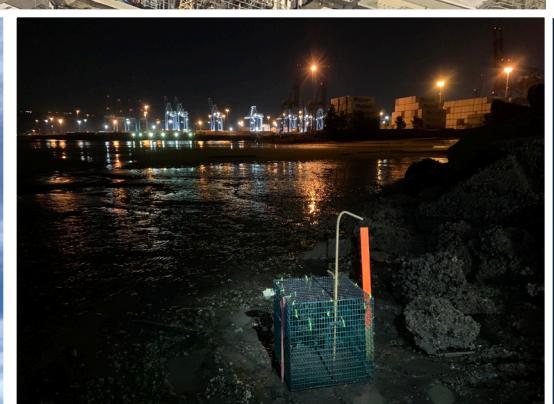
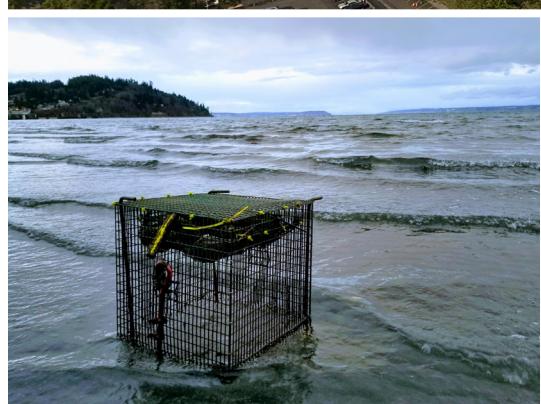


# NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM

## A 2019 Assessment of Trace Metals in the Pacific Northwest



January 2025



NOAA TECHNICAL MEMORANDUM NOS NCCOS 351  
NOAA NCCOS Monitoring and Assessment Branch

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# NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM

## A 2019 Assessment of Trace Metals in the Pacific Northwest

January 2025

Authors:

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NOAA Technical Memorandum NOS NCCOS 351

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## EXECUTIVE SUMMARY

In 2019, the National Oceanic and Atmospheric Administration (NOAA) Mussel Watch Program (MWP) conducted an assessment of the presence, distribution, and concentrations of trace metals in mussels (*Mytilus* species) from the Pacific Northwest's coastal waters in collaboration with the Washington Department of Fish and Wildlife (WDFW) and the Oregon Department of Fish and Wildlife (ODFW). Like the national MWP, the WDFW Toxics-focused Biological Observation System (TBiOS) program utilizes a sentinel-based monitoring approach by collecting and analyzing bivalves as surrogates for coastal water pollution. Mussels and oysters are sessile organisms that filter and accumulate particles from water; therefore, measuring contaminant levels in their tissue is a good indicator of local chemical contamination. Mussel tissue samples from 17 Mussel Watch monitoring sites across the Pacific Northwest coasts of Washington and Oregon were analyzed in this study. Samples were analyzed for 14 trace metals including aluminum (Al), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), nickel (Ni), selenium (Se), silver (Ag), tin (Sn), and zinc (Zn).

The results indicated that trace metals were present at a range of concentrations in mussel tissue collected from waters of the Pacific Northwest. The accumulation of metals in organisms are often analyte and location dependent and can often be influenced by the natural presence of many metals in the environment. In this study, results indicated that the presence and concentration of a specific metal are heavily influenced by its chemistry, sources, fate, and transport. The information presented here may be useful to regional managers because of the long-term nature and geographic spread of the dataset, which could further compliment data from other regional monitoring programs.

The MWP provides unique data that is essential for assessing the health of the Nation's coastal waters through temporal and spatial evaluation of chemical contamination. Studies such as this not only provide needed data and information for the MWP but also address contamination data gaps that are relevant for coastal management. Therefore, coastal managers within the Pacific Northwest would benefit from this study as they develop long-term policies with the aim to protect their managed ecosystems.

## KEY FINDINGS

- Aluminum (Al), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), nickel (Ni), selenium (Se), and zinc (Zn) were detected at 100% of sites.
- Silver (Ag) and tin (Sn) were detected at 0% and 18% of sites, respectively.
- Selenium (Se) was detected at high concentrations relative to historic national data.
- There were no instances of decreasing temporal regional-wide trends.
- Selenium (Se) showed a significantly increasing temporal regional-wide trend.
- Increasing site-specific temporal trends occurred at sites:
  - CBCH (Coos Bay Coos Head) for cadmium
  - PSPT (Puget Sound Port Townsend) for copper
  - YHFC (Yaquina Bay Fogarty Creek) for mercury
  - BBSM (Bellingham Bay Sqalicum Marina Jetty), PSPT (Puget Sound Port Townsend), PSEH (Puget Sound Everett Harbor), PSMF (Puget Sound Mukilteo), EBFR (Elliot Bay Four-Mile Rock), EBDH (Elliot Bay Duwamish Head), SIWP (Sinclair Inlet Waterman Point), CBTP (Commencement Bay Tahlequah Point), and SSBI (South Puget Sound Budd Inlet) for selenium
  - PSHC (Puget Sound Hood Canal) and EBFR (Elliot Bay Four-Mile Rock) for tin
- Decreasing site-specific temporal trends occurred at sites:
  - PSMF (Puget Sound Mukilteo) and PSEF (Puget Sound Edmonds Ferry) for aluminum
  - PSMF (Puget Sound Mukilteo) for copper
  - PSMF (Puget Sound Mukilteo) and PSEF (Puget Sound Edmonds Ferry) for iron
  - BBSM (Bellingham Bay Sqalicum Marina Jetty), PSMF (Puget Sound Mukilteo), PSEF (Puget Sound Edmonds Ferry), and EBFR (Elliot Bay Four-Mile Rock) for lead
  - PSMF (Puget Sound Mukilteo) and PSEF (Puget Sound Edmonds Ferry) for manganese
  - BBSM (Bellingham Bay Sqalicum Marina Jetty) and EBFR (Elliot Bay Four-Mile Rock) for mercury
  - PSMF (Puget Sound Mukilteo) and PSEF (Puget Sound Edmonds Ferry) for nickel
  - WBNA (Willapa Bay Nahcotta) for selenium
  - EBFR (Elliot Bay Four-Mile Rock) for silver

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## COMMONLY USED ACRONYMS

dw	dry weight
EPA	Environmental Protection Agency
FDA	Food and Drug Administration
g	gram
MDL	method detection limit
MWP	Mussel Watch Program
NCCOS	National Centers for Coastal Ocean Science
NOAA	National Oceanic and Atmospheric Administration
NS&T	National Status and Trends
ODFW	Oregon Department of Fish and Wildlife
OR	Oregon
PSMM	Puget Sound Mussel Monitoring
SD	standard deviation
$\mu$ g	microgram
US	United States
TBiOS	Toxics-focused Biological Observing System
WA	Washington
WDFW	Washington Department of Fish and Wildlife

# Introduction

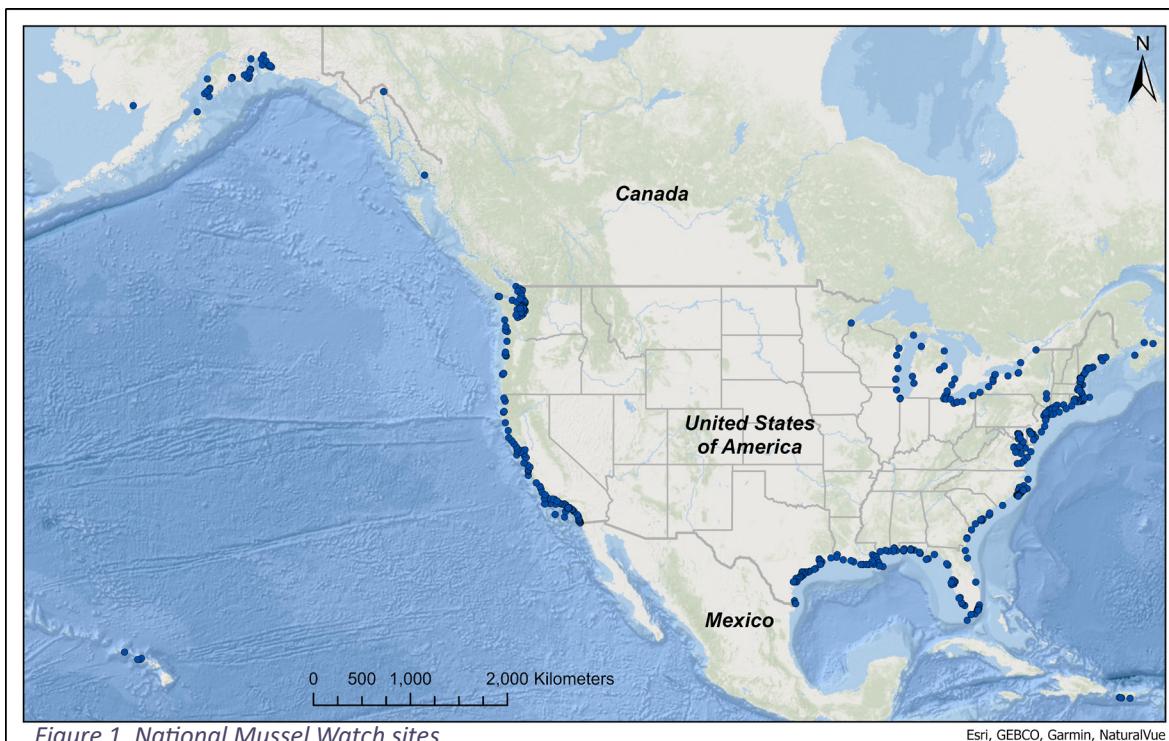
## 1.0 HISTORY OF MUSSEL WATCH PROGRAM

The National Mussel Watch Program (MWP), which began in 1986, was designed by the National Oceanic and Atmospheric Administration (NOAA) to monitor the Nation's coastal waters for chemical contaminants and biological indicators of water quality. The MWP was established in response to a legislative mandate under Section 202 of Title II of the Marine Protection, Research and Sanctuaries Act (MPRSA) (33 USC 1442), which called on the Secretary of Commerce to initiate a continuous monitoring program, among other activities. The MWP design is based on the periodic collection and analysis of bivalves (oysters and mussels) and sediment from a network of monitoring sites located throughout the nation's coastal zones. To date, NOAA's MWP is one of the longest-running, continuous coastal monitoring programs.

The MWP monitoring sites are found along all of the United States (US) coastlines including Alaska, the Great Lakes, Hawaii, and in territories such as Puerto Rico. Different target bivalves are used as sentinel species. Mussels and oysters are sessile organisms that filter and accumulate particles from water and their body burden reflects ambient concentrations; therefore, measuring contaminant levels in their tissue is a good indicator of local chemical contamination (Farrington, 1983). Mussels (*Mytilus* species) are collected from the North Atlantic and Pacific coasts, oysters (*Crassostrea virginica*) are collected from the mid-Atlantic (Delaware Bay) southward and along the Gulf Coast, the invasive zebra and quagga mussels (*Dreissena* species) are collected from the Great Lakes, mangrove oysters (*Crassostrea rhizophorae*) are collected from Puerto Rico, and Hawaiian oysters (*Dendostrea sandvicensis*) are collected from Hawaii.

A fundamental challenge faced by any long-term environmental monitoring program is how to evolve in response to changing environmental conditions and anthropogenic drivers. In 2013, due to budgetary constraints, the National Centers for Coastal Ocean Science (NCCOS) undertook the task of re-designing the MWP, moving from a nationwide annual monitoring approach to the rotating regional monitoring model that is currently employed. The regional approach allows the program to improve its presence in coastal communities by increasing interaction with local stakeholders, integrating inputs from coastal resource managers, and providing specific data needs to help fill local data gaps. By making adaptive changes and leveraging regional partnerships, the program has increased its scientific relevance and reputation and has evolved to include more than 300 monitoring sites (Figure 1) and nearly 600 chemical contaminants including metals, legacy organic compounds, and contaminants of emerging concern (CECs).

The MWP provides unique data that is vital to evaluating the health of the nation's estuarine and coastal waters and bivalves, particularly describing the levels of chemical contamination. The MWP dataset allows for temporal and spatial evaluation of regional and national changes in chemical distribution, including CECs as their potential risks are identified. The programs' long-term data supports the assessment of impacts of unforeseen events such as oil spills and hurricanes, the evaluation of sanctuary statuses, the analysis of resource and ecosystem service trends, and the evaluation of the effectiveness of regulations that ban toxic chemicals or support legislation such as the Clean Air and Clean Water Acts.



# Introduction

## 2.0 INTRODUCTION

The MWP has a network of long-term monitoring sites in the Pacific Northwest that were analyzed in this study. Sites spanned the Pacific coastlines of the states of Washington and Oregon and many were clustered within Puget Sound, WA. Puget Sound is the Washington portion of the Salish Sea and is an economically, culturally, and ecologically important region, lending to the importance of monitoring its health. The Puget Sound region is home to over 100,000 registered recreational vessels and over 3,000 commercial vessels, therefore supporting a large watersport culture as well as one of the top shellfish-producing industries in the US (WSDE, 2024). While highly critical to the local lifestyle and economy, Puget Sound is also uniquely sensitive to pollution due to its naturally narrow shape, which limits water circulation throughout the region (WSDE, 2024). Contaminants enter Puget Sound by a number of pathways including nonpoint sources such as runoff, air deposition, and marina and ferry terminals, as well as from point sources such as stormwater and wastewater outfalls (WSDE, 2024). With such high economic outputs of aquatic resources, it is critically important to track chemical contamination in Puget Sound, especially in regards to recovery and remediation efforts (Puget Sound Partnership, 2024). Specifically, it is important to track contamination in nearshore areas where many toxins are able to enter the waterways as land and water intersect.

The MWP sites in Washington and Oregon have been assessed and monitored by the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends Program (NS&T) for resource and ecosystem management and production since 1986 (Kimbrough et al., 2008; Figure 2). The MWP has forged a long-term collaboration with state resource managers, particularly the Oregon and Washington Departments of Fish and Wildlife (ODFW and WDFW, respectively), both of which support local and regional ecosystem monitoring programs. In Oregon, ODFW monitors shellfish stock and harvest, and in collaboration with their Department of Agriculture (ODA), routinely conducts shellfish monitoring for biological toxins such as domoic acid, which causes amnesic shellfish poisoning (ASP), and saxitoxins, which cause paralytic shellfish poisoning (ODFW, 2024). In Washington, WDFW manages shellfish beds and their harvest and also routinely collects and uses mussels as surrogates for nearshore environmental health monitoring in their Toxics-focused Biological Observation System (TBiOS) program (WDFW, 2024). TBiOS is a Puget Sound monitoring and research program based on a biological framework that evaluates the extent and magnitude of toxic contaminants, and their health effects, in marine biota (WDFW, 2024). In partnership with a number of other state, county, and city agencies and programs, as well as several tribes and NGO partners, TBiOS leads the cooperative Washington State Mussel Watch (WAMW) program which utilizes active biomonitoring of transplanted mussels to track contaminants at a range of sites in the Puget Sound nearshore (Langness and Nordstrom, 2024; Langness et al., 2022; Langness and West, 2020; Lanksbury et al., 2017; Lanksbury et al., 2014).

Programs such as these, which value the collection and assessment of long-term water quality monitoring, have provided relevant data and information to coastal managers and the scientific community with a primary focus on legacy organic contaminants. These legacy pollutants are routinely monitored and regulated and include trace elements ("trace metals") and persistent organic pollutants such as butyltins, chlordanes, chlorobenzenes, dichlorodiphenyltrichloroethane (DDTs), dieldrins, endosulfans, hexachlorocyclohexane (HCHs), mirex, polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs). Trace metals occur naturally in earth's crust, so their sources of contamination are not always anthropogenic or of concern, and high concentrations can often be linked to natural enrichment or biogenic sources.

In 2019, the MWP leveraged their long-term partnerships with ODFW and WDFW to conduct a comprehensive assessment of contaminants in the Pacific Northwest. The study was designed within the MWP regional monitoring approach framework, which balances the importance of a large geographic site network with the cost of broad contaminant surveys. Additionally, in Washington, the survey benefited from stakeholder inputs particularly from groups involved in the WAMW program, including the Washington Department of Ecology's Stormwater Action Monitoring (SAM) program, the Washington Department of Natural Resources' (DNR) Aquatic Resources Division, Snohomish County Surface Water Management, King County DNR Water and Land Resources Division, Kitsap County Public Works, Jefferson County Public Health, City of Bellingham Natural Resources Division, Port of Tacoma, Jamestown S'Klallam Tribe, Makah Tribe, Sustainable Bainbridge, and the University of Washington. Through these partnerships, study capacity was increased for all parties, as scientific activities of mutual interest were coordinated for efficient delivery of needed data and information. The objectives of this study were to 1) assess the magnitude and distribution of trace metals historically assessed by the MWP; 2) compare contamination in the Pacific Northwest in 2019 to previous studies in the same region and nationally; and 3) make data electronically available to coastal resource managers in the Pacific Northwest region.

# Methods

## 3.0 METHODS

### 3.1 Study Area and Sampling Design

The MWP has 41 long-term monitoring sites in coastal waters in the Pacific Northwest. Historically, monitoring sites were selected in locations with abundant bivalve populations to allow for repetitive sampling and to convey information about the degree of chemical contamination in the general area over time. The sites were not randomly selected nor designed to target specific pollution sources. The sampling design for this study included the selection of sites from both the MWP and the WDFW TBiOS program that met both programs' monitoring needs.

Sample collection at 19 MWP sites and 40 WDFW TBiOS sites was conducted by the Oregon and Washington Departments of Fish and Wildlife (ODFW and WDFW) following standard protocols utilized by the MWP (Apeti et al., 2012) in primarily January - February 2020. However, these samples were collected as part of fiscal year 2019 and sampling was only conducted in early 2020 due to resource availability and scheduling. Thus, the data found in this study is reported as 2019 data for continuity within the MWP's long term data collection pattern. Mussel samples (*Mytilus* species) were collected from 59 sites (Figure 2). Nine additional MWP sites were initially identified for collection but were unsuccessful due to lack of bivalve abundance, adverse site conditions, or lack of resources/permission (Table 1). In this study, blue mussels from the 17 of the 19 MWP sites were collected and analyzed for 22 trace metals, 14 of which have been assessed historically and are included in the main body of this report. Mussel samples collected at sites CRSJ and CBRP did not yield enough material to conduct metals analysis and so are not included in this report. The 40 WDFW sites were not analyzed for trace metals by the MWP; however, raw data obtained from the Northwest Fisheries Science Center laboratory for the WDFW sites is summarized in the Appendices (Table A5-A7).

In this study, MWP incorporated a sampling design that combines the collection of wild mussels in Oregon and transplanted mussels in Washington. The WDFW has efficiently adopted a transplanted-mussel monitoring design as part of their WAMW program and in order to capitalize on the resources available to both programs, the MWP transitioned their historically wild mussel sites to transplanted mussel sites (Lanksbury et al., 2014). More research needs to be conducted regarding the appropriateness of temporal comparisons when this transition from wild to transplanted mussels is a factor, but in this report, it is just important to note that data from sites in Washington used transplanted mussels and sites in Oregon used wild mussels, denoted in figures as dark and light gray, respectively. The transplanted mussel deployment and retrieval followed the WAMW program's standard operating procedure and used both scientists and citizen science volunteer groups to conduct field operations (Lanksbury and Lubliner, 2015). A clean aquaculture source of native bay mussels (*Mytilus trossulus*) were transplanted to their monitoring locations in anti-predator cages during the fall/winter of 2019/2020 (Langness et al., 2022; PSEMP Toxics Work Group, 2023). Mussel cages were deployed at approximately the 0.0 (zero) foot mean lower low water mark during low tides in late October 2019 and retrieved in late January 2020, for a four month exposure period. WDFW also collected samples from the aquaculture source at the start of the study (predeployment). These samples are referred to as the Baseline mussel sample and was reported within the WDFW reports and summarized in the appendices of this report to provide an initial condition/concentration of contaminants in mussels (Table A5; Langness et al., 2022).

# Methods

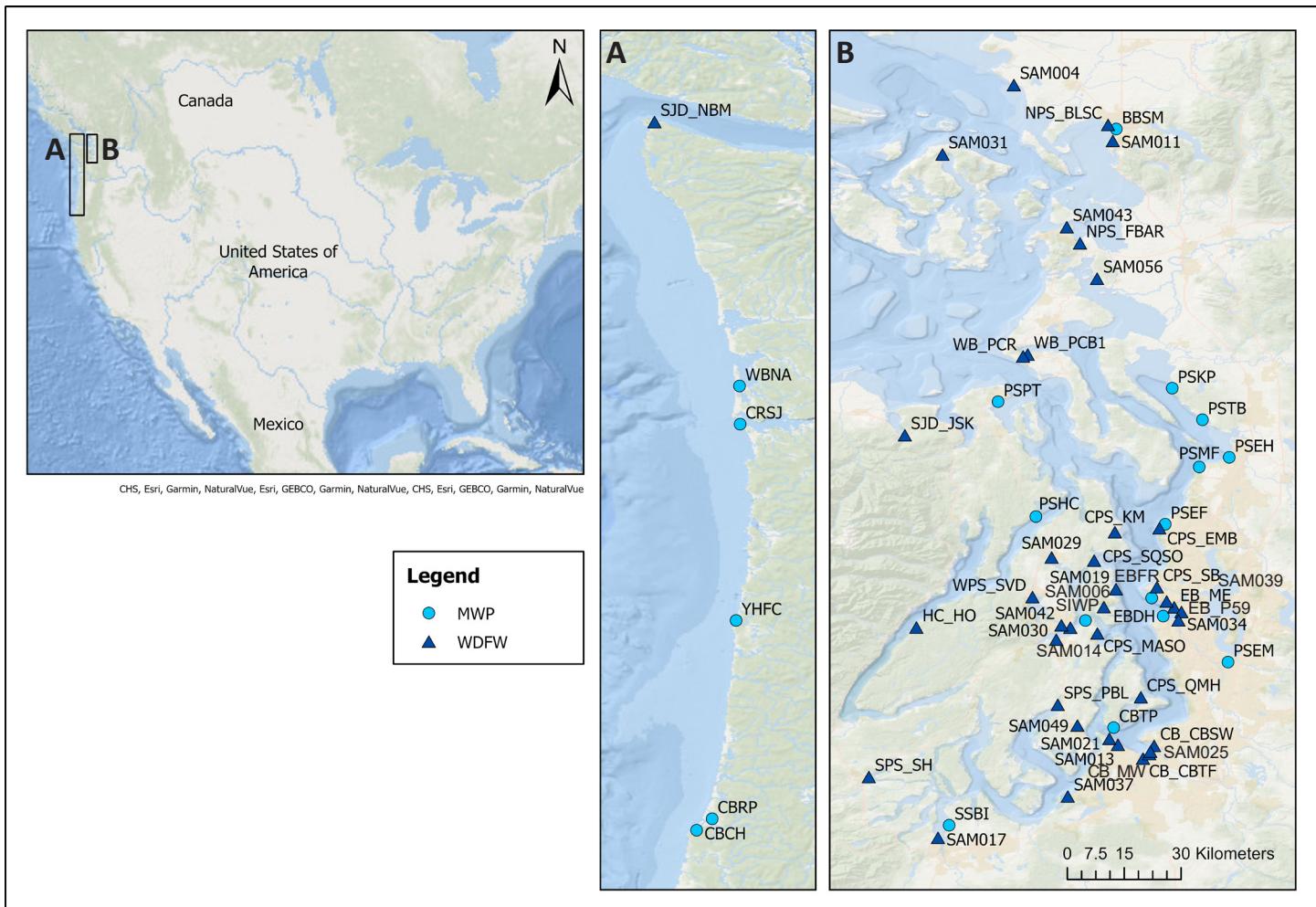


Figure 2. Map of sites in the Pacific Northwest region sampled in 2019 and their respective program origins.

## 3.2 Analytical Methods

Chemical analyses in mussel tissue followed procedures established by the NOAA National Status and Trends (NS&T) Program. Detailed descriptions of analytical methods for trace metals analyzed in this study by NCCOS' Ecotoxicology Laboratory in Charleston can be found in EPA (1992, 1994, 1996). Laboratory results were subjected to regular NS&T performance-based quality assessment and quality control procedures for data accuracy and precision. Trace metals data is presented in micrograms per gram of dry tissue ( $\mu\text{g/g dw}$ ). The laboratory at which analyses were conducted has changed over time; however, results from laboratory intercomparisons did not identify any significant concerns regarding data comparability. In the MWP, trace metal contaminants have been analyzed by TDI-Brooks International since 2000, before being taken over by the NOAA Ecotoxicology lab in Charleston in 2014. The data in this report will be archived and publicly available at NOAA's National Centers for Environmental Information (NCEI) (<https://www.ncei.noaa.gov/>). For all contaminant classes, a background summary ("Chemical Description") and analysis summary ("Results Summary") can be found throughout this document.

## 3.3 Data Analysis

Data management and analysis were conducted using a combination of R version 4.2.1 (R Core Team, 2022), Microsoft Excel (Microsoft Corporation, 2018), ArcGIS (ESRI, 2011), and JMP12 Software (JMP, 2022) following the standards detailed in the Quality Assurance Project Plan. The NCCOS Ecotoxicology Laboratory (Ecotox Lab) in Charleston, SC report metals data in dry weight ( $\mu\text{g/g dw}$ ), therefore, all contaminant concentrations are reported as such throughout this document.

Concentrations of all compounds were blank corrected and any values below the Method Detection Limit (MDL) were categorized as undetected and were assigned a value of 0. The MDL is defined as the lowest concentration able to be

# Methods

detected by the analytical instrument or method. Metals that are not routinely measured (antimony, barium, beryllium, cobalt, lithium, thallium, uranium, vanadium), and therefore don't relate to any historical data, were not included in the main body of this report but their concentrations can be found in the appendices, when detected (Figures A1 - A4). Data for each metal from sites in 2019 were statistically analyzed at site and regional scales using a Spearman's Rank test. Spearman's rank correlation was used to evaluate whether concentrations co-varied predictably as a function of time (Zar, 1999), and therefore imply a significant increasing or decreasing temporal trend. The Spearman's rank correlation procedure is a nonparametric technique that is free of assumptions about concentrations being normally distributed with a common variance about sites (Kimbrough et al., 2008). The variables used for the Spearman's test were year and concentration. Regional temporal trends were also depicted using a three-point moving average of the yearly mean for all sites combined.

Additionally, contamination levels detected at sites in 2019 were compared to national long-term NOAA NS&T monitoring data. The NS&T data used for comparison comprises mussel tissue sample concentrations collected by the national MWP since the program's initiation in 1986. This comparison was done using a multivariate cluster analysis (the Ward method) on the concentrations detected for each compound. This analysis clusters contaminant concentrations into significantly different groups such that values contained within a group are more like each other than any other value of a different group. The categories derived from the clusters were not representative measurements that have exceeded any regulatory thresholds; rather, they denoted concentrations that were significantly higher than the preceding category. For each contaminant class, sites were clustered into three groups to represent high, medium, and low contamination relative to all data collected within the MWP. If the resulting high cluster had fewer than 10 data points, those points were temporarily removed as "outliers" (due to the large sample size of this dataset), the data were re-clustered, and the outliers were added back in as part of the "high" cluster before resulting boxplots were created. Clustering results were reported in a map on the Summary page of each compound. Any instances of "not detected" contaminants were differentiated from low contamination on the map.

To provide further context, cluster analysis results were summarized for all metals per site assessed in this study in two heatmaps showing (1) cluster results of the 2019 study sites respective to concentrations found within the sites assessed in this study and (2) cluster results of the 2019 study sites respective to national long-term NOAA NS&T monitoring data. The clustering process for comparing contaminant data between sites only analyzed within the 2019 Pacific Northwest study was similar to the process described for the nationwide, historic clustering with two exceptions to account for the smaller sample size. The concentrations were similarly clustered into groups of different contamination levels, but "outliers" were not removed (as described previously) and sites were not forced into three clusters (high, medium, and low) if they statistically defaulted to fewer than three clusters, in which case they were labeled as lower and higher groups.

After this cluster analysis, cluster values of each compound for each site for both datasets were summed and normalized by the maximum value possible at that site, taking the number of compounds assessed at each site and the maximum cluster value possible based on the cluster analysis results into account (i.e.,  $(\text{sum cluster values}) / (\# \text{ chemical classes analyzed} * \text{maximum cluster value possible}) * 100$ ) (Table A2; Table A3). These normalized values were then clustered using the Ward method to generate groups of sites with statistically different degrees of overall contamination and were presented in a map. The 2019 sites only dataset was, again, treated slightly differently than the nationwide, historic dataset due to the difference in sample size. The nationwide, historic dataset was clustered into five groups of sites with statistically different degrees of overall contamination (not detected, low, medium, high, and very high), whereas the 2019 sites only dataset was not forced into five clusters if they statistically defaulted to fewer than five clusters, in which case they were labeled just as lower and higher groups.

# Methods

Table 1. Mussel Watch sites selected for 2019 Pacific Northwest survey. ● signifies the site was analyzed for metals in 2019. An attempt was made to sample 68 sites in this survey, but only 59 sites yielded mussels to be analyzed (\*). Only 17 were analyzed for metals. OR - Oregon, WA - Washington.

Site	State	Program	General Location	Specific Location	Latitude	Longitude	Tissue Sampled?
BBSM	WA	MW	Bellingham Bay	Squalicum Marina Jet.	48.75312	-122.49865	●
CB_CBSW	WA	WDFW	Commencement Bay	Skookum	47.29000	-122.41000	
CB_CBTF	WA	WDFW	Commencement Bay	Thea Foss Waterway	47.25919	-122.43469	
CB_MW	WA	WDFW	Commencement Bay	Milwaukee Waterway	47.26813	-122.42163	
CBCH	OR	MW	Coos Bay	Coos Head	43.35000	-124.33083	●
CBRP	OR	MW	Coos Bay	Russell Point	43.43133	-124.22117	
CBTP	WA	MW	Commencement Bay	Tahlequah Point	47.33101	-122.50498	●
CPS_EMB	WA	WDFW	Central Puget Sound	Edmonds Marina Beach	47.80604	-122.39670	
CPS_KM	WA	WDFW	Central Puget Sound	Kingston Marina	47.79470	-122.49986	
CPS_MASO	WA	WDFW	Central Puget Sound	Manchester	47.55622	-122.54281	
CPS_QMH	WA	WDFW	Central Puget Sound	Quartermaster Harbor	47.40500	-122.44070	
CPS_SB	WA	WDFW	Central Puget Sound	Salmon Bay Commodore Park	47.66630	-122.40180	
CPS_SQSO	WA	WDFW	Central Puget Sound	Suquamish Stormwater Outfall	47.72961	-122.55062	
CRSJ	OR	MW	Columbia River	South Jetty	46.22867	-124.02317	
EB_ME	WA	WDFW	Elliott Bay	Myrtle Edwards	47.61862	-122.36108	
EB_P59	WA	WDFW	Elliott Bay	Seattle Aquarium Pier 59	47.60700	-122.34200	
EBDH	WA	MW	Elliott Bay	Duwamish Head	47.59543	-122.38760	●
EBFR	WA	MW	Elliott Bay	Four-Mile Rock	47.63888	-122.41280	●
GHWJ*	WA	MW	Grays Harbor	Westport Jetty	46.91222	-124.11757	
HC_HO	WA	WDFW	Hood Canal	Holly	47.57058	-122.97154	
JFCF*	WA	MW	Strait of Juan de Fuca	Cape Flattery	48.34000	-124.68000	
NPS_BLSC	WA	WDFW	North Puget Sound	Bellingham Little Squalicum Creek	48.76390	-122.51750	
NPS_FBAR	WA	WDFW	North Puget Sound	Fidalgo Bay Aq Reserve	48.48245	-122.58388	
PSCC*	WA	MW	Puget Sound	Cavalero County Park	48.17611	-122.47883	
PSEC*	WA	MW	Puget Sound	Everett Cemex	48.02000	-122.22000	
PSEF	WA	MW	Puget Sound	Edmonds Ferry	47.81407	-122.38250	●
PSEH	WA	MW	Puget Sound	Everett Harbor	47.97260	-122.22977	●
PSEM	WA	MW	Puget Sound	Edmonds Marina	47.81108	-122.38813	●
PSHC	WA	MW	Puget Sound	Hood Canal	47.83252	-122.68741	●
PSKP	WA	MW	Port Susan	Kayak Point	48.13675	-122.36728	●
PSMF	WA	MW	Puget Sound	Mukilteo	47.94968	-122.30158	●
PSPT	WA	MW	Puget Sound	Port Townsend	48.10454	-122.77775	●
PSSS*	WA	MW	Puget Sound	South Seattle	47.52993	-122.40157	
PSTB	WA	MW	Puget Sound	Tulalip Bay	48.06170	-122.29390	●

\*GHWJ: Deployed and found unanchored and washed upland the next day. Deemed unsuitable due to the high energy environment.

\*JFCF: No attempt to deploy was made. Deemed unsuitable due to bedrock substrate preventing cage anchoring and the high energy environment.

\*PSCC: Deployed but was not found during the retrieval period. Presumed lost in a winter storm.

\*PSEC: No attempt to deploy was made. During site evaluations, unable to gain permission from the landowners to access the site area.

# Methods

Table 1 cont. Mussel Watch sites selected for 2019 Pacific Northwest survey. ● signifies the site was analyzed for metals in 2019. An attempt was made to sample 68 sites in this survey, but only 59 sites yielded mussels to be analyzed (\*). Only 17 were analyzed for metals. OR - Oregon, WA - Washington.

Site	State	Program	General Location	Specific Location	Latitude	Longitude	Tissue Sampled?
SAM004	WA	WDFW	North Puget Sound	Cherry Point	48.85755	-122.73630	
SAM006	WA	WDFW	Central Puget Sound	Eagle Harbor Dr	47.61871	-122.52759	
SAM011	WA	WDFW	Bellingham Bay	South Bay Trail	48.72568	-122.50606	
SAM013	WA	WDFW	Commencement Bay	Ruston Way	47.29253	-122.49510	
SAM014	WA	WDFW	Central Puget Sound	Point Heron East	47.57101	-122.60648	
SAM017	WA	WDFW	South Puget Sound	Budd Inlet, West Bay	47.06878	-122.91975	
SAM019	WA	WDFW	Central Puget Sound	Skiff Point	47.66154	-122.49952	
SAM021	WA	WDFW	Central Puget Sound	Point Defiance Ferry	47.30376	-122.51146	
SAM025	WA	WDFW	Commencement Bay	Blair Waterway	47.27578	-122.41737	
SAM029	WA	WDFW	West Puget Sound	Liberty Bay	47.74626	-122.65216	
SAM030	WA	WDFW	Sinclair Inlet	Kitsap St Boat Launch	47.54111	-122.64058	
SAM031	WA	WDFW	North Puget Sound	Eastsound Fishing Bay	48.69258	-122.91127	
SAM034	WA	WDFW	Elliott Bay	Harbor Island, Pier 17	47.58771	-122.35063	
SAM037	WA	WDFW	South Puget Sound	Saltar's Point	47.16998	-122.61066	
SAM039	WA	WDFW	Elliott Bay	Smith Cove Terminal 91	47.63237	-122.37869	
SAM042	WA	WDFW	West Puget Sound	Evergreen Rotary Park	47.57617	-122.62899	
SAM043	WA	WDFW	North Puget Sound	N Avenue Park	48.52109	-122.61104	
SAM049	WA	WDFW	Central Puget Sound	Donkey Creek Delta	47.33837	-122.59049	
SAM056	WA	WDFW	North Puget Sound	Fidalgo Island Swinomish Res	48.39735	-122.53921	
SIWP	WA	MW	Sinclair Inlet	Waterman Point	47.58447	-122.57039	●
SJD_JSK	WA	WDFW	Strait of Juan de Fuca	Jamestown S'Klallam	48.02479	-122.99814	
SJD_NBM	WA	WDFW	Strait of Juan de Fuca	Neah Bay Marina	48.37693	-124.62760	
SPS_PBL	WA	WDFW	South Puget Sound	Purdy Burley Lagoon	47.38698	-122.63671	
SPS_SH	WA	WDFW	South Puget Sound	Shelton	47.21469	-123.08535	
SSBI	WA	MW	South Puget Sound	Budd Inlet	47.09929	-122.89466	●
TBHP*	OR	MW	Tillamook Bay	Hobsonville Point	45.54720	-123.90750	
WB_PCB1	WA	WDFW	Whidbey Basin	Penn Cove Baseline #1	48.21863	-122.70797	
WB_PCR	WA	WDFW	Whidbey Basin	Penn Cove Reference	48.21423	-122.71897	
WBNA	WA	MW	Willapa Bay	Nahcotta	46.49819	-124.02704	●
WIPP*	WA	MW	Whidbey Island	Possession Point	47.90535	-122.37787	
WPS_SVD	WA	WDFW	West Puget Sound	Silverdale Dyes Inlet	47.64279	-122.69671	
YBOP*	OR	MW	Yaquina Bay	Oneatta Point	44.57520	-123.98900	
YHFC	OR	MW	Yaquina Bay	Fogarty Creek	44.83700	-124.05200	●
YHYH*	OR	MW	Yaquina Bay	Yaquina Head	44.67630	-124.07800	

\*PSSS: Deployed but was not found during the retrieval period. Presumed lost in a winter storm.

\*WIPP: Deployed but found unanchored and far away from original site during the retrieval period. All the mussels were dead.

