Water Quality Assessment and Monitoring Study: Contaminants of Emerging Concern

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King County

Department of Natural Resources and Parks
Water and Land Resources Division
Science and Technical Support Section
King Street Center, KSC-NR-0600
201 South Jackson Street, Suite 600
Seattle, WA 98104
206-477-4800 TTY Relay: 711
www.kingcounty.gov/EnvironmentalScience

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Water Quality Assessment and Monitoring Study: Contaminants of Emerging Concern

Prepared for:

Wastewater Treatment Division
Department of Natural Resources and Parks

Submitted by:

Richard Jack and Martin Grassley
King County Water and Land Resources Division
Department of Natural Resources and Parks

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Department of
Natural Resources and Parks
Water and Land Resources Division

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Table of Contents

Figures

Tables

EXECUTIVE SUMMARY

King County completed a study of the presence in the surface waters of organic chemicals originating from pharmaceutical and personal care products. The study was prepared as part of a Water Quality Assessment and Monitoring Study, undertaken to explore ways to optimize water quality improvements in the three waterbodies where the County is planning combined sewer overflow (CSO) control projects—Lake Union/Ship Canal, Elliott Bay, and the Duwamish Estuary.

Background

King County updates its CSO control plan about every five years. Before each update, the Wastewater Treatment Division reviews its entire CSO Control Program against conditions that have changed since the last update. In September 2012, the King County Council passed Ordinance 17413 approving an amendment to King County's long-term CSO control plan. The amended plan includes nine projects to control the County's remaining 14 uncontrolled CSO locations in Lake Union/Ship Canal, Elliott Bay, and the Duwamish Estuary by 2030 to meet the Washington State standard of no more than one overflow per year on average. The recommended projects involve construction of underground storage tanks, green stormwater infrastructure, and/or wet weather treatment facilities.

Ordinance 17413 also calls for completion of a Water Quality Assessment and Monitoring Study (assessment) to inform the next CSO control plan update due to the Washington State Department of Ecology in 2018. The ordinance specified that the assessment answer the following questions:

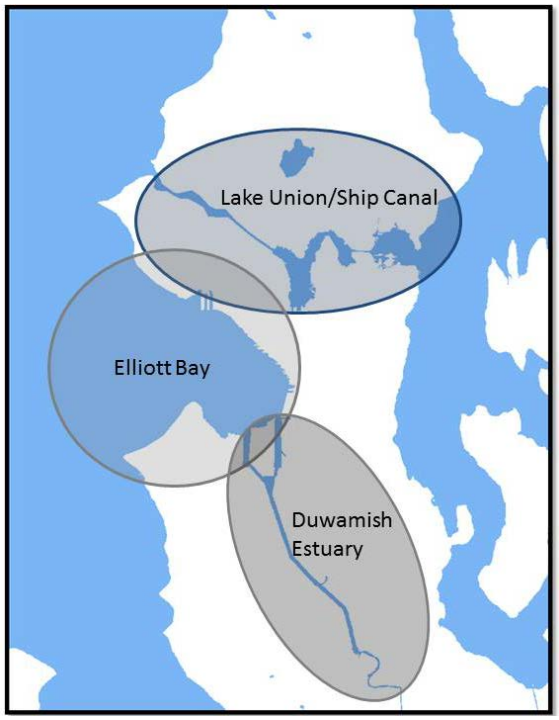
1. What are the existing and projected water quality impairments in receiving waters (waterbodies) where King County CSOs discharge?
2. How do county CSOs contribute to the identified impairments?
3. How do other sources contribute to the identified impairments?
4. What activities are planned through 2030 that could affect water quality in the receiving waters?

Three additional questions will be addressed by the County's CSO planning team based partly on the results of the assessment:

5. How can CSO control projects and other planned or potential corrective actions be most effective in addressing the impairments?
6. How do various alternative sequences of CSO control projects integrated with other corrective actions compare in terms of cost, schedule, and effectiveness in addressing impairments?
7. What other possible actions, such as coordinating projects with the City of Seattle and altering the design of planned CSO control projects, could make CSO control projects more effective and/or help reduce the costs to WTD (King County Wastewater Treatment Division) and the region of completing all CSO control projects by 2030?

Study Areas

The Water Quality Assessment and Monitoring Study focused on three study areas: Lake Union and the Lake Washington Ship Canal (Lake Union/Ship Canal), Elliott Bay, and the Duwamish Estuary.



- Lake Union/Ship Canal includes the waters flowing out of Lake Washington into the Montlake Cut, Portage Bay, Lake Union, the Fremont Cut, and Salmon Bay upstream of the Hiram M. Chittenden Locks (Ballard Locks). The Locks separate the salt water of Puget Sound from the fresh water of Lake Union.
- Elliott Bay encompasses the area east of a line drawn between Duwamish Head in West Seattle and Magnolia Bluff near Smith Cove, including the downtown Seattle waterfront. This area, also known as Inner Elliott Bay, is open to Outer Elliott Bay and Puget Sound to the west and receives freshwater inflows from the Duwamish Estuary.
- The Duwamish Estuary includes the East, West, and Lower Duwamish waterways. The estuary receives freshwater flows from the Duwamish River and Green River watershed. The Duwamish Estuary is influenced by tidal exchange with Elliott Bay.

Study Approach and Results

A growing body of research is revealing the persistence and ubiquity of contaminants originating from prescription drugs, drug residues, and personal care products. These contaminants are called contaminants of emerging concern (CECs) to distinguish them from more conventional pollutants (such as motor oil and dissolved copper, which have been studied for some time). Advancing analytical techniques allow researchers and chemists to monitor CECs in the environment. Some CECs like DEET (mosquito repellant) have been in existence for decades but have only recently become of concern as the result of development of analytical methods to measure them in the environment. Few regulatory standards are in place for most CECs.

The primary goal of this study was to assess the presence or absence of CECs in ambient waters in the study areas. A secondary goal was to describe the relative magnitude of the detected CECs. A total of 51 samples were collected across multiple depths within the three study areas and were analyzed for 140 CECs including the following:

- 100 pharmaceuticals

- 12 pharmaceutical metabolites
- 20 pyrethroid insecticides
- DEET
- Cocaine and nicotine (and their metabolites)

As shown in Figure ES-1, the analysis detected 26 different CECs out of the 140 that were analyzed. For the CECs found, the frequency of detection ranged from 1 out of 51 samples (1.9 percent) to up to 48 of the 51 samples (94.1 percent).

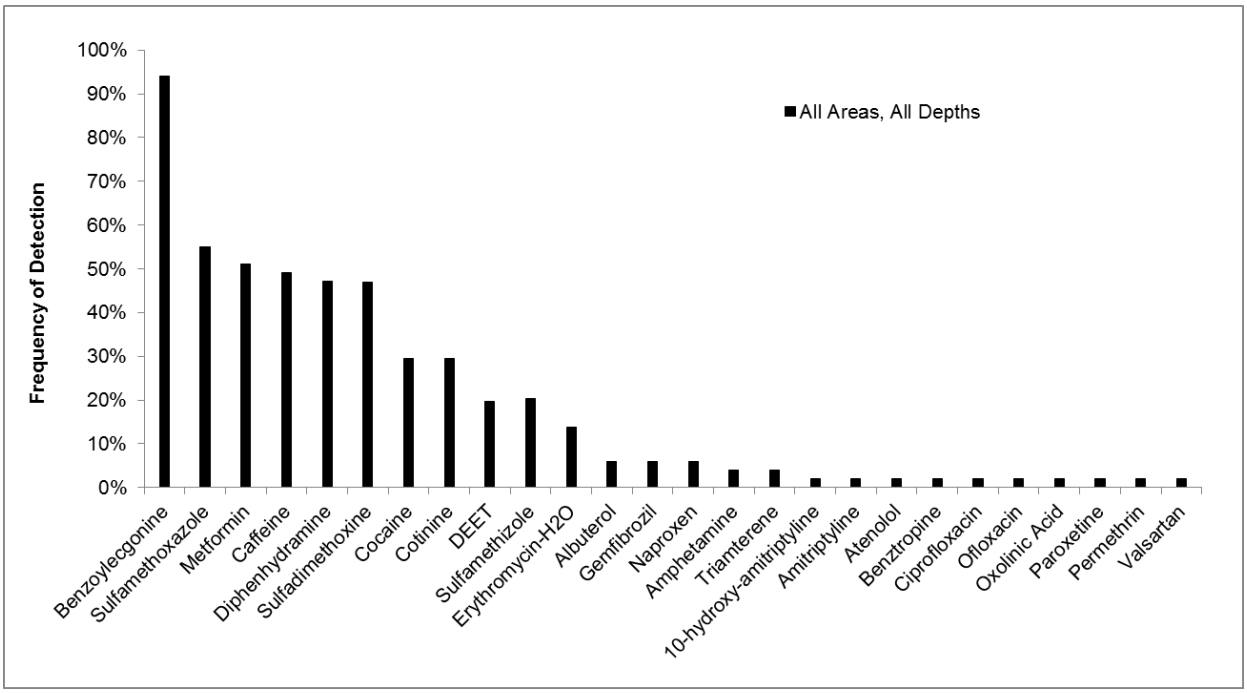


Figure ES-1. Frequency of detection of CECs in Lake Union/Ship Canal, Elliott Bay, and the Duwamish Estuary.

Concentrations of the detected CECs ranged from less than 1 ng/L (parts per trillion) for 10-hydroxy-amitriptyline, a metabolite of the antidepressant amitriptyline, to 786 ng/L for the type 2 diabetes drug metformin. Findings across all study areas were as follows:

- Metformin had the highest concentrations (up to 786 ng/L) and a high frequency of detection (27 of the 51 samples, or 52.9 percent).
- Caffeine and sulfamethoxazole (an antibiotic) were also commonly detected in 25 (50 percent) and 28 (55 percent) of the 51 samples, respectively.
- The most commonly detected CEC was benzoylecgonine (the metabolite of cocaine), which was found in 48 of the 51 samples (94.1 percent).
- The maximum detected concentration of benzoylecgonine was 1.1 ng/L; this is just above the method detect limits, which ranged from 0.2 ng/L to 0.4 ng/L.

Some CECs were detected in analytical method and equipment blank samples, which limited the ability to detect and distinguish some CECs in some samples even when

accounting for these quality assurance/quality control issues using standard U.S. Environmental Protection Agency protocols and best professional judgment. This is not unusual for the low analytical detection limits associated with this study.

Detected concentrations were compared to results from CEC studies of ambient waters in other parts of the country. All detected CECs in this study were generally comparable and well within an order of magnitude of the other studies. In some cases, detected results from this study were the highest (for example, 9.1 ng/L for albuterol); in other cases, detections were the lowest of all studies (for example, 0.929 ng/L for sulfadimethoxine).

Maximum detected CEC concentrations from this study were also compared to published ecological toxicology databases and (human and veterinary) prescribing information to help characterize the relative magnitude of these detections. Adverse impacts to ecological receptors or people exposed to the ambient waters in the study areas are not expected even at the reported maximum concentrations:

- All detected concentrations in this study are orders of magnitude below literature-reported toxicity values, which were mostly associated with mortality endpoints and other obvious impacts. This finding suggests that overt impacts to aquatic organisms associated with exposure to the detected CECs are not likely.
- For all detected CECs, drinking water doses (2 L) were assessed as a conservative way to screen out the likelihood that people who incidentally ingest waters from the study areas could acquire an adverse effect. The drinking water doses were still four or more orders of magnitude lower than a therapeutic dose. While this analysis was unable to account for sensitive groups, subpopulations, or the interactions of mixtures, it appears that human health impacts through CEC exposures in ambient waters are highly unlikely.

Results from this study may be useful if these CECs are monitored again in the future. Monitoring CECs in the future will help us to better understand if the concentrations of these chemicals are changing over time or if compounds that weren't detected this time might be in the future or vice versa. These results may also be used in designing future targeted CEC monitoring efforts.

Other Assessment Reports

This report is one of several reports that have been prepared as part of King County's Water Quality Assessment and Monitoring Study. Other reports are as follows:

- Three reports describe existing conditions and long-term trends in the Lake Union/Ship Canal, Elliott Bay, and Duwamish Estuary study areas.
- Two reports discuss the methodology and results of selected new studies to improve understanding of existing conditions: a study of bacteria in wet and dry weather and a literature review of potential conservative sewage tracers.
- A loadings report discusses present-day contributions of pollutants from various pathways, including stormwater runoff and CSOs, into the study areas and evaluates water quality impairments.

- A future loadings report assesses the potential of planned actions such as CSO control to improve water quality.
- A final report summarizes these analyses and implications.

King County will use the information from the Water Quality and Assessment and Monitoring Study to inform the next CSO control plan update, including looking for opportunities to improve water quality outcomes, possibly reduce costs of CSO control projects, establish baseline conditions for post-construction monitoring of CSO control projects, and decide whether to pursue an integrated CSO control plan. The information from the assessment can also be used to inform regional efforts to continue to improve water and sediment quality.

ABBREVIATIONS AND ACRONYMS

ACTC	anhydrochlortetracycline
Ballard Locks	Hiram M. Chittenden Locks
BPA	bisphenol-A
CEC	contaminants of emerging concern
CRQL	contract required reporting limit
CSO	combined sewer overflow
CTD	conductivity-temperature-depth
EPA	U.S. Environmental Protection Agency
FOD	frequency of detection
HRGC-MS	high-resolution gas chromatograph-mass spectrometry
HRLC-MS/MS	high-resolution liquid chromatography mass spectrometer
LMCL	lower method calibration limit
LOEC	lowest observed effect concentration
MDL	method detection limits
MNPCA	Minnesota Pollution Control Agency
NOAA	National Oceanic and Atmospheric Administration
PFC	perfluorinated chemical
PPCP	personal care products
QSAR	quantitative structure-activity relationship model
SAP	sampling and analysis plan
SFEI	San Francisco Estuary Institute
SSRI	selective serotonin reuptake inhibitor
STRT	Scientific and Technical Review Team
USGS	U.S. Geological Survey

1.0 INTRODUCTION

This report documents a King County study of the presence in surface waters of organic chemicals originating from pharmaceutical and personal care products. The study was prepared as part of a Water Quality Assessment and Monitoring Study, undertaken to explore ways to optimize water quality improvements in the three waterbodies where the County is planning combined sewer overflow (CSO) control projects—Elliott Bay, Lake Union/Ship Canal, and the Duwamish Estuary.

