

AN ASSESSMENT OF TRIBUTYLTIN AND METALS IN SEDIMENT CORES FROM THE ST. THOMAS EAST END RESERVES



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Center for Coastal Monitoring and Assessment

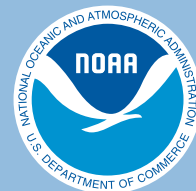
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An Assessment of Tributyltin and Metals in Sediment Cores from the St. Thomas East End Reserves

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ABSTRACT

This is the sixth report in a series from a project to assess land-based sources of pollution (LBSP) and their effects, and to characterize the biological community within the St. Thomas East End Reserves (STEER) in St. Thomas, USVI. Here we summarize the results of a study to assess the distribution and depositional history of tributyltin (TBT), heavy metals and other trace elements in the sediments in the Benner Bay sub-region of the STEER.

Tributyltin and copper concentrations in Benner Bay on St. Thomas USVI were found to be elevated relative to other areas in the larger study of the STEER. At the request of the USVI Coastal Zone Management Program, sediment cores and surface sediment samples were collected to better define the extent and history of TBT deposition in the vicinity of Benner Bay. The sediment cores were sectioned into 2 cm intervals and dated using ^{210}Pb and ^{137}Cs . The core sections and the surface samples were analyzed for butyltins and 16 metals and trace elements. Sediment deposition rates varied from 0.7 -5.0 mm/yr, and were highest in the marina complex. Core ages ranged from 54 to 200 yrs. The inner reaches of northern Benner Bay are severely degraded by marina operations, shoreline development and watershed changes. Sediment dynamics and sediment quality are, and have been for decades, heavily impacted. The bottoms of the cores contained shell hash from a once thriving benthic community. That community has vanished and has been smothered by very fine sediment that accumulates at rates an order of magnitude above normal rates. The sediment is highly contaminated with butyltin paint residues, copper, and other toxic metals. Surface concentrations of TBT exceeded 2,000 ppb at two locations. At a depth of 8 cm TBT exceeded 8,800 ppb in the marina complex sediment. Based on the ratio of tributyltin to total butyltins, it appears the marina sediments are the source of contamination of the surrounding area. There is evidence that vessels from neighboring islands may also be a source of fresh TBT. Gradients of virtually all metals and trace elements extended away from the marina complex. NOAA sediment quality guidelines were exceeded for As, Pb, Cu, Zn and Hg. Pile driving and dredging new or deeper channels will spread these contaminants over a wide area.

INTRODUCTION

The St. Thomas East End Reserves or STEER is a collection of Marine Reserves and Wildlife Sanctuaries (MRWS) located on the southeastern end of the island of St. Thomas, US Virgin Islands. It is made up of four MRWS including the Inner Mangrove Lagoon, Cas Cay/Mangrove Lagoon, St James Bay, and Compass Point Salt Pond Marine Reserves and Wildlife Sanctuaries (Figure 1). Within the STEER, there are extensive mangroves and seagrass beds, along with coral reefs, lagoons and cays. The value of the natural resources in the Reserves has long been recognized. In 1979, the area was identified by NOAA's National Marine Sanctuary Program as a "marine area of national significance, deserving of marine sanctuary designation". The same year, the Mangrove Lagoon/Benner Bay area along with Vessup Bay were designated by the USVI government as Areas of Particular Concern, due to the abundance of important but threatened natural resources, and the desire to preserve and as needed, restore these areas.

With the recognition of the connectivity of the natural, cultural, and economic resources of the Reserves, it was decided that a comprehensive management plan was needed, in order to protect and properly manage the natural resources in the area (STEER, 2011). In 2008, DPNR (Department of Planning and Natural Resources), along with the University of the Virgin Islands, The Nature Conservancy, and a community group called the Friends of Christmas Cove began development of a management plan for the STEER, which was completed in May 2011.

In 2011 NOAA's National Centers for Coastal Ocean Science (NCCOS), in partnership with DPNR's Divisions of Coastal Zone Management, Fish and Wildlife, and Environmental Protection, along with the University of the Virgin Islands and The Nature Conservancy initiated a project to develop an integrated assessment of chemical and biological conditions within the STEER. Sediments were collected for chemical contaminant analysis, bioeffects assays, and benthic infaunal analyses.



Figure 1. Individual Reserves that comprise the STEER.

Results from the initial characterization of sediment contaminants in the area (Pait et al., 2013) indicated unusually elevated concentrations of tributyltin (TBT) and metals in sediments in the Benner Bay region of the STEER. After reviewing these results, the USVI DPNR requested that NCCOS conduct a follow-up study to better assess the extent of contamination in surface sediments and to determine contaminant concentrations in the deeper underlying sediments in the area. In 2013 we conducted the follow-up study to determine the distribution of TBT and heavy metal residues in the area in more detail, and also to take sediment cores at a number of locations to assess the record of TBT concentrations and metals over time.

BACKGROUND

For 40 years tributyltin (TBT) was used as a prime ingredient in antifouling paint applied to the hulls of boats and ocean going vessels. The antifouling properties of TBT compounds were discovered in the 1950s. The function of the biocide in the antifouling paint is to prevent the settling of organisms on the hull and to poison the organisms that do. By the mid-1960s it became the most popular antifouling paint worldwide. Although the paints were effective, the TBT slowly leaches out into the marine environment where it was highly toxic to a wide range of organisms.

The formulation of TBT paints changed over time. Initially it worked by contact leaching to release the TBT to water. However, the release rate proved inconsistent and unpredictable, and as a result self-polishing co-polymer paints were developed. These paints used a polymer base through which the biocide discharge rate is regulated by reacting with water, and resulted in the TBT being slowly released in water. Once a surface covering was worn away, TBT release continued with the next layer. TBT-based paints were extremely effective and long lasting.

Bottom paint improves ship performance and durability by reducing biofouling on the ship's hull (Bray and Langston, 2006). This allowed increased ship speed and lower fuel consumption due to the lack of fouling organisms on the hull, and increased the time between scraping and repainting, all of which were economically advantageous. By the late 1970s TBT paints were commonly used on commercial and recreational vessels.

Negative aspects of TBT were suspected in the late 1960s when it was recognized that the release of organotin into aquatic environments was impacting non-target organisms. Toxic effects in some species occur at a

concentration as low as one part per trillion (i.e., 1 nanogram per liter (ng/L)) of water (Bray and Langston, 2006). TBT is toxic to bacteria, algae, fungi, mollusks and crustaceans (Cruz et al., 2015). There are implications of effects on cetaceans and bioaccumulation of TBT in the human food chain. A galvanizing event occurred in the late 1970s and early 1980s, when oyster crops in Arcachon Bay, France, failed. Subsequent research identified that TBT had caused decreased spatfall, unnatural shell thickening and abnormal structure that weakened the shells (Bray and Langston, 2006). Similar observations were seen in UK oysters stocks. Away from hull cleaning operations, TBT sediment concentrations were higher in harbors with many small boats than in industrial harbors with commercial ships (Bryan and Gibbs, 1991; Page et al, 1996). In 1982, France banned TBT use on recreational vessels less than 25 meters long. Subsequent work showed that TBT was an endocrine disruptor in marine gastropods causing masculinization (imposex) in females and widespread population decline. By the early 1990s, many nations had partial or complete bans on TBT. In 1999, the International Maritime Organization (IMO) a United Nations agency, came to agreement that TBT would be phased out between 2003 and 2008, with a total ban of organotin antifouling coatings by January 2008. The U.S. ratified the agreement in 2012. However, while the agreement requires compliance from the developed nations, much of the developing world are not signatories. Also, TBT is still used as a slimicide in power plant cooling towers and other industrial heat exchange equipment, as a wood preservative, and as a molluscicide. It was reportedly still available in bottom paint as recently as 2014 in the Caribbean and Central America through U.S. outlets (Turner and Glegg, 2014).

TBT is a persistent and bioaccumulative compound. The degradation pathways proceed from tributyltin to dibutyl, to monobutyl-, and finally to elemental tin. The reported half-life in estuarine waters range from days to weeks (Omae, 2005). TBT is strongly sorbed to sediments particles, however, and the half-life of TBT in sediments is years, and in anaerobic sediments extends into decades (Matthiessen, 2013). Also, paint chips from boat hulls, for example as might occur during the scraping and repainting process, can ultimately be flushed into a water body, and serve as a reservoir for release into the sediment. Thus high concentrations may persist in older buried sediments where uncontrolled releases have occurred in the past. This material may or may not become bioavailable depending on local sediment deposition rates and subsequent disturbance (e.g. dredging).

MATERIALS AND METHODS

Sampling

Surface Sediments

Surficial sediment samples were collected at six locations in Benner Bay leading away from the main marina facility on two transects, one out the approach channel and one through an offshore boat anchoring area (Figure 2). Water quality measurements and sediments were collected using standard NOAA National Status and Trends (NS&T) protocols (Apeti et al., 2012). A PONAR grab was deployed to collect the samples, and retrieved by hand. All equipment was rinsed with acetone and then distilled water just prior to use at a site. The top 3 cm of sediment were collected from the grab using a stainless steel sediment scoop. Surface sediment was used for chemical analyses to assess current depositional conditions.

Sediments were placed into a certified clean (I-Chem®) 250 ml labeled jar, capped and then kept on ice in a cooler. Sediments for grain size analysis were placed in a WhirlPack® bag, sealed and then kept on ice in a cooler. At the end of each day, sediment samples for contaminant analysis were frozen. The WhirlPack® bags for the grain size analysis were placed in a refrigerator rather than frozen, to avoid altering the grain size structure.

