DEPARTMENT OF
ECOLOGY
State of Washington

# Lake Washington Area Regional Background

## Seattle, WA

*Data Evaluation and Summary Report*

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# Regional Background

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## Data Evaluation and Summary Report

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# Acronyms and Abbreviations

| | |
|---|---|
| AICc | Aikake Information Criterion |
| As | arsenic |
| CERCLA | Comprehensive Environmental Response, Compensation, and Liability Act |
| CoCs | chemicals of concern |
| cPAH | carcinogenic polycyclic aromatic hydrocarbon |
| CSL | Cleanup Screening Level |
| CSO | combined sewer overflow |
| DOT | Washington State Department of Transportation |
| Ecology | Washington State Department of Ecology |
| EIM | Environmental Information Management System |
| EM | expectation-maximization |
| Hg | mercury |
| i.i.d | independently and identically distributed |
| KM | Kaplan-Meier |
| km | kilometer |
| LPL | lower prediction limit |
| m | meter |
| NADA | Nondetects and Data Analysis statistical software package |
| NOAA | National Oceanic and Atmospheric Administration |
| PCA | principal components analysis |
| PCB | polychlorinated biphenyl |
| ppb | parts per billion |
| ppm | parts per million |
| PQL | practical quantitation limit |
| ProUCL | statistical software package for analyzing environmental data sets |
| QQ | Quantile-Quantile |
| SCO | Sediment Cleanup Objective |
| SCUM II | Sediment Cleanup User’s Manual II |
| SMS | Sediment Management Standards |
| TBT | tributyltin |
| TEC | toxic equivalent concentration |
| TEQ | toxic equivalent quotient |
| TOC | total organic carbon |
| UCL | upper confidence limit |
| UTL | upper tolerance limit |
| UPL | upper prediction limit |
| WAC | Washington Administrative Code |

# 1.0 Introduction

In early 2013, the Washington State Department of Ecology (Ecology) revised the Sediment Management Standards (SMS) (Chapter 173-204 WAC) to establish a new framework for identifying and cleaning up contaminated sediment sites. A key component of this framework is the concept of regional background sediment concentrations, which can serve as the Cleanup Screening Level (CSL) for sediment sites. During the rule revision, the advisory group recommended that Ecology be responsible for establishing regional background sediment concentrations for areas of the state. This report provides Ecology's evaluation of existing data for the Lake Washington Area to establish regional background.

## 1.1 Regional Background

For a number of bioaccumulative chemicals, risk-based values protective of human health and upper trophic levels fall below the natural and regional background concentrations defined in the SMS (WAC 173-204-505). Sediments receive chemicals from potentially hundreds of sources, including a mix of permitted and unpermitted stormwater, atmospheric deposition, and historical releases from industrial activities. In urban areas with developed shorelines, chemical concentrations in sediment are frequently higher than natural background concentrations.

The SMS rule includes a two-tiered framework used to establish sediment cleanup levels. It incorporates *natural background* as one component of the Sediment Cleanup Objective (SCO), and a new term and concept—*regional background*—as a component of the CSL. The SMS rule provides a definition for regional background in WAC 173-204-505(16) and parameters for establishing it in WAC 173-204-560(5):

> **"Regional Background"** means the concentration of a contaminant within a department defined geographic area that is primarily attributable to diffuse sources, such as atmospheric deposition or storm water, not attributable to a specific source or release. (WAC 173-204-505(16))

The SMS is intended to provide flexibility in establishing regional background on a case-by-case basis and does not prescribe specifically how regional background should be established. Ecology's approach to establishing regional background has evolved over time by working on the first bays, and receiving comments afterward from stakeholders and tribes. Current guidance for establishing regional background is based on these discussions. Completed studies can be found in Chapter 10 of the *Sediment Cleanup User's Manual II* (SCUM II) (Ecology 2015a).

## 1.2 Lake Washington Area Regional Background

To date, Ecology has established regional background concentrations for Port Gardner, Bellingham Bay, and the North Olympic Peninsula (Ecology 2014, 2015b, 2016) using methods that rely primarily on collecting new data. However, SCUM II also allows regional background to be established using existing data if the data are sufficient and statistically robust. Regional background proposed in this report is based on existing sediment data collected from Lake Washington, Union Bay, the Montlake Cut, Portage Bay, and Lake Sammamish, collectively called the Lake Washington Area. This evaluation was limited to those chemicals for which there are adequate existing data: arsenic, mercury, and carcinogenic polycyclic aromatic hydrocarbons (cPAHs). If there is a need for regional background concentrations for additional bioaccumulative chemicals in this area, new sediment data will need to be collected and analyzed.

In addition, in cases where an entire water body may be directly influenced by identifiable sites and sources, the SMS includes a provision to establish regional background using data from an alternative but similar geographic area(s) that is not directly influenced by sources as a substitute:

> **WAC 173-204-560 (5)(d):** Calculation of regional background for a contaminant must exclude samples from areas with an elevated level of contamination due to the direct impact of known or suspected contaminant sources, including areas within a sediment cleanup unit or depositional zone of discharge.
>
> **WAC 173-204-560 (5)(f):** If a water body is not beyond the direct influence of a significant contaminant source, the department may use alternative geographic approaches to determine regional background for a contaminant. Several factors must be evaluated when determining an alternate geographic approach including:
>
> > (i) Proximity of sampling to the site;
> > (ii) Similar geologic origins as the site sediment;
> > (iii) Similar fate and transport and biological activities as the site; and
> > (iv) Chemical similarity with the site.

Consistent with this provision, the Lake Washington Area was selected as a surrogate for freshwater urban lakes in Water Resource Inventory Area 8 (WRIA 8) that may be within the direct influence of sites and sources, such as Lake Union. The Lake Washington Area is an appropriate surrogate because it a) receives diffuse urban sources, b) is relatively less impacted from chemical contamination than other urban lakes such as Lake Union, c) is geographically proximate, and d) within the same watershed and geologic units.

Regional background from the Lake Washington Area is considered applicable to urban lakes in King County WRIA 8, including Lake Union, Lake Washington, Lake Sammamish, and the Lake Washington Ship Canal area east of the Hiram Chittenden Locks. Note that regional background established in this report is not applicable to river systems, or less developed suburban or rural lakes.

This report represents Ecology's approach for a) using existing data to establish regional background concentrations in a lake system, and b) establishing the first regional background concentrations for freshwater urban lakes. The draft of this report (released September 2016) included Section 5, summarized guidance for using this approach in other areas. Chapter 10 in SCUM II (Ecology 2015a) will include an updated summary of the recommended approach, along with important limitations for using this approach in other areas. The SCUM II updates were informed by public comments received both in writing and during a technical workshop conducted in Seattle in October 2016.

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# 2.0 Conceptual Lake Model

The Lake Washington Area, in which existing data were evaluated to determine regional background concentrations, was defined as Lake Washington, Union Bay, the Montlake Cut, Portage Bay, and Lake Sammamish (Figure 1). These water bodies are hydraulically connected, considered representative of mixed urban uses, and connected to the more industrial areas of Lake Union, the Ship Canal, and Salmon Bay before discharging to Puget Sound.

Lake Sammamish is predominantly fed by Issaquah Creek and drains to Lake Washington via the Sammamish River. Lake Washington receives inputs from the Sammamish and Cedar Rivers, which are developed watersheds, and from mixed residential, commercial, and urban water-dependent uses characteristic of dense urban areas.

Areas within Lake Union were excluded because the majority of the lake is directly influenced by numerous sources (industry, residential and industrial stormwater, water-dependent uses, and cleanup sites) and due to the highly altered nature of Lake Union, the Ship Canal, and Salmon Bay.

## 2.1 Geography and Land Use

Lake Washington is the largest lake in King County, with a surface area of 87.6 $km^2$ and draining an area of 1448 $km^2$ (King County 2015). The lake is approximately 35.4 km long and 3.2 km wide and surrounded by cities: Seattle to the west; Kenmore to the north; Kirkland, Bellevue, Medina, and several smaller cities to the east; and Renton to the south (Figure 1). Mercer Island, a large residential island approximately 34 $km^2$ in total area, occupies the southeast area of the lake. Lake Sammamish is located east of Lake Washington and drains an area of 250 $km^2$. At approximately 11 km long and 2 km wide, Lake Sammamish has a surface area of 21 $km^2$ and is surrounded by Bellevue to the west; Redmond to the north; and Issaquah and Sammamish to the south and east, respectively.

Land use around Lake Washington and Lake Sammamish is largely high- and medium-density urban residential, with some commercial/industrial and urban parkland (King County 2008). Typical nonresidential uses include marinas, shopping centers, restaurants, and recreational areas such as beaches and parks. On Lake Washington, a floatplane base is located in Kenmore, and the Renton Municipal Airport and Boeing are located at the south end. Historic land uses on Lake Washington were more industrial than today, which included boatyards and shipyards; landfills, sawmill and log rafting; wood treating facilities; coal loading and barging; the Shuffleton power plant; and U.S. Navy and NOAA facilities. Two freeways cross Lake Washington on floating bridges: Interstate 90 to the south and State Route 520 to the north.

There are federally authorized ship navigation channels at the mouths of the Cedar River and Sammamish River, which are infrequently dredged for navigation and flood control purposes.

Located on the west-central side of Lake Washington, Union Bay is surrounded by residential areas, the University of Washington, and wetlands. Linking Union Bay to Portage Bay is the Montlake Cut: a 760-meter long, human-made channel completed in 1916 to allow passage between Lake Washington and Puget Sound via Lake Union and the Lake Washington Ship Canal. Portage Bay contains yacht clubs, marinas, and numerous houseboats. Surrounding areas include residential neighborhoods, the NOAA Fisheries Science Center, and the University of Washington. West of Portage Bay, I-5 runs north-south. South of Portage Bay, SR 520 runs east-west across Lake Washington, and connects I-5 to I-405.

Both Lake Washington and Lake Sammamish are within the Usual and Accustomed Fishing Areas of the Muckleshoot, Suquamish, and Tulalip Tribes. They are also used for recreational fishing, boating, swimming, and other recreational and commercial activities.

## 2.2 Hydrology and Bathymetry

Lake Washington receives the majority of its inflows from the Cedar River in Renton (57%) and the Sammamish River in Kenmore (27%), with numerous smaller creeks providing the rest. The watershed is primarily developed (67%), with the exception of the upper Cedar River watershed, which provides Seattle's water supply. Lake Washington’s outlet is through Union Bay, the Montlake Cut, and Portage Bay into Lake Union, flowing then through the Hiram M. Chittenden Locks and Salmon Bay to Puget Sound (King County 2015). Historically, Lake Washington was landlocked before construction of the Montlake Cut and Lake Washington Ship Canal in 1916. The new canal lowered the lake by 3 meters and diverted Cedar River into the lake. Lake Sammamish is predominantly fed by Issaquah Creek (~70%) and the main drainage to Lake Washington is via the Sammamish River.

Lake Washington is a glacially formed lake with steeply sloping sides, averaging 33 meters deep and 65.2 meters at its deepest point. Water levels in the lake are controlled by the Hiram M. Chittenden Locks, with an average about 7 meters above mean lower low tide in Puget Sound. The lake has a residence time of about 2.4 years (King County 2015). Lake Washington is strongly thermally stratified in the summer, with distinct upper, middle, and lower layers. Convection and wind mixing produce isothermal conditions in the lake in winter. No information on currents in the lake is available (Ecology 2014).

## 2.3 Sedimentation, Grain Size, and Organic Carbon

Figures 2 and 3 show total organic carbon and grain size for all data in this geographic region, which were downloaded from Ecology's Environmental Information Management System (EIM). Lake Washington and Lake Sammamish sediments are typically a fine silt or mud, with generally coarser sediments near river mouths, high-traffic areas of the Montlake Cut, and nearshore areas. Non-native clean sand has been imported in some shoreline areas to create swimming beaches and parks.

There is very little specific data on sedimentation in the lakes. While much of these two lakes likely receive little sedimentation (especially since source control has reduced eutrophication), areas near the river mouths receive periodic siltation and require occasional dredging for flood control and navigation. Deeper lake areas likely receive slow siltation through deposition and erosion from nearshore areas.

## 2.4 Unrepresentative Areas

Some areas were not considered representative due to unusual sediment grain size or organic carbon. (See Figures 2 and 3). These unrepresentative areas include:

- <u>Swimming Beaches</u>. In a number of areas, sediments were imported to enhance swimming beaches. These sediments are not native, coarser-grained, generally very clean, and more in the range of natural background than regional background. King County has sampled many of these beaches over the years. The distribution of samples from these areas was generally within the range of Puget Sound natural background and freshwater sediment reference areas.

- <u>High TOC Areas.</u> The following areas were identified and considered not representative of sediments in the Lake Washington as a whole:

    - Wetlands. Some areas around the shoreline of Lake Washington contain wetlands or aquatic vegetation such as milfoil that could result in elevated TOC.

    - Other areas. Other areas with unusually high TOC were identified and then determined to be unrepresentative.

## 2.5 Sites and Sources

The SMS rule states that samples within or immediately adjacent to cleanup sites cannot be used to establish regional background for site-related CoCs. Samples along the shoreline with the same site CoCs (As, Hg, cPAHs) were excluded based on potential sources and known locations of sites, regardless of chemical concentrations. A number of sediment sites and other sources have historically been or are currently located in Lake Washington. Lake Sammamish does not have cleanup sites along or near the shoreline that are considered significant contaminant sources to sediment. Consistent with the SMS rule, Ecology focused on identifying the sites and sources that had a relatively high potential to directly influence existing data concentrations. These sites and sources are described below from north to south and shown in Figures 4-6. This list is not intended to include all potential sources, but rather those with the high potential to directly influence sediment with nearby existing data. Other potential sources in the region, such as other stormwater drainages and nonpoint sources, are not included here.

- <u>Kenmore marinas.</u> The area including North Lake Marina and Harbour Village Marina at the northeast end of Lake Washington, as a result of known PAH, TBT, phthalates, and dioxins/furans; boat repair and refueling activities; and large storm drains that empty into these enclosed areas (Ecology 2013, DMMP 2013). Harbour Village Marina is a MTCA cleanup site.

- <u>Kenmore Air Harbor.</u> One of the largest seaplane bases in the world, Kenmore Air Harbor conducts plane refueling and maintenance at its Kenmore location between the marinas and the barge area at Lakepointe. Minimal data is available for this area.

- <u>Former landfill and barge area around Kenmore Industrial Park (Lakepointe).</u> Kenmore Industrial Park was an upland MTCA cleanup site historically used as a landfill for industrial debris. Petroleum hydrocarbons and metals were found in soils and groundwater at this site, and PAHs have been found in sediments in the barge area north of it (Ecology 2001).

- <u>Former Naval Station and NOAA facilities at Sand Point.</u> Both of these facilities had docking areas at which low levels of metals and PAHs were found in early sediment investigations in the 1990s.

- <u>Quendall Terminals.</u> Elevated PAH concentrations were found in sediments offshore of this former wood-treating site along the eastern shore of Lake Washington (Anchor and Aspect 2012). This is a CERCLA cleanup site.

- Renton Coal Terminal. Early in Seattle's history, a coal terminal was located at the southeast end of the lake, along the eastern shoreline (Bagley 1916). Sediments in this area continue to have elevated PAHs, although much of the area has been redeveloped as a waterfront park.

- Puget Power & Light Shuffleton Power Plant. Studies in the 1990s found higher concentrations of PAHs and PCBs near this former oil-fired power plant at the southeast end of Lake Washington.

- SR 522 stormwater outfalls. Areas outside the marina.

- SR 520 runoff. Areas at the end of a runoff channel into Yarrow Bay from an SR 520 storm drain through a swale north of the highway.

- I-90 runoff. Areas within a swale south of the I-90 Bridge receiving runoff from I-90 storm drains at the south end of Mercer Slough Park.

- Boeing/Renton Airport runoff. Areas immediately offshore of the runways.

- King County Montlake CSO/Montlake Bridge**.** Areas near the Montlake Bridge CSO and on either side of the bridge.

- King County University Regulator CSO. Areas near the University Regulator CSO on the north side of Portage Bay.

- City of Seattle CSO and storm drain in Portage Bay. Areas near a City of Seattle CSO and storm drain at the base of Brooklyn Avenue on the northwest side of Portage Bay.

- City of Seattle CSO near I-5. Areas near a City of Seattle CSO just east of I-5 along Northlake Way.

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# 3.0 Data Screening and Analysis

To ensure consistency with the SMS, existing data for the geographic region were downloaded from EIM and screened (excluded) from the data set used to calculate regional background (Figures 4-6, Tables 4a – c). Some of the screening steps described below are the same as those used for previous regional background studies in which new data was collected (such as determination of geographic scope; exclusion of unrepresentative areas; exclusion of areas under the direct influence of sites and sources; outlier analysis; and precision analysis). Additional or modified screening steps (Steps 1 and 3 below) addressed issues with existing data. Taken together, these three screening steps were performed to exclude data that was not considered representative of a regional background distribution:

1. First screen of the data set was to ensure samples met adequate quality control and assurance specific to recency, depth, replicates, and detection limit issues (Sections 3.1.1 through 3.1.3, Figures 4–6, Tables 4a–c).

2. Second screen of the data set was to ensure potential and known sources were not directly influencing samples and that samples with high TOC ($\geq$ 15%) were excluded (Section 3.2, Figures 4–6, and Tables 4a–c).

3. Third screen of the data set was through statistical analysis and included analysis for independence, population separation to obtain a representative distribution (normally addressed during sampling design for new studies), precision, principal components analysis, and identifying outliers (Tables 1 & 2; Figure 8; Appendix B).

It was determined that sufficient data existed in EIM to evaluate if regional background could be established for cPAHs, arsenic, and mercury. However, the congener data for PCBs and dioxins/furans outside known cleanup sites are insufficient for calculating regional background. New data would need to be collected to calculate regional background values for these CoCs.