This chapter provides background on the Water Quality Assessment and Monitoring Study, study areas, contaminants of emerging concern, and the content of this report.

1.1 Water Quality Assessment and Monitoring Study

King County owns and operates 38 CSO outfalls in the City of Seattle. The County's 2012 CSO control plan includes nine projects to control 14 uncontrolled CSOs by 2030 to meet the Washington State standard of no more than one overflow per year on a 20-year moving average. The recommended projects involve construction of underground storage tanks, green stormwater infrastructure, and/or wet weather treatment facilities. Four projects are in the Lake Union/Ship Canal area and five are in the Duwamish Estuary and Elliott Bay areas.

Ordinance 17413, approving the CSO control plan, also calls for completion of a Water Quality Assessment and Monitoring Study (assessment) to inform the next plan update, which is due to regulators in 2018. In September 2013, the King County Council approved the assessment's scope of work through Motion 13966. The assessment includes a comprehensive scientific and technical analysis of water quality of the receiving waters ("study areas") where uncontrolled county CSOs discharge (Elliott Bay, Lake Union/Ship Canal, and the Duwamish Estuary).

The Water Quality Assessment and Monitoring Study set out to generate information that will help answer the following study questions:

1. What are the existing and projected water quality impairments in receiving waters (waterbodies) where King County CSOs discharge?[1]
2. How do county CSOs contribute to the identified impairments?
3. How do other sources contribute to the identified impairments?
4. What activities are planned through 2030 that could affect water quality in the receiving waters?
5. How can CSO control projects and other planned or potential corrective actions be most effective in addressing the impairments?

[1] "Impairments" is defined as water quality-related concerns.

6. How do various alternative sequences of CSO control projects integrated with other corrective actions compare in terms of cost, schedule, and effectiveness in addressing impairments?
7. What other possible actions, such as coordinating projects with the City of Seattle and altering the design of planned CSO control projects, could make CSO control projects more effective and/or help reduce the costs to WTD (King County Wastewater Treatment Division) and the region of completing all CSO control projects by 2030?

The assessment addresses Questions 1 through 4. King County will use the information to inform the 2018 CSO control plan update, prioritize and sequence CSO control projects, establish baseline conditions for post-construction monitoring of CSO control projects, and decide whether to pursue an integrated plan based on U.S. Environmental Protection Agency (EPA) guidelines. Questions 5 through 7 will be addressed during the CSO control program update.

An external Scientific and Technical Review Team (STRT) was assembled to review methodology and results. Depending on assessment findings, the King County Council may decide to approve formation of an Executive's Advisory Panel of approximately 10 regional leaders. The panel would develop independent recommendations to the King County Executive on how planned county CSO control projects can best be sequenced and integrated with other projects to maximize water quality gains and minimize costs to ratepayers.

Table 1-1 shows elements of the assessment and their associated study questions, deliverables, and estimated timeframes. Figure 1-2 illustrates the flow of reports and how they will inform the CSO program review process. More information on the assessment is available at http://www.kingcounty.gov/environment/wastewater/CSO/WQstudy.aspx.

Table 1-1. Elements of the Water Quality Assessment and Monitoring Study.

Element	Applicable Study Question	Deliverable	Timeframe
Review and analyze existing scientific and technical data on impairments in Lake Union/Ship Canal, Duwamish Estuary, and Elliott Bay.	1	Area reports: • Elliott Bay • Lake Union/Ship Canal • Duwamish Estuary	2013–2016
Conduct targeted data gathering and monitoring to fill some of the identified gaps in scientific data on water quality in these receiving waters.	1,2,3	Data gaps analysis report[a] Data gap study reports: • Bacteria • Contaminants of emerging concern • Literature review of conservative sewage tracers	2014–2016
Identify and quantify the current (2015) pathways of contaminants into the receiving waters.	2,3	Loadings Report	2015–2016
Identify changes in contaminant loadings between 2015 and 2030, including the potential impact of planned corrective actions on identified impairments in the waterbodies.	1,2,3,4	Future Loadings Report	2015–2016
Summarize scientific and technical data collected and reviewed during the assessment.	1,2,3,4	Synthesis Report	2015–2016

[a] *Identification and Assessment of New Studies to Improve Understanding of Existing Conditions.*

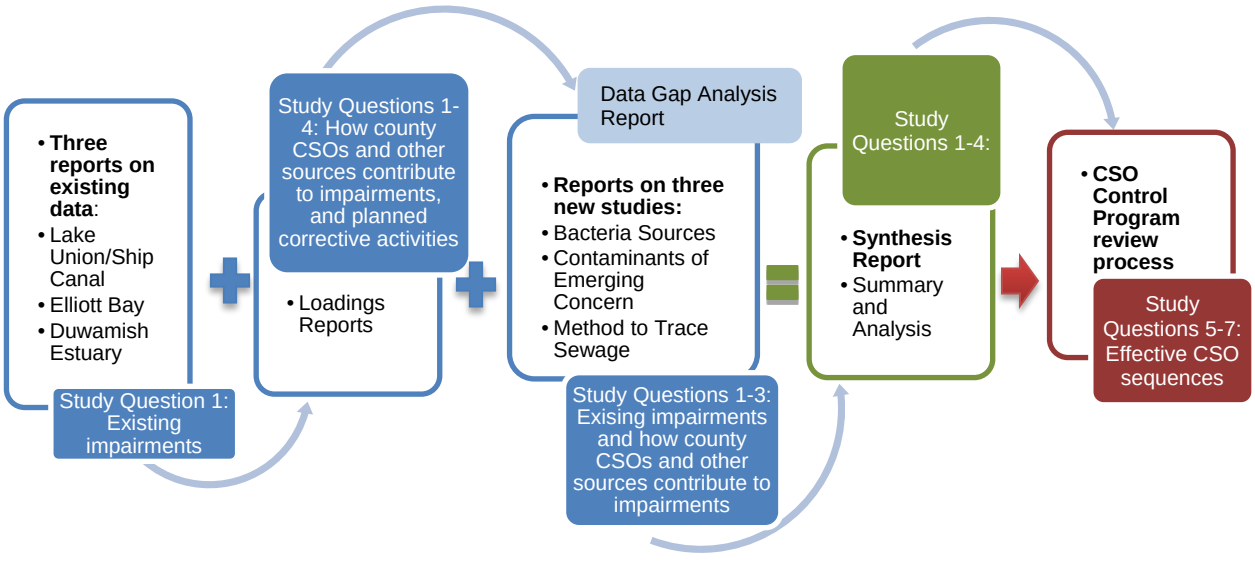


Figure 1-1. Reports and study questions answered as part of the Water Quality Assessment and Monitoring Study.

A key component of the Water Quality Assessment and Monitoring Study was to complete water quality characterizations of the study areas using data previously collected from a variety of monitoring programs and studies. The characterizations included assessment of current water, sediment, and fish and shellfish tissue quality and other indicators of ecological health; evaluation of long-term trends in conditions over time; and comparison to Washington State water and sediment quality standards to help identify impairments. The assessment also included estimations of loadings to these waterbodies from contaminant pathways and expected future loadings following planned water quality improvement actions.

Early in the assessment, a number of gaps were identified in the existing data that if filled, would provide critical information to answer study questions. Studies were identified to fill the data gaps and three studies were selected for implementation: sources and pathways of bacteria, chemical sewage tracers, and contaminants of emerging concern.

1.2 Study Areas

The three study areas are defined as follows (Figure 1-1):

- Lake Union/Ship Canal includes the waters flowing out of Lake Washington into the Montlake Cut, Portage Bay, Lake Union, the Fremont Cut, and Salmon Bay upstream of the Hiram M. Chittenden Locks (Ballard Locks). The Ballard Locks separate the saltwater of Puget Sound from the freshwaters of Lake Union.
- Elliott Bay encompasses the area east of a line drawn between Duwamish Head in West Seattle and Magnolia Bluff near Smith Cove, including the downtown Seattle waterfront. This area, also known as Inner Elliott Bay, is open to Outer Elliott Bay and Puget Sound to the west and receives freshwater inflows from the Duwamish Estuary.
- The Duwamish Estuary includes the East, West, and Lower Duwamish waterways, extending upstream to the turning basin located near the south end of the King County airport. The Duwamish Estuary receives freshwater flows from the Duwamish River and Green River watershed. At the mouth of the estuary, tidal exchanges occur with Elliott Bay.

Treated wastewater, stormwater, CSOs, illicit boat discharges, and other surface waters drain to these highly urbanized receiving waters.

1.3 Contaminants of Emerging Concern

A growing body of research is showing the persistence and ubiquity of contaminants such as prescription drugs, drug residues, and personal care products in the environment. Most of these contaminants of emerging concern (CECs) are relatively new; some are older chemicals such as DEET (a mosquito repellant). Advancing analytical techniques allow researchers and chemists around the world to monitor CECs, but there is no defined list of CECs and few regulatory standards for most of these chemicals (EPA, 2015a).

Interest in CECs has been increasing (Petrie et al., 2015). A recent effort in California set priorities for which CECs to monitor (Klosterhaus et al., 2013), and the Puget Sound Ecosystem Monitoring Program (James et al., 2015) and the Columbia River Toxics Reduction Working Group (EPA, 2015b) have conducted literature reviews of CECs.

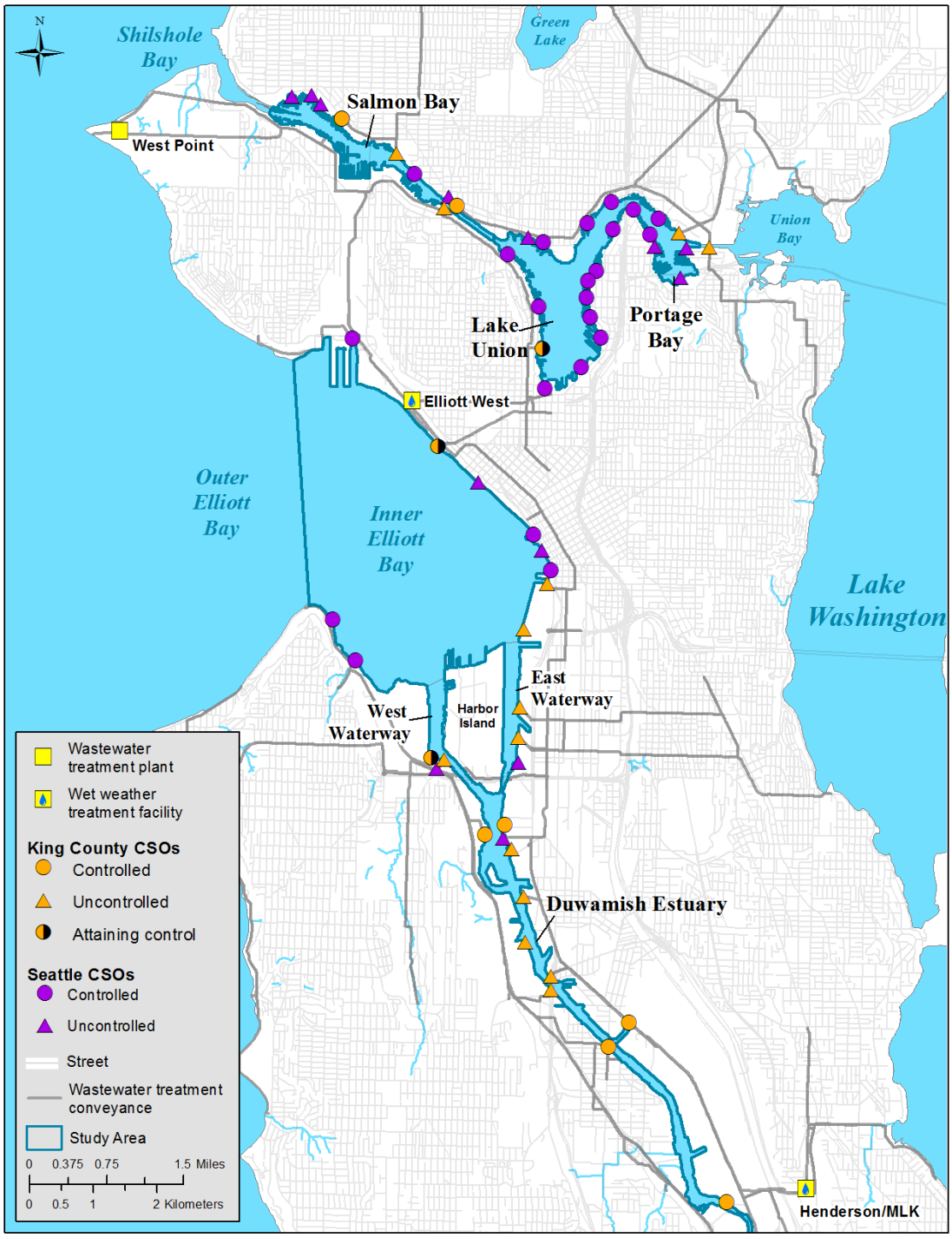


Figure 1-2. Lake Union/Ship Canal, Elliott Bay, and Duwamish Estuary study areas.

