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Lake Washington Area Regional Background

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For more information contact:

Toxics Cleanup Program
P.O. Box 47600
Olympia, WA 98504-7600

Phone: 800-826-7716

Washington State Department of Ecology - www.ecy.wa.gov

- Headquarters, Olympia 360-407-6000
- Northwest Regional Office, Bellevue 425-649-7000
- Southwest Regional Office, Olympia 360-407-6300
- Central Regional Office, Yakima 509-575-2490
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Regional Background

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Washington State Department of Ecology
Toxics Cleanup Program
Olympia, WA

Lorraine Read, TerraStat Consulting Group

Toxics Cleanup Program
Washington State Department of Ecology
Olympia, Washington

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Acknowledgements

The Department of Ecology (Ecology) would like to thank the following authors of this study:

- Chance Asher, Ecology
- Lorraine Read, TerraStat Consulting Group
- Laura Inouye, Ecology

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List of Acronyms

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
KM	Kaplan-Meier
LPL	lower prediction limit
PCB	polychlorinated biphenyl
PQL	practical quantitation limit
SCO	sediment cleanup objective
SCUM II	Sediment Cleanup Users Manual II
SMS	Sediment Management Standards
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; Washington Administrative Code [WAC] 173-204) to establish a new framework for identification and cleanup of 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 advisory group process for the rule revisions, it was recommended that Ecology be responsible for establishing regional background sediment concentrations for areas of the state. This draft report includes 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 natural and regional background concentrations, as 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, which 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 includes a definition for regional background (WAC 173-204-505(16)) and parameters for establishing regional background (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.*

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 through working on initial bays and after receiving comments from stakeholders and tribes. Current guidance for establishing regional background based on these discussions and completed studies can be found in the Sediment Cleanup Users Manual II (SCUM II; Ecology 2015a), Chapter 10.

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 collection of 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 draft report is based on existing sediment data collected from Lake Washington, Union Bay, the Montlake Cut, and Portage Bay, 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 Resources Area (WRIA 8) that may be within the direct influence of sites and sources, such as Lake Union. The Lake Washington Area is an applicable surrogate because it receives diffuse urban sources, is relatively less impacted from chemical contamination than other urban lakes such as Lake Union, is geographically proximate, and 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, as well as the Lake Washington Ship Canal area east of the Hiram Chittenden Locks. Regional background established in this report is not applicable to river systems, or less developed suburban or rural lakes.

This draft report represents Ecology's approach for using existing data to establish regional background concentrations, as well as the first proposed regional background concentrations for freshwater urban lakes. Section 5.0 includes a summary of this approach, important limitations, and suggested guidance for using this approach in other areas. This approach and the resulting regional background concentrations for the Lake Washington Area may be further refined through the public review process prior to finalization

2.0 Conceptual Lake Model

The Lake Washington Area in which existing data was evaluated to determine regional background concentrations was defined as Lake Washington, Union Bay, the Montlake Cut, and Portage Bay (Figure 1). These water bodies are hydraulically linked, considered representative of mixed urban uses, and are connected to the more industrial areas of Lake Union, the Ship Canal, and Salmon Bay before discharging to Puget Sound. Lake Washington receives inputs from the Sammamish and Cedar Rivers, which are predominantly developed watersheds, and inputs 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 such as industry, residential and industrial stormwater, water-dependent uses, cleanup sites, as well as 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, covering 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. It is surrounded by 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). A large residential island of approximately 34 km^2, Mercer Island, occupies the southeast area of the lake.

Land use around Lake Washington 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. A floatplane base is located in Kenmore and the Renton Municipal Airport and Boeing are located at the south end of Lake Washington. Historic land uses on the lake were more industrial and 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 the lake on floating bridges, I-90 to the south and SR 520 to the north. There are federally authorized navigation lanes 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. The 760-m long Montlake Cut connects Union Bay with Portage Bay, and is a man-made channel created in 1916 to provide 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. I-5 crosses north-south to the west and SR 520 extends from I-5 across Lake Washington to I-405.

Lake Washington is within the Usual and Accustomed Fishing Areas of the Muckleshoot, Suquamish, and Tulalip Tribes. It is 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 remainder. The watershed is primarily developed (67%), with the exception of the upper Cedar River watershed, which provides Seattle's water supply. Its outlet is through Union Bay, the Montlake Cut, and Portage Bay, into Lake Union, then through the Hiram M. Chittenden Locks and Salmon Bay to Puget Sound (King County 2015). Historically, Lake Washington was landlocked prior to construction of the Montlake Cut and the Lake Washington Ship Canal in 1916. The construction of the canal lowered the lake by 3 m and diverted the Cedar River into the lake.

Lake Washington is a glacially formed lake with steeply sloping sides, averaging 33 m deep and 65.2 m deep at its deepest point. Water levels in the lake are controlled by the Hiram M. Chittenden Locks and average about 7 m 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 that were downloaded from Ecology's Environmental Information Management System (EIM). Lake Washington 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 of the lake. Non-native clean sand has been imported in some shoreline areas to create swimming beaches and parks.

Very little specific data on sedimentation in the lake is available. While much of the lake likely receives little sedimentation, particularly since source control has reduced eutrophication of the lake, areas near the river mouths receive periodic siltation and require occasional dredging for flood control and navigation. Deeper areas of the lake likely receive slow siltation through deposition and erosion from nearshore areas.

2.4 Unrepresentative Areas

Areas that were considered unrepresentative of Lake Washington sediments from a grain size or organic carbon standpoint included (Figures 2 and 3):

- *Swimming Beaches.* In a number of areas around Lake Washington, sediments were imported to enhance swimming beaches. These sediments are not native to Lake Washington, are coarser-grained, are generally very clean, and more in the range of natural background than regional background. Many of these beaches have been sampled by King County 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.
- *High TOC Areas.* The following areas were identified and considered not representative of sediments in the lake 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. Consistent with the SMS rule, Ecology focused on identifying the sites and sources that had relatively high potential to directly influence existing data concentrations, described below from north to south (Figures 4-6). There are other potential sources in the region that are not included in the below list, such as other stormwater drainages and nonpoint sources. This list is not intended to include all potential sources, but rather sources that had high potential to directly influence sediment with nearby existing data.

- *Kenmore Marinas.* The area including the North Lake Marina and Harbour Village Marina at the northeast end of Lake Washington, due to 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.
- *Kenmore Air Harbor.* One of the largest seaplane bases in the world, Kenmore Air Harbor conducts refueling and maintenance of its planes at its Kenmore location between the marinas and the barge area at LakePointe. Minimal data is available for this area.

- *Former Landfill and Barge Area around Kenmore Industrial Park (LakePointe).* Kenmore Industrial Park was an upland MTCA cleanup site that was historically used as a landfill for industrial debris. Petroleum hydrocarbons and metals were found in soils and groundwater at the site, and PAHs have been found in sediments in the barge area north of the site (Ecology 2001).
- *Former Naval Station and NOAA Facilities at Sand Point***.** Both of these facilities had docking areas at which low levels of metals and PAHs were found in early sediment investigations in the 1990s.
- *Quendall Terminals.* 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 from an SR 520 storm drain through a swale north of the highway into Yarrow Bay.
- *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.

3.0 Data Screening and Analysis

Existing data for the geographic region were downloaded from EIM and screened (excluded) from the data set to be used to calculate regional background to ensure consistency with the SMS (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 (1 and 3 below) were used to address the issues with existing data. In total the following screening steps were performed to exclude data considered not representative of a regional background distribution:

- 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).
- 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).
- 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, identifying outliers (Tables 1 & 2; Appendix B and Figure 8).

It was determined that sufficient data existed in EIM to evaluate if regional background could be established for cPAHs, arsenic, and mercury. The congener data for PCBs and dioxins/furans outside known cleanup sites is 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 from 2005 and earlier 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 2005 data set for cPAHs had a smoother distribution with fewer high-end outliers. Therefore, it was determined that data sampled from 2005 and earlier 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, 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, including:

- 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 retained.
- 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 was 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 further evaluated from a statistical standpoint to address issues specific to using existing data, as described below and in Appendix B. Tables 4a - c and Figures 4 – 6 provide the data set for these analytes and differentiates which data were screened out by age (older than 10 years), depth (non-surface sediment), high TOC, non-detect issues, and potential and known sources. This screened data was then used to conduct statistical analysis (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 above was appropriate for all three chemicals. This is because the data tended to have similar trends in the 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 long and varied history of sources of these chemicals within the lake system. As anticipated, the autocorrelation distances were different for each analyte: 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, and generally followed the methods proposed by Singh et al. (1994). Modifications to this process were made due to the lack of a dominant signal from regional background within the data set and the high percentage of undetected values at the low end of the cPAH TEQ data distribution.

The following steps were carried out:

1. Preliminary robust prediction limits were estimated and used to identify distinct subpopulations using only the independent samples (i.e., those samples not part of a cluster), based on the minimum autocorrelation distance identified in Section 3.5.

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, the population separation analysis was repeated including both the independent samples and the cluster averages calculated in Step 3.

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. Seventeen 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 five values similar to or exceeding the Puget Sound natural background 90/90 UTL, ranging from 10 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 slight signal for arsenic that may represent regional background.

The mercury dataset had eight 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 could be considered a single population and 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.

Following identification of 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, and 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 5 and 8 respectively; Table 2). Using the samples with values between 29 to 205 ppb for cPAHs, the precision is 33%. Precision becomes worse using the samples with values higher than 205. A precision of 25% or less is ideal, but may not be achievable with all data sets, particularly existing data sets. For example, Ecology determined that a precision of 30% was acceptable for a previous regional background study using new data (Ecology, 2015b). Considering the patchiness of this existing data set, a precision of 33% was determined generally acceptable but not ideal.

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 for 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 evaluate the safety of swimming beaches, to monitor sediment quality near stormwater and combined sewer overflow outfalls, to evaluate general sediment quality, and to collect data for dredged material evaluations or remedial investigations. This tended to bias the data set to nearshore areas and to 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 one obvious outlier (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 past 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 suggesting 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 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 180 µg TEQ/kg. The data set on which this values is based is fairly limited in size (n = 17) approximately two-thirds of the Ecology recommended minimum sample size for newly collected data, 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 propose regional background values for these CoCs until additional data is collected.

5.0 Guidance for Using Existing Data to Calculate Regional Background Concentrations

This section provides Ecology's guidance on developing regional background values using existing data, based on the Lake Washington area effort. Ecology recognizes that other geographic areas may warrant departures from the approach described below, based on area-specific conditions and the nature and quality of the existing data. Similar to the approach Ecology has developed for new sampling data, this approach using existing data may evolve over time based on public comment and experience with implementation.

5.1 Similarities to Developing Regional Background using New Data

Section 10.3.1 of SCUM II includes details of the approach Ecology has developed to establish regional background values using new data, consistent with the SMS rule. Many of the same steps and guiding principles should be used when calculating regional background based on existing data, for example:

- Develop a conceptual site model that guides the area from which sediment data will be selected. As part of the model, describe relevant features of the water body, including land use, bathymetry, hydrology, grain size/TOC, known sites and sources, and presence of bioaccumulative chemicals.
- Once the overall area of interest has been selected, exclude areas near known sites and sources with high potential to directly influence sediment concentrations.
- Exclude areas that are considered natural background, have unusually high TOC, or are otherwise unrepresentative of the water body as a whole.
- Determine whether different areas of interest should be identified for different chemicals or whether different samples should be included for different chemicals.
- Ensure that the data are of acceptable quality, screening out data of unacceptable quality.
- Identify the sample independence distance and ensure that the data retained for analysis meet this criterion to avoid sample bias (especially important for existing data sets).
- Depending on how the data set is described, conduct an outlier analysis and remove outliers as appropriate.
- Conduct a population separation analysis to obtain a distribution representative of regional background.
- Conduct a principal components analysis, if it is determined useful.
- Calculate and report precision for the final data set, screening out analytes that do not meet precision targets or screening out samples that unduly degrade precision.