## 3.1 First Data Screen – Quality Control/Assurance

### 3.1.1 Data Recency

Initially, all data dating back to the year 2000 were downloaded from EIM for evaluation. Inspection and subsequent statistical analysis of the data using a population comparison identified that data sets earlier than 2004 were elevated throughout their distribution compared to more recent data (Figure 9). Substantial source control efforts by King County and the City of Seattle over the last 20 years, along with small but measurable deposition of cleaner sediments during that time, may account for the observed lower concentrations in more recent data sets. In addition, the post 2003 data set for cPAHs had a smoother distribution with fewer high-end outliers. Therefore, it was determined that data sampled earlier than 2004 were not representative of relatively current conditions and a standard ~10-year recency cutoff was used.

### 3.1.2 Detection Limits

Data that were undetected above the PQL-based cleanup level identified in SCUM II's Chapter 11 were excluded to avoid bias from elevated detection limits.

### 3.1.3 Depths, Time Series, and Replicates

Several initial screens were applied to obtain the most recent surface samples at each location:

- Data that were not from surface samples, or that were composited over more than 2 feet in depth, were excluded.

- When multiple samples were collected at the same station over time, only the most recent sample was kept.

- When replicate samples were collected at the same station at the same time, the data were averaged.

## 3.2 Second Screen – Sites and Sources

The intent of the SMS definition of regional background is to avoid the *direct* influence of known sites and sources from the calculation of regional background. Therefore, data near sources that Ecology determined had high potential to directly influence samples were excluded. As described in detail in Section 2.5, the following sources were identified:

- Current and historic sites with PAHs and/or metals.
- Areas potentially directly influenced by historic uses but not formally designated as cleanup sites.
- CSOs and storm drains associated with elevated concentrations and a decreasing gradient away from the source.
- Swales and channels containing concentrated stormwater runoff from the major roadways including from bridges.
- Areas associated with airport runoff.
- Areas with high TOC ($\geq$ 15%) which included wetlands.

## 3.3 Third Screen – Statistical Analysis

The resulting data set was evaluated further from a statistical perspective to address issues specific to using existing data, as described below and in Appendix B. Tables 4a–c in Appendix A and Figures 4–6 provide the data set for these analytes, and differentiate which data were screened out by age (older than ~10 years, e.g., from 2003 or older); depth (non-surface sediment); high TOC; non-detect issues; and potential and known sources. This screened data was then used to conduct statistical analyses (Tables 4a–c). The entire unscreened data set from this geographic region can be downloaded from EIM. A review of station concentrations for cPAHs, arsenic, and mercury indicated that the screening approach described in Sections 3.1 and 3.2 was appropriate for all three chemicals. This is because the data tended to have similar trends in generally the same areas.

### 3.3.1 Sample Independence

A spatial autocorrelation analysis was conducted to identify the autocorrelation distance, which is the minimum distance required between samples to consider the results statistically independent. Samples that were spatially isolated or clear outliers were temporarily removed for this analysis to reduce variability that would disproportionately affect the model.

The three analytes were not expected to have identical spatial concentration distributions, because of the chemicals' long and varied history of sources within the lake system. As anticipated, the autocorrelation distances were different for each analyte: 50 meters (50-m) for mercury, 100-m for arsenic, and 250-m for cPAH TEQ. A detailed description of the autocorrelation analysis methods and results can be found in Appendix B.

Generally, clusters of samples within the autocorrelation distance are assumed to have been influenced by the same sources and would be expected to have similar concentrations. This was not always the case, however, and there were some sample clusters within which concentrations varied by more than an order of magnitude. Additional analysis was required to evaluate these clusters and determine how to select or average sample results to include in the final data set (see Section 3.6 and Appendix B).

### 3.3.2 Identification of Subpopulations and Outliers

The studies that make up the data set include samples from several distinct and sometimes overlapping distributions. A detailed analysis of the data set for each analyte was conducted to exclude outliers and isolate the subset of data that most closely represents the SMS definition of regional background (Tables 1 & 2; Appendix B).

A population separation analysis was conducted to identify the regional background population from within the mixture of subpopulations present in the data set. This analysis used likelihood methods to find the most likely breakpoints for mixtures of subpopulations, and prediction intervals on independent data to interpret results that were spatially clustered. The following steps were carried out:

1. Preliminary distinct subpopulations were identified using likelihood methods, using only the independent samples (i.e., those samples not part of a cluster), and based on the minimum autocorrelation distance identified in Section 3.5. Robust prediction limits were calculated for each subpopulation. Prediction limits are the expected upper and lower limits for individual future observations from each population. These limits are robust because the effects of extreme values are down-weighted in the estimation process.

2. These limits were then applied to each sample cluster. Individual samples within each sample cluster were allocated to their appropriate subpopulation.

3. Samples located closer together than the autocorrelation distance and within the same subpopulation were averaged and the average treated as an independent data point.

4. Finally, a population separation analysis based on likelihood methods was repeated on the combination of the independent samples and the cluster averages calculated in Step 3 (above).

For cPAH TEQs, three primary subpopulations and four higher-concentration samples were identified (Table 1). The lowest concentration subpopulation was made up mainly of swim beach samples that were believed to contain imported clean sand. Nine additional samples fell within the range of Puget Sound natural background concentrations (SCUM II, Table 10-1) and are presumed to fall within natural background of the area of interest. Twenty-three samples found in depositional areas and near urban shorelines were considered to appropriately represent regional background as defined in the SMS. Finally, four additional samples represent high-concentration samples associated with potential sources and are considered outliers.

For the metals, there were very few samples with concentrations above Puget Sound natural background concentrations (Table 2). The arsenic data set only had nine values similar to or exceeding the Puget Sound natural background 90/90 UTL, ranging from 13 to 70 ppm. The arsenic concentrations within the Puget Sound natural background distribution were fairly homogeneous and similar between the clean swim beach samples and the non-swim beach samples. There appeared to be a signal for arsenic with concentrations > 11 ppm that may represent regional background.

The mercury data set had thirteen samples with values similar to or exceeding the Puget Sound natural background 90/90 UTL, ranging from 0.14 to 0.39 ppm (Table 2). Within the mercury data set, there was some distinction between swim beach samples and non-swim beach samples. The mercury concentrations similar to or exceeding Puget Sound natural background represent a range that may be representative of regional background.

However, with the limited number of samples for both mercury and arsenic, conclusive regional background values cannot be established without more data. Ecology prefers a sample size of approximately 25 for each CoC to establish regional background.

### 3.3.3 Precision

Throughout the evaluations above, the precision of the resulting data set was used as one measure of whether the data set a) could be considered a single population, and b) was sufficiently cohesive to provide a reasonable representation of regional background. This is important because a data set with low precision will have broader tails and higher upper percentiles. While it was considered unlikely that a sample population made up of existing data would be as precise as one resulting from a single synoptic sampling event, it was considered important to improve the precision as much as possible through the steps described above to obtain the best measure of regional background.

After identifying the regional background data sets through the evaluations described above, the precision of each data set was calculated as the width of the 95 percent upper confidence limit (95 UCL) on the mean, divided by the mean. Precision of the mean expressed in this way is a common method for quantifying uncertainty in the data set used to calculate the 90/90 UTL.

The data set representing regional background for cPAH TEQ was evaluated in ProUCL to determine the most appropriate distribution, then associated summary statistics were calculated (Table 3). The analysis was not conducted for arsenic or mercury because there was too little data within the range of regional background (sample sizes of six and ten respectively; Table 2). Using the samples with values between 38 to 240 ppb for cPAHs, the precision is 25%.

### 3.3.4 cPAH Summing

Kaplan-Meier (KM) TEQs were calculated for the cPAHs in each sample consistent with the recommendations in SCUM II. The KM sums reported for the retained TEQ data were calculated using R version 3.2.2 (R Core Team 2015) using the *cenfit* function from the NADA package (Lee 2013). The KM sum was calculated as the KM mean multiplied by the number of congeners (Helsel 2012). The following rules were applied to calculate and qualify the final KM TEQs:

- If the number of non-detected cPAHs for a sample exceeded 50 percent (4 or more out of

7), the KM TEQ was qualified as a "less than" value (L-qualified), followed by the number of non-detected values. For example, if 4 of the 7 cPAHs were undetected, the detection frequency would be 57% and the KM TEQ would be calculated and qualified with "L4."

- If the lowest toxic equivalent concentration (TEC) was based on a non-detected value, the positive bias in the KM estimate was adjusted downwards using Efron’s bias correction (Klein and Moeschberger 2003). This method treats the lowest ranked value as detected even if it was reported as a non-detected value.

- Normally, if the highest value is a non-detect, it is excluded by the statistical software used to conduct KM calculations. However, all of the cPAHs must be included when calculating a TEQ value. Therefore, the highest TEC value was always treated as a detected value (at the detection limit) for calculating the KM TEQ. The TEQ was qualified with an L if the highest TEC was originally a non-detected value.

- All L-qualified TEQ values were treated as censored (upper-bound) values in the distributional assessments and when calculating summary statistics across samples.

# 4.0 Regional Background Concentrations

## 4.1 Data Distributions

Overall, the following observations regarding the chemistry data set can be made. Many of these observations may apply to other urban areas and existing data sets.

- For this geographic area, there was a relatively large amount of existing data for chemicals of concern that were sampled to assess benthic toxicity, including PAHs and metals. There was very limited data (number of samples and geographic coverage) for chemicals of bioaccumulative concern, such as PCB congeners and dioxins/furans. Because of the past emphasis on benthic toxicity, historic data sets may not include data for bioaccumulative chemicals that are most relevant to establish regional background.

- In this existing data set, much of the data was collected for specific monitoring objectives other than establishing regional background. For example, data were collected to a) evaluate the safety of swimming beaches, b) monitor sediment quality near stormwater and combined sewer overflow outfalls, c) evaluate general sediment quality, and d) collect data for dredged material evaluations or remedial investigations. This tended to bias the data set to nearshore areas and areas that were unusually clean (swimming beaches) or with variable and higher concentration stations (near sources and sites) (Figures 4-6).

  In contrast, the previous regional background studies using newly collected data had the objective to characterize general concentrations in a bay or other area by sampling in an unbiased, systematic manner with good spatial coverage. For this area of interest, it would have been preferable to have more data in offshore areas of the lake where concentrations are expected to be more consistent and representative of long-term influences from the surrounding urban areas.

- The cPAH data set, in particular, was determined through statistical analysis to be composed of several independent populations and had a number of unrepresentative high-concentration samples and two clear outliers (1900 ppb, Lake Washington and 1500 ppb, Lake Sammamish; Table 1). While nearly all sources of higher concentration samples could be identified, the historic use of this area for coal mining and transport, industrial and water-dependent uses, and the patchy station locations made it difficult to be certain of sources in all cases. Professional judgment was carefully used to select the data population that best reflected the SMS definition of regional background (Table 1).

- While metals concentrations were generally lower than expected given past reports of metals enrichment in the lake, the reasons for this are unclear. One possibility is that concerted source control efforts by the City of Seattle and King County have reduced concentrations in the lake over the last several decades. In addition, the Asarco smelter was reported as the source of high metals concentrations in lake sediments in the 1970s, including arsenic and mercury (Barnes and Schell 1973; Crecelius 1975; Crecelius and Piper 1973), and that source has been discontinued for 30 years. However, as was the case for cPAH data, most of the metals data set was in nearshore areas. There are insufficient data in the offshore areas of the lake to draw strong conclusions. The few data that exist for mercury and arsenic suggest that offshore areas may have higher concentrations than nearshore areas, confirming past reports that suggested settling of finer-grained, higher-concentration sediments in the deeper areas of the lake. However, the highest concentrations in the current data set are still substantially lower than those reported in the 1970s (Crecelius 1975).

## 4.2 Lake Washington Area Regional Background Values

Table 3 presents the Lake Washington area 90/90 UTL value for cPAH TEQs alongside the Puget Sound 90/90 UTL natural background value (SCUM II, Chapter 10). While Puget Sound natural background concentrations may not be directly applicable to freshwater urban lakes, they are presented here for general comparison and discussion. The 90/90 UTL value was calculated in ProUCL 5.0 (USEPA 2013) and is consistent with the recommendations in SCUM II, Chapter 10.

The following conclusions regarding regional background can be drawn from these results:

- The regional background value for cPAHs based on the 90/90 UTL for cPAHs was calculated as 210 µg TEQ/kg. The data set on which this value is based is fairly limited in size (n = 23) for the area it is intended to characterize and is best described by a skewed gamma distribution.

- The data set for arsenic and mercury included a limited number of samples that may be representative of regional background (Table 2). This is because most of the data was within the range of concentrations for Puget Sound natural background and freshwater reference sites. Due to the limited data set, Ecology will not establish regional background values for these CoCs until additional data is collected.

- Ecology will remain receptive to reviewing new data that may be analyzed in the Lake Washington Area in the future. If new data is received, and Ecology deems it sufficient, we will consider reviewing and revising regional background.

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# Tables

Tables 1–3 (pp. 24–25)

Tables 4a–c are found online in Appendix A (p. 36)

Tables 5–15 are found in Appendix B of this report (pp. 57–65)

**Table 1:** cPAH TEQ values for subpopulations within the data set.

Population 1 was used to calculate regional background. See Figure 8 for more detail.

| Location Description | cPAH TEQ(ppb) | % Fines |
|---|---|---|
| **Lowest Concentration Samples (mainly swim beach samples)** | | |
| 29 independent observations or cluster averages | <2.6 – <6.7 | <1 – 21 |
| **Apparent Natural Background Samples** | | |
| 15 independent observations or cluster averages | 11 – 31 | 2 – 85 |
| **Selected Regional Background Samples (Population 1)** | | |
| Boeing (average) | 38 | 79 |
| Middle of Lake Sammamish, northern portion | 38 | 80 |
| Portage Bay (near the University of Washington; average) | 42 | -- |
| Near Newport Yacht Club | 45 | 19 |
| Western shoreline of Lake Sammamish, offshore near Squibbs Creek (average) | 50 | 1 |
| Harbor Village Marina | 58 | 6 |
| Lake Sammamish State Park, nearshore west of boat launch | 72 | 78 |
| Middle of Lake Washington, west of Mercer Island | 72 | 77 |
| Middle of Lake Washington, between the southwest shoreline of Mercer Island /Rainier Beach | 75 | 75 |
| Kenmore Navigational Channel (average) | 76 | 41 |
| Boeing (average) | 88 | 64 |
| May Creek | 92 | 14 |
| Offshore of the northeastern shoreline of Lake Sammamish | 94 | 64 |
| South of Pleasure Point | 100 | 16 |
| Middle of Lake Washington between Magnuson Park and Kirkland | 100 | 80 |
| Montlake Cut | 110 | -- |
| Pleasure Point | 130 | 20 |
| Middle of Lake Washington, north end, between Lake Forest park and Inglewood Golf Club | 130 | -- |
| Nearshore, Idylwood Park in Lake Sammamish | 150 | <1 |
| McAleer Creek | 160 | 12 |
| South of Newcastle Beach Park | 170 | 56 |
| Boeing | 220 | 53 |
| South of Newcastle Beach Park | 240 | 41 |
| **High-Concentration Samples** | | |
| 5 independent observations or cluster averages | 330 – 1,900 | 5 – 70 |

**Table 2:** Arsenic and mercury data near or above Puget Sound natural background concentrations.

| Location Description | ppm | % Fines |
|---|---|---|
| **Samples near or above Puget Sound Natural Background 90/90 UTL for Arsenic (11 ppm)** | | |
| South of Newcastle Beach Park | 10 | 41 |
| West of I-5 | 13 | N/A |
| North end of Lake Sammamish, offshore at Marymoor Park | 15 | |
| Lake Sammamish State Park, nearshore west of boat launch | 19 | |
| Middle of Lake Sammamish, northern portion | 22 | |
| Offshore of the northeastern shoreline of Lake Sammamish (average) | 23 | |
| Middle of Lake Sammamish, southern portion | 24 | |
| Middle of Lake Washington west of Mercer Island | 46 | 77 |
| Middle of Lake Washington between Magnuson Park and Kirkland | 46 | 80 |
| Near Boeing, nearshore | 70 | 42 |
| **Samples near or above Puget Sound Natural Background 90/90 UTL for Mercury (0.21 ppm)** | | |
| North Lake Marina, Kenmore (average) | 0.14 | 56 |
| Middle of Lake Washington between southwest Mercer Island & Rainier Beach | 0.16 | 75 |
| Middle of Lake Sammamish | 0.16 | 67 |
| Middle of Lake Sammamish, southern portion | 0.17 | 85 |
| Near Boeing, nearshore | 0.21 | 42 |
| South of Newcastle Beach Park | 0.21 | 56 |
| Kenmore, inner navigational channel | 0.24 | 31 |
| Offshore of the northeastern shoreline of Lake Sammamish (average) | 0.25 | 64 |
| Middle of Lake Sammamish | 0.25 | 100 |
| Middle of Lake Sammamish, northern portion | 0.26 | 80 |
| Middle of Lake Washington west of Mercer Island | 0.37 | 77 |
| Middle of Lake Washington between Magnuson Park and Kirkland | 0.38 | 80 |
| West of I-5 | 0.39 | N/A |

**Table 3:** Summary statistics and precision for the Lake Washington Area regional background.

| Analyte | N | Detection Frequency | Distribution | Mean | SD | Lake WA Area Regional Background 90/90 UTL | Puget Sound Natural Background 90/90 UTL | Precision |
|---|---|---|---|---|---|---|---|---|
| cPAHs | 23 | 23/23 | Gamma | 104 | 56 | 210 µg TEQ/kg | 21 µg TEQ/kg | 25% |