In general, CECs include the following:

- Over-the-counter and prescription pharmaceuticals
- Ingredients in personal care products such as the insect repellent DEET, parabens, and titanium dioxide nanoparticles
- Illicit drugs
- Metabolites of over-the-counter, prescription, and illicit pharmaceuticals and drugs
- Newer generation pesticides typically without water quality standards such as pyrethroid and neonicotinoid pesticides
- Some industrial chemicals without established water quality standards such as the plasticizer bisphenol-A (BPA), the antimicrobial triclosan, or long-chain perfluorinated chemicals (PFCs)

Studying the presence of CECs in the environment is of interest because environmental exposures to CECs can cause adverse human health or ecological impacts. Wastewater treatment plants and stormwater treatment facilities are not designed to remove many of these chemicals, and removal rates vary widely depending on the chemical, treatment technology, and operating conditions (Kostich et al., 2014). Work by universities and water resource agencies in North America have demonstrated that some CECs may impact aquatic life at relatively low levels of exposure (in the parts per trillion or parts per billion [ng/L or µg/L] range).

CECs that are likely to be most concentrated in wastewater may be useful as indicators of wastewater incursion and as a baseline from which to measure CSO control (Dickenson et al., 2011). CECs that are present in stormwater or that are not removed by wastewater treatment may be of interest because input of stormwater-associated contaminants will continue regardless of CSO controls.

1.4 Scope of this Study

No prior investigations into CECs in study area waterbodies have been done. The primary objective of this study was to conduct an initial, broad assessment by identifying the presence or absence of CECs in the study areas during non-storm conditions. A secondary study objective was to describe the relative magnitude of the detected CECs.

The study broadly defines CECs as any synthetic or naturally occurring chemical that is not regulated or commonly monitored in the environment. The study scope did not allow for the characterization of CECs in other weather conditions (storm sampling) or in other media (sediments, tissue, CSO discharges, stormwater). Identifying the CECs that people and aquatic life are routinely exposed to during ambient conditions was considered as an important first step. Future studies that characterize episodic exposures during storms without or during CSO events would enhance understanding of CECs in these waterbodies.

2.0 METHODOLOGY

This chapter describes the methodologies used to complete this CEC study. The study consisted of two main components: sampling/analysis and literature review.

2.1 Sampling

The sampling and analysis plan (SAP) outlined the general objectives of the study and the field and analytical methods to be used (King County, 2014). This section describes the sampling that was done.

2.1.1 Locations, Depths, and Equipment

A total of 11 station locations were used to collect samples in the study areas. Six of these stations included two depths (1 meter deep and 1 meter above the bottom), for a total of 17 samples collected during each sampling event. Four sampling stations were located in Lake Union/Ship Canal, three in Elliott Bay, and four in the Duwamish Estuary (Figure 2-1 and Table 2-1). Samples were collected from multiple depths to ensure that stratified waterbodies were adequately characterized. Either the Niskin bottles that are part of the conductivity-temperature-depth (CTD) profiler (KCEL SOP #221v2) or Scott bottles (KCEL SOP #213v2) were used.[2]

2.1.2 Frequency

A total of 51 samples were collected during three sampling events by the King County Environmental Laboratory Field Services Unit and submitted for analysis. Samples were collected during fall and winter 2014–2015 to span both dry and wet seasons (Table 2-2). Each of the three sampling events occurred over multiple days to accommodate other sampling activities and vessel schedules. Sampling dates were selected to avoid CSO discharge events and the 48 hours immediately after. Duwamish Estuary locations were sampled during high flows and/or outgoing tides.

Two field blanks were collected on September 22, 2014, in addition to the field samples collected for the study. Field blanks are a way of checking for contamination that may be introduced into environmental samples from sampling equipment and can confound the results. Field blanks were collected by pouring laboratory-grade deionized water through the Scott or Niskin bottle samplers. This reinstate water was analyzed for the full list of CECs.

[2] KCEL SOP = King County Environmental Laboratory standard operating procedure.

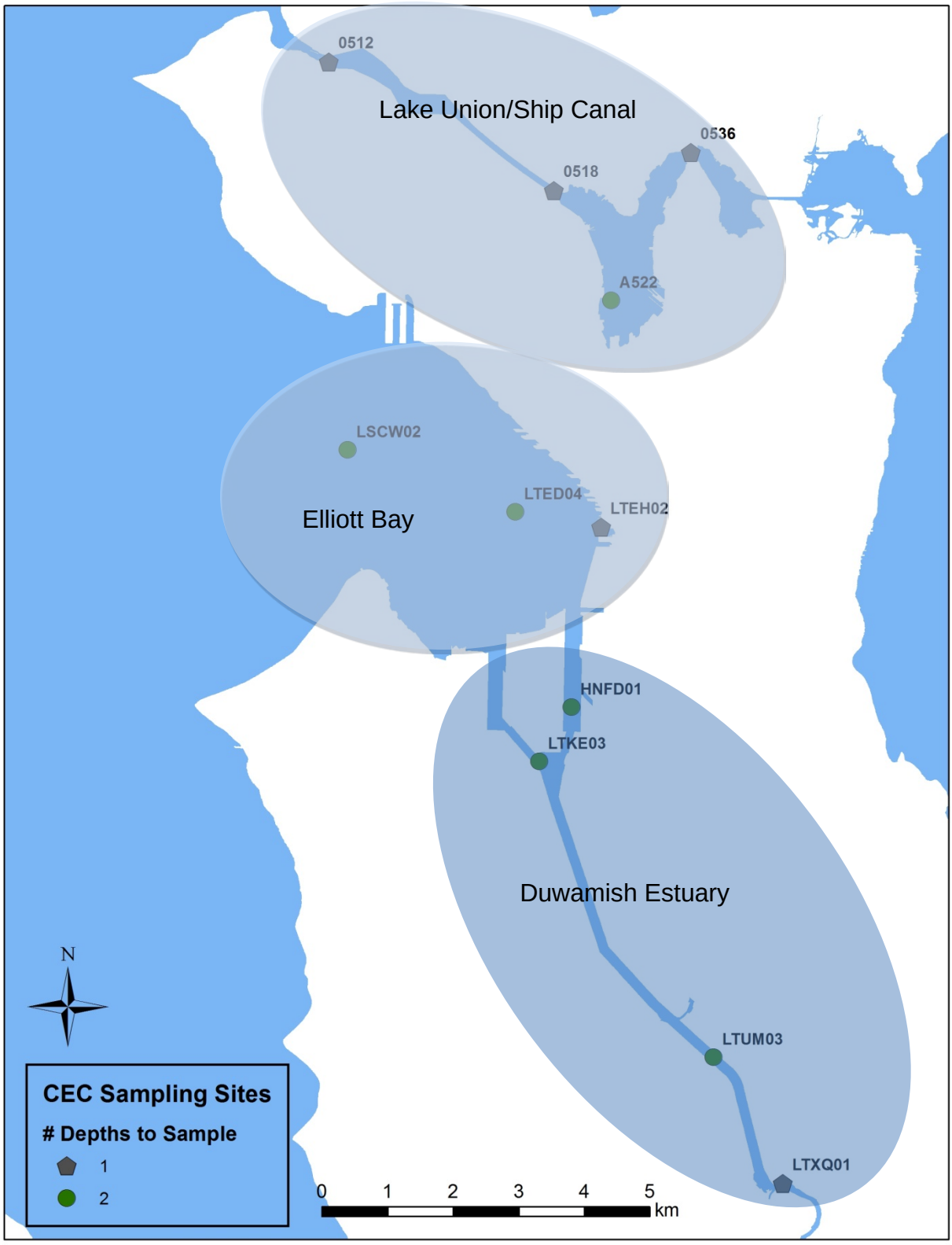


Figure 2-1. CEC sampling locations in the study areas.

Table 2-1. CEC sampling locations, target depths, and equipment.

Locator	Description	State Plane Easting	State Plane Northing	Depth	Sampling Equipment
LTKE03	Duwamish Estuary, Spokane Street Bridge over West Waterway	1265871	211418	2 depths, surface (1 m) and near bottom (~11 m)	Niskin/ conductivity-temperature-depth profiler (CTD)
LTUM03	Duwamish Estuary, 16th Ave. S. Bridge	1274591	196629	2 depths, surface (1 m) and near bottom (~6 m); target upper and lower layers	Niskin/CTD
HNFD01	Duwamish Estuary, East Waterway	1267486	214139	2 depths, surface (1 m) and near bottom (~7 m); target upper and lower layers	Niskin/CTD
LTXQ01	Duwamish Estuary, upstream of the turning basin	1278053	190313	1 m	Scott
A522	Lake Union/Ship Canal, western shore of Lake Union	1269458	234484	2 depths, surface (1 m) and near bottom (10 m)	Scott
0512	Lake Union/Ship Canal, Ballard Locks	1255339	246408	1 m	Scott
LTEH02	Elliott Bay Shoreline, Pier 52	1268961	223134	1 m	Niskin/CTD
LSCW02	Outer Elliott Bay	1247399	223360	2 depths, 1 m and 175 m	Niskin/CTD
LTED04	Inner Elliott Bay	1264675	223909	2 depths, 1 m, and 75 m	Niskin/CTD
0518	Lake Union/Ship Canal, Fremont Bridge	1266596	239972	1 m	Scott
0536	Lake Union/Ship Canal, at Interstate-5	1273447	241871	1 m	Scott

Table 2-2. CEC sampling dates by location.

Event	Dates	Locations Sampled
September	September 15, 2014	0512, 0518, A522, and 0536
	September 22, 2014	LTKE03, LSCW02, LTED04, HNFD01, LTUM03, Niskin and Scott bottle equipment blanks
November	November 10, 2014	0512, 0518, A522, and 0536
	November 17, 2014	LTKE03, LSCW02, LTED04, HNFD01, LTUM03
January	January 6, 2015	0512, 0518, A522, and 0536
	January 20, 2015	LTKE03, LSCW02, LTED04, HNFD01, LTUM03

2.2 Analysis

The following sections describe the CECs that were analyzed and the methods used in the analyses.

2.2.1 Analytes

An objective of the study was to analyze for a broad suite of likely CECs, preferably those detected in surface waters in other regions and those that may pose the greatest ecological or human health concerns. The list of CECs analyzed for this study was based on two analytical methods used—one for pharmaceuticals and a few personal care products (PPCPs) and one for pyrethroid insecticides. (See the section below for descriptions of these methods.)

The methods produced an analyte list of 140 CECs (Table 2-3), including the following:

- 100 pharmaceuticals
- 12 pharmaceutical metabolites
- 20 pyrethroid insecticides
- DEET
- Cocaine and nicotine (and their metabolites)

Both natural and synthetic pyrethroid insecticides were identified for analysis. Pyrethroids were originally extracted from chrysanthemum flowers; second and third generation pyrethroids are manufactured synthetically. Long-chain perfluorinated chemicals, hormonally active compounds, and nonylphenols are some of the chemicals not analyzed by these methods. King County (2007) conducted surveys for hormonally active compounds, including natural and synthetic estrogens, in the study areas. The objectives of these surveys were similar to those of this study, and these compounds were not included on the analyte list.

Table 2-3. CECs analyzed in this study including the common purpose for drugs, type of pyrethroid, or parent compound for metabolites.

CEC Analyte	Use, Type, or Parent Compound
1,7-Dimethylxanthine	Stimulant, caffeine metabolite, antispasmodic
10-hydroxy-amitriptyline	Antidepressant, amitriptyline metabolite
2-hydroxy-ibuprofen	Anti-inflammatory metabolite
4-epianhydro-chlortetracycline [EACTC]	Chlorotetracycline degradate
4-Epianhydrotetracycline [EATC]	Chlorotetracycline degradate
4-epichlortetracycline [ECTC]	Chlorotetracycline degradate
4-epioxytetracycline [EOTC]	Oxytetracycline degradate
4-epitetracycline [ETC]	Tetracycline degradate
Acetaminophen	Antipyretic, analgesic
Albuterol	Antiasthmatic
Allethrin	Synthetic pyrethroid
Alprazolam	Benzodiazepine anxiety medication
Amitriptyline	Tricyclic antidepressant
Amlodipine	Calcium channel blocker, antihypertensive
Amphetamine	Stimulant, medicinal and recreational drug
Anhydrochlortetracycline [ACTC]	Chlorotetracycline degradate
Anhydrotetracycline [ATC]	Chlorotetracycline degradate
Atenolol	Beta blocker, antihypertensive
Atorvastatin	Lipid regulator

CEC Analyte	Use, Type, or Parent Compound
Azithromycin	Macrolide antibiotic
Benzoylecgonine	Main metabolite cocaine
Benztropine	Anticholinergic, parkinson's medication
Betamethasone	Glucocorticoid steroid, both topical and injectable
Bifenthrin	Synthetic pyrethroid
Bisphenol A	Plasticizer
Caffeine	Stimulant
Carbadox	Livestock antibacterial
Carbamazepine	Anticonvulsant
Cefotaxime	Cephalosporin antibiotic
Chlortetracycline [CTC]	Tetracyline anitibiotic, first generation
Cimetidine	Antacid
Cinerin I	Natural pyrethroid
Cinerin II	Natural pyrethroid
Ciprofloxacin	Fluoroquinolone antibiotic
Clarithromycin	Macrolide antibiotic
Clinafloxacin	Fluoroquinolone antibiotic
Clonidine	Centrally acting α2 adrenergic agonist and imidazoline receptor agonist
Cloxacillin	Antibiotic
Cocaine	Opiate
Codeine	Prescribed opiate
Cotinine	Nicotine metabolite
Cyfluthrin	Synthetic pyrethroid
Cypermethrin	Synthetic pyrethroid
DEET	Insect repellent
Dehydronifedipine	Metabolite of nifedipine, an anti angina and antihypertensive drug
Deltamethrin/ Tralomethrin	Synthetic pyrethroid
Demeclocycline	Tetracycline antibiotic
Desmethyldiltiazem	Metabolite of diltiazem a calcium channel blocker
Diazepam	Benzodiazepine anxiety medication
Digoxigenin	Plant steroid
Digoxin	Cardiac glycoside
Diltiazem	Antihypertension
Diphenhydramine	Antihistamine
Doxycycline	Tetracycline antibiotic
Enalapril	Angiotensin-converting-enzyme (ACE) inhibitor for hypertension and CHF
Enrofloxacin	Veterinary fluoroquinolone antibiotic
Erythromycin-H2O	Macrolide antibiotic metabolite
Fenpropathrin	Synthetic pyrethroid
Fenvalerate	Synthetic pyrethroid
Flucythrinate	Synthetic pyrethroid insecticide, not approved in U.S.
Flumequine	Fluoroquinolone antibiotic
Fluocinonide	Topical corticosteroid
Fluoxetine	SSRI (serotonin reuptake inhibitor)
Fluticasone propionate	corticosteroid, topical and oral use
Furosemide	loop diuretic for edema and congestive heart failure
Gemfibrozil	Antilipemic
Glipizide	Short acting diabetes drug partially block potassium channels
Glyburide	Sulfonylurea, antidiabetic potassium channel inhibibitor

