Historical sediment record and levels of PCBs in sediments and mangroves of Jobos Bay, Puerto Rico

by

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DEDICATION

To my family for supporting me while I achieve my goals.

To my husband, Hunter, for always believing in me.

Finally, to my daughter Ridley, you provided me the inspiration that I needed to finish this journey.

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There are so many people that helped me along my way. This has been one of the hardest things that I have done. Words are not enough to express my sincere gratitude to everyone who I have encountered during this process.

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ABSTRACT

Polychlorinated biphenyls (PCBs) were quantified in 18 surface sediment samples, 1 sediment core, and several mangrove tissue samples collected in Jobos Bay, Puerto Rico in September 2013. Total PCBs in surface sediments ranged from 0.42 to 1232 ng g^{-1} dw. Generally, higher levels were observed near the shore, close to urban and industrial areas. The levels suggest significant pollution in Jobos Bay with respect to PCBs. Two-thirds of the sites were dominated by lighter PCB congeners (tri- to penta-chlorinated PCBs) while one-third had heavy PCB congeners (hexa- to octa-chlorinated PCBs) dominant. Total PCBs in a sediment core indicated levels fluctuating according to historical usage patterns. Total PCBs were measured in mangal leaves (14 – 747 ng g^{-1} dw), roots (0.26 – 120 ng g^{-1} dw), and seeds (16 – 93 ng g^{-1} dw), suggesting bioaccumulation from sediments. This is the first report of a historical profile of PCBs in the study area and of PCB bioaccumulation in mangroves. This article provides new and useful information on PCBs in the Caribbean area.

INTRODUCTION

Persistent organic pollutants (POPs) are a group of toxic, halogenated organic chemicals. Their capability to resist photolytic, biological, and chemical degradation allows for them to persist in the environment, with half-lives often measured in years or even decades. POPs can be found all over the world, even reaching remote locations where use has never occurred, including Arctic regions. Molecular weights of POPs range from 200 to 500 Da and they have vapor pressures lower than 1000 Pa (Ross, 2004). POPs are also lipophilic and therefore accumulate in the adipose tissue of animals (Jacob and Cherian, 2013).

POPs bioaccumulate throughout food chains as the trophic levels increase, leading to a high bioconcentration factor in quaternary consumers. They are found in high concentrations in sediments due to their high affinity towards organic matter (absorption) and clay-size sediments (adsorption) (Kruitwagen et al., 2008; Oyo-Ita et al., 2013). Clay particles have a relatively large surface area and thereby a high cation exchange capacity for contaminants (De Paiva et al., 2008). Their (re)mobilization and subsequent fate and bioavailability is dependent upon water solubility, persistence, temperature, salinity, and redox conditions. For example, organochlorines are less soluble than organophosphates but they are more persistent in the environment (Edwards, 1977). The sorption potential and solubility of POPs and other contaminants (*e.g.*, heavy metals) can either increase or decrease with different salinity gradients (Bayen, 2012; Martinez-Colon et al., 2009).

When POPs were first introduced, their persistence in the environment was not known and they were used extensively in a variety of applications. The Monsanto Company first produced POPs in 1929 with the production of PCBs in St. Louis, Missouri (De Boer, 2012). In 1939 DDT was produced as an insecticide to treat malaria (De Boer, 2012). POPs became widely distributed during World War II due to their beneficial use in pest and disease control, industrial uses, and crop production ("Persistent organic," 2012). It was not until the environmental movement in the 1960s that POPs were found to be widely present in the environment (De Boer, 2012).

