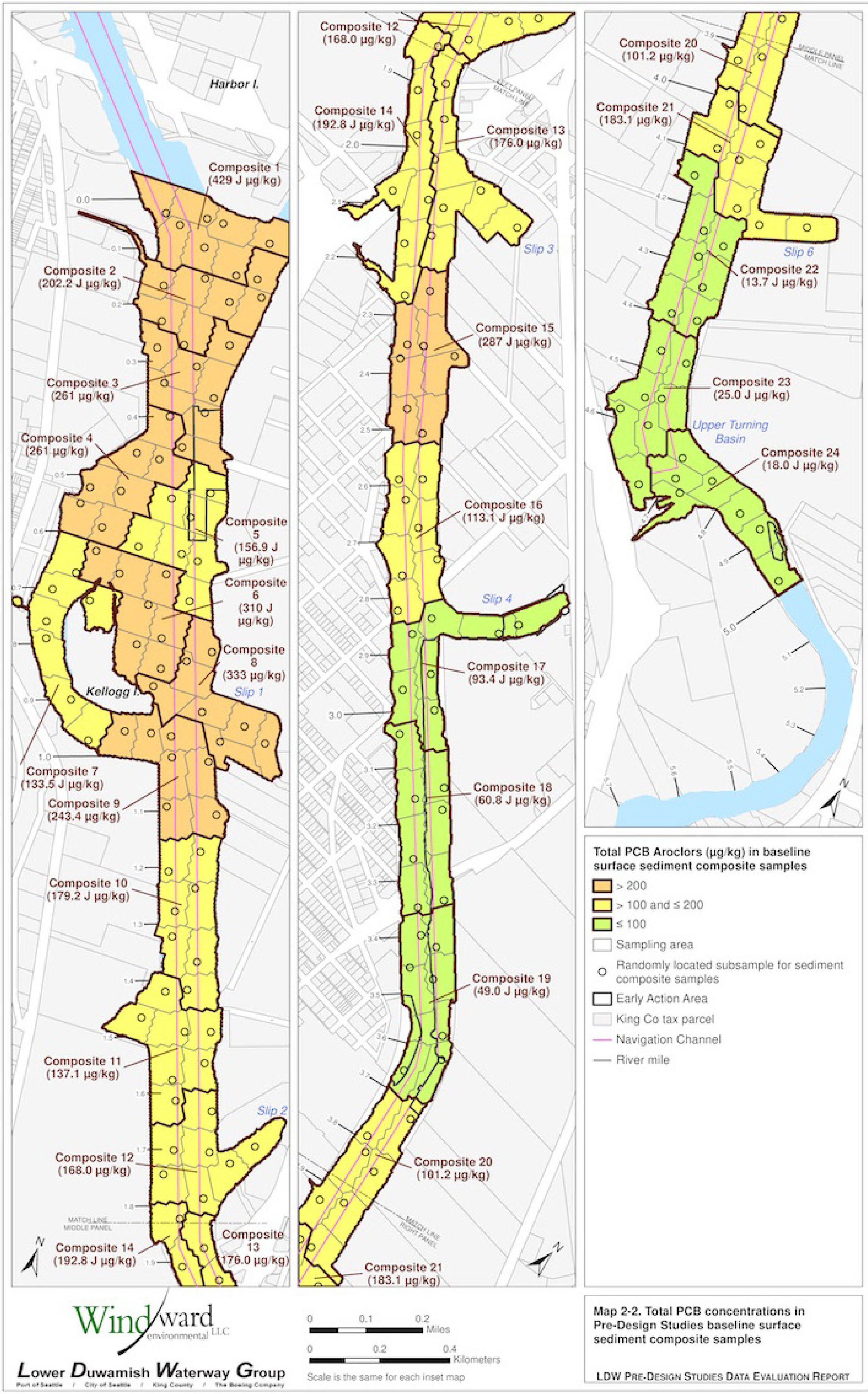

Figure 1. Total PCB concentrations in composite samples analyzed in 2018 to establish baseline conditions prior to full sediment remedy.

Sediment Sampling to Support the Remedy Design of the Upper Reach of the Lower Duwamish Waterway

Kathy Godtfredsen[1], Susan McGroddy[1], Suzanne Replinger[1], Craig Hanson[1], Tom Wang[2], John LaPlante2, Katy Gross[2], Todd Thornburg[2], Jeffrey Stern[3], Debra Williston[3], David Schuchardt[4], Allison Crowley[4], Pete Rude[4], Joe Flaherty[5], Joanna Florer[6]
1. Windward Environmental LLC, 2. Anchor QEA, 3. King County, 4. City of Seattle, 5. The Boeing Company, 6. Port of Seattle
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- **Pre-design sediment sampling occurred in the LDW upper reach in 2020 and 2021, resulting in 14.6 acres needing remedial action based on 30% remedial design.**
- **Areas with remedial action level exceedances will be remediated via dredging, partial dredging and capping, or enhanced natural recovery likely starting fall 2024, after 100% remedial design.**

The Lower Duwamish Waterway (LDW), located in Seattle, Washington, is a 5-mile estuarine site that was listed as a Superfund site in 2001 and as a Washington State Model Toxics Control Act site in 2002. The Record of Decision (ROD) detailing the cleanup plan was issued by the Environmental Cleanup Agency (EPA) in late 2014, and early action sediment cleanups were completed by 2016. EPA has divided the LDW into three reaches for remedial design and cleanup actions. In 2020 and 2021, the Lower Duwamish Waterway Group conducted two phases of pre-design investigation sampling in the 2-mile upper reach (river miles 3.0 to 5.0) to refine the cleanup areas based on sediment remedial action levels (RALs) in the ROD.

In total, the two phases of pre-design investigation sampling comprised 260 surface sediment (0–10 cm) locations, 147 intertidal subsurface sediment (0–45 cm) locations, 135 subtidal subsurface sediment (0–60 cm) locations, and 42 shoaling locations in the Federal Navigation Channel to determine where contaminants exceed RALs. In addition, 88 vertical extent core (variable depth) locations were sampled to assess the depth of contamination within the areas where RALs were exceeded.

Using both existing data collected before 2020 and the pre-design investigation sampling data collected in 2020/2021 and two interpolation methods (indicator kriging for polychlorinated biphenyls and Thiessen polygons for other contaminants), a total of 7.4 acres of the 113 acres that compose the upper reach area (excluding the already remediated early action areas) were designated as RAL exceedance areas. These areas formed the basis for remedial action areas (RAAs), which total 14.6 acres once engineering considerations were addressed in 30% remedial design (Figure 1). The RAAs will be dredged, partially dredged and capped, or addressed through enhanced natural recovery (ENR), depending on requirements defined in the ROD.

A third phase of pre-design investigation sampling was conducted in December 2022 to address remaining data gaps; the results will be incorporated into 90% remedial design. Sediment remedial action (i.e., constructed cleanup) is anticipated to begin in fall 2024, with monitoring to be conducted during remediation, following remediation, and periodically thereafter to track compliance with cleanup goals.

Pre-design investigation sediment sampling within the middle reach (river miles 1.6 to 3.0) is being conducted in winter 2022/spring 2023, with completion of remedial design in 2026 and remedial action anticipated to begin in 2027. The lower reach (river miles 0.0 to 1.6) will be the last reach to undergo remedial design and action.

Processing a sediment core to determine depth of contamination in the upper reach of the Lower Duwamish Waterway. Photo: Brandi Quinlisk, Windward Environmental LLC

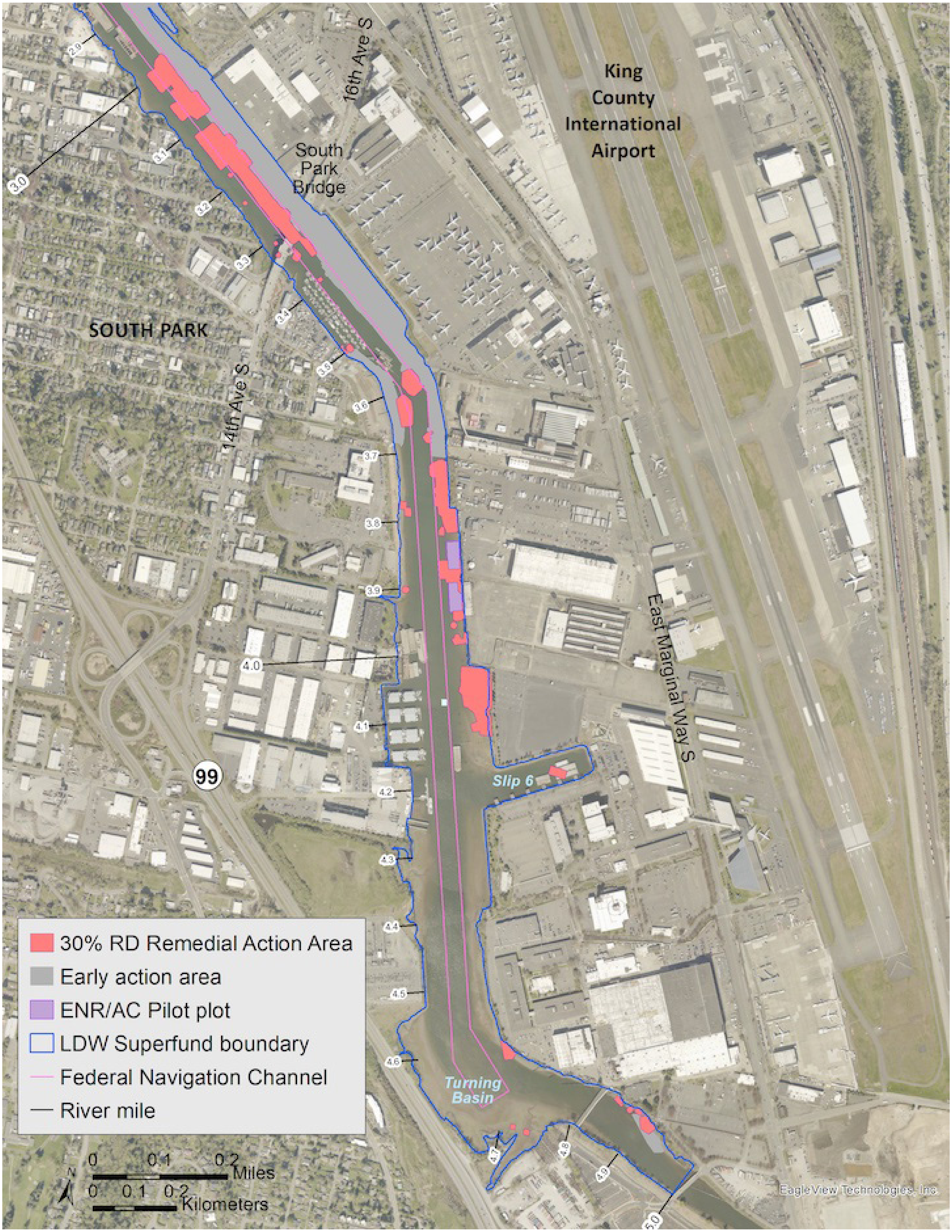

Figure 1. 30% design remedial action areas in the upper reach of the Lower Duwamish Waterway in Seattle WA.

SECTION 2: RESEARCH

Rosario Strait, North Puget Sound. Photo: William Hobbs

Using a Hierarchical Bayesian Model to Predict Stormwater Pollutant Reduction of Four Bioretention Mixes

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- **Combining realistic stormwater concentrations with pollutant removal trials results in credible bioretention media pollutant outflow concentrations.**
- **Bioretention mixes with lower compost content are more likely to reduce nutrient concentrations and meet high-performance goals.**

Four bioretention mixes have been evaluated for pollutant reduction at the Washington State University Puyallup campus. Bioretention mixes were distinguished by their compost content as Mix15 (60% sand, 15% compost, 10% water treatment residuals,5% shredded bark), Mix20 (80%, 20%, 0%, 0%), Mix30 (60%, 30%, 10%, 0%), and Mix40 (60%, 40%, 10%, 5%). Pollutants of interest were chemical oxygen demand, ammonia, nitrite, nitrate, total Kjeldahl nitrogen, and total and dissolved forms of phosphorus, copper, lead, and zinc. Water quality data were collected for eleven storms and cumulative rainfall after construction was used to measure the media age.

Sampling of effluent water quality using replicated bioretention mesocosms using automated water samplers. Photo: Kimberlie Gridley

Pollutant reduction was assessed using a hierarchical Bayesian model combined with data from stormwater outfall samples collected by Phase 1 municipal stormwater permittees between 2007 and 2013 (Hobbs et al., 2015). The Phase 1 permittee data allowed predicting pollutant removal for stormwater concentrations typical for Western Washington State. We showed that this Bayesian modeling approach has advantages over computing bootstrapping confidence intervals typically done on similar water quality data sets. Bootstrapping typically only uses data collected during a study; therefore, the results and outcome of the study are entirely dependent on the suite of observed stormwater concentrations and the experimental design. In our work, pollutant reduction predictions were based on the voluminous and representative Phase 1 permittee data, making this method more objective than a simple bootstrapping approach.

The data were evaluated for two probabilities: 1) the probability of observing a simple pollutant reduction; 2) the probability of a high-performance reduction (HPR), where HPR was defined as a minimum pollutant reduction of 60%. The chemical oxygen demand had a probability of reduction of P = 0.09 and HPR of P = 0.00, resulting in pollutant export (an increase of chemical oxygen demand in the effluent) in most storms. HPR reduction goals were met for total lead, zinc, and copper, and reduction probabilities were generally greater than 0.5 for all total and dissolved metals. Age and media composition affected phosphorus and nitrogen removal, with increased pollutant retention observed as the media aged. Lower compost percentages resulted in less nutrient export.

Mesocosm with vegetation; PVC pipe is the stormwater inlet. Photo: Carly Thompson

In terms of a specific example, a young bioretention mix has only a low chance of total phosphorus removal ($P < 0.08$). However, this probability increases to $P = 0.71$, $P = 0.61$, $P = 0.44$, and $P = 0.58$ as the media ages for Mix15, Mix20, Mix30, and Mix40, respectively. High-performance goals (minimum reduction of 60%) are possible in the long term, specifically after receiving cumulative precipitation greater than 3,000 mm (1181 in) for Mix15 and Mix20. However, Mix30 and Mix40 failed this performance goal even in the long term.

This probabilistic model of realistic stormwater concentrations in combination with bioretention pollutant removal experiments results in a holistic view of credible pollutant outflow concentrations that stormwater managers can use to alleviate the effects of stormwater on surface water quality.

This research was funded by the Puget Sound National Estuary Program Watershed Grant Program grant number G1200452 and was recently published (Knappenberger et al., 2022).

Biochar and Fungi as Bioretention Amendments for PAH Removal from Stormwater

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Website: https://www.wastormwatercenter.org/project/pah-bacteria-project

- **PAHs were removed from stormwater equally well across treatments.**
- **PAHs in bioretention media declined over time despite inputs from stormwater.**

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous organic contaminants comprising multiple fused benzene rings. They are present in petroleum and petroleum-based products and are generated by the incomplete combustion of fossil fuels or biomass. Vehicles release PAHs through exhaust emissions, fluid leaks, and tire wear - stormwater is a significant vector of PAH pollution to aquatic environments (Hwang et al., 2019). Some impervious surfaces, like coal tar seal-coated pavements, can also release PAHs into stormwater at elevated levels (McIntyre et al., 2016). Several PAHs are carcinogenic, and many are toxic to humans (Guo et al., 2011) and aquatic organisms (McIntyre et al., 2016). Because of their toxicity and ubiquity in stormwater, PAHs are essential targets for stormwater treatment using Green Stormwater Infrastructure (GSI). In this study, we evaluated novel bioretention amendments, biochar, and fungi, for their impact on PAH removal and accumulation in bioretention media.

Biochar and fungi possess properties that offer promise as bioretention amendments for PAH treatment. Because biochar is a porous substance, like charcoal, generated via pyrolysis of organic wastes, it contains an enormous internal surface area that facilitates the adsorption of organic contaminants (Mohanty et al., 2018). In addition, ligninolytic fungi, which degrade woody biomass, excrete enzymes capable of initiating the breakdown of high molecular weight PAHs, which may otherwise resist degradation by bacteria (Haritash & Kaushik, 2009). We compared PAH treatment performance of Washington's standard 60:40 bioretention mix of 60% sand: 40% compost (by volume) and three other mixtures. The first amended mixture contained biochar (20% biochar, 20% compost, 60% sand), the second contained fungi (40% compost and 60% sand inoculated with *Stropharia rugosoannulata*), and the third contained biochar and fungi (20% biochar, 20% compost, and 60% sand inoculated with *S. rugosoannulata*). All mesocosms were planted with *Carex oshimensis*. Mesocosms were dosed with highway runoff during eight simulated storm events over two years.

Experimental bioretention columns located in a greenhouse at the WSU Puyallup Research & Extension Center. Images on right show fruiting bodies (top) and mycelial growth (bottom) of *Stropharia rugosoannulata.*

PAH concentrations in the highway runoff (influent) ranged from 0.09-5.08 µg/L (mean = 1.21 µg/L) (Figure 1A). Every mix removed PAH from highway runoff at high rates (>97% for all but 2 samples) - no significant difference was detected in % removal rates across treatments. Bioretention soils amended with biochar had lower initial (day 0) soil PAH concentrations than those in the standard 60:40 mix. PAH concentrations in initial media samples scaled with % compost volume, suggesting that compost was the primary source of media PAHs. The concentrations of PAHs in soil decreased throughout the study despite the repeated

addition of PAHs via influent stormwater (Figure 1B). Our results suggest that microbial bioremediation or plant uptake of PAHs is the primary mechanism for PAH removal in bioretention mixes. Greater losses of media PAHs (69-79%) were observed in the fungi amended bioretention mix compared with the standard mix (54-70%); however, further replication and a longer study period are needed to determine if fungi improve PAH bioremediation in bioretention soils. Sand and compost mixtures are sufficient for mitigating PAH concentrations in stormwater. A partial replacement of compost with biochar seems to provide similar, if not improved, treatment. A partial replacement of compost with biochar could be beneficial for reducing pollutant export, especially for bioretention mixes with a high compost fraction.

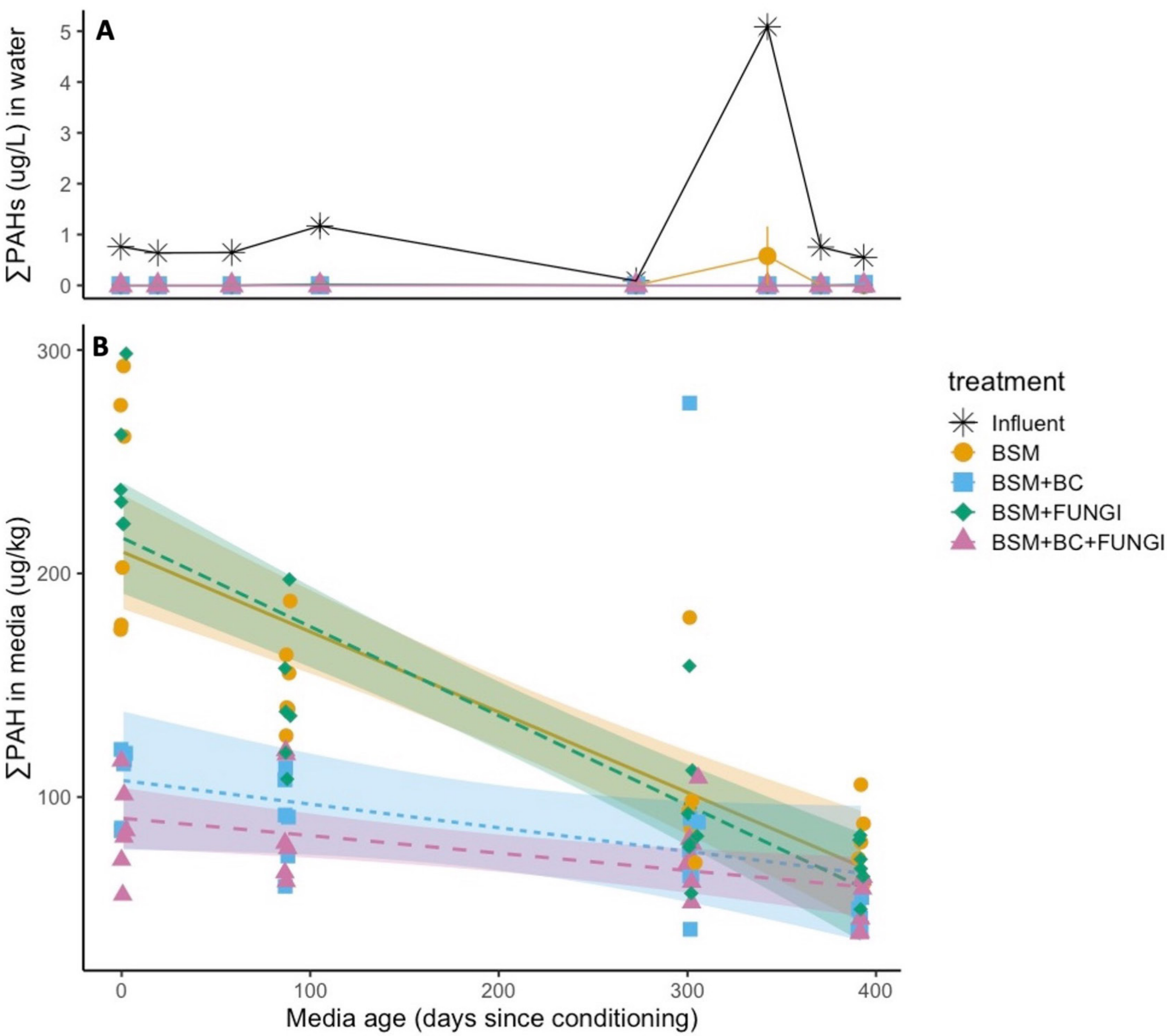

Figure 1 (A) Mean ± SE concentrations of PAHs in influent (solid black line) and effluent samples by treatment (colored lines) for the 8 dosing events in the study. (B) Concentrations of PAH (μg/kg) in bioretention media samples by time since conditioning the columns by flushing them with clean water to release leachable contaminants. BSM = Bioretention soil media (60% sand: 40% compost); BSM + BC = 60% sand, 20% compost, 20% biochar; BSM + Fungi = 60% sand and 40% compost inoculated with *S. rugosoannulata*; BSM + BC + Fungi = 60% sand, 20% compost, and 20% biochar innoculated with *S. rugosoannulata*.