CEC Analyte	Use, Type, or Parent Compound
Hydrochlorothiazide	Thiazide diuretic
Hydrocodone	Semi-synthetic opioid
Hydrocortisone	Natural steroid hormone also given orally, intravenous injection, or topically
Ibuprofen	Analgesic
Isochlortetracycline [ICTC]	Chlorotetracycline degradate
Jasmolin I	Natural pyrethroid
Jasmolin II	Natural pyrethroid
L-Cyhalothrin	Synthetic pyrethroid
Lincomycin	Lincosamide antibiotic
Lomefloxacin	Fluoroquinolone antibiotic
Meprobamate	Anti-anxiety agent, largely replaced by benzodiazepines
Metformin	Diabetes management
Methylprednisolone	Synthetic corticosteroid
Metoprolol	Selective beta blocker for high blood pressure
Miconazole	Antifungal
Minocycline	Tetracycline antibiotic
Naproxen	Non-steroidal anti-inflammatory
Norfloxacin	Fluoroquinolone antibiotic
Norfluoxetine	SSRI
Norgestimate	Synthetic progesterone, hormone
Norverapamil	Calcium channel blocker, main metabolite of verapamil
Ofloxacin	Fluoroquinolone antibiotic
Ormetoprim	Veterinary macrolide antibiotic
Oxacillin	Beta lactam antibiotic
Oxolinic Acid	Quinolone antibiotic
Oxycodone	Semi-synthetic opioid
Oxytetracycline [OTC]	Second tetracycline
Paroxetine	SSRI
Penicillin G	Beta lactam antibiotic (benzylpenicillin, by injection)
Penicillin V	Beta lactam antibiotic (phenoxymethylpenicillin, oral)
Permethrin	Synthetic pyrethroid
Phenothrin	Synthetic pyrethroid, also known as sumithrin
Piperonyl butoxide	Synthetic pesticide synergist
Prallethrin	Natural pyrethroid
Prednisolone	Synthetic glucocorticoid, also active metabolite of prednisone
Prednisone	Synthetic corticosteroid and immunosuppressant
Promethazine	Antihistamine, first generation
Propoxyphene	Opioid, no longer marketed in U.S. since 2010
Propranolol	Nonselective beta blocker
Pyrethrin I	Natural pyrethroid
Pyrethrin II	Natural pyrethroid
Ranitidine	Antacid
Resmethrin	Synthetic pyrethroid
Roxithromycin	Macrolide antibiotic, only available in australia
Sarafloxacin	Fluoroquinolone antibiotic
Sertraline	SSRI
Simvastatin	Lipid regulator
Sulfachloropyridazine	Veterinary sulfonamide antibiotic
Sulfadiazine	Sulfonamide antibiotic
Sulfadimethoxine	Veterinary sulfonamide antibiotic
Sulfamerazine	Veterinary sulfonamide antibiotic
Sulfamethazine	Sulfonamide antibiotic, growth promotor in livestock and fowl

CEC Analyte	Use, Type, or Parent Compound
Sulfamethizole	Sulfonamide antibiotic
Sulfamethoxazole	Sulfonamide antibiotic
Sulfanilamide	Sulfonamide antibiotic
Sulfathiazole	Sulfonamide antibiotic now only veterinary use
Tetracycline [TC]	Tetracyline antibiotic
Tetramethrin	Synthetic pyrethroid
Theophylline	Methylxanthine drug for copd and asthma
Thiabendazole	Fungicide and parasiticide
Trenbolone	Active form of veterinary steroid used illicitly by body builders
Trenbolone acetate	Delivered form of veterinary steroid used illicitly by body builders
Triamterene	Sodium channel blocking diuretic
Triclocarban	Antimicrobial
Triclosan	Antimicrobial
Trimethoprim	Antibiotic
Tylosin	Macrolide antibiotic and feed additive
Valsartan	Angiotensin receptor blocker for high blood pressure and CHF
Verapamil	Phenylalkylamine calcium channel blocker for hypertension, angina pectoris, cardiac arrhythmia, and most recently, cluster headaches
Virginiamycin M1	Macrolide antibiotic feed additive
Warfarin	Anticoagulant

2.2.2 Analytical Methods

AXYS Analytical Services in Sidney, British Columbia, conducted the CEC analysis.

A total of 3 L of ambient water was collected at each sampling location to allow sufficient volume for analysis using the following methods:

- AXYS Analytical's SOP MLA-075 was used to analyze PPCPs. The method is based on the U.S. Environmental Protection Agency (EPA) Method 1694, which is a high-resolution liquid chromatography method with triple quadrupole mass spectrometer (HRLC-MS/MS) (EPA, 2007):
    - EPA method 1694 is one of the most robust methods developed for any contaminant. It uses a combination of internal and external calibrations and standards to ensure highly accurate identification of compounds with no misidentified peaks. The method also uses isotopically labeled internal standards to ensure the most precise quantification of detected compounds possible.
    - SOP MLA-075 uses isotope dilution and internal standards to identify and quantify compounds to ng/L (parts per trillion) concentrations or less. Isotope dilution methods adjust for the performance of each sample throughout the processing and analysis steps.
- Pyrethroid insecticides were analyzed by a high-resolution gas chromatograph-mass spectrometry (HRGC-MS) method (AXYS proprietary SOP MLA-046). Twenty pyrethroids were analyzed. This method is not an isotopic dilution method; it uses

retention times to identify individual pyrethroids and internal standards to quantify them to the ng/L level.

Because performance may vary slightly from sample to sample as the result of extraction volumes, matrix interferences, and other sample or analytical effects, the analytical results show the range of detected concentrations, method detection limits (MDLs), and lower method calibration limits (LMCLs) for all samples. LMCLs are the lowest method calibration limit used for the particular compound in that sample, accounting for dilution and other sample preparation influences. Detected concentrations below the LMCL are considered as estimates and are flagged with a "J".

2.3 Data Verification and Validation

Because MDLs are so low (sometimes less than 1 part per trillion), numerous equipment and method blanks were collected and analyzed. In some cases, environmental concentrations were very low and difficult to differentiate from possible field or laboratory contamination.

To ensure that the reported data adequately describe the presence or absence of the suite of analytes and provide an initial perspective on the relative magnitude of their concentrations, laboratory narratives and electronic data deliverables were reviewed. This review was conducted according to EPA (2009) at the "2A" level. Method performance criteria were reviewed along with method and equipment blanks. Most of the validation qualifiers applied to the results were due to method and equipment blank contamination.

No pyrethroids were found in method or equipment blanks. Concentrations of pharmaceuticals found in method and equipment blanks were always greater than the LMCL. The LMCL is the functional equivalent of the "contract required reporting limit" (CRQL) used in EPA (2014). Because the sample results were also greater than the LMCL, EPA recommends the use of best professional judgment during verification and validation. While a method blank was analyzed along with every batch of samples (total of five), only one Niskin bottle and one Scott bottle blank sample were analyzed. Therefore, the following blank qualification rules were developed based on typical analytical practices and best professional judgment:

- When concentrations in the environmental sample were within five times the respective workgroup's method blank, the reported concentrations were validated as non-detect with an elevated detection limit at the reported concentration.
- When the concentrations in the environmental sample were less that two times the respective equipment blank concentration, the sample result was qualified as non-detect at an elevated detection limit equal to the reported concentration.

Seven pharmaceuticals were detected in the five different method blanks analyzed (anhydrochlortetracycline [ACTC], metformin, atenolol, amphetamine, DEET, albuterol, and benztropine):

- Four pharmaceuticals were detected in the Niskin bottle equipment blank (metformin, atenolol, albuterol, and DEET).
- Only DEET was detected in the Scott bottle equipment blank.

Blank contamination led to requalification of 61 detected concentrations as non-detects with elevated detection limits. These concentrations represented 18.7 percent of all (325) detected values and 0.8 percent of the 7,256 individual sample-compound analyses. Because the primary study objective was to identify the presence or absence of CECs, the requalification of some results should not adversely impact study outcomes.

2.4 Literature Search and Comparisons to Toxicological and Therapeutic Values

To focus study resources on the CECs of greatest potential concern, a literature search was conducted on all detected CECs by generic name, with an emphasis on studies evaluating their concentrations in the environment and/or their risk to aquatic organisms. Literature and databases of ecological toxicity and prescribed human doses were also reviewed and synthesized to provide additional context and understanding to the detected CEC results. Neither the aquatic life nor the human health based concentrations should be viewed as a comprehensive risk assessment. Many other factors may be addressed in a risk assessment, such as an understanding of target organs and synergistic or antagonistic effects.

Individual peer-reviewed or government documents were reviewed and the National Oceanic and Atmospheric Administration (NOAA) database of CEC properties and lowest observed effect concentrations (LOECs) for aquatic life was queried for all detected CECs (NOAA, 2015). To put the detected chemicals and their concentrations into perspective relative to human health concerns, the Epocrates database published by Athena Health (2015) was queried to determine the current range of prescribed doses for all human pharmaceuticals. The lowest daily therapeutic dose prescribed was then compared to an assumed ingested dose calculated from the maximum detected concentration from this study multiplied by 2 liters of water (a typical ingestion rate used in human health risk assessment; Table 6-11 in EPA, 1989). This approach was taken to reduce the likelihood of overlooking a CEC with potential human or ecological impacts.

Comparing MDLs for all CECs with potential toxicological thresholds and understanding differences in numbers of prescriptions written and doses per patient, both of which may vary by an order of magnitude or more, were not considered vital to answering of study questions and therefore were not effective uses of project resources.

CECs that were not detected might be of future interest because of improved detection limits and concern for potential local impacts. However several of the CECs on the list of analytes are not approved for human or veterinary use in the United States and are unlikely candidates for illicit use or importation (for example, roxithromycin, a macrolide antibiotic approved in Australia). Other CECs that were undetected and are rarely prescribed (for

example, flumequine, a first-generation fluoroquinolone antibiotic) are also unlikely to be found in future studies.

3.0 RESULTS OF SAMPLING AND ANALYSIS

This chapter presents the results of CEC sampling and analysis. A summary of results across all study areas is presented first, followed by results for each study area. Because only three sampling events took place in less than six months, the results are considered as a snapshot of a select group of potential CECs in study area waters. Details of all samples for all compounds including non-detected CECs can be found here: http://green2.kingcounty.gov/ScienceLibrary/default.aspx.

Results of the literature search are presented in Chapter 4.

3.1 Across All Study Areas

CECs were detected 254 times across all stations and depths out of a total of 7,256 possible analyses. A total of 26 different CEC were detected out of the 140 that were analyzed. The overall frequency of detection (FOD) was 3.6 percent. As Figure 3-1 illustrates, benzoylecgonin (a metabolite of cocaine) was the most commonly detected CEC, with an FOD of 94 percent. Other CECs with FODs greater than 50 percent include sulfamethoxazole (an antibiotic) and metformin (a type 2 diabetes drug).

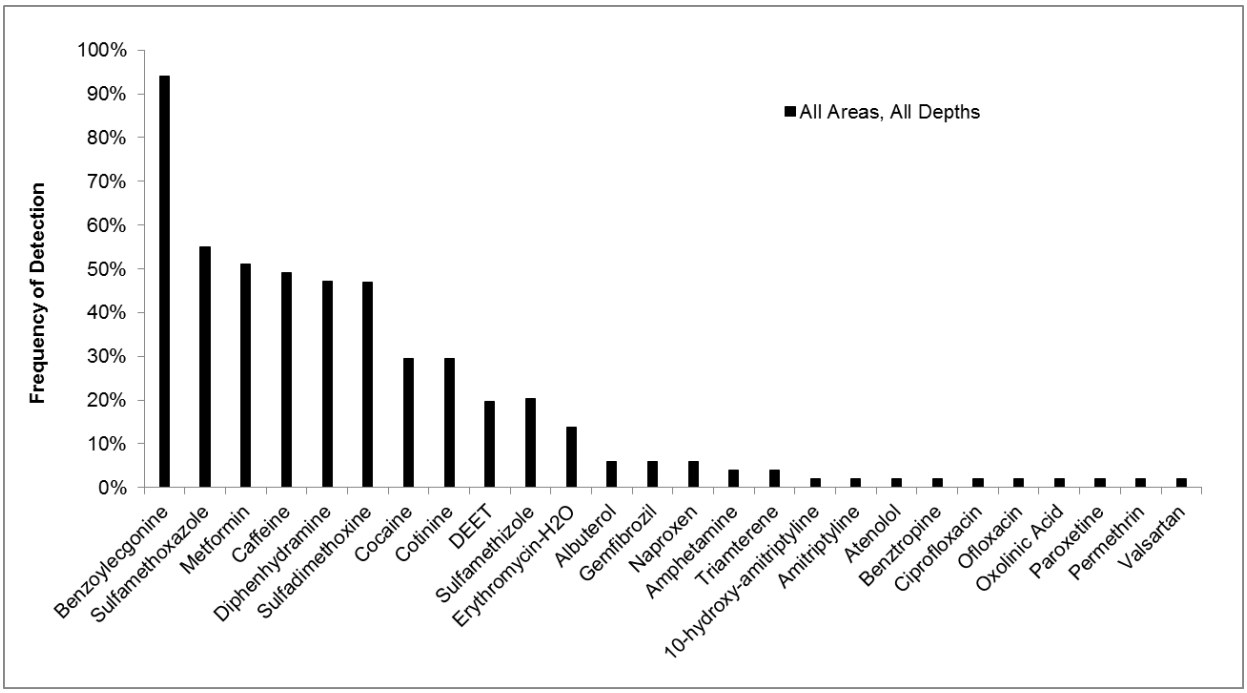


Figure 3-1. Frequency of detection of the 26 different CECs found across all stations and depths.

