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Cover Photo
Top left: Ecology staff collecting sediments from sampling vessel in Puget Sound.
Bottom left: Ecology staff scooping sediment out of vanVeen grab sampler and into a collection bucket.
Right: Estuarine sediments overlain with photos of products that contribute PPCPs and PFAS to the estuarine environment, including medical tablets and capsules, personal care product dispensers, firefighting foams, coatings for non-stick cookware, stain-resistant chemicals in carpeting, and grease-resistant and waterproof coatings in food packaging.

Pharmaceuticals, Personal Care Products, and Per- and Polyfluoroalkyl Substances in Puget Sound Sediments: 2010-2019 Data Summary

by

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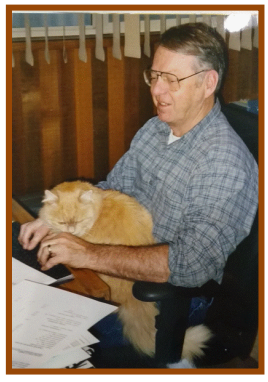
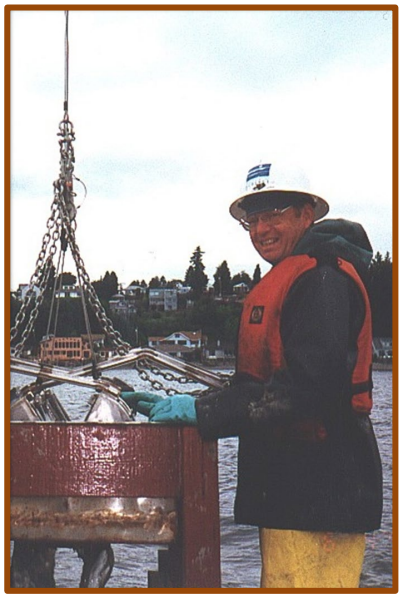
Dedication

This report is dedicated to the memory of our colleague of 18 years, Ed Long. Ed taught our team much about monitoring estuarine sediments for toxic contaminants and the Sediment Quality Triad of parameters. He was the lead author for many of our publications, including our first report on PPCPs and PFAS in sediments collected in 2010 (Long et al., 2013).





Ed was a cherished friend and mentor and is missed by his team.

Abstract

Surface sediments collected from 2010 through 2019 for the Puget Sound Sediment Monitoring Program were analyzed to determine concentrations of 118 pharmaceuticals and personal care products (PPCPs) and 24 per- and polyfluoroalkyl substances (PFAS), Chemicals of Emerging Concern (CECs) in Puget Sound. Stations sampled included 50 within our Puget Sound-wide sampling frame and 30 to 33 from each of six routinely-monitored urban bay sampling frames.

Occurrence and concentrations for both PPCPs and PFAS were low, incidence and spatial extent values differed widely among sampling frames, and the spatial distribution of detected chemicals also differed from one sampling frame to the next, reflecting unique chemical "signatures" in each bay.

Forty-three (36.4%) of 118 PPCPs were detected in 571 (3.0%) of 19,228 results. Diphenhydramine (an antihistamine), triclocarban (an antibiotic), and triamterene (a diuretic) were detected most frequently. Spatial patterns of detected PPCPs indicated that they often occurred in depositional locations with fine-grained sediments, at the heads of bays, and along portions of shoreline likely near discharge sources. More PPCPs were detected in Commencement Bay, the Bainbridge Basin, and Budd Inlet than in other locations.

Detectable levels of nine (37.5%) of 24 PFAS were measured in 71 (1.7%) of 4,189 results. Perfluorooctane sulfonate (PFOS) and perfluorohexanoate (PFHxA) were detected most frequently. PFOS occurred in depositional locations with fine-grained sediments, in some industrialized urban waterways, and in terminal inlets, and was not detected in the Budd Inlet or Pt. Gardner/Everett Harbor sampling frames. PFHxA was detected only in Budd Inlet and Pt. Gardner/Everett Harbor, with no obvious spatial patterns.

A literature review revealed that some PPCPs and PFAS detected in our sediments have been detected in other environmental media in Puget Sound and other eastern Pacific estuaries from British Columbia to southern California, in freshwater river and lake systems in Washington State, and elsewhere in the United States. The presence of these chemicals throughout the ecosystem suggests that future work to understand and manage PPCPs and PFAS in Puget Sound is necessary.

Introduction

Background

Chemical contaminants in the Puget Sound estuary and sediments have been the focus of intense regulatory, cleanup, and ambient monitoring activity since the inception of the Clean Water Act and Model Toxic Control Act in the 1970s (Dutch, et al., 2009; Norton et al., 2011; Roberts, 2017; PSP, 2018; Ecology, 2021). Traditional contaminant lists have included a suite of over 120 metals and organic chemicals based on the United States Environmental Protection Agency's (EPA) Toxic and Priority Pollutant lists developed in 1977 (EPA, 2021a).

Scientists, however, realize that thousands of other chemicals, developed for both consumer and industrial applications, are known or suspected to be released into aquatic environments without regulation or monitoring, with unknown or poorly understood impacts to ecological and human health (Kolpin et al., 2002; Diamond et al., 2011; James et al., 2015). Over the past two decades, a growing emphasis has been placed on quantifying the presence and understanding the impact of these *Chemicals of Emerging Concern*, or CECs, on the receiving environment and the organisms that live in them (Nilsen et al., 2019; Diamond and Burton, 2021).

The Washington State Department of Ecology's (Ecology) Puget Sound Sediment Monitoring Program (Sediment Program) was initiated in 1989 to measure levels of USEPA Priority Pollutant contaminants in sediments collected throughout the Sound (Puget Sound Water Quality Authority, 1988; Striplin, 1988). A traditional list of Priority Pollutant contaminants, which does not include PPCPs and PFAS, has been surveyed from 1989 to the present, with modifications made over time based on our findings. Our evolving contaminant lists can be viewed in our program's succession of quality assurance plans (Striplin, 1988; Dutch et al., 1998, 2009, 2018).

In the late 2000s, Ecology, along with the Puget Sound Partnership and other regional stakeholders, intensified efforts to better understand toxic chemical loadings to Puget Sound, including both traditional Priority Pollutants and CECs, with the goal of developing better strategies to resolve toxic contamination issues (Hart Crowser et al., 2007; Ecology, 2008; Ecology and Environment, Inc. 2009; Ecology and Herrera, 2010; Herrera, 2011).

In keeping with this renewed regional focus on toxics, our Marine Sediment Monitoring Team (MSMT) sought to expand the Sediment Program's contaminant list to include CECs of interest to Ecology scientists, regulatory staff, and other regional stakeholders. Our own survey of these stakeholders indicated high interest in gaining an understanding of two suites of CECs in Puget Sound sediments: (1) pharmaceuticals and personal care products (PPCPs), and (2) per- and polyfluoroalkyl substances (PFAS). With this in mind, we began scoping an expanded parameter list, while seeking both approval and supplementary funding to conduct these analyses.

Pharmaceuticals and personal care products (PPCPs)

PPCPs include thousands of prescription and over-the-counter drugs, nutritional supplements, shampoos, cosmetics, and lotions used by humans, and also includes antibiotics and other drugs used in veterinary practices and administered to commercial livestock. Use of these products is intense. Data from the National Health and Nutrition Examination Survey conducted in 2015 and 2016 indicated that 45.8% of the population in the United States (U.S.) used prescription drugs in the past 30 days, including 46.7% of adults aged 20-59 and 85% of those aged 60 and over (Martin et al., 2019). The same survey indicated that nearly 7 in 10 adults (69%) aged 40-79 used at least 1 prescription drug in the past 30 days in the U.S. while around 1 in 5 (22.4%) used at least 5 prescription drugs (Hales et al, 2019). Additionally, market size of beauty and personal care products in the United States was valued at $81.1 billion in 2019, and expected to reach $128.7 billion by 2030 (Prescient and Strategic Intelligence, 2020).

While PPCPs are formulated to elicit specific pharmacological effects, or responses, in humans or other animals they are administered to, varying percentages of the active ingredients passing through the body are ultimately excreted as either the parent compound or its metabolites (Thomas, 2003). They then make their way into both municipal and private wastewater treatment systems, where some are removed from the waste stream and others pass through untreated and can be detected in groundwater, freshwater systems, and wastewater treatment plant (WWTP) effluent and receiving waters (Kümmerer, 2004).

Prior to the inception of the work conducted for this report, studies conducted in the 2000s found PPCPs in measurable concentrations in wastewater treatment plant influent, effluent, and biosolids in Washington (Puget Sound and other waterbodies) and Oregon (Hope, 2012; Johnson et al., 2004; Lubliner et al., 2010; Morace, 2012), reclaimed water (Johnson and Marti, 2012), and surface and groundwater (Dougherty et al., 2010). Biological effects due to exposure to estrogenic and endocrine-disrupting chemicals had also been observed in Puget Sound fish (Johnson et al., 2008; Peck et al., 2011; daSilva et al., 2013). While PPCPs had been detected in sediments from the streambed of the lower Columbia River and selected tributaries (Nilsen et al, 2007), no sediment work had yet been conducted in Puget Sound or other Pacific Northwest estuaries.

Per- and Polyfluoroalkyl substances (PFAS)

PFAS are a group of human-made chemicals consisting of carbon atoms for which some or all hydrogen atoms have been replaced with fluorine. All hydrogen atoms have been replaced with fluorine in the perfluoralkyl substances, while polyfluoroalkyl substances are not fully fluorinated (Ecology, 2021). The carbon-fluorine bond exhibits extreme chemical and thermal stability, making these chemicals highly persistent — no known natural processes are able to break down these substances. Configuration of their atoms and bonds also make PFAS extremely water- and fat-repellent, as well as heat-resistant (Buck et al., 2011). The EPA's *PFAS Master List of PFAS Substances (Version 2)* includes 9252 PFAS chemicals, 6330 of which are registered in the Chemical Abstract Service and are of potential interest based on environmental occurrence, manufacturing process data, and testing within EPA research programs (EPA, 2020). The numbers are increasing as new forms are invented (NIEHS, 2021).

Since the 1940s, PFAS have been manufactured and used extensively in fabrication of products such as non-stick cookware, water- and stain-resistant clothing, fabrics, and carpeting; grease-resistant food packaging; aqueous film-forming foams (AFFFs) used to extinguish fires involving highly flammable liquids; and a variety of other products (EPA, 2021b). PFAS and PFAS-containing products are used extensively by the average consumer; by the military; and by the aerospace, aviation, automotive, construction, electronics, energy, food processing, paper production, textiles and leather treatment, metal-plating, petroleum and mining, and medical products industries (Ecology, 2021; NIEHS, 2021).

PFAS can be released into the environment during primary manufacturing of the chemicals themselves and during manufacturing of products containing them. As PFAS-containing products are used and ultimately disposed of, these persistent chemicals gradually find their way into the environment (Ecology, 2021). Humans are exposed to them via food, drinking water, breast milk, and airborne dust. Point- and nonpoint-source wastewater discharges carry PFAS into streams, rivers, and estuaries, where they accumulate and are detected in water, sediment, and aquatic organisms (Buck et al., 2011).

The two most-studied PFAS chemicals, PFOA (perfluorooctanoic acid) and PFOS (perfluorooctanesulfonic acid), have been associated with low infant birth weights, effects on the immune system, cancer, and thyroid hormone disruption in humans (Lau, 2015). These two chemicals have also been linked to reproductive and developmental, liver and kidney, and immunological effects, as well as tumors in laboratory animals (EPA, 2016). Based on their properties, PFAS are chemicals considered to be persistent, bioaccumulative, and toxic (Kümmerer, 2004).

As with PPCPs, a limited number of studies had been conducted in the 2000s to assess whether PFAS were present in the Puget Sound ecosystem. Prior to the inception of the surveys conducted in Puget Sound sediments for this report, PFAS were detected in surface water, fish tissue, and osprey eggs in and around Washington rivers and lakes, five of which drain into the Puget Sound watershed (Furl and Meredith, 2010). Additionally, PFAS were detected in effluent from 10 Puget Sound area WWTPs (Ecology and Herrera, 2010). Again, as with PPCPs, no surveys had yet been conducted on the presence and concentrations of PFAS in Puget Sound sediments.

PPCPs and PFAS prioritized for Puget Sound sediment sampling

With both the growing set of data indicating the presence of PPCPs and PFAS in Washington State ecosystems and the desire of stakeholders to better understand the presence and concentrations of these chemicals in Puget Sound, the MSMT received supplemental funding from the EAP in 2010 to expand our annual Sediment Program's analyte list for that year's survey of 10 sentinel[1] Puget Sound stations and 30 stations sampled in Bellingham Bay (Long et

[1] "Sentinel" stations – We collect sediments from 10 "sentinel" Puget Sound stations in locations with differing habitat and benthic invertebrate assemblage characteristics. These stations have been sampled annually since the inception of the program in 1989 to look for changes over time in their benthic assemblages.

al., 2013) to include PPCPs and PFAS. A suite of 118 PPCPs and 13 (later expanded to 24) PFAS were selected for analyses to establish a baseline dataset of their concentrations.

In subsequent years, interest in understanding sources, fate, and transport of toxic chemicals, including CECs, remained high with Puget Sound stakeholders (e.g., the Puget Sound Partnership Action Agenda and associated Implementation Plan for 2018 – 2022 (PSP, 2018), and previous versions generated in 2008, 2012, 2014, 2016). Based on these priorities, supplemental funding was secured periodically through 2020 from both the NEP and from the EAP to conduct similar analyses on sediments from five additional Puget Sound urban bays and at 50 of the Sediment Program's Long-Term monitoring stations collected during the MSMT's routine annual sediment monitoring cruises (Dutch et al., 2018). These 50 Long-Term stations included the 10 sentinel stations sampled in 2010.

Study objectives

The objectives of the Sediment Program's supplemental PPCP and PFAS analyses were to:

- Establish a baseline record of concentrations of selected PPCPs and PFAS for our program's 10 sentinel stations, six urban bay sampling frames, and Puget Sound-wide sampling frame.
- Compare the incidence, spatial extent, spatial patterns, and concentrations of PPCPs and PFAS in sediments collected from our stations and sampling frames.
- Determine whether PPCPs and PFAS detected in our sediment samples are also detected in other components of the Puget Sound ecosystem and other estuaries.
- Provide high-quality PPCP and PFAS sediment data and data summaries to Puget Sound scientists, managers, regulators, and the general public.

Methods

Sediments were collected by the MSMT during eight monitoring events conducted from 2010 through 2019 at 10 sentinel long-term stations and at stations distributed throughout six urban bays and our Puget Sound-wide sampling frame (Figure 1). Summary information for each event is provided in Table 1. The quality assurance plans referenced with each project provide detailed sampling information for each event, including maps and geographic coordinates for all stations.

Station locations

Stations sampled for the PPCP/PFAS surveys include those drawn from different Sediment Program monitoring elements developed over the life of the program. The locations of the 10 sentinel stations[2] were hand-selected at the inception of the program (Striplin, 1988). Each represents an area of seafloor with differing sediment and benthic community characteristics. Stations sampled from the urban bay and Puget Sound-wide sampling frames were selected using a simple stratified random (SSR) sampling design, a generalized random tessellation stratified (GRTS) multi-density category-weighted design, or an intensive GRTS equally-weighted survey design (Dutch et al., 2009, 2018). The differing designs reflect the evolution of the Sediment Program over time. The SSR and the GRTS designs allow our team to characterize the spatial extent (km^2) of sediment quality parameters, including PPCP and PFAS concentrations, measured in the associated urban bay or Puget Sound-wide sampling frame.

Field sampling

A double 0.1-m^2 van Veen grab sampler was deployed multiple times during each sampling event to collect sediments from each station. The top 2-3 cm of sediment were removed from one side of the grab and composited. Sediments retained for PPCP and PFAS analyses were preserved and transported to the appropriate laboratory for analysis. Standard Operating Procedures for sediment sample collection are summarized in detail in Weakland, 2021, while sampling containers, preservation, and holding times for PPCP and PFAS samples are summarized in Table 2.

Extraction, treatment, and analysis of samples

Sample extraction, cleanup, analysis, and quantification were conducted by AXYS Analytical Services Ltd., Sidney, BC, Canada (AXYS) for a suite of 118 PPCPs and 13 PFAS (Table 3) for sediment samples collected in 2010, 2013, 2014, and 2015. SGS AXYS Analytical Services, Ltd. (SGS AXYS[3]) also conducted PPCP analysis for sediments collected in 2019. In 2019 and 2020[4], PFAS analyses were conducted at Ecology's Manchester Environmental Laboratory

[2] Station names, numbers, and locations for the 10 sentinel stations are in Table 9 of Dutch et al., 2018.

[3] AXYS Analytical Services, Ltd. became SGS AXYS Analytical Services, Ltd. in 2016.

[4] In 2020, analyses were conducted on archived sediment samples collected in 2019 from the Sediment Program's 50 Puget Sound-wide stations.

(MEL), Manchester, WA, on expanded suites of PFAS analytes (21 and 24, respectively) (Table 3).
A summary of sample extraction, cleanup, and analysis methods conducted each year is provided in Table 2. Further details can be found in the case narrative/data packages and quality assurance reviews provided by AXYS/SGS AXYS and MEL. These are included in Appendix A.

For analyses conducted at AXYS/SGS AXYS, analyte concentrations in sediments were measured with AXYS Method MLA-075 (an extension of EPA 1694) for PPCPs and MLA-041 for PFAS. The methods use both acidic and basic extractions, spiking with isotopically labeled surrogates or standards, and analysis and quantification with high performance liquid chromatography-tandem mass spectrometry. The PPCP analyses include use of both positive and negative electrospray ionization in multiple reaction monitoring mode, while the PFAS analyses use negative electrospray ionization mode only. All methods used by AXYS/SGS AXYS were consistent from 2010 through 2019, ensuring comparability of their data among years. Minor changes to procedures over time were validated to ensure data continuity.

As indicated in Table 2, sample preservation, hold time, extraction, clean-up, and analysis methods for PFAS measured by MEL differed in some ways from those used by AXYS. In 2019, samples were immediately frozen rather than refrigerated after collection, with holding times extended from 14 days to 1 year. This change was based on MEL's use of EPA methods 8321B and 8327 for analysis of PFAS in our sediment samples. These methods indicate that while no formal holding time studies have been completed, freezing PFAS samples can prevent losses and degradation of some target and non-target analytes into other target PFAS.