\*TBHP: Inadequate bivalve abundance / inaccessible.

\*YBOP: Inadequate bivalve abundance / inaccessible.

\*YHYH: Inadequate bivalve abundance / inaccessible.

# Results - Aluminum (Al)

## 4.0 RESULTS - ALUMINUM (Al)

### 4.1 Aluminum Chemical Description

Aluminum is widely distributed in the environment and is the most abundant metal in the Earth's crust (ATSDR, 2008). As a common crustal element not bioaccumulated to a significant degree in shellfish, aluminum was measured, in part, to help assess whether elevated concentrations of other metals were associated with tissue uptake or with ingestion of contaminated particles (LeBlanc et al., 2011). Aluminum is highly reactive and is never found as a free metal but rather combined with other elements, mostly oxygen, silicon, and fluorine. These compounds are typically found in soil, minerals, rocks, and clays. Aluminum occurs naturally in soil, water, and air. High levels in the environment can be caused by the mining and processing of aluminum ores or the production of aluminum metal, alloys, and compounds. Small amounts of aluminum are released into the environment from coal-fired power plants and incinerators. Products that contain aluminum include beverage cans, pots and pans, airplanes, siding and roofing, foil, explosives, fireworks, antacids, aspirin, food additives, antiperspirants, and cosmetics (ATSDR, 2008).

Human exposure to aluminum is commonplace and typically occurs by the consumption of processed foods that include flour, baking powder, coloring agents, or anticaking agents (ATSDR, 2008). An average US adult consumes approximately 7-9 mg of aluminum per day. Occupational exposure is the primary cause of exposure to high levels of aluminum in humans. High levels of exposure can result in lung problems (largely addressed by wearing masks) and decreased nervous system function. Accretion of aluminum happens in humans with kidney disease, as it is not removed through the urinary tract like normal, and can sometimes result in the development of bone or brain diseases (ATSDR, 2008).

Aluminum cannot be destroyed in the environment, it can only change its form or become attached/separated from other particles (ATSDR, 2008). Aluminum particles in air settle to the ground or are washed out of the air by rain. However, very small aluminum particles can stay in the air for many days. Most aluminum-containing compounds do not dissolve in water unless the water is acidic or very alkaline (ATSDR, 2008).

### 4.2 Magnitude and Distribution of Aluminum in Mussel Tissue in 2019

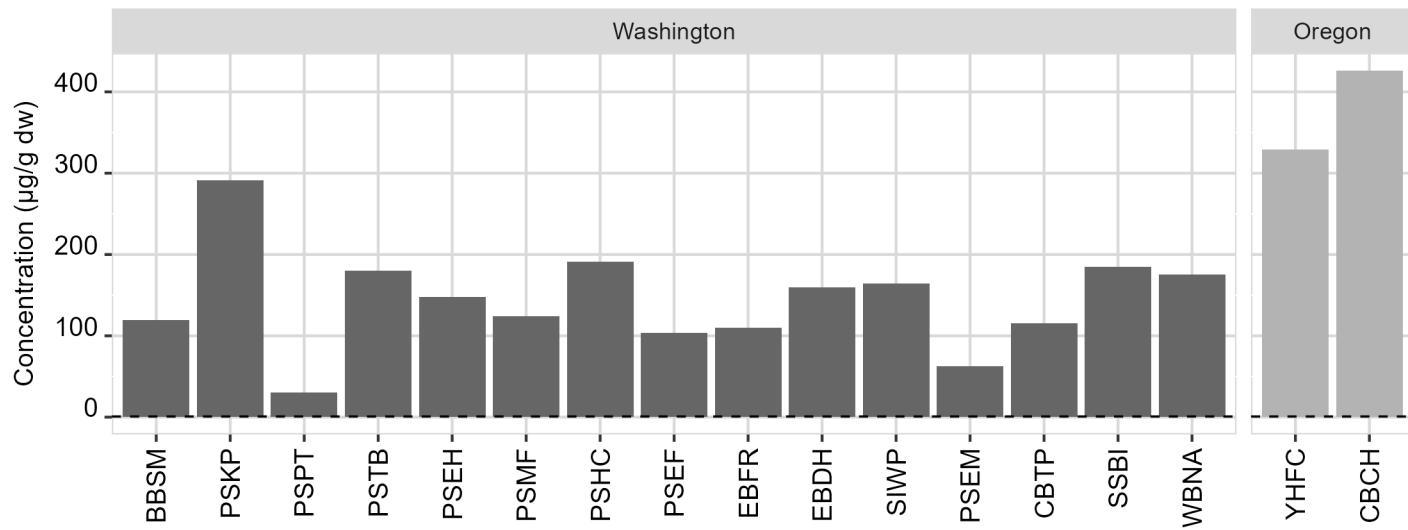


Figure 3. Bar graph showing magnitude of aluminum ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Aluminum (Al)

## 4.3 Historical Context of Aluminum Magnitude and Distribution in Mussel Tissue

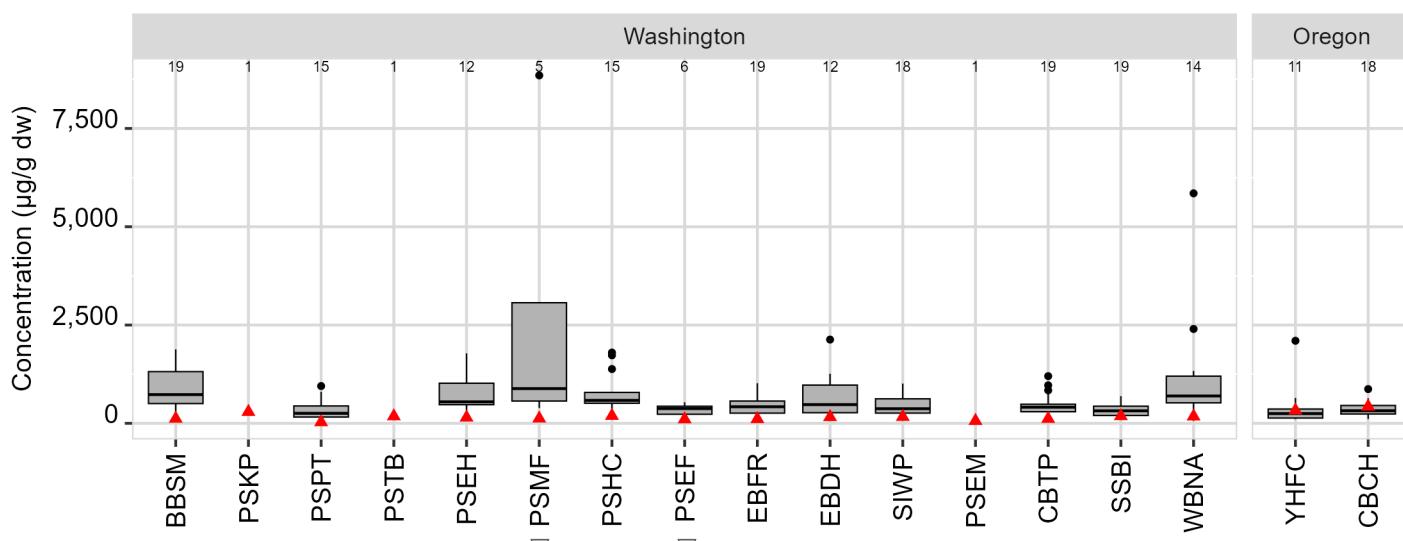


Figure 4. Aluminum concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic aluminum concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

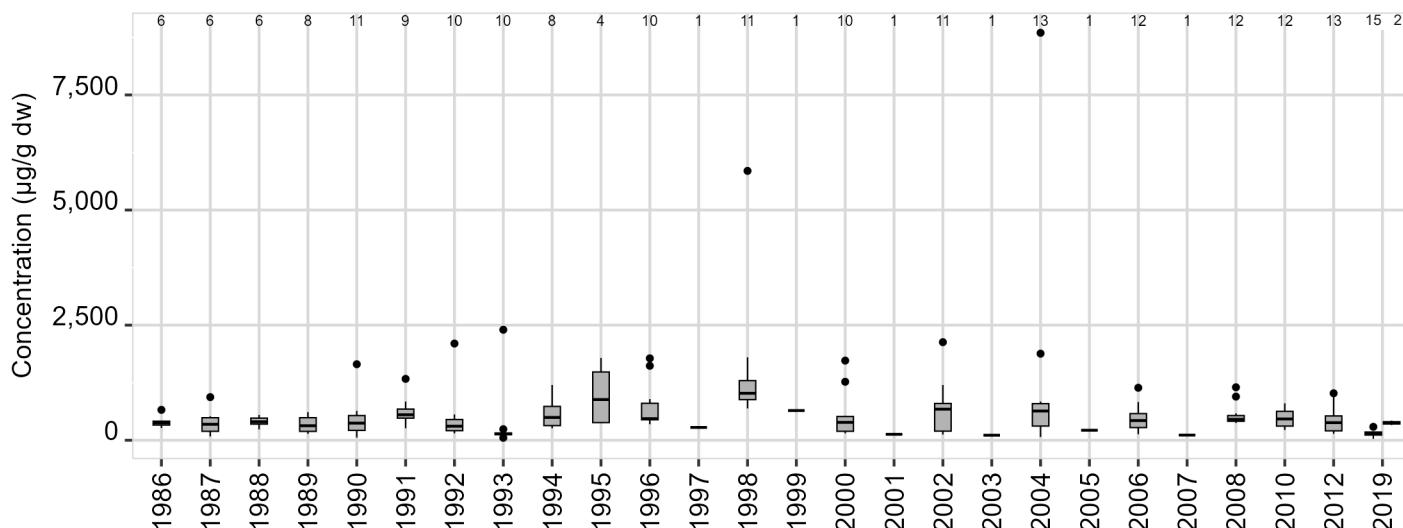


Figure 5. Boxplots representing the historic aluminum concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

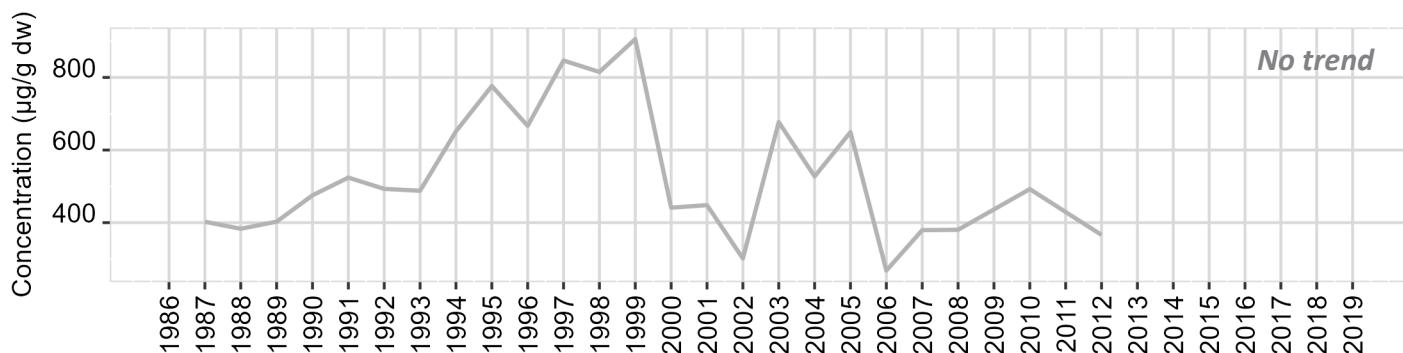


Figure 6. Three-point moving average of the yearly mean aluminum concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Aluminum (Al)

## 4.4 Aluminum Summary

### 2019 Mussel Tissue Results:

- Al was detected at 100% of 17 sites surveyed (Figure 3)
- Al concentration descriptive statistics (Figure 3):
  - Range: 29.95 – 426.42  $\mu\text{g/g dw}$
  - Minimum Al concentration was detected at PSPT
  - Maximum Al concentration was detected at CBCH
  - Median: 159.66  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $171.52 \pm 98.04 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 14% of sites surveyed were above their historic median concentrations in 2019 (Figure 4)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Al concentrations in the Pacific Northwest ( $p = 0.66$ ,  $\rho = -0.09$ ) (Figure 5; Figure 6)
- 2 sites (PSMF, PSEF) in the Pacific Northwest showed significant decreasing temporal trends of Al concentrations at  $\alpha = 0.05$  (Figure 4; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of Al concentrations at  $\alpha = 0.05$  (Figure 4; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 7):
  - 100% of sites in low cluster (0.00 – 583.00  $\mu\text{g/g dw}$ )
  - 0% of sites in medium cluster (590.00 – 1,460.00  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (1,490.00 – 8,850.00  $\mu\text{g/g dw}$ )

### General Observations:

- Al concentrations were uniformly low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Regionally, Al concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Al pollution.

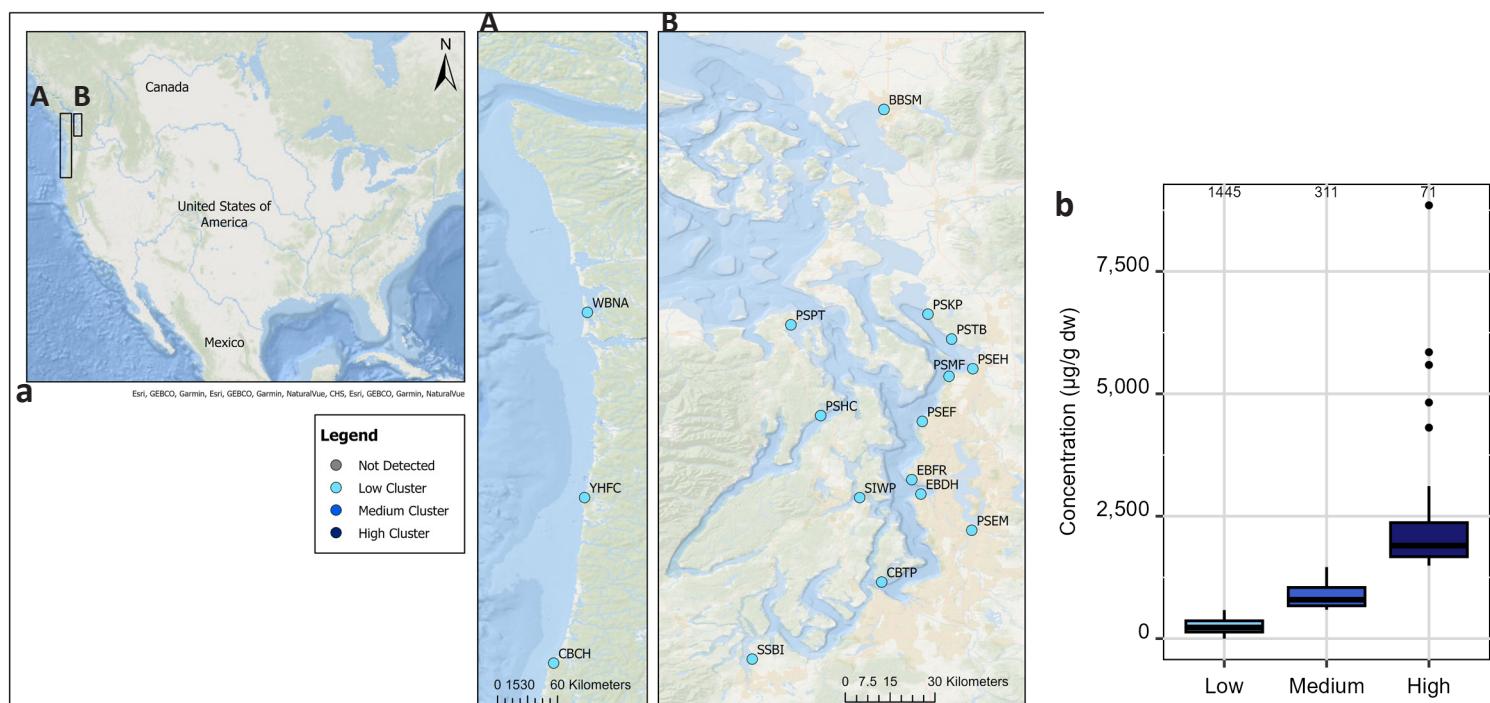


Figure 7. Aluminum concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP Mytilus species aluminum concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples.

# Results - Arsenic (As)

## 5.0 RESULTS - ARSENIC (As)

### 5.1 Arsenic Chemical Description

Arsenic is found in high levels in the environment as a result of both natural sources and industrial production. It is naturally present in low concentrations in the Earth's crust and in rock, soil, water, and air and is highly prevalent in the bedrock of the Pacific Northwest region. Additionally, 30% of atmospheric arsenic comes from volcanoes and other natural sources and a major source responsible for apparent elevated levels of arsenic in the Nation is natural crustal rock, which varies by region (Welch et al., 1988). Generally, with respect to trend analysis, continuous natural sources are associated with neither decreasing nor increasing trends (Kimbrough et al., 2008). Industrial products that contain arsenic include preserved wood, semiconductors, pesticides, defoliants, pigments, antifouling paints, and veterinary medicines. In the recent past, as much as 90% of arsenic in industrial production was used for wood preservation (ATSDR, 2007a). Atmospheric sources of arsenic include smelting, fossil fuel combustion, power generation, and pesticide application contribute to high environmental levels of arsenic (ATSDR, 2007a).

Arsenic is an essential trace nutrient for many animals; however, dietary requirements for humans are unknown (Uthus, 1992). Arsenic is toxic at high concentrations to fish, birds and plants. In animals and humans prolonged chronic exposure is linked to cancer (Goyer, 1986). Inorganic arsenic, the most toxic form, represents approximately 10% of total arsenic in bivalves. Less harmful organic forms, such as arsenobetaine, predominate in seafood (Edmonds and Francesconi, 1977, 1988, 1993; Phillips, 1990; FDA, 1993). The MWP measures total arsenic, including both the inorganic and the organic forms. Safety guidance levels for arsenic in fish and shellfish are no longer listed by the US FDA (FDA, 2011).

Centuries of human activities have changed the natural biogeochemical cycle of arsenic resulting in contamination of land, water, and air. Movement of arsenic to coastal and estuarine water occurs primarily from river runoff and atmospheric deposition.

### 5.2 Magnitude and Distribution of Arsenic in Mussel Tissue in 2019

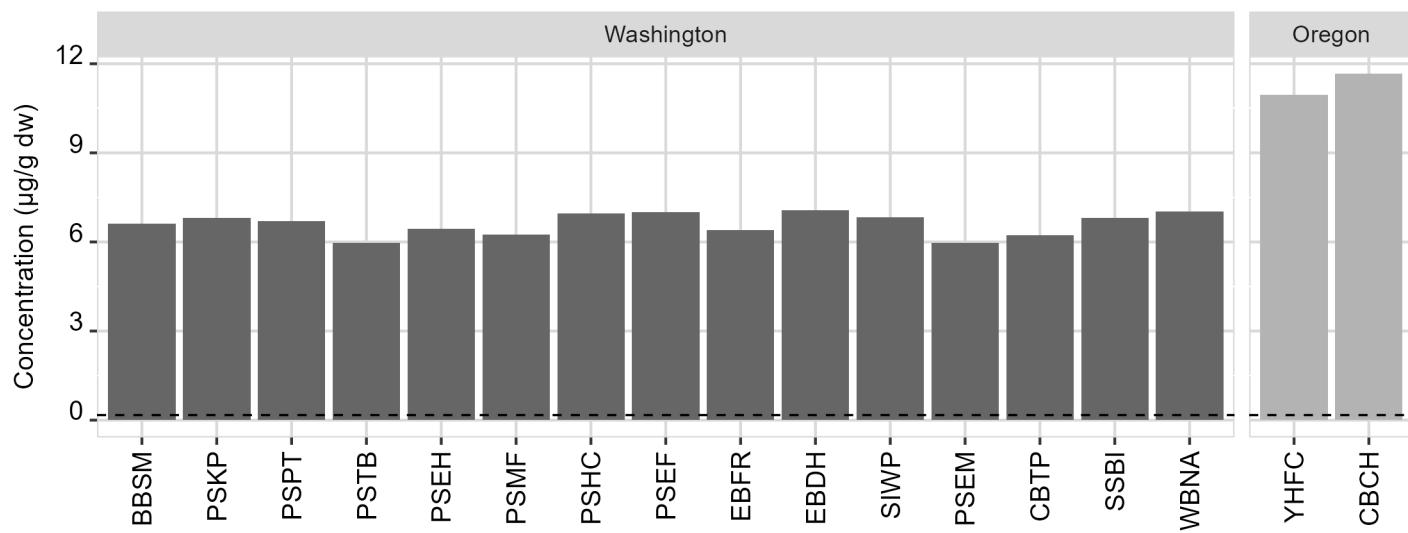


Figure 8. Bar graph showing magnitude of arsenic ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Arsenic (As)

## 5.3 Historical Context of Arsenic Magnitude and Distribution in Mussel Tissue

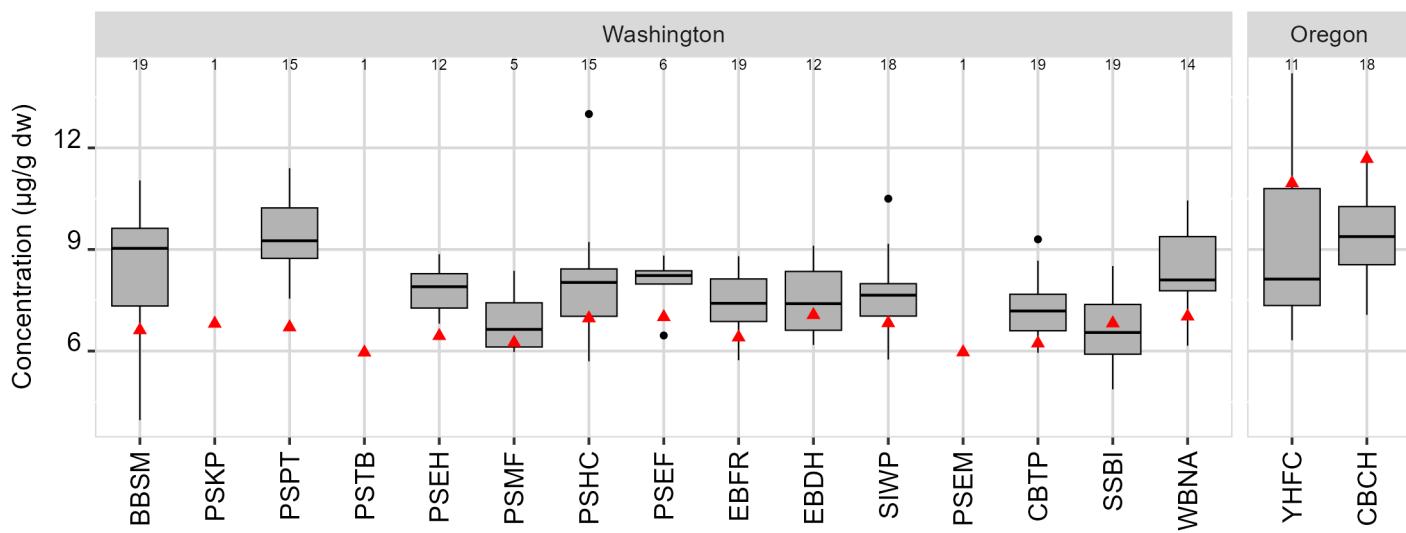


Figure 9. Arsenic concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic arsenic concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

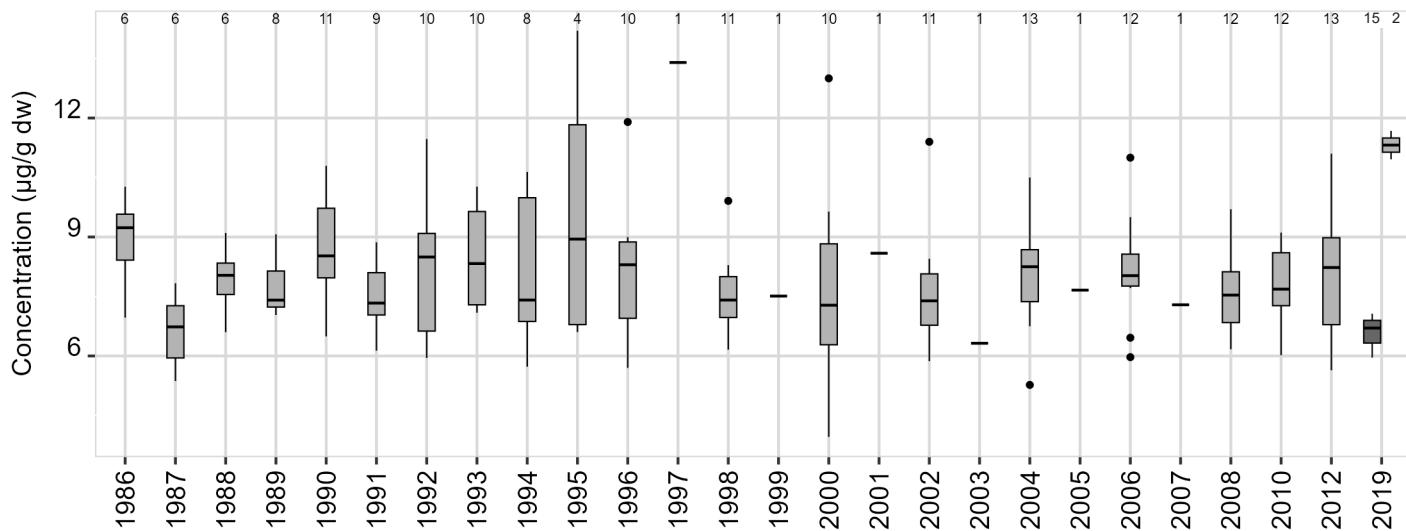


Figure 10. Boxplots representing the historic arsenic concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels, light gray indicates that wild mussels were used.

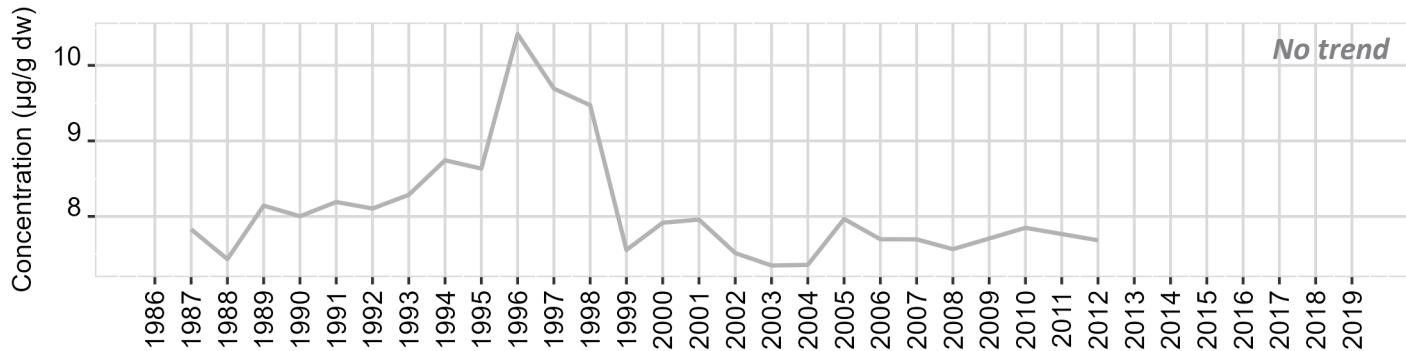


Figure 11. Three-point moving average of the yearly mean arsenic concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Arsenic (As)

## 5.4 Arsenic Summary

### 2019 Mussel Tissue Results:

- As was detected at 100% of 17 sites surveyed (Figure 8)
- As concentration descriptive statistics (Figure 8):
  - Range: 5.96 – 11.68  $\mu\text{g/g dw}$
  - Minimum As concentration was detected at PSTB and PSEM
  - Maximum As concentration was detected at CBCH
  - Median: 6.81  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $7.16 \pm 1.61 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 21% of sites surveyed were above their historic median concentrations in 2019 (Figure 9)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of As concentrations in the Pacific Northwest ( $p = 0.15$ ,  $\rho = -0.29$ ) (Figure 10; Figure 11)
- 0 sites in the Pacific Northwest showed significant decreasing temporal trends of As concentrations at  $\alpha = 0.05$  (Figure 9; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of As concentrations at  $\alpha = 0.05$  (Figure 9; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 12):
  - 88% of sites in low cluster (1.97 – 8.42  $\mu\text{g/g dw}$ )
  - 12% of sites in medium cluster (8.43 – 15.17  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (15.33 – 41.34  $\mu\text{g/g dw}$ )

### General Observations:

- As concentrations were generally low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Medium As concentrations were detected in mussel tissue off the Oregon coast, possibly indicating offshore natural sources or localized point sources of contamination.
- Regionally, As concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic As pollution.

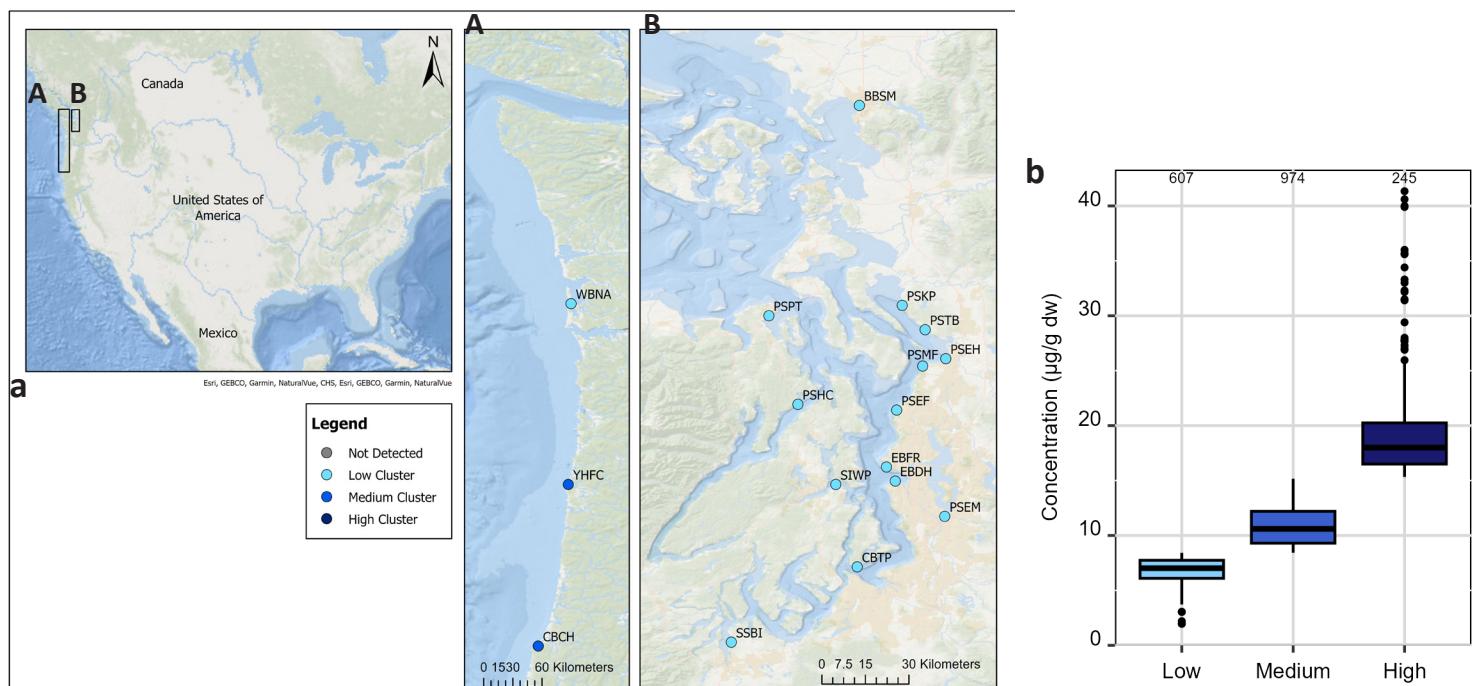


Figure 12. Arsenic concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP *Mytilus* species arsenic concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1826 samples.

# Results - Cadmium (Cd)

## 6.0 RESULTS - CADMIUM (Cd)

### 6.1 Cadmium Chemical Description

Cadmium found in coastal and estuarine environments can be linked to both natural and non-point anthropogenic sources (Roesijadi, 1984). Cadmium occurs naturally in the Earth's crust as complex oxides and sulfides in ores (Plachy, 2003) and natural sources can be linked to river runoff from cadmium rich soils, leaching from bedrock, and upwelling from marine sediment deposits (Sokolova et al., 2005). Industrial sources and uses include zinc, lead, and copper production, electroplating and galvanizing, smelting, mining, fossil fuel burning, waste slag, and sewage sludge (ATSDR, 1999a; FDA, 1993). In addition to abundant industrial applications, other products that contain cadmium include batteries, color pigment, plastics, and phosphate fertilizers. Generally, anthropogenic emissions exceed natural emissions and elevated cadmium levels are primarily located in freshwater-dominated estuaries (e.g., Mississippi Delta, Great Lakes, Chesapeake Bay).

Cadmium is a non-essential and toxic element. Cadmium is toxic to fish, especially salmonid species and juveniles, and chronic exposure can result in reductions in growth. Cadmium has been shown to impair development and reproduction in several invertebrate species and osmoregulation in herring larvae (ATSDR, 1999a; Eisler, 1985). Respiration and consumption represent the two major exposure pathways for humans to cadmium with exposure to high levels resulting primarily from occupational exposure. Safety guidance levels for cadmium in fish and shellfish are no longer listed by the US FDA (FDA, 2011).

As a result of fossil fuel burning, erosion, and biological activities, cadmium becomes airborne and is transported by atmospheric processes. Land-based runoff, river transport, and ocean upwelling are the main conveyors of cadmium into coastal environments.

