



# Tracking Riverborne Sediment and Contaminants in Commencement Bay, Washington, Using Geochemical Signatures



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U.S. Department of the Interior  
U.S. Geological Survey

**Cover.** Photograph of the Puyallup River, Washington, on August 21, 2013 (U.S. Geological Survey photograph by Renee Takesue).

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By Renee K. Takesue, Kathleen E. Conn, and Richard S. Dinicola

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# Conversion Factors

## International System of Units to U.S. customary units

<b>Multiply</b>	<b>By</b>	<b>To obtain</b>
Length		
centimeter (cm)	0.3937	inch (in.)
millimeter (mm)	0.03937	inch (in.)
meter (m)	3.281	foot (ft)
kilometer (km)	0.6214	mile (mi)
Area		
square kilometer (km <sup>2</sup> )	247.1	acre
square kilometer (km <sup>2</sup> )	0.3861	square mile (mi <sup>2</sup> )
Volume		
milliliter (mL)	0.06102	cubic inch (in <sup>3</sup> )
Flow rate		
cubic meter per second (m <sup>3</sup> /s)	70.07	acre-foot per day (acre-ft/d)
cubic meter per second (m <sup>3</sup> /s)	35.31	cubic foot per second (ft <sup>3</sup> /s)
cubic meter per second (m <sup>3</sup> /s)	22.83	million gallons per day (Mgal/d)
Mass		
microgram (μg)	0.00003527	ounce, avoirdupois (oz)
gram (g)	0.03527	ounce, avoirdupois (oz)
kilogram (kg)	2.205	pound avoirdupois (lb)

## U.S. customary units to International System of Units

<b>Multiply</b>	<b>By</b>	<b>To obtain</b>
Length		
mile (mi)	1.609	kilometer (km)
Mass		
ton, short (2,000 lb)	0.9072	metric ton (t)

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as °F = (1.8 × °C) + 32.

Temperature in degrees Fahrenheit (°F) may be converted to degrees Celsius (°C) as °C = (°F - 32) / 1.8.

## Acronyms and Abbreviations

CB	Commencement Bay
dpm/g	disintegrations per minute per gram of dry sediment
EF	enrichment factor
ERL	effects range low
ERM	effects range median
FNU	Formazin Nephelometric Units
GS	sediment grain-size distributions (GS)
IAEA	International Atomic Energy Association
ICP-AES	inductively coupled plasma atomic emission spectrometry
ICP-MS	inductively coupled plasma mass spectrometry
keV	kiloelectronvolts
LOW	Puget Lowland
µg/g	micrograms per gram
µg/kg	micrograms per kilogram
MLLW	mean lower low water
NASC	North American shale composite
NOAA	National Oceanic and Atmospheric Administration
NWQL	USGS National Water Quality Laboratory
PAH	polycyclic aromatic hydrocarbons
PBDE	polybrominated diphenyl ether
PCB	polychlorinated biphenyl
PCMSC	USGS Pacific Coastal and Marine Science Center
ppm	parts per million
pcSDSZ	PCMSC in-house statistical software
ppm	parts per million
RIV	Puyallup and White River
TC	total carbon
TIC	total inorganic carbon
TOC	total organic carbon
USGS	U.S. Geological Survey

## Chemical Symbols Used

Al	aluminum
As	arsenic
Ba	barium
<sup>7</sup> Be	beryllium-7
<sup>214</sup> Bi	bismuth-214
Cd	cadmium
Co	cobalt
Cr	chromium
<sup>137</sup> Cs	cesium-137
Cu	copper
(La/Yb) <sub>N</sub>	normalized lanthanum/ytterbium ratio
Nb	niobium
Ni	nickel
Pb	lead
<sup>210</sup> Pb	lead-210
<sup>210</sup> Pb <sub>ex</sub>	excess <sup>210</sup> Pb
<sup>214</sup> Pb	lead-214
<sup>226</sup> Ra	radium-226
Rb	rubidium
REE	rare earth elements
Sc	scandium
Si	silica
Th	thorium
Tm	thulium
Zn	zinc
Zr	zirconium

# Tracking Riverborne Sediment and Contaminants in Commencement Bay, Washington, Using Geochemical Signatures

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## Abstract

Large rivers carry terrestrial sediment, contaminants, and other materials to the coastal zone where they can affect marine biogeochemical cycles and ecosystems. This U.S. Geological Survey study combined river and marine sediment geochemistry and organic contaminant analyses to identify riverborne sediment and associated contaminants at shoreline sites in Commencement Bay, Puget Sound, Washington, that could be used by adult forage fish and other marine organisms. Geochemical signatures distinguished the fine fraction (<0.063 millimeter, mm) of Puyallup River sediment—which originates from Mount Rainier, a Cascade volcano—from glacial fine sediment in lowland bluffs that supply sediment to beaches. In combination with activities of beryllium-7 ( $^7\text{Be}$ ), a short-lived radionuclide, geochemical signatures showed that winter 2013–14 sediment runoff from the Puyallup River was transported to and deposited along the north shore of Commencement Bay, then mixed downward into the sediment column. The three Commencement Bay sites at which organic contaminants were measured in surface sediment did not have measurable  $^7\text{Be}$  activities in that layer, so their contaminant assemblages were attributed to sources from previous years. Concentrations of organic contaminants (the most common of which were polycyclic aromatic hydrocarbons, polychlorinated biphenyls, and fecal sterols) were higher in the <0.063-mm fraction compared to the <2-mm fraction, in winter compared to summer, in river suspended sediment compared to river bar and bank sediment, and in marine sediment compared to river sediment. The geochemical property barium/aluminum (Ba/Al) showed that the median percentage of Puyallup River derived fine surface sediment along the shoreline of Commencement Bay was 77 percent. This finding, in combination with higher concentrations of organic contaminants in marine rather than river sediment, indicates that riverborne sediment-bound contaminants are retained in shallow marine habitats of Commencement Bay. The retention of earlier inputs complicates efforts to identify recent inputs and sources. Understanding modern sources and fates of riverborne sediment and contaminants and their potential ecological impacts will therefore require a suite of targeted geochemical studies in such marine depositional environments.

## Introduction

Urbanized coastal watersheds discharge land-derived sediment and contaminants to the coastal ocean from point sources (ditches, outfalls) and nonpoint sources (overland runoff, atmospheric deposition). Particle-bound contaminants can persist for decades to centuries in the

marine environment, can be transported long distances, and can affect organisms that live in or on the seabed or that ingest sediment while feeding. Thus, understanding the transport and deposition of riverborne sediment and contaminants in the nearshore region is the first step in understanding the risks these materials pose to nearshore organisms, their community structure, and food webs. Geochemical properties of sediment, such as elemental ratios and short-lived radionuclide activities, can distinguish sediments from different geologic sources (Prego and others, 2012) and land-use activities (Matisoff and others, 2002; Fernandez and others, 2006). Once riverborne sediment enters the coastal zone, geochemical tracers can provide insights about dispersal patterns and the recency of deposition (Sommerfield and others, 1999; Fernandez and others, 2006; Prego and others, 2009).

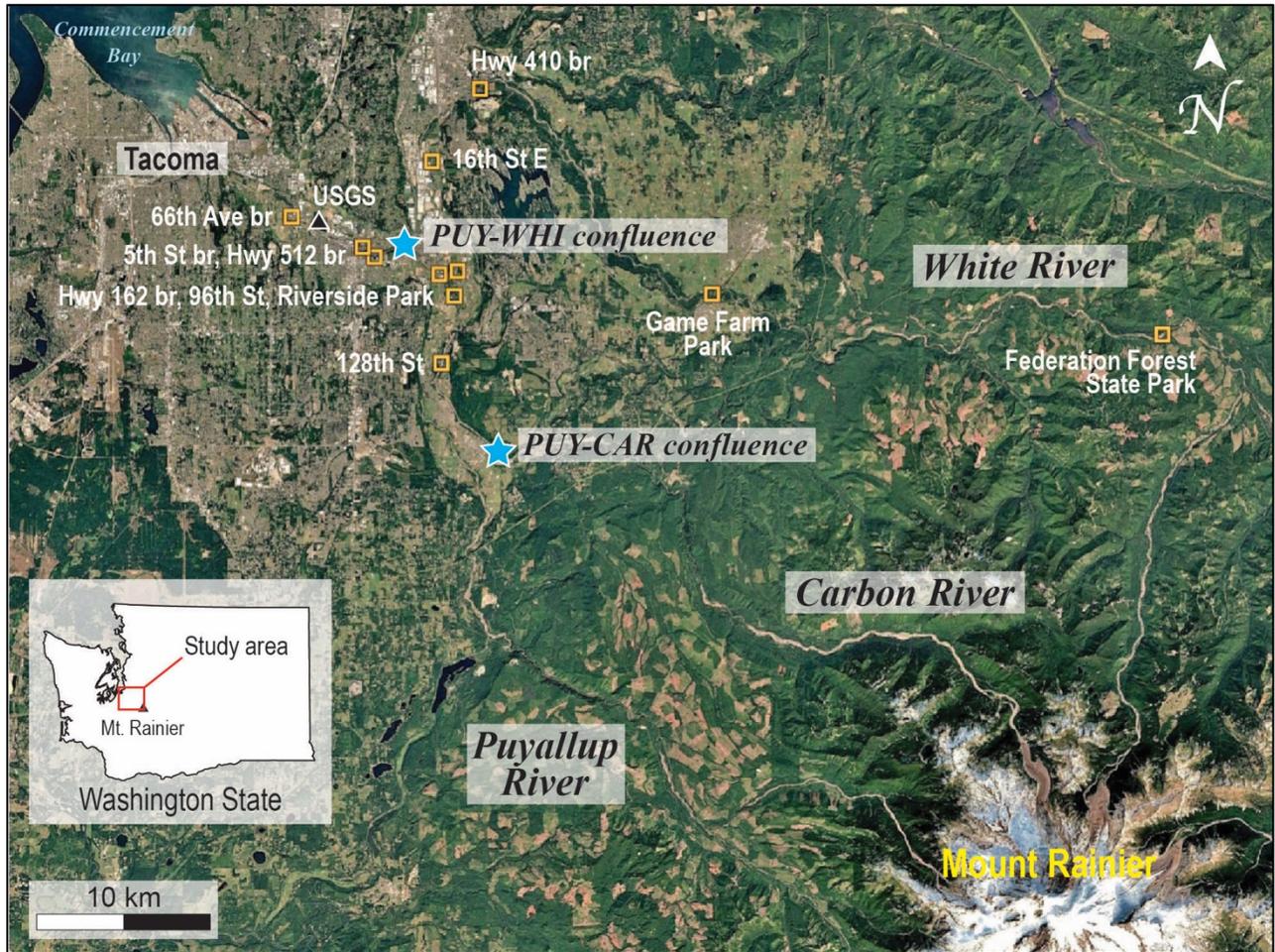
This report describes a study with goals to (1) identify sediment-geochemical signatures that distinguish riverborne fine sediment from the Puyallup River, Washington, from nearshore sediment, (2) determine how fine sediment from the Puyallup River was dispersed in shallow areas of Commencement Bay in Puget Sound following a winter storm, and (3) quantify current-use and legacy urban contaminants associated with Puyallup River sediment in shallow areas that could be used by beach associated forage fish such as sand lance (*Ammodytes personatus*) and surf smelt (*Hypomesus pretiosus*). Forage fish eggs and/or adult fish were found in unconsolidated sediment at eight beaches in and near Commencement Bay during a qualitative site reconnaissance in January 2014. We also report concentrations of (so-called) anthropogenic metals (cadmium, Cd; copper, Cu; lead, Pb, and zinc, Zn) and arsenic (As) in nearshore fine-sediment. Understanding about the fate of riverborne sediment and contaminants in Commencement Bay can identify nearshore areas where nearshore organisms could be at risk from contaminants. Ecological impacts of contaminants are a growing concern (Schwarzenbach and others, 2006) as coastal development increases around Puget Sound (Washington Office of Financial Management, 2016) and worldwide (Mee, 2012).

