Title
A survey of benthic sediment contaminants in reaches of the Columbia River Estuary based on channel sedimentation characteristics

Journal
Science of The Total Environment, 484

ISSN
00489697

Authors
Counihan, Timothy D
Waite, Ian R
Nilsen, Elena B
et al.

Publication Date
2014-06-01

DOI
10.1016/j.scitotenv.2014.03.013

Peer reviewed

Science of the Total Environment 484 (2014) 331–343

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

A survey of benthic sediment contaminants in reaches of the Columbia River Estuary based on channel sedimentation characteristics

Timothy D. Counihan [a,*], Ian R. Waite [b], Elena B. Nilsen [b], Jill M. Hardiman [a], Edwin Elias [c], Guy Gelfenbaum [c], Steven D. Zaugg [d]

[a] U.S. Geological Survey, Western Fisheries Research Center, 5501-A Cook-Underwood Road, Cook, WA 98605, United States
[b] U.S. Geological Survey, Oregon Water Science Center, 2130 S.W. 5th Avenue, Portland, OR 97201, United States
[c] U.S. Geological Survey, Pacific Coastal and Marine Science Center, 400 Natural Bridges Drive, Santa Cruz, CA 95060, United States
[d] U.S. Geological Survey, National Water Quality Laboratory, West 6th Ave. & Kipling Street, Lakewood, CO 80225, United States

HIGHLIGHTS

- We modeled sedimentation patterns in three reaches in the Columbia River Estuary.
- Sediment transport model is used to guide bottom sediment sample allocation.
- Organic carbon levels are correlated with predicted sedimentation patterns.
- Sediment contaminants are correlated with predicted sedimentation patterns.
- Longitudinal contaminant trends comport to trends in resident bird and fish tissues.

ARTICLE INFO

Available online 3 April 2014

Keywords:
Sediment contaminants
Columbia River Estuary
Hydrodynamic model
Sediment transport
Survey design

ABSTRACT

While previous studies have documented contaminants in fish, sediments, water, and wildlife, few specifics are known about the spatial distribution of contaminants in the Columbia River Estuary (CRE). Our study goal was to characterize sediment contaminant detections and concentrations in reaches of the CRE that were concurrently being sampled to assess contaminants in water, invertebrates, fish, and osprey (*Pandion haliaetus*) eggs. Our objectives were to develop a survey design based on sedimentation characteristics and then assess whether sediment grain size, total organic carbon (TOC), and contaminant concentrations and detections varied between areas with different sedimentation characteristics. We used a sediment transport model to predict sedimentation characteristics of three 16 km river reaches in the CRE. We then compartmentalized the modeled change in bed mass after a two week simulation to define sampling strata with depositional, stable, or erosional conditions. We collected and analyzed bottom sediments to assess whether substrate composition, organic matter composition, and contaminant concentrations and detections varied among strata within and between the reaches. We observed differences in grain size fractions between strata within and between reaches. We found that the fine sediment fraction was positively correlated with TOC. Contaminant concentrations were statistically different between depositional vs. erosional strata for the industrial compounds, personal care products and polycyclic aromatic hydrocarbons class (Indus–PCP–PAH). We also observed significant differences between strata in the number of detections of Indus–PCP–PAH (depositional vs. erosional; stable vs. erosional) and for the flame retardants, polychlorinated biphenyls, and pesticides class (depositional vs. erosional, depositional vs. stable). When we estimated mean contaminant concentrations by reach, we observed higher contaminant concentrations in the furthest downstream reach with a decreasing trend in the two upstream reaches. Contaminant survey designs that account for sedimentation characteristics could increase the probability that sampling is allocated to areas likely to be contaminated.

Abbreviations: CRE, Columbia River Estuary; FR–PCB–Pest, flame retardants, polychlorinated biphenyls, and pesticides; Indus–PCP–PAH, fragrances, surfactants, industrial compounds, personal care products, and polycyclic aromatic hydrocarbons; TOC, total organic carbon.

[*] Corresponding author. Tel.: +1 509 538 2299.
E-mail address: tcounihan@usgs.gov (T.D. Counihan).

http://dx.doi.org/10.1016/j.scitotenv.2014.03.013
0048-9697/Published by Elsevier B.V.

1. Introduction

Exposure of fish, wildlife, and people to contaminants within the Columbia River Basin has caused concern (USEPA, 2009). Contaminants measured in Columbia River fish included PCBs, dioxins, furans, arsenic, mercury, and DDE, a toxic breakdown product of the pesticide DDT (USEPA, 2009). In 1992, a USEPA contaminants survey suggested a potential health threat to tribal and other people who eat fish from the Columbia River Basin (USEPA, 1992). The consumption survey showed that 92 contaminants were detected in fish with some above levels of concern for aquatic life or human health. More recently, the Oregon Department of Environmental Quality (2012) found concentrations of DDTs and PCBs that grossly exceed DEQ's human health criteria, both in smallmouth bass (*Micropterus dolomieu*) and largescale suckers (*Catostomus macrocheilus*). Exposure of aquatic life to contaminants, including emerging contaminants such as flame retardants, pharmaceuticals, cosmetics, personal care products, hormones, antibiotics, and other drugs, can occur through various routes such as permitted effluent. Since contaminants can affect fish, human, and wildlife health and there are long standing concerns about contaminant exposure in the CRE, a more thorough investigation of contaminants in the CRE is warranted.

Contaminants in sediments and water in the CRE could confound efforts to recover threatened and endangered anadromous salmon and to manage other fish and wildlife species. Current and past industrial discharges into the CRE have resulted in contamination of sediments and water (USEPA, 2009). All anadromous fish species produced in the Columbia and Snake Rivers, many that are listed as endangered or threatened under the Endangered Species Act, have to migrate through the CRE (NMFS, 2014). In a study examining the body burdens of contaminants in juvenile salmon in the CRE, researchers documented exposure levels for some contaminants approaching concentrations that could affect their health and survival and that were among the highest levels measured from Pacific Northwest estuaries (Johnson et al., 2007). Sediment deposition is one way compounds may persist in the aquatic environment and enter the foodweb via benthic organisms (Nakata et al., 2007). Understanding what habitats are contaminated may help identify mitigation opportunities.

While previous studies have documented contaminants in fish, sediments, water, and wildlife, little is known about the specific spatial distribution of contaminants in habitats of the Columbia River and CRE, despite its designation as a priority Large Aquatic Ecosystem (USEPA, 2010). In a 2007 contaminants survey, approximately 16% of the Columbia River estuarine area was in poor condition with respect to sediment contaminants (USEPA, 2007). The USEPA (2007) study employed a design intended to assess the status and trends of ecological resources at a regional scale. Few other probabilistic surveys of sediment contaminants have been conducted in the Columbia River. As part of the Columbia River Toxics Reduction Plan, the USEPA (2010) recognizes the need for a coordinated effort to identify research and monitoring priorities and the integration of water, land, air, sediment and biota monitoring for contaminants in this system.

The goal of our study was to provide a characterization of benthic sediment contaminant detections and concentrations in three reaches of the CRE that were concurrently being sampled to assess contaminants in water, invertebrates, fish, and osprey (*Pandion haliaetus*) (Nilsen and Morace, 2014-in this issue) to provide context to these studies. Our objectives were to develop a survey design based on sedimentation characteristics and then assess whether sediment grain size, total organic carbon (TOC), and contaminant concentrations and detections varied between areas with different sedimentation characteristics and between reaches that were hypothesized to represent a gradient in contamination.

1.1. Study area

The Columbia River is the fourth largest river in the United States (US), draining an area of approximately 670,800 km^2, from Canada to the northwest of the US. The Columbia River is highly affected by dams that have altered the river's hydrograph and sediment load (Naik and Jay, 2011; Gelfenbaum et al., 2001). Our study was conducted in the Columbia River Estuary (CRE), an un-impounded reach of the Columbia River below Bonneville Dam, which is located at river kilometer 378. The entire CRE is affected by ocean tides as well as by a seasonally changing hydrograph.

We chose three river reaches hypothesized to provide a potential gradient of contaminant concentrations (Nilsen and Morace, in this issue). The three reaches selected were the Columbia River near Longview (river kilometer 106), near Columbia City (river kilometer 132), and near Skamania (river kilometer 225) (Fig. 1). Results from contaminant studies using passive samplers, fish tissues, and osprey eggs in 2008–2010 (Henny et al., 2009, 2011; Alvarez et al., 2014-in this issue; Christiansen et al., 2014–in this issue; Nilsen et al., in this issue; Torres et al., in this issue) were used to help select these three study reaches. Sample reaches were made approximately 16 km in length and were located to encompass sampling sites for largescale suckers (Nilsen et al., 2014-in this issue), osprey eggs (Henny et al., 2011), and passive contaminant samplers (Alvarez et al., in this issue).

The Longview reach is located near the Port of Longview that consists of eight marine terminals and waterfront industrial property dominated by forest products and steel industries and has inputs from municipal and industrial sources. The Columbia City reach is located near two small towns in Oregon, Columbia City on the downstream end and St. Helens on the upstream end. The Multnomah Channel, which branches off of the Willamette River, drains into the Columbia River at the upstream end of this site. The Skamania reach is the most upstream reach and is located downstream of Bonneville Dam, and upstream of the urban areas of Portland and Vancouver. Areas immediately upstream of the Skamania reach have been shown to have relatively low levels of contaminants (Fuhrer et al., 1996; Johnson et al., 2007; Morace, 2006).

Fluvial processes, modified by tidal processes, control the hydrodynamics and sediment transport in the CRE. Flow is directed downstream in all three reaches, but slows and even reverses direction with the tides, even as far upriver as the Skamania reach. Even though salt water never reaches this far upriver, the tides still influence water elevation and velocity. The magnitude of flow reversal is a function of seasonally-varying river discharge. The highest river flows occur during the spring freshet while low flows typically occur during late summer. Sediments and contaminants were sampled during late summer — early fall low flow conditions when flow reversals are typically greatest. Sedimentation patterns in the CRE depend spatially on convergences and divergences in sediment transport flux that can occur near river meanders, in side channels, or in embayments. Sedimentation or erosion can also result from temporal changes in the sediment concentration, typically during rapid decreases or increases in river discharge associated with flooding.

