

WASHINGTON STATE
DEPARTMENT OF
E C O L O G Y

The Use of Sediment Cores to Track Persistent Pollutants in Washington State

A Review

January 2001

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The Use of Sediment Cores to Track Persistent Pollutants in Washington State

A Review

*by
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Environmental Assessment Program
Olympia, Washington 98504-7710

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Abstract

Measurement of persistent chemical concentrations in sediment cores can provide a valuable historical record of contamination. This method of evaluating trends in pollution may be particularly useful in association with the Washington State Department of Ecology's Persistent, Bioaccumulative, Toxic (PBT) Chemical Initiative.

This publication reviews 11 studies that, with varying success, evaluated sediment cores collected from lakes and marine waters in Washington State. It notes the purpose, responsible agency and funding source (if included) for each study. It summarizes the number and location of samples, as well as sampling and analytical methods. Results, including reported sedimentation rates, are recapped. Selected results from the three most successful coring studies are graphed and discussed in some detail.

Information from these studies is used to summarize issues that should be considered by those planning to use cores from Washington State sediments to evaluate historical contamination trends.

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Introduction

Historical information about environmental contamination is a useful, even necessary, component of efforts to reduce this contamination. This history can be used to establish “background” or “baseline” conditions. It can also be used to evaluate how contaminant levels have responded to changes in population, land use, and human activities – including actions (e.g., laws, chemical bans) designed to reduce that contamination. This knowledge can be valuable in assessing the efficacy of past pollution-control efforts and in designing and implementing more effective programs in the future.

Data to determine trends is commonly gathered by long-term programs that monitor environmental media: air, water, fish, and surface sediments. These monitoring programs can continue for years or even decades. Although valuable, this long-term monitoring can suffer from several shortcomings:

- These programs cannot evaluate conditions prior to the time they were initiated. Thus, even the oldest continuous programs generally provide information about only the past few decades.
- Changes (often improvements) in sampling and analytical methods can void or compromise comparisons with earlier data.

In some cases, analyses of sediment cores can be used to overcome these shortcomings. Concentrations of persistent pollutants (especially metals, radionuclides, and refractory organic compounds) can be measured in successive layers within cores. Dating techniques, including use of radioactive isotopes like lead²¹⁰ and cesium¹³⁷, can be used to estimate the age of these layers. By combining the age and contaminant concentration data, extended records of pollutant concentrations in sediments can be reconstructed.

The length of these records is limited only by the age of the sediment deposits and the depth to which undisturbed cores can be collected. Cores collected in Washington State have provided records that extend well back into the 19th century.

Problems with changes in sampling and analytical techniques are largely eliminated because subsamples (layers) are taken from a single core and can be analyzed at one time using a single laboratory with identical techniques.

Core Dating

Sedimentation rates and the age of specific layers of a core can be estimated by several techniques. A brief summary of these techniques follows:

1. **The Use of Markers:** Various *markers* can be used to identify the age of specific strata. The top layer of the core is assigned to the year of collection; the bottom layer of a core from a reservoir may be assigned the year the reservoir was first filled. Measurements of cesium¹³⁷ concentrations in layers of a core are often used to establish markers. For instance:
 - ◇ 1952 – when cesium¹³⁷ was first produced in large quantities by the Ivy test at Eniwetok Atoll (Schell, 1986).
 - ◇ 1958 – when there was an early peak in cesium¹³⁷ fallout (Charles and Hites, 1987).
 - ◇ 1963 – when subsequent nuclear tests resulted in the maximum cesium¹³⁷ fallout (Schell, 1986; Charles and Hites, 1987).

Another marker that is likely to be useful throughout much of Washington State in the Mt. St. Helens ash layer (1980).

The ages of strata are usually estimated by assuming a constant rate of sediment deposition between two markers.

2. **Decay of Radioactive Materials:** Aerosols deposited on the surface of the earth (or water) contain certain radioisotopes at predictable concentrations. The subsequent decrease in the concentration of these radioisotopes in older strata can be used to date these strata. Lead²¹⁰ has characteristics that make it useful for dating strata that are less than 100-150 years old. Lead²¹⁰ is created by the radioactive decay of atmospheric radium²²⁶ and radon²²². After it is deposited in sediments, because it is no longer in equilibrium with its source gases, its concentration decreases with age (half-life of 22 years). The rate of decrease in lead²¹⁰ concentrations can be predicted and used to estimate the age of the strata in which it is measured (Schell and Nevissi, 1980). This method requires establishing “background activity,” which is variously interpreted as loss of 95-99% of the original activity (approximately 100-150 years).

Sedimentation rates, often expressed in cm/yr, can be estimated by dividing the core length by the number of years it took to lay down that depth of sediment. Compression of cores, either by natural accumulation of overburden or compression generated by the coring device, can complicate this determination, as can activities that disturb sediments such as dredging, filling, and bioturbation.

Purpose of this Review

The Washington State Department of Ecology (Ecology) is developing a strategy to address persistent, bioaccumulative, toxic chemicals (PBTs) (Gallagher, 2000). At present, the following PBTs are listed in the strategy: dioxins/furans, DDT and metabolites, PCBs, benzo(a)pyrene, mercury, alkyl-lead, hexachlorobenzene, aldrin/dieldrin, chlordane, and toxaphene.

As one measure of progress, this PBT strategy proposes the “continued reduction in the concentrations of PBTs recorded in sediments deposited in key water bodies throughout the state.” Also, the strategy states, “Ecology proposes to establish a baseline monitoring program that will be used in combination with existing monitoring and reporting programs to measure progress in reducing PBT chemicals.”

Simultaneously, Ecology’s Environmental Assessment Program is reevaluating efforts to monitor toxics in Washington State’s environment.

This review of existing information was undertaken because the use of sediment cores offers substantial promise for both of these efforts.

This document reviews selected publications that deal with sediment coring in Washington State. It presents figures that show selected results from three of the most successful studies. Finally, based on these reviews, it summarizes key considerations in designing and implementing future monitoring efforts.

Review of 11 Studies

Eleven studies were located that report concentrations of persistent chemicals in dated sediment cores collected from Washington State.

These studies are presented in chronological order by date of publication. In some cases, authors or others were contacted to clarify certain points or provide additional information. These contacts are cited as “personal communication.”

Each review notes the purpose, responsible agency, and funding source (if included) for the study. It summarizes the number and location of samples, as well as sampling and analytical methods. Results, including reported sedimentation rates, are recapped. Additional information is noted if it seemed relevant to the design of future coring studies.

Selected results from the three most successful coring studies (8, 9, and 11) are graphed and discussed in some detail. Compounds chosen for graphing and discussion were: dioxins/furans, total PCBs, total DDT, benzo(a)pyrene, mercury, cadmium, lead, and arsenic. Metals concentrations are reported as milligrams per kilogram dry weight (mg/Kg). Concentrations of organic contaminants are expressed as micrograms per kilogram dry weight (ug/Kg), except in the case of dioxins and furans, which are expressed as nanograms per kilogram dry weight (ng/Kg). These concentrations are equivalent to parts per million (ppm), parts per billion (ppb) and parts per trillion (pptr), respectively.

Although not the primary focus of this review, these discussions are augmented by information not contained in the original publications. This includes the use of sediment standards and guidelines to provide context (e.g., are the concentrations in these sediments high enough to cause adverse effects?). Standards and guidelines are shown only in those figures in which concentrations found in the cores approach or exceed these values. For this purpose Puget Sound sediment standards and “consensus-based sediment quality guidelines for freshwater ecosystems” proposed by MacDonald et al., 2000 are used.

Graphs and trend discussions also include some limited information on the timing of events that might help explain the trends revealed by the cores. In many cases this information is limited, and a more extensive review of the history of events linked to specific contaminants would, no doubt, reveal additional insights.

1. 1974, Crecelius, E.A. *The Geochemistry of Arsenic and Antimony in Puget Sound and Lake Washington, Washington*. Ph.D. dissertation. University of Washington. pp. 64-77.

The purpose of this study appears to have been to evaluate the disposition and cycling of arsenic in Lake Washington, especially arsenic from the ASARCO smelter in Tacoma. Several surface samples and five cores were collected. The cores were collected, apparently in 1972, and were sited along the north-south axis of Lake Washington, from south of Seward Park to north of Sand Point. A single core, collected near the Evergreen Point Floating Bridge, was both dated using lead²¹⁰ and analyzed for arsenic. Other cores were either dated or analyzed for arsenic, but not both.

Arsenic concentrations in surface sediments ranged from 15 to 210 parts per million (ppm = milligrams per kilogram, mg/Kg). Based on graphs contained in the report, background (historical) concentrations in the cores were less than 10 mg/Kg, while recent (mid-1970s) concentrations ranged up to 200 mg/Kg. Graphs of five cores generally show rising arsenic concentrations, although in several cases these concentrations may be leveling off. The thesis states that arsenic concentrations began to rise “about the time when the Tacoma smelter began operations in 1890.”

A sedimentation rate of about 0.3 cm/yr is reported from the dated cores. This rate is compared to those found in other studies and found to be in “good agreement.”

Single cores were also collected in Lake Sammamish, Lake Samish, and Lake Quinault. These were apparently analyzed only for arsenic. Lake Sammamish and Lake Samish showed some increase in arsenic levels in the top 2-20 cm and reached concentrations of 50-60 mg/Kg in surface sediments. Arsenic concentrations in Lake Quinault remain low (<15 mg/Kg).

2. 1975, Crecelius, E.A. M.H. Bothner, and R. Carpenter. *Geochemistries of arsenic, antimony, mercury, and related elements in sediments of Puget Sound.* Environmental Science and Technology. Vol. 9(4):325-333.

This work was conducted by the Department of Oceanography, University of Washington.

The purpose of this study was to determine distributions of arsenic, antimony, mercury, chromium, and cobalt in surface sediments and arsenic in sediment cores from Puget Sound, “and to relate these distributions to sources, transport paths, and geochemical reactions these metals may undergo.” Discussion focuses on arsenic and mercury.

Samples were collected between 1970 and 1972. Results of arsenic analyses in four cores are shown graphically and results from other cores are referred to, but there are no tables or maps enumerating or locating coring sites. Three sampling devices were used. Most samples were collected with a “a special coring apparatus” with “hydrostatically slowed rate of penetration...A few samples were collected with a conventional gravity corer or with a Van Veen grab.”

Metals analyses were by neutron activation and flameless atomic absorption (mercury only).

Sediment accumulation rates were determined using lead²¹⁰. Calculated sedimentation rates ranged from 0.22 cm/yr (Quartermaster Harbor) to 2.0 cm/yr (East Passage).

Arsenic results are graphed for four cores: Quartermaster Harbor, East Passage, Fox Island, and North Seattle. Only the Quartermaster Harbor sample penetrated to background concentrations which appear to have started rising in about 1890. Concentrations prior to this appear to be less than 10 mg/Kg. Concentrations in all cores, except the one collected near North Seattle, appear to be rising with current (as of 1972) concentrations of 35-50 mg/Kg.

The Tacoma smelter (ASARCO) is prominently mentioned as a likely source of the elevated arsenic found in the sediments. The smelter and a chlor-alkali plant in Bellingham are mentioned as likely mercury sources.

Mercury results from cores are mentioned but not specifically reported.

3. 1977, Schell, W.R. and A. Nevissi. *Heavy metals from waste disposal in central Puget Sound.* Environmental Science and Technology. Vol. 11(9):887-893.

This work was supported by a contract from the Municipality of Metropolitan Seattle (METRO) and conducted by the Laboratory of Radiation Ecology and the College of Fisheries at the University of Washington.

The purpose of study appears to have been to evaluate heavy metals concentrations in water, sediment, and biota in Puget Sound as they might relate to the West Point municipal wastewater discharge.

Three cores were taken with a 5-cm diameter gravity corer and a 3.5-cm damped piston corer. Samples were digested with nitric and perchloric acid and analyzed by atomic absorption spectrophotometry for copper, cobalt, chromium, mercury, manganese, nickel, lead, and zinc. These cores were dated using lead²¹⁰, which was analyzed by neutron activation.

The results of dating 14 additional cores are “reported elsewhere” but used here to interpret the results. Sedimentation rates in Puget Sound of 0.1-2 cm/yr are reported.

The three cores were located as follows: one near Alki Point, one near the West Point outfall, and one north of Blake Island. The lead²¹⁰ results are equivocal for the first two of these cores – indicating high rates of recent deposition – due, perhaps, to the dumping of dredge spoils and/or high rates of deposition near the West Point outfall.

Results for most metals are erratic. This may, in part, reflect the apparent disturbance of at least two of the cores. Little can be concluded from the metals analyzed in these cores.

4. 1985, Carpenter, R., M.L. Peterson, and J.T. Bennett. *²¹⁰Pb-derived sediment accumulation and mixing rates for the Greater Puget Sound Region.* Marine Geology Vol. 64:291-312.

This work was supported by a U.S. Department of Energy grant and performed by the University of Washington School of Oceanography.

The implied purposes of the study were to report recent accumulation and mixing rates for Puget Sound sediments. These were calculated from excess lead²¹⁰ activity profiles from 44 sediment cores collected at 27 sites in Puget Sound.

Sample sites were located from near the Nisqually delta in the south, to near Cherry Point to the north. A map showing the locations is included.

Cores were collected between 1976 and 1983 with a box corer (23 x 30 cm cross-sectional area) and a multiple corer with damped penetration that simultaneously collected four, 5.6 cm diameter cores. Lead²¹⁰, total carbon, aluminum, and manganese were analyzed. Although cores from the multiple corer were shortened by 36% in one case, the authors conclude “that shortening or other coring artifacts have not detectably influenced our results.”

Calculated sediment accumulation rates ranged from 46 to 1200 mg/cm²/yr and 0.12 to 2.4 cm/yr. Sedimentation rates are highest in the central basin between Seattle and Tacoma. These sites also have the lowest sediment mixing layer residence time (1.5-8.7 years). These data are tabulated by station.