Sediment Cores

The sediment core samples were collected at four locations starting from the approach channel leading from Mangrove Lagoon to the southwest, and up into the head of Benner Bay in the heart of the area where the marina facilities are located (Figure 2). The boat was anchored at the bow and stern to minimize drift so the core could be deployed vertically. The corer was designed to collect undisturbed cores of the sediment and mud-water interface. The corer drove a seven cm diameter polycarbonate tube into the sediment with a hand-held weight. A one-way check valve seated in the core head allowed water and sediment to move through the core

barrel. During retrieval the check valve automatically seats, creating a partial vacuum which retains the sample in the core barrel. The core was returned to the dock where it was extruded in 2 cm sections. Each section was placed into a certified clean (I-Chem®) 250 ml labeled jar and homogenized. A sub-sample was removed for grain size analysis. Sediments for grain size analysis were placed in a WhirlPack® bag, sealed and then kept refrigerated. Sediment samples for metals and TBT analyses were frozen. An additional surface sediment sample was collected for TBT analysis with the PONAR grab at station BB2 as a check on potential gear bias.

Chemical Contaminants Analyzed

The sediments were analyzed for a suite of 16 major and trace elements, grain size analysis and for four butyltin compounds by TDI-Brooks International, using protocols established by the NS&T Program. The major and trace elements were analyzed using inductively coupled plasma mass spectrometry and atomic-fluorescence spectroscopy. Detailed descriptions of the NS&T protocols, including quality assurance/quality control (QA/QC) used in the analysis can be found in Kimbrough and Lauenstein (2006). The four butyltins were analyzed using gas chromatography/flame photometric detection after derivatization with hexyl-MgBr.

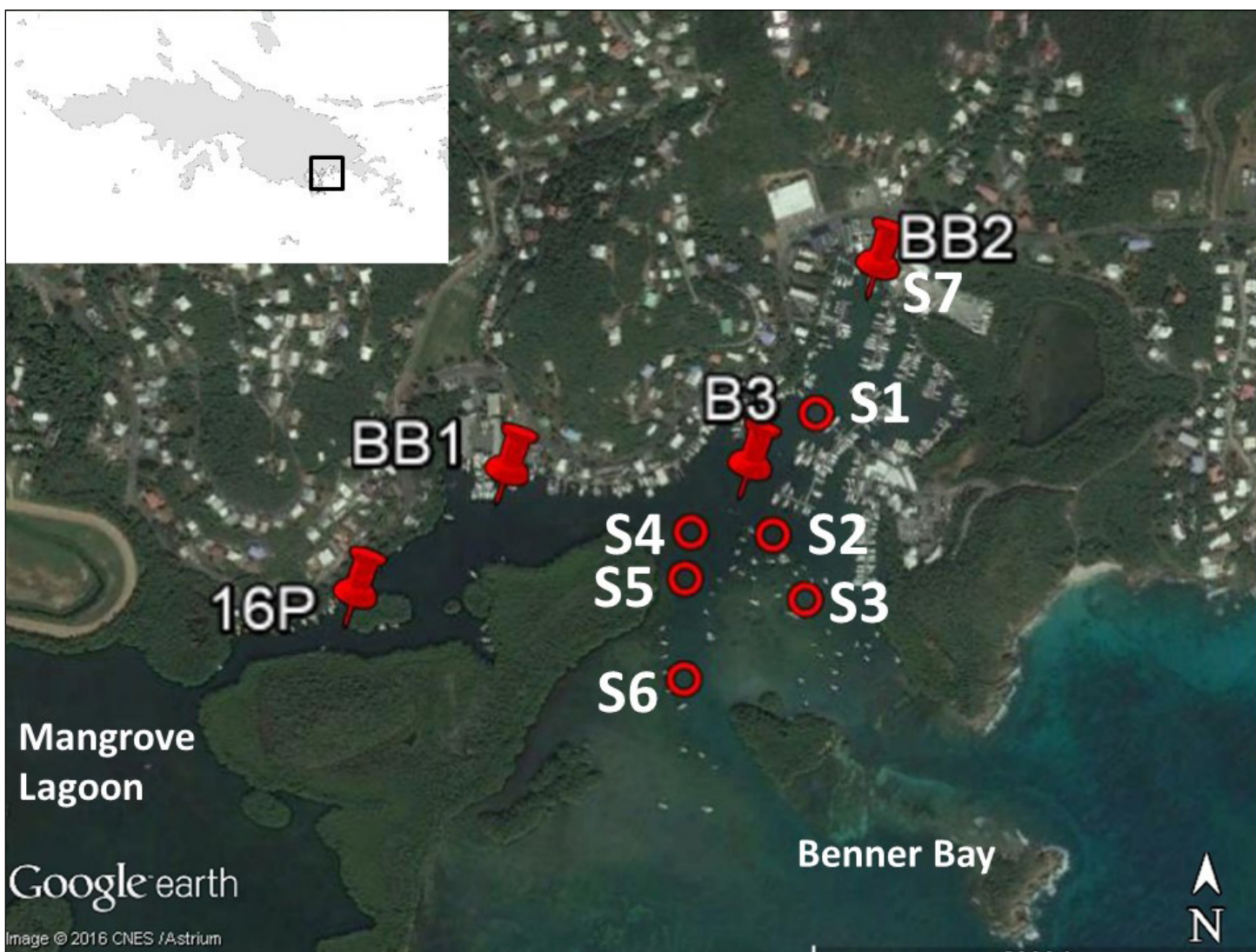


Figure 2. Map showing the north shoreline of Benner Bay. Sediment cores were collected at locations marked with pins, surface sediment samples were collected at locations marked with circles.

Radiochemical Dating

The core sections were also analyzed for two isotopes in order to estimate the age of the sediments down the core length. In depositional environments, sediments are continually laid down and compacted by overlying sediment. In a continuous sequence, the age of the sediment can be estimated and the history of chemical

contamination can be recreated. Depending on the rate of sediment deposition, changes in contaminant inputs will be obvious. Bioturbation by burrowing organisms will tend to mix the surface layers as they accumulate and may blur the history. Also, powerful storms, like hurricanes, may scour out whole sections, or, conversely bury layers with large deposits from runoff or resuspension. Also human activities such as dredging and spoil disposal may cause breaks in the historical record. Activities on land that alter sediment delivery from runoff may increase or decrease the rate of sedimentation over time.

The age of the sediment sections were estimated by measuring an isotope of lead (^{210}Pb) and of cesium (^{137}Cs). Briefly, Uranium-238 (^{238}U) decays to Uranium-234 (^{234}U) and then to thorium-230 (^{230}Th) which then decays to radium-226 (^{226}Ra). ^{226}Ra has a half-life of 16,000 yr, so the background supply is virtually constant. Radium eroded from rocks is deposited in sediments. Radium decays into radon-222 (^{222}Rn) which has a half-life of only 3.8 days. Finally, radon decays to ^{210}Pb with a moderate half-life of 22.3 yr. So, a constant background level of ^{210}Pb is present in rock and eroded sediments. Because radon is a gas, some of it escapes from the earth into the atmosphere to yield atmospheric ^{210}Pb . This is washed out of the air by rain and is deposited into fresh sediments. Thus sediment deposits have two sources of ^{210}Pb , the background amount constantly derived from slow radium decay, and from atmospheric deposition. The older a layer of sediment is, the more of the atmospherically derived ^{210}Pb will have decayed until only background levels remain. So, looking at ^{210}Pb levels down the length of a core you would expect to see concentrations decline in proportion to the sediment deposition rate, until only background levels are seen. ^{210}Pb is used to determine the age of sediments and accumulation rate of sediments in water bodies. In a typical application, the average accumulation rate over a period of 100 - 200 years is obtained. From the accumulation rate, the age of sediment from a particular depth in the sediment column can be estimated.

Cesium-137 (^{137}Cs) measurements are used as a check on calculated age profiles by providing date “markers” rather than concentration slopes. ^{137}Cs is derived from atomic bomb testing. The first appearance of ^{137}Cs in sediments marks the year 1954, which is the year when global concentrations generally achieved detectable levels. The other ^{137}Cs marker is the concentration maximum in the year 1963, after which atmospheric testing ceased.

Lead and cesium measurements were performed at the University of Maryland, Horn Point Environmental Laboratory. ^{210}Pb measurements were carried out via analysis of its short-term daughter product polonium-210 (^{210}Po), measured by alpha spectroscopy (Palinkas and Nitttrouer, 2007). Briefly, 1-2 g of dry, ground sediment samples were spiked with a known volume of polonium-209 (^{209}Po), leaching first in 15.8 N HNO_3 , then in 6 N HCl. ^{210}Po and ^{209}Po were then electroplated onto silver planchets and counted for 24 hours in a Canberra Alpha Analyst alpha spectrometer (Nitttrouer et al., 1979). Age models were fit to the data, as appropriate, to determine sediment accumulation rates (Appleby and Oldfield, 1978; Carroll and Lerche, 2003; Hancock et al., 2000). ^{137}Cs measurements were performed using gamma spectroscopy. Dry, ground sediment from each sampling interval (~3-5 g) was sealed in 60-mL plastic jars. The gamma emissions from each sample was counted for approximately 24 hr with a calibrated Canberra germanium detector, using the 661 KeV photopeak of the gamma spectrum. Accumulation rates were calculated from both the depth of first appearance and maximum activity, corresponding to the onset of atmospheric weapons testing in 1954 and maximum fallout in 1963, respectively.