### 6.2 Magnitude and Distribution of Cadmium in Mussel Tissue in 2019

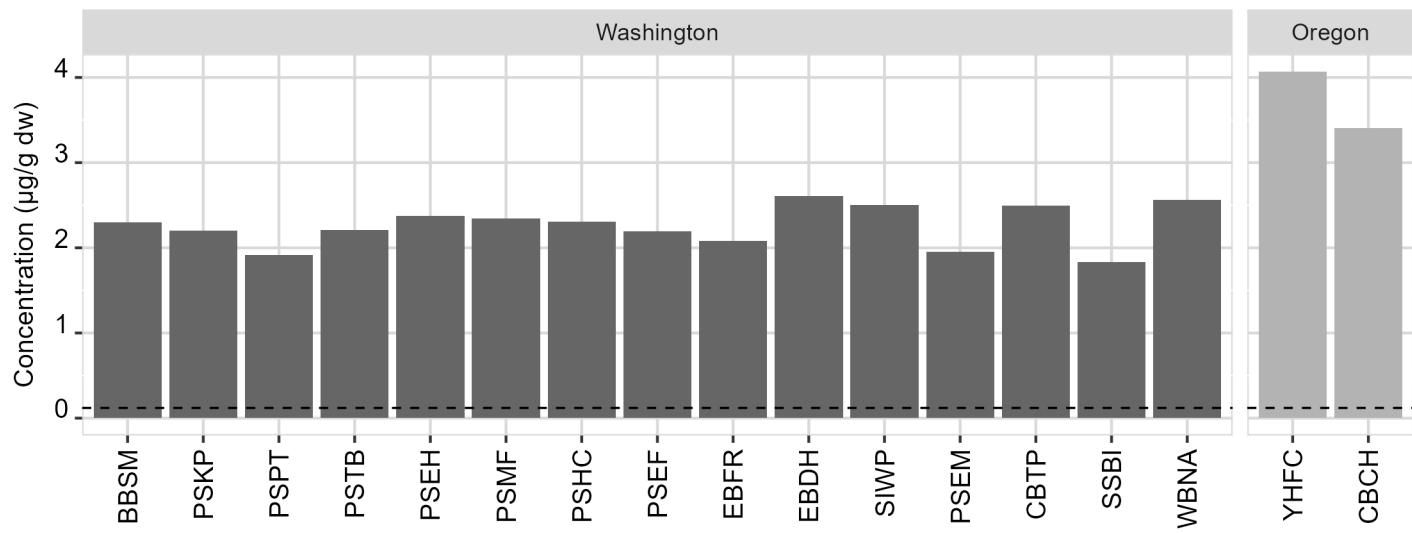


Figure 13. Bar graph showing magnitude of cadmium ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Cadmium (Cd)

## 6.3 Historical Context of Cadmium Magnitude and Distribution in Mussel Tissue

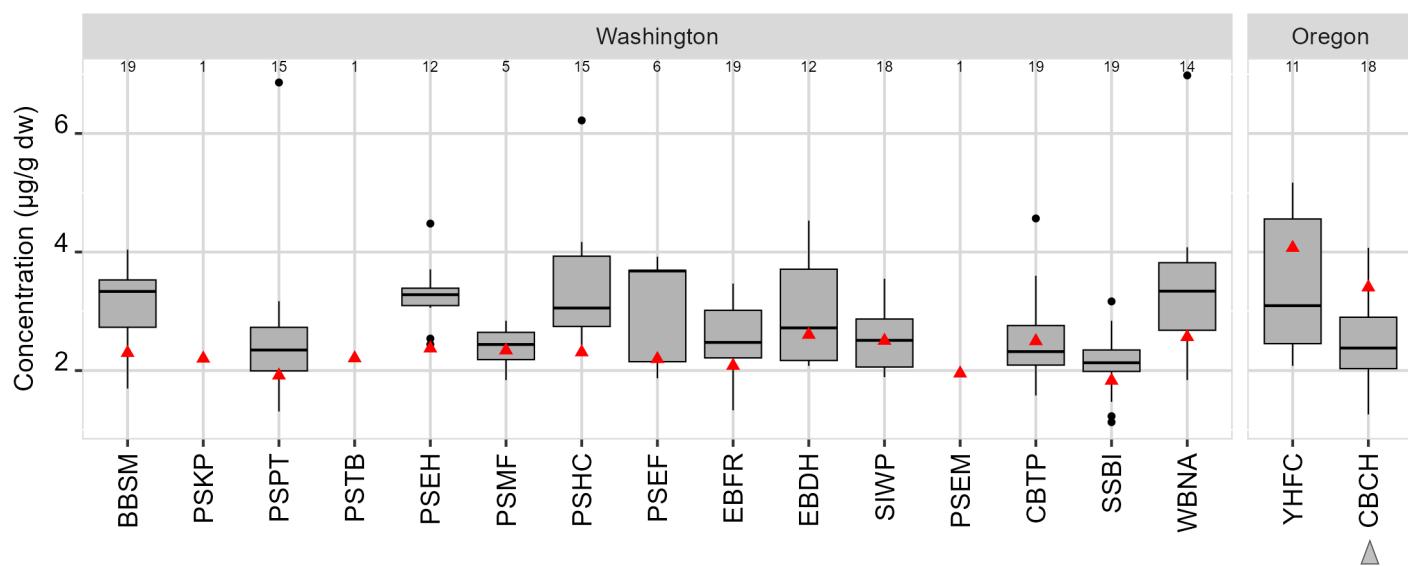


Figure 14. Cadmium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic cadmium concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

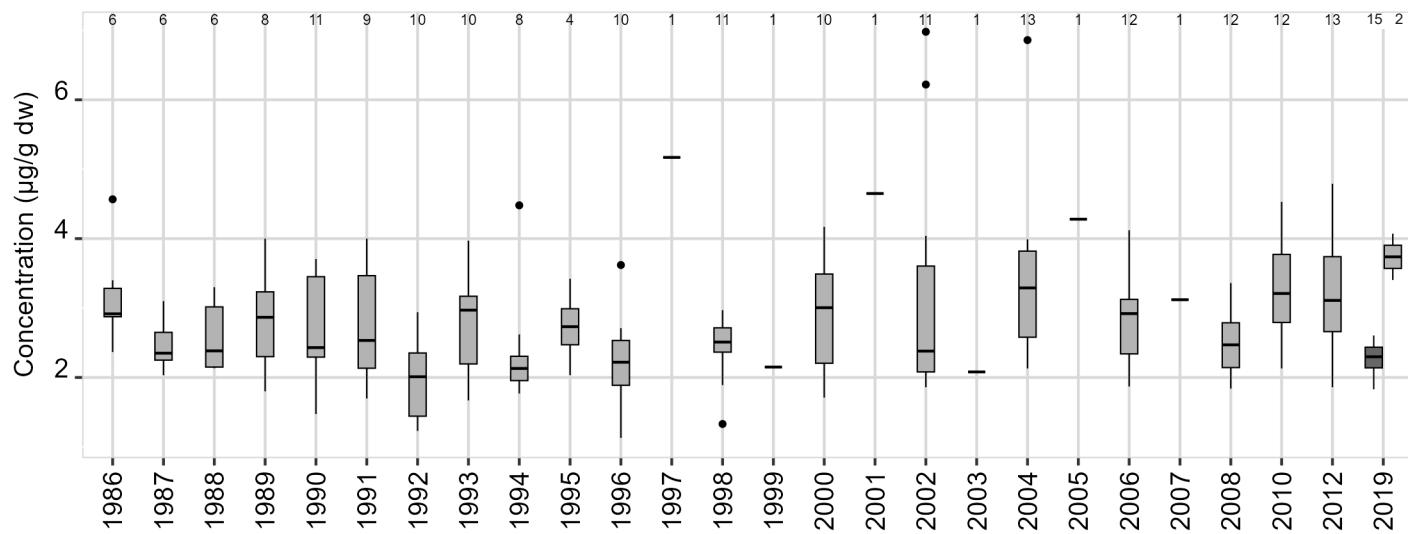


Figure 15. Boxplots representing the historic cadmium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

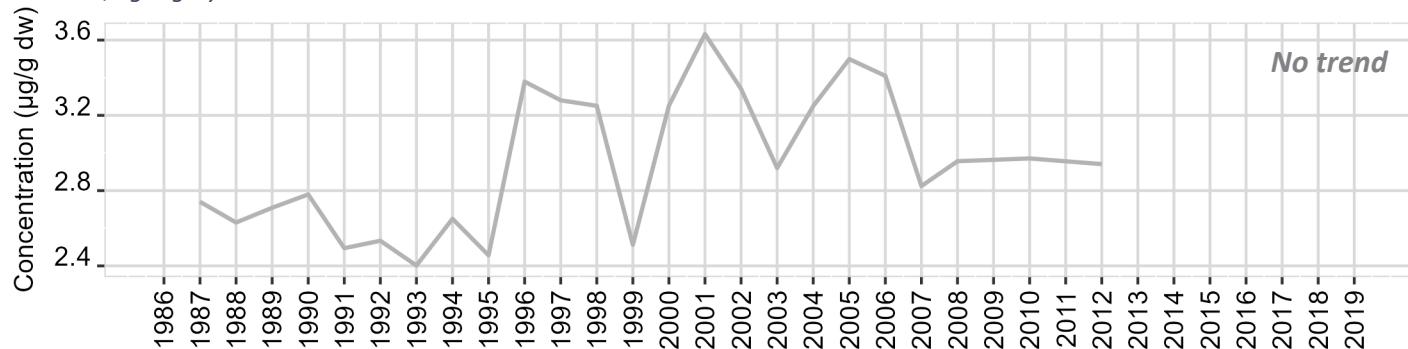


Figure 16. Three-point moving average of the yearly mean cadmium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Cadmium (Cd)

## 6.4 Cadmium Summary

### 2019 Mussel Tissue Results:

- Cd was detected at 100% of 17 sites surveyed (Figure 13)
- Cd concentration descriptive statistics (Figure 13):
  - Range: 1.83 – 4.07  $\mu\text{g/g dw}$
  - Minimum Cd concentration was detected at SSBI
  - Maximum Cd concentration was detected at YHFC
  - Median: 2.31  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $2.43 \pm 0.55 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 21% of sites surveyed were above their historic median concentrations in 2019 (Figure 14)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Cd concentrations in the Pacific Northwest ( $p = 0.25$ ,  $\rho = 0.24$ ) (Figure 15; Figure 16)
- 0 sites in the Pacific Northwest showed significant decreasing temporal trends of Cd concentrations at  $\alpha = 0.05$  (Figure 14; Table A4)
- 1 site (CBCH) in the Pacific Northwest showed a significant increasing temporal trend of Cd concentrations at  $\alpha = 0.05$  (Figure 14; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 17):
  - 76% of sites in low cluster (0.00 – 2.51  $\mu\text{g/g dw}$ )
  - 24% of sites in medium cluster (2.53 – 6.63  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (6.67 – 20.40  $\mu\text{g/g dw}$ )

### General Observations:

- Cd concentrations were generally low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Medium Cd concentrations were detected in mussel tissue off the Oregon coast and at Puget Sound site EBDH, possibly indicating offshore natural sources or localized point sources of contamination.
- Regionally, Cd concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Cd pollution.

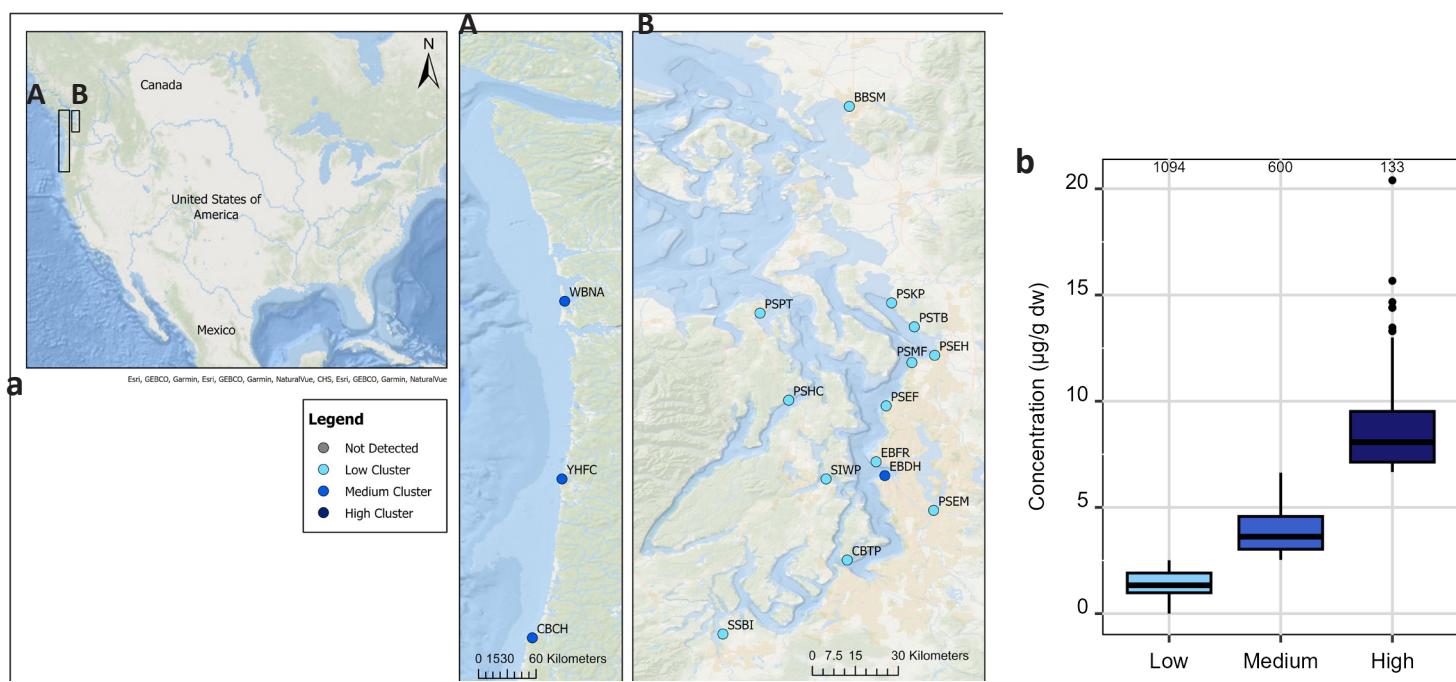


Figure 17. Cadmium concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP *Mytilus* species cadmium concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples.

# Results - Chromium (Cr)

## 7.0 RESULTS - CHROMIUM (Cr)

### 7.1 Chromium Chemical Description

Chromium is a naturally occurring metal found in rocks, animals, plants and soils in three main forms: chromium(0), chromium(III), and chromium(VI). Chromium is widely used in industries involved in electroplating, leather tanning, textile production, and the manufacture of chromium-based products including treated wood, tanned leather, stainless steel cookware, and metal-on-metal hip replacements (ATSDR, 2012a). Chromium can also be released into the environment from the burning of natural gas, oil, or coal (ATSDR, 2012a). The EPA has established a maximum contaminant level of 0.1 mg/L for total chromium in drinking water and sought voluntary monitoring of chromium(VI) in drinking water by municipalities. OSHA has set a legal limit for atmospheric exposure over an 8-hour workday of 1.0 mg/m<sup>3</sup> for chromium, 0.005 mg/m<sup>3</sup> for chromium(VI), and 0.5 mg/m<sup>3</sup> for chromium(III).

Chromium is an essential trace nutrient but can be toxic at high levels. Exposure to chromium can result from inhalation, ingestion, and physical contact. Inhalation primarily occurs near industries using or manufacturing chromium or through cigarette smoke. Ingestion typically occurs through drinking or bathing water. The general population is most likely to be exposed to trace levels of chromium in food that is eaten, including fruits, vegetables, nuts, beverages, and meats, but exposure at high levels occurs primarily resulting from occupational exposure (ATSDR, 2012a). In humans, exposure to chromium most impacts the respiratory tract, specifically causing irritation to nasal lining, runny nose, and breathing problems. In animals, ingestion of chromium(VI) has been shown to cause negative effects on the stomach, small intestine, blood, and male reproductive system. The International Agency for Research on Cancer (IARC) has determined that chromium(VI) compounds are carcinogenic to humans. In animals, some studies show that exposure to high doses during pregnancy may cause miscarriage, low birth weight, and some changes in development of the skeleton and reproductive system (ATSDR, 2012a).

Chromium does not typically remain in the atmosphere, but is deposited into the soil and water. In soil and water, chromium can change from one form to another depending on external conditions (ATSDR, 2012a).

### 7.2 Magnitude and Distribution of Chromium in Mussel Tissue in 2019

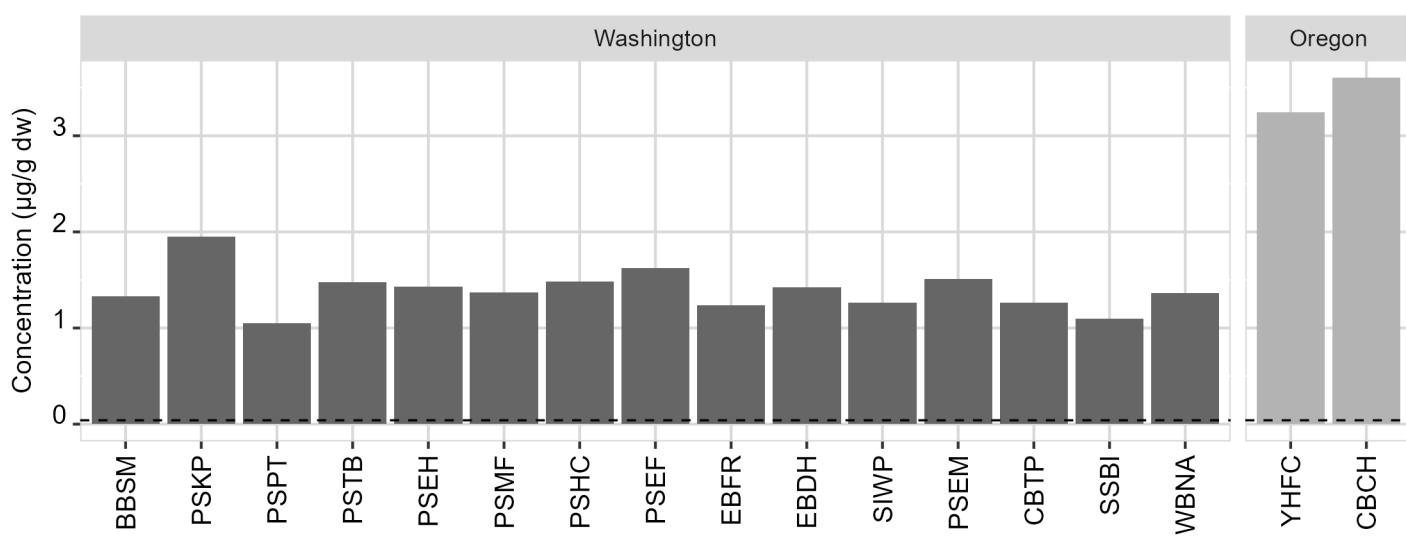


Figure 18. Bar graph showing magnitude of chromium ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Chromium (Cr)

## 7.3 Historical Context of Chromium Magnitude and Distribution in Mussel Tissue

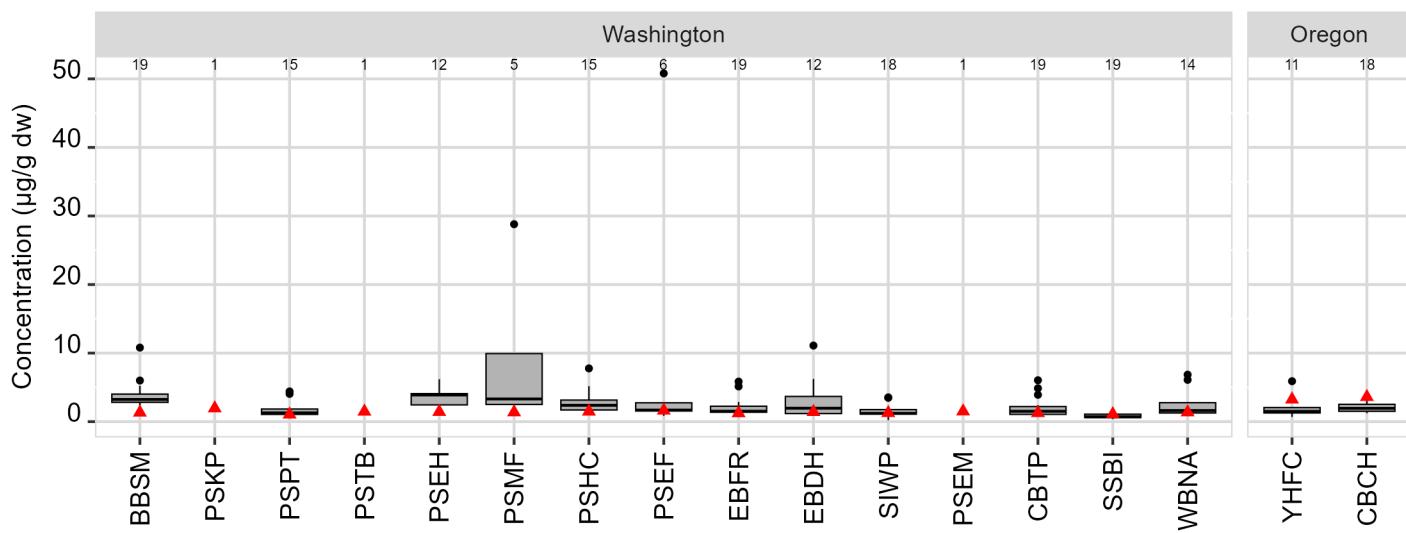


Figure 19. Chromium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic chromium concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

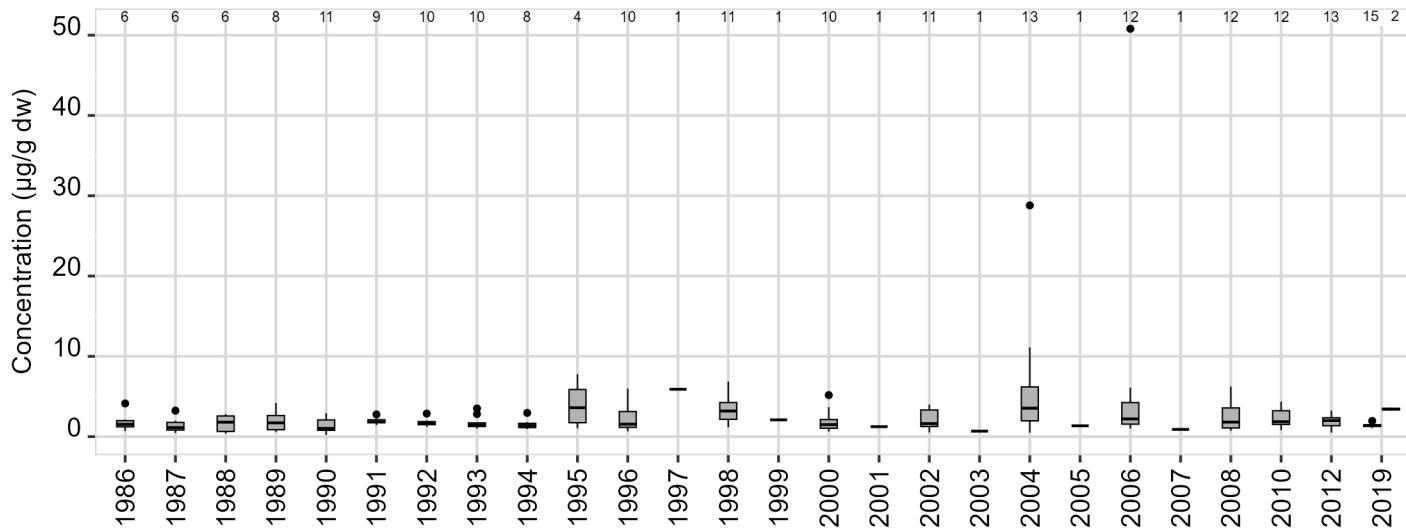


Figure 20. Boxplots representing the historic chromium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.



Figure 21. Three-point moving average of the yearly mean chromium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Chromium (Cr)

## 7.4 Chromium Summary

### 2019 Mussel Tissue Results:

- Cr was detected at 100% of 17 sites surveyed (Figure 18)
- Cr concentration descriptive statistics (Figure 18):
  - Range: 1.05 – 3.61  $\mu\text{g/g}$  dw
  - Minimum Cr concentration was detected at PSPT
  - Maximum Cr concentration was detected at CBCH
  - Median: 1.42  $\mu\text{g/g}$  dw
  - Mean  $\pm$  SD:  $1.63 \pm 0.71 \mu\text{g/g}$  dw

### Historic Context for Mussel Tissue:

- 29% of sites surveyed were above their historic median concentrations in 2019 (Figure 19)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Cr concentrations in the Pacific Northwest ( $p = 0.57$ ,  $\rho = 0.12$ ) (Figure 20; Figure 21)
- 0 sites in the Pacific Northwest showed significant decreasing temporal trends of Cr concentrations at  $\alpha = 0.05$  (Figure 19; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of Cr concentrations at  $\alpha = 0.05$  (Figure 19; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 22):
  - 82% of sites in low cluster (0.00 – 1.80  $\mu\text{g/g}$  dw)
  - 18% of sites in medium cluster (1.81 – 5.70  $\mu\text{g/g}$  dw)
  - 0% of sites in high cluster (5.78 – 290.00  $\mu\text{g/g}$  dw)

### General Observations:

- Cr concentrations were generally low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Medium Cr concentrations were detected in mussel tissue off the Oregon coast and at Puget Sound site PSKP, possibly indicating offshore natural sources or localized point sources of contamination.
- Regionally, Cr concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Cr pollution.

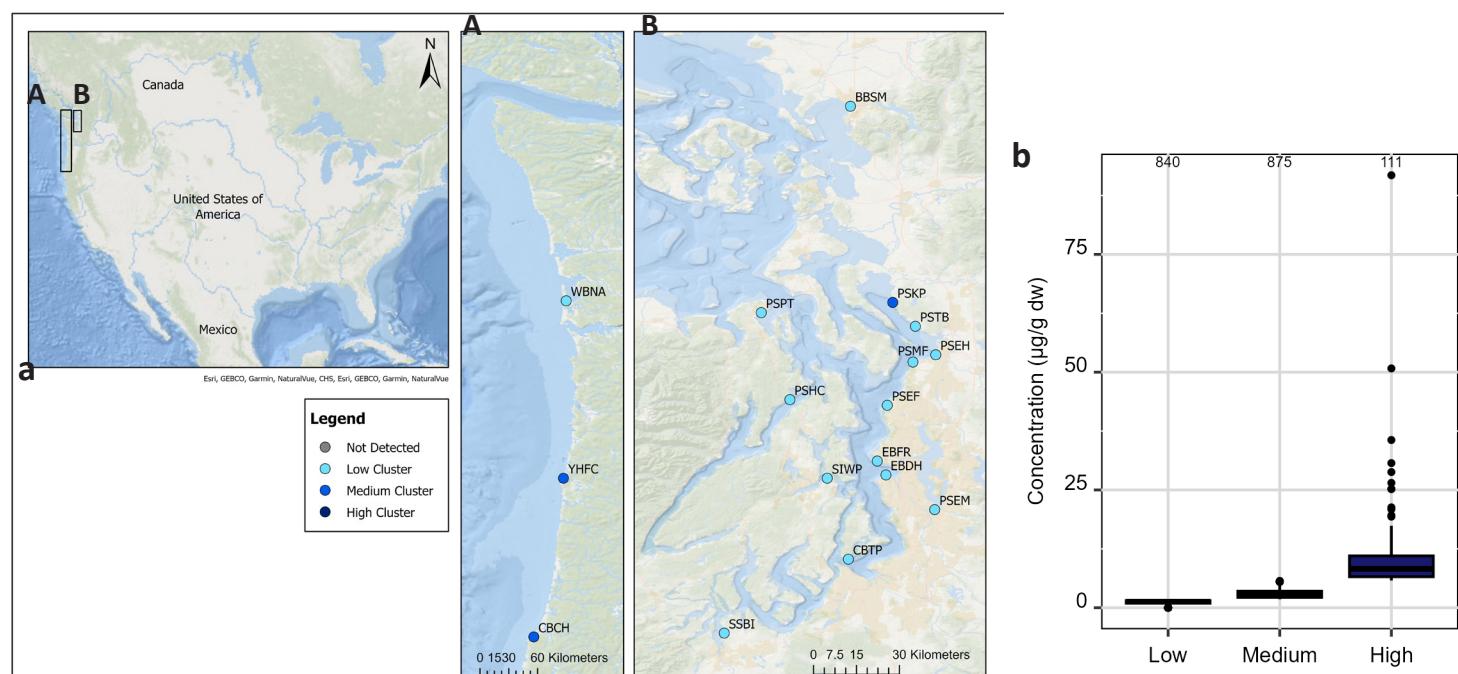


Figure 22. Chromium concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP *Mytilus* species chromium concentrations ( $\mu\text{g/g}$  dw). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples. Site HRUB in 2004 (290.00  $\mu\text{g/g}$  dw) was removed from 'b' to aid in visualization.

# Results - Copper (Cu)

## 8.0 RESULTS - COPPER (Cu)

### 8.1 Copper Chemical Description

Copper is a naturally occurring element that is ubiquitous in the environment. Anthropogenic sources include: mining, manufacturing, agriculture, sewage sludge, antifouling paint, fungicides, wood preservatives, and vehicle brake pads (ATSDR, 2004; Denier van der Gon et al., 2007). The U.S. ranks third in the world for utilization and second in production. The EPA phase-out of chromated copper arsenate (CCA) wood preservatives and the 1980s restrictions on tributyltin marine antifouling paint has stimulated a transition to copper-based wood preservatives and marine antifouling paint.

Trace amounts of copper are an essential nutrient for plants and animals. High concentrations of copper can be toxic to aquatic organisms, with juvenile fishes and invertebrates much more sensitive than adults. Elevated levels of copper can impact aquatic organisms including the functioning of gills, reproduction, and development (Eisler, 1998). Although copper is not highly toxic to humans, chronic effects occur as a result of prolonged exposure to high doses and can cause eye irritation and damage to the digestive tract (ATSDR, 2004). There is no recommended US FDA safety level for copper in fish and fish products (FDA, 2020).

The most common form of copper in water is Cu (II) which is mostly found bound to organic matter. Transport of copper to coastal and estuarine water occurs as a result of runoff and river transport. Atmospheric transport (Denier van der Gon et al., 2007) and deposition of particulate copper into surface waters may also be a significant source of copper to coastal waters.

### 8.2 Magnitude and Distribution of Copper in Mussel Tissue in 2019

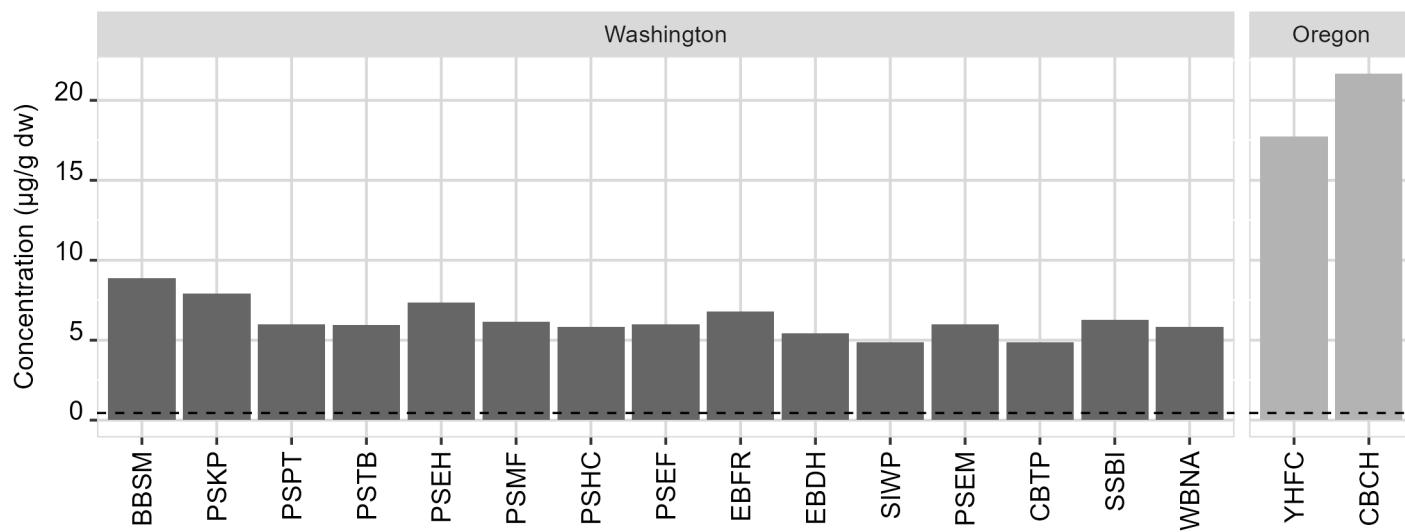


Figure 23. Bar graph showing magnitude of copper ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Copper (Cu)

## 8.3 Historical Context of Copper Magnitude and Distribution in Mussel Tissue

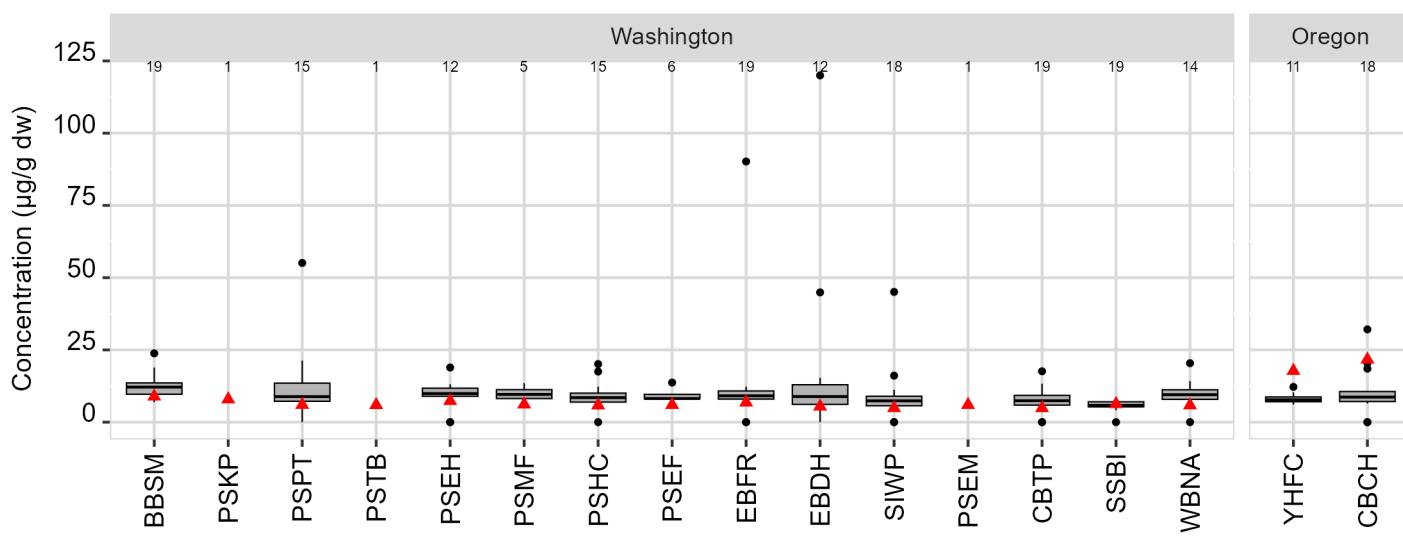


Figure 24. Copper concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic copper concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

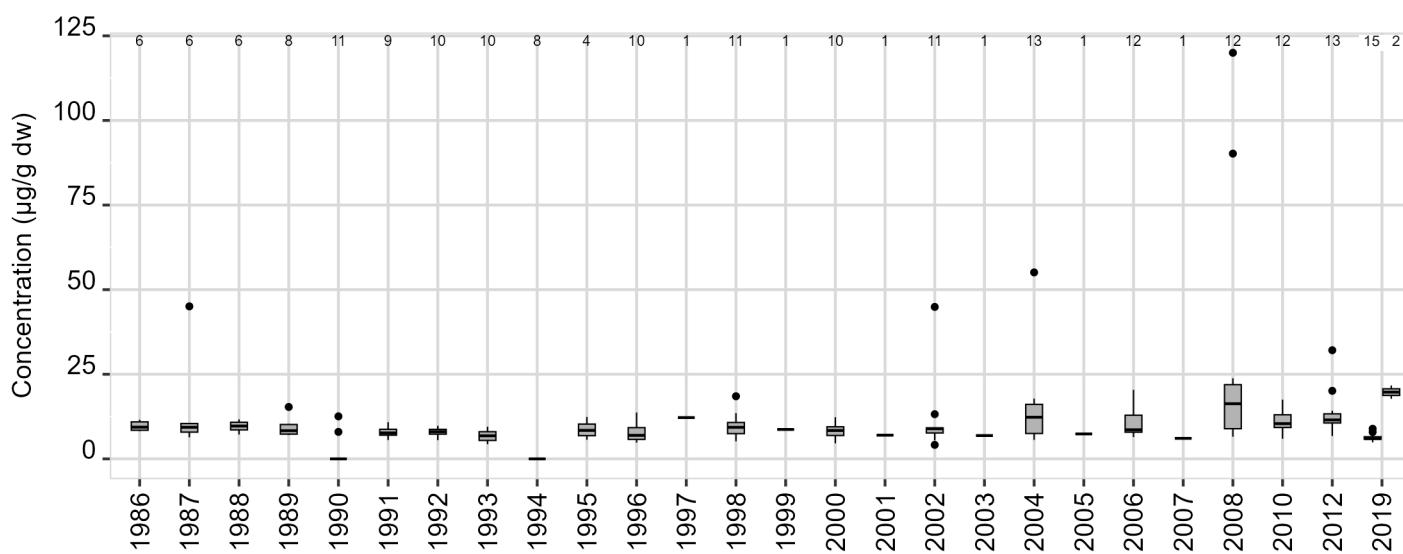


Figure 25. Boxplots representing the historic copper concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

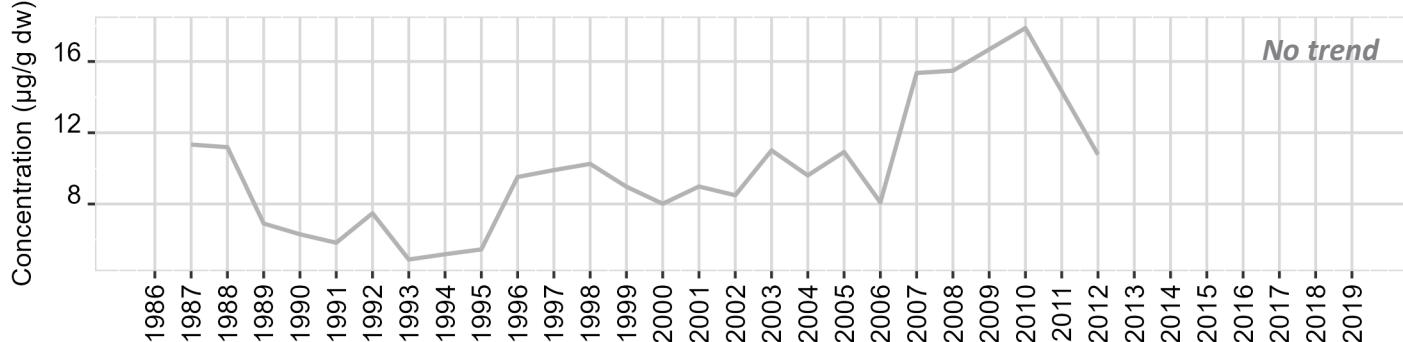


Figure 26. Three-point moving average of the yearly mean copper concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Copper (Cu)

## 8.4 Copper Summary

### 2019 Mussel Tissue Results:

- Cu was detected at 100% of 33 sites surveyed (Figure 23)
- Cu concentration descriptive statistics (Figure 23):
  - Range: 4.87 – 21.69  $\mu\text{g/g dw}$
  - Minimum Cu concentration was detected at CBTP
  - Maximum Cu concentration was detected at CBCH
  - Median: 6.00  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $7.85 \pm 4.63 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 21% of sites surveyed were above their historic median concentrations in 2019 (Figure 24)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Cu concentrations in the Pacific Northwest ( $p = 0.42$ ,  $\rho = 0.17$ ) (Figure 25; Figure 26)
- 1 site (PSMF) in the Pacific Northwest showed a significant decreasing temporal trend of Cu concentrations at  $\alpha = 0.05$  (Figure 24; Table A4)
- 1 site (PSPT) in the Pacific Northwest showed a significant increasing temporal trend of Cu concentrations at  $\alpha = 0.05$  (Figure 24; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 27):
  - 100% of sites in low cluster (0.00 – 29.50  $\mu\text{g/g dw}$ )
  - 0% of sites in medium cluster (31.42 – 69.80  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (90.20 – 857.00  $\mu\text{g/g dw}$ )

### General Observations:

- Cu concentrations were uniformly low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Regionally, Cu concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Cu pollution.

