# Impacts of Dredging Activities on the Accumulation of Dioxins in Surface Sediments of the Houston Ship Channel, Texas

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## ABSTRACT

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The Houston Ship Channel (HSC) and upper Galveston Bay (GB), Texas, are known to be contaminated with dioxins (polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans), the majority of which are associated with sediments. Since 1914, dredging operations to establish, sporadically expand, and consistently maintain a navigable channel for large ships has been and remains continuous here. The objectives of this research focus on determining if dredging activities have any significant impact on the quantities of dioxins associated with surface sediments in the HSC and GB. Four transects were sampled, located on the dredged and undredged sides of two dredge-spoil islands. Sediment samples were characterized in terms of their organic carbon contents, grain size fractions, indicator dioxin concentrations (2,3,7,8-tetrachlorinated dibenzo-*p*-dioxin [TCDD], 2,3,7,8-tetrachlorodibenzofuran [TCDF], and toxic equivalents), and fallout radionuclide activities. The physical and geochemical data were examined using a combination of principal components analysis and one-way analysis of variance. Results of the statistical tests show that (1) sedimentary dioxin concentrations are significantly higher adjacent to the northernmost dredge-spoil island, which is located closer to a recently identified dioxin point source (San Jacinto waste pits); and (2) while mean sedimentary dioxin concentrations were slightly higher for undredged as compared to dredged transect samples as a whole, these differences were not significant ($p > 0.05$). Mean fallout isotope ratio values ($^7Be/^{137}Cs$, $^7Be/^{210}Pb_{xs}$) were greater for the dredged sample population, indicating that dredged sediments contain more of the shorter-lived radionuclides (*i.e.,* $^7Be$), as their buildup is more rapid after dredging than those isotopes with longer half-lives ($^{210}Pb$) or those for which a longer time has passed since introduction into the environment ($^{137}Cs$).

**ADDITIONAL INDEX WORDS:** *Dioxin, sediment, radionuclides, dredging, ship channel.*

## INTRODUCTION

Members of the organochlorine class of compounds are generally characterized by their environmental persistence, their ability to be transported long distances in the atmosphere or in surface waters, a tendency to bioaccumulate, and, for many, being toxic to many types of organisms, including humans. Twelve of the most toxic of these compounds (or groups of compounds) were identified at the United Nation's Stockholm Convention (2001) as persistent organic pollutants (POPs), resulting in a multinational agreement to implement plans aimed to reduce and ultimately eliminate anthropogenically sourced POPs. Dioxins are a family of compounds (polychlorinated dibenzo-*p*-dioxins [PCDDs] and polychlorinated dibenzofurans [PCDFs]) that were among those POPs identified.

Seven of the seventy-five PCDD congeners and ten of the one hundred thirty-five PCDF congeners are known to be highly toxic (Liem, 1997). These compounds cause a variety of biochemical, immunological, and reproductive effects in animals and are suspected human carcinogens (Bailar, 1991; Krishnan *et al.*, 1995; Poland and Knudson, 1982; Safe, 1986). Dioxins bioaccumulate in the aquatic food chain and consequently pose a significant threat to human health (USEPA, 2000). While dioxins are known to be produced naturally from processes including forest fires and have been quantified in sediment and soil samples which predate the industrial revolution (*e.g.,* Alcock *et al.*, 1998; Green *et al.*, 2001; Green *et al.*, 2004), anthropogenic sources, particularly since the 1920s, have become much more significant. Primary contemporary sources of dioxins include metal smelting (Quass, Fermann, and Broker, 2004), herbicide production (Stellman *et al.*, 2003), pulp and paper mills, and waste incineration (Fiedler, 1996; Hagenmaier *et al.*, 1992; Sonnenberg and Nichols, 1995; Wilken *et al.*, 1992).

Elevated concentrations of dioxins were detected in fish and crabs from the Houston Ship Channel (HSC) in 1990 (Crocker

and Young, 1990). As a result, the Texas Department of State Health Services issued a seafood consumption advisory for the HSC and upper Galveston Bay (GB), which is still in effect. The HSC was subsequently placed, and remains, on the Texas §303(d) list of impaired water bodies, as required by the federal Clean Water Act (1977). Results from a total maximum daily load (TMDL) study (2002–2003) showed that on average, dioxin concentrations exceeded the Texas surface water quality standard (0.093 pg L$^{-1}$) in >80% of all samples (sediment, fish, and crabs), and that 2,3,7,8-TCDD (tetrachlorinated dibenzo-*p*-dioxin) was the major contributor to total toxic equivalents (TEQs) in all samples (Rifai, Palachek, and Jensen, 2003). Toxic equivalents are calculated to be the sum of the seventeen 2,3,7,8-substituted dioxin congeners on the basis of their toxic equivalence to 2,3,7,8-TCDD. More recent work has shown that TEQ concentrations in suspended and bottom sediments remain elevated and that bottom sediments are the most significant dioxin reservoir (Suarez *et al.*, 2005; Yeager *et al.*, 2007). The central and western portions of the HSC have the highest concentration of industrial sources (pulp and paper mills) and the highest concentrations of dioxins (Suarez *et al.*, 2005; Yeager *et al.*, 2007). Because of their hydrophobicity, dioxins are thought to rapidly partition to organic carbon in suspended solids and become buried (Broman, Naf, and Zebuhr, 1992; Muir *et al.*, 1992; Servos, Muir, and Webster, 1992). Sediment resuspension, such as that associated with dredging activities, may reintroduce dioxins into the food chain; however, this process has not been thoroughly investigated.

This work expands upon research focused on the fate and impacts of dioxins in this system, including their bioaccumulation in waterfowl (Frank *et al.*, 2001) and representative benthic fauna (Suarez *et al.*, 2005); their dry and wet atmospheric deposition (Correa *et al.*, 2006; Raun *et al.*, 2005); their distributions in the water column, suspended sediments, and bottom sediments (Suarez *et al.*, 2006); and their accumulation chronologies, sedimentary inventories, and associations (Yeager *et al.*, 2007). The preponderance of evidence from these and other studies clearly shows that the HSC and upper GB system is significantly contaminated with dioxins, a fact reinforced by the listing of the San Jacinto River waste pits (a likely major, contemporary point source) on the U.S. Environmental Protection Agency's National Priorities List of Superfund sites in 2008 (USEPA, 2009). A considerable body of research shows that dredging operations, *via* the resuspension of large quantities of bottom sediment, can result in marked increases in particulate matter–associated contaminant concentrations in overlying or adjacent waters; often this occurs with corresponding deleterious impacts on organisms that occupy those areas (*e.g.,* Bellas *et al.*, 2007; Bocchetti *et al.*, 2008; Engwall *et al.*, 1998; Sundberg *et al.*, 2007; Sturve *et al.*, 2005). This research is focused on the impacts of channel-maintenance dredging on the accumulation of dioxins in surface sediments. To the best of our knowledge, dredging is not currently being used as a remediation mechanism anywhere in the HSC or GB.

We tested the hypothesis that active removal, by dredging, of surface sediments contaminated with dioxins results in a net decrease in the amount of dioxins found in HSC surface sediments. The null hypothesis is that dredging results in no

significant reduction in the amount of dioxins in HSC surface sediments.

