

## Depositional history of organic contaminants in Narragansett Bay, Rhode Island, USA

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### Abstract

Sediment cores were taken at three locations in Narragansett Bay, Rhode Island, USA in 1997 and analyzed for a variety of organic contaminants including polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), chlordanes, linear alkyl benzenes (LABs), benzotriazoles (BZTs) and dichlorodiphenyltrichloroethanes (DDTs) and metabolites. The distributions of these chemicals at Apponaug Cove and in the Seekonk River indicate that there was a disturbance in the depositional environment relative to cores collected at these locations in 1986 demonstrating the potential for buried contaminants to be remobilized in the environment even after a period of burial. Sharp breaks in the concentration of several organic markers with known dates of introduction were successfully used to determine the sedimentation rate at Quonset Point. Both the Quonset Point and Seekonk River cores had subsurface maximums for DDTs, PCBs, PAHs and BZTs, which are consistent with expected inputs to the environment. The Apponaug Cove core showed an increase of most contaminants at the surface indicating a recent event in which more contaminated sediments were deposited at that location.

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**Keywords:** PCBs; PAHs; Sediments; Narragansett bay; Linear alkyl benzenes; Benzotriazoles

### 1. Introduction

A wide range of organic contaminants have entered the environment since the beginning of the industrial revolution. An increase in petroleum hydrocarbons in sediment cores has been associated with the increase in use of petroleum at the end of the 19th century (Hurt and Quinn, 1979; Gschwend and Hites, 1981). Other synthetic organic contaminants, such as dichlorodiphenyltrichloroethanes (DDTs) and polychlorinated biphenyls (PCBs), have also been used as time markers in sediment cores to reconstruct a depositional history of an area (Hom et al., 1974; Latimer and Quinn, 1996). A number of these chemicals have been banned or decreased in use over the last 30–50 years resulting in lower

concentrations in the surface sediments, yet they may still pose a risk if contaminants in deeper sections are mobilized through dredging or erosional processes.

Some of the chemicals examined in this study are well recognized to have deleterious effects in the environment. Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous trace contaminants in the marine environment and they vary both in their acute toxicity and sublethal effects to aquatic organisms (Neff, 1979); in addition, they may also bioaccumulate in shellfish and other benthic organisms (Stegeman and Teal, 1973; Harris et al., 1977; Fucik and Neff, 1977), which gives them a pathway to humans. Some of these PAHs, such as benzo[*a*]pyrene, 7,12-dimethylbenzo[*a*]anthracene, 3-methylcholanthrene, and benzo[*a*]phenanthrene, have been shown to be carcinogenic to mammals (Neff, 1979). PCBs were produced in the United States from 1929 through 1977 with peak production in 1970 when approximately 39,000 metric tons were produced

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(Versar, 2000). Since the 1977 ban on the production and use of PCBs in the US, the major sources of PCB releases to the air, soil and water are from cycling of PCBs between reservoirs through processes such as sediment resuspension, runoff, or volatilization (Versar, 2000). PCBs have numerous health effects including acting as endocrine disrupters, having neurological, immune and reproductive effects, and in addition are classified as probable human carcinogens (EPA, 1999). DDTs and chlordanes (first produced in 1940 and 1946, respectively) were also banned in the US in 1972 and 1988, respectively, due to environmental and health concerns. In the environment, DDTs are highly toxic to aquatic invertebrates and fish and were thought to be responsible for eggshell thinning in predator species of birds (EXTOXNET, 1996). They are also classified by the EPA as probable human carcinogens as well as having acute toxicity (Environmental Health Center, 2000). Chlordane is of concern to humans because it is both acutely toxic and a carcinogen (Environmental Health Center, 2000). The DDTs and chlordanes are important to study because they are persistent in the environment, bioaccumulate and are now found globally at all trophic levels including humans (EPA, 1996; Finizio et al., 1998; Muller and Buser, 1994; Buser and Muller, 1992).