Investigating a Trophic Connection between a Putative Local PCB Source and Broader Contamination of Puget Sound's Pelagic Food Web

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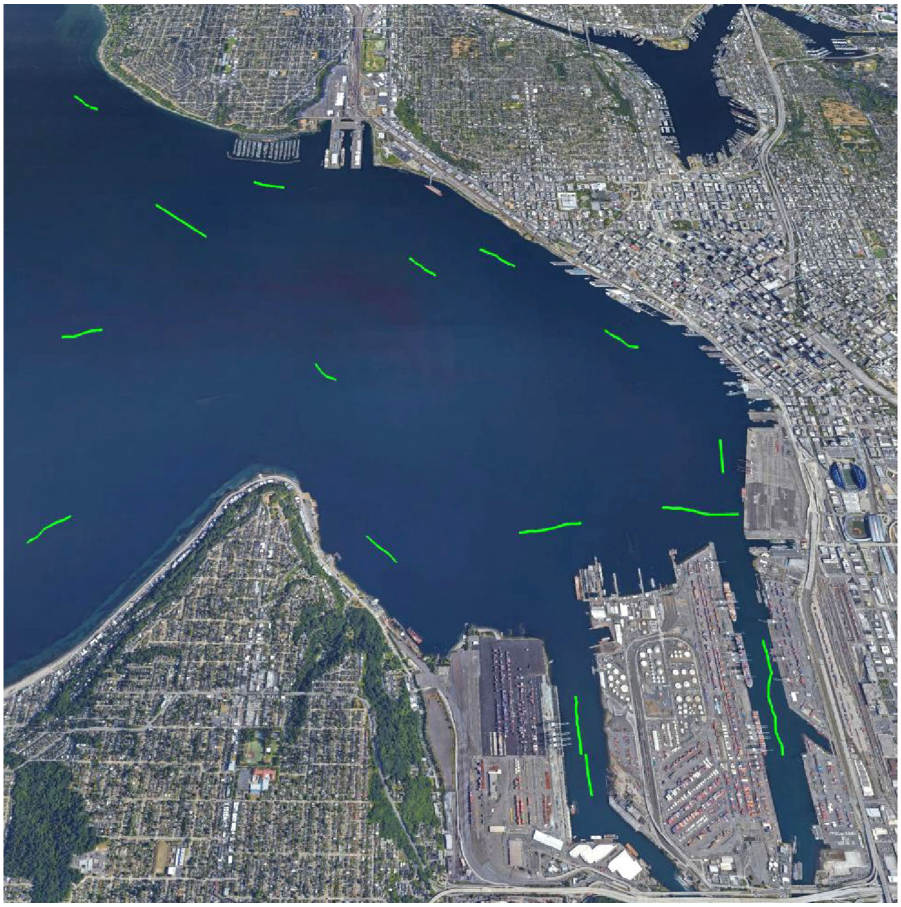
- **PCBs in particulate organic matter exhibited a strongly increasing gradient from outer Elliott Bay to the Lower Duwamish Waterway.**
- **Results suggest an entry point of PCBs into Puget Sound's pelagic food web from phytoplankton to primary consumers such as krill.**

The pelagic food web is widely contaminated with (polychlorinated biphenyls) PCBs in Puget Sound's southern and central basins, as evidenced by high PCB concentrations in Pacific herring (*Clupea palassii*, West et al., 2008), their predators (Chinook salmon, *Oncorhynchus tshawytscha*, O'Neill and West 2009; harbor seals *Phoca vitulina*, Ross et al., 2004) and apex predators feeding in these basins (Southern Resident Killer Whales *Orcinus orca*, Cullon et al., 2009). Biomagnification of PCBs in these pelagic predators prompted investigations into PCB distribution in lower trophic levels -- primary producers (e.g., diatoms), and primary consumers (e.g., the North Pacific krill, *Euphausia pacifica*; West et al., 2011) to evaluate where PCBs are entering the pelagic food web (*sensu* Frouin et al., 2013). The following describes recent efforts to evaluate the distribution of PCB concentrations in one of the lowest trophic levels of the pelagic food web, phytoplankton, relative to a putative PCB source, the Lower Duwamish Waterway (LDW).

The LDW and its surrounding watershed are highly contaminated with PCBs; it is designated by U.S. Environmental Protection Agency (EPA) as a PCB Superfund Site under the U.S. Comprehensive Environmental Response, Compensation and Liability Act. This study examines the possible link between this local putative source of PCBs to broader geographic



Sampling locations for particulate organic matter (POM—green lines on map, created in Google Earth Pro).

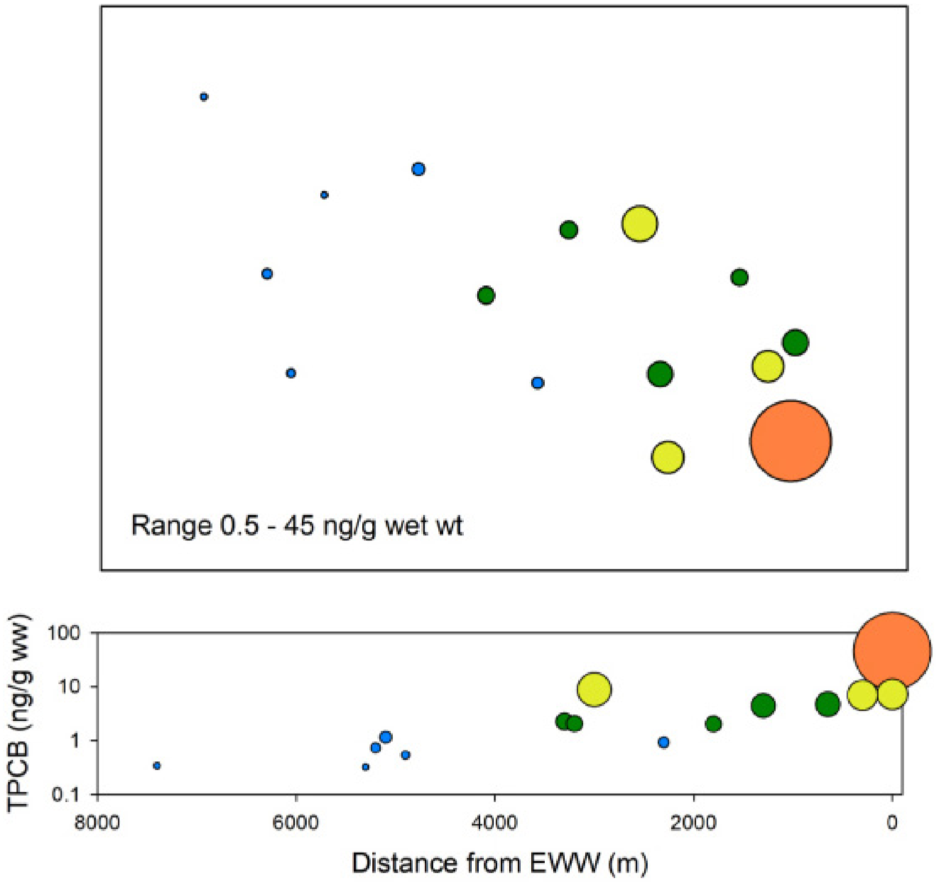


Figure 1. Relative concentration of total PCBs in POM (top); and total PCB concentration in POM plotted against distance from the LDW East Waterway, a putative PCB source (bottom).

contamination in the ecosystem. We targeted diatoms in Puget Sound waters during a fall bloom in 2021, in the East and West Waterways of the LDW, and moving outward into Elliott Bay, with the outermost sites approximately 8,000-m from the LDW (figure 1). Although we intended to sample diatoms it was unfeasible to selectively collect a single plankter of such small size in quantities sufficient to analyze for PCBs. We used a Puget Sound plankton net with 20-µm Nitex mesh and subsequent sieving to size-select particles we termed "particulate organic matter" (POM). Nets were towed horizontally at each station from the surface to approximately 9-m depth. Although there were several species of diatoms and some heterotrophs in these samples, the dominant plankter was *Coscinodiscus* sp., a relatively large centric diatom.

Overall, PCBs analyzed from 15 POM samples showed a strong declining gradient with distance from the LDW, with the greatest total PCB concentrations observed in the East Waterway of the LDW (45 ng/g ww), followed by the West Waterway of the LDW and two locations along the Seattle waterfront. POM from outer Elliott Bay exhibited total PCB concentrations as low as 0.5 ng/g ww (figure 1). This PCB gradient was reflected in krill as well (results to be reported elsewhere), illustrating a trophic linkage to the pelagic food web. These early results provide data to document the transfer of PCBs from the water column to POM, and into the pelagic food web – a key step towards developing a predictive hydrodynamic fate and transport model for PCBs in the Salish Sea Model (Khangaonkar, 2012; 2018). These results have also motivated additional investigations, including a recent (Sept 2022) Puget Sound-wide POM and krill survey to compare the gradient observed in Elliott Bay with other putative PCB sources in Puget Sound.

Maternal Transfer of PCBs in Pacific Sand Lance

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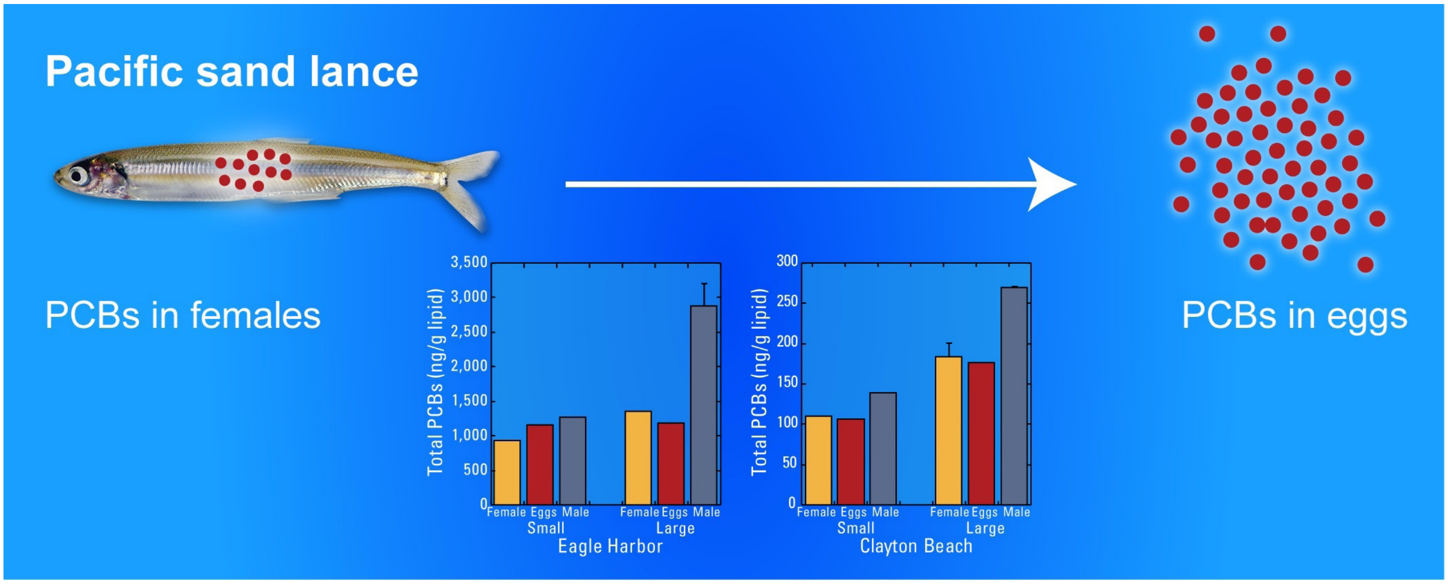
- **Egg/female PCB concentration ratios were close to 1, confirming maternal transfer.**
- **Young of year juveniles were contaminated before exposure to exogenous sources of PCBs through feeding.**
- **Male sand lance in two size classes had higher PCBs than comparable females.**

Forage fish are small, schooling planktivores that form a critical link in marine food webs by transferring energy from plankton up to larger fishes, birds, and marine mammals. In Puget Sound, the forage fish guild includes Pacific sand lance (Ammodytes personatus) which depend on nearshore habitats. Spawning occurs November to February on upper intertidal beach areas and eggs hatch into planktonic larvae that reside in the nearshore during late winter (Penttila, 2007). Juveniles occupy nearshore habitats throughout the summer and fall (Penttila, 2007). Sand lance bury in bottom sediments to avoid predators and conserve energy because they lack a swim bladder (Winslade, 1974; Pearson et al., 1984). Their burying behaviors are not well documented, but it is assumed that they remain buried during the winter as part of an overwintering stage (Quinn, 1999).

Our study used congener specific analyses to evaluate the maternal transfer pathway for polychlorinated biphenyls (PCBs) in sand lance at two sites in Puget Sound. PCBs are toxic, bioaccumulative, and biomagnify up the food chain. Specifically, we (1) selected two sites from the reconnaissance survey of Conn et al. (2020) with known differences in the PCB levels of adult sand lance, (2) evaluated PCB levels in young-of the year (YOY), for which no data existed, and (3) defined two size/age classes of adult sand lance and replicated them at each study site to compare PCB concentrations in males, females and eggs.

We collected sand lance at an urban embayment (Eagle Harbor) and a state park along an open shoreline (Clayton Beach) during spring and fall. Similar to Conn et al. (2020), we found lipid-normalized concentrations of PCBs in sand lance at Eagle Harbor were 5–11 times higher than PCB concentrations in comparable samples at Clayton Beach. This was true for every life stage and size class of sand lance, including eggs removed from females. The same trend was observed in environmental samples. In Eagle Harbor, PCB concentrations in unfiltered water (0.19 ng/L), sieved (<63 μm) nearshore bed sediments (0.78 ng/g dw) and suspended particulate matter (1.69 ng/g dw) were 2–3 times higher than equivalent samples from Clayton Beach.



Graphical abstract of the study, depicting the transfer of PCBs to eggs within females to PCBs in eggs in the environment along with figures of the total PCBs from fish at Eagle Harbor and Clayton Beach. The figures are total PCBs (ng/g lipid) in composites of whole-body Pacific sand lance tissue (males or females) of two size classes, or eggs that had been removed from collected small and large females. Yellow bars are females, red bars are eggs removed from females and gray bars are males. Note the approximate 10-fold-higher y-axis scale on Eagle Harbor graph compared to the Clayton Beach graph.

Pacific sand lance captured in a beach seine. Photo: David Ayers

Sand lance collected in the fall (buried in sediment during presumed winter dormancy) had lower lipid content and up to four times higher PCB concentrations than comparably sized fish collected in the water column by beach seine during the spring. Lipid content was 5–8% in spring fish and was reduced in fall fish (1–3%). Male sand lance had higher PCB concentrations than comparable females. All egg samples contained PCBs, and the lipid normalized egg/female concentration ratios were close to 1 (0.87–0.96), confirming that maternal transfer of PCBs occurred, resulting in sand lance eggs and early life stages being contaminated with PCBs even before they were exposed to exogenous sources. These early life stages are prey for an even wider range of species than adult sand lance, creating additional exposure pathways for biota and increasing the challenges for mitigation of PCBs in the food web.

Highly Contaminated River Otters (*Lontra canadensis*) Are Effective Biomonitors of Environmental Pollutant Exposure

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- **Toxics in otter scat reflect regional conditions, making otters excellent biomonitors that should be included in evaluating the Lower Duwamish Waterway cleanup.**
- **River otter scat from the Lower Duwamish Waterway showed evidence of all toxics measured, and PCB and PAH levels reported are among the highest published.**

River otters (*Lontra canadensis*) are apex predators that play important roles in aquatic ecosystems (e.g., Ben-David et al., 1998; Cote et al., 2008; Larsen, 1984; Roemer et al., 2009). They prey primarily on fish and crustaceans but are opportunistic predators with a diet ranging from insects to birds and mammals (see Boyle, 2006). They are vulnerable to biomagnification of persistent pollutants, and the correlation between polychlorinated biphenyl (PCB) levels found in otter scat to those found in liver tissue has been established through modeling and empirical studies (see Guertin et al., 2010). Given this toxicological profile and their relatively localized home ranges (Blundell, 2000; Bowyer et al., 1995), river otters are considered biomonitors of wildlife exposure to toxics and environmental health (Carpenter et al., 2014; Guertin et al., 2010).

Game camera photo of river otters at a latrine site along the Green-Duwamish River, King County, WA. Photo: Michelle Wainstein

River otters reside throughout the Green-Duwamish River, WA (USA), a watershed encompassing an extreme urbanization gradient, including a U.S. Superfund site slated for a 17-year remediation (including a post-construction recovery period). The objectives of this study were to document baseline contaminant levels in river otters; assess otters' utility as top trophic-level biomonitors of contaminant exposure; and evaluate the potential for health impacts on this species. To meet these objectives, we measured a suite of contaminants of concern, lipid content, and nitrogen stable isotopes ($\delta^{15}N$) in 69 otter scat samples collected from 12 sites. Landcover characteristics were used to group sampling sites into industrial (Superfund Site), suburban, and rural development zones.

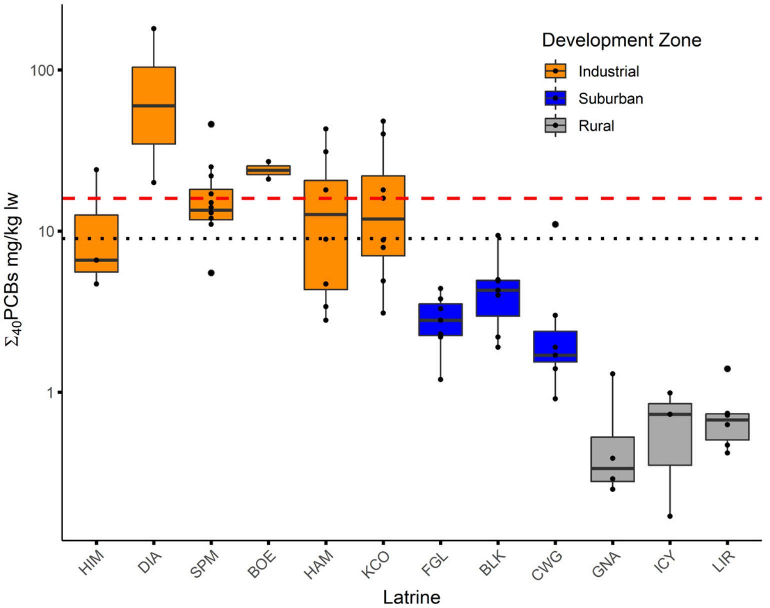

Figure 1. Median concentrations of $\sum_{40}$PCBs (mg/kg lw) in river otter scat collected at each of 12 latrines located in three different development zones along the Green-Duwamish River, Washington, USA as compared to published values for level of concern (9 mg/kg lw; black dotted line) and critical level (16 mg/kg lw; red dashed line).

Concentrations of PCBs, polybrominated diphenyl ether flame-retardants (PBDEs), dichlorodiphenyltrichloroethanes (DDTs) and polycyclic aromatic hydrocarbons (PAHs) increased significantly with increasing urbanization (Figure 1) and were best predicted by models that included development zone, suggesting that river otters are effective biomonitors, as defined in this study. Diet also played an important role, with lipid content, $\delta^{15}N$, or both included in best models. The data are contemporaneous with other baseline studies conducted for the Lower Duwamish Waterway Superfund remediation effort.

River otter scat is recommended for inclusion in evaluating restoration efforts in this Superfund Site, and as a potentially useful monitoring tool wherever otters are found. The ΣPCB and ΣPAH exposures reported are among the highest published for wild river otters, with almost 70% of samples in the Superfund Site exceeding established levels of concern. See full article: https://doi.org/10.1007/s10661-022-10272-9.

Occurrence of Contaminants of Emerging Concern in the Lower Columbia River

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- **Confident detection of 119 chemicals in the Lower Columbia and Willamette River representing a variety of consumer and industrial use products.**
- **Results showed spatial and seasonal differences in chemical occurrence, potentially explained by wastewater and stormwater dominance.**

Contaminants of emerging concern (CECs) are a class containing thousands of compounds that are not well described in terms of distribution, environmental fate and transport, or regulation. CECs include pharmaceuticals and personal care products, hormones or endocrine disrupting compounds, flame retardants, and agrichemicals present in surface and ground water. They generally occur at low levels (µg/L or ng/L) and are unregulated when found in the environment. Importantly, there is growing evidence that they may adversely affect biota. Although direct toxicity and mortality is generally not associated with exposure to CECs, endocrine disruption, reproductive alteration, and behavior modification have all been observed in organisms exposed to CECs, including in studies performed in the region (Lubliner et al. 2010, Meador et al. 2016, WDFW TBiOS 2016, Miller-Schulze et al. 2017). Advances in analytical methods over the last decade have facilitated the investigation and quantification of CECs (Wu et al. 2010, Richardson 2012).

We collected samples covering a 100 river-mile length of the Lower Columbia River during four events in 2021 representing traditionally wet (February, March) and dry (August, September) seasons. These samples were extracted and analyzed using high resolution mass spectrometry (HRMS) with non-target screening. Observed mass spectra were compared against curated spectral libraries using the Global Natural Products Social Molecular Network (GNPS, Wang et al. 2016), resulting in 886 potential chemical matches. Accurate mass, retention time, isotopic ratios, and additional criteria were used to validate 119 chemical matches. These chemicals are representative of a wide variety of consumer and industrial use categories.

Our results show that there are seasonal differences in chemical occurrences (Figure 1). Irbesartan, an antihypertensive beta-blocker drug, was detected in the Willamette River during both wet and dry seasons. It was only detected in the main Columbia River during dry season, downstream of the City of St. Helens wastewater treatment plant effluent zone. Irbesartan has been shown to bioaccumulate in some species of aquatic moss and is suspected to pose a high risk to aquatic species (Sossey Alaoui et al. 2021, Nannou et al. 2022). Overall, the results indicate that organisms in the Columbia River are exposed to a wide variety of CECs. Waterways that flow into the Salish Sea face many of the same pressures as the Columbia River from surrounding land use and urbanization, as well as wastewater and stormwater effluents. These results can help to interpret emerging contaminants from multiple sources in the Salish Sea.

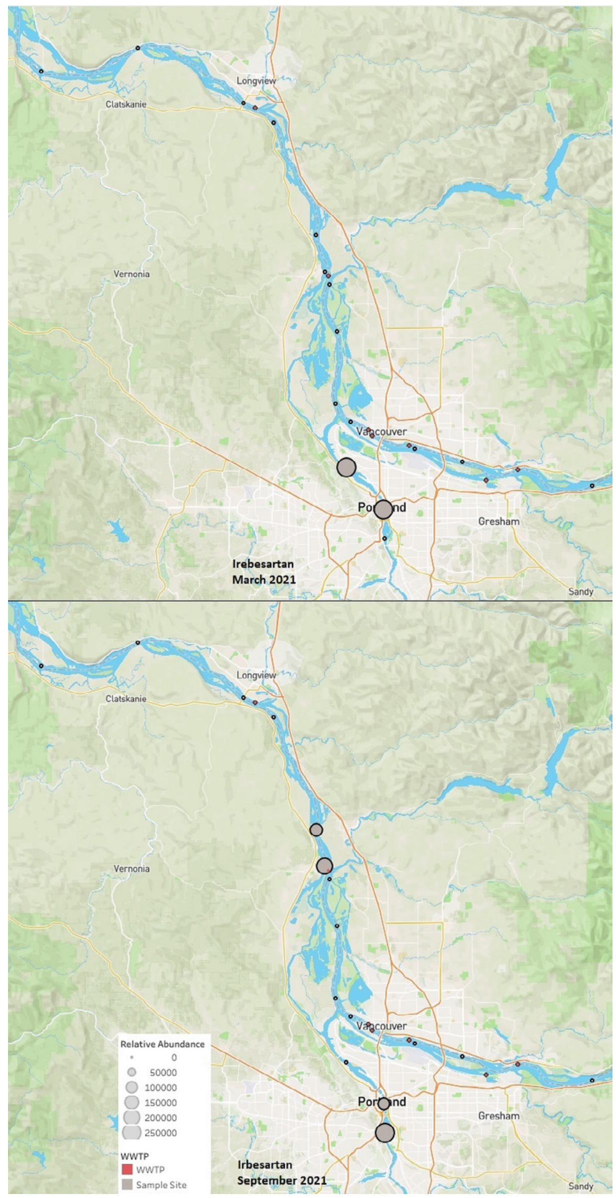

Figure 1. Irbesartan occurrence in the Columbia and Willamette River during March and September 2021. WWTP effluent zones indicated with red diamonds. Sample sites with a "0" result indicate non-detect values (peak height less than 1,000). Relative abundance values are peak area taken from HRMS analysis.