FODs for 6 of the 10 more commonly detected CECs (sulfamethoxazole, caffeine, diphenhydramine, sulfadimethoxine, cotinine, and sulfamethizole) vary by study area. For example, sulfamethoxazole was detected in as many as 80 percent of the samples from Elliott Bay but in as little as 13 percent of the samples in Lake Union/Ship Canal. Cotinine was detected in 100 percent of the Lake Union/Ship Canal samples but in none of the Elliott Bay and Duwamish Estuary samples. Figure 3-2 illustrates these differences.

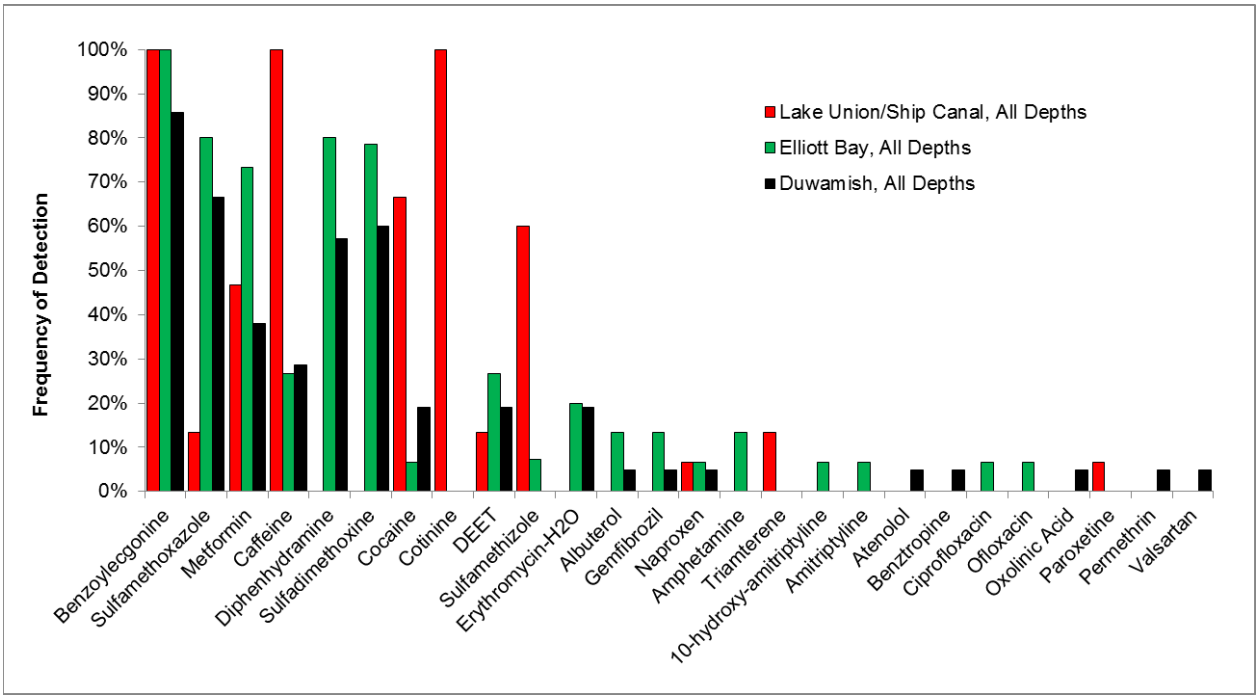


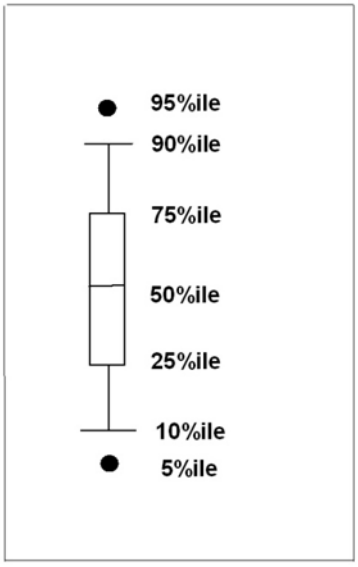
Figure 3-2. Frequency of detection of CECs by study area.

3.2 Lake Union Ship Canal

Fifteen samples were taken from Lake Union/Ship Canal. Figure 3-3 shows the concentrations and number of detections of CECs, and Table 3-1 presents summary statistics for each sampling station and compound.

A total of 11 different CECs were detected in Lake Union/Ship Canal. Detected CECs with high FODs were as follows:

- Caffeine was detected at the highest concentration. Concentrations were between 20 ng/L and 70 ng/L. The FOD was 100 percent.
- Benzoylecgonine and continine (metabolites of cocaine and nicotine, respectively) also had FODs of 100 percent.



Legend for box plots in Figures 3-3, 3-4, and 3-5.

- Cocaine (an illicit drug), sulfamethizole (a sulfonamide antibiotic), and metformin were commonly detected with FODs of 66, 60, and 47 percent, respectively.

Stations in Lake Union/Ship Canal were sampled at the surface only (1 m), except for A522 along the western shore of Lake Union that was sampled at both 1-m and 10-m depths. Twice as many samples were collected at this location to account for both depths. There were few notable differences between the twelve surface samples from this study area and the three samples taken at the 10-m depth from A522. Findings by station are as follows:

- Out of the 140 compounds analyzed over three sampling events, CECs were detected 34 times at station A522 at the surface and at depth. Naproxen (a non-steroidal anti-inflammatory) was detected only at the 10-m depth. The other detected CECs were in both the surface and the 10-m samples.
- CECs were detected 17 times each at the Fremont Bridge (0518) and Ballard Locks (0512).
- Paroxetine (a selective serotonin reuptake inhibitor [SSRI], trade name Paxil) was detected once at 0518.
- Sulfamethoxazole was detected twice in surface samples at A522 and 0512
- CECs were detected 11 times at the upstream boundary of the study area at 0536 under the Interstate-5 Bridge.

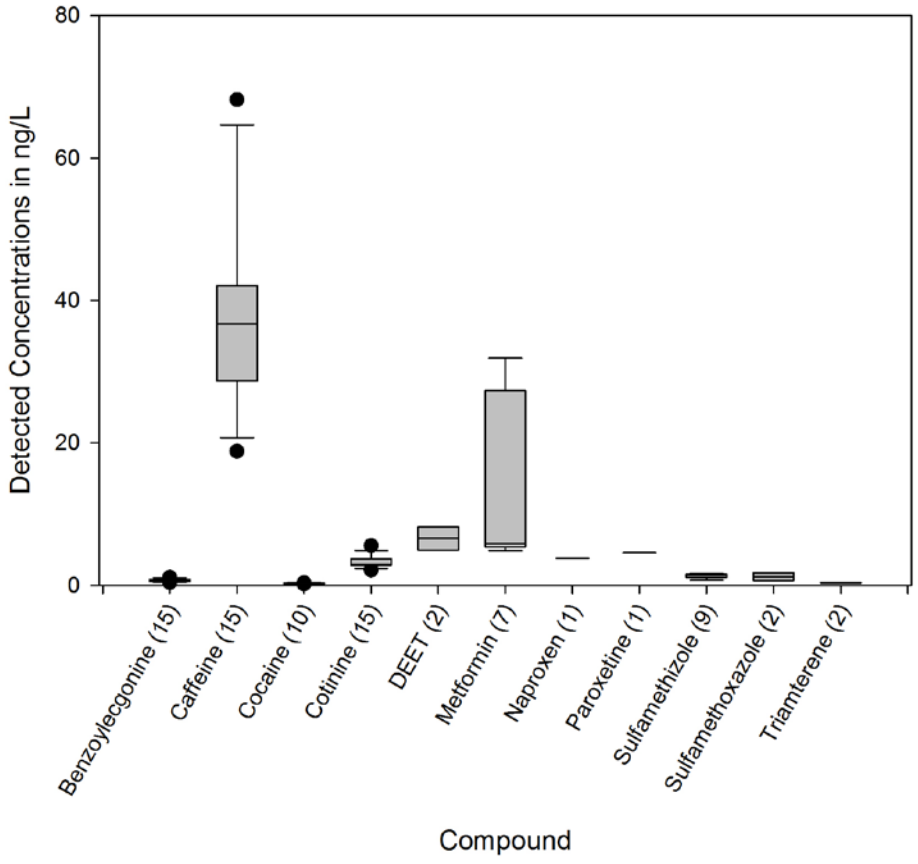


Figure 3-3. Concentrations and number of detections of CECs in Lake Union/Ship Canal (n =15).

Table 3-1. Detected CECs in Lake Union/Ship Canal by sampling location and depth.

Location	Depth	Chemical	FOD	Detected Concentration			Method Detection Limit			Lower Method Calibration Limit		
				Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Ship Canal, Ballard Locks												
0512	Surface	Benzoylecgonine	3/3	0.488	1.020	0.749	0.30	0.30	0.30	0.28	0.30	0.30
		Caffeine	3/3	35.0	42.1	39.3	14	15	15	14	15	15
		Cocaine	3/3	0.182	0.231	0.211	0.14	0.15	0.15	0.14	0.15	0.15
		Cotinine	3/3	2.95	5.57	3.83	1.5	1.6	1.5	1.5	1.6	1.5
		Metformin	2/3	5.83	31.9	18.2	3.0	17	7.7	3.0	3.1	3.1
		Sulfamethizole	2/3	1.53	1.56	1.55	0.56	0.77	0.65	0.56	0.61	0.59
		Sulfamethoxazole	1/3	1.75	1.75	1.75	0.56	1.2	0.80	0.56	0.61	0.59
Ship Canal, Fremont Bridge												
0518	Surface	Benzoylecgonine	3/3	0.455	1.100	0.729	0.30	0.39	0.33	0.30	0.35	0.32
		Caffeine	3/3	24.7	68.2	42.5	15	18	16	15	18	16
		Cocaine	3/3	0.226	0.332	0.270	0.15	0.18	0.16	0.15	0.18	0.16
		Cotinine	3/3	2.70	4.40	3.61	1.53	1.7	1.62	1.53	1.70	1.62
		Metformin	2/3	5.41	21.6	14.0	3.1	15	7.4	3.1	3.4	3.2
		Paroxetine	1/3	4.59	4.59	4.59	4.0	4.7	4.3	4.0	4.7	4.3
		Sulfamethizole	2/3	1.46	1.69	1.58	0.62	0.87	0.73	0.60	0.70	0.64
Ship Canal, at Interstate-5												
0536	Surface	Benzoylecgonine	3/3	0.349	0.595	0.454	0.31	0.40	0.34	0.31	0.31	0.31
		Caffeine	3/3	18.80	41.70	27.50	15.3	15.6	15.4	15.3	15.6	15.4
		Cotinine	3/3	2.55	3.01	2.80	1.55	1.64	1.59	1.55	1.64	1.59
		Metformin	1/3	5.39	15.40	9.28	3.09	15.4	8.51	3.09	3.28	3.18
		Sulfamethizole	1/3	1.370	1.370	1.370	0.61	0.88	0.70	0.61	0.62	0.62
Lake Union, Western Shore												
A522	Surface	Benzoylecgonine	3/3	0.595	0.920	0.721	0.30	0.45	0.35	0.30	0.32	0.31
		Caffeine	3/3	28.7	61.3	42.1	15	16	15	15	16	15
		Cocaine	2/3	0.177	0.241	0.209	0.15	0.21	0.18	0.15	0.16	0.15
		Cotinine	3/3	2.11	2.95	2.62	1.5	1.6	1.6	1.5	1.6	1.6
		DEET	1/3	4.24	8.20	6.49	0.81	7.0	4.0	0.79	0.86	0.82
		Sulfamethizole	2/3	0.695	1.45	1.07	0.59	1.2	0.80	0.59	0.64	0.61
		Sulfamethoxazole	1/3	0.680	0.680	0.680	0.59	1.1	0.77	0.59	0.64	0.61
		Triamterene	1/3	0.300	0.300	0.300	0.29	0.32	0.31	0.29	0.32	0.31

Location	Depth	Chemical	FOD	Detected Concentration			Method Detection Limit			Lower Method Calibration Limit		
				Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Lake Union, Western Shore												
A522	Bottom	Benzoylecgonine	3/3	0.539	0.801	0.646	0.30	0.32	0.30	0.30	0.32	0.30
		Caffeine	3/3	36.7	62.3	45.4	15	16	15	15	16	15
		Cocaine	2/3	0.267	0.287	0.277	0.15	0.18	0.16	0.15	0.16	0.15
		Cotinine	3/3	2.96	3.78	3.37	1.5	1.6	1.6	1.5	1.6	1.6
		DEET	1/3	3.79	6.74	5.16	0.79	6.74	3.77	0.79	0.84	0.81
		Metformin	2/3	4.85	27.3	12.8	3.0	6.4	4.2	3.0	3.2	3.1
		Naproxen	1/3	3.77	3.77	3.77	3.0	3.2	3.0	3.0	3.2	3.0
		Sulfamethizole	2/3	0.825	1.57	1.20	0.59	0.63	0.61	0.59	0.63	0.61
		Triamterene	1/3	0.347	0.347	0.347	0.30	0.32	0.31	0.30	0.32	0.31

FOD = frequency of detection.