The presence of POPs throughout the world is due to atmosphere transport, a process known as global distillation. This accounts for the occurrence of POPs in remote regions where their use never occurred. The global distillation of POPs operates under the same concept of distillation that occurs in an industrial setting, just on a global scale. The distillation process begins when POPs are deposited into the earth system under warm climatic conditions. Due to the tendency of many POPs to adsorb to particles and their relatively low vapor pressures, they slowly get released into the air. After POPs volatize in the air they migrate to colder areas through wind currents. They remain in the atmosphere until atmospheric temperatures drop to a temperature low enough for the POP to condense on atmospheric particles and return to the earth. POPs are then re-volatized into the atmosphere when the temperature heats up again. This process continues until they reach the poles (Bard, 1999). The overall effect of global distillation is a result of evaporation in warmer regions and deposition by condensation in colder regions. Even when POPs are eliminated they still continue to be deposited into cold environments because of global distillation combined with their persistence (Holmqvist et al., 2006).

The rate of global distillation of different POPs depends upon several factors. POPs with low volatilities migrate at a slower rate than those with higher volatilities. Their lipophilicity also plays a role in environmental transport of POPs. POPs with low lipophilicity move more readily into the atmosphere, thereby moving faster towards the poles than those with higher lipophilicity (Bard, 1999). Temperature also affects the transport of POPs. As the temperature increases so will vapor pressures, thereby facilitating movement. Therefore, when POPs get disbursed to warm, temperate climates they dissipate at a faster rate and begin the global distribution process. (MacKay & Wania, 1995).

The toxicity of POPs makes it important to understand and determine the fate and transport and cycling of these chemicals. POPs can undergo biomagnification along the food chain because of their high lipid solubility (“Persistent organic”, 2012). The result is increasing concentrations of POPs with each trophic level, leading to a potential hazard for species at the top of the food chain. In humans, POPs have been known to cause certain types of cancers, a decrease in intelligence, and damages to both the central and peripheral nervous systems (WHO, 2010). POPs have been associated with a depression in an organism’s immune systems, causing allergies and hypersensitivity (Hansen et al., 2015). A study conducted by Hansen et al. (2015) demonstrated an increase in rates of chronic obstructive lung disease in children of mothers who had measurable concentrations of POPs in their systems. POPs have also been found to be endocrine disruptors, altering the hormonal system in both humans and wildlife. This can cause damage to the reproductive system and birth defects in offspring (Elise et al., 2013). Therefore, it is important to continuously monitor POPs concentration levels and to implement reduction policies.

Polychlorinated biphenyls

Polychlorinated biphenyls (PCBs) are considered a “legacy” POP. They are a class of organochlorinated compounds made up of isomer mixtures with 1 to 10 chlorine atoms attached to a biphenyl (Souza, et al, 2008). The different configurations that the PCB molecule can form are called congeners.

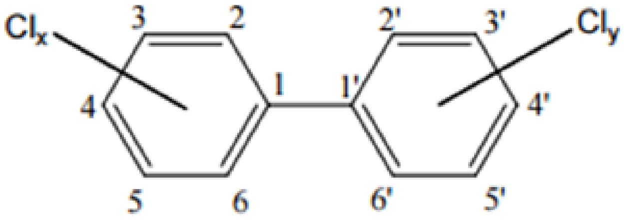


Figure 1. The general structure of PCBs (Faroon & Olson, 2000).

There are 209 different PCB congeners. PCB congeners are identified by an assigned number ranging from 1 to 209, with monochlorobiphenyls being PCB-1 and the one decachlorobiphenyl identified as PCB-209. The PCB’s chlorination degree determines the substance’s toxicity and biological activity (Faroon & Olson, 2000). Co-planar PCBs, PCB congeners with four hydrogens located next to the pheynl bond, are more toxic than other PCB congeners because they can adhere more readily to certain biological enzymes (Hites and Raff, 2012). The different PCB congeners exhibit slightly different physiochemical properties but, generally, they have high n-octanol/water partition coefficients, low water solubility, low vapor pressure, and a high fat solubility (Geyer et al., 1984).