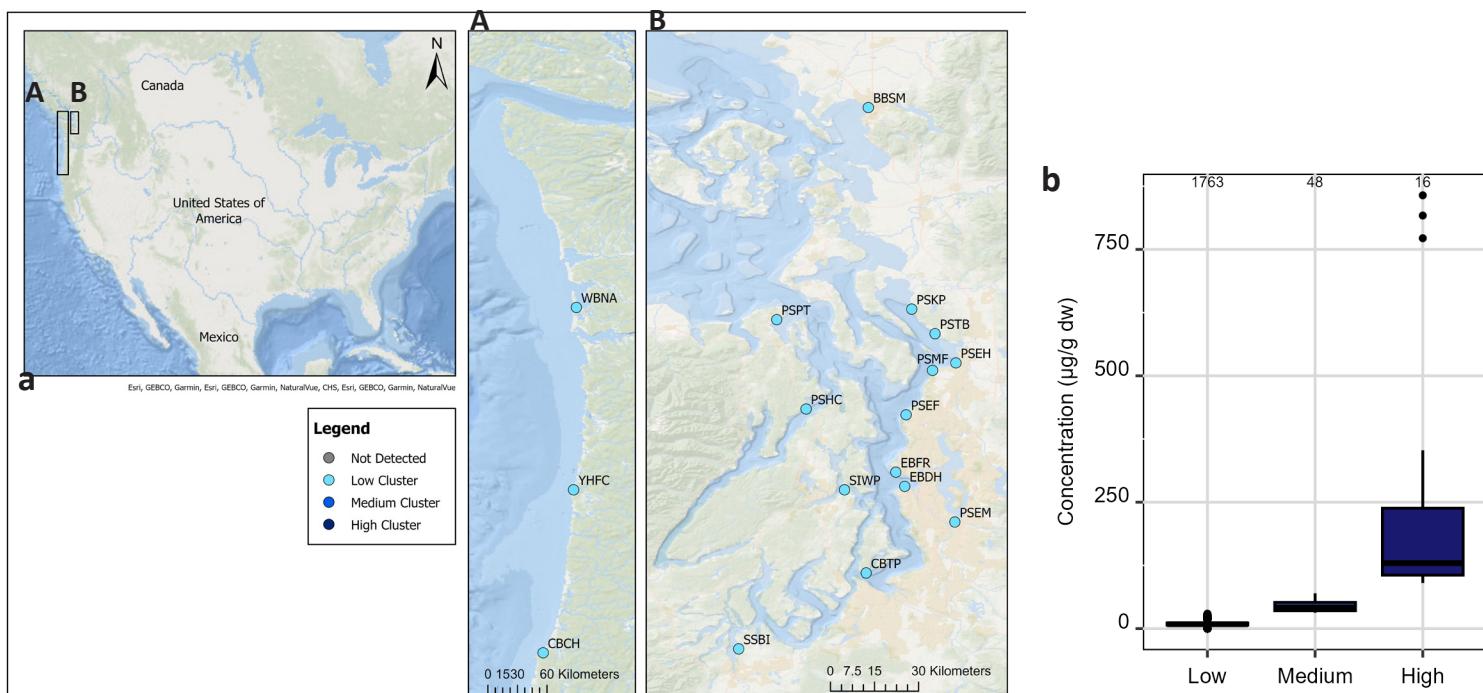


Figure 27. Copper concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP Mytilus species copper concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples.

# Results - Iron (Fe)

## 9.0 RESULTS - IRON (Fe)

### 9.1 Iron Chemical Description

Iron is the second most abundant metal in the Earth's crust but tends to have low concentrations in water due to low solubility (Xing and Liu, 2011). Iron concentrations are generally low in natural freshwater bodies but contamination can occur from anthropogenic sources and result in eutrophication and algal blooms (Xing and Liu, 2011). Natural sources of iron in the environment are primarily from weathered rocks and soils and anthropogenic sources of iron input are primarily from fossil fuel combustion and incorporation of iron into foods and nutritional supplements (Liu et al., 2022). More recently, increasing usage of iron-based nanoparticles has raised concerns regarding their environmental behavior and ecological effects (Lei et al., 2018).

Iron is an essential nutrient for humans, with adverse effects caused both by deficiency and exposure to high concentrations (EPA, 2006). Iron deficiency, known as anemia, can cause fatigue, weakness, and cold extremities in humans. Conversely, exposure to high levels of iron in animals and humans typically results in injury to the gastrointestinal mucosa which can result in nausea, vomiting, abdominal pain, and diarrhea (Yuen and Becker, 2022). Exposure at these levels is typically caused by accidental ingestion (Lei et al., 2018). At the cellular level, exposure to high levels of iron can impair cellular metabolism in the heart, liver, and central nervous system (Yuen and Becker, 2022). In the environment, the toxicity of iron-based nanoparticles is increasingly observed (Lei et al., 2018).

In the environment, iron-based nanoparticles undergo physical and chemical transformations influenced by environmental factors including pH, ions, dissolved oxygen, natural organic matter, and biota (Lei et al., 2018). Iron in the environment is often taken up by both land-based and aquatic plants to the extent that excess iron is often stored by plants as iron plaque to use in times of iron deficiency. Potentially harmful environmental effects of iron are a direct result of pollution caused by iron ore mining.

### 9.2 Magnitude and Distribution of Iron in Mussel Tissue in 2019

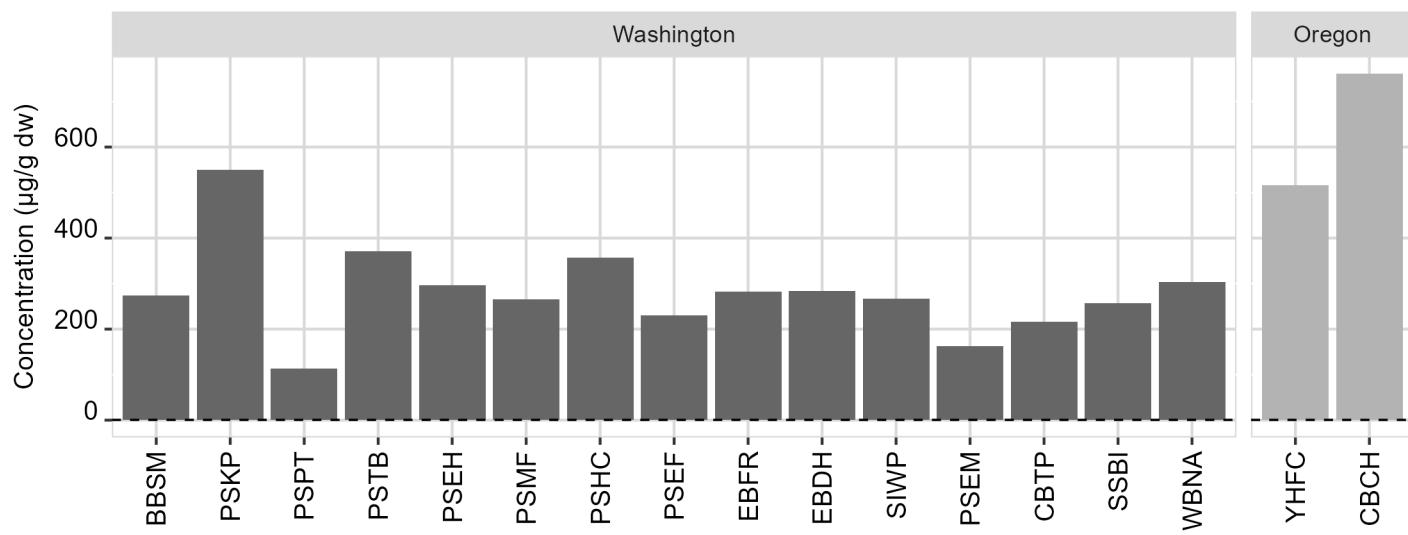


Figure 28. Bar graph showing magnitude of iron ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Iron (Fe)

## 9.3 Historical Context of Iron Magnitude and Distribution in Mussel Tissue

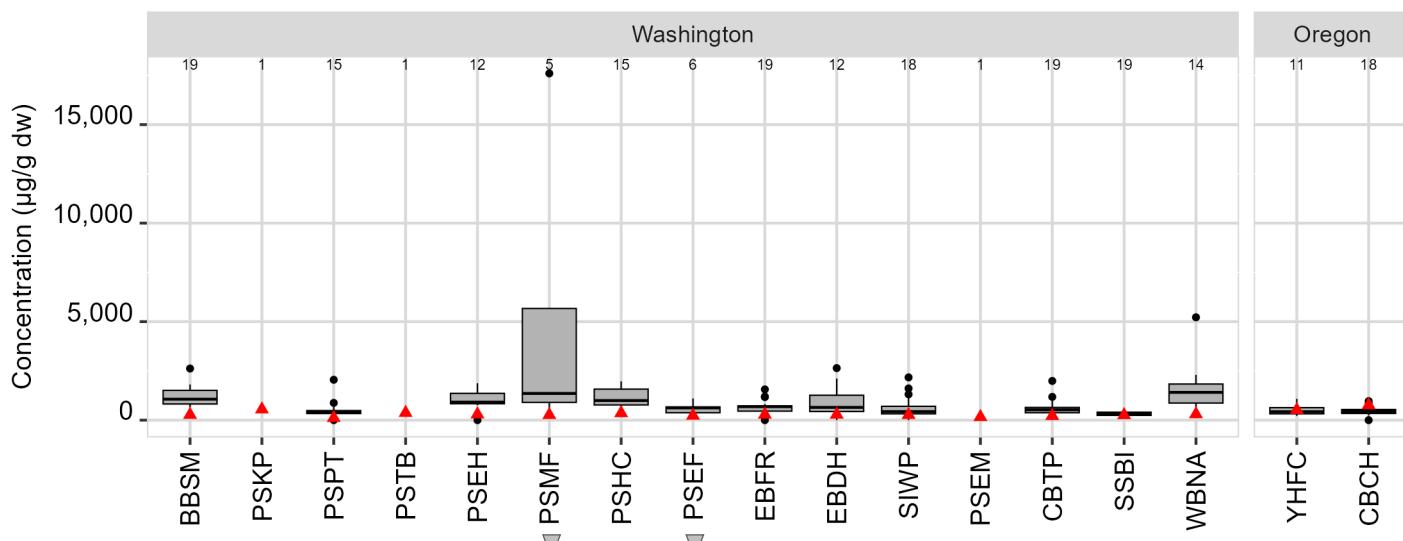


Figure 29. Iron concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic iron concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

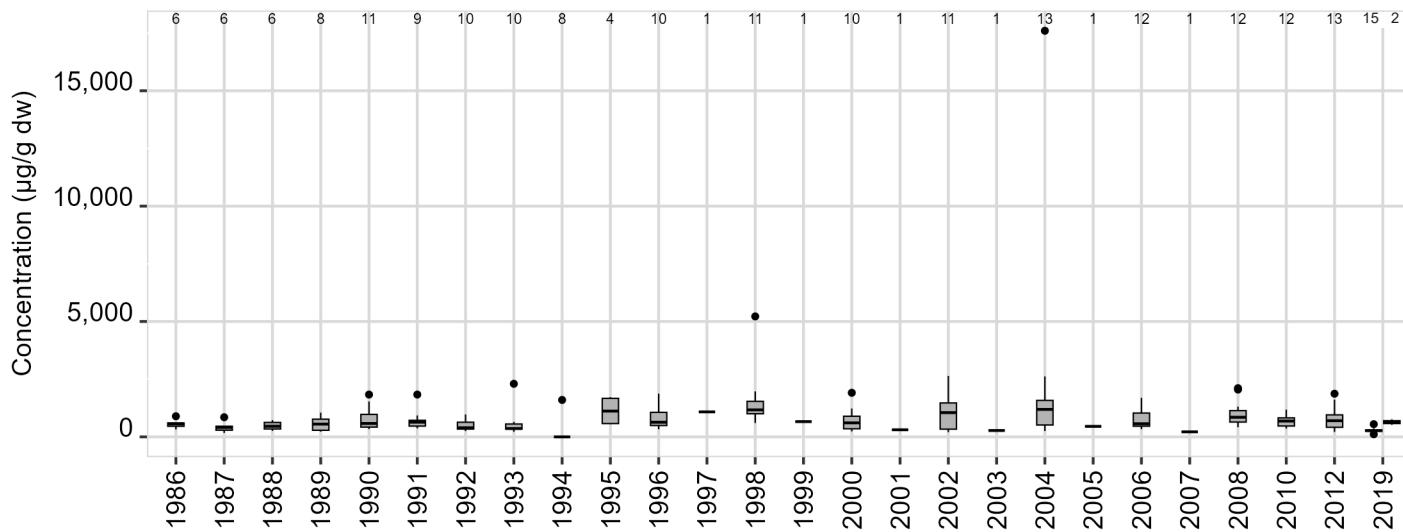


Figure 30. Boxplots representing the historic iron concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

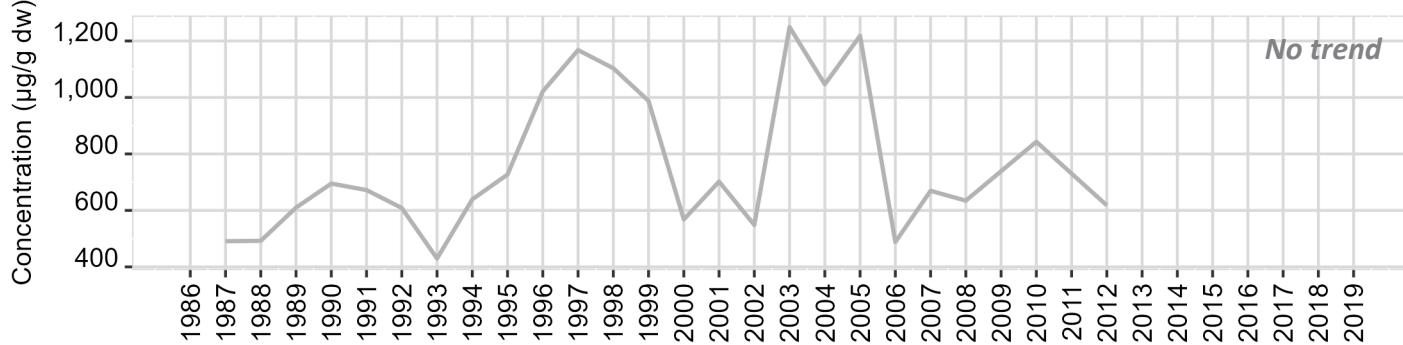


Figure 31. Three-point moving average of the yearly mean iron concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Iron (Fe)

## 9.4 Iron Summary

### 2019 Mussel Tissue Results:

- Fe was detected at 100% of 17 sites surveyed (Figure 28)
- Fe concentration descriptive statistics (Figure 28):
  - Range: 112.96 – 761.86  $\mu\text{g/g}$  dw
  - Minimum Fe concentration was detected at PSPT
  - Maximum Fe concentration was detected at CBCH
  - Median: 282.47  $\mu\text{g/g}$  dw
  - Mean  $\pm$  SD:  $323.75 \pm 156.66 \mu\text{g/g}$  dw

### Historic Context for Mussel Tissue:

- 14% of sites surveyed were above their historic median concentrations in 2019 (Figure 29)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Fe concentrations in the Pacific Northwest ( $p = 0.64$ ,  $\rho = 0.10$ ) (Figure 30; Figure 31)
- 2 sites (PSMF, PSEF) in the Pacific Northwest showed significant decreasing temporal trends of Fe concentrations at  $\alpha = 0.05$  (Figure 29; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of Fe concentrations at  $\alpha = 0.05$  (Figure 29; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 32):
  - 100% of sites in low cluster (0.00 – 1,270.00  $\mu\text{g/g}$  dw)
  - 0% of sites in medium cluster (1,280.00 – 3,778.00  $\mu\text{g/g}$  dw)
  - 0% of sites in high cluster (3,991.00 – 17,600.00  $\mu\text{g/g}$  dw)

### General Observations:

- Fe concentrations were uniformly low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Regionally, Fe concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Fe pollution.

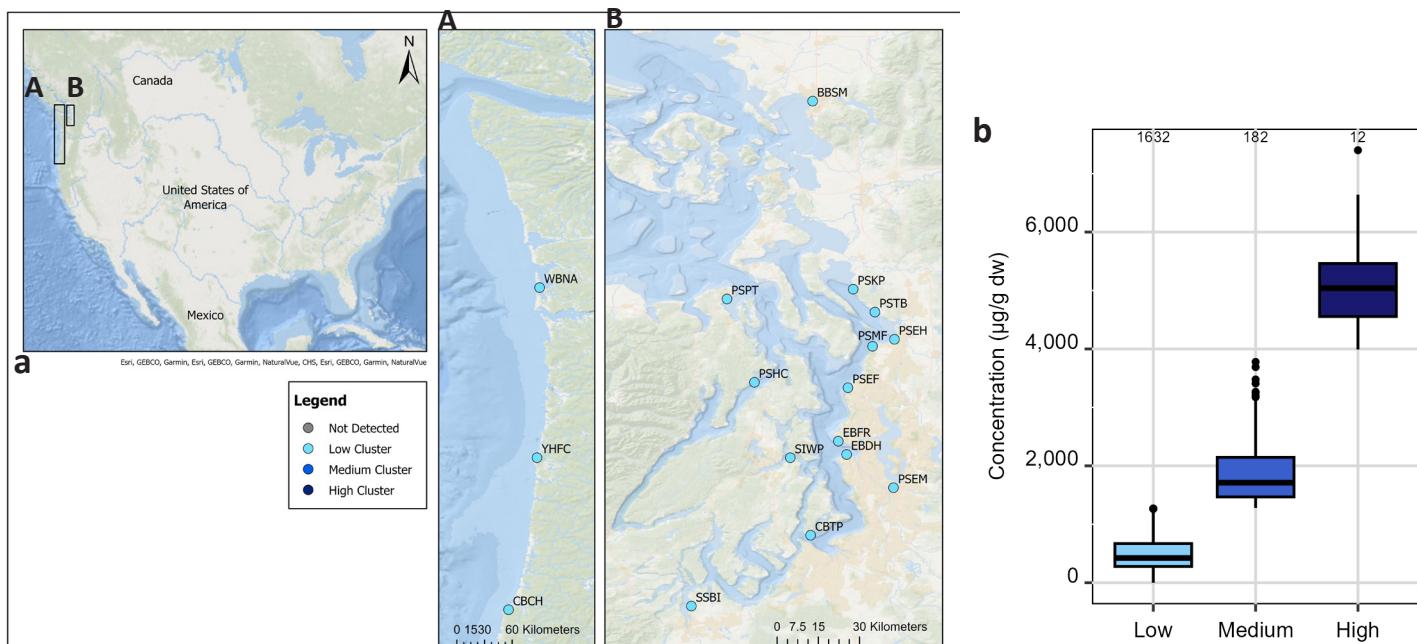


Figure 32. Iron concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP *Mytilus* species iron concentrations ( $\mu\text{g/g}$  dw). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples. Site PSMF in 2004 (17,600  $\mu\text{g/g}$  dw) was removed from 'b' to aid in visualization.

# Results - Lead (Pb)

## 10.0 RESULTS - LEAD (Pb)

### 11.1 Lead Chemical Description

Lead is a ubiquitous metal that occurs naturally in the Earth's crust. Release of lead into coastal waters are primarily linked with wastewater discharge, river runoff, atmospheric deposition and natural weathering of rock. Lead can be found in air, soil and surface water (ATSDR, 2007b). Environmental levels of lead increased worldwide over the past century because of leaded gasoline use (ATSDR, 2007b). Significant reductions in source and load resulted from the regulation of lead in gasoline and lead based paints. High levels found in the environment are usually linked to anthropogenic activities such as manufacturing processes, paint and pigment, solder, ammunition, plumbing, incineration and fossil fuel burning. In the communications industry, lead is still used extensively as protective sheathing for underground and underwater cables, including transoceanic cable systems (USGS, 2008).

Lead has no biological use and is toxic to many organisms, including humans. Exposure of fish to elevated concentrations of lead results in neurological deformities and black fins in fish (Mance, 1987). Lead primarily affects the nervous system, which results in decreased mental performance and inhibits typical mental development in humans. Exposure to lead may also cause brain and kidney damage and cancer (IARC, 2006). Safety guidance levels for lead in fish and shellfish are no longer listed by the US FDA (FDA, 2011).

Loadings of lead into coastal waters are primarily linked with wastewater discharge, river runoff, atmospheric deposition and natural weathering of rock. Lead can be found in air, soil and surface water. (ATSDR, 2007b).

### 10.2 Magnitude and Distribution of Lead in Mussel Tissue in 2019

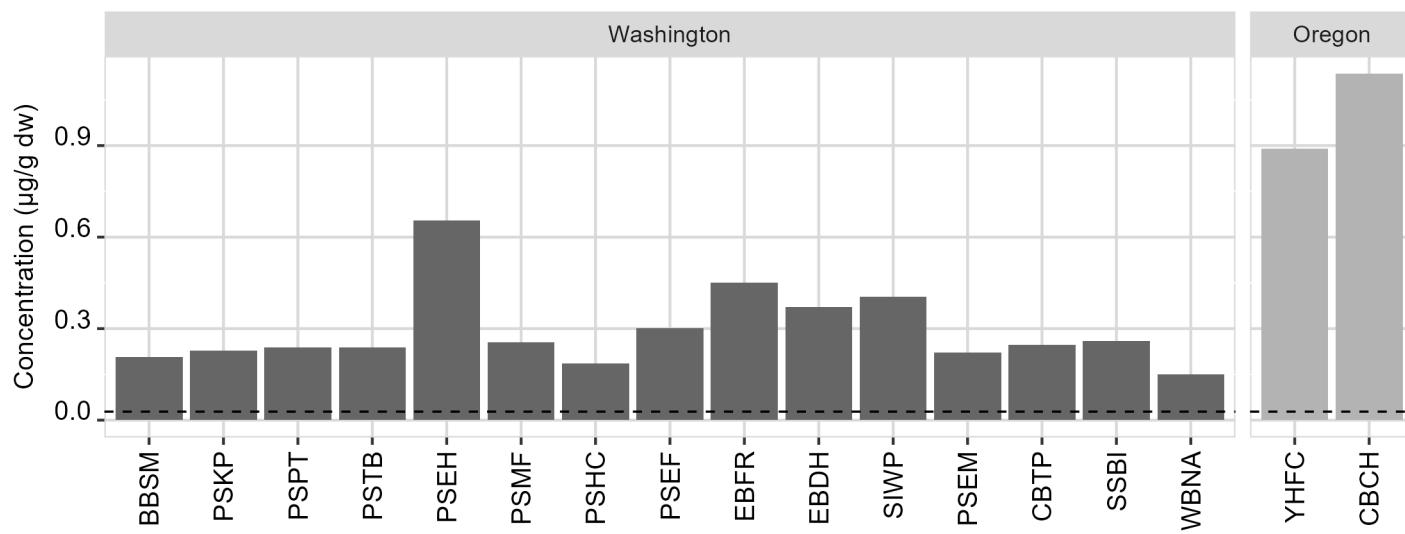


Figure 33. Bar graph showing magnitude of lead ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Lead (Pb)

## 10.3 Historical Context of Lead Magnitude and Distribution in Mussel Tissue

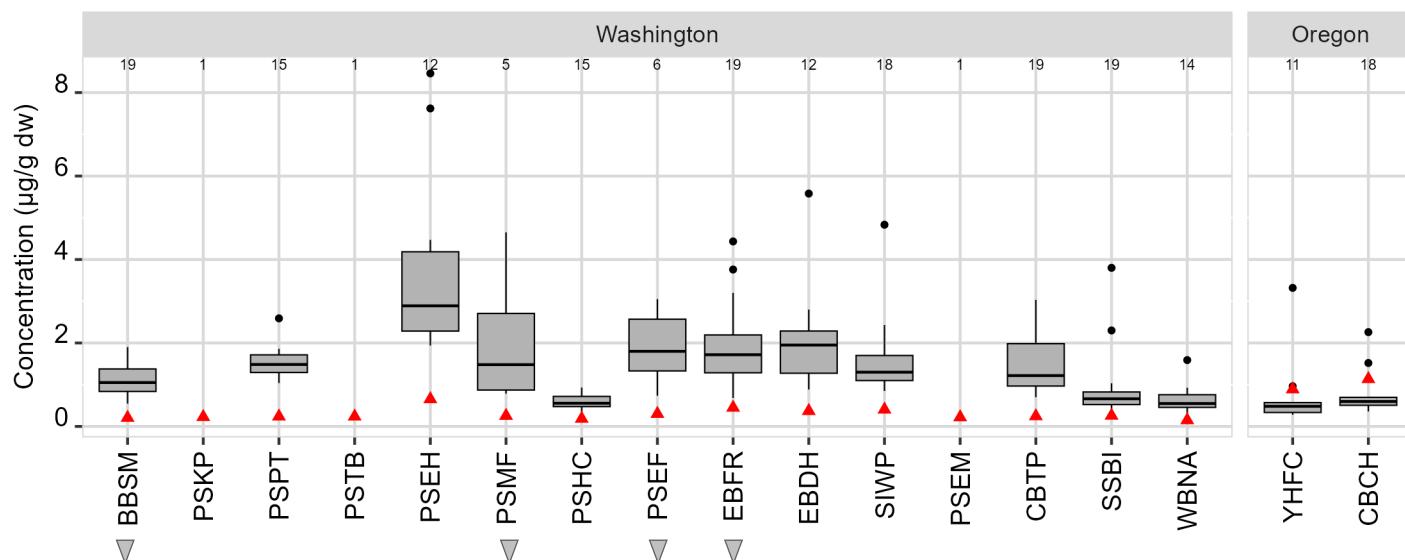


Figure 34. Lead concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic lead concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

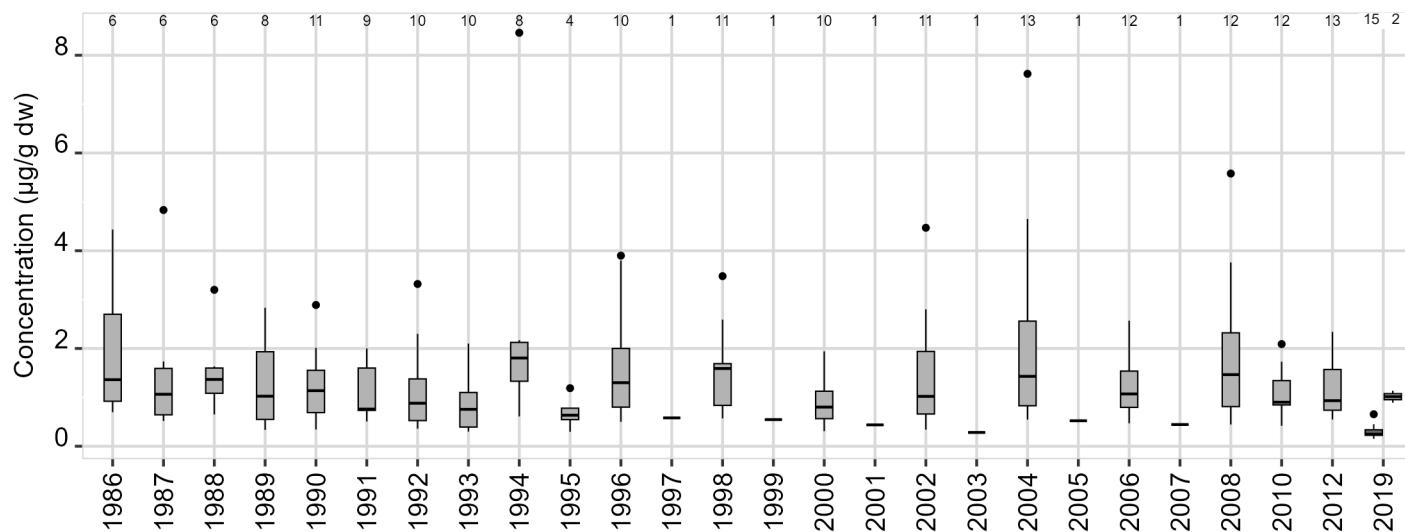


Figure 35. Boxplots representing the historic lead concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

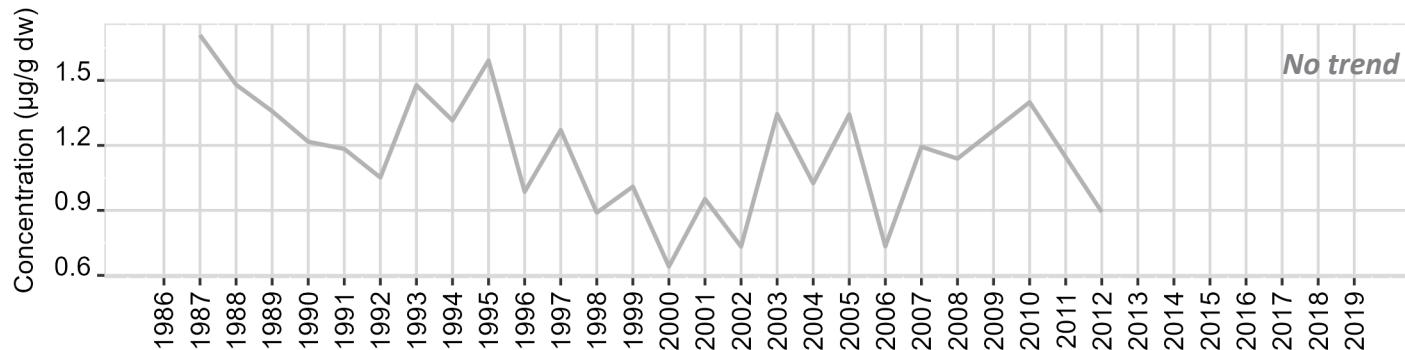


Figure 36. Three-point moving average of the yearly mean lead concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Lead (Pb)

## 10.4 Lead Summary

### 2019 Mussel Tissue Results:

- Pb was detected at 100% of 17 sites surveyed (Figure 33)
- Pb concentration descriptive statistics (Figure 33):
  - Range: 0.15 – 1.14  $\mu\text{g/g dw}$
  - Minimum Pb concentration was detected at WBNA
  - Maximum Pb concentration was detected at CBCH
  - Median: 0.25  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $0.38 \pm 0.27 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 14% of sites surveyed were above their historic median concentrations in 2019 (Figure 34)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Pb concentrations in the Pacific Northwest ( $p = 0.06$ ,  $\rho = -0.38$ ) (Figure 35; Figure 36)
- 4 sites (BBSM, PSMF, PSEF, EBFR) in the Pacific Northwest showed significant decreasing temporal trends of Pb concentrations at  $\alpha = 0.05$  (Figure 34; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of Pb concentrations at  $\alpha = 0.05$  (Figure 34; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 37):
  - 100% of sites in low cluster (0.00 – 4.35  $\mu\text{g/g dw}$ )
  - 0% of sites in medium cluster (4.38 – 12.00  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (12.67 – 44.60  $\mu\text{g/g dw}$ )

### General Observations:

- Pb concentrations were uniformly low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Regionally, Pb concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Pb pollution.

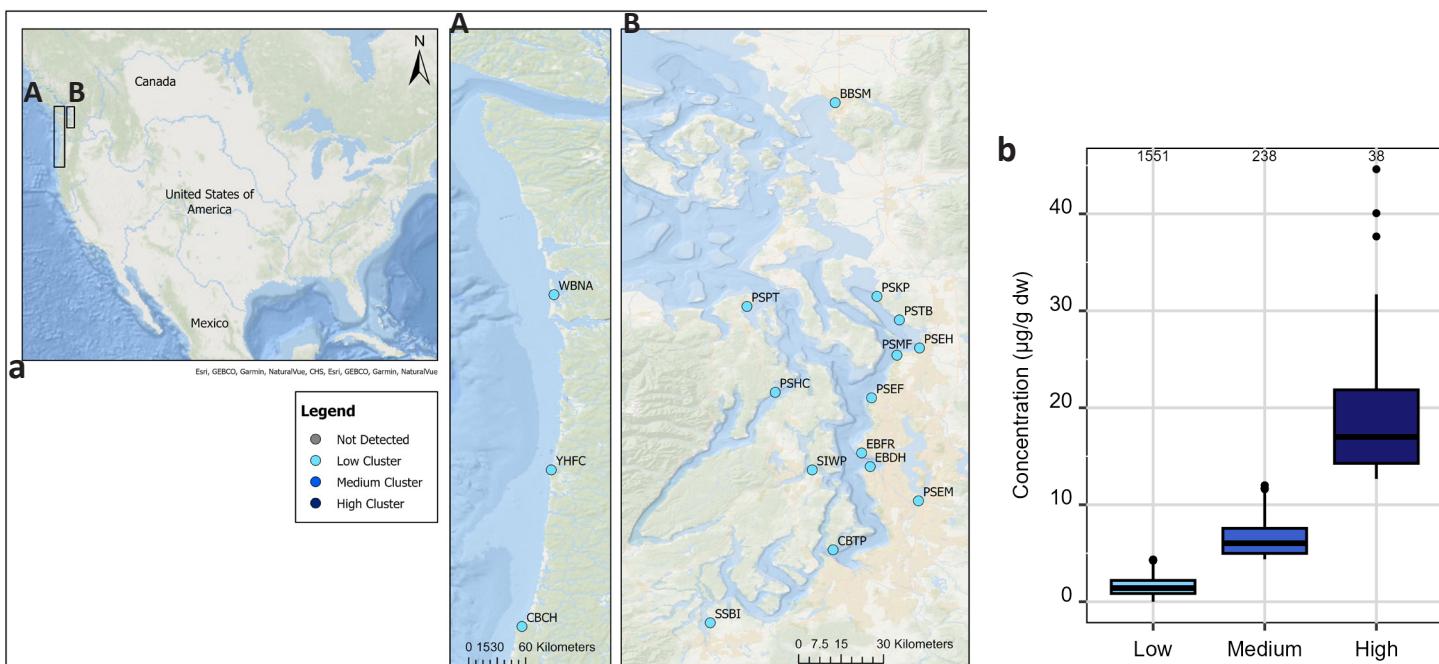


Figure 37. Lead concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP Mytilus species lead concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples.

# Results - Manganese (Mn)

## 11.0 RESULTS - MANGANESE (Mn)

### 11.1 Manganese Chemical Description

Manganese is found in the environment both naturally and from anthropogenic sources including industrial production, pharmaceuticals, and gasoline (ATSDR, 2012b). Manganese occurs naturally in Earth's crust combined with other substances like oxygen, sulfur, and chlorine and is also a normal component of air, soil, water, and food. Humans are routinely exposed to low levels of manganese through drinking water, ground water, and soil. Anthropogenic sources of manganese include use in steel production to improve the hardness, stiffness, and strength of carbon steel, stainless steel, high-temperature steel, tool steel, cast iron, and superalloys. Manganese can also be found in products such as fireworks, dry-cell batteries, fertilizer, paints, medical imaging agents, cosmetics, and gasoline. Atmospheric manganese can result from industrial activities, mining, automobile exhaust, and cigarette smoke (ATSDR, 2012b).

While manganese is an essential nutrient, it can have negative effects at high doses (ATSDR, 2012b). Typically, manganese enters the body through inhalation, ingestion, and dermal contact. Primarily, humans are exposed to manganese through manganese-containing nutritional supplements or food, especially grains, beans, nuts, and tea. Additionally, occupations such as welding or steel-making may increase the chances of high manganese exposure. High exposure to manganese often impacts the nervous system in the form of behavioral changes and slow, clumsy movements – referred to as “manganism”. Additional observed effects include lung irritation resulting in pneumonia and negative changes in the male reproductive system. Typically, the concentrations required to produce these negative effects are twenty thousand times higher than concentrations normally found in the environment. The EPA has established that exposure to manganese in drinking water at concentrations of 1 mg/L for 1 or 10 days is not expected to cause any adverse effects and OSHA set a legal limit of 5 mg/m<sup>3</sup> manganese in air averaged over an 8-hour work day (ATSDR, 2012b).

Manganese does not break down in the environment and can only change form or become attached or separated from particles (ATSDR, 2012b). The chemical state of manganese and the type of soil determine how fast it moves through the soil and how much is retained in the soil. Most manganese in water tends to attach to particles or settle into the sediment and may be significantly bioconcentrated at lower trophic levels and fish. Manganese found in gasoline additives may degrade in the environment quickly when exposed to sunlight, releasing manganese (ATSDR, 2012b).