3.3 Elliott Bay

In Elliott Bay, samples were collected at two depths from two stations and one depth from another station during three sampling events, for a total of 15 samples collected from Elliott Bay marine waters. Across all samples, 18 different CECs were detected (Figure 3-4). Table 3-2 presents summary statistics for each sampling station and compound in the bay.

Many of the detected CECs were found in only one or two samples. Benzoylecgonine, diphenhydramine (an antihistamine), sulfadimethoxine (a veterinary antibiotic), sulfamethoxazole (a human antibiotic), and metformin were consistently detected in most samples, with FODs ranging from 73 to 100 percent. There were no obvious patterns of these detections with depth.

Metformin concentrations were over 10 times higher than the next highest CEC (caffeine). Even with blank qualifications, metformin concentrations were so much higher than other detected CECs that the box plot in Figure 3-4 is presented with a split scale.

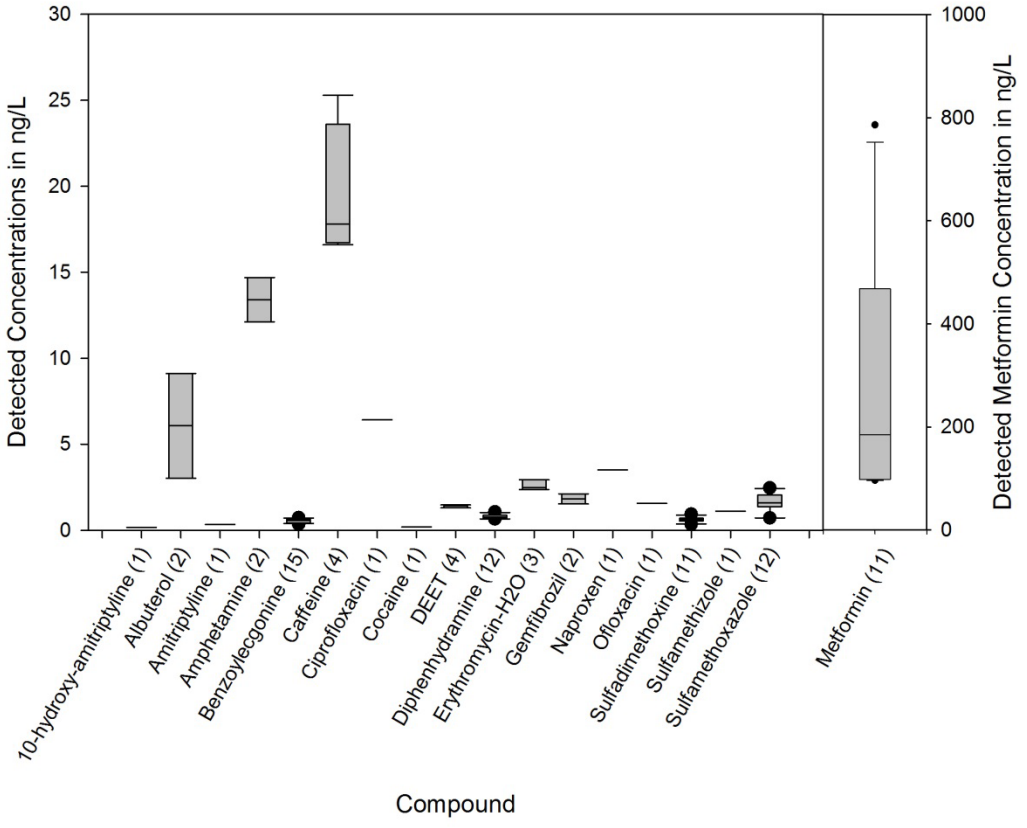


Figure 3-4. Concentrations and number of detections of CECs in Elliott Bay (n =15).

Table 3-2. Detected CECs in Elliott Bay by sampling location and depth.

Location	Depth	Chemical	FOD	Detected Concentrations			Method Detection Limit			Lower Method Calibration Limit		
				Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Outer Elliott Bay												
LSCW02	Surface	Albuterol	1/3	9.11	9.11	9.11	1.6	13	5.7	1.6	12.60	5.7
		Amphetamine	1/3	14.7	14.7	14.7	15	63	31	7.9	63	28
		Benzoylecgonine	3/3	0.336	0.643	0.518	0.29	0.31	0.30	0.29	0.31	0.30
		DEET	1/3	1.43	2.41	1.89	0.83	2.4	1.7	0.77	0.83	0.81
		Diphenhydramine	3/3	0.688	0.747	0.711	0.57	0.62	0.61	0.57	0.62	0.61
		Metformin	2/3	131	786	459	19	126	62	16	126	57
		Sulfadimethoxine	2/3	0.683	0.691	0.687	0.30	0.31	0.31	0.29	0.31	0.30
		Sulfamethoxazole	3/3	0.708	2.13	1.44	0.62	0.82	0.71	0.57	0.62	0.61
Outer Elliott Bay												
LSCW02	Bottom	Benzoylecgonine	3/3	0.445	0.692	0.603	0.29	0.33	0.31	0.29	0.33	0.31
		DEET	1/3	1.29	2.02	1.72	0.81	2.02	1.6	0.78	0.89	0.83
		Diphenhydramine	2/3	0.743	1.06	0.902	0.59	0.67	0.62	0.59	0.67	0.62
		Gemfibrozil	1/3	2.11	2.11	2.11	1.5	1.7	1.6	1.5	1.7	1.6
		Metformin	2/3	96.2	468	282	33	103	68	16	103	49
		Ofloxacin	1/3	1.56	1.56	1.56	1.5	1.7	1.6	1.5	1.7	1.6
		Sulfadimethoxine	2/2	0.342	0.417	0.380	0.30	0.33	0.32	0.30	0.33	0.32
		Sulfamethoxazole	2/3	1.57	2.46	2.02	0.61	12	4.4	0.59	0.67	0.62
Inner Elliot Bay												
LTED04	Surface	Benzoylecgonine	3/3	0.423	0.624	0.518	0.29	0.32	0.30	0.29	0.32	0.30
		Caffeine	1/3	16.6	16.6	16.6	15	16	15	15	16	15
		DEET	1/3	1.47	2.52	1.98	0.79	2.5	1.8	0.78	0.86	0.81
		Diphenhydramine	3/3	0.647	0.794	0.714	0.58	0.64	0.61	0.58	0.64	0.61
		Erythromycin-H2O	1/3	2.36	2.36	2.36	2.2	2.5	2.3	2.2	2.5	2.3
		Gemfibrozil	1/3	1.53	1.53	1.53	1.5	1.6	1.5	1.5	1.6	1.5
		Metformin	2/3	118	396	257	35	230	132	17	131	59
		Sulfadimethoxine	2/3	0.702	0.928	0.815	0.29	0.32	0.30	0.29	0.32	0.30
		Sulfamethoxazole	2/3	1.39	1.72	1.56	0.64	1.7	0.98	0.58	0.64	0.61

Location	Depth	Chemical	FOD	Detected Concentrations			Method Detection Limit			Lower Method Calibration Limit		
				Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Inner Elliot Bay												
LTED04	Bottom	Benzoylecgonine	3/3	0.416	0.725	0.592	0.29	0.32	0.30	0.29	0.32	0.30
		DEET	1/3	1.40	3.13	2.11	0.78	3.1	1.9	0.77	0.86	0.81
		Diphenhydramine	2/3	0.856	0.880	0.868	0.58	0.65	0.60	0.58	0.65	0.60
		Erythromycin-H2O	2/3	2.47	2.93	2.70	2.2	2.5	2.3	2.2	2.5	2.3
		Metformin	2/3	97.2	620	359	48	133	78	15	133	59
		Naproxen	1/3	3.50	3.50	3.50	2.9	3.2	3.1	2.9	3.2	3.1
		Sulfadimethoxine	2/3	0.551	0.604	0.578	0.29	0.33	0.32	0.29	0.32	0.30
		Sulfamethoxazole	2/3	1.34	1.80	1.57	0.59	0.90	0.71	0.58	0.65	0.60
Elliott Bay Shoreline, Pier 52												
LTEH02	Surface	10-hydroxy-amitriptyline	1/3	0.164	0.164	0.164	0.15	0.15	0.15	0.15	0.15	0.15
		Albuterol	1/3	3.03	3.03	3.03	1.6	2.1	1.8	1.6	2.1	1.8
		Amitriptyline	1/3	0.332	0.332	0.332	0.30	0.33	0.31	0.29	0.30	0.30
		Amphetamine	1/3	12.1	12.1	12.1	7.9	11	9.2	7.9	11	9.2
		Benzoylecgonine	3/3	0.426	0.574	0.524	0.29	0.31	0.30	0.29	0.30	0.30
		Caffeine	3/3	17.1	25.3	20.3	15	20	17	15	15	15
		Ciprofloxacin	1/3	6.41	6.41	6.41	6.0	6.6	6.2	5.9	6.1	6.0
		Cocaine	1/3	0.184	0.184	0.184	0.15	0.16	0.15	0.15	0.15	0.15

FOD = frequency of detection.

3.4 Duwamish Estuary

In the Duwamish Estuary, samples were collected from two depths at three stations and one depth at a fourth station during three sampling events, for a total of 21 samples collected in the Duwamish Estuary. Seventeen different CECs were detected across the 21 samples out of the 140 CECs analyzed. (Figure 3-5). Table 3-3 presents summary statistics for each sampling station and compound in the Duwamish Estuary.

CECs were detected at all sites and at all depths sampled. Not all CECs were detected at all stations and depths. Many of the detected CECs, such as metformin (38 percent FOD), bezoylecgonine (76 percent FOD), and sulfamethoxazole (66 percent FOD), were also frequently detected in Lake Union/Ship Canal and Elliott Bay. There was no obvious pattern to the detections based on depth or spatial distribution and no obvious patterns related to salinity. The absence of measurable differences between depths and salinities may indicate a true lack of any relationship or may reflect the limited number of samples collected and/or the number of detections.

While all study areas had detectable concentrations of DEET (N, N-diethyl-meta-toluamide), the Duwamish Estuary was the only study area where a pyrethroid pesticide (permethrin) was detected. Permethrin was detected once at LTUM03 (at the 16th Avenue South Bridge). This pesticide is used in agriculture, to treat livestock, in forestry, to prevent fleas and ticks in pets, and to treat scabies (skin infection from the mite *Sarcoptes scabiei*).

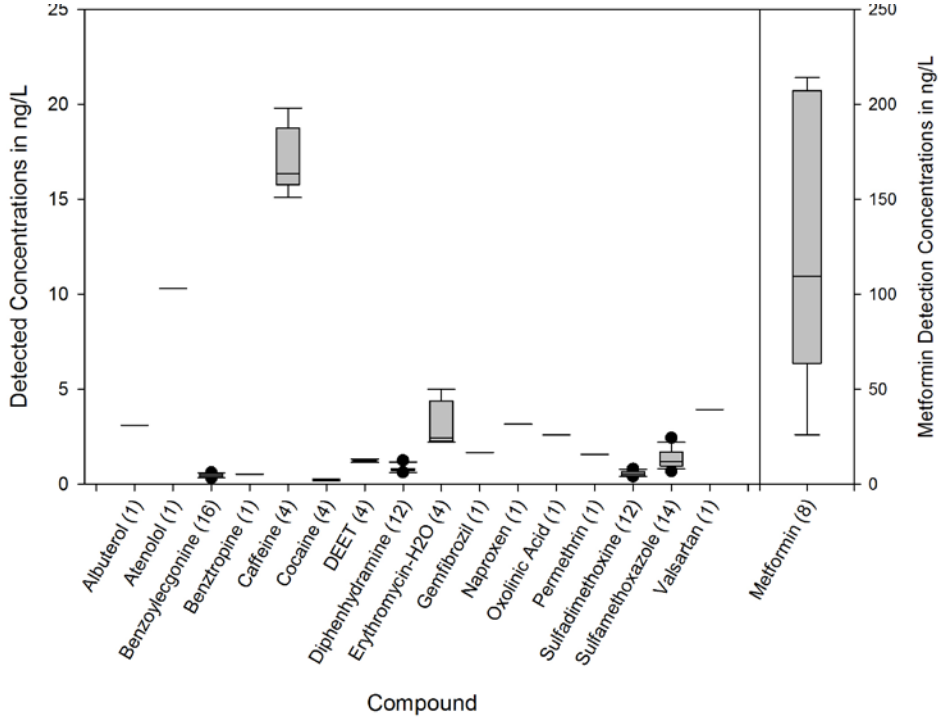


Figure 3-5. Concentrations and number of detections of CECs in the Duwamish Estuary (n=21).

Table 3-3. Detected CECs in the Duwamish Estuary by sampling location and depth.