PCBs were first produced in 1929 and were used commercially until the late 1980s (Bayona & Albaigés, 2006). Unlike some POPs, PCBs were never intended to be released into the environment. They were used extensively in industrial application because of their chemical stability and nonflammable properties. PCBs are also resistant to acids, bases and microbial degradation (Geyer et al., 1984). Some insurance companies and city codes even required that certain equipment be made using PCB materials. Commercial PCB items include plastics, transformers, hydraulic and heat-exchange items, and carbonless duplicating paper (Bayona & Albaigés, 2006; Pepper et al, 2006). PCBs were marketed and used extensively worldwide under trade names like Aroclor, Sovol, and Therminol (Faroon & Olson, 2000; Tehrani & Van Aken, 2014). Peak production of PCBs in the U.S. occurred in 1970 by the Monsanto Company, with a total production of 85 million pounds (Faroon & Olson, 2000). Worldwide, approximately 1.5 million tons of PCBs were produced, leading to a significant amount being released into the environment (Tehrani & Van Aken, 2014). PCBs can be released through the combustion of coal in power plants, burning of plastics and buildings, and migration from landfills (Wolska et.al., 2014). This widespread use has made it difficult to determine a point source of PCB emission.

During the 1970s the U.S. began to restrict PCB production as concern grew over their presence in the environment. In 1976 the Toxic Substances Control Act (TSCA) regulated PCB production. By 1979 the US EPA had banned PCB production and placed strict regulations on their continued use (Ross, 2004). In 2004 the Stockholm Convention on Persistent Organic Pollutants listed them as one of the "dirty dozen" POPs to be slated for elimination (Combi et al., 2016). Since their ban, the concentrations of PCBs in various media have declined. The greatest decline was noticed in the 1980s as many of the regulations and bans took effect. Even though PCB levels are relatively low, some studies have shown that concentrations have changed very little over the past 25 years. This low but stable concentration is indicative of the complete PCB removal from the environment being a slow process (Hites & Raff, 2012). One of the factors contributing to the recent concentration of PCBs in the environment is leaching from landfills, urban runoff, and volatilization into the atmosphere (Combi et al, 2016). PCB-containing equipment is still being used in closed systems around the world (Ross, 2004). Therefore, there is still considerable interest in studying the fate of PCBs in the environment, especially in areas such as Latin America and the Caribbean where they were extensively used and where there is a dearth of studies on their environmental fate and transport.

While the chemical stability of PCBs made them great for industrial use, this characteristic made them highly persistent in the environment. The more chlorinated the PCB, the more likely it is to bioaccumulate and persist in the environment (Geyer et.al., 1984). Sediments, specifically fine grained sediments, act as a sink for organic contaminants and are the main source of PCB exposure (Ross, 2004). The high lipophilicity and the ability to resist degradation through abiotic or biotic processes allow PCBs to readily bind to sediment (Geyer et.al., 1984). PCBs accumulate mainly on clay sized particles that have a high surface to volume ratio and that are rich in organic matter (Tam & Yao, 2002; Zhao et al., 2012). A study quantifying the concentrations of PCBs in mangrove sediments of Hong Kong demonstrated that PCB concentration in sediment increases as the total organic carbon (TOC) content increases (Tam & Yao, 2002). However, other factors can affect PCB uptake. In a study conducted by Zhao et al. (2012), there was a weak correlation between PCB concentration and organic carbon within a mangrove ecosystem. This is attributed to the other intrinsic factors that affect absorption, like grain size and accumulation patterns.

Estuaries

Estuarine areas are important ecosystems globally due to their high productivities (Aldarondo-Torres et al., 2010). They receive pollutants (including POPs such as PCBs) from both point and nonpoint sources. The ultimate fate of such pollutants depends strongly on their physico-chemical properties (Mackay et al., 1992). POPs have high organic carbon partition coefficients (Koc), which causes them to adsorb to sediments in estuarine areas (Pozo et al., 2014). Therefore, sediments serve as a good matrix to determine the extent of PCB pollution of estuarine areas. Some reports have also suggested that POPs may be bioaccumulated in mangroves in estuarine areas (Shete et al., 2009; Hellstrom, 2004; Patterson et al., 1990; Walsh et al., 1974; Walsh, 1973).