Other changes included MEL's use of the "Quick, Easy, Cheap, Effective, Rugged, Safe" (QuEChERS) extraction method (AOAC 2007.01) and Agilent Enhanced Matrix Removal cleanup protocols to prepare samples, followed by analysis with EPA-modified method 8321B in 2019 and the similar EPA-modified method 8327 in 2020.

All PFAS methods included use of high performance liquid chromatography with triple quadrupole mass spectrometry in negative electrospray ionization mode. Modified methods 8321B and 8327 used by MEL includes isotopic dilution and internal surrogate standard quantitation, as does AXYS Method MLA-041. While instrument methods are broadly comparable, the extraction and cleanup steps differ between these methods. While the two labs worked to achieve similar levels of accuracy and precision in quantifying these chemicals, the use of different methods indicates the need for some caution when comparing 2010-2015 to 2019-2020 PFAS data.

In addition, it is important to note that the analytical methods for these chemicals are still undergoing refinement. For example, with PFAS, it is important to note that method 8327 was designed for non-drinking water aqueous samples. Draft 8328 was released later to allow for solid-phase extraction, and EPA now has draft method 1633 as the latest for non-aqueous matrices. These advances in analytical methods will certainly result in refinement of detection and reporting limits for these chemicals, resulting in potential increase in percent detection over

time. Ultimately, more caution in comparison of future data over time will be necessary (*Arianne Fernandez, Ecology's Toxics Cleanup Program – personal communication; December 2021*).

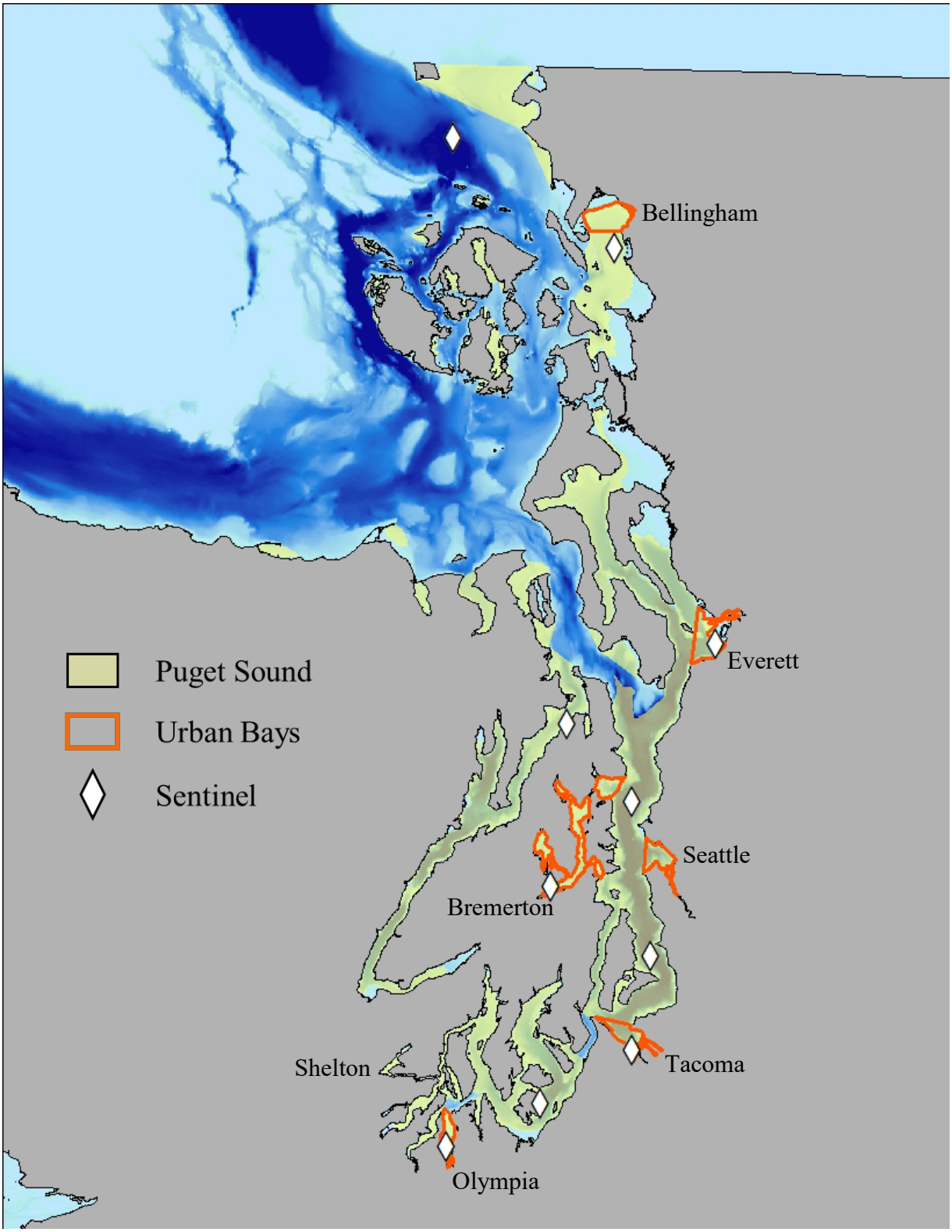


Figure 1. Location of the Puget Sound Sediment Monitoring Program's Sound-wide sampling frame, six urban bay sampling frames, and 10 sentinel stations.

Table 1. PPCP and PFAS sampling and analysis summary for the Puget Sound Sediment Monitoring Program, 2010 through 2020.

Sampling Year	Sampling Frame	No. of Stations	Parameters Analyzed	Analytical Laboratory	Quality Assurance Project Plan & addendum
2010	Long-Term sentinel stations[a]	10	118 PPCPs, 13 PFAS	AXYS[b]	Dutch et al., 2009; Dutch et al., 2010
2010	Bellingham Bay	30	118 PPCPs, 13 PFAS	AXYS	Dutch et al., 2009; Dutch et al., 2010
2013	Elliott Bay	30	118 PPCPs, 13 PFAS	AXYS	Dutch et al., 2009; Dutch et al., 2012[c]
2014	Commencement Bay	30	118 PPCPs, 13 PFAS	AXYS	Dutch et al., 2009; Dutch et al., 2014; 5/6/2014 Project Work Plan Memo
2015	Bainbridge Basin	33	118 PPCPs, 13 PFAS	AXYS	Dutch et al., 2009; Dutch et al., 2015
2019	Budd Inlet	30	118 PPCPs, 21 PFAS	SGS-AXYS[d] (PPCPs), MEL[e] (PFAS)	Dutch et al., 2018; Dutch et al., 2020
2019	Pt Gardner/ Everett Harbor	30	21 PFAS	MEL	Dutch et al., 2018; Dutch et al., 2020
2019[f]	Puget Sound	50	24 PFAS	MEL	Dutch et al., 2018; Dutch et al., 2020; 9/8/2020 Project Work Plan Memo

[a]10 "sentinel" stations located Puget Sound-wide from the Sediment Program's Long-Term Temporal monitoring element (Dutch et al., 2009)

[b]AXYS Analytical Services Ltd., Sidney, BC, Canada

[c]Station information only, all analytical methods for 2013 are taken from 2010 QAPP addendum

[d]SGS AXYS Analytical Services, Ltd., Sidney, BC, Canada

[e]Manchester Environmental Laboratory, Manchester, Washington

[f]Analysis conducted in 2020 on archived sediment samples collected in 2019 from the Sediment Program's 50 Puget Sound-wide stations

Table 2. Sample collection, preservation, and analyses for PPCPs and PFAS in homogenized sediment collected for the Sediment Program.

Parameter	Container	Preservation	Maximum Holding Time[a]	Extraction Method	Clean-up Method	Analysis Method	Technique/ Instrument
PPCPs (2010 – 2019, AXYS)	8 oz HDPE internally certified by lab	Wrap in aluminum foil and place in ice chest with dry ice or in field freezer immediately after collection. Store in dark at less than -10°C until analyzed.	Extract within 48 hours if not frozen or within 7 days of collection if frozen. Extract within 48 hours of removal from freezer. Analyze extracts within 40 days of extraction.	Two extractions – one basic and one acidic. The samples are pH adjusted, then sonication with aqueous buffered acetonitrile and pure acetonitrile, concentrate then dilute with ultra-pure water.	Solid-phase extraction cartridge then filtered.	AXYS Method MLA-075 (an extension of EPA 1694)	HPLC/ESI-MS/MS. High performance liquid chromatography with triple quadrupole mass spectrometer in positive and negative electrospray ionization modes using isotopic dilution and internal standard quantitation techniques.
PFAS (2010 – 2015, AXYS)	8 oz HDPE internally certified by lab	Refrigerate at 4°C±2°C	14 days to extraction	Shake extraction with dilute acetic acid solution then methanolic ammonium hydroxide solution (pH 9). Combine supernatants and treat with ultrapure carbon powder and diluted with ultrapure water.	Weak anion exchange (WAX) sorbent solid-phase extraction	AXYS Method MLA-041	HPLC/ESI-MS/MS. High performance liquid chromatography with triple quadrupole mass spectrometer in negative electrospray ionization mode using isotopic dilution and internal standard quantitation techniques.
PFAS (2019, 2020 MEL)	8 oz HDPE internally certified by lab	Freeze	1 year	AOAC2007.01 - *Quick, Easy, Cheap, Effective, Rugged, Safe* extraction (QuEChERS) with acetonitrile/ammonium hydroxide solution (pH 9).	Agilent Enhanced Matrix Removal cleanup protocol. Then dilute with ultra-pure LCMS water for analysis.	EPA 8321B (2019), 8327 (2020)	HPLC/ESI-MS/MS. High performance liquid chromatography with triple quadrupole mass spectrometer in negative electrospray ionization mode using isotopic dilution quantitation techniques.

[a]These are suggested holding times only. Formal holding time studies have not been performed or published for these analyses.

Table 3. List of 118 personal care products and pharmaceuticals (PPCPs) and 24 per- and polyfluoroalkyl substances (PFAS) measured in Puget Sound sediments for the Puget Sound Sediment Program, 2010 through 2020.

PPCPs

List 1

Acetaminophen
Azithromycin
Caffeine
Carbadox
Carbamazapine
Cefotaxime
Ciprofloxacin
Clarithromycin
Clinafloxacin
Cloxacillin
Dehydronifedipine
Digoxigenin
Digoxin
Diltiazem
1,7-Dimethylxanthine
Diphenhydramine
Enrofloxacin
Erythromycin-H20
Flumequine
Fluoxetine
Lincomycin
Lomefloxacin
Miconazole
Norfloxacin
Norgestimate
Ofloxacin
Ormetoprim
Oxacillin
Oxolinic acid
Penicillin G
Penicillin V
Roxithromycin
Sarafloxacin
Sulfachloropyridazine
Sulfadiazine
Sulfadimethoxine
Sulfamerazine
Sulfamethazine
Sulfamethizole
Sulfamethoxazole
Sulfanilamide
Sulfathiazole
Thiabendazole
Trimethoprim
Tylosin
Virginiamycin

List 2

Anhydrochlortetracycline
Anhydrotetracycline
Chlortetracycline
Demeclocycline
Doxycycline
4-Epianhydrochlortetracycline
4-Epianhydrotetracycline
4-Epichlortetracycline
4-Epioxytetracycline
4-Epitetracycline
Isochlortetracycline
Minocycline
Oxytetracycline
Tetracycline

List 3

Bisphenol A
Furosemide
Gemfibrozil
Glipizide
Glyburide
Hydroclorothiazide
2-hydroxy-ibuprofen
Ibuprofen
Naproxen
Triclocarban
Triclosan
Warfarin

List 4

Albuterol
Amphetamine
Atenolol
Atorvastatin
Cimetidine
Clonidine
Codeine
Cotinine
Enalapril
Hydrocodone
Metformin
Oxycodone
Ranitidine
Triamterene

List 5

Alprazolam
Amitriptyline
Amlodipine
Benzoylecgonine
Benztropine
Betamethasone
Cocaine
DEET
Desmethyldiltiazem
Diazepam
Fluocinonide
Fluticasone propionate
Hydrocortisone
10-hydroxy-amitriptyline
Meprobamate
Methylprednisolone
Metoprolol
Norfluoxetine
Norverapamil
Paroxetine
Prednisolone
Prednisone
Promethazine
Propoxyphene
Propranolol
Sertraline
Simvastatin
Theophylline
Trenbolone
Trenbolone acetate
Valsartan
Verapamil

List 1 - Acid Extraction in Positive Ionization
List 2 - Tetracyclines in Positive Ionization
List 3 - Acid Extraction in Negative Ionization
List 4 - Basic Extraction in Positive Ionization
List 5 - Acid Extraction in Positive Ionization

PFAS

N-ethylperfluorooctane-sulfonamidoacetate (N-EtFOSAA)[b]
N-methylperfluorooctane-sulfonamidoacetate (N-MeFOSAA)[b]
Perfluorobutane sulfonate (PFBS)[a]
Perfluorobutanoate (PFBA)[a]
Perfluorodecane sulfonate (PFDS)[b]
Perfluorodecanoate (PFDA)[a]
Perfluorododecanoate (PFDoA)[a]
Perfluoroheptane sulfonate (PFHpS)[b]
Perfluoroheptanoate (PFHpA)[a]
Perfluorohexane sulfonate (PFHxS)[a]
Perfluorohexanoate (PFHxA)[a]
Perfluorononane sulfonate (PFNS)[b]
Perfluorononanoate (PFNA)[a]
Perfluorooctanesulfonamide (PFOSA)[a]
Perfluorooctane sulfonate (PFOS)[a]
Perfluorooctanoate (PFOA)[a]
Perfluoropentanoate (PFPeA)[a]
Perfluorotetradecanoate (PFTeDA)[b]
Perfluorotridecanoate (PFTrDA)[b]
Perfluoroundecanoate (PFUnA)[a]
Perfluoropentane sulfonate (PFPeS)[b]
8:2 fluorotelomer sulfonate (8:2 FTS)[c]
6:2 fluorotelomer sulfonate (6:2 FTS)[c]
4:2 fluorotelomer sulfonate (4:2 FTS)[c]

[a] 13 PFAS measured by AXYS (2010-2015) and MEL (2019-2020)
[b] 8 additional PFAS measured by MEL (2019)
[c] 3 additional PFAS measured by MEL (2020)

Data availability

All PPCP and PFAS data generated for sediments collected from 2010 through 2019 for our Monitoring Program surveys are available in the following formats:

- Raw data that can be downloaded from the MSMT Marine Sediments portal to Ecology's Environmental Information Management (EIM) database.
- Analyte concentration and spatial distribution summaries provided in the Results and Appendix sections of this report.
- A companion story map containing an interactive, on-line dashboard where you may search for specific PPCP and PFAS results and generate your own custom maps and summary graphics.

Data summary, standardization, and comparison

The incidence, or number and percent of detected chemicals, was calculated for each survey, as were the number and percentage of stations with detected values for each chemical. Spatial patterns for detected chemicals were geographically plotted and examined, and the spatial extent (km^2 and percent of area) of chemicals detected for each urban bay and the Puget Sound-wide sampling frame was calculated per the Sediment Program study design (Dutch et al., 2018).

Minimum and maximum detected concentrations were summarized for each chemical, along with the minimum and maximum reporting limits (RLs) and the ratio of the minimum and maximum concentrations to the RLs. Regression on order statistics (ROS; Helsel, 2012) was used to estimate mean and median concentrations for chemicals where both detected and non-detected concentrations were observed.

Our findings were compared among the sentinel stations, urban bays, and the Puget Sound-wide surveys. To facilitate comparison of chemicals and sampling frames, chemical concentrations were standardized to be on a common scale. For each chemical, all valid results, including undetected results reported at their RL, were divided by the maximum detected value across all surveys, resulting in *percents of the maximum detected concentration*.

Additionally, the *weighted mean detected concentration* was calculated for each survey for each chemical. Based on our sampling designs, this procedure properly adjusts (weights) the results from each station proportionally to characterize the entire sampling frame, allowing comparison among sampling frames (Olsen et al., 2012). Detected chemical concentrations were then standardized by the *maximum weighted survey mean* for each chemical, to enable comparisons across chemicals.

We also conducted a review of 33 published government agency reports and peer-reviewed literature reporting PPCP and PFAS monitoring results from aquatic media sampled in other locations. They included marine, estuarine, and freshwater surveys that had been conducted in Puget Sound; Eastern Washington State; the Pacific Northwest, including Southwest Washington, Oregon, and British Columbia; California; and elsewhere in the United States. We compared the results from these surveys with our own to determine other media and locations in which the PPCPs and PFAS detected in our survey occurred.

Data Quality

Case narratives, data packages, and quality review

Case narratives with detailed PPCP and PFAS data packages were provided by AXYS/SGS AXYS to MEL for quality review. Similar narratives, with electronic data packages, were provided by MEL for their PFAS work. These case narratives, included in Appendix A, provide detailed information on:

- Methods
- Sample receipt and storage
- Sample preparation, extraction, and analysis
- Target analyte concentration calculation methods
- Reporting conventions
- QA/QC notes
- Analytical discussion

Every AXYS/SGS AXYS case narrative and data package was reviewed by either MEL's Quality Assurance officer Karin Feddersen or by MEL Organics Unit Supervisor John Weakland. These MEL experts examined raw data for qualitative and quantitative precision, accuracy, and bias. They also reviewed quality control conditions flagged by AXYS that might affect the data and added or changed qualifiers, as appropriate, to be consistent with MEL and EIM guidelines. Their MEL quality review reports are included in Appendix A.

Minor analytical and data quality issues occurred in every set of analyses. These are recorded in the AXYS/SGS AXYS and MEL case narratives and in the MEL data review documents, and their impact on data quality or comparability are discussed. Data were flagged with the appropriate qualifier codes. Most of the issues were deemed to have low or no impact to overall data quality. Larger issues related to analytical challenges, differences in sample handling, and data qualification methods are summarized below.