The Initial Screening of Contaminants of Emerging Concern in the Marine Environment Based on Multiple Biological Response Measures

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- **NORMAN data, ToxCast high-throughput in vitro data, and published screening values can be used in screening assessment of CECs to overcome gaps in available ecotoxicology data.**
- **Results from CEC screening and prioritization can inform future environmental management decisions.**

Contaminants of emerging concern (CECs) are chemicals used in daily life, such as pharmaceuticals, personal hygiene products, steroids, plasticizers, and flame retardants. The environmental occurrence and toxicology of CECs are poorly characterized, and they are generally unregulated. Traditionally, risk characterization relies on in vivo methods to test whole organisms for apical endpoints, including survival, reproduction, and growth. This is time-consuming and costly, both financially and in terms of laboratory animal well-being, limiting ecotoxicological data. To overcome this challenge, we are utilizing alternative approaches to perform a screening-level evaluation of CECs present in Puget Sound to identify CECs most likely to elicit a biological response. Considering the number and diverse range of CECs introduced to the environment, it is important to identify and prioritize chemicals most likely to cause environmental harm.

Regional CEC data were compiled from 20 individual studies across multiple environmental matrices, including water, wastewater, and tissue. Over 950 samples were analyzed for a total of 433 unique compounds, 240 of which were detected at least once. Screening was performed with three different methods to provide unique lines of evidence and improve the confidence of prioritization. In each case, the measured concentration was compared to a biological effects threshold; if the concentration was greater than the threshold it suggested the potential for biological impact. For fish and mussel data, tissue concentrations were also converted to marine water concentrations by dividing the measured concentrations by the bioconcentration factor.

The first approach used data from the NORMAN ecotoxicology database as a source of Predicted No Effects Concentrations (PNECs). PNECs are based on in vivo experimental data or modeled (NORMAN Network, 2022). The second approach utilized the ToxCast database to access results from high-throughput in vitro assays, which expose mammalian cell or cell components (rather than whole organisms) to CECs and measure responses. In vitro assays may not measure apical endpoints but do identify events that may lead to an adverse health outcome. The ToxCast database is a source for activity cutoff concentrations (ACC), which are the concentrations at which the exposure level yields a biological response. As the third and final line of evidence, screening values derived by Gefell et al. (2019) for 14 CECs were used. Population-relevant screening values include an SVLOW which is the concentration in the water below which adverse impacts are not anticipated, and an SVHIGH above which adverse impacts are anticipated.

Detected environmental concentrations were compared to the measures of biological response to calculate a biological response ratio (BRR): a Toxicity Quotient for compounds with published PNECs, and an Exposure Activity Ratio, as defined by Corsi et al (2019), for compounds with ACCs, and a Hazard Quotient for chemicals with screening values. BRRs were then evaluated against an effects threshold, with larger BRRs suggesting greater likelihood of biological response. Priority chemicals were identified as those that exceeded the effects threshold by a factor of 100. Watch-list chemicals, low-priority chemicals, and chemicals with insufficient information were also identified. As a result of this phase of the prioritization, 56 chemicals were classified as priority chemicals (figure). These results, along with additional planned prioritization phases are meant to focus further monitoring and research efforts and inform management actions to mitigate the potential impacts of high-priority CECs.

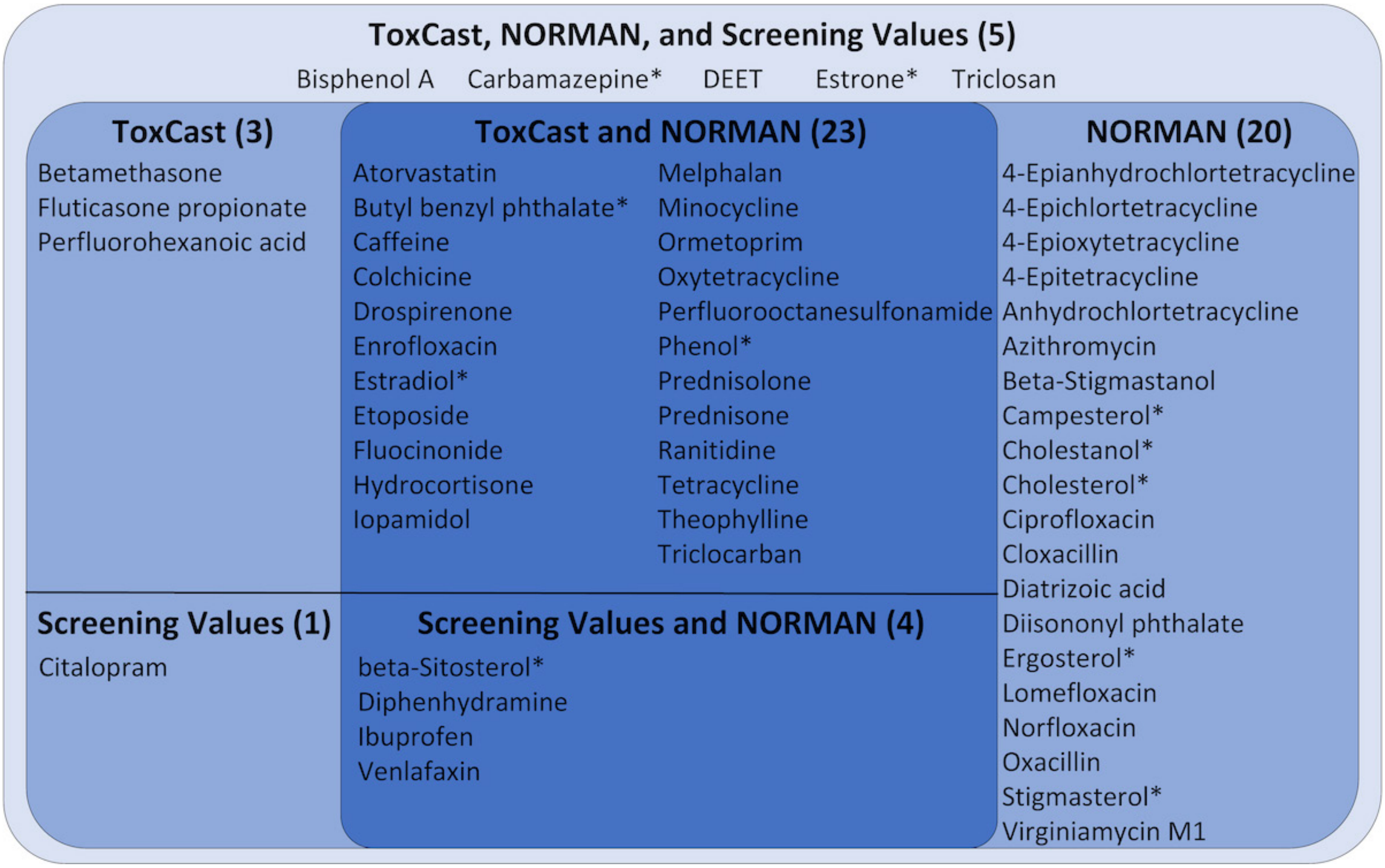

Figure 1. High-priority chemicals were identified based on information from either ToxCast, NORMAN, screening values, or multiple lines of evidence. The number of chemicals represented in each category is shown in parentheses. *indicates that the CEC was included as a high-priority chemical solely due to the measured concentration in wastewater treatment plant effluent samples. Chemical categories include antibiotics (n=19), hormones (n=9), industrial chemicals (n=2), current use pesticides (n=1), per- and poly-fluoroalkyl substances (PFAS) (n=2), pharmaceuticals (n=14), phthalates (n=2), and sterols (n=7). Figure adapted from Alvarez et al., 2021.

Physiological Responses of Chinook Salmon to Wastewater Exposure

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- **Wastewater effluent exposure affects the endocrine system, stress axis, metabolism and brain function in juvenile Chinook salmon.**

Municipal wastewater treatment plants release toxicants into receiving waters, including pharmaceuticals, personal care products, metals, legacy compounds, and industrial compounds. Many of these toxicants can result in adverse physiological effects in aquatic animals, and the impact of mixtures of contaminants in wastewater effluent (WWE) is still poorly understood. Chinook salmon (*Oncorhynchus tshawytscha*) are a culturally important species in the Pacific Northwest and a vital food resource for critically endangered southern resident killer whales (*Orcinus orca*) (Ford & Ellis, 2006; Hanson et al., 2021). Chinook populations have declined drastically (~60% since 1984) across the Pacific Northwestern U.S. in recent decades, and one source of stress is chemical pollution from WWE (Ecology & King County, 2011).

In this study, we investigated the impact of WWE on juvenile Chinook health in a ten-day exposure. Exposure water was secondary-treated King County WWE in five dilutions from 0.1% to 20%. Hundreds of contaminants were measured in whole WWE and samples from several dilutions. At the end of the exposure, we measured endpoints associated with endocrine disruption, brain function, osmoregulation, stress, and metabolism. Additional analyses were conducted but are not detailed in this discussion, including estuarine water sampling, targeted and non-targeted chemistry for water and tissue samples, bioaccumulation modeling, and metabolomics. Exposure to WWE significantly ($\alpha = 0.1$) induced vitellogenesis, indicating endocrine system disruption. We also saw significant reductions in glucose (a stress indicator) and brain Na+/K+-ATPase activity (an enzyme essential for neuronal signaling). Lastly, metabolism was affected as evidenced by altered total protein, cholesterol, and albumin in plasma, a drastic decrease in lipid content, and a significant increase in visible liver anomalies.

We compared contaminant concentrations in exposure water with effects concentrations from the literature for vitellogenin induction and brain Na+/K+-ATPase inhibition. These endpoints were compared because they elicit specific responses when exposed to certain toxicants. For most measured contaminants, concentrations were several orders of magnitude below effect concentrations for vitellogenin induction in the literature. The exception was estrogenic hormones (estrone, 17α-ethinylestradiol, and 17β-estradiol), which were detected at similar concentrations in exposure water as concentrations that induced vitellogenesis in previous studies (Bjerregaard et al., 2008; Thorpe et al., 2003). Contaminant classes known to affect brain Na+/K+-ATPase in fish include biocides, phthalates, and pharmaceuticals such as selective serotonin reuptake inhibitors and calcium channel blockers (Lajeunesse et al., 2011; Ajima et al., 2017; Das & Mukherjee, 2003; Poopal et al., 2017), but these were not detected in water at concentrations that explained brain Na+/K+-ATPase inhibition. Many toxicants in WWE are acting together to cause harmful effects in Chinook. In addition to contaminants affecting vitellogenin and brain Na+/K+-ATPase, many classes of toxicants were detected in water and plasma at concentrations potentially capable of causing adverse physiological responses. Additionally, some of the effects seen in this short-term exposure at higher concentrations of WWE could be similar to chronic exposures at lower concentrations in the field. This research highlights the need for improved wastewater treatment to improve aquatic health and help threatened species such as Puget Sound Chinook salmon.

Fish tissue processing at the WSU-Puyallup research facility. Photo: Jason Berg

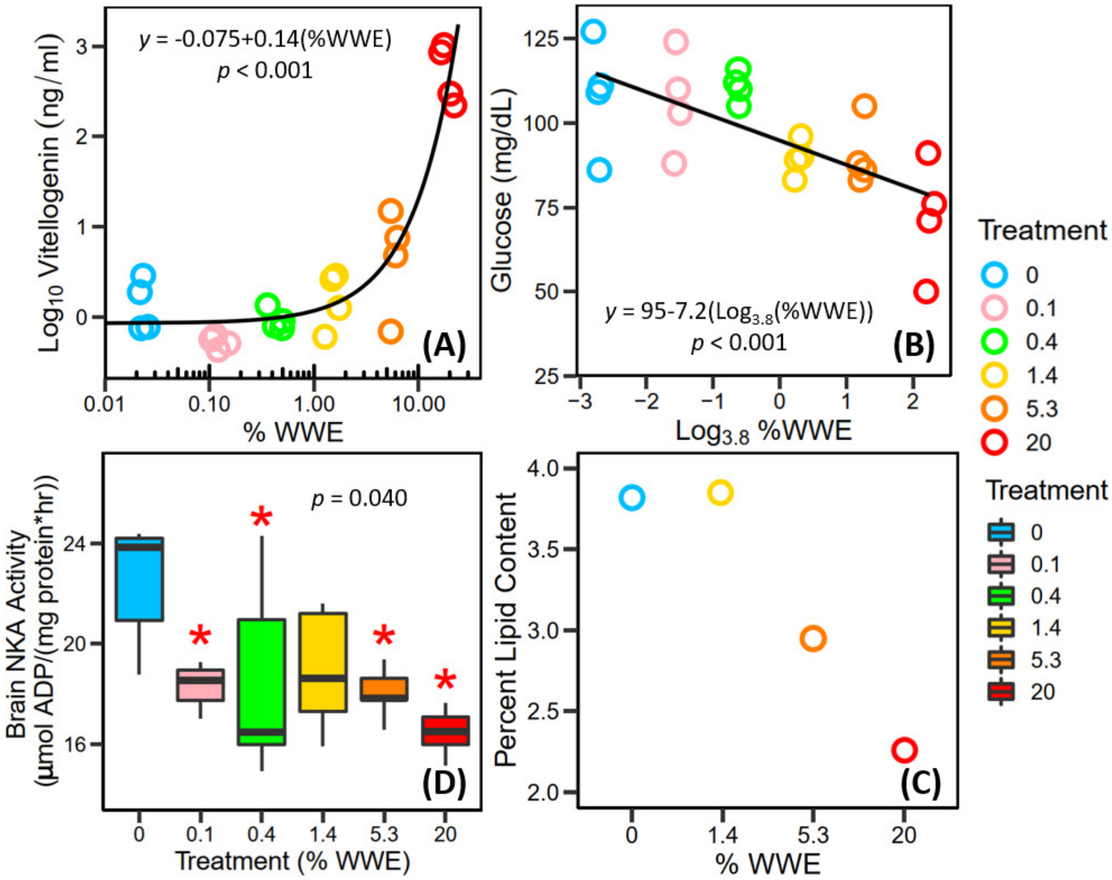

Figure 1. Changes in physiological endpoints compared to WWE treatment. (A) Log of plasma vitellogenin versus % WWE with equation and p-value from simple linear regression; (B) plasma glucose versus log3.8 % WWE with equation and p-value from simple linear regression; (C) whole body percent lipid content versus % WWE; (D) brain Na+/K+-ATPase activity versus % WWE with p-value from Kruskal-Wallis test.

Monitoring Xenoestrogen Exposure and Endocrine Disruption in a Puget Sound Benthic Flatfish, English Sole (*Parophrys vetulus*)

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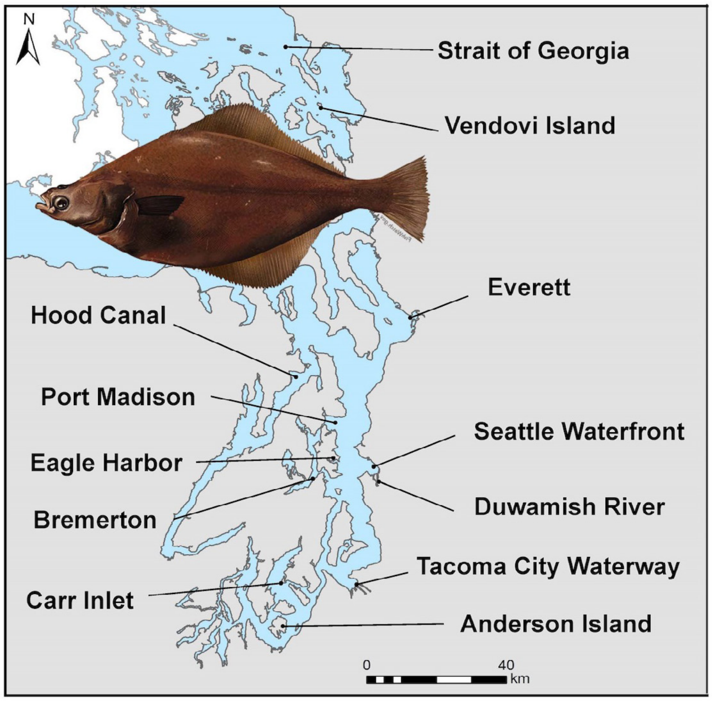
- **English sole are exposed to xenoestrogens throughout much of Puget Sound.**
- **Endocrine disruption from xenoestrogen exposure is likely linked to sewage inputs to Puget Sound.**

Xenoestrogens, including human-derived natural estrogens found in sewage and synthetic chemicals that mimic estrogens such as bisphenol-A, can disrupt endocrine functions and reproduction in exposed fish. Vitellogenin is an egg-yolk protein precursor synthesized in the liver of female fish during their normal reproductive cycle. However, vitellogenin can be abnormally induced in male fish exposed to xenoestrogens, making this protein a suitable and widely used biomarker of xenoestrogen

English sole collected near Anderson Island in south Puget Sound by the Washington Department of Fish and Wildlife Toxics Biological Observation System (TBiOS) team and the crew of the F.V. Chasina as part of their toxics monitoring program. Photo: WDFW TBiOS



Map of 12 English sole monitoring stations throughout Puget Sound, Washington.

exposure and potential endocrine disruption (Sumpter and Jobling, 1993). In the late 1990s, researchers from Washington Department of Fish and Wildlife and National Oceanic and Atmospheric Administration observed evidence of xenoestrogen exposure and endocrine disruption in English sole, including vitellogenin induction in males and altered reproductive timing in females (Johnson et al., 2008). We report here the current geographic extent and severity of xenoestrogen exposure and reproductive effects in English sole, and the xenoestrogens potentially responsible.

Adult English sole were sampled from 12 monitoring locations throughout Puget Sound in May of 2017 and 2019, after the spawning season when English sole have returned to their foraging grounds. Vitellogenin levels were measured in male fish plasma using a commercial multi species vitellogenin ELISA kit (TECO). Gonads were collected for histological analysis to confirm sex and determine reproductive stage. Lastly, bile was collected and analyzed for xenoestrogens including 3 natural steroid estrogens and 5 bisphenols as described in da Silva et al (2013). Estradiol equivalencies for each sample were estimated based on methods described by Vega-Morales et al. (2013).

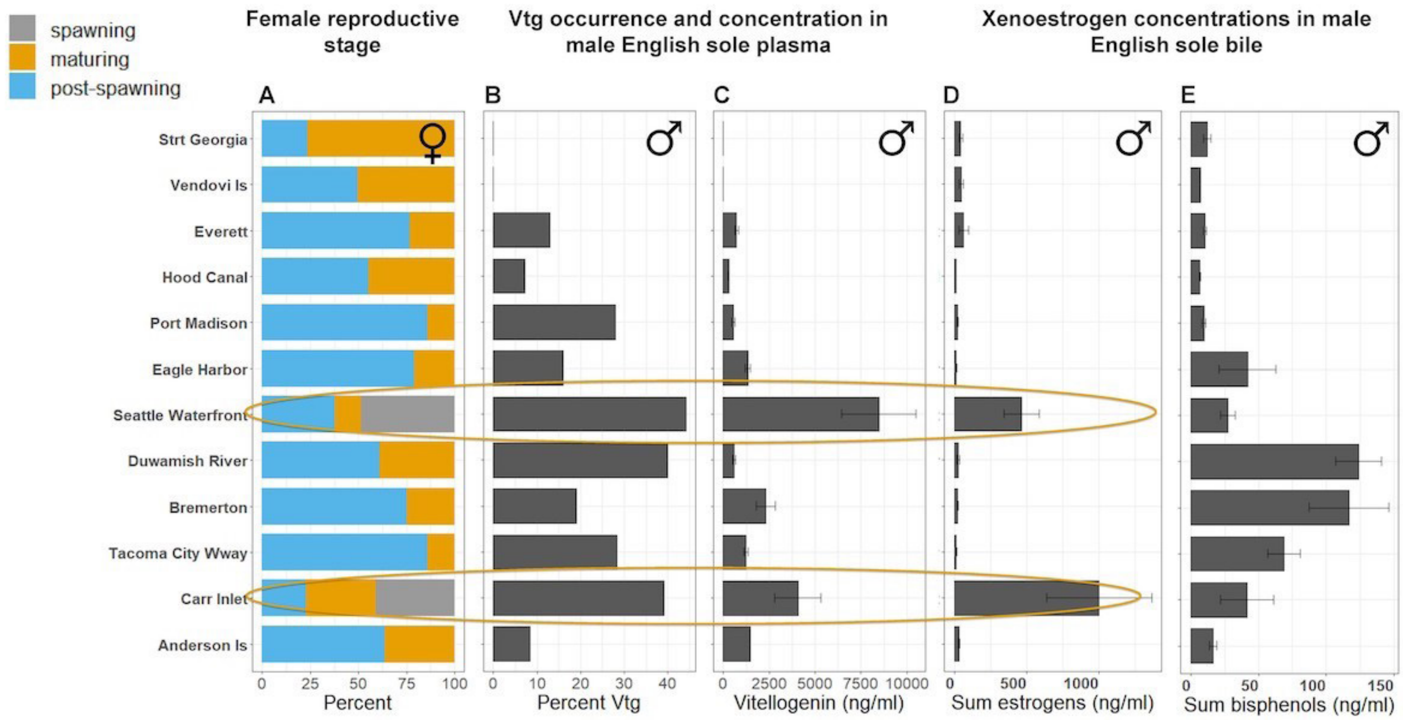


Figure 1. Plots showing lines of evidence of endocrine disruption in English sole resulting from xenoestrogen exposure. A) Percent of female English sole at different stages of reproductive development. B) Percentage of male English sole with detectable levels of vitellogenin (Vtg) in their plasma. C) The mean detected Vtg concentration in male English sole (error bars = standard error). D) The estradiol equivalency (EEQ) of 3 steroid estrogens measured in male English sole bile and E) The sum of 5 bisphenols measured in male English sole bile. English sole image credit: NOAA Fisheries.

Briefly, estradiol equivalencies were calculated as the sum of steroid estrogens measured in male English sole bile multiplied by their estradiol equivalency factor as follows: EEQ = estradiol*1 + estrone*0.11 + estriol*0.11.

Vitellogenin expression in male English sole was observed throughout most of Puget Sound, consistent with previous work by Johnson et al (2010). A high proportion of female English sole were in spawning condition at Seattle Waterfront and Carr Inlet, compared to the other locations. This unusual reproductive timing in females co-occurred with high plasma vitellogenin occurrence and concentrations in males at Seattle Waterfront and Carr Inlet providing good evidence of xenoestrogen exposure and endocrine disruption at these two locations. Male fish collected from Seattle Waterfront and Carr Inlet had high levels of steroid estrogens in their bile and low to moderate levels of bisphenols, suggesting steroid hormones from sewage or other chemicals not measured are a more likely cause of endocrine disruption in English sole than bisphenols. These results suggest endocrine disruption may occur in English sole where sewage enters Puget Sound, from on-site sewage systems (rural) or wastewater treatment plant effluent and combined sewer overflows (urban) – a problem that could increase as population growth continues throughout the region.

The authors thank Kurt Dobszinksy and the crew of the Chasina for fish collection, Denis da Silva for bile xenoestrogen analysis, Mark Myers for histological analysis and Lyndal Johnson and other former WDFW and NOAA staff who contributed to this work.