Mangroves are the dominant plant species in estuaries. They provide many economic and ecological benefits. The global economic value of mangrove ecosystems is estimated at 181 billion dollars. (Lewis, et al., 2011). They generate income and jobs through tourism opportunities and fisheries (Bayen, 2012). Mangroves act as a nursery ground for a variety of juvenile organisms, providing food sources and protection against predation. Mangrove root systems prevent shoreline erosion by soil stabilization and decreasing wave energy. They reduce water movement and retain suspended materials, thereby generating organic-rich soil (Lewis et al., 2011). Mangroves act as a natural filtration system preventing contaminants in runoff from reaching marine environments. They are able to take up pesticides, excess nutrients, and other pollutants in surrounding soil and plant tissue, thereby maintaining marine water clarity and quality (Shete et al., 2009). Mangroves have unique sediments that are rich with organic carbon and detritus (Bayen, 2012). The fine-grain sediment and organic rich matter in this ecosystem provides a large surface area for PCBs to adhere to (Kruitwagen et al., 2008).

Mangroves are a threatened tropical ecosystem, with many different species at risk of extinction (Bayen, 2012). Mangroves are susceptible to stress, which has resulted in a reduction in size and diversity. Since the 1980s the world's mangrove ecosystems have decreased by 35 percent at a rate of 2.1 percent per year (Martinuzzi, 2009). It is well known that mangrove forest coverage has changed as a response to natural (e.g., sea level) and anthropogenic (e.g., deforestation) factors (Tschirley, 1969). Limited studies have linked the effect of POPs mainly to sediment-water quality in the Caribbean (Rawlins et al., 1998; Potter et al., 2013) and other locations (e.g., Asia, Australia, etc.)

(Bayen, 2012). One study investigated the transport and fate of atrazine within an estuary system in Jobos Bay, Puerto Rico. It determined that surface runoff was the main pathway for atrazine transport into the bay, thereby leaching into groundwater and negatively affecting the surrounding water quality. Mangrove forests may be sensitive to POPs due to high surface-to-volume ratio of the rhizosphere (root-sediment-porewater) and the chemical variability in this zone (e.g., pH, redox potential, salinity, etc.). It is known that mangrove forest distribution is directly related to porewater and/or soil salinity gradients and that mangroves do not grow at salinities greater than 90 ppt (Lugo et al., 1974; Orge et al., 2000; Cintron et al., 1978). Mangroves recover slowly from damage taking around 20 to 50 years to fully recover (Lewis et al., 2011).

Plant tissues serve as “reservoirs” of chemicals, and mangrove tissues are no exception (Patterson et al., 1990; Hellstrom, 2004). POPs are not readily biodegradable, so when they are deposited into an ecosystem they are capable of transferring from the sediment and into plant tissues. Due to the hydrodynamic conditions in which they thrive, mangroves are also capable of concentrating POPs within their tissues (Shete et al., 2009). Agricultural and industrial chemicals have been found in seedlings (Lewis et al., 2011; Walsh et al., 1974; Youssef, 2002; Bell & Duke, 2005), leafs and/or roots (Bayen 2012; Shete et al., 2009; Walsh et al., 1974), and stems (Walsh et al., 1973) of several mangrove trees (*Avicenia marina*, *Rhizosphora mangle*, *Rhizophora sylosa, Ceriops australis*, and *Kandelia candel*). A report by Shete et al. (2009) documented the presence of organochlorine (OC) pesticides in the roots and leaves of *A. marina* (white mangrove) in India as well as the sediments associated with this mangrove species. In India, DDT and HCH were shown to bioaccumulate within the *A. marina* mangrove system even after regulations limited use. DDTs (p,p'-DDT, p,p'-DDE, p,p'-DDD) were found in the mangrove samples with p,p'-DDE concentrations being the highest. Within the mangrove ecosystem, p,p'-DDT was found in the highest concentration in the root with a concentration of 77.713 ng/g. The high concentrations of p,p'-DDE even after its restricted use is indicative of the degradation of p,p'-DDT in the mangrove sediments once it has been deposited, as well as before it reaches this ecosystem. Within the mangrove plant system, the roots were found to have the highest concentrations of OC pesticides. The sediment associated with *A. marina* exhibited the lowest concentration of OC pesticides. This is attributed to the low water solubility of organic carbon (OC) pesticides and their lipophilic nature (Shete et al., 2009).