The paper points out that the relatively deep and active mixing layers in Puget Sound sediments will blur changes in sediment concentrations in cores and will delay the response of sediments to changes in pollutant input.

5. 1987, Bloom, N.S. and E.A. Crecelius. *Distribution of silver, mercury, lead, copper and cadmium in central Puget Sound sediments.* Marine Chemistry Vol. 21: 377-390.

This work was supported by contracts from the U.S. Department of Energy, Seattle METRO, and the National Oceanographic and Atmospheric Association (NOAA), and was conducted by the Marine Science Institute at the University of Connecticut and the Battelle Marine Research Laboratory in Sequim, Washington.

Data were used to show:

- Contamination of fine sediments in the central (deeper) portion of the Sound with mercury, lead, copper, and silver.
- That these sediments are quite consistent in quality and represent a major sink for metals enrichment in the Sound.
- Historical patterns of contamination for the five metals studied.

Twenty-one cores were collected in Puget Sound in 1982, all but one using a 3-m long Kasten corer with a large (225 cm²) cross section. Sediments were digested using a hot mixture of nitric and sulfuric acids. Silver, copper, cadmium, mercury, and lead were analyzed using Zeeman-corrected graphite-furnace atomic adsorption spectroscopy. Cores were dated using lead²¹⁰. Cesium¹³⁷ analysis was also conducted on some of the cores and found “chronologically consistent with those of lead²¹⁰.” The quality assurance and quality control techniques for all aspects of sample collection and analysis appear to have been rigorous.

Sedimentation rates of as high as 2.5 cm/yr were calculated. Complete data for four cores are presented: off Edmonds, off Meadow Point, in the outer portion of Elliott Bay, and between Maury Island and Brown’s Point.

“Background” concentrations for the five metals at these four stations are summarized here as ranges (units = mg/Kg dw): silver (0.015-0.07), copper (19-31), cadmium (0.19-0.47), mercury (0.028-0.063), and lead (4.0-7.2).

Historical concentrations are shown for “mean core profiles” dated from 1848 to 1979. Only cadmium shows no apparent anthropogenic enrichment. “Subsurface maxima” are shown for lead, mercury, and “possibly” silver, implying that concentrations of these metals had begun to fall prior to 1979.

6. 1990, Johnson, A., D. Norton, B. Yake, and S. Twiss. *Transboundary metal pollution of the Columbia River (Franklin D. Roosevelt Lake)*. Bulletin of Environmental Contamination and Toxicology. Vol. 45:703-710.

This work was conducted by the Washington State Department of Ecology. The publication reports results of analyses of Lake Roosevelt bottom sediments for metals.

Several surface sediment samples and one sediment core were collected in 1986. The core was collected using a 2-inch gravity corer off Frenchman Point Rocks near Kettle Falls.

Subsamples were analyzed for arsenic, cadmium, copper, mercury, lead, zinc, grain size, and organic carbon. Cesium¹³⁷ was measured “for purposes of dating the core.” Sediments were digested using nitric acid and hydrogen peroxide, and metals analyzed by atomic adsorption.

As noted in the report, “The high metals concentrations in Lake Roosevelt sediments are thought to be primarily due to discharges from the Cominco Limited lead-zinc smelter and refinery at Trail, BC...which has been in existence since the turn of the century.”

Cesium¹³⁷ dating implies a sedimentation rate of less than 1 cm/yr between a 1954 marker (apparently based on information in Schell and Nevissi, 1980) and sample collection in 1986. The core extended 30 cm below the 1954 cesium¹³⁷ marker, but did not reach the bottom of the deposited sediments. Because Grand Coulee dam was completed in 1941, the bottom 30-cm of the core must have been laid down in less than 13 years (Johnson, personal communication). This means that the average sedimentation rate between 1941 and the mid-1950s exceeded 2.3 cm/yr.

This high rate of initial sedimentation may have been caused by bank sloughing in the early years of the reservoir. Additionally, Hugh Keenleyside Dam, located upstream of Castlegar B.C. was completed in 1968; and the Boundary Dam on the Pend Oreille River was completed in 1967. These and other dams on the upper Columbia system probably decreased sediment loads to Lake Roosevelt from rates that were initially higher (Johnson, personal communication).

Substantial changes in the sources of sediment and rate at which this sediment was deposited make it difficult to interpret the historical record. Metals concentrations were low in sediments from the bottom 20 cm of the core. This may imply high deposition rates of clean sediments from bank sloughing with the resulting dilution of metals discharges from Cominco. Metals concentrations are relatively high in the top 25 cm. A reduced sedimentation rate in the later record may accentuate the effect of metals discharged between 1954 to 1986. Although lead and arsenic had subsurface maxima, other metals (zinc, copper, cadmium, and mercury) did not. No general downturn in recent (1986) metals concentrations was apparent.

7. 1995, Norton, D. and T. Michelsen. Elliott Bay Waterfront Recontamination Study. Volume 1: Field Investigation Report. Washington State Department of Ecology, Olympia, WA. Publication No. 95-335.

This work was conducted by the Washington State Department of Ecology to identify sources of recontamination along the Seattle waterfront, mechanisms of contaminant transport and resuspension, and rates of sedimentation.

Three cores were collected in 1994 along the Seattle waterfront between Piers 48 and 57.

These cores were collected using a barrel gravity corer. Lead²¹⁰ and cesium¹³⁷ were used to date the cores. Metals (aluminum, copper, iron, mercury, lead, manganese, and zinc) and PCBs (Aroclors) were analyzed. Metals analyses were done by graphite furnace and cold vapor atomic absorption and ICP; PCBs were done by GC/ECD. Settling particulate matter was also collected using sediment traps.

Estimated sedimentation rates varied from 0.1 to 0.72 cm/yr. Difficulties in interpreting the age of the core segments and a likely history of dredging in the study area make these data problematic. One core (#3, located north of Pier 48) seems, at depth, to approach baseline concentrations for lead (5-7.5 ppm), mercury (0.035-0.055 ppm) and PCBs (<50 ppb).

8. Cleverly, D., M. Monetti, L. Phillips, P. Cramer, M. Heit, S. McCarthy, K. O'Rourke, J. Stanley, and D. Winters, 1996. *A time-trends study of the occurrences and levels of CDDs, CDFs, and dioxin-like PCBs in sediment cores from 11 geographically distributed lakes in the United States.* Organohalogen Compounds Vol. 28:77-82.

This work was conducted by the U.S. Environmental Protection Agency (EPA) and the U.S. Department of Energy (USDOE) to address the following questions:

- When did dioxins, furans, and PCBs initially appear in the general environment and are they related to anthropogenic activities?
- What has been the chronology of environmental burden in terms of environmental concentrations and fluxes to the sink?
- Is there evidence of any trends in environmental burden with time?

Eleven lakes/reservoirs from the USDOE archive of cores were evaluated. Two of these lakes (Ozette and Beaver) are located on Washington State's Olympic Peninsula and are shown in Figure 1. Lakes were chosen for:

- remoteness from specific anthropogenic source activity
- geographical coverage of the continental U.S.
- existence of adequate dating records

Sediment cores were originally collected using a 21-cm diameter "sphincter" launched from a boat and drilled into the lake sediment. Cores up to 1-m length were extruded and sectioned in 2-10 cm lengths. Cores were tested for target PCDDs, PCDFs, and cp-PCBs by stable isotope dilution and GC/MS (EPA Method 1613B). "Radiometric techniques, together with counting laminations, were used to date the sediment cores."

Specific data for Lake Ozette and Beaver Lake were provided in spreadsheet format by David Cleverly (personal communication.)

Sedimentation rates for the past century are about 0.4 cm/yr for Beaver Lake and about 0.18 cm/yr for Lake Ozette. These data appear to show clear historical trends for dioxins and PCBs in Lake Ozette and Beaver Lake sediments.

Figure 2 shows dioxin results displayed as toxic equivalents (TEQs) in ng/Kg (parts per trillion, ppt). Concentrations prior to 1925 are low and constant at less than 0.2 ng/Kg. From approximately 1930-1955, concentrations rise to about 1.7 ppt (Beaver Lake) and 2.5 ppt (Lake Ozette). By the mid-1970s concentrations had fallen to about 1 ppt at Beaver Lake and 1.4 ppt at Lake Ozette.

Cleverly et al. make no inferences in this paper about reasons for these trends, but they do note similar patterns from cores collected in other parts of the country (except Alaska). It is probable that these trends reflect the production and dissemination of dioxins and furans upwind and upstream of the lakes tested.

Figure 3 shows total PCB results (sum of congeners tested, recovery corrected, non-detected congeners assumed to be present at ½ the detection limit) in ug/kg (or ppb).

Prior to 1935 concentrations were very low at less than 2 ppb. This is consistent with the 1929 beginning of production of PCBs in the U.S. From approximately 1935-1965 concentrations rose, peaking at about 60 ug/Kg (Beaver Lake) and 175 ug/Kg in Lake Ozette. By the mid-1970s concentrations had fallen to about 40 ug/Kg at Beaver Lake and 100 ug/Kg at Lake Ozette. EPA restricted production and use of PCBs in 1977. Studies discussed later in this review (Lefkovitz et al., 1997; USGS, 2000) show similar patterns. Because PCB concentrations appear to have been falling prior to EPA's action, it is probable that the production and decimation of PCBs to the environment were already decreasing by the mid-60s.

Included in Figure 2 is a "consensus-based threshold effect concentration" (TEC) reported by MacDonald et al., 2000. This is a level "below which adverse effects [to sediment dwelling organisms] are not expected to occur."

The first appearance of PCBs in these cores coincides well with what is known about production of PCBs in the U.S. The Puget Sound sediment results (see following entry) seem to show PCBs appearing in the sediment record before they were produced in the U.S. This is one of several similar examples that point to "smearing" in marine sediments, which implies that mixing of surface sediments, probably by bioturbation, is much more prevalent in Puget Sound sediments than in lake sediments.

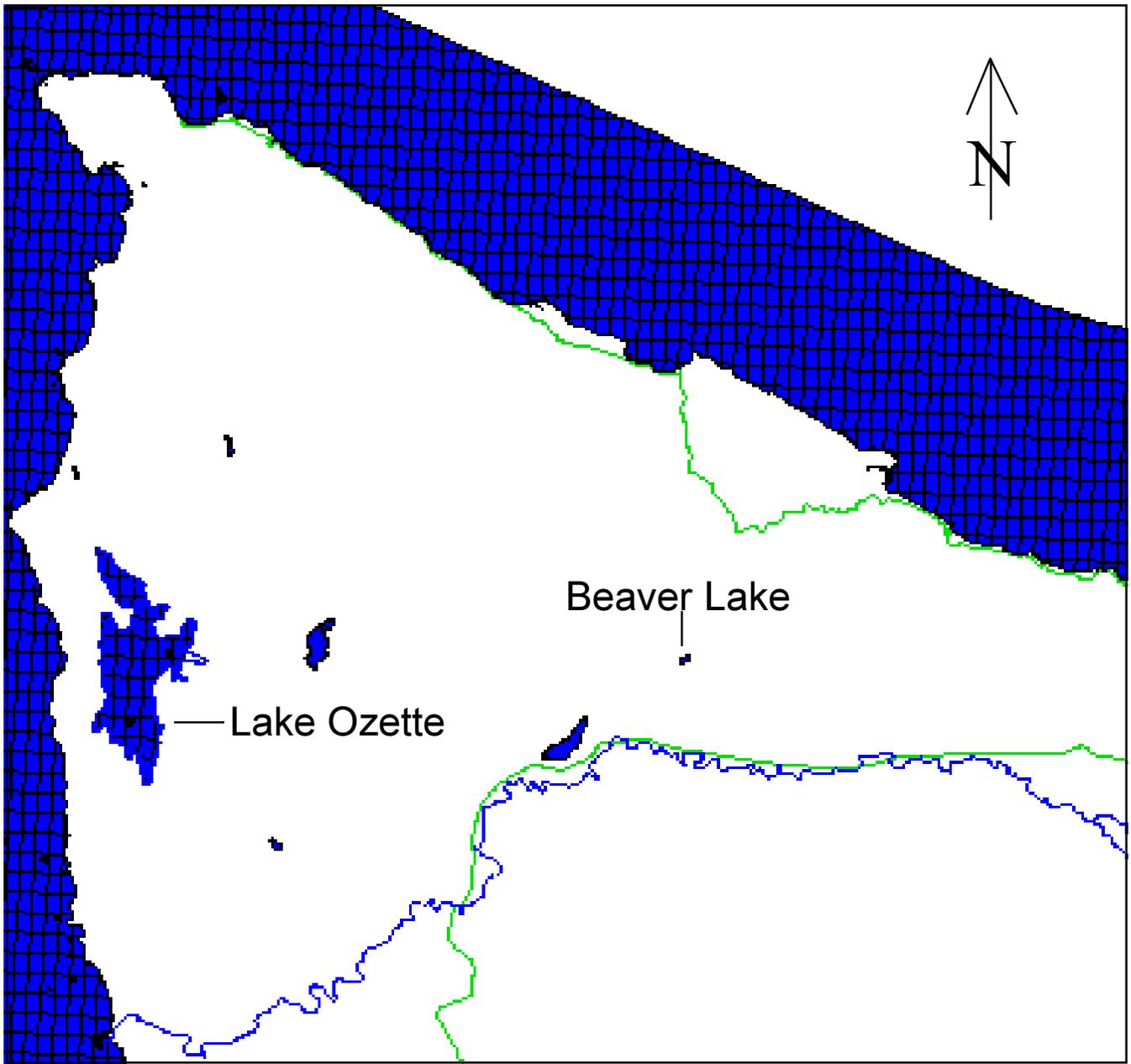
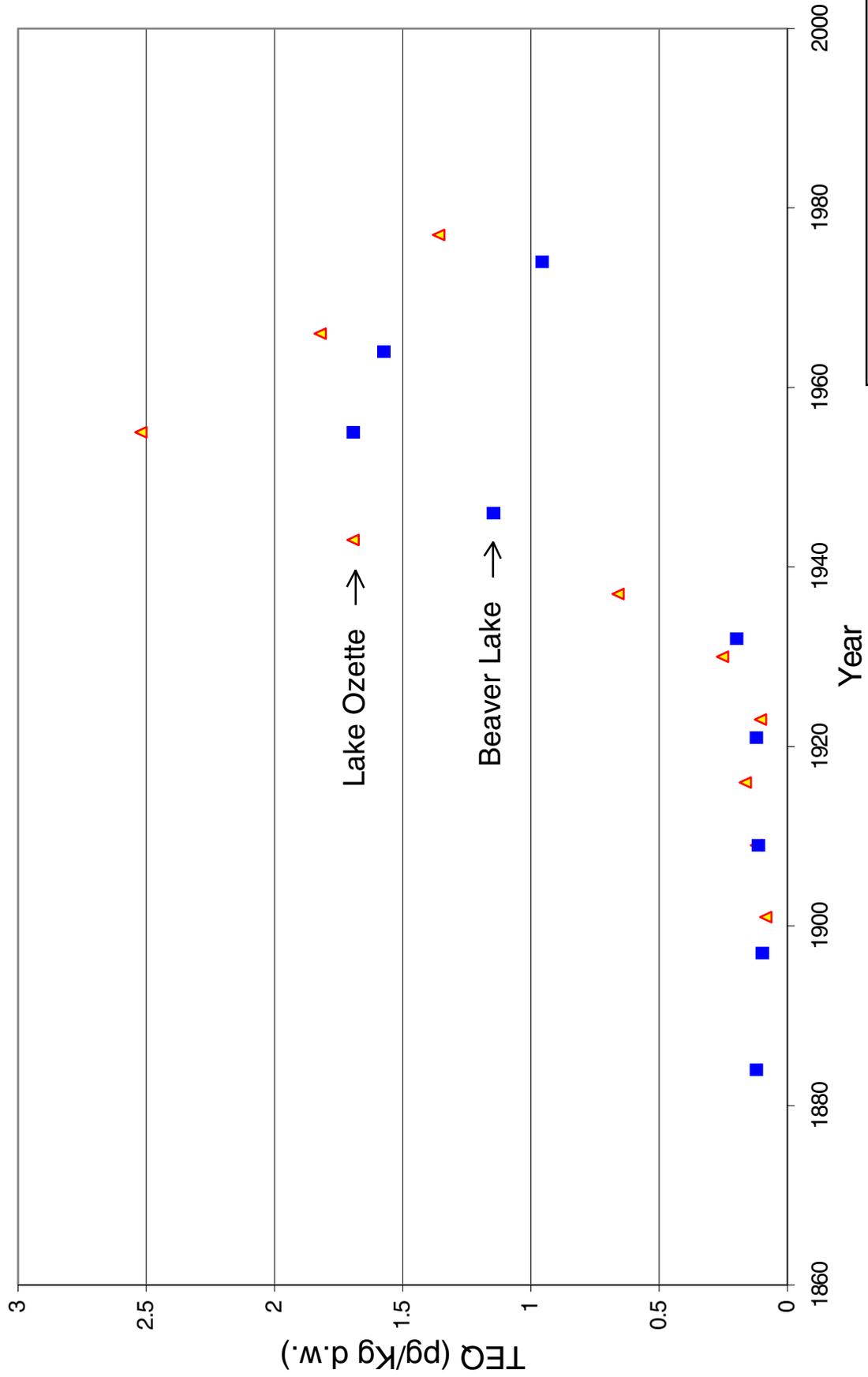