RESULTS AND DISCUSSION

Core depth varied and was limited by the depth of dense and/or shell hash layers which was the depth of refusal for the corer (Figure 3). Two cores were taken at BB2. The first core was 28 cm deep, but the core above 18 cm was disturbed by air bubbles during handling. A second core sample was taken which was only 14 cm deep so there is a break in the data. There was only enough material to do butyltins and the radiochemical analyses on the 18-20cm section.

Contaminants in Surface Sediments

Metals and butyltin concentrations in the surface samples and the top layer of the cores are shown in Table 1. The concentration of total butyltins (TotBT, the sum of the butyltins analyzed) at BB2 was orders of magnitude above all other stations. There is a clear gradient of butyltins from BB2 out into Benner Bay and down the channel toward Mangrove Lagoon. There was good agreement between the top core section at BB2 and sample S7, indicating no gear bias. The concentration of Cu was also orders of magnitude higher at station BB2 than all other stations. Copper concentrations exceeded the ERM by 5X at BB2. Zinc also exceeded the ERM at BB2. Impacts to benthic organisms in this area are likely as a result of these concentrations.

Isotope Analysis of Cores

Plots of ^{210}Pb and ^{137}Cs activities with depth are shown for each core in Figures 4-7. Core 16P had the best profile of all the cores, showing the characteristic shape of logarithmic ^{210}Pb decay with depth until the background activity (0.1 dpm/g) is reached (Figure 4). For ^{137}Cs , the depth of first appearance (1954) is 6-8 cm; the depth of maximum activity (1963) is 4-6 cm. The accumulation rate calculated from the ^{210}Pb data is 0.073 cm/yr. Calculated accumulation rates based on the ^{137}Cs data are 0.10-0.14 cm/y and 0.08-0.12 cm/y, respectively. All of these rates are in rough agreement, given errors and assumptions inherent in both methods, and the profiles are very clear. The accumulation rates using ^{137}Cs are slightly higher than from the ^{210}Pb data, indicating somewhat younger sediment at depth. This becomes more obvious when the TBT data is plotted vs YBP (years before present).



Figure 3. Photograph of core BB2 showing shell hash in the lower part of the core, grading to fine mud up the core in more recent sediments.

Figure 5 shows the profile for core BB1. The accumulation rate calculated from the ^{210}Pb data is 0.06 cm/y. The profile is straightforward – ^{210}Pb decays logarithmically with depth to a background level of 0.1 dpm/g. For ^{137}Cs , the depth of first appearance (1954) is 6-8 cm, yielding an accumulation rate of 0.10-0.14 cm/y. The depth of maximum ^{137}Cs activity (1963) is 2-4 cm, yielding an accumulation rate of 0.04-0.08 cm/y. There is good agreement between the ^{137}Cs and ^{210}Pb data. There is some evidence for increased accumulation rates over time, but increased depth resolution would be needed for further evaluation.

Figure 6 shows the profile for core B3. The ^{210}Pb data for this core are subject to interpretation. It does not appear that the core was long enough to reach the background ^{210}Pb level, so the background activity was assumed to be equal to that used for 16P and BB1 (0.1 dpm/g). The background level within a small region should be the same. If a higher background activity is used, the accumulation rate will decrease – for example, using 0.5 dpm/g as the background level yields a rate of 0.085 cm/y. Also, there is some question as to whether all data points reflect sedimentation, and thus should be used in the rate calculation, or whether the upper 4 data points reflect mixing and should be neglected in the rate calculation. The accumulation rates are 0.32 cm/y (all data) or 0.16 cm/y (only the lower 4 data points).

For ^{137}Cs data, the depth of first appearance (1954) is 8-10 cm, yielding an accumulation rate of 0.14-0.17 cm/y. This is in agreement with the ^{210}Pb rate from lower four points. There are two depths, above and below the 6-8 cm interval, with identically high activities that could be considered the depth of maximum ^{137}Cs activity

Table 1. Concentrations of butyltins (ng Sn/g) and metals in surface samples of sediments and the top layer of the cores.

Site	Monobutyltin	Dibutyltin	Tributyltin	TotBT	%TriBT	Ag	As	Cd	Pb	Sb	Sn	Al
16P 0-2	6.61	10.43	66.4	83.5	79.6	0.00	11.00	0.156	15.70	0.46	1.99	38,600
B3 0-2	13.3	11.2	12.4	36.9	33.6	0.00	10.00	0.085	17.40	0.22	1.76	21,400
BB1 0-2	12.6	28.2	40.0	80.8	49.5	0.00	9.64	0.230	25.40	1.00	3.32	48,900
BB2 0-2	940	700	1102	2741.2	40.2	0.27	16.40	0.281	129.00	0.73	22.20	64,700
S1	72.6	68.7	134	275.0	48.6	0.08	9.67	0.078	119.00	1.49	5.25	29,500
S2	25.7	9.74	12.2	47.6	25.6	0.00	6.96	0.080	24.10	0.19	3.06	26,500
S3	7.61	3.93	4.01	15.6	25.8	0.00	3.44	0.066	7.77	0.10	0.82	7,660
S4	6.25	5.07	6.24	17.6	35.5	0.00	5.08	0.073	10.20	0.13	1.21	13,300
S5	11.2	8.29	5.57	25.0	22.2	0.00	3.96	0.000	15.90	0.12	1.20	12,400
S6	8.33	6.21	217	231.3	93.7	0.00	4.46	0.067	7.51	0.09	0.90	9,450
S7	692	592	993	2277.4	43.6							
ERM						3.7	70.0	9.6	218.0			
ERL						1.0	8.2	1.2	46.7			
			Cr	Cu	Fe	Mn	Ni	Zn	Si	Se	Hg	
16P 0-2			16.80	92.9	23,200	176.0	5.28	118.0	140,000	0.75	0.061	
B3 0-2			10.30	97.4	11,700	94.1	3.12	92.9	57,000	0.34	0.050	
BB1 0-2			19.70	88.7	27,000	200.0	5.09	145.0	222,000	0.61	0.096	
BB2 0-2			61.50	1520.0	36,200	225.0	8.86	574.0	141,000	0.52	0.410	
S1			18.90	373.0	17,100	115.0	4.40	206.0	62,600	0.24	0.126	
S2			15.90	115.0	15,000	124.0	3.32	104.0	76,400	0.24	0.083	
S3			2.50	51.7	4,760	38.9	2.59	47.5	22,700	0.00	0.030	
S4			6.13	50.6	7,810	61.7	3.01	52.7	32,600	0.24	0.032	
S5			5.13	54.8	6,840	55.3	2.57	49.4	29,800	0.15	0.033	
S6			4.43	41.6	6,210	49.3	2.37	45.4	26,200	0.09	0.033	
S7												
ERM			370	270			51.6	410			0.71	
ERL			81	34			20.9	150			0.15	

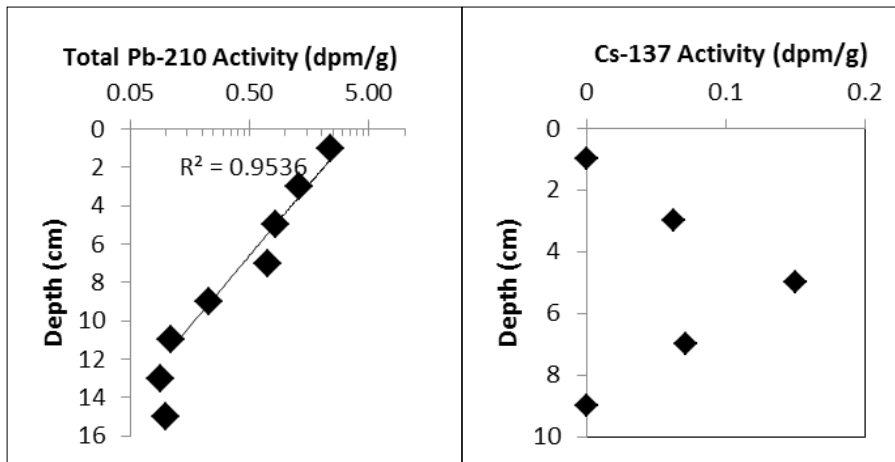


Figure 4. Plots of ^{210}Pb and ^{137}Cs activities with depth for core 16p.

into the marina facilities. Figure 7 shows the profile for core BB2. The ^{210}Pb profile for this core is also subject to interpretation because of the break in the data set. Accumulation rates were calculated both using all points and the lower points. Accumulation rates are 0.35 cm/y for all data points and 0.12 cm/y for only the lower points. A supported value of 0.1 dpm/g was used, consistent with cores 16P and BB1, since the core does not appear to be long enough for ^{210}Pb activities to have reached background activities. As noted previously, a higher background value would yield a lower accumulation rate. There is an anomaly at the 6-8 cm depth interval, which may be an event-related disturbance of the sediment layer similar to core B3. Station BB2 was in the heart of the marina facilities, adjacent to the boat ramp and travel lift.