- Use the 90/90 UTL to calculate regional background (but see Section 5.2.5 – 5.2.7 below).

Several of these steps require modification to work with existing data sets and some additional steps are needed, discussed below.

5.2 Additional or Modified Steps for Developing Regional Background using Existing Data

The following sections include additional or modified steps that may be necessary when using existing data to calculate regional background.

5.2.1 Minimum Data Requirements

Data for the area of interest may be downloaded from EIM or other available sources. The chemicals for which regional background can be calculated will depend on the availability of sufficient data once all screening steps have been completed. When calculating regional background based on new data, Ecology estimated that at least 25 samples were preferred, with an equal number of samples archived in case additional data were needed to fill in part of the distribution. Therefore, it was anticipated that at least this many data points would be needed to calculate regional background using existing data, once all screening steps were completed. However, data sets as small as the dataset used for cPAH TEQs could be sufficient if the data were generally well behaved (symmetric and with adequate precision) and/or appeared to be representative of the regional background population of interest.

Furthermore, the data set should:

- Encompass the range of concentrations found in the water body away from sites and sources, to adequately define the 90/90 UTL.
- Be of adequate quality.
- Be geographically representative of the water body, to the degree possible.
- Not include anomalous samples or data sets that are distinctly different from the rest of the distribution.

It may be helpful to conduct additional statistical analyses to determine whether multiple distributions are present in the data set, since conceptually, regional background would be a single population. This may assist in determining whether certain samples should be included or excluded. Statistical evaluation of excluded data should be accompanied by a clear rationale that provides a logical explanation for why the samples are different.

For the Lake Washington area, initially there were substantially more than 25 samples in the data set. However, distributional and precision analysis indicated that the data set contained several distinct distributions. These distributions were carefully analyzed to select data that were considered most representative of the geographic area as a whole, represented a single distribution, and had good precision. As a result, the final data set had fewer than the expected minimum number of samples, but met all of the other conceptual, regulatory, and statistical requirements for regional background. Therefore, the recommended minimum number of samples of 25 is considered ideal, but is not a hard and fast rule as long as other requirements are met.

5.2.2 Evaluation of Older Data

A recency cutoff for the data set (e.g., 10 years for this study) should be established to ensure that the calculated regional background represents relatively current conditions, to the extent possible. Selection of the recency cutoff should include consideration of:

- The conceptual model for the area, e.g., the sedimentation rate compared to the depth of the samples.
- Other changes that may have occurred in the area, such as source control efforts.
- Changes in analytical methods that may have affected the existing data values.
- Data quality and the ability to obtain backup documentation of methods and quality assurance.
- The results of any statistical evaluations showing breakpoints in the data set.

In many cases, older data will be co-located with or nearby more recent data. In all such instances, the more recent data should be used unless there is a specific reason for excluding the more recent data.

5.2.3 Sample Depths

A cutoff should similarly be established for sample depth. This will depend in part on the conceptual model for the area. Samples should not be used with depths that extend well below relatively recent sediments, as determined by the sedimentation rate and the date of sampling. However, samples need not necessarily be limited to 2 to 10 cm in depth, as this would likely limit the amount of useable data in many areas.

5.2.4 Data Quality

Under the SMS, regional background can define the CSL, which is considered regulatory criteria. Therefore, it is important that data used to calculate regional background be of good quality. Ideally, data will have undergone QA2 review (also known as EPA Level III/IV, SCUM II Chapter 5) prior to or as part of the regional background calculation. However, Ecology will use professional judgment in accepting data that have undergone QA1 review (also known as EPA Level I/II, SCUM II Chapter 5) if there is no evidence of bias or concern.

5.2.5 Representativeness

Representativeness is a challenge when working with existing data, as most existing data sets were not collected with the goal of evenly characterizing general conditions in a water body.

Best professional judgment will need to be used along with the conceptual model to evaluate potential biases in the data set. If those biases are substantial enough, then collection of new data may be preferable to fill important data gaps.

For example, in Lake Washington, it is generally believed that deeper sediments are finer-grained and likely serve as the ultimate repository for chemicals entering the lake. Ideally, characterization of regional background would include deeper lake samples. However, relatively recent data were limited in these areas. It is therefore possible that the existing data set is biased low in terms of characterizing the entire lake. On the other hand, using primarily shoreline samples collected closer to sites and sources could introduce unrepresentative high concentrations and increase the variability in the data set, thus increasing the 90/90 UTL. All of these issues were considered. Careful screening of the data and confirming data independence were relied on to ensure that high-concentration samples and proximity to sources did not bias the data high. Similarly, unrepresentative samples at swimming beaches that were coarser and cleaner than others were clearly identified as a different population and removed. The limited number of deeper lake samples remains a concern, offset by the reality that regional background would apply to sites predominantly located at the shoreline, where the majority of the data to establish regional background were collected.

5.2.6 Data Independence

Data independence is especially important for existing data sets. Many existing data sets may have been designed for biased sampling of sites and sources. This presents several problems. First, the data may be biased toward areas with higher concentrations. Second, the data may be too close together and not independent of one another. Together, these challenges contribute to an overall lack of representativeness of the water body as a whole, particularly in those areas that would meet the SMS definition of regional background.

Therefore, an evaluation of the autocorrelation distance should be conducted as described in Section 3.5 and Appendix B. Once the autocorrelation distance is determined, it should be applied to the screened data set to further remove (or average) any samples that are too close together, minimizing the bias toward heavily sampled areas.

Decision rules may need to be developed to determine which samples to remove. Older samples should in general be removed first. However, clusters of samples may remain that were sampled at the same time. For this data set, simulations were used to determine the effect of randomly selecting stations from clusters for removal. These simulations showed that due to the heterogeneity of the data, the specific samples retained could have a substantial effect on the 90/90 UTL. Therefore, clusters of autocorrelated samples were identified and concentrations within the same subpopulation were averaged, but kept separate from autocorrelated samples from different subpopulations. Other alternatives could be considered in areas with different data distributions.

5.2.7 Precision and Distributional Analysis

As described in Section 3.3.3, precision is a measure of the spread of the data set. If precision is poor (% is high), the 90/90 UTL will be higher than if precision is good (% is low). Because compilations of existing data sets have been collected for varying purposes, they will likely have poorer precision than those that are synoptically collected and analyzed for a specific purpose. Therefore, it is particularly important to calculate precision for existing data sets and evaluate whether it is sufficiently low to be useable. The target Ecology has established for the purpose of establishing regional background with synoptically collected data sets is 25%. Existing data sets may or may not be able to meet this target, but it should serve as a general goal to ensure that regional background values calculated for various geographic regions have a similar degree of conservativism regardless of the type of data set used.

The various screening steps described in Section 3 have a substantial effect on precision. If the decision is uncertain, it can be helpful to calculate precision throughout the process to evaluate the appropriateness of screening data. If precision is substantially improved by screening out specific data, it is likely that these data were unrepresentative of the rest of the population or introduced substantial variability into the data set.

It may be the case, as with Lake Washington area data set, that the data set is made up of several different clearly identifiable distributions, therefore reducing the precision and increasing the variability of the overall data set (even when none of the individual values qualifies as an outlier in the combined data set). Where this is the case, the individual distributions should be carefully evaluated for screening, both at the low and high end. The goal of this screening is to obtain a data set that is 1) representative of the geographic area being evaluated, 2) consistent with the SMS definition of regional background, and 3) represents a single statistical distribution with reasonably good precision.

If all of the above screening steps have been attempted and precision is still very high, it may be appropriate to reconsider whether the data set is usable for this purpose.

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Tables

Tables 1 – 3 pages 24 - 25

Tables 4a - c are in Appendix A, page 36

Tables 5 – 15 are in Appendix B pages 55 - 62

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

Population 1 was used to calculate regional background. *Sample removed from regional background calculation due to high potential influence from a source. See Figure 8 for additional detail.

Location Description	cPAH TEQ(ppb)	% Fines
Lowest Concentration Samples (mainly swim beach samples)		
25 independent observations or cluster averages	<2.6 – <6.7	<1 – 21
Apparent Natural Background Samples		
9 independent observations or cluster averages	11 – <23	2 – 74
Selected Regional Background Samples (Population 1)		
Lyon Creek Waterfront Preserve	29	3
Houghton Beach Park	29	12
South of Newcastle Beach Park	38	21
Portage Bay (near the University of Washington; average)	42	--
Near Newport Yacht Club	45	19
Harbor Village Marina (average)	46	6
Boeing (average)	58	73
Middle of the lake west of Mercer Island	72	77
Middle of lake between the southwest shoreline of Mercer Island /Rainier Beach	75	75
Kenmore Navigational Channel (average)	76	41
May Creek	92	14
South of Pleasure Point	100	16
Middle of the lake between Magnuson Park and Kirkland	100	80
Montlake Cut	110	--
Pleasure Point	130	20
McAleer Creek	160	12
South of Newcastle Beach Park (average)	205	49
Boeing*	220*	53
High-Concentration Samples		
3 independent observations or cluster averages	330 – 1,900	16 – 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
Middle of the lake west of Mercer Island	46	77
Middle of the lake 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 the lake between the southwest shoreline of Mercer Island /Rainier Beach	0.16	75
Near Boeing, nearshore	0.21	42
South of Newcastle Beach Park	0.21	56
Kenmore, inner navigational channel	0.24	31
Middle of the lake west of Mercer Island	0.37	77
Middle of the lake 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	17	17/17	Gamma	83	48	180 µg TEQ/kg	21 µg TEQ/kg	33%