## Site Description

Puget Sound watersheds fall into two general categories—large river basins with headwaters in the Cascade or Olympic Ranges or local stream drainages. Because the geology of the Cascades and the lowlands differ, so too will the compositions of clastic sediment originating from these source areas. Cascade volcanoes consist of recent intermediate (andesitic) lava flows and tephras overlying intermediate to mafic (basaltic) continental arc basement rocks (Condie and Swenson, 1973; Smith and Leeman, 1993; Sisson and Vallance, 2009). The Puget Lowlands consist of glacial deposits whose sediment was derived from felsic (granitic) batholiths in western British Columbia (Jones, 1999; Crawford and others, 2005). Expected geochemical differences between volcanic upland sediment and glacial lowland sediment form the basis of using sediment-geochemical signatures as a means of discriminating riverborne Puyallup sediment from other sediment in Commencement Bay.

The sediment-geochemical study was undertaken in the Puyallup River watershed in western Washington, and its coastal outlet Commencement Bay in South Puget Sound (fig. 1). The Puyallup River has headwaters in alpine glaciers of Mount Rainier, an active Cascade stratovolcano approximately 65 kilometers (km) (40 miles) from the City of Tacoma (Sisson and Vallance, 2009). The Puyallup River has two main tributaries, the White and Carbon Rivers (fig. 1), and in combination the three rivers drain an area of approximately 2,700 square kilometers (km<sup>2</sup>) (Czuba and others, 2010). The geology of the Puyallup watershed uplands consists of

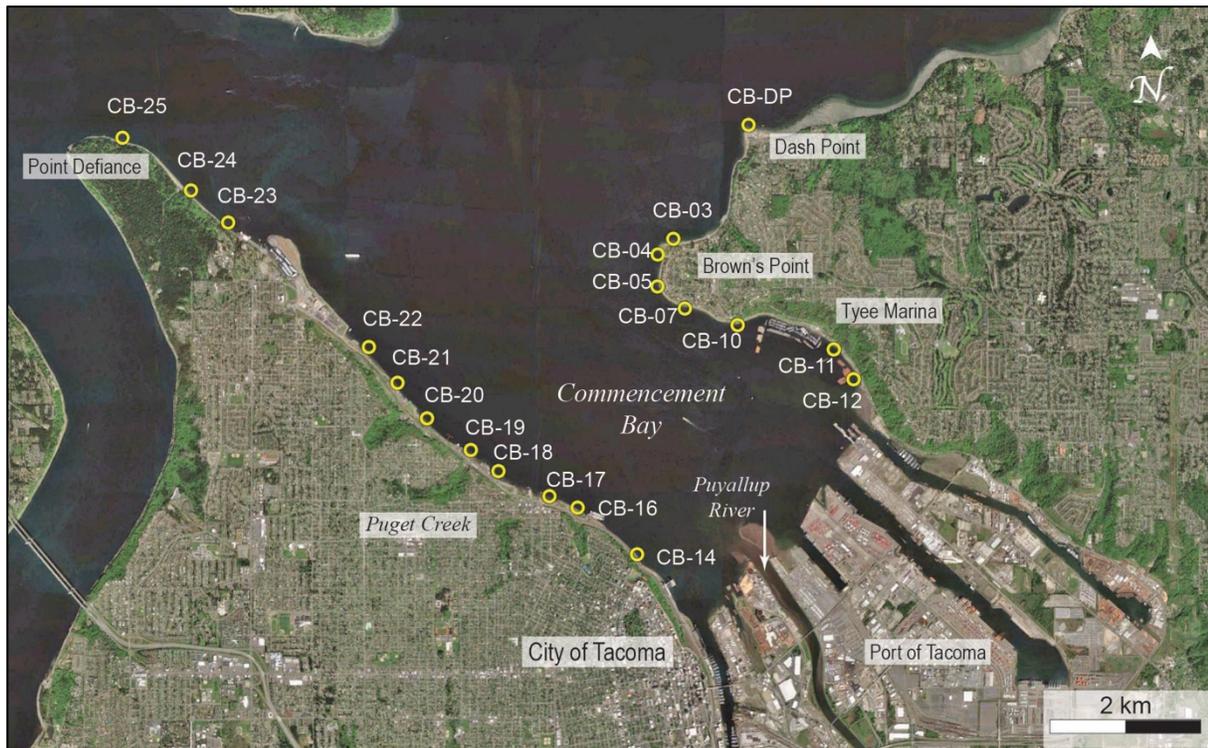
granodiorite basement rocks and Mount Rainier andesite and basaltic-andesite lava flows (Fiske and others, 1964).



**Figure 1.** Annotated Google Earth satellite image of the Puyallup River, Washington, and its tributaries. Orange squares show river sediment collection sites, blue stars show the confluences of major tributaries, and the black triangle shows U.S. Geological Survey gaging station 12101500. St, Street; br, bridge; PUY, Puyallup River; WHI, White River; CAR, Carbon River.

Owing to the volcanic headwaters and the steepness of the watershed (from 4,392 meters, m, elevation at Mount Rainier to sea level within a distance of 65 km), the Puyallup River transports large amounts of sediment to the lowlands and Puget Sound. The Puyallup River contributes the third largest annual river sediment load to Puget Sound, an estimated 1,000,000 tons per year (Czuba and others, 2010). Peak discharge of the Puyallup River occurs in winter during storm events, and there is a secondary discharge peak in summer associated with melting snowpack of Mount Rainier glaciers (Czuba and others, 2012). Agricultural, residential, urban, and industrial activities occur in the lower Puyallup River watershed. Runoff in winter is associated with rainfall and consists of sediment and contaminants from the lowlands; runoff in summer is associated with meltwater from glaciers on Mount Rainier and includes glacial flour and contaminants adsorbed during transport.

Commencement Bay is an approximately 20-km<sup>2</sup>, 165-m-deep urban embayment (Ebbesmeyer and others, 1986) at the mouth of the Puyallup River (fig. 2). Tidal flats of the Puyallup River delta were channelized and filled to accommodate railroad, lumber, and shipping industries in the early 1900s and now comprise the Port of Tacoma. A large percentage of the shoreline of Commencement Bay has been armored with rip-rap, eliminating intertidal beach habitat. Cannon and Grigsby (1982) observed the Puyallup River plume exiting Commencement Bay along the northeast shore during ebb tide, whereas near-bottom return flow was along the southwest shore. Ebbesmeyer and others (1986) calculated a water residence time of approximately 15 days for Commencement Bay.



**Figure 2.** Annotated Google Earth satellite image of Commencement Bay in Puget Sound, Washington, and vicinity showing 2013–14 sediment collection sites (yellow circles).

## Methods

### River Discharge and Turbidity

Time series of daily river discharge and turbidity were obtained from U.S. Geological Survey (USGS) gaging station 12101500 on the Puyallup River at Puyallup, (see [https://waterdata.usgs.gov/nwis/uv?site\\_no=12101500](https://waterdata.usgs.gov/nwis/uv?site_no=12101500), accessed May 10, 2017). Turbidity was measured with an infrared optical sidescatter nephelometer (Forest Technology Systems, Digital Turbidity Sensor-12) and reported in Formazin Nephelometric Units (FNU).

### Sediment Collection

River bar and river bank sediment was collected for determination of sediment geochemical signatures and contaminants in the Puyallup and White Rivers (fig. 1) on January 20–21, 2013, August 19–21, 2013, and January 29, 2014 (table 1). Sampling sites on river bars were targeted to areas where fine-grained sediment had accumulated. Particles >2 millimeters (mm) in diameter were excluded during sampling. River-bank sediment was collected at the waterline. Sampling was carried out for elemental analyses with a polypropylene scoop and wire-top plastic bags and for organic contaminant analyses with a Teflon™ spatula and amber glass jars. All samples were stored frozen.

River suspended sediment was collected for contaminant analysis on five occasions between October 2012 and August 2013 from the 66th Avenue bridge on the lower Puyallup River, including during two autumn/winter storm events and three summer glacial melt events. Suspended sediment was collected by pumping large volumes of water from the river through a continuous-flow centrifuge (CFC Express, Scientific Methods, Inc.). The centrifuge captures sediment in the centrifuge bowl and continuously discharges low-sediment water back to the river, as described in Conn and others (2016).

**Table 1.** Location, collection date, and types of analyses performed on specified sediment size fractions from the Puyallup River watershed and Commencement Bay nearshore sites, Puget Sound, Washington. [GEO, geochemistry; RAD, radionuclides; CB, contaminants in bank or bar sediment; CS, contaminants in suspended sediment; GS, grain size; OC, organic carbon content. Date format is Month/Day/Year. mm, millimeter]

Site	Latitude	Longitude	Date	GEO	RAD	CB	CS	GS	OC
Puyallup River sediment									
66th Avenue bridge	47.21378	-122.34159	10/31/12				bulk		
			01/09/13			bulk			
			01/20/13	<0.063 mm					
			06/21/13			bulk			
			07/01/13			bulk			
			08/06/13			bulk			
			08/21/13	<0.063 mm	<2 mm, <0.063 mm		bulk	bulk, <2 mm, <0.063 mm	
01/29/14	<0.063 mm	<2 mm		bulk	bulk				
5th Street bridge	47.19812	-122.28708	01/20/13	<0.063 mm					
Hwy 512 bridge	47.19321	-122.28090	01/20/13	<0.063 mm					
Hwy 162 bridge	47.18479	-122.22899	08/21/13	<0.063 mm					
			01/21/13	<0.063 mm					
			08/21/13	<0.063 mm					
01/29/14	<0.063 mm	<2 mm		bulk	bulk				
96th Street	47.17334	-122.21849	01/21/13	<0.063 mm					
Riverside Park	47.18634	-122.21563	01/29/14	<0.063 mm			bulk	bulk	
128th Street	47.13927	-122.22780	01/21/13	<0.063 mm					
			08/21/13	<0.063 mm					
White River sediment									
16th Street East	47.24289	-122.23514	1/21/13	<0.063 mm					
			1/29/14	<0.063 mm	<2 mm		bulk	bulk	
Hwy 410 bridge	47.17408	-122.02303	8/21/13	<0.063 mm					
			1/29/14	<0.063 mm			bulk	bulk	
Game Farm Park	47.27991	-122.19853	1/21/13	<0.063 mm					
			8/21/13	<0.063 mm					
			1/29/14	<0.063 mm			bulk	bulk	
Federation Forest	47.15200	-121.68239	8/21/13	<0.063 mm					
Commencement Bay and Nearshore sediment									
CB-Dash Point	47.32032	-122.42918	1/28/14	<0.063 mm				bulk	bulk
CB-03	47.30684	-122.44231	1/28/14	<0.063 mm				bulk	bulk
CB-04 deep	47.30498	-122.44503	1/28/14	<0.063 mm	bulk			bulk	bulk
CB-04 shallow	47.30485	-122.44471	1/28/14	<0.063 mm	bulk	<0.063 mm		bulk	bulk
CB-05.5	47.30121	-122.44508	1/28/14	<0.063 mm				bulk	bulk
CB-07	47.29859	-122.44030	1/28/14	<0.063 mm				bulk	bulk
CB-10	47.29662	-122.43109	1/28/14	<0.063 mm				bulk	bulk
CB-11	47.29374	-122.41436	1/28/14	<0.063 mm	bulk			bulk	bulk
CB-12.5	47.29021	-122.41090	1/28/14	<0.063 mm	bulk	<0.063 mm		bulk	bulk
CB-14	47.26951	-122.44864	1/28/14	<0.063 mm				bulk	bulk
CB-16	47.27504	-122.45899	1/28/14	<0.063 mm				bulk	bulk
CB-17	47.27639	-122.46391	1/28/14	<0.063 mm		<0.063 mm		bulk	bulk
CB-18	47.27936	-122.47279	1/28/14	<0.063 mm	bulk			bulk	bulk