Narragansett Bay is an urban estuary that has been severely impacted by pollution in the past, although conditions have improved dramatically in the past 30 years. Historically, the worst of the pollution has been in the urban upper reaches of the bay; however, shifting populations toward the more rural areas further south along the shoreline of the bay have the potential to negatively impact the mid and lower bay areas. The Narragansett Bay watershed, particularly the Blackstone and Pawtuxet Rivers, has a history of industrial activity, such as textiles, jewelry and metal working in particular (Desbonnet and Lee, 1991) that have contributed large quantities of PCBs to the bay's sediments, (Latimer et al., 1990; Latimer and Quinn, 1996). As in many marsh areas around the country, DDT was sprayed locally around Narragansett Bay to control mosquitoes. Chlordane was extensively used for pest control in homes and agriculture, possibly reaching the bay through run-off. A previous study of the bay (Quinn et al., 1992), with samples collected in 1985–1986, found PCBs and DDTs in surface sediments and sediment cores. Understanding how PCBs and organochlorine pesticides (OCPs) are distributed in the bay and how they are changing temporally is important for assessing the risk to human populations that depend on Narragansett Bay. For example, Narragansett Bay is also an economically important fishery (e.g. quahogs and lobsters) that could provide a source of these contaminants to man. Continued long term monitoring programs for various pollutants are essential to understanding how the bay is being affected by changes in population pressures and industrial develop-

ment. Towards that end, this study used three sediment cores to assess temporal changes in the geochemical history of contaminants in Narragansett Bay.

## 2. Methods

### 2.1. Sampling

Sediment cores were collected in 1997 with a piston corer at three locations, stations 27, 36 and 42 in Narragansett Bay (Fig. 1). The cores were stored upright until they were returned to the laboratory and then stored at 4 °C for up to 1 week. Two of the cores (stations 27 and 42) were split into 2 cm sections, and the third core (station 36) was split into 10 cm sections. An aliquot of each section was placed in a solvent cleaned glass jar, homogenized and stored at –20 °C until analyzed.

### 2.2. Sample extraction and analysis

The extraction and analysis has previously been described in detail (Hartmann, 2001; Hartmann et al.,

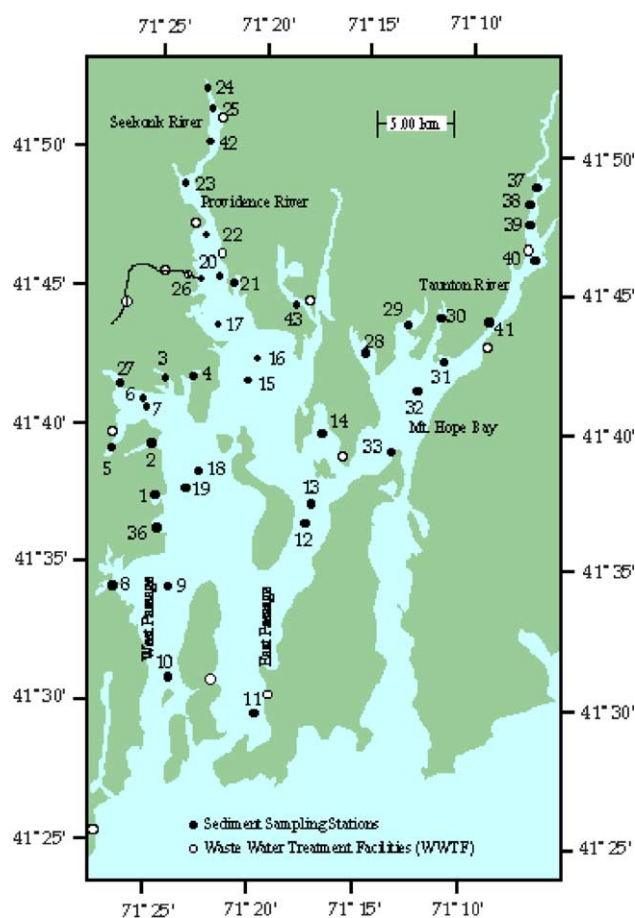


Fig. 1. Sediment core were taken at stations 27, 36 and 42. Surface sediments were taken at all other stations and are discussed elsewhere (Hartmann et al., 2000, 2004a,b).