Figures

Figures 1 – 9 pages 27 - 35

Figures 10 – 15 are in Appendix B pages 49-- 54

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

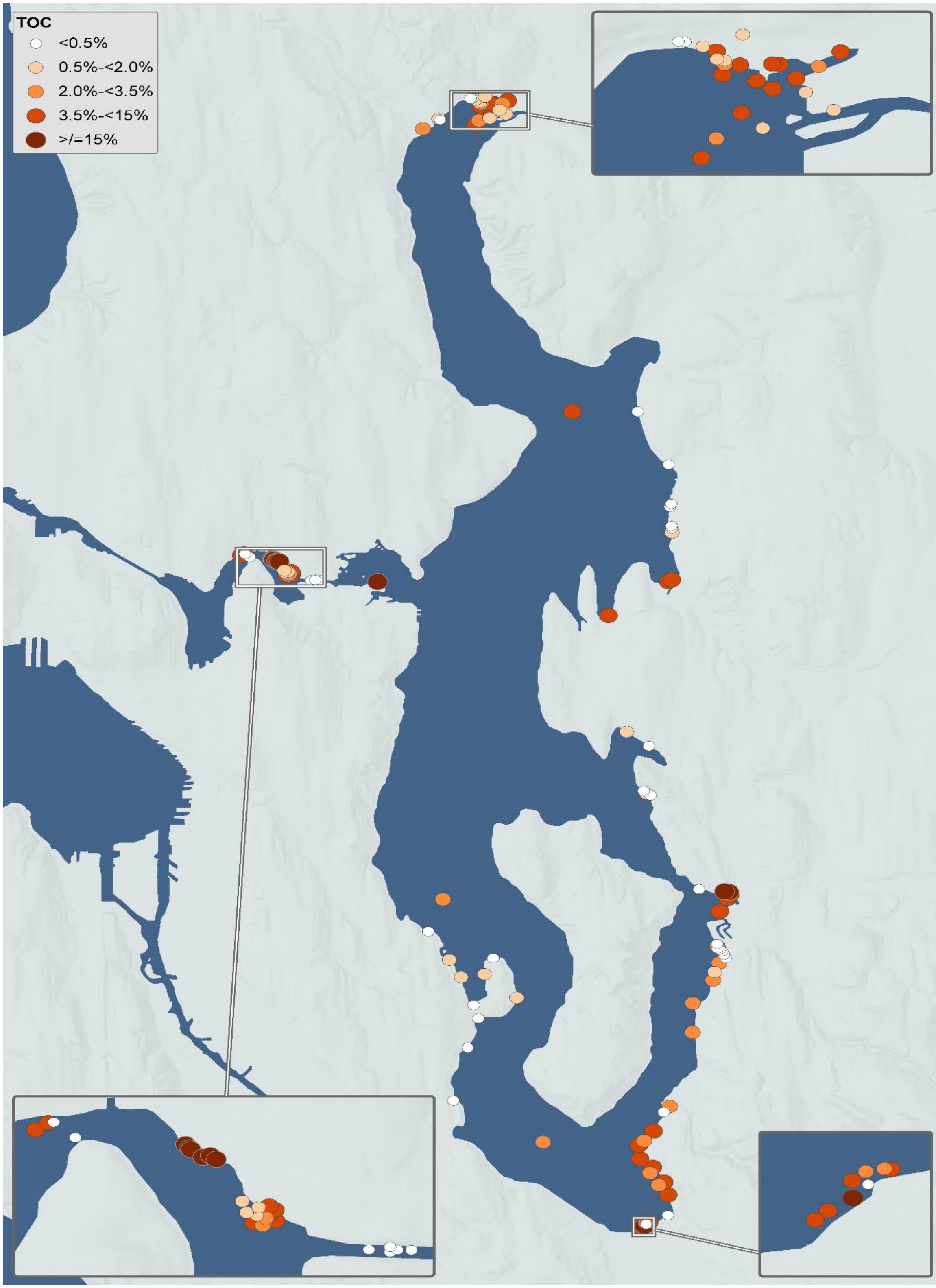

Figure 2: Total organic carbon throughout the Lake Washington area of interest.

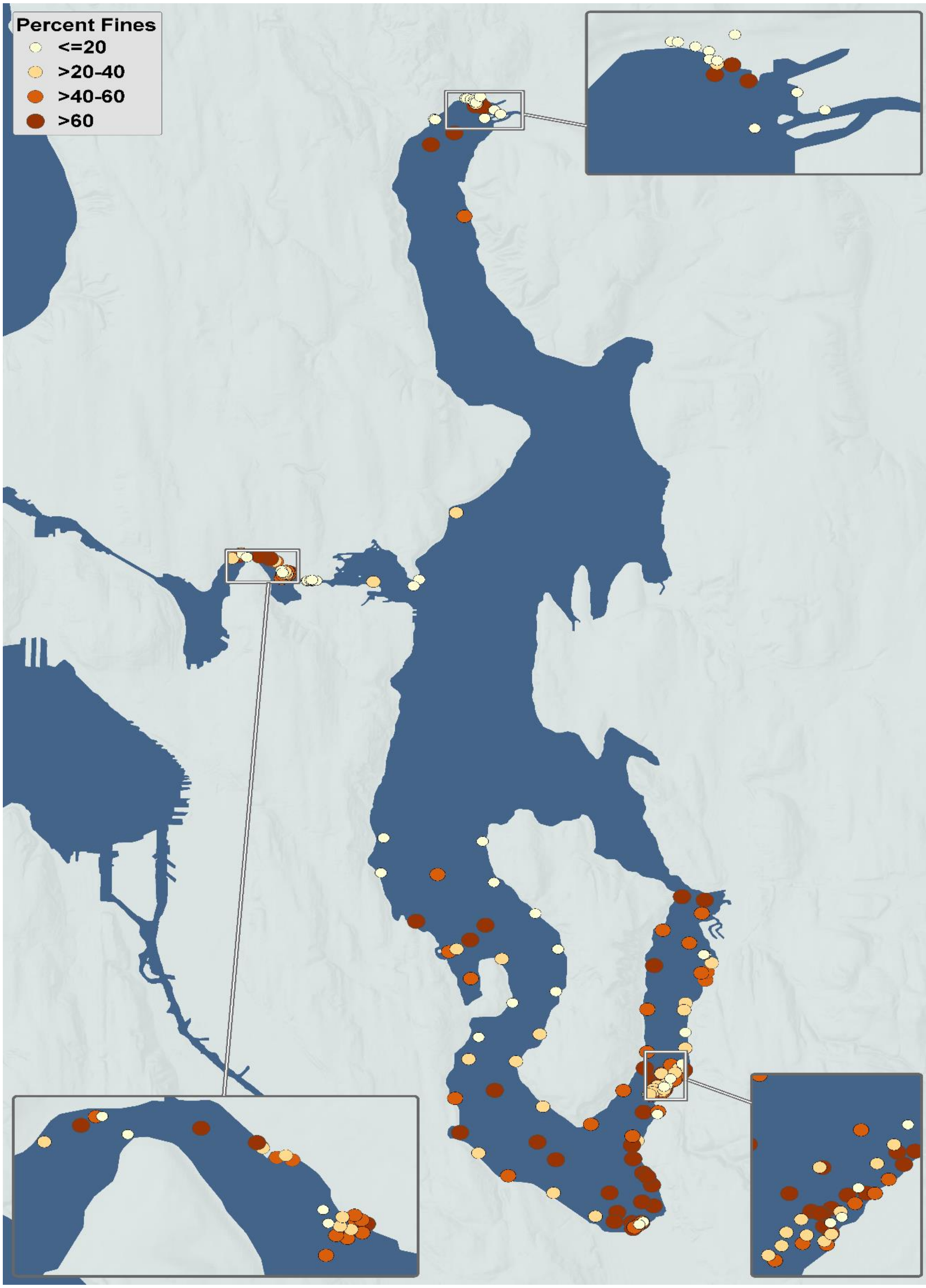

Figure 3: Percent fines throughout the Lake Washington area of interest.

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

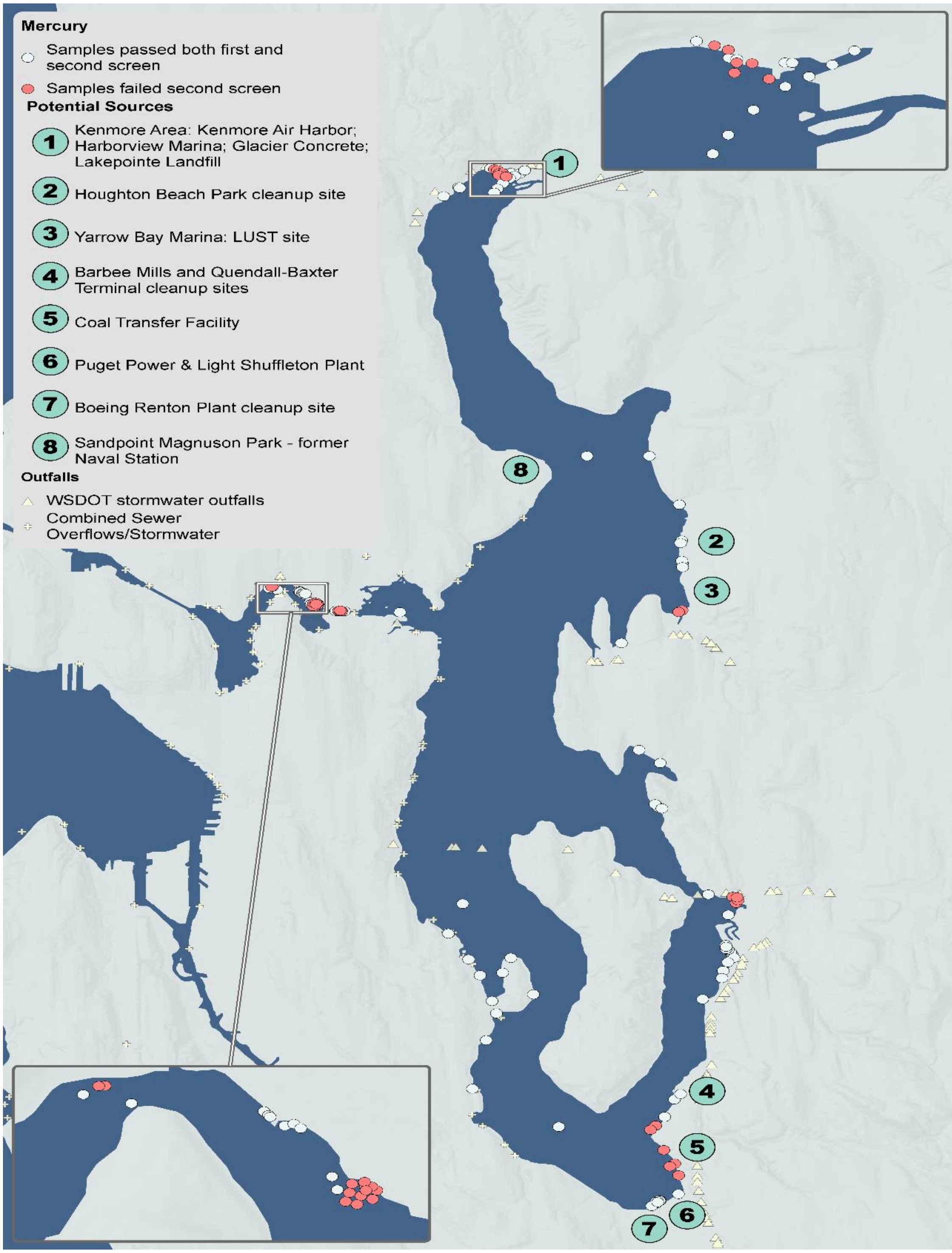

Figure 5: The mercury data remaining after being filtered by the second screen (orange circles) for potential impact from sites and sources and high TOC.

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.