^{137}Cs samples down to 12 cm were originally analyzed, with no detectable ^{137}Cs . The reason for this is unclear; one potential explanation is that sediment properties differ between this core and the others. The remaining

(1963). The first, at the top of the higher-activity layer, is 4-6 cm and yields an accumulation rate of 0.08-0.12 cm/y. One could also assume a typical profile shape and assume that the sediment in the 6-8 cm interval has anomalously low activities. If 6-8 cm is used as the depth of maximum activity, the accumulation rate would be 0.12-0.16 cm/y. However, the anomaly in the ^{210}Pb data occurs at the 6-8 cm depth interval, so it may be an event-related disturbance of the sediment layer is responsible. Station B3 is in the center of the channel leading

samples were counted with detections for the 20-22 cm and 22-24 cm intervals. However, these detections occurred at the 659 keV photopeak (part of gamma spectrum); the photopeak that represents ^{137}Cs -is 662 keV. In practice, a deviation of ± 2 keV is accepted when evaluating gamma spectra, using information about sample and site to guide decisions.

Thus, the 20-22 and 22-24 sections are considered borderline; they would likely be recorded as detectable ^{137}Cs , unless other evidence suggested otherwise. It is difficult to determine whether these detections represent the depth of first appearance or maximum activity. Using the point as a first occurrence (1954) yields an accumulation rate of 0.42 cm/yr. Using the point as a maximum (1963) occurrence yields an accumulation rate of 0.50 cm/yr.

The age of each layer of the cores as calculated by the ^{210}Pb and ^{137}Cs methods are shown in Table 2. The sections representing 1963 (upper) and 1954 (lower) are highlighted. These represent 50 and 59 years before 2013 respectively. The ages determined by the ^{137}Cs method appear to more closely reflect years before present (YBP). Years at 16P, BB1 and BB2 are overestimated while B3 is underestimated by the ^{210}Pb method. It is unclear which ^{137}Cs accumulation rate to use at BB2 so the most recent marker (1963) was used.

Grain Size in the Sediment Cores

The characteristics of the sediment in the area have changed dramatically over the years. Figures 8 and 9 show the proportion of fine grained (silt + clay) and gravel-sized particles in the cores over time and down the length of the cores. All the cores show a much higher percentage of gravel-sized material at the bottom of the cores than the top. These particles, however, were not gravel but were in fact shell hash (Figure 3). The shift toward fine-grained material is most dramatic at BB2. Station BB2 is surrounded by bulkheads and sits beneath constant boat activity. It is next to the main marina ramp. While BB2 was the deepest core, it covers the shortest time span due to the much higher accumulation rate (Table 2). Recall that there were anomalies in the cores from BB2 and B3 and that the calculated accumulation rates in the lower half of the cores were much lower than in the top half. A much lower accumulation rate in the deeper, older, layers at BB2 is logical as human activity has clearly and drastically altered the bottom sediment characteristics. Note also that the most dramatic shifts have occurred in the last 50-75 years.

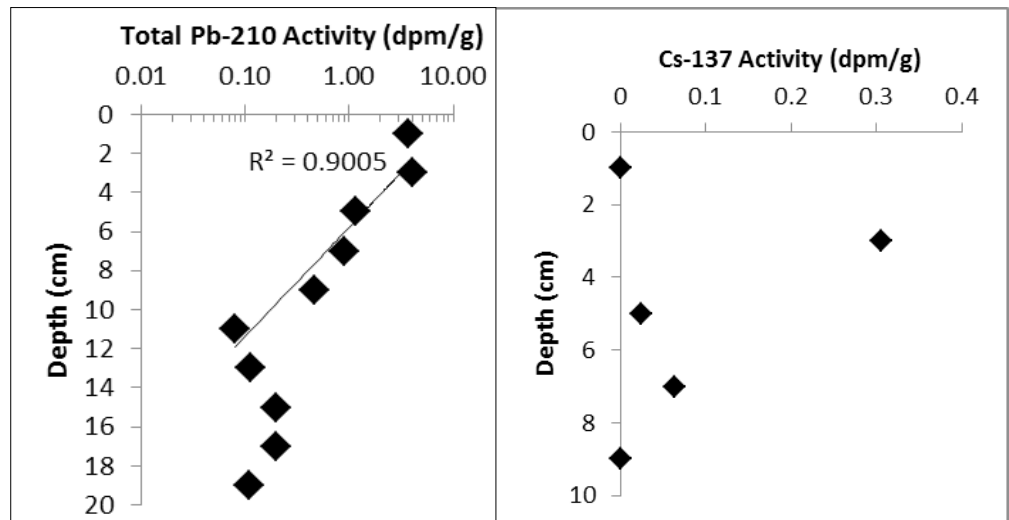


Figure 5. Plots of ^{210}Pb and ^{137}Cs activities with depth for core BB1.

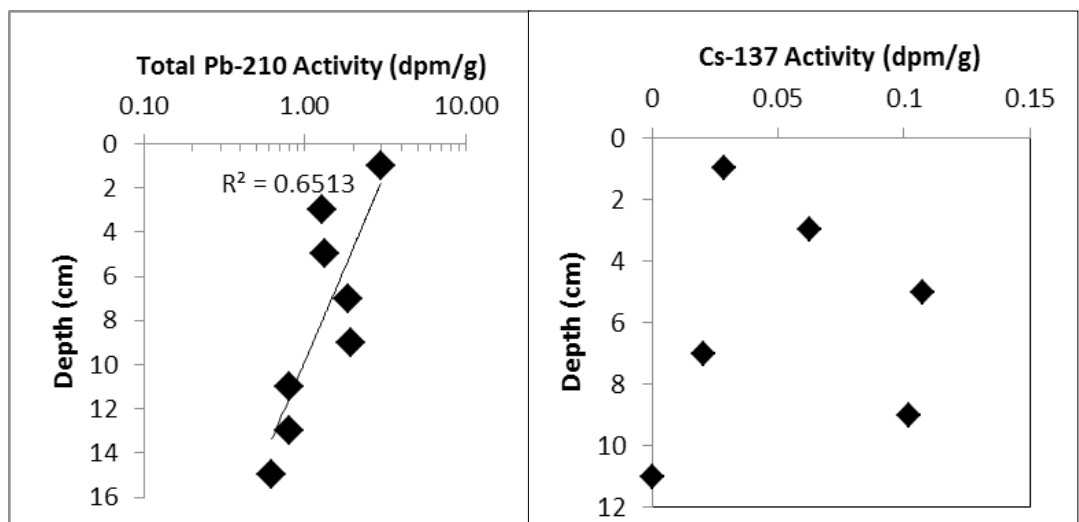


Figure 6. Plots of ^{210}Pb and ^{137}Cs activities with depth for core B3

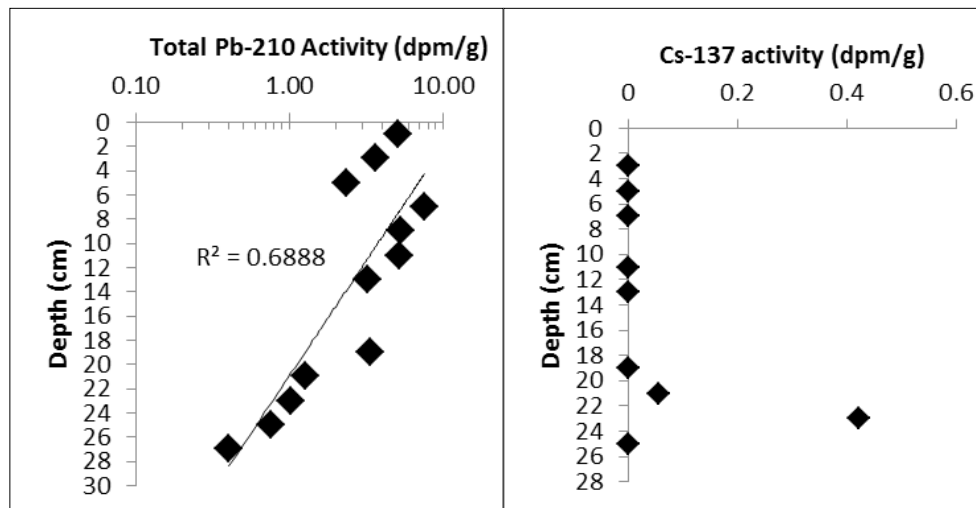


Figure 7. Plots of ^{210}Pb and ^{137}Cs activities with depth for core BB2.

Butyltin Concentrations in the Sediment Cores

The concentrations of total butyltins are shown in Figure 10. The peak concentrations occur at less than 50 years ago, which is consistent with historical use patterns. There are low levels of butyltins in sections 2-4 cm below 50 years ago which may reflect the initial buildup of TBT contamination and/or an indication of how deep sediments are churned by storms and bioturbation.