2000). For the PAHs, 1–6 ml of four deuterated PAHs, naphthalene- $d_8$ , acenaphthene- $d_{10}$ , anthracene- $d_{10}$ , and benzo[*e*]pyrene- $d_{12}$ , each at  $\sim 2000$  ng/ml in acetonitrile, were added as internal standards. The internal standard for the PCBs and pesticides contained DBOFB (dibromooctachlorobiphenyl) 0.09  $\mu\text{g/ml}$ , CB103 0.10  $\mu\text{g/ml}$ , and CB198 0.39  $\mu\text{g/ml}$ , in acetonitrile. PAHs, total petroleum hydrocarbons (TPHs), benzotriazoles (BZTs) and linear alkyl benzenes (LABs) were analyzed on Hewlett Packard HP5890 gas chromatograph (GC) fitted with a 5971A quadrupole mass spectrometer (MSD) using condition previously described (Hartmann et al., 2000) or with an Agilent 6890/5973 GC/MSD (Hartmann, 2001). Data was collected using selected ion monitoring (SIM), quantified on the primary ion and confirmed with secondary and tertiary ions. Twenty parent PAHs, five mono-, di-, or tri-methylated PAHs and 20 PAH alkyl homologs were analyzed and were reported as the  $\Sigma$  PAHs. A response identical to the parent compound was assumed except in cases where we had an authentic standard of the substituted PAHs.

PCBs and OCPs were analyzed on Hewlett Packard (HP) 5890 gas chromatograph with an electron capture detector (ECD). A J&W Scientific Durabond DB-5, 60 m fused silica capillary column, 0.25 mm i.d. and 0.25  $\mu\text{m}$  film thickness was used with helium as the carrier gas at a linear velocity of about 36 cm/s. Injection port was maintained at 315  $^{\circ}\text{C}$ , and the sample was injected in splitless mode. Column temperature was held at 100  $^{\circ}\text{C}$  for initial 1.0 min, and then programmed at 11  $^{\circ}\text{C/min}$  to 130  $^{\circ}\text{C}$ , held for 2 min, ramped at 5  $^{\circ}\text{C/min}$  to 190  $^{\circ}\text{C}$ , held for 6 min, ramped at 5  $^{\circ}\text{C/min}$  to 310  $^{\circ}\text{C}$  and held for 5 min. The peaks were identified by matching the retention time of the peaks in the sample with those of authentic standards in the calibration solution. PCB/OCPs in selected samples were confirmed by GC/MS using primarily a HP 5890 gas chromatograph fitted with an HP 5971A quadrupole mass spectrometer and a J&W Scientific Durabond DB-XLB 30 m fused silica capillary column, 0.25 mm i.d. and 0.25  $\mu\text{m}$  film thickness using helium as the carrier gas with a linear velocity of about 30 cm/s.

### 2.3. Quality control

Calibration and quality control was rigorous throughout the study. A three-point calibration curve was used for 24 CBs and 13 OCPs covering the range of material expected in the samples. The  $\Sigma$  PCBs are reported as two times the sum of the individual chlorinated biphenyls (CBs) measured based on the analysis of standard mixtures of Aroclor 1242, 1254, and 1260 in our laboratory. The  $\Sigma$  chlordanes is the sum of  $\alpha$ -chlordane, *trans*-nonachlor, heptachlor, and heptachlor epoxide and the  $\Sigma$  DDTs are the sum of the *p,p'*-DDT, *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDE and *p,p'*-DDD.

The mid level calibration solution was injected at least once a week to ensure the response factors remained constant. Quality control for the analysis was done by analyzing blanks, spiked samples, duplicates and analyzing a standard reference sediment, SRM1941a marine sediment, with certified values from the National Institute of Standards and Technology (Schantz et al., 1995). One or more of these was done with each batch of samples with a minimum of four QC samples per twenty field samples. The acceptable limits for recovery in the spiked blanks was from 70% to 130% for each analyte, the duplicates agreed within 30% of one another and the recoveries of analytes were  $\pm 30\%$  of the certified values for the SRM1941a. All values reported for the sediments are on a dry weight basis.