2. Material and methods

2.1. Delft3D sediment transport modeling

To investigate physical processes and sediment patterns in the CRE a Delft3D flow and sediment transport model was created that encompasses the Columbia River downstream of Bonneville Dam and includes the estuary, mouth, and open coast to the continental shelf break. The model used in this study is based on a Delft3D hydrodynamic and sediment transport model originally developed specifically for the mouth of the Columbia River and which has been validated for waves and currents against field data (Elias et al., 2012). World-wide case studies indicate that Delft3D is applicable to a wide range of coastal and fluvial environments including ocean, estuary and river settings (Grunnet et al., 2004; Van Maren, 2004; Elias et al., 2006; Lesser et al., 2004; Mulligan et al., 2010). The base model for the CRE has a grid resolution varying between 50 to 100 m^2, with an average of 20 grid cells

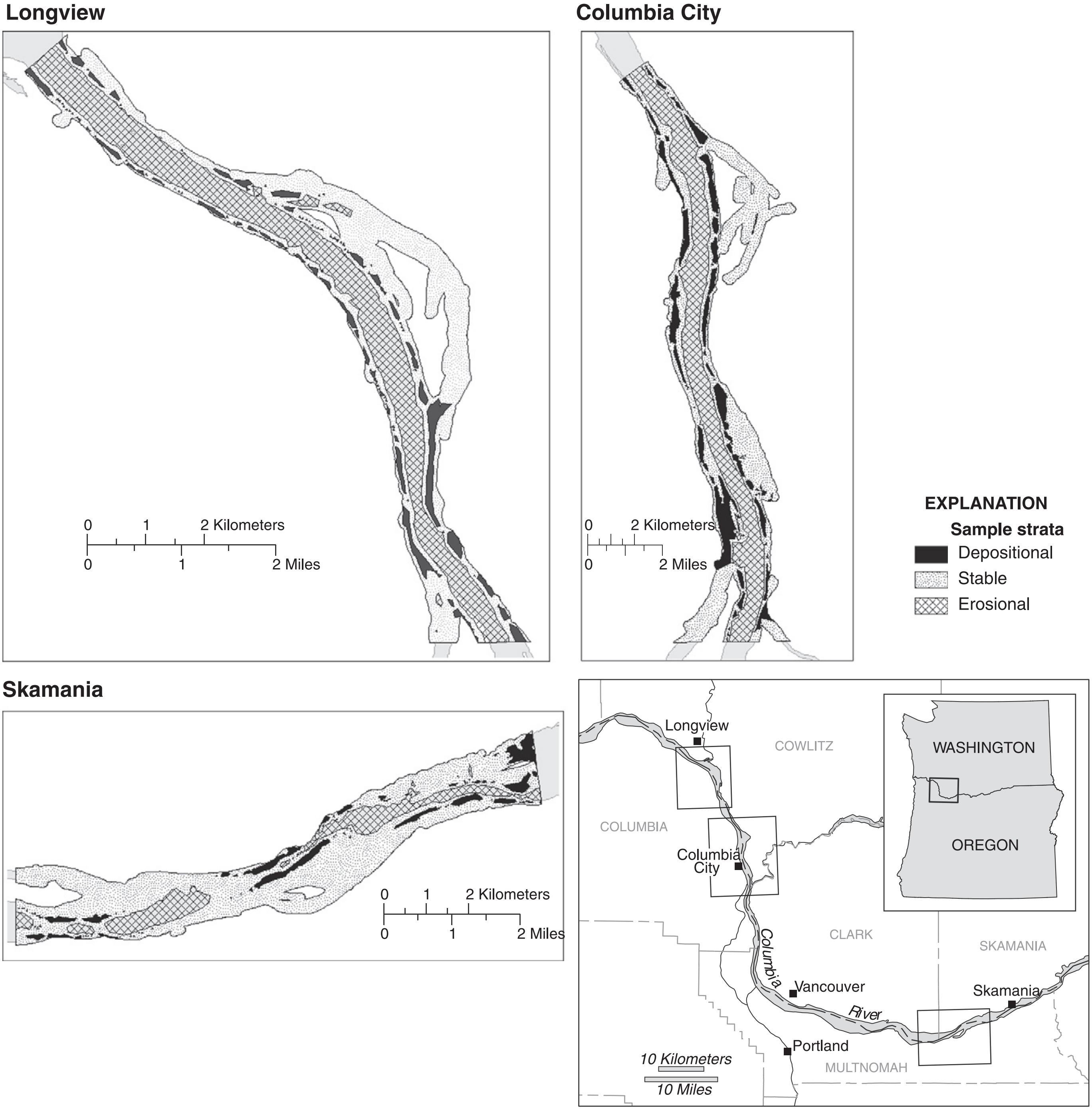

Fig. 1. Map of the study area and the delineation of the three sedimentation strata, depositional (black), stable (stipule), and erosional (cross hatch) for the Longview, Columbia City, and Skamania reaches.

covering the main channel of the river. This resolution was found to be sufficient to represent the base characteristics of the main channel on the large scale and enable relatively fast yet accurate simulations over the entire CRE for model calibration. For detailed patterns of fine sediment dispersal, this resolution is however not sufficient and higher resolution modeling was performed within the three study reaches of interest. For a complete description of the estuary hydrodynamic model, validation procedures, and limitations see Elias et al. (2012).

Sediment transport formulae from van Rijn (1993) and van Rijn et al. (2004) are used to model non-cohesive sediment fractions. The sediment transport is separated into bed load and suspended load components. The suspended sediment transport is computed by the advection–diffusion solver, and includes the effect of sediment in suspension on the fluid density. Bed load transports represent the transport of sand particles in a layer in close contact with the bed surface, and include an estimate of the effect of wave orbital velocity asymmetry when waves are present (Lesser et al., 2004).

The hydrodynamic and sediment transport model for silt distributions, used to formulate the sediment collection sample design, started from an initially uniformly mixed sediment bed composed of two size fractions, 50% sand and 50% silt. The hypothetical bed was composed of a 10-m thick sediment layer, with a specific density of 2650 kg/m^3 and a dry bed density of 1600 kg/m^3. From this initial uniform distribution of sediment on the bed, a space varying sediment distribution was estimated over the CRE. Results were derived from a quasi-equilibrium state (2-weeks of runtime) and based on a constant (average) river discharge of 3500 m^3/s. The output from these simulations formed a grid of latitude, longitude, and change in sediment mass value.

2.2. GIS integration, sampling stratum delineation, and site selection

We developed a Geographic Information System (GIS) to integrate the Delft3D modeling results into our sampling design. We imported the data generated from the Delft3D models into ArcGIS and then used an inverse distance weighted interpolation method to create a continuous surface (or grid) with a 30 × 30 m resolution. The 30 × 30 m resolution was selected to comport with the resolution of a previously generated sample frame formulated for the CRE. We then delineated sampling strata (strata) by evaluating the distribution of sediment transport change in mass from the modeling results. A median value of 0.00 indicated no change in mass on the bed during the simulation, with a range from -33.6 to 34.0 kg/m^2. We established strata such that we could identify areas most likely to be depositional (>20.0 kg/m^2) or erosional (<-20.0 kg/m^2), with the remaining values of the distribution categorized as indicating a stable bed. The grid files were classified into the three strata and were converted into polygon shape files in which total area for each stratum type could be calculated for each reach (Table 1).

2.3. Site selection and allocation

Sample sites were selected by drawing points from each of the three strata within each reach from a sample frame developed for the entire CRE that was generated using a Generalized Random Tessellation Stratified (GRTS) algorithm (Stevens and Olsen, 2004). The CRE sample frame encompasses the entire CRE including the floodplain extent; essentially establishing a grid of sample points at a resolution of 30 × 30 m. We then attributed the sample frame within each reach with the stratum designations (i.e., depositional, erosional, or stable). We extended the sample frame 100 m beyond the static depiction of the river shoreline in our GIS (Fig. 1). The sample frame was extended to ensure that we did not exclude sites if river discharge was higher when we sampled than under the flow conditions represented in the GIS. Within each reach and stratum type, a list of primary sampling sites (15 sites) and oversample sites (50 sites) was generated. Oversample sites were used in the event that primary sampling sites were unable to be sampled or were determined not to be representative of the assigned stratum.

We collected both composite and non-composite samples to assess sediment characteristics and contaminant concentrations within and between the strata and within and between the study reaches. Sampling effort was allocated equally across strata and reaches for the composite samples we collected. To assess differences in sediment characteristics and contaminant concentrations and detections between and within the reaches, we collected three composite samples per stratum per reach for a total of nine composite samples per reach. The composite samples were each comprised of five individual samples from different sampling locations within each of the strata, for a total of 45 unique sampling locations per reach. Additional individual (non-composite) samples were taken in the Columbia City reach and consisted of seven samples from each of the erosional and depositional strata within this reach. We collected these samples to bolster our sample size for assessing the hypotheses that there were differences in sediment grain size, percent organics, and contaminant concentrations between the erosional and depositional strata.

2.4. Field methods

Sampling for sediments and contaminants occurred from September 14–27, 2010, when river discharge was low (Fig. 2). Sediment samples were collected from a boat using a standard ponar benthic grab sampler, deployed from a bow-mounted crane and winch. Samples were typically collected in a downstream to upstream order within each distinct stratum; if a site was deemed inaccessible by boat a replacement site was chosen from a list of oversample points previously generated from the GRTS sample frame for each reach. Furthermore, a sample site was rejected if the site clearly displayed characteristics that suggested the site's stratum designation was incorrect.