# Figures

Figures 1–9 (pp. 27–35)

Figures 10–15 are found in Appendix B of this report (pp. 51-56)

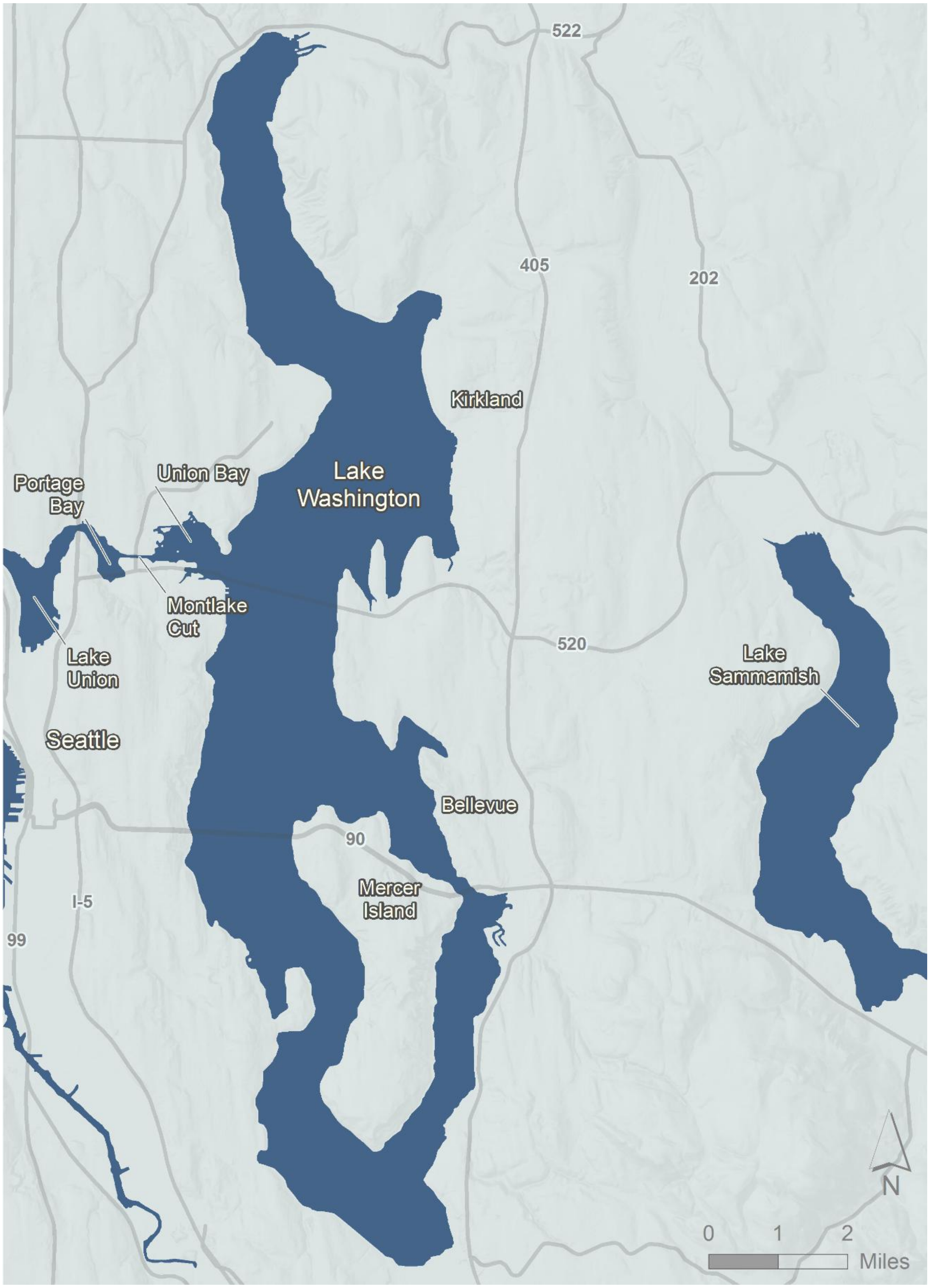

**Figure 1:** Geographic location of the Lake Washington area of interest.

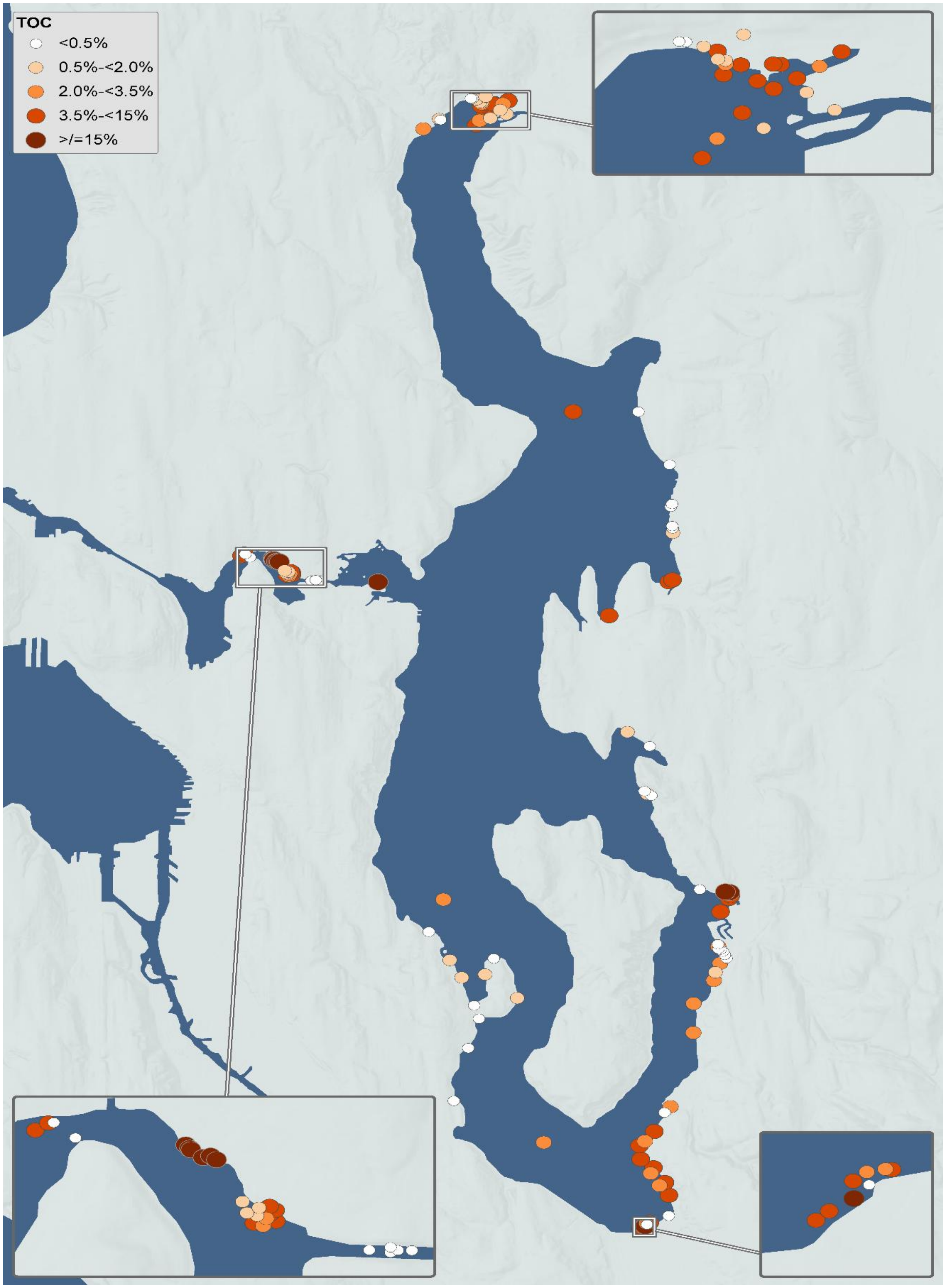

**Figure 2:** Total organic carbon throughout the Lake Washington area of interest.

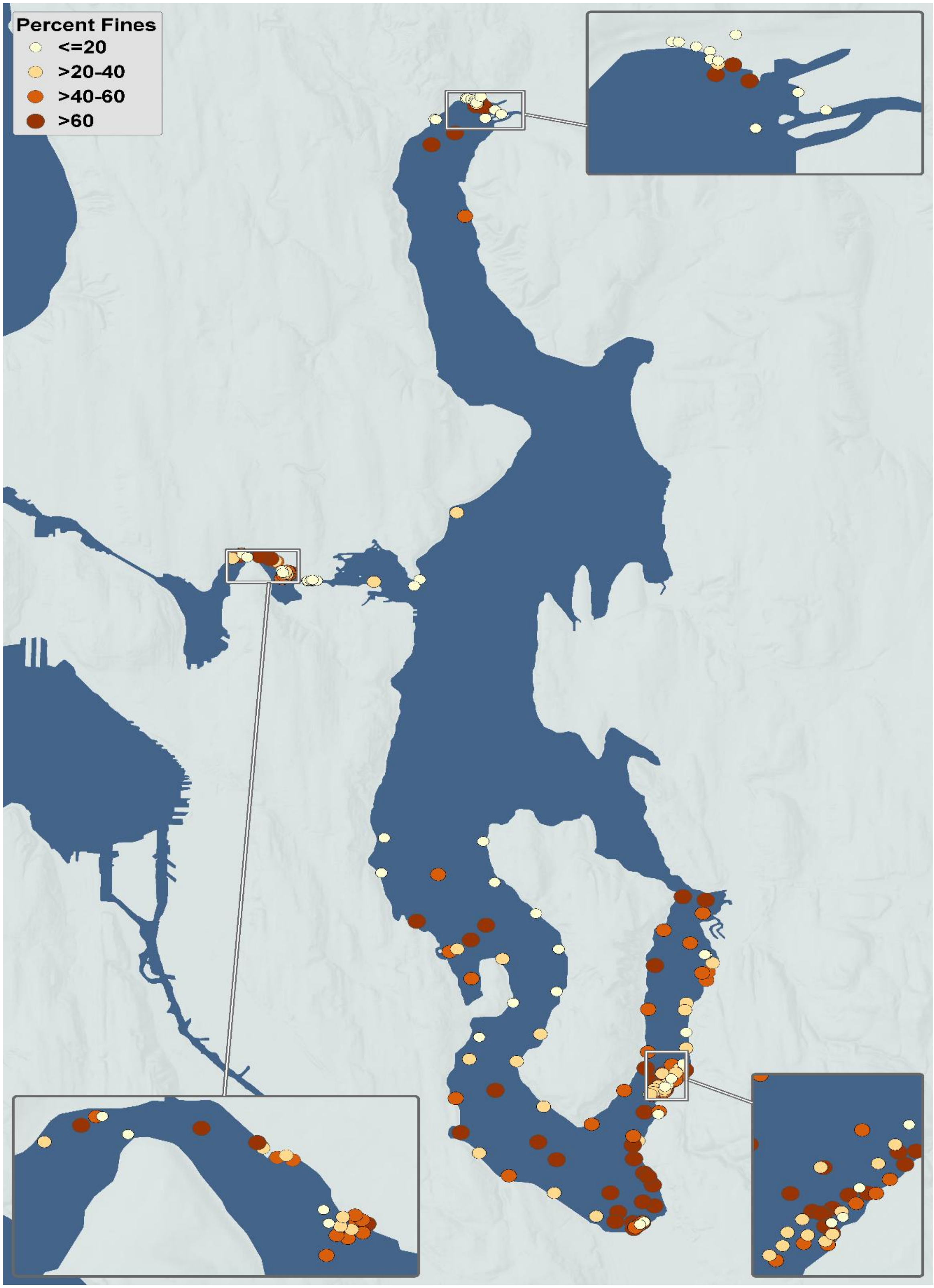

**Figure 3:** Percent fines throughout the Lake Washington area of interest.

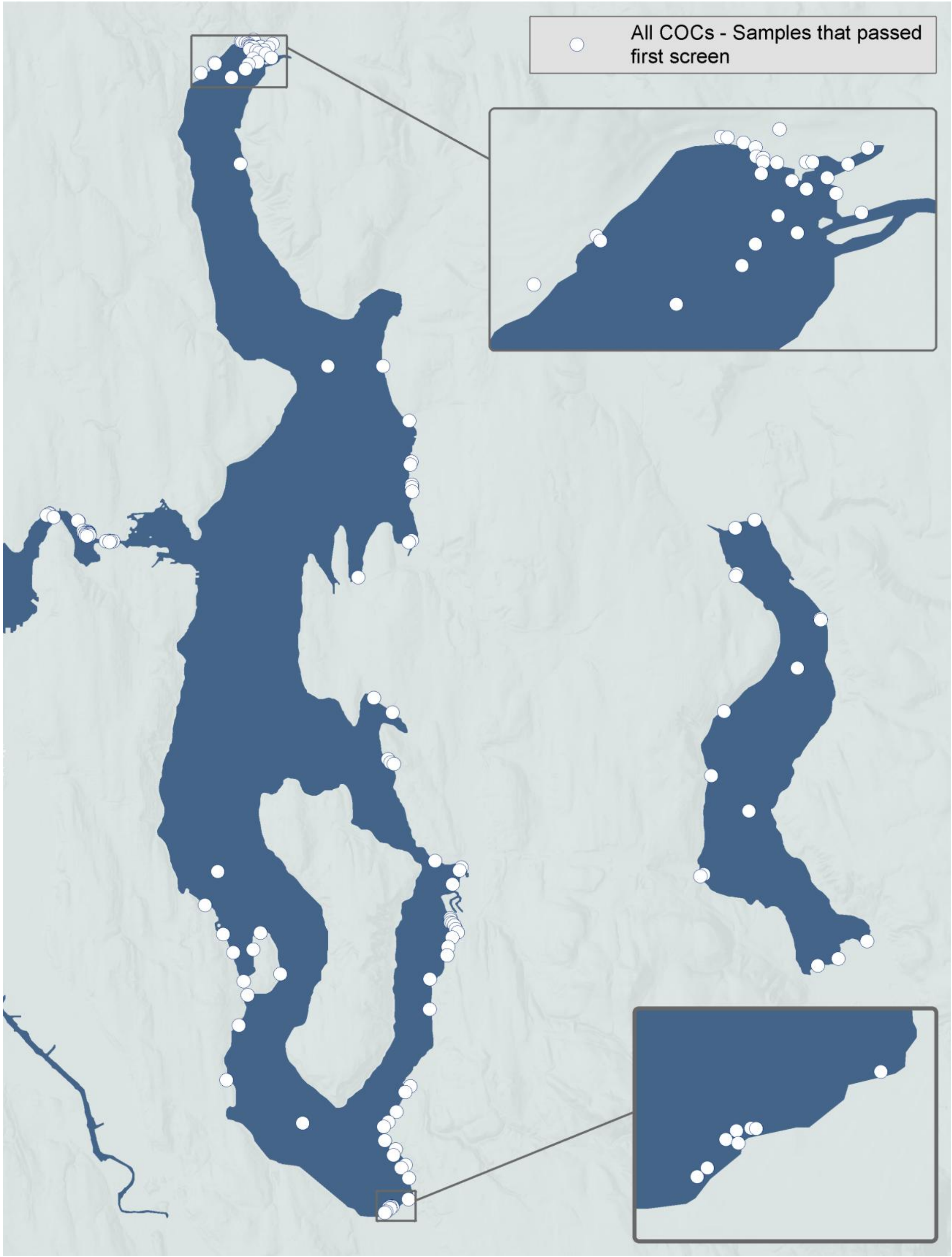

**Figure 4:** The cPAH, mercury, and arsenic data remaining after the first screen for age, depth, duplicates, and non-detect issues.

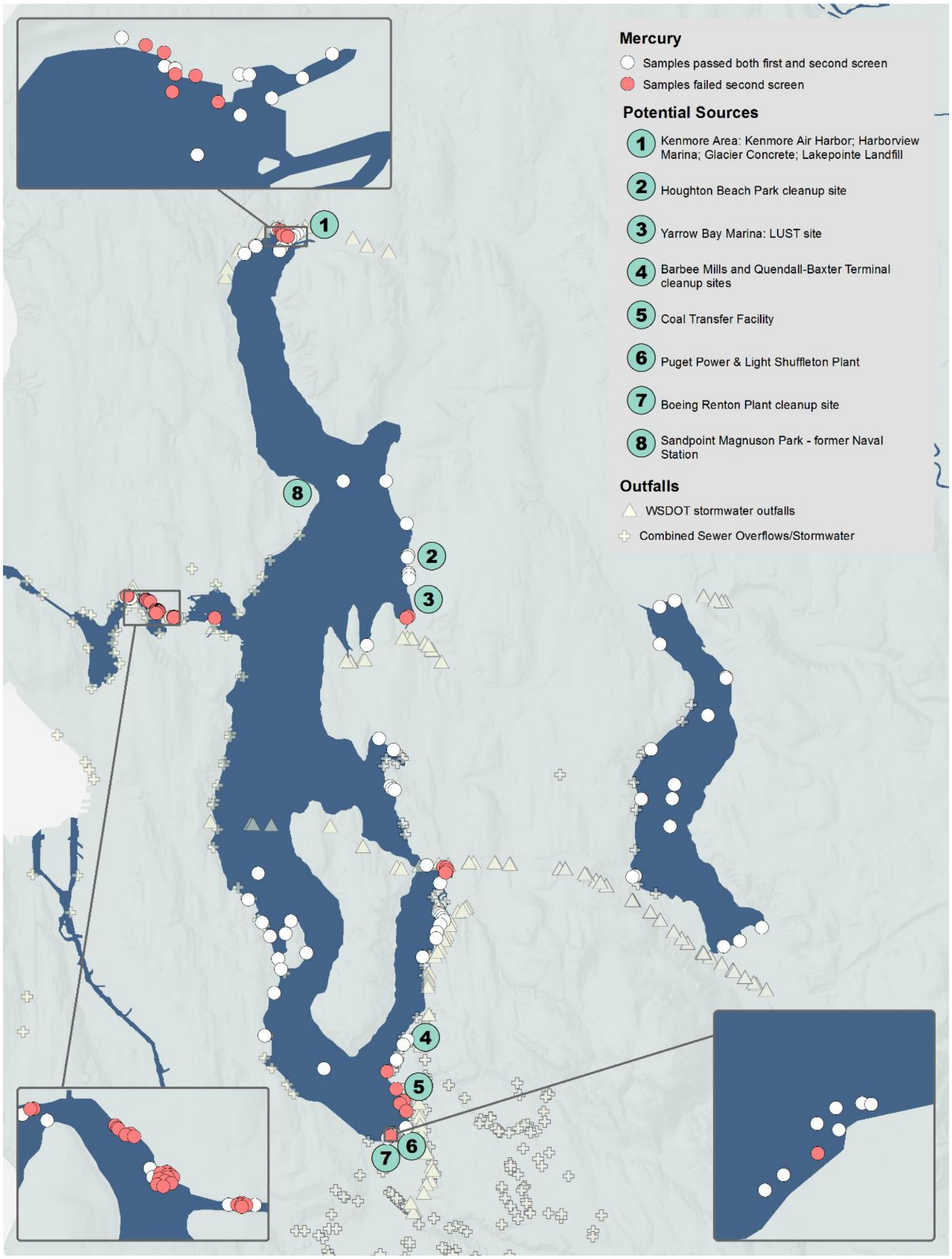

**Figure 5:** The mercury data remaining after being filtered by the second screen (orange circles) for potential impact from sites and sources and high TOC.