## STUDY SITE

Galveston Bay is the largest estuary in Texas and seventh largest in the United States. It is shallow (approximately 7–9 ft) and covers approximately 600 mi$^2$ (Figure 1); most of the bay's freshwater is supplied from the Trinity (83%) and San Jacinto (8%) rivers (Wen *et al.*, 1999). The Port of Houston is a 25-mile-long complex of public and private facilities in close proximity to the Gulf of Mexico, and it is situated on the NW end of the 52-nautical-mile-long HSC (Figure 1). The Port of Houston is among the busiest in the nation; as of 2006, in excess of 200 million tons of cargo moved through the port, and it is estimated that approximately \$118 billion of statewide economic impact is realized due to ship channel–related business activities (Port Of Houston Authority, 2009). Since the opening of the HSC in 1914, dredging operations to establish, sporadically expand, and consistently maintain a navigable channel for large ships has been and remains continuous. Predominant dredging techniques currently in use in the HSC include both mechanical and hydraulic cutter suction. Between 2002 and 2007, the U.S. Army Corps of Engineers enlarged the HSC from a 40 ft (12.2 m) depth by 400 ft (122 m) width to a 45 ft (13.7 m) depth by 530 ft (162 m) width (Tate and Berger, 2006). The impacts of this expansion on HSC sediment dynamics and the adequacy of existing dredged-sediment storage volumes are both issues of ongoing investigation.

Four sample transects were chosen for this research, located on the dredged and undredged sides of Hog and Alexander islands (Figure 1), two of many dredge-spoil islands that have been created since the opening of the HSC. This series of islands stretches from the lower San Jacinto River, just below the Interstate 10 overpass, through the HSC and Port of Houston facilities between the cities of Houston to the west and Baytown to the east; the islands continue to the south a considerable distance into Galveston Bay. These transect sites were chosen due to their location north of Galveston Bay, the likelihood that representative quantities of dioxins would be present in sediments there, and the physical proximity of dredged versus undredged portions of the system and their relationship to each other (*i.e.,* sediments resuspended from dredging or ship traffic on the dredged side cannot be deposited on the undredged side).

## METHODS

### Sediment Sampling, Sedimentology, and Organic Matter

Surface sediment samples were collected in August 2007 using a small, ponar-type sampler. Care was taken to collect only the upper 3–4 cm of sediment at each station. All sediment samples were stored at 4°C until processing and analyses. Aliquots of each sample were then separated to provide materials for grain size and particulate organic carbon (POC) determinations and dioxin and radiochemistry analyses. For the determination of grain size, samples were dried at approximately 75°C for 24 hours, gently disaggregated, and
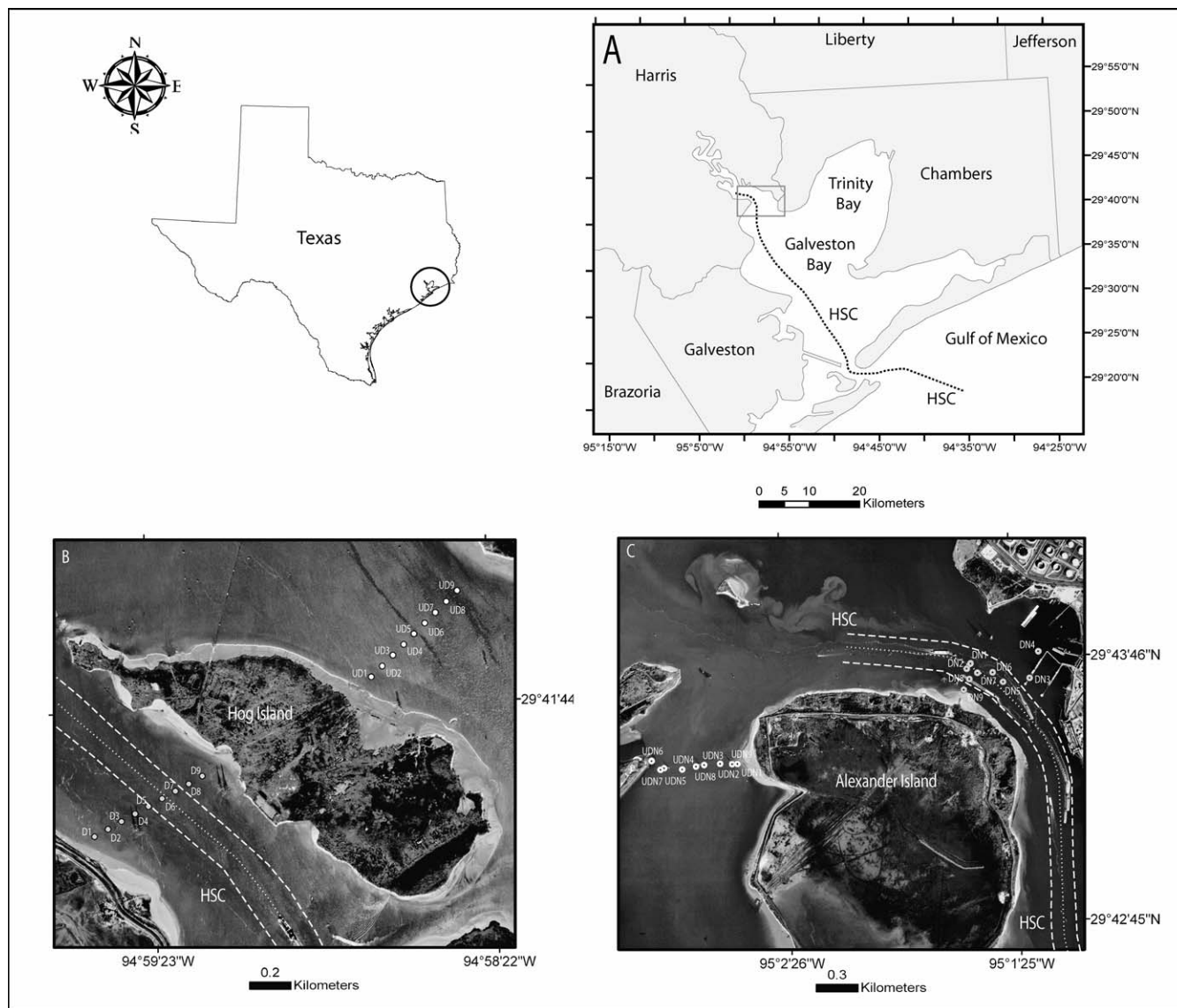
Figure 1. Houston Ship Channel study areas, primary dredged channels, dredged (red) and undredged (white) transects, and sampling stations.

passed through 2 mm and 0.5 mm sieves. Grain size distributions (sand, silt, clay) were determined by the standard Folk settling method. Sedimentary POC was determined by first treating samples with HCl acid fumes to destroy any carbonates present (Harris, Horwath, and Van Kessel, 2001; Hedges and Stern, 1984) followed by elemental analyses using a 4010 CHN/SO Analyzer (Costech Analytical Technologies, Valencia, California).

## Determination of Sedimentary Dioxins

Sample aliquots for dioxin determination were stored in precleaned amber glass jars with a Teflon seal at $4°C$ until analysis. Dioxins were quantified by high resolution gas chromatography/high resolution mass spectrometry using Environmental Protection Agency Method 1613B at a commercial laboratory (Uni-Prove Technology). Samples were homogenized, spiked with fifteen $^{13}C_{12}$-labeled PCDD/PCDF internal standards, and extracted using Soxhlet extraction. The extract was evaporated to 10 mL, with 9 mL loaded on the gel permeation chromatograph (GPC) and 1 mL taken for lipid determination. The GPC extracts were concentrated to 1 mL for cleanup. Extracts were spiked with a $^{37}Cl_4$-labeled 2,3,7,8-TCDD enrichment efficiency standard and subjected to acid/base washes and multilayer silica, alumina, and carbon column cleanup procedures. After cleanup, extracts were concentrated and spiked with recovery standards ($^{13}C_{12}$-1,2,3,4-TCDD and $^{13}C_{12}$-1,2,3,7,8,9-Hexachlorodibenzo-p-dioxin) prior to injection. Chromatographic separation was achieved with a DB-5 capillary chromatography column (60 m, 0.25 mm internal diameter, 0.25 μm film

thickness). A second column DB-225 (30 m, 0.25 mm internal diameter, 0.25 μm film thickness) was used for confirmation of tetrachlorodibenzofuran (TCDF) identification.