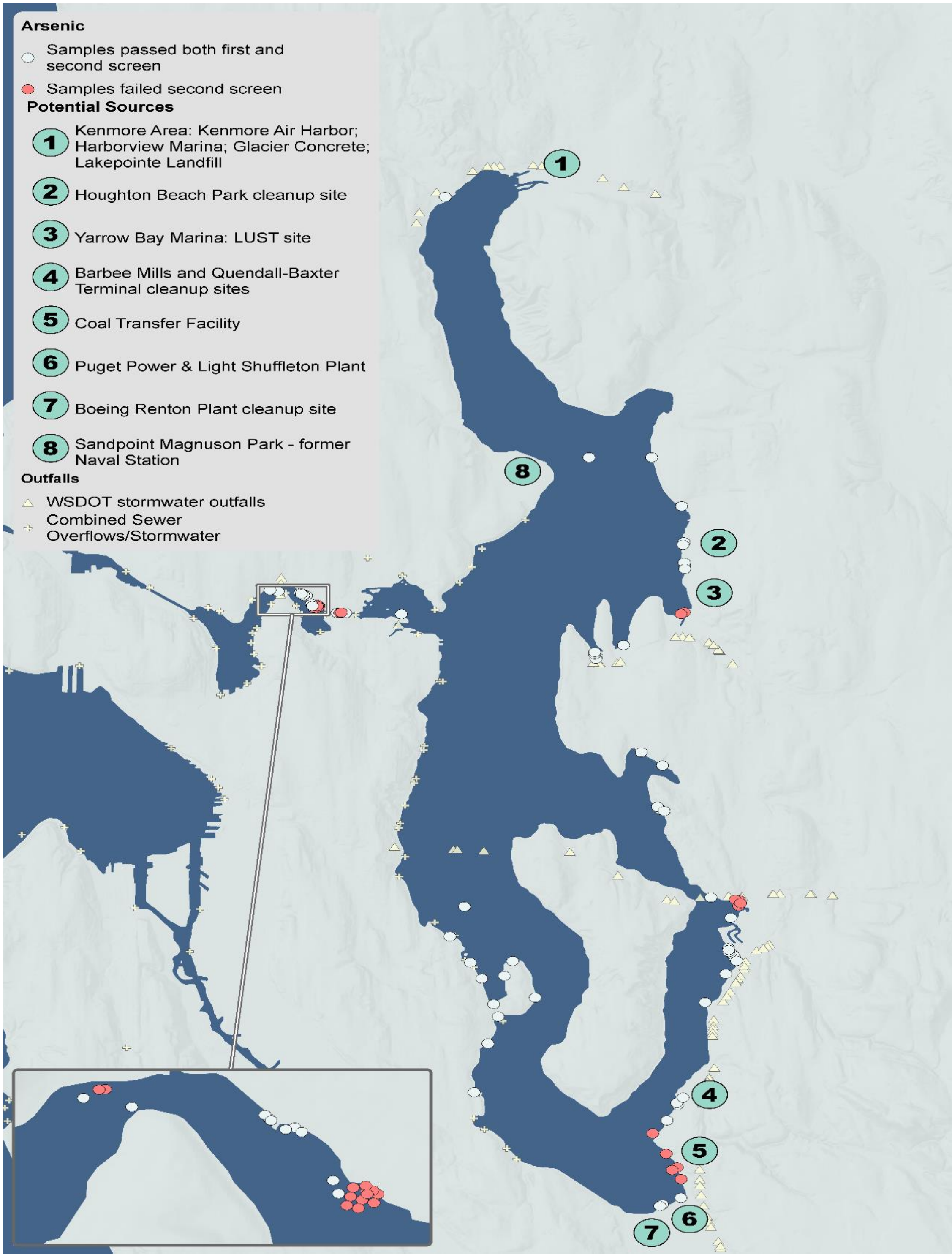

Figure 6: The arsenic data remaining after being filtered by the second screen (orange circles) for potential impact from sites and sources and high TOC.

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.

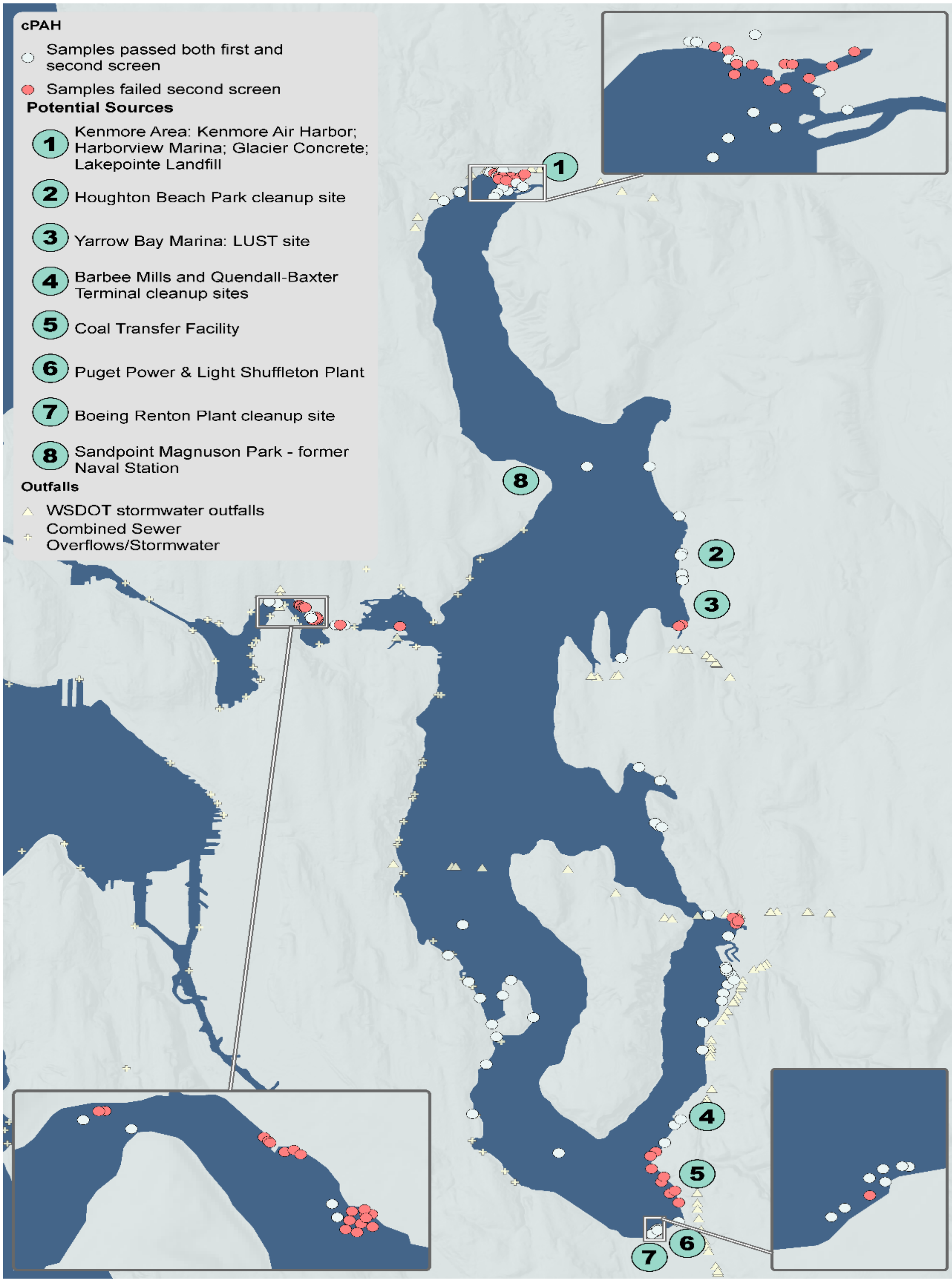

Figure 7: The cPAH data set remaining after being filtered by the second screen (orange circles) for potential impact from known sources and sites and high TOC**.**

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.

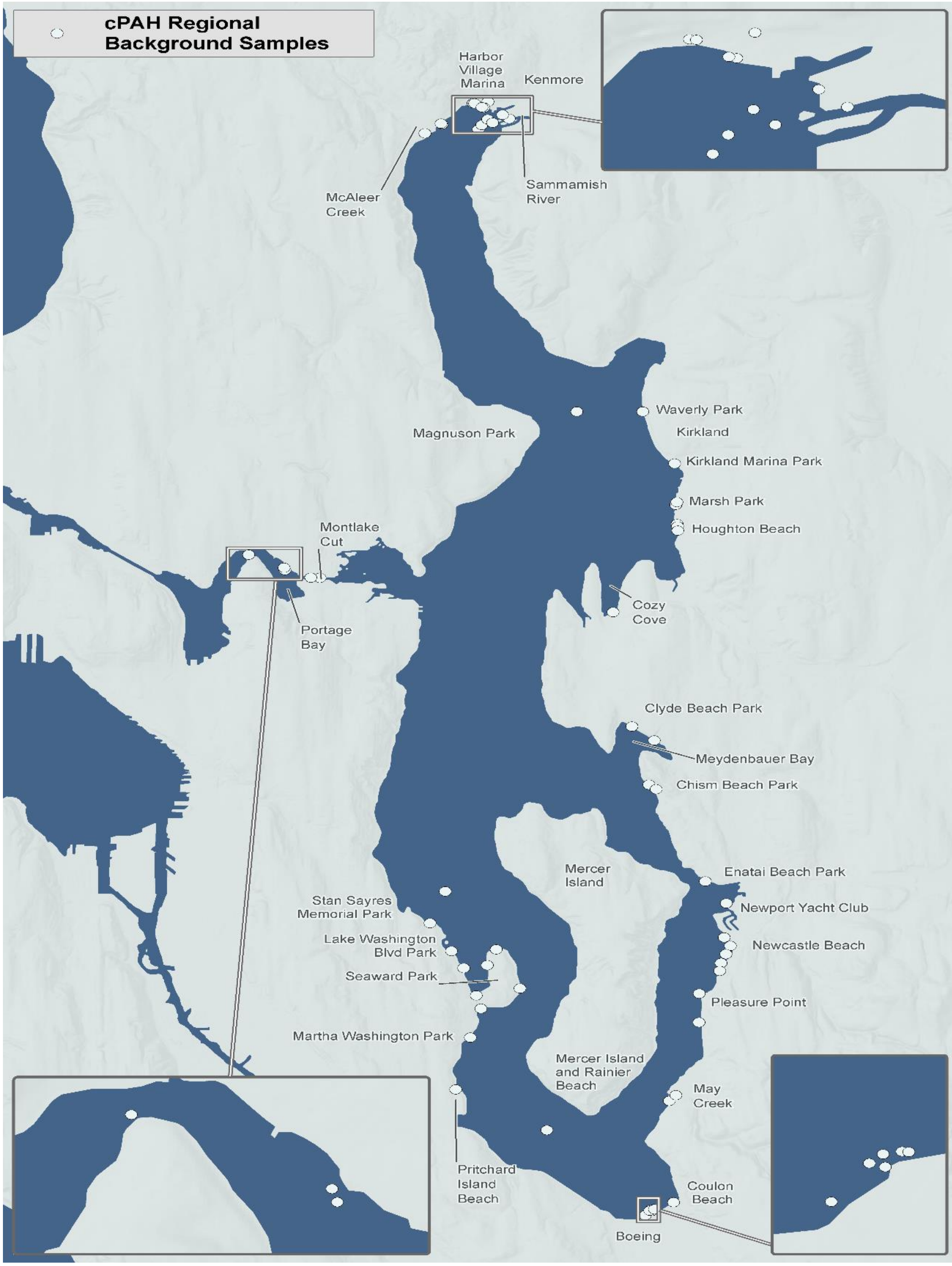

Figure 8: The remaining cPAH data set after statistical analysis.

This data set 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.