Location	Depth	Chemical	FOD	Detected Concentrations			Method Detection Limit			Lower Method Calibration Limit		
				Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
East Waterway at Hanford												
HNFD01	Surface	Benzoylecgonine	3/3	0.343	0.576	0.467	0.28	0.32	0.30	0.28	0.32	0.30
		Caffeine	1/3	16.1	16.1	16.1	14	16	15	14	16	15
		Cocaine	1/3	0.183	0.183	0.183	0.14	0.16	0.15	0.14	0.16	0.15
		DEET	1/3	1.15	2.62	1.87	0.80	2.62	1.7	0.76	0.86	0.81
		Diphenhydramine	2/3	0.712	1.24	0.976	0.57	0.65	0.60	0.57	0.65	0.60
		Erythromycin-H2O	1/3	2.21	2.21	2.21	2.2	2.5	2.3	2.2	2.5	2.3
		Sulfadimethoxine	2/3	0.407	0.601	0.504	0.28	0.32	0.30	0.28	0.32	0.30
		Sulfamethoxazole	3/3	0.961	1.63	1.22	0.60	0.81	0.69	0.57	0.65	0.60
East Waterway at Hanford												
HNFD01	Bottom	Benzoylecgonine	3/3	0.420	0.612	0.522	0.29	0.31	0.30	0.29	0.31	0.30
		DEET	1/3	1.15	2.21	1.75	0.77	2.21	1.6	0.77	0.84	0.79
		Diphenhydramine	3/3	0.748	0.806	0.784	0.58	0.63	0.59	0.58	0.63	0.59
		Gemfibrozil	1/3	1.67	1.67	1.67	1.4	1.6	1.5	1.4	1.6	1.5
		Metformin	1/3	103	103	103	20	114	55	16	114	53
		Sulfadimethoxine	2/3	0.623	0.756	0.690	0.29	0.48	0.36	0.29	0.31	0.30
		Sulfamethoxazole	2/3	1.83	2.43	2.13	0.58	3.3	1.5	0.58	0.63	0.59
West Waterway, Spokane St Bridge												
LTKE03	Surface	Benzoylecgonine	3/3	0.416	0.464	0.444	0.29	0.30	0.30	0.29	0.30	0.30
		Caffeine	2/3	15.1	16.6	15.9	14	15	15	14	15	15
		Cocaine	1/3	0.222	0.222	0.222	0.14	0.15	0.15	0.14	0.15	0.15
		DEET	1/3	1.30	2.57	2.02	0.81	2.57	1.9	0.77	0.81	0.79
		Diphenhydramine	2/3	0.631	0.711	0.671	0.58	0.60	0.59	0.58	0.60	0.59
		Erythromycin-H2O	1/3	2.46	2.46	2.46	2.2	2.3	2.3	2.2	2.3	2.3
		Metformin	1/3	23.2	214	119	23	107	75	15	95	47
		Sulfadimethoxine	1/2	0.552	0.552	0.552	0.30	0.36	0.33	0.30	0.30	0.30
		Sulfamethoxazole	2/3	1.14	1.16	1.15	0.60	8.06	3.25	0.58	0.60	0.59

Location	Depth	Chemical	FOD	Detected Concentrations			Method Detection Limit			Lower Method Calibration Limit		
				Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
West Waterway, Spokane St Bridge												
LTKE03	Bottom	Benzoylecgonine	3/3	0.399	0.645	0.526	0.29	0.31	0.30	0.29	0.31	0.30
		Benztropine	1/3	0.525	0.525	0.525	0.49	0.52	0.50	0.49	0.52	0.50
		Caffeine	1/3	19.8	19.8	19.8	15	16	15	15	16	15
		DEET	1/3	1.31	2.24	1.76	0.78	2.24	1.6	0.78	0.83	0.80
		Diphenhydramine	3/3	0.732	0.949	0.808	0.58	0.62	0.60	0.58	0.62	0.60
		Metformin	2/3	116.0	213.0	164.5	15	144	93	15	120	55
		Naproxen	1/3	3.15	3.15	3.15	2.9	3.1	3.0	2.9	3.1	3.0
		Oxolinic Acid	1/3	2.58	2.58	2.58	0.62	2.6	1.8	0.62	2.0	1.5
		Sulfadimethoxine	2/3	0.659	0.797	0.728	0.29	0.31	0.30	0.29	0.31	0.30
		Sulfamethoxazole	2/3	0.903	1.96	1.43	0.62	2.46	1.28	0.58	0.62	0.60
		Valsartan	1/3	3.93	3.93	3.93	3.9	4.1	4.0	3.9	4.1	4.0
Duwamish, 16th Ave South Bridge												
LTUM03	Surface	Albuterol	1/3	3.10	3.10	3.10	1.5	2.9	2.3	1.5	2.9	2.3
		Atenolol	1/3	10.3	10.3	10.3	3.1	16	8.2	3.1	16	8.2
		Benzoylecgonine	2/3	0.320	0.349	0.335	0.29	0.30	0.30	0.29	0.30	0.30
		Cocaine	1/3	0.161	0.161	0.161	0.15	0.15	0.15	0.15	0.15	0.15
		Erythromycin-H2O	1/3	2.38	2.38	2.38	2.3	2.3	2.3	2.3	2.3	2.3
		Metformin	2/3	26.0	190	108	15	39	26	15	29	23
		Sulfadimethoxine	1/3	0.393	0.393	0.393	0.29	0.30	0.30	0.29	0.30	0.30
		Sulfamethoxazole	2/3	0.926	1.21	1.07	0.60	0.81	0.67	0.59	0.61	0.60
Duwamish, 16th Ave South Bridge												
LTUM03	Bottom	Benzoylecgonine	3/3	0.343	0.605	0.504	0.29	0.32	0.31	0.29	0.32	0.31
		Diphenhydramine	2/3	0.615	0.715	0.665	0.58	0.65	0.61	0.58	0.65	0.61
		Erythromycin-H2O	1/3	5.00	5.00	5.00	2.2	2.5	2.3	2.2	2.5	2.3
		Metformin	2/3	63.1	65.6	64.4	17	40	29	17	30	23
		Permethrin	1/3	1.57	1.57	1.57	0.52	0.69	0.63	---	---	---
		Sulfadimethoxine	2/3	0.488	0.548	0.518	0.29	0.32	0.31	0.29	0.32	0.31
		Sulfamethoxazole	2/3	1.22	1.24	1.23	0.65	1.2	0.85	0.58	0.65	0.61

Location	Depth	Chemical	FOD	Detected Concentrations			Method Detection Limit			Lower Method Calibration Limit		
				Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Duwamish, Upstream of Turning Basin at Boeing Pedestrian Bridge												
LTXQ01	Surface	Benzoylecgonine	1/3	0.339	0.339	0.339	0.30	0.32	0.31	0.30	0.32	0.31
		Caffeine	2/3	16.0	18.4	17.2	15	16	15	15	16	15
		Cocaine	1/3	0.253	0.253	0.253	0.16	0.18	0.17	0.15	0.16	0.15
		Sulfadimethoxine	2/3	0.427	0.502	0.465	0.30	0.37	0.33	0.30	0.32	0.31
		Sulfamethoxazole	1/3	0.674	0.674	0.674	0.60	1.2	0.82	0.60	0.65	0.62

FOD = frequency of detection.

4.0 DISCUSSION

This chapter discusses issues and uncertainties associated with analytical results and compares detected CECs with concentrations found by other monitoring programs, with ecological toxicity values, and with human therapeutic doses.

4.1 Blank Issues and Data Validation Concerns

4.1.1 Method Blanks

Some CECs were detected in method blanks, partly because of the very low detection limits provided by high resolution liquid chromatography/high resolution mass spectroscopy methods. Sample concentrations were requalified when they were within five times the concentrations reported in the analytical workgroups associated with the method blank. Table 4-1 shows the CECs with detected concentrations in method blanks. Most concentrations were close to the MDL for the method, indicating that method blank contamination is an unlikely source of substantive bias to the reported FODs and detected concentrations except at the very lowest concentrations.

Table 4-1. Detected CECs in method blank samples (ng/L).

Contaminant of Emerging Concern	Detected Concentration	Method Detection Limit
Albuterol	0.706	0.3
Albuterol	0.561	0.3
Amphetamine	1.52	1.5
Anhydrochlortetracycline (ACTC)	16.2	15
Atenolol	1.97	0.6
Benztropine	0.509	0.5
DEET	1.66	0.8
DEET	1.56	0.8
DEET	1.11	0.8
DEET	0.954	0.8
Metformin	3.9	3
Metformin	3.31	3

4.2 Equipment Blanks

Because only two equipment blank samples were collected and they were not collected for every sampling event day (six days), only concentrations within a factor of two were requalified as non-detect based on equipment blanks (Table 4-2). The detected concentrations in equipment blanks were much higher than those in method blanks. Equipment contamination, therefore, could influence the reported results and FODs for these CECs, particularly at low to modest concentrations (single to tens of ng/L).

Table 4-2. Detected CECs in equipment blank samples by equipment type (ng/L).

Equipment	Parameter	Detected Concentration	Method Detection Limit
Niskin	Albuterol	4.97	3.17
Niskin	Atenolol	58.2	6.34
Scott	DEET	2.83	0.955
Niskin	DEET	1.93	0.837
Niskin	Metformin	74.5	31.7

4.3 Comparison to Ambient Levels Found in Other Monitoring Programs

To help understand the relative magnitude of the CEC detections in the study areas, the detected concentrations were compared with the concentrations from the following studies done in the United States (Table 4-3):

- A well-known U.S. Geological Survey (USGS) publication on data from Boulder Creek in Colorado, an effluent-dominated stream (Barber et al., 2003)
- Fifty lakes sampled by the Minnesota Pollution Control Agency (MNPCA) in cooperation with EPA (Ferrey, 2013)
- Values reported by the San Francisco Estuary Institute (SFEI) (Klosterhaus et al., 2013)
- Delaware River ambient concentrations (MacGillivray, 2013)
- Lake Michigan (Blair et al., 2013)
- Puget Sound (Miller-Schultze et al., 2014)
- Ambient concentrations from the Santa Ana River in Orange County, California (Intertox, 2009)

Methods, detection limits, and analyte lists were not entirely in common with this study.

Detected concentrations in this study do not appear noteworthy or unusual compared to the other published results. Across all studies and all CECs, the maximum detections from this study were in all cases comparable and either within the range or in the same order of magnitude relative to detections in the other studies. Findings from the comparison are as follows:

- In most cases, concentrations of the detected CECs in the study areas are similar to or substantially less than CEC concentrations measured elsewhere in the United States. For example, gemfibrozil was detected in five of the studies at concentrations ranging from 2.11 ng/L to 41.03 ng/L; the maximum concentration for this study was the lowest of all of these values (2.11 ng/L).
- In some cases, the detected concentration in this study was the highest; albuterol was analyzed in four of the six studies with concentrations ranging from non-detect (no MDL given) to 9.11 ng/L. The highest value (9.11 ng/L) was detected in this study.

Table 4-3. Maximum detected CECs in this study compared to other ambient water values in the literature (ng/L). Bold values in italics represent the highest concentration across the studies.

Parameter	This Study[a]	Puget Sound[a]	Lake Michigan[a]	Boulder Creek, CO[a]	50 Lakes in Minnesota[a]	San Francisco Estuary[a]	Santa Ana River, Orange County, CA[a]	Delaware River[b]
10-hydroxy-amitriptyline	0.164	N/A	N/A	N/A	ND	***0.3***	N/A	N/A
Albuterol	***9.11***	N/A	5.9	ND	N/A	1	N/A	0.84
Amitriptyline	0.332	N/A	N/A	N/A	***4.1***	0.6	N/A	1.39
Amphetamine	***14.7***	N/A	N/A	N/A	N/A	9.7	N/A	5.53
Atenolol	58.2	N/A	N/A	N/A	N/A	37	N/A	***58.8***
Benzoylecgonine	1.1	N/A	N/A	N/A	9.5	7.2	N/A	***39.6***
Benztropine	0.525	N/A	N/A	N/A	0.7	< 0 .3	N/A	N/A
Caffeine	68.2	~200	190	510	42.9	132	***1,255***	158.7
Ciprofloxacin	6.41	N/A	ND	N/A	19.4	***1,300***	22	N/A
Cocaine	0.332	N/A	N/A	N/A	5.3	2.4	N/A	***2.58***
Cotinine	5.57	~10.5	21	***200***	N/A	25	N/A	36.75
DEET	8.2	N/A	N/A	N/A	125	21	***136***	42.8
Diphenhydramine	1.24	N/A	43	***82.5***	1.1	1.9	N/A	4.93
Erythromycin-H2O	5	N/A	N/A	N/A	ND	***41.6***	N/A	9.69
Gemfibrozil	2.11	N/A	***43***	ND	13.2	38	23	41.03
Metformin	786	N/A	***9,200***	ND	N/A	NQ	N/A	2,355.0
Naproxen	3.77	N/A	31	N/A	ND	8.2	N/A	***46.23***
Ofloxacin	1.56	N/A	***21***	N/A	8.9	< 15	N/A	ND
Oxolinic Acid	***2.58***	N/A	N/A	N/A	ND	< 3.5	N/A	N/A
Paroxetine	***4.59***	N/A	N/A	ND	ND	< 4.0	N/A	N/A
Permethrin	***1.57***	N/A	N/A	N/A	N/A	< 0.40	N/A	N/A
Sulfadimethoxine	0.928	~6	ND	N/A	3.5	***223***	N/A	1.64
Sulfamethizole	1.69	N/A	ND		2.5	***16***	N/A	ND
Sulfamethoxazole	2.46	~7	ND	220	57.1	***1,060***	84	116.93
Triamterene	0.347	N/A	N/A	N/A	N/A	***9.6***	N/A	4.47
Valsartan	3.93	N/A	N/A	N/A	5	92	N/A	***97.6***

See the text for citations for these studies.

[a] Maximum reported values.

[b] Maximum and arithmetic mean values depending on the parameter, per Table 4 in MacGillivray (2013).

ND = not detected, no detection limit given; <# = not detected at the given maximum detection limit; N/A = not analyzed.

4.4 Detected CECs of Potential Concern

4.4.1 Effects on Aquatic Organisms

The NOAA database of pharmaceuticals in the environment was queried for toxicological thresholds for detected CECs to better understand their potential to cause adverse effects to aquatic life and to help prioritize future investigations. The database is compiled from a broad suite of literature that includes direct measurements of toxicity to representative organisms and predicted toxicity values based on the chemical mode of action. The predicted toxicity values are based on quantitative structure-activity relationship models (QSARs) of CECs that show the functional chemical groups attached to each molecule and how these functional groups are known to interact chemically with biological tissues.