## Radiochemistry

High-resolution gamma spectrometry was employed to resolve $^7$Be (t$_{1/2}$ = 53 d, E$\gamma$ = 477 keV) and $^{137}$Cs (t$_{1/2}$ = 30 y, E$\gamma$ = 661 keV) using HPGe well detectors and a DSA 1000 multichannel analyzer (Canberra, Meriden, Connecticut). Samples were contained in plastic test tubes (inner diameter 1.3 cm, height 9.4 cm), and standards ($^{137}$Cs: NIST, SRM #4357; $^7$Be: Isotope Products Laboratories CN #6007, Valencia, California) were prepared and run on each detector in geometries identical to those for sediment samples to determine representative efficiencies. Efficiency errors based on standards were less than ±2%; samples were counted, on average, for 3–4 days to reach a standard deviation (SD) for both isotopes on the order of approximately 3–5%, and activities were decay-corrected to the date of collection.

Alpha spectrometry was employed to resolve $^{210}$Pb (t$_{1/2}$ = 22.4 y) *via* $^{210}$Po using a 7200 integrated alpha spectrometer (Canberra). Lead-210 samples were spiked with a certified $^{209}$Po tracer (Isotope Products Laboratories #6209-100N) and then leached using the method described by Aalto and Dietrich (2005). In summary, 20 mL of concentrated HNO$_3$ was added to samples over low heat to near dryness, followed by 20 mL of 6 N HCl over low heat to near dryness. Samples were then brought up in 40 mL of 1.5 N HCl and centrifuged. The supernatant was decanted, and ascorbic acid was added to bind free Fe(III). Finally, a silver disk was added to the solution over heat to provide a substrate for the spontaneous deposition of polonium isotopes (Santschi *et al.*, 1999; Yeager *et al.*, 2004; Yeager, Santschi, and Rowe, 2007). This method selectively removes the $^{210}$Po daughter (assumed to be in equilibrium with $^{210}$Pb) from the grain exterior to measure only mobile, exogenic, unsupported $^{210}$Pb activity, not endogenic activity, which can represent significant noise in radium-bearing sediments (Brenner, Schelske, and Kenney, 2004).

## Data Analysis

In order to address our hypothesis, we first examined the physical and geochemical data sets. Kolmogorov-Smirnov one-sample tests were performed against the normal distribution on data pooled across all 36 sampling sites using two forms of the three sediment dioxin variables: untransformed and log$_{10}$-transformed. All three dioxin variables fit the normal distribution much better in their log-transformed forms, so these values were used for statistical analyses. Eight nondetect (ND) values for dioxins (five TCDD and three TCDF) were replaced by appropriately small values (*i.e.,* mean − 2.5 SD) for the log-transformed variables. Kolmogorov-Smirnov values for all three log-transformed dioxin variables were nonsignificant ($p = 0.29 - 0.75$) relative to the normal distribution. The POC data were not different from normal ($p = 0.79$) in their raw form (mg/g) and thus were not transformed. As percent values, the three sediment grain size variables were used in their arcsin-square root–transformed form for statistical analyses.

Because 14 of 21 bivariate correlations were significant (unadjusted $p < 0.05$), and at least one bivariate correlation was significant for every one of the test variables (three log-transformed dioxin variables, raw POC data, and three arcsin-square root–transformed grain size variables), a principal components analysis (PCA) was conducted. The PCA used the correlation matrix and employed a varimax rotation to maximally resolve the contributions of the original variables to respective orthogonal composite variables (PCA axes). Only those principal components having associated eigenvalues greater than 1 (representing more than one variable worth of information) were considered meaningful. Because these data originated from transects located in four distinct spatial areas, representing combinations of undredged or dredged conditions on opposing sides of two islands (Figure 1), one-way analysis of variance (ANOVA) was performed on differences in a transect factor representing four levels: Hog Island, undredged and dredged; and Alexander Island, undredged and dredged. One-way ANOVAs were run for each of the first two PCA components, which represented most of the meaningful variation in the seven original variables. Levene's tests of equality of error variances among groups accompanied these ANOVAs, which were done separately, as the two PCA components were orthogonal. In addition to the one-way ANOVAs, two customized, special contrasts examined whether PCA scores for the two components were different with respect to islands or with respect to dredging. Since the two special contrasts as well as the two responses (PCA axes) were orthogonal, no corrections of significance levels were required.

## RESULTS AND DISCUSSION

Tabular data for all variables are presented in Table 1. Only the most toxic of the dioxin congeners (2,3,7,8-TCDD and 2,3,7,8-TCDF) and TEQ values are discussed here. The maximum contaminant level (MCL) concentration for 2,3,7,8-TCDD in drinking water is 0.03 parts per trillion (ppt; USEPA, 2008). Applying this standard to sediment (in pg/g) indicates that concentrations of 2,3,7,8-TCDD are significantly above this level in surface sediments (≤4 cm) at all stations where it was detected (Table 1 and Figure 2). The mean 2,3,7,8-TCDD, 2,3,7,8-TCDF, and TEQ values for the undredged sample population are consistently slightly higher than those for the dredged sample population (Figure 2).

Sedimentary concentrations of these dioxin congeners and TEQs were generally higher at transects near Alexander Island than at those near Hog Island; they were somewhat higher at undredged than at dredged transects (Figure 2, Table 2). As indicated by associated, meaningful eigenvalues, the first two PCA dimensions effectively explain 83.5% of the variation in the seven test variables (50% and 33.5% for rotated principal component 1 [PC$_1$] and principal component 2 [PC$_2$], respectively). As shown by PC loadings as a reflection of the magnitudes of associated correlations, PC$_1$ mainly represents joint variation in the three dioxin variables as well as sediment clay content, whereas PC$_2$ represents orthogonal joint variation in POC as well as sediment silt and sand contents (Table 3). High scores on PC$_1$ reflect relatively high amounts of sedimentary dioxins and clay; high scores on PC$_2$ reflect

Table 1. *Summary physical, geochemical, and radiochemical data for all stations. ND denotes concentrations below the detection limit for that compound.*