2.4.1. Composite and non-composite sample collection and processing

Individual ponar grab samples collected within a strata were deposited in a stainless steel bin and then composited. Individual samples were homogenized with a stainless steel spoon, subsampled, and transferred to a whirlpak for grain size and organic carbon fraction analysis; the remaining sample portion was then transferred to one of three bins set up for the composite samples for each stratum. Each bin was labeled for each composite and covered between sample sites. Once all five samples were collected for the composite sample, the sample was homogenized again and a portion was transferred to a 500 ml glass jar for contaminant analysis. The individual stainless steel collection bins were rinsed thoroughly with native water between samples. After the full composite was collected for each stratum, the bins were rinsed with native water, cleaned with Liquinox soap and deionized water, rinsed two more times with deionized water, and finally rinsed with methanol from a squirt bottle and allowed to dry before the next composite sample was collected. For non-composite samples, material was transferred directly into a 500 ml glass jar and whirlpak for analysis.

2.5. Lab methods

2.5.1. Sediment grain size and organic carbon

Sediment samples were analyzed according to procedures modified from Carver (1971) and Folk (1966, 1974). Sediment samples (5–25 g) were place in beakers and soaked overnight in a solution of water (300 ml) and 35% hydrogen peroxide (10 ml), in order to digest organic matter and initiate the process of sample disaggregation. The samples were then heated gently for several hours to drive off excess hydrogen peroxide, washed twice via centrifugation and decantation to remove soluble components. The samples were then wet-sieved through a 2 mm and 0.063 mm sieves in order to segregate the samples into three size classes: gravel (>2 mm); sand (0.063–2 mm) and mud (<0.063 mm). The gravel and sand fractions were dried and weighed and the mud was collected in 1000 ml graduated cylinders. The weight of the mud fraction was determined by calculation. Simply, a 20 ml aliquot of suspended sample was taken from a graduated cylinder and dried and weighed. The dry weight of the 20 ml aliquot multiplied by

Table 1
Area (percent of total) contained within the depositional, erosional, and stable strata and the total area of each of Longview, Columbia, and Skamania reaches (m^2).

Reach	Depositional Area m^2 (%)	Stable Area m^2 (%)	Erosional Area m^2 (%)	Total area m^2
Skamania	2,161,000 (7.8)	20,798,000 (75.6)	4,568,000 (16.6)	27,527,000
Columbia City	3,081,000 (13.3)	12,224,000 (52.7)	7,885,000 (34.0)	23,190,000
Longview	1,847,000 (8.1)	11,580,000 (50.8)	9,359,000 (41.1)	22,786,000
All reaches	7,089,000 (9.6)	44,603,000 (60.7)	21,812,000 (29.7)	73,504,000

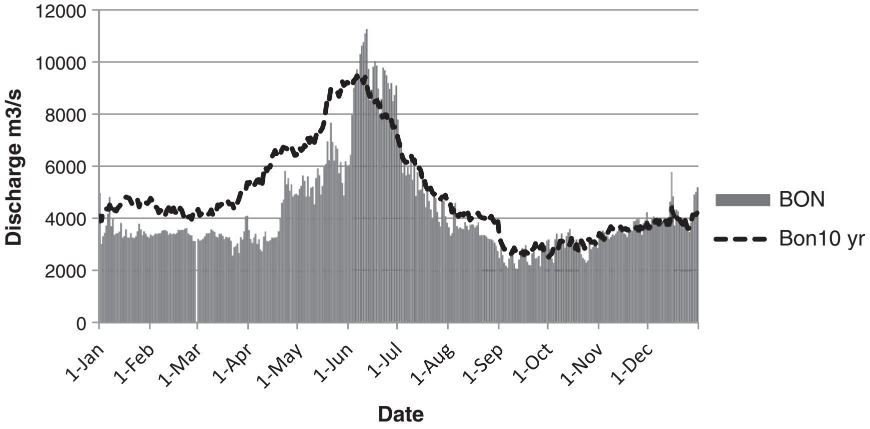

Fig. 2. Discharge (BON) during 2010 and the 10-year average (Bon10 yr) discharge of the Columbia River at Bonneville Dam; Bonneville Dam discharges into the Columbia River Estuary. Sampling for sediments occurred on September 14–27, 2010.

50 gives the total weight of the 1000 ml sample in the graduated cylinder. The gravel fraction is analyzed by sieve and the sand and mud fractions were analyzed with a Beckman Coulter LS230 laser diffraction particle size analyzer at 1/4 phi size intervals.

Inorganic carbon was measured by coulometric technique (UIC, Inc., Coulometrics Model 5012) and reported as weight percent. Total carbon was also measured by this technique and organic carbon was determined by difference. Each sample was measured in duplicate and reported as average weight percent. Relative standard deviation of the mean of multiple measures of a pure calcium carbonate standard was <1%, and detection limits for typical sample sizes of 5–10 mg were <1 wt.%.

2.5.2. Contaminants

All samples were shipped to the USGS National Water Quality Laboratory (NWQL) for contaminants analyses. Once received at the NWQL, samples were frozen at −20 °C and thawed just prior to sample preparation. For the composite samples, the compounds of interest (Appendix A) were extracted from 5 to 10 g of homogenized sediment (by wet weight) using a pressurized liquid extraction system (Dionex ASE™ 200, Sunnyvale, CA, USA). Two analyses were performed. First, sample extracts were prepared as described by Nilsen et al. (in this issue) and halogenated compounds were separated by capillary column gas chromatography and detected by negative ion mass spectrometry (MS), with ammonia as the reaction gas, using selected ion monitoring (Agilent Technologies, Model 5975 GC/MS). Then, a separate extraction was performed on all samples for analysis of 62 anthropogenic waste indicator compounds; these extracts were prepared and analyzed by positive electrospray ionization gas chromatograph–mass spectrometry using methods previously described (Burkhardt et al., 2005, 2006). For the non-composite samples collected in the depositional and erosional strata in the Columbia City reach, sediment samples were analyzed for 60 halogenated organic contaminants (see Zaugg et al., 2006).

2.6. Data analysis

Due to the large number of non-detects, we summarized the chemistry data by compound type and method into the following seven categories: fragrances, surfactants, industrial and personal care products (Indus–PCP), polycyclic aromatic hydrocarbons (PAH), flame retardants (FR), polychlorinated biphenyls (PCB), and pesticides (Pest). Compounds were then further grouped into two summary classifications, Indus–PCP–PAH and FR–PCB–Pest, to compare among strata and study reaches (see Appendix A for individual compound classifications). We used an analysis of variance (ANOVA) followed by the Tukey's pair-wise comparison test to test for statistical differences among strata and reaches. Using the values for the seven contaminant compound categories above, a non-metric dimensional scaling ordination (NMDS) was performed on a Euclidean based similarity matrix among all samples for the three reaches and strata (n = 27). The ordination was performed in Primer-e (Clarke and Gorley, 2006) and all other analyses and graphics were completed in R (R Development Core Team, 2007).

We used estimates of metrics describing chemical compound concentrations, sediment grain size characteristics, and percent TOC arising from the analyses of the composite samples to estimate weighted means by study reach (Cochran, 1977). We used the proportion of the areal extent of the individual strata to the entire area within each study reach as stratum weights.

3. Results

3.1. Hydro-geomorphic characterization

The process-based modeling indicated that the three reaches had different sediment transport characteristics, in terms of the percentage of area that was depositional, erosional, or stable (Table 1). The estimated total area was similar for the three reaches, ranging from 27,527,000 m^2 in the Skamania reach to 22,786,000 m^2 in the Longview reach. Based on the criteria established for formulating the strata, the percent area classified as depositional was highest in the Columbia City reach (13%) and lowest in the Longview and Skamania reaches (8%). Conversely, the Longview reach had the highest percentage of area contained within the erosional strata (41%) while the Skamania reach had the highest percentage of area classified as stable (76%). Some sample sites were rejected in the field because the sediment grabs indicated that the stratum assignment was incorrect (i.e. if a depositional site contained cobble or bedrock). Assignment errors while few overall (<5%) occurred primarily in areas classified as depositional.

The regions of the river that are classified as erosional and depositional are related to the river channel morphology (Fig. 1). In the deepest part of the channel the flows are greatest and sediment transport rates are high. These regions tend to have coarser sediments (see Section 3.2 below) and are consistently classified as erosional. The banks of the river channel are regions where the flow slows down dramatically, the sediments tend to be finer grained, and the classification is predominantly depositional. These patterns are consistent with the Level 5 geomorphic catena classification of Simenstad et al. (2011) in their estuary ecosystem classification scheme.

3.2. Sediment grain size analyses

While the predominant sediment type in the composite samples was sand and gravel we observed differences in the other sediment grain size fractions among strata within and between reaches (Fig. 3).

For the composite samples, the mean percent sediment characterized as sand or gravel by strata ranged between 85 and 99% across the Columbia City, Longview, and Skamania study reaches; the mean percent sediment characterized as sand in the composite samples ranged from 70 to 93%. The amount of silt and mud (percent fines) and percent total organic carbon (TOC) were higher on average at Longview and Columbia City than those at Skamania and depositional and stable strata had higher percent fines and organic carbon than the erosional strata in all reaches (Fig. 3). We found that the amount of fine sediment was positively correlated with TOC; the log of percent fines and percent TOC had a strong linear response and a spearman rank correlation of 94% (Fig. 4). There was also a large difference in the average sediment grain sizes between the seven non-composite depositional and erosional samples taken in the Columbia City reach; the average percent sand plus gravel, percent fines and percent TOC for the depositional samples were 82, 18 and 0.44%, respectively versus 99, 1 and 0.04% for the erosional samples.

3.3. Contaminants analyses

The number of detections and total contaminant concentrations were highly variable among strata within and between reaches (Table 2). The only pairwise comparisons for sediment contaminants that were statistically significant (Tukey test; $p < 0.05$) were at Columbia City: one for total concentration for Indus–PCP–PAH between depositional versus erosional strata (Fig. 5) and two pairwise comparisons each for number of detections for Indus–PCP–PAH (depositional versus erosional, stable versus erosional strata) and for FR–PCB–Pest (depositional versus erosional, depositional versus stable strata; Tukey test; $p < 0.05$) (Fig. 6). Even though the pairwise comparisons for Longview were not statistically significant, the averages follow the expected trend of higher concentrations and more detections at the depositional strata, intermediate values in the stable strata and then the lowest values in the erosional strata (Figs. 5 and 6 and Table 1).