### 11.2 Magnitude and Distribution of Manganese in Mussel Tissue in 2019

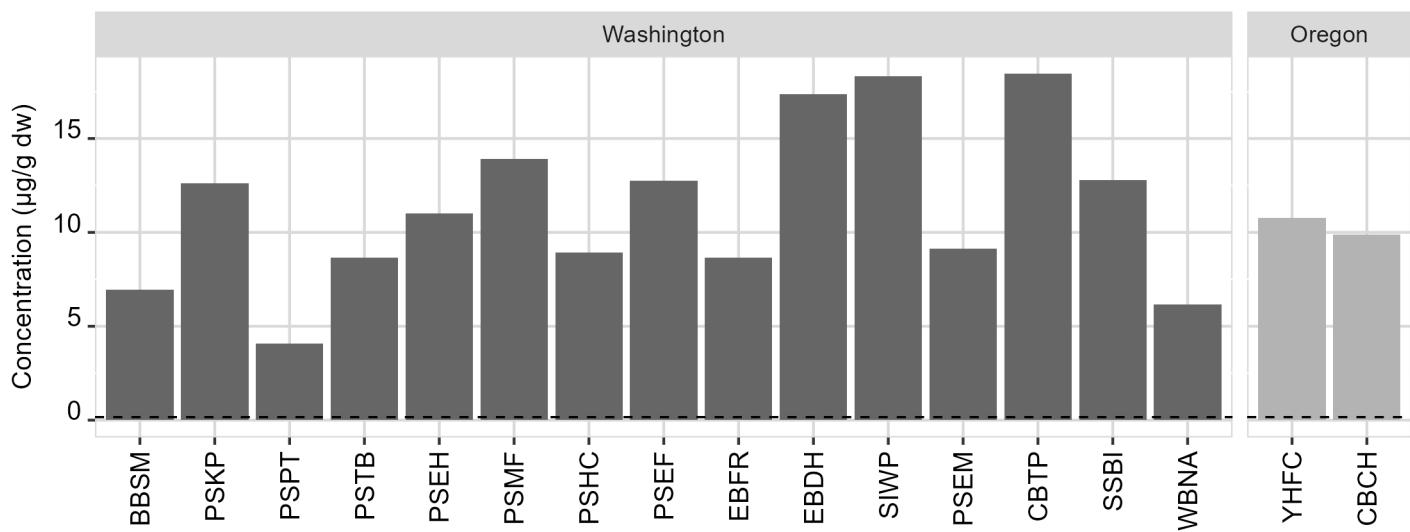


Figure 38. Bar graph showing magnitude of manganese (µg/g dw) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Manganese (Mn)

## 11.3 Historical Context of Manganese Magnitude and Distribution in Mussel Tissue

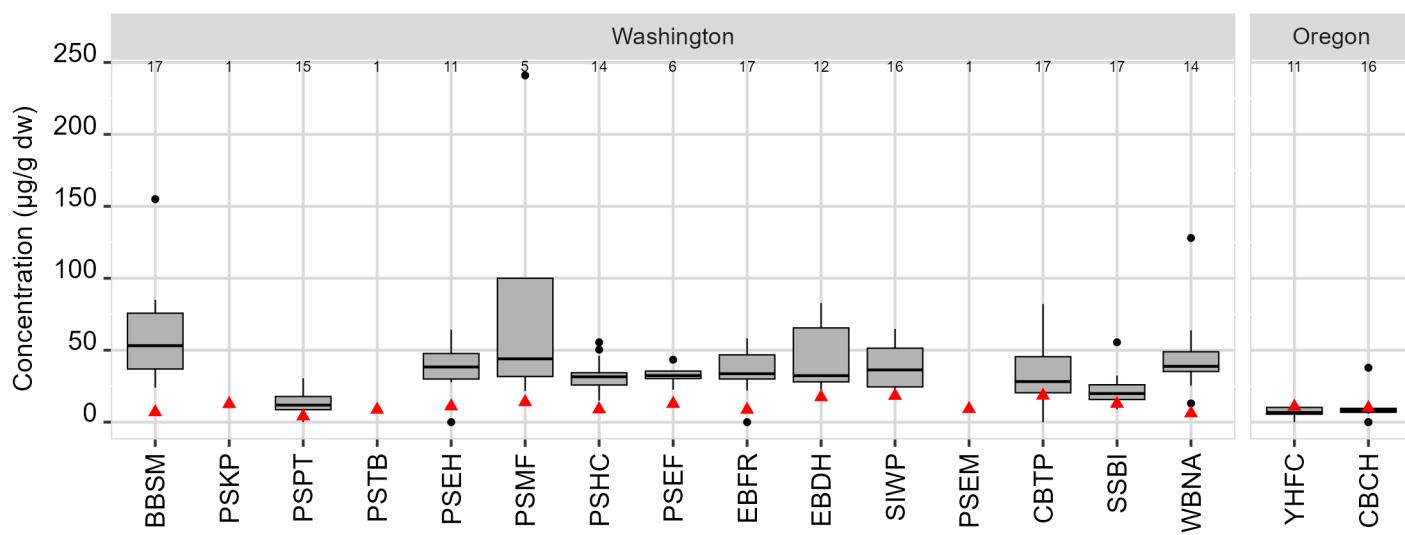


Figure 39. Manganese concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic manganese concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

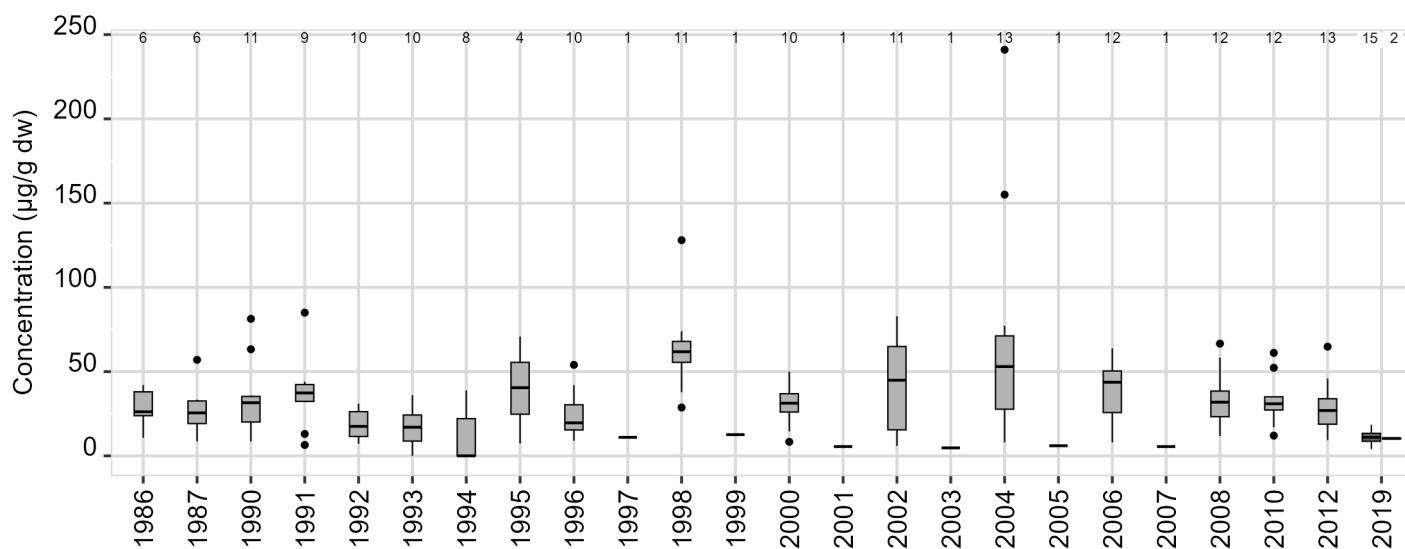


Figure 40. Boxplots representing the historic manganese concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

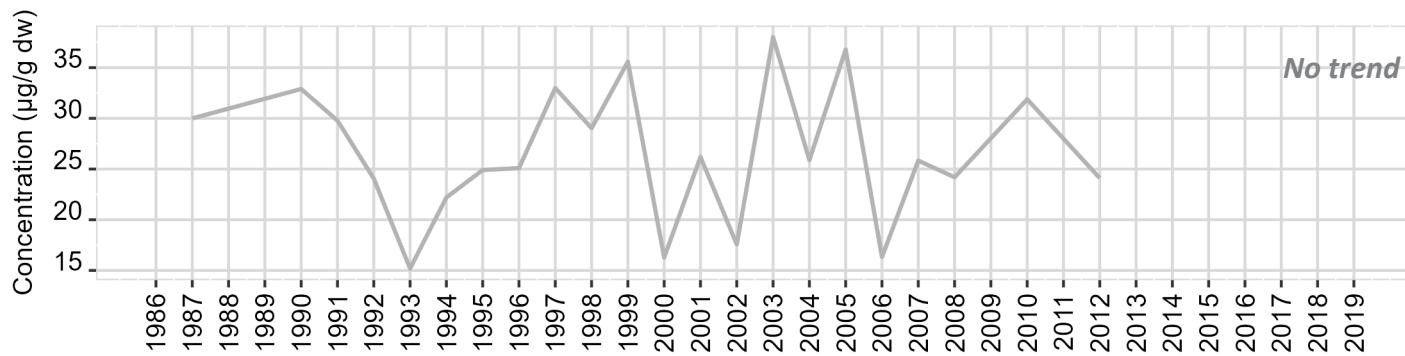


Figure 41. Three-point moving average of the yearly mean manganese concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Manganese (Mn)

## 11.4 Manganese Summary

### 2019 Mussel Tissue Results:

- Mn was detected at 100% of 17 sites surveyed (Figure 38)
- Mn concentration descriptive statistics (Figure 38):
  - Range: 4.06 – 18.47  $\mu\text{g/g dw}$
  - Minimum Mn concentration was detected at PSPT
  - Maximum Mn concentration was detected at CBTP
  - Median: 10.78  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $11.20 \pm 4.14 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 14% of sites surveyed were above their historic median concentrations in 2019 (Figure 39)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Mn concentrations in the Pacific Northwest ( $p = 0.78$ ,  $\rho = -0.06$ ) (Figure 40; Figure 41)
- 2 sites (PSMF, PSEF) in the Pacific Northwest showed significant decreasing temporal trends of Mn concentrations at  $\alpha = 0.05$  (Figure 39; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of Mn concentrations at  $\alpha = 0.05$  (Figure 39; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 42):
  - 100% of sites in low cluster (0.00 – 44.10  $\mu\text{g/g dw}$ )
  - 0% of sites in medium cluster (44.80 – 121.00  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (125.00 – 580.00  $\mu\text{g/g dw}$ )

### General Observations:

- Mn concentrations were uniformly low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Regionally, Mn concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Mn pollution.

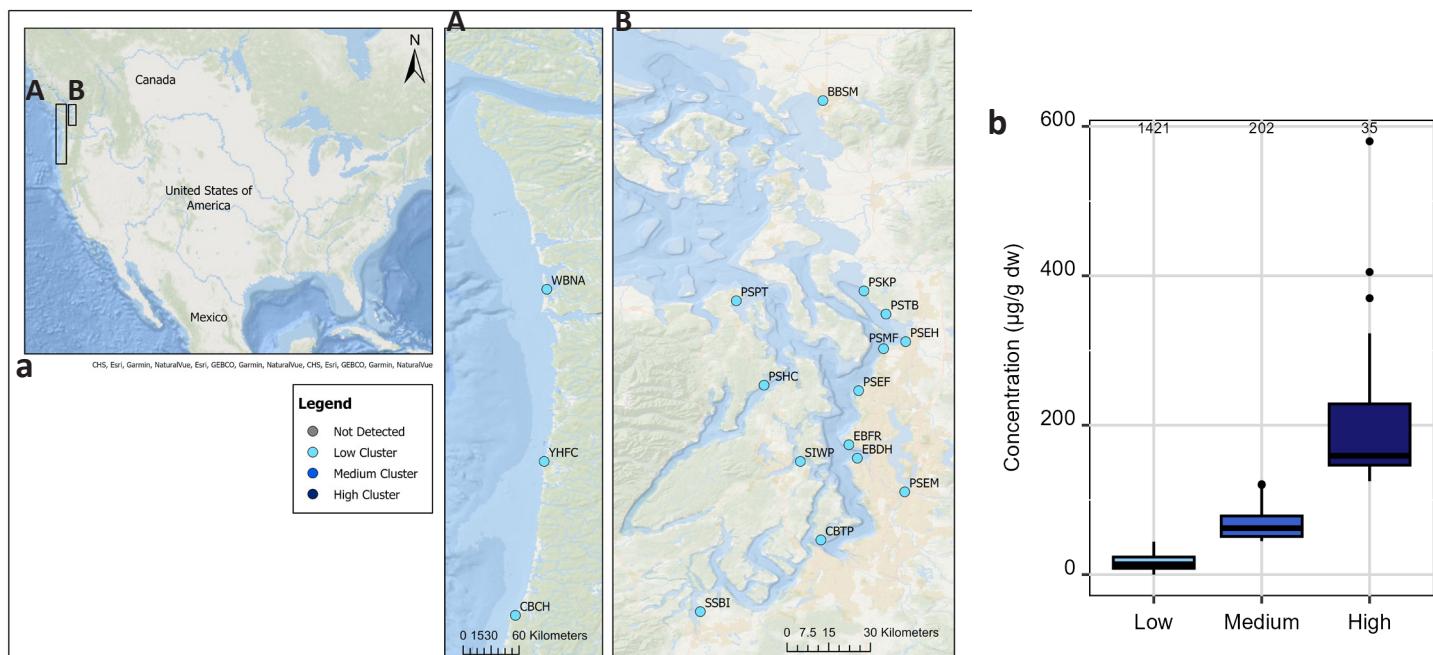


Figure 42. Manganese concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP Mytilus species manganese concentrations ( $\mu\text{g/g dw}$ ). 211 national sites sampled between 1986 - 2021 for a total of 1658 samples.

# Results - Mercury (Hg)

## 12.0 RESULTS - MERCURY (Hg)

### 12.1 Mercury Chemical Description

Mercury is found naturally as mercuric sulfide from forest fires, crustal ores, fossil fuels, and volcanoes. Anthropogenic sources of mercury are from mining, gold refining, coal fired power plants, and the wood pulp industry. Elevated levels occur as a result of human activity (ATSDR, 1999b). In the US, coal fired-electric turbines, municipal and medical waste incinerators, mining, landfills, and sewage sludge are the primary emitters of mercury into the air.

Mercury is a highly toxic, non-essential trace metal that occurs naturally. Mercury is a human neurotoxin that also affects the kidneys and developing fetuses. The most common human exposure route for mercury is the consumption of contaminated food. The US FDA has not established a safety level for mercury but has set a safety level of 1.0 ppm wet weight for methyl mercury, the form most likely to impact animal and humans (FDA, 2011). The Mussel Watch Program measures total mercury, of which methyl mercury is only one component. Children, pregnant women, or women likely to become pregnant are advised to avoid consumption of swordfish, shark, king mackerel and tilefish and should limit consumption to fish and shellfish recommended by US FDA and US EPA (EPA, 1979; FDA, 2011). Mercury exposure can also cause reduced growth and reproduction rates in other phyla such as copepods (Eisler, 1987).

In the environment, mercury may change forms between elemental, inorganic and organic. Natural sinks, such as sediment and soil, represent the largest source of mercury to the environment. Estimates suggest that wet and dry deposition accounts for 50-90% of the mercury load to many estuaries, making atmospheric transport a significant source of mercury worldwide. Long range atmospheric transport is responsible for the presence of mercury at, or above, background levels in surface waters in remote areas.

### 12.2 Magnitude and Distribution of Mercury in Mussel Tissue in 2019

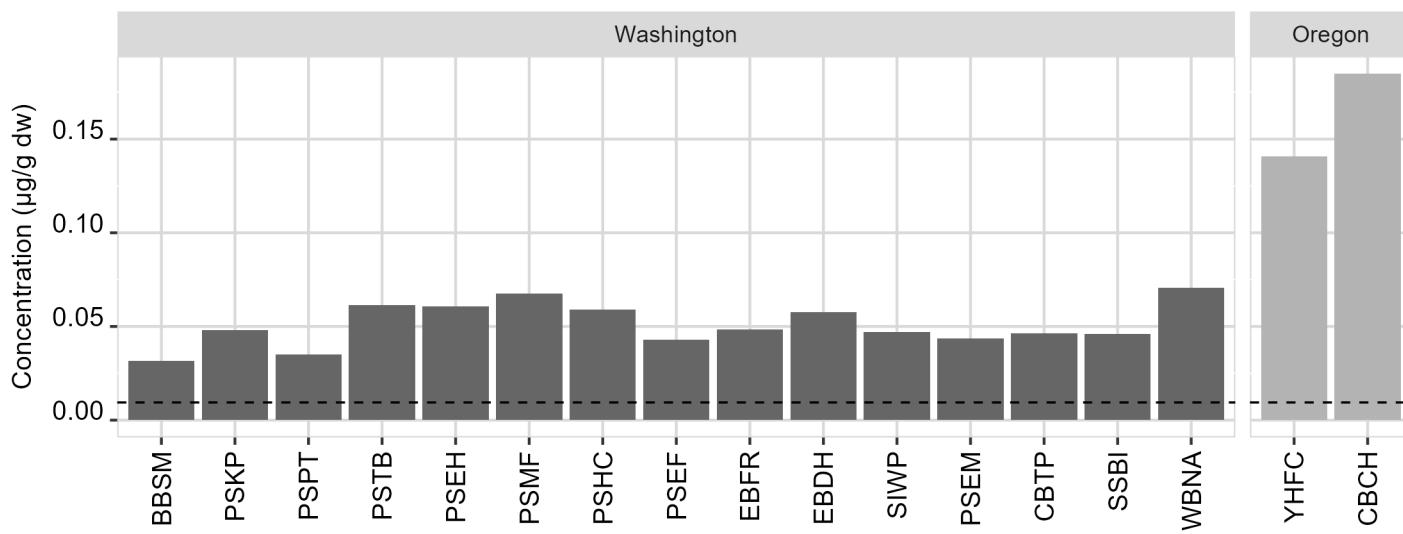


Figure 43. Bar graph showing magnitude of mercury ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Mercury (Hg)

## 12.3 Historical Context of Mercury Magnitude and Distribution in Mussel Tissue

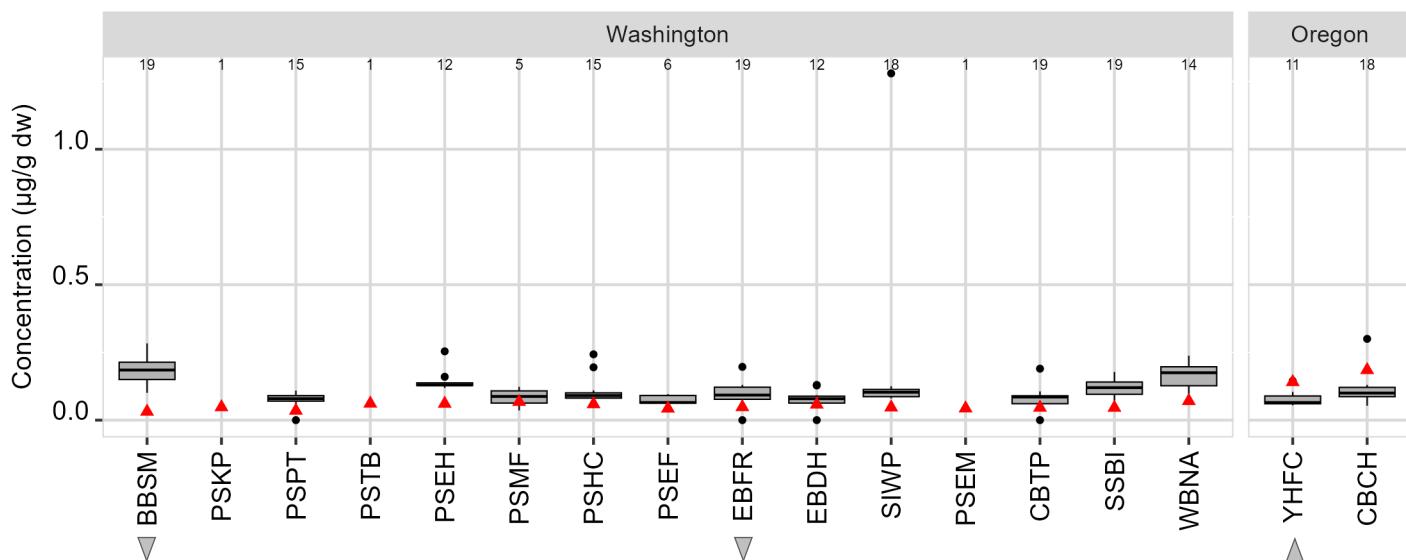


Figure 44. Mercury concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic mercury concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

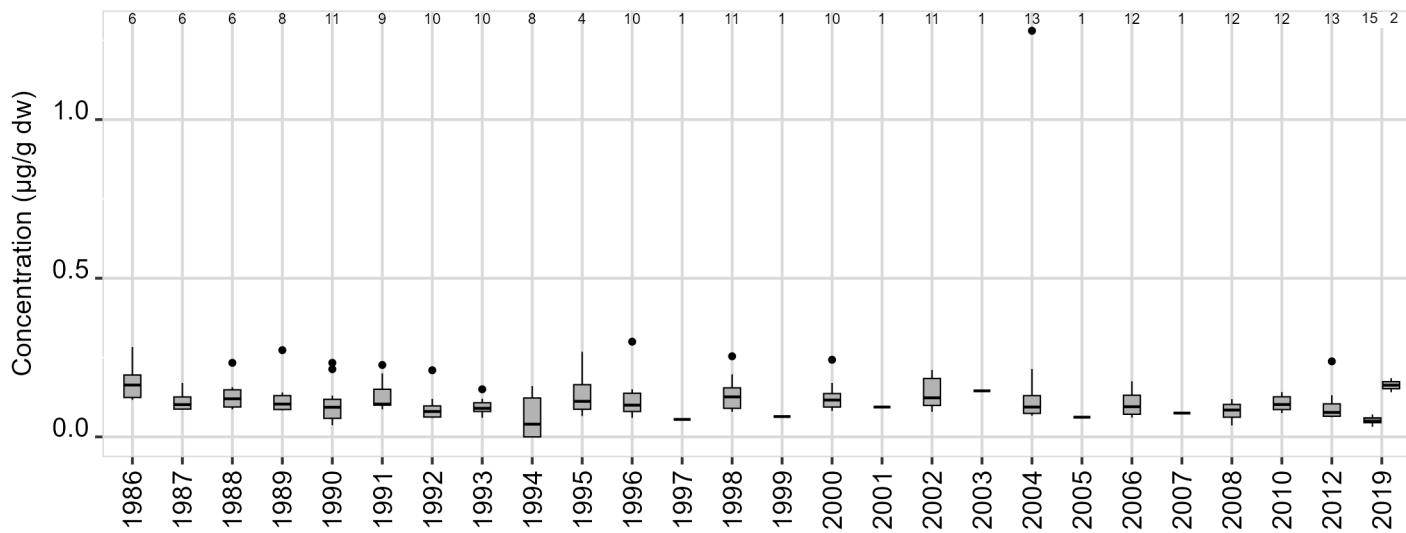


Figure 45. Boxplots representing the historic mercury concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

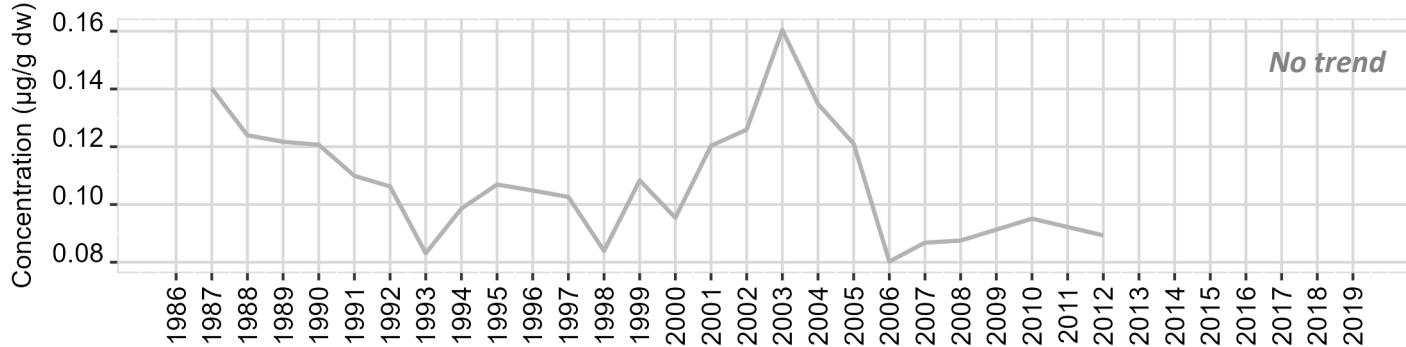


Figure 46. Three-point moving average of the yearly mean mercury concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Mercury (Hg)

## 12.4 Mercury Summary

### 2019 Mussel Tissue Results:

- Hg was detected at 100% of 17 sites surveyed (Figure 43)
- Hg concentration descriptive statistics (Figure 43):
  - Range: 0.03 – 0.19  $\mu\text{g/g dw}$
  - Minimum Hg concentration was detected at BBSM
  - Maximum Hg concentration was detected at CBCH
  - Median: 0.05  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD: 0.06  $\pm$  0.04  $\mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 14% of sites surveyed were above their historic median concentrations in 2019 (Figure 44)
- 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Hg concentrations in the Pacific Northwest ( $p = 0.13$ ,  $\rho = -0.30$ ) (Figure 45; Figure 46)
- 2 sites (BBSM, EBFR) in the Pacific Northwest showed significant decreasing temporal trends of Hg concentrations at  $\alpha = 0.05$  (Figure 44; Table A4)
- 1 site (YHFC) in the Pacific Northwest showed a significant increasing temporal trend of Hg concentrations at  $\alpha = 0.05$  (Figure 44; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 47):
  - 94% of sites in low cluster (0.00 – 0.16  $\mu\text{g/g dw}$ )
  - 6% of sites in medium cluster (0.16 – 0.29  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (0.30 – 2.07  $\mu\text{g/g dw}$ )

### General Observations:

- Hg concentrations were generally low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Medium Hg concentrations were detected in mussel tissue off the Oregon coast, possibly indicating offshore natural sources or localized point sources of contamination.
- Regionally, Hg concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Hg pollution.

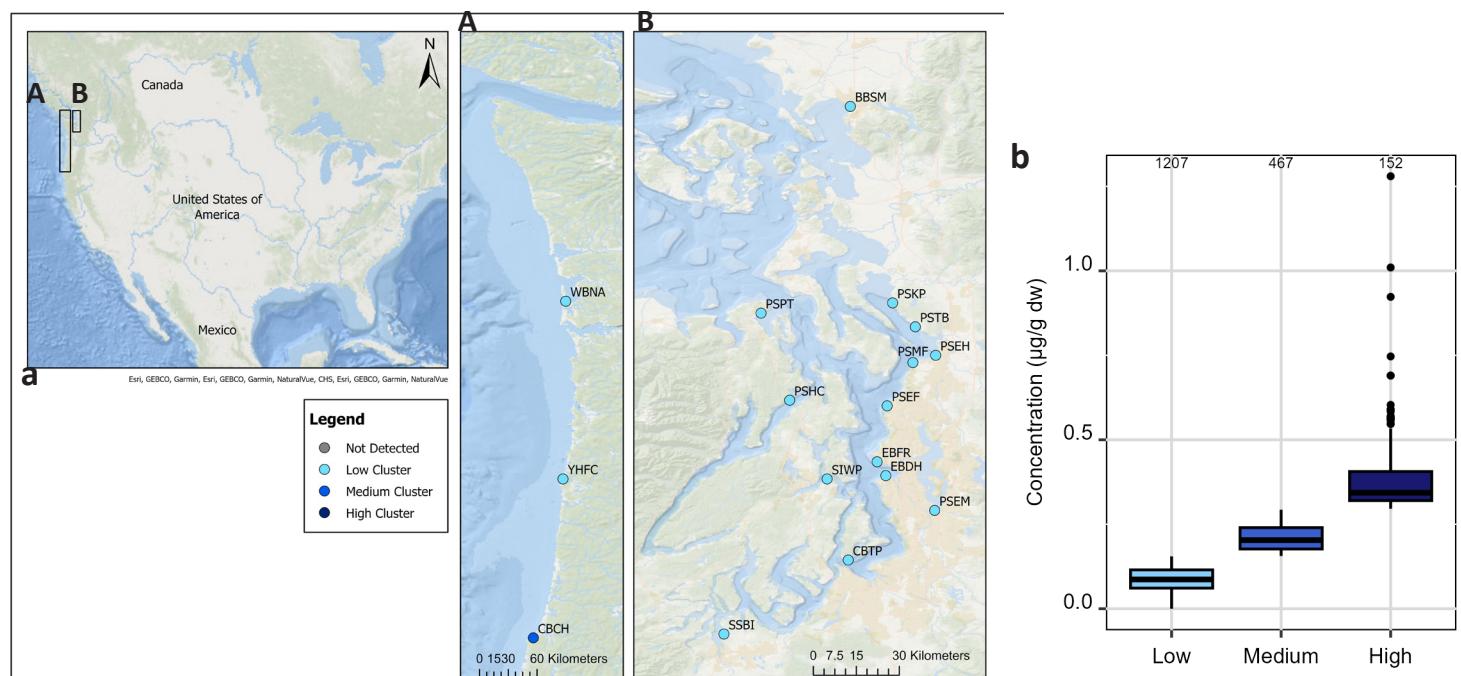


Figure 47. Mercury concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP Mytilus species mercury concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples. Site SMOH in 2018 (2.07  $\mu\text{g/g dw}$ ) was removed from 'b' to aid in visualization.

# Results - Nickel (Ni)

## 13.0 RESULTS - NICKEL (Ni)

### 13.1 Nickel Chemical Description

Nickel is naturally occurring and widely distributed in the environment. It exists in alloy form in combination with other metals and as a soluble element. Naturally, nickel is derived from weathering rocks and soil and is transported to streams and rivers by runoff. River and stream input of nickel are the largest sources for oceans and coastal waters. Anthropogenic sources of nickel can be attributed to the presence of nickel in stainless steel, nickel-cadmium batteries, pigments, computers, wire, coinage, and electroplating (ATSDR, 2005a). Atmospheric sources are usually not significant, with the exception of in the Great Lakes, where the atmospheric input of nickel accounts for 60-80% of the total anthropogenic input to Lake Superior, and 20-70% of total inputs to Lakes Erie and Ontario (Nriagu et al., 1995). In Lakes Erie and Ontario, most of the nickel is derived from municipal and industrial wastewaters and thus is likely to be complexed with organic matter (Sweet et al., 1998).

Nickel is a biologically essential trace element for animals and plants and is widely distributed in the environment at very low concentrations. Exposure to large doses of nickel can cause serious health effects, such as bronchitis, and long-term exposure can result in cancer. Food is the major source of human exposure to nickel (ATSDR, 2005a). Safety guidance levels for nickel in fish and shellfish are no longer listed by the US FDA (FDA, 2011).

Nickel derived from weathering rocks and soil is transported to streams and rivers by runoff. It accumulates in sediment and becomes inert when it is incorporated into minerals. There is no evidence that nickel biomagnifies in the food chain (McGeer et al., 2003; Suedel et al., 1994). Complexation reduces the chemical reactivity and bioavailability of Ni and may account for the high concentrations and long residence times in the water column (Nriagu et al., 1995).

### 13.2 Magnitude and Distribution of Nickel in Mussel Tissue in 2019

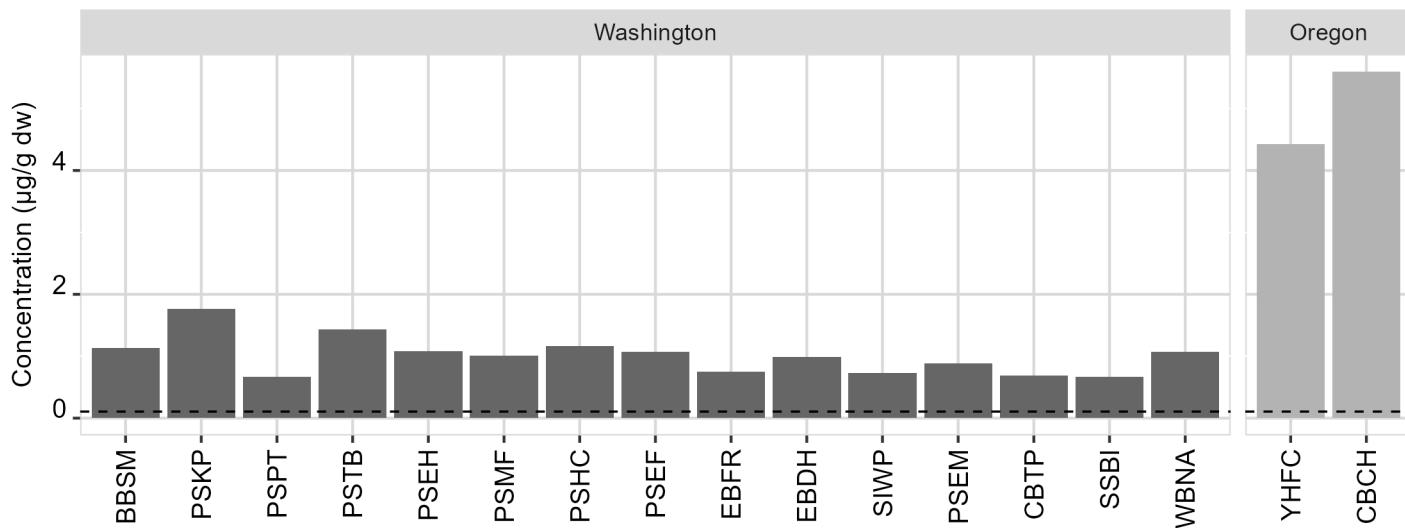


Figure 48. Bar graph showing magnitude of nickel ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Nickel (Ni)

## 13.3 Historical Context of Nickel Magnitude and Distribution in Mussel Tissue

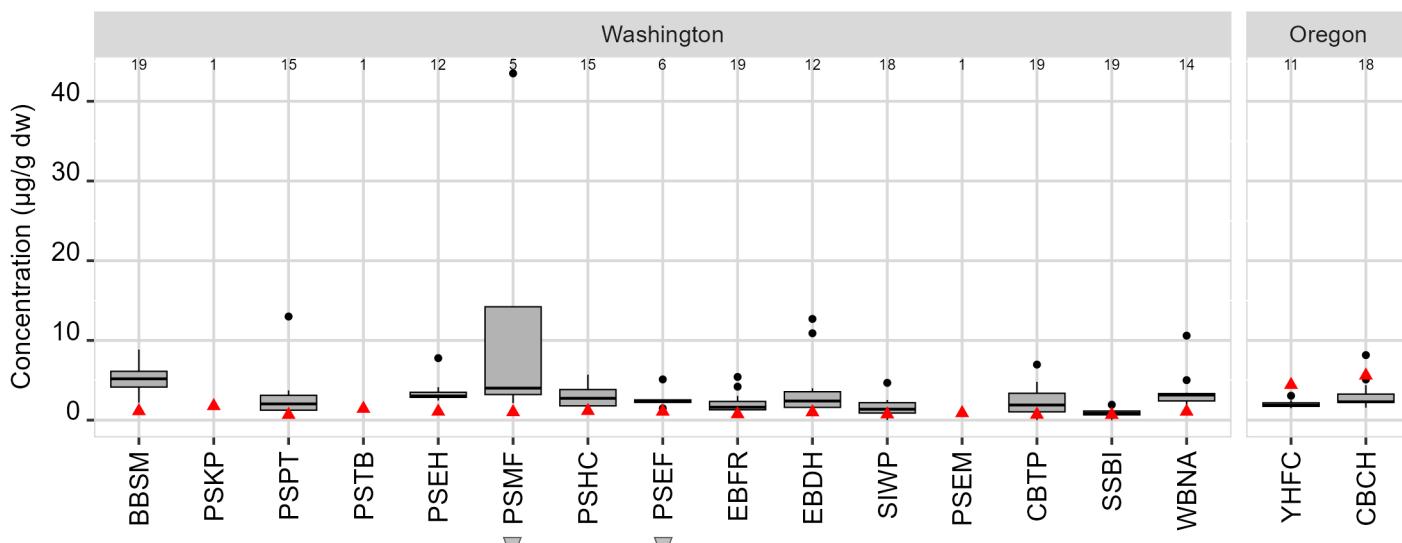


Figure 49. Nickel concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic nickel concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

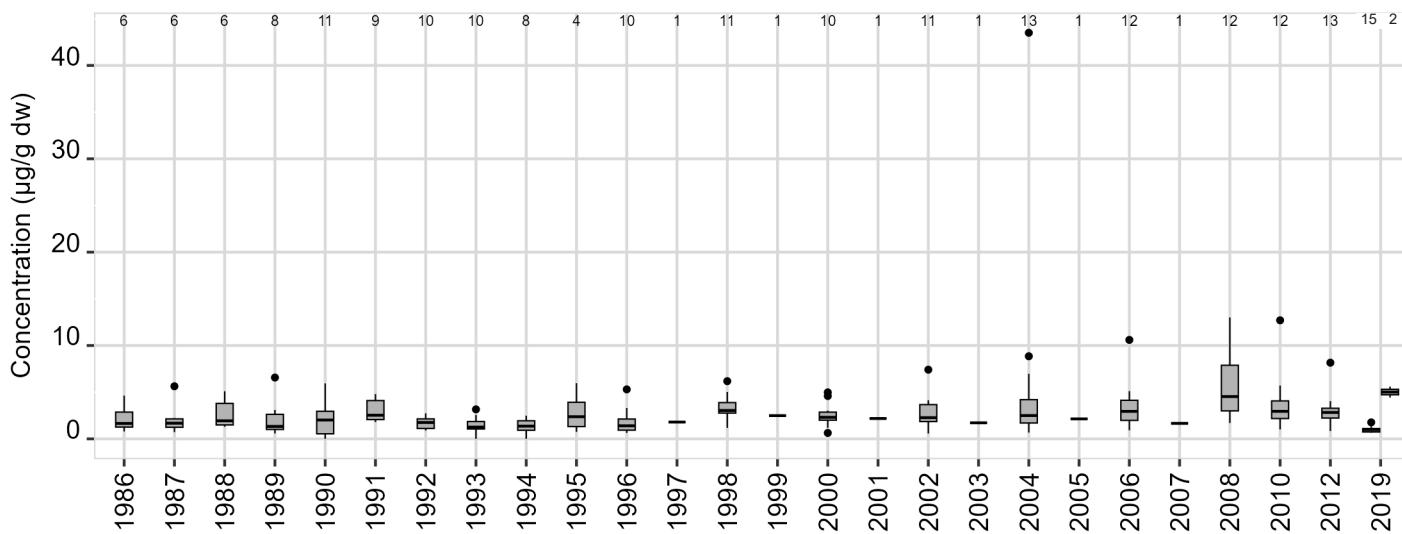


Figure 50. Boxplots representing the historic nickel concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

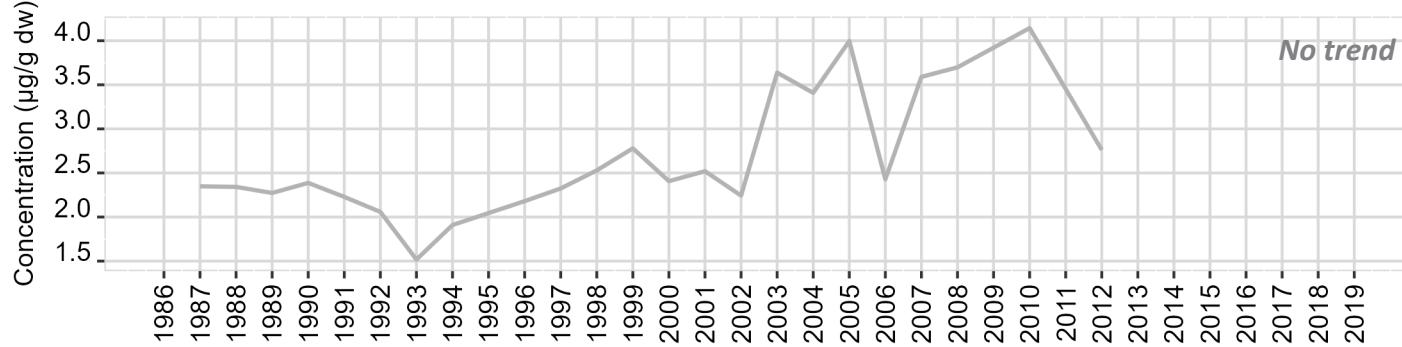


Figure 51. Three-point moving average of the yearly mean nickel concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Nickel (Ni)

## 13.4 Nickel Summary

### 2019 Mussel Tissue Results:

- Ni was detected at 100% of 17 sites surveyed (Figure 48)
- Ni concentration descriptive statistics (Figure 48):
  - Range: 0.67 – 5.60  $\mu\text{g/g dw}$
  - Minimum Ni concentration was detected at SSBI
  - Maximum Ni concentration was detected at CBCH
  - Median: 1.07  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $1.48 \pm 1.38 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 14% of sites surveyed were above their historic median concentrations in 2019 (Figure 49)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Ni concentrations in the Pacific Northwest ( $p = 0.27$ ,  $\rho = 0.22$ ) (Figure 50; Figure 51)
- 2 sites (PSMF, PSEF) in the Pacific Northwest showed significant decreasing temporal trends of Ni concentrations at  $\alpha = 0.05$  (Figure 49; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of Ni concentrations at  $\alpha = 0.05$  (Figure 49; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 52):
  - 88% of sites in low cluster (0.00 – 2.27  $\mu\text{g/g dw}$ )
  - 12% of sites in medium cluster (2.28 – 6.80  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (6.96 – 43.50  $\mu\text{g/g dw}$ )

### General Observations:

- Ni concentrations were generally low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Medium Ni concentrations were detected in mussel tissue off the Oregon coast, possibly indicating offshore natural sources or localized point sources of contamination.
- Regionally, Ni concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Ni pollution.