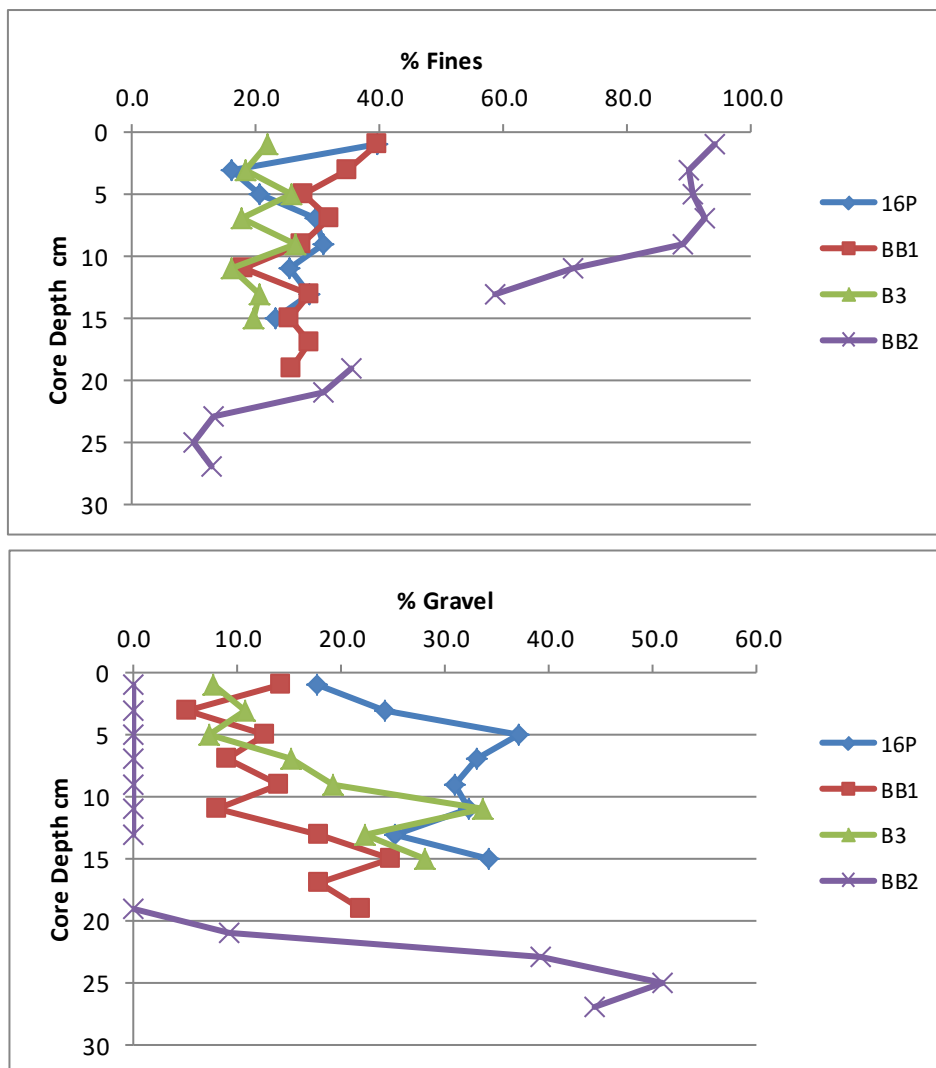


Figure 8. Proportion of fine grained (silt + clay) and gravel-sized particles down the length of the cores.

Butyltin concentrations are vastly higher at BB2 which also shows a peak in the past. The break in the data unfortunately occurs where the use of TBT was initiated so it is impossible to conclude if concentrations were even higher in the past, or if TBT use continued into more recent years. Concentrations have declined in recent years. Nevertheless, the observed concentrations at depth are vastly higher than anywhere else in Benner Bay or the rest of the STEER. Concentrations of this magnitude have only been observed in a few places (Page et al., 1996; Shim et al., 2002). Also, our data is reported as ng/g of tin (ng Sn/g), as opposed to TBT which has a 60% higher molecular weight than elemental tin. Of 1,506 data points in the NS&T data base with TBT sediment analyses, the median total butyltin value is 0.95 ng/g, the average is 8.9 ng Sn/g. The highest value is 990 ng Sn/g from Elliott Bay, a Superfund site in Puget Sound sampled in 1989.

Table 2. Estimated ages of sections within sediment cores from the STEER and the calculated accumulation rates. Sections indicating markers for the years 1954 and 1963 are highlighted.

Mean Core Depth cm	16P		BB1		B3		BB2		
	²¹⁰ Pb age	¹³⁷ Cs age	²¹⁰ Pb age	¹³⁷ Cs age	²¹⁰ Pb age	¹³⁷ Cs age	²¹⁰ Pb age	¹³⁷ Cs age 1954	¹³⁷ Cs age 1963
1	13.7	9.1	16.7	11.1	3.1	6.7	2.9	2.4	2.0
3	41.1	27.3	50.0	33.3	9.4	20.0	8.7	7.1	6.0
5	68.5	45.5	83.3	55.6	15.6	33.3	14.5	11.8	10.0
7	95.9	63.6	116.7	77.8	21.9	46.7	20.3	16.5	14.0
9	123.3	81.8	150.0	100.0	28.1	60.0	26.1	21.3	18.0
11	150.7	100.0	183.3	122.2	34.4	73.3	31.9	26.0	22.0
13	178.1	118.2	216.7	144.4	40.6	86.7	37.7	30.7	26.0
15	205.5	136.4	250.0	166.7	46.9	100.0			
17			283.3	188.9					
19			316.7	211.1			55.1	44.9	38.0
21							60.9	49.6	42.0
23							66.7	54.4	46.0
25							72.5	59.1	50.0
27							78.3	63.8	54.0
Rate mm/yr	0.73	1.10	0.60	0.90	3.20	1.50	3.45	4.23	5.0

There was relatively little butyltin in the B3 core. Neither is there evidence of historical changes over the years. Relative to the ¹³⁷Cs date markers, the ²¹⁰Pb analyses severely underestimated the age of the B3 sediments unlike the other cores (Table 2). There are obvious discontinuities in both the ¹³⁷Cs and ²¹⁰Pb records (Figure 6). It's location is in the middle of the channel, perhaps the natural channel, leading into inner Benner Bay. What events or processes (e.g. storms, dredging, etc.) may have contributed to the history of deposition at this location is unknown. The concentrations of the major soil elements of aluminum, iron and silicon all show a major shift at B3 in the mid-1940s (Figure 11), which coincides with the discontinuity in the ²¹⁰Pb and ¹³⁷Cs anomalies (Figure 6). Interestingly, the same is true for all the trace metals as well. Clearly, some event or change in circulation or terrestrial input occurred then.

Another puzzling aspect is the percentage of tributyltin as a proportion of total butyltins in the sediment. While the concentration of butyltins is decreasing over time, the proportion of tributyltin is increasing at 16P, BB1 and B3 up to the present, indicating continuing fresh inputs (Figure 12). The peak concentrations at BB2 in the marina are only 4-10 cm deep. The proportion of tributyltin at those depths is 50-70%. Prop wash from boats and storm-driven tidal currents may be the source of fresh tributyltin at the outer stations. Unlike BB2, the proportions below 10 cm depth in the cores at 16P, BB1 and B3 are meaningless as the concentrations are essentially zero and they date back to before TBT existed. Station 16P was located behind a mangrove island in the middle of the waterway between Benner Bay and Mangrove lagoon (Figure 2). There are moored boats, but no marina facilities in the immediate vicinity, so the TBT load there must be drifting in from other areas. The currents in the waterway are highly variable and dependent on tidal flux and wind driven currents. The last tropical storm to hit St. Thomas was Irene in 2011, which passed just south of the island with sustained gale force winds and heavy rain.

Gradients in Chemical Contamination

Chemical results from the top sections in the cores and the surface sediment grabs illustrate consistent gradients away from inner Benner Bay for almost all constituents. Metals and butyltin concentrations for the surface samples and the top layer of the cores are shown in Table 1. With three exceptions (Se, Sb, Si) the highest metals concentrations are all found at BB2. Metals concentrations were higher in general at the stations located closest to shore and declined further out into Benner Bay. Mercury was an order of magnitude higher at BB2 than all other stations except S1. Zinc exceeded the ERM. As noted above, the copper concentration was more than five times the ERM. Arsenic, Pb, Hg, and Zn exceeded the ERL at multiple stations. Aluminum, silicon, and iron are the most common elements in the earth's crust. Decreasing gradients of these elements leading away from Benner Bay all indicate a greater contribution of land-based sediment material close to the shoreline, as opposed to marine sources. The pattern of calcium decreasing from offshore toward Benner Bay and Mangrove Lagoon locations in the 2011 data are consistent with this interpretation (Pait et al., 2013). All of these observations, plus the historical patterns revealed in the cores indicate a disturbed habitat that is heavily and increasingly impacted by land-based sediment input and chemical contamination from boating related activities.

The gradient of butyltins in the surface from BB2 out into Benner Bay and down the channel toward Mangrove Lagoon clearly illustrate the impact of boating related activities (Table 1). Surface concentrations in the marina complex are one to two orders of magnitude higher than anywhere else. Butyltins at S1 leading out of the harbor were higher than at core B3 and those stations further out. Stations 2-5 were laid out on two transects leading away from core B3