Poor fluoroquinolone recovery, all PPCP analyses in sandy sediments

In 2010, AXYS reported difficulties with quantification of seven List 1 PPCPs (see AXYS, 2010 report in Appendix A). The lab observed consistently low and variable recovery of the ^{13}C-^{15}N-ciprofloxacin surrogate standard in sediment samples, particularly in those with high sand content. This resulted in data quality issues for the fluoroquinolone target analytes quantified against this surrogate. Since this surrogate is used to correct for recovery and to quantify all fluoroquinolones, the seven List 1 fluoroquinolones – ciprofloxacin, ofloxacin, clinafloxacin, enrofloxacin, lomeofloxacin, norfloxacin, and sarafloxacin – were either flagged as not quantifiable or were quantified against instrument standards with appropriate flagging and narration. This issue persisted for all years of our survey.

Maximum holding times exceeded, 2019 PPCP analyses

The specified 7-day maximum PPCP holding times were exceeded by SGS AXYS for analysis of many of the 2019 urban bay sediment samples. While no formal holding time studies have been completed for these methods, these circumstances may have affected comparability between the 2019 PPCP urban bay data and data generated in earlier years from samples analyzed within this specified holding time. During quality review of these data, therefore, MEL qualified 2,992, or 86.7%, of the results either as "J" (estimated) or "UJ" (not detected, sample quantitation limit estimated).

Maximum holding times exceeded and sample container type changed, 2020 PFAS analyses

In late 2020, personnel time and funding for analysis of PFAS became available at MEL. While no sampling was conducted that year, archived sediments collected during our 2019 Puget Sound-wide monitoring at 50 stations were available for analysis. These samples had been frozen at -18°C after collection, but were in glass jars with Teflon-lined lids[5], rather than the recommended high density polyethylene (HDPE) jars, and were between 8 and 9 months beyond the one-year maximum holding time adopted by MEL. However, due to Ecology's strong interest in PFAS in the environment, there was agreement to analyze these archived samples for PFAS. All data for these samples were qualified as estimates and assigned either a "J" or "UJ" qualifier due to these deviations from protocol. Other reasons for "J" or "UJ" qualifiers may have existed for some of these 2019 data values, and are so noted in the case narrative.

To attain some comparative information on the effects of container type and hold time on sample variability, six quality assurance (QA) samples were run along with the 2019 Puget Sound-wide samples analyzed in 2020. These included sediment samples from three 2019 urban bay stations that were archived in HPDE and three from the same stations that were archived in glass jars with Teflon lids. Same-station results were compared to one another to examine differences in concentrations from samples held in different jar types. Additionally, the 2020 results from sediments in the HPDE containers were compared with the results from the same samples analyzed in 2019 within the 1-year hold time.

While there were too few samples to make definitive statements, comparison graphics (Appendix B) suggest that container type and hold time may have affected some of the PFAS results. From the limited sample measurements, all of the PFOS and PFHxA results that were analyzed over hold time were either undetected (1 of 12 results) or were measured at lower concentrations than the same sample analyzed within the holding time (11 of 12 results). These circumstances may

[5] The Teflon-lined lids from the glass jars may have been a possible source of background contamination for these 2019 PFAS samples. To minimize the possibility of analyzing contaminated sample, MEL staff removed and discarded approximately one-half inch of sediment from the top of the thawed sample. The remaining sediments were composited and analyzed following the designated methods.

affect comparability between the 2019 PFAS urban bay and 2020 Puget Sound-wide data, and comparison to PFAS data collected in previous years.

A recent study on the effect of sample type and storage temperature on the stability of 29 PFAS in water and effluent samples showed both increasing and decreasing concentration trends in samples stored at different temperatures for up to 180 days. This suggests there may be some analyte conversion based on storage time and temperature (Woudneh et al., 2019). It is unknown whether these changes may have occurred in the PFAS measured in our sediments archived for 20 to 21 months.

Lab differences in qualifying detected PFAS values

Reporting of qualifiers for the PFAS data differed markedly between AXYS/SGS AXYS and MEL. From 2010 through 2015, AXYS/SGS AXYS reported concentrations as "not detected" if values fell below the lowest point on the measured calibration range of the instrument, that is, below the RL[6]. These results were qualified as "U", or undetected, in their data sets. For the 2019 and 2020 analyses conducted by MEL, all results with values below the RL were qualified as estimates and given a "J" qualifier. These two systems for qualifying data with values below the RL are not comparable, so all MEL PFAS data qualified as "J" for this reason are treated in this report (both text and graphics) as if they were "U" qualified for comparison with the 2010-2015 data set. The original "J" qualifier code will be retained, however, in EIM.

Lab differences in detection limits

The majority of analyses conducted for these surveys resulted in undetected concentrations for most PPCPs and PFAS. Undetected values were reported at the RL, and differences in RLs were found for some chemicals both between surveys and between analytical labs. Among-survey comparisons should therefore be made with caution.

[6] The RL was defined by AXYS as the concentration equivalent to the lowest calibration standard or the sample specific detection limit, whichever was greater.

Results and Discussion

This survey of Puget Sound sediments establishes a baseline of PPCP and PFAS concentrations at 10 sentinel stations, 50 Puget Sound-wide stations[7], and 30-33 stations in each of six major urban bays[8].

PPCP

Detected PPCPs and their biochemical classes

Over the course of this project, concentrations of 118 PPCPs were measured in sediments from 163 stations. Forty-three of these chemicals were detected in our samples, including three – diphenhydramine, triclocarban, and triamterene – which were detected in over 100 samples each. Nine were detected in 10 to 31 samples, 12 in two to six samples, and 19 in only one sample each (Table 4). The detected PPCPs belong to 23 different classes with a wide range of biochemical functions. Each class makes up 0.2 to 25.2% of 571 total detected results (Figure 2). Over 65% of the total detected results fell into three biochemical classes based on the function of the three most-frequently detected chemicals. This included the antihistamine diphenhydramine (21.7%), a suite of antibiotics – primarily triclocarban (19.4%), but also 12 others which were less frequently detected, and the diuretic triamterene (18.4%).

[7] PFAS only at 50 Puget Sound-wide stations

[8] PFAS only in Pt Gardner/Everett Harbor

Table 4. Incidence of 43 PPCPs, and their associated biochemical classification, detected in six Puget Sound surveys, 2010 through 2019.

PPCP	*Chemical Class*	No. of Detected Results	Percent (%) of 571 Detected Results	PPCP	*Chemical Class*	No. of Detected Results	Percent (%) of 571 Detected Results
Diphenhydramine	*Antihistamine*	124	21.7	Prednisolone	*Steroid*	2	0.4
Triclocarban	*Antibiotic*	111	19.4	Simvastatin	*Statin*	2	0.4
Triamterene	*Diuretic*	105	18.4	4-Epitetracycline	*Antibiotic*	1	0.2
Sertraline	*Selective serotonin reuptake inhibitor (SSRI)*	31	5.4	Alprazolam	*Sedative*	1	0.2
Amitriptyline	*Antidepressant & nerve pain*	29	5.1	Amlodipine	*Calcium channel blocker*	1	0.2
Fluoxetine	*SSRI*	21	3.7	Anhydrochlortetracycline	*Antibiotic*	1	0.2
Miconazole	*Antifungal*	21	3.7	Anhydrotetracycline	*Antibiotic*	1	0.2
Verapamil	*Antihypertensive & calcium channel blocker*	19	3.3	Carbadox	*Antibiotic*	1	0.2
Azithromycin	*Antibiotic*	17	3.0	Ciprofloxacin	*Antibiotic*	1	0.2
Norverapamil	*Calcium channel blocker*	16	2.8	Cocaine	*Stimulant*	1	0.2
Erythromycin-H2O	*Antibiotic & gut motility stimulator*	11	1.9	Codeine	*Opioid pain reliever*	1	0.2
Amphetamine	*Stimulant & anorectic*	10	1.8	Cotinine	*Metabolite of nicotine*	1	0.2
Metformin	*Anti-diabetic medication*	6	1.1	Dehydronifedipine	*Calcium channel blocker*	1	0.2
Diltiazem	*Antihypertensive & calcium channel blocker*	5	0.9	Enrofloxacin	*Antibiotic*	1	0.2
DEET	*Insect repellent*	4	0.7	Ibuprofen	*Nonsteroidal anti-inflammatory*	1	0.2
Desmethyldiltiazem	*Calcium channel blocker*	4	0.7	Norgestimate	*Progestin*	1	0.2
Sarafloxacin	*Antibiotic*	3	0.5	Ofloxacin	*Antibiotic*	1	0.2
Sulfadimethoxine	*Antimicrobial*	3	0.5	Paroxetine	*SSRI*	1	0.2
Albuterol	*Bronchodilator*	2	0.4	Propoxyphene	*Opioid pain reliever*	1	0.2
Benztropine	*Anti-tremor*	2	0.4	Sulfamethoxazole	*Antibiotic*	1	0.2
Caffeine	*Stimulant*	2	0.4	Valsartan	*Antihypertensive*	1	0.2
Oxytetracyclin	*Antibiotic*	2	0.4				

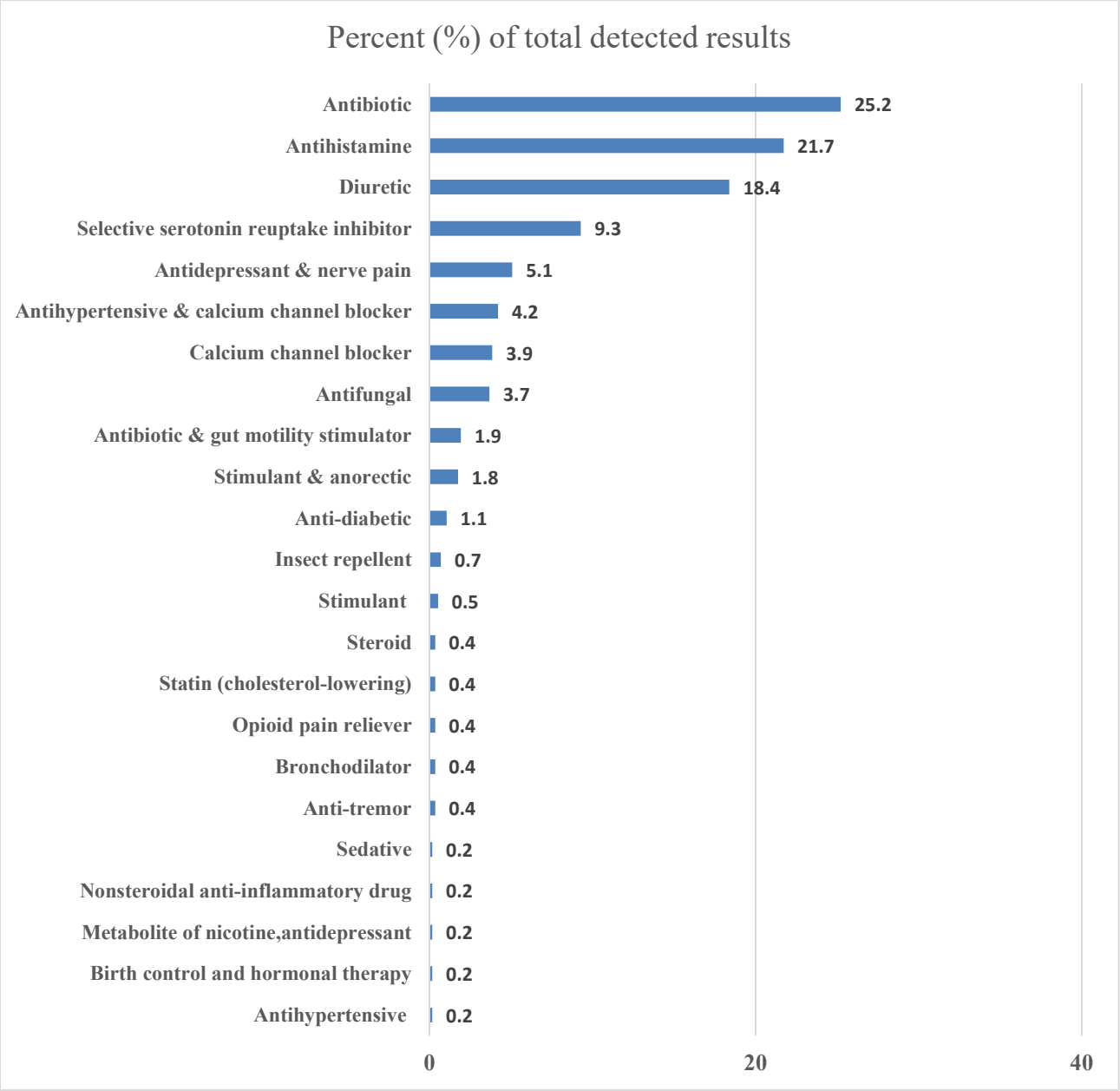


Figure 2. Percent of total detected results for the 23 biochemical classes of PPCP function detected in six Puget Sound surveys, 2010 through 2019.

Incidence and spatial patterns for all detected PPCPs and results

The incidence of detected PPCPs and total detected results varied among the study areas, likely due to the quantity, persistence, and treatment of contaminants in waste streams that discharge into each of the study areas and their interaction with conditions in the receiving environment (e.g., currents, temperature, sunlight). Detected concentrations of 43 (36.4%) of the 118 PPCPs occurred in 571 (3.0%) of 19,228 results generated. The highest number of PPCP analytes detected in a survey, 24 (20.3%), was measured in the Bainbridge Basin in 2015; the lowest was 7 (5.9%), measured at the 10 sentinel stations in 2010. The highest number of detected PPCP results, 159 (4.5%), was found in Budd Inlet in 2019; the lowest, 18 (1.5%), was again at the 2010 sentinel stations (Table 5).

Table 5. Incidence of total PPCPs and total results detected for 118 personal care products and pharmaceuticals in six Puget Sound sediment monitoring surveys.

Survey Year/ Location	Number of Detected PPCPs	Percent (%) of 118 PPCPs	Number of Detected Results	Percent (%) of Total Results	Total Number of Results
2010 Sentinel stations	7	5.9	18	1.5	1,180
2010 Bellingham Bay	13	11.0	68	1.9	3,540
2013 Elliott Bay	13	11.0	75	2.1	3,540
2014 Commencement Bay	20	16.9	142	4.0	3,540
2015 Bainbridge Basin	24	20.3	109	2.8	3,888
2019 Budd Inlet	17	14.4	159	4.5	3,540
Overall	**43**	**36.4**	**571**	**3.0**	**19,228**

Spatial patterns of the number of PPCPs detected at each station displayed both similarities and differences among the six surveys (Figure 3).

In 2010, no PPCPs were detected at the Sinclair Inlet sentinel station near the city of Bremerton, while one to three were detected at eight of the other sentinel stations. Six were detected at the Point Pully sentinel station located at 200 meters depth near the city of Des Moines, in Puget Sound's Central Basin. The sediments at this deep station consist of silt-clay particles onto which contaminants may have adsorbed while suspended in the water column.

Zero to four PPCPs were measured in stations sampled in Bellingham Bay in 2010, with the highest number of analytes found either in stations closest to the eastern shoreline or in deeper stations in the center of the bay.

In the 2013 Elliott Bay survey, zero to four PPCPs were found at stations in the Duwamish River, the East and West Waterways (WW), and along the northwestern shoreline, while five to eight were found in the deepest central stations with sediments consisting of high percent fines.

One to 12 PPCPs were measured in stations from Commencement Bay in 2014. Stations with the highest numbers of chemicals were found in the bay's industrial waterway, including the western-most Thea Foss WW, the eastern-most Hylebos WW, and the adjacent Blair WW. The

number of detected values declined north and westward of these waterways toward the mouth of Commencement Bay.

Bainbridge Basin sediments, sampled in 2015, had zero to 11 detected chemicals. The highest numbers of detected analytes were found near the heads of the multiple terminal inlets of the basin, including Sinclair Inlet, Dyes Inlet, Liberty Bay, and Phinney Bay, and at a few stations in Rich Passage.

Two to 11 PPCPs were detected in Budd Inlet sediments in 2019. The highest numbers of detected analytes were again near the head of the inlet in the East Bay WW near discharge from the Deschutes River, along the eastern shoreline, and in some of the deeper, central inlet stations. Most of the lower values were along the western shoreline and at the mouth of the inlet.

Overall, PPCPs detected in the five urban bays and at the 10 Long-Term sentinel stations displayed the following spatial patterns:

- The sentinel station with the highest number of detected PPCPs was a deep, depositional Central Basin station with fine-grained sediments.
- Most sentinel, Bellingham Bay, and Elliott Bay stations had fewer detected chemicals than stations in Commencement Bay, the Bainbridge Basin, and Budd Inlet.
- Bellingham Bay and Elliott Bay had the highest numbers of chemicals detected at their deepest stations with fine-grained sediments.
- In Commencement Bay, the Bainbridge Basin, and Budd Inlet, larger numbers of chemicals were detected near the head of each bay or along portions of shoreline likely near discharge sources. Chemicals were also found in the path of currents likely carrying particulate contaminants and in deeper, depositional locations that accumulate fine-grained sediments.