Site	Latitude	Longitude	Date	GEO	RAD	CB	CS	GS	OC
CB-19	47.28186	-122.47761	1/28/14	<0.063 mm				bulk	bulk
CB-20	47.28560	-122.48524	1/28/14	<0.063 mm				bulk	bulk
CB-21	47.28981	-122.49039	1/28/14	<0.063 mm				bulk	bulk
CB-22	47.29404	-122.49538	1/28/14	<0.063 mm				bulk	bulk
CB-23	47.30876	-122.51992	1/28/14	<0.063 mm				bulk	bulk
CB-25	47.31879	-122.53837	1/28/14	<0.063 mm				bulk	bulk
Point Defiance	47.31473	-122.53002	8/19/13	<0.063 mm				bulk	bulk
Dockton	47.37151	-122.45670	8/20/13	<0.063 mm		<2 mm, <0.063 mm		bulk	bulk, <2 mm, <0.063 mm
Glacial sediment									
Point Bolin, bluff	47.69066	-122.59193	6/25/13	<0.063 mm				bulk	
Point Bolin, slide	47.69646	-122.60348	6/25/13	<0.063 mm				bulk	
Point Bolin, bluff	47.69646	-122.60348	6/25/13	<0.063 mm				bulk	
Point Defiance, clay	47.31761	-122.53459	8/19/13	<0.063 mm				bulk	

Nearshore sediment was collected at 18 sites along the shoreline of Commencement Bay (fig. 2, table 1), including Browns Point, on January 28, 2014, shortly after a winter storm. A stainless steel van Veen-type benthic sampler was deployed from a small boat to collect bottom sediment close to the shore. Water depth from the vessel's depth finder and the predicted tidal height indicated that bottom sediment was collected at depths ranging from -1.0 m relative to mean lower low water (MLLW) to -15.7 m MLLW. The median sampling depth was -3.1 m MLLW. The upper 0–2 centimeters (cm) of the intact sediment surface was subsampled for geochemistry and contaminants from the material in the benthic sampler. Sediment for radionuclide analyses was collected at five sites whose sediment appeared muddy at the time of sampling—CB-04 (shallow), CB-04 (deep), CB-11, CB-12.5, and CB-18 (fig. 2) by emplacing a 3.5-cm-diameter polycarbonate core barrel in the benthic sampler to recover a short sediment column (as long as 12 cm). A reconnaissance sample of nearshore sediment was also collected on August 20, 2013, in Quartermaster Harbor (not shown), a nearby Puget Sound embayment that receives sediment from the Puyallup River plume.

Glacial sediment was collected from three subaerial bluffs above the high-tide line on the west side of Point Bolin, Kitsap County, and a clay layer exposed in the intertidal beach face at Point Defiance, Pierce County (table 1).

## Sediment Analyses

### Grain-Size Distribution and Organic-Carbon Content

Sediment grain-size distributions (GS) were determined at the USGS Pacific Coastal and Marine Science Center (PCMSC) sediment laboratory in Santa Cruz, California, on organic- and salt-free sediment using a combination of techniques. Particles coarser than gravel (>2 mm) were quantified by dry sieving at quarter-phi intervals (size in millimeters= $2^{-\text{phi}}$ ). Sand- and mud-sized particles (2–0.063 mm and <0.063 mm, respectively) were separated by wet sieving and quantified using a laser particle diffraction counter (Beckman Coulter Life Sciences). GS parameters were calculated with in-house statistical software (pcSDSZ) according to the methods

of Folk and Ward (1957). Total carbon (TC) and total inorganic carbon (TIC) were determined coulometrically (UIC, Inc.) in the surface 0–2 cm layer at each site. Total organic carbon (TOC) was calculated as the difference between TC and TIC. Grain size and organic carbon results are tabulated in appendix 1.

## Elemental Composition

A subsample of bulk sediment was oven-dried at 60 degrees Celsius (°C), disaggregated in a mortar and pestle, and dry-sieved in stainless steel sieves to obtain 2 grams (g) of the <0.063-mm fine fraction (silt and clay). The sediment fine fraction was sent to SGS, Inc., a nationally recognized testing laboratory, for quantification of major, minor, and trace elements, including anthropogenic metals and rare earth elements (REEs), using a sodium peroxide sinter followed by quantification by inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma atomic emission spectrometry (ICP-AES). The reproducibility of three replicate samples submitted to SGS, Inc., was better than 10 percent for all target elements except the heavy REE thulium (Tm), which had a reproducibility of 12 percent. A second procedure that targeted trace elements used a four-acid near-total sediment decomposition followed by quantification by ICP-MS (Briggs and Meier, 2002). Neither analytical method quantified silica (Si) contents. Major element contents are reported in units of percent (%) and minor and trace element contents as parts per million (ppm), which is equivalent to micrograms per gram (µg/g). Total element contents were normalized to Al to account for differences in grain size. REE contents were normalized to a North American shale composite (NASC), which represents the composition of average sedimentary rock (McLennan, 1989). Normalized values were used to calculate REE ratios. Contents of anthropogenic elements in a glacial clay bed in Commencement Bay were used to define natural geologic levels, or backgrounds, of these elements because there were no anthropogenic influences on their contents at the time of deposition. Elemental compositions are tabulated in appendix 2.

## Short-Lived Radionuclide Activities

Sediment cores as long as 12 cm were sectioned into 2-cm-thick intervals 1 day after collection and refrigerated. At the PCMSC radionuclide laboratory, sediment was oven-dried (60 °C), disaggregated, packed into 10-milliliter (mL) vials, and counted on an ultra-low-background, high-purity germanium well detector for 24 hours. Lead-210 ( $^{210}\text{Pb}$ ) was measured at 46.5 kiloelectronvolts (keV); radium-226 ( $^{226}\text{Ra}$ ) was measured at 351.87 (lead-214,  $^{214}\text{Pb}$ ) and 609.31 (bismuth-214,  $^{214}\text{Bi}$ ) keV; cesium-137 ( $^{137}\text{Cs}$ ) was measured at 661 keV; and  $^7\text{Be}$  was measured at 477.56 keV. Detectors were regularly calibrated with standards from the International Atomic Energy Association (IAEA)—IAEA-RGU-1, IAEA-RGTH-1, and IAEA 300 prepared in the same geometry as samples. Geologically unsupported, or excess,  $^{210}\text{Pb}$  (abbreviated as  $^{210}\text{Pb}_{\text{ex}}$ ) was calculated by subtracting supported  $^{210}\text{Pb}$  (from its parent  $^{226}\text{Ra}$ ) from the total  $^{210}\text{Pb}$  activity. Radionuclide activities are tabulated in appendix 3.

## Current-Use and Legacy Contaminants

Sediment samples were analyzed at the USGS National Water Quality Laboratory (NWQL) for a suite of organic contaminants including PAHs by NWQL schedule 5506 (Zaugg and others, 2006), waste indicator chemicals by NWQL schedule 5433 (Burkhardt and others, 2006), hormones by NWQL schedule 6434 modified from Foreman and others (2012), and

halogenated organic compounds including select PCB congeners and polybrominated diphenyl ether (PBDE) congeners by NWQL lab code 8093, a research method described in Wagner and others (2014). A complete list of analyzed parameters is in appendix 4. When a parameter was analyzed by multiple methods, results are reported from the most sensitive method. In this report, detections and maximum concentrations are reported when the concentration is above the long-term NWQL method detection limit and at least three times greater than the associated laboratory blank sample. Results are reported on a dry-weight basis. Contaminant data can be found at the USGS National Water Information System—Web Interface (see <http://dx.doi.org/10.5066/F7P55KJN>, accessed September 25, 2017).

## Sediment-Geochemical Sourcing and Aging

Immobile and relatively immobile elements, those that are minimally altered during sediment erosion and transport, form the most effective geochemical signatures because they reflect the source-rock compositions. These elements include the trace elements barium, cobalt, chromium, nickel, niobium, rubidium, scandium, thorium, and zirconium (Ba, Co, Cr, Ni, Nb, Rb, Sc, Th, Zr), as well as the REE and their ratios (McLennan and others, 1990). Elements that had statistically different mean values in river and glacial sediment were identified with two tailed Student's *t*-tests and *p*-values <0.01 using StatPlusPro software. Geochemical signatures in river sediment collected during January 2013 and 2014 (*n*=13; table 1) were used to define a river end-member representative of terrestrial sediment deposited in Commencement Bay in January 2014. Four samples from glacial deposits were used to define an end-member representative of Puget Lowland sediment. It was assumed that the inorganic fraction of nearshore sediment in Commencement Bay was primarily a mixture of material derived from riverine and glacial sources.

Activities of short-lived radionuclides  $^7\text{Be}$ ,  $^{137}\text{Cs}$ , and  $^{210}\text{Pb}$  in bulk sediment can indicate how recently terrestrial sediment was deposited in the coastal region.  $^7\text{Be}$ , with a 53.2-day half-life ( $t_{1/2}$ ), is primarily used as a runoff indicator to track sedimentation events that occurred as many as 5 months before sampling (Sommerfield and others, 1999).  $^{137}\text{Cs}$  ( $t_{1/2}$ =30.2 years) is a radionuclide produced by thermonuclear weapons testing between the mid-1950s and mid-1980s (Garcia-Agudo, 1998). Its presence indicates sediment deposited during that interval or mixtures of such sediment. Excess  $^{210}\text{Pb}$  ( $t_{1/2}$ =22.2 years) in the sediment column indicates the presence of sediment deposited within the past 100 years (Carpenter and others, 1985). Radionuclide activities were assessed in surface sediments (depth of 0–2 cm), the interval in which organic contaminants and anthropogenic metals were measured, to link nearshore contaminant assemblages with terrestrial runoff from recent winter storms. To account for potential vertical redistribution of radionuclides by mixing processes, radionuclide activities were also summed in the sediment mixed layer, here limited to a depth of 0–8 cm, the maximum core length recovered in four of five sediment cores. To account for differences in TOC contents among sites, radionuclide activities were normalized to the bulk sediment TOC content of the surface 0–2 cm layer at each site, the only interval in which TOC was determined. The sediment core at CB-04-D was only 4 cm long, so the radionuclide inventory calculated there is a minimum estimate of the total activity over an 8-cm-long sediment column.

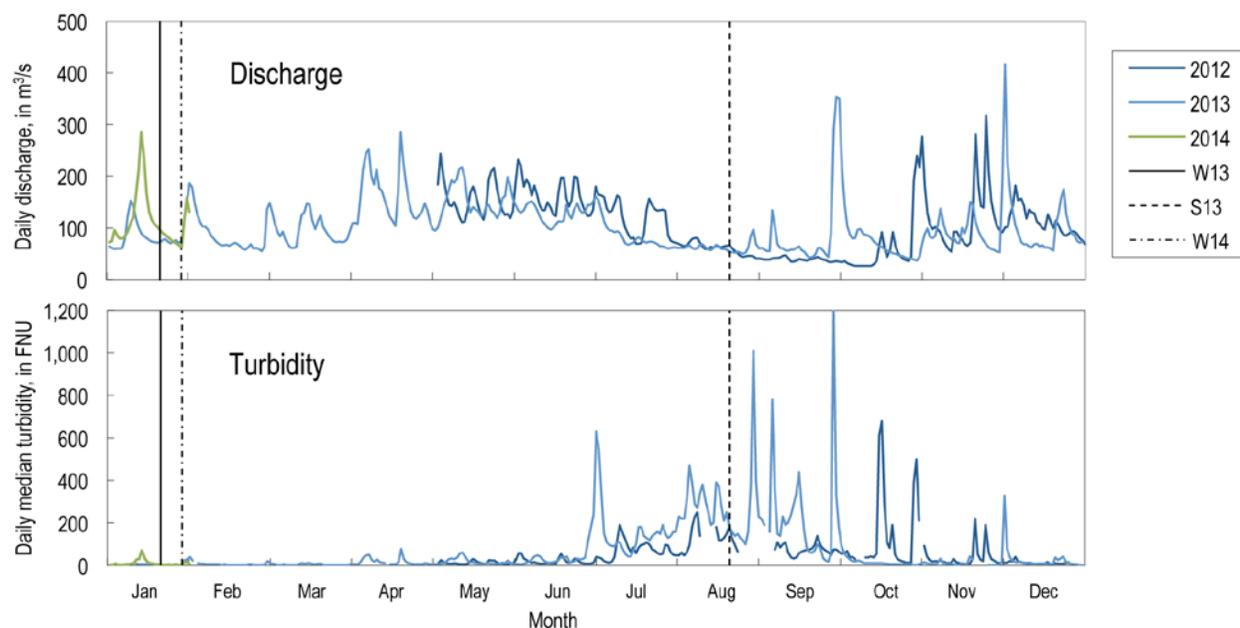
## Results

### River and Lowland Sediment

The resulting data for river and lowland sediment include river discharge and turbidity, elemental composition of river and lowland sediment, and organic contaminants in river sediment.