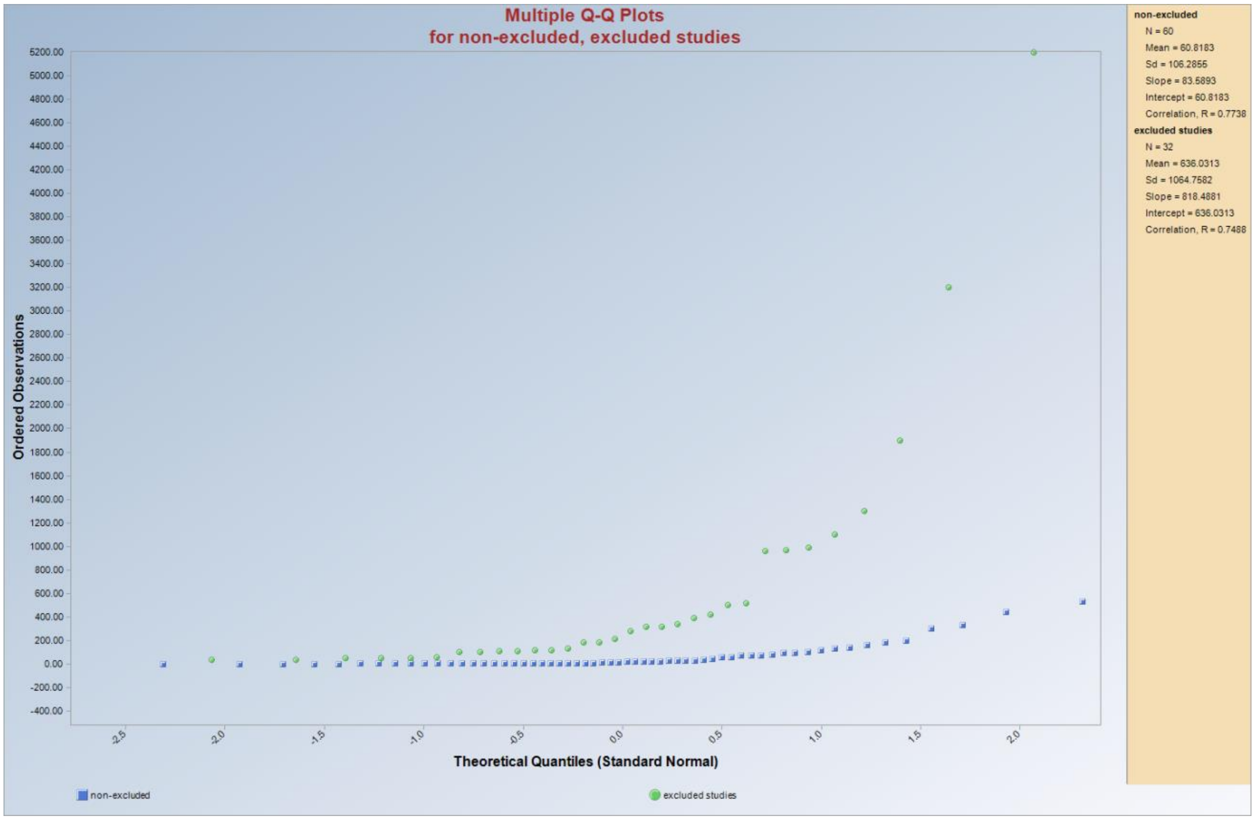

Figure 9: Quantile-Quantile plot comparing 2005 and earlier data (**green dots**) to post-2005 cPAH data (**blue dots**).

The decision for the first screen to exclude data older than 2005 was based on the results of this analysis.

Appendix A. Data Tables

To view Tables 4 a – c as an excel spreadsheet, see:
https://fortress.wa.gov/ecy/publications/SummaryPages/1609064.html

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) and the reasons for excluding specific samples.

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.

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.

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 intensities. 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 computing 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 from within the lake. This iterative process involved multiple steps listed below, summarized in Table 5 and described in more detail in Section B.2:

- *Step 1.* 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.

- *Step 2.* A population separation analysis (Singh et al. 1994) was initially applied only to 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 present in the data set.

- *Step 3.* 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 population based on concentration. Subsequently, any samples closer than the autocorrelation distance within each subpopulation were averaged.

- *Step 4.* The population separation analysis in Step 2 was repeated, this time including the cluster averages generated in Step 3 along with the independent samples. This produced a final set of prediction limits for each subpopulation present within the data set. Once the subpopulations were separated, the specific subpopulation representing regional background was identified.

- *Step 5.* Precision and 90/90 UTL estimates were calculated for the identified regional background subpopulation.

B.2 Methods

The sections below describe each of the above steps and statistical methods used in greater detail.

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 5^{th} order polynomial). The six polynomial regression models were compared using the Aikake Information Criterion corrected for sample size (AIC_c;

Burnham and Anderson 2002), and patterns in the residual diagnostic plots. The model with the lowest AIC_c 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

The Singh et al. (1994) approach was applied for population separation within the data set. This approach includes visual inspection of QQ distribution plots and calculation of robust statistical limits to define the boundaries of apparent individual subpopulations within a mixed data set.

Singh et al. (1994) recommend calculating the robust limits on the full data set after excluding any obvious elevated values. One of their primary assumptions is that there is a single dominant population present (natural background in their examples), and the objective is to partition out the smaller subpopulations that are influencing the mixture. This data set is fairly limited in sample size with multiple populations present. Regional background appears to be bounded on two sides, with Puget Sound natural background on the lower end and smaller populations with elevated values on the high end. The separation analysis was approached by working down from the highest concentrations, sequentially separating out each subpopulation in turn. The preliminary thresholds between subpopulations were identified by the natural breaks in the QQ plot. 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 plots. Chester Morse Reservoir is in the upper region of the Cedar River Watershed, the watershed for Lake Washington (King County, 2016); 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). Any obvious outliers were removed and the QQ plots were evaluated to identify obvious breaks in the data. This provided preliminary thresholds between adjacent but separate 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, 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 (including obvious outliers that appeared to belong to a subpopulation once the clustered samples were included) plus the cluster averages of samples within 250-m calculated as described above. Using the full data set, the process was repeated: remove obvious outliers, evaluate the QQ plot for obvious breaks in the data, and estimate robust 95% prediction limits for each of the preliminary populations. The new prediction limits were used to re-evaluate samples within clusters. If samples would be allocated to different subpopulations than was done in the preliminary pass, they were moved, cluster averages were recalculated, and the process was repeated until no samples changed to a different subpopulation

B.2.4 Principal Components Analysis

A principal components analysis (PCA) was conducted on the dataset 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 clear enough, these chemical pattern distinctions could then be used to classify samples as being 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 dataset. 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 Analysis

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:

- *cPAH TEQ:* One station from Portage Bay/Lake Union, just west of I-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.

- *Arsenic:*
  - 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 dataset was almost 8-km away.
- *Mercury:*
  - 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. 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 correlations that were negative, strongly influenced by single data points, and/or 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 dataset is limited in the number of samples and in their spatial separations. The locations also are 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 dataset 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 dataset.

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: 11 from swim beaches and 17 others scattered around the lake, all of which were more than 250-m from all other stations.

The QQ plot for these independent samples (Figure 10a) was examined, identifying one elevated value at 330 ppb and a preliminary threshold between two adjacent subpopulations around 26 ppb based on an obvious break in the QQ plot. The robust 95% prediction limits calculated for the seven data points within this subset labeled "Population 1" were (LPL_1 = 32 ppb and UPL_1 = 160 ppb).

The QQ plot for the remainder of the data (all samples with concentrations less than the LPL_1 of 32 ppb, Figure 10b) was examined and a preliminary threshold between subpopulations was apparent at 10 ppb based on an obvious break in the QQ plot. The robust 95% prediction limits calculated for the four data points within this subset labeled "Population 2" were (LPL_2 = 10 ppb and UPL_2 = 26 ppb). Values between 32 ppb (the lower limit from Population 1) and 26 ppb (the upper limit from Population 2) would be considered ambiguous, requiring further evaluation. There were no samples in this overlapping region.

The QQ plot for the remainder of the data (all samples with concentrations less than the LPL_2 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 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 10.

The final pass of the process used the full data set, including the 28 observations used in the first pass plus 27 values that were included in the clusters (Table 10). The extreme value at 1900 ppb was excluded as a clear outlier. The QQ plot for the remaining 54 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. These data were not included in any of the statistical calculations or decisions about break points in the QQ plots. In Figure 11a, there was a large separation in the QQ plot between the two 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. The small sample size and patchy spatial distribution of sampling locations in this composite data set are inadequate to interpret the nature of localized trends or local hotspots (e.g., 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). In light of the generally high sampling uncertainty associated with this composite data set, the values $\geq$ 330 ppb which would have a strong influence on regional background were excluded until more information becomes available.

A clear change of slope is apparent in the QQ plot (Figure 11a) at 10 ppb, but the scale of this plot makes it difficult to identify where other slope changes may occur. A QQ plot using just the data with cPAH TEQ values ranging from 11 to 22 ppb (Figure 11b) offers a clearer picture of the distributional features, including where the breaks between multiple populations may occur. The skewness in these data is apparent. These data could represent a single skewed distribution (e.g., gamma or log-normal) indicative of a spatially heterogeneous concentrations distribution, or a mixture of overlapping distributions (e.g., one or more normal or slightly skewed distributions and one or more high concentrations). A dominant feature of the QQ plot is the apparent change in slope around 28 to 30 ppb which suggests the presence of two potentially overlapping subpopulations. Two samples with cPAH TEQ values of 29 ppb are uncertain as to whether they belong to the upper or the lower sub-populations. Comparisons of the possible sub-populations (e.g., 11 to 29 ppb and 38 to 220 ppb versus 11 to 23 ppb and 29 to 220 ppb) were made using QQ plots and robust prediction limits for the two sets of two sub-populations. All of the robust limits were self-affirming (i.e., the robust limits based on each subset of data captured all the values within the subset and no others, signifying that the two values at 29 ppb are statistically valid members of either the lower or the upper population. Preference was made to include these two values in the upper populations based on higher QQ plot correlation coefficients for sub-populations 11 – 23 ppb and 29 – 220 ppb (Table 11).

The 18 samples with concentrations between 29 and 220 ppb are Population 1 (Figure 11b). The distribution of these data is skewed such that the normal distribution is abandoned and the lognormal distribution is used instead to provide a reasonable fit to these data (Table 11 and Figure 11c). The robust 95% upper and lower prediction limits were calculated (LPL_1 = 23.5 ppb, UPL_1 = 242 ppb), showing that these limits include all values within the preliminary thresholds and no other values.

The QQ plot for the data in Population 2 (samples with concentrations from 11 – 23 ppb; Figure 11d) is indicative of a normal distribution. The robust 95% prediction limits were calculated on these 9 data points (Figure 11d; LPL_2 = 10.7 ppb, UPL_2 = 24.6 ppb).

For the remaining 25 values (with concentrations $\leq$ 11 ppb), all but three of the TEQ sums were "less than" values. This population contains all but one of the swim beach samples, and all; samples had percent fines < 21%. Because of the dominance of non-detects in this population, these data were not used to calculate an upper prediction limit and the lower limit from Population 2 was used to separate the populations.

Examination of the sample types and sampling locations for the complete cPAH data set (Figure 11a, Table 12) suggested there were three primary subpopulations plus a group of higher concentration samples (> 242 ppb) as follows:

- Very low-concentration samples of clean sand (Population 3).
- Mainly nearshore samples within the range of the Puget Sound natural background data set (Population 2).
- Samples found in depositional areas and near urban shorelines representing regional background (Population 1).
- A smaller set of increasingly higher concentration samples (330 ppb and above).

Further evaluation of the 220 ppb sample in Population was located near Boeing and spatially adjacent to two samples with very different results:

- A 43 ppb sample less that 45-m to the southwest.
- A 260 ppb sample approximately 70-m to the northeast was omitted early on because of a high TOC value of 15%.

This level of spatial heterogeneity for these samples suggests a highly localized source of contamination influencing the higher concentrations. Due to the high level of sampling uncertainty present in the data set, and the influence the 220 ppb sample had on the 90/90 UTL for regional background, this sample was screened from the regional background data set. A regional background value for cPAH TEQ was estimated from the Population 1 data with concentrations between 29 and 205 ppb (Table 1).

B.3.3.2 Arsenic

The arsenic data set included 37 independent samples: 11 from swim beaches and 26 others scattered around the lake, 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. These two samples were treated as independent in the population separation analysis despite their geographic proximity.