This data set (white circles) was then analyzed statistically to determine if it was suitable to establish regional background. See Section 3 and Table 4c for further detail on screening data out.

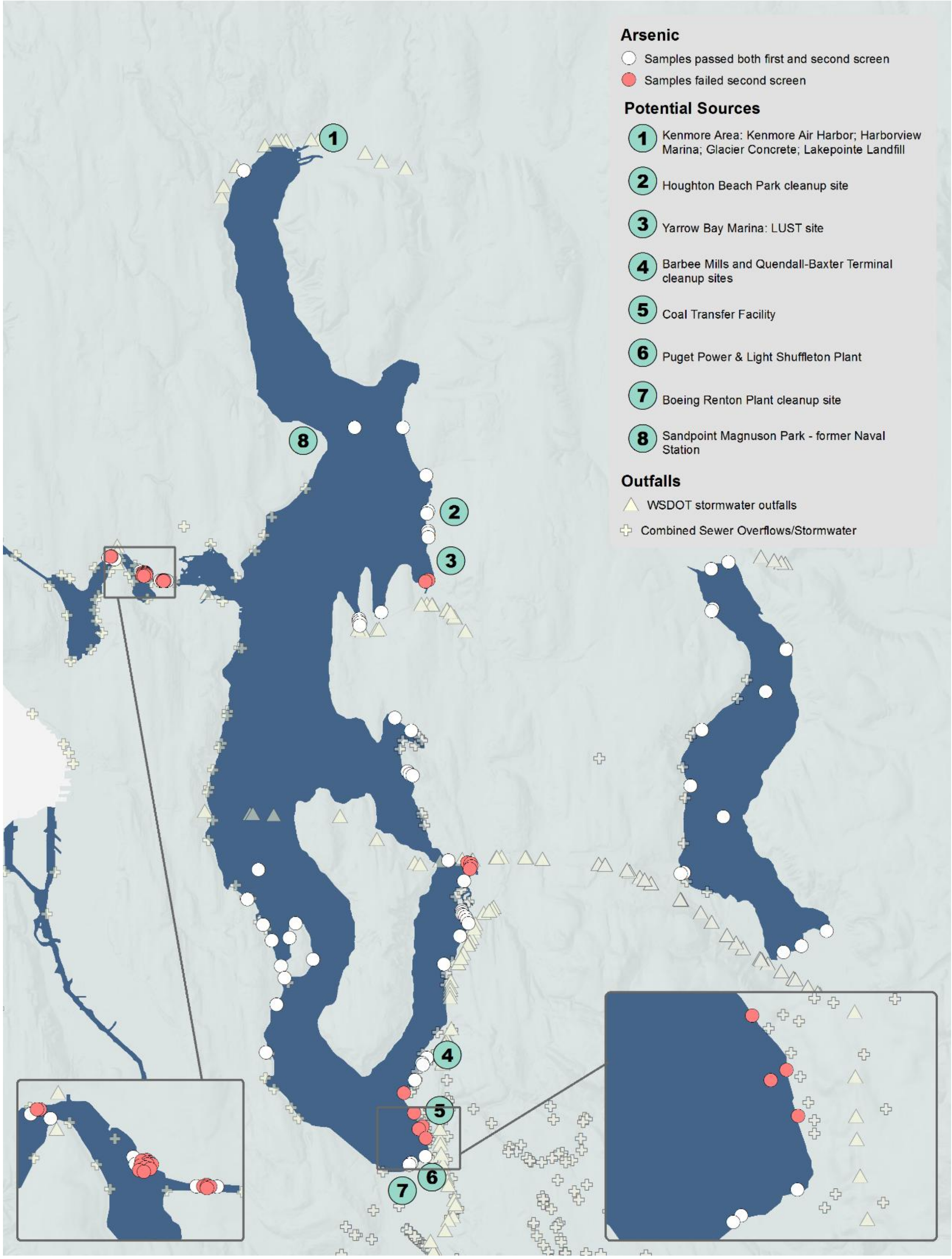

**Figure 6:** The arsenic data remaining after being filtered by the second screen (orange circles) for potential impact from sites and sources and high TOC.

This data set (white circles) was then analyzed statistically to determine if it was suitable to establish regional background. See Section 3 and Table 4b for further detail on screening data out.

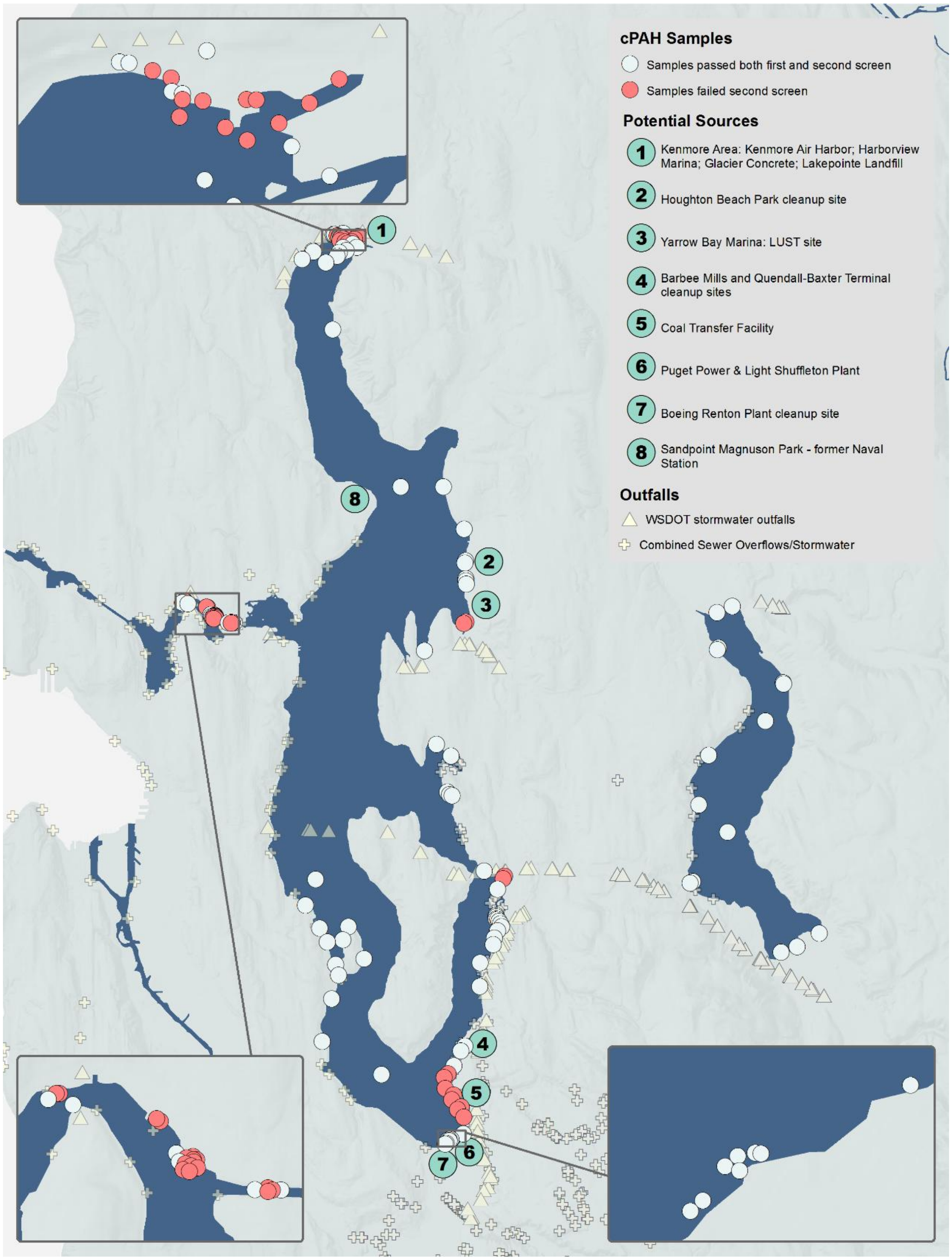

**Figure 7:** The cPAH data set remaining after being filtered by the second screen (orange circles) for potential impact from known sources and sites and high TOC**.**

This data set (white circles) was then analyzed statistically to determine if it was suitable to establish regional background. See Section 3 and Table 4a for detailed information about samples and reasons for screening data out.

**Figure 8:** The remaining cPAH data set after statistical analysis.

This data set (individual samples, prior to cluster averaging) was used to calculate the 90/90 UTL to establish regional background. See Table 2 for specific samples and Section 3 and Table 4a for specific reasons for screening data out of the regional background calculation.

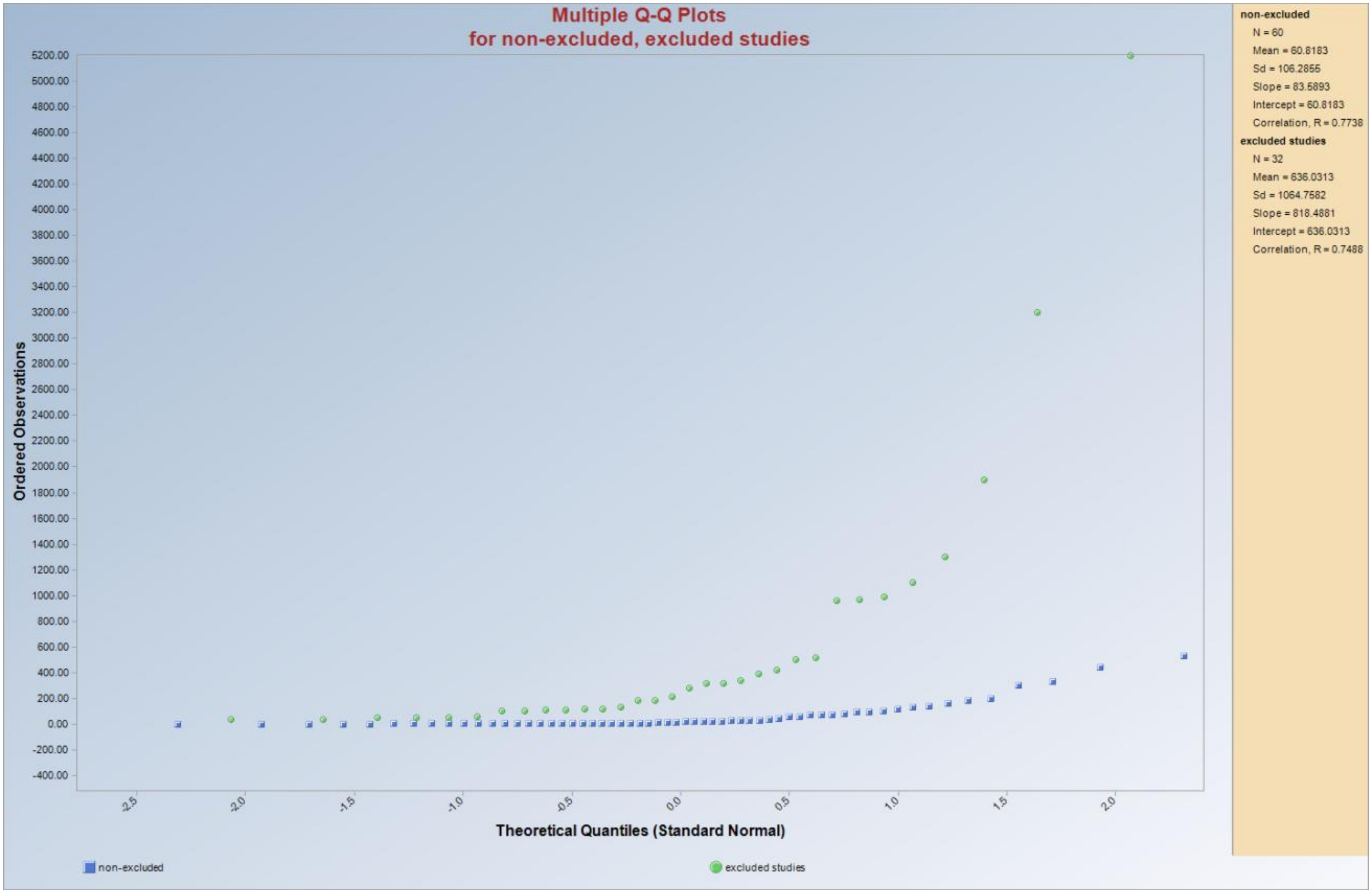

**Figure 9:** Quantile-Quantile plot comparing 2003 and earlier data (green dots) to post-2003 cPAH data (blue dots). The decision for the first screen to exclude data from 2003 and older was based on the results of this analysis.

# Appendix A. Data Tables

To view Tables 4 a **-** c as an Excel spreadsheet, see the links below.

**Table 4a:** cPAH data downloaded from EIM and examined to establish regional background. The table shows data results from the first, second, and third screens (Section 3 of this report) and the reasons for excluding specific samples.

https://fortress.wa.gov/ecy/publications/othersupplements/1609064other.zip

**Table 4b:** Arsenic data downloaded from EIM and examined to establish regional background**.** The table shows data results from the first, second, and third screens (Section 3) and the reasons for excluding specific samples.

https://fortress.wa.gov/ecy/publications/othersupplements/1609064other.zip

**Table 4c:** Mercury data downloaded from EIM and examined to establish regional background. The table shows data results from the first, second, and third screens (Section 3) and the reasons for excluding specific samples.

https://fortress.wa.gov/ecy/publications/othersupplements/1609064other.zip

# Appendix B. Statistical Methods and Analysis Used to Characterize the Lake Washington Area Regional Background Data Set

## B.1 Introduction

The Lake Washington area data set is a compilation of relatively recent studies with differing objectives and sampling designs. A data set consisting of multiple studies requires careful screening and spatial analysis to isolate those results that best represent the regional background concentration distribution before calculating summary statistics. Data that passed the first and second screens (Figures 4 – 6) were used in this analysis. As part of this process, the spatial relationships among samples were evaluated to identify independent samples, avoiding over-emphasis on areas of the lake with greater sampling intensity. The data set was then statistically evaluated to determine if it represented a single homogenous population, or multiple overlapping subpopulations. This iterative process involved multiple steps listed below, summarized in Table 5, and described in more detail in Section B.2 of this report:

> <u>Step 1.</u> A spatial autocorrelation analysis was conducted to identify the autocorrelation distance, which is the minimum distance between samples required to consider them independent. Samples that were spatially isolated or clear outliers were temporarily removed for this analysis, to reduce variability that would disproportionately affect the model. Clusters of samples within the autocorrelation distance can be assumed to have been influenced by the same sources and can be expected to have similar concentrations.
>
> <u>Step 2.</u> A population separation analysis of finite mixture models (Benaglia et al. 2009) followed by estimation of robust prediction limits for the subpopulations (Singh et al. 1994) was applied to just the independent samples (i.e., clusters of samples within the autocorrelation distance were excluded from this step). This step resulted in preliminary prediction limits used to identify subpopulations.
>
> <u>Step 3.</u> The prediction limits from Step 2 were then applied to each sample cluster, and individual samples within each cluster were allocated to their appropriate subpopulation. In some clusters, all of the samples were assigned to the same subpopulation when concentrations were similar. For more heterogeneous clusters, samples within the cluster were assigned to different subpopulations. If a cluster had samples with greatly dissimilar concentrations, this was an indication that the assumption that these samples were affected by the same sources was incorrect. Despite the physical proximity of these samples, these heterogeneous clusters appeared to have had multiple influences that affected their

concentrations, such as a sharply defined boundary of a swimming beach with imported sand, or a highly localized source of chemicals. In this step, the samples within each cluster were allocated to the most appropriate subpopulation based on concentration. Subsequently, any samples closer than the autocorrelation distance within each subpopulation were averaged.

Step 4. The population separation analysis of finite mixture models used in Step 2 (Benaglia et al. 2009) was repeated, this time including the cluster averages generated in Step 3 along with the independent samples used in Step 2. This step produced a final set of subpopulations, from which the specific subpopulation representing regional background was identified.