| Stations | Water Depth (m) | POC (mg/g) | % Sand | % Silt | % Clay | 2,3,7,8-TCDD (pg/g) | 2,3,7,8-TCDF (pg/g) | Total TEQ (pg/g) | $^7$Be (mBq/g) | $^{137}$Cs (mBq/g) | $^{210}$Pb$_{xs}$ (mBq/g) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| UD1 | 0.490 | 4.52 | 22.9 | 49.1 | 28.0 | 2.08 | 4.70 | 6.38 | 0 | 1.84 ± 0.23 | 20.3 ± 0.89 |
| UD2 | 0.610 | 3.71 | 36.7 | 35.3 | 28.0 | 2.07 | 4.78 | 6.32 | 5.78 ± 0.64 | 1.14 ± 0.11 | 17.8 ± 0.71 |
| UD3 | 0.610 | 3.31 | 51.4 | 28.8 | 19.8 | 1.75 | 4.85 | 5.42 | 4.64 ± 0.75 | 0.71 ± 0.07 | 15.4 ± 0.69 |
| UD4 | 0.870 | 4.63 | 42.9 | 28.5 | 28.6 | 2.71 | 12.1 | 8.44 | 7.83 ± 0.74 | 1.56 ± 0.14 | 22.5 ± 1.01 |
| UD5 | 0.900 | 7.31 | 3.90 | 46.5 | 49.6 | 4.36 | 10.9 | 12.5 | 1.82 ± 0.88 | 2.42 ± 0.23 | 37.6 ± 1.38 |
| UD6 | 0.660 | 7.84 | 8.70 | 41.5 | 49.8 | 3.60 | 10.2 | 12.9 | 0.89 ± 0.54 | 2.47 ± 0.23 | 42.8 ± 1.76 |
| UD7 | 0.570 | 8.79 | 2.00 | 42.2 | 55.8 | 3.39 | 12.0 | 12.3 | 2.84 ± 1.39 | 3.46 ± 0.36 | 45.8 ± 1.92 |
| UD8 | 0.410 | 9.16 | 2.40 | 40.5 | 57.1 | 3.20 | 10.7 | 13.7 | 2.40 ± 0.79 | 2.57 ± 0.27 | 50.3 ± 2.05 |
| UD9 | 0.310 | 5.85 | 53.0 | 15.8 | 31.2 | 1.89 | 5.11 | 6.60 | 29.1 ± 1.87 | 0.94 ± 0.10 | 25.5 ± 1.04 |
| D1 | 1.05 | 2.48 | 44.0 | 34.4 | 21.6 | 1.14 | 3.64 | 4.39 | 2.43 ± 0.30 | 1.02 ± 0.10 | 15.1 ± 0.52 |
| D2 | 1.36 | 0.95 | 85.6 | 6.00 | 8.40 | ND | 1.85 | 1.54 | 0 | 0 | 8.43 ± 0.32 |
| D3 | 2.57 | 2.79 | 38.8 | 30.8 | 30.4 | 2.03 | 5.04 | 7.02 | 6.93 ± 1.13 | 1.23 ± 0.12 | 23.8 ± 0.84 |
| D4 | 4.38 | 4.98 | 33.8 | 33.3 | 32.9 | ND | 6.06 | 5.24 | 5.59 ± 0.44 | 1.06 ± 0.09 | 32.4 ± 1.18 |
| D5 | 8.61 | 12.1 | 0.90 | 74.7 | 24.4 | 1.02 | 3.15 | 7.69 | 8.17 ± 1.98 | 4.16 ± 0.33 | 60.1 ± 2.13 |
| D6 | 11.57 | 9.81 | 4.30 | 65.8 | 29.9 | ND | 1.38 | 3.64 | 62.0 ± 5.74 | 2.80 ± 0.32 | 51.1 ± 1.80 |
| D7 | 9.48 | 9.06 | 16.7 | 76.6 | 6.70 | 0.628 | 1.64 | 5.77 | 43.8 ± 2.79 | 2.01 ± 0.21 | 43.8 ± 1.43 |
| D8 | 4.74 | 3.32 | 49.2 | 26.4 | 24.4 | 6.30 | 32.2 | 13.3 | 0 | 1.06 ± 0.09 | 17.7 ± 0.64 |
| D9 | 1.95 | 4.29 | 55.9 | 22.0 | 22.1 | 11.4 | 23.9 | 22.9 | 0 | 0.65 ± 0.06 | 13.7 ± 0.53 |
| UDN1 | 0.579 | 12.3 | 77.8 | 11.3 | 10.9 | ND | ND | 1.18 | 129 ± 5.81 | 4.03 ± 0.41 | 67.6 ± 3.95 |
| UDN2 | 1.13 | 5.35 | 52.7 | 16.5 | 30.8 | 4.10 | 10.9 | 8.44 | 7.02 ± 0.68 | 1.75 ± 0.20 | 28.2 ± 1.07 |
| UDN3 | 1.19 | 7.36 | 34.1 | 4.40 | 61.5 | 8.10 | 28.3 | 17.5 | 26.0 ± 2.09 | 0.67 ± 0.08 | 42.2 ± 2.25 |
| UDN4 | 1.28 | 2.66 | 1.50 | 26.3 | 72.2 | 13.6 | 28.9 | 26.5 | 5.74 ± 0.51 | 0.34 ± 0.04 | 16.3 ± 0.79 |
| UDN5 | 1.16 | 10.4 | 1.30 | 34.9 | 63.8 | 12.8 | 32.7 | 24.3 | 20.3 ± 1.83 | 1.84 ± 0.22 | 62.7 ± 2.95 |
| UDN6 | 0.579 | 6.30 | 37.4 | 26.6 | 36.0 | 2.90 | 8.90 | 5.60 | 63.8 ± 5.79 | 0.81 ± 0.09 | 32.8 ± 1.31 |
| UDN7 | 1.16 | 10.1 | 1.40 | 31.7 | 66.9 | 10.2 | 31.0 | 21.5 | 25.8 ± 1.52 | 1.72 ± 0.16 | 61.0 ± 2.45 |
| UDN8 | 1.31 | 11.1 | 6.10 | 28.2 | 65.7 | 9.20 | 69.0 | 43.9 | 27.9 ± 2.39 | 2.47 ± 0.25 | 69.9 ± 2.86 |
| UDN9 | 1.00 | 4.31 | 47.2 | 21.6 | 31.2 | 2.60 | 9.00 | 5.77 | 23.1 ± 2.12 | 0.21 ± 0.05 | 24.1 ± 1.05 |
| DN1 | 15.4 | 10.9 | 6.50 | 33.6 | 59.9 | 4.30 | 11.4 | 13.2 | 123 ± 7.82 | 2.73 ± 0.28 | 59.2 ± 2.80 |
| DN2 | 16.2 | 11.2 | 1.90 | 27.5 | 70.6 | 3.50 | ND | 12.0 | 84.0 ± 7.44 | 2.38 ± 0.26 | 60.8 ± 2.92 |
| DN3 | 13.9 | 12.5 | 3.10 | 26.9 | 70.0 | 5.20 | 11.3 | 15.9 | 60.2 ± 4.10 | 1.86 ± 0.20 | 72.6 ± 2.86 |
| DN4 | 8.41 | 12.5 | 6.20 | 28.0 | 65.8 | 5.20 | 15.0 | 18.6 | 89.1 ± 5.33 | 1.77 ± 0.19 | 74.0 ± 3.41 |
| DN5 | 16.3 | 13.6 | 6.50 | 32.0 | 61.5 | 4.60 | 14.1 | 15.4 | 142 ± 7.37 | 1.15 ± 0.13 | 64.1 ± 2.86 |
| DN6 | 16.5 | 14.5 | 4.50 | 34.2 | 61.3 | 6.90 | 12.7 | 16.9 | 185 ± 12.06 | 2.67 ± 0.29 | 62.3 ± 2.44 |
| DN7 | 17.8 | 12.8 | 5.40 | 26.1 | 68.5 | 3.40 | 10.7 | 14.6 | 108 ± 8.52 | 1.69 ± 0.21 | 52.8 ± 2.17 |
| DN8 | 15.2 | 11.3 | 21.7 | 26.2 | 52.1 | 3.50 | 9.80 | 10.9 | 83.6 ± 6.54 | 1.51 ± 0.18 | 44.1 ± 2.59 |
| DN9 | 1.65 | 0.55 | 95.8 | 1.60 | 2.60 | ND | ND | 0.226 | 2.24 ± 0.48 | 0 | 4.24 ± 0.34 |

relatively high amounts of POC and silt and relatively low amounts of sand. One-way ANOVA on variation among the four transects as a main factor was significant for $PC_1$ ($p = 0.019$) as a response but nonsignificant for $PC_2$ ($p = 0.527$), indicating that dioxins and clay varied significantly in some fashion among the four transects, whereas POC, silt, and sand did not. Levene's tests were nonsignificant for both principal components. A substantial portion of the variation in $PC_1$ was described by the transect factor ($\eta^2 = 0.263$). Special orthogonal contrasts of island and dredging effects within one-way ANOVAs revealed markedly significant ($p = 0.008$) island location effects and insignificant (at $p = 0.05$) dredging effects for $PC_1$, and nonsignificant contrast effects for $PC_2$. Considerable variation in $PC_1$ was explained by the two contrasts (island $\eta^2 = 0.201$; dredging $\eta^2 = 0.091$). A plot of mean $PC_1$ scores for the four transects illustrate a parallel pattern corresponding with the foregoing contrast differences (Figure 3A); no such pattern in mean $PC_2$ scores was apparent (Figure 3B). Sedimentary dioxin concentrations and sediment clay contents were markedly and significantly higher at Alexander Island as compared with Hog Island transects; however, while mean dioxin concentrations were visibly slightly higher for undredged as compared

with dredged transect samples as a whole (Figure 2), these differences were not significant.