There are limited studies that look at the accumulation of the PCBs in mangrove tissues. One study conducted by Walsh et al. (1974) reported a concentration of 0.18 ng/g dry wt. of the Aroclor 1242 within the Rhizophora mangrove tissues from samples located within the Virgin Islands. This study demonstrated that PCBs are capable of bioaccumulating in the mangrove ecosystem. Another study quantified the concentrations of Aroclor 1242 in the roots, hypocotyl, stems, and leaves of R. mangle from Jupiter, Florida that had been exposed to varying concentrations of the PCB over a period of six weeks in a lab setting (Forester et al., 1974) . This study measured the uptake of the PCBs in the hypocotyls and leaves at a rate of greater than 0.56 kg/ha, showing that, if present in the environment, they can pass throughout the trophic levels via species that eat mangrove seedlings (Forester et al., 1974).

The bioaccumulation of PCBs and their presence in mangroves could also pose long term negative effects on the mangrove plant (Nowrouzi, 2012). There are limited published reports on the effects of bioaccumulation on plant physiology. However, a few studies have demonstrated that trace pollutants have been found to cause a reduction of photosynthesis, growth, and induce mortality in the mangrove plant (Bayen, 2012). Chemical pollution can even lead to a change in the diversity and structure of a mangrove ecosystem, subsequently affecting biota that associate with it. Mangroves exposed to trace pollutants have shown to have a change in activity, enzyme activity inhibition, and eventually death. These alterations to the mangrove ecosystem have been known to cause imposex in snails, where female snails develop male sex organs, an alteration to the ratio of RNA to DNA in crabs, and abnormalities in fish and oysters, (Bayen, 2012). Rhee et al. (2009) conducted a study that demonstrated the gene expression of metallothionein (MT), a protein that is important in plant and animal metal detoxification. This study showed that when exposed to metal pollutants (copper, cadmium, and zinc), mangrove killfish induced MT. However, when exposed to bispheol A, nonylphenol, and octylphenol, MT was suppressed, thereby demonstrating the effects of different pollutants on gene expression in the mangrove ecosystem. Knowledge of bioaccumulation and bioconcentration within the mangrove ecosystem can help alert coastal communities of possible impacts on food webs associated with them (Qui et al., 2011).

Puerto Rico

Puerto Rico has undergone many changes to its economy and land use over the last 200 years, including significant industrialization, urbanization and agriculture.

Mangrove coverage in Puerto Rico has evolved over four distinct periods. The time-period from 1800 to 1938 is identified as the agricultural period. During this time-period mangrove coverage decreased from 11,791 ha to 6475 ha, equating to a decrease of 45 percent. Land was converted to agricultural fields and natural drainage patterns were altered for irrigation purposes (Martinuzzi, 2009). Mangroves underwent a natural rebound in the second period. This was a period falling between the agricultural period and the industrial period when mangrove land cover increased by 12% to 7285 ha. The third period of mangrove coverage change occurred between 1959 and 1971 when rapid urbanization led to a decline in the mangrove population. Mangrove coverage decreased to 6745 ha as mangrove forests were converted into housing developments and other urban developments. The fourth (and current) period began in 1971. This period focuses on mangrove recovery through protection and has led to an increase in mangrove coverage to 8323 ha. This increase is credited to the end of the sugar cane industry, increase protection of mangroves, and restoration of wetland ecosystems (Martinuzzi, 2009).