Toxicological Interpretation and Demonstrated Application of DGT Labile Copper and Zinc Measurements in Marine Waters

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- **The DGT measured labile copper EC50 (µg/L) of *Mytilus galloprovincialis* in marine waters = 1.07(DOC concentration [mg/L]) + 3.38, (R2 = 0.880).**
- **The DGT measured labile zinc EC50 (µg/L) of *Strongylocentrotus purpuratus* in marine waters = 19.9(DOC concentration [mg/L]) + 30.5, (R2 = 0.872).**

As novel speciation techniques become available to environmental monitoring programs there is a need to ascertain the correlations to sensitive toxicological endpoints. Diffusive Gradients in Thin-films (DGT) passive samplers are designed to accumulate the operationally defined labile fractions of target analytes over time; performing in situ ligand competition and allowing continuous surveillance over sensitive life stages of aquatic species. The LSNM-NP DGT is designed to accumulate labile transition metals, including copper and zinc. These two metals are persistent in coastal seawaters and while they are essential micronutrients required by many aquatic organisms, excess copper and zinc can cause toxicological impairment during early growth stages. In this article, we summarize DGT half maximal effective concentrations (EC50s) and suggested toxicological thresholds for protection of mussel (*Mytilus galloprovincialis*) larvae from labile copper and urchin (*Strongylocentrotus purpuratus*) larvae from labile zinc (Strivens et al., 2020a; Farrell et al., 2021). We then present the status of nearshore waters in Sinclair and Dyes inlets, monitored under the Puget Sound Naval Shipyard's (PSNS) Environmental Investment (ENVVEST) Program (Strivens et al., 2020b).

For proof of concept, we first determined DGT labile mussel copper EC50s by conducting simultaneous EPA/600/R-95/136 tests and DGT measurements in the laboratory, with dissolved organic carbon (DOC) concentrations (ligand competition) adjusted by Suwannee River natural organic matter. We validated the results by repeating the laboratory study with field

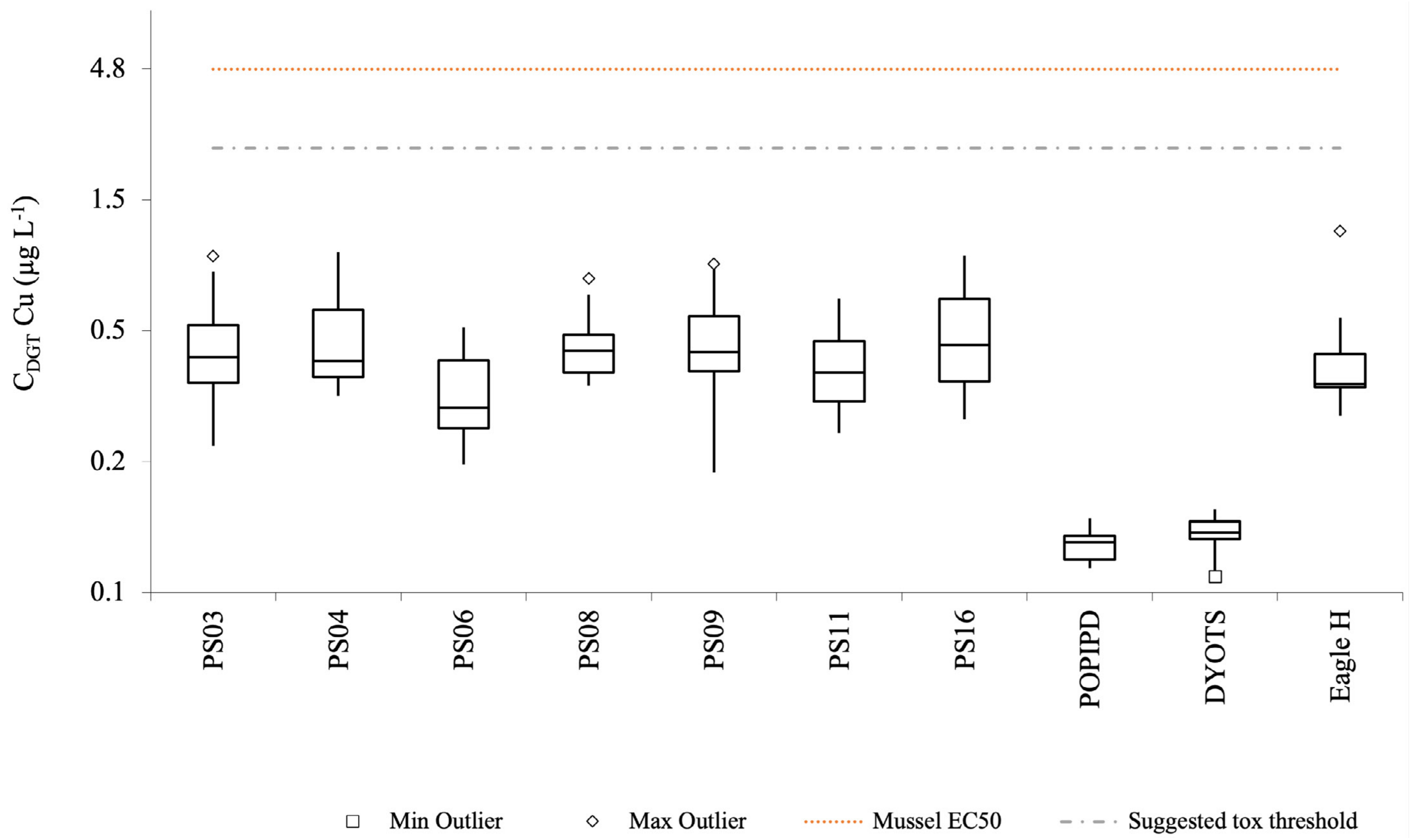

Figure 1. Statistics of DGT Labile copper in the PSNS Nearshore, and reference stations within the Kitsap Basin, during 5 campaigns between December 2016 and July 2019 (*n* = 28/ PSNS station [3-d] and 5–7/ reference station [14-d]). Reprinted from Strivens et al. 2020b.

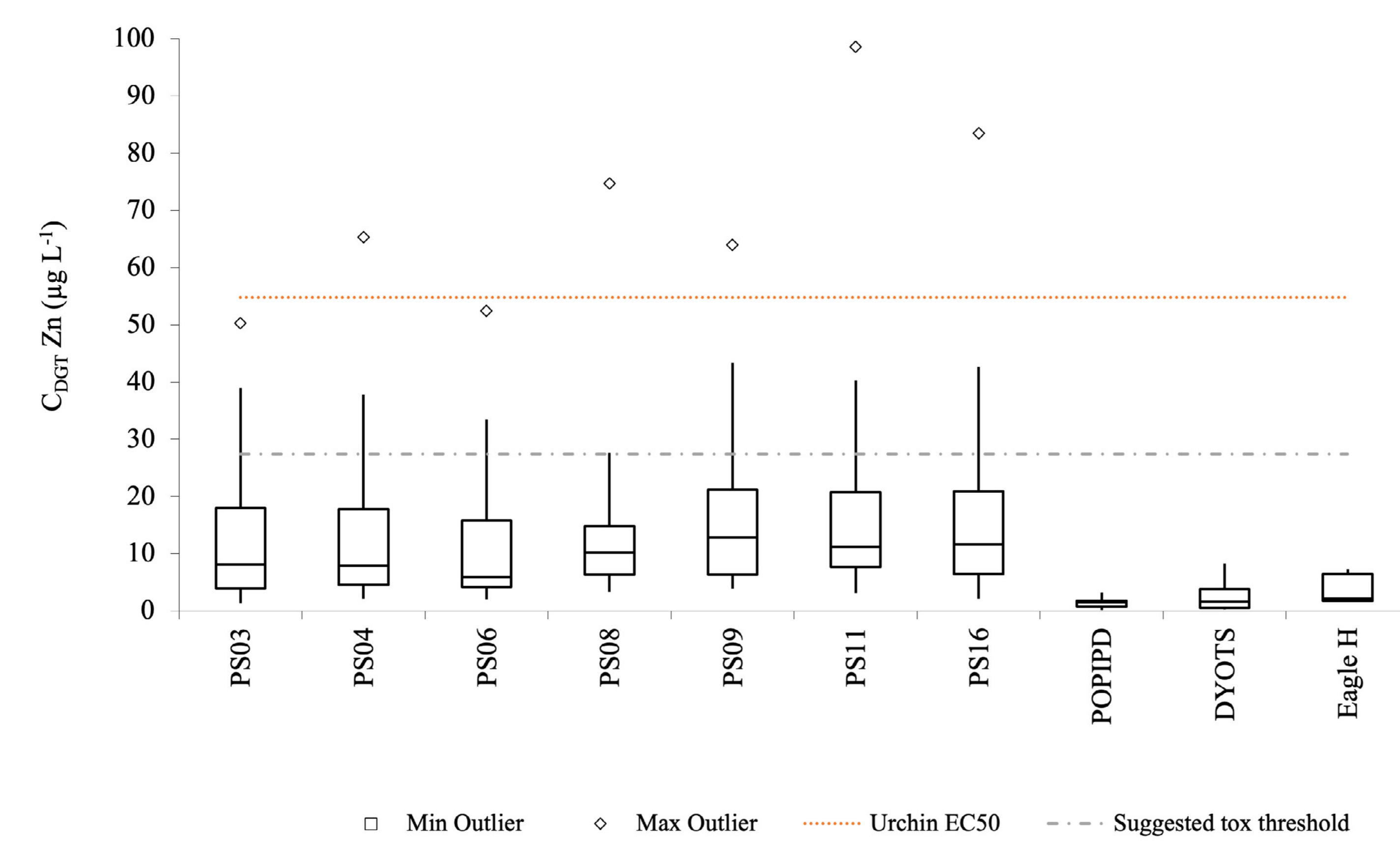

Figure 2. Statistics of DGT Labile zinc in the PSNS Nearshore, and reference stations within the Kitsap Basin, during 5 campaigns between December 2016 and July 2019 (*n* = 28/ PSNS station [3-d] and 5–7/ reference station [14-d]). Reprinted from Strivens et al. 2020b.

collected marine waters with a range of fluorescence properties. For determination of DGT labile zinc urchin EC50s we again conducted simultaneous EPA/600/R-95/136 tests and DGT measurements in the laboratory, using field collected marine waters. The range of fluorescence properties indicated that the mix of ligands present in varying marine organic matter did not significantly change the ratio of binding strengths. We then derived EC50 equations to account for the ranges of DOC concentrations. For suggested toxicological thresholds, we followed the EPA guidelines for deriving numerical national water quality criteria. The suggested thresholds, in our study area, for protection of mussels by DGT labile copper is 2.4 µg/L and for protection of urchins by DGT labile zinc is 27.4 µg/L.

ENVVEST field collected data consisted of 3-day DGT deployments (the average of sensitive mussel and urchin life stages) inside of the PSNS (PS stations) and 14-day deployments at reference stations (POPIPD is the Port of Illahee, DYOTS is the Port of Silverdale, and Eagle H is near the WA State Ferry Maintenance Facility). The results show that all areas had labile copper levels below the suggested threshold and illustrate the differences between industrial and rural/commercial areas (Figure 1). We also observed that labile zinc may be affecting urchin survival (13 percent of datapoints, 4 percent during potential spawning periods) near industrial shipyard activities (Figure 2), which is expected as urchin embryos are known to exhibit one of the narrowest thresholds between essential nutrient and zinc impairment in marine waters; a fact that is not of primary consideration in EPA Aquatic Life Criteria. Overall, the data demonstrate ecosystem health and allow for an improved understanding of status.

Occurrence and Implications of the Tire Rubber-Derived, Acute Coho Salmon Toxicant 6PPD-Quinone in Urban Receiving Waters

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- **6PPD-quinone, a ubiquitous transformation product of the tire rubber antiozonant 6PPD, causes adult and juvenile coho salmon acute mortality**
- **Fall and spring storms transport 6PPDQ into urban receiving waters at concentrations at/near the coho salmon LC50, indicating year-round exposures**

For decades, an unexplained acute mortality phenomenon linked to stormwater and roadway runoff has impacted adult coho salmon (Oncorhynchus kisutch) in Pacific Northwest (PNW) urban streams (Scholz et al., 2011). Building from prior studies, we first analyzed vehicle-linked contaminant sources (e.g., tire wear particles, antifreeze, motor oil) using liquid chromatography coupled to high-resolution mass spectrometry (LC-HRMS) to find that tire wear particle (TWP) leachates were most chemically similar to waters that killed coho (Peter et al., 2018). Follow-up studies indicated that chemicals leached from TWPs re-created the mortality syndrome in juvenile coho salmon (Chow et al., 2019; McIntyre 2021). Subsequent fractionation and effect-direct analyses reduced the observed chemical complexity of TWP leachate from >2000 chemicals to just four, enabling toxicant identification (Tian et al., 2021). We then identified 6PPD-quinone (6PPDQ; C18H22N2O2), a transformation product of the ubiquitous tire rubber antioxidant 6PPD (N-(1,3- dimethylbutyl)-N'-phenyl-p- phenylenediamine) as the primary causal toxicant for coho mortality and confirmed its formation pathway from tires by ozonating commercial 6PPD (Tian et al., 2021). Fish exposures confirmed the acute lethality of 6PPDQ to coho salmon; subsequent availability of a commercial standard yielded a 24-h LC50 of 95 ng/L (95% C.I. = 80-110 ng/L) for juvenile coho salmon (Tian et al., 2022). 6PPDQ is

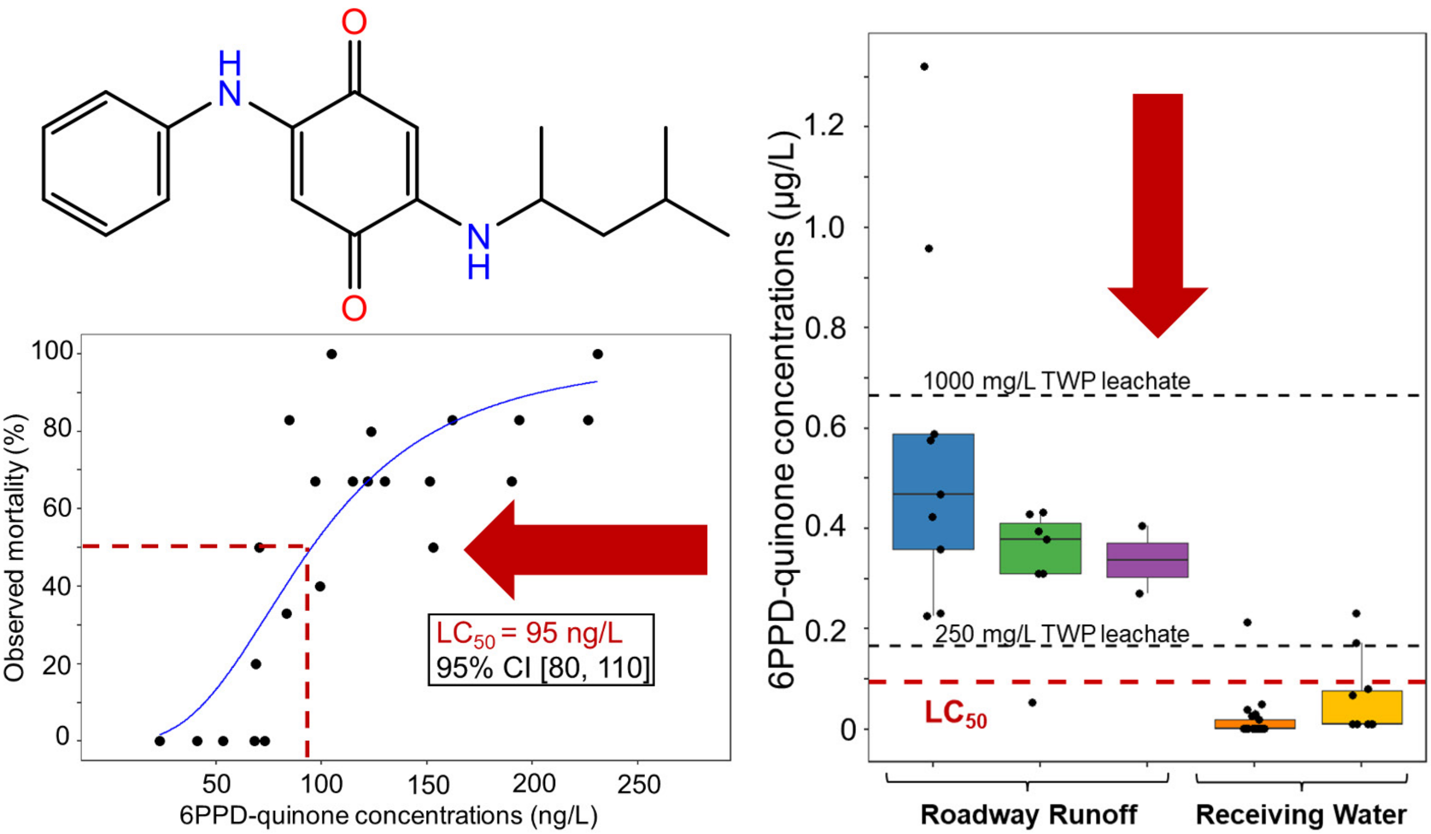

Figure 1. Chemical structure, toxicity estimate, and environmental occurrence of the tire rubber-derived, toxic transformation product 6PPD-quinone in roadway runoff and runoff-impacted receiving waters. For complete details see (Tian et al., 2021, 2022).

A female coho that died due to stormwater exposure, showing egg retention characteristic of the acute urban runoff mortality syndrome. Photo: Zhenyu Tian

among the most lethal known aquatic toxicants, at least for coho salmon (Tian et al., 2022), although many aquatic organisms are insensitive.

Retrospective analyses revealed the ubiquity of 6PPDQ in Seattle-area roadway runoff samples from busy highways (n=18/18 samples; ~50-1200 ng/L) and receiving waters during storm events (n=6/7 storms; <20-200 ng/L). Detections were near or above LC50 values in both Los Angeles roadway runoff and San Francisco receiving waters (Tian et al., 2021, 2022). Sampling in Miller Creek (Burien, WA, USA) during storm events in Fall 2020 (n=4) and Spring 2021 (n=3) revealed peak 6PPDQ concentrations of 33 – 100 ng/L (median 59 ng/L). Importantly, no significant difference was observed between fall and spring peak concentrations (Wilcoxon rank-sum text, p=0.36), indicating that storm events transport 6PPDQ to urban receiving waters year-round, with potential toxic exposure implications for both adult spawners (fall) and juvenile coho salmon (spring/summer). Time-resolved sampling in Miller Creek during Fall 2021 indicated the 6PPDQ concentration profile echoed that of the storm hydrograph, with peak concentrations (at or above the LC50) occurring concurrent with peak flow, followed by extended periods of elevated concentrations (~10-60 ng/L) during the hydrograph tail. Additional research is needed to better understand environmental dynamics, fate, and toxicological implications of the ubiquitous compound 6PPDQ. Concurrent with the 6PPDQ detections, we have also detected many other 6PPD transformation products and quinone transformation products of other PPD antioxidants (Hu et al., 2022). Although the toxicity characteristics of these additional transformation products remain unknown, their detection underscores the substantial impact of tire-derived contaminants on water quality and aquatic species in urban receiving waters.

More than a First Flush: Urban Creek Storm Hydrographs Demonstrate Broad Contaminant Pollutographs

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- **Low-intensity rainfall ("drizzle events") can drive substantial, rapid water quality degradation in urban receiving waters.**
- **In small urban watersheds, more runoff mobilizes more contaminant mass (vs. diluting concentration), especially for vehicle/tire-derived chemicals.**

Stormwater runoff transports complex contaminant mixtures, impacting receiving water quality and ecological health. Adverse impacts are particularly relevant in smaller urban watersheds with limited dilution capacity and "flashy" hydrology. Management efforts are often guided by a conceptual "first flush", with elevated contaminant concentrations and/or mass loads early in storm hydrographs (Bach et al., 2010; Bertrand-Krajewski et al., 1998; Saget et al., 1996). First flush dynamics are primarily reported for "traditional" stormwater contaminants, like dissolved metals and suspended solids, from point sources (Bach et al., 2010; Surbeck et al., 2006). Although urban stormwater systems might be expected to reflect the nonpoint source dynamics of agricultural systems, where increased contaminant mobilization during high runoff flows has been observed (Carpenter et al., 2019), previous studies lacked necessary temporal resolution and a focus on urban/roadway-derived contaminants.

To address this data gap, we collected water samples before, during, and after three fall storm events (~48 h period; October, November, December 2018) in Miller Creek, a representative, small urban watershed in the Puget Sound (WA, USA) region (Peter et al., 2020). October and November storms were concurrent with documented coho salmon (Oncorhynchus kisutch) mortality by urban runoff mortality syndrome (Tian et al., 2021). To evaluate contaminant dynamics along the storm hydrograph (the "pollutograph"), 35 stormwater-derived organic contaminant concentrations were quantified by liquid chromatography coupled to tandem mass spectrometry (Hou et al., 2019), with complementary non-targeted analysis by high resolution mass spectrometry (HRMS). We estimated tire wear particle concentrations as a surrogate metric for urban runoff mortality syndrome risk (the causal toxicant, N-(1,3-dimethylbutyl)-N′ -phenyl-p-phenylenediamine [6PPD-quinone] (Tian et al., 2021) was not yet identified at the time of this study).

During storms, nine contaminants were consistently detected at >500 ng/L: 1,3-diphenylguanidine, hexa(methoxymethyl) melamine, dicyclohexylurea (tire rubber-derived chemicals); benzotriazole, 5-methylbenzotriazole, and 2-hydroxybenzothiazole (corrosion inhibitors); diuron, ibuprofen, and caffeine. All storms exhibited sharp hydrographs and broad pollutographs for quantified analytes (Figure 1), and the total number/peak area of HRMS detections, which are broader metrics of contaminant loads. A rapid first flush occurred, with elevated concentrations early in each storm event notable relative to low first-flush runoff volumes (even during light drizzle), indicating high-strength, readily transportable contaminant sources. We estimated ~0.5-170 mg/L tire wear particles during storms, mirroring first-flush and middle-flush trends observed for individual tire wear particle-derived chemicals (Peter et al., 2020).

Water sampling equipment set up next to Miller Creek on January 9, 2018, in preparation for storm sampling. Photo: Katherine Peter

Contaminant mass mobilization continued during and after peak in-stream flows, with contaminant concentrations undiluted by additional runoff – a so-called "middle flush" (Qin et al., 2016). These observations indicated "semi-infinite" mass reservoirs of abundant/pervasive contaminants relative to available runoff volumes may exist in urban watersheds, yielding transport-limited (vs. mass-limited) dynamics in some locations. Additional runoff transports additional contaminant mass into receiving waters, rather than depleting available reservoirs. Supporting this observation, per-storm tire wear particle loads were equivalent to the tread wear generated by 0.5x106 –7.8x106 vehicle miles traveled (Peter et al., 2020), indicating the presence of large tire wear particle "source reservoirs" (e.g., road dust, detention systems, creek sediments) from which contaminants were mobilized by runoff throughout storm events. Such stagnant, high-strength sources represent opportunities for focused identification and management or treatment efforts.