The full arsenic data set included the 37 independent samples and 7 values for the clusters (Table 13). In the QQ plot for these 44 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 (all samples with concentrations < 45 ppm; Figure 12b), there appears to be a change in slope at 2.9 ppm. Above this preliminary threshold, the data could represent a skewed distribution or multiple overlapping approximately normal distributions. The lognormal QQ plot fit to these data (Figure 12c) fails to adequately capture the skewness in the data even within this truncated range (i.e., excluding the three values > 13 ppm). The robust 95% upper and lower prediction limits calculated for this (log-transformed) subset labeled "Population 1" were LPL_1 = 2.6 ppm, and UPL_1 = 9.6 ppm. These limits exclude two samples with concentrations at 10 and 13 ppm, and capture five additional samples with concentrations between 2.6 and 2.9 ppm. However, those lower values appear to be more consistent with the distribution of data below 2.9 ppm. It appears likely that there are multiple, potentially overlapping distributions within this concentration range, but with insufficient data to be able to properly segregate the sub-populations.

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 four 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 2.0 to 3.9 ppm (shown in green on Figure 12b), and the concentrations in the non-swim beach samples ranged from 1.4 to 13 ppm. There was no distributional distinction between the clean swim beach samples and the non-swim beach samples (Figure 12b).

There is insufficient data in this data set 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 56 independent samples: 11 from swim beaches and 45 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, 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 56 independent samples and 8 values that fell within clusters (Table 15). In the QQ plot for these 64 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.

The five samples from Population 1 (with mercury concentrations between 0.11 and 0.24 ppm; Figure 13b) appear to be normally distributed. These data have 95% lower and upper robust prediction limits of LPL_1 = 0.13 ppm and UPL_1 = 0.26 ppm and include all samples within Population 1.

The QQ plot for the 13 samples from Population 2 (with mercury concentrations below the LPL_1 = 0.13 ppm and the preliminary threshold of 0.05 ppm; Figure 13c) appear normally distributed. These data have 95% lower and upper robust prediction limits of LPL_2 = 0.057 ppm and UPL_2 = 0.0.11 ppm and include all samples within Population 2.

The remaining 43 samples with mercury concentrations below the LPL_2 = 0.057 ppm may be a mixture of overlapping distributions. However, distinguishing sub-populations at concentrations in this range may not be particularly relevant to regional background. Therefore prediction limits were not calculated and any sample with concentrations < 0.057 ppm is considered “Population 3”. This subset of data includes all of the swim beach samples.

Most of the mercury concentrations within the data set (58 out of 64) 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 92% (59/64) 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 3 vs Population 2, Figure 13a). The regional background signal is expected to be in the concentration region above 0.11 ppm (Population 1, 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, there is insufficient data in this population 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 first two principal components explained over 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, *kmeans* in R).

The PCA showed samples also identified as elevated or influential in the univariate QQ plots (Section B.3.3) or 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 source or site influenced. The concentration patterns among samples from near sites and from the data set at large were generally similar. Concentrations were the main distinguishing characteristic, with values ranging from non-detect or very low in the clean swim beach samples to samples with high concentrations for one or more chemicals.

The *a priori* screening of samples considered 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 presupposed that samples were properly assigned to the 'site' category. 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: that sufficient distance from a source does not automatically indicate a lack of site influence. By screening samples near sources may have excluded samples with concentrations in the range of regional background. However, this is the best screening tool to avoid including too many samples in the regional background data set that are directly influenced by sites or sources. All samples beyond a safe 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 from further analysis.

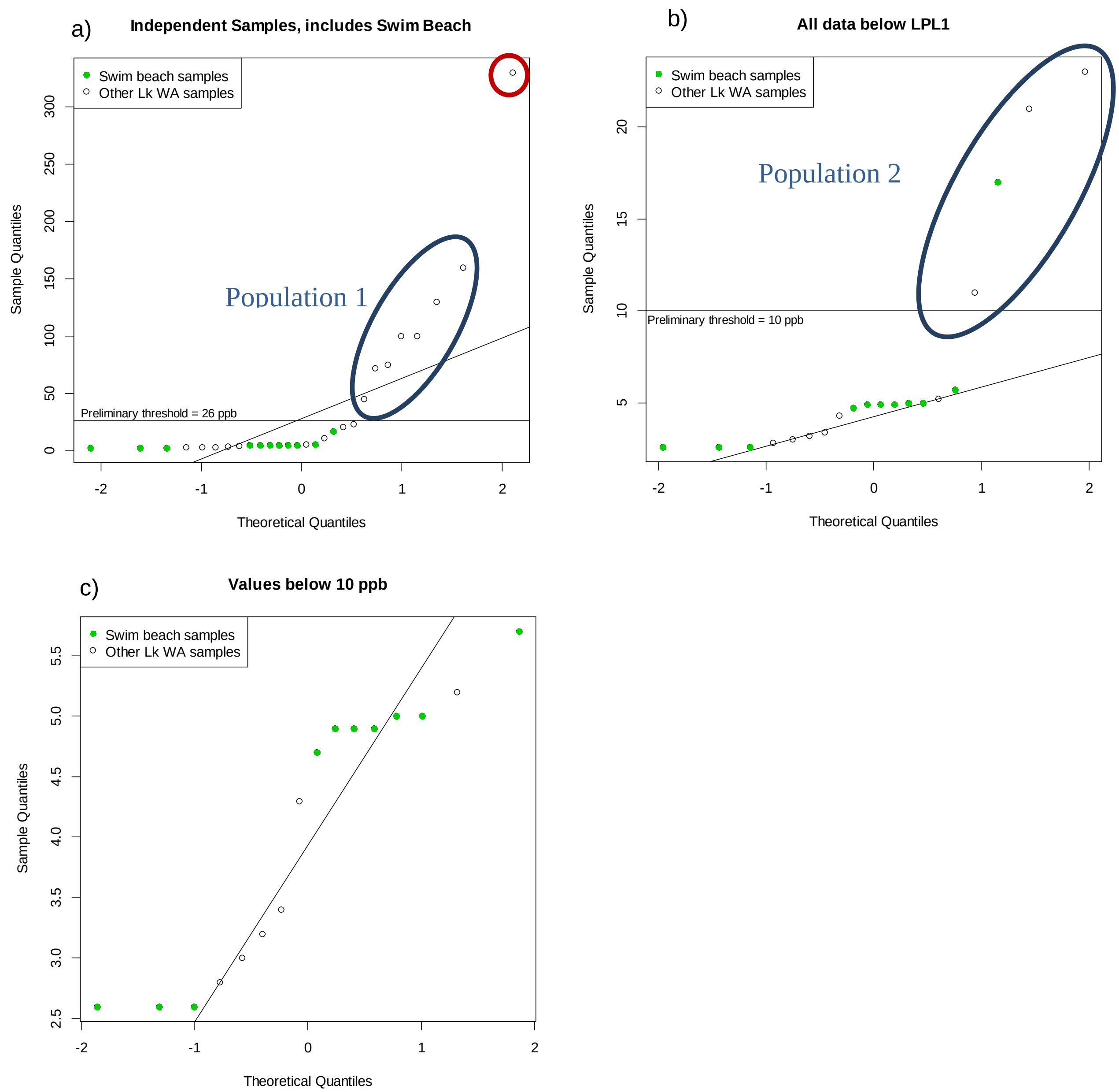

Figure 10: Sequential QQ plots for the cPAH TEQ (µg/Kg, dw) data, excluding samples within clusters (n = 28).

a) All data with one elevated value (**circled in red**) and Population 1 identified subjectively based on breaks in the QQ plot.
b) QQ plot for data excluding elevated values and Population 1, with Population 2 identified based on a break in the QQ plot.

c) QQ plot for the remainder of the data.

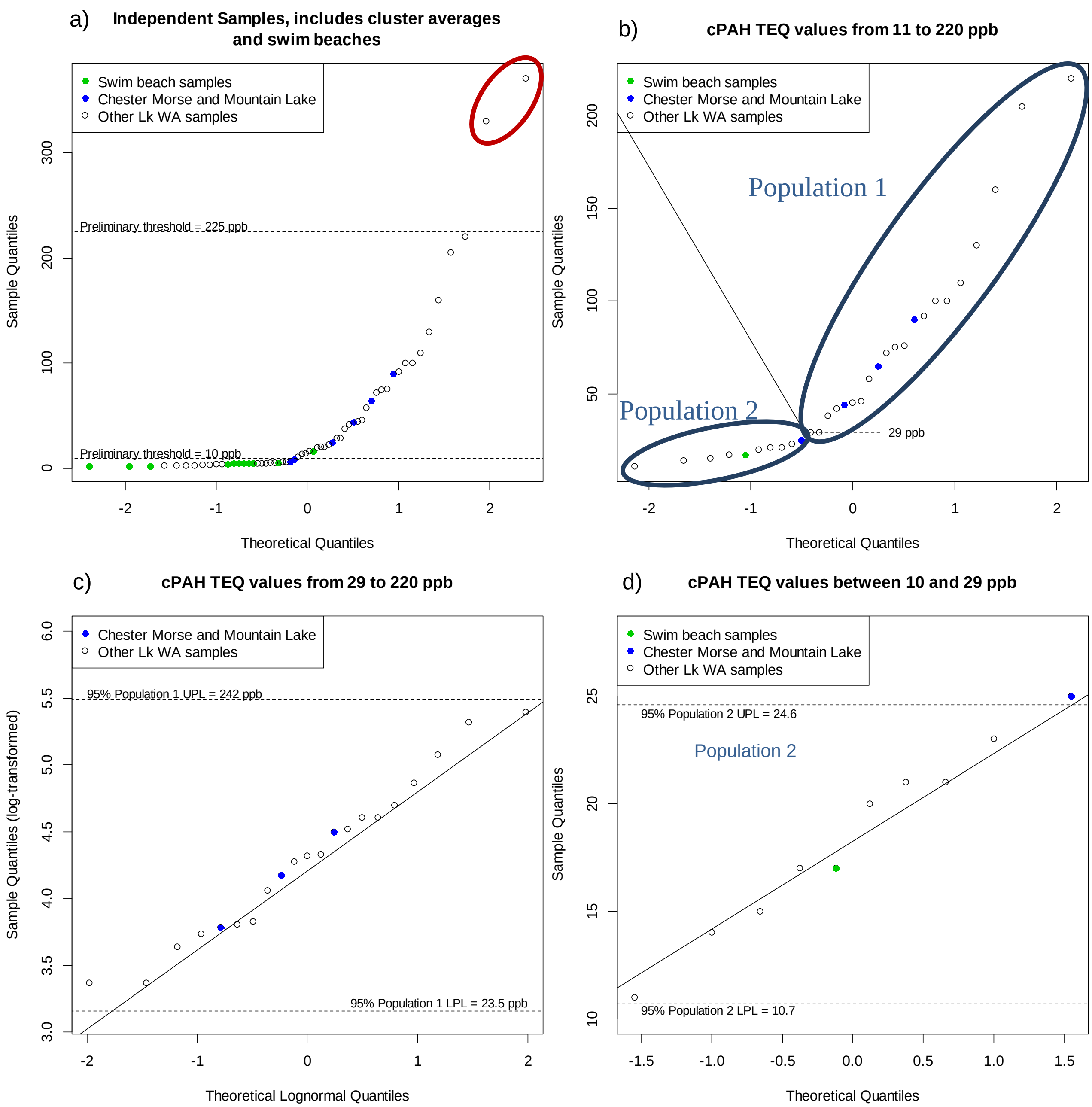

Figure 11: Sequential QQ plots for the cPAH data.