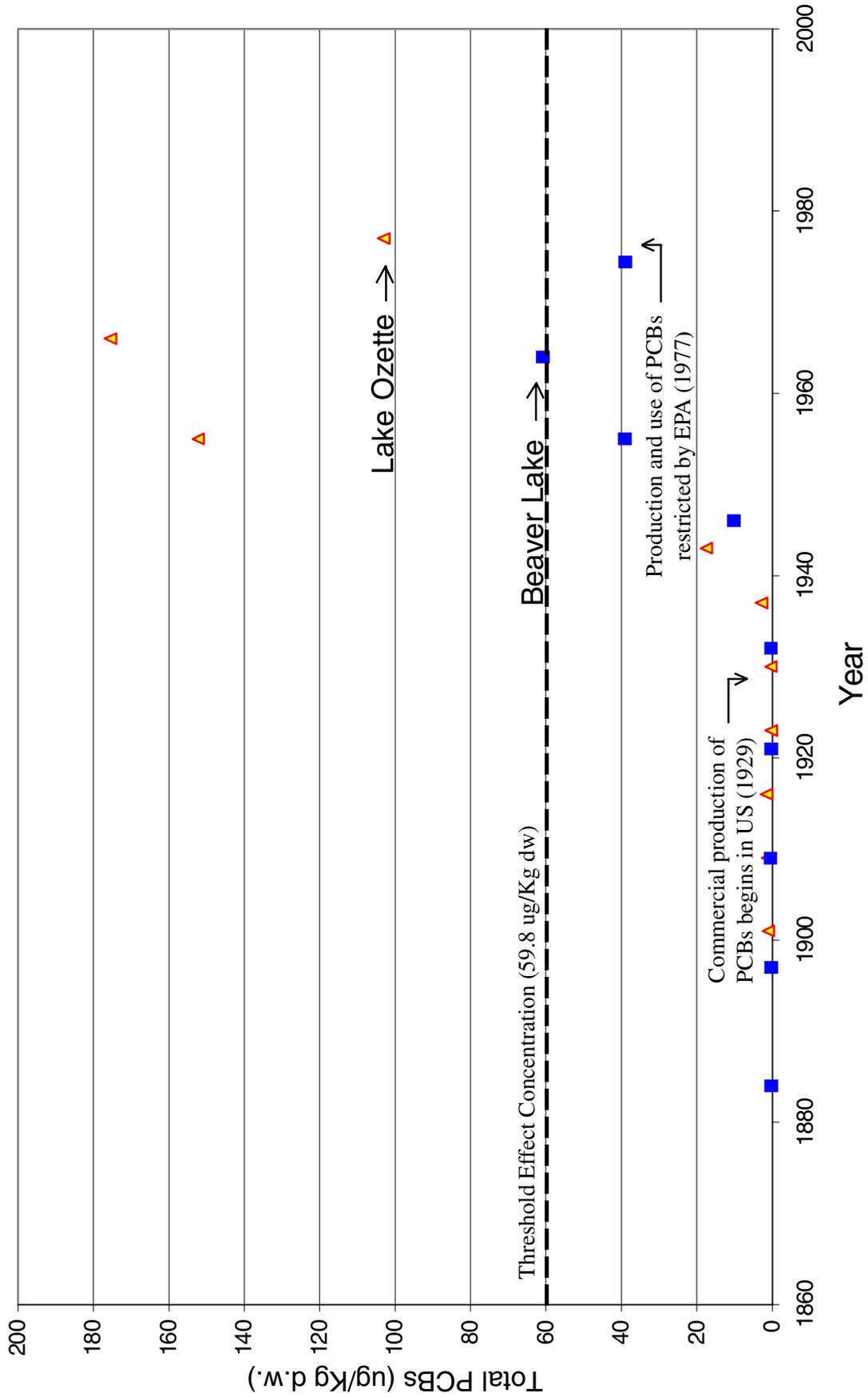
Figure 1. Olympic Peninsula Lakes

Figure 2.
History of Dioxin Contamination in Sediments of
Two Olympic Peninsula Lakes



Graphic by Bill Yake
 Sediment Core Data - Cleverly, et al., 1996.

Figure 3.
History of PCB Contamination in Sediments of
Two Olympic Peninsula Lakes



Graphic by Bill Yake
Sediment Core Data - Cleverly, et al., 1996.

9. 1997, Lefkovitz, L.F., V.I. Cullinan, and E.A. Crecelius. Historical Trends in the Accumulation of Chemicals in Puget Sound. Nat. Ocean. Atmos. Admin., Silver Springs, MD. NOAA Technical Memorandum NOS ORCA 111. 60 pp. + appendices.

This National Oceanic and Atmospheric Administration (NOAA) study was conducted by Battelle/Marine Sciences Laboratory in Sequim as part of NOAA's Status and Trends Program.

The study purposes were to:

- Continue monitoring historical trends in the concentration of contaminants in Puget Sound sediment.
- Quantify recent trends in the recovery of contaminated sediments.

Sediment cores were collected from six locations in the main basin of Puget Sound in 1991. Results of three of those cores provided long-term records – in one case reaching back to the early 1800s. The cores with long-term records are located as follows:

- off Elliott Bay
- off Seahurst Park
- north of Dash Point

Samples were collected using a stainless steel, open barrel gravity corer. Cores were dated using lead²¹⁰ and cesium¹³⁷. Contaminant analyses included 16 metals (graphite furnace atomic absorption, ICP/MS, CVAA, x-ray fluorescence), PAHs (GC/MS), PCBs and chlorinated pesticides (GC/ECD), nutrients, and butyltins (GC/FPD). NOAA Status and Trends organics data are surrogate-recovery-corrected (Pevin, personal communication). Core segments were also analyzed for TOC, grain size, and percent moisture. Data quality appears to be excellent.

Sites showed little physical disturbance other than bioturbation. Sedimentation rates ranged from about 1 to 2 cm/yr.

Material from these cores is held in archive (Crececius, personal communication).

The data provided by this study are particularly valuable for several reasons:

- Cores were long and undisturbed, providing records of 80-180 years.
- A wide range of possible contaminants were analyzed.
- Data quality is excellent, including low detection limits.
- Many subsegments of each core were analyzed, providing good spatial resolution.

The results of analysis for seven analytes (PCBs, DDT and metabolites, benzo(a) pyrene, lead, arsenic, mercury, and cadmium) are shown in Figures 4 – 10.

The PCB results are summarized in Figure 4. Prior to 1915 concentrations of total PCBs are very low – less than 1 ug/Kg. Concentrations then rise, reaching their peaks (25-35 ug/Kg) between 1960 and 1965. Concentrations then begin declining and by 1990 have fallen to 5–10 ug/Kg.

There are several interesting aspects of this record.

- Historically, there appears to be a north-south concentration gradient, with PCB concentrations near Seattle being somewhat higher than those near Tacoma.
- PCB concentrations in Puget Sound appear to begin increasing about 20 years earlier than they do in the Olympic Peninsula lakes discussed above. Because commercial production of PCBs did not begin in the U.S. until 1929, the Beaver and Ozette lakes records seem more reasonable. The apparent early appearance of PCBs in the Puget Sound record is probably an artifact caused by the relatively deep mixed sediment layer found in Puget Sound sediments. Bioturbation by benthic invertebrates mixes newly deposited sediments down into older sediments “smearing” the record. Without this “smearing,” peak (1960) concentrations in these sediment records would probably have been higher.
- Peak PCB concentrations appear in the 1960-1965 period for each of the three Puget Sound cores and both of the Olympic Peninsula lake cores. Additional information about PCB production rates and handling practices prior to EPA restrictions in 1977 would help explain the timing of this peak.

The total DDT (DDD + DDE + DDT) results are summarized in Figure 5. With one anomaly, concentrations of total DDT are undetected prior to 1915. Concentrations then rise, reaching peaks (4-5 ug/Kg) between 1945 and 1965. These peaks appear to coincide well with the peak production of DDT in 1959. Concentrations then began declining at two sites and by 1990 had fallen to 1-3 ug/Kg. Concentrations at one site (near Seahurst Park) show recent (and puzzling) increases to over 5 ug/Kg.

Like the PCB record, DDT shows apparent increases in sediment DDT about 30 years prior to the date that it was first produced in the U.S. This appears to be another case of “smearing” due to deep sediment-mixing layers.

Figure 6 shows benzo(a)pyrene [B(a)P] results. B(a)P is a byproduct of combustion, and the sediment record is probably heavily influenced by the history of coal burning in western Washington. Concentrations appear to increase from a baseline of less than 5 ug/Kg prior to 1880, to peaks at two sites of about 300 and 670 ug/Kg around 1943. The third site, near Dash Point, has a broader peak of 100-150 ug/Kg between 1920 and 1955. Like PCBs, but unlike DDT, there is an historical north-south gradient with peak concentrations being higher near Seattle and lower near Tacoma. Recent (1990) concentrations appear to have leveled off at about 100 ug/Kg.

Figure 7 shows cadmium results. Of the contaminants evaluated here, only cadmium shows little fluctuation over the past 200 years. With only one exception, all concentrations reported in these cores were between 0.2 and 0.5 mg/Kg.

Figure 8 displays arsenic results. As noted in earlier studies (Crecelius, 1974; Crecelius et al., 1975) the Tacoma smelter (ASARCO), which operated from 1890 until 1986, was a prominent source of arsenic to the Puget Sound basin. Prior to 1905, sediment concentrations are less than 10 mg/Kg. They then rise to a broad peak of 15-25 mg/Kg between 1925 and 1960, after which there appears to be a modest decline to 12-17 mg/Kg by 1990. Several measurements off Seahurst Park around 1960 exceed 25 mg/Kg. Note that, unlike patterns for PCBs and B(a)P, arsenic concentrations are generally higher at the sites near Tacoma and lower near Seattle. Information about production rates, pollution control efforts and, most specifically, environmental discharge rates of arsenic from ASARCO to air and water could help explain specific aspects of Figures 8, 9, and 10.

Lead results are shown in Figure 9. These results generally track the arsenic results. Prior to about 1890 they establish a baseline of about 10 ug/Kg, then increase until about 1920. From 1920 to 1960 lead concentrations form a broad peak of 40-70 ug/Kg, then begin to decline. By 1990, concentrations are in the 30-45 ug/Kg range. Like arsenic, peak lead concentrations are higher near Tacoma than near Seattle.

Mercury results (Figure 10) generally track those of arsenic and lead. Prior to 1890 they establish a baseline of less than 0.1 ug/Kg, then climb steeply reaching peaks of 0.3-0.46 ug/Kg between 1915 and 1965. Subsequent decreases bring 1990 concentrations down to 0.18-0.28 ug/Kg.