Step 5. Precision and 90/90 UTL estimates were calculated for the identified regional background subpopulation (Section 4).

# B.2 Methods

The following sections describe the above steps in greater detail, as well as the statistical methods that were used.

## B.2.1 Outlier Analysis

Prior to trend analysis and estimating the autocorrelation distance, certain samples were excluded from the data set for each analyte. These samples were either spatially isolated and/or chemically distinct (i.e., samples with unusual concentrations that were dissimilar to neighboring samples). Such samples unduly influence the trend model and disrupt the pattern of the residuals in the area.

Identification of potential outliers was conducted using boxplots and Quantile-Quantile (QQ) plots. These diagnostic tools generally assume independent and identically distributed (i.i.d.) data, an assumption that is not confirmed for this data set. However, the intent was to identify elevated values that might be indicative of an unsuspected source and exclude data points that could bias the autocorrelation analysis due to higher or spatially isolated values. Outliers were subsequently added back into the data set for the final population analysis, since they may not be elevated when viewed in the context of a homogenous sub-population.

## B.2.2 Autocorrelation Analysis

The autocorrelation distance is estimated based on data that do not exhibit a trend and have a zero mean, specifically the residuals from the best-fit model to the concentration surface. A simplified approach to evaluating trends was used. Multiple surface trend models were used to evaluate potential trends in concentrations, including least squares polynomial surface models of orders 0 to 5 (i.e., from no trend up to a 5th order polynomial). The six polynomial regression models were compared using the Aikake Information Criterion (AICc) corrected for sample size (Burnham and Anderson 2002), and patterns in the residual diagnostic plots. The model with the lowest AICc and best fitting residuals plots was considered to be the best trend model.

Lacking a regularly spaced grid of samples, the autocorrelation boundary was estimated by evaluating the correlation among pairs of points within various distances of each other. Pairs of sample points were grouped into bins of similar distances. For example, using a test distance of 50-m between samples, all pairs of samples within 0 to 50-m, 50 to 100-m, 100 to 150-m, etc. were grouped. Pearson's linear correlation coefficient between residuals for all possible station pairs within each distance bin provided an estimate of autocorrelation.

The distance bins considered were required to have a minimum of six pairs per bin, considered the smallest number of pairs that can reasonably be used to test for autocorrelation (e.g., Journel and Huijbregts 1978). When the sample size is small ($n < 10$), a significance test of the autocorrelation within each distance bin was applied using $\alpha = 0.20$ to limit Type II errors (i.e., failing to reject the null hypothesis when autocorrelation is present). This binned hypothesis testing approach was useful given the data limitations (i.e., insufficient pairs of samples at sequentially increasing distances) and the objective of estimating the minimum distance between independent samples.

### B.2.3 Population Separation

Two methods were used to separate the composite data set into individual (and possibly overlapping) subpopulations. First, likelihood methods were used to identify the breakpoints between subpopulations of the independent samples, and robust prediction limits for subpopulations were calculated for interpreting the results within groups of spatially clustered samples. Finally, the likelihood methods were repeated to separate the complete composite data set (the combination of independent samples and cluster averages) into the subpopulations of a finite mixture model of either normal, or gamma distributions.

The composite data sets were sometimes fairly limited in sample size with multiple populations present. The objective was to identify the subpopulation representative of regional background. In the composite data sets used for this analysis, regional background was expected to be bounded on two sides, with what would likely represent natural background (presumably similar to Puget Sound natural background) on the low end and portions of elevated populations on the high end. Breakpoints between subpopulations were identified using an expectation-maximization (EM) algorithm implemented in the following functions in the *mixtools* package in R (Benaglia et al. 2009): *normalmixEM* for mixtures of $k$ normal distributions and *gammamixEM* for mixtures of $k$ gamma distributions ($k$ = 1, 2, or 3). The breakpoints were readily confirmed by natural breaks or slope changes in the QQ plots. The fit of a single gamma distribution was also evaluated, using the *fitdistr* function in the *MASS* package in R (Venables and Ripley, 2002).

The mixture model alternatives were compared using Akaike Information Criterion (AICc), with correction for small sample sizes (Burnham and Anderson 2002). The model with the smallest AICc was preferred.

Where

= the log-Likelihood for the specified model

k = number of parameters (2 for each gamma or normal distribution in the mixture, so a mixture of 2 gamma distributions would have k=4)

n = number of observations

The AICc values for alternative models were compared, and any model with an AICc within 2 units of the minimum AICc was considered a reasonable alternative.

When more than one mixture model was considered appropriate, the models were reviewed to determine which was most suitable to identify regional background. For example, when comparable models differed only in how they differentiated concentrations at or below natural background, but the upper breakpoint which differentiated regional background was unchanged, both would be considered suitable.

For reference, Puget Sound natural background and chemical results for sediments collected from two freshwater reference lakes (Chester Morse and Mountain Lake, Ecology 2009) are also shown on the probability plots. These values were <u>not</u> included in the mixture model analysis. Chester Morse Reservoir is in the upper region of the Cedar River Watershed, the watershed for Lake Washington (King County 2015); Mountain Lake is located in the San Juan Islands, in the Puget Lowlands eco-region.

The first pass of the process used only the samples that were identified as independent based on the autocorrelation analysis (Section B.2.2 of this report). Any obvious outliers were removed and the EM algorithm was used to identify breakpoints between adjacent but separation subpopulations.

Robust 95% prediction limits for each of the preliminary subpopulations were then estimated. Robust limits are $(1 - \alpha) \times 100\%$ prediction limits for individual observations ($x_i$ for $i = 1, 2, \ldots n_g$) within a population $g$, i.e., Prob ($LPL_g \leq x_i$) = $1 - \alpha$ and Prob ($x_i \leq UPL_g$) = $1 - \alpha$. The prediction limits are "robust" because they use estimates of the mean, variance, and degrees of freedom derived after invoking the PROP influence function (Singh et al. 1994). The influence function down-weights the effect of extremely high or low values on the parameter estimates. The functions used to calculate the robust parameter estimates and prediction limits were written in R (R Core Team 2015).

Next, the individual samples within clusters were evaluated using the estimated prediction limits, and each of the samples within clusters was allocated to the most appropriate subpopulation based on concentration. Any values within the autocorrelation distance from the same subpopulation were averaged together.

The final pass of the process used the full data set, which included all of the data used in the first pass plus the cluster averages of samples within the autocorrelation distance calculated as described above (Section B.2.2). Using the full data set, the process was to:

- Evaluate the QQ plot and remove obvious outliers based on large jumps in the data distribution. Use the EM algorithm to identify maximum likelihood breakpoints between multiple normal or gamma subpopulations, and calculate the AICc associated with each mixture.

- The finite mixture with the lowest AICc was deemed the most appropriate description for the data set and the subpopulations were interpreted using location information (e.g., swim beaches versus depositional areas) and reference points (i.e., Puget Sound natural background and freshwater reference lakes, Ecology 2009) to name the subpopulation indicative of Lake Washington Area regional background.

### B.2.4 Principal Components Analysis

A principal components analysis (PCA) was conducted on a broader Lake Washington Area data set including 11 swim beach samples, 43 samples associated with sites around the lake, and 66 samples from the lake at large. The intent of this analysis was to look for patterns in chemical concentration that may distinguish different subsets of the data. If partitioning of the samples in this data set was distinct, it would shed light on how samples could be classified as influenced by sites or sources, or not.

PCA is an exploratory data analysis tool that can be used to investigate relationships between samples, and for data reduction of a multivariate data set. Sample relationships are illustrated using graphical representations of the data in terms of a small number of *principal components*, or linear combinations of the original variables. Correlations between the principal components and the original variables allow for the interpretation of which variables drive the primary differences among samples.

Computationally, the objective of PCA is to summarize the covariance or correlations structure of the original data set using a set of principal components constructed as linear combinations of the original variables. The first principal component is constructed to summarize most of the variability. The second principal component summarizes most of the residual variability and is constrained to be uncorrelated with the first principal component. The third principal components summarizes most of the residual variability remaining after the first two principal components and is constrained to be uncorrelated with each of the preceding principal components, and so on. Constraining the set of principal components to be uncorrelated allows us to interpret them as providing independent information about the variability in the data set. When a set of principal components cumulatively summarizes "most" (e.g., 80 to 90%) of the total sample variance then these principal components can "replace" the original variables without much loss of information. When a set of principal components summarizes only a moderate proportion of the total sample variance (e.g., 50 to 70%), then these results should be used primarily for interpretation of how the original variables contribute to the sample variance structure.

When the original variables have widely differing ranges or units of measure (e.g., mercury concentrations ranging from 0.006 to 0.9 ppm, and phenanthrene concentrations ranging from 2 to 1600 ppb) the PCA should be based on the correlation matrix rather than the covariance matrix. If the covariance matrix were used on a data set with widely disparate units of measure, the variables with the widest range and therefore largest variability would drive the principal component results.

In the PCA for this data set, all individual samples were used, some of which may be close enough together to be autocorrelated. This does not invalidate the results, but will increase the clustering of samples that have a spatial dependence. The physical and chemical endpoints included in the PCA were: TOC, fines, metals (arsenic, mercury, copper, lead, nickel, and zinc), and PAHs (phenanthrene, fluoranthene, pyrene, chrysene, and cPAH TEQ). The PCA was based on the correlation matrix and samples with any missing values were omitted, leaving 113 samples for the analysis.

## B.3 Results

### B.3.1 Outlier Identification

Identification of "outliers," or simply highly influential samples, was performed prior to performing the autocorrelation analysis. Samples that were spatially isolated or chemically distinct were removed prior to the autocorrelation step. All of the swim beach samples were excluded from the autocorrelation analysis because these samples represent a separate stratum (imported sand) and are not expected to have the same spatial relationships as the native sediments. The following non-swim beach samples were also excluded from the autocorrelation analysis:

- <u>cPAH TEQ:</u>
    - One station from Portage Bay/Lake Union, just west of Interstate 5 (Survey = KC_CSO_2013, Location ID = CSO13_B535). This sample had a cPAH TEQ value of 1900 ppb, more than 5 times the next highest concentration anywhere in the lake. Its nearest neighbor was approximately 175-m away with a TEQ value of < 6.7 ppb. This sample strongly influenced the trend surface, which subsequently affected the autocorrelation distance calculation.

- <u>Arsenic:</u>
    - One station near Boeing (Survey = AQSLWA082010, Location ID = COMP08102010). This sample had an arsenic concentration of 70 ppm. Its nearest neighbor was 72-m away with an arsenic concentration of 5.1 ppm. This sample strongly influenced the trend surface, which subsequently affected the autocorrelation distance calculation.

    - Two stations from the middle of the lake (Survey = KingLakeSeds, Location IDs = KCM-0826 and KCM-0890). These samples both had arsenic concentrations of 46 ppm. They were both spatially isolated and chemically distinct and were very influential to the trend surface.

    - One station in the north end of the lake from McAleer Creek (Survey = KingStrmsSeds, Location ID = 432). This sample was spatially isolated; the closest sample within the arsenic data set was almost 8-km away.

- <u>Mercury:</u>
    - One sample from Portage Bay/Lake Union just west of I-5 (Survey = KC_CSO_2013, Location ID = CSO13_B535). This sample had a mercury value of 0.392 ppm which was more than 24 times the concentration at its nearest neighbor. This sample strongly influenced the trend surface, which subsequently affected the autocorrelation distance calculation.

    - Two samples from deeper areas in the middle of the lake (Survey = KingLakeSeds, Location IDs = KCM-0826 and KCM-0890). These samples had mercury concentrations of 0.38 and 0.37 ppm, respectively. They were both spatially isolated and chemically distinct and were very influential to the trend surface.

## B.3.2 Autocorrelation Distance

For this data set, samples were collected unevenly through space and time. This analysis did not include the Lake Sammamish data set. These clusters of sampling locations emphasized sub-areas of the lake (Figure 4) such as:

- The Seward Park area (sampled in 2008)
- Certain eastside beaches (sampled in 2009 and 2010)
- The Renton Boeing plant shoreline (2010)
- Sub-areas within Portage Bay (2013)
- The north end of the lake near Kenmore (2012).

A summary of the data used in this analysis is shown in Table 6.

The residuals from the best fit model for each chemical (Table 6) were grouped based on distance between sampling locations. For example, if the distance interval under evaluation was 50-m, then all sample pairs within 0 to 50-m, 50 to 100-m, 100 to 150-m (etc.) were grouped, and the Pearson correlation was calculated between the values among all sample pairs within each distance bin.

Finding the most appropriate minimum autocorrelation distance was exploratory. For example, when the cPAH residuals were binned at 50-m intervals, all but one of the 50-m intervals up to 250-m had positive and statistically significant correlations ($p < 0.20$), while all other intervals had a) correlations that were negative, b) strongly influenced by single data points, and/or c) were non-significant ($p < 0.20$). When the residuals were binned at 250-m intervals, only the first interval was positive and statistically significant ($p < 0.20$). The estimate of the minimum autocorrelation distance is not considered precise, because the data set is limited in the number of samples and in their spatial separations. The locations are also highly clustered, so that the autocorrelation estimates at smaller distance intervals can be influenced by a single geographic area. The final autocorrelation results for the Lake Washington data set are shown in Tables 7 – 9. The starred autocorrelation distance for each chemical is assumed to be representative of the minimum autocorrelation distance within this data set.

## B.3.3 Population Separation

The following sections describe the results of the population separation analysis for the cPAH, arsenic, and mercury data sets. The station locations describing regional background may be different for each analyte.

### B.3.3.1 cPAHs

Two iterations of the process were performed, first using only the independent samples (13 from swim beaches and 26 others scattered around the two lakes, all of which were more than 250-m from all other stations). Data from both Lake Washington and Lake Sammamish were included.

The QQ plot for these independent samples (Figure 10a) was examined, identifying one elevated value at 330 ppb and one or two possible breakpoints between subpopulations. The data below the elevated value of 330 ppb (n=38) were evaluated as a mixture of two or three normally distributed data sets, and as a mixture of one, two or three gamma distributed data sets. The AICc among the five competing mixture models indicated that a mixture of three normal distributions was preferred (Table 10). However, mixtures of two or three gamma distributions showed comparable results (difference in AICc < 2, Table 10). The breakpoint for the upper distribution was identical whether mixtures of three normal or gamma distributions were considered. The mixture of two gamma distributions identified the upper distribution as the values ranging from 11 ppb to 160 ppb, which overlaps substantially with Puget Sound natural background (21 ppb) and the reference lakes (6.7 to 90 ppb). The mixture of three normal distributions was selected to move forward, because it had the lowest AICc and was most consistent with the conceptual site model (i.e., concentrations above Puget Sound natural background, and individual distributions that were not excessively skewed).

For the mixture of three normal distributions, the first breakpoint was identified between the observations of 45 and 72 ppb, and the second breakpoint between 5.7 and 11 ppb (breakpoints shown at 50 and 10 ppb, Figure 10).

Using the nine values with concentrations greater than 50 ppb (“Population 1” in Figure 10a), the robust 95% prediction limits were ($LPL_1$ = 50 ppb and $UPL_1$ = 170 ppb).

The QQ plot for the data below the breakpoint of 50 ppb is shown with the second breakpoint at 10 ppb (Figure 10b). Using the nine values between 10 and 50 ppb (“Population 2” in Figure 10b), the robust 95% prediction limits were ($LPL_2$ = 8.3 ppb and $UPL_2$ = 45 ppb).

The QQ plot for the remainder of the data (all samples with concentrations less than the population breakpoint of 10 ppb, Figure 10c) was then examined. All but two of these samples had “less than” values for the TEQ sum. This population contained all but one of the swim beach samples and represented very low values. Because of the dominance of non-detects, the upper bound for this population could not be adequately determined. Therefore, the lower limit from Population 2 was used as the statistical limit to separate the populations.

The preliminary population boundaries identified above were subsequently applied to the data within clusters and any sample within 250-m of another sample whose concentration fell within the same population limits were averaged. Assignment of the results for the clustered samples to each populations is shown in Table 11.

The final pass of the process used the full data set from both lakes (n=73), including the 39 observations used in the first pass, plus 2 stations in Lake Sammamish whose field reps were averaged and 32 values that were part of the spatial clusters (Table 11). The extreme values at 1900 ppb (Lake Washington) and 1500 ppb (Lake Sammamish) were excluded as clear outliers. The QQ plot for the remaining 71 independent values (Figure 11a) was examined. For reference purposes, the six freshwater sediment cPAH TEQ values from Chester Morse Reservoir and Mountain Lake (ranging from 6.7 to 90 ppb) are also shown on these plots. Note that these data were not included in any of the statistical calculations or decisions about break points in the QQ plots. In Figure 11a, there were large breaks in the QQ plot between the two highest values in Lake Sammamish (two values greater than 600 ppb along the west shoreline) and the two next highest values (Kennydale Beach and Chism Beach at 330 and 370 ppb, respectively) and the remainder of the data set.

In a well-mixed environment, the sediment chemistry from the regional background signal may be expected to follow a normal distribution without excessive skewness. When the sediment chemistry data has a skewed probability distribution, this may be an indicator of an environmental setting that has multiple regional sources that are not well-mixed or of overlapping distributions with the upper concentrations representing very localized contaminant sources. A skewed distribution that is supported by a smooth QQ plot without large breaks in the distribution of values may reasonably be modeled as a gamma distribution.

Any samples with concentrations substantially elevated relative to the remainder of the data set should be carefully evaluated, in case they represent unique local conditions rather than a more general regional background. The relatively small sample size and irregular spatial distribution of sampling locations in this composite data set are inadequate to interpret the nature of localized trends or local hotspots (i.e., the 370 ppb value at Chism beach is within 100-m of a sample with a concentration < 5 ppb; while the Kennydale Beach sample with a value of 330 ppb was in the vicinity of potential sources from the Quendall-Baxter Terminal and Coal Transfer facility sites; and the two elevated samples from Lake Sammamish were the result of averaging field replicates that had vastly different concentrations). In light of the generally high sampling uncertainty associated with this composite data set, the four values ≥ 330 ppb which would have a strong influence on regional background were excluded until more information becomes available.