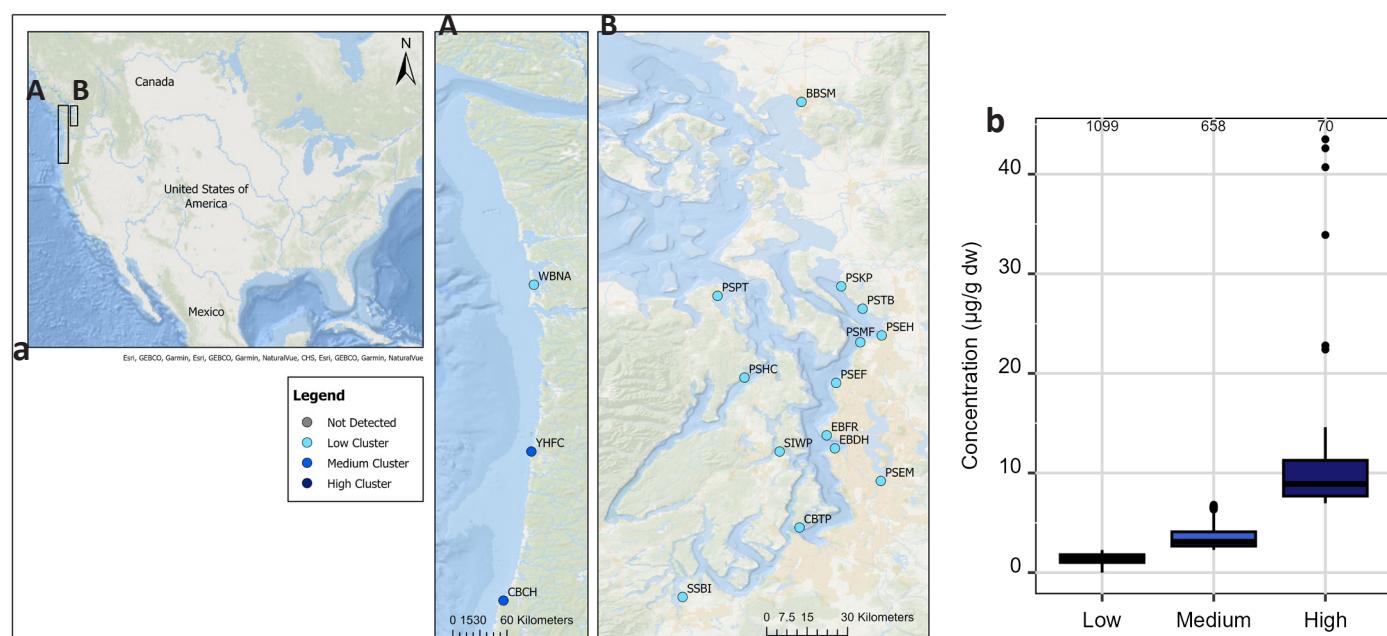


Figure 52. Nickel concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP *Mytilus* species nickel concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples.

# Results - Selenium (Se)

## 14.0 RESULTS - SELENIUM (Se)

### 14.1 Selenium Chemical Description

Selenium naturally occurs in the Earth's crust and is commonly found in rocks and soil (ATSDR, 2003). Weathering of rocks and soil can result in low levels of selenium in water and in the air as dust-like particles. Additionally, volcanic eruptions may release atmospheric selenium. Selenium is also commercially produced as a byproduct of copper refining or from burning coal or oil. Other products that selenium compounds can be found in include photographic devices, gun bluing, plastics, paints, anti-dandruff shampoos, vitamin and mineral supplements, fungicides, glass, drug preparation, and livestock feed supplements. Regardless of source, selenium is not often found in its elemental form in the environment, but rather combined with sulfide, silver, copper, lead, or nickel minerals (ATSDR, 2003).

Selenium is an essential nutrient in low concentrations and is usually ingested by humans through drinking water, food, and air (ATSDR, 2003). Selenium is often contained in soils and varies by region, so depending on where food is grown, humans may be exposed to differing levels of selenium. Humans are not typically exposed to high selenium levels but this can vary with occupation, specifically for individuals employed in the metal industries, selenium-recovery processes, paint manufacturing, and special trades. When exposed at high levels, dizziness, fatigue, and irritation of mucous membranes can occur and even mild exposure over long time periods can result in brittle hair, deformed nails, and even loss of feeling and control in extremities (called selenosis). Conditions this extreme usually occur in areas with locally high levels of selenium where the population eats locally. For most people in the US who eat food from many different areas, overexposure to selenium is unlikely to occur. There is no support that selenium causes cancer except for one specific form called selenium sulfide which has been shown to cause cancer in animals. FDA regulations allow a level of 50 ppb of selenium in bottled water and OSHA set the exposure limit for selenium compounds in the air for an 8-hour period at 0.2 mg selenium/m<sup>3</sup> (ATSDR, 2003).

The forms and fate of selenium in soil depend largely on the acidity of the surroundings and its interaction with oxygen (ATSDR, 2003). In the absence of oxygen when the soil is acidic, the amount of selenium that can enter plants and organisms should be low. Elemental selenium that cannot dissolve in water and other insoluble forms of selenium are less mobile and will usually remain in the soil, posing a smaller risk of exposure. Selenium compounds that can dissolve in water are sometimes very mobile; thus, there is an increased chance of exposure to these compounds. Some evidence indicates that selenium can be taken up by and bioaccumulate in tissues of aquatic organisms. Selenium concentrations in aquatic organisms have been a problem as a result of irrigation runoff in some areas of the US (ATSDR, 2003).

### 14.2 Magnitude and Distribution of Selenium in Mussel Tissue in 2019

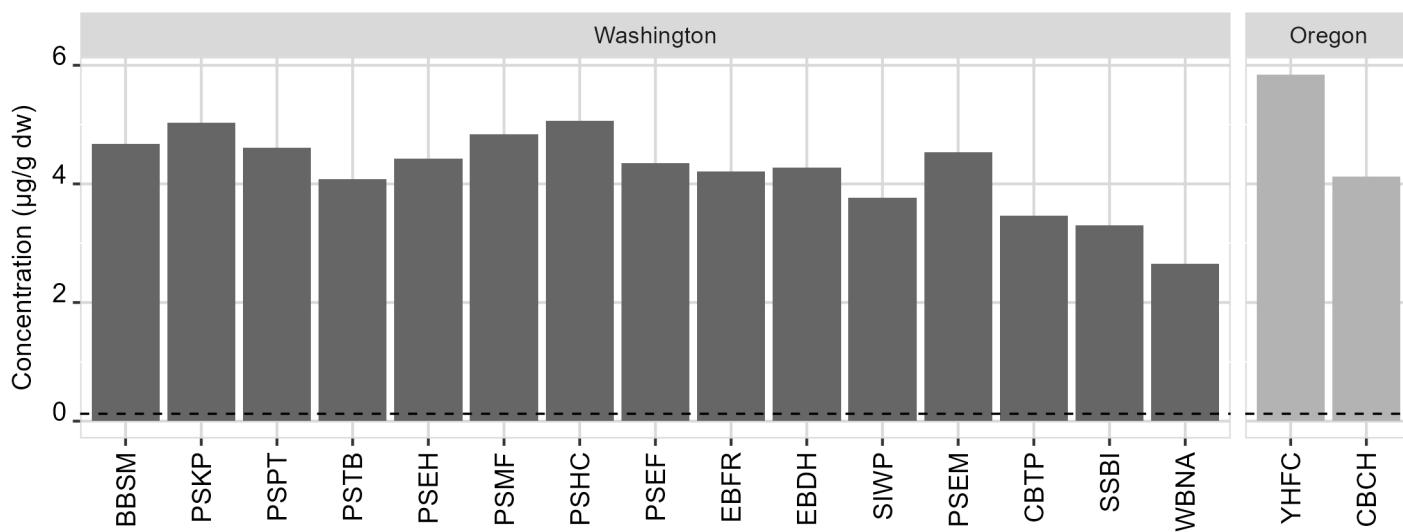


Figure 53. Bar graph showing magnitude of selenium (µg/g dw) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Selenium (Se)

## 14.3 Historical Context of Selenium Magnitude and Distribution in Mussel Tissue

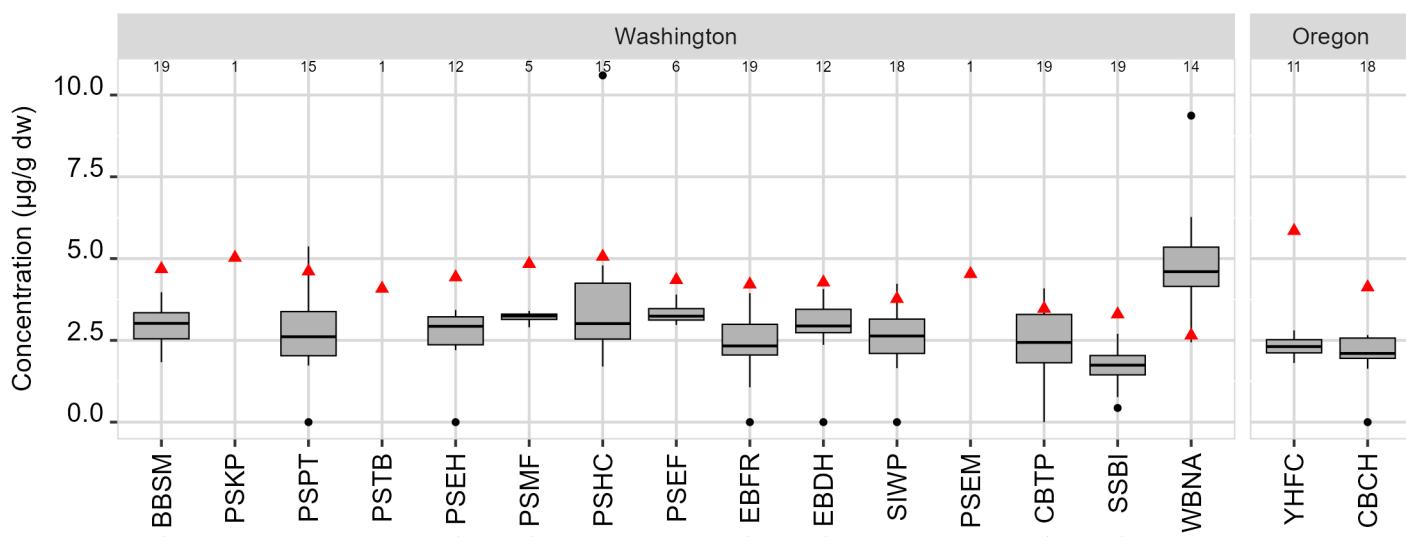


Figure 54. Selenium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic selenium concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

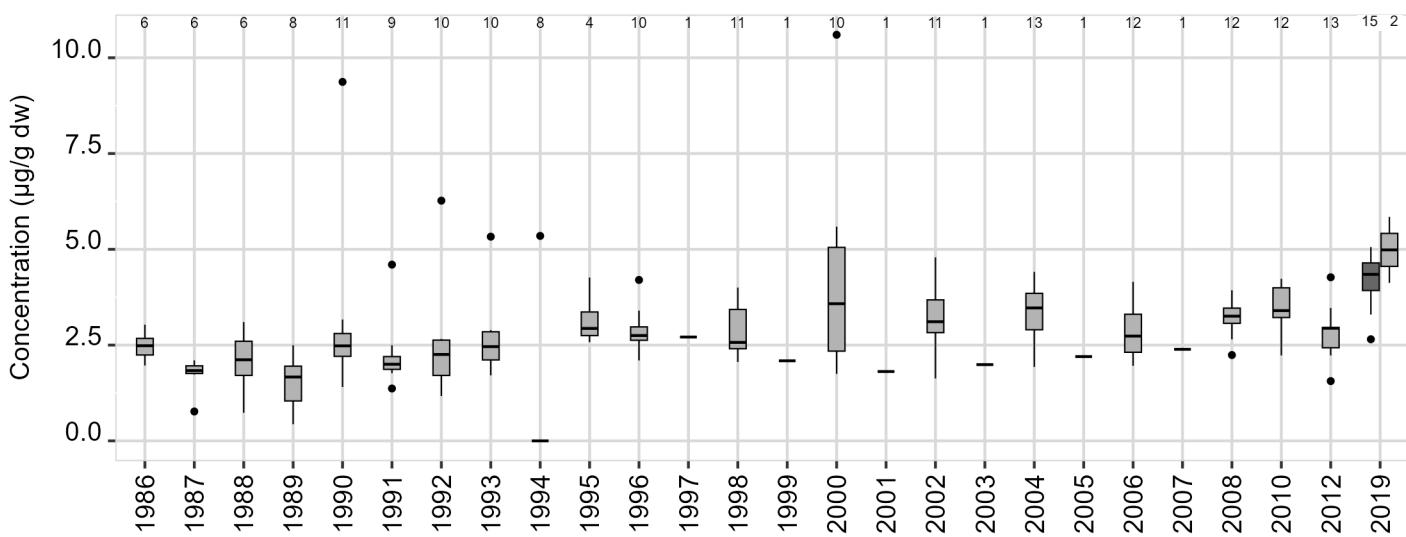


Figure 55. Boxplots representing the historic selenium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

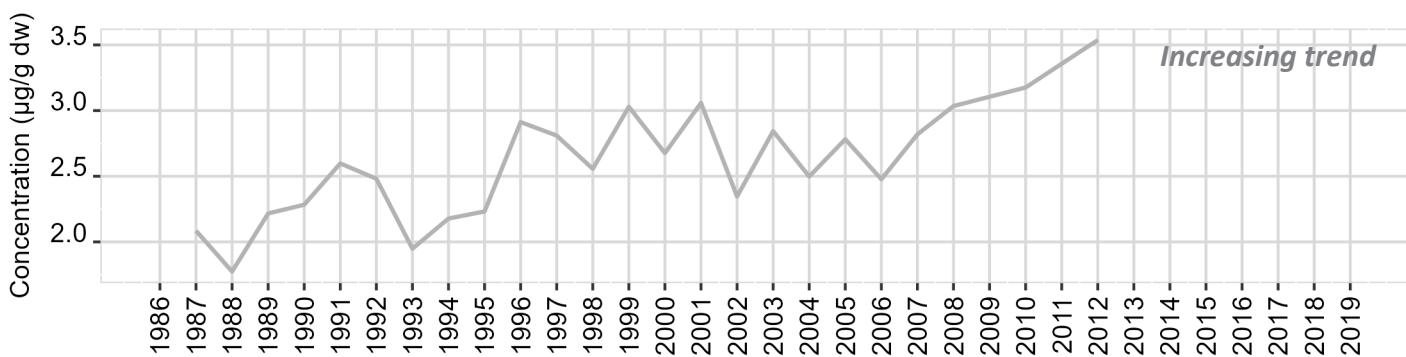


Figure 56. Three-point moving average of the yearly mean selenium concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Selenium (Se)

## 14.4 Selenium Summary

### 2019 Mussel Tissue Results:

- Se was detected at 100% of 17 sites surveyed (Figure 53)
- Se concentration descriptive statistics (Figure 53):
  - Range: 2.65 – 5.85  $\mu\text{g/g dw}$
  - Minimum Se concentration was detected at WBNA
  - Maximum Se concentration was detected at YHFC
  - Median: 4.35  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $4.31 \pm 0.74 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 93% of sites surveyed were above their historic median concentrations in 2019 (Figure 54)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was a significant increasing regional temporal trend of Se concentrations in the Pacific Northwest ( $p = 0.01$ ,  $\rho = 0.48$ ) (Figure 55; Figure 56)
- 1 site (WBNA) in the Pacific Northwest showed a significant decreasing temporal trend of Se concentrations at  $\alpha = 0.05$  (Figure 54; Table A4)
- 9 sites (BBSM, PSPT, PSEH, PSMF, EBFR, EBDH, SIWP, CBTP, and SSBI) in the Pacific Northwest showed significant increasing temporal trends of Se concentrations at  $\alpha = 0.05$  (Figure 54; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 57):
  - 6% of sites in low cluster (0.0 – 3.17  $\mu\text{g/g dw}$ )
  - 53% of sites in medium cluster (3.18 – 4.44  $\mu\text{g/g dw}$ )
  - 41% of sites in high cluster (4.46 – 10.60  $\mu\text{g/g dw}$ )

### General Observations:

- Se concentrations were generally medium to high in mussel tissue both in Puget Sound and along the Washington and Oregon coasts.
- Regionally, Se concentrations have increased significantly over time in mussel tissue, possibly indicating local point sources of Se contamination.

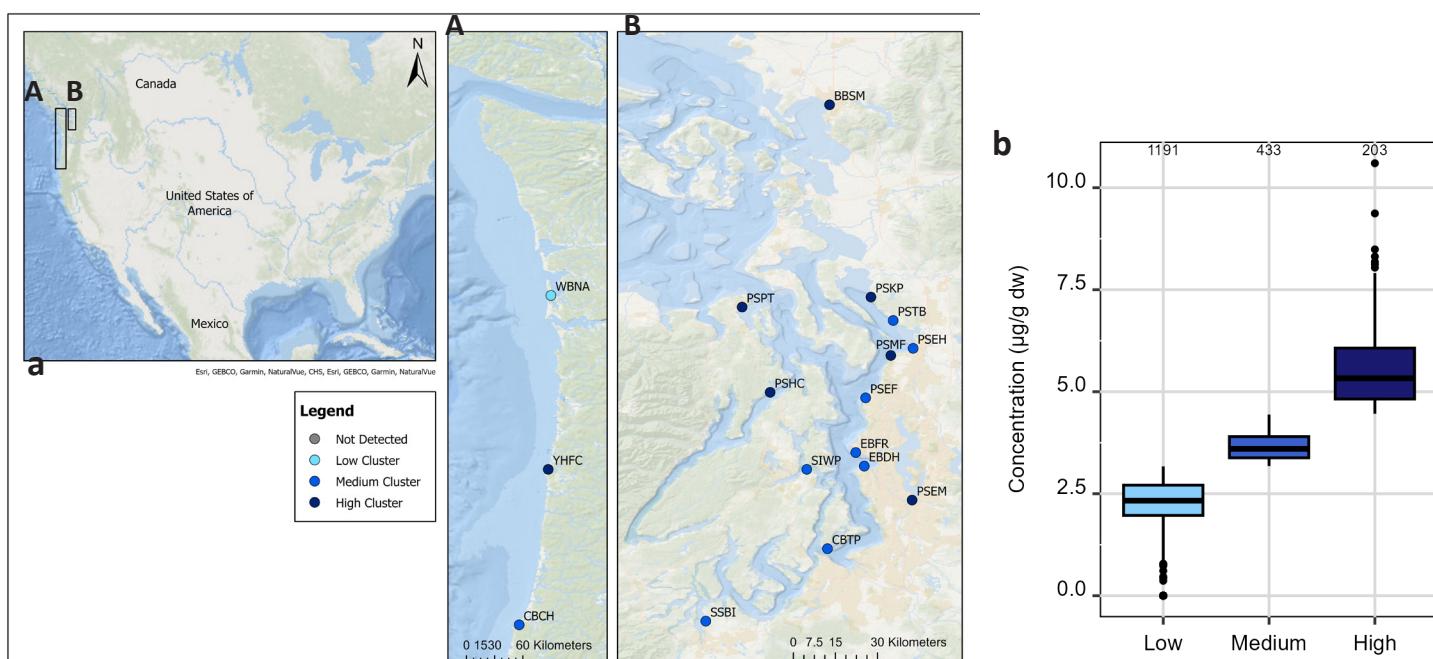


Figure 57. Selenium concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP Mytilus species selenium concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples.

# Results - Silver (Ag)

## 15.0 RESULTS - SILVER (Ag)

### 15.1 Silver Chemical Description

Silver is a naturally occurring element but elevated levels of contamination are typically from anthropogenic sources (ATSDR, 1990). The natural wearing down of silver-bearing rocks and soil by the wind and rain releases large amounts of silver into the environment. Anthropogenically, products that contain silver are photographic technology, dental applications, electronics, silverware, jewelry, and solders. Photographic materials are the major anthropogenic source of silver that is released into the environment (ATSDR, 1990). Silver has been regulated in the US since the 1960s, but initial criteria were conservative, as the realized risk of silver was not known at the time. The US EPA downgraded silver from a primary to a secondary maximum contaminant level in the 1990s due to its low realized risk from exposure (Purcell, 2009).

Silver is not an essential nutrient to humans and is mostly harmless at low levels and infrequent exposure (ATSDR, 1990). Humans are generally exposed to silver through food and drinking water, and minimally through the air. Silver can cause some areas of the skin and other body tissues to turn gray or gray-blue in a condition known as "argyria". This condition can occur in humans who eat or breathe in silver compounds over a long period. Long or high exposure to silver compounds such as silver nitrate or silver oxide may cause breathing problems, lung and throat irritation, and stomach pain. These conditions typically manifest with occupational exposure. Skin contact with silver compounds has been found to cause mild allergic reactions, such as rash, swelling, and inflammation in some people. Tests in animals have shown that exposure to silver is only likely to be life threatening when very high amounts are swallowed, which is unlikely. Conversely, silver does have beneficial medicinal uses including as eye drops to prevent blindness, in salves for burn victims, and to kill bacteria in water (ATSDR, 1990).

Silver that is released into the environment may be carried long distances in air and water (ATSDR, 1990). Rain washes silver compounds out of many soils so that it eventually moves into the groundwater. Silver is stable and remains in the environment in one form or another until it is taken out again by people. Silver does not break down, but it can change its form by combining with other substances and the form it is found in depends on environmental conditions (ATSDR, 1990).

### 15.2 Magnitude and Distribution of Silver in Mussel Tissue in 2019

Silver (Ag) was not detected at any sites in 2019.

# Results - Silver (Ag)

## 15.3 Historical Context of Silver Magnitude and Distribution in Mussel Tissue

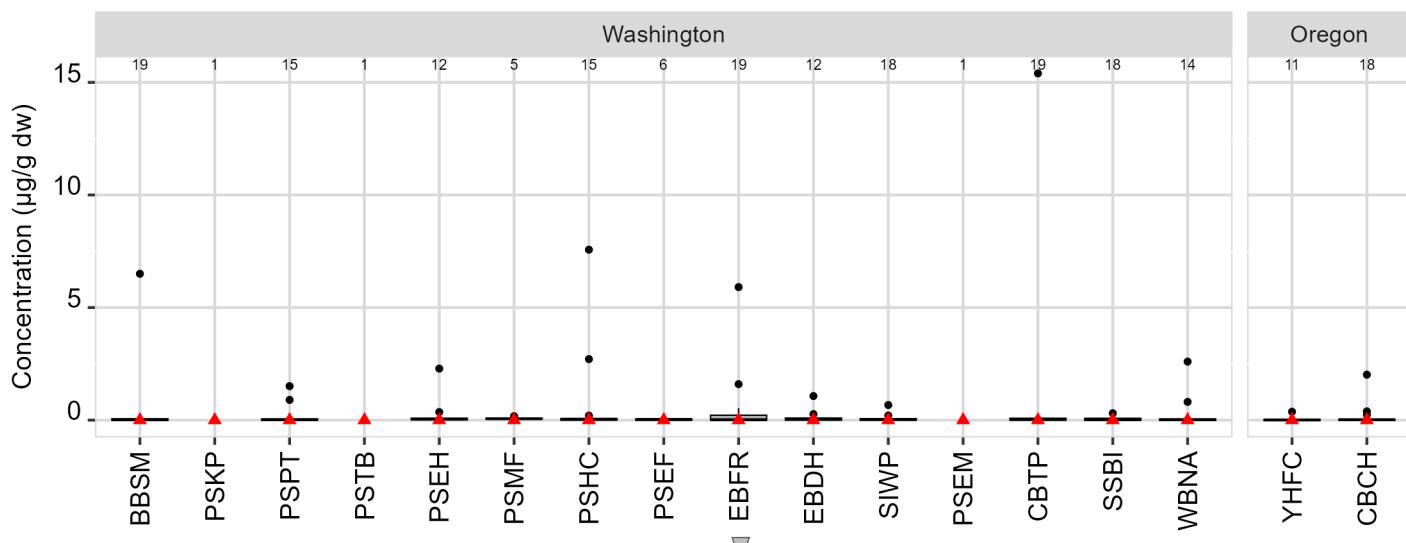


Figure 58. Silver concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic silver concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

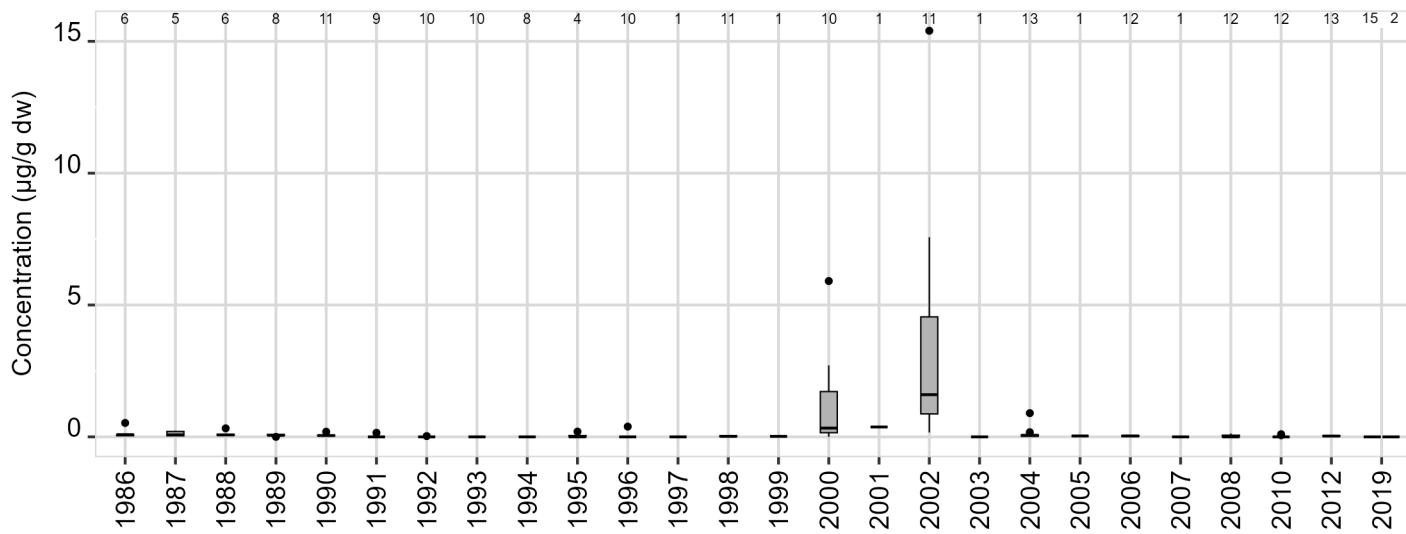


Figure 59. Boxplots representing the historic silver concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

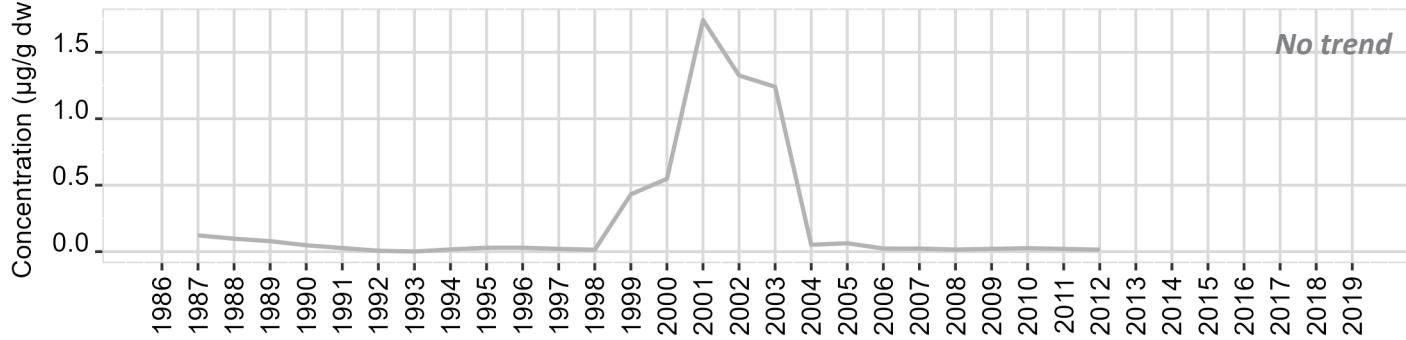


Figure 60. Three-point moving average of the yearly mean silver concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Silver (Ag)

## 15.4 Silver Summary

### 2019 Mussel Tissue Results:

- Ag was detected at 0% of 17 sites surveyed

### Historic Context for Mussel Tissue:

- 0% of sites surveyed were above their historic median concentrations in 2019 (Figure 58)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Ag concentrations in the Pacific Northwest ( $p = 0.20$ ,  $\rho = -0.26$ ) (Figure 50; Figure 51)
- 1 site (EBFR) in the Pacific Northwest showed a significant decreasing temporal trend of Ag concentrations at  $\alpha = 0.05$  (Figure 58; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of Ag concentrations at  $\alpha = 0.05$  (Figure 58; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 61):
  - 100% of sites in low cluster (0.00 – 1.41  $\mu\text{g/g dw}$ )
  - 0% of sites in medium cluster (1.45 – 4.54  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (4.71 – 33.76  $\mu\text{g/g dw}$ )

### General Observations:

- Ag was not detected at any sites in the Pacific Northwest in mussel tissue in 2019.
- Regionally, Ag concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Ag pollution.

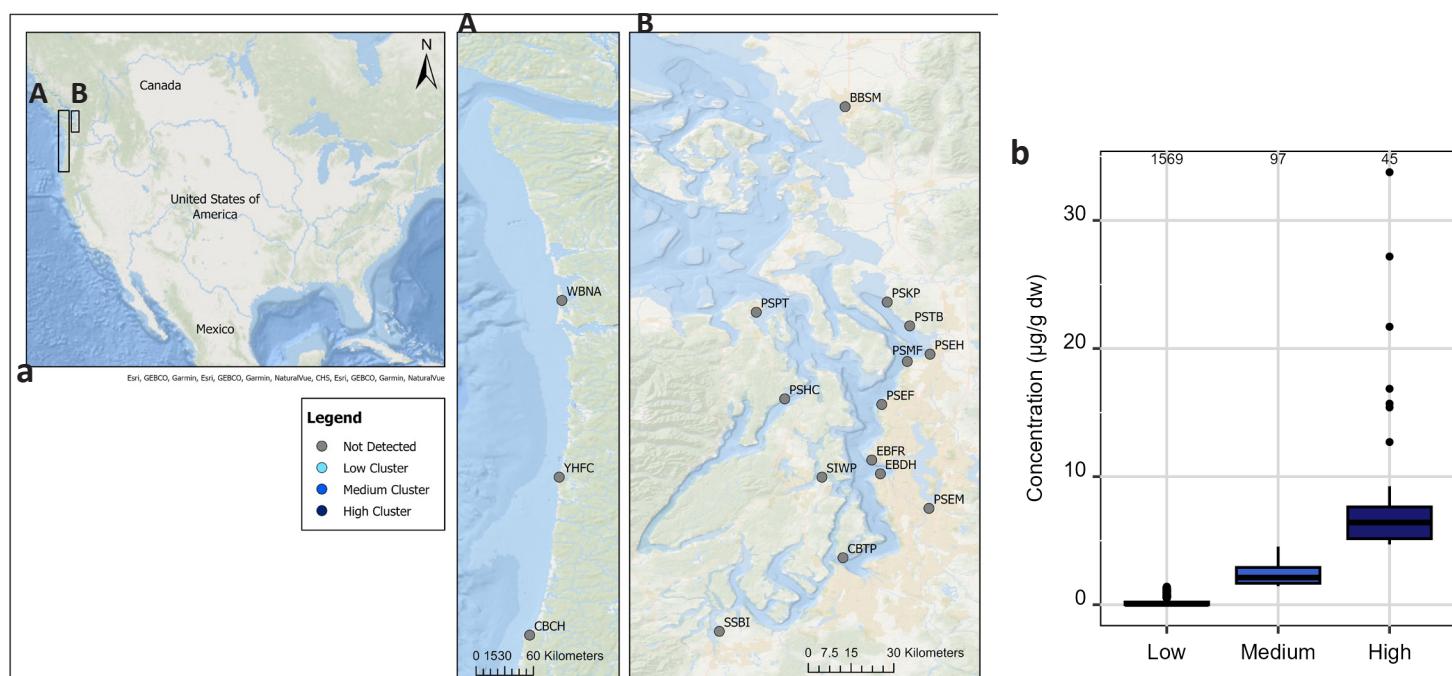


Figure 61. Silver concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP *Mytilus* species silver concentrations ( $\mu\text{g/g dw}$ ). 211 national sites sampled between 1986 - 2021 for a total of 1711 samples.