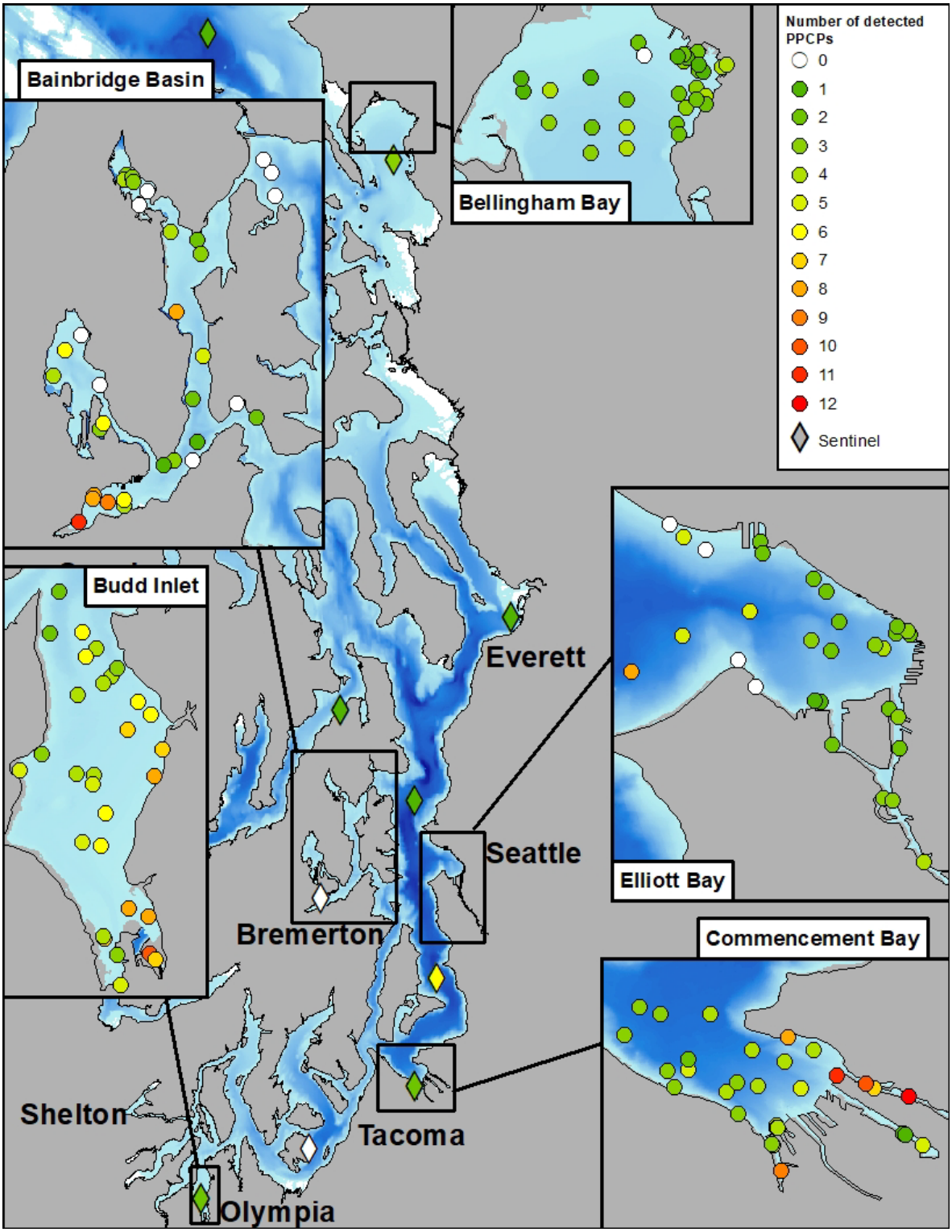


Figure 3. Spatial patterns for the number of PPCPs detected in sediments from 10 sentinel stations and in 5 urban bays, 2010 through 2019.

In all Figures, one of three stations in Commencement Bay's Middle WW (second WW from left) is not visible.

Three dominant PPCPs – incidence, spatial extent and patterns, and summary statistics

As indicated earlier, diphenhydramine, triclocarban, and triamterene were detected most frequently in the sediments sampled in our surveys. Incidence and spatial extent values for these three PPCPs are summarized below for each survey, along with all measured and estimated chemical concentrations and ratio-to-RL values. Spatial patterns of the concentrations of each of these chemicals are also depicted below. Concentration values for all 43 detected PPCPs are available in Appendix C-1, and spatial patterns for all can be interactively explored by our readers in our companion story map.

Diphenhydramine

Diphenhydramine, an antihistamine, was the most frequently detected chemical in our surveys. This chemical was detected in 93.3% of the Bellingham Bay samples, representing 87.9% of the study area. It was also detected in 90% of all Budd Inlet samples, representing 90% of the study area. Incidence and spatial extent were lower in Elliott Bay and Commencement Bay, and lowest in the Bainbridge Basin, where it was detected in 57.6% of samples, representing 44.7% of the study area (Figure 4, Table 6).

Concentrations in all surveys ranged from 0.4 to 14.5 ng/g dry weight (dry wt) with the greatest range of values measured in Budd Inlet. Highest concentrations were found in West Bay at the southern end of Budd Inlet and in the middle and head of the Hylebos WW in Commencement Bay, possibly near sources of contamination. High levels were also found in two of the deep, depositional central stations in Elliott Bay, which may accumulate silts and clays contaminated with these chemicals (Figure 4, Table 7).

Triclocarban

Triclocarban, an antibacterial used in some soaps and cleaning products, was the second-most commonly detected PPCPs in our survey. This chemical was detected in 90.0% and 96.7% of the samples from Commencement Bay and Budd Inlet, respectively, and in both bays represented approximately 96.7% of the study area. Incidence was lowest in Bellingham Bay and the Bainbridge Basin, detected in 30.0% and 48.5% of samples and representing approximately 30.5% and 35.7% of these sampling frames, respectively (Figure 5, Table 6).

Concentrations in all surveys ranged from 1.1 to 96.5 ng/g dry wt, with the greatest range of values measured in Elliott Bay. Highest concentrations were observed in the Duwamish River and associated waterways, near the eastern shoreline, and in the deep, depositional stations in Elliott Bay; in the Thea Foss WW in Commencement Bay; in terminal Sinclair Inlet in the Bainbridge Basin; and in the terminal East Bay in Budd Inlet (Figure 5, Table 7). Again, all areas with highest concentrations were likely near potential sources of contaminants or were in deep, depositional locations.

Triamterene

Triamterene, a diuretic, was detected in 96.7% and 100% of the sediment samples collected from the Commencement Bay and Budd Inlet sampling frames, representing 98.4% and 100% of these study areas, respectively. Incidence was lowest at the sentinel stations (20% of 10 samples), and

in Bellingham Bay and the Bainbridge Basin, where it was detected in 40.0% and 48.5% of samples, representing 35.7% of each of these study areas (Figure 6, Table 6).

Concentrations in all surveys ranged from 0.2 to 2.2 ng/g dry wt, with the greatest range of values measured in Budd Inlet. Highest concentrations were observed in both East Bay and West Bay and along the eastern Budd Inlet shoreline; at the head of the Thea Foss WW and along the north eastern shoreline of Commencement Bay; in the East WW, near the shoreline, and in deeper stations in Elliott Bay; and in stations at the head and middle of Sinclair Inlet in the Bainbridge Basin (Figure 6, Table 7).

In summary, all three of these most-frequently detected chemicals were found in the highest percentage of stations and covering the highest percentage of study area in Budd Inlet and Commencement Bay. Incidence and spatial extent values were also high in Bellingham Bay for diphenhydramine. Lowest incidence and spatial extent values for triclocarban and triamterene were generally found in the Bainbridge Basin and Bellingham Bay, and in the Bainbridge Basin for diphenhydramine.

Table 6. Incidence and spatial extent of the three most frequently detected PPCPs in six Puget Sound sediment surveys.

Parameter Year, Sampling Frame	**No. of Stations**	**No. of Stations with Detected Values**	**% of Stations with Detected Values**	**Area (km²)**	**% of Study Area**
Diphenhydramine					
2010 Sentinel stations	10	7	70.0	NA[a]	NA
2010 Bellingham Bay	30	28	93.3	36.3	87.9
2013 Elliott Bay	30	18	60.0	19.5	74.0
2014 Commencement Bay	30	25	83.3	18.4	76.4
2015 Bainbridge Basin	33	19	57.6	36.6	44.7
2019 Budd Inlet	30	27	90.0	15.6	90.0
Triclocarban					
2010 Sentinel stations	10	5	50.0	NA	NA
2010 Bellingham Bay	30	9	30.0	12.6	30.5
2013 Elliott Bay	30	25	83.3	23.3	88.3
2014 Commencement Bay	30	27	90.0	23.3	96.7
2015 Bainbridge Basin	33	16	48.5	29.2	35.7
2019 Budd Inlet	30	29	96.7	16.8	96.7
Triamterene					
2010 Sentinel stations	10	2	20.0	NA	NA
2010 Bellingham Bay	30	12	40.0	14.7	35.7
2013 Elliott Bay	30	16	53.3	18.9	71.7
2014 Commencement Bay	30	29	96.7	23.7	98.4
2015 Bainbridge Basin	33	16	48.5	29.2	35.7
2019 Budd Inlet	30	30	100.0	17.3	100.0

[a]NA - The 10 sentinel stations are not part of a sampling frame with a stratified random sampling design, so no spatial extent calculations for the measured PPCPs were determined for this survey.

Table 7. Summary statistics for the most frequently detected PPCPs in six Puget Sound surveys.

Parameter Year, Sampling Frame	**Minimum Detected Concentration (ng/g dry wt)**	**Maximum Detected Concentration (ng/g dry wt)**	**Estimated Mean Concentration (ng/g dry wt)[a]**	**Estimated Median Concentration (ng/g dry wt)[a]**	**Minimum RL ng/g dry wt)[b]**	**Maximum RL (ng/g dry wt)[b]**	**Min. Conc./RL (ng/g dry wt)[c]**	**Max. Conc./RL (ng/g dry wt)[c]**
Diphenhydramine								
2010 Sentinel stations	0.6	4.8	1.6	1.4	0.5	0.6	1.1	8.5
2010 Bellingham Bay	1.0	2.7	1.7	1.7	0.4	0.6	1.7	5.7
2013 Elliott Bay	0.6	12.7	2.4	1.3	0.4	1.4	1.1	23.9
2014 Commencement Bay	0.7	12.3	2.4	1.5	0.4	0.6	1.3	22.4
2015 Bainbridge Basin	0.8	9.2	2.9	1.6	0.5	0.6	1.3	15.8
2019 Budd Inlet	0.4	14.5	2.7	2.3	0.2	0.3	1.4	61.7
Triclocarban								
2010 Sentinel stations	3.1	16.6	5.6	2.4	2.4	3.0	1.0	5.9
2010 Bellingham Bay	3.3	9.6	2.7	1.6	2.3	3.1	1.1	4.1
2013 Elliott Bay	2.8	96.5	19.8	17.1	1.6	3.4	1.1	31.0
2014 Commencement Bay	3.4	60.2	8.7	5.5	2.1	3.1	1.1	22.6
2015 Bainbridge Basin	3.3	25.9	5.3	1.9	2.5	3.0	1.2	8.6
2019 Budd Inlet	1.1	18.0	4.8	3.9	0.9	1.6	1.1	12.8
Triamterene								
2010 Sentinel stations	0.8	0.8	NA	NA	0.3	0.3	2.7	2.7
2010 Bellingham Bay	0.3	0.5	0.3	0.2	0.2	0.3	1.0	1.8
2013 Elliott Bay	0.4	1.5	0.6	0.4	0.2	1.0	1.2	5.0
2014 Commencement Bay	0.3	1.2	0.5	0.5	0.2	0.3	1.0	4.5
2015 Bainbridge Basin	0.3	1.2	0.4	0.2	0.2	0.3	1.1	4.2
2019 Budd Inlet	0.2	2.2	0.9	0.8	0.1	0.2	2.1	15.6

RL = Reporting Limit
[a] Estimated by regression on order statistics (ROS) when non-detected data were present (Helsel, 2012)
[b] from all values
[c] from detected values only

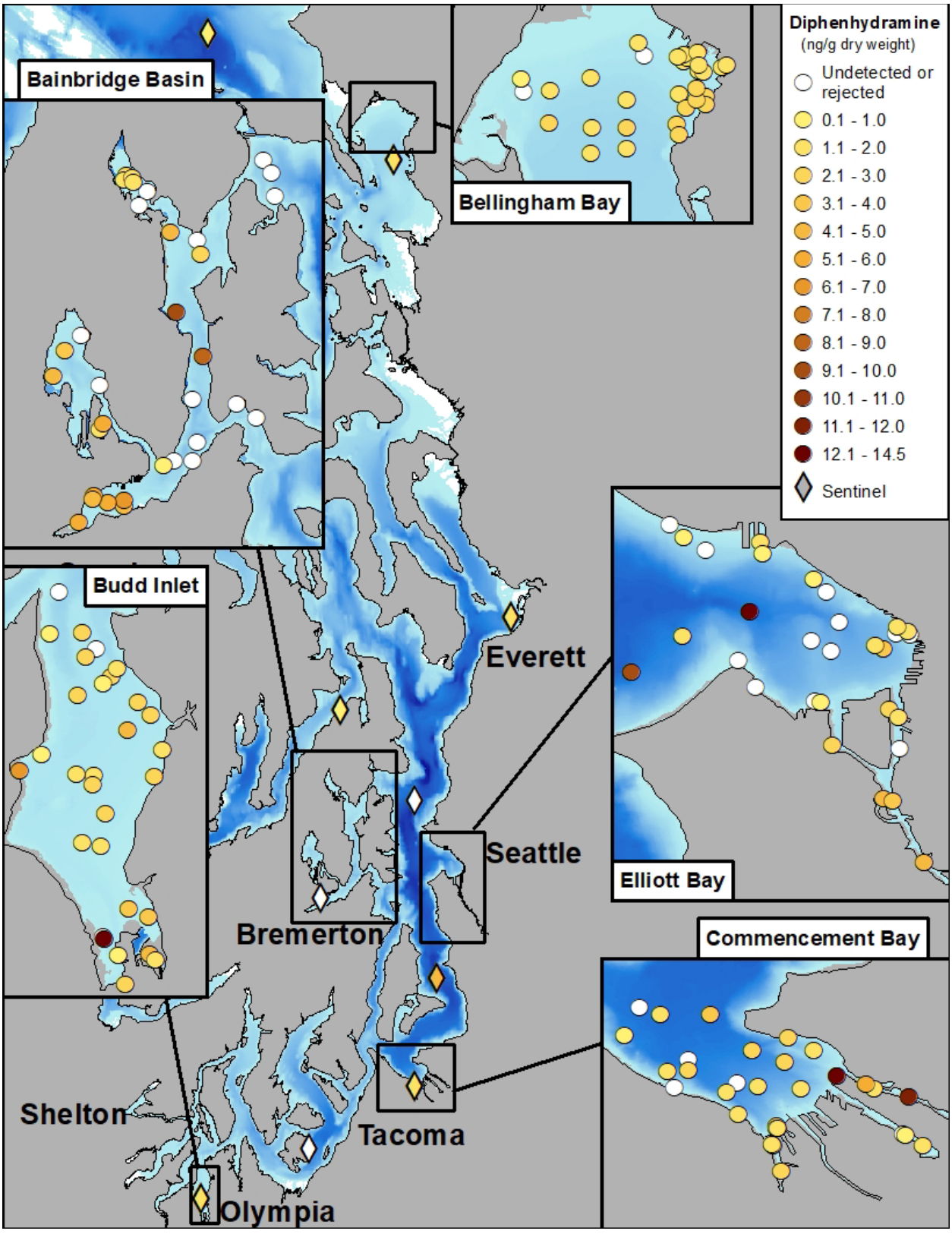


Figure 4. Spatial patterns for diphenhydramine concentrations (ng/g) detected in sediments from 10 sentinel stations and in 5 urban bays, 2010 through 2019.

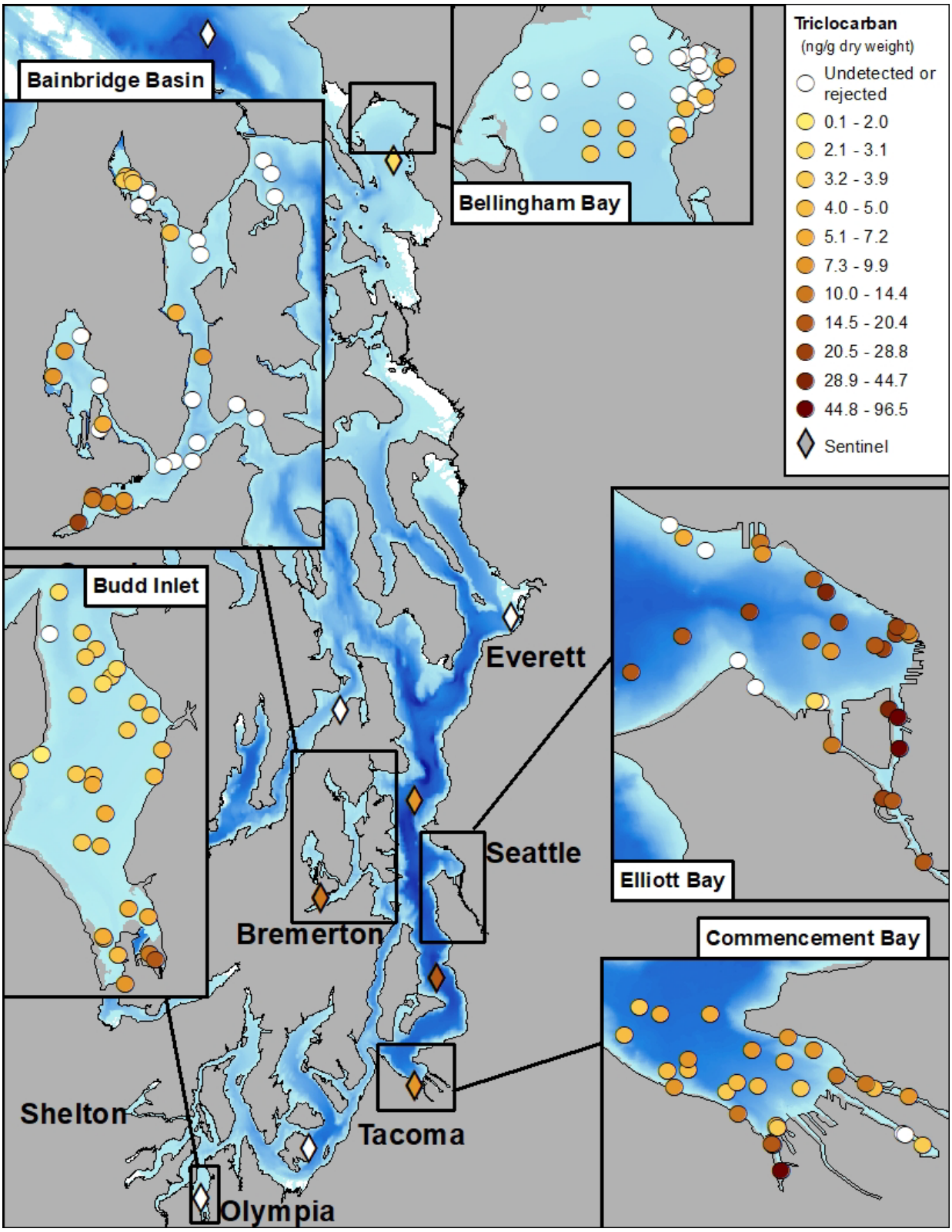


Figure 5. Spatial patterns for triclocarban concentrations (ng/g) detected in sediments from 10 sentinel stations and in 5 urban bays, 2010 through 2019.