Measured and QSAR-based toxicity thresholds for fish, algae/cyanobacteria, and crustaceans were compared with maximum concentrations of CECs detected in this study (Table 4-4). The results indicate that the concentrations of CECs detected in this study are not likely to cause adverse effects to aquatic life. Most of the toxicity data included in the NOAA database were based on exposure to much higher concentrations than detected in this study; toxicity endpoints evaluated were generally limited to acute effects. Subtle effects may be associated with other endpoints (such as reproduction, predator avoidance, olfaction), but they are difficult to measure and describe for ambient concentrations of CECs in the ng/L range. Unlike endocrine disrupting compounds such as estrogens and phytoestrogens, most of the CECs detected in this study have different modes of action and are unlikely to behave profoundly synergistically with one another and amplify each other's effects in a mixture.

Table 4-4. Maximum detected CECs from this study compared to toxicological effects thresholds for aquatic organisms complied by National Oceanographic and Atmospheric Administration (NOAA) (ng/L).

Parameter	Maximum Detected in this Study	Lowest Reported Effect Level (QSAR, NOEC, LOEC, EC10, or EC50)		
		Fish	Algae/ Cyanobacteria	Crustacean
10-hydroxy-amitriptyline	0.164	Not reported		
Albuterol	9.11	1,158,000	5,837,000	5,730,000
Amitriptyline	0.332	20,900,000	11,985,000	5,000,000
Amphetamine	14.7	53,000,000	1,400,000	3,800,000
Atenolol	58.2	1,461,000,000	77,700	83,000,000
Benzoylecgonine	1.1	Not reported		
Benztropine	0.525	Not reported		
Caffeine	68.2	Not reported		
Ciprofloxacin	6.41	8,096,293,000	2,970,000	991,485,000
Cocaine	0.332	Not reported		
Cotinine	5.57	Not reported		

Parameter	Maximum Detected in this Study	Lowest Reported Effect Level (QSAR, NOEC, LOEC, EC10, or EC50)		
		Fish	Algae/ Cyanobacteria	Crustacean
DEET	8.2	Not reported		
Diphenhydramine	1.24	Not reported		
Erythromycin-H2O	5	17,109,000	3,898,000	49,800
Gemfibrozil	2.11	930,000	2,179,000	440,000
Metformin	786	33,170,000,000	39,000,000	130,000,000
Naproxen	3.77	24,279,000	22,952,000	2,620,000
Ofloxacin	1.56	Not reported	16,000	26,700,000
Oxolinic Acid	2.58	Not reported		
Paroxetine	4.59	2,000,000	15,699,000	3,000,000
Permethrin	1.57	Not reported		
Sulfadimethoxine	0.928	Not reported		
Sulfamethizole	1.69	Not reported		
Sulfamethoxazole	2.46	3,869,000	51,328,000	Not reported
Triamterene	0.347	Not reported		
Valsartan	3.93	Not reported		

QSARs = quantitative structure–activity relationship models; NOEC = no observed effect concentration; LOEC = lowest observed effect concentration; EC10 or EC50 = 10% or 50% effect concentration (the concentration in a laboratory or field toxicity test at which it is expected 10% or 50% of the test organisms would show an adverse effect).

4.4.2 Effects on Humans

Human prescribing information was queried from the Epocrates database (athenahealth, 2015) to assess the human health effects from exposure to detected CECs from ambient waters. This online reference is published by athenahealth and is commonly used by medical practitioners. Methodology was as follows:

- The lowest single therapeutic dose, regardless of administration route, was identified for each detected pharmaceutical (for example, albuterol is inhaled while ciprofloxin may be administered either intravenously or orally). For topical medications, the lowest therapeutic dose was assumed to be 0.1 mL of the lowest marketed product strength. Sulfadimethoxine is registered for veterinary use only. Chickens (and other animals) are given sulfadimethoxine to treat respiratory infections. One "dose" was calculated as a maintenance dose of 12.5 mg/lb in a typical 6.5-lb chicken (12.5 mg x 6.5 = 81.25 mg) (Petcarerx, 2015). Although the Food and Drug Administration has not approved sulfadimethoxine for humans, its mode of action and potency are comparable to other sulfa-based antibiotics.
- The lowest therapeutic doses were then compared to the potential ingested dose that would be consumed by drinking 2 L of ambient surface water containing the maximum detected CEC concentration in this study. This amount is a typical upper bound of adult daily consumption of water, reconstituted juice, cooking uses, and so forth. The comparison is shown in Table 4-5.

While the comparison cannot account for sensitive individuals who might be allergic to a particular drug or the possible side effects from exposure during sensitive life stages (such as in utero), it is a useful yardstick for understanding how detected CEC concentrations compare to doses of the same drugs that people and animals in King County take every day.

The comparison shows that ambient water exposures are at least six orders of magnitude below prescribed therapeutic doses, indicating that human health effects from exposure to detected CECs in ambient waters is very unlikely. Other researchers have used safety factors of 1,000 when conducting similar comparisons (Bull et al., 2011), but use of such factors appears unnecessary given the wide disparity observed between environmental doses through water ingestion and therapeutic doses.

Table 4-5. Single lowest therapeutic doses of detected CECs compared to potential dose from ingestion of 2 L of drinking water.

Parameter	Prescribing Information	A Single Lowest Therapeutic Dose (mg)[a]	B Lowest Single Therapeutic Dose (ng) (A x 1,000,000)	C Maximum Ambient Concentration Found (ng/L)	D Dose Consumed per 2L Water Consumed (C ÷ B)
10-hydroxy-amitriptyline	Amitriptyline metabolite	None, metabolite	N/A	0.164	N/A
Albuterol	2–4 mg PO tid–qid	2	2,000,000	9.11	0.0000091100
Amitriptyline	50–150 mg PO qhs	50	50,000,000	0.332	0.0000000133
Amphetamine	5 mg	10	10,000,000	14.7	0.0000029400
Atenolol	50–100 mg PO qd	50	50,000,000	58.2	0.0000023280
Benzoylecgonine	Cocaine metabolite	None, metabolite	N/A	1.1	N/A
Benztropine	1–2 mg PO/IM/IV qhs	1	1,000,000	0.525	0.0000010500
Caffeine	~100 mg per 8 oz coffee	100	100,000,000	68.2	0.0000013640
Ciprofloxacin	250–750 mg PO q12h	250	250,000,000	6.41	0.0000000513
Cocaine	Illicit drug	None	N/A	0.332	N/A
Cotinine	Nicotine metabolite	None, metabolite	N/A	5.57	N/A
DEET	Variable concentrations available (assumed 0.1 mL of 25% solution applied)	0.025	25,000	8.2	0.0006560000
Diphenhydramine	25–50 mg PO/IM/IV q4–6h prn	25	25,000,000	1.24	0.0000000992
Erythromycin-H2O	1,000 mg/day PO divided q6–12h	250	250,000,000	5	0.0000000400
Gemfibrozil	600 mg PO bid	600	600,000,000	2.11	0.0000000070
Metformin	850–1,000 mg PO bid	850	850,000,000	786	0.0000018494
Naproxen	250–500 mg PO q12h	250	250,000,000	3.77	0.0000000302
Ofloxacin	200–400 mg PO q12h	200	200,000,000	1.56	0.0000000156
Oxolinic Acid	12–20 mg/kg	12	12,000,000	2.58	0.0000004300

Parameter	Prescribing Information	A Single Lowest Therapeutic Dose (mg)[a]	B Lowest Single Therapeutic Dose (ng) (A x 1,000,000)	C Maximum Ambient Concentration Found (ng/L)	D Dose Consumed per 2L Water Consumed (C ÷ B)
Paroxetine	20–50 mg PO qam	20	20,000,000	4.59	0.0000004590
Permethrin	1% lotion and 5% cream available (assumed 0.1 mL of lotion applied)	0.001	1,000,000	1.57	0.00000314
Sulfadimethoxine	In fowl, initial dose of 25 mg/lb followed by four maintenance doses of 12.5 mg/lb/day (assumed maintenance dose in average 6.5-lb chicken)	81	81,000,000	0.928	0.0000000229
Sulfamethizole	1.5 to 4 mg/day	1.5	1,500,000	1.69	0.0000022533
Sulfamethoxazole	160 mg TMP PO q12h	400	400,000,000	2.46	0.0000000123
Triamterene	100 mg PO bid	100	100,000,000	0.347	0.0000000069
Valsartan	80–320 mg PO qd	80	80,000,000	3.93	0.0000000983

Prescribing abbreviations: IM = intramuscular; IV = intravenous; q = every; qd = once per day; bid = twice per day; tid = three times per day; qid = four times per day; qhs = once, at bedtime; pd = per day; qam = every morning; PO = orally; prn = as needed; TMP = dosing form (includes trimethoprim and sulfamethoxazole).

[a] One-tenth of a mL applied for topical use.

5.0 CONCLUSIONS AND RECOMMENDATIONS

This study describes the general presence-absence of a number of CECs in three different areas of King County surface waters: Lake Union/Ship Canal, Elliott Bay, and the Duwamish Estuary. Results from this study may be useful if these CECs are monitored again in the future. Monitoring CECs in the future and comparing them to this data set will help us to better understand if the concentrations of these chemicals are changing over time or if compounds that weren't detected this time might be in the future or vice versa. They may also be used to help address future questions about CECs, sensitive species, and potential human health concerns or changes in pharmaceutical use patterns.

The following sections present the conclusions and recommendations from this study in more detail.

5.1 Conclusions

A total of 26 different CEC were detected out of the 140 that were analyzed. No obvious patterns of detection by study area or sample depth emerged, and the (statistical) power to detect differences between depths or study subareas was generally weak. One exception was the notably higher concentrations of metformin detected in Elliott Bay compared to levels found in the Duwamish Estuary and Lake Union/Ship Canal.

Findings across all study areas were as follows:

- Metformin had the highest concentrations (up to 786 ng/L) and a high frequency of detection (27 of the 51 samples, or 52.9 percent).
- Caffeine and sulfamethoxazole (an antibiotic) were also commonly detected in 25 (50 percent) and 28 (55 percent) of the 51 samples, respectively.
- The most commonly detected CEC was benzoylecgonine (the metabolite of cocaine), which was found in 48 of the 51 samples (94.1 percent).
- The maximum detected concentration of benzoylecgonine was 1.1 ng/L; this is just above the MDLs, which ranged from 0.2 ng/L to 0.4 ng/L

In general, CECs detected in this study were comparable and within the range of concentrations found elsewhere. For most CECs detected in this study, the concentrations were lower than those detected in Boulder Creek (Colorado), Minnesota lakes, the Delaware River, the Santa Ana River in California, and San Francisco Bay. One exception was albuterol, an inhaled bronchodilator; concentrations appear to be slightly higher in King County waters (9.11 ng/L) compared to the next highest detected concentration (1 ng/L) in San Francisco Bay.

Adverse impacts to ecological receptors or people exposed to the ambient waters in the study areas are not expected even at the reported maximum concentrations:

- All detected concentrations in this study are orders of magnitude below literature-reported toxicity values, which were mostly associated with mortality endpoints and other obvious impacts. This finding suggests that overt impacts to aquatic organisms associated with exposure to the detected CECs are not likely.
- For all detected CECs, drinking water doses (2 L) were four or more orders of magnitude lower than a therapeutic dose. While this analysis was unable to account for sensitive groups, subpopulations, or the interactions of mixtures, it appears that human health impacts through CEC exposures in ambient waters are highly unlikely.

5.2 Recommendations

While a relatively new concern, these CECs are of great interest to the public and now many researchers are publishing thousands of papers annually. As with any topic, future work to better understand the extent and impacts of these CECs would best be guided by robust and specific study questions and hypotheses developed during the planning stages.

While the concentrations found in the study areas do not appear to be cause for concern for impacts to fish, other aquatic life, or humans, future investigators should consider doing a more focused literature review of available sub-lethal endpoints and recent publications describing genetic, behavioral, or other potential important ecological endpoints. Such a review may wish to assess modes of action of select CECs to enhance understanding of relevant effects and to identify data gaps (Escher et al., 2005).

This study used numerous secondary sources to describe the relative magnitude of ecological effects. Additional literature reviews of primary literature sources may help in prioritizing management attention on CECs and potential receptors of greatest concern. King County may wish to partner with agencies, such as USGS and NOAA, with expertise in understanding the impacts of contaminants on tissues and organisms (for example, the potential behavioral impacts of CECs on endangered species like salmon or steelhead). The current dataset is unable to rule out the potential impact of some CECs on especially sensitive or high priority aquatic organisms such as salmon.

With a refined understanding of possible ecological impacts, such as behavioral or olfactory changes in adult and juvenile fish, more refined and targeted sampling and analysis of specific CECs and habitats during sensitive life stages would provide information on ecological problems and help to focus management solutions.

Also, detected CECs, and possibly other CECs, may be present in sediments or tissues in the study areas. Future investigations could examine a broader range of environmental media for CECs, including sediment, fish, and other animals, especially sessile animals such as mussels. These other media may have different concentrations and potential effects on organisms from different uptake pathways.

Other researchers have used patterns and shifts in illicit drug and drug metabolite concentrations in sewers and ambient waters to help answer public health questions about

illicit drug use and the effectiveness of interventions and other actions (Boles and Wells, 2010). The main metabolite of cocaine (benzoylecgonine) and cocaine itself were commonly detected in ambient waters (with frequencies of detection of 94 percent and 29 percent, respectively). King County may wish to partner with public health agencies to examine cocaine and other illicit drugs in both ambient waters and wastewaters to help target drug intervention strategies. King County Environmental Laboratory's research chemists may wish to develop in-house capabilities to analyze CECs of particular public interest (such as illicit drugs and drug metabolites) to help maintain their regional leadership.

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