Includes cluster averages (n = 54). 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) Data with preliminary thresholds identified subjectively based on breaks/slope changes in the QQ plot.

b) Data between the two preliminary thresholds (10 ppb and 225 ppb), with two sub-populations identified.

c) Data within Population 1 prediction limits. Data were log-transformed to accommodate the skewness in these data.

d) Data within Population 2 prediction limits.

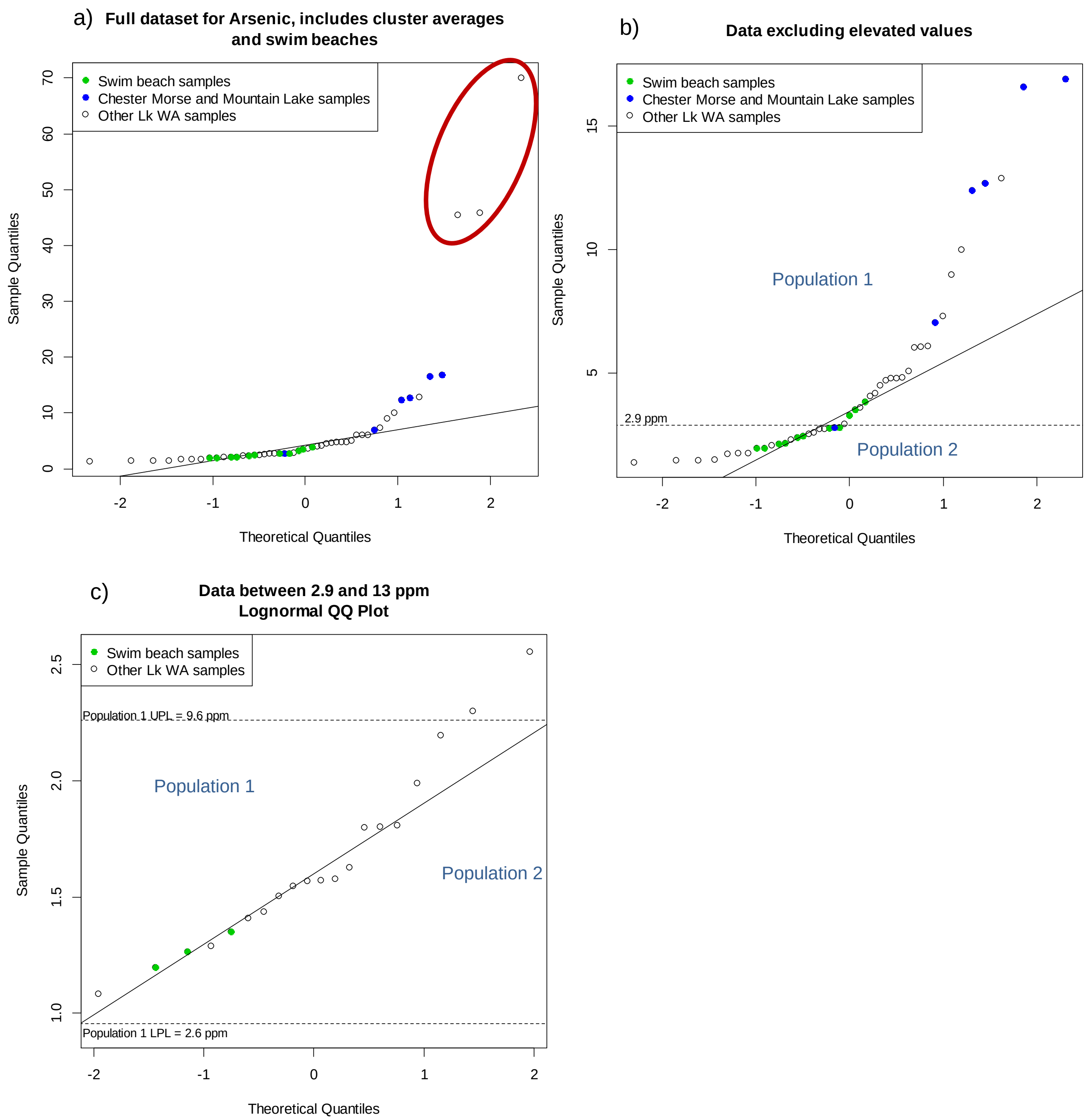

Figure 12: Sequential QQ plots for the arsenic data, including cluster averages (n = 42).

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.

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

b) All data excluding the elevated and influential values, showing a preliminary threshold of 2.9 ppm based on an apparent slope change in the QQ plot

c) Data within Population 1, and its associated robust prediction limits. Data were log-transformed to accommodate the skewness in these data; Chester Morse and Mountain Lake samples not shown on this panel).

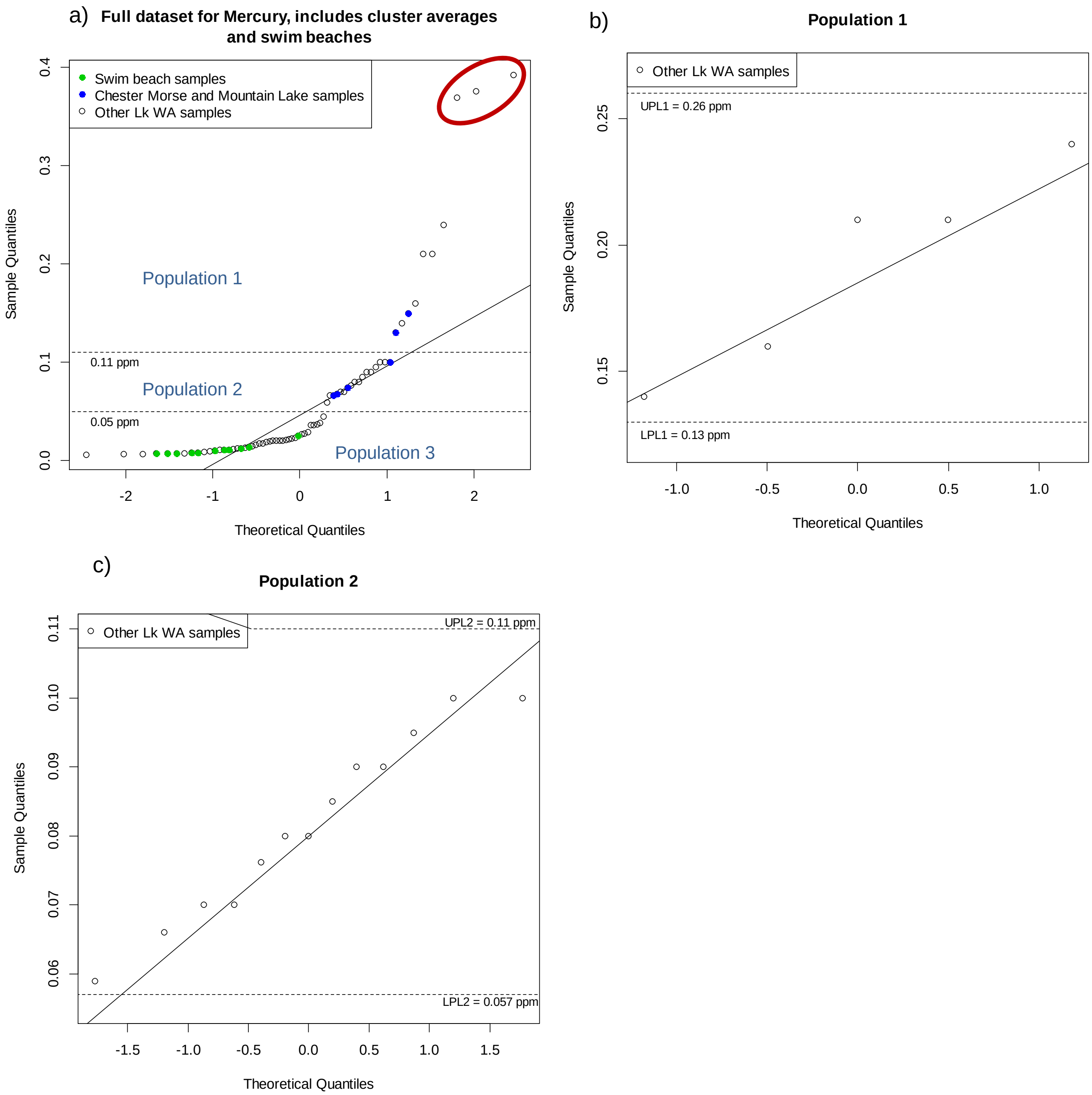

Figure 13: Sequential QQ plots for mercury, including cluster averages (n = 64).

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 preliminary thresholds based on breaks in the QQ Plot. Influential elevated values are circled in red. Preliminary subpopulations are identified.

b) Data from Population 1, with its associated robust prediction limits..

c) Data from Population 2, with its associated robust prediction limits.

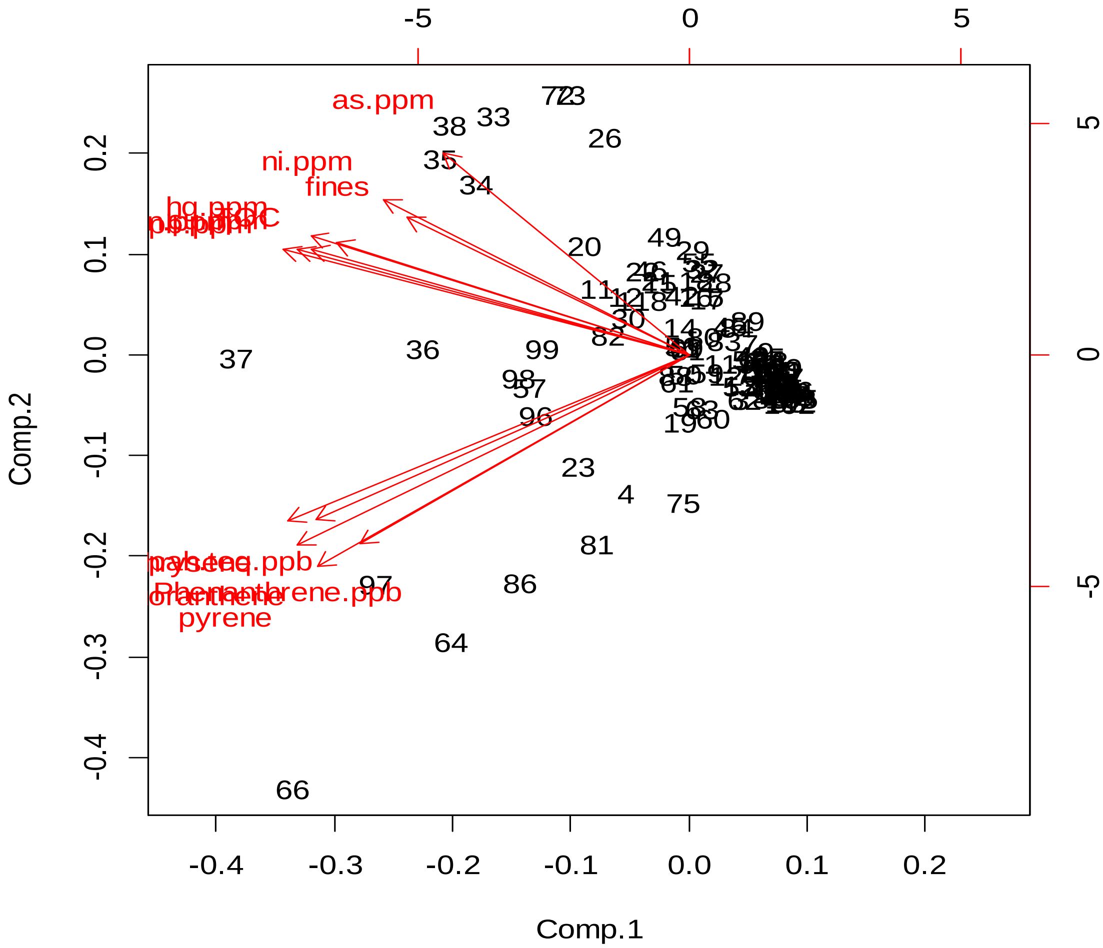

Figure 14: Principal Components Analysis. Bi-plot showing the direction that the original variables load onto the first two principal components. PCA was done on 113 samples with no missing values for 13 chemical or physical endpoints**.**