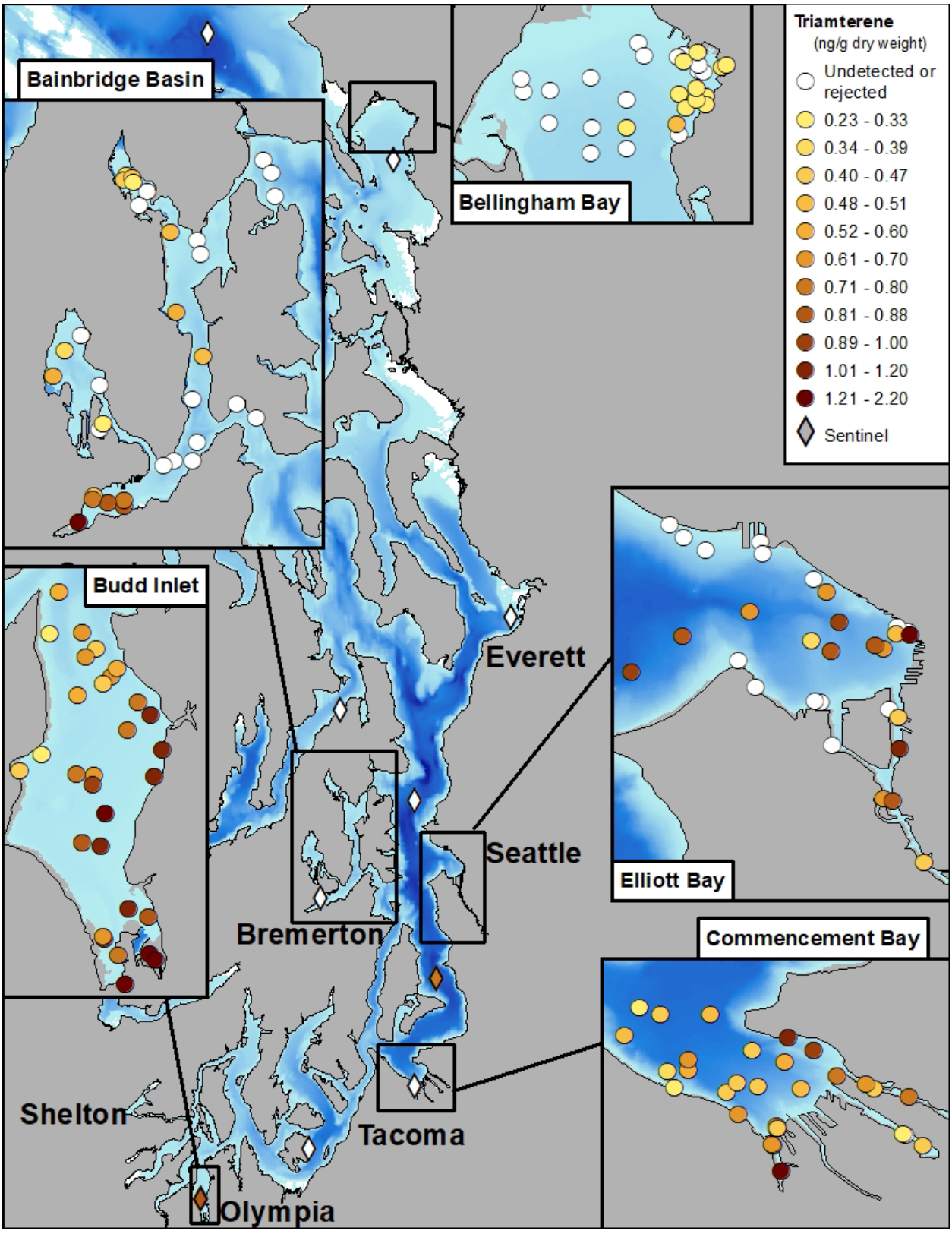


Figure 6. Spatial patterns for triamterene concentrations (ng/g) detected in sediments from 10 sentinel stations and in 5 urban bays, 2010 through 2019.

Comparisons with other PPCP surveys

We conducted a limited literature review examining PPCP measurements from 18 agency reports and peer-reviewed journal articles. Results of these analyses were summarized for nine types of freshwater and estuarine media sampled along the west coast and elsewhere in the United States and were compared to results from our Puget Sound sediment surveys. Media tested included estuarine sediments, surface waters, fish, and mussels; wastewater treatment plant influent, effluent, biosolids, and reclaimed water; groundwater and wells; and surface waters from rivers and streams.

Other Puget Sound surveys

Comparison of the presence of the 43 PPCPs detected in our sediment surveys to those detected in seven surveys conducted in Puget Sound (Table 8) indicated the following:

- Three to 36 PPCPs were detected in the nine additional types of media examined.
- Metformin, a drug used in the control of diabetes, while detected in only six of the samples collected for our sediment surveys, was detected in all 10 types of media.
- Diphenhydramine, triclocarban, and triamterene, the most commonly detected PPCPs in our sediment surveys, were detected in eight, six, and three of the 10 media types, respectively:
    - Diphenhydramine was detected in estuarine sediments, water, fish, and mussels; and in WWTP influent, effluent, biosolids, and reclaimed water; but was absent from ground/well water and freshwater samples.
    - Triclocarban was detected in estuarine sediments and fish; and in WWTP influent, effluent, biosolids, and reclaimed water.
    - Triamterene was detected in our estuarine sediments, and in WWTP effluent and reclaimed water.
- Thirty-six of the 43 PPCPs detected in our sediment surveys were detected in Puget Sound WWTP effluent, while 20, 17, and 18 were detected in WWTP influent, biosolids, and reclaimed water, respectively.
- Fewer than half (19) of the 43 PPCPs detected in our sediment surveys were detected in tissue from Chinook salmon and Pacific staghorn sculpin collected from Puget Sound.
- One-third or fewer (11, 11, 8, and 3) of the 43 PPCPs detected in our sediment surveys were detected in estuarine sediments, mussels, ground water and wells, and freshwater rivers and streams, respectively.

These comparisons highlight the PPCPs that have been most- and least-frequently detected across a variety of environmental media in the Puget Sound watershed. This information may be useful to stakeholders interested in understanding the fate and transport of these chemicals throughout the Sound. Caution must be taken in interpretation of this information, however, as these studies were conducted in different years and locations, with overlapping but not identical analyte lists, and in some cases with differing analytical methods.

Other location surveys

Literature summarizing PPCP monitoring in geographic regions beyond Puget Sound, including Eastern Washington, the Pacific Northwest (i.e., Southwest Washington, Oregon, and British Columbia), California, and other locations throughout the United States was also reviewed. The regions, media types, and surveys in which each of the 43 PPCPs found in our sediments were detected are summarized in Appendices D-1 and D-2.

Diphenhydramine and triclocarban, the two most frequently detected PPCP in our surveys, and five others – caffeine, DEET, erythromycin-H_2O, Miconazole, and sertraline – were detected in Puget Sound, the Pacific Northwest, and California. All seven were detected in WWTP effluent in Puget Sound and the Pacific Northwest, and most were detected in the tissue of bivalves collected in these three regions. The geographically widespread presence of these PPCPs in effluent, biota, and sediments should be considered in future monitoring and environmental management activity.

Table 8. Presence (X, blue cell) of the 43 PPCPs detected in our surveys in comparison to their presence in seven surveys of other environmental media sampled from the Puget Sound watershed.

The three most frequently detected PPCPs in our sediment surveys are in blue text.
White cell = chemical not detected.
Footnotes indicate each comparison survey report. See References section for full citations.

Detected PPCPs	No. detected results in this study	No. media types	Estuarine sediment[a]	Estuarine water[b,c]	Estuarine fish[b]	Estuarine mussels[d]	WWTP influent[e]	WWTP effluent[b,e,f]	Biosolids[e]	Reclaimed water[g]	Ground-water/ wells[f,g,h]	Fresh water[f,h]
Metformin	6	10	X	X	X	X	X	X	X	X	X	X
Diphenhydramine	*124*	*8*	X	X	X	X	X	X	X	X		
Diltiazem	5	7	X	X	X	X	X	X	X			
Caffeine	2	7	X	X	X		X	X			X	X
Sulfamethoxazole	1	7	X	X			X	X	X	X	X	
Ciprofloxacin	1	7	X	X	X	X	X	X	X			
Triclocarban	*111*	*6*	X		X		X	X	X	X		
Fluoxetine	21	6	X		X	X	X	X	X			
Miconazole	21	6	X		X	X	X	X	X			
DEET	4	6	X	X	X			X		X	X	
Ibuprofen	1	6	X				X	X	X		X	X
Sertraline	31	5	X		X	X		X		X		
Amitriptyline	29	5	X		X	X		X		X		
Verapamil	19	5	X		X	X		X		X		
Erythromycin-H2O	11	5	X				X	X	X	X		
Amphetamine	10	5	X	X	X			X			X	
Sulfadimethoxine	3	5	X	X	X		X	X				
Dehydronifedipine	1	5	X				X	X		X	X	
Cotinine	1	5	X				X	X	X	X		
Alprazolam	1	5	X		X			X		X	X	
Azithromycin	17	4	X			X	X	X				
Norverapamil	16	4	X		X			X		X		
Albuterol	2	4	X	X			X	X				
Cocaine	1	4	X		X			X		X		
Ofloxacin	1	4	X				X	X	X			
Codeine	1	4	X				X	X	X			

Detected PPCPs	No. detected results in this study	No. media types	Estuarine sediment[a]	Estuarine water[b,c]	Estuarine fish[b]	Estuarine mussels[d]	WWTP influent[e]	WWTP effluent[b,e,f]	Biosolids[e]	Reclaimed water[g]	Ground-water/ wells[f,g,h]	Fresh water[f,h]
4-Epitetracycline	1	4	X				X	X	X			
Enrofloxacin	1	4	X			X		X	X			
Valsartan	1	4	X	X				X		X		
Triamterene	*105*	*3*	X					X		X		
Benztropine	2	3	X		X			X				
Amlodipine	1	3	X		X			X				
Propoxyphene	1	3	X					X		X		
Desmethyldiltiazem	4	2	X							X		
Simvastatin	2	2	X					X				
Oxytetracyclin	2	2	X						X			
Paroxetine	1	2	X					X				
Carbadox	1	2	X					X				
Norgestimate	1	2	X				X					
Anhydro-chlortetracycline	1	2	X						X			
Sarafloxacin	3	1	X									
Prednisolone	2	1	X									
Anhydro-tetracycline	1	1	X									
Total of the 43 sediment PPCPs detected in other media:			**43**	**11**	**19**	**11**	**20**	**36**	**17**	**18**	**8**	**3**

[a] This study
[b] Meador, 2016
[c] Tian et al., 2019
[d] James et al., 2020
[e] Lubliner et al., 2010
[f] Johnson et al., 2010
[g] Johnson and Marti, 2012
[h] Dougherty et al., 2010

PFAS

Detected PFAS

Over the course of this project, concentrations of 13 to 24 PFAS were measured in sediments from 50, 110, or 243 stations[9]. Nine of these chemicals were detected in one to 34 samples, or 1.4% to 47.9% of 71 detected results. Perfluorooctane sulfonate (PFOS) and perfluorohexanoate (PFHxA) were detected the most frequently, in 34 and 19 samples, respectively (Table 9).

Table 9. Incidence of nine PFAS detected in eight Puget Sound sediment surveys, 2010 through 2019.

Parameter	No. Detected Results	Percent (%) of 71 Detected Results
Perfluorooctane sulfonate (PFOS)	34	47.9
Perfluorohexanoate (PFHxA)	19	26.8
Perfluorobutanoate (PFBA)	7	9.9
Perfluorooctanesulfonamide (PFOSA)	4	5.6
Perfluorooctanoate (PFOA)	2	2.8
Perfluoroundecanoate (PFUnA)	2	2.8
Perfluorodecane sulfonate (PFDS)	1	1.4
Perfluorodecanoate (PFDA)	1	1.4
Perfluorododecanoate (PFDoA)	1	1.4

Incidence and spatial patterns for all detected PFAS and results

Detectable levels of nine (37.5%) of the 13 to 24 chemicals were measured in 71 (1.7%) of 4,189 results generated. The incidence of detected PFAS in each of the eight surveys was low. The highest number of PFAS detected in a survey, four (30.8%), was measured in Commencement Bay in 2014; the lowest was one (4.8%), measured in both Budd Inlet and Pt Gardner/Everett Harbor in 2019. The highest number of detected PFAS results, 19 (4.4%), was found in Budd Inlet in 2019; the lowest, three (0.3%), was measured for the 2019 Puget Sound-wide stations (Table 10).

[9] Dependent on year that the parameter was added to the analyte list

Table 10. Incidence of total PFAS and total results detected for 13 to 24 per- and polyfluoroalkyl substances in eight sediment monitoring surveys in Puget Sound.

Survey Year/ Location	Number of Detected PFAS	Percent (%) of 13 to 24 PFAS	Number of Detected Results	Percent (%) of Total Results	Total Number of Results
2010 Sentinel stations	3	23.1	6	4.6	130
2010 Bellingham Bay	2	15.4	7	1.8	390
2013 Elliott Bay	3	23.1	9	2.3	390
2014 Commencement Bay	4	30.8	8	2.1	390
2015 Bainbridge Basin	3	23.1	19	4.4	429
2019 Budd Inlet	1	4.8	5	0.8	630
2019 Pt Gardner/Everett Harbor	1	4.8	14	2.2	630
2019 Puget Sound-wide	2	8.3	3	0.3	1,200
Overall	**9**	**37.5[a]**	**71**	**1.7**	**4,189**

[a]Calculated for 24 total PFAS

Spatial patterns for the number of PFAS detected at each station differed among the eight surveys (Figure 7 and 8).

Zero to two PFAS were detected at the 10 sentinel stations sampled in 2010. Two PFAS were detected at the Point Pully station, a deep (200m) depositional location between the cities of Seattle and Tacoma. Four stations each had one detected PFAS: (1) Sinclair Inlet station, a shallow location in the Bainbridge Basin, (2) Shilshole station, a deep (200m) depositional location north of Seattle, (3) Bellingham Bay station, a shallow location near Chuckanut Bay, and (4) Strait of Georgia station, over 220 meters deep, near the Canadian border. No PFAS were detected in the remaining five sentinel stations.

Only one PFAS was detected at each of seven stations in the Bellingham Bay survey of 2010. Six of these stations were located near the western urban shoreline, while one was in a deeper station in the middle of the bay.

One station, sampled in 2013 in a deeper, depositional location in outer Elliott Bay, had three detected PFAS. Six other stations, located in a curved path from the Duwamish River to the deeper shoreline and central bay stations, each had one detected PFAS.

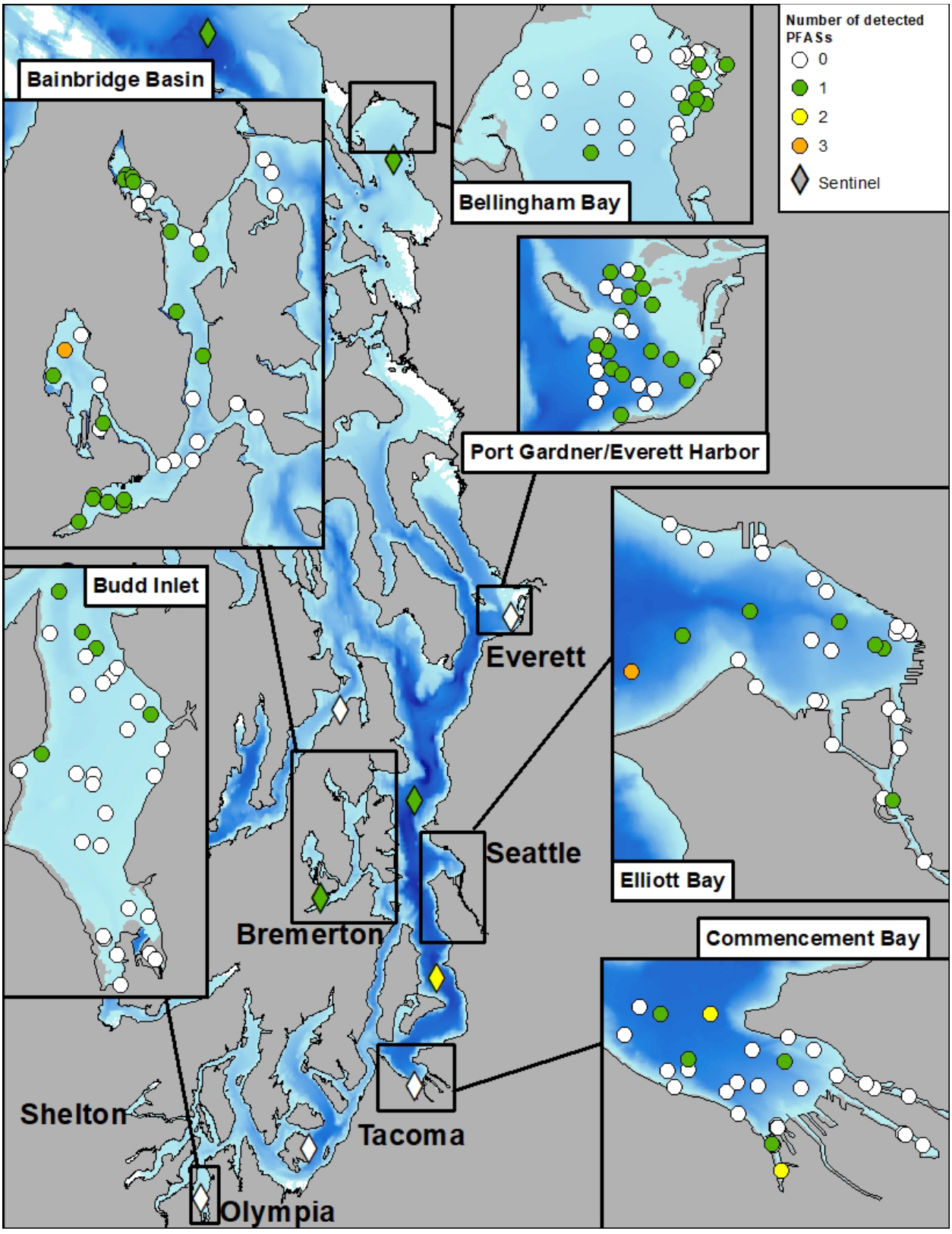


Figure 7. Spatial patterns for the number of PFAS detected in sediments from 10 sentinel stations and in 6 urban bays, 2010 through 2019.