The Longview depositional strata had the highest mean concentration for Indus–PCP–PAH but Columbia City depositional strata had the highest mean concentration for FR–PCB–Pest (Table 2). Across all reaches, there were more detections and higher mean concentrations in the depositional strata than the erosional strata for both summary contaminant classes; the stable stratum values were intermediate to these two extremes in each reach. For the non-composite samples collected in the Columbia City reach, there were no detections of the 60 contaminant compounds analyzed from seven individual erosional samples while there was at least one detection of 17 of the 60 contaminant compounds in all seven depositional samples; two of the seven depositional samples had detections for flame retardants, two for pesticides and four samples for PCB's.

Our analyses suggest a positive relationship between the percent substrate classified as fines and the number of contaminants detected. We documented a relatively strong relationship across all samples between the log transformed number of detections for both Indus–PCP–PAH and for FR–PCB–Pest and the percentage of fine substrate in samples (Fig. 4). The relation between detections of FR–PCB–Pest and percent fines followed an increasing curvilinear response (Spearman rho = 0.65), while the relationship of Indus–PCP–PAH and percent fines followed a more linear response (rho = 0.75) (Fig. 4). The multivariate ordination of all samples (NMDS; n = 27) based on the seven contaminant compound categories revealed three general groupings of samples: 1) all the Skamania samples loosely grouped together on the left of the ordination along with the erosional samples from the other two sites, 2) a group of depositional and stable stratum samples from Longview and Columbia City together in the middle of the ordination, and 3) four individual samples separated from all other samples along the margins of the ordination. The four individual samples separated from all other samples along the margins of the ordination included one Columbia City stable stratum sample and three Longview samples, one of each from the three strata (Fig. 7).

We observed a trend towards higher mean total contaminant concentrations in the Longview reach with a decreasing trend in the reaches upstream of Longview; the lowest concentrations occurring in the Skamania reach (Table 3). We also observed a concomitant trend in percent TOC in sediment from the Longview reach to the Skamania reach. The FR–PCB–Pest group did not conform to the observed trend; the highest levels were observed in the Columbia City reach and not Longview. However, the trend was evident for 2 of the 3 individual components of the metric, PCB's and pesticides (Table 3). Upon further examination we discovered that the flame retardant values observed for the Columbia City reach were due to the influence of a single detection of triphenyl phosphate in one of the composite samples.

4. Discussion

The importance of understanding sedimentation patterns in estuaries when evaluating the distribution of contaminants is underscored by research on sediment contamination conducted in estuary and river systems with known contamination issues (Olsen et al., 1978, 1993; Ankley et al., 1992; Steuer et al., 1995; Hirschberg et al., 1996). The

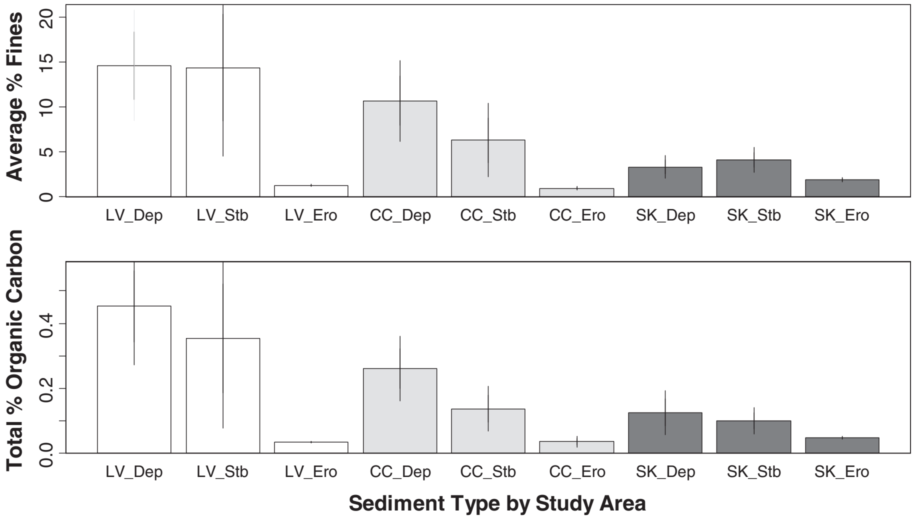

Fig. 3. Average percent silt and mud (% Fines) and percent total organic carbon (% Organic Carbon) in samples collected in erosional (Ero), depositional (Dep), and stable (Stb) sampling strata by reach in the Columbia River Estuary (see text for site codes).

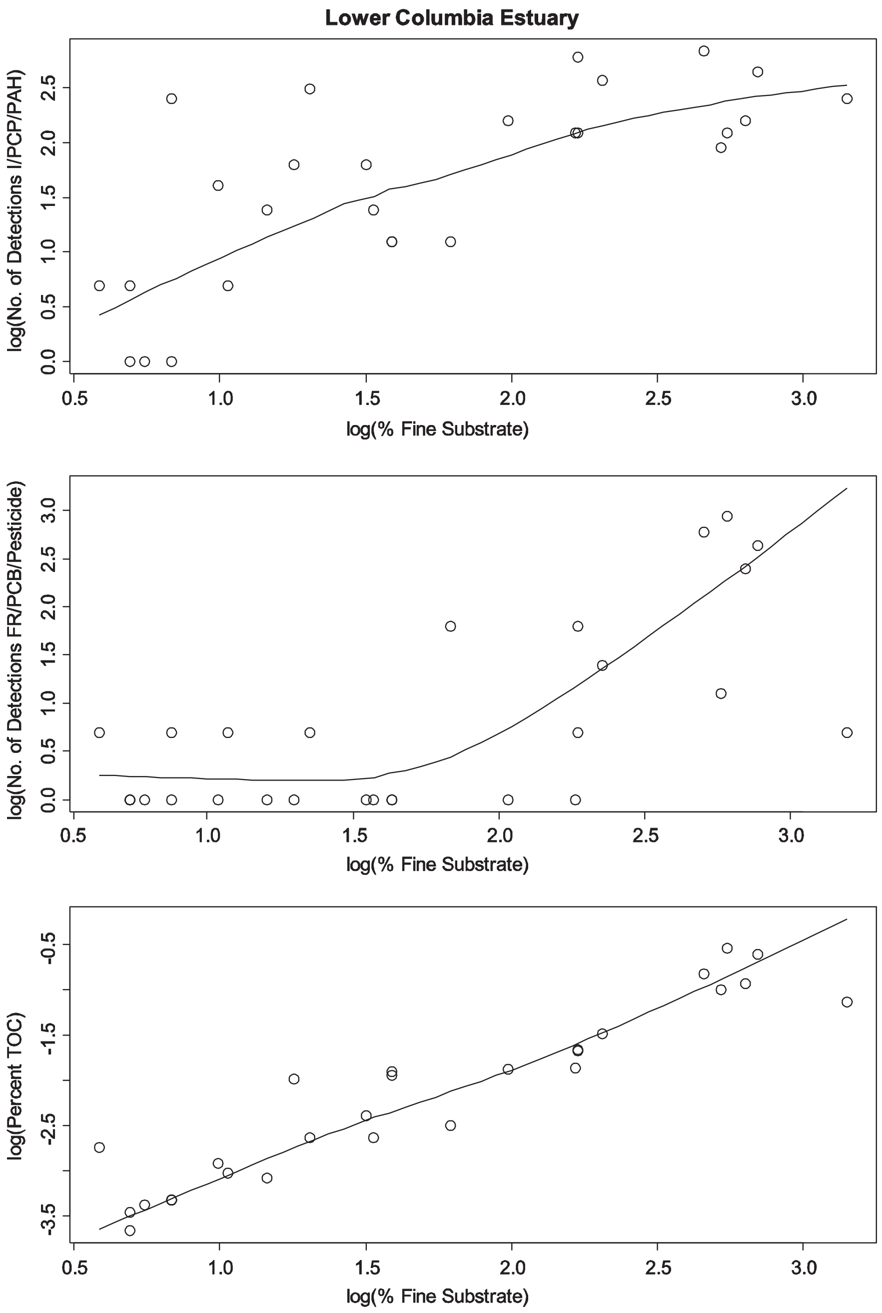

Fig. 4. Two-way plots of the log of percent fine substrate vs. a) log of number of detections of sum of Indus-PCP-PAH compounds, b) log of number of detections of sum of FR–PCB–Pest compounds and c) log of percent total organic carbon (TOC) for all 27 samples from three reaches in the Lower Columbia. See Appendix A for chemicals included in the metrics Indus–PCP–Pest and FR–PCB–Pest.

Table 2
Mean number of detections and total concentration of two summary classes of contaminant compounds: Indus–PCP–PAH and FR–PCB–Pest (nanogram per gram) (see text for complete explanation of compounds).