Of the contaminants evaluated, only mercury exceeded Puget Sound Sediment Standards (Chapter 174-204 WAC) during the period of record. Mercury concentrations at the station off Seahurst Park approached or exceeded the standard of 0.41 ug/Kg between about 1915 and 1965. It is interesting that the station located roughly midway between Tacoma and Seattle showed the highest mercury levels. This may reflect different dispersion and deposition characteristics with mercury, which is more volatile than arsenic or lead. Alternatively, it may suggest that other mercury sources influence the distribution of mercury at these sites.

Figure 4.
History of PCB Contamination in Puget Sound Sediments

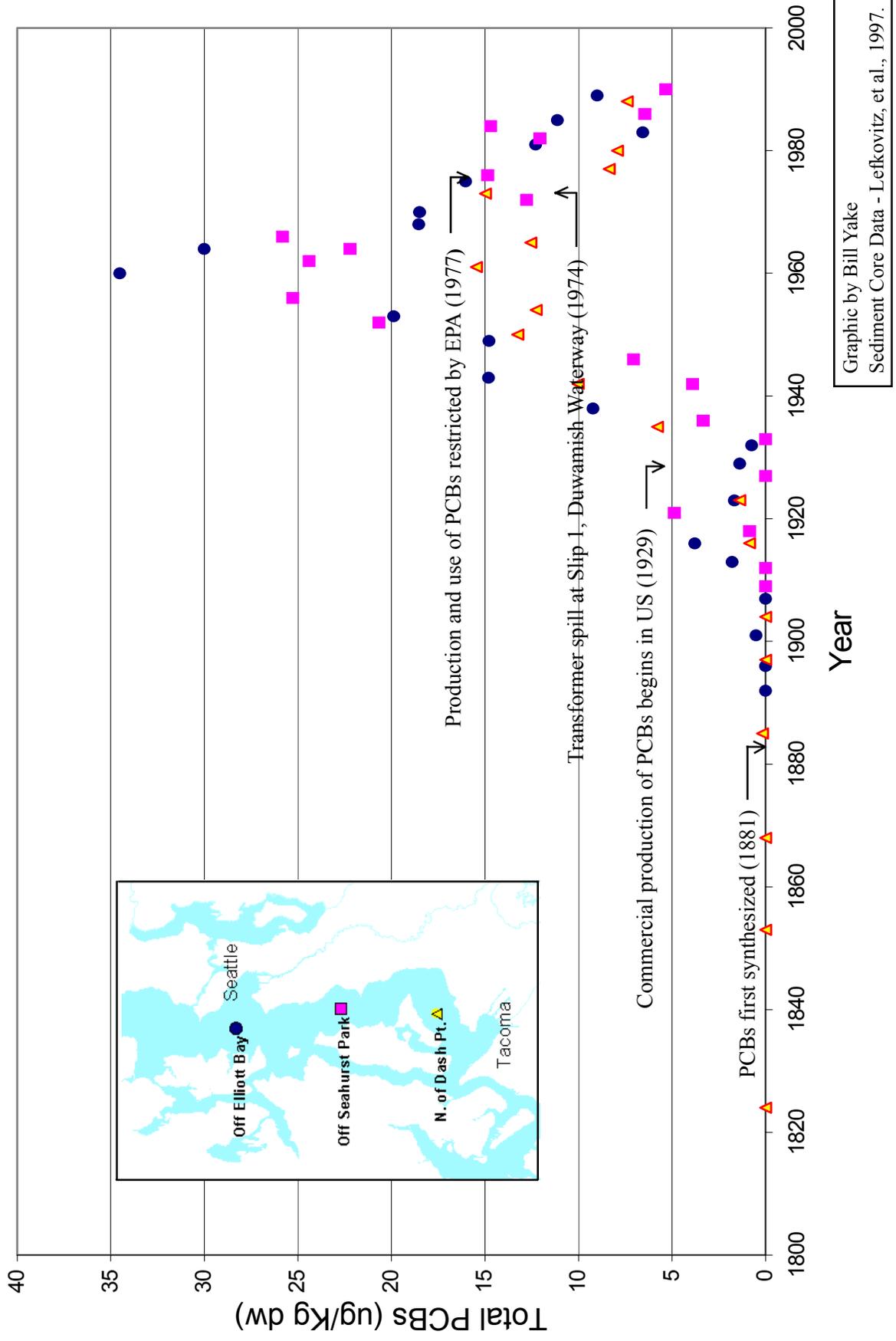


Figure 5.
History of Total DDT Contamination in Puget Sound Sediments

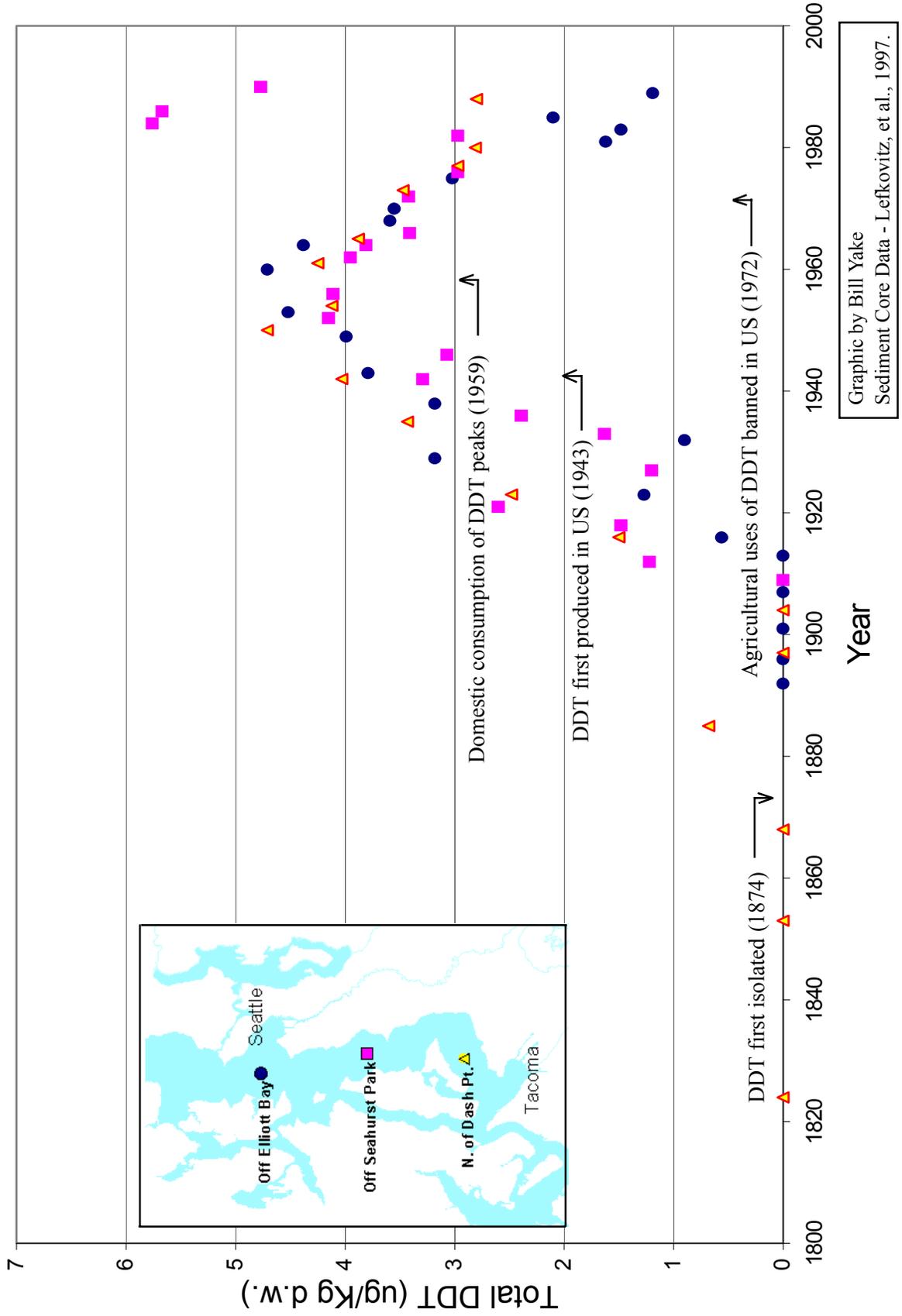


Figure 6.
History of Benzo(a)Pyrene Contamination in Puget Sound
Sediments

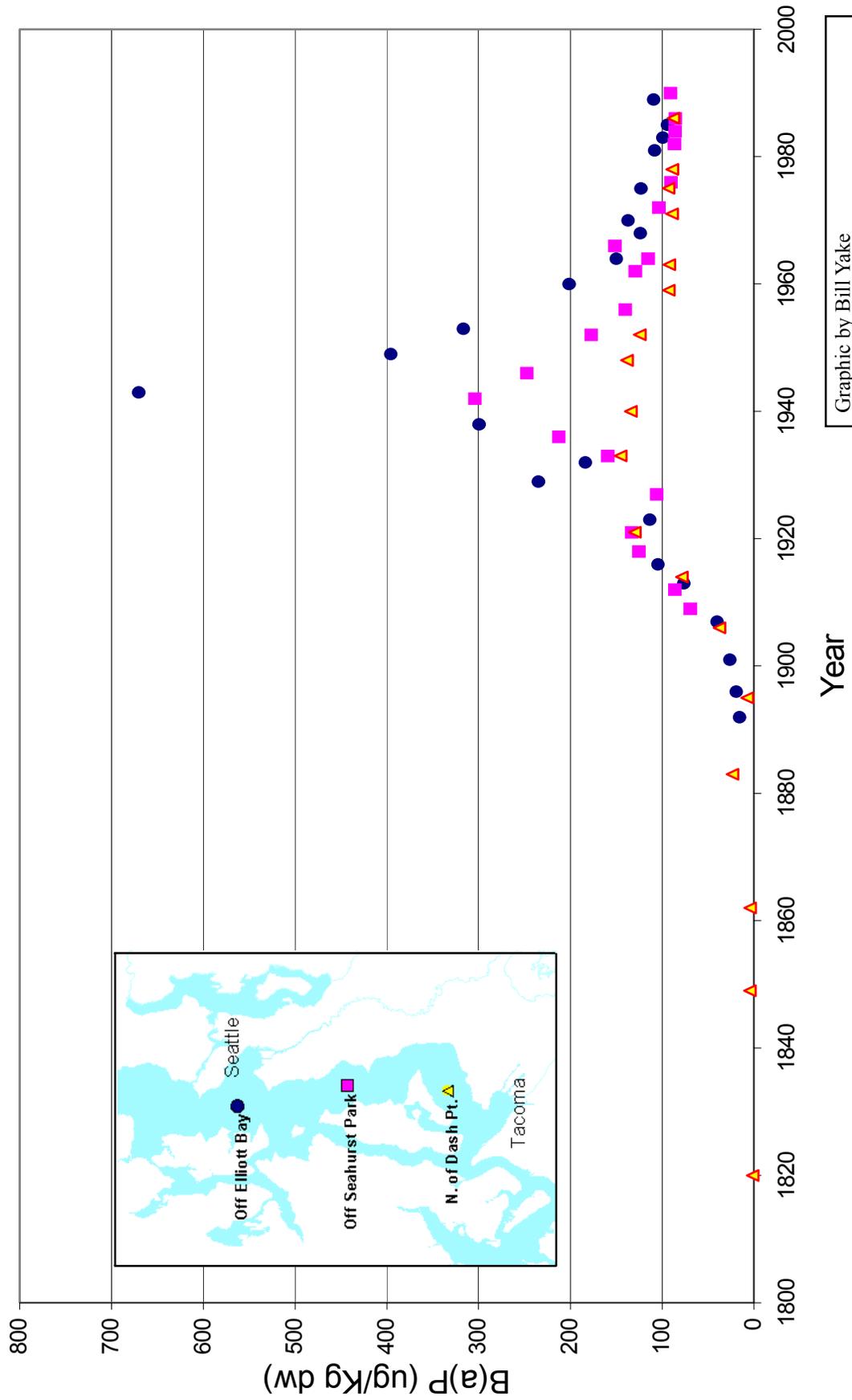
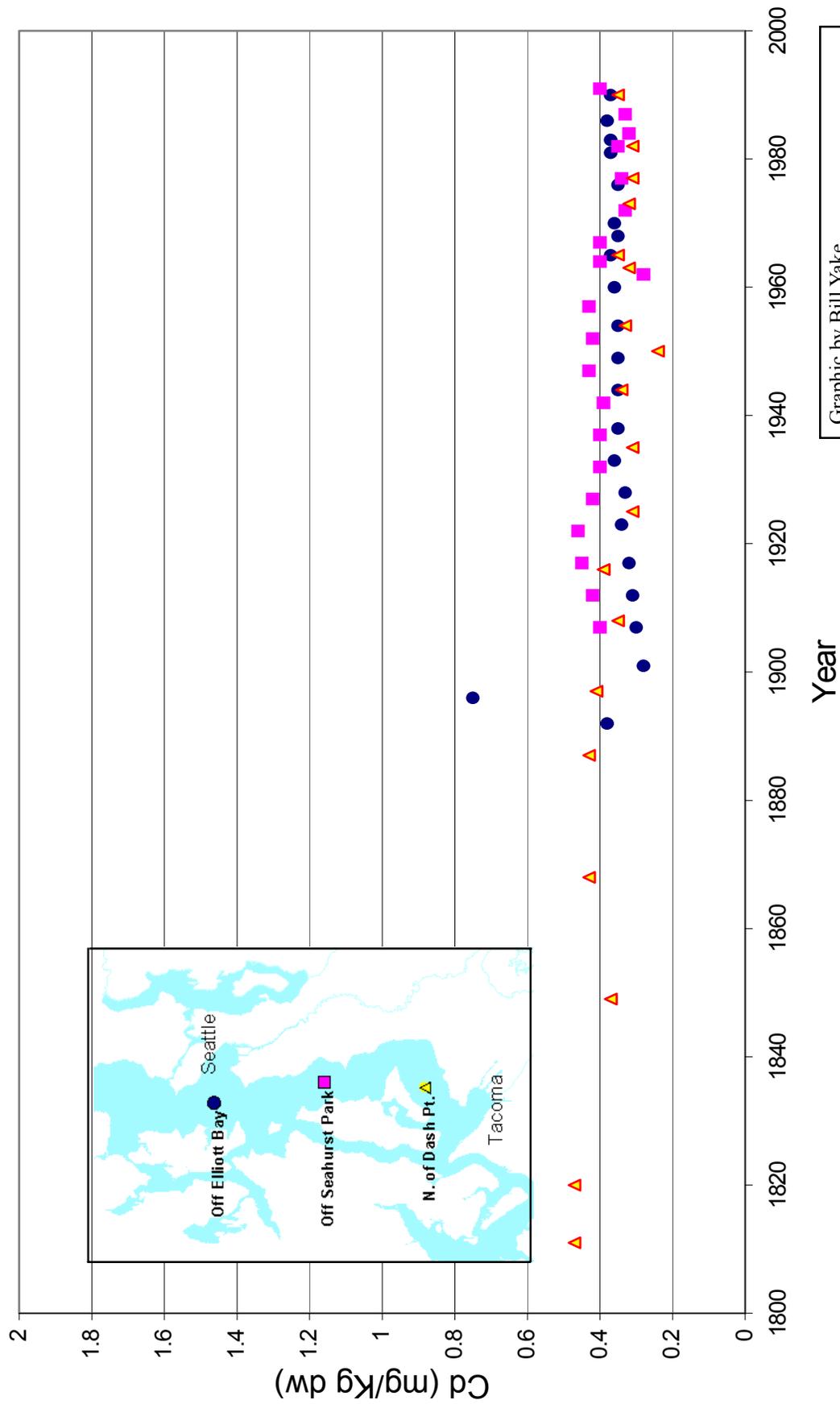