Most studies in Puerto Rico have focused on documenting levels of pollutants, including metals and POPs. One study examined the concentrations of organic and metal pollutants in sediments from Guanica Bay, Puerto Rico (Whitall et al., 2014). It showed that historical PCB use from the surrounding area was persistent in the environment. Levels of total PCB in this area had an average value of 336.068 ng/g, exceeding the National Oceanic and Atmospheric Administration's (NOAA) numerical sediment guidelines for ELR (effects low-range) and ERM (effects range-median), which is 22.7 ng/g and 180 ng/g. This is indicative of sediment toxicity having a negative effect on the surrounding biota (Whitall et al., 2014).

In Jobos Bay, similar studies have focused on documenting such levels of pollutants. Whitall et al. (2011) analyzed PCBs from seven sample sites from Jobos Bay in 2012 and determined a mean PCB value of 2.09 ± 0.50 ng/g, a concentration that is below the ELR value. A separate study conducted in 2010 from different sampling sites within Jobos Bay showed total PCB concentration values ranging from 1.6 ng/g to 11.21 ng/g. They classified the sediments of Jobos Bay as moderately polluted, possibly due to the proximity of different anthropogenic sources (Aldarondo-Torres et al., 2010). While there have been studies that quantify levels of different POPs in sediments, no studies have attempted to examine the temporal distribution of POPs and their uptake by mangroves.

There are no peer-reviewed studies related to mangrove tissue and POPs concentrations from Puerto Rico except for unpublished data reported by Walsh et al. (1974). This study showed p,p’-DDD in roots (0.022 ppm), hypocotyls (0.220 ppm), stems (0.032 ppm), and leaves (0.0019 ppm) on R. mangle seedlings from Joyuda Bay on the west coast of Puerto Rico. This study was carried out to determine the temporal fluctuation in PCBs in Jobos Bay and the bioaccumulation of these chemicals in mangroves.

STUDY AREA

Jobos Bay, a semi-enclosed estuary-bay located in the southeastern coast of Puerto Rico, is part of the National Estuarine Research Reserve. It is within the Central Aguirre watershed (Fig. 2). This coastal plain estuary, with a surface area around 25 km^2 (Morellock & Bunkley, 2006) and maximum water depth of 30 feet, is the second largest on the island of Puerto Rico (Laboy et al., 2002). It has three times the amount of shoreline as any other estuary in Puerto Rico (Laboy et al., 2002). Its topography is comprised mainly of sea grass beds, coral reefs, and mangrove forests (NOAA, 2002). Mangrove forests comprise approximately 25% of the Bay's surface area (Lugo et al., 2007). Sediments in Jobos Bay are dominated by (1) sandy mud (most common); (2) silty clay (deeper areas); and (3) poorly sorted terrigenous and carbonate sediments (reefs).

Jobos Bay is composed of two main areas: Mar Negro and Cayos Caribe. The study area was located within the landside area of the Bay known as Mar Negro. The Mar Negro ecotone comprises 72% of the bay's surface area and consists of critical mangrove habitats with a restricted tidal exchange. Sediments within this area are deposited under relatively stable accumulation rates making it an excellent site to study temporal and spatial changes in PCB pollution (Laboy et al., 2002).

Mangrove species are the dominant plant species within Jobos Bay. There are three physiographic types of mangroves in Jobos Bay: basin, fringe, and overwash mangroves, which are of the *Rhizophora mangle* (red mangrove), *Avicennia germinans* (black mangrove), and *Laguncularia racemosa* (white mangrove) species. Tidal changes and flood frequency create depth gradients that determine the zonation of the different mangrove species. Jobos Bay mangrove forests make up 42.6% of the mangrove habitat on the southern coast of Puerto Rico. This extensive coverage makes it