### River Discharge and Turbidity

Daily discharge of the Puyallup River at Puyallup, Washington (USGS gaging station 12101500), approximately 11 km upstream of the river mouth, was generally similar in January 2013 (winter 2013, W13) and 2014 (winter 2014, W14) in the months preceding sediment collection, whereas median daily turbidity was higher in W13 (fig. 3). Discharge and turbidity were seasonally out of phase: median turbidity levels were low in winter when discharge was high and vice versa in summer. Sediment sampling in the summer 2013 (S13) occurred during the period of glacial runoff (fig. 3).

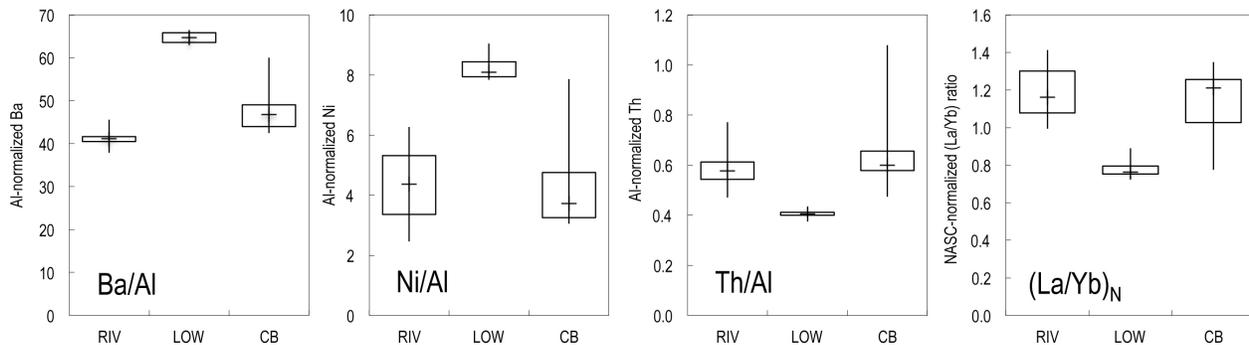


**Figure 3.** Graphs showing time series of median daily discharge in cubic meters per second ( $\text{m}^3/\text{s}$ ) and median turbidity values in Formazin Nephelometric Units (FNU) in the Puyallup River at Puyallup, Washington (U.S. Geological Survey gaging station 12101500). Vertical lines show sediment collection dates—January 20, 2013 (winter 2013, W13), August 21, 2013 (summer 2013, S13), and January 28, 2014 (winter 2014, W14).

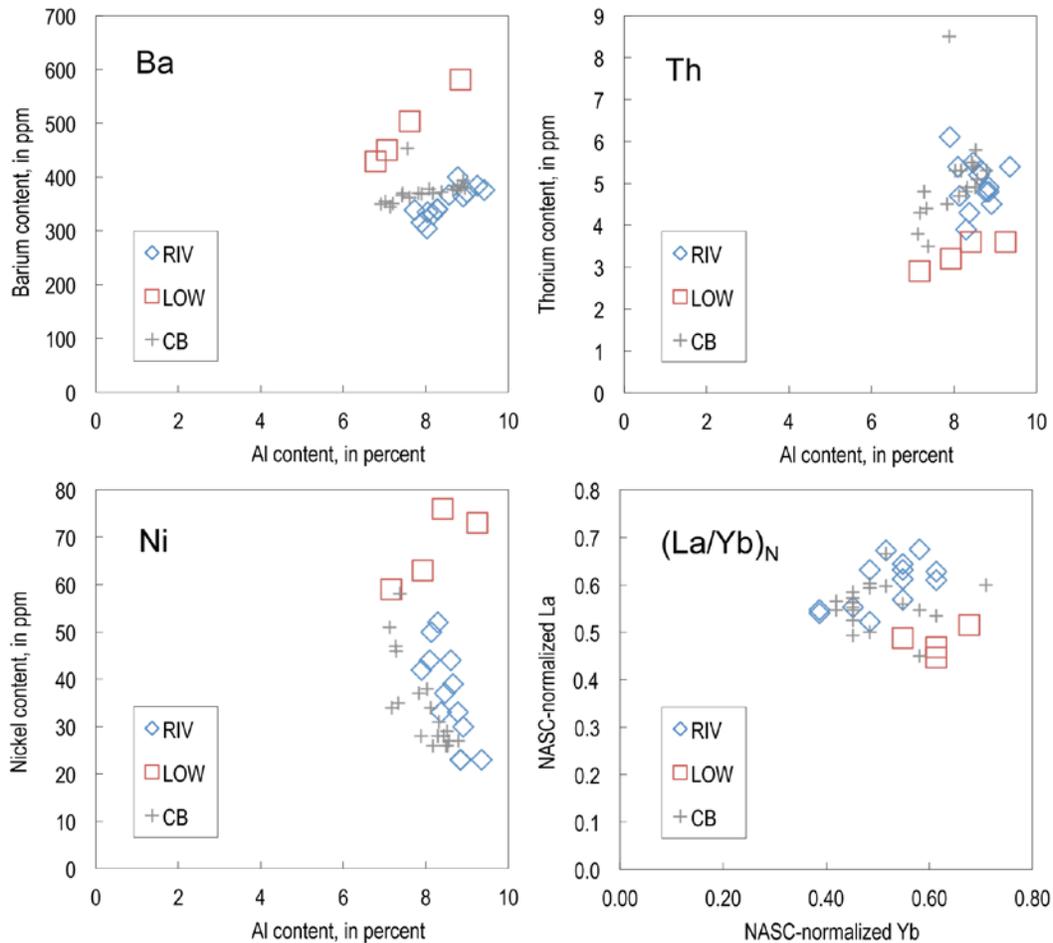
### Elemental Compositions of River and Lowland Sediment

Among the immobile and relatively immobile elements used to identify geochemical source signatures, four had significantly different means between winter river sediment (RIV, Puyallup and White Rivers) and Puget Lowland sediment (LOW, glacial bluffs and clay)—Al-normalized contents of Ba, Ni, and Th, and the NASC-normalized REE ratio lanthanum/ytterbium— $(\text{La}/\text{Yb})_N$ . Descriptive statistics for these parameters are summarized in

figure 4, and the raw data are shown in figure 5. Ba/Al had the lowest coefficients of variation (CV, defined as ratio of the standard deviation to the mean), in RIV (0.05) and LOW (0.02), indicating that sediment sources were very well constrained with this parameter. In comparison, the CVs were more than two times higher for Ni/Al (0.31, 0.06), Th/Al (0.14, 0.04), and  $(La/Yb)_N$  (0.12, 0.09) than for Ba/Al. Some of the geochemical variability was related to compositional differences between Puyallup and White River sediment and some to differences between the two years. Ba/Al values were normally distributed in the two end members (Shapiro-Wilk test,  $p=0.54$ ,  $\alpha=0.05$ ), so mean Ba/Al values of RIV and LOW were used as geochemical signatures [ $Ba/Al_{RIV}=41\pm 2$  (mean $\pm 1\sigma$ ),  $Ba/Al_{LOW}=65\pm 1$ ] to estimate fractions of river-derived sediment in the nearshore.



**Figure 4.** Box and whisker plots of aluminum (Al) normalized barium, nickel, and thorium (Ba, Ni, and Th) and the North American shale composite (NAS) normalized lanthanum/ytterbium  $[(La/Yb)_N]$  ratio in Puyallup and White River (RIV), Puget Lowland (LOW), and Commencement Bay (CB) fine sediment, Puget Sound region, Washington. Box heights show the interquartile ranges, whiskers show maxima and minima, and lines crossing whiskers show medians.



**Figure 5.** Scatter plots of barium, nickel, and thorium contents relative to aluminum (Al) contents and North American shale composite (NASC) normalized lanthanum (La) to normalized ytterbium (Yb) contents in winter fine sediment from the Puyallup and White Rivers (RIV), Puget Lowland (LOW), and Commencement Bay (CB), Puget Sound region, Washington. ppm, parts per million.

### Current-Use and Legacy Contaminants in River Sediment

Table 2 lists the 14 compounds that were detected in more than one river-bar or riverbank sediment or suspended-sediment sample. Two fecal sterols were the most frequently detected organic chemicals in river sediment—cholesterol (6/7 detections, concentrations ranging from an estimated 223 micrograms per kilogram,  $\mu\text{g}/\text{kg}$ , to 678  $\mu\text{g}/\text{kg}$ ) and  $\beta$ -sitosterol (6/6 detections, concentrations ranging from an estimated 1,520  $\mu\text{g}/\text{kg}$  to an estimated 2,350  $\mu\text{g}/\text{kg}$ ). In addition, the biocide degradate pentachloroanisole was detected in three winter samples. Other chemicals were detected singly and (or) sporadically.

Ten (of 157) organic chemicals were detected in one or more of the four samples for which the <2-mm fraction was analyzed—four PAHs, three waste indicators, two hormones, and one halogenated compound. Nine of the ten compounds were detected in river-bank sediment collected during winter 2013 at the Highway 162 bridge (Puyallup River upstream of the confluence with the White River). One to four chemicals were detected in the <2-mm fraction of river-bar and riverbank sediment collected from the White River in winter 2014 and the 66th Avenue bridge (lower Puyallup River below the White River confluence) in summer 2013 and

winter 2014. When a fine-sediment (<0.063 mm) subsample was analyzed and compared to the corresponding <2-mm fraction at the 66th Avenue bridge during the summer sampling, three additional compounds were detected at low concentrations—the hormone 17- $\alpha$ -estradiol and the flame retardants PBDE 47 and PBDE 99.

More organic chemicals were detected in river suspended sediment than in river-bar or riverbank sediment. Twenty-seven (of 147) organic chemicals were detected in one or more river suspended-sediment samples collected from the 66th Avenue bridge on the lower Puyallup River. This included 13 PAHs, 7 waste indicators, and 7 halogenated organic compounds (suspended sediment samples were not analyzed for hormones). Similar to the river-bar and riverbank sediment samples, more compounds were detected in the winter samples (12 to 14 compounds detected) than in the summer samples (1, 2, and 8 compounds detected).

**Table 2.** Chemicals detected in more than one Puyallup River, Washington, bar, bank, or suspended sediment sample.

[ $\mu\text{g}/\text{kg}$ , microgram per kilogram dry weight; E, estimated.]