## 3. Results and discussion

### 3.1. Quonset Point

For station 36, Quonset Point, the 0–2 cm section was from the surface grab sample taken at the same time as the core while the 0–10 cm section is the top section of the core. At this station,  $\Sigma$  PCBs were seen in all core sections and the  $\Sigma$  DDTs were seen in all but the bottom core sections (detection limit 0.63 ng/g) (Fig. 2). The  $\Sigma$  chlordanes were below the detection limit (0.79 ng/g) in all core sections (data not shown). The  $\Sigma$  PCBs show a maximum at 40–50 cm. Based on the estimated sedimentation rate at this site of  $\sim 2$  cm/y (see following discussion and Fig. 3), this would represent approximately the years 1972–1977 which is just after the peak in PCB production in 1970 (Versar, 2000). The concentration in the 50–60 cm section representing  $\sim 1967$ –1972 also was not significantly lower, given the analytical uncertainty. The  $\Sigma$  DDTs peak in the 50–60 cm section just prior to their ban in 1972. This station had a relatively small range (3200–5400 ng/g) in total PAH concentration from the 0–2 cm section down through the 100–110 cm section. The concentrations are relatively low in the surface with the values increasing through 30–40 cm and then decreasing below 70 cm to a low of 383 ng/g in the 119–129 cm section. Somewhat lower concentrations in the top 30 cm may reflect the diminished use of Quonset Point over the past 15 years. The rapid increase in PAHs between the 119–129 cm and 100–110 cm section likely reflects the beginning of Quonset Point as a Navy base in the 1939. The 119–129 cm section is from a second sediment core taken at the same location at the same time and was matched to the first sediment core. The TPHs (data not shown) have a similar pattern to the PAHs, but the changes are more dramatic. There is large increase in TPHs up core to a 60–70 cm subsurface maximum, and then a decrease to the top. The overall pattern of TPH may also