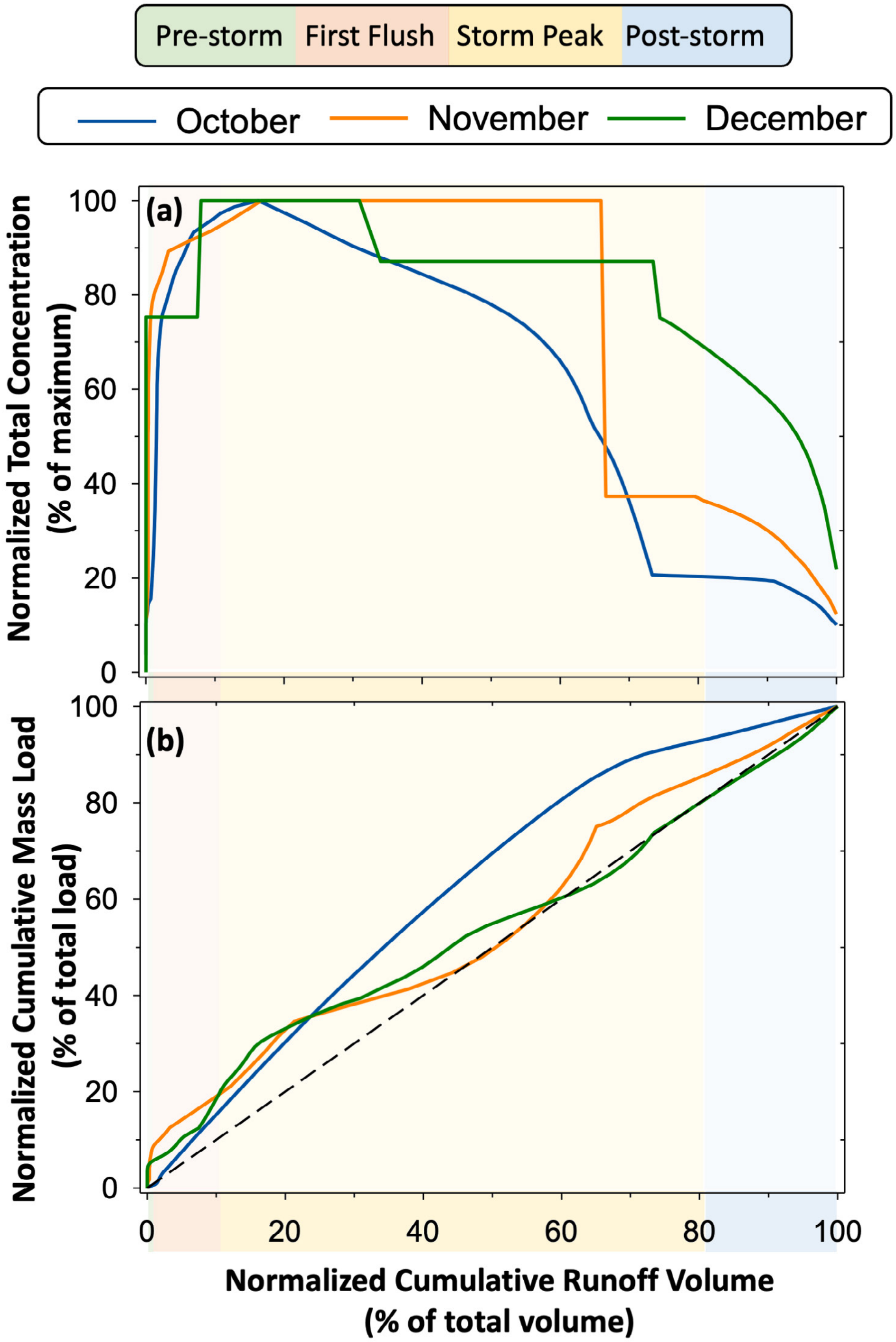

Figure 1. Pollutographs (of the 35 targeted stormwater-derived organic contaminants) during 3 storm events in Miller Creek (Burien, WA, USA) show both (a) high contaminant concentrations and (b) continued mass transport into receiving water through the majority of the storm hydrographs. Reproduced from Figure 2 in Peter et al. (2020).

Leachate from Tire Wear Particles Recreates Acute Toxicity from Roadway Runoff in Coho but Not Chum salmon

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- **Chemicals from tires are responsible for acute coho spawner mortalities.**
- **As with whole roadway runoff, chum salmon do not appear sensitive to tire-derived chemicals.**

In developed areas of the Pacific Northwest, acute mortality of adult coho salmon (Oncorhynchus kisutch) follows rain events (Scholz et al., 2011) and is correlated with roadway density (Feist et al., 2017). Roadway runoff experimentally triggers behavioral symptoms (lethargy to loss of equilibrium) and associated changes in blood (low ions, high hematocrit) indicative of cardiorespiratory distress prior to death in coho adults (Spromberg et al., 2016) and juveniles (McIntyre et al., 2018). Closely related chum salmon (O. keta) lack an equivalent response (McIntyre et al., 2018). Acute mortality of juvenile coho was recently experimentally linked to 6PPD-quinone (p-phenylenediamine)– a transformation product of a tire-derived chemical (Tian et al., 2021). We evaluated whether chemicals leaching from tire wear particles (TWP) is sufficient to trigger the acute mortality syndrome in adult coho salmon, published as McIntyre et al. (2021) in Environmental Science & Technology. Well water at the Grovers Creek Indian Hatchery (Suquamish, WA) was recirculated for 24 h through a mixture of particles from nine unique tires (7 used, 2 new). Adult coho were exposed to well water or tire wear particle (TWP) leachate at 0.1 mg/L, 0.32 mg/L or 1.0 mg/L for up to 24 h. Some mortality was observed at 0.1 mg/L but all coho died at the two higher concentrations. We characterized the acute response (survival, behavior, blood physiology) of adult coho and chum salmon co-exposed to 0.32 mg/L TWP leachate and compared it with that caused by roadway runoff. TWP leachate was acutely lethal to coho at concentrations of chemicals expected in roadway runoff, with the same behaviors and blood parameters impacted. Concentrations of 6PPD-quinone at 320 mg/L TWP leachate were on the order of 2 mg/L. Within 3 h of exposure, coho were severely symptomatic; behaviorally unresponsive, with significantly increased hematocrit, and decreased plasma pH, sodium and chloride. As with runoff, chum salmon appeared insensitive to TWP leachate even at concentrations lethal to coho. Our results confirm that environmentally relevant TWP exposures cause acute mortalities of a keystone aquatic species, lending support to the finding that 6PPD-quinone is acutely lethal to juvenile coho at concentrations measured in runoff and receiving waters (Tian et al., 2021).

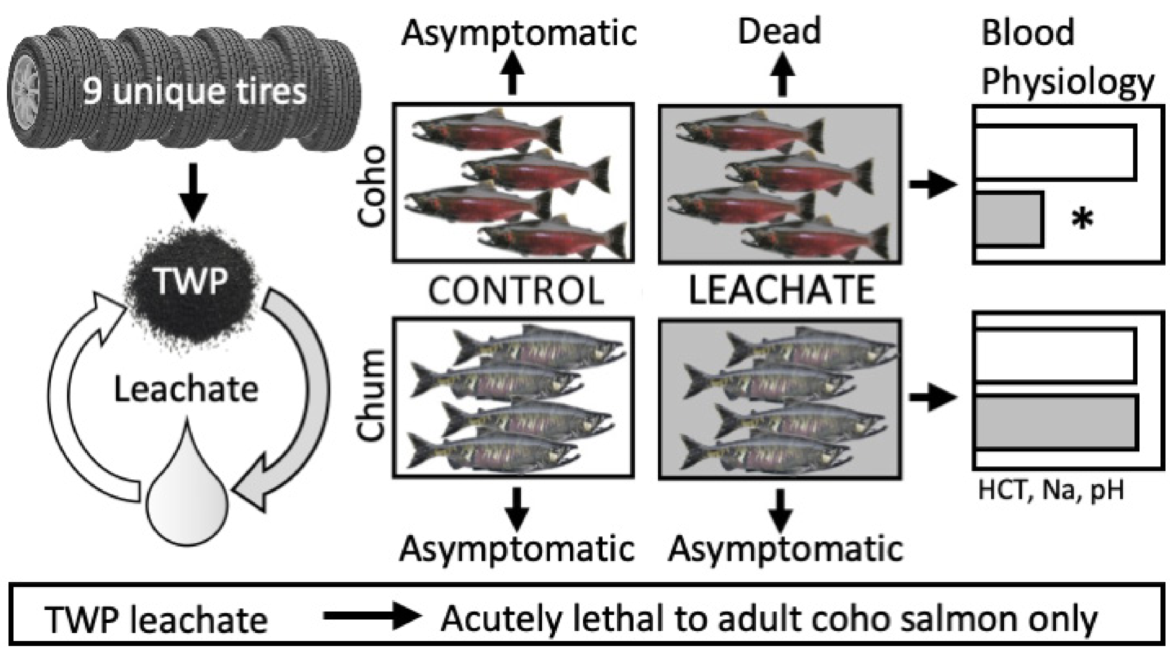

Figure 1. Overview of study

An Urban Stormwater Runoff Mortality Syndrome in Juvenile Coho Salmon

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- **Juvenile coho exposed to runoff exhibit similar changes in behavior and blood chemistry as adult spawners, and also die within a few hours.**
- **Symptomatic fish did not recover when transferred to clean water, suggesting a single runoff event to stream habitats can be lethal.**

Adult coho salmon (*Oncorhynchus kisutch*) suffer from acute mortality following rain events in urban streams across the Pacific Northwest (Scholz et al., 2011) and when exposed directly to collected untreated road runoff (Spromberg et al., 2016, McIntyre et al., 2018). Urban runoff mortality syndrome in adult coho spawners has been documented for decades, however except for anecdotal evidence, it was previously not known whether juvenile coho, which spend over a year in freshwater streams, are also affected. We conducted a detailed assessment of behavior and physiology in juvenile coho exposed to roadway runoff, which was published in Aquatic Toxicology as Chow et al. (2019).

Freshwater-stage juveniles were exposed to clean water (control) or road runoff collected from a high traffic volume urban arterial roadway. Symptoms characteristic of the mortality syndrome were evaluated using digital image analysis, and discrete stages of abnormal behavior were characterized as the syndrome progressed. At a subset of these stages, blood was analyzed for ion homeostasis, hematocrit, pH, glucose, and lactate.

The progression of behavioral changes observed in exposed juvenile coho were similar to those previously observed in exposed spawners. Within 45 minutes of exposure, time spent at the water surface had significantly increased. Over 7 hours, surface swimming progressed from occasional to continuous surface swimming, fish eventually lost equilibrium, and finally lost buoyancy and settled to the bottom of the tank in a moribund state (23 of 24 exposed fish). No mortality was observed in control fish exposed to fish lab water.

The observed effects on the blood chemistry of juvenile coho exposed to urban runoff were also comparable to the effects reported for symptomatic coho adults (McIntyre et al., 2018). At the point of equilibrium loss, both life history stages had a measurable increase in blood glucose, lactate, and hematocrit, and a decrease in blood ion content. Furthermore, affected fish did not recover when transferred to clean water, suggesting a single runoff event to stream habitats could be lethal if resident coho become symptomatic.

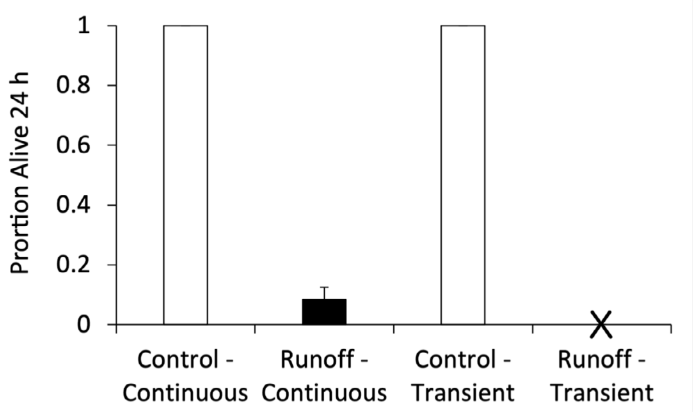

Figure 1. Average survival of juvenile coho at 24h following continuous exposure to runoff or after transfer to clean water upon showing surface swimming. Controls were fish exposed only to clean water that were time-matched for transfer to another tank containing clean water. Three fish were exposed in each treatment for each exposure. Error bar is one standard error of the mean of four exposures.

Among coho life stages, our findings indicated the urban runoff mortality syndrome is not unique to adult spawners. The above core finding suggests a common toxic mechanism across life stages such that juveniles represent an expanded platform for understanding and eventually mitigating stormwater toxicity to wild coho. Over the past two decades, the logistical challenges associated with studies on adults have slowed the pace of research and limited the range of experimental questions that can be addressed in a given year. By contrast, juveniles are convenient to handle, amenable to small volume exposures, and available year-round. The establishment of juveniles coho as a model for urban runoff mortality syndrome in this study has allowed more recent studies to utilize juveniles in advancing this body of research (Blair et al., 2021; Tian et al., 2021).

Acute Cerebrovascular Effects in Juvenile Coho Salmon Exposed to Roadway Runoff

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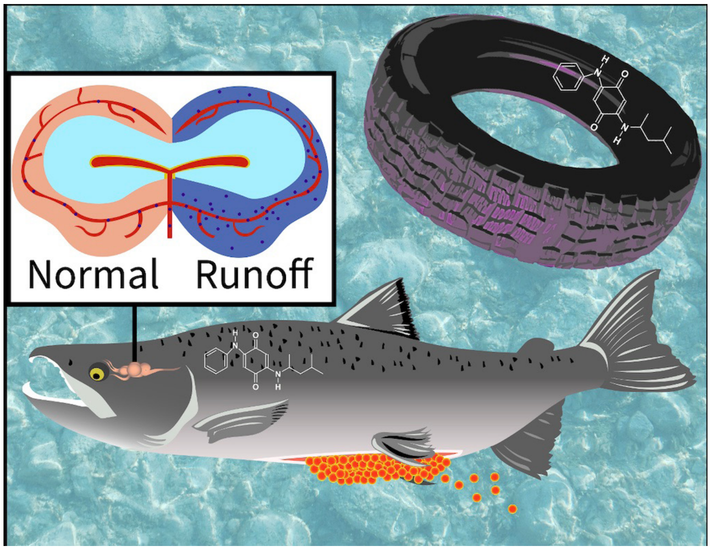
- **A severe hematocrit rise is a recurrent symptom in coho salmon lethally exposed to urban stormwater runoff.**
- **Hematocrit rises may be explained by plasma leakage from the blood-brain barrier in runoff-exposed coho salmon.**

Stormwater runoff conveys a complex mixture of known and unknown contaminants to salmon-bearing streams that threaten the survival of coho salmon in the urban Pacific Northwest. The tire chemical known as 6PPD-quinone has been recently identified as the putative toxicant driving acute toxicity in coho salmon exposed to urban stormwater runoff (Tian et al., 2020). Deaths are accompanied by a symptomology indicative of cardiorespiratory distress (e.g., surface swimming, gaping) and neurological impairment (e.g. fin splaying, loss of orientation, loss of equilibrium) in prespawner adults (McIntyre et al., 2018) and juvenile coho (Chow et al., 2019) exposed to urban runoff. A severe hematocrit rise, or increased concentration of red blood cells by volume, was a notable symptom upon the development of loss of equilibrium behavior prior to death.



The tire chemical 6PPD-quinone causes blood-brain barrier disruption and acute mortality in coho salmon.

Several potential mechanisms of hematocrit rise were investigated to explore the toxic mode of action in the acute mortality of juvenile coho salmon exposed to roadway runoff. These included possible combinations of released stored red blood cells in the spleen, red blood cell swelling, dehydration of blood plasma from an osmoregulatory disturbance, and/or loss of circulating blood plasma (Randall, 1982). Juvenile coho salmon were exposed to roadway runoff diluted to 50% using fish rearing water until demonstrating loss of equilibrium behavior. Blood samples were analyzed for hematocrit and total hemoglobin concentration. Mean cell hemoglobin concentration was determined from total hemoglobin concentration and hematocrit. Blood plasma was isolated and analyzed for total protein, total antioxidant power and thiols.

The aim of the study was to distinguish between pathways related to hematocrit rise that would lead to death and non-lethal fish responses to stress, based on the severity of responses to runoff exposure. Runoff-exposed coho demonstrated dramatic increased total hemoglobin and slightly reduced mean cell hemoglobin concentration of the blood, relative to controls, indicating severely increased concentration of red blood cells and slight swelling of red cells, respectively. Osmoregulatory dysfunction and plasma loss were tested as mechanisms of increased red blood cell concentration. Because we did not observe a significant change in blood plasma constituents (i.e., antioxidant power, thiols), plasma dehydration due to osmoregulatory dysfunction was discounted as a major driver of hemoconcentration. To test for plasma leakage from blood vessels, Evans Blue dye complexed with bovine serum albumin (EDB-BSA) was introduced into circulation of anesthetized juvenile coho, followed by a saline washout of the blood vessels. Runoff-exposed coho showed pervasive accumulation of EBD-BSA in the brain, relative to controls, demonstrating plasma leakage from the cerebrovasculature.

Our results suggest a breakdown of the blood-brain barrier underlies extreme hematocrit rises, based on the accumulation of EBD-BSA in the brain of runoff-exposed coho. Blood-brain barrier disruption allows passive diffusion of toxic substances to pass from the blood into the central nervous system, which may lead to impairment of neuronal function and death (Abbott et al., 2006). Future toxic mode of action studies should examine pathways of blood-brain barrier disruption to evaluate lethal and sublethal effects in coho salmon exposed to roadway runoff.

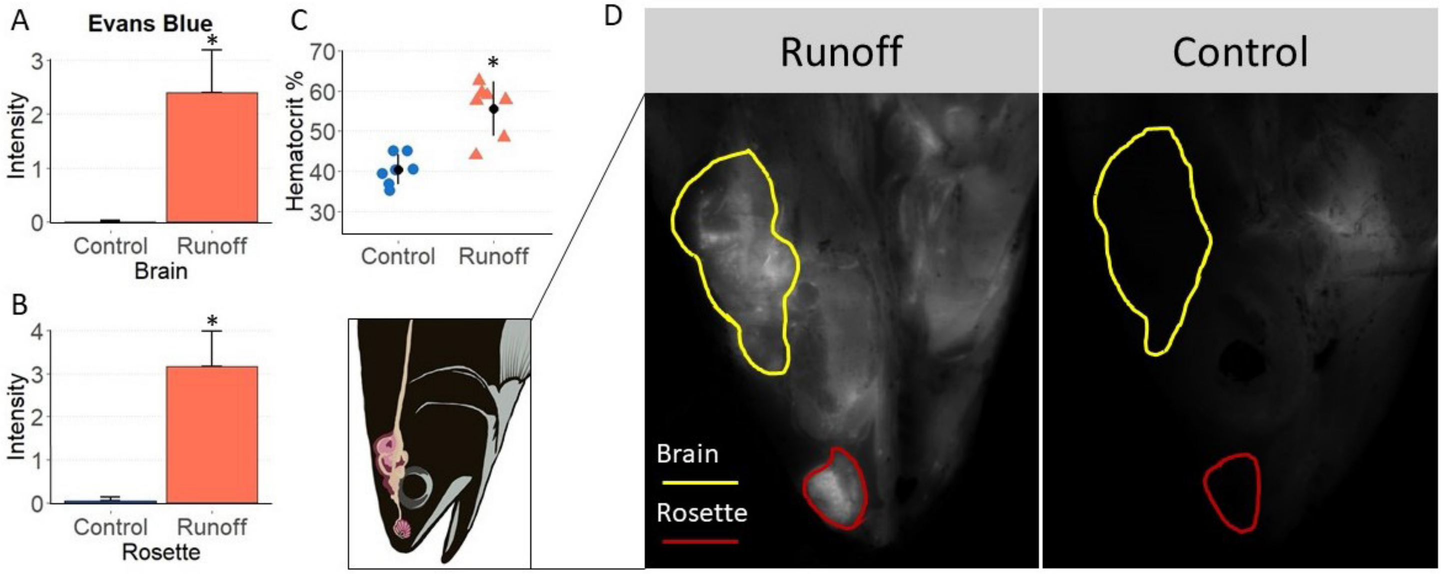
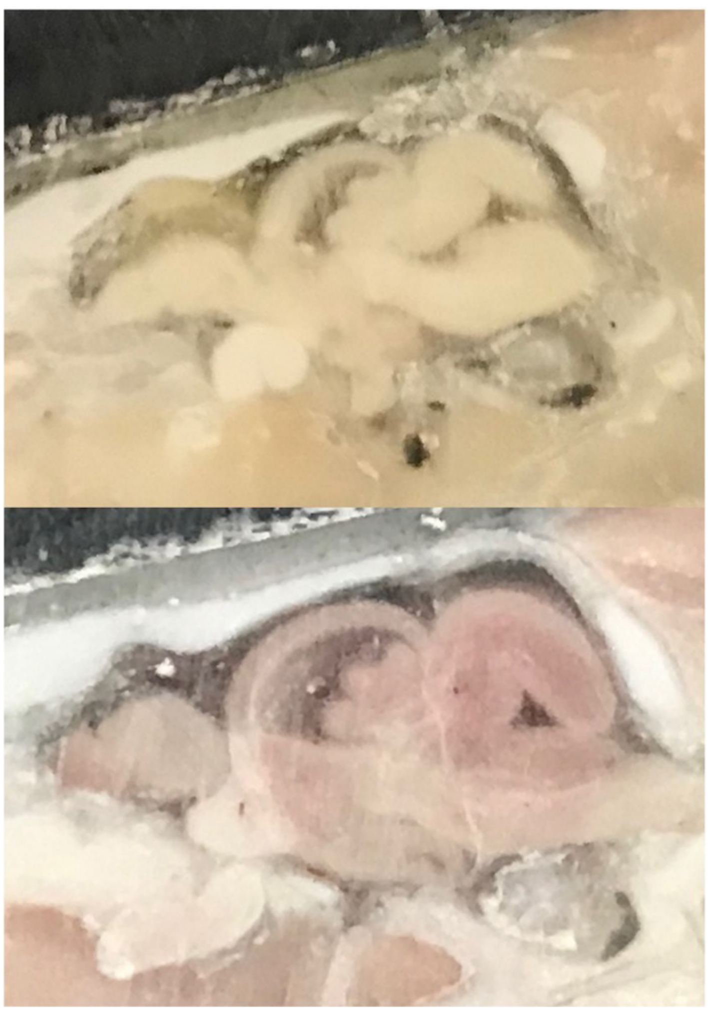
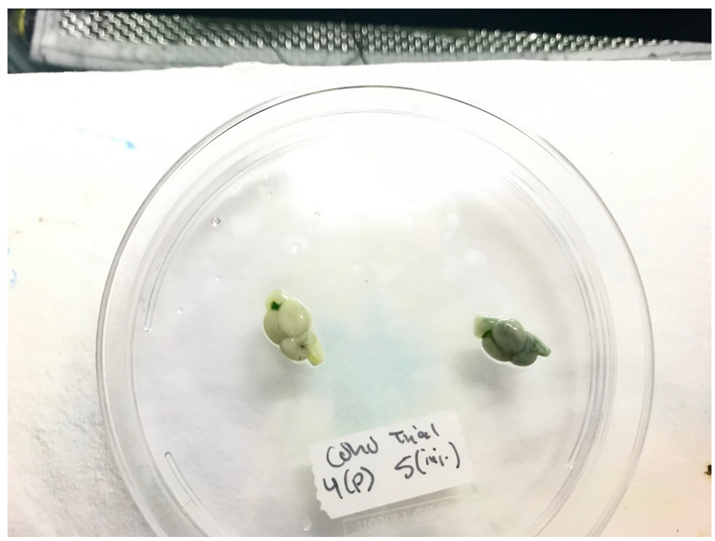


Figure 1. Hematocrit and Evans Blue bound to bovine serum albumin (EBD-BSA) results for juvenile coho lethally exposed to roadway runoff (n=7) and controls (n=7). Mean fluorescence intensities of blood-brain barrier tracer (EBD-BSA) for brain (A) and olfactory rosette (B) regions show pervasive accumulation of the tracer in runoff-treated fish. Runoff-exposed coho also demonstrated severe hematocrit rises (bars show mean ± s.d.) (C), relative to controls. * indicates p-value < 0.001 based on Bonferroni confidence intervals. EBD-BSA fluorescence imaging of whole head cryosections (D) demonstrate post-washout retention of tracer in brain and olfactory rosette regions of runoff-treated fish, while tracer was nearly absent in controls. Figure from Blair et al. (2021).