Chemical	Maximum detected concentration ( $\mu\text{g}/\text{kg}$ )	Number of detects or samples	
		River-bar or riverbank sediment	River suspended sediment
Polycyclic aromatic hydrocarbons			
1,6-Dimethylnaphthalene	E15.2	2/5	2/5
1-Methylphenanthrene	17.1	1/5	2/5
2,6-Dimethylnaphthalene	18.8	1/5	3/5
Fluoranthene	34.1	0/5	2/5
Naphthalene	E5.9	0/5	2/5
Waste indicators and hormones			
4-Cumylphenol	71.9	0/3	2/3
Bisphenol A	E489	0/3	2/2
Cholesterol	E678	5/5	2/3
Indole	142	1/5	1/3
Phenol	E548	1/5	1/3
$\beta$ -sitosterol	E2350	3/3	2/2
Halo-organic compounds			
Pentachloroanisole	0.14	2/4	1/5
Polybrominated diphenyl ether 47	E0.93	1/4	1/5
Polybrominated diphenyl ether 99	0.16	1/5	1/5

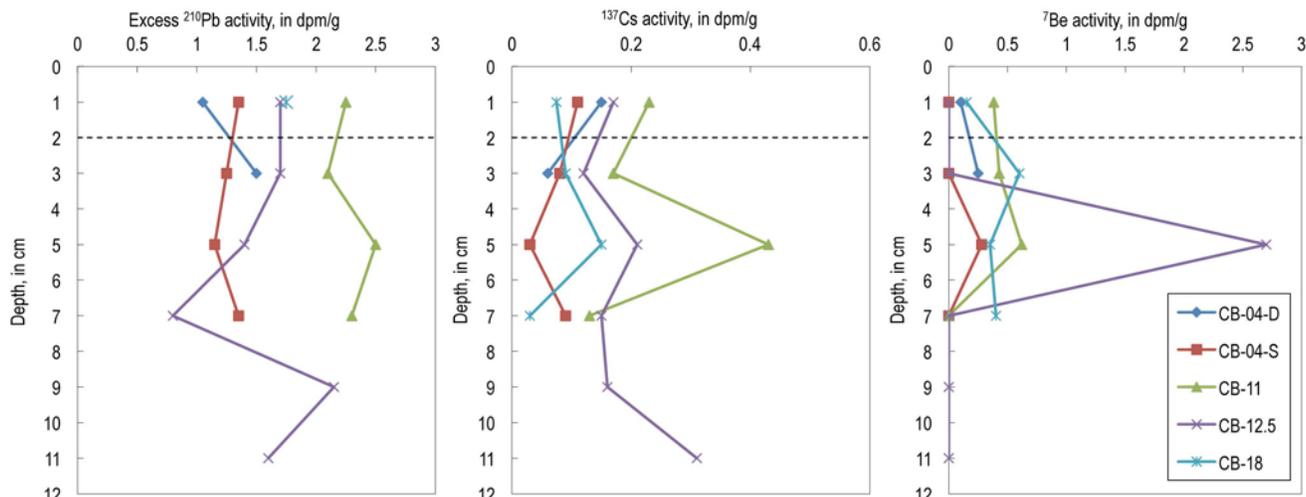
## Nearshore Sediment

The resulting data for nearshore sediment include short-lived radionuclide activities, elemental composition, organic contaminants, and anthropogenic metals and arsenic.

### Short-lived Radionuclide Activities in Nearshore Sediment

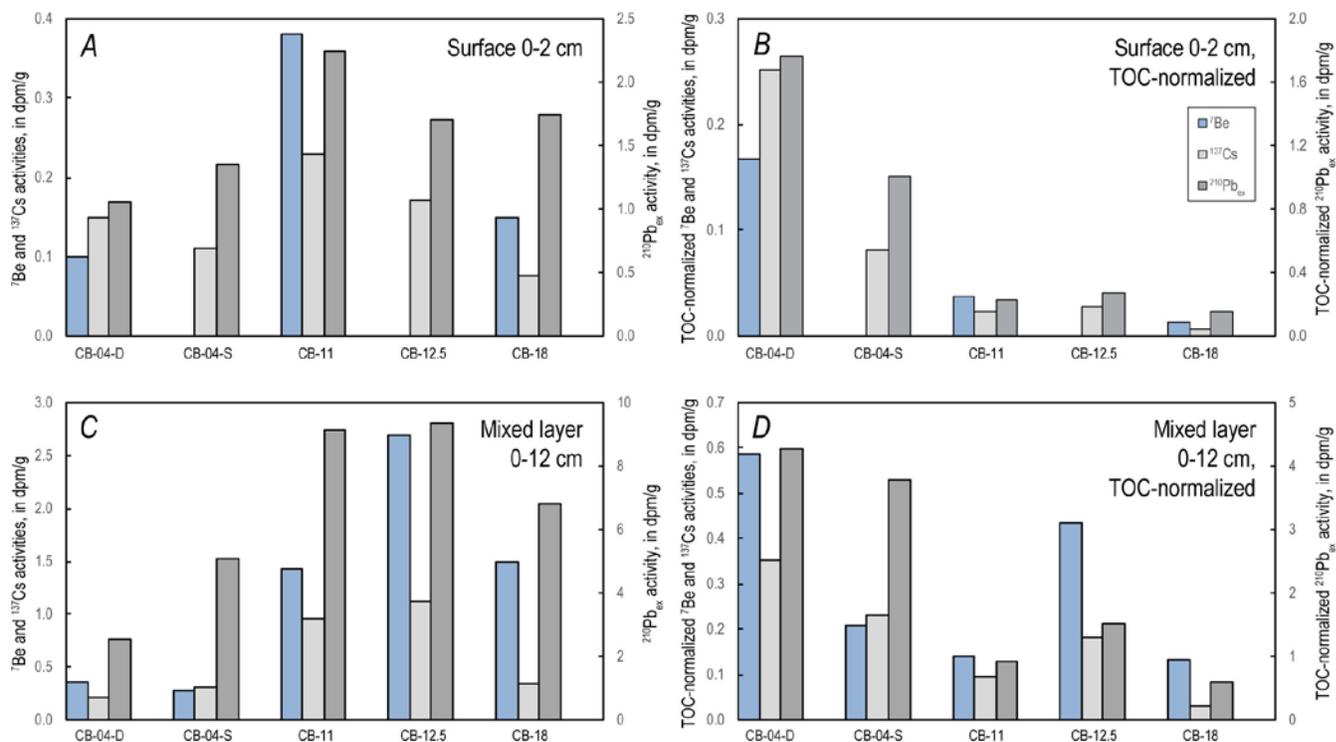
Profiles of excess  $^{210}\text{Pb}$  showed no trend with depth in sediment cores, indicating that the sediment column up to a depth of 12-cm was vertically mixed (fig. 6) and thus not useful for estimating  $^{210}\text{Pb}$ -based sediment accumulation rates.  $^{137}\text{Cs}$  activities in the sediment column were

low and akin to background levels measured by Davis and others (1997) in Commencement Bay (CB) waterways.



**Figure 6.** Graphs showing variations with depth of short-lived radionuclide activities in disintegrations per minute per gram of dry sediment (dpm/g) in sediment cores from Commencement Bay (CB), Puget Sound, Washington. Downcore values were measured in 2 centimeter (cm) thick sections and are plotted at midpoints of intervals. Dashed lines indicate the bottom of the surface layer of sediment.

Three sites had surface sediment with measurable <sup>7</sup>Be activities—CB-04-D (–15.7 m MLLW) at Browns Point, CB-11 (–2.7 m MLLW) on the north shore near Tye Marina, and CB-18 (likely –5 m MLLW) offshore of Puget Creek on the south shore (fig. 7A). Sites CB-04-S (–7.6 m MLLW) and CB-12.5 (–2.6 m MLLW, adjacent to a floating log corral) had no measurable <sup>7</sup>Be in the surface layer. Among these sites, the bulk-sediment TOC varied from 0.6 percent at CB-04-D to 11.5 percent at CB-18. TOC-normalized radionuclide activities were highest at CB-04-D (Browns Point) (fig. 7B), suggesting that this site experienced the most recent sediment deposition. Normalized radionuclide activities increased from the inner bay to the outer bay along the north shore (fig. 7B), similar to the manner in which the Puyallup River runoff plume is often observed to flow. There are no local drainages (<sup>7</sup>Be sources) along the north shore of Commencement Bay between CB-04 and CB-12.5 suggesting that <sup>7</sup>Be in surface sediment at CB-04-D and CB-11 was from recent Puyallup River outflow and deposition. Measurable <sup>7</sup>Be-containing sediment at CB-18 may have originated in whole or in part from Puget Creek, a local drainage on the south shore of the bay.

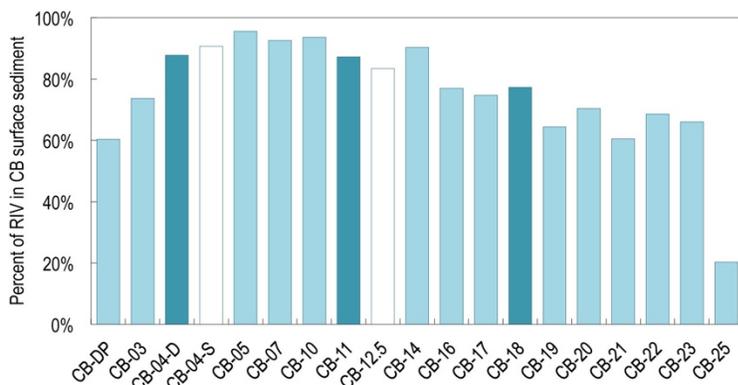


**Figure 7.** Graphs showing short-lived radionuclide activities in (A) bulk surface layer sediment, (B) surface layer sediment normalized by the total organic carbon content (TOC), (C) in the mixed layer, and (D) in the mixed layer normalized by TOC at five sites in Commencement Bay (CB), Puget Sound, Washington.  $^7\text{Be}$ , beryllium-7;  $^{137}\text{Cs}$ , cesium-137;  $^{210}\text{Pb}_{\text{ex}}$ , excess lead-210; dpm/g, disintegrations per minute per gram of dry sediment; cm, centimeter.

TOC-normalized radionuclide inventories in the mixed-layer were generally similar to surface patterns, having highest values at Browns Point (CB-04-D) and lowest values offshore of Puget Creek. The mixed layer at CB-12.5 (fig. 7D) had a high  $^7\text{Be}$  inventory and could reflect recent runoff from nearby Hylebos Creek.

### Elemental Composition of Nearshore Sediment

Based on the Ba/Al geochemical signature, nearshore fine sediment in Commencement Bay was more similar in composition to Puyallup River sediment than to Puget Lowland sediment. Assuming that the nearshore sediment sampled in this study was primarily a mixture of material from RIV and LOW, then the fraction of RIV in surface (0–2 cm) sediment in Commencement Bay calculated using a linear two end-member mixing equation ranged from 20–96 percent (fig. 8) and the median value was 77 percent. Surface sediment on the north shore of Commencement Bay (CB-03 to CB-12.5) generally had a greater fraction of riverborne sediment than that on the south shore of the bay (CB-14 to CB-23).



**Figure 8.** Plot showing the fraction of Puyallup and White River sediment (RIV) in surface fine sediment from Commencement Bay (CB), Puget Sound, Washington, on January 28, 2014, calculated from two end-member mixing. Dark blue columns show samples with measurable beryllium-7 ( $^7\text{Be}$ ); white columns show samples with no detectable  $^7\text{Be}$ . Samples shown by medium blue columns were not analyzed for radionuclides. Locations of sampling sites are shown in figure 2. %, percent.