# Results - Tin (Sn)

## 16.0 RESULTS - TIN (Sn)

### 16.1 Tin Chemical Description

Tin may be released to the environment from both natural and anthropogenic sources. Tin occurs in water in trace amounts naturally and can combine with other chemicals to form various inorganic and organic compounds. Sources of tin in coastal water and soil include manufacturing and processing facilities and mining. Tin has not been mined in the US since 1993 (USGS, 2008); however, Canadian tin mining occurs in the Great Lakes Region. Tin metal is mainly used as liner in cans for food and aerosols and is also used in paint, plastic, pesticide brass, bronze, and some soldering products (ATSDR, 2005b). Concentrations in unpolluted waters and the atmosphere are often near analytical detection limits.

Humans are exposed to elevated levels of tin by eating from tin-lined cans and by consuming contaminated seafood (ATSDR, 2005b). Exposure to elevated levels of tin compounds by humans leads to liver damage, kidney damage, and cancer. There is no US FDA recommended guideline for tin in seafood.

Tin enters coastal waters bound to particulates and from riverine sources derived from soil and sediment erosion. Bioconcentration factors for inorganic tin were reported to be 1,900 and 3,000 for marine algae and fish (Seidel et al., 1980; Thompson et al., 1972). Inorganic tin can be transformed into organometallic forms by microbial methylation and is correlated with increasing organic content in sediment (Hadjispyou et al., 1998). Tin is regarded as being relatively immobile in the environment and is rarely detected in the atmosphere. It is mainly found in the atmosphere near industrial sources as particulates from combustion of fossil fuels and solid waste (Gerritse et al., 1982; WHO, 1980).

### 16.2 Magnitude and Distribution of Tin in Mussel Tissue in 2019

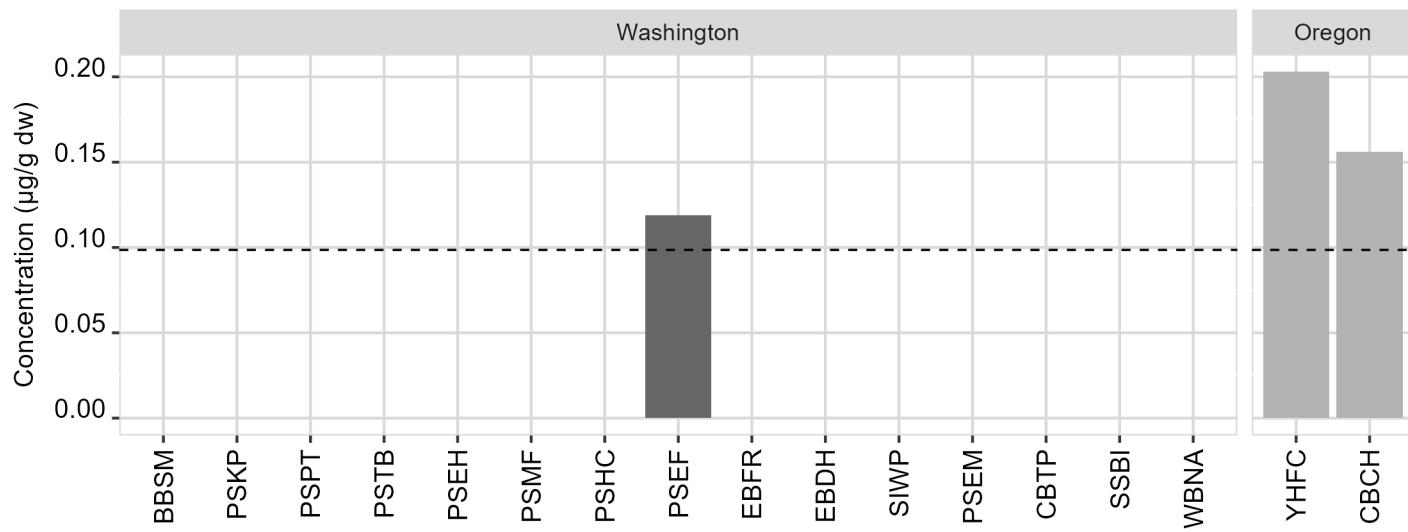


Figure 62. Bar graph showing magnitude of tin ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Tin (Sn)

## 16.3 Historical Context of Tin Magnitude and Distribution in Mussel Tissue

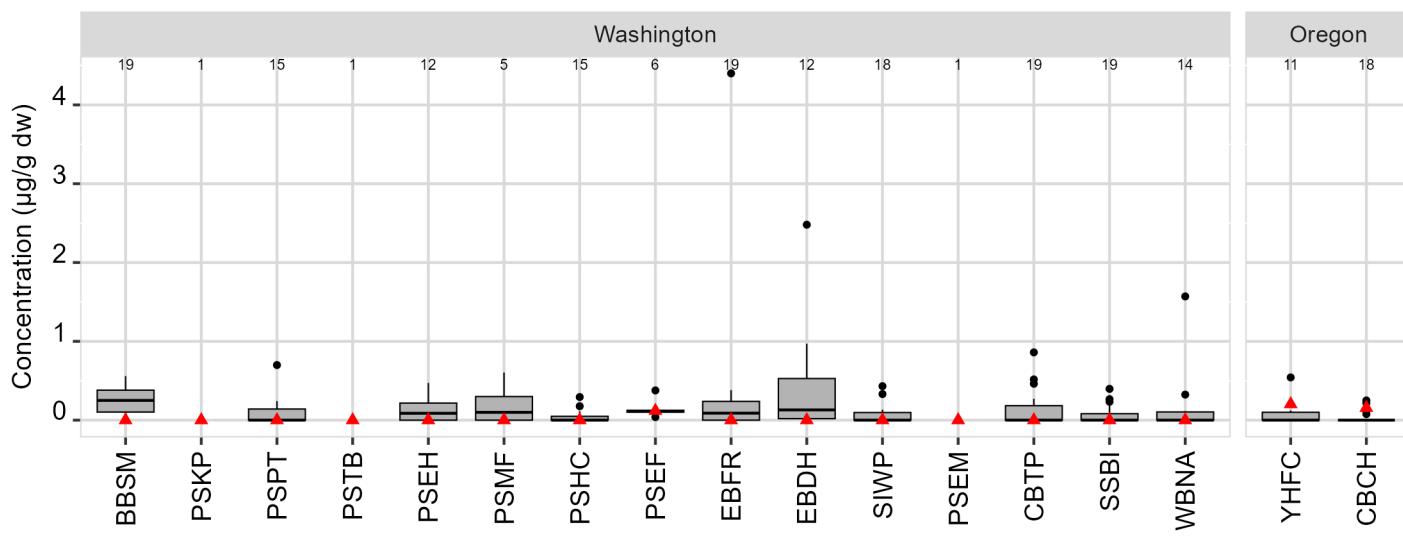


Figure 63. Tin concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic tin concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

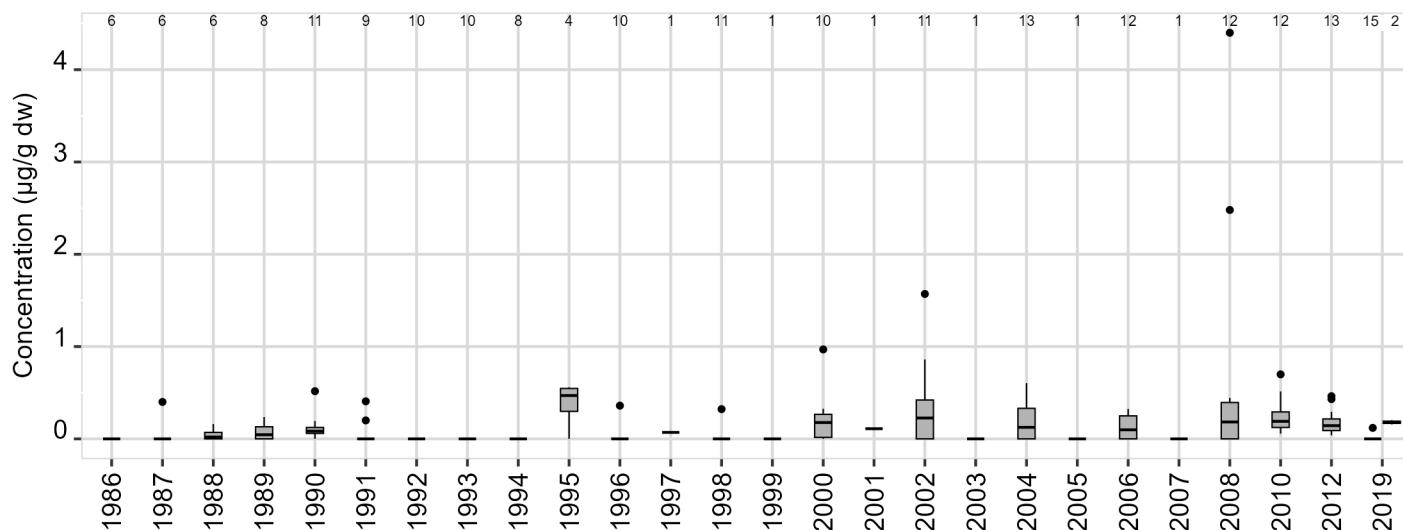


Figure 64. Boxplots representing the historic tin concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

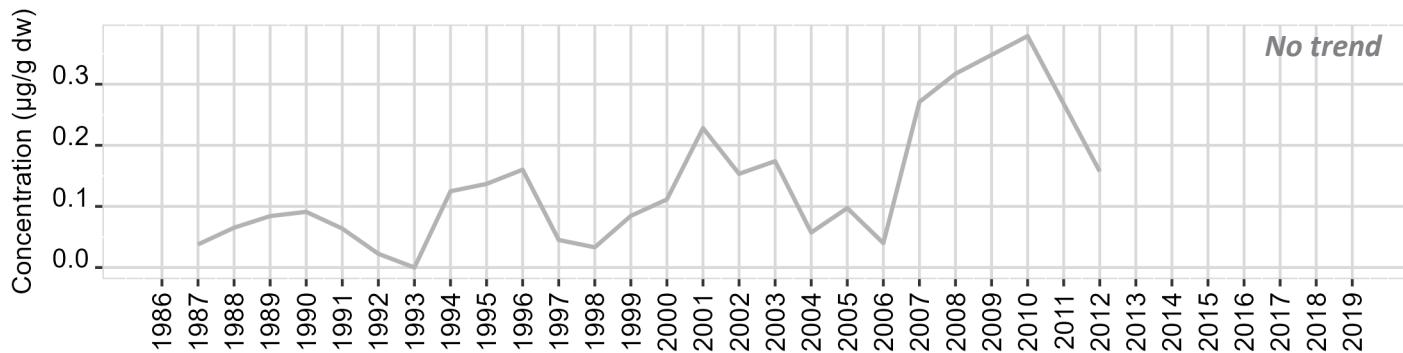


Figure 65. Three-point moving average of the yearly mean tin concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Tin (Sn)

## 16.4 Tin Summary

### 2019 Mussel Tissue Results:

- Sn was detected at 18% of 17 sites surveyed (Figure 62)
- Sn concentration descriptive statistics (Figure 62):
  - Range: 0.00 – 0.20  $\mu\text{g/g dw}$
  - Minimum detectable Sn concentration was detected at PSEF (0.12  $\mu\text{g/g dw}$ )
  - Maximum Sn concentration was detected at YHFC
  - Median: 0.00  $\mu\text{g/g dw}$
  - Mean  $\pm$  SD:  $0.03 \pm 0.06 \mu\text{g/g dw}$

### Historic Context for Mussel Tissue:

- 21% of sites surveyed were above their historic median concentrations in 2019 (Figure 63)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Sn concentrations in the Pacific Northwest ( $p = 0.21$ ,  $\rho = 0.26$ ) (Figure 64; Figure 65)
- 0 sites in the Pacific Northwest showed significant decreasing temporal trends of Sn concentrations at  $\alpha = 0.05$  (Figure 63; Table A4)
- 2 sites (PSHC, EBFR) in the Pacific Northwest showed significant increasing temporal trends of Sn concentrations at  $\alpha = 0.05$  (Figure 63; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 66):
  - 100% of sites in low cluster (0.00 – 0.49  $\mu\text{g/g dw}$ )
  - 0% of sites in medium cluster (0.50 – 1.91  $\mu\text{g/g dw}$ )
  - 0% of sites in high cluster (2.10 – 5.13  $\mu\text{g/g dw}$ )

### General Observations:

- Sn was generally not detected in the Pacific Northwest in mussel tissue in 2019.
- Low Sn concentrations were detected in mussel tissue off the Oregon coast and at Puget Sound site PSEF, notable as the only sites where Sn was detected.
- Regionally, Sn concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Sn pollution.

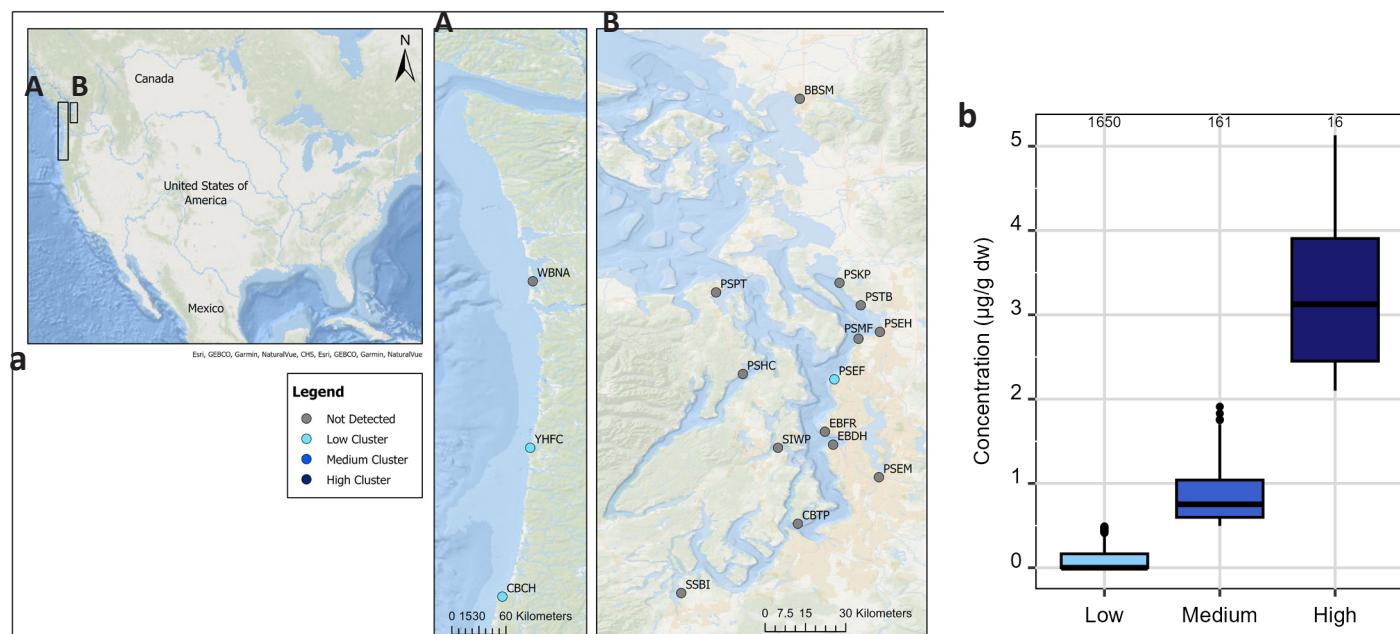


Figure 66. Tin concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP Mytilus species tin concentrations ( $\mu\text{g/g dw}$ ). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples.

# Results - Zinc (Zn)

## 17.0 RESULTS - ZINC (Zn)

### 17.1 Zinc Chemical Description

Zinc occurs naturally in the Earth's crust and can be released into the environment by natural rock weathering, but its anthropogenic sources far exceed its natural ones. Major industrial sources include electroplating, smelting, and drainage from mining operations (Mirenda, 1986). The greatest use of zinc is as an anti-corrosive coating for iron and steel products (sheet and strip steel, tube and pipe, wire, and wire rope). Canada is one of the largest producers and exporters of zinc. The US is the largest customer for Canadian refined zinc and the automobile industry is the largest user of galvanized steel.

Zinc is an essential nutrient and particles can be found in air, water, soil, and most foods. Human exposure to high doses of zinc may cause anemia or damage to the pancreas and kidneys (ATSDR, 2005c). However, zinc does not bioaccumulate in humans; therefore, toxic effects are uncommon and associated with excessively high doses. Fish exposed to low zinc concentrations can sequester it in some cases (McGeer et al., 2003). There is no FDA recommended safety level for zinc in fish and fish products.

Dissolved zinc occurs as the free hydrated ion and as dissolved complexes. Changes in water conditions (pH, redox potential, chemical speciation) can result in dissolution from or sorption to particles (EPA, 1979). In air, zinc is primarily found in the oxidized form bound to particles. Zinc precipitates as zinc sulfide in anaerobic or reducing environments (such as wetlands) and thus is less mobile, but typically remains as a free ion at lower pHs. Zinc is found in all environmental media (air, water, soil, and biota) as a result of natural and anthropogenic activities.

### 17.2 Magnitude and Distribution of Zinc in Mussel Tissue in 2019

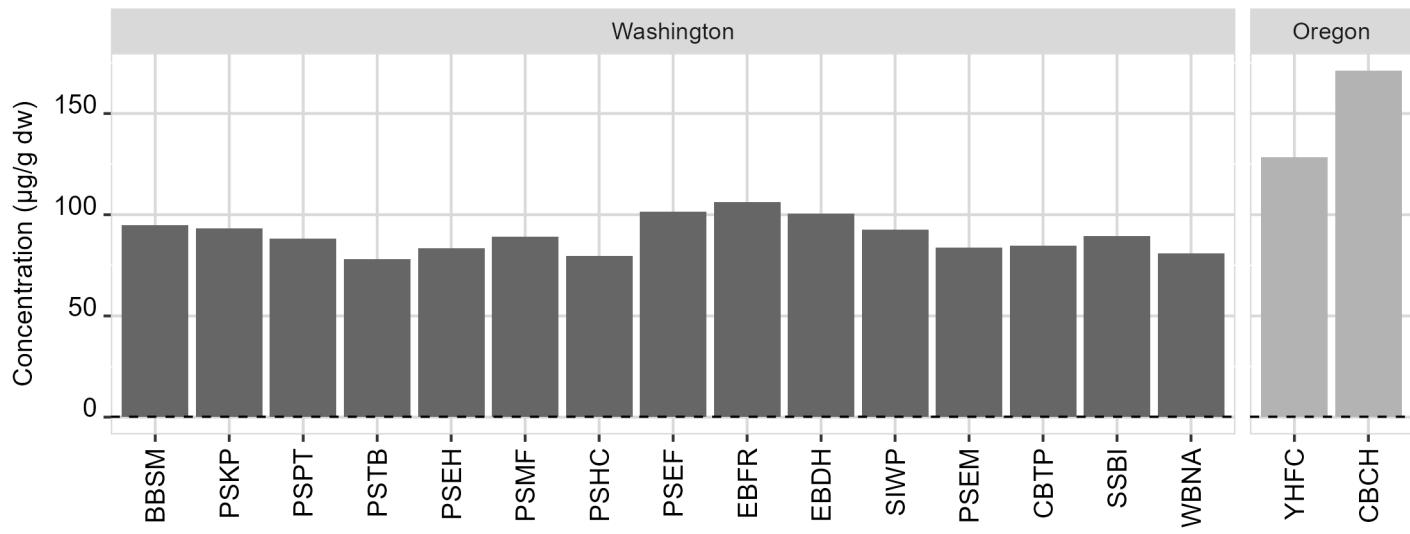


Figure 67. Bar graph showing magnitude of zinc ( $\mu\text{g/g dw}$ ) detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

# Results - Zinc (Zn)

## 17.3 Historical Context of Zinc Magnitude and Distribution in Mussel Tissue

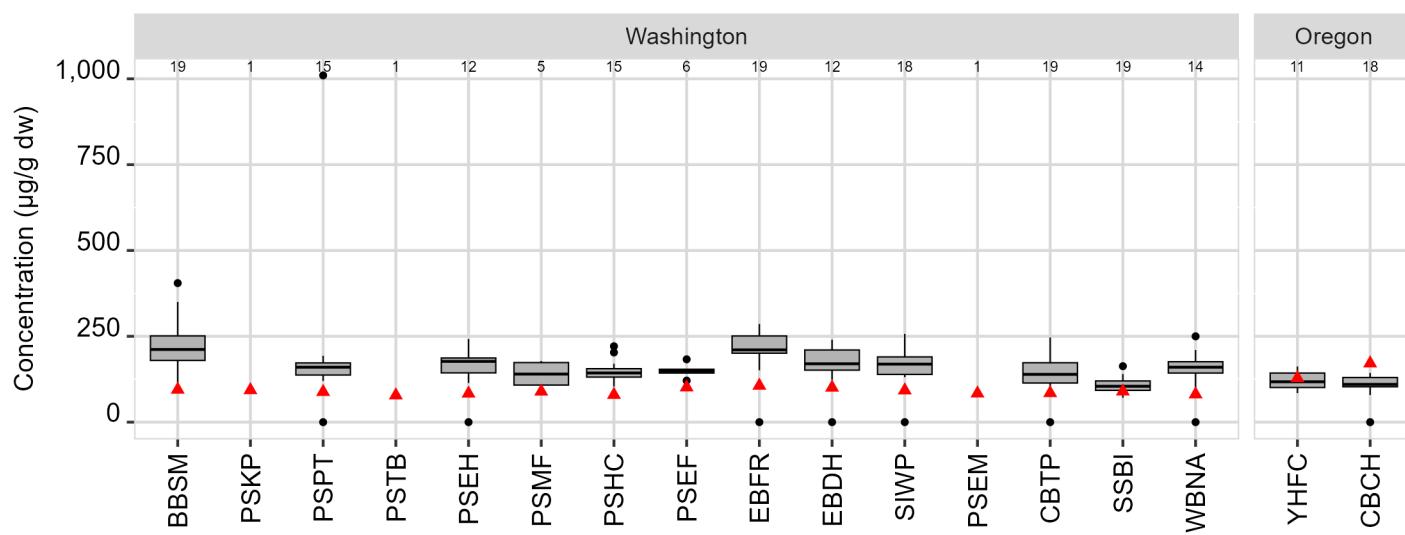


Figure 68. Zinc concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in 2019 (red triangles) compared to boxplots of the historic zinc concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing site-specific contamination trends over time (gray triangles). Sites are listed geographically from north to south, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

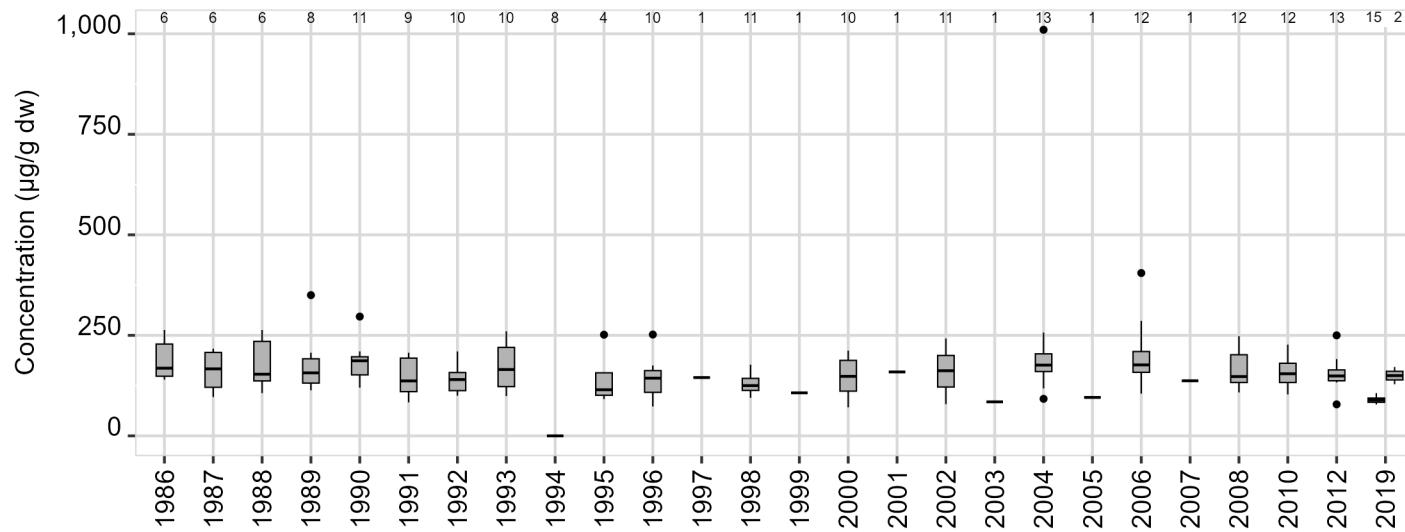


Figure 69. Boxplots representing the historic zinc concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. In 2019, dark gray indicates transplanted mussels were used, light gray indicates that wild mussels were used.

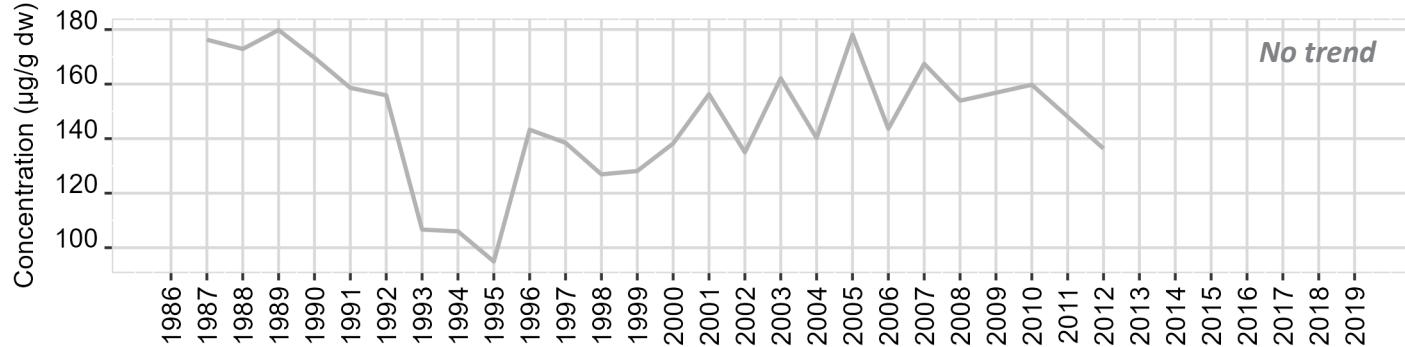


Figure 70. Three-point moving average of the yearly mean zinc concentrations ( $\mu\text{g/g dw}$ ) in mussel tissue in the sites analyzed in this study.

# Results - Zinc (Zn)

## 17.4 Zinc Summary

### 2019 Mussel Tissue Results:

- Zn was detected at 100% of 17 sites surveyed (Figure 67)
- Zn concentration descriptive statistics (Figure 67):
  - Range: 78.14 – 171.31  $\mu\text{g/g}$  dw
  - Minimum Zn concentration was detected at PSTB
  - Maximum Zn concentration was detected at CBCH
  - Median: 89.62  $\mu\text{g/g}$  dw
  - Mean  $\pm$  SD:  $96.84 \pm 22.75 \mu\text{g/g}$  dw

### Historic Context for Mussel Tissue:

- 14% of sites surveyed were above their historic median concentrations in 2019 (Figure 68)
  - 2 sites were not assessed in 2019 (CRSJ, CBRP) and 3 sites (PSEM, PSKP, PSTB) were only assessed in 2019, so were not included in this analysis
- There was no significant regional temporal trend of Zn concentrations in the Pacific Northwest ( $p = 0.19$ ,  $\rho = -0.26$ ) (Figure 69; Figure 70)
- 0 sites in the Pacific Northwest showed significant decreasing temporal trends of Zn concentrations at  $\alpha = 0.05$  (Figure 68; Table A4)
- 0 sites in the Pacific Northwest showed significant increasing temporal trends of Zn concentrations at  $\alpha = 0.05$  (Figure 68; Table A4)
- The percentage of sites in the Pacific Northwest in 2019 grouped into each cluster based on historic data (Figure 71):
  - 88% of sites in low cluster (0.00 – 107.33  $\mu\text{g/g}$  dw)
  - 12% of sites in medium cluster (108.00 – 180.00  $\mu\text{g/g}$  dw)
  - 0% of sites in high cluster (181.73 – 11,500.00  $\mu\text{g/g}$  dw)

### General Observations:

- Zn concentrations were generally low in mussel tissue, likely reflecting natural ambient concentrations in the Pacific Northwest.
- Medium Zn concentrations were detected in mussel tissue off the Oregon coast, possibly indicating offshore natural sources or localized point sources of contamination.
- Regionally, Zn concentrations have not changed significantly over time in mussel tissue, indicating no obvious anthropogenic Zn pollution.

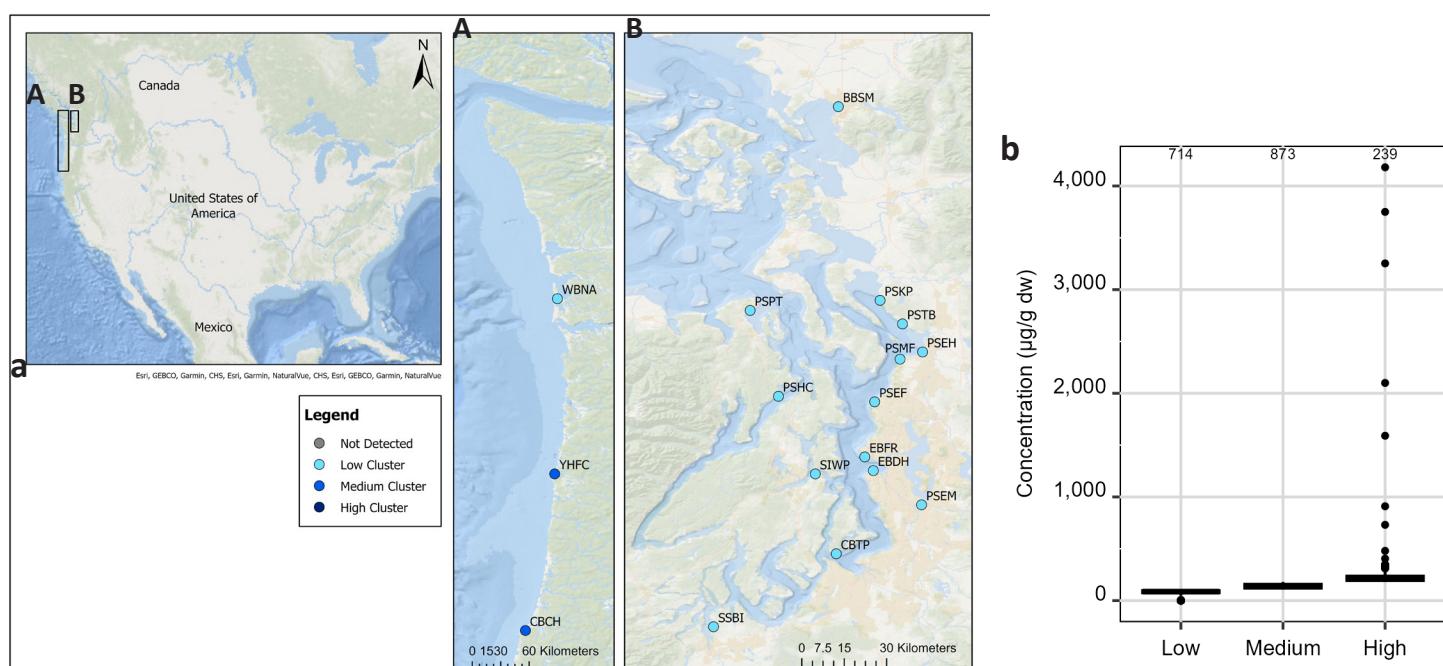


Figure 71. Zinc concentrations in mussel tissue in 2019 (a) spatially and (b) numerically compared to the historic national MWP. Mytilus species zinc concentrations ( $\mu\text{g/g}$  dw). 212 national sites sampled between 1986 - 2021 for a total of 1827 samples. Site DBHC in 2005 (11,500.00  $\mu\text{g/g}$  dw) was removed from 'b' to aid in visualization.

# Summary

## 18.0 SUMMARY

Mussels are good indicators of water quality; hence, they have been used worldwide as sentinel species for chemical pollution in aquatic systems. In this study, mussel tissue samples (*Mytilus* species) were assessed for a suite of trace metals. The mussel samples were collected at historic MWP monitoring sites located within the Pacific Northwest. Sample collection was conducted by WDFW and ODFW following standard protocols (Apeti et al., 2012; Lanksbury and Lubliner, 2015). Trace metals were analyzed in mussel tissue for 17 sites. Separate result summaries for each trace metal can be found in the Summary subsection of each trace metal section within this document. This summary attempts to integrate all contamination results to assess overall contamination of sites in the Pacific Northwest, both with respect to the other sites analyzed within this study and to the larger historical Mussel Watch dataset. For both analyses, a multivariate cluster analysis was first conducted for each contaminant group at each site and represented in a heatmap, and then a second multivariate cluster analysis was conducted using the total cluster value for each site to group sites into groups with statistically different degrees of overall contamination. This two-pronged analysis was conducted (1) as respective of sites within this study and (2) as respective to all historic MWP contamination analyzed in mussels.

The first observation of note is that all trace metals are highly ubiquitous and were detected within the Pacific Northwest in 2019 in mussel tissue at detection frequencies ranging from 18% (Sn) to 100% (all other metals), with the exception of Ag which was not detected at any sites in this survey (Table 2; Table 3). Trace metals are naturally occurring crustal elements, so detecting them at a high proportion of sites is expected. There were no sites where no trace metals were detected.

For analyses conducted respective only to sites assessed for mussel tissue in this 2019 study, 88% of sites were clustered into the lower contamination group and 12% were clustered into the higher contamination group (Figure 73). Sites in 2019 only generated two statistically distinct contamination levels, both within each contaminant group (Figure 72) and when combined and summarized (Figure 73). The two sites in the higher contamination group occurred along the Oregon coastline, possibly due to natural coastal sources or localized point sources (Figure 73).

Table 2. Frequency of detection (% of total) of trace metals in mussel tissue in 2019 at sites within the Pacific Northwest.

Contaminant Group	Sites with Detects	Sites Analyzed	Detection Frequency
Aluminum (Al)	17	17	100%
Arsenic (As)	17	17	100%
Cadmium (Cd)	17	17	100%
Chromium (Cr)	17	17	100%
Copper (Cu)	17	17	100%
Iron (Fe)	17	17	100%
Lead (Pb)	17	17	100%
Manganese (Mn)	17	17	100%
Mercury (Hg)	17	17	100%
Nickel (Ni)	17	17	100%
Selenium (Se)	17	17	100%
Silver (Ag)	0	17	0%
Tin (Sn)	3	17	18%
Zinc (Zn)	17	17	100%

# Summary

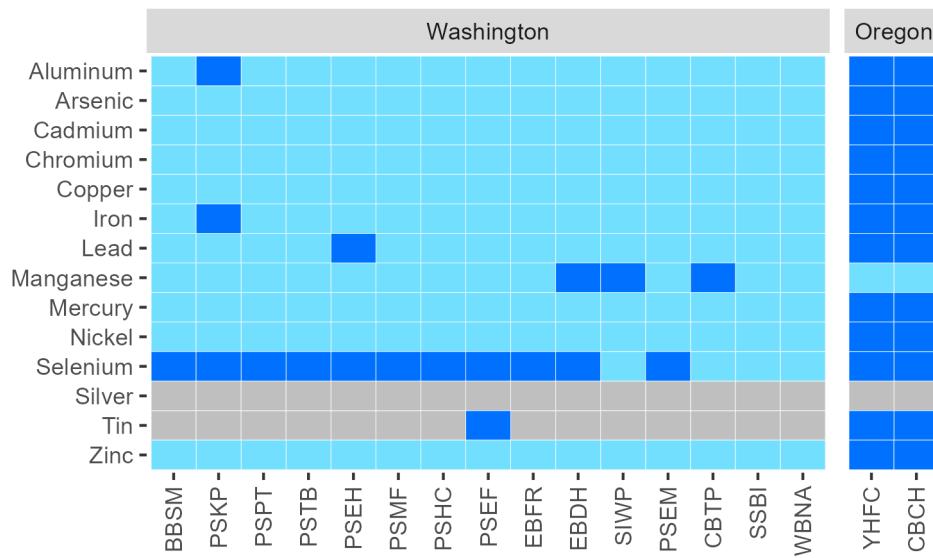


Figure 72. Distribution map showing the distribution and magnitude of 2019 concentrations detected in the Pacific Northwest in mussel tissue, respective to one another in this study. Colors represent contamination magnitude as follows: lower group ( ), higher group ( ), and not detected ( ).