The normal QQ plot for the remaining data (Figure 11b) indicates the presence of two or three possible subpopulations. These data below 250 ppb (n=67) were evaluated as a mixture of two or three normally distributed data sets, and as a mixture of one, two or three gamma distributed data sets. Among the five competing mixture models, model selection based on the AICc showed a clear preference for a mixture of three gamma distributions (Table 10). This mixture model identified a breakpoint between 6.7 and 11 ppb; and another breakpoint somewhere between 31 and 50 ppb. There were four values (38, 38, 42, and 45 ppb) with a 30% or greater probability of being associated with more than one of these subpopulations. These values are feasibly members of both the first and second subpopulations (Figures 11b and 11c).

The objective to identify the data set most likely representative of regional background does not require that each value be assigned to only one population. When population distributions overlap, the regional background data set may include some samples that have a non-trivial probability (e.g., >30%) of being associated with the adjacent natural background distribution. That appears to be the case for these cPAH TEQ data. There are four values that appear to represent both the upper end of natural background and the lower end of regional background.

Examination of the sample types and sampling locations for the complete cPAH data set (Figures 11a and 11b, Table 12) suggested there were three primary subpopulations plus a group of higher concentration samples (≥ 330 ppb) as follows:

- Very low-concentration samples of clean sand (Population 3; concentrations ≤ 6.7 ppb).
- Mainly nearshore samples overlapping with the Puget Sound natural background data set and reference lakes west of the Cascades (Population 2; concentrations between 11 and 45 ppb, inclusive).
- Samples found in depositional areas and near urban shorelines representing regional background (Population 1; concentrations between 38 and 240 ppb, inclusive).
- A smaller set of increasingly higher concentration samples (330 ppb and above).

A regional background value for cPAH TEQ was estimated from the Population 1 data with concentrations between 38 and 240 ppb (Table 1).

### B.3.3.2 Arsenic

The arsenic data set included 50 independent samples: 13 from swim beaches and 37 others scattered around the two lakes, all of which were more than 100-m from all other stations. The samples within clusters were homogeneous, with the exception of the cluster near Boeing (Table 13). The values within each cluster (other than Boeing) were so similar that these values were averaged. The two samples near Boeing, however, were within 70-m of one another with more than an order of magnitude difference in concentration. Despite their geographic proximity, these two samples were treated as independent in the population separation analysis, on the assumption that they were influenced differently.

The full arsenic data set included the 50 independent samples and 8 values for the clusters (Table 13). In the QQ plot for these 58 independent values (Figure 12a), the most dominant feature is the presence of three elevated and influential values at 45.5, 45.9, and 70 ppm. Similar to the cPAH TEQ distribution, the values $\geq$ 45 ppm would have a strong influence on the 90/90 UTL for regional background, so they will be excluded from the regional background data set until more information becomes available.

In the QQ plot for the remainder of the data (55 samples with concentrations < 45 ppm; Figure 12b), there appear to be two or three possible subpopulations. These data were evaluated as a mixture of two or three normal distributions, or a mixture of one, two or three gamma distributions. Among the five competing mixture models, model selection based on the AICc showed a preference for a mixture of gamma distributions over normal distributions, with mixtures of two or three gamma distributions showing comparable results (difference in AICc < 2, Table 10).

The two alternative models both identified a separation subpopulation above 11 ppm, and differed only in how they distinguished the number of subpopulations below 11 ppm. Puget Sound natural background is 11 ppm, and Lake Washington area regional background is assumed to be above this threshold. The competing mixture models identified a breakpoint between 10 and 12.9 ppm (Figure 12b).

Puget Sound natural background for arsenic is 11 ppm. The six freshwater sediment values from Chester Morse Reservoir and Mountain Lake ranged from 2.8 to 17 ppm. Distinguishing sub-populations at concentrations below approximately 11 ppm (the concentration range for all but nine of the Lake Washington samples) may not be particularly relevant to regional background. All of the samples in this data set had detected arsenic concentrations. The concentrations in the swim beach samples ranged from 1.6 to 3.9 ppm (shown in green on Figure 12b), and the concentrations in the non-swim beach samples ranged from 1.4 to 24 ppm. There was no distributional distinction between the clean swim beach samples and the non-swim beach samples (Populations 2 and 3, Figure 12b).

With only six samples in the range of regional background (> 11 ppm and excluding the extreme values), there is insufficient data to describe the regional background distribution. Therefore, a regional background value for arsenic was not calculated.

### B.3.3.3 Mercury

The mercury data set comprised 70 independent samples: 13 from swim beaches and 57 others scattered around the lake, all of which were more than 50-m from all other stations. The samples within clusters were homogeneous, with the exception of one of the clusters near Boeing (Table 15). The values within each cluster (except the cluster near Boeing) were so similar that values within each cluster were averaged. However, one cluster near Boeing had values approximately an order of magnitude different from the next closest mercury concentration. For this cluster near Boeing, three similar concentrations were averaged and the two remaining samples (one higher and one lower than the averaged values) were treated as independent despite their geographic proximity (Table 15).

The full mercury data set included 70 independent samples and 9 values that fell within clusters (Table 15). In the QQ plot for these 79 independent values (Figure 13a), the most dominant feature is three elevated and influential values at 0.37, 0.38, and 0.39 ppm. Similar to the cPAH TEQ and arsenic distributions, the values $\geq$ 0.37 ppm would have a strong influence on 90/90 UTL regional background calculation, so they have been excluded from any regional background data set until more information becomes available.

In the QQ plot for the remainder of the data (all samples with concentrations < 0.3 ppm; Figure 13b), there appear to be two, three, or four possible subpopulations. These data below 0.3 ppm (n=76) were evaluated as a mixture of two to four normally distributed data sets, or as a mixture of two to four gamma distributed data sets. Among the six competing mixture models, model selection based on the AICc showed a preference for a mixture of four normal distributions (Table 10). This mixture model identified breakpoints around 0.015 ppm, 0.029 ppm, and 0.12 ppm (Figure 13b).

Most of the mercury concentrations within the data set (70 out of 79) were within the Puget Sound natural background range (< 0.2 ppm) and also within the range of values found in Chester Morse Reservoir and Mountain Lake (0.07 ppm to 0.15 ppm). The detection frequency was 94% (74/79) with a maximum detection limit of 0.02 ppm. Concentrations for the swim beach samples ranged from 0.007 to 0.03 ppm, and from 0.006 to 0.39 ppm for the non-swim beach samples. There was a distributional distinction between the swim beach samples and the non-swim beach samples (Population 4 vs. Population 3, Figure 13b). The regional background signal is expected to be in the concentration region above 0.12 ppm (Population 1, Figure 13b, and possibly including the three values > 0.3 ppm, Figure 13a) due to the apparent distributional separation of these data from all of the swim beach samples and most of the Chester Morse and Mountain Lake samples. However, with only ten data points in this subpopulation, there is insufficient data to adequately characterize the regional background distribution. Therefore, regional background for mercury was not calculated.

### B.3.4 Principal Components Analysis Including Site Data

The PCA analysis was undertaken to look for chemical patterns that may be present and distinct in the samples that were known to be associated with sites and point sources. If distinct patterns were present, this would allow some samples to be excluded from the regional background investigation due to their chemical similarity to site-related samples.

The first two principal components explained more than 75% of the total variability of the data. The bi-plot for the PCA is shown in Figure 14. The first principal component (PC1) was an overall (negative) average of all individual variables. The second principal components (PC2) had proportionally higher metals, TOC, and fines in the positive direction of PC2 versus proportionally higher PAHs in the negative direction. The cluster plot (Figure 15) shows how the samples clustered into 5 groups (using the k-means clustering algorithm, *means* in R).

In the PCA plots, the same samples are called out. These were previously identified as elevated or influential in the univariate QQ plots (Section B.3.3), or were called out because they had extremely high TOC (Bryant samples). The PCA did not show distinct chemistry patterns that could be used to classify samples as being directly influenced by source or site. . The concentration patterns among samples from near sites and from the data set at large were generally similar, and concentration was the main distinguishing characteristic among the identified sample groups (i.e., swim beach samples to expected regional background groups, and above).

The initial screening out of samples considered to be “directly site- or source-influenced” was based strictly on geographic proximity to known or potential sources. The success of this effort to define a chemical pattern for “site” samples relies on samples having been properly assigned to the “site” category in the first place. The spatial distribution of chemicals in the lake is very patchy, so it is possible that proximity to a site does not uniquely determine direct site influence. The opposite is also true: sufficient distance from a source does not automatically indicate a lack of site influence. By screening out samples located near sources, some samples with concentrations in the range of regional background may have been inappropriately excluded. However, this screening tool is the best, to avoid including too many samples in the regional background data set that are directly influenced by sites or sources, even if some regional background samples are excluded. All samples beyond a safe (generally not directly influenced) distance from known sites and sources were included in the population separation analysis, and if samples were identified as being elevated and highly influential, they were excluded the regional background population.

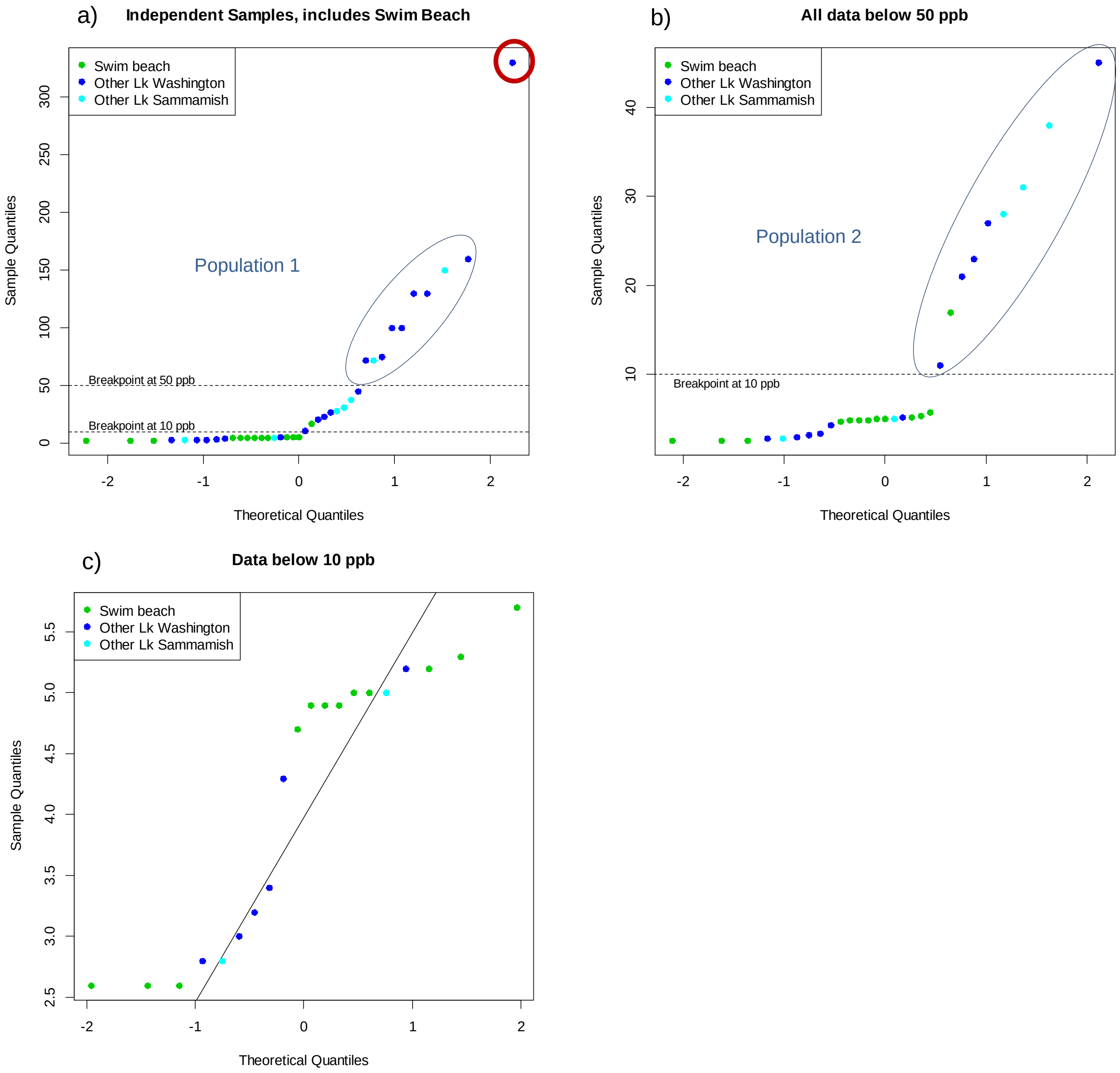

**Figure 10:** Sequential QQ plots for the cPAH TEQ (µg/Kg, dw) data, excluding samples within clusters (n = 39).

a) All data with one elevated value (**circled in red**) and subpopulation breakpoints supported by likelihood methods.

b) QQ plot for data excluding elevated value and Population 1, with a population break supported by likelihood methods.

c) QQ plot for the remainder of the data.

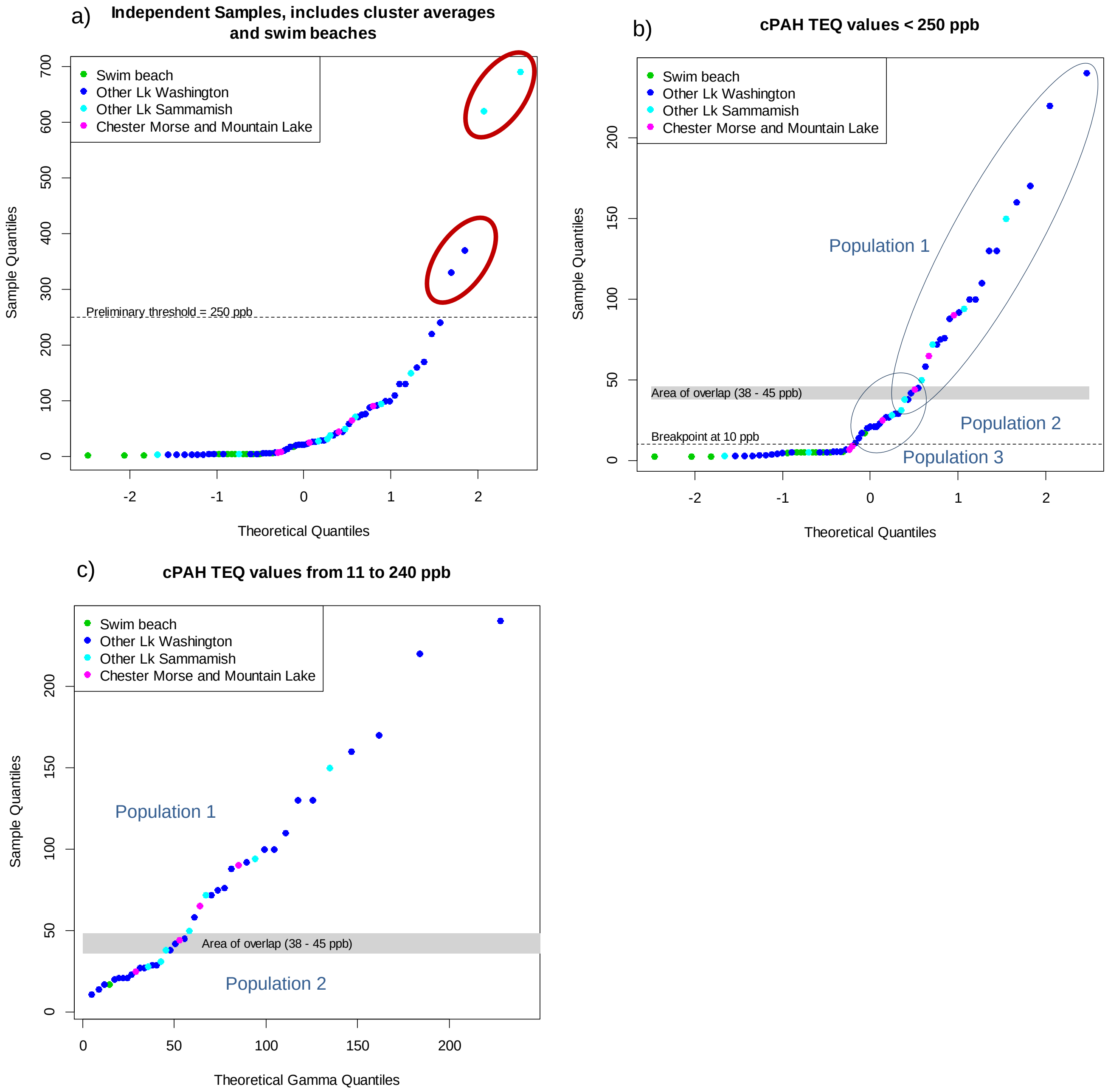

**Figure 11:** Sequential QQ plots for the cPAH data.

Includes cluster averages (n = 66). Concentrations in freshwater sediment from Chester Morse Reservoir and Mountain Lake are shown on the plots for reference. These values were not used in the prediction limits calculation.

a) All data with several elevated values (**circled in red**) and preliminary thresholds subjectively identified based on large breaks in the QQ plot.

b) Data < 250 ppb, with subpopulations identified using the breakpoints supported by likelihood methods.

c) Gamma QQ plot for the data within Populations 1 and 2, showing the area of overlap between the two populations.