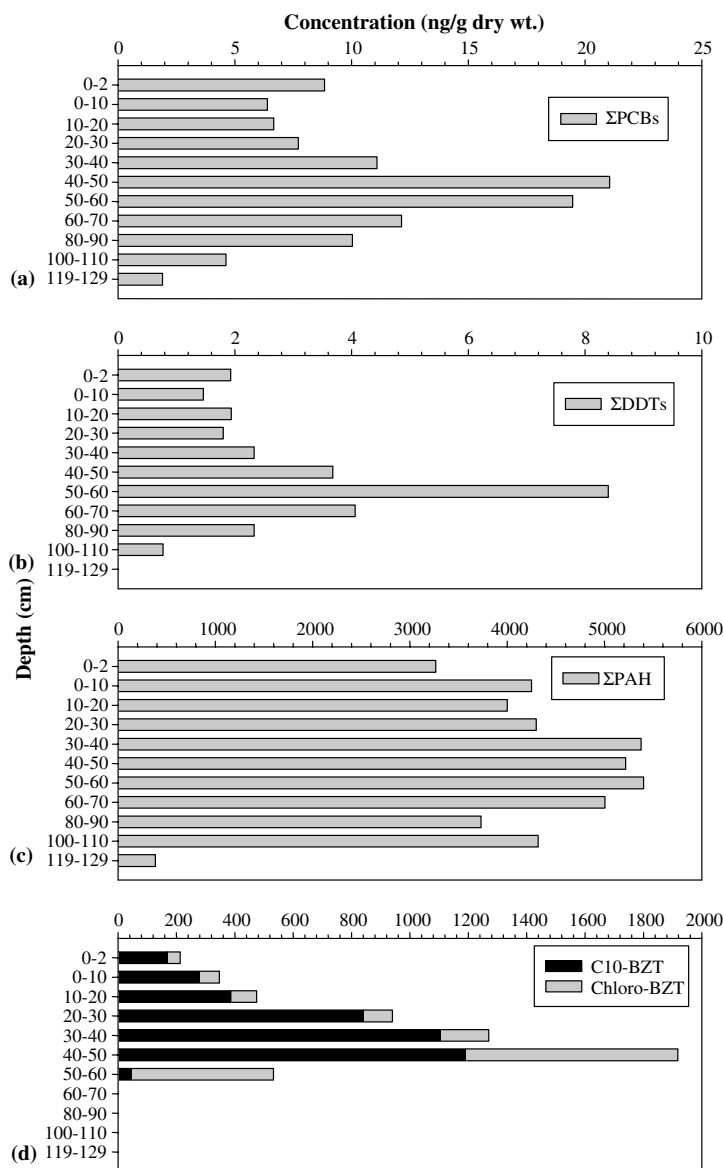


Fig. 2. Station 36, Quonset Point sediment core profile: (a) PCBs, (b) DDTs, (c) PAHs and (d) BZTs.

reflect the level of activity at Quonset Point over time while the difference in the relative change in PAH and TPHs may indicate a change in energy sources.

The BZT profile increases down core through the 40–50 cm section before decreasing in the 50–60 cm section (Fig. 2). Below the 50–60 cm section, the BZTs were below the detection limit ( $\sim 10$  ng/g). In the 50–60 cm section, the Chloro-BZT is much more prominent than the C10-BZT. Moving up core, the C10-BZT progressively accounts for more of the  $\Sigma$  BZTs. This reflects the earlier introduction (1963) and subsequent earlier discontinuation (1972) of the Chloro-BZTs relative to the C10-BZTs (1970 and 1985, respectively). Trends for the LABs have been reported by Hartmann et al. (2000). Like the BZTs, the LABs are not detected below the 50–60 cm section. Organic carbon ranged from 3.1%

at the surface down to 2.5% in the lower sections. The trends in the rest of the analytes did not change when normalized to organic carbon. The sharp breaks in several organic markers with known dates of introduction were used to estimate a sedimentation rate a linear sedimentation rate of about  $\sim 2.1$  cm/y (Fig. 3).

### 3.2. Apponaug Cove

At station 27, both the  $\Sigma$  PCBs and  $\Sigma$  DDTs had a maximum concentration at 10–12 cm and then dropped off sharply at 20–22 cm for the  $\Sigma$  PCBs along with the disappearance of the  $\Sigma$  DDTs (and  $\Sigma$  chlordanes) below 10–12 cm (Fig. 4). Based on a published sedimentation rate for Apponaug Cove of 0.51–0.85 cm/y (Latimer and Quinn, 1996), DDTs (1940) and Chlordanes

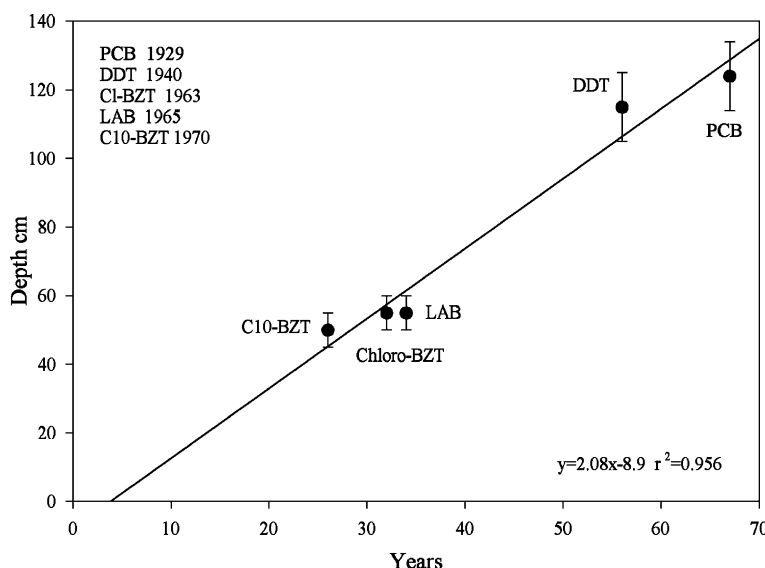


Fig. 3. Estimate of the sedimentation rate at Quonset Point based on first date of introduction of organic markers. The depth is based on the deepest section in which the markers appear. Polycyclic aromatic hydrocarbons and total petroleum hydrocarbons were seen throughout the core and therefore not included.