Figure 7.
History of Cadmium Contamination in Puget Sound Sediments



Graphic by Bill Yake
Sediment Core Data - Lefkowitz, et al., 1997.

Figure 8.
History of Arsenic Contamination in Puget Sound Sediments

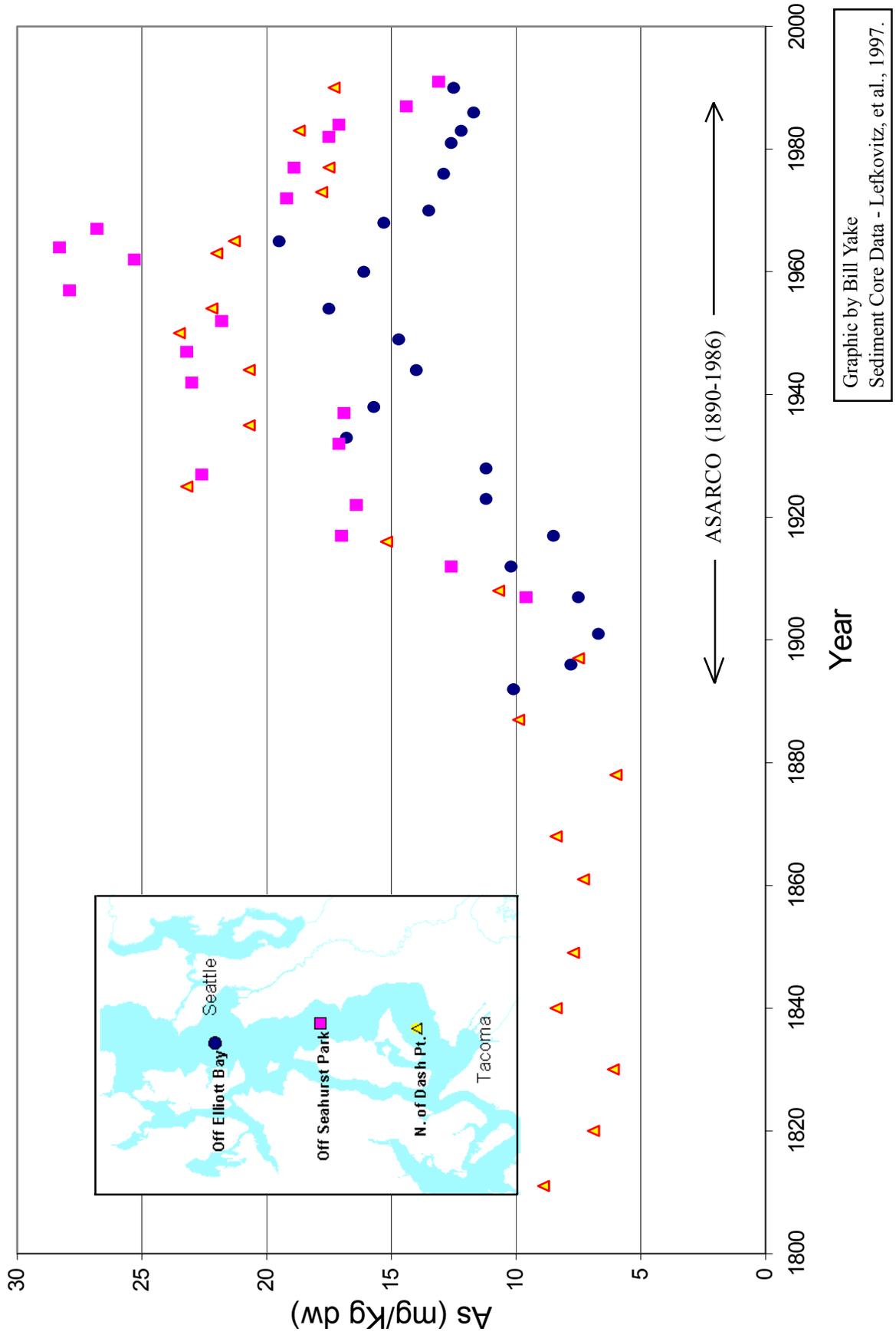
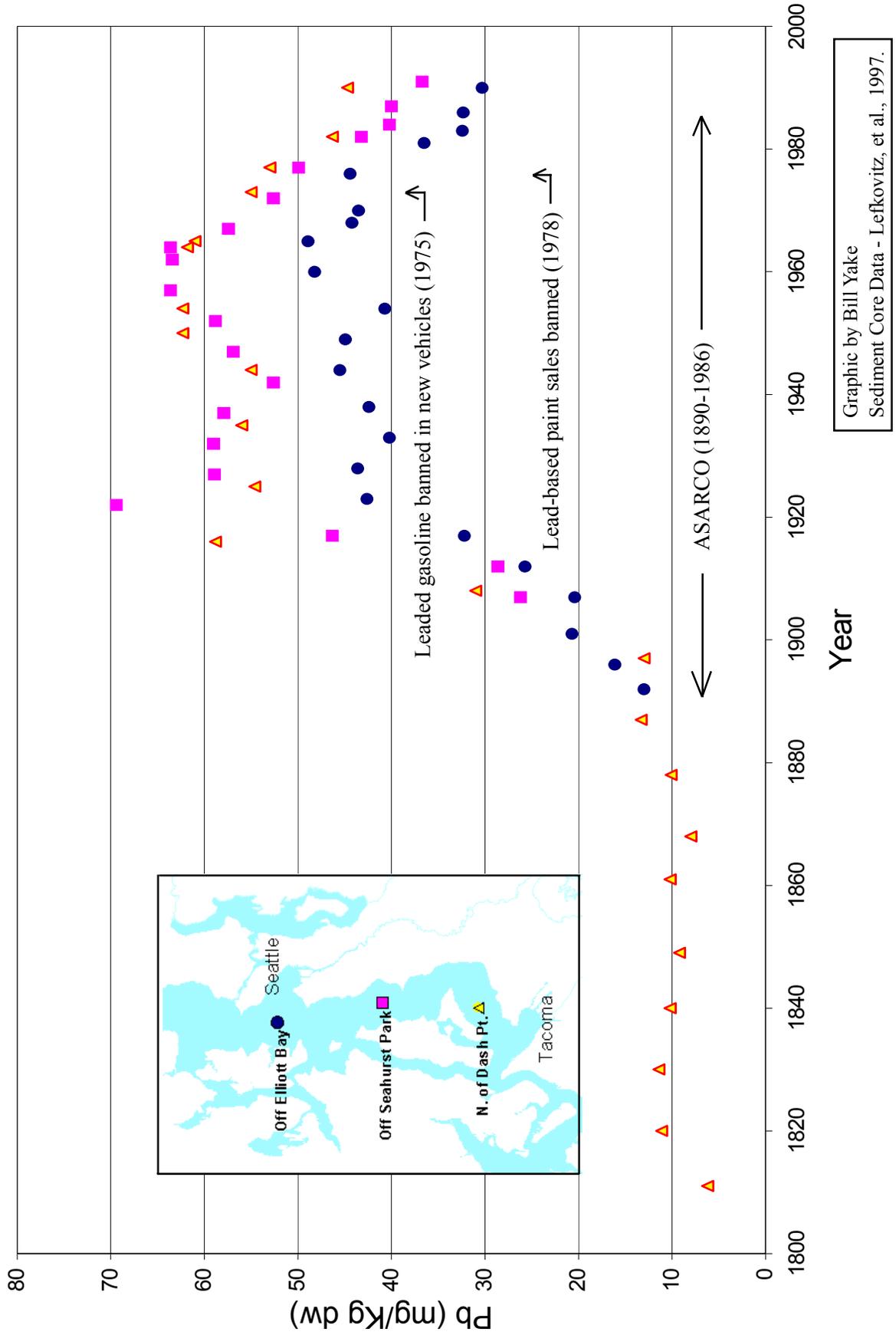
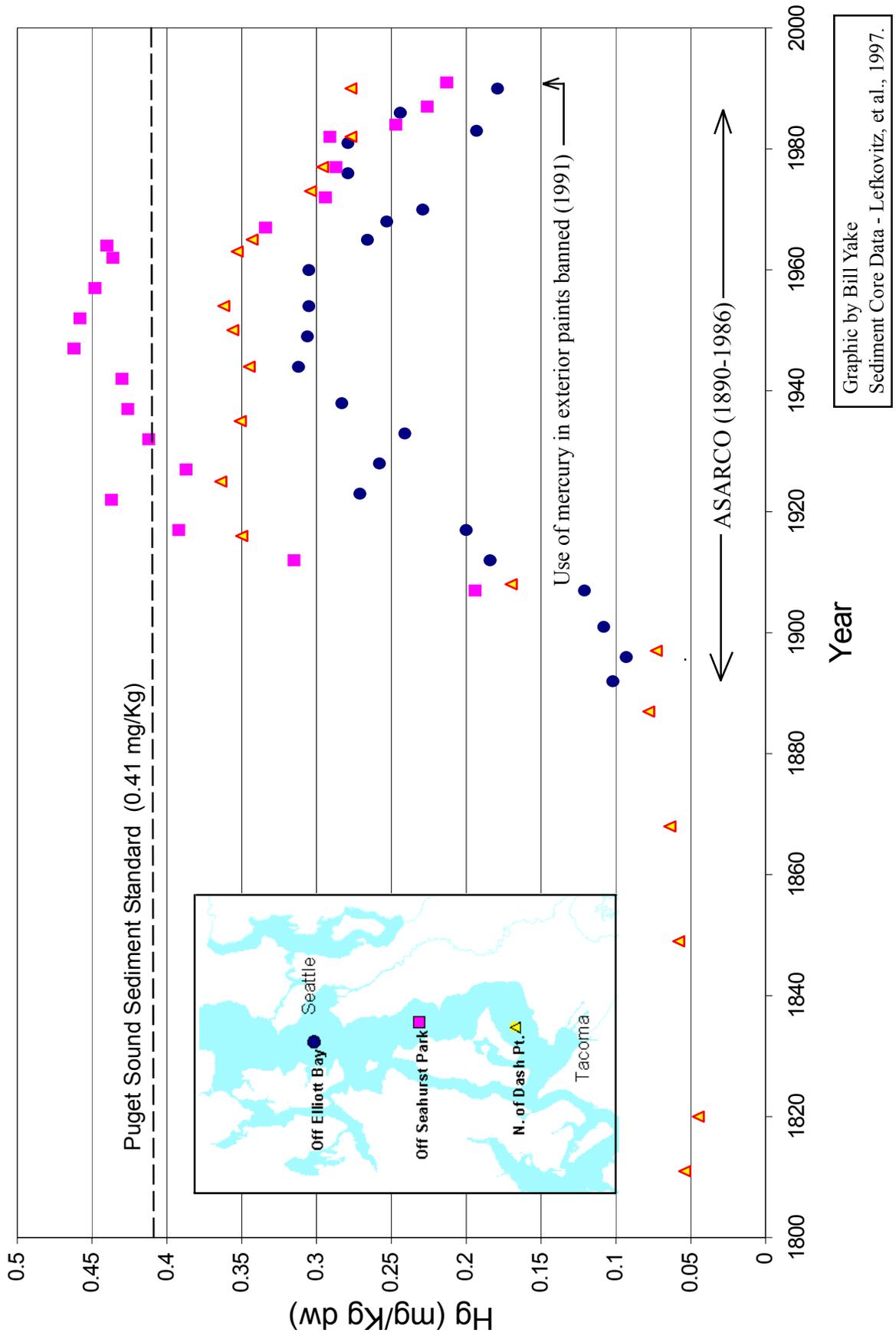


Figure 9.
History of Lead Contamination in Puget Sound Sediments



Graphic by Bill Yake
Sediment Core Data - Lefkowitz, et al., 1997.

Figure 10.
History of Mercury Contamination in Puget Sound Sediments



10. 2000, Johnson, A. and R. Plotnikoff. Review of Sediment Quality Data for the Similkameen River. Washington State Department of Ecology, Olympia, WA. Publication No. 00-03-027.