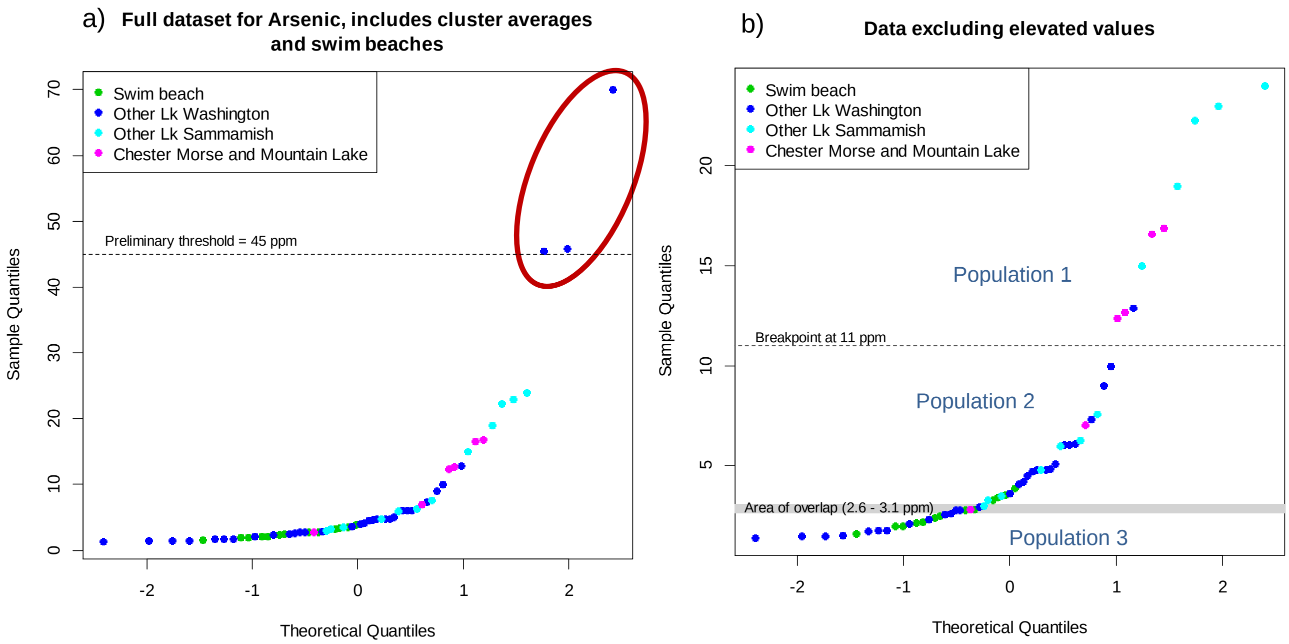

**Figure 12:** Sequential QQ plots for the arsenic data, including cluster averages (n = 58).

Concentrations in freshwater sediments from Chester Morse Reservoir and Mountain Lake are shown on the plots for reference. These values were not used in the calculation of prediction limits.

All data with three elevated and influential values (**circled in red**).

All data excluding the elevated and influential values, with subpopulations identified using the breakpoints supported by likelihood methods.

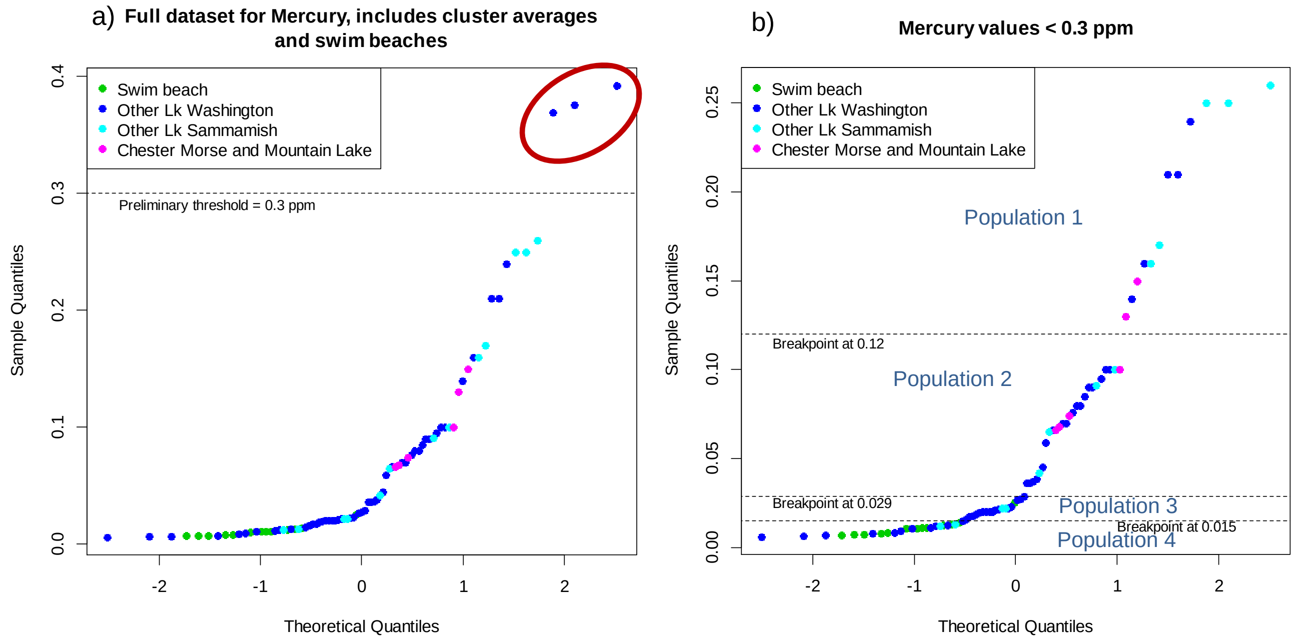

**Figure 13:** Sequential QQ plots for mercury, including cluster averages (n = 79).

Concentrations in freshwater sediments from Chester Morse Reservoir and Mountain Lake are shown on the plots for reference, but these values were not used in the calculation of Lake Washington prediction limits.

a) All data and a preliminary threshold based on a break in the QQ Plot. Influential elevated values are (**circled in red**).

b) All data excluding the elevated and influential values, with subpopulations identified using the breakpoints supported by likelihood methods.

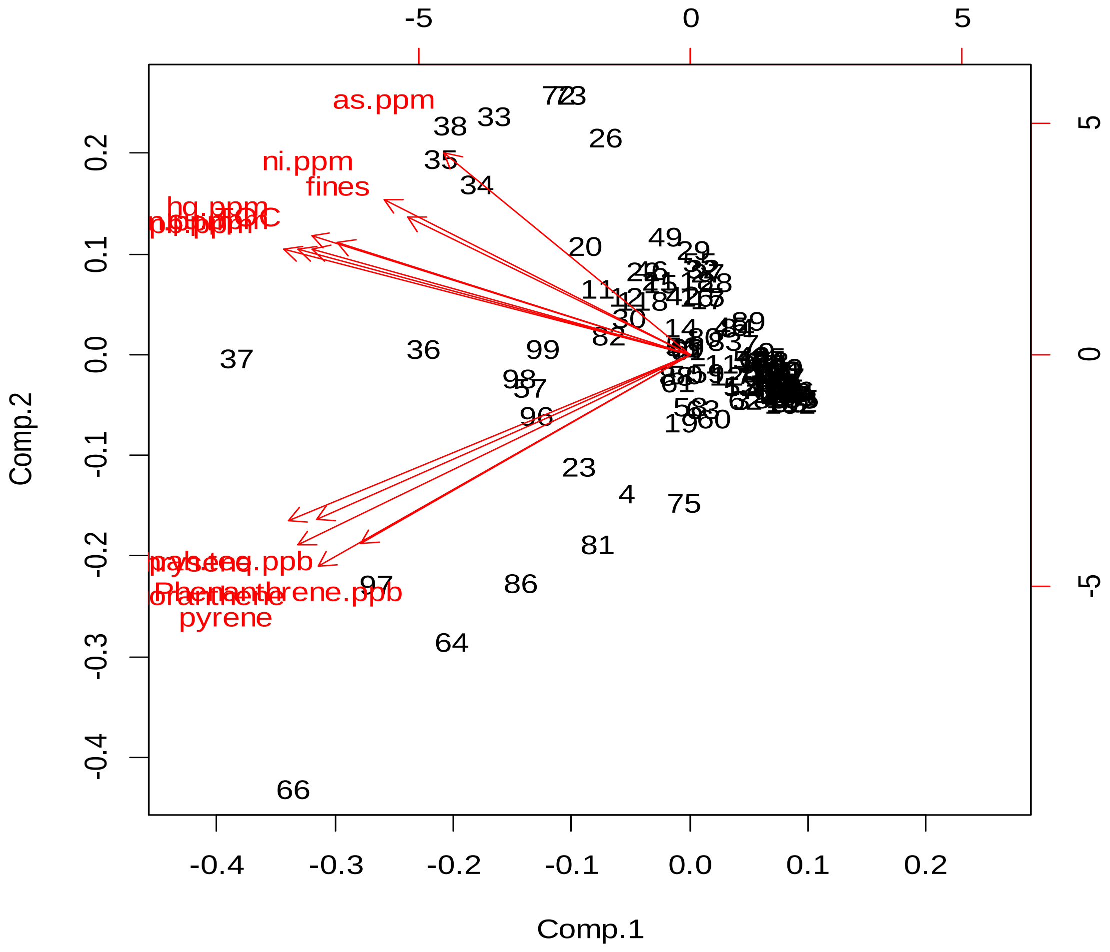

**Figure 14:** Principal Components Analysis. Bi-plot showing the direction that the original variables load onto the first two principal components. PCA was done on 113 samples with no missing values for 13 chemical or physical endpoints**.**

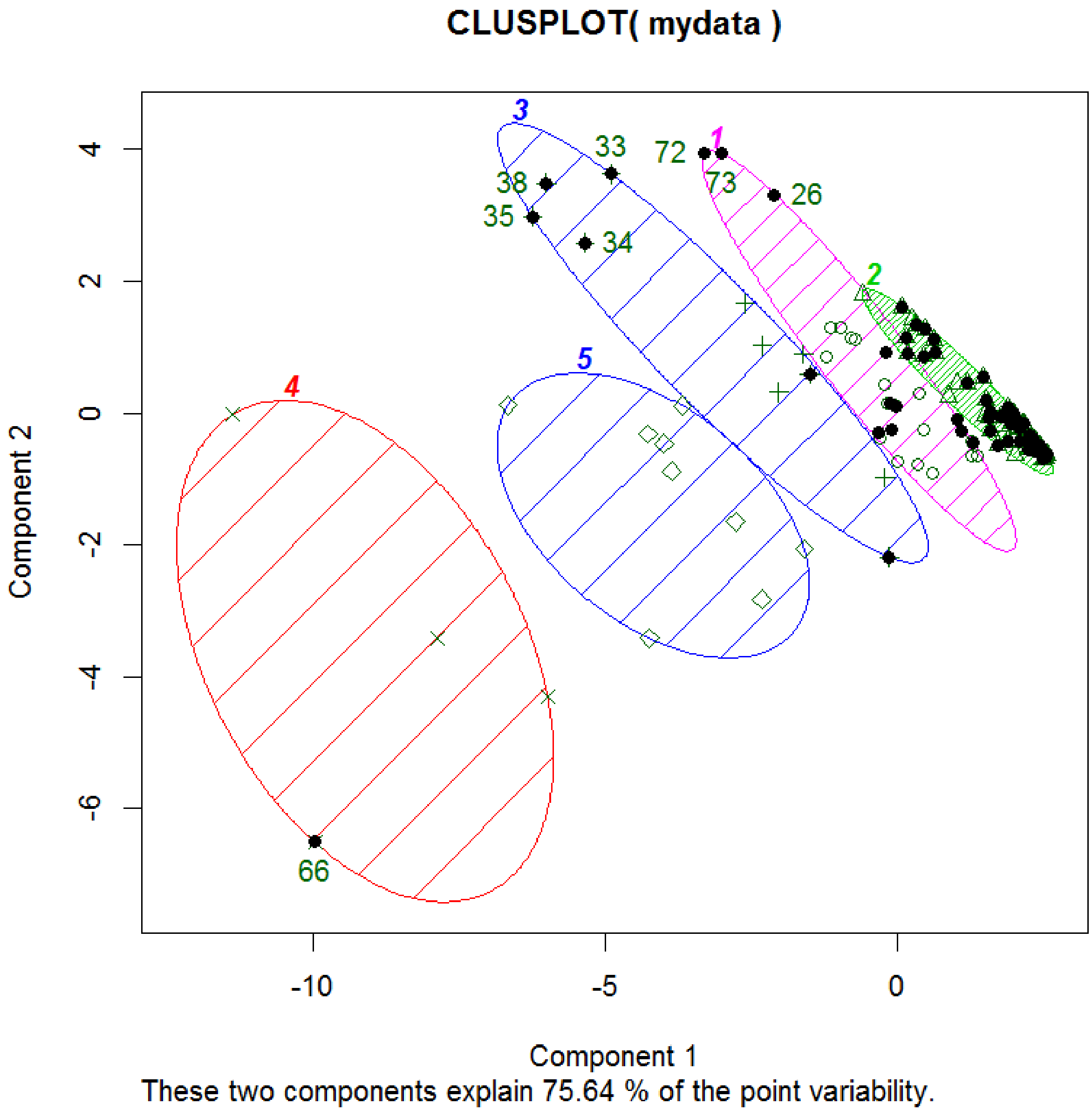

**Figure 15:** Plot of the 113 samples on the first two principal components (see Figure 14).

Ellipses indicate 5 clusters established using k-means clustering. Samples identified with black dot were part of the Lake Washington data set evaluated for the regional signal. Numbered samples were extreme in one way or another (#66 had cPAH TEQ value of 1900 ppb. Samples near the top of group 3 (#33, #34, #35, and #38) are Bryant samples with high TOC. Samples near the top of group 1 are in the middle of the lake (#72 and #73) and near Boeing (#26) with proportionally higher metals than PAHs.)

**Table 5:** Stepwise approach to identify the regional background data set from the compiled data set.

| Step | Data Set | Objective |
|---|---|---|
| 1 | All samples, excluding swim beach samples and chemical/spatial outliers. Lake Sammamish data were not included. | Identify autocorrelation distance. |
| 2 | Independent samples, including swim beach samples and chemical/spatial outliers, excluding clusters. | Identify subpopulation concentration prediction limits. |
| 3 | Each cluster. | Separate samples within clusters into their respective subpopulations. Average samples within the same subpopulation within each cluster. |
| 4 | Independent samples plus cluster averages, including swim beach samples and outliers. | Finalize subpopulation concentration prediction limits. Identify regional background subpopulation. |
| 5 | Regional background subpopulation. | Calculate precision and 90/90 UTL. |

**Table 6:** Summary of the trend surface analysis for each analyte, not including Lake Sammamish data.

| Analyte | Sample Size | Concentration Range of the Data Used to Fit the Trend Surface | Polynomial Order of the Best-Fit Trend Surface |
|---|---|---|---|
| cPAH TEQ | 59 | <2.4 to 370 ppb | 2nd |
| Arsenic | 39 | 1.15 to 12.9 ppm | No trend |
| Mercury | 57 | 0.0061 to 0.24 ppm | 2nd |

**Table 7:** Autocorrelation results for cPAH data, not including Lake Sammamish data.

| Bin Number | Bin Endpoints | *N* | Pearson's Correlation Coefficient | One-Tailed *p* Value for Parametric Test |
|---|---|---|---|---|
| 1 | 0 to < 250-m | 55 | 0.348 | 0.005 |
| 2 | 250 to < 500-m | 37 | 0.008 | 0.482 |
| 3 | 500 to < 750-m | 38 | -0.574 | 1.000 |
| 4 | 750 to < 1000-m | 34 | -0.206 | 0.878 |

**Table 8:** Autocorrelation results for arsenic data, not including Lake Sammamish data.

| Bin Number | Bin Endpoints | *N* | Pearson's Correlation Coefficient | One-Tailed *p* Value for Parametric Test |
|---|---|---|---|---|
| 1 | 0 to < 100-m | 10 | 0.831 | 0.001 |
| 2 | 100 to < 200-m | 13 | -0.109 | 0.639 |
| 3 | 200 to < 300-m | 6 | -0.217 | 0.660 |

**Table 9:** Autocorrelation results for mercury data, not including Lake Sammamish data.

| Bin Number | Bin Endpoints | *N* | Pearson's Correlation Coefficient | One-Tailed *p* Value for Parametric Test |
|---|---|---|---|---|
| 1 | 0 to < 50-m | 10 | 0.738 | 0.007 |
| 2 | 50 to < 100-m | 9 | -0.161 | 0.661 |
| 3 | 100 to < 150-m | 17 | -0.072 | 0.609 |
| 4 | 150 to < 200-m | 13 | 0.197 | 0.260 |
| 5 | 200 to < 250-m | 10 | 0.021 | 0.477 |
| 6 | 250 to < 300-m | 12 | 0.529 | 0.038 |
| 7 | 300 to < 350-m | 9 | 0.552 | 0.062 |

**Table 10:** AICc values and relative likelihoods for competing mixture models[1] for cPAH TEQ, arsenic and mercury data sets.

| Distribution of Mixtures | AICc for cPAH TEQ data (preliminary) | Difference from Min(AICc) | AICc for cPAH TEQ data (final) | Difference from Min(AICc) | AICc for Arsenic data | Difference from Min(AICc) | AICc for Mercury data | Difference from Min(AICc) |
|---|---|---|---|---|---|---|---|---|
| 2 Normals | 311.7 | 6 | 604 | 27 | 281 | 10 | -293 | 20 |
| 3 Normals | 305.8* | 0 | 583.3 | 6 | 274.4 | 3 | -299 | 14 |
| 4 Normals | n/a | -- | n/a | -- | n/a | -- | -313* | 0 |
| 1 Gamma | 351.1 | 45 | 632 | 55 | 292.2 | 21 | -282 | 31 |
| 2 Gammas | 306.7 | 1 | 583.6 | 7 | 272.2 | 1 | -310 | 3 |
| 3 Gammas | 306.9 | 1 | 577* | 0 | 271.5* | 0 | -307 | 6 |

[1] Evaluated using *mixtools* package in R (Benaglia et al 2009).