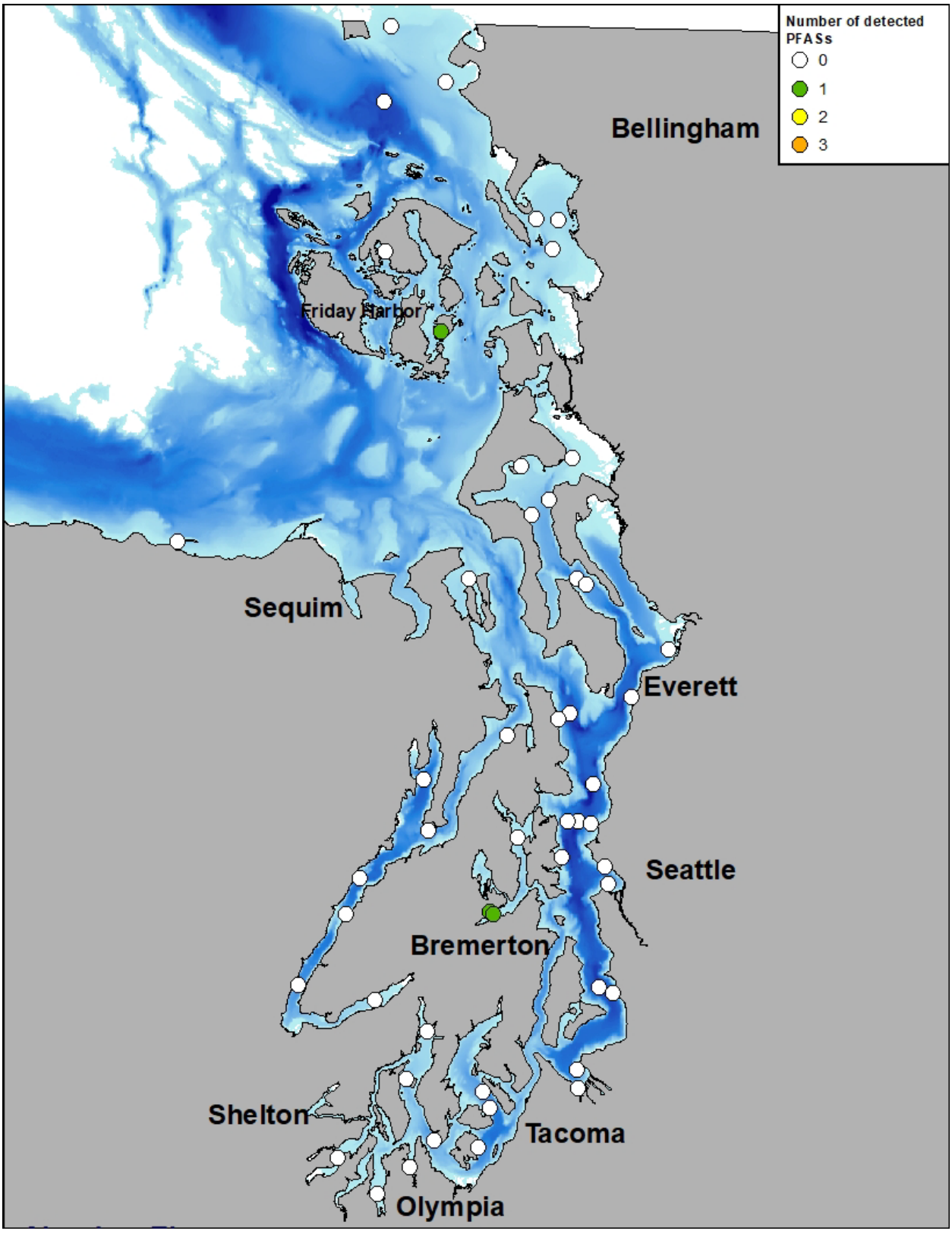


Figure 8. Spatial patterns for the number of PFAS detected in sediments from 50 Puget Sound-wide stations, 2019.

In 2014, two stations in Commencement Bay, one at the head of the Thea Foss WW and one in the northeastern mouth of the bay, had two detected PFAS, while four stations scattered around the bay each had one. Stations in the other waterways did not have any detected PFAS.

In 2015, PFAS were detected in the highest number of stations in the Bainbridge Basin. Sixteen stations located in the terminal inlets, including Sinclair Inlet, Dyes Inlet, Liberty Bay, and Phinney Bay, and in narrow Rich Passage that connects them, had one PFAS each, while a 17th station had three.

In 2019, five stations in the northern portion of Budd Inlet and 14 stations scattered throughout the Pt. Gardner/Everett Harbor sampling area had one detected PFAS each. Three of 50 stations sampled Puget Sound-wide also had one PFAS each: two in Sinclair Inlet near Bremerton, and one in Lopez Sound in the San Juan Islands.

Overall, the occurrence of PFAS, detected in the six urban bays, 10 Long-Term sentinel stations, and 50 Puget Sound-wide stations displayed the following spatial patterns:

- Most stations in our study with detected PFAS had only one detected chemical, but a small number had two or three.
- One to three PFAS were detected in some deep, depositional stations and in some industrialized urban waterways.
- The Bainbridge Basin had the highest number of stations with one detected PFAS each. Most of these were in the terminal inlets.
- Pt. Gardner/Everett Harbor had the second largest number of stations with one detected PFAS. No spatial patterns in the distribution of PFAS were observed.

Two dominant PFAS – incidence, spatial extent and patterns, and summary statistics

The PFAS detected most frequently in the sediments sampled in our surveys were perfluorooctane sulfonate (PFOS) and perfluorohexanoate (PFHxA). Incidence and spatial extent values for these two chemicals are summarized below for each survey, along with all measured and estimated chemical concentrations and ratio-to-RL values. Spatial patterns of the concentrations of PFOS and PFHxA are also depicted below. Values for all nine PFAS are available in Appendix C-2 and spatial patterns for all can be interactively explored by our readers in our companion story map.

Perfluorooctane sulfonate (PFOS)

Perfluorooctane sulfonate was the most frequently detected PFAS in our surveys, occurring in sediments from six of the eight study areas. This chemical was detected in 51.5% of the stations sampled in the Bainbridge Basin, representing 41% of the study area. Detection occurred primarily in terminal inlets and a few in the passage stations. It was also detected in 23.3% of the stations sampled in Elliott Bay, primarily in deeper, depositional stations, and represented 58.6% of the study area. PFOS was detected at 40% of the sentinel stations, and in 4.0%, 3.3%, and 10% of the Puget Sound-wide study area, Bellingham Bay, and Commencement Bay, representing 4.1%, 4.2%, and 15.9% of those survey areas, respectively (Figures 9 and 10, Table 11). Concentrations ranged from a minimum of 0.2 ng/g dry wt at the sentinel stations, and in

Bellingham Bay, Elliott Bay, and Commencement Bay, to a maximum of 1.6 ng/g dry wt in the Bainbridge Basin (Table 12).

Perfluorohexanoate (PFHxA)

While PFHxA was the second-most frequently detected PFAS in our surveys, it was measurable only in sediments collected in the two study areas sampled in 2019. This chemical was detected in 46.7% of the stations sampled in Port Gardner/Everett Harbor, representing 48.2% of the study area. In Budd Inlet, PFHxA was detected in 16.7% of both the samples and study area (Figures 11, Table 11). Concentrations across both study areas ranged from 0.3 ng/g dry wt to 1.8 ng/g dry wt (Table 12).

Table 11. Incidence and spatial extent of the most frequently detected PFAS in eight Puget Sound sediment surveys.

Parameter Year, Sampling Frame	**No. of Stations**	**No. of Stations with Detected Values**	**% of Stations with Detected Values**	**Area (km²)**	**% of Study Area**
Perfluorooctane sulfonate (PFOS)					
2010 Sentinel stations	10	4	40.0	NA[a]	NA
2010 Bellingham Bay	30	1	3.3	1.7	4.2
2013 Elliott Bay	30	7	23.3	15.4	58.6
2014 Commencement Bay	30	3	10.0	3.8	15.9
2015 Bainbridge Basin	33	17	51.5	33.5	41.0
2019 Puget Sound-wide	50	2	4.0	90.1	4.1
Perfluorohexanoate (PFHxA)					
2019 Budd Inlet	30	5	16.7	2.9	16.7
2019 Pt Gardner/Everett Harbor	30	14	46.7	18.4	48.2

[a]NA - The 10 sentinel stations are not part of a sampling frame with a stratified random sampling design, so no spatial extent calculations for the measured PFAS were determined for this survey.

Table 12. Summary statistics for the most frequently detected PFAS in eight Puget Sound surveys.

Parameter Year, Sampling Frame	**Minimum Detected Conc. (ng/g dry wt)**	**Maximum Detected Conc. (ng/g dry wt)**	**Estimated Mean Conc. (ng/g dry wt)[a]**	**Estimated Median Conc. (ng/g dry wt)[a]**	**Minimum RL (ng/g dry wt)[b]**	**Maximum RL (ng/g dry wt)[b]**	**Minimum Conc./RL (ng/g dry wt)[c]**	**Maximum Conc./RL (ng/g dry wt)[c]**
Perfluorooctane sulfonate (PFOS)								
2010 Sentinel stations	0.2	1.5	0.3	0.1	0.2	0.2	1.1	7.9
2010 Bellingham Bay	0.2	0.2	n/a	n/a	0.2	0.2	1.1	1.1
2013 Elliott Bay	0.2	0.5	0.2	0.1	0.2	0.3	1.2	2.0
2014 Commencement Bay	0.2	0.6	0.1	0.0	0.2	0.2	1.0	3.1
2015 Bainbridge Basin	0.3	1.6	0.5	0.3	0.2	0.2	1.5	7.9
2019 Puget Sound-wide	0.5	0.7	n/a	n/a	0.1	0.5	1.2	2.1
Perfluorohexanoate (PFHxA)								
2019 Budd Inlet	0.6	1.7	0.4	0.2	0.4	1.0	1.1	2.2
2019 Pt Gardner/Everett Harbor	0.3	1.8	0.5	0.3	0.2	0.7	1.1	4.2

Conc. = Concentration
RL= Reporting Limit
[a] Estimated by regression on order statistics (ROS) when non-detected data were present (Helsel, 2012)
[b] from all values
[c] from detected values only

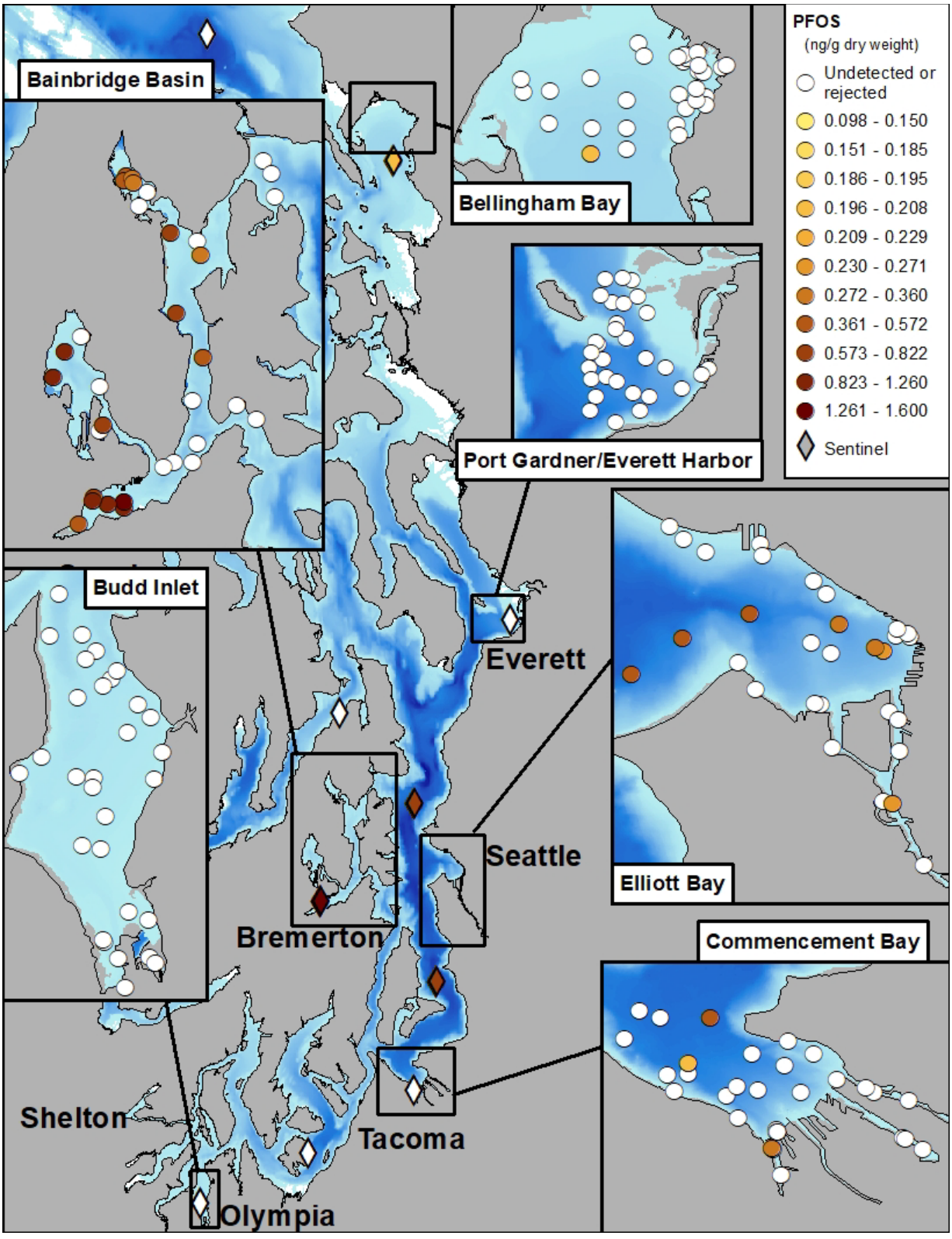


Figure 9. Spatial patterns for perfluorooctane sulfonate (PFOS) concentrations (ng/g) detected in sediments from 10 sentinel stations and in 6 urban bays, 2010 through 2019.

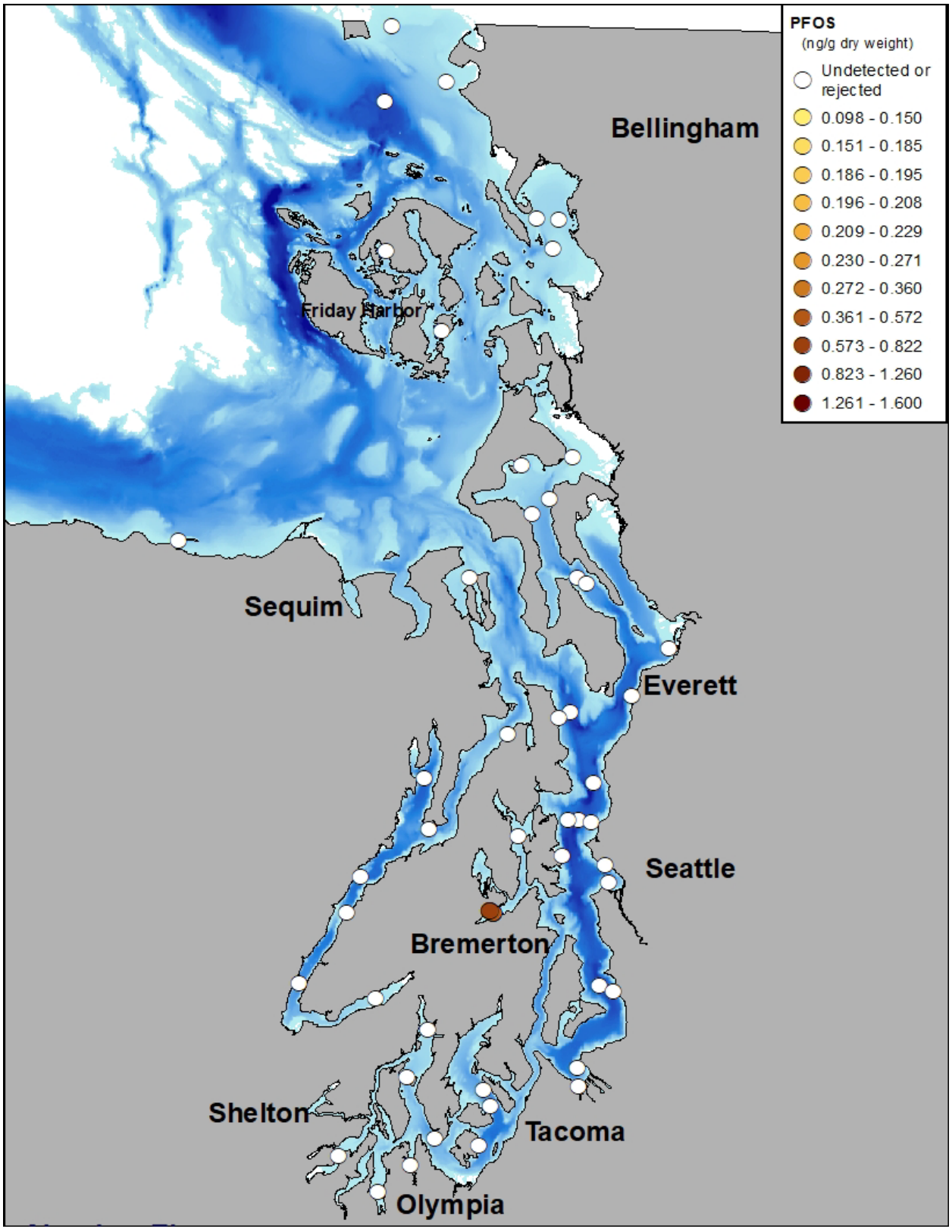


Figure 10. Spatial patterns for perfluorooctane sulfonate (PFOS) concentrations (ng/g) detected in sediments from 50 Puget Sound-wide stations, 2019.