A cryosection of two juvenile coho salmon exposed to roadway runoff until loss of equilibrium (bottom) and clean water (top). The brain of the runoff exposed coho is abnormally pink colored (bottom), relative to the control (top), indicating runoff exposure causes cerebrovascular effects, such as inflammation, coagulation, or hemorrhaging. Photo: Stephanie Blair



Dissected brains of juvenile coho exposed to clean water (left) and roadway runoff (right) before injection with a high concentration of Evans Blue dye and saline perfusion. In the coho exposed to roadway runoff, the brain shows high accumulation of Evans Blue dye following a washout of the blood vessels with saline, indicating for severe blood-brain barrier disruption. Photo: Stephanie Blair


Shark Reef, Lopez Island, Washington/ Photo: Sylvia Kantor

SECTION 3: MANAGEMENT

Steelhead on redd at Boise Creek restoration site.
Photo: Josh Latterell

Safer Products: An Opportunity for Pollution Prevention

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https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Green-chemistry
https://ecology.wa.gov/Regulations-Permits/Guidance-technical-assistance/Safer-alternatives

- **Consumer products are significant sources of toxic chemicals to Puget Sound.**
- **Regulatory and voluntary programs at Ecology are reducing hazardous chemicals in consumer products and preventing their release into the environment.**

Many consumer products we use at home, work, or school contain toxic chemicals that can harm our health and contaminate the environment. For most chemicals used in consumer products, there is inadequate hazard or exposure information to understand the risks they pose to people and the environment. Yet epidemiological and environmental monitoring studies often find impacts from chemicals used in consumer products. One way to prevent risks from chemicals in consumer products is to avoid the use of hazardous chemicals. This approach reduces risks across the lifecycle of the product by reducing exposures to toxic chemicals during the manufacturing, use, and disposal or reuse phases (Figure 1).

The Department of Ecology works to reduce the use of hazardous chemicals in consumer products through regulatory and voluntary efforts.

One key regulatory program is Safer Products for Washington, which implements The Pollution Prevention for Healthy People and Puget Sound Act . This program aims to promote transparency and reduce hazardous chemicals in consumer products. The statute requires identifying hazardous chemicals or chemical classes and consumer products that are significant sources or uses of those chemicals (Figure 2). If safer alternatives are feasible and available, Ecology can restrict a chemical or chemical class in a consumer product through rulemaking.

To implement this law, Ecology developed methods for identifying alternatives that are safer than hazardous classes of chemicals. Together with stakeholders, the program set a criteria for safer, which can be used as a transparent bar for hazard reduction (Ecology, 2022). The criteria aligns with other

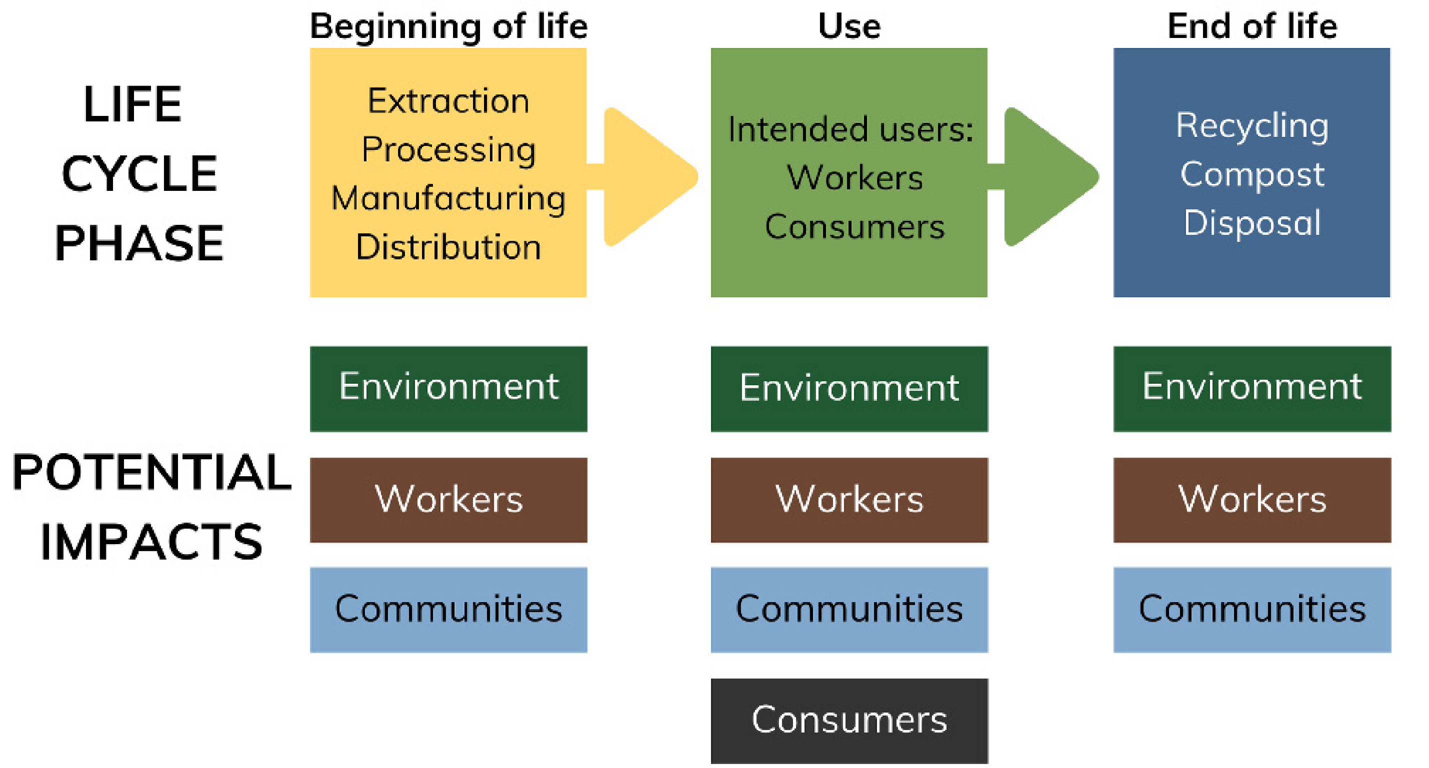

Figure 1. Life cycle phases for consumer products and the potential impacts of toxic chemicals on the environment, workers, communities, and consumers.

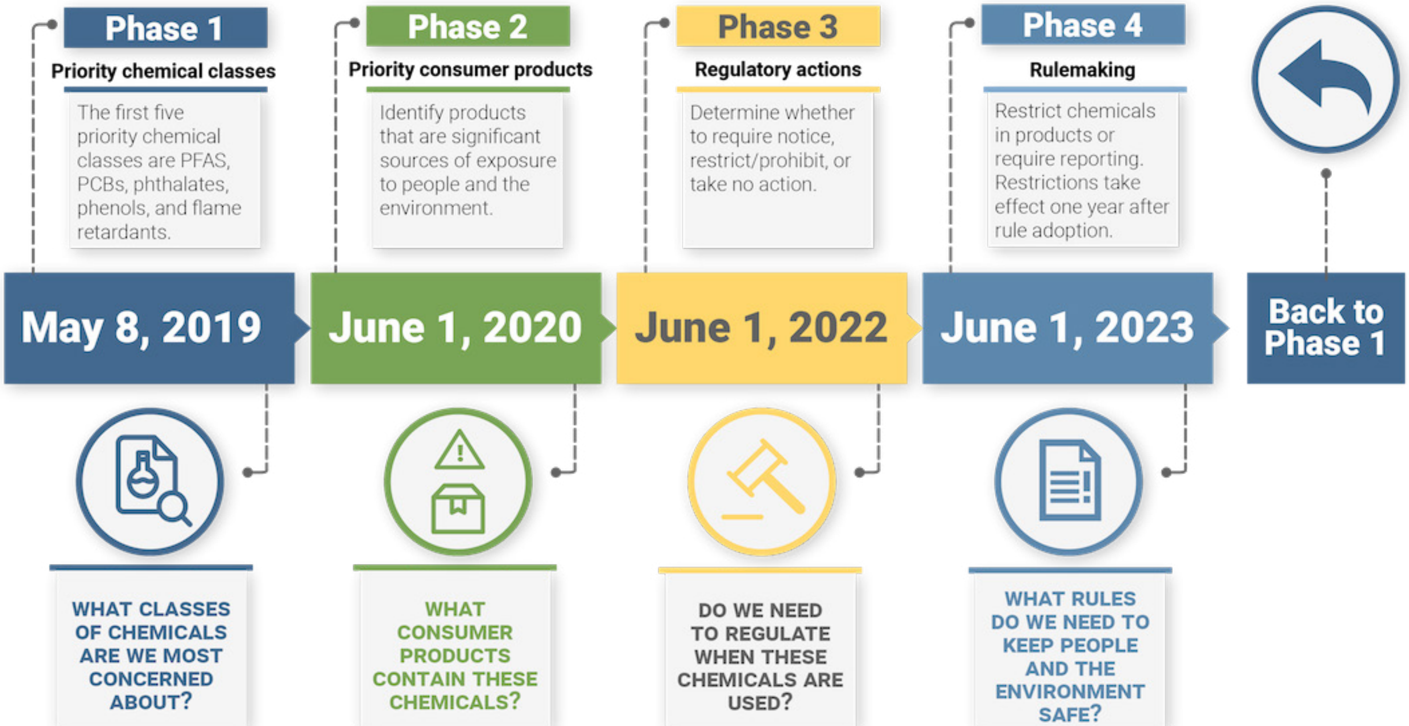

Figure 2. The four implementation phases of the Safer Products for Washington cycle. These four phases repeat on a five year cycle.

chemical hazard assessment methods, such as GreenScreen® for safer chemicals and the Environmental Protection Agency's Safer Chemical Ingredients List Master Criteria. This approach allowed Ecology to leverage existing hazard assessments and certifications. Since 2019, the program identified safer, feasible, and available alternatives to per- and polyfluoroalkyl substances, phthalates, organohalogen flame retardants, specific organophosphate flame retardants, bisphenols, alkylphenol ethoxylates, and polychlorinated biphenyls in a wide range of consumer products (Ecology, 2022). Efforts to adopt restrictions and transparency requirements are currently underway.

Ecology advances the awareness and voluntary adoption of safer alternatives and green chemistry practices by developing tools and trainings and providing technical assistance. Some examples include collaborating to develop and update the Interstate Chemicals Clearinghouse Alternatives Assessment Guide, funding chemical and product assessments to increase the availability of alternatives, and connecting businesses with resources to help them switch to safer alternatives. Ecology educates current and future scientists by developing trainings on green chemistry and safer alternatives and creating curriculum on safer certifications. These efforts help both the current and the next generation of scientists learn to design chemistries with the health of people and the environment in mind.

These regulatory and voluntary pollution prevention efforts are important for reducing continued sources of toxic chemicals to Puget Sound and the environment more broadly.

The Puget Sound Stormwater Heatmap Identifies Hotspots of Pollution Generation and Runoff throughout the Puget Sound Watershed

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- **The Puget Sound Stormwater heatmap is an interactive tool built to identify where stormwater pollution and runoff is generated on the landscape.**
- **The heatmap is available at www.stormwaterheatmap.org; data are open source.**

The stormwater heatmap is an interactive mapping tool, report generator, and data repository that quantitatively visualizes hotspots of pollution and runoff generation throughout the Puget Sound watershed. By merging local monitoring data, statistical modeling, and cloud computing technologies with compelling visuals and flexible scaling, the heatmap improves our collective ability to prioritize stormwater infrastructure investments by mapping predicted pollutant loads across the Puget Sound landscape. The heatmap provides a runoff and pollution loading "threat" map that can be coupled with social-ecological data to generate action maps for stormwater intervention.

To build the heatmap, we conducted a Design Thinking (DT) exercise to identify what type of tool would be useful to stormwater managers, particularly the 85 Phase II jurisdictions recently required to develop science-based stormwater management plans. The DT project identified 4 critical elements:

- **Compelling Visuals**: tools should help stormwater managers tell a story to different audiences.
- **Multiple Scales**: stormwater planning takes place at the parcel, neighborhood, watershed and regional scale. Data need to be flexibly aggregated at all scales.
- **Make it Mine-able**: serve as a data platform and resource for use with other tools. Land cover, soils, hydrology, and climate change impacts data would help meet multiple modeling needs.
- **Grounded in Science**: data and calculations should be apparent and meet current best practices.

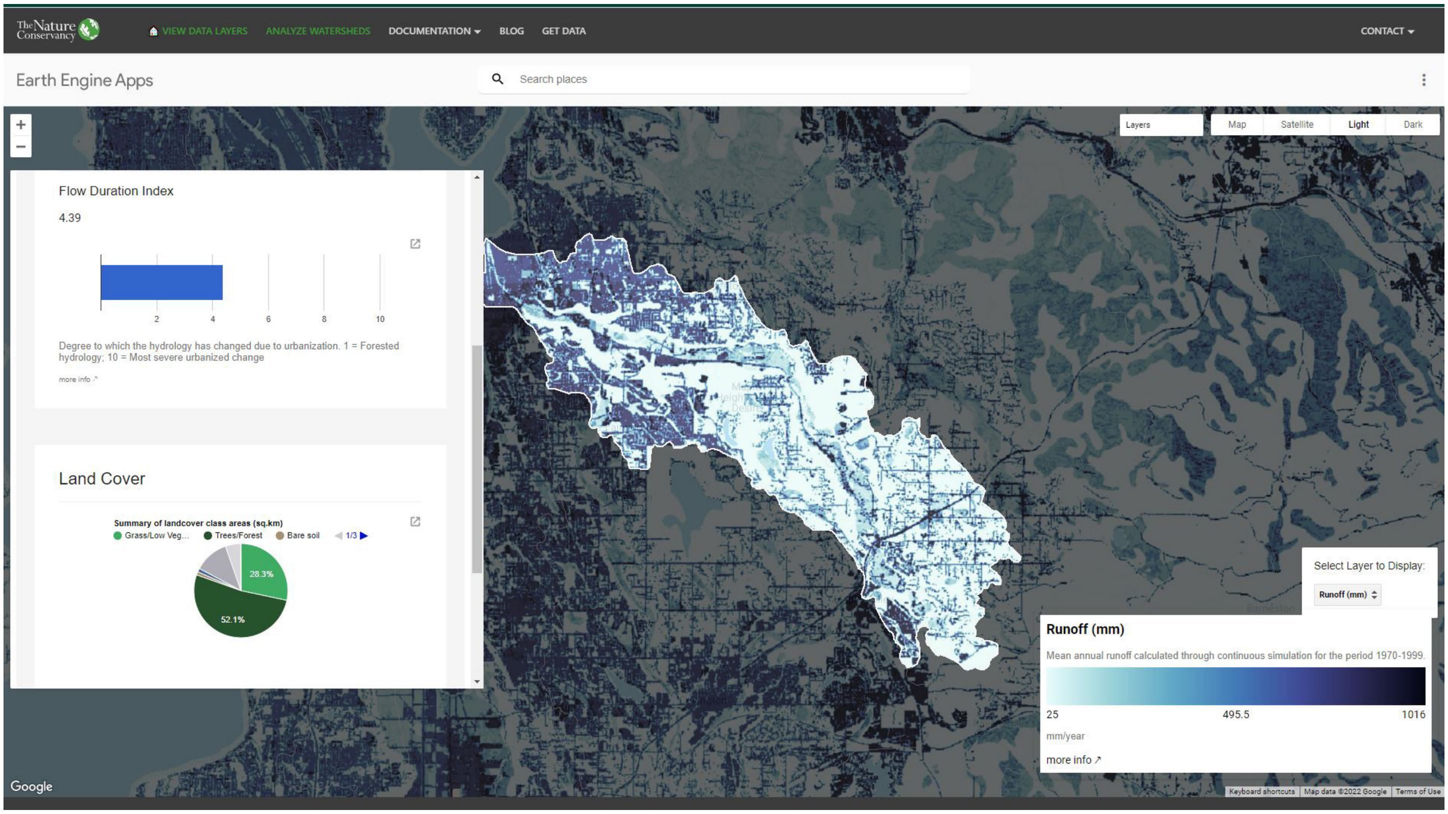

Figure 1. Example of output metrics provided by the stormwater heatmap for a user-specified watershed unit. Metrics depicted include mean annual runoff (calculated through continuous simulation from 1970-1990), the flow duration index (unitless metric characterizing the degree to which hydrology has changed due to urbanization), and land cover class areas for the specified watershed.

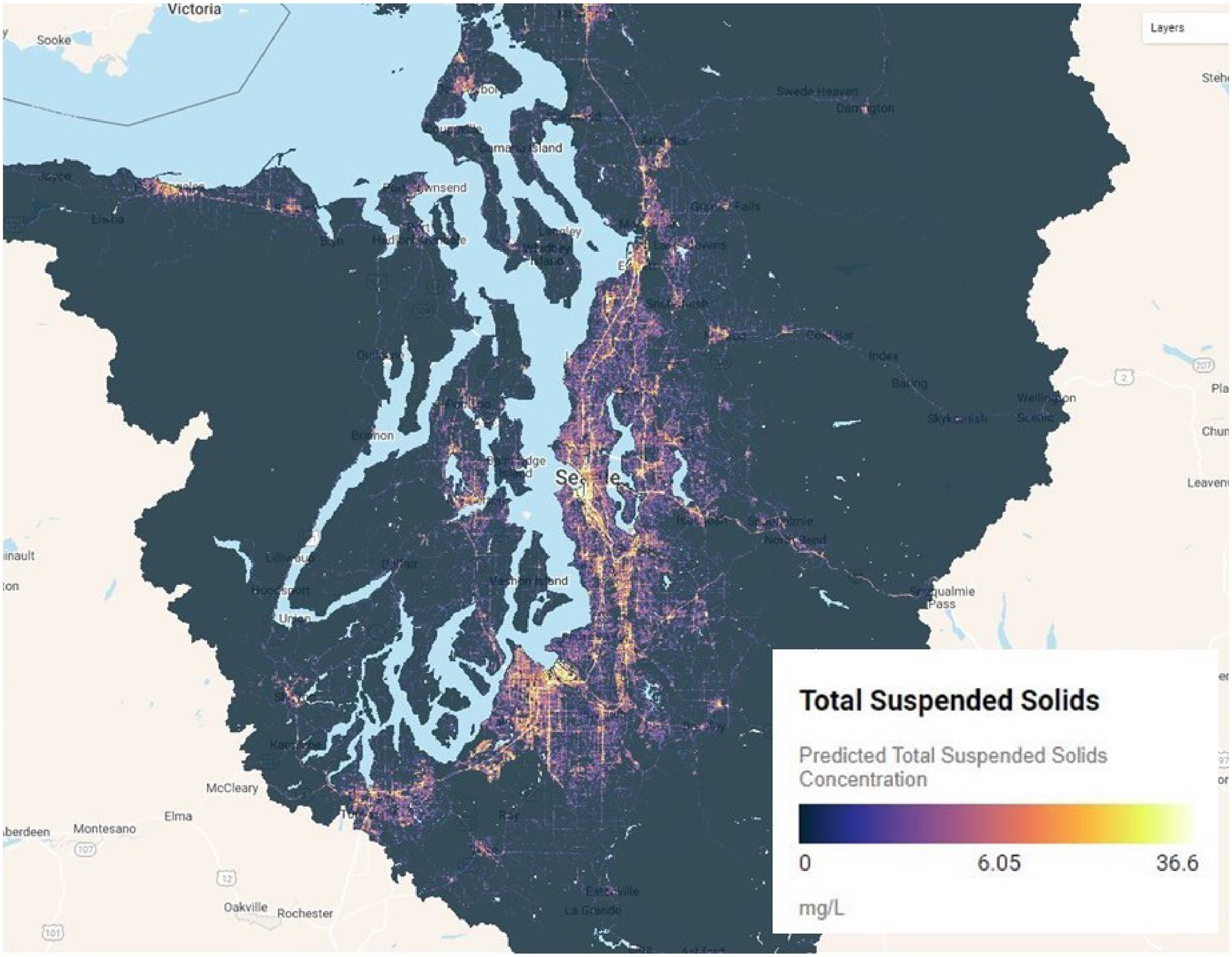

Figure 2. Example output data layer depicting the predicted concentration (mg/L) of total suspended solids across the Puget Sound watershed.

The heatmap is comprised of three major components:

1. Landcover Refinement: we generated a high resolution (1-m^2) landcover dataset. This is a critical level of resolution for urban runoff modeling because impervious surfaces strongly drive hydrologic response, and therefore pollution loading.

2. Hydrology: we conducted continuous hydrology simulations for the 32 hydrologic response units (HRU = combination of landcover, soils, & slope) found within Puget Sound. Using regional precipitation datasets provided by the Climate Impacts Group, we simulated current and future hydrology in order to assess climate change impacts on runoff and pollution loading, generating more than 311 billion rows of data. This dataset provides an efficient way to model rainfall-runoff relationships using the Western Washington Hydrology Model.

3. Pollution Statistics: We developed a series of linear mixed effects models to predict concentration of pollutants across the Puget Sound landscape. These models link monitoring data to landuse and land cover characteristics, as well as rainfall, seasonality, and other spatial covariates, such as air quality (PM2.5). Concentration predictions were then combined with hydrology output to calculate pollution load across the Puget Sound landscape at a 1-m^2 spatial resolution.

The resulting interactive tool enables users to visualize and aggregate stormwater pollution loading data at several spatial resolutions for local, watershed, and regional-scale planning. Figure 1 shows an example of output metrics provided by the stormwater heatmap for a user-specified watershed unit. The project reveals that areas with high percent cover of impervious surfaces, such as hard cityscapes, as well as industrial and commercial zones, tend to produce higher pollutant loads than high-density residential, low-density residential, and rural areas. Transportation networks–roads and highways–generate very high levels of stormwater contaminants, especially those with higher traffic intensity as illustrated for total suspended solids in Figure 2.

'Salmon Friendly' Stormwater Projects in King County Watersheds

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- **Salmon Friendly is on track to install 30 green stormwater projects in King County by the end of 2022. Up to 200,000 gallons of stormwater will be available for reuse during the growing season from cistern projects and an estimated 130,000 gallons of runoff will be treated by Salmon Friendly rain gardens.**
- **New methods of pollution monitoring and data collection will need to be explored to accurately track the pollution mitigating effects of rain gardens and other Salmon Friendly projects.**

Chinook salmon (*Oncorhynchus tshawytscha*) are the largest species of Pacific salmon. They are a keystone species in the Salish Sea region, playing a critical role in indigenous cultures and natural ecosystem processes. Puget Sound Chinook salmon have been listed as threatened under the Endangered Species Act since 1999, however to date their populations are showing little sign of recovery and are considered "in crisis" despite the significant local and regional efforts being invested in Chinook recovery (Governor's Salmon Recovery Office, 2021).

Non-point source pollution from stormwater runoff is among the many threats to Chinook population viability, especially during their freshwater life phases. In part because of the difficulty in identifying and addressing nonpoint sources of pollution, they continue to endanger public health, natural resources, and aquatic ecosystems (Wong and Pickett, 2014). Chinook salmon are affected by the cumulative impacts of these nonpoint sources, which is why the implementation of green stormwater best management practices (BMPs) has been recommended by experts at every level and geographic scale.