		Detections		Total concentration	
		Indus–PCP–PAH	FR–PCB–Pest	Indus–PCP–PAH	FR–PCB–Pest
Longview	Depositional	12	10	821.4	1.830
	Stable	8	6	388.0	1.596
	Erosional	3	0	258.3	0.863
Columbia City	Depositional	11	6	485.6	1.377
	Stable	8	1	252.5	5.073
	Erosional	1	0	2.3	1.420
Skamania	Depositional	4	0	310.9	0.000
	Stable	2	2	33.0	0.056
	Erosional	3	0	120.8	2.037

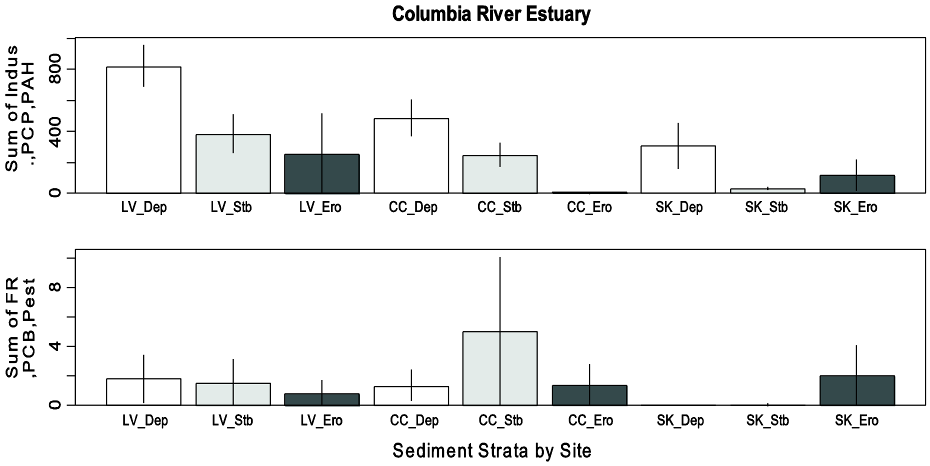

Fig. 5. Barplots of the sum (nanograms per gram) of Indus–PCPvPAH compounds (upper) and FR–PCB–Pest compounds (lower) for three sediment strata from three reaches in the Lower Columbia. Bars represent mean concentration (nanogram per kilogram); vertical lines 90% standard error. See Appendix A for chemicals included in the metrics Indus–PCP–Pest and FR–PCB–Pest.

sedimentation characteristics we observed in our three study reaches are likely related to numerous physical, chemical and biological factors that can affect estuarine sedimentary processes and patterns (Olsen et al., 1978). Research has shown that factors affecting sedimentation in estuaries include alternating tidal currents, two-layer (non-tidal) estuarine circulation, estuary morphology, flocculation and bioagglomeration of fine particles in the water column, mixing of sediments by organisms, human activities and multiple sediment sources (Olsen et al., 1978). The river reaches we sampled in the Columbia River estuary occur above the upper extent of salt water intrusion so sedimentation processes related to the mixing of salt and fresh water are not relevant in these reaches; however tidal cycles do affect the river current in all of the reaches we examined. Most important seems to be channel geomorphology, with the deepest part of the channels classified as erosional and the channel banks classified predominantly as depositional.

We demonstrated that the distribution of fine sediments, TOC, and contaminants in reaches of the Columbia River was correlated with reach- and stratum-specific sedimentation characteristics. The transport and deposition of particle-associated contaminants in a river estuary are controlled by a number of factors including the distribution of sources, seasonal variation in river flow and particle transport, and spatial patterns in sediment deposition (Hirschberg et al., 1996). Research on the distribution and fate of contaminants in estuarine and coastal environments has shown that many chemically reactive contaminants become associated with fine particles and that the rate, pattern, and extent of contaminant accumulation can be extremely variable (Troup and Bricker, 1975; Hites et al., 1977; Schubel and Meade, 1977; Turekian, 1977; Olsen et al., 1982; Sharp et al., 1982; Brush, 1989; Dyer, 1989; Smith and Levy, 1990; Sugai, 1990; Tanaka et al., 1991). Contaminant distribution variability has been attributed to variations in input sources, the reactivity of different particle types, the net rate and pattern of fine-particle deposition, and the extent of surface sediment mixing (Turekian et al., 1980; Bopp et al., 1982; Olsen et al., 1984; Schropp et al., 1990; Venkatesan and Kaplan, 1990). In a paper summarizing the results obtained from 15 yr of research on contaminant distributions in estuaries along the eastern coastline of the United States, Olsen et al. (1993) demonstrated that fine-particle deposition is the most important factor affecting contaminant accumulation in coastal areas. Olsen et al. (1993) further concluded that a large fraction of the total fine-grained sediment surface area in these estuaries is relatively uncontaminated. The latter point suggests the importance of understanding sedimentation characteristics when designing contaminants surveys given that sample sizes are often extremely limited by the cost of collecting and analyzing samples for contaminants.

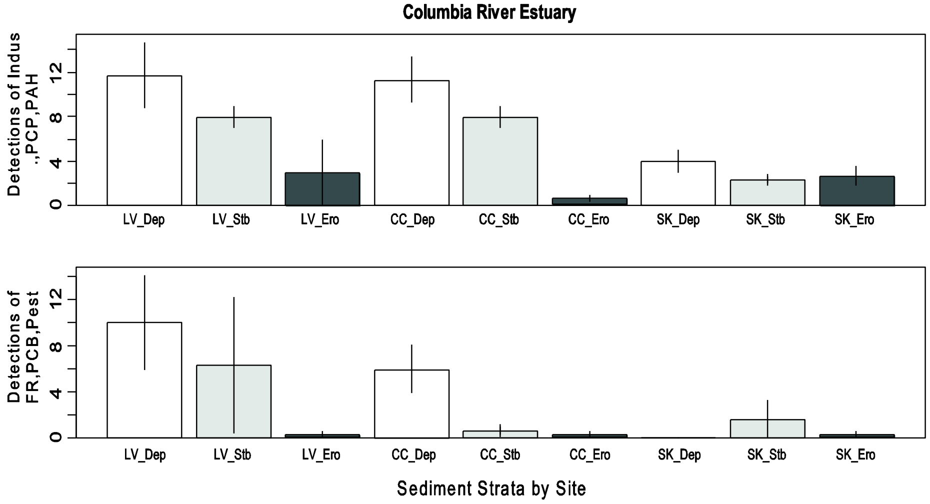

Fig. 6. Barplots of the number of detections of Indus–PCP–PAH compounds (upper) and FR–PCB–Pest compounds (lower) for three sediment strata from three reaches in the Lower Columbia. Bars represent mean concentration (nanogram per kilogram); vertical lines 90% standard error. See Appendix A for chemicals included in the metrics Indus–PCP–Pest and FR–PCB–Pest.

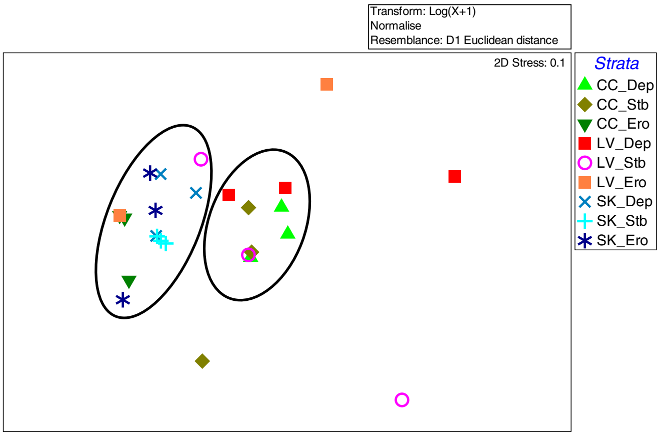

Fig. 7. Non-metric dimensional scaling ordination across all samples (n = 27) based on seven contaminant summary groups (log transformed and normalized). CC = Columbia City, LV = Longview, SK = Skamania; Dep = depositional strata, Ero = erosional strata, Stb = Stable strata. Three samples per stratum, 1–3.

Random sampling and spatially balanced designs (Stevens and Olsen, 2004) that do not account for factors affecting the potential distribution of contaminants in the environment can potentially allocate too few samples to areas that have a high probability of accumulating contaminants. Weighting the allocation of samples based on the probability of habitats to be contaminated could help to ensure that benthic sediment contamination in the CRE is documented by surveys; especially if efforts expended during surveys are limited because of fiscal constraints.

Using different stratum delineation criteria could help better identify areas likely to be contaminated within the CRE. Since this was the first time a sediment transport model has been used to develop a survey design for contaminants in the CRE, we purposively chose to delineate our strata to increase the probability of capturing areas that were depositional or erosional. Consequently, the stable strata, which consistently composed the majority of the area of each reach, contained areas that were either erosional or depositional. We therefore observed, as expected, that the substrate characteristics of sites sampled in the stable strata had variable sediment grain size compositions that included areas with very fine substrates that contained relatively high amounts of TOC and also areas with coarser substrates. Disparate conditions can cause areas within river channels to be stable or marginally erosional or depositional. For instance, main channel areas that are subjected to uniformly higher water velocities are going to contain substrates that are not easily erodible and water velocities that are not conducive to sediment deposition. Conversely, areas off the main channel will infrequently be subjected to higher water velocities but can be areas where very fine sediments are deposited because of the slow water velocities. Given that we observed a positive relation between percentage of substrate classified as fines and the numbers of detections of contaminants, further discriminating portions of the channel that were classified as stable during this study that were more likely to be depositional or erosional could help to better assess the probability of contamination.

Accounting for human induced changes to riverine processes and acquiring regularly updated bathymetric data may provide a more accurate description of the areal extent of areas with different sedimentation characteristics in the CRE. Some man-made structures that affect sediment characteristics such as bridge pilings, dike pilings, and commercial piers were not accounted for in the hydrodynamic and sediment transport models. Increased deposition can occur in areas where the tidal or estuarine flow is hindered or dissipated, such around piers and docks (Ippen, 1966; Schubel, 1968). Furthermore, while the bathymetric data we used for our hydrodynamic and sediment transport models were the most current available, there were recent channel dredging activities not accounted for. Large sand waves regularly form on the Columbia River bed impinging into required channel depths needed to accommodate barge and shipping traffic making routine dredging necessary (Levin et al., 1992). Dredging modifies the bathymetry and has also been shown to focus deposition in other systems (Olsen et al., 1993; Klingbeil and Sommerfield, 2005; Nitsche et al., 2010). Smaller sediment variations include debris fields of anthropogenic deposits, trenches for pipelines and cables (Nitsche et al., 2005).