This work was conducted by the Washington State Department of Ecology and evaluated sediment chemistry and bioassay results from the Washington portion of the Similkameen River.

Three cores were collected in 1999 from sediments from the upper, middle, and lower reservoir impounded behind Enloe Dam. These cores were collected using a 4-inch vibra-corer fitted with an acrylic liner. The upper and middle reservoir cores were sectioned into two sections (0-30 cm and 30-60 cm) while the lower reservoir sample was a single 0-30 cm section. No dating was done. Sediments were quite coarse – mostly sand. Johnson (personal communication) reports that penetration of this substrate with the vibra-corer was difficult.

Sediments were analyzed for a wide range of metals (including arsenic, copper, mercury, lead, cadmium), cyanide, semivolatile organics, PCBs, pesticides, dioxins, and furans. Concentrations were quite low, generally being highest in the top 30-cm of the upper reservoir sample.

11. U.S. Geological Survey Reconstructive Trends Project.

11-A. 1997, Van Metre, P.C. and E. Callender. *Study plan for the reconstructed trends project of the National Water-Quality Assessment Program.* U.S. Geological Survey. Posted on Internet at <http://txwww.cr.usgs/coring/approach.html>

This sampling plan describes work subsequently conducted by the U.S. Geological Survey (USGS) and published, in part, by Van Metre et al., 2000 (see below). Selected data from this work is also presented graphically and discussed in section 11-C below.

The sampling plan describes work to be conducted as part of the USGS National Water-Quality Assessment Program (NAWQA). One of the objectives of NAWQA is to “define trends (or lack of trends) in our nation’s water quality.” A pilot study has been underway since 1992 investigating the use of cores to “describe natural and anthropological changes in water quality in drainage basins of reservoirs.”

Analyses have included radiochemistry, trace elements (metals), chlorinated organics, and PAHs. Special attention seems to be paid to total DDT, PCBs, B(a)P, lead, and cesium¹³⁷.

The proposed study is designed to build on the pilot study. The proposed study objectives are to:

- Identify trends, or lack of trends, in the nation’s surface waters for hydrophobic constituents including trace metals, hydrophobic organic compounds, and, to some extent, nutrients.
- Characterize relationships between trends and major land use settings including urban and agricultural, and to a limited extent, mining.

Six principles for site selection are given including targeted land uses. These are, in order of importance: suburban, “old” urban, agricultural, mining, and background.

Characteristics of reservoirs sampled include:

- ideally built before 1960
- not dredged
- not drained for any length of time
- defined drainage with known land use

Characteristics of urban indicator sites include:

- ideally relatively small drainage basins (10-100 km²)
- relatively homogeneous land use
- as a group, represent a range of population densities
- industrial areas and point sources will be avoided

Agricultural indicator sites “can be larger in most areas and still be representative of a relatively homogeneous land use.”

Mining indicator sites “will be selected on a case-by-case basis.”

11-B. 2000, Van Metre, P.C., B.J. Mahler, and E.T. Furlong. *Urban sprawl leaves its PAH signature. Environmental Science and Technology. Vol. 34:4064-4070.*

This study was conducted by the USGS National Water-Quality Assessment Program (NAWQA) under the work plan referenced above.

The publication reports and interprets polycyclic aromatic hydrocarbon (PAH) data for ten cores (three from lakes, seven from reservoirs) collected across the U.S. Watershed land use for these drainages is “largely mixed residential and commercial with percent urban land use ranging from 25 to nearly 100%.” Included are data for Lake Ballinger in north Seattle.

Sections of cores were analyzed for cesium¹³⁷ and PAHs (GC/MS).

The core collected from Lake Ballinger represents the record from 1947-1998. The rate of sediment deposition in Lake Ballinger is reported at 0.45 cm/yr.

General findings reported here are:

- “Modest to dramatic increases in total PAH concentrations are seen in sediments deposited in all 10 watersheds over the last 20-40 years” with recent concentrations 1 to 2 orders of magnitude above pre-development concentrations.
- “A general shift in PAH source over the last 40 years from uncombusted to combusted fossil fuels.”
- An implied linkage between increased traffic volumes in urban areas and increased PAH concentrations in sediments in these areas. Tire wear, crankcase oil, roadway wear, car soot, and exhaust are mentioned as specific possibilities.
- PAH concentrations at surface of cores exceed Canadian interim freshwater sediment quality guidelines (ISQG) for protection of aquatic life (Environment Canada, 1995) for three prevalent PAHs in all 10 lakes. Six or more of the lakes exceed the probable effects levels (PELs, the concentrations above which adverse biological effects are expected to occur) – in some cases by as much as 12-25 times. “Clearly, increases in PAHs in urban water bodies have degraded sediment quality to the point that it is an ecological concern.”

The publication also notes the “presence of a 1950s [PAH] peak and subsequent decreasing trend” that is attributed to “the transition from home heating with coal to the use of oil and natural gas and the increasing efficiency of centralized power plants.”

11-C. 2000, USGS. *Reconstructed Trends National Synthesis Study*. U.S. Geological Survey. Posted at <http://tx.usgs.gov/coring/>

USGS results from the analysis of Lake Ballinger and Lake Washington sediment cores have recently been posted at this Internet site.

The location of these lakes is shown in Figure 11.

Although detailed information on sampling, analysis, and quality assurance techniques is pending, personal communication with Van Metre (2000) yielded the following sampling information. When cores of <50 cm length are needed, a 14x14 cm box corer is used. This is preferred as it yields a large volume sample with little disturbance. When a core with greater than 50-cm length is needed, a 2.5 inch diameter gravity or piston corers is used. These allow greater penetration without disturbing (shortening) the core.

The results of analysis for seven analytes (PCBs, DDT and metabolites, B(a)P, lead, arsenic, mercury, and cadmium) are shown in Figures 12-18. For these graphs a sedimentation rate of 0.45 cm/yr was used for Lake Ballinger as reported by Van Metre et al., 2000. For Lake Washington, using the peak cesium¹³⁷ concentration as a marker (1964) yields a sedimentation rate of 0.3 cm/yr. This is the same rate estimated by Crecelius (1974).

The PCB results are summarized in Figure 12. Total PCB concentrations begin to rise in the early 1940s. This corresponds well with US PCB production beginning in 1929. Concentrations in both lakes rise, reaching their peaks (220-265 ug/Kg) during the late 1960s. Concentrations then fall steeply and by 1980 have reached 40-75 ug/Kg, where they level off near the “threshold effects level of approximately 60 ug/Kg. Inexplicably, during the most recent years, concentrations in Ballinger sediments appear to be once again increasing. With the exception of this late record rise, the shape of these curves is very similar to those in Olympic Peninsula lakes discussed above, although the concentrations are somewhat higher.

The total DDT (DDD + DDE +DDT) results are summarized in Figure 13. Like PCBs, the concentrations in these lake sediments are much higher (10-30 times) than those reported from the center of Puget Sound (Lefkovitz et al., 1997). Concentrations prior to 1940 are low. This contrasts with the record in Puget Sound sediments which shows increasing concentrations starting about 1915. Because DDT was not produced in the U.S. until 1943, it is probable that the relative lack of vertical mixing in lake sediment makes these records accurate.

Total DDT concentrations peak in the late 1950s and early 1960s, with those in Lake Washington reaching about 130 ug/Kg, and those in Lake Ballinger about 75 ug/Kg. These peaks coincide well with those in Puget Sound sediments and the 1959 peak in domestic consumption of DDT. By 1980 concentrations in both lakes appear to have decreased and leveled off in the 10-25 ug/Kg range, still well above the threshold effects

level (the concentration below which adverse effects are unlikely to occur) of approximately 5 ug/Kg.

A major source of DDT in Lake Washington sediments can be inferred from a photograph (Figure 14) from the *Seattle Times* (1998) showing a 1957 DDT application in Lake Washington's Union Bay.

Figure 15 shows benzo(a)pyrene results. Because B(a)P is a PAH, the discussion of PAH trends above (Van Metre et al., 2000) holds. Lake Ballinger is adjacent to high traffic volumes, being located in Edmonds directly between highways I-5 and SR-99. This may well account for the B(a)P trend in Lake Ballinger sediments, which begins to climb steeply in the mid-1960s. Concentrations in Lake Ballinger sediments over the past 15 years have ranged from 1000-3000 ppb, while Lake Washington sediment concentrations peak at 104 ppb in 1973.

In addition to the threshold effects level, a "consensus probable effects concentration" (PEC) is included in the graph. This PEC is reported by MacDonald et al. (2000), and identifies "concentrations above which harmful effects on sediment-dwelling organisms are expected to occur frequently." Recent B(a)P concentrations in Lake Ballinger sediments exceed this guideline.

Lead results are shown in Figure 16. Lake Washington sediments show initial increases early in the 20th century; these may be associated with the Tacoma ASARCO smelter. Lead concentrations rise more steeply in both lakes from the 1960s through the middle-to-late 1970s. Lake Ballinger lead concentrations are particularly high, peaking at about 660 mg/Kg in 1975. This is the year the use of leaded gas was banned in new cars. Since 1980 lead concentrations have fallen in sediments in both lakes, leveling off at about 275 mg/Kg in Lake Ballinger and about 140 mg/Kg in Lake Washington.

Since approximately 1950, lead concentrations in the sediments of both lakes have exceeded the PEC of 128 mg/kg. Recent concentrations in Lake Ballinger sediments remain more than double this guideline.

Figure 17 shows arsenic results. Like earlier work by Crecelius (1975), concentrations in Lake Washington sediments rise from a baseline of less than 10 mg/Kg starting in the earlier 1900s. They reach a peak of about 36 mg/Kg, slightly above the PEL, in the late 1930s. The post-1950 record is unusual in both lakes, with arsenic concentrations fluctuating between 10 and 25 mg/Kg. The early (1910-1950) record in Lake Washington appears to have been influenced by the Tacoma smelter.

Mercury concentrations are shown in Figure 18. The record for Lake Washington is similar to the Puget Sound records for metals, which appear to have been influenced by the Tacoma smelter as discussed above. Concentrations rise from a baseline of less than 0.2 ppm beginning early in the 20th century. From about 1930 to 1970 they plateau (0.5-0.75 ppm) and then begin to fall. The most recent concentrations are about

0.35 ppm. Lake Ballinger sediments fluctuate between about 0.3 and 0.7 ppb, with lower concentrations in the early 1970s and higher concentrations around 1990. The source(s) of mercury to Lake Ballinger are currently unknown.

The cadmium record (Figure 19) for Lake Washington shows an increase from about 0.2 ppm to about 2.5 ppm in the first half of the 20th century. After that, concentrations fluctuate between 1 and 2 ppm, with recent concentrations being about 1.4 ppm. The Lake Ballinger record shows an apparent drop from 1950 to 1960 (1.7 to 1.1 ppm) which corresponds closely with the Lake Washington record. This is followed by an increase, with recent concentrations reaching 2.0-2.4 ppm. Both the presence of apparent trends and the elevated (>0.4 mg/Kg) concentrations in these lake sediments contrast with the history recorded in Puget Sound sediments. This implies local (airshed or watershed scale) sources that are currently unidentified.