* Mixture model preferred by smallest AICc value.

**Table 11:** Clustered cPAH samples allocated to preliminary populations or left unassigned due to outlier status.

When more than one sample from a cluster fell within the same population, their values were averaged (underlined).

| | | | Value(s) used for each cluster | | |
|---|---|---|---|---|---|
| **Cluster Location** | **Individual Concentrations (ppb)** | **Unassigned> 170 ppb** | **Pop 1 [50, 170]** | **Pop 2 [8.3, 45]** | **Pop 3 <8.3** |
| West of I-5 | <6.7, 1900 | 1900 | | | <6.7 |
| Portage Bay (near UW) | 38, 46 | | | 42 | |
| Kenmore (near mouth of Sammamish River) | 3.8, 6.3 | | | | 5.1 |
| Kenmore Navigational Channel | 21, 66, 72, 89 | | 76 | 21 | |
| Marsh Park | <5.6, 20 | | | 20 | <5.6 |
| Lyon Creek Waterfront Preserve | <5.5, 29 | | | 29 | <5.5 |
| Houghton Beach Park | <4.7, 29 | | | 29 | <4.7 |
| Newcastle Beach Park | <2.4, <3, <3.7, <4, 6.1, 15, 38 | | | 27 | <3.8 |
| South of Newcastle Beach Park | 170, 240 | 240 | 170 | | |
| Montlake Cut | 17, 110 | | 110 | 17 | |
| May Creek | 14, 92 | | 92 | 14 | |
| Harbour Village Marina | <4.8, 5, 8.8, 33, 58 | | 58 | 21 | <4.9 |
| Chism Beach | <5.4, 370 | 370 | | | <5.4 |
| Boeing | <2.9, 34, 36, 43, 56, 120, 220 | 220 | 88 | 38 | <2.9 |
| Western shoreline of Lake Sammamish, near Squibbs Creek | 50, 1500 | 1500 | 50 | | |
| Offshore of the northeastern shoreline of Lake Sammamish | 91, 97 | | 94 | | |
| Number of independent observations in clusters: | | 5 | 8 | 10 | 9 |

**Table 12:** Samples with cPAH values, broken into sub-populations.

| cPAH TEQ (ppb) | Location |
|---|---|
| **Population 3 (cPAH TEQ < 11 ppb)** | |
| <2.6 | Coulon Beach Park, shallow swim beach |
| <2.6 | Pritchard Island Beach Park, shallow swim beach |
| <2.6 | Seward Park Beach, shallow nearshore park |
| <2.8 | South of Stan Sayres Memorial park, very nearshore |
| <2.8 | South end of Lake Sammamish, offshore at Lake Sammamish State Park |
| <2.9 | Boeing |
| <3.0 | Seward Park, north side of peninsula, shallow nearshore park |
| <3.2 | Opposite Seward Park, west side of Andrews Bay, nearshore |
| <3.4 | Lake Washington Blvd Park, nearshore |
| <3.8 | Newcastle Beach Park (average) |
| <4.3 | Seward Park, west side of peninsula, nearshore |
| <4.7 | Waverly Park swim beach |
| <4.7 | North Houghton Beach Park |
| <4.9 | Harbour Village Marina (average) |
| <4.9 | Houghton Beach Park, swim beach |
| <4.9 | Meydenbauer Bay, nearshore swim park |
| <4.9 | Newcastle Beach park, swim beach |
| <5.0 | Chism Beach park, swim beach |
| <5.0 | Kirkland Marina park, inlet with swim beach |
| <5.0 | North end of Lake Sammamish, offshore at Marymoor Park |
| 5.1 | Kenmore, near mouth of Sammamish River (average) |
| 5.2 | Lake Sammamish Idylwood Park swim beach |
| 5.2 | South of Seward Park, nearshore |
| 5.3 | Lake Sammamish State Park swim beach |
| <5.4 | Chism Beach |
| <5.5 | Lyon Creek Waterfront Preserve |
| <5.6 | Marsh Park |
| 5.7 | Enatai Beach park, swim beach |
| <6.7 | West of I-5 |
| **Population 2 (11 ppb ≤ cPAH TEQ ≤ 31 ppb)** | |
| 11 | Martha Washington Park |
| 14 | May Creek |
| 17 | Clyde Beach Park, swim beach |
| 17 | Montlake Cut |
| 20 | Marsh Park |
| 21 | Harbour Village Marine (average) |
| 21 | Seward Park, east side of peninsula, nearshore |
| 21 | Kenmore (near the mouth of the Sammamish River) |

| cPAH TEQ (ppb) | Location |
|---|---|
| <23 | Cozy Cove wetland area |
| 27 | Newcastle Beach (average) |
| 27 | Saint Edward State Park, offshore |
| 28 | North end of Lake Sammamish, offshore at Marymoor Park |
| 29 | Lyon Creek Waterfront Preserve |
| 29 | Houghton Beach Park |
| 31 | Middle of Lake Sammamish, southern portion |
| **Overlap between Population 1 and Population 2 (38 ppb ≤ cPAH TEQ ≤ 45 ppb), included with regional background** | |
| 38 | Middle of Lake Sammamish, northern portion |
| 38 | Boeing (average) |
| 42 | Portage Bay, near UW (average) |
| 45 | Near Newport Yacht Club |
| **Population 1 (50 ppb ≤ cPAH TEQ ≤ 240 ppb), representing regional background** | |
| 50 | Western shoreline of Lake Sammamish, offshore near Squibbs Creek (average of field reps) |
| 58 | Harbour Village Marina |
| 72 | Lake Sammamish State Park, nearshore west of boat launch |
| 72 | Middle of Lake Washington west of Mercer Island |
| 75 | Middle of Lake Washington between the southwest shoreline of Mercer Island and Rainier Beach |
| 76 | Kenmore Navigational Channel (average) |
| 88 | Boeing (average) |
| 92 | May Creek |
| 94 | Offshore of the northeastern shoreline of Lake Sammamish |
| 100 | South of Pleasure Point |
| 100 | Middle of Lake Washington between Magnuson Park and Kirkland |
| 110 | Montlake Cut |
| 130 | Pleasure Point |
| 130 | Middle of Lake Washington, north end, between Lake Forest park and Inglewood Golf Club |
| 150 | Nearshore, Idylwood Park in Lake Sammamish |
| 160 | McAleer Creek |
| 170 | South of Newcastle Beach Park |
| 220* | Boeing |
| 240 | South of Newcastle Beach Park |
| **Samples with elevated and influential cPAH TEQ concentrations 330 ppb ≤ cPAH TEQ)** | |
| 330 | Kennydale Beach swim park, very high fines |
| 370 | Chism Beach |
| 620 | Off a residential dock, western shore of Lake Sammamish (average of field reps) |
| 690 | Off a residential dock, western shore of Lake Sammamish (average of field reps) |
| 1500 | Western shoreline of Lake Sammamish, nearshore south of Squibbs Creek |
| 1900 | West of Interstate5 |

**Table 13:** Average arsenic concentrations for clustered samples and one outlier.

| Location of the Cluster | Individual Concentrations (ppm) | Average | Outlier |
|---|---|---|---|
| Near Boeing, nearshore | 5.1, 70 | 5.1 | 70 |
| May Creek | 1.4, 2.8 | 2.1 | |
| Fairweather Bay Residential inlet | 5.0, 5.0, 6.3, 8.1 | 6.1 | |
| Portage Bay, near UW | 2.4, 3.1 | 2.8 | |
| Marsh Park | 2.1, 3.0 | 2.6 | |
| Newcastle Beach Park | 1.2, 1.9, 2.0, 3.1, 3.6 | 2.4 | |
| Lake Sammamish | 22, 24 | 23 | |
| Number of Independent Observations | | 7 | 1 |

**Table 14:** Samples with arsenic concentrations, broken into sub-populations (Figure 12). Samples are from Lake Washington unless otherwise noted.

| Location | Concentration (ppm) | Percent Fines |
|---|---|---|
| **Populations 2 or 3, in the range of natural background (< 11 ppm)** | | |
| Under I-5 | 1.4 | NA |
| Chism Beach | 1.5 | 6 |
| South of Stan Sayres Memorial park | 1.5 | 7 |
| Seward Park - south of park. | 1.5 | 17 |
| Lake Sammamish Idylwood Park swim beach | 1.6 | |
| Martha Washington Park | 1.8 | 8 |
| Seward Park - north side of peninsula | 1.8 | 9 |
| Montlake Cut | 1.8 | NA |
| Chism Beach park swimming beach | 2 | 4 |
| Pritchard Island Beach Park swim beach | 2 | 4 |
| Enatai beach park; swim beach | 2.1 | 3 |
| May Creek, cluster average | 2.1 | 10 |
| Meydenbauer Bay beach park, swim beach | 2.2 | 3 |
| Newcastle Beach Park, cluster average | 2.3 | 13 |
| Waverly Park swim beach | 2.4 | 3 |
| Seward Park Beach swim beach | 2.5 | 4 |
| Newcastle Beach Park | 2.6 | 5 |
| Marsh Park, cluster average | 2.6 | 4 |
| Coulon Beach Park swim beach | 2.8 | 4 |
| Kirkland Marina park swim beach | 2.8 | 3 |
| South Houghton Beach Park | 2.8 | 12 |
| Portage Bay (near UW), cluster average | 2.8 | NA |
| Nearshore, Idylwood Park in Lake Sammamish | 3.0 | |
| Opposite Seward Park (w side of Andrews Bay) | 3.0 | 21 |

| Location | Concentration (ppm) | Percent Fines |
|---|---|---|
| Newcastle Beach park swimming beach | 3.3 | 19 |
| Western shoreline of Lake Sammamish, offshore south of Squibbs Creek (average of field reps) | 3.3 | |
| Lake Sammamish State Park swim beach | 3.4 | |
| South end of Lake Sammamish, offshore at Lake Sammamish State Park | 3.5 | |
| Seward Park (w side of peninsula) | 3.6 | 15 |
| Clyde Beach Park swim beach | 3.6 | 3 |
| Houghton Beach Park swim beach | 3.9 | 3 |
| Seward Park (e side of peninsula) | 4.1 | 20 |
| Chism Beach | 4.2 | 16 |
| From a stream (May Creek) | 4.5 | 14 |
| From a stream (McAleer Creek) | 4.7 | NA |
| Lake Washington Blvd Park | 4.8 | 13 |
| Near Newport Yacht Club (south of I-90). | 4.8 | 19 |
| Off a residential dock, western shore of Lake Sammamish (average of field reps) | 4.8 | |
| Montlake Cut | 4.9 | NA |
| Near Boeing | 5.1 | 3 |
| North end of Lake Sammamish, offshore at Marymoor Park | 6.0 | |
| Fairweather Bay Residential inlet, cluster average | 6.1 | NA |
| Cozy Cove wetland area | 6.1 | 74 |
| North Houghton Beach Park | 6.1 | 4 |
| Western shoreline of Lake Sammamish, south of Squibbs Creek | 6.3 | |
| Kennydale Beach Park | 7.3 | 70 |
| Off a residential dock, western shore of Lake Sammamish (average of field reps) | 7.6 | |
| Pleasure Point | 9.0 | 20 |
| **Population 1 (greater than 12 ppm), representing regional background** | | |
| South of Newcastle Beach Park | 10 | 41 |
| West of I-5 | 13 | NA |
| North end of Lake Sammamish, offshore at Marymoor Park | 15 | |
| Lake Sammamish State Park, nearshore west of boat launch | 19 | |
| Middle of Lake Sammamish, northern portion | 22 | |
| Offshore of the northeastern shoreline of Lake Sammamish (average) | 23 | |
| Middle of Lake Sammamish, southern portion | 24 | |
| Middle of Lake Washington between Magnuson Park and Kirkland | 46 | 80 |
| Middle of Lake Washington west of Mercer Island | 46 | 77 |
| Near Boeing, nearshore | 70 | 42 |

**Table 15:** Average mercury concentrations for clustered samples, and two elevated values.

| Location of the Cluster | Individual Concentrations (ppm) | Average | Independent Low and High Values |
|---|---|---|---|
| Harbor Village Marina | < 0.02, < 0.02 | < 0.02 | |
| Lyon Creek Waterfront Preserve | < 0.02, < 0.02 | < 0.02 | |
| North Lake Marina, Kenmore | 0.10, 0.18 | 0.14 | |
| Boeing | 0.018, 0.08, 0.08, 0.11, 0.21 | 0.09 | 0.018, 0.21 |
| Boeing | 0.08, 0.09 | 0.085 | |
| Newcastle Beach Park | 0.0076, 0.0076 | 0.0076 | |
| Lake Sammamish | 0.22, 0.27 | 0.25 | |
| Number of Independent Observations | | 7 | 2 |

**Table 16:** Samples with concentrations, broken into sub-populations (see Figure 13). Samples are located in Lake Washington unless otherwise noted.

| Location | Mercury Concentration (ppm) | Percent Fines |
|---|---|---|
| **Population 3 or 4 (mercury < 0.03 ppm)** | | |
| 44 samples | 0.0061 – 0.029 | <1 – 52 |
| **Population 2 (0.03 ppm < mercury < 0.11 ppm)** | | |
| Portage Bay (near UW) | 0.036 | NA |
| Montlake Cut | 0.036 | NA |
| Seward Park (East side of peninsula; nearshore) | 0.037 | 20 |
| From a stream (McAleer Creek) | 0.038 | NA |
| Western Lake Sammamish, nearshore south of Squibbs Creek | 0.042 | 4 |
| Montlake Cut | 0.045 | NA |
| Cozy Cove wetland area | 0.059 | 74 |
| Off a residential dock, western Lake Sammamish (average) | 0.065 | 5 |
| Portage Bay (near UW) | 0.066 | NA |
| Kenmore - navigational channel | 0.07 | 49 |
| South of Newcastle Beach Park | 0.07 | 21 |
| Kennydale Beach Park | 0.076 | 70 |
| Kenmore - navigational channel | 0.08 | 48 |
| Kenmore - navigational channel | 0.08 | 60 |
| Near Boeing (average) | 0.085 | 73 |
| Pleasure Point | 0.09 | 20 |
| Near Boeing (average) | 0.09 | 77 |
| Off a residential dock, western Lake Sammamish (average) | 0.091 | 14 |
| Kenmore - navigational channel | 0.095 | 45 |
| Lake Sammamish State Park, nearshore west of boat launch | 0.10 | 78 |

| Location | Mercury Concentration (ppm) | Percent Fines |
|---|---|---|
| Kenmore - navigational channel | 0.10 | 55 |
| South of Newcastle Beach Park | 0.10 | 41 |
| **Population 1 (0.11 ppm < mercury < 0.26 ppm), expected regional background** | | |
| North Lake Marina, Kenmore | 0.14 | 56.25 |
| Middle of Lake Washington – SW of Mercer Island | 0.16 | 74.5 |
| Middle of Lake Sammamish | 0.16 | 67 |
| Middle of Lake Sammamish, southern portion | 0.17 | 85 |
| Near Boeing | 0.21 | 41.9 |
| South of Newcastle Beach Park | 0.21 | 56.3 |
| Kenmore – inner navigational channel | 0.24 | 30.6 |
| Offshore of northeastern Lake Sammamish (average) | 0.25 | 64 |
| Middle of Lake Sammamish | 0.25 | 100 |
| Middle of Lake Sammamish, northern portion | 0.26 | 80 |
| **Samples with elevated and influential mercury concentrations (> 0.3 ppm)** | | |
| Middle of Lake Washington - west of Mercer Island | 0.37 | 77 |
| Middle of Lake Washington - between Magnuson Park and Kirkland | 0.38 | 80 |
| West of Interstate 5 | 0.39 | NA |

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# Appendix C. Summary of Revisions in this Report

A draft of this report was released for comment in September 2016. Comments were received from September 15 to November 4, 2016, as well as during a technical workshop in Seattle, Washington, on October 20, 2016. Following is a summary of revisions and clarifications in this final report that were made in response to those comments.

- Clarified that Ecology is willing to add to this dataset as new data becomes available. Clarified that, if the new data are deemed sufficient, we will consider reviewing and revising regional background.

- Reviewed existing data and sources to determine if more than one regional background population exists within the Lake Washington Area. Concluded there is only one appropriate regional background population.

- Included additional data and conducted new analyses for mercury, arsenic, and cPAHs to establish regional background. The additional data included 1) data from the Lake Washington area from 2004, and 2) data from Lake Sammamish from 2004 and newer.
    - Even with the additional data, sample numbers remained insufficient to establish regional background for mercury and arsenic.
    - The new analyses of the additional data resulted in a cPAH regional background value changing from 180 to 210 μg TEQ/kg.

- Reviewed all identified sources (including historic sources as well as current and historical outfalls) to determine if the impact of these sources reflected current conditions.

- Reviewed all samples near current and historical sources that had been originally screened out of the data set in the second screen. (Ecology had previously determined that these samples had been directly influenced by current and historical sources). Ecology carefully reviewed the data to determine if the potential for these sources to directly influence elevated concentrations had been accurately judged. It was determined that these samples had been screened out appropriately.

- Re-analyzed data using a different statistical population separation method. In the population separation analysis, a likelihood-based method for separating finite mixture models was used to identify the breakpoints between adjacent populations (e.g., between natural and regional background). Robust prediction limits were used only to interpret results within geographically clustered samples.

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