The reach-specific trends in contaminants we observed in our sediment samples agree with trends in tissue concentrations observed in birds and fish reported for other components of this study (Henny et al., 2011; Nilsen et al., in this issue) suggesting that sediment contamination may contribute to bioaccumulation in resident birds and fish. Nilsen et al. (in this issue) found that concentrations of halogenated compounds in tissues sampled in 2009 ranged from <1 to >400 ng g^{-1} wet tissue weight. PBDEs, organochlorine pesticides, DDT and its degradates (ΣDDT), and PCBs generally showed an increasing trend moving downstream, with the lowest concentrations for most contaminants in fish collected in the Skamania reach and the highest concentrations in fish collected in the Longview reach. Concentrations of brominated flame retardants analyzed in osprey eggs were uniformly higher at the downstream sites than at the upstream site, as was observed in previous studies (Johnson et al., 2006; Henny et al., 2011). Similarly, bioaccumulation of PBDE congeners 47, 99, 100, 153 and 154 at all levels of the foodweb was demonstrated to occur to a greater degree for organisms collected at the sites located in the downstream reaches compared to upstream reaches (Nilsen et al., in this issue). Organic carbon is an important food source for invertebrates that inhabit large river systems (Junk et al., 1989; Thorp and Delong, 2002; Zeug and Winemiller, 2008). Given that we observed a positive relationship between percent fines and TOC and a positive relation between percent fines and detection of contaminants, examining whether invertebrate production is concentrated in contaminated areas could provide insight into how and where sediment contaminants enter the food web.

Table 3
Weighted mean (standard error) of the sums of Indus–PCP–PAH, Indus–PCP, PAH and FR–PCB–PEST and flame retardant, polychlorinated biphenyl (PCB), pesticide concentrations (nanogram per gram), and percent total organic carbon (TOC) detected in sediments in the Longview, Columbia City, and Skamania reaches in the Columbia River Estuary. The stratum weights consisted of the proportion of the areal extent of areas characterized as erosional, depositional, or stable based on hydrodynamic model outputs to the total reach area.

	Longview	Columbia City	Skamania
	Weighted mean (SE)	Weighted mean (SE)	Weighted mean (SE)
Indus–PCP–PAH	369 (124)	198 (44.2)	69.4 (21.6)
Indus–PCP	59.6 (23.6)	51.3 (15.8)	26.9 (5.00)
PAH	310 (137)	147 (38.7)	42.5 (20.6)
FR–PCB–PEST	1.31 (0.89)	3.34 (2.69)	0.38 (0.34)
Flame retardants	0.06 (0.05)	3.30 (2.69)	0.36 (0.34)
PCB	0.73 (0.69)	0.02 (0.01)	No detection
Pesticide	0.52 (0.37)	0.02 (0.01)	0.02 (0.02)
TOC	0.23 (0.06)	0.12 (0.02)	0.09 (0.02)

Although studies examining this wide a suite of contaminants, especially including chemicals of emerging concern, in bed sediments are rare, some comparisons to previous literature can be made. Concentrations of PBDEs measured in this study were on the low end of ranges measured in sediments of the San Francisco Bay, CA (Oros and Ross, 2004) and other locations in the US and Canada (Anderson et al., 2012). Concentrations of nonylphenol and diethylhexyl phthalate were comparable to those measured in the Southern California Bight (Maruya et al., 2012), while concentrations of the surfactants NP1EO and NP2EO, and the PCP triclosan were 3–25 times higher in CRE sediments measured here than measured in the Southern California Bight (Maruya et al., 2012) and San Francisco Bay (Klosterhaus, 2010). Concentrations of PCPs, fragrances, surfactants, and other industrial compounds were higher in bed sediments of tributaries to the Columbia River, but comparable to concentrations in mainstem bed sediments previously measured (Nilsen et al., 2014). Concentrations of legacy contaminants measured as part of this study were comparable to those measured in a previous study at nearby sites (McCarthy and Gale, 1999), but over an order of magnitude lower than concentrations measured in other parts of the Columbia River (e.g., Hermann, 2008).

By using information from hydrodynamic models to allocate sampling effort to habitats with different sedimentation characteristics, we were able to provide a cursory documentation of how contaminant concentrations and detections vary with the hydraulics and sedimentation patterns of the Columbia River. Understanding how riverine processes affect the distribution of contaminants in the CRE could help identify river reaches or areas within the river channel where contaminants are likely to accumulate. To identify and scope contaminant remediation efforts, hydrodynamic models have been used in other river systems to help predict the effects of channel disturbance (i.e., dredging), flow, and other factors affecting river hydrodynamics on the distribution of contaminants within the river channel. For instance, hydrodynamic models have been used to predict bed shear stress at varying river flows in sections targeted for remediation of the Fox River, WI (Shaw Environmental and Infrastructure, Inc. et al., 2006). As part of the Contamination Assessment and Reduction Project (CARP) in the Hudson River, NY, hydrodynamic models are being used to help evaluate the fate and transport of contaminated sediments throughout the New York and New Jersey Harbor Estuary system (Hydroqual, 2007). Flow hydraulic and sediment transport models of the Coeur d'Alene River, ID were developed to better understand the effects of proposed recovery actions to mitigate the effects of mining on the transport and deposition of metal-enriched sediments (Berenbrock and Tranmer, 2008). More research is needed to increase our understanding of the relation of river hydraulics to contaminant distributions in the Columbia River. Our research will lead to a better understanding of transport pathways of contaminants from source, through sediments and into the food web.

5. Conclusions

In this study we used a sediment transport model to predict sedimentation characteristics of reaches in the Columbia River Estuary. By allocating samples based on channel sedimentation characteristics in our study reaches, we found that the distribution of fine sediments, TOC, and contaminants in reaches of the Columbia River Estuary was correlated with reach- and stratum-specific sedimentation characteristics. The reach-specific trends in contaminants we observed in our sediment samples agree with trends in tissue concentrations observed in birds and fish reported for other components of this study (Nilsen and Morace, in this issue) suggesting that sediment contamination in the Columbia River may contribute to bioaccumulation in resident birds and fish. By using information from a sediment transport model to allocate sampling effort to habitats with different sedimentation characteristics, we were able to provide a cursory documentation of how contaminant concentrations and detections vary with the hydraulics and sedimentation patterns of the Columbia River.

Acknowledgments

We would like to acknowledge Glen Holmberg, USGS Western Fisheries Research Center, and Michael Torresan and Angela Lam in the USGS Menlo Park Sediment laboratory for technical assistance with this study. Funding for this study was provided by USGS Regional and REX-flex funds administered by Anne Kinsinger, Leslie Dierauf, and Nancy Lee. Use of trade, product, or firm names is for descriptive purposes only and does not imply endorsement by the U.S. Government. With deep sadness we note that our coauthor, Steven D. Zaugg, passed away while this article was in press.

Appendix A. Chemicals tested for in sediment samples collected in reaches of the Columbia River Estuary during September 10–17, 2010 and their classification per this manuscript

Chemical	Compound category	Summary classification
Triclosan	Personal care product	Indus–PCP–PAH
Isoquinoline	Industrial	Indus–PCP–PAH
Anthraquinone	Industrial	Indus–PCP–PAH
N,N-diethyl-meta-toluamide (deet)	Personal care product	Indus–PCP–PAH
Isopropylbenzene (cumene)	Industrial	Indus–PCP–PAH
Phenol	Industrial	Indus–PCP–PAH
3-Tert-butyl-4-hydroxyanisole (bha)	Industrial	Indus–PCP–PAH
Benzophenone	Personal care product	Indus–PCP–PAH
Isophorone	Industrial	Indus–PCP–PAH
Para-cresol	Industrial	Indus–PCP–PAH
1,4-Dichlorobenzene	Industrial	Indus–PCP–PAH
Pentabromotoluene	Flame retardant	FR–PCB–Pest
1,2-Bis(2,4,6-tribromophenoxy)ethane	Flame retardant	FR–PCB–Pest
Dechlorane plus	Flame retardant	FR–PCB–Pest
Tributyl phosphate	Flame retardant	FR–PCB–Pest
Tri(2-butoxyethyl) phosphate	Flame retardant	FR–PCB–Pest
Tri(2-chloroethyl) phosphate	Flame retardant	FR–PCB–Pest
Tri(dichloroisopropyl) phosphate	Flame retardant	FR–PCB–Pest
Triphenyl phosphate	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 100	Flame retardant	FR–PCB–Pest

Appendix A (*continued*)

Chemical	Compound category	Summary classification
Polybrominated diphenyl ether 138	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 153	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 154	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 183	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 47	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 66	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 71	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 85	Flame retardant	FR–PCB–Pest
Polybrominated diphenyl ether 99	Flame retardant	FR–PCB–Pest
3-Methyl-1H-indole (skatol)	Fragrance	Indus–PCP–PAH
Acetophenone	Fragrance	Indus–PCP–PAH
Acetyl-hexamethyl-tetrahydronaphthalene (ahtn)	Fragrance	Indus–PCP–PAH
Camphor	Fragrance	Indus–PCP–PAH
D-limonene	Fragrance	Indus–PCP–PAH
Hexahydrohexamethyl-cyclo-pentabenzopyran (hhcb)	Fragrance	Indus–PCP–PAH
Indole	Fragrance	Indus–PCP–PAH
Isoborneol	Fragrance	Indus–PCP–PAH
Menthol	Fragrance	Indus–PCP–PAH
1-Methylnaphthalene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
2-Methylnaphthalene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
2,6-Dimethylnaphthalene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
Anthracene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
Benzo[a]pyrene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
Fluoranthene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
Naphthalene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
Phenanthrene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
Pyrene	Polycyclic aromatic hydrocarbon	Indus–PCP–PAH
Polychlorinated biphenyl 101	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 110	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 118	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 138	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 146	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 149	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 151	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 170	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 174	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 177	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 180	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 183	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 187	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 194	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 206	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 49	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 52	Polychlorinated biphenyl	FR–PCB–Pest
Polychlorinated biphenyl 70	Polychlorinated biphenyl	FR–PCB–Pest
Oxyfluorfen	Pesticide	FR–PCB–Pest
Pendimethalin	Pesticide	FR–PCB–Pest
Trifluralin	Pesticide	FR–PCB–Pest
Fipronil	Pesticide	FR–PCB–Pest
Tetradifon	Pesticide	FR–PCB–Pest
Desulfinyl fipronil	Pesticide	FR–PCB–Pest
Fipronil sulfide	Pesticide	FR–PCB–Pest
Chlorpyrifos	Pesticide	FR–PCB–Pest
Cyfluthrin	Pesticide	FR–PCB–Pest
Cyhalothrin	Pesticide	FR–PCB–Pest
Tefluthrin	Pesticide	FR–PCB–Pest
Pentachloroanisole (pca)	Pesticide	FR–PCB–Pest
Hexachlorobenzene (hcb)	Pesticide	FR–PCB–Pest
Pentachloronitrobenzene	Pesticide	FR–PCB–Pest
dcpa (dacthal)	Pesticide	FR–PCB–Pest
Octachlorostyrene	Pesticide	FR–PCB–Pest
Oxychlordane	Pesticide	FR–PCB–Pest
Benfluralin (benefin)	Pesticide	FR–PCB–Pest
ddd, p,p′	Pesticide	FR–PCB–Pest
dde, p,p′	Pesticide	FR–PCB–Pest
Nonachlor, cis	Pesticide	FR–PCB–Pest
Nonachlor, trans	Pesticide	FR–PCB–Pest
Endosulfan sulfate, solids	Pesticide	FR–PCB–Pest
Chlordane, cis	Pesticide	FR–PCB–Pest
Chlordane, trans	Pesticide	FR–PCB–Pest
ddt, p,p′	Pesticide	FR–PCB–Pest
Dieldrin	Pesticide	FR–PCB–Pest
Endosulfan i	Pesticide	FR–PCB–Pest
Endosulfan II, solids	Pesticide	FR–PCB–Pest
Atrazine	Pesticide	FR–PCB–Pest
Bromacil	Pesticide	FR–PCB–Pest