Dredging significantly impacts and disrupts natural sedimentary processes and associations in the HSC *via* the repeated, physical removal of large quantities of sediment from within the navigation channel and by resuspending large quantities of sediment which are not actually removed by dredging. This scenario is exacerbated by the continuous movements of large ships within the HSC, which generate considerable turbulence, resulting in frequent sediment resuspension. These results show that surface sediments ($\leq 4$ cm) in dredged portions of the HSC carry dioxin concentrations which are not significantly different from those carried by sediments found in undredged parts of the system. Those channel-bottom sediments which are being reworked and resuspended by a combination of dredging and ship traffic are then able to mix with sediments in the water column, which are sourced both from lateral erosion (adjacent to the active shipping channel) and from locations upstream. Relationships between the fallout radionuclides associated with these sediments are supportive of this scenario. The use of isotope ratios, as opposed to discrete activity concentrations, has been shown to be an effective way to mitigate extrinsic variability in
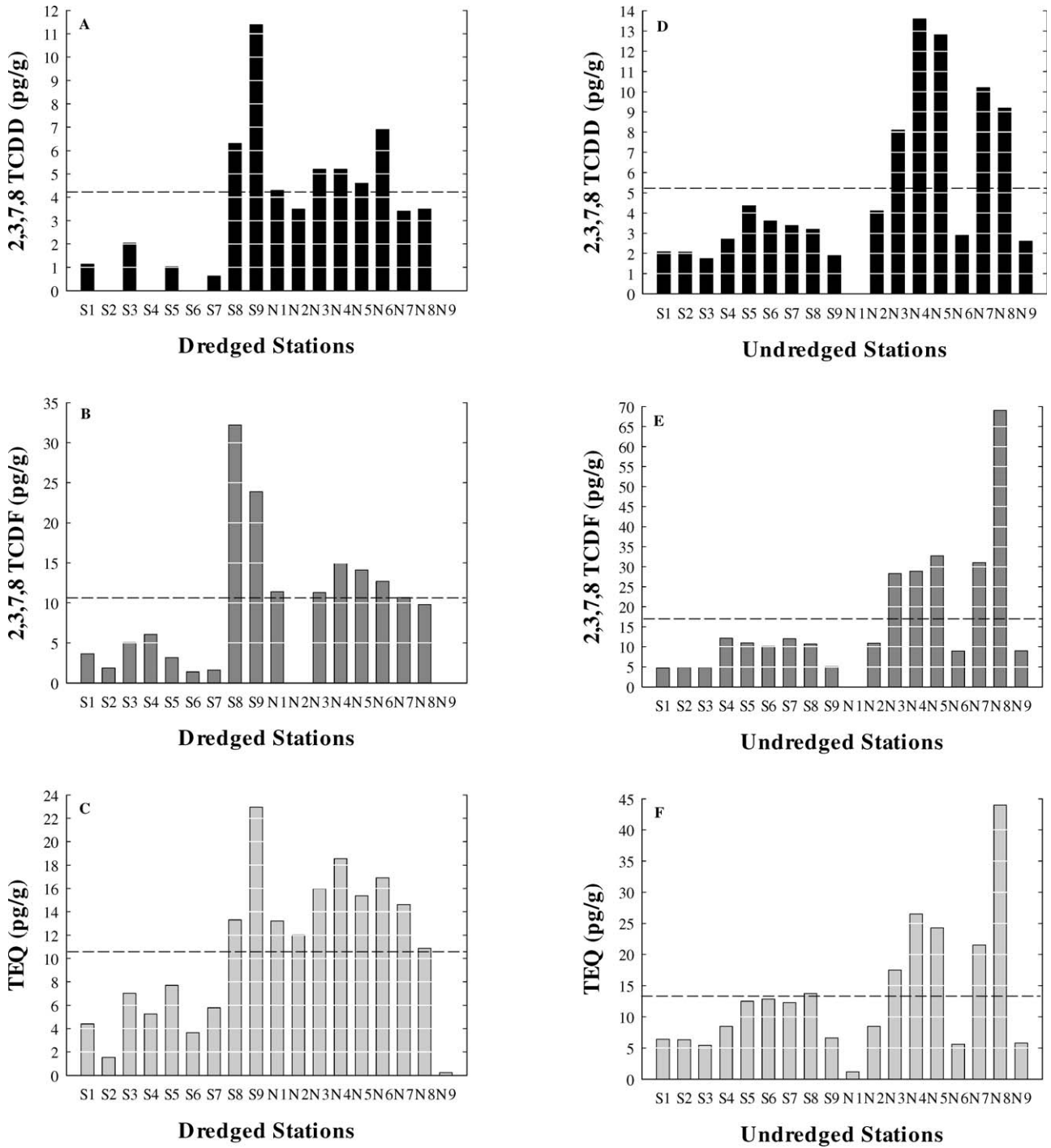
Figure 2. Concentrations of 2,3,7,8-TCDD (black), 2,3,7,8-TCDF (dark gray), and TEQ (gray) for stations along both transects (S and N denote stations from the southern [Hog Island] and northern [Alexander Island] transects, respectively). Dashed lines indicate population means, which do not include nondetects, or NDs (in pg/g, A = 4.22; B = 10.24; C = 10.52; D = 5.21; E = 17.30; F = 13.30). Missing data in panels A, B, D, and E represent NDs.

terrestrial or aquatic sediments (Yeager and Santschi, 2003). This approach has been used when considering natural and fallout radionuclides, including $^{210}$Pb, $^{137}$Cs, $^7$Be, and $^{234}$Th (Bonniwell, Matisoff, and Whiting, 1999; Feng, Cochran, and Hirschberg, 1999; Yeager *et al.*, 2005). While variability is in evidence from station to station, the mean isotope ratio values for each transect ($^7$Be/$^{137}$Cs, $^7$Be/$^{210}$Pb$_{xs}$) exhibit a consistent trend, with the dredged transect values being greater for both ratios (Figure 4). This indicates that dredged sediments contain more of the shorter-lived radionuclides (*i.e.,* $^7$Be), as

Table 2. *Mean (±SD), raw values for contaminant and physical sediment variables from all four transect locations. Nondetects (NDs) are not included in these calculations. For all variables, N = 9; except Hog Island dredged, TCDD = 6, Alexander Island undredged, TCDD = 8 and TCDF = 8, Alexander Island dredged, TCDD = 8 and TCDF = 7.*

| Transect | 2,3,7,8-TCDD (pg/g) | 2,3,7,8-TCDF (pg/g) | TEQ (pg/g) | POC (mg/g) | % Sand | % Silt | % Clay |
|---|---|---|---|---|---|---|---|
| Hog Island undredged | 2.78 (0.90) | 8.38 (3.40) | 9.39 (3.39) | 6.12 (2.22) | 24.88 (21.49) | 36.47 (10.52) | 38.66 (14.22) |
| Hog Island dredged | 3.75 (4.28) | 8.76 (11.24) | 7.95 (6.52) | 5.53 (3.85) | 36.58 (26.72) | 41.11 (25.06) | 22.31 (9.23) |
| Alexander Island undredged | 7.94 (4.33) | 27.34 (19.74) | 17.20 (13.56) | 7.76 (3.35) | 28.83 (27.79) | 22.39 (9.95) | 48.78 (21.74) |
| Alexander Island dredged | 4.57 (1.19) | 12.14 (1.88) | 13.08 (5.38) | 11.11 (4.13) | 16.84 (30.17) | 26.23 (9.77) | 56.92 (21.20) |

their buildup is more rapid after dredging than those with longer half-lives ($^{210}$Pb) or those for which a longer time has passed since introduction into the environment ($^{137}$Cs). Thus, a higher value of $^{7}$Be/$^{137}$Cs suggests that over short time scales (months), sediment mixing and contact with the overlying water column is likely more significant than lateral mixing of deeper, older sediments, since $^{7}$Be is a short-lived, fallout isotope with a near-constant atmospheric delivery, and concentrations of $^{137}$Cs are highest in older, deeper sediments and are near detection limits in overlying waters. A higher value of $^{7}$Be/$^{210}$Pb$_{xs}$ is also supportive, as $^{7}$Be activity concentrations tend to be higher than $^{210}$Pb$_{xs}$ activity concentrations in actively mixed surface sediments, given the higher atmospheric flux of $^{7}$Be versus $^{210}$Pb (*e.g.,* Baskaran, Coleman, and Santschi, 1993; Dominik, Burrus, and Vernet, 1987; Le Cloarec *et al.*, 2007).