(1947) should be seen down to at least ~30 cm and the PCB to ~35 cm. It appears that there may have been a disturbance that removed some layers of sediment after the introduction of DDTs (1940) and Chlordanes (1947), and that  $\sum$ PCBs (1929) were mixed down deeper in the sediments when they were first introduced to Apponaug Cove. The highest concentration of  $\sum$ DDTs being in the 10–12 cm section and subsequent decrease in more recent sediment is consistent with its ban in 1972, after which time a decrease in concentration would be expected.

PAHs generally decreased down core with the exception of the 20–22 cm and the bottom 40–42 cm section (Fig. 4). Organic carbon ranged from 4.9% at the surface to 2.5% in the 30–32 cm section with a slight increase in the bottom of the core. Again, there was not change in the trends when normalized to organic carbon.

The BZT profile indicates a much higher surface concentration than the lower sections of the core. The production of C10-BZTs was discontinued 12 years before the core section was taken (1985–1997) and the Chloro-BZT 25 years before that date (1972–1997). The higher concentration in the surface likely represents resuspended sediment transport and deposition of materials in Apponaug Cove with relatively high concentrations of BZTs, possibly coming from Greenwich Bay (Hartmann, 2001). The Ratio of the Chloro-BZT to C10-BZT also increases in the surface section and may indicate a disturbance of older sediments having higher Chloro-BZT levels. As the BZTs are buried, they may also be substantially degraded in the oxic surface layer of the sediments, which is reflected in the lower concen-

tration in the deeper sections. Based on published sedimentation rates for this site of 0.51–0.85 cm/y (Latimer and Quinn, 1996), BZT would have been in production at the time these two deeper sections (6–8 and 10–12 cm) were deposited, possibly resulting in the higher values. Below the 10–12 cm section, BZTs are below the method detection limit (~10 ng/g). Based on a sedimentation rate of 0.85 cm/y, Chloro-BZTs should be seen down to 29 cm. The ratio of Chloro/C10 is higher in the 10–12 cm section as would be expected for earlier inputs of BZTs. Since they are absent in the 20–22 cm section and deeper, the 0.51 cm/y rate seems more reasonable. This would put the lower limit at about 17 cm assuming no bioturbation. The relatively high concentration of PAHs (input starting ~1880) in the 40–42 cm section would argue for a lower sedimentation rate (~0.35 cm/y), a disturbance of sediments below 20–22 cm as stated above, or a more recent increase in the sedimentation rate. Since the C10-BZTs are seen through 12 cm, this would set a minimum sedimentation rate of about 0.5 cm/y. Data from a core taken in 1986 has a profile more consistent with the appearance of the different analytes (Quinn et al., 1992). The LABs were below the detection limit in this core.

### 3.3. Seekonk River

Station 42, in the Seekonk River, had a  $\sum$ PCB maximum concentration at 2–4 cm and decreased sharply between the 12–14 cm and 20–22 cm sections to low levels throughout the rest of the core (Fig. 5). A similar pattern was seen for the  $\sum$ DDTs, which had high concentrations in both the 2–4 and 6–8 cm section