(*continued on next page*)

Appendix A (*continued*)

Chemical	Compound category	Summary classification
Carbazole	Pesticide	FR–PCB–Pest
Chlorpyrifos	Pesticide	FR–PCB–Pest
Diazinon	Pesticide	FR–PCB–Pest
Metolachlor	Pesticide	FR–PCB–Pest
Prometon	Pesticide	FR–PCB–Pest
4-Cumylphenol	Surfactant	Indus–PCP–PAH
4-N-octylphenol	Surfactant	Indus–PCP–PAH
4-Nonylphenol monoethoxylate, total, np1eo	Surfactant	Indus–PCP–PAH
4-Octylphenol diethoxylate-(opeo2)	Surfactant	Indus–PCP–PAH
4-Octylphenol monoethoxylate-(opeo1)	Surfactant	Indus–PCP–PAH
4-Tert-octylphenol	Surfactant	Indus–PCP–PAH
Nonylphenol, diethoxy-(total,npeo2)	Surfactant	Indus–PCP–PAH
Para-nonylphenol (total)	Surfactant	Indus–PCP–PAH

References

Alvarez DA, Perkins S, Nilsen E, Morace J. Spatial and temporal trends in occurrence of emerging and legacy contaminants in the lower Columbia River 2008-2010. Sci Total Environ 2014;484:322–30. [in this issue].

Anderson PD, Denslow ND, Drewes JE, Olivieri AW, Schlenk D, Scott GI Snyder SA. Monitoring strategies for chemicals of emerging concern (CECs) in California's aquatic ecosystems. *Recommendations of a Science Advisory Panel*, Southern California Coastal Water Research Project Report Technical Report 692; 2012.

Ankley GT, Lodge K, Call D, Balcer MD, Brooke LT, Cook PM, et al. Integrated assessment of contaminated sediments in the Lower Fox River and Green Bay, Wisconsin. Ecotoxicol Environ Saf 1992;23:46–63.

Berenbrock Charles, Tranmer AW. Simulation of flow, sediment transport, and sediment mobility of the Lowers Coeur d'Alene River, Idaho. U.S. Geological Survey Scientific Investigations Report 2008–5093; 2008 [164 pp.].

Bopp RF, Simpson HJ, Olsen CR, Trier RM, Kostyk DN. Chlorinated hydrocarbons and radionuclide chronologies in sediments of the Hudson River and estuary, New York. Environ Sci Tech 1982;16:666–76.

Brush GS. Rates and patterns of estuarine sediment accumulation. Limnol Oceanogr 1989; 34:1235–46.

Burkhardt MR, Zaugg SD, Burbank TL, Olson MC, Iverson JL. Pressurized liquid extraction using water/isopropanol coupled with solid-phase extraction cleanup for semivolatile organic compounds, polycyclic aromatic hydrocarbons (PAH), and alkylated PAH homolog groups in sediment. Anal Chim Acta 2005;549:104–16.

Burkhardt MR, Zaugg SD, Smith SG, ReVello RC. Determination of wastewater compounds in sediment and soil by pressurized solvent extraction, solid-phase extraction, and capillary-column gas chromatography/mass spectrometry. U.S. Geological Survey Techniques and Methods, Report 5-B2; 2006. p. 40.

Carver RE. Procedures in Sedimentary Petrography. New York: John Wiley and Sons; 1971 [653 pp.].

Christiansen H, Mehinto AC, Yu F, Perry RW, Denslow ND, Maule AG, Mesa MG. Correlation of gene expression and contaminant concentrations in wild largescale suckers: A field-based study. USA. Sci Total Environ 2014;484:379–89. [in this issue].

Clarke KR, Gorley RN. PRIMER ver. 6—User manual: Plymouth Routines in Multivariate Ecological Research. PRIMER-E, Ltd.; 2006 [190 pp.].

Cochran WG. Sampling Techniques. 3rd ed. New York: John Wiley& Sons, Inc.; 1977.

Dyer KR. Sediment processes in estuaries: future research requirements. J Geophys Res 1989;C10:14327–40.

Elias EPL, Cleveringa J, Buisman MC, Stive MJF, Roelvink JA. Field- and model data analysis of sand transport patterns in Texel Tidal inlet (the Netherlands). Coast Eng 2006; 53(5-6):505–29. http://dx.doi.org/10.1016/j.coastaleng.2005.11.006.

Elias E, Gelfenbaum G, Van der Westhuysen A. Validation of a coupled wave-flow model in a high-energy setting: the Mouth of the Columbia River. J Geophys Res Oceans 2012;117:C9.

Folk RL. A review of grain-size parameters. Sedimentology 1966;6:73–93.

Folk RL. Petrology of Sedimentary Rocks. Austin, TX: Hemphill; 1974. p. 1–159.

Fuhrer GJ, Tanner DQ, Morace JL, McKenzie SW, Skach KA. Water quality of the Lower Columbia River basin—analysis of current and historical water-quality data through 1994. U.S. Geological Survey Water-Resources Investigations Report 95-4294; 1996 [157 pp., http://or.water.usgs.gov/pubs_dir/Pdf/columbia_bistate.pdf].

Gelfenbaum G, Buijsman MC, Sherwood CR, Moritz HR, Gibbs AE. Coastal evolution and sediment budget at the mouth of the Columbia River, USA. Proceedings of Coastal Dynamics '01, ASCE; 2001. p. 818–27.

Grunnet NM, Walstra D-JR, Ruessink BG. Process-based modeling of a shoreface nourishment. Coast Eng 2004;51:581–607.

Henny CJ, Kaiser JL, Grove RA, Johnson BL, Letcher RJ. Polybrominated diphenyl ether flame retardants in eggs may reduce reproductive success of ospreys in Oregon and Washington, USA. Ecotoxicology 2009;18:802–13.

Henny CJ, Grove RA, Kaiser JL, Johnson BL, Furl CV, Letcher RJ. Wastewater dilution index partially explains observed polybrominated diphenyl ether flame retardant concentrations in osprey eggs from Columbia River Basin, 2008–2009. Ecotoxicology 2011; 20:682–97.

Hermann RM. Polychlorinated Biphenyl Presence in the Columbia River Corridor. Washington Closure Hanford, report WCH-208; 2008.

Hirschberg DJ, Chin P, Feng H, Cochran JK. Dynamics of Sediment and Contaminant Transport in the Hudson River Estuary: Evidence from Sediment Distributions of Naturally Occurring Radionuclides Estuaries. 1996;19(4):931–49. [Dedicated Issue: The Hudson River Estuary].

Hites RA, LaFlamme RF, Farrington JW. Sedimentary polycyclic aromatic hydrocarbons: the historical record. Science 1977;198:829–31.

Hydroqual. A model for the evaluation and management of contaminants of concern in water, sediment, and biota in the NY/NJ Harbor Estuary, sediment transport/organic carbon production sub-model. Contamination Assessment and Reduction Project (CARP); 2007 [1383 pp.].

Ippen AT. Estuary and Coastline Hydrodynamics. New York: McGraw-Hill; 1966.

Johnson LL, Anulacion BF, Arksooh M, Buzitis J, Bolton J, Boyd D, et al. Chemical contaminants and endangered salmon in the Lower Columbia River and Estuary—results of the fish monitoring component of the Lower Columbia River Water Quality Monitoring Project, 2004–2006. In: Lower Columbia River Estuary Partnership, editor. Seattle, WA: Northwest Fisheries Science Center; 2006.

Johnson LL, Ylitalo GM, Sloan CA, Anulacion BF, Kagley AN, Arkoosh MR, et al. Persistent organic pollutants in outmigrant juvenile chinook salmon from the Lower Columbia Estuary, USA. Sci Total Environ 2007;374:342–66.

Junk WJ, Bayley PB, Sparks RE. The flood pulse concept in river-floodplain systems. In: Dodge DP, editor. Proceedings of the International Large River Symposium (LARS), 14–21 September 1986. Canadian special publication of fisheries and aquatic sciences number 106. Ottawa, Ontario, Canada.: Department of Fisheries and Oceans; 1989. p. 110–27.

Klingbeil AK, Sommerfield CK. Latest Holocene evolution and human disturbance of a channel segment in the Hudson River Estuary. Mar Geol 2005;218:135–53.

Klosterhaus S. Update on contaminants of emerging concern. Regional Monitoring Program for Water Quality in the San Francisco Bay Estuary. Oakland, CA: San Francisco Estuary Institute; 2010.

Lesser GR, Roelvink JA, Van Kester TM, Stelling GS. Development and validation of a three-dimensional morphological model. Coast Eng 2004;51:883–915. http://dx.doi.org/10.1016/j.coastaleng.2004.07.014.