### Current-Use and Legacy Contaminants in Nearshore Sediment

Nearshore sediment was sieved for contaminant analysis of the fine-sediment fraction (<0.063 mm) based on the low number and concentrations of chemicals detected on bulk river sediment, the increased number and concentrations of chemicals detected on suspended sediment, which has a finer grain-size distribution, and the knowledge that many organic chemicals preferentially sorb to finer rather than coarser sediment (Karickhoff and others, 1979). Sixty-one (of 157) organic chemicals were detected in winter nearshore fine sediment. This included 27 PAHs, 3 waste indicators, 7 hormones, and 24 halo-organic compounds. Table 3 lists the 36 compounds that were detected in more than one nearshore fine-sediment sample. PAHs were regularly detected in all three nearshore sediment samples; the lowest concentration for all but three individual PAHs was measured at CB-12.5 as compared to CB-04-S and CB-17. Among these three sites, CB-04-S and CB-12.5 were analyzed for short-lived radionuclides and contained no detectable  $^7\text{Be}$  activity in the surface 0–2 cm layer, an indication that organic contaminants in surface sediment were deposited more than 5 months before sample collection in January 2014. Measurable  $^7\text{Be}$  was found below the surface layer at CB-12.5 showing reworking of recently deposited sediment; however, contaminant assemblages were not measured in the corresponding subsurface layer. Four waste indicators and hormones were detected in all three nearshore samples, including  $\beta$ -sitosterol, indole, 3- $\beta$ -coprostanol and cholesterol. A few legacy chlorinated pesticides were detected at low concentrations, including cis- and trans-chlordane, dieldrin, trans-nonachlor, and hexachlorobenzene. Twelve (of 18) PCB congeners were detected; the summed PCB concentration ranged from 1.83 to 2.77  $\mu\text{g}/\text{kg}$ , which when normalized to the median TOC content were well below the Washington State marine-sediment quality standard of 12 mg/kg organic carbon (Washington Department of Ecology, 2013). Seven (of 10) PBDE congeners were detected. CB-04-S had a summed PBDE concentration of 9.87  $\mu\text{g}/\text{kg}$ , representing 6 congeners. In contrast, the summed PBDE concentration was 1.2  $\mu\text{g}/\text{kg}$  and 0.35  $\mu\text{g}/\text{kg}$  at CB-12.5 and CB-17, respectively, each representing the sum of 2 congeners. Washington State does not publish sediment quality standards for PBDEs.

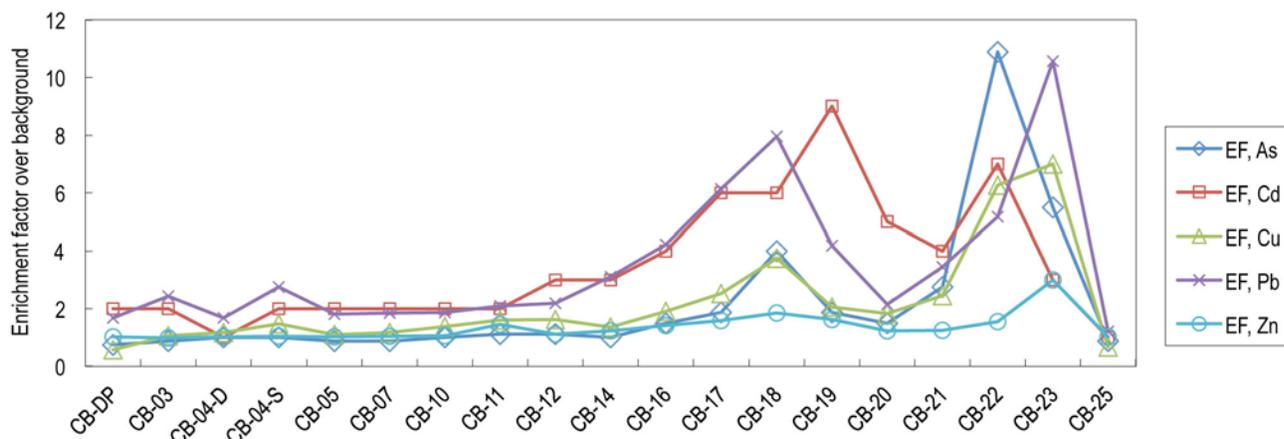
**Table 3.** Chemicals detected in more than one Commencement Bay, Puget Sound, Washington, fine-sediment sample.

[ $\mu\text{g}/\text{kg}$ , microgram per kilogram dry weight; PBDEs, polybrominated diphenyl ethers; PCBs, polychlorinated biphenyls; E, estimated.]

Chemical	Maximum detected concentration ( $\mu\text{g}/\text{kg}$ )	Number of detects or samples
Polycyclic aromatic hydrocarbons		
1,6-dimethylnaphthalene	53	3/3
1-methyl-9h-fluorene	21	2/3
1-methylphenanthrene	79.9	3/3
1-methylpyrene	45.4	2/3
2,3,6-trimethylnaphthalene	25.1	2/3
2,6-dimethylnaphthalene	149	3/3
4,5-methylenephenanthrene	74.2	3/3
9h-fluorene	83.7	2/3
Acenaphthene	47.6	2/3
Acenaphthylene	57	2/3
Anthracene	145	3/3
Anthraquinone	134	3/3
Benz[a]anthracene	194	3/3
Benzo[a]pyrene	150	3/3
Benzo[b]fluoranthene	E246	3/3
Benzo[e]pyrene	126	3/3
Benzo[g,h,i]perylene	E54.7	3/3
Benzo[k]fluoranthene	E85.7	3/3
Carbazole	36.8	3/3
Chrysene	269	3/3
Fluoranthene	748	3/3
Indeno[1,2,3-cd]pyrene	E48.9	3/3
Naphthalene	259	3/3
Perylene	54.2	3/3
Phenanthrene	416	2/3
Pyrene	724	3/3
Waste indicators and hormones		
3- $\beta$ -coprostanol	E227	3/3
4-androstene-3,17-dione	0.91	2/3
Cholesterol	2765	3/3
Indole	565	3/3
$\beta$ -sitosterol	E6580	3/3
Halo-organic compounds		
Chlordane, cis	0.15	3/3
Chlordane, trans	0.13	2/3
Hexachlorobenzene (HCB)	E2.2	3/3
PBDEs (sum of 10)	9.87	3/3
PCBs (sum of 18)	2.77	3/3

## Anthropogenic Metals and Arsenic in Nearshore Sediment

Nearshore fine-sediment contents of As, Cd, Cu, Pb, and Zn in Commencement Bay were enriched as much as ten-fold relative to geologic background levels in glacial clay (fig. 9), likely a legacy of ore smelting operations on the south shore of the bay and subsequent reuse of smelter slag as fill material along the south shore and in the industrial area at the head of the bay. Arsenic and Pb had maximum enrichment factors (EF) more than ten times higher than background; however, the median EF for As was approximately 1 ( $EF_{As}=1.1$ , same as background), whereas that for Pb was  $EF_{Pb}=2.4$ . Arsenic was the only element whose content in Commencement Bay fine sediment exceeded the effects range median (ERM), the level determined by the National Oceanic and Atmospheric Administration (NOAA) at which adverse biological effects frequently occur in marine and estuarine sediment (Long and others, 1995), and this was found at only one site ( $ERM_{As}=70$  ppm;  $CB-22_{As}=87$  ppm). At no sites were the State of Washington's sediment-quality standards exceeded for anthropogenic elements in marine-sediment impact zones affected by discharges, which are higher than the NOAA ERMs for As (by 133 percent), Cu (by 144 percent), Pb (by 243 percent), and Zn (by 234 percent) and lower than the NOAA ERM for Cd (by 30 percent) (Washington Department of Ecology, 2013). The median sedimentary contents of anthropogenic metals and As measured in shallow areas of Commencement Bay in January 2014 on the <0.063-mm fine fraction were within 23 percent of the values at 30 stations surveyed by the Washington Department of Ecology in Commencement Bay in June 2014 (after those values were adjusted for the median fine-sediment content) (Weakland and others, 2016).



**Figure 9.** Graph showing enrichment factors (EF) of anthropogenic elements in fine sediment from Commencement Bay (CB), Puget Sound, Washington, over the geologic background in glacial deposits. As, arsenic; Cd, cadmium; Cu, copper; Pb, lead; Zn, zinc. Locations of sampling sites are shown in figure 2.

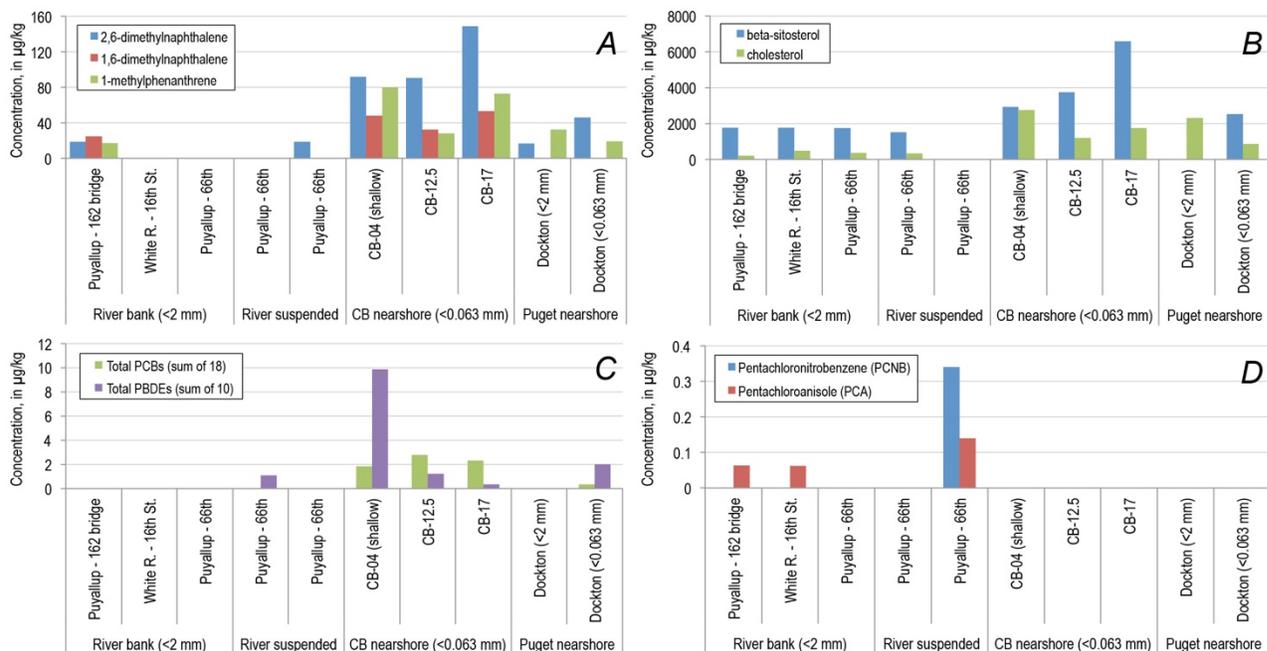
## Discussion

The high proportion of river-derived sediment in the fine fraction of Commencement Bay nearshore sediment suggests that the Puyallup River is the dominant sediment source to the bay, a finding that is consistent with a deltaic environment. Spatial distributions of Al-normalized Ba contents of fine sediment show that the northeast shore of Commencement Bay received a higher proportion of fine sediment from the Puyallup River than the southwest shore and Dash Point (CB-DP), where glacial bluffs played a greater role as a secondary sediment source.

The predominance of river-derived sediment in the nearshore region of Commencement Bay precluded event-scale runoff sourcing with composition-based geochemical signatures alone because riverine sediment entering the bay during a storm had only a slight compositional difference compared to preexisting sediment. In combination with  $^7\text{Be}$  activities (TOC-normalized), it was determined that the sediment mixed layers at CB-04-D and CB-12.5 contained the most recently deposited riverborne fine sediment. A higher  $^7\text{Be}$  inventory at the deeper site at Browns Point (CB-04-D, -16 m MLLW) compared to the shallower site (CB-04-S, -8 m MLLW) could be indicative of downslope transport of riverborne sediment or a greater retention of fine sediment at deeper depths.

The elemental composition of sediment in summer runoff, which was not investigated in detail in this study, may allow discrimination of riverborne and nearshore sediment; however, because summer runoff largely originates in the forested upper watershed, it is not expected to be associated with contaminants of ecological concern. Indeed, few contaminants were detected in river suspended sediment collected in the summer, when the sediment primarily originates from glacial meltwater in the upper watershed. Most detections of river sediment-bound contaminants occurred in winter samples, suggesting that contaminants are introduced into the river during lowland runoff events.