Mid Sound Fisheries Enhancement Group started Salmon Friendly, a green stormwater infrastructure retrofit program, as a Puget Sound Action Agenda Near Term Action (NTA) in 2020. We committed to implementing a total of 30 such BMPs over two years in Water Resource Inventory Areas (WRIAs) 8 and 9,

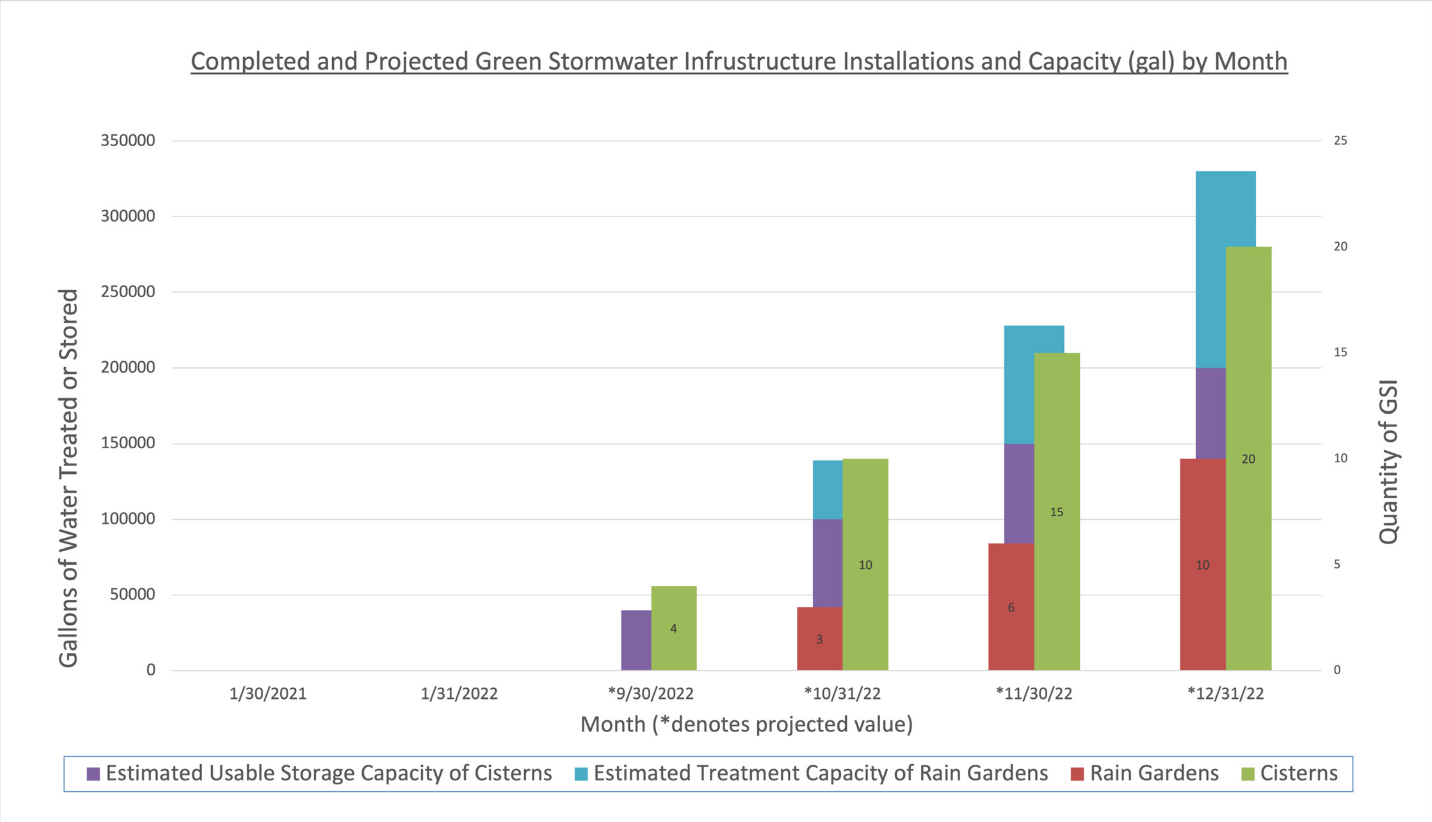

Figure 1. A bar graph showing the number of projects installed, and the amount of stormwater treated in gallons.

Salmon migrating through the Ballard Locks, September 2022. Photo: Tracy Banaszynski

the watersheds at the core of the most populous metropolitan region in the state. The rapidly urbanizing landscape of these watersheds is home to some of the highest levels of pollutants and impervious surfaces, a common source of stormwater runoff, in the state. It is also home to remnants of historically important populations of Chinook salmon, a species of focus for restoration and conservation efforts throughout the region. In response, Mid Sound created the Salmon Friendly program as a targeted approach to voluntarily address nonpoint source pollution from stormwater on private, typically residential, properties near Chinook salmon streams.

BMPs offered to participants include rain gardens and cisterns, which both provide opportunities to reuse stormwater and infiltrate it onsite rather than allowing it to leave as untreated pollution. We are able to estimate how many gallons of stormwater are treated by a BMP in a calendar year, and by our program in total, given the types of BMP installed and the areas of contributing impervious surface from which they collect stormwater. However, there is currently no mechanism in place to measure the types and quantities of pollution being imported to and exported from a specific BMP. Existing pollution monitoring infrastructure is not designed to measure the relatively small and diffuse stormwater benefits of Salmon Friendly projects, and new and novel data collection methods will need to be identified to properly calculate the treatment of heavy metals and other pollutants by Salmon Friendly projects.

Low-cost Biomonitoring and High-resolution, Scalable Models of Urban Metal Pollution across the Puget Sound Watershed

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- **New low-cost protocol for X-Ray analysis of moss detects zinc and copper concentrations >10 times over background levels in Puget Sound urban areas.**
- **Pollution mapping at 5 m resolution with traffic & land use data shows 50% of metal pollution on 3.3% of land area in Puget Sound region.**

Metals are pollutants of high concern in urban areas due to their persistence, bioavailability, and toxicity. High concentrations of metals threaten aquatic ecosystem functioning and biodiversity, as well as human health. In cities, metals reach waterways directly through atmospheric deposition and indirectly through rain runoff by being leached or picked up as dust after depositing and building up on impervious surfaces. Road dust naturally contains metals from eroded soil but anthropogenic sources like building and road materials, vehicle brake, tire wear, and tailpipe emissions are the main non-point sources of metal pollution in urban areas.

High-resolution estimates of pollutant sources are required to mitigate exposure to toxic compounds by identifying the specific locations and associated site characteristics where the deposition of metals is greatest. Pollution predictions must also be available for large geographic areas to match the spatial scale of funding, management, and some environmental issues. Low resolution (coarse) models cannot identify emission hot spots, as pollution sources are averaged over large spatial units of analysis, and models developed for small spatial extents may not match the ecological scale relevant to species targeted by management. However, the few models that focus on metals usually lack the combination of resolution and scale for this. Therefore, high-resolution, scalable models that describe the relative magnitude of metal pollution sources across space are needed to guide investments across large areas.

This study tackled this gap by demonstrating novel scalable methods to monitor and predict urban metal pollution at high resolution (<5 m) across large areas (10,000–100,000 km^2) to guide pollution reduction and stormwater management. We showcased and calibrated predictive models of zinc, copper, and a synthetic index of pollution for the Puget Sound region of Washington State, U.S., and exemplified their transferability across the entire United States. We leveraged widely and freely available datasets of car traffic characteristics and land use as predictor variables and trained the models with biological monitoring data of metal concentrations in moss growing on tree trunks from >100 trees by in-situ portable X-Ray Fluorescence spectroscopy, a new rapid and low-cost protocol introduced in this study.

Sampling of moss revealed the existence of metal emission hotspots in densely urbanized areas of the Puget Sound watershed with concentrations exceeding background levels by an order of magnitude. Our model predictions show that the pollutant footprint of human development spans across the entire landscape, with 27% of the watershed's land area exhibiting metal concentrations above background levels for at least one of the indicators (copper, zinc, and the pollution index), with copper affecting the greatest area due to further spread away from roads. Our work also reveals that metal pollution is very localized: 50% of the total pollution deposited on land across the Puget Sound watershed is deposited over only 3.3% of the land area for both copper and zinc. Combined with open access datasets to train predictive models, this inexpensive and rapid approach, can support cities in targeting toxic hotspots, thus maximizing environmental returns on mitigation investments.

Metal concentrations were measured by in-situ portable X-Ray Fluorescence (XRF) spectroscopy for moss mats growing on >100 trees based on new protocols introduced in this study. Photo: Mathis Messager

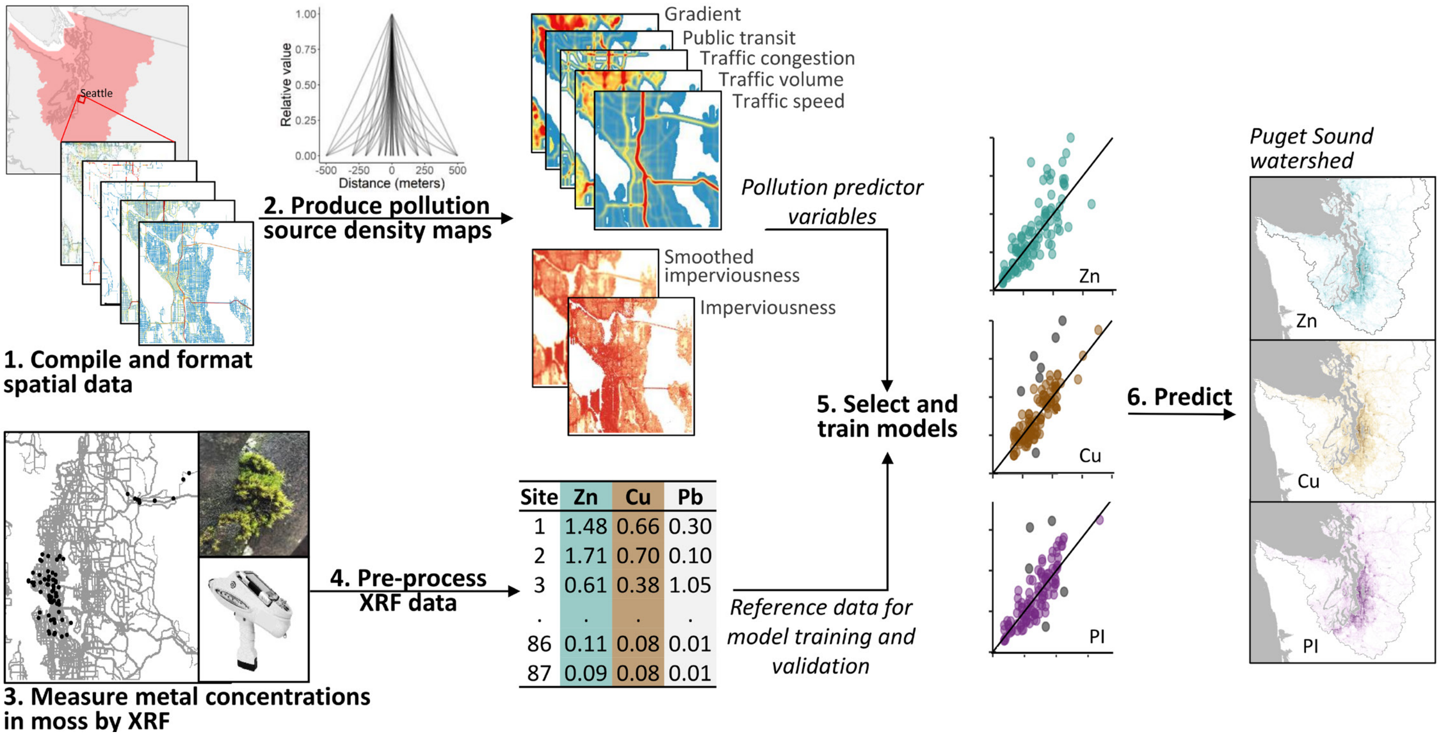

Figure 1. (1) We compiled spatial data for the main variables reported to drive the spatial distribution of urban metal pollution. (2) We generated pollution source “density maps” that simulate the cumulative and diffuse emissions from each road-based source of pollution (3) We measured the concentrations of metals in epiphytic moss at 87 sites across the greater Seattle area using portable X-Ray Fluorescence (XRF). (4) A set of statistical models were developed using the density maps values and land use at the sampling sites as candidate predictor variables and the XRF measurements as response variables (i.e., training data). (5) Three models were developed separately for zinc (Zn), copper (Cu), and a synthetic pollution index (PI). Zn, Cu, and PI were selected as our pollutants of study as these elements are among the most commonly studied and reported metal pollutants in urban environments. (6) Trained models were subsequently used to make spatially continuous predictions of the relative concentrations of each pollutant across the Puget Sound watershed.

Regulating the West Coast Cruise Industry: Canada at the Low Water Mark

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- **Canada is the low water mark for regulating the cruise industry.**
- **Pollution from cruise ships pose significant risks to marine ecosystems and need to be addressed.**

Sewage, greywater, and scrubber washwater from cruise vessels amount to 32 billion liters of dumping off the coast of BC every cruise season that is comparable to the 2019 season (Stand, 2020). When comparing regulations on the West Coast from Alaska to California in the report "Regulating the West Coast Cruise Industry", Canada is found to be the low water mark when it comes to regulating the cruise industry (Stand et al, 2021). Discrepancies in regulations create a perverse incentive for ships to hold their waste to dump off the waters of BC instead of treating it to standards outlined in other jurisdictions.

In the Salish Sea, Puget Sound is protected by a sewage no discharge zone and greywater discharge, that is known to be more contaminated than raw municipal sewage, is limited to fecal coliforms up to 14 Colony Forming Units (CFU)/100mL on average by the US EPA. Once ships cross the water border out of Washington State to Canada, they are legally able to dump sewage contaminated with fecal coliforms up to 250 CFU/100mL within 12 nautical miles of shore. In Canada greywater does not legally have a pollution limit, but must simply be passed through a marine sanitation device in ships built after 2013 within 3 nautical miles of shore, older ships are legally exempt. Voluntary measures asking that sewage and greywater be treated to 14 CFU/100mL fecal coliforms within 12 nautical miles of shore were introduced in April of 2022 by Transport Canada (Transport Canada, 2022), but the voluntary measures are not enforced with audits, and information is limited to self-reporting about compliance without publicly available discharge data.

Both sewage and greywater contain personal care products and pharmaceuticals, which are contaminants of concern for Chinook Salmon according to Canada's Southern Resident Killer Whale Contaminants Technical Working Group (ECCC, 2020). Land based sources of this pollution are being addressed through increased municipal sewage treatment (Canada, n.d.), but pollution from vessels represent a loophole in the regulations and ocean protections.

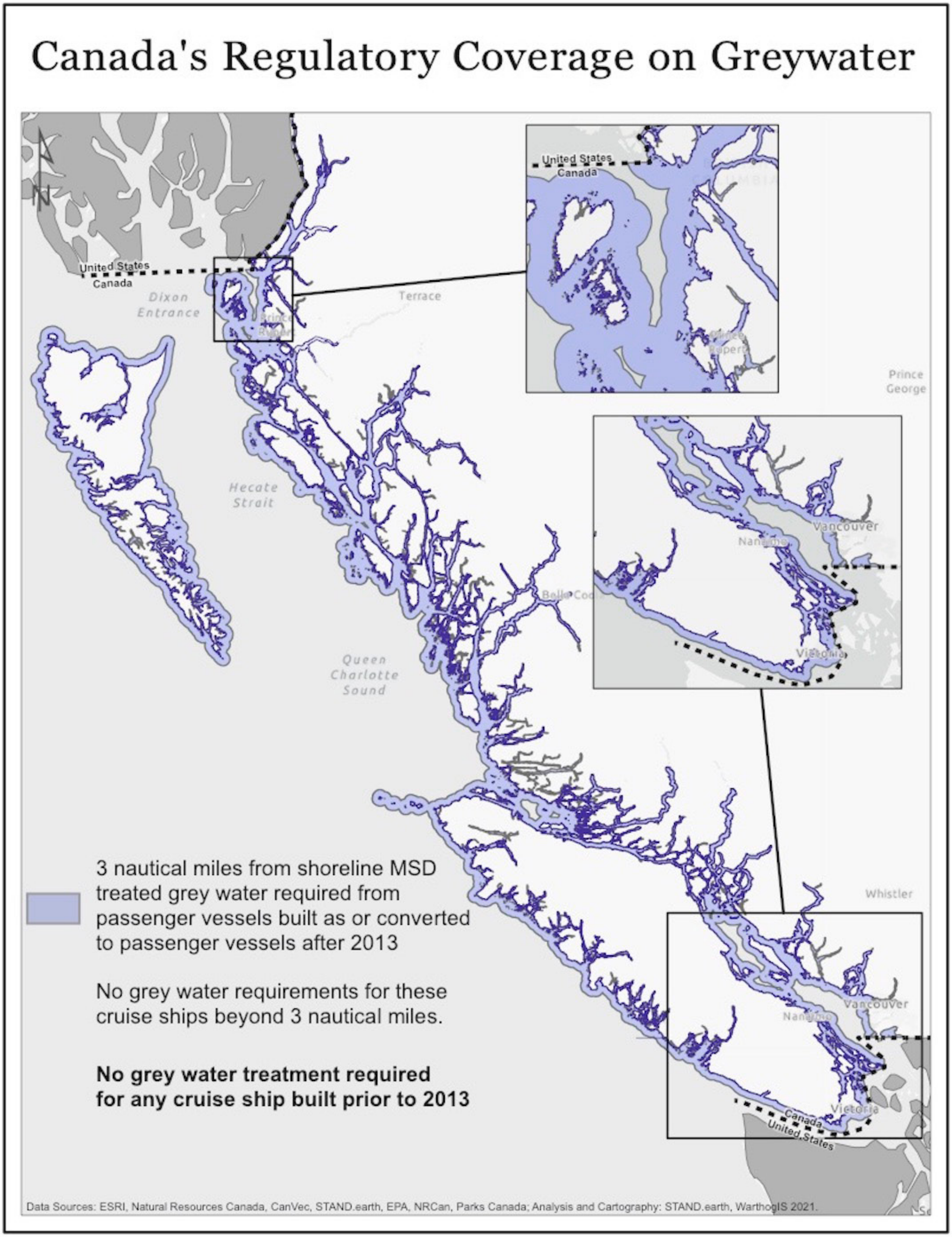
Scrubber washwater makes up over 90% of the volume of dumping from cruise ships (Stand, 2020). Scrubbers take an air pollution problem and turn it into a water pollution issue. Scrubbers primarily rinse the exhaust created by burning high sulfur fuel, like heavy fuel oils, with naturally basic fresh sea water. The resulting washwater is acidic as a result of the newly dissolved sulfur, and also contains PAHs and heavy metals associated with high sulfur fuels (ICCT, 2019).

The washwater contributes directly to ocean acidification and has been shown to be immediately lethal to plankton, and contains toxins that accumulate in the marine environment and wildlife (ICES, 2020). This waste stream is unique in that it is preventable. Ships simply need to burn low sulfur fuels to avoid creating this acidic, toxic and warm waste stream instead of continuing to burn the cheaper heavy fuel oil.

Pollution from cruise ships pose significant risks to marine ecosystems and need to be addressed through prevention, strict treatment requirements that match-up across borders and are designed to protect important marine environments, and independent onboard observers to ensure that regulations are followed away from port inspections.

Please note: in June of 2023 an Interim Order made the voluntary measures described in this publication mandatory. This significantly reduces the low-water mark effect of Canadian regulators. The Interim Order is a mandatory but temporary measure until regulatory changes are made on a permanent basis.

The Zuiderdam cruise ship of the Holland America Line (owned by Carnival Corporation) taken at Canada Place in Vancouver on the 27th of April, 2022. Photo: Solaye Snider, Stand.earth



Map showing Canada's regulatory coverage area for greywater treatment and discharge from large vessels built after 2013.

Pollutants Affecting Endangered Whales and their Prey: The Science behind a New Web Application for Environmental Monitoring Data

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Website: https://www.canada.ca/en/environment-climate-change/services/wildlife-habitat/conservation-funding-success-stories/reducing-contaminants-threat-southern-resident-killer-whales/polluant-affecting-whales-prey-tool.html

Government of Canada Gouvernement du Canada

Government of Canada
Gouvernement du Canada
Pollutants Affecting Whales and their Prey Inventory Tool

- **The Government of Canada has an innovative, interactive tool to estimate contaminants released to the habitats of endangered whales and their prey and to visualize the location of pollutant hot spots.**

Canada's Recovery Strategy and Action Plan under the Species at Risk Act for the Northern and Southern Resident Killer Whales (SRKW, *Orcinus orca*) lists environmental contaminants as a key threat to viability and recovery, and recommends identifying and prioritizing key contaminants, and their sources. The RS also identifies the need to close certain data gaps, such as all potential anthropogenic environmental contaminants to which killer whales and their prey are exposed over time and in space.

To address this recommendation, ECCC developed The Pollutants Affecting Whales and their Prey Inventory Tool (PAWPIT), which shows estimates of pollutant releases by all identified sources within the habitats of Northern and Southern Resident Killer Whales and their primary prey, Chinook salmon. When the development of PAWPIT began, not all sources of pollution within the spatial extent (the area where pollution could affect SRKW, including Resident Killer Whale critical habitat and Chinook salmon distribution in the Fraser Basin and coastal areas) were known, and many locations did not have associated wastewater or ambient monitoring data. To address this knowledge gap, we collated and analyzed available environmental monitoring, literature, and geospatial data. For locations for which data were not available, we used modelling, statistical approaches, and extrapolation to fill data gaps, creating a comprehensive geospatial database of characteristic contributions per sector/activity and ambient contaminant loads.

Visualized using an online mapping tool, this inventory addresses several recommendations for SRKW recovery by identifying sources of priority pollutants, contaminant hot-spots, locations of exceedances of environmental quality guidelines, and enabling a comparison of releases to loads which assists in identifying where data gaps and uncharacterized sources remain. Estimates indicate contaminants of concern are being released by all the sources identified, including runoff, and determined exceedances of environmental quality guidelines for PCBs, some pesticides, and priority metals in the Lower Fraser River.

This information can help guide risk management and future controls. The certainty of the estimates relies on availability of source-specific monitoring data, and some major contributors of contaminants to the spatial extent, such as storm-water runoff, have very little directly associated data, instead relying on estimates derived from modelling and data gathered from the literature. As more data becomes available, work will continue to improve the estimates and update the inventory.