Results presented here are in congruence with other work on sedimentary distributions of dioxins in this system. Yeager *et al.* (2007) presented data on the vertical distributions and chronology of dioxins in HSC sediments and their modern fluxes. Two sediment cores presented in that work are located in close proximity to the undredged test transects presented here (station 16499 on Alexander Island and station 13337 on Hog Island). Station 16499 had a dioxin concentration peak at the sediment surface and a much larger peak at a depth of 30–35 cm, corresponding to sediment ages of approximately 1 year and 18–25 years prior to sampling in 2004, respectively (Yeager *et al.*, 2007). Station 13337 had elevated dioxin concentrations at the sediment surface and a sharp increase in concentrations beginning at a depth of 30 cm, corresponding to sediment ages of approximately 1 year and 20 years prior to sampling in 2004, respectively (Yeager *et al.*, 2007). While both stations had

similar modern fluxes of dioxins to surface sediments (*e.g.,* TEQ of $1.26 \times 10^5$ pg m$^{-2}$ y$^{-1}$ and $1.44 \times 10^5$ pg m$^{-2}$ y$^{-1}$ for 16499 and 13337, respectively), sedimentary dioxin inventories were much different (*e.g.,* TEQ of $4.14 \times 10^6$ pg m$^{-2}$ and $2.28 \times 10^6$ pg m$^{-2}$ for 16499 and 13337, respectively; Yeager *et al.*, 2007). The higher sedimentary dioxin inventories at station 16499 reflect higher, long-term delivery of dioxins to sediments near Alexander Island as compared with Hog Island, supporting the statistical results presented here. Persistently high sedimentary dioxin concentrations at depths beginning at approximately 30 cm may be an important source of dioxins to

Table 3. *Principal component (PC) loadings (after varimax rotation) on three contaminant and four physical variables from all 36 stations distributed along the four transects. Values in bold were used to interpret components. For the most part, PC$_1$ describes variation in the three contaminant variables and percent clay; the orthogonal PC$_2$ mainly describes variation in POC, percent silt, and percent sand.*

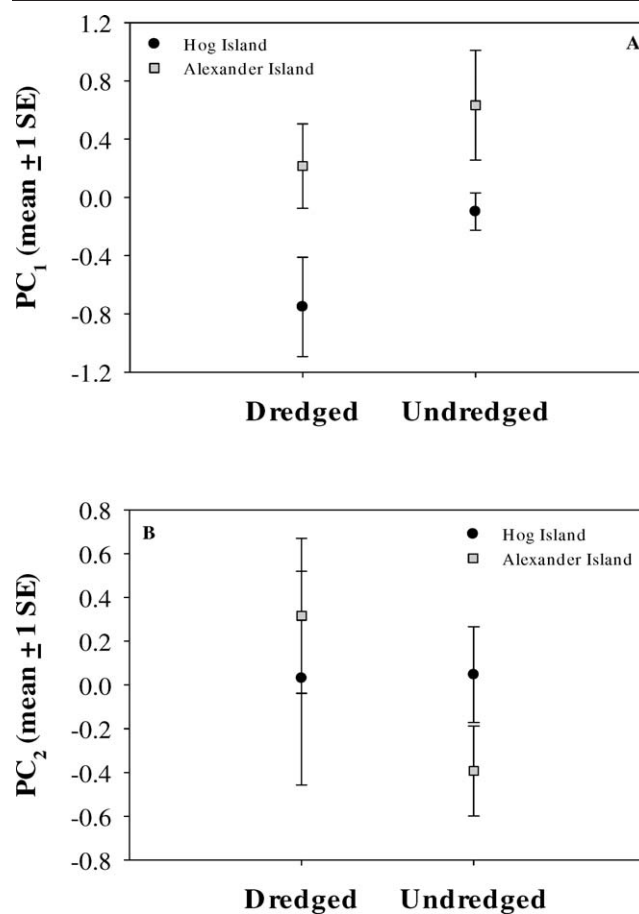| Variable | PC$_1$ | PC$_2$ |
|---|---|---|
| Log TCDD | **0.966** | 0.030 |
| Log TCDF | **0.925** | −0.036 |
| Log TEQ | **0.858** | 0.413 |
| POC | 0.261 | **0.738** |
| Arcsin % Sand | −0.475 | **−0.848** |
| Arcsin % Clay | **0.806** | 0.435 |
| Arcsin % Silt | −0.176 | **0.846** |



Figure 3. Plots of PC$_1$ (A) and PC$_2$ (B) scores for the four transects (island-dredging conditions).
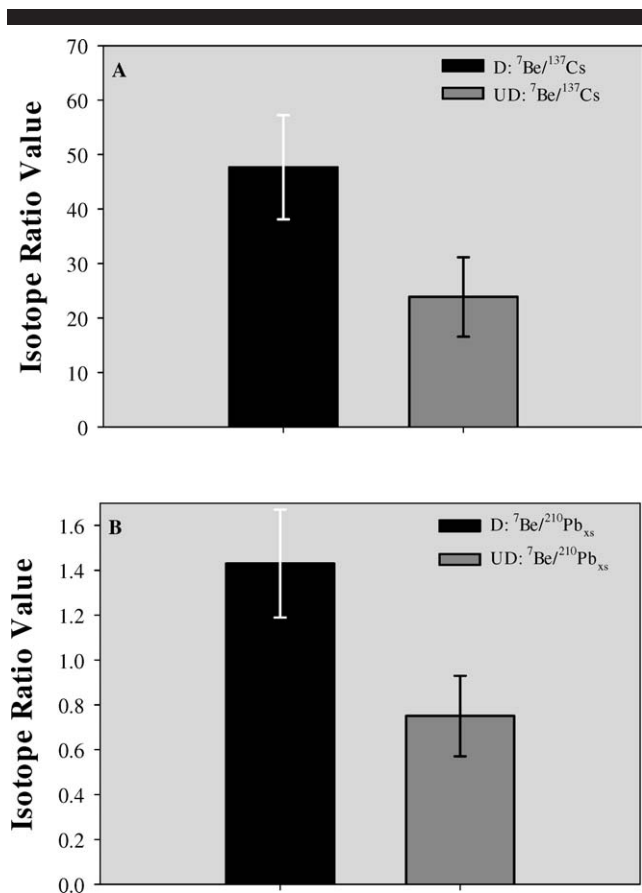
Figure 4. Plots of mean isotope ratio values for dredged (D) *versus* undredged (UD) sample populations (errors were propagated as discussed in Choppin, Liljenzin, and Rydberg [2001]; zero values and discrete values with errors in excess of 15% were not considered; error bars represent 1 standard error of the mean).

dredged parts of the channel, as these sediments will be exposed as portions of the channel walls. These deeper, dioxin-enriched sediments can then be eroded, locally deposited, and resuspended cyclically, providing a continuous source of dioxin-enriched sediment to dredged parts of the system. Additionally, it is likely that the dredge-spoil islands themselves provide a continuous source of dioxins to the system as well.