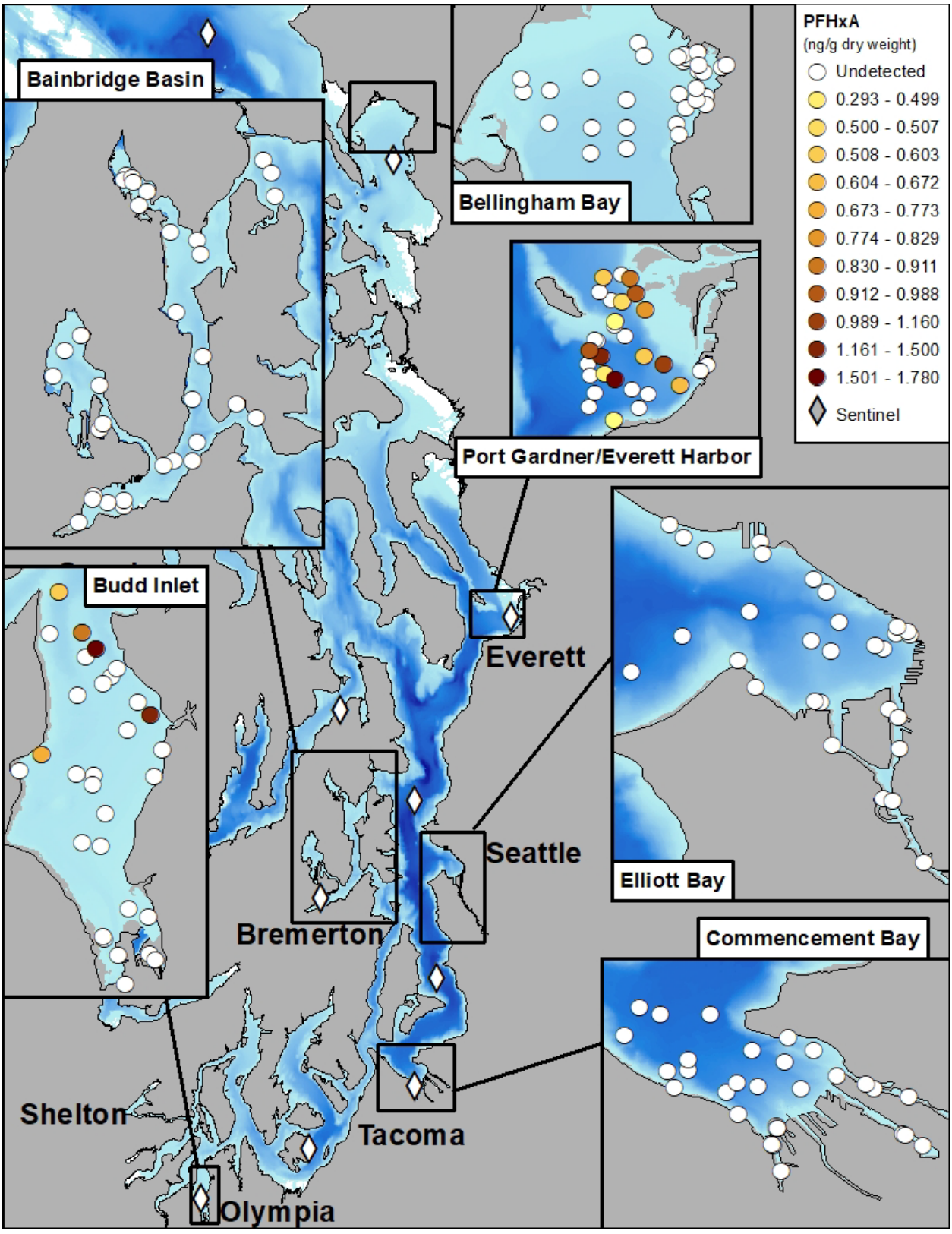


Figure 11. Spatial patterns for perfluorohexanoate (PFHxA) concentrations (ng/g) detected in sediments from Budd Inlet and Port Gardner/Everett Harbor, 2019.

Comparison between years

While each of the urban bay survey areas was sampled only once during 2010 through 2019, the 10 sentinel stations sampled in 2010 were resampled for PFAS in 2019, allowing for between-year comparison of results. PFAS were detected in sediments at five of the 10 sentinel stations in 2010, including PFBA at the station in the Strait of Georgia; PFOS at the stations in Bellingham Bay, Shilshole, Sinclair Inlet, and Point Pully (3-Tree Point); and PFOSA at the Point Pully station. PFOS was detected again in 2019 at the Sinclair Inlet station, at less than half the concentration found in 2010. This was the only PFAS detected at any of the 10 sentinel stations in 2019. Laboratory RLs for these chemicals were higher in 2019 than in 2010, and may explain some of the 2019 undetected results (Table 13).

Table 13. Comparison of detected PFAS concentrations in five sentinel stations sampled in 2010 and 2019.

Sampling year (lab):		2010 (SGS AXYS)			2019 (MEL)		
Sentinel station	PFAS	No. detected	Conc. (ng/g dry wt)	RL (ng/g dry wt)	No. detected	Conc. (ng/g dry wt)	RL (ng/g dry wt)
St. of Georgia	PFBA	1	0.22	0.09	0	--	0.24
Bellingham Bay	PFOS	1	0.21	0.19	0	--	0.40
Shilshole	PFOS	1	0.60	0.20	0	--	0.36
Sinclair Inlet	PFOS	1	1.50	0.19	1	0.67	0.32
Point Pully	PFOS	1	0.64	0.19	0	--	0.47
Point Pully	PFOSA	1	0.18	0.09	0	--	0.47

Conc = concentration RL = reporting limit

Comparison with other PFAS surveys

We conducted a limited literature review examining PFAS measurements from 14 agency reports and peer-reviewed journal articles. Results of these analyses were summarized for 13 types of freshwater and estuarine media sampled along the west coast and elsewhere in the United States and were compared to results from our Puget Sound sediment surveys. Media tested included estuarine sediments, surface waters, fish, bivalves, eggs of Ospreys and Double-crested cormorants, and marine mammals; WWTP effluent; stormwater and sediments in stormwater catch basins; groundwater and wells; and surface waters, sediments, and fish from rivers and streams.

Other Puget Sound surveys

Comparison of the presence of the nine PFAS detected in our sediment surveys to those detected in 10 surveys conducted in Puget Sound (Table 14) indicated the following:

- One to six PFAS were detected in the nine additional types of media examined.
- PFOS, the most commonly detected PFAS in our sediment surveys, was detected in nine of the 10 media types examined. This included marine and estuarine sediment, water, and fish; osprey eggs; WWTP effluent; groundwater and wells; and freshwater sediments, water, and

fish. This chemical was not detected in the surveys of marine and estuarine mussels and Olympia oysters.

- PFHxA, the second-most commonly detected PFAS in our sediment surveys, was detected in six of the 10 media types examined. This included marine and estuarine sediment, water, and fish; WWTP effluent; ground water and wells; and freshwater.
- PFDA, found in only one sample in our surveys, was detected in seven of the 10 media types examined.
- PFOSA and PFOA were detected in six, PFBA and PFUnA in five, PFDoA in four, and PFDS in one of the 10 media types examined.

As with the PPCP comparisons, these PFAS comparisons highlight the PFAS that have been most- and least-frequently detected across a variety of environmental media in the Puget Sound watershed. They may provide insight regarding the fate and transport of these chemicals throughout the Sound. Again, caution must be taken in interpretation of this information, as these studies were conducted in different years and locations, with overlapping but not identical analyte lists, and in some cases with differing analytical methods.

Other location surveys

Again, literature summarizing PFAS monitoring in geographic regions beyond Puget Sound, including Eastern Washington, the Pacific Northwest (i.e., Southwest Washington, Oregon, and British Columbia), California, and other locations throughout the United States, was reviewed. The regions, media types, and surveys in which each of the nine PFAS found in our sediments were detected are summarized in Appendix D-3 and D-4.

It is notable in the surveys examined that six of the PFAS detected in our Puget Sound sediment surveys, including PFOS, PFOA, PFDA, PFUnA, PFDoA, and PFOSA, were also detected in sediments in a California survey. Additionally, PFOS, PFDA, PFUnA, and PFDoA were detected in osprey or cormorant eggs sampled in Puget Sound, the Pacific Northwest, Eastern Washington, and California. PFOS and PFDA were also detected in water samples from rivers and streams in the same four regions. The geographically widespread presence of these PFAS in freshwater, biota, and sediments should be considered in future monitoring and environmental management activity.

An additional resource for PFAS survey information is the extensive review of data collected from various ecosystems and environmental media sampled throughout Washington State and summarized in Appendices 5 and 7 of Ecology's Per- and Polyfluoroalkyl Substances Chemical Action Plan (Ecology, 2021). Knowing which PFAS are detected in other parts of Washington's ecosystem provides perspective on the fate and transport of these chemicals and their potential for bioaccumulation in Puget Sound and State-wide.

Table 14. Presence (X, blue cell) of the nine PFAS detected in our surveys in comparison to their presence in 10 surveys of other environmental media sampled from the Puget Sound watershed.

The two most frequently detected PFAS in our sediment surveys are in blue text.
White cell = chemical not detected.
Footnotes indicate author(s) and date of the comparison survey reports. See References section for full citations.

PFAS detected in this study	No. detected results in this study	No. media types	Marine/ estuarine sediment [a]	Marine/ estuarine water [b,c]	Marine/ estuarine fish [d,e]	Marine/ estuarine mussels, oysters [f]	Osprey eggs[g]	WWTP effluent [e,g,h,i]	Ground water/ wells [d]	Fresh-water [g,h]	Fresh water sedi-ment [j]	Fresh-water fish [g,h,j,k]
PFOS	*34*	*9*	X	X	X		X	X	X	X	X	X
PFDA	1	7	X	X	X		X	X		X		X
PFHxA	*19*	*6*	X	X	X			X	X	X		
PFOSA	4	6	X		X	X		X			X	X
PFOA	2	6	X	X				X	X	X		X
PFBA	7	5	X	X	X			X		X		
PFUnA	2	5	X		X		X				X	X
PFDoA	1	4	X				X				X	X
PFDS	1	1	X									
Total of the 9 sediment PFAS detected in other media:			**9**	**5**	**6**	**1**	**4**	**6**	**3**	**5**	**4**	**6**

[a] This study
[b] Tian et al., 2019
[c] Dinglasan-Panlilio et al., 2014
[d] Ecology, 2020
[e] Meador et al., 2016
[f] James et al., 2020
[g] Mathieu and McCall, 2017
[h] Furl and Meredith, 2010
[i] Ecology and Herrera, 2010
[j] Mathieu, 2013
[k] Johnson and Friese, 2012

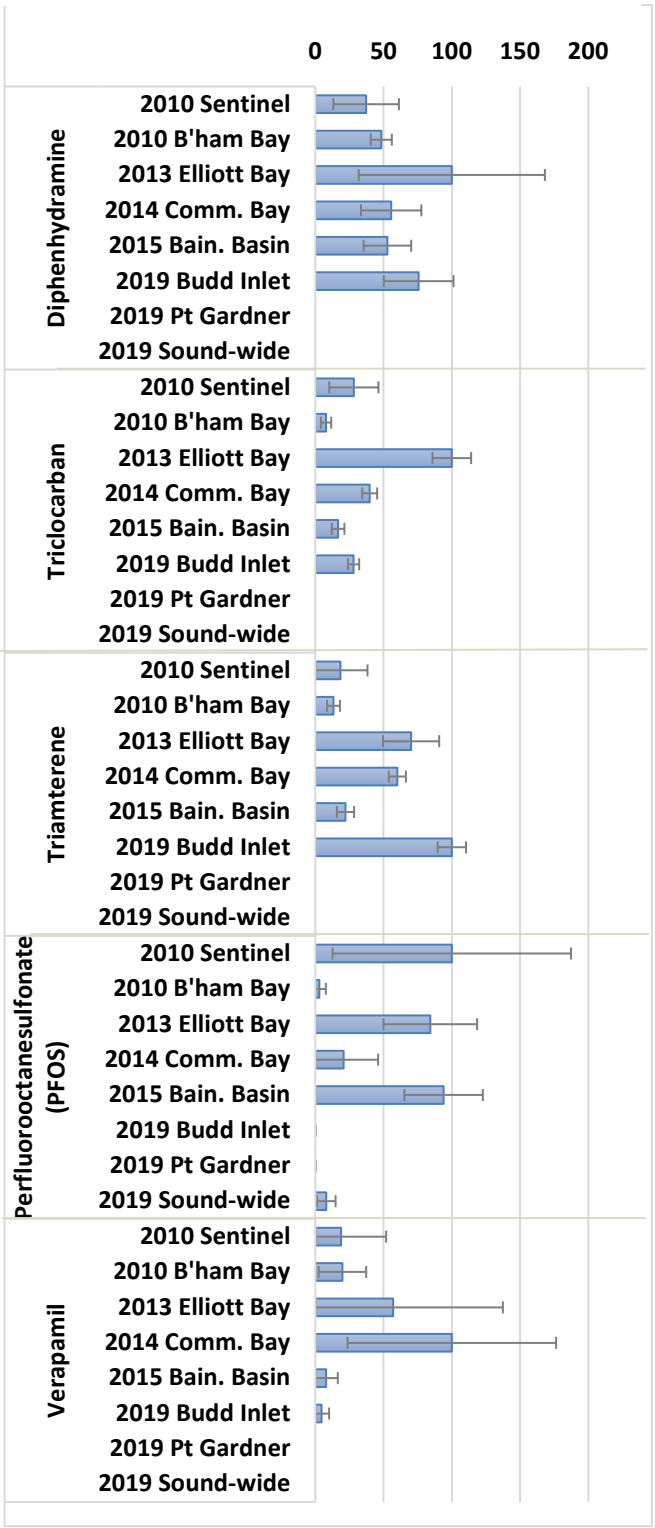
Concentration and spatial extent comparisons among study areas

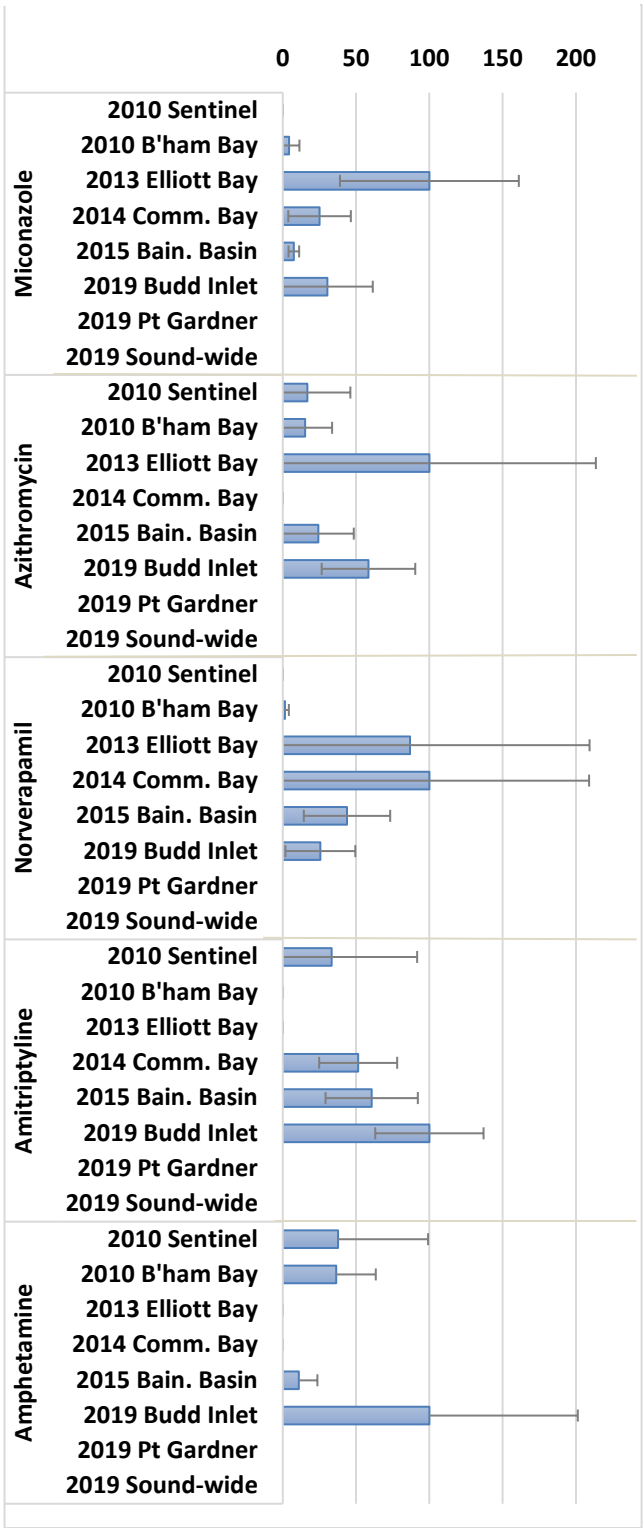
Results for all chemicals, standardized by the maximum detected value across all study areas, are summarized in Appendix E. Among-survey differences for these standardized detected concentrations can be seen for many of the chemicals. As indicated earlier in this report, the reporting limits and definitions of undetected differed among surveys and analytical labs. Comparisons among surveys and between labs should be made with caution due to these differences.