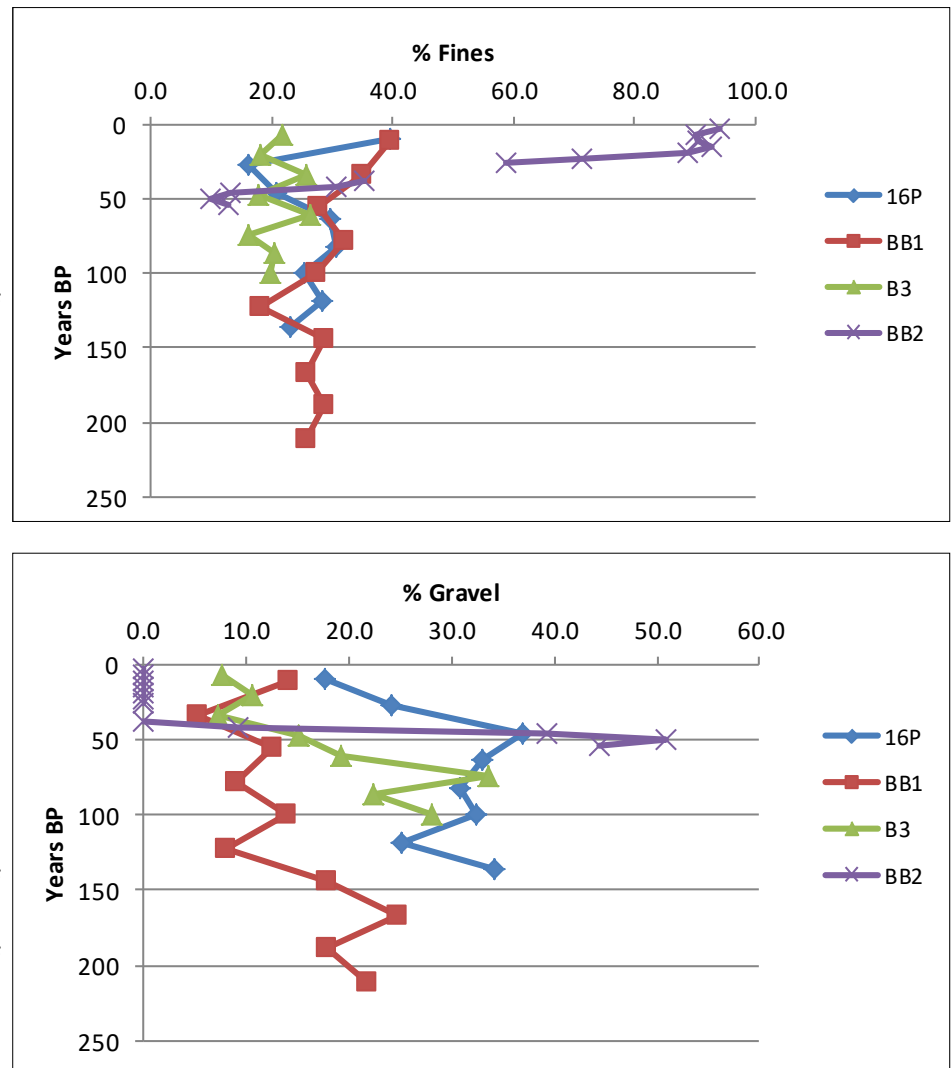


Figure 9. Percentage of fine grained sediment and gravel sized material on different routes. Concentrations (shell hash) as a function of time.

drop off rapidly further out. The one exception was station S6 that had a total butyltins concentration of 231 ug/g. Station S6 was taken on the edge of the channel leading out to open waters. The spike in concentration at that point, was almost as high as seen at S1. Notably, the percentage of tributyltin (relative to total butyltins) at that site was 93.7%, indicating fresh contamination. This strongly suggests a recent spill or perhaps grounding of a freshly painted vessel. It is unclear why butyltins are elevated at 16P and BB1, unless there are other sources in the area as well.

There is also a strong gradient of copper leading away from the marina complex (Table 1). Copper-based bottom paints were used before the advent of TBT, and copper-based paints have replaced the TBT paints. Note also, that copper at Station S6 is not elevated, in contrast to TBT. The concentration of copper in the cores show a

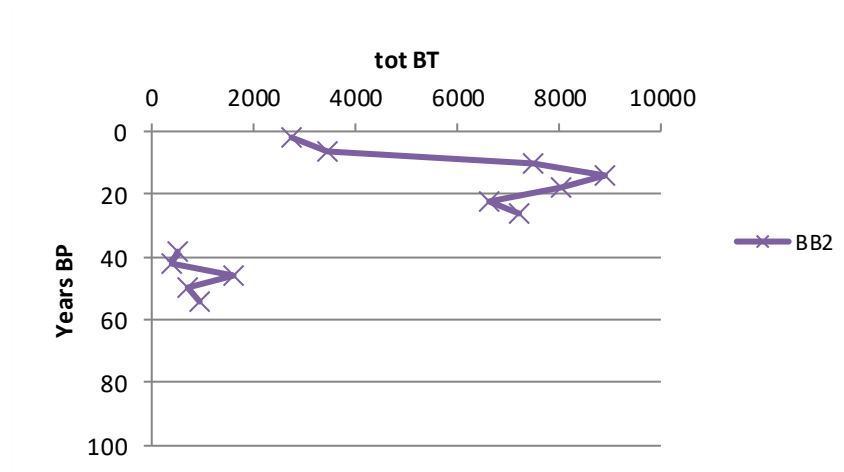
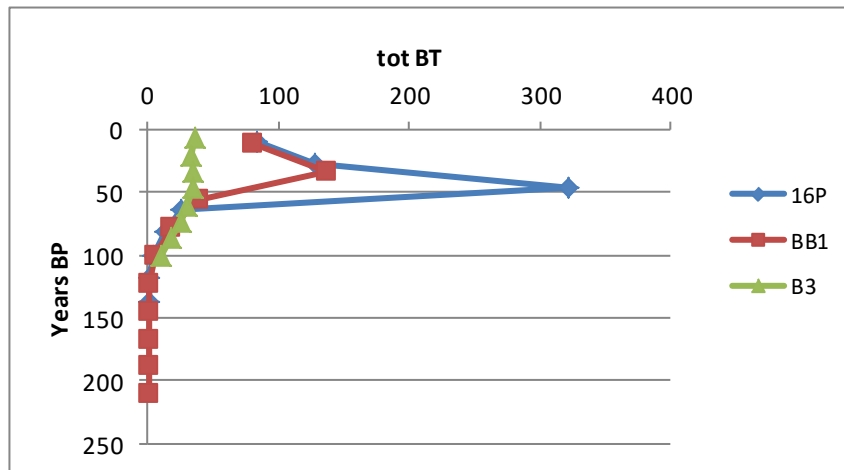


Figure 10. Concentration (ng Sn/g) of total butyltins as a function of time.

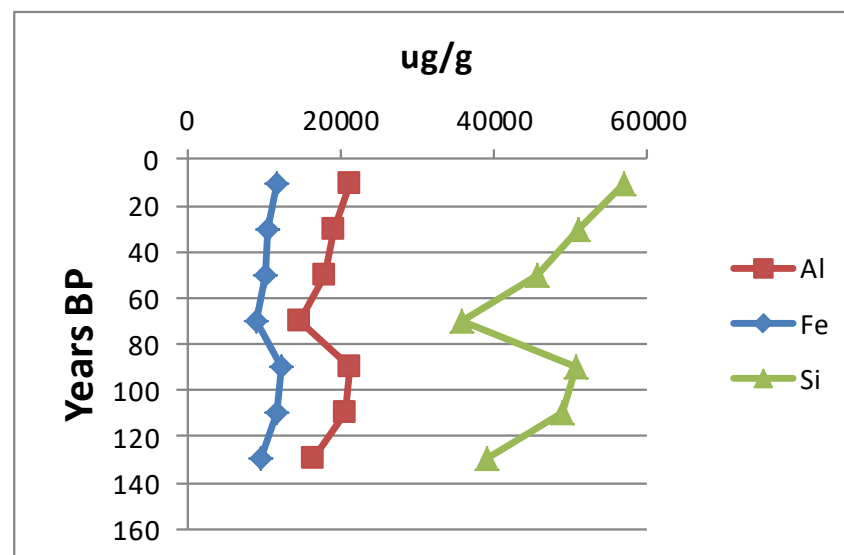


Figure 11. Changes in major element concentrations over time at station B3.

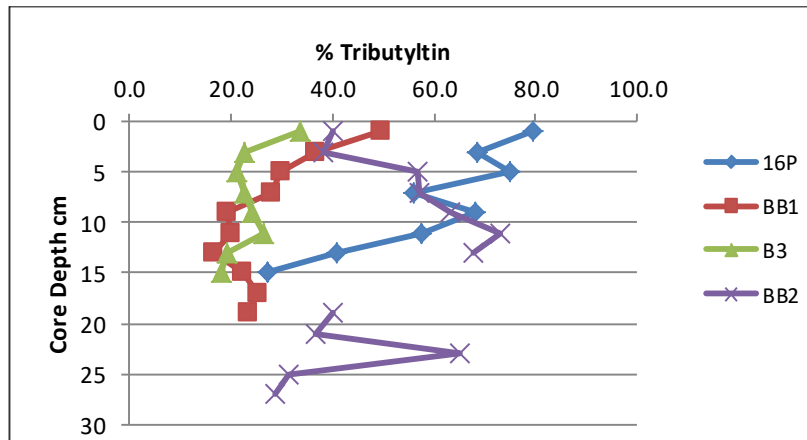
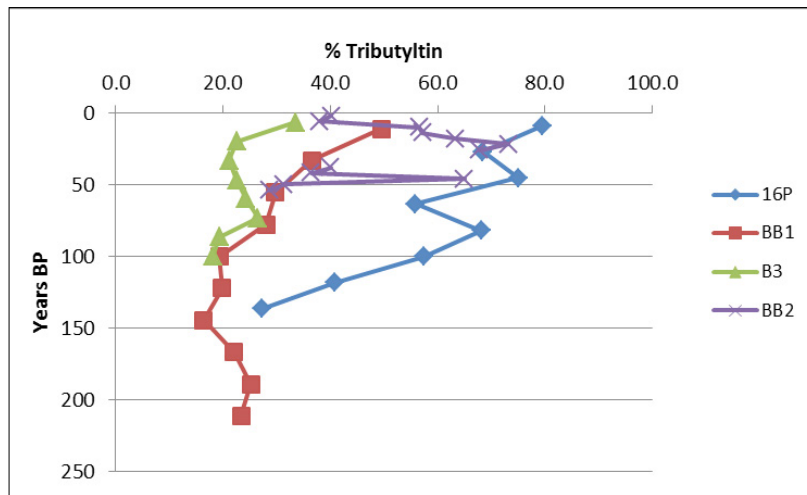


Figure 12. Percent tributyltin as a proportion of the total butyltins as a function of time and core depth.