## CONCLUSIONS

While this research focused on the sampling of surface ($\leq 4$ cm) sediments at only four transects in the HSC, the data presented provide strong support for a number of conclusions: (1) sedimentary dioxin concentrations are significantly higher at transects sampled adjacent to Alexander Island as compared with Hog Island, which is most likely due to the closer proximity of Alexander Island to the San Jacinto waste pits; (2) while mean dioxin concentrations were slightly higher for undredged as compared with dredged transect samples as a whole, these differences were not significant; and (3) mean fallout isotope ratio values ($^{7}$Be/$^{137}$Cs, $^{7}$Be/$^{210}$Pb$_{xs}$) were significantly higher for dredged *versus* undredged sample popula-

tions, indicating that the buildup of shorter-lived radionuclides (*i.e.,* $^{7}$Be) is more rapid after dredging than those with longer half-lives ($^{210}$Pb) or those for which a longer time has passed since introduction into the environment ($^{137}$Cs). Thus, resuspension and mixing of surface sediments with those in the water column prior to redeposition is an important process in dredged channel settings and may be more important than mixing of surface sediments with those sourced from lateral erosion. The data set as a whole supports our null hypothesis, which is that dredging results in no significant reduction in the concentration of dioxins in HSC surface sediments.

Given the large geographic area and volume of sediment continually impacted by both dredging operations and shipping, the pervasive nature of dioxin contamination here, the recognition that bottom sediments are the most significant dioxin reservoir, the identification of at least one major upstream dioxin point source (San Jacinto waste pits), and the relevance of these studies to numerous other bays and harbors throughout the world, we recommend that further research be conducted to (1) increase understanding of the impacts of dredging and shipping activities on the distribution, concentration, and fate of sedimentary dioxins here; (2) characterize the concentrations of dioxins in sediments which comprise the many dredge-spoil islands in this system; and (3) review the practice of using dredge spoil as a resource in the construction and maintenance of wetlands, intertidal zones, and wildlife areas.

## ACKNOWLEDGMENTS

## LITERATURE CITED

Aalto, R. and Dietrich, W., 2005. Sediment accumulation with $^{210}$Pb geochronology for Strickland River flood plains, Papua New Guinea. *In: Sediment Budgets I: Proceedings of the International Symposium on Sediment Budgets: Held during the Seventh International Scientific Assembly of the Association of Hydrological Sciences* (Foz do Iguacüu, Brazil, April 2005), pp. 303–309.

Alcock, R.E.; McLachlan, M.S.; Johnston, A.E., and Jones, K.C., 1998. Evidence for the presence of PCDD/Fs in the environment prior to 1900 and further studies on their temporal trends. *Environmental Science and Technology*, 32(11), 1580–1587.

Bailar, J.C., III, 1991. How dangerous is dioxin? *New England Journal of Medicine*, 324, 260–262.

Baskaran, M.; Coleman, C.H., and Santschi, P.H., 1993. Atmospheric depositional fluxes of $^{7}$Be and $^{210}$Pb at Galveston and College Station, Texas. *Journal of Geophysical Research*, 98(D11), 20555–20571.

Bellas, J.; Ekelund, R.; Halldorsson, H.P.; Berggren, M., and Granmo, A., 2007. Monitoring of organic compounds and trace metals during a dredging episode in the Göta Älv Estuary (SW Sweden) using caged mussels. *Water, Air and Soil Pollution*, 181(1–4), 265–279.

Bocchetti, R.; Fattorini, D.; Pisanelli, B.; Macchia, S.; Oliviero, L.; Pilato, F.; Pellegrini, D., and Regoli, F., 2008. Contaminant accumulation and biomarker responses in caged mussels, *Mytilus galloprovincialis*, to evaluate bioavailability and toxicological

effects of remobilized chemicals during dredging and disposal operations in harbour areas. *Aquatic Toxicology*, 89(4), 257–266.

Bonniwell, E.C.; Matisoff, G., and Whiting, P.J., 1999. Determining the times and distances of particle transit in a mountain stream using fallout radionuclides. *Geomorphology*, 27, 75–92.

Brenner, M.; Schelske, C.L., and Kenney, W.F., 2004. Inputs of dissolved and particulate Ra-226 to lakes and implications for Pb-210 dating recent sediments. *Journal of Paleolimnology*, 32(1), 53–66.

Broman, D.; Naf, C., and Zebuhr, Y., 1992. Occurrence and dynamics of polychlorinated dibenzo-p-dioxins and dibenzofurans and other combustion related organic pollutants in the aquatic environment of the Baltic. *Chemosphere*, 25, 125–128.

Choppin, G.R.; Liljenzin, J.-O., and Rydberg, J., 2001. *Radiochemistry and Nuclear Chemistry*. Woburn, Massachusetts: Elsevier, 720p.

Correa, O.; Raun, L.; Rifai, H.; Suarez, M.; Holsen, T., and Koenig, L., 2006. Depositional flux of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans in an urban setting. *Chemosphere*, 64(9): 1550–1561.

Crocker, P.A. and Young, C., 1990. *Tetrachlorodibenzo-p-dioxins and Dibenzofurans in Edible Fish Tissue at Selected Sites in Arkansas, Louisiana, and Texas*. Dallas, Texas: U.S. Environmental Protection Agency Region 6 unpublished report.

Dominik, J.; Burrus, D., and Vernet, J.-P., 1987. Transport of the environmental radionuclides in an alpine watershed. *Earth and Planetary Science Letters*, 84, 165–180.

Engwall, M.; Naf, C.; Broman, D., and Brunstrom, B., 1998. Biological and chemical determination of contaminant levels in settling particulate matter and sediments—a Swedish river system before, during, and after dredging of PCB-contaminated lake sediments. *Ambio*, 27(5), 403–410.

Feng, H.; Cochran, J.K., and Hirschberg, D.J., 1999. $^{234}$Th and $^{7}$Be as tracers for transport and sources of particle-associated contaminants in the Hudson River Estuary. *Science of the Total Environment*, 237/238, 401–418.

Fiedler, H., 1996. Sources of PCDD/PCDF and impact on the environment. *Chemosphere*, 32, 55–64.

Frank, D.S.; Mora, M.A.; Sericano, J.L.; Blankenship, A.L.; Kannan, K., and Giesy, J.P., 2001. Persistent organochlorine pollutants in eggs of colonial waterbirds from Galveston Bay and East Texas, USA. *Environmental Toxicology and Chemistry*, 20(3), 608–617.

Green, N.J.L.; Hassanin, A.; Johnston, A.E., and Jones, K.C., 2004. Observations on historical, contemporary, and natural PCDD/Fs. *Environmental Science and Technology*, 38(3), 715–723.

Green, N.J.L.; Jones, J.L.; Johnston, A.E., and Jones, K.C., 2001. Further evidence for the existence of PCDD/Fs in the environment prior to 1900. *Environmental Science and Technology*, 35(10), 1974–1981.

Hagenmaier, H.; She, J.; Benz, T.; Dawidowsky, N.; Dusterhoft, L., and Lindig, C., 1992. Analysis of sewage sludge for polyhalogenated dibenzo-p-dioxins, dibenzofurans, and diphenylethers. *Chemosphere*, 25, 1457–1462.

Harris, D.; Horwath, W.R., and Van Kessel, C., 2001. Acid fumigation of soils to remove carbonates prior to total organic carbon or carbon-13 isotopic analysis. *Soil Science Society of America Journal*, 65, 1853–1856.

Hedges, J.I. and Stern, J.H., 1984. Carbon and nitrogen determinations of carbonate-containing solids. *Limnology and Oceanography*, 29(3), 657–663.