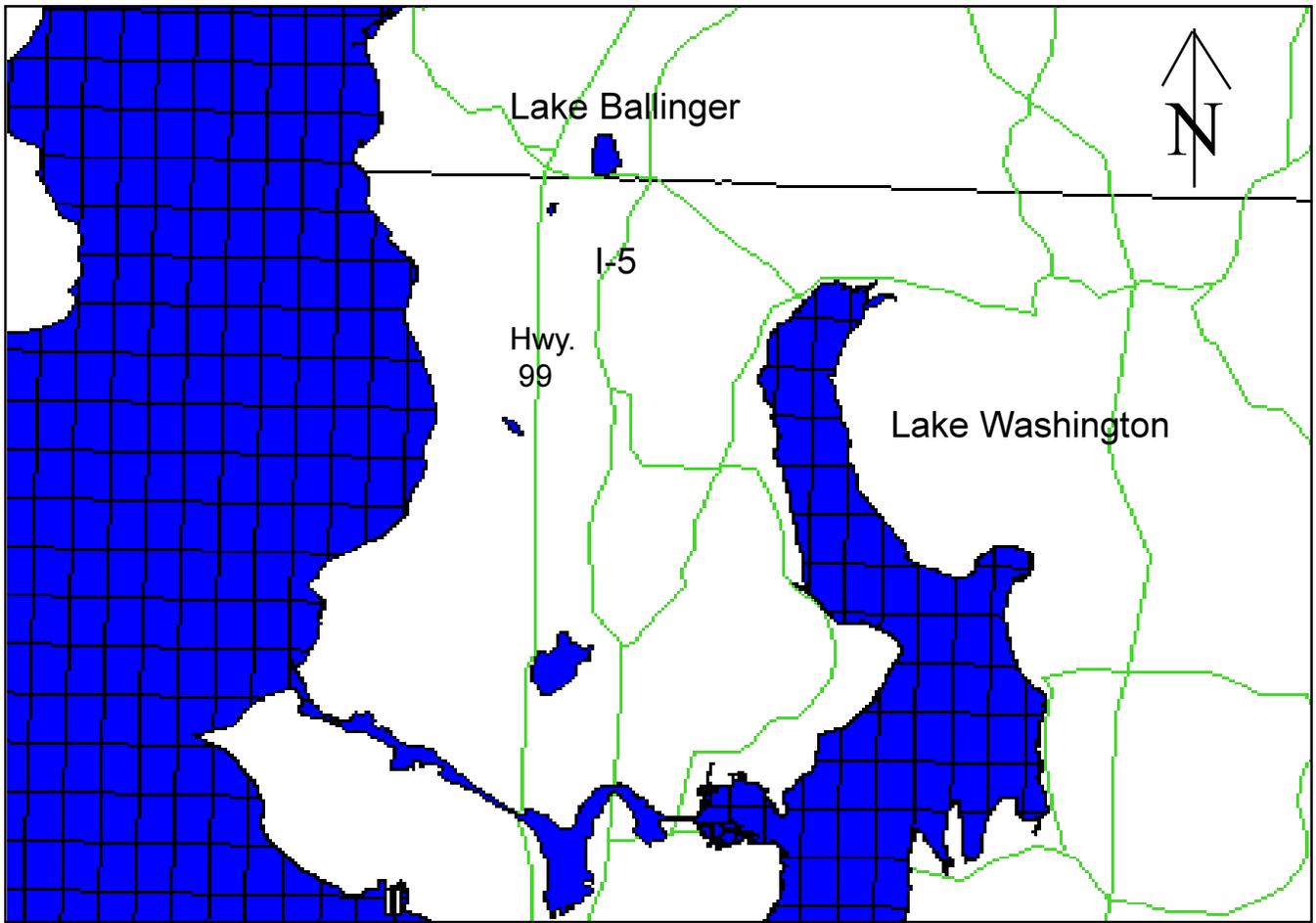
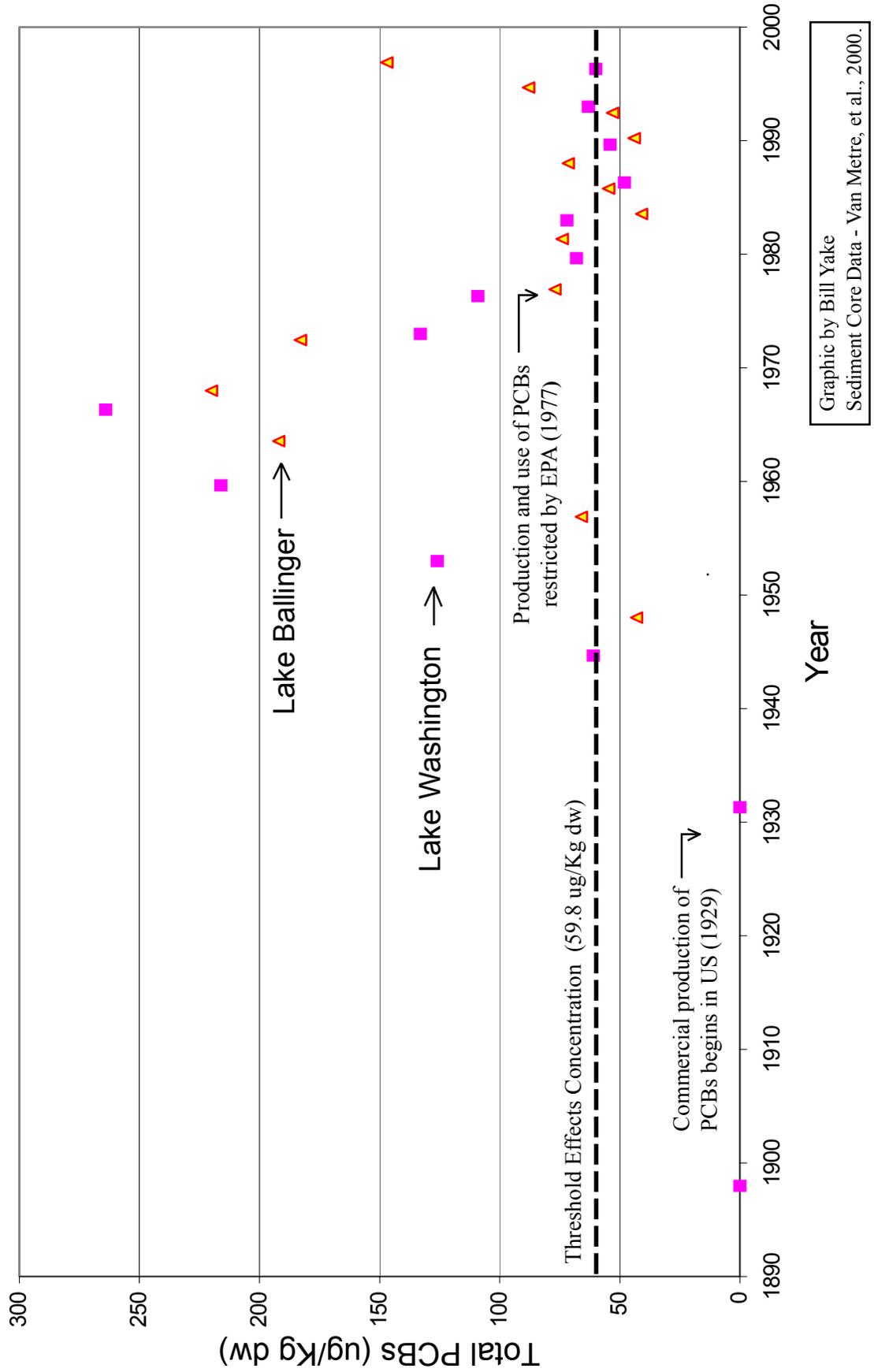


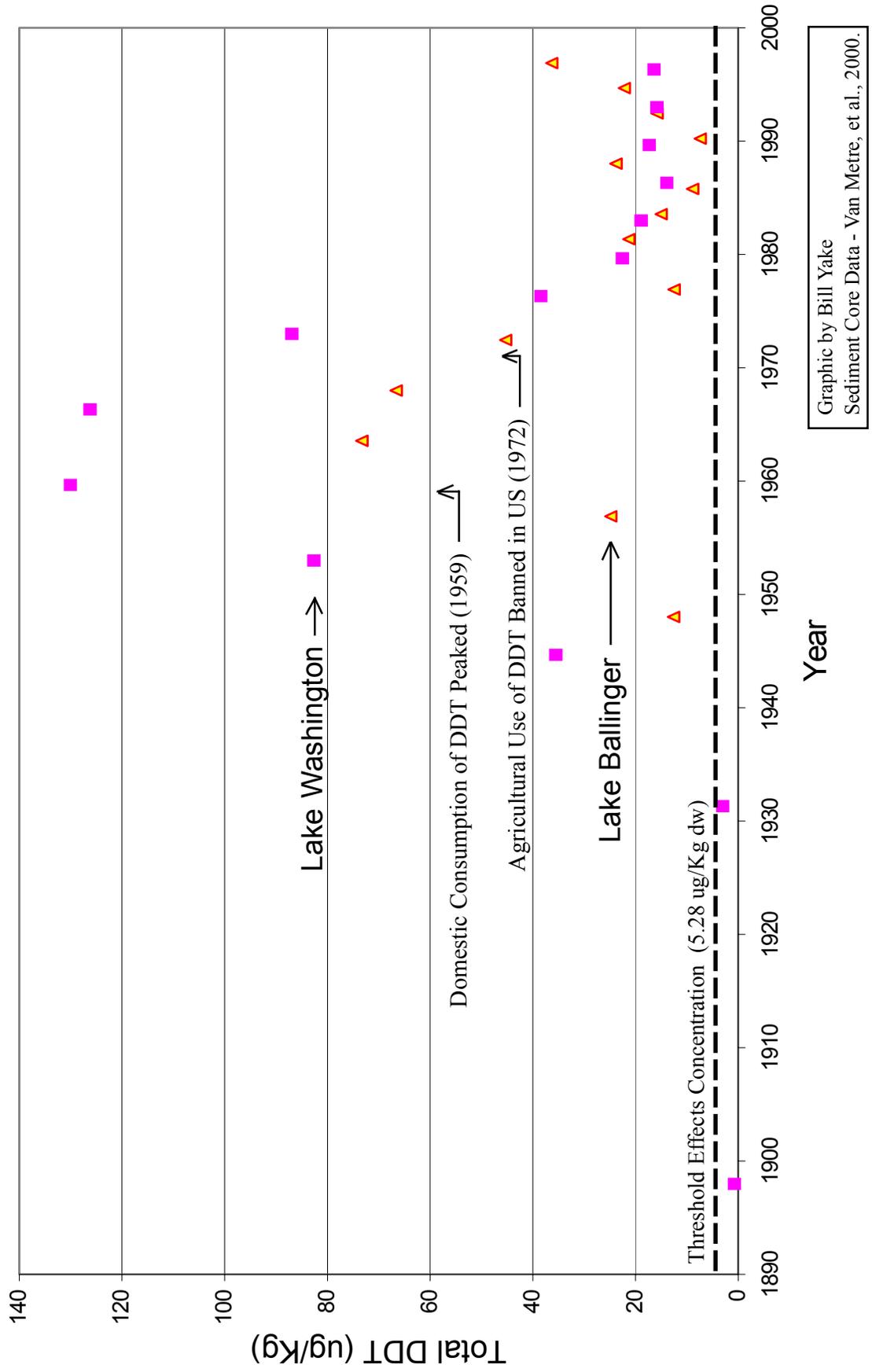
Figure 11. Seattle Area Lakes

Figure 12.
 History of PCB Contamination in Lake Washington
 and Lake Ballinger Sediments



Graphic by Bill Yake
 Sediment Core Data - Van Metre, et al., 2000.

Figure 13.
History of Total DDT Contamination in Lake Washington and Lake Ballinger Sediments

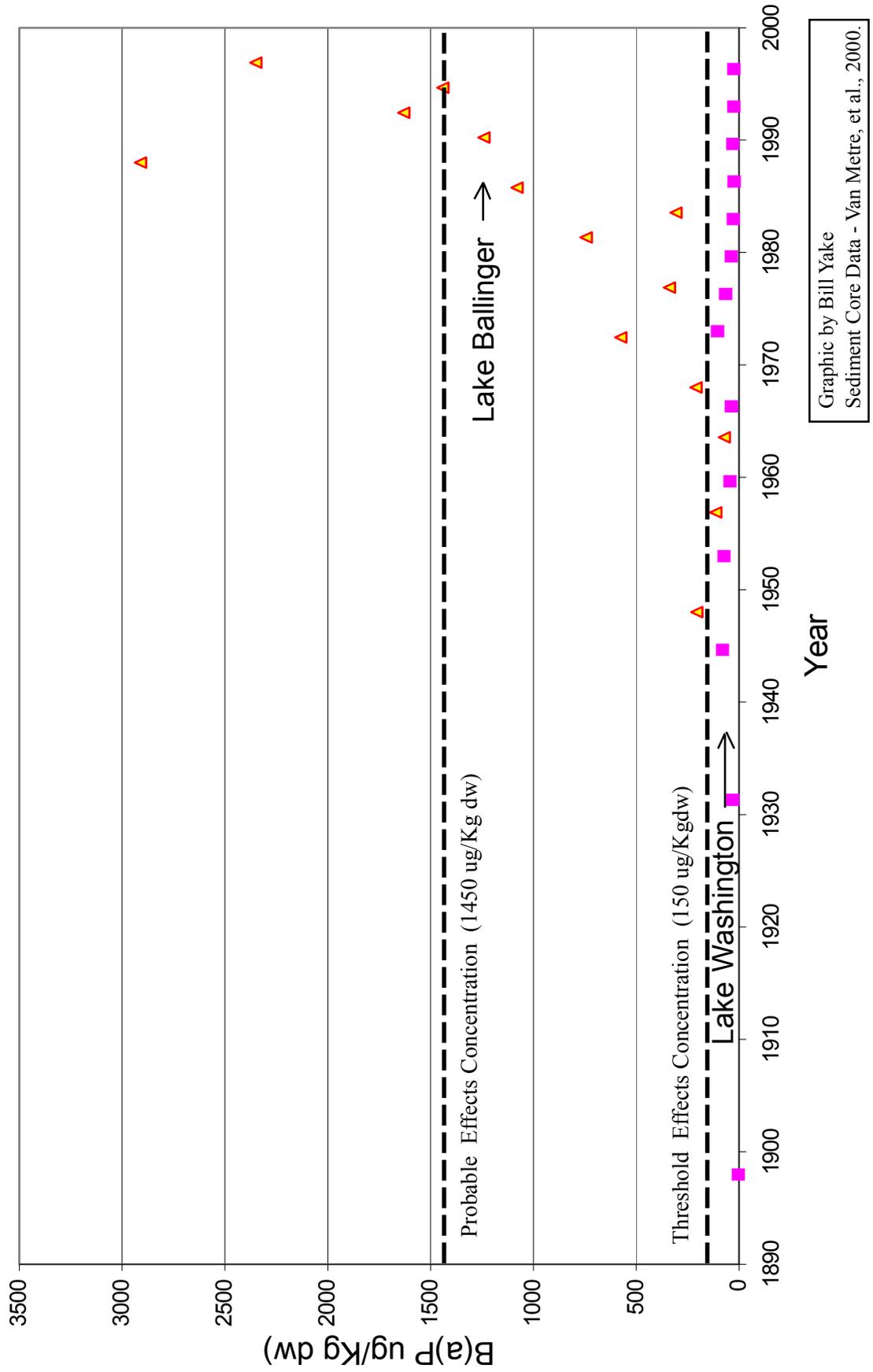


Graphic by Bill Yake
Sediment Core Data - Van Metre, et al., 2000.