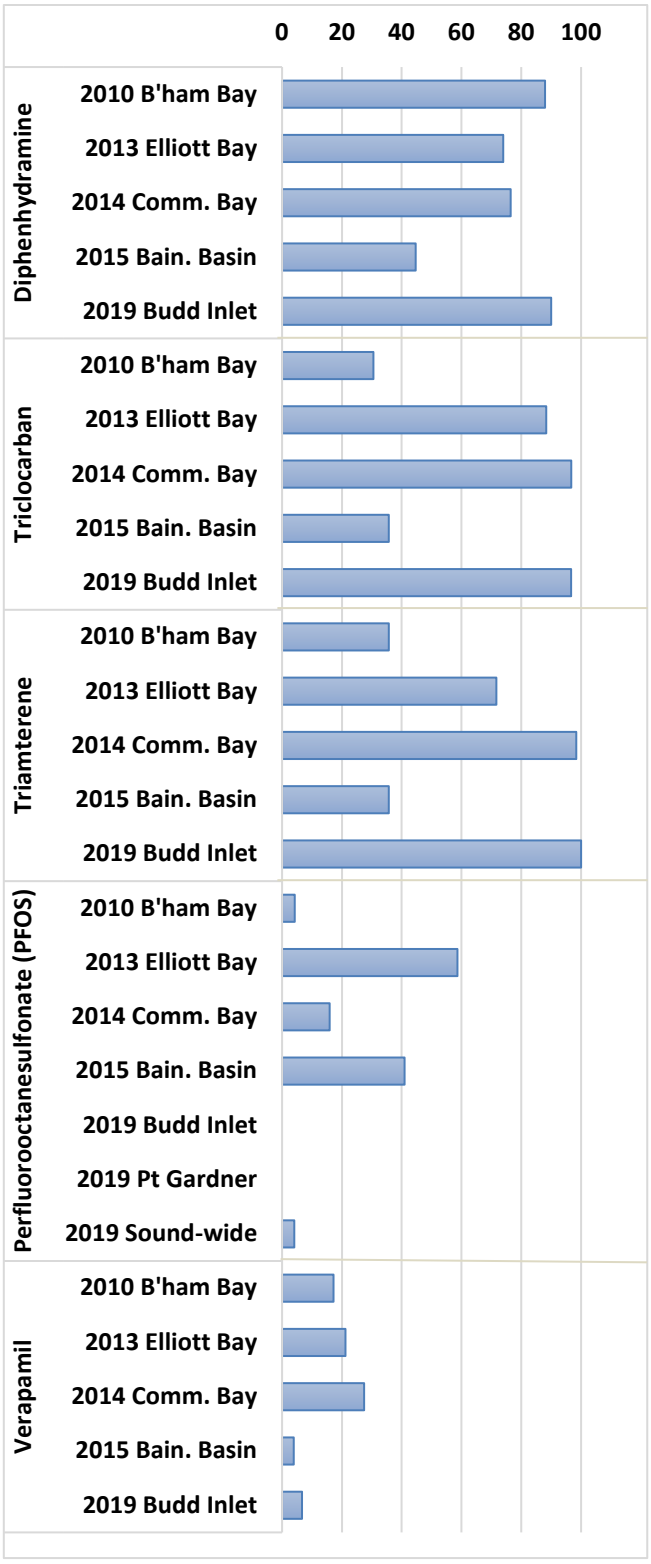
The standardized percent of maximum weighted mean concentrations for all detected chemicals are summarized in Figure 12 for the 10 chemicals that were found in the most surveys and at the highest number of stations. These concentrations are summarized in Appendix F for all detected chemicals. Additionally, the percent of the study area represented by each chemical is displayed in Figure 13 for the same 10 chemicals and in Appendix F for all detected chemicals.

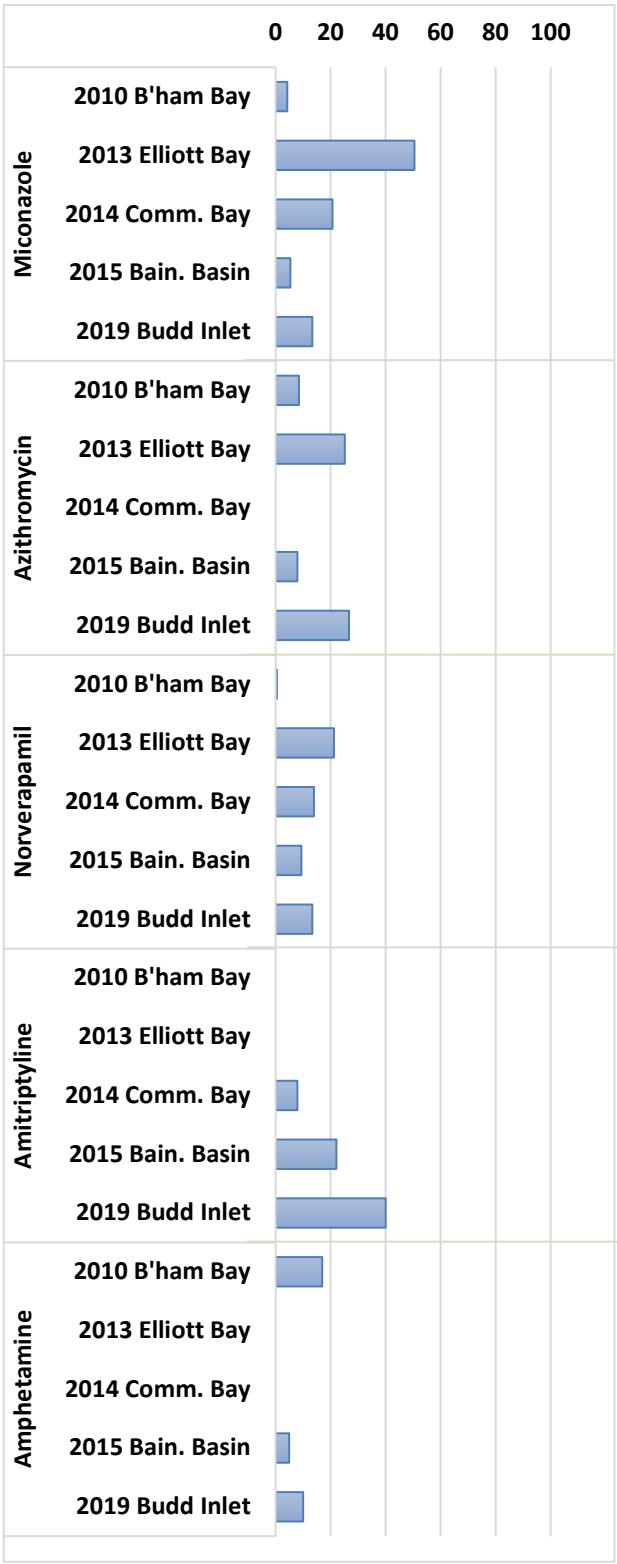
Comparison of both sets of standardized values for the 43 PPCPs and nine PFAS detected in our surveys indicated that:

- Diphenhydramine, triclocarban, and triamterene were detected at the 10 sentinel stations and in all five bays in which they were tested, with the highest combined spatial area. Verapamil, Miconazole, and norverapamil also occurred in all five bays, but covered a lower percent of the study areas.
- Of the 24 PFAS measured, PFOS had the highest percent of maximum weighted mean and greatest spatial extent, having been measured at six of eight sampling surveys.
- Occurrence, relative concentrations, and spatial extent values for each chemical varied among study areas. No clear patterns emerged, giving each study area a unique chemical signature.


Percent of Maximum Weighted Mean
0
50
100
150
200
Diphenhydramine
Triclocarban
Triamterene
Perfluorooctanesulfonate (PFOS)
Verapamil
2010 Sentinel
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
2019 Pt Gardner
2019 Sound-wide


Percent of Maximum Weighted Mean
0
50
100
150
200
Miconazole
Azithromycin
Norverapamil
Amitriptyline
Amphetamine
2010 Sentinel
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
2019 Pt Gardner
2019 Sound-wide


Percent of study area
0
20
40
60
80
100
Diphenhydramine
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
Triclocarban
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
Triamterene
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
Perfluorooctanesulfonate (PFOS)
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
2019 Pt Gardner
2019 Sound-wide
Verapamil
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet


Percent of study area
0
20
40
60
80
100
Miconazole
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
Azithromycin
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
Norverapamil
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
Amitriptyline
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet
Amphetamine
2010 B'ham Bay
2013 Elliott Bay
2014 Comm. Bay
2015 Bain. Basin
2019 Budd Inlet

Summary and Conclusions

Surface sediments collected from 2010 through 2019 for the Sediment Program were analyzed to determine the concentrations of 118 PPCPs and 24 PFAS, CECs in Puget Sound. Stations sampled included 50 that were located throughout our Puget Sound-wide sampling frame, 10 of which are considered sentinel stations sampled since the inception of the program. Additionally, 30 to 33 stations were also sampled from each of six routinely monitored urban bay sampling frames.

Overall, occurrence and concentrations for both the PPCPs and PFAS was low, and spatial distribution of detected chemicals differed from one sampling frame to the next, reflecting unique chemical "signatures" in each bay.

PPCPs in Puget Sound sediments

Forty-three (36.4%) of the 118 PPCPs were detected in 571 (3.0%) of 19,228 results generated. The detected chemicals belonged to a diverse array of classes with over 20 different biochemical functions. The highest number of PPCP analytes detected in a survey, 24 (20.3%), was measured in the Bainbridge Basin in 2015; the lowest was seven (5.9%), measured at the 10 sentinel stations in 2010. The highest number of detected PPCP results, 159 (4.5%), was found in Budd Inlet in 2019; the lowest, 18 (1.5%), was again at the 2010 sentinel stations.

Three PPCPs – diphenhydramine (an antihistamine), triclocarban (an antibiotic), and triamterene (a diuretic) – were detected most frequently in the sediments sampled. Spatial patterns of detected PPCPs indicated that they tended to occur in depositional locations with fine-grained sediments, at the heads of bays, and along portions of shoreline likely near discharge sources. More chemicals were detected in Commencement Bay, the Bainbridge Basin, and Budd Inlet than in the other sampled locations.

PFAS in Puget Sound sediments

Detectable levels of nine (37.5%) of the 13 to 24 PFAS were measured in 71 (1.7%) of 4,189 results. The highest number of PFAS analytes detected in a survey, four (30.8%), was measured in Commencement Bay in 2014; the lowest was one (4.8%), measured in both Budd Inlet and Pt Gardner/Everett Harbor in 2019. The highest number of detected PFAS results, 19 (4.4%), was found in Budd Inlet in 2019; the lowest, three (0.3%), was measured for the 2019 Puget Sound-wide stations.

Two PFAS – PFOS and PFHxA – were detected most frequently in our sediment samples. PFOS was found in depositional locations with fine-grained sediments, in some industrialized urban waterways, and in terminal inlets, and was not detected in the Budd Inlet or Pt Gardner/Everett Harbor sampling frames. PFHxA was detected only in Budd Inlet and Pt Gardner/Everett Harbor, but with no discernible spatial patterns.

PFAS were measured at our 10 sentinel stations in 2010 and again in 2019. At the five stations in which PFAS were measured in 2010, PFOS was the only chemical detected again in 2019, at our monitoring station in Sinclair Inlet. However, higher reporting limits in the 2019 analysis

hampered our ability to compare detections between the two sampling periods. Further monitoring is needed to determine whether any change over time is occurring in PFAS concentrations throughout Puget Sound.

Comparisons with other surveys

Comparison of our PPCP and PFAS sediment results to those in surveys of other types of environmental media in Puget Sound, the Pacific Northwest, Eastern Washington, California, and elsewhere in the United States indicates a number of chemicals that are common across media and locations. In Puget Sound surveys, the diabetes drug metformin was detected in all 10 types of media samples, while diphenhydramine and triclocarban, the most commonly detected PPCPs in our sediment surveys, were detected in eight and six of the 10, respectively. Across geographic locations, diphenhydramine and triclocarban, along with caffeine, DEET, erythromycin-H_2O, Miconazole, and sertraline, were detected in Puget Sound, the Pacific Northwest, and California WWTP effluent, bivalves, and sediments.

Our literature review indicated that PFOS, the most commonly detected PFAS in our surveys, was detected in nine of the 10 media types examined in Puget Sound. PFOS, along with PFOA, PFDA, PFUnA, PFDoA, and PFOSA were all detected in sediments in Puget Sound and California surveys, while PFOS, PFDA, PFUnA, and PFDoA were detected in osprey or cormorant eggs sampled in all of the regions, while PFOS and PFDA were detected in freshwater samples from all regions.

Spatial patterns and extent

Finally, when comparing the spatial extent of concentrations for all detected chemicals standardized across our Puget Sound survey areas, we determined that diphenhydramine, triclocarban, triamterene, and PFOS had the highest combined spatial area values. Occurrence, relative concentrations, and spatial extent values for each chemical varied among study areas. No clear patterns emerged, reflecting a unique chemical signature in each study area.

The PPCP and PFAS data from our sediment surveys contribute to the growing baseline of information on the spatial extent and spatial patterns of these CECs in Puget Sound. These data lay a foundation for understanding the sources, transport patterns, and fate of these chemicals in discharged waste, sediments, water, and biota throughout the Puget Sound ecosystem.

Recommendations

Recommendations for future work on PPCPs and PFAS in Puget Sound include:

- Measurement of an equivalent suite of PPCPs in sediment collected from the Port Gardner/ Everett Harbor sampling frame as part of our Sediment Program's annual sampling, creating baseline data for this area. It is the only routinely-sampled study area for our program that has not yet been tested for PPCPs.
- Continued monitoring of PPCP and PFAS analytes prioritized by the Puget Sound Ecosystem Monitoring Program's Toxics Workgroup. This workgroup is currently conducting a CEC prioritization exercise to inform and guide future environmental monitoring and management in the Puget Sound watershed.
- Development of coordinated multimedia surveys for these chemicals in Puget Sound that simultaneously examine the same suites of PPCPs and PFAS in sediments, ambient water, and biota from the same locations. Such information would better inform stakeholders on the fate and transport of these chemicals.
- Development of PPCP and PFAS biotic thresholds and endpoints for sediments for use in future regulatory work and status and trends monitoring. This includes current efforts by Ecology's Toxics Cleanup Program staff to develop cleanup levels for a limited number of PFAS.
- Development of educational information to ensure that regional and local consumers and stakeholders are aware of products containing these chemicals, consider reduction of their use, and are knowledgeable of and use the most appropriate disposal and waste-handling methods available.

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Glossary, Acronyms, Abbreviations

Glossary

Association of Official Analytical Chemists (AOAC) –The analytical methods adopted by the Association of Official Analytical Chemists are used by government agencies concerned with the analysis of fertilizers, foods, feeds, pesticides, drugs, cosmetics, hazardous substances, and other materials related to agriculture, health and welfare, and the environment.

AXYS, SGS-AXYS – AXYS Analytical Services Ltd, Sydney, British Columbia.
Renamed as SGS AXYS Analytical Services Ltd in 2016.

Generalized Random Tessellation Stratified (GRTS) **–** Multi-density category-weighted design or intensive equally-weighted survey design. Two survey designs developed by Tony Olsen, EPA, for the Sediment Program's Regional and Urban Bays monitoring efforts beginning in 2004.

Holding time – The maximum times that samples may be held prior to analyses and still be considered valid or not compromised.

Incidence – In this study, the number and percent of detected chemical values at a station or in a study area.

Quick, Easy, Cheap, Effective, Rugged, Safe (QuEChERS) – A chemistry extraction method used for our studies.

Sampling frame – A defined geographic area sampled by the Sediment Program. We have 6 Urban Bay sampling frames and a Puget Sound-wide sampling frame.

Sentinel stations – 10 sediment monitoring stations sampled by the Sediment Program. These stations have differing sediment quality characteristics and unique benthic invertebrate assemblage characteristics. They have been sampled annually since the inception of the program in 1989 to look for changes over time in their benthic assemblages.

Simple stratified random sampling design (SSR) **–** The Sediment Program's survey design developed for surveys conducted in partnership with Ed Long and NOAA's National Status and Trends Sediment Monitoring Program from 1997 – 1999.

Spatial extent – In this study, the area (km^2) or percent of study area that is estimated to be associated with a measured environmental parameter.

Acronyms and Abbreviations

AOAC	(see Glossary above)
AXYS	(see Glossary above)
CECs	Chemicals of Emerging Concern
Ecology	Washington State Department of Ecology
EAP	Environmental Assessment Program (Washington State Department of Ecology)
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency

GRTS	(see Glossary above)
MEL	Manchester Environmental Laboratory, Manchester, WA
MSMT	Marine Sediment Monitoring Team
PFAS	Per- and polyfluoroalkyl substance/substances
PPCP/PPCPs	Pharmaceuticals and personal care product/products
QuEChERS	(see Glossary above)
RL	Reporting limit
Sediment Program	– Puget Sound Sediment Monitoring Program
SGS-AXYS	(see Glossary above)
SSR	(see Glossary above)
WW	Waterway
WWTP	Wastewater Treatment Plant

Appendices

Appendix A. Case narratives, data packages, and quality review reports for all PPCP and PFAS chemical analyses conducted by AXYS/SGS-AXYS and MEL.

Appendix B. Holding time and container type comparisons, 2020 PFAS analyses.

Appendix C. Incidence, spatial extent, and summary statistics for concentrations of 43 PPCPs and 9 PFAS detected in Puget Sound surveys, 2010 through 2019.

C-1. 43 detected PPCPs

C-2. 9 detected PFAS

Appendix D. Comparison with PPCP and PFAS surveys in other aquatic ecosystems from the west coast and elsewhere in the United States.

D-1. Geographic regions in the western United States in which the 43 PPCPs detected in sediments from our surveys were measured.

D-2. Presence of 43 PPCPs detected in sediments from our surveys as compared with their presence in 32 surveys of other environmental media sampled from the Puget Sound watershed.

D-3. Geographic regions in the western United States in which the nine PFAS detected in sediments from our surveys were measured.

D-4. Presence of nine PFAS detected in sediments from our surveys as compared with their presence in 32 surveys of other environmental media sampled from the Puget Sound watershed.

Appendix E. Scatterplots of chemical concentrations, standardized as the percent of the maximum detected value, for 43 PPCPs and 9 PFAS detected in Puget Sound surveys, 2010 through 2019.

Appendix F. Chemical concentrations, standardized as the percent of maximum weighted mean, and the spatial extent (percent of the study area) of detections, for 43 PPCPs and 9 PFAS detected in Puget Sound surveys, 2010 through 2019.