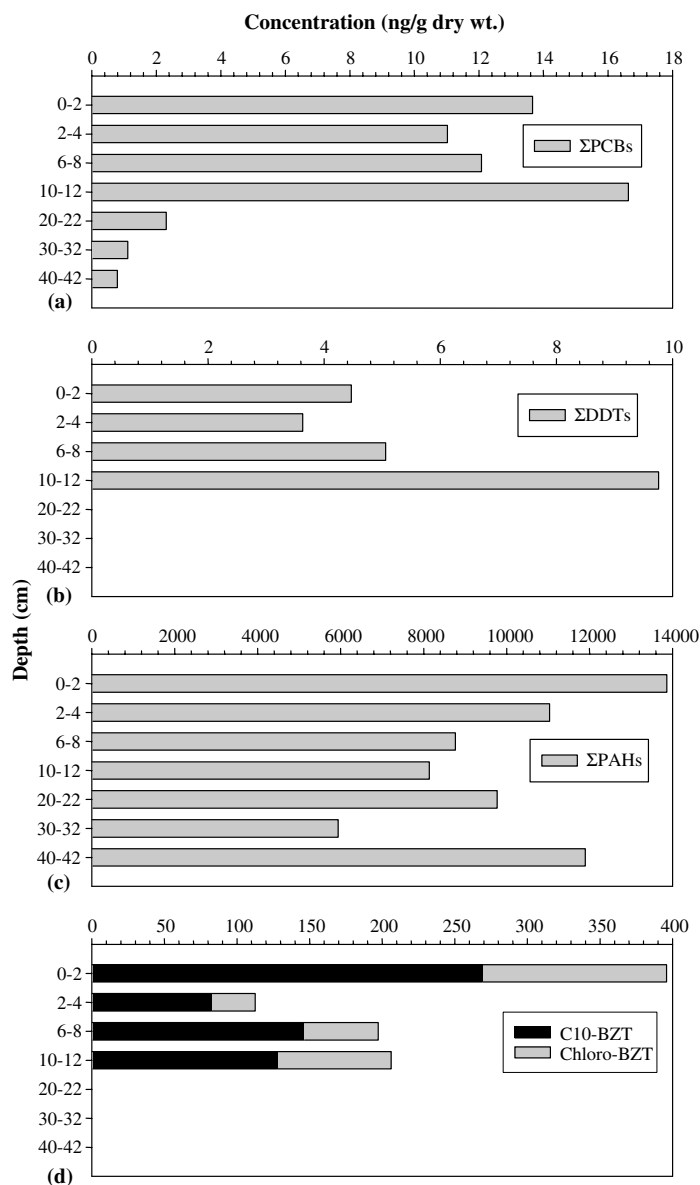


Fig. 4. Station 27, Apponaug Cove sediment core profile: (a) PCBs, (b) DDTs, (c) PAHs and (d) BZTs.

followed by a sharp decrease to the 20–22 cm section. The  $\Sigma$  chlordanes (not shown) have a maximum in the 6–8 cm section, decrease by about half in the 10–12 cm section and then were not detected in any of the lower sections. The sharp decrease below 12–14 cm, along with other data for PAHs, BZTs, and LABs (Hartmann et al., 2000; Hartmann, 2001) compared to another core taken in the same area in 1986 that shows a more orderly decrease down to 70–80 cm (Quinn et al., 1992), would suggest that some sedimentary layers were removed. Additional evidence of a disturbance is found in the BZT ratio. The lowest core section with BZTs (12–14 cm) should have a high ratio of Chloro-BZT to C10-BZT due to their production history as discussed earlier. The 12–14 cm section in this case actually has

a lower ratio of the Chloro-BZT to C10-BZT than the sections above it. The low background concentration of PAH and TPHs is indicative of a time before ~1880 (Latimer and Quinn, 1996) when industrial activity led to an increase in petroleum contamination. Organic carbon is elevated in the surface and 2–4 cm sections relative to the deeper sections of the core below 20–22 cm. OC was not measured in the 6–8 and 12–14 cm sections. The concentrations of organic carbon below 12–14 cm are the lowest seen in this study and may be additional evidence that these sediments represent older sediments that have less anthropogenic influence. The relatively low but constant levels of  $\Sigma$  PCBs in the lower sections also suggest that the sediments were mixed during the event that removed the overlying