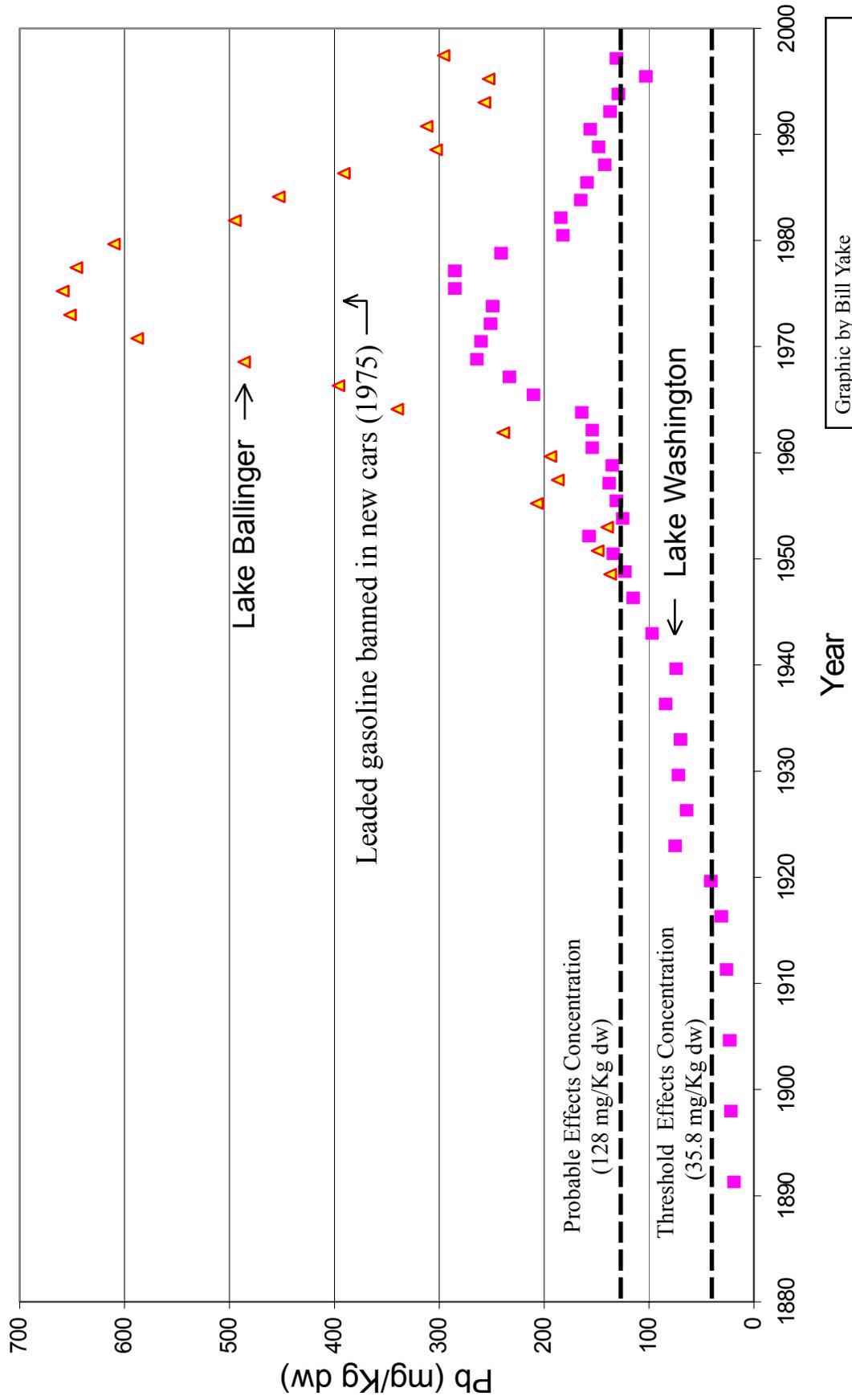
Figure 14. Fog-spraying (smoke and DDT) swampy areas of Lake Washington's Union Bay - 1957. (Seattle Times photo.)

Figure 15.
 History of Benzo(a)Pyrene Contamination in Lake Washington and
 Lake Ballinger Sediments



Graphic by Bill Yake
 Sediment Core Data - Van Metre, et al., 2000.

Figure 16.
 History of Lead Contamination in Lake Washington
 and Lake Ballinger Sediments



Graphic by Bill Yake
 Sediment Core Data - Van Metre, et al., 2000.

Figure 17.
 History of Arsenic Contamination in Lake Washington
 and Lake Ballinger Sediments

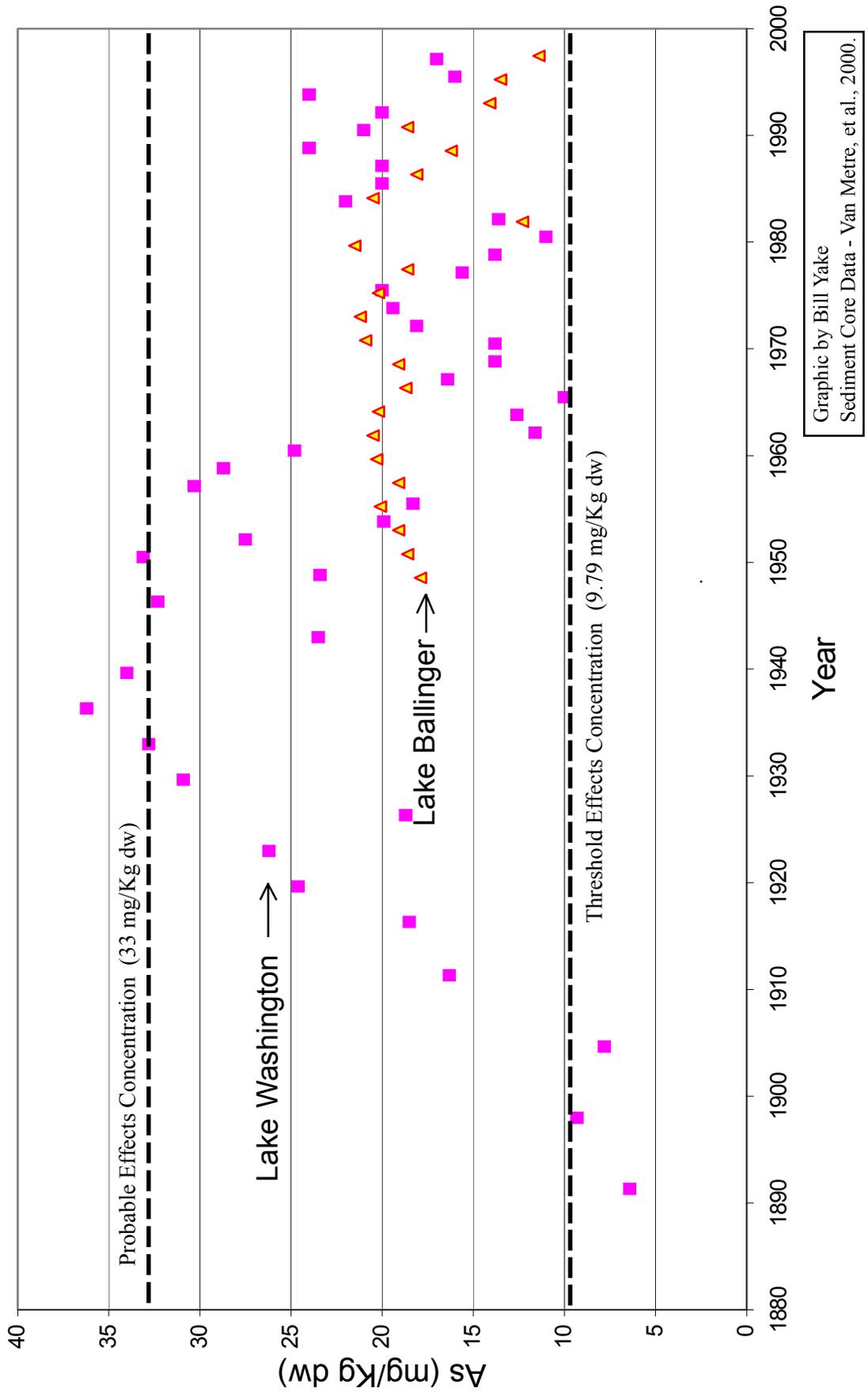
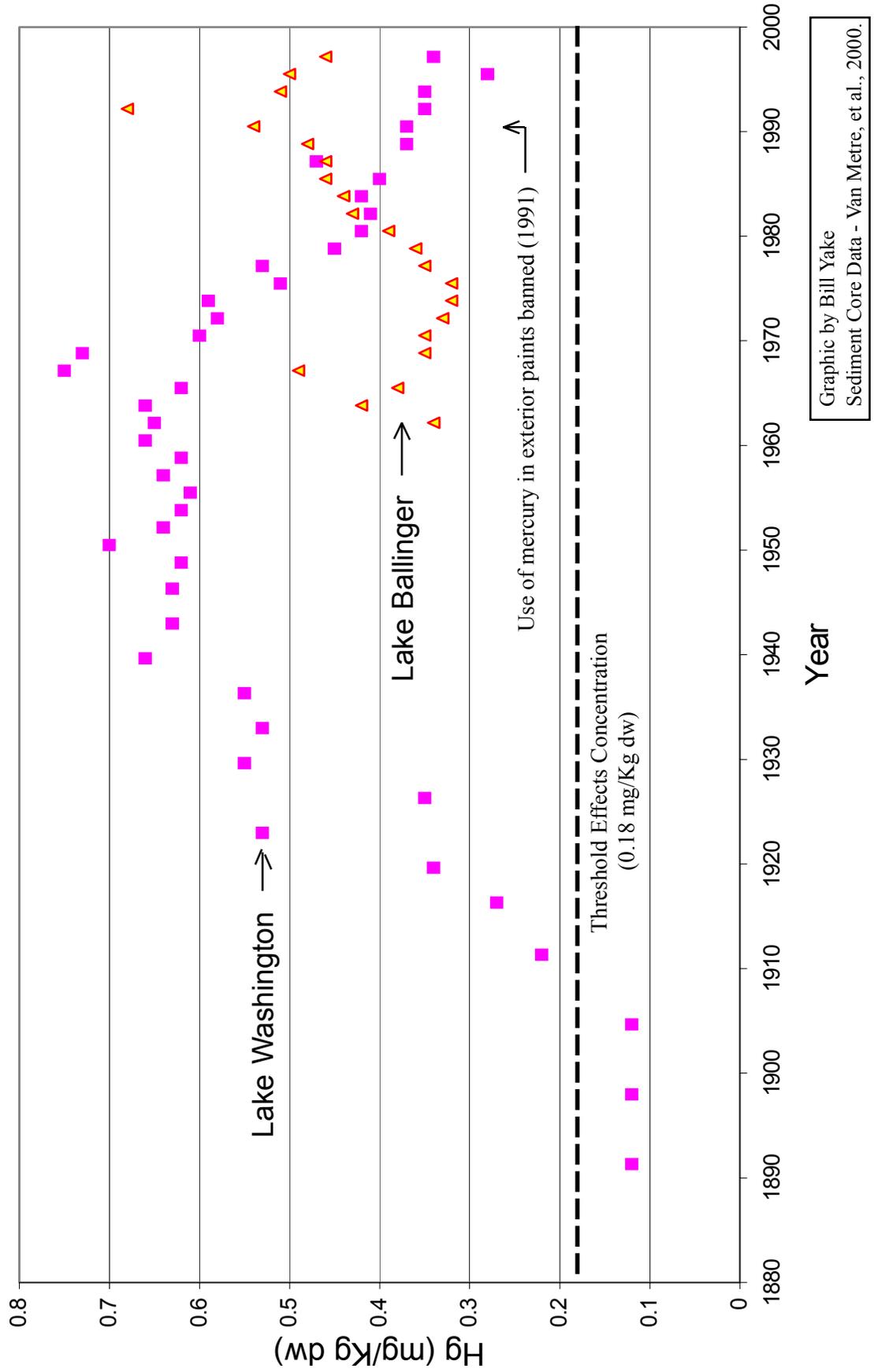


Figure 18.
 History of Mercury Contamination in Lake Washington
 and Lake Ballinger Sediments



Graphic by Bill Yake
 Sediment Core Data - Van Metre, et al., 2000.

Conclusions Derived from this Review; Considerations for Core Sampling

1. Sediment cores can provide a powerful tool for determining the history of persistent pollutant contamination in lakes and marine waters. The trends revealed by analysis of these cores often coincide with historical events and actions (e.g., peak of DDT use, leaded gas switchover, industrial activities). In other cases, the trends revealed by these cores raise new questions and provide clues about potential sources in the contributing airshed or watershed.
2. With the exception of two cores collected in Olympic Peninsula lakes (PCB, dioxin and furan analyses only), cores providing detailed information on persistent pollutant trends have been collected only from the highly urbanized Seattle-Tacoma corridor. Based on the limited results available, some contaminants (e.g., PCBs, DDT) show similar trends in cores taken from different locations. Others (lead, cadmium, and mercury) show different trends in different locations.
3. A sediment core monitoring program that evaluates cores from lakes and reservoirs with a range of land uses (urban, suburban, agricultural, mining, and wilderness) in both eastern and western Washington is likely to reveal both general trends and airshed and watershed-specific trends for persistent pollutants. Results from cores collected from this wider range of drainages will substantially improve our understanding of the statewide history and current status of persistent pollutants.
4. Coring studies require careful planning to produce useful results.
5. Generally, fine-grained sediments provide better core samples. As long as these sediments are not extremely fine (clays), it is easier to get penetration. In addition, these sediments often have higher contamination levels.
6. Large bore core samplers are less likely to compress or shorten sediment cores, although they can require a larger “platform” (vessel) and can have more difficulty penetrating deep sediments. Large bore corers also yield larger sample sizes for a given core length; this can make for better resolution of time periods, an expanded analyte list, and improved detection limits. Vibra-corers may be required to penetrate sandy sediments, although Johnson (personal communication) reported difficulties with this application. Vibra-corers can disturb the core (Van Schell, 1986).
7. The following markers may be useful in determining sedimentation rates:
 - ◇ Surface of sediments coincides with sample collection.
 - ◇ Mt. St. Helens ash layer in 1980.
 - ◇ Lead peak in 1975 due to leaded gas switchover (Van Metre, personal communication).

- ◇ Cs¹³⁷ from nuclear tests (e.g., peak in 1963).
 - ◇ The bottom of the sediments may coincide with the date that the impoundment was constructed.
8. The length of core required for a specific study depends on the objectives of the study, the history of water body and surrounding land use, method of core dating, and sedimentation rates. Establishing historical contaminant and lead²¹⁰ baselines may require cores reaching into the 1800s.
 9. Consider the potential for disturbance. If looking for an undisturbed historical record, avoid areas where dredging, turbulence, or dumping may have occurred.
 10. Estuarine sediments appear to show considerably more bioturbation than lake sediments. Lake sediments, therefore, can provide “cleaner” historical records. For the same reason, surface samples of lake sediments would reflect changes in contaminant input more quickly than would surface samples of estuarine sediments.

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