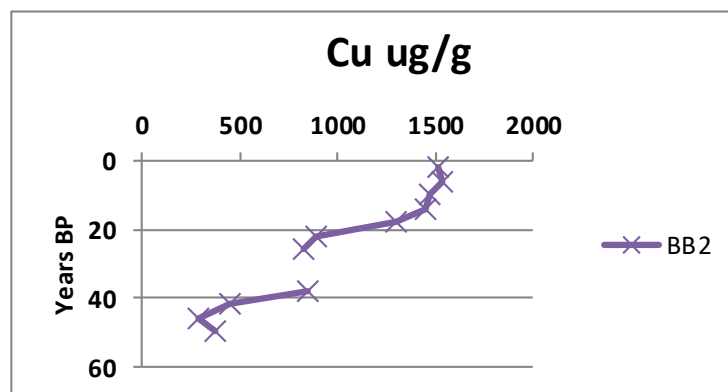
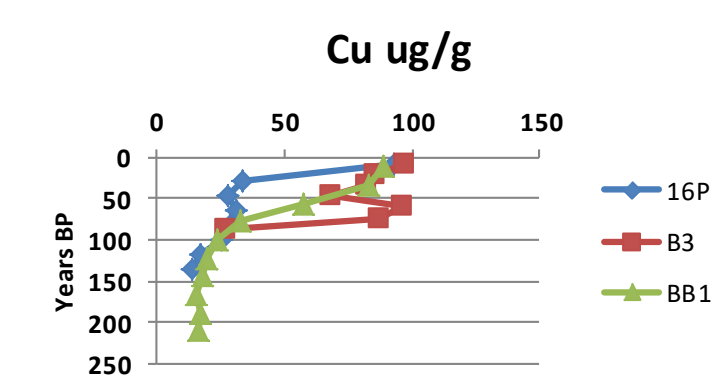


Figure 13. Copper concentrations as a function of time.

clear increase in concentrations over time at all locations (Figure 13), including at B3. Copper concentrations exceeded the ERL at all surface stations.

CONCLUSIONS

The inner reaches of northern Benner Bay are severely degraded by marina operations, shoreline development and watershed changes. Sediment dynamics and sediment quality are, and have been for decades, heavily impacted. The benthic community was once a thriving ecosystem as evidenced by the remnants of shelled species, that, at the bottom of some cores exceeded the volume of sediment present. That community has vanished and was smothered by very fine sediment that accumulates at rates an order of magnitude above normal rates. The sediment is highly contaminated with butyltin paint residues, copper, and other toxic metals. The sediment is contaminated below the surface as well. Dredging new or deeper channels will spread these contaminants over a wide area. Dredging for remediation purposes is advisable, but will be expensive to do with methods that will properly prevent dredge spoil release to the water column. Otherwise, the benthic community will likely never recover, and environmental damage will expand as contaminants are diffused over an even increasing area. Matthiessen (2013) reported that benthic communities do not recover from TBT contamination until the concentration of butyltins is reduced to 10-40 ng/g. The concentration of butyltins at a depth of 6-8 cm was 8,871 ng/g at station BB2. The system may be receiving fresh inputs of TBT as evidenced by the percentage of tributyltin residues at Station S6. The system is dynamic. Between sampling in 2011 and 2013, the concentration of TBT had more than doubled at station BB2. Copper had increased by 50%. What event(s) may have caused such large changes in such a short time are unknown. Boat groundings, prop wash, and pile driving are all likely activities present at a marina that would stir up the bottom, and bring up contaminants from deeper in the sediment column.

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APPENDICES

Appendix A. Butyltin concentrations (ng Sn/dry g) in core and surface sediment samples from the STEER in 2013.

Site	Depth (cm)	Monobutyltin	Dibutyltin	Tributyltin	Tetrabutyltin	%Tributyltin
16P	0-2	6.61	10.43	66.43	0.31	79.58
16P	2-4	7.49	32.87	87.62	0.22	68.46
16P	4-6	12.96	67.61	241.98	0.29	75.02
16P	6-8	3.16	8.13	14.32	0.10	55.92
16P	8-10	1.29	3.19	9.60	0.11	68.19
16P	10-12	0.59	1.54	2.87	0.06	57.39
16P	12-14	0.53	0.55	0.75	-	40.83
16P	14-16	0.62	0.36	0.37	-	27.23
BB1	0-2	12.60	28.19	40.02	0.14	49.52
BB1	2-4	42.09	45.15	50.22	0.72	36.53
BB1	4-6	13.24	14.64	11.80	0.09	29.75
BB1	6-8	5.91	6.80	4.95	-	28.04
BB1	8-10	2.45	2.70	1.23	-	19.32
BB1	10-12	0.79	0.81	0.40	-	19.86
BB1	12-14	0.79	0.60	0.27	-	16.46
BB1	14-16	0.52	0.46	0.28	-	22.08
BB1	16-18	0.47	0.56	0.35	-	25.19
BB1	18-20	0.50	0.37	0.26	-	23.40
B3	0-2	13.32	11.20	12.39	0.20	33.56
B3	2-4	16.90	8.42	7.38	0.13	22.58
B3	4-6	18.01	9.19	7.29	0.14	21.14
B3	6-8	17.82	9.81	8.06	0.13	22.59
B3	8-10	13.66	9.35	7.37	0.16	24.27
B3	10-12	9.90	8.63	6.70	0.13	26.56
B3	12-14	9.40	4.88	3.43	0.08	19.36
B3	14-16	5.03	3.60	1.91	-	18.12
BB2	0-2	939.63	699.68	1,101.90	37.57	40.20
BB2	2-4	1,257.45	882.26	1,316.91	47.97	38.10
BB2	4-6	1,580.24	1,655.57	4,227.50	96.59	56.64
BB2	6-8	1,974.14	1,823.88	5,072.91	141.42	57.19
BB2	8-10	1,050.79	1,894.26	5,082.79	74.79	63.31
BB2	10-12	494.72	1,274.96	4,830.40	9.69	73.19
BB2	12-14	731.32	1,587.29	4,900.30	9.15	67.88
BB2	18-20	147.90	148.23	198.48	3.18	40.13
BB2	20-22	126.64	123.16	143.47	2.57	36.48
BB2	22-24	287.73	275.67	1,047.10	3.89	65.02
BB2	24-26	240.11	244.04	221.06	1.13	31.35
BB2	26-28	322.10	358.41	274.19	1.74	28.72
S1	/	72.58	68.69	133.73	1.20	48.63
S2	/	25.71	9.74	12.19	0.39	25.59
S3	/	7.61	3.93	4.01	0.10	25.77
S4	/	6.25	5.07	6.24	0.11	35.54
S5	/	11.18	8.29	5.57	0.11	22.24
S6	/	8.33	6.21	216.82	1.82	93.72
S7	/	691.80	592.41	993.15	12.79	43.61

Appendix B. Elemental concentrations (ug/ dry g) in core and surface sediment samples from the STEER in 2013.