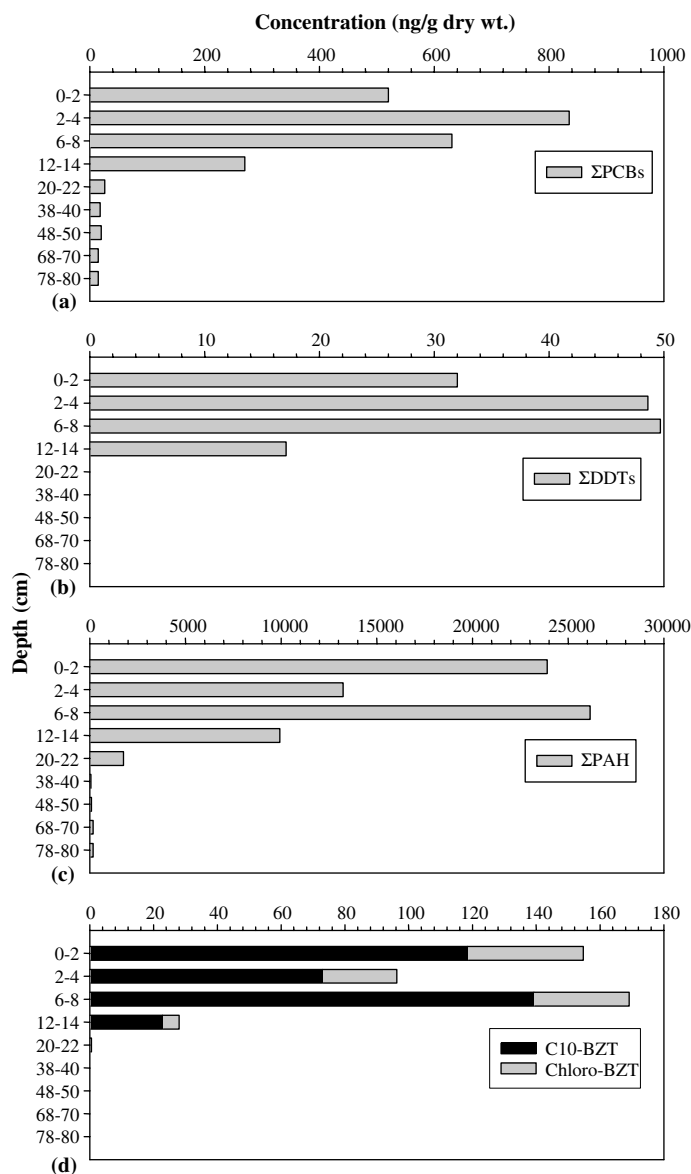


Fig. 5. Station 42, Seekonk River sediment core profile: (a) PCBs, (b) DDTs, (c) PAHs and (d) BZTs.

layers. Assuming the sediment layers above 20–22 cm are more recent than 1986, the date of the last core collection, the sedimentation rate at this site is at greater than 1 cm/y which is agreement with a previously estimated rate for this site of 1.2 cm/y (Corbin, 1989). Since PCBs and DDTs were long since out of production by the time these sedimentary layers were deposited, we would expect that the 12–14 cm section would have the highest concentrations with decreasing values toward the surface, however this is clearly not the case. A possible explanation would be the erosion of more highly contaminated sediments upstream (e.g. station 25) near the Narragansett Bay Commission waste water treatment plant that have recently been deposited at this site (Hartmann, 2001).

### Acknowledgments

The authors thank John Peck, Sheldon Pratt, and Elizabeth Lacy who collected the samples. The authors also thank Emily Chen, Abby Aukerman (deceased), Mike Kittredge, Matt Dimatteo, Christopher Reddy and Sean Sylva for assistance with sample analysis. Thanks to Steve Wise and Michelle Shantz at NIST for providing several standard reference materials. This study was funded by the URI/NOAA Cooperative Marine Education Research (CMER) Program grant #NA77FE0493. The 5890/5971 GC/MSD was donated to our laboratory through the Hewlett-Packard University Grant Program and the 6890/5973 GC/MS was purchased with funds from the RI Department of

Environmental Management. Graduate fellowship support was provided for P.C. Hartmann through a University of Rhode Island assistantship and a Narragansett Electric Coastal Fellowship.

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