Although a direct comparison between contaminant concentrations in river sediment and nearshore sediment is not possible because different particle size fractions were analyzed, a qualitative comparison suggests that nearshore sediment in Commencement Bay contains more contaminants, including persistent, legacy chemicals, than river sediment (fig. 10). For example, four PAHs at concentrations less than  $25 \mu\text{g}/\text{kg}$  were detected in one river-bar sediment sample, and only during the winter. PAHs were not detected in two other winter river-bar and riverbank sediment samples or in  $<2\text{-mm}$  or fine-sediment samples collected during the summer. As many as 11 PAHs at concentrations less than  $35 \mu\text{g}/\text{kg}$  were detected in river suspended sediment samples, which have a greater fraction of fine sediment than river-bar and riverbank sediment. In contrast, 27 PAHs were detected in winter nearshore fine sediment at concentrations as much as  $750 \mu\text{g}/\text{kg}$ . Based on a comparison of fine and  $<2\text{-mm}$  sediment from a nearby Puget Sound nearshore sediment sample (collected at Dockton in Quartermaster Harbor on Vashon Island, Puget Sound, Washington, August 20, 2013), the same suite of PAHs was present in both fine and  $<2\text{-mm}$  sediment. Fine sediment PAHs concentrations were similar to  $<2\text{-mm}$  sediment PAHs concentrations, with fine to  $<2\text{-mm}$  concentration ratios ranging from 0.4 to 1.5 (in exception, 2.8 for 2,6-dimethylnaphthalene).



**Figure 10.** Graphs comparing contaminant concentrations in winter Puyallup River, Washington, sediment and nearshore sediment for (A) three polycyclic aromatic hydrocarbons, (B) two fecal sterols, (C) two industrial chemicals, and (D) two agricultural chemicals. A summer sample from a nearby Puget Sound nearshore location (Dockton) is also shown to relate chemical concentrations in <2-millimeter (mm) and <0.063-mm fractions. CB, Commencement Bay. dpm/g, disintegrations per minute per gram of dry sediment.

Similar to PAHs (fig. 10A), more contaminants at higher concentrations were measured in nearshore fine sediment than in river-bar, riverbank, or suspended sediment for fecal sterols (fig. 10B), legacy industrial chemicals (fig. 10C), and agricultural chemicals (fig. 10D). The higher occurrence of persistent, legacy compounds including PCBs, PBDEs, and chlorinated pesticides in the Commencement Bay nearshore than in the Puyallup River suggests that the nearshore sediment is impacted by other sources of contaminants in addition to contaminants transported by the Puyallup River, such as direct discharges of stormwater or wastewater to Commencement Bay and current and (or) historical human activities occurring directly in Commencement Bay.

Potential indicators of Puyallup River watershed overland run-off include PAHs, fecal indicators (cholesterol,  $\beta$ -sitosterol, indole, phenol), and pentachloroanisole. Of these, pentachloroanisole was unique to river sediment, as it was detected in both sediment from the Puyallup and White Rivers and not in nearshore sediment (fig. 10D). Pentachloroanisole is an environmental metabolite of pentachlorophenol (PCP), which is a general biocide, which has been used, for example, in agriculture and other industries for preserving wood. Other agricultural chemicals also may be indicators of recent sediment derived from the Puyallup River watershed, such as pentachloronitrobenzene (crop fungicide) and chlorinated pesticides like chlordane and chlorpyrifos, which were each detected in a single winter river suspended-sediment sample (not shown). Additional sampling of river suspended sediment over a range of flow, sediment, and seasonal conditions would aid in identifying additional indicator chemicals for the Puyallup River watershed.

## Summary

Understanding the fate, transport, and burial of riverborne sediment and sediment-bound contaminants in the coastal zone is an important step in assessing how these materials could affect coastal biogeochemical cycles and organisms. This USGS study combined river and marine sediment geochemistry and current-use and legacy contaminant analyses to identify riverborne sediment and associated contaminants at shoreline sites in Commencement Bay. Beaches in and near Commencement Bay are used by forage fish and other organisms that could be at risk of exposure to sediment-bound contaminants. The findings of the study are as follows:

- Winter fine sediment from the Puyallup and White Rivers was compositionally distinct from Puget Lowland glacial fine sediment based on ratios of immobile and relatively immobile elements—Ba/Al, Ni/Al, Th/Al, and (La/Yb)<sub>N</sub> ratios.
- Based on Ba/Al ratios, surface sediment (0–2 cm) in Commencement Bay was predominantly composed of Puyallup River sediment compared to glacial bluff sediment. The northeast shore of the bay contained a larger proportion of Puyallup River sediment than did the southwest shore, where glacial bluffs were a larger secondary sediment source.
- <sup>7</sup>Be activities showed that sediment from the Puyallup River was present in the nearshore sediment mixed layer at all five sites where it was measured and that the highest TOC-normalized activities (most recent deposition) occurred at –16 m depth (MLLW) offshore of Browns Point and west of the mouth of Hylebos Creek.
- Contaminants associated with Puyallup River sediment primarily originated from runoff from the Puget Lowlands in winter rather than the upper watershed where glacial meltwater is the main component of flow in summer.
- Nearshore sediment in Commencement Bay is impacted by other sources of contaminants in addition to those carried by Puyallup River sediment, such as legacy embayment contamination from past activities, such as railroad and sawmill operations and ore smelting, and current urban stormwater.

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## Appendix 1. Contents of Anthropogenic Metals, Grain-Size Parameters, and Organic Carbon Contents of Nearshore Fine Sediment Collected in Commencement Bay, Washington, January 28, 2014

[Sediment-quality guidelines (ERL, effects range low; ERM, effects range median) and the geologic background (BKGD) are shown for comparison. As, arsenic; Cd, cadmium; Cu, copper; Pb, lead; Zn, zinc; D50, median grain size; OC, organic carbon;  $\mu\text{g/g}$ , micrograms per gram; %, percent; mm, millimeter]

Sample no.	As ( $\mu\text{g/g}$ )	Cd ( $\mu\text{g/g}$ )	Cu ( $\mu\text{g/g}$ )	Pb ( $\mu\text{g/g}$ )	Zn ( $\mu\text{g/g}$ )	Gravel (%)	Sand (%)	Silt (%)	Clay (%)	Mud (%)	D50 (mm)	OC (%)
CB-03	7	0.2	28	19	73	1.9	92.6	3.0	2.5	5.4	0.285	0.2
CB-04-D	8	0.1	31	13	76	2.9	65.3	20.4	11.4	31.8	0.140	1.3
CB-04-S	8	0.2	38	22	77	7.0	74.6	11.8	6.6	18.4	0.226	0.6
CB-05	7	0.2	28	14	76	0.4	89.3	6.2	4.1	10.3	0.271	0.2
CB-07	7	0.2	30	14	77	2.5	73.3	16.1	8.1	24.2	0.185	0.6
CB-10	8	0.2	36	15	79	7.2	63.9	18.6	10.2	28.9	0.247	0.5
CB-11	9	0.2	42	16	107	1.7	7.3	58.7	32.4	91.1	0.009	10.1
CB-12	9	0.3	42	17	82	0.2	9.8	61.0	29.0	90.1	0.011	6.2
CB-14	8	0.3	36	24	91	21.7	56.8	15.6	5.9	21.6	0.338	1.5
CB-16	12	0.4	50	33	105	24.5	51.3	16.8	7.4	24.2	0.289	3.1
CB-17	15	0.6	66	48	117	0.0	86.7	9.3	4.0	13.3	0.169	2.7
CB-18	32	0.6	97	62	137	0.0	62.0	27.1	10.9	38.0	0.090	11.5
CB-19	15	0.9	54	33	120	0.2	85.2	9.8	4.8	14.7	0.265	2.9
CB-20	12	0.5	48	17	91	0.0	93.4	4.3	2.3	6.6	0.227	0.2
CB-21	22	0.4	64	27	92	0.0	93.2	4.6	2.3	6.8	0.184	0.3
CB-22	87	0.7	163	40	114	0.0	82.4	12.4	5.3	17.6	0.133	8.4
CB-23	44	0.3	182	82	222	0.5	96.0	1.9	1.6	3.5	0.284	0.2
CB-25	7	0.1	17	10	76	0.9	93.2	4.3	1.6	5.9	0.493	0.1
CB-DP	6	0.2	15	13	76	0.0	90.8	6.5	2.7	9.2	0.164	0.6
Sediment-quality guidelines and geologic background												
ERL <sup>1</sup>	8.2	1.2	34	46.7	150							
ERM <sup>2</sup>	70	9.6	270	218	410							
BKGD <sup>3</sup>	8	0.1	26	7.8	74							
Number of samples >ERL	10	0	13	3	1							
Number of samples >ERM	1	0	0	0	0							

<sup>1</sup>ERL values from Long and others (1995).

<sup>2</sup>ERM values from Long and others (1995).

<sup>3</sup>Values from Point Defiance, Tacoma, Washington, glacial clay were used for the geologic background.

## Appendix 2. Contents of Aluminum, Barium, and Trace Elements in Fine Sediment Collected from the Puyallup Watershed, Commencement Bay, and Point Bolin, Washington

[PU, Puyallup River; WH, White River; CB, Commencement Bay; PB, Point Bolin; Al, aluminum; Ba, barium; Ni, nickel; Th, thorium; (La)<sub>N</sub>, North American shale composite normalized lanthanum; (Yb)<sub>N</sub>, North American shale composite normalized ytterbium; %, percent; µg/g, micrograms per gram]

Sample type	Sample no.	Date collected	Al (%)	Ba (µg/g)	Ni (µg/g)	Th (µg/g)	(La) <sub>N</sub>	(Yb) <sub>N</sub>
River	PU13-01	1/20/13	8.27	340	31.7	4.7	0.64	0.55
River	PU13-02	1/20/13	7.73	339	33.0	4.7	0.68	0.58
River	PU13-03	1/20/13	8.04	335	34.4	4.7	0.67	0.52
River	PU13-05	1/21/13	7.89	315	39.6	5.2	0.61	0.61
River	PU13-06	1/21/13	8.02	304	44.7	4.4	0.57	0.55
River	PU14-01	1/29/14	9.42	376	27.5	4.5	0.54	0.39
River	PU14-02	1/29/14	8.91	364	34.9	4.9	0.63	0.48
River	PU14-03	1/29/14	8.17	331	35.4	4.6	0.61	0.55
River	WH13-01	1/21/13	8.77	400	16.3	5.6	0.63	0.55
River	WH13-02	1/21/13	8.56	367	23.4	4.9	0.63	0.61
River	WH14-01	1/29/14	8.28	341	27.1	4.1	0.52	0.48
River	WH14-02	1/29/14	8.99	372	17.9	4.5	0.55	0.39
River	WH14-03	1/29/14	9.24	384	19.6	4.6	0.55	0.45
Nearshore	CB-03	1/28/14	7.61	362	24.6	4.6	0.55	0.45
Nearshore	CB-04-D	1/28/14	8.90	394	24.1	5.1	0.58	0.45
Nearshore	CB-04-S	1/28/14	8.65	377	24.3	5.0	0.59	0.48
Nearshore	CB-05	1/28/14	8.95	380	27.0	4.8	0.56	0.45
Nearshore	CB-07	1/28/14	8.85	382	22.7	4.9	0.57	0.45
Nearshore	CB-10	1/28/14	8.76	376	21.8	4.9	0.60	0.48
Nearshore	CB-11	1/28/14	8.38	372	21.5	4.7	0.57	0.42
Nearshore	CB-12.5	1/28/14	8.17	370	22.5	5.0	0.67	0.52
Nearshore	CB-14	1/28/14	8.77	383	24.1	4.6	0.55	0.42
Nearshore	CB-16	1/28/14	8.08	378	26.0	4.6	0.55	0.45
Nearshore	CB-17	1/28/14	7.82	370	32.9	4.6	0.60	0.52
Nearshore	CB-18	1/28/14	7.90	369	30.2	4.4	0.56	0.55
Nearshore	CB-19	1/28/14	7.44	370	30.9	4.1	0.49	0.45
Nearshore	CB-20	1/28/14	7.14	345	41.9	4.7	0.60	0.71
Nearshore	CB-21	1/28/14	7.01	355	41.8	4.1	0.55	0.58
Nearshore	CB-22	1/28/14	7.20	351	30.1	3.9	0.50	0.48
Nearshore	CB-23	1/28/14	7.42	366	32.7	4.0	0.53	0.45
Nearshore	CB-25	1/28/14	7.55	453	57.0	3.5	0.45	0.58
Nearshore	CB-DP	1/28/14	6.91	350	46.9	4.7	0.53	0.61
Glacial bluff	PB13-01	6/25/13	8.84	581	61.2	4.2	0.47	0.61
Glacial bluff	PB13-10	6/25/13	7.06	450	45.6	2.7	0.52	0.68
Glacial bluff	PB13-11	6/25/13	6.78	429	42.9	2.4	0.49	0.55
Glacial clay	Point Defiance	8/19/13	7.60	503	58.1	3.3	0.45	0.61