Site	Depth cm	Ag	Al	As	Cd	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Sb	Se	Si	Sn	Zn
16P	0-2	0.00	38,600	11	0.156	16.8	92.9	23,200	0.061	176.0	5.3	15.70	0.46	0.75	140,000	1.99	118.0
16P	2-4	0.00	26,200	8.85	0.141	11	33.5	14,700	0.052	115.0	4.0	9.04	0.48	0.50	118,000	1.10	88.3
16P	4-6	0.00	30,400	12	0.094	13.2	27.7	17,700	0.041	150.0	4.1	8.11	0.61	0.53	131,000	1.08	50.5
16P	6-8	0.00	37,400	16.3	0.117	13.6	31.1	21,800	0.037	176.0	4.6	6.59	0.61	0.68	158,000	2.11	51.7
16P	8-10	0.00	40,200	16.8	0.074	13.1	28.5	22,800	0.028	191.0	4.4	4.90	0.70	0.87	164,000	2.56	49.0
16P	10-12	0.00	33,900	17.5	0.284	12.9	25.2	20,500	0.021	163.0	5.2	3.41	0.79	1.11	138,000	0.18	41.6
16P	12-14	0.00	21,500	14.1	0.483	11.5	17.4	13,300	0.018	96.9	5.1	2.08	0.59	1.45	80,200	0.00	28.9
16P	14-16	0.00	14,400	11.3	0.613	6.76	14.1	9,280	0.017	60.3	4.6	2.13	0.51	1.32	55,200	0.00	22.0
BB1	0-2	0.00	48,900	9.64	0.230	19.7	88.7	27,000	0.096	200.0	5.1	25.40	1.00	0.61	222,000	3.32	145.0
BB1	2-4	0.00	50,500	9.56	0.315	21.8	83.0	26,900	0.092	205.0	5.0	22.60	0.56	0.65	227,000	2.71	141.0
BB1	4-6	0.00	42,000	11.6	0.215	17.6	57.7	22,500	0.096	166.0	3.9	20.10	0.60	0.63	204,000	1.94	103.0
BB1	6-8	0.00	37,900	12.9	0.127	14.4	32.8	19,900	0.071	146.0	3.6	11.70	0.61	0.46	192,000	1.00	64.8
BB1	8-10	0.00	34,100	14	0.097	11.9	23.8	17,900	0.046	133.0	3.5	9.00	0.52	0.43	174,000	0.54	46.8
BB1	10-12	0.00	36,600	18.2	0.096	13.9	19.8	20,700	0.026	153.0	3.4	21.40	0.64	0.64	189,000	0.13	37.2
BB1	12-14	0.00	31,200	17	0.141	10.7	18.1	17,800	0.021	126.0	3.8	3.49	0.78	0.85	161,000	0.19	32.9
BB1	14-16	0.00	25,300	14.5	0.185	9.65	15.4	14,800	0.017	98.9	4.1	2.13	0.79	1.05	125,000	0.00	28.8
BB1	16-18	0.00	26,300	14.9	0.188	13.5	17.2	15,000	0.016	108.0	4.8	2.55	0.74	1.02	135,000	0.00	32.3
BB1	18-20	0.00	27,100	12.9	0.172	11.5	16.3	14,500	0.015	108.0	3.7	1.93	0.67	1.17	103,000	0.29	29.5
B3	0-2	0.00	21,400	10	0.085	10.3	97.4	11,700	0.050	94.1	3.1	17.40	0.22	0.34	57,000	1.76	92.9
B3	2-4	0.00	19,300	5.24	0.085	8.42	85.3	10,400	0.050	82.7	3.1	15.00	0.18	0.25	50,900	1.75	81.8
B3	4-6	0.00	18,100	5.19	0.067	9.21	81.9	10,200	0.054	79.6	3.1	13.70	0.17	0.30	45,800	1.44	79.9
B3	6-8	0.00	14,700	5.63	0.091	5.48	68.2	8,990	0.045	64.5	2.8	12.60	0.17	0.27	35,700	1.25	67.0
B3	8-10	0.00	21,400	6.43	0.097	11.1	95.8	12,400	0.060	92.6	3.8	15.80	0.22	0.29	50,800	1.84	90.5
B3	10-12	0.00	20,600	5.62	0.094	8.91	87.1	11,600	0.053	87.8	3.3	15.70	0.27	0.34	48,800	1.63	85.6
B3	12-14	0.00	16,500	8.24	0.070	6.74	27.3	9,790	0.028	72.7	2.6	6.86	0.32	0.34	39,000	0.77	35.3
B3	14-16																
BB2	0-2	0.27	64,700	16.4	0.281	61.5	1520.0	36,200	0.410	225.0	8.9	129.00	0.73	0.52	141,000	22.20	574.0
BB2	2-4	0.24	73,700	18.4	0.329	63.4	1540.0	41,200	0.473	251.0	9.4	125.00	0.78	0.73	162,000	25.00	2460.0
BB2	4-6	0.24	72,100	19.7	0.539	61.3	1470.0	41,700	0.594	262.0	12.4	134.00	0.87	0.81	162,000	31.10	664.0
BB2	6-8	0.29	69,700	20.2	0.553	63.3	1450.0	43,500	0.637	296.0	10.7	139.00	0.85	0.86	155,000	34.30	615.0
BB2	8-10	0.15	69,700	19.2	0.599	52.6	1300.0	42,300	0.552	283.0	10.5	126.00	0.84	0.98	223,000	29.20	565.0
BB2	10-12	0.18	68,400	18.1	1.030	44	885.0	41,800	0.627	281.0	10.6	124.00	0.77	0.87	218,000	22.80	506.0
BB2	12-14	0.00	62,500	13.5	0.499	31	824.0	35,100	0.600	241.0	8.2	114.00	0.67	0.75	224,000	16.50	338.0
BB2	18-20	/	/	/	/	/	/	/	/	/	/	/	/	/	/	/	/
BB2	20-22	0.00	52,800	0	0.114	27.3	842.0	24,600	0.267	201.0	4.6	74.30	0.58	0.46	184,000	11.40	330.0
BB2	22-24	0.07	41,300	12.4	0.101	13.6	444.0	16,300	0.176	233.0	2.3	40.40	0.60	0.32	193,000	6.11	812.0
BB2	24-26	0.00	35,900	8.99	0.130	8.95	284.0	14,800	0.202	134.0	2.0	38.20	0.64	0.35	165,000	6.50	144.0
BB2	26-28	0.00	36,100	8.42	0.183	10.7	374.0	16,600	0.361	122.0	2.8	89.50	1.13	0.44	141,000	6.10	157.0
S1	/	0.08	29,500	9.67	0.078	18.9	373.0	17,100	0.126	115.0	4.4	119.00	1.49	0.24	62,600	5.25	206.0
S2	/	0.00	26,500	6.96	0.080	15.9	115.0	15,000	0.083	124.0	3.3	24.10	0.19	0.24	76,400	3.06	104.0
S3	/	0.00	7,660	3.44	0.066	2.5	51.7	4,760	0.030	38.9	2.6	7.77	0.10	0.00	22,700	0.82	47.5
S4	/	0.00	13,300	5.08	0.073	6.13	50.6	7,810	0.032	61.7	3.0	10.20	0.13	0.24	32,600	1.21	52.7
S5	/	0.00	12,400	3.96	0.000	5.13	54.8	6,840	0.033	55.3	2.6	15.90	0.12	0.15	29,800	1.20	49.4
S6	/	0.00	9,450	4.46	0.067	4.43	41.6	6,210	0.033	49.3	2.4	7.51	0.09	0.09	26,200	0.90	45.4

Appendix C. Percent grainsize distributions in core and surface sediment samples from the STEER in 2013.

Site	Depth cm	%GRAVEL	%SAND	%SILT	%CLAY
16P	0-2	17.6	42.9	11.6	27.9
16P	2-4	32.3	42.3	7.7	17.6
16P	4-6	25.0	46.5	11.9	16.6
16P	6-8	34.1	42.7	13.4	9.7
16P	8-10	24.2	59.7	5.0	11.1
16P	10-12	37.0	42.4	9.2	11.5
16P	12-14	33.0	37.5	11.4	18.2
16P	14-16	30.8	38.3	11.3	19.6
BB1	0-2	14.2	46.2	19.8	19.8
BB1	2-4	17.8	53.5	18.1	10.7
BB1	4-6	9.0	59.1	21.3	10.6
BB1	6-8	21.8	52.5	15.6	10.1
BB1	8-10	17.9	53.5	18.6	10.0
BB1	10-12	13.9	58.7	17.4	10.0
BB1	12-14	12.6	59.6	18.7	9.0
BB1	14-16	5.2	59.9	29.2	5.7
BB1	16-18	24.7	49.7	20.4	5.1
BB1	18-20	8.1	73.8	13.4	4.8
B3	0-2	7.7	70.5	12.4	9.4
B3	2-4	33.7	50.3	5.9	10.1
B3	4-6	22.3	57.3	9.2	11.2
B3	6-8	28.0	52.3	8.6	11.1
B3	8-10	10.7	71.2	8.5	9.6
B3	10-12	7.3	66.9	12.1	13.7
B3	12-14	15.1	67.2	5.8	11.9
B3	14-16	19.3	54.5	12.4	13.9
BB2	0-2	0.0	10.0	37.5	52.5
BB2	2-4	0.0	7.3	41.4	51.3
BB2	4-6	0.0	9.5	39.8	50.7
BB2	6-8	0.0	11.1	38.7	50.2
BB2	8-10	0.0	5.9	44.9	49.2
BB2	10-12	0.0	28.6	27.2	44.2
BB2	12-14	0.0	41.2	20.7	38.1
BB2	18-20	9.2	60.0	13.7	17.1
BB2	20-22	0.0	64.6	19.0	16.4
BB2	22-24	39.2	47.5	6.8	6.5
BB2	24-26	44.4	42.9	6.8	5.9
BB2	26-28	50.8	39.4	5.5	4.4
S1	/	27.0	46.1	11.4	15.6
S2	/	4.2	51.7	27.5	16.6
S3	/	5.6	80.1	6.0	8.3
S4	/	11.4	74.7	9.6	4.3
S5	/	8.5	71.1	14.5	5.9
S6	/	2.0	72.7	18.3	7.0
S7	/	0.0	4.4	54.8	40.8



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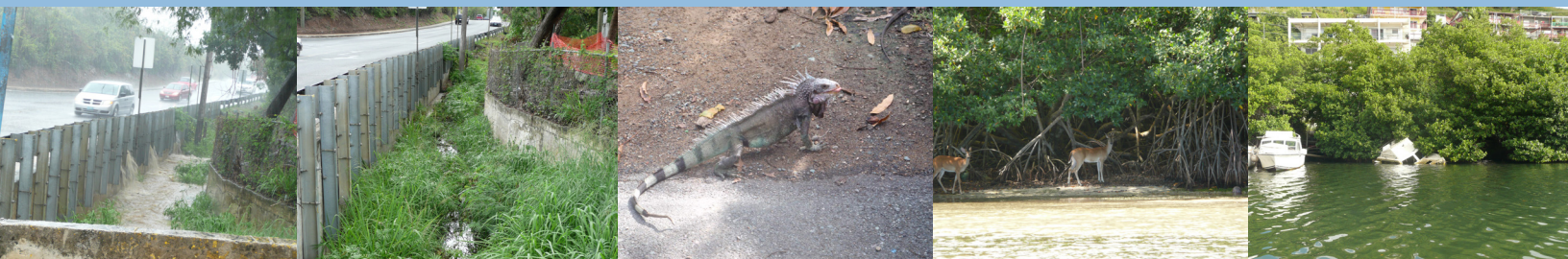
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Benjamin Friedman, *Acting Administrator*

National Ocean Service

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