Krishnan, V.; Porter, W.; Santostefano, M.; Wang, X., and Safe, S., 1995. Molecular mechanism of inhibition of estrogen-induced cathepsin D gene expression by 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) in MCF-7 cells. *Molecular and Cellular Biology*, 15, 6710–6719.

Le Cloarec, M.-F.; Bonte, P.; Lefevre, I.; Mouchel, J.-M., and Colbert, S., 2007. Distribution of $^{7}$Be, $^{210}$Pb and $^{137}$Cs in watersheds of different scales in the Seine River basin: inventories and residence times. *Science of the Total Environment*, 375, 125–139.

Liem, A.K.D., 1997. Dioxins: Chemical Analysis, Exposure and Risk Assessment. Utrecht, The Netherlands: Research Institute of Toxicology, Utrecht University, Ph.D. thesis, 262p.

Muir, D.C.G.; Lawrence, S.; Holoka, M.; Fairchild, W.L.; Segstro,

M.D.; Webster, G.R.B., and Servos, M.R., 1992. Partitioning of polychlorinated dioxins and furans between water, sediments and biota in lake mesocosms. *Chemosphere*, 25, 199–224.

Poland, A. and Knudson, J.C., 1982. 2,3,7,8-Tetrachlorodibenzo-*p*-dioxin and related aromatic hydrocarbons: examination of the mechanism of toxicity. *Annual Reviews in Pharmacology*, 22, 517–554.

Port of Houston Authority, 2009. Home page. http://www.portofhouston.com/geninfo/overview1.html (accessed December 2008).

Quass, U.; Fermann, M., and Broker, G., 2004. The European dioxin air emission inventory project—final results. *Chemosphere*, 54(9), 1319–1327.

Raun, L.H.; Correa, O.; Rifai, H.; Suarez, M., and Koenig, L., 2005. Statistical investigation of polychlorinated dibenzo-p-dioxins and dibenzofurans in the ambient air of Houston, Texas. *Chemosphere*, 60(7), 973–989.

Rifai, H.; Palachek, R., and Jensen, P., 2003. Total Maximum Daily Loads for Dioxins in the Houston Ship Channel. Austin, Texas: unpublished report prepared for Texas Commission on Environmental Quality Total Maximum Daily Load Program, 125p. http://163.234.20.106/assets/public/implementation/water/tmdl/26hscdioxin/26-progrpt-q4-fy06.pdf (accessed August 2009).

Safe, S.H., 1986. Comparative toxicology and mechanism of action of polychlorinated dibenzo-*p*-dioxins and dibenzofurans. *Annual Reviews in Pharmacology*, 26, 371–399.

Santschi, P.H.; Allison, M.; Asbill, S.; Perlet, A.B.; Cappellino, S.; Dobbs, C., and McShea, L., 1999. Sediment transport and Hg recovery in Lavaca Bay, as evaluated from radionuclide and Hg distributions. *Environmental Science and Technology*, 33, 378–391.

Servos, M.R.; Muir, D.C.G., and Webster, G.R.B., 1992. Environmental fate of polychlorinated dibenzo-*p*-dioxins in lake enclosures. *Canadian Journal of Fisheries and Aquatic Sciences*, 49, 722–734.

Sonnenberg, L.B. and Nichols, K.M., 1995. Emissions of hydrochloric acid, PCDD and PCDF from the combustion of chlorine-containing kraft pulp mill bleach plant waste. *Chemosphere*, 31, 4207–4223.

Stellman, J.M.; Stellman, S.D.; Christian, R.; Weber, T., and Tomassalo, C., 2003. The extent and patterns of usage of Agent Orange and other herbicides in Vietnam. *Nature*, 4222, 681–687.

Sturve, J.; Berglund, A.; Balk, L.; Broeg, K.; Böhmert, B.; Massey, S.; Savva, D.; Parkkonen, J.; Stephensen, E.; Koehler, A., and Förlin, L., 2005. Effects of dredging in Goteborg Harbor, Sweden, assessed by biomarkers in eelpout (*Zoarces Viviparus*). *Environmental Toxicology and Chemistry*, 24(8), 1951–1961.

Suarez, M.P.; Rifai, H.S.; Palachek, R.M.; Dean, K.E., and Koenig, L., 2005. Polychlorinated dibenzo-*p*-dioxins and dibenzofurans in Houston Ship Channel tissue and sediment. *Environmental Engineering Science*, 22(6), 891–906.

Suarez, M.P.; Rifai, H.S.; Palachek, R.; Dean, K., and Koenig, L., 2006. Distribution of polychlorinated dibenzo-p-dioxins and dibenzofurans in suspended sediments, dissolved phase and bottom sediment in the Houston Ship Channel. *Chemosphere*, 62(3), 417–429.

Sundberg, H.; Hanson, M.; Liewenborg, B.; Zebuhr, Y.; Broman, D., and Balk, L., 2007. Dredging associated effects: maternally transferred pollutants and DNA adducts in feral fish. *Environmental Science and Technology*, 41(8), 2972–2977.

Tate, J.N. and Berger, R.C., 2006. Houston-Galveston Navigation Channels, Texas Project: Navigation Channel Sedimentation Study, Phase 1, Final Report. Vicksburg, Mississippi: U.S. Army Corps of Engineers, Engineer Research and Development Center, Coastal and Hydraulics Laboratory, 136p.

USEPA (U.S. Environmental Protection Agency), 2000. Exposure and Human Health Reassessment of 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) and Related Compounds. Part I: Estimating Exposure to Dioxin-Like Compounds. Volume 3: Properties, Environmental Levels, and Background Exposures. Washington, DC: U.S. Environmental Protection Agency, 321p.

USEPA, 2008. Technical Factsheet on: Dioxin (2,3,7,8-TCDD). http://www.epa.gov/ogwdw/dwh/t-soc/dioxin.html (accessed December 2008).

USEPA, 2009. National Priorities List Sites in the United States.

http://www.epa.gov/superfund/sites/npl/npl.htm (accessed December 2008).

Wen, L.-S.; Santschi, P.H.; Gill, G., and Paternostro, C., 1999. Estuarine trace metal distributions in Galveston Bay: importance of colloidal forms in the speciation of the dissolved phase. *Marine Chemistry*, 63, 185–212.

Wilken, M.; Neugebauer, F.; Fabarius, G.; Zeschmar-Lahl, B., and Jager, J., 1992. PCDD/PCDF contamination of different pesticides produced in former GDR. *Chemosphere*, 25, 1525–1531.

Yeager, K.M. and Santschi, P.H., 2003. Invariance of isotope ratios of lithogenic radionuclides: more evidence for their use as sediment source tracers. *Journal of Environmental Radioactivity*, 69, 159–176.

Yeager, K.M.; Santschi, P.H.; Phillips, J.D., and Herbert, B.E., 2005.

Suspended sediment sources and tributary effects in the lower reaches of a coastal plain stream as indicated by radionuclides, Loco Bayou, Texas. *Environmental Geology*, 47(3), 382–395.

Yeager, K.M.; Santschi, P.H.; Rifai, H.; Suarez, M.; Brinkmeyer, R.; Hung, C.C.; Schindler, K.J.; Andres, M., and Weaver, E.A., 2007. Dioxin chronology and fluxes in sediments of the Houston Ship Channel, Texas: influences of non-steady state sediment transport and total organic carbon. *Environmental Science and Technology*, 41(15), 5291–5298.

Yeager, K.M.; Santschi, P.H., and Rowe, G.T., 2004. Sediment accumulation and radionuclide inventories ($^{239,240}$Pu, $^{210}$Pb and $^{234}$Th) in the northern Gulf of Mexico, as influenced by organic matter and macrofaunal density. *Marine Chemistry*, 91(1–4), 1–14.