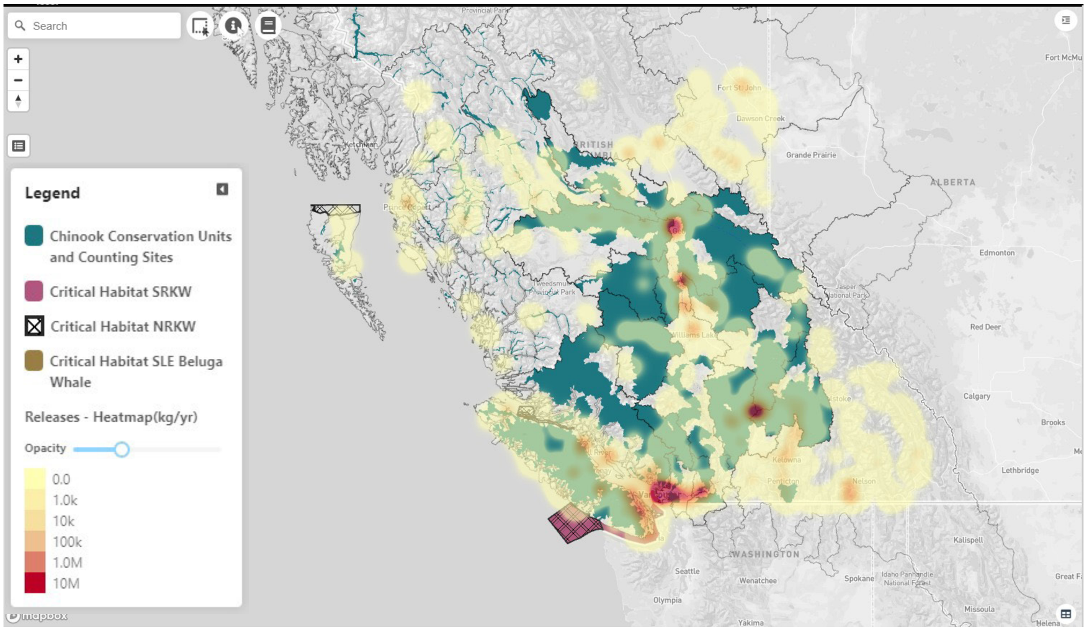

Figure 1. PAWPIT map of British Columbia, Canada showing critical habitat for Southern and Northern Resident Killer Whales, Chinook salmon areas, and a heat map of contaminant releases from point and area sources.

Southern Resident Orca Task Force Report Goal 3: Reduce Exposure of Southern Resident Orcas and their Prey to Contaminants

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The Southern Residents have dwindled to only 73 orcas and are faced with several primary threats. The Southern Resident Orca Task Force identified 49 recommendations within 6 goals or threat areas. Toxics are a primary threat to orcas.

"Southern Residents are exposed to pollutants primarily through their prey and also through transfers from their mothers. Their prey (salmon) are exposed to pollutants in their freshwater and marine habitats throughout their lives. Many pollutants are poorly metabolized, persist in the environment and bioaccumulate and biomagnify in the food web. These toxins can reduce salmon survival by making them more susceptible to disease, which in turn means less food available for the orcas. Toxic contaminants can also reduce immunity and cause reproductive disruption in orcas (Southern Resident Orca Task Force, 2019)."

The purpose of this summary is to highlight contaminant priorities in the WA State Recreation and Conservation Office, Southern Resident Task Force, Final Report and Recommendations (Southern Resident Orca Task Force, 2019) so that participants in the Puget Sound Ecosystem Monitoring Program see where their related toxics monitoring data is important to and overlaps with orca recovery efforts. For more details on recommendations, progress and responsible agencies, see https://www.orca.wa.gov.

Southern Resident Killer Whales. Photo: NOAA Fisheries.

The following is a list of contaminant related recommendations.

- Accelerate the implementation of the ban on polychlorinated biphenyls in state-purchased products and make information available online for other purchasers.
- Identify, prioritize and take action on chemicals that impact orcas and their prey.
- Reduce stormwater threats and accelerate clean-up of toxics harmful to orcas.
- Improve effectiveness, implementation and enforcement of National Pollutant Discharge Elimination System permits to address direct threats to Southern Resident orcas and their prey.
- Increase monitoring of toxic substances in marine waters; create and deploy adaptive management strategies to reduce threats to orcas and their prey.
- Provide sustainable funding for implementation of all recommendations.
- Conduct research, science, and monitoring to inform decision-making, adaptive management and implementation of actions to recover Southern Residents.
- Monitor progress of implementation and identify needed enhancements.
- Protect against regulatory rollbacks at the federal and state level.
- Explore setting minimum standards for local stormwater funding to ensure that all programs have the resources necessary to protect water quality.
- Develop a National Pollutant Discharge Elimination System permit framework for advanced wastewater treatment in Puget Sound to reduce nutrients in wastewater discharges to Puget Sound by 2022.
- Better align existing nonpoint programs with nutrient reduction activities and explore new ways to achieve the necessary nonpoint source nutrient reductions.
- Collect high-quality nutrient data in watersheds to fill key knowledge gaps of baseline conditions.
- Identify and mitigate increased threats to Southern Residents from contaminants due to climate change and ocean acidification. Prioritize actions that proactively reduce exposure where the increased impacts are expected to be most severe.

Current Use Pesticides that Drain into Canadian Tributaries: A Potential Threat to Whale Habitats

Environment and Climate Change Canada / Environnement et Changement climatique Canada
Fisheries and Oceans Canada / Pêches et Océans Canada

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- **High pesticide yields and exceedances of environmental quality guidelines were found in tributaries across the Great Lakes Region, the Fraser River Basin, and the St. Lawrence Region.**
- **Yields, exceedances of environmental quality guidelines, and contaminant runoff estimates were used to identify contaminant hot spots and to inform potential recovery actions.**

Elevated contaminant concentrations in odontocete cetaceans within Canadian waters have been well documented. The Endangered transboundary Southern Resident killer whales (*Orcinus orca*) and St. Lawrence Estuary beluga whales (*Delphinapterus leucas*) face significant threats from high levels of contaminants, both directly and indirectly (reduced prey availability). The Recovery Strategy for the St. Lawrence Estuary belugas, Southern Resident killer whales, and the Threatened Northern Resident killer whales lists contaminants as a key threat to these populations and identifies urban and agricultural runoff and stormwater as contaminant sources. Impervious surfaces, and stormwater drainage systems, allow for efficient delivery of runoff to receiving waters, exposing the whales and their prey to contaminants.

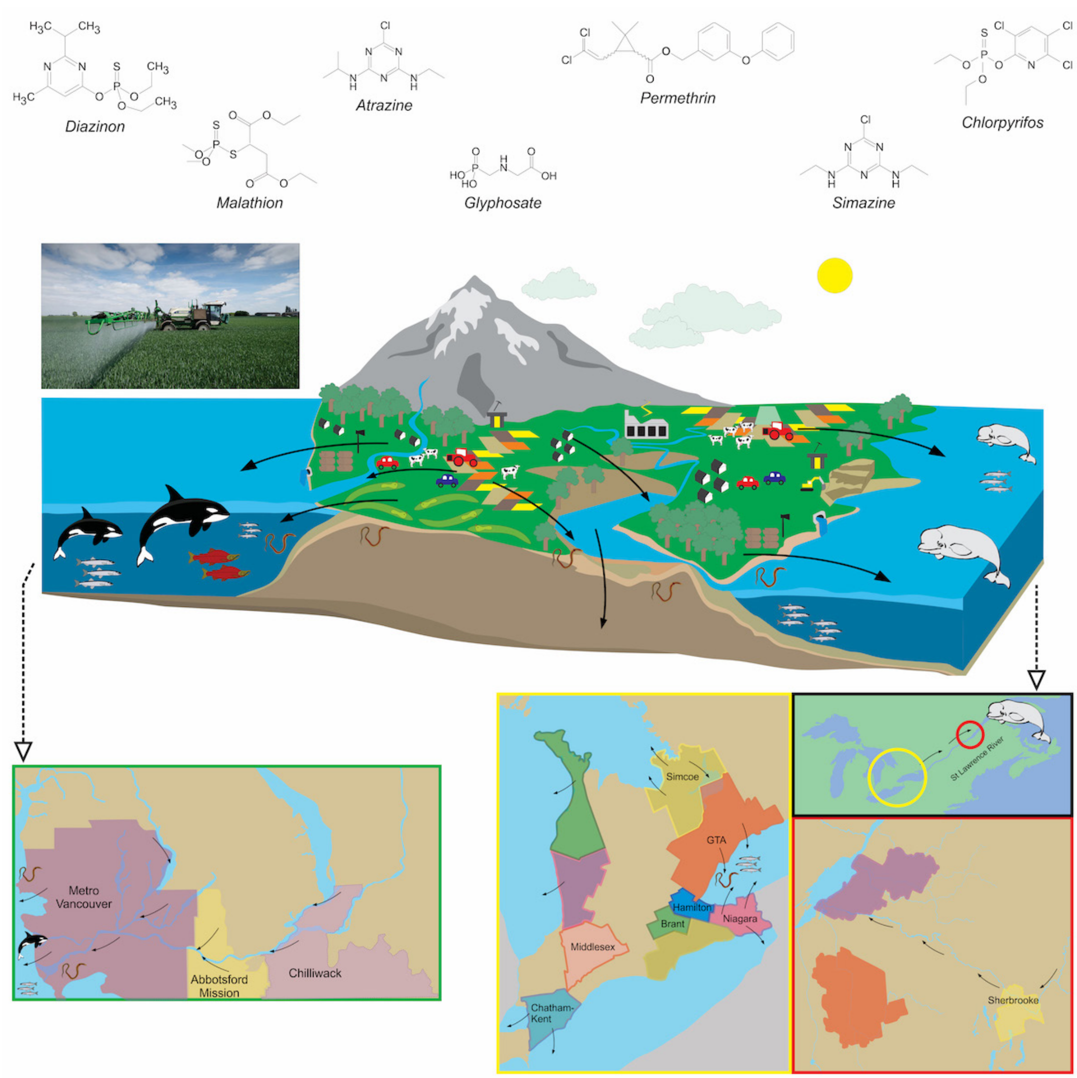

Figure 1. An illustration of our sampling sites and the major tributaries.

Our main objective was to compare levels, loads, and yields of seven current use pesticides (atrazine, chlorpyrifos, diazinon, glyphosate, malathion, permethrin, and simazine) in tributaries within urban and agricultural areas that could impact the habitats of the whales and their prey. Our study design includes three major Canadian metropolitan areas: 1) the Great Lakes Region (15 monitoring stations), 2) the Fraser River Basin (5 monitoring stations), and 3) the St. Lawrence Region (3 monitoring stations). The Great Lakes stations drain into Lake Ontario, which discharges into the St. Lawrence River.

Time series of contaminant concentrations from 2012 to 2019, and corresponding water

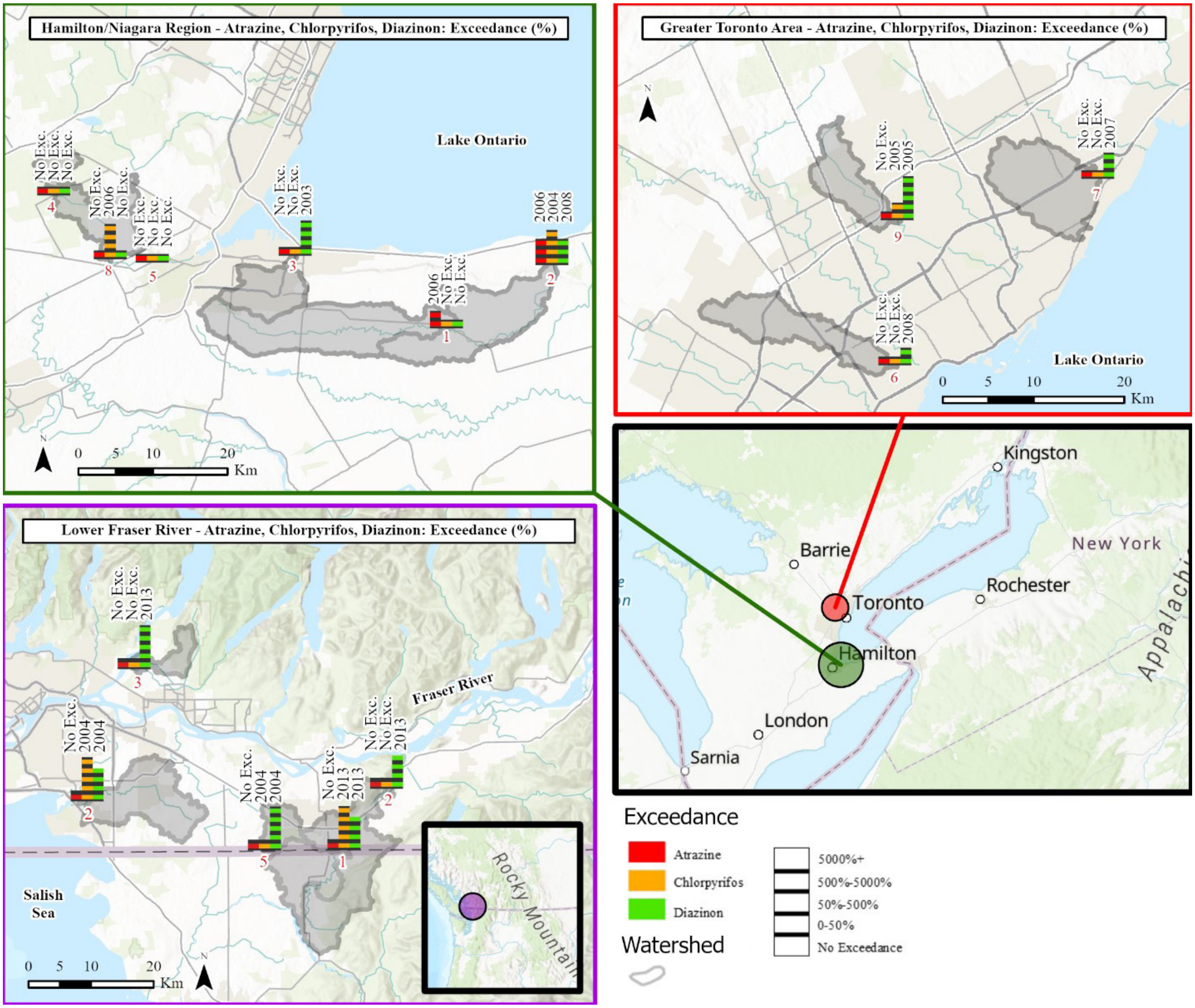

Figure 2. Map of concentration exceedances for atrazine, chlorpyrifos, and diazinon for the Hamilton/Niagara region (top-left panel), the Greater Toronto Area (top-right panel), and the Lower Fraser (bottom-left panel). When exceedances are observed, the year with the maximum exceedance is shown.

flows (discharges, in m^3/s) were used to estimate loads. Water flows were downloaded from Environment and Climate Change Canada's (ECCC's) Hydrometric Station and Network Data (https://wateroffice.ec.gc.ca). Average yields for each pesticide-station combination were calculated by dividing each load by the respective upstream area. The Great Lakes had the highest pesticide yields, followed by the Fraser River, and the St. Lawrence where yields were about 11 times lower than in the Great Lakes. Glyphosate had the highest yields across all sites followed by simazine (Fraser River Basin) and atrazine (Great Lakes and St. Lawrence).

For each pesticide, ECCC's Environmental Quality Guidelines (EQGs) that focused on freshwater and aquatic life with fish as the highest trophic level receptor were assessed. In the Great Lakes, the highest exceedances were observed for diazinon, ranging from roughly 75% to 28,000%. Chlorpyrifos has the second highest exceedance levels ranging from 20% to 2,540%, and atrazine showed lower exceedances, ranging from roughly 20% to 260%. In contrast, the St. Lawrence Region had the lowest exceedance levels, where exceedances were only observed for atrazine and ranged from 35% to 70%. In the Fraser River Basin, diazinon had the highest exceedances, ranging from 62% to 900%.

Contaminant runoff estimates were also generated for average precipitation conditions. Runoff was used to fill in data gaps of contaminant releases from different land types (urban versus agricultural). The Fraser River basin had the highest runoff (40 km^3/yr), while runoff for main tributaries to the St. Lawrence River ranged from 0.2 to 2 km^3/yr. Yields, exceedances of EQGs, and contaminant runoff estimates were used to identify contaminant hot spots and to inform potential recovery actions for the endangered whales and their prey.

GET INVOLVED

Please help us keep our region and you healthy and beautiful.

Use the following links to check on fish, shellfish, and drinking water quality near you and to report oil and other spills.

Washington State

Fish consumption advisories
https://doh.wa.gov/data-and-statistical-reports/washington-tracking-network-wtn/fish-advisories/fish-consumption-advisories-washington-state

Shellfish advisories
https://fortress.wa.gov/doh/biotoxin/biotoxin.html

Drinking water alerts
https://doh.wa.gov/community-and-environment/drinking-water/active-alerts

How to test your water
https://doh.wa.gov/community-and-environment/drinking-water/contaminants/testing-your-water

Report a spill including oil spills
https://ecology.wa.gov/Footer/Report-an-environmental-issue/Report-a-spill

Pollution prevention | Washington

Don't drip and drive
https://www.stormwaterpartners.com/dont-drip-and-drive

Safe medication return (Drug take back)
https://doh.wa.gov/you-and-your-family/healthy-home/safe-medication-return

Household waste disposal sites
https://ecology.wa.gov/waste-toxics/community-waste-toxics/household-hazardous-waste-mrw/find-a-household-hazardous-waste-site

British Columbia

Seafood safety
https://www2.gov.bc.ca/gov/content/industry/agriculture-seafood/fisheries-and-aquaculture/seafood-safety

Shellfish safety
https://www.pac.dfo-mpo.gc.ca/fm-gp/rec/shellfish-coquillages-eng.html

Drinking water alerts
https://www2.gov.bc.ca/gov/content/environment/air-land-water/water/water-quality/drinking-water-quality

Report a spill including oil spills
https://www.crd.bc.ca/education/concerns/report-a-spill

Report All Poachers and Polluters (RAPP) form
https://forms.gov.bc.ca/environment/rapp/

Pollution prevention | British Columbia

Careless Recycling Can Kill
https://recyclebc.ca/hazardous/

Safe medication return
https://www.bcpharmacy.ca/resource-centre/health-issues/medication-return

ACRONYMS AND ABBREVIATIONS

6PPD	N-(1,3- dimethylbutyl)-N'-phenyl-p- phenylenediamine
6PPDQ	6PPD-quinone
ACC	activity cutoff concentration
AVS	acid volatile sulfide
BB	Bellingham Bay
BBAMP	Boundary Bay Ambient Monitoring Program
BC	British Columbia
BCMCA	British Columbia Marine Conservation Analysis
BIBI	benthic index of biotic integrity
BMP	best management practice
BRR	biological response ratio
BSM	bioretention soil media
BWG	Bioretention Work Group
CABS	compost-amended biofiltration swales
CB	Commencement Bay
CBR	critical body residue
CEC	contaminant of emerging concern
CFU	colony forming unit
cPAH	carcinogenic polycyclic aromatic hydrocarbon
CSL	cleanup screening level
CSO	combined sewer overflow
CTWP	creosote-treated wood pilings
CWA	Clean Water Act
DDT	dichlorodiphenyltrichloroethane
DEET	diethyltoluamide
DGT	diffusive gradients in thin-film
DMMP	Dredged Material Management Program
DOC	dissolved organic carbon
DT	design thinking
dw	dry weight
EBD-BSA	Evans Blue dye complexed with bovine serum albumin
EBO	emergency bypass outfall
EC50	half maximal effective concentration
ECCC	Environment and Climate Change Canada
Ecology	Washington State Department of Ecology
EDC	endocrine disrupting compound
eDNA	environmental DNA
EEA	early action area
ELISA	enzyme-linked immunosorbent assay
ENVVEST	EPA's Environmental Investment program; for this document, specific to a project for Sinclair and Dyes Inlets
EPA	U S Environmental Protection Agency
EQG	environmental quality guideline
ESA	Endangered Species Act
FTIR	Fourier transform infrared spectrometer
GIS	geographic information system
GSI	green stormwater infrastructure
HCB	hexachlorobenzene
HCBD	hexachlorobutadiene
HCH	hexachlorocyclohexane
HLB	hydrophilic-lipophilic-balanced
HPAH	high-molecular-weight polycyclic aromatic hydrocarbon
HRMS	high resolution mass spectrometry
KC DNRP	King County Department of Natural Resources and Parks
KCEL	King County Environmental Lab
L	ligand concentration
LC	liquid chromatography
LC50	Lethal concentration for 50% of exposed organisms
LDW	Lower Duwamish Waterway
LID	low impact development
MP	microplastic
NBK	Naval Base Kitsap
NGO	non-governmental organization
NMFS	National Marine Fisheries Service (NOAA)
NOAA	National Oceanic and Atmospheric Administration
NPDES	National Pollutant Discharge Elimination System
NRKW	Northern Resident killer whale
NWFSC	Northwest Fisheries Science Center (NOAA)
OC	organic carbon
OPEO	octylphenol ethoxylates
ORCA	Ocean Research College Academy at Everett Community College
PAH	polycyclic aromatic hydrocarbon
PB	procedural blank
PBDE	polybrominated diphenyl ether
PBT	persistent bioaccumulative and toxic contaminant
PCA	principal component analysis
PCB	polychlorinated biphenyl
PCP	pentachlorophenol
PEG	polyethylene glycol
PES	polyethersulfone
PFAS	per- and poly-fluoroalkyl substances
PFNS	perfluorononanesulfonic acid
PFOA	perfluorooctanoic acid
PFOS	perfluorooctane sulfonate
PI	pollution index
PNEC	predicted no effects concentration
PNNL	Pacific Northwest National Laboratory
PNW	Pacific Northwest
POCIS	polar organic chemical integrative sampler
POM	particulate organic matter
POP	persistent organic pollutant
PPCP	pharmaceutical and personal care product
PPG	polypropylene glycol
PSDDA	Puget Sound Dredged Disposal Analysis program
PSEMP	Puget Sound Ecosystem Monitoring Program
PSNS & IMF	Puget Sound Naval Shipyard and Intermediate Maintenance Facility
PSSST	Puget Sound Stormwater Science Team
RAL	remedial action level
ROD	Record of Decision
RoR	run of the river dam
RSMP	Regional Stormwater Monitoring Program (currently called SAM)
SAM	Stormwater Action Monitoring (previously called RSMP)
SFU	Simon Fraser University
SMS	sediment management standards
SOG	Strait of Georgia
SPAWAR	Systems Command Space and Naval Warfare
SPMD	semi-permeable membrane device
SQS	sediment quality standard
SRKW	Southern Resident killer whale
SSRI	selective serotonin reuptake inhibitor
SVOC	semi-volatile organic compound
SWAC	spatially weighted average concentration
SWMMWW	Stormwater Management Manual for Western WA
TBiOS	WDFW's Toxics-focused Biological Observation System
TCEP	tris-(2-chloroethyl) phosphate
TCP	Toxics Cleanup Program
TEQ	toxic equivalency quotient or toxic equivalent
TM	toxic metal
TOC	total organic carbon
TOP	total oxidizable precursor
TPCBs	total polychlorinated biphenyls
TSS	total suspended solids
TTL	target tissue level
TWP	tire wear particle
UBC	University of British Columbia

UGA	urban growth area
USFWS	United States Fish and Wildlife Service
USGS	United States Geological Survey
UW	University of Washington
UW CEE	UW Civil & Environmental Engineering
UWT CUW	UW Tacoma Center for Urban Waters
UWT SIAS	UW Tacoma School of Interdisciplinary Arts and Sciences
WAMSQS just SMS)	Washington State Marine Sediment Quality Standards (or
WDFW	Washington Department of Fish and Wildlife
WDNR	Washington Department of Natural Resources
WDOH	Washington Department of Health
WQG	water quality guideline
WSC	Washington Stormwater Center
WSDA	Washington State Department of Agriculture
WSDOT	Washington State Department of Transportation
WSU	Washington State University
WWE	wastewater effluent
WWHM	Western Washington hydrology model
WWTP	wastewater treatment plant
XRF	X-Ray fluorescence
YOY	young of the year

Ship Harbor, Anacortes, Washington. Photo: Sylvia Kantor

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