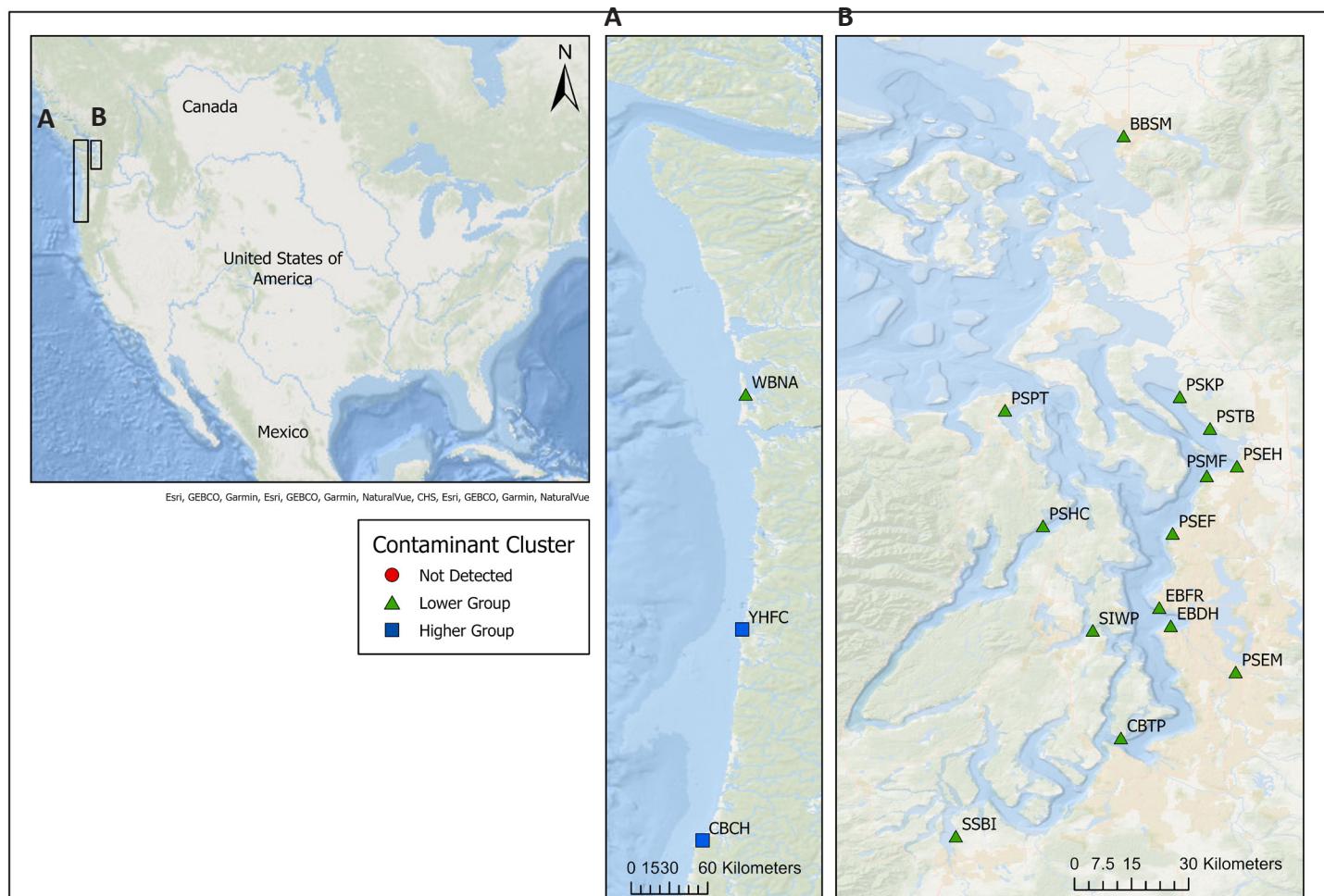


Figure 73. Map of sites in the Pacific Northwest highlighting locations with lower and higher degrees of contamination in mussel tissue in 2019, respective to one another in this study (Table A2).

# Summary

While it is useful to know which sites in the Pacific Northwest are most contaminated relative to other sites in the region, it is equally important to put contamination of a region into perspective with other available data nationwide and historically. When compared to all trace metal data collected by the MWP for mussels since 1986, the resulting clusters of the 2019 sites were largely categorized as having low contamination in mussel tissue. In mussel tissue, 82% of sites were clustered into the low contamination category, 6% in the medium contamination category, 12% in the high contamination category, and 0% in the very high contamination category (Figure 75).

Most trace metals have shown no discernable trend in concentration over time, with only selenium (Se) showing a significantly increasing regional temporal trend (Table 3). Decreasing trends in trace metal contamination over time are largely seen in the same sites throughout the Pacific Northwest, namely PSMF and PSEF (Table 3), implying that localized point sources of contamination have possibly been addressed at these sites. This analysis of trends seen in trace metals demonstrates the importance of long-standing monitoring programs and provide a point of reference for current sampling. These trends allow us to observe the effects of regulations on the concentrations of these contaminants in the environment and to put into perspective any recently detected concentrations. Specifically, the trace metal selenium was detected at high levels throughout the Pacific Northwest. The elevated levels of some of these elements can be largely attributed to their prevalence in the bedrock and crust of the region (Edwards et al., 2014). Additional causes of elevated contamination may be attributed to high urban land use in the area (i.e., Puget Sound) from specific sources such as tire wear, agriculture, and wood treatment (Edwards et al., 2014). Further analysis and sampling may be beneficial to determine the current point sources of elevated selenium contamination seen in this region.

Some additional context can be determined by comparing the data from this survey to monitoring data obtained by other programs such as the Toxics-focused Biological Observation System (TBiOS) program (Table A5; Table A6; Table A7). While this additional data was collected at the same time as the sites analyzed in this report, the analyses were conducted by a different laboratory and for separate projects, so the data was not integrated into this report. However, the study design and data are theoretically comparable, so raw concentration sum data was reported in the appendices of this report for WDFW Stormwater Action Monitoring (SAM) survey sites (Table A5), WDFW partner sites (Table A6), and MW sites that were analyzed by the same laboratory (Table A7) to improve the spatial distribution of available data. Broadly, the SAM sites were selected using a probabilistic random stratified sampling design, whereas MW sites were originally selected due to the presence of bivalves, despite these sites now being assessed using a caged study design (Langness et al., 2022). Across all chemical groups, the average concentrations of MW sites were generally comparable to concentrations of SAM sites and partner sites (Table A5-A7).

The different classifications of site contamination when respective to 2019 only versus respective to nationwide NS&T data support the need for consistent monitoring programs that can contextualize any given result. A successful monitoring program must both achieve an understanding of background contamination levels and capture the variability and range of possible contamination. Existing contaminant data has provided an understanding of general background contamination including range, trends, and variability. Further monitoring should aim to continue analyzing the temporal trends of these contaminants on a regional scale (i.e., through periodic sampling and biennial sampling at select sites) but focus on areas of interest that could give further insight on range, variation, and potential localized point sources of contamination. For example, further assessments into the potential sources of elevated levels of selenium could help to clarify the high overall contamination seen there, relative to other regions. Through an understanding of both the temporal and spatial variations, a monitoring program can serve its purpose of assessing potential contaminant exposure. This study provides needed data and information for the MWP and provides contamination data required by coastal resource managers as they develop long-term policies to protect the services provided by the coastal environment within this region.

# Summary

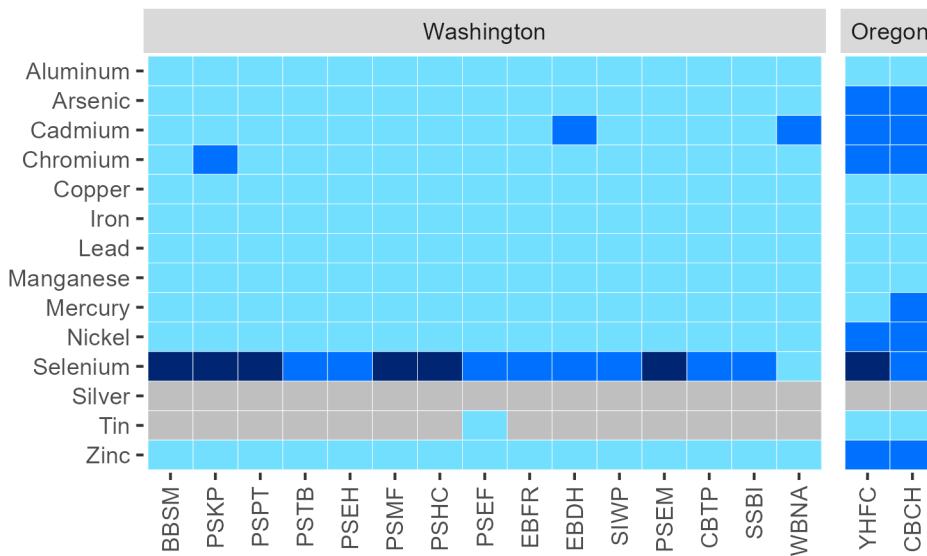


Figure 74. Distribution map showing the distribution and magnitude of 2019 concentrations detected in the Pacific Northwest in mussel tissue, respective to all historic MWP contamination analyzed in blue mussels. Colors represent contamination magnitude as follows: low (light blue), medium (medium blue), high (dark blue), and not detected (gray).

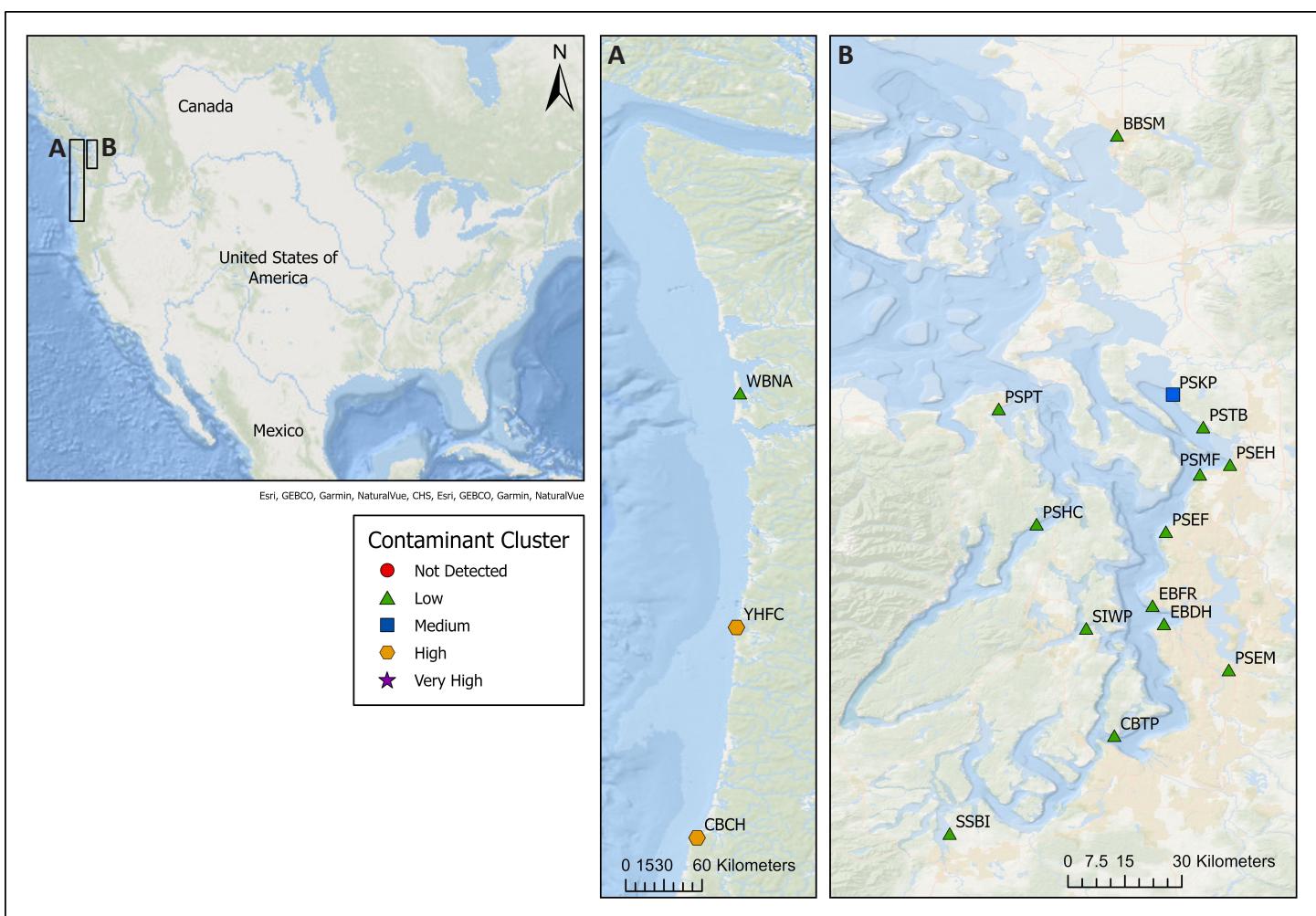


Figure 75. Map of sites in the Pacific Northwest highlighting locations with Low, Medium, High, and Very High degrees of contamination in mussel tissue in 2019, respective to all historic MWP contamination analyzed in blue mussels (Table A3).

# Summary

Table 3. Summary of site-based trend analysis for trace metals in mussel tissue; D = decreasing trend, I = increasing trend. Sites PSKP, PSTB, and PSEM were only assessed in 2019 so could not be analyzed for temporal trends (denoted with --).

Site	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Se	Ag	Sn	Zn
BBSM							D		D		I			
CBCH			I											
CRTP											I			
EBDH											I			
EBFR							D		D		I	D	I	
PSEF	D					D	D	D		D				
PSEH											I			
PSEM	--	--	--	--	--	--	--	--	--	--	--	--	--	--
PSHC													I	
PSKP	--	--	--	--	--	--	--	--	--	--	--	--	--	--
PSMF	D				D	D	D	D		D	I			
PSPT					I						I			
PSTB	--	--	--	--	--	--	--	--	--	--	--	--	--	--
SIWP											I			
SSBI											I			
WBNA											D			
YHFC									I					
<b>Regional</b>											I			

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# Appendices

## APPENDICES

Table A1. Percent dry values for tissue samples at each site collected in the Pacific Northwest in 2019.

Site	% Dry of Tissue Samples*
BBSM	13.48
CBCH	14.53
CBTP	16.03
EBDH	16.77
EBFR	12.99
PSEF	15.75
PSEH	14.61
PSEM	16.04
PSHC	14.88
PSKP	14.75
PSMF	16.15
PSPT	15.48
PSTB	13.66
SIWP	18.50
SSBI	15.44
WBNA	14.46
YHFC	14.12

\* conc. (ng/g ww) = conc. (ng/g dw) x (% dry / 100)

# Appendices

Table A2. Breakdown of cluster analysis for mussel tissue in the Pacific Northwest respective to one another in this 2019 study. The first section of the table is the cluster value assigned for each compound. The second section of the table is the calculation conducted to normalize the compound cluster sums by number of compounds assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site. 0 = not detected, 1 = lower group, 2 = higher group.

Site	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Se	Ag	Sn	Zn	Cluster Sum	#Classes Analyzed	Normalized Cluster	Overall Cluster	
BBSM	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	46.43	1	
CBCH	2	2	2	2	2	2	2	1	2	2	2	0	2	2	25	14	89.29	2	
CBTP	1	1	1	1	1	1	1	2	1	1	1	0	0	1	13	14	46.43	1	
EBDH	1	1	1	1	1	1	1	2	1	1	2	0	0	1	14	14	50.00	1	
EBFR	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	46.43	1	
PSEF	1	1	1	1	1	1	1	1	1	1	2	0	2	1	15	14	53.57	1	
PSEH	1	1	1	1	1	1	1	2	1	1	2	0	0	1	14	14	50.00	1	
PSEM	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	46.43	1	
PSHC	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	46.43	1	
PSKP	2	1	1	1	1	1	2	1	1	1	1	2	0	0	1	15	14	53.57	1
PSMF	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	46.43	1	
PSPT	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	46.43	1	
PSTB	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	46.43	1	
SIWP	1	1	1	1	1	1	1	2	1	1	1	0	0	1	13	14	46.43	1	
SSBI	1	1	1	1	1	1	1	1	1	1	1	0	0	1	12	14	42.86	1	
WBNA	1	1	1	1	1	1	1	1	1	1	1	0	0	1	12	14	42.86	1	
YHFC	2	2	2	2	2	2	2	1	2	2	2	0	2	2	25	14	89.29	2	

Table A3. Breakdown of cluster analysis for mussel tissue in the Pacific Northwest respective to all historic MWP contamination analyzed in blue mussels. The first section of the table is the cluster value assigned for each compound. The second section of the table is the calculation conducted to normalize the compound cluster sums by number of compounds assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site. 0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high.

Site	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Se	Ag	Sn	Zn	Cluster Sum	#Classes Analyzed	Normalized Cluster	Overall Cluster
BBSM	1	1	1	1	1	1	1	1	1	1	3	0	0	1	14	14	33.33	1
CBCH	1	2	2	2	1	1	1	1	2	2	2	0	1	2	20	14	47.62	3
CBTP	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	30.95	1
EBDH	1	1	2	1	1	1	1	1	1	1	2	0	0	1	14	14	33.33	1
EBFR	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	30.95	1
PSEF	1	1	1	1	1	1	1	1	1	1	2	0	1	1	14	14	33.33	1
PSEH	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	30.95	1
PSEM	1	1	1	1	1	1	1	1	1	1	3	0	0	1	14	14	33.33	1
PSHC	1	1	1	1	1	1	1	1	1	1	3	0	0	1	14	14	33.33	1
PSKP	1	1	1	2	1	1	1	1	1	1	3	0	0	1	15	14	35.71	2
PSMF	1	1	1	1	1	1	1	1	1	1	3	0	0	1	14	14	33.33	1
PSPT	1	1	1	1	1	1	1	1	1	1	3	0	0	1	14	14	33.33	1
PSTB	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	30.95	1
SIWP	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	30.95	1
SSBI	1	1	1	1	1	1	1	1	1	1	2	0	0	1	13	14	30.95	1
WBNA	1	1	2	1	1	1	1	1	1	1	1	0	0	1	13	14	30.95	1
YHFC	1	2	2	2	1	1	1	1	1	2	3	0	1	2	20	14	47.62	3

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*Table A4. Results of statistically significant Spearman's Rank tests on site specific temporal trends in mussel tissue for sites sampled in the Pacific Northwest in 2019.*

Chemical Group	Site	p value	rho value	Increasing / Decreasing
Al	PSMF	0.04	-0.90	D
Al	PSEF	0.04	-0.83	D
Cd	CBCH	0.004	0.65	I
Cu	PSPT	0.04	0.54	I
Cu	PSMF	0.04	-0.90	D
Fe	PSMF	0.04	-0.90	D
Fe	PSEF	0.04	-0.83	D
Pb	BBSM	0.05	-0.46	D
Pb	PSMF	0.04	-0.90	D
Pb	PSEF	< 0.001	-1.00	D
Pb	EBFR	0.01	-0.57	D
Mn	PSMF	0.04	-0.90	D
Mn	PSEF	0.005	-0.94	D
Hg	BBSM	< 0.001	-0.74	D
Hg	EBFR	< 0.001	-0.77	D
Hg	YHFC	0.01	0.72	I
Ni	PSMF	0.04	-0.90	D
Ni	PSEF	0.04	-0.83	D
Se	BBSM	0.03	0.51	I
Se	PSPT	0.02	0.60	I
Se	PSEH	0.01	0.71	I
Se	PSMF	0.04	0.90	I
Se	EBFR	0.001	0.70	I
Se	EBDH	0.005	0.76	I
Se	SIWP	0.001	0.72	I
Se	CBTP	0.003	0.64	I
Se	SSBI	0.02	0.52	I
Se	WBNA	0.004	-0.71	D
Ag	EBFR	0.05	-0.46	D
Sn	PSHC	0.05	0.52	I
Sn	EBFR	0.04	0.48	I

# Appendices

Table A5. Dry weight concentrations ( $\mu\text{g/g}$ ) of trace metals detected in mussels in 2019 at SAM sites analyzed by the NOAA NWFSC Laboratory. Values were not blank corrected.

Site Type	Site	As	Cd	Cu	Pb	Hg	Zn
Baseline	PCB_MEAN	6.59	2.45	4.49	0.142	0.0177	88.1
Reference	WB_PCR	7.22	2.10	5.43	0.241	0.0302	92.0
Reference	HC_HO	6.86	2.25	5.17	0.113	0.0304	93.5
Monitoring	Site #2	7.42	2.62	4.97	0.470	0.0334	96.7
Monitoring	Site #3	7.78	2.49	4.90	0.203	0.0364	95.1
Monitoring	Site #4	6.73	2.19	4.01	0.198	0.0271	85.9
Monitoring	Site #5	5.57	2.44	3.37	0.189	0.0248	77.1
Monitoring	Site #6	7.68	2.70	6.63	0.930	0.0504	106.0
Monitoring	Site #8	6.21	2.21	4.04	0.225	0.0304	94.3
Monitoring	Site #10	7.04	2.19	5.07	0.423	0.0315	101.0
Monitoring	Site #11	7.13	2.23	4.72	4.520	0.0320	102.0
Monitoring	Site #13	6.37	2.30	4.46	0.349	0.0283	86.9
Monitoring	Site #14	6.62	2.19	4.26	0.421	0.0319	97.4
Monitoring	Site #15	6.79	2.08	4.40	0.239	0.0254	91.5
Monitoring	Site #16	6.71	2.19	4.44	0.237	0.0311	98.7
Monitoring	Site #17	6.95	2.17	5.05	0.251	0.0307	85.0
Monitoring	Site #18	6.81	2.28	4.56	0.298	0.0316	95.0
Monitoring	Site #19	7.41	2.14	4.62	0.246	0.0304	90.7
Monitoring	Site #21	6.28	2.77	5.40	0.524	0.0277	141.0
Monitoring	Site #22	6.88	2.29	5.12	0.508	0.0353	111.0
Monitoring	Site #23	7.35	2.15	5.37	0.351	0.0356	117.0
Monitoring	Site #24	6.42	2.13	4.60	0.521	0.0287	95.0
Monitoring	Site #25	6.02	2.14	4.18	0.220	0.0305	79.5
Monitoring	Site #26	7.12	2.36	4.08	0.450	0.0366	101.0
Monitoring	Site #27	8.00	2.81	4.67	0.243	0.0315	103.0
Monitoring	Site #29	6.89	2.28	5.78	0.606	0.0341	116.0
Monitoring	Site #30	6.84	2.25	6.35	0.910	0.0337	130.0
Monitoring	Site #31	7.69	2.27	4.22	0.299	0.0278	97.4
Monitoring	Site #34	6.94	2.51	6.75	0.418	0.0263	109.0
Monitoring	Site #35	8.29	2.32	5.60	0.525	0.0344	91.8
Monitoring	Site #37	7.21	2.62	4.26	0.224	0.0293	87.1
Monitoring	Site #38	7.39	2.17	5.15	0.501	0.0314	107.0
Monitoring	Site #39	7.45	2.70	7.38	0.851	0.0350	130.0
Monitoring	Site #42	8.00	2.52	6.08	0.935	0.0421	140.0
Monitoring	Site #43	7.04	2.45	4.62	0.458	0.0259	108.0
Monitoring	Site #46	7.18	2.35	5.08	0.244	0.0301	111.0
Monitoring	Site #47	8.27	2.55	4.55	0.198	0.0357	93.2
Monitoring	Site #48	7.64	2.15	4.77	0.215	0.0288	98.2
Monitoring	Site #49	8.46	2.69	6.11	0.755	0.0341	110.0
Monitoring	Site #54	7.78	2.12	5.36	0.654	0.0339	110.0
Monitoring	Site #56	7.18	2.22	5.15	0.267	0.0308	85.2
AVERAGE		7.13	2.34	5.01	0.502	0.0315	101.2

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Table A6. Dry weight concentrations ( $\mu\text{g/g}$ ) of trace metals detected in mussels in 2019 at WDFW partner sites analyzed by the NOAA NWFSC Laboratory. Values were not blank corrected.

Site	As	Cd	Cu	Pb	Hg	Zn
AI_MMB	6.59	1.93	4.30	0.272	0.0255	94.6
AI_OB	7.11	2.39	4.46	0.220	0.0279	106.0
CB_CBSW	6.83	2.22	5.61	0.419	0.0293	89.4
CB_CBTF	7.82	2.77	7.55	0.563	0.0265	102.0
CB_DGL	6.63	2.07	5.30	0.497	0.0259	110.0
CB_MW	6.07	2.18	4.33	0.252	0.0239	81.5
CPS_EMB	7.57	2.34	4.63	0.306	0.0397	112.0
CPS_KM	6.94	1.95	5.04	0.305	0.0281	92.9
CPS_MASO	6.52	2.12	4.32	0.388	0.0293	135.0
CPS_PNP	7.20	2.44	4.82	0.183	0.0373	96.2
CPS_QMH	8.06	2.16	5.84	1.050	0.0467	83.6
CPS_RP	7.16	2.27	3.89	0.306	0.0273	87.7
CPS_SB	7.14	2.58	7.36	0.572	0.0408	101.0
CPS_SQSO	7.34	2.23	4.73	0.303	0.0352	106.0
EB_ME	5.70	2.20	4.11	0.255	0.0267	98.2
EB_P59	7.08	2.29	5.76	0.554	0.0352	110.0
NPS_BLSC	7.21	2.00	4.62	0.178	0.0276	80.1
NPS_CPAR4	8.00	2.30	4.33	0.241	0.0356	97.0
NPS_FBAR	6.88	2.18	4.55	0.246	0.0255	81.8
SJD_JSK	6.59	2.16	4.10	0.204	0.0233	76.1
SJD_NBM	6.92	2.03	4.86	0.403	0.0326	96.2
SPS_HIMP	6.56	2.18	3.95	0.236	0.0303	80.1
SPS_PBL	7.35	2.50	4.47	0.236	0.0278	95.9
SPS_SH	8.06	2.77	5.48	0.671	0.0355	95.3
WPS_SVD	7.08	2.19	5.09	0.714	0.0358	106.0
<b>AVERAGE</b>	<b>7.06</b>	<b>2.26</b>	<b>4.94</b>	<b>0.383</b>	<b>0.0312</b>	<b>96.6</b>

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*Table A7. Dry weight concentrations (μg/g) of trace metals detected in mussels in 2019 at MW sites analyzed by the NOAA NWFSC Laboratory. Values were not blank corrected. Oregon sites CBCH and YHFC were not analyzed by the NOAA NWFSC Laboratory.*

Site	As	Cd	Cu	Pb	Hg	Zn
BBSM	6.99	2.44	6.58	0.217	0.0286	102.0
CBTP	6.75	2.54	4.05	0.262	0.0326	91.3
EBDH	7.46	2.56	4.60	0.344	0.0461	104.0
EBFR	7.31	2.34	6.43	0.514	0.0499	129.0
PSEF	7.67	2.23	4.49	0.300	0.0423	103.0
PSEH	7.22	2.51	5.26	0.709	0.0509	94.7
PSEM	6.54	2.22	4.83	0.256	0.0448	97.4
PSHC	7.69	2.56	4.87	0.218	0.0500	93.2
PSKP	7.08	2.30	6.21	0.234	0.0397	102.0
PSMF	6.79	2.64	4.94	0.295	0.0533	101.0
PSPT	7.16	2.16	4.78	0.272	0.0367	93.8
PSTB	6.54	2.48	5.04	0.270	0.0428	88.0
SIWP	6.93	2.52	4.22	0.408	0.0404	93.3
SSBI	7.85	2.13	5.85	0.310	0.0394	103.0
WBNA	6.58	2.47	4.35	0.145	0.0372	79.9
<b>AVERAGE</b>	<b>7.10</b>	<b>2.41</b>	<b>5.10</b>	<b>0.317</b>	<b>0.0423</b>	<b>98.4</b>

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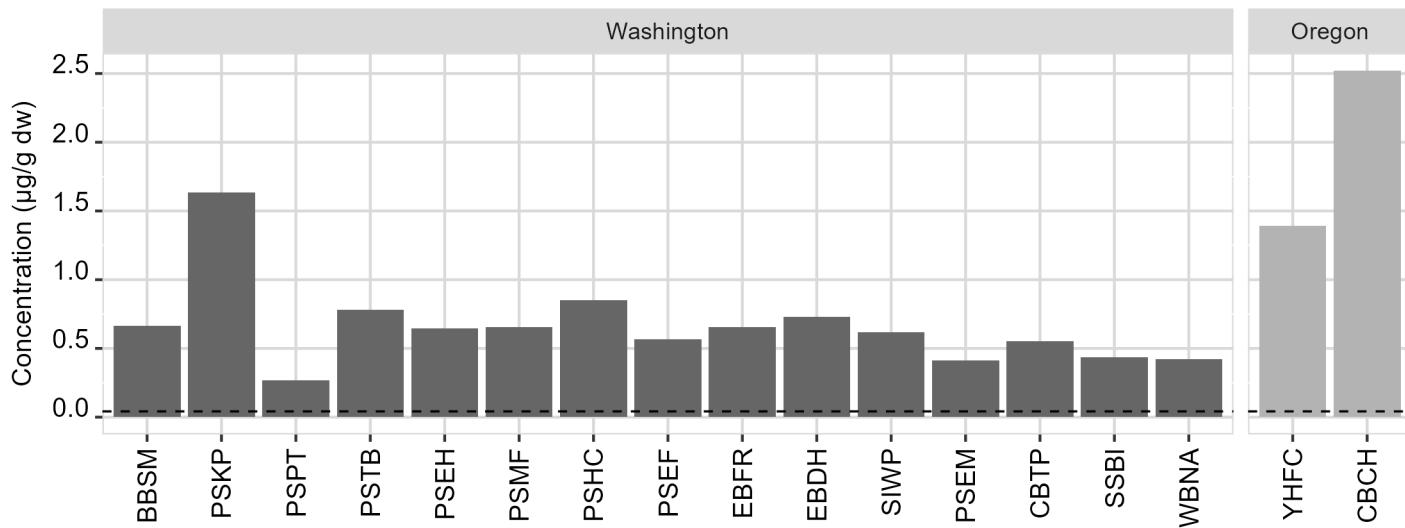


Figure A1. Bar graph showing magnitude of barium detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Barium was not sampled historically so is not included in the main body of this report.

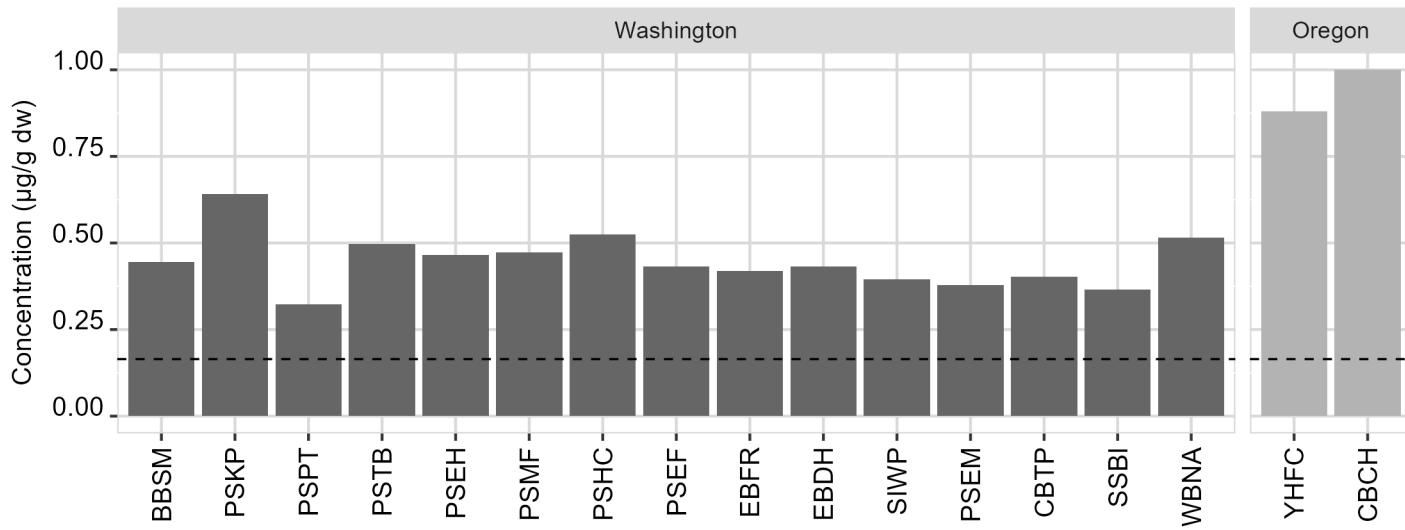


Figure A2. Bar graph showing magnitude of cobalt detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Cobalt was not sampled historically so is not included in the main body of this report.

# Appendices

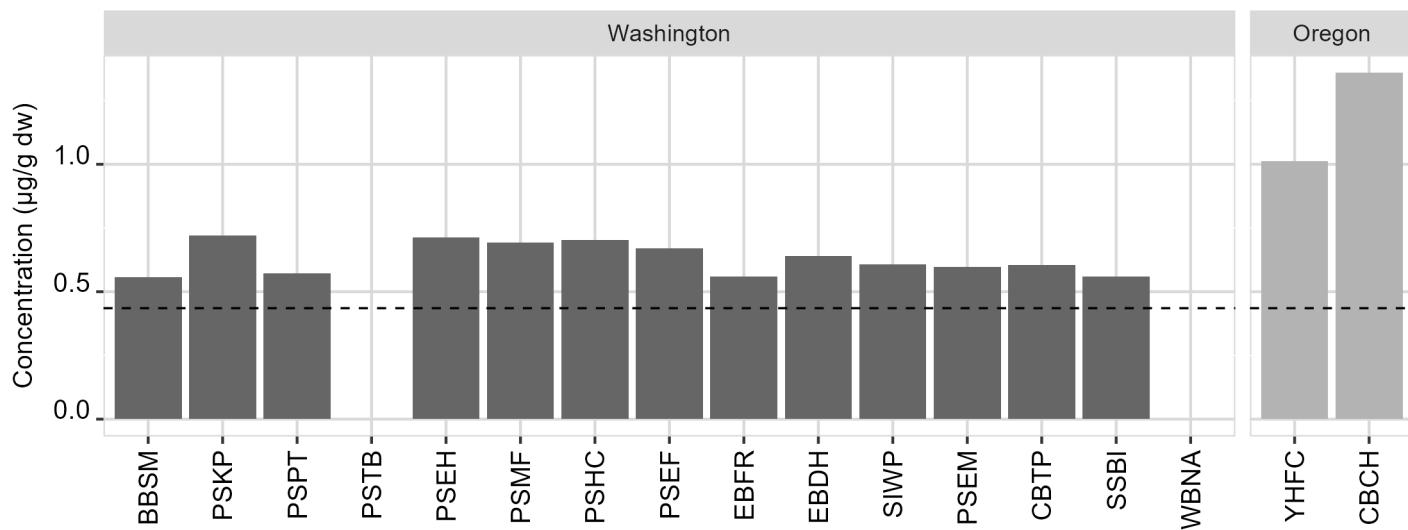


Figure A3. Bar graph showing magnitude of lithium detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Lithium was not sampled historically so is not included in the main body of this report.

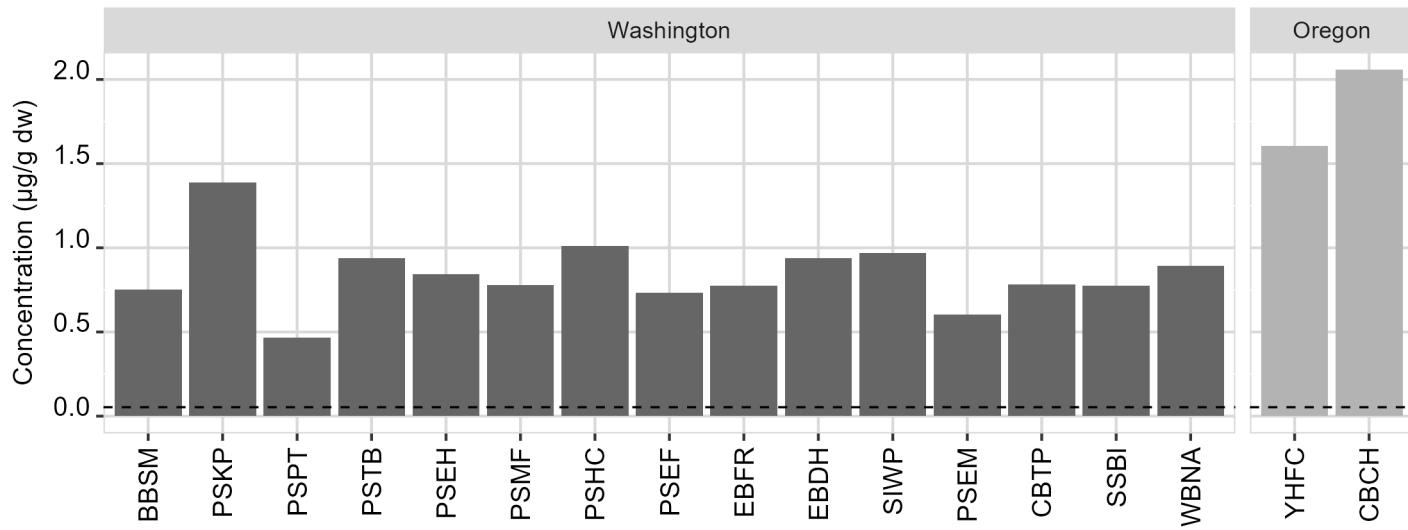


Figure A4. Bar graph showing magnitude of vanadium detected in mussel tissue in the Pacific Northwest in 2019. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline. Vanadium was not sampled historically so is not included in the main body of this report.



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National Ocean Service  
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The mission of the National Centers for Coastal Ocean Science is to provide managers with scientific information and tools needed to balance society's environmental, social and economic goals. For more information, visit: <http://www.coastalscience.noaa.gov/>