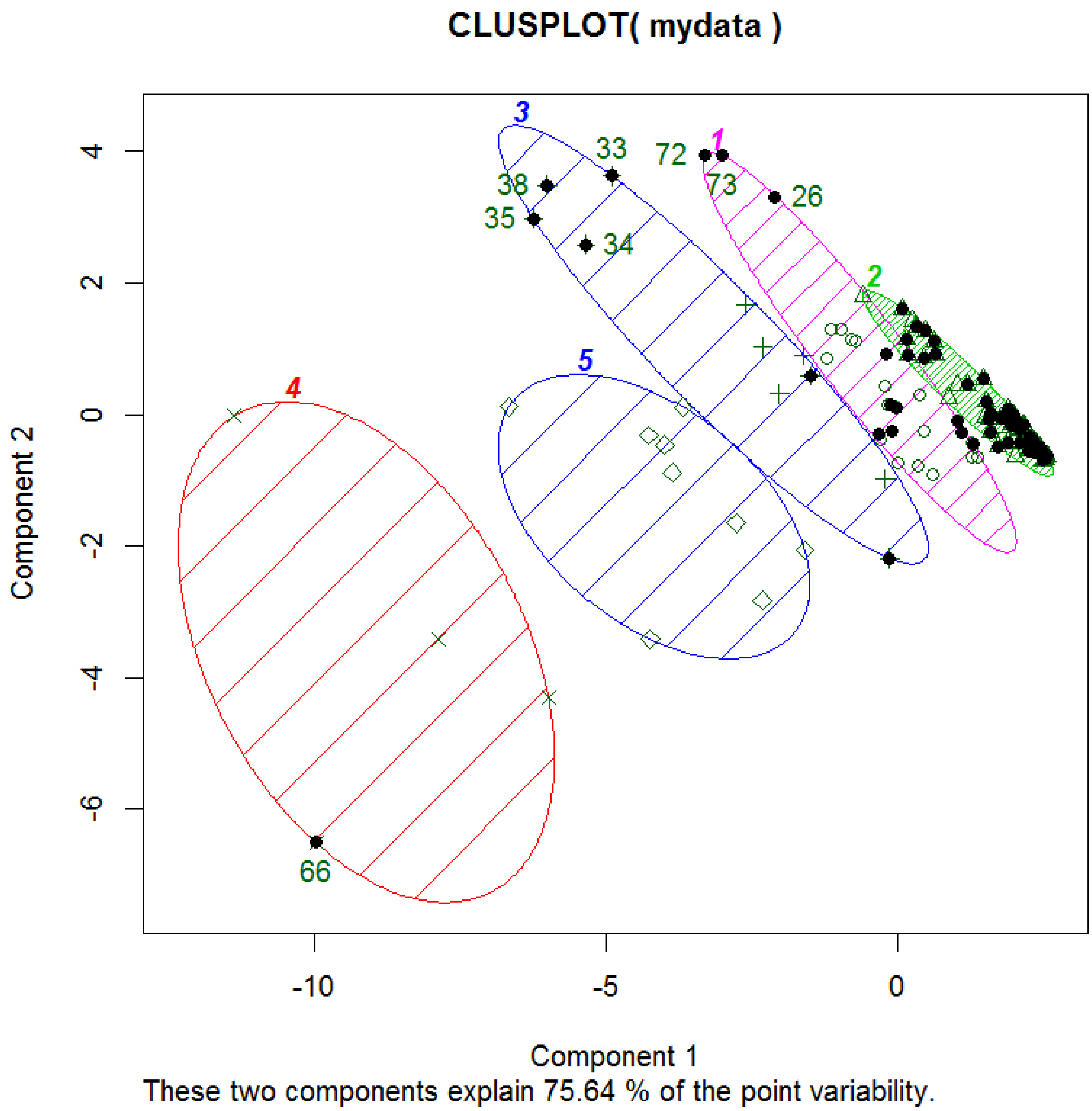

Figure 15: Plot of the 113 samples on the first two principal components (see Figure 14).

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. The numbered samples were extreme in one way or another (#66 had cPAH TEQ value of 1900 ppb; the samples near the top of group 3 (#33, #34, #35, and #38) are Bryant samples with high TOC; the 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.	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.

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	2^{nd}
Arsenic	39	1.15 to 12.9 ppm	No trend
Mercury	57	0.0061 to 0.24 ppm	2^{nd}

Table 7: Autocorrelation results for cPAH 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.

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.

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: 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 > 160 ppb**	**Pop 1 [32, 160]**	**Pop 2 [10, 26]**	**Pop 3 <10**
West of I-5	6.7, 1900	1900			6.7
Portage Bay (near UW)	38, 46		42		
Kenmore (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		38	15	<3.8
South of Newcastle Beach Park	170, 240	205			
Montlake Cut	17, 110		110	17	
May Creek	14, 92		92	14	
Harbour Village Marina	<4.8, 5, 8.8, 33, 58		46		<6.2
Chism Beach	<5.4, 370	370			<5.4
Boeing	<2.9, 34, 36, 43, 56, 120, 220	220	57.8		<2.9
Number of elevated values or independent observations		4 (elevated)	9	5	9

Table 11: QQ plot correlation coefficients for possible sub-populations of the cPAH TEQ values (Figure 11b).

*Not significantly different (α=0.05) from the specified statistical distribution**.**

Two values at 29 ppb included in the	**Data Subset**	**Normal**	**Log-Normal**
Upper Population	11 to 23 ppb	0.982	
Upper Population	29 to 220 ppb	0.972	0.989
Lower Population	11 to 29 ppb	0.977*	
Lower Population	38 to 220 ppb	0.942	0.985

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

*Sample removed from regional background calculation due to potential influence from a source.

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
<2.9	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	Houghton Beach Park
<4.9	Houghton Beach Park, nearshore
<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
5.1	Kenmore, mouth of Sammamish River (average)
5.2	South of Seward Park, nearshore
<5.4	Chism Beach
<5.5	Lyon Creek Waterfront Preserve
<5.6	Marsh Park
5.7	Enatai Beach park, swim beach
<6.2	Harbour Village Marina (average)
<6.7	Under I-5 near houseboats
Population 2 (11 ppb ≤ cPAH TEQ ≤ 25 ppb)	
11	Martha Washington Park
14	May Creek
15	Newcastle Beach Park
17	Clyde Beach Park, swim beach
17	Montlake Cut
20	Marsh Park
21	Seward Park, east side of peninsula, nearshore
21	Near the mouth of the Sammamish River
<23	Cozy Cove wetland area
Population 1 (24 ppb ≤ cPAH TEQ ≤ 240 ppb), representing regional background	
29	Lyon Creek Waterfront Preserve
29	Houghton Beach Park
38	South of Newcastle Beach Park

Population 1 (24 ppb ≤ cPAH TEQ ≤ 240 ppb), representing regional background	
42	Portage Bay, near UW (average)
45	Near Newport Yacht Club
46	Harbor Village Marina (average)
58	Boeing (average)
72	Middle of the lake west of Mercer Island
75	Middle of the lake between the southwest shoreline of Mercer Island and Rainier Beach
76	Kenmore Navigational Channel (average)
92	May Creek
100	South of Pleasure Point
100	Middle of the lake between Magnuson Park and Kirkland
110	Montlake Cut
130	Pleasure Point
160	McAleer Creek
205	South of Newcastle Beach Park (average)
220*	Boeing
Samples with elevated and influential cPAH TEQ concentrations 242 ppb ≤ cPAH TEQ)	
330	Kennydale Beach swim park, very high fines
370	Chism Beach
1900	West of I-5

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	
Number of Independent Observations		6	1

Table 14: Samples with arsenic concentrations, broken into sub-populations (Figure 12).

	Location	Concentration (ppm)	Percent Fines
	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
	Martha Washington Park	1.8	8
	Seward Park (n side of peninsula)	1.8	9
	Montlake Cut	1.8	NA
	Chism Beach park swimming beach	2	4
	Pritchard Island Beach Park swimming beach	2	4
	Enatai beach park; swimming beach	2.1	3
	May Creek, cluster average	2.1	10
	Meydenbauer Bay beach park, swimming beach	2.2	3
	Newcastle Beach Park, cluster average	2.3	13
	Waverly Park swimming beach	2.4	3
	Seward Park Beach swimming beach	2.5	4
	Newcastle Beach Park	2.6	5
	Marsh Park, cluster average	2.6	4
	Coulon Beach Park swimming beach	2.8	4
	Kirkland Marina park swimming beach	2.8	3
	South Houghton Beach Park	2.8	12
	Portage Bay (near UW), cluster average	2.8	NA
	Opposite Seward Park (w side of Andrews Bay)	3	21
	Newcastle Beach park swimming beach	3.3	19
	Seward Park (w side of peninsula)	3.6	15
	Clyde Beach Park swimming beach	3.6	3
	Houghton Beach Park swimming 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
	Lk Washington Blvd Park	4.8	13
	near Newport Yacht Club (south of I-90).	4.8	19
	Montlake Cut	4.9	NA
	by Boeing	5.1	3
	Fairweather Bay Residential inlet, cluster average	6.1	NA
	Cozy Cove wetland area	6.1	74
	North Houghton Beach Park	6.1	4
	Kennydale Beach Park	7.3	70
	Pleasure Point	9.0	20

Samples exceeding Population 1 UPL for Arsenic (9.6 ppm)			
	south of Newcastle Beach Park	10	41
	W. of I-5	13	NA
	Middle of the lake between Magnuson Park and Kirkland	46	80
	Middle of the lake 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	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	
Number of Independent Observations		6	2

Table 16: Samples with concentrations, broken into sub-populations (see Figure 13).

	Location	Mercury Concentration (ppm)	Percent Fines
Population 3 (mercury < 0.05 ppm)			
	43 samples	0.0061 – 0.045	3 – 21
Population 2 (0.05 ppm < mercury < 0.11 ppm)			
	Cozy Cove wetland area	0.059	74
	Portage Bay (near UW)	0.066	NA
	Kenmore - navigational channel	0.07	49
	South of Newcastle Beach Park	0.07	21
	Kennydale Beach Park	0.0762	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
	Kenmore - navigational channel	0.095	45
	Kenmore - navigational channel	0.1	55
	South of Newcastle Beach Park	0.1	41
Population 1 (0.11 ppm < mercury < 0.26 ppm)			
	North Lake Marina, Kenmore	0.14	56.25
	Middle of the lake – SW of Mercer Island	0.16	74.5
	Near Boeing	0.21	41.9
	South of Newcastle Beach Park	0.21	56.3
	Kenmore – inner navigational channel	0.24	30.6
Samples with elevated and influential mercury concentrations (> 0.3 ppm)			
	Middle of the lake - west of Mercer Island	0.37	77
	Middle of the lake - between Magnuson Park and Kirkland	0.38	80
	West of I-5	0.39	NA