Levin D, Lillycrop WJ, Alexander MP. Sand Waves: Sand wave shoaling in navigation channels. Waterways Experiment Station Technical Report HL-90-17; 1992 [56 p.].

Maruya KA, Vidal-Dorsch DE, Bay SM, Kwon JW, Xia K, Armbrust KL. Organic contaminants of emerging concern in sediments and flatfish collected near outfalls discharging treated wastewater effluent to the southern California Bight. Environmental Toxicology and Chemistry 2012;31:2683–8.

McCarthy KA, Gale RW. Investigation of the distribution of organochlorine and polycyclic aromatic hydrocarbon compounds in the lower Columbia River using semipermeable membrane devices U.S. Geological Survey, Prepared in cooperation with the Lower Columbia River Estuary Program and the National Stream Quality Accounting Network Program. Water-Resources Investigations Report 99–4051; 1999 [136 pp.].

Morace JL. Water-quality data, Columbia River Estuary, 2004–05. U.S. Geological Survey Data Series, 213; 2006 [http://pubs.water.usgs.gov/ds213].

Mulligan RP, Hay AE, Bowen AJ. A wave-driven jet over a rocky shoal. J Geophys Res 2010. http://dx.doi.org/10.1029/2009JC006027. [115 pp., C10038].

Naik PK, Jay DA. Human and climate impacts on Columbia River hydrology and salmonids. River Res Appl 2011;27(10):1270–6.

Nakata H, Sasaki H, Takemura A, Yoshioka M, Tanabe S, Kannan K. Bioaccumulation, temporal trend, and geographical distribution of synthetic musks in the marine environment. Environ Sci Technol 2007;41:2216–22.

Nilsen EB, Morace J. Foodweb transfer, sediment transport, and biological impacts of emerging and legacy organic contaminants in the lower Columbia River, Oregon and Washington, USA: USGS Contaminants and Habitat (ConHab) Project. Sci Total Environ 2014;484:319–21. [in this issue].

Nilsen EB, Furlong E, Rosenbauer R. Reconnaissance of pharmaceuticals and wastewater indicators in streambed sediments of the lower Columbia River basin, Oregon and Washington. USA. J Am Water Resour Assoc 2014;50(2):219–301.

Nilsen EB, Zaugg S, Alvarez D, Morace J, Waite I, Counihan T, et al. Contaminants of legacy and emerging concern in largescale sucker (*Catostomus macrocheilus*) and the foodweb in the lower Columbia River, Oregon and Washington, USA. Sci Total Environ 2014;484:344–52. [in this issue].

Nitsche FO, Bell RE, Carbotte SM, Ryan WBF, Flood RD, Ferrini V, et al. Integrative acoustic mapping reveals Hudson River sediment processes and habitats. EOS Trans Am Geophys Union 2005;86:225–9.

Nitsche FO, Kenna TC, Haberman M. Quantifying 20th century deposition in complex estuarine environment: an example from the Hudson River. Estuar Coast Shelf Sci 2010;89(2010):163–74.

NMFS (National Marine Fisheries Service). Endangered Species Act - Section 7(a)(2) Supplemental Biological Opinion. Consultation on remand for operation of the Federal Columbia River Power System. NWR-2013-9562; January 17, 2014 [610 pp.].

Olsen CR, Simpson HJ, Bopp RF, Williams SC, Peng TH, Deck BL. A Geochemical analysis of the sediments and sedimentation in the Hudson Estuary. J Sediment Petrol 1978; 48(2):401–18.

Olsen CR, Cutshall NH, Larsen IL. Pollutant-particle associations and dynamics in coastal marine environments: a review. Mar Chem 1982;11:501–33.

Olsen CR, Larsen IL, Brewster RH, Cutshall NH, Bopp RF, Simpson HJ. A Geochemical assessment of sedimentation and contaminant distributions in the Hudson-Raritan Estuary. National Oceanic and Atmo-spheric Administration Technical Report OMS NOS 2, Rock-ville, Maryland; 1984 [101 pp.].

Olsen CR, Larsen IL, Mulholland PJ, Von Damm KL, Grebmeier JM, Schnaffner LC, et al. The concept of an equilibrium surface applied to particle sources and contaminant distributions in estuarine sediments. Estuaries 1993;16:683–96.

Oregon Department of Environmental Quality. Regional Environmental Monitoring and Assessment Program: Lower mid-Columbia River Ecological Assessment Final Report: 2009. Publication No. 12/LAB/006 ; 2012 [219 pp.].

Oros DR, Ross JRM. Polycyclic aromatic hydrocarbons in San Francisco Estuary sediments. Mar Chem 2004;86:169–84.

R. Development Core Team. R: A Language and Environment for Statistical Computing. Version 2.13.1. Vienna, Austria: R Foundation for Statistical Computing; 2007. http://www.R-project.org].

Schropp SJ, Lewis FG, Windom HL, Ryan JD, Calder FD, Burney LC. Interpretation of metal concentrations in estuarine sediments of Florida using aluminum as a reference element. Estuaries 1990;13:227–35.

Schubel JR. Turbidity maximum of the northern Chesapeake Bay. Science 1968;161: 1013–6.

Schubel JR, Meade RH. Man's impact on estuarine sedimentation. In Estuarine Pollution, Control, and Assessment. Washington, D.C.: United States Environmental Protection Agency; 1977. p. 193–209.

Sharp JH, Culberson CH, Church TM. The chemistry of the Delaware Estuary: general considerations. Limnol Oceanogr 1982;27:1015–28.

Shaw Environmental & Infrastructure, Inc., Anchor Environmental, LLC, Foth & Van Dyke, and Limno-Tech. Final basis of design report. Prepared for: Fort James Operating Company, Inc. and NCR Corporation. For Submittal to: Wisconsin Department of Natural Resources and U.S. Environmental Protection Agency, 2006, 271 pp.

Simenstad CA, Burke JL, O'Connor JE, Cannon C, Heatwole DW, Ramirez MF, et al. Columbia River Estuary ecosystem classification—concept and application. U.S. Geological Survey Open-File Report 2011-1228; 2011 [54 pp. http://pubs.usgs.gov/of/2011/1228/].

Smith JN, Levy EM. Geochronology for poly-cyclic aromatic hydrocarbon contamination in sediments of the Saguenay Fjord. Environ Sci Tech 1990;24:874–9.

Steuer JJ, Jaeger S, Patterson D. A deterministic PCB transport model for the Lower Fox River between Lake Winnebago and DePere, Wisconsin. WR: Wisconsin Department of Natural Resources, Publ.; 1995. p. 389–95 [283 pp.].

Stevens Jr DL, Olsen AR. Spatially-balanced sampling of natural resources. J Am Stat Assoc 2004;99(465):262–78.

Sugai SF. Transport and sediment accumulation of 210Pb and '37Cs in two southeast Alaskan fjords. Estuaries 1990;13:380–92.

Tanaka N, Turekian KK, Rye DM. The radiocarbon, b'3C, 21'Pb, and '37Cs record in box cores from the continental margin of the Middle Atlantic Bight. Am J Sci 1991;291:90–105.

Thorp JH, Delong MD. Dominance of autochthonous autotrophic carbon in food webs of heterotrophic rivers. Oikos 2002;96:543–50.

Torres L, Nilsen E, Grove R, Patiño R. Health condition of largescale sucker (*Catostomus macrocheilus*) collected along a contaminant gradient in the Lower Columbia River, Oregon and Washington, USA. Sci Total Environ 2014e. [in this issue].

Troup BH, Bricker OP. Processes affecting the transport of materials from continents to oceans. In: Church TM, editor. Marine Chemistry in the Coastal Environment. Washington, D.C: American Chemical Society; 1975. p. 133–57.

Turekian KK. Fate of metals in estuaries. Estuaries, Geophysics and the Environment. Washington, D.C.: National Academy of Sciences; 1977. p. 121–7.

Turekian KK, Ccohran JK, Benninger LK, Aller RC. The sources and sinks of nuclides in Long Island Sound. In: Saltzman B, editor. Estuarine Physics and Chemistry: Studies in Long Island Sound. Advances in GeophysicsNew York: Academic Press; 1980. p. 129–64.

USEPA. National Study of Chemical Residues in Fish, EPA 823-R-92-008a; 1992.

USEPA. Ecological Condition of the Columbia River Estuary: an Environmental Monitoring and Assessment Program (EMAP) Report. U.S. Environmental Protection Agency Region 10; 2007 [EPA 910-R-07-004 2007; 48 pp.].

USEPA. Columbia River Basin: State of the River Report for Toxics, EPA 910-R-08-004; 2009 [60 pp.].

USEPA. Columbia River Toxics Reduction Action Plan. Prepared by: U.S. Environmental Protection Agency, Region 10 and the Columbia River Toxics Reduction Working Group. U.S. Environmental Protection Agency Region 10; 2010. 20 p.

Van Maren DS. Morphodynamics of a Cyclic Prograding Delta: The Red River, Vietnam. PhD thesis Utrecht, Netherlands: Utrecht Univ.; 2004.

van Rijn LC. Principles of Sediment Transport in Rivers. Amsterdam, The Netherlands: Estuaries and.Coastal Seas, Aqua Publications; 1993 [650 pp.].

van Rijn LC, Walstra DJRM, van Ormondt M. Description of TRANSPOR2004 and Implementation in Delft3D-ONLINE, WL |Delft Hydraulics Report, Z3748.00; 2004 [77 pp.].

Venkatesan MI, Kaplan IR. Sedimentary coprostanol as an index of sewage addition in Santa Monica Basin, Southern California. Environ Sci Technol 1990;24:208–14.

Zaugg SD, Burkhardt MR, Burbank TL, Olson MC, Iverson JL, Schroeder MP. Determination of semivolatile organic compounds and polycyclic aromatic hydrocarbons in solids by gas chromatography/mass spectrometry. U.S. Geological Survey Techniques and Methods; 2006 [book 5, chap. B3, 44 pp.].

Zeug SC, Winemiller KO. Evidence supporting the importance of terrestrial carbon in a large river food web. Ecology 2008;89(6):1733–43.