### Appendix 3. Radionuclide Activities in Sediment Cores Collected in Commencement Bay, Washington, January 28, 2014

[CB, Commencement Bay; <sup>7</sup>Be, beryllium-7; <sup>137</sup>Cs, cesium-137; <sup>210</sup>Pb, lead-210; <sup>226</sup>Ra, radium-226; <sup>210</sup>Pb<sub>ex</sub>, excess lead-210; cm, centimeters; dpm/g, disintegrations per minute per gram]

Site no.	Depth interval (cm)	<sup>7</sup> Be (dpm/g)	<sup>137</sup> Cs (dpm/g)	<sup>210</sup> Pb (dpm/g)	<sup>226</sup> Ra (dpm/g)	<sup>210</sup> Pb <sub>ex</sub> (dpm/g)
CB-04-S	0-2	0.1	0.2	1.8	0.8	1.1
CB-04-S	2-4	0.3	0.1	2.4	0.9	1.5
CB-04-S	4-6	0.0	0.1	2.2	0.9	1.4
CB-04-S	6-8	0.0	0.1	2.1	0.9	1.3
CB-04-D	0-2	0.3	0.0	2.0	0.9	1.2
CB-04-D	2-4	0.0	0.1	2.3	1.0	1.4
CB-11	0-2	0.4	0.2	3.6	1.4	2.3
CB-11	2-4	0.4	0.2	3.4	1.3	2.1
CB-11	4-6	0.6	0.4	3.6	1.1	2.5
CB-11	6-8	0.0	0.1	3.4	1.1	2.3
CB-12.5	0-2	0.0	0.2	2.8	1.1	1.7
CB-12.5	2-4	0.0	0.1	3.1	1.4	1.7
CB-12.5	4-6	2.7	0.2	2.5	1.1	1.4
CB-12.5	6-8	0.0	0.2	2.4	1.6	0.8
CB-12.5	8-10	0.0	0.2	2.9	0.8	2.2
CB-12.5	10-12	0.0	0.3	2.8	1.2	1.6
CB-18	0-2	0.2	0.1	2.7	0.9	1.8
CB-18	2-4	0.6	0.1	2.7	0.8	1.9
CB-18	4-6	0.4	0.2	2.9	0.7	2.3
CB-18	6-8	0.4	0.0	1.6	0.7	0.9

## Appendix 4. Chemical Parameters Analyzed by the U.S. Geological Survey National Water Quality Laboratory in Sediment Collected from the Puyallup and White Rivers and Commencement Bay, Washington

Parameter	Parameter code
Polycyclic aromatic hydrocarbons by schedule 5506	
2,6-dimethylnaphthalene <sup>1</sup>	63167
anthracene <sup>1</sup>	63180
anthraquinone <sup>1</sup>	63181
benzo[a]pyrene <sup>1</sup>	63183
bis(2-ethylhexyl) phthalate <sup>1</sup>	63187
carbazole <sup>1</sup>	63194
diethyl phthalate <sup>1</sup>	63202
fluoranthene <sup>1</sup>	63208
naphthalene <sup>1</sup>	63220
phenanthrene <sup>1</sup>	63224
pyrene <sup>1</sup>	63227
benz[a]anthracene	63610
hexachlorobenzene <sup>2</sup>	63631
pentachloronitrobenzene <sup>2</sup>	63650
1,2,4-trichlorobenzene	64095
1,2-dimethylnaphthalene	64097
1,6-dimethylnaphthalene	64099
1-methyl-9h-fluorene	64100
1-methylphenanthrene	64101
1-methylpyrene	64102
2,3,6-trimethylnaphthalene	64103
2-ethylnaphthalene	64104
2-methylantracene	64105
4,5-methylenephenanthrene	64106
9h-fluorene	64107
acenaphthene	64108
acenaphthylene	64109
benzo[b]fluoranthene	64111
benzo[e]pyrene	64112
benzo[g,h,i]perylene	64113
benzo[k]fluoranthene	64114
chrysene	64115
dibenz[a,h]anthracene	64116
dibenzothiophene	64117
indeno[1,2,3-cd]pyrene	64118
pentachloroanisole <sup>2</sup>	64119
perylene	64120
phenanthridine	64121
2-fluorobiphenyl (surrogate)	90754
nitrobenzene-d5 (surrogate)	90755
terphenyl-d14 (surrogate)	90756
Waste-indicator compounds by schedule 5433	
1,4-dichlorobenzene	63163
1-methylnaphthalene	63165
polybrominated diphenyl ether 47 <sup>2</sup>	63166
2,6-dimethylnaphthalene <sup>2</sup>	63167
2-methylnaphthalene	63168
3-beta-coprostanol <sup>2</sup>	63170

Parameter	Parameter code
3-methyl-1h-indole (skatol)	63171
3-tert-butyl-4-hydroxyanisole (bha)	63172
4-cumylphenol	63173
4-n-octylphenol	63174
para-nonylphenol (total)	63175
4-tert-octylphenol	63176
acetophenone	63178
acetyl-hexamethyl-tetrahydronaphthalene (ahtn)	63179
anthracene <sup>2</sup>	63180
anthraquinone <sup>2</sup>	63181
atrazine	63182
benzo[a]pyrene <sup>2</sup>	63183
benzophenone	63184
beta-sitosterol	63185
beta-stigmastanol	63186
bis(2-ethylhexyl) phthalate <sup>2</sup>	63187
bisphenol a <sup>2</sup>	63188
bromacil	63189
camphor	63192
carbazole <sup>2</sup>	63194
chlorpyrifos <sup>2</sup>	63195
cholesterol <sup>2</sup>	63196
diazinon	63198
nonylphenol, diethoxy- (total,np2eo)	63200
4-octylphenol diethoxylate-(op2eo)	63201
diethyl phthalate <sup>2</sup>	63202
d-limonene	63203
4-octylphenol monoethoxylate-(op1eo)	63206
fluoranthene <sup>2</sup>	63208
hexahydrohexamethyl-cyclo-pentabenzopyran (hhcb)	63209
indole	63210
isoborneol	63211
isophorone	63212
isopropylbenzene (cumene)	63213
isoquinoline	63214
menthol	63215
metolachlor	63218
n,n-diethyl-meta-toluamide (deet)	63219
naphthalene <sup>2</sup>	63220
4-nonylphenol monoethoxylate, total, (np1eo)	63221
para-cresol	63222
phenanthrene <sup>2</sup>	63224
phenol	63225
prometon	63226
pyrene <sup>2</sup>	63227
tri(2-butoxyethyl) phosphate	63229
tri(2-chloroethyl) phosphate	63230
tributyl phosphate	63231
triclosan <sup>2</sup>	63232
triphenyl phosphate	63234
tris(dichloroisopropyl) phosphate	63235
bisphenol a-d3 (surrogate)	90735
decafluorobiphenyl (surrogate)	90737
fluoranthene-d10 (surrogate)	90738

Parameter	Parameter code
Steroid hormones by schedule 6434	
17-beta-estradiol	63164
3-beta-coprostanol <sup>1</sup>	63170
bisphenol a <sup>1</sup>	63188
cholesterol <sup>1</sup>	63196
equilenin	63204
estrone	63205
17-alpha-ethynylestradiol	63207
cis-androsterone	63607
trans-diethylstilbestrol	63620
mestranol	63638
norethindrone	63644
progesterone	63657
11-ketotestosterone	64467
17-alpha-estradiol	64468
4-androstene-3,17-dione	64473
epitestosterone	64477
equilin	64479
estriol	64480
dihydrotestosterone	64484
testosterone	64485
bisphenol a-d16 (surrogate)	67310
progesterone-2,3,4-13c3 (surrogate)	90512
cholesterol-d7 (surrogate)	90772
estriol-2,4,16,17-d4 (surrogate)	91617
16-epiestriol-d2 (surrogate)	91684
medroxyprogesterone-d3 (surrogate)	91686
nandrolone-d3 (surrogate)	91687
17-b-estradiol-13c6 (surrogate)	91757
estrone-13c6 (surrogate)	91758
17-alpha-ethynylestradiol-d4 (surrogate)	91805
cis-androsterone-2,2,3,4,4-d5 (surrogate)	91808
trans-diethylstilbestrol-d8 (surrogate)	91809
mestranol-2,4,16,16-d4 (surrogate)	91813
Organohalogens by lab code 8093	
polybrominated diphenyl ether 47 <sup>1</sup>	63166
triclosan <sup>1</sup>	63232
endosulfan i	63259
endosulfan ii, solids	63260
benfluralin (benefin)	63265
chlordane, cis	63271
chlordane, trans	63272
chlorpyrifos <sup>1</sup>	63273
cyfluthrin	63279
cyhalothrin	63280
dcpa (dacthal)	63282
dieldrin	63289
endosulfan sulfate, solids	63298
fipronil	63313
fipronil sulfide	63314
desulfanyl fipronil	63316
nonachlor, cis	63338
nonachlor, trans	63339
oxyfluorfen	63341
ddt, p,p'	63345

Parameter	Parameter code
ddd, p,p'	63346
dde, p,p'	63347
pendimethalin	63353
tefluthrin	63377
trifluralin	63390
hexachlorobenzene <sup>1</sup>	63631
triclosan, methyl-	63639
pentachloronitrobenzene <sup>1</sup>	63650
tetradifon	63665
pentachloroanisole <sup>1</sup>	64119
polychlorinated biphenyl 49	64725
polychlorinated biphenyl 52	64726
polychlorinated biphenyl 70	64727
polychlorinated biphenyl 101	64729
polychlorinated biphenyl 110	64730
polychlorinated biphenyl 118	64731
polychlorinated biphenyl 138	64732
polychlorinated biphenyl 146	64733
polychlorinated biphenyl 149	64734
polychlorinated biphenyl 151	64735
polychlorinated biphenyl 170	64736
polychlorinated biphenyl 174	64737
polychlorinated biphenyl 177	64738
polychlorinated biphenyl 180	64739
polychlorinated biphenyl 183	64740
polychlorinated biphenyl 187	64741
polychlorinated biphenyl 194	64742
polychlorinated biphenyl 206	64743
polybrominated diphenyl ether 66	64852
polybrominated diphenyl ether 71	64853
polybrominated diphenyl ether 85	64854
polybrominated diphenyl ether 99	64855
polybrominated diphenyl ether 100	64856
polybrominated diphenyl ether 138	64857
polybrominated diphenyl ether 153	64858
polybrominated diphenyl ether 154	64859
polybrominated diphenyl ether 183	64860
oxychlorane	64866
pentabromotoluene	64867
1,2-bis(2,4,6-tribromophenoxy)ethane	64868
octachlorostyrene	65217
dechlorane plus	65220
pcb 202-13c12 (surrogate)	90802
dibromooctafluorobiphenyl (surrogate)	91785
p,p'-ddt-d8 (surrogate)	91828

<sup>1</sup>Analyzed by two methods; results from this method are reported.

<sup>2</sup>Analyzed by two methods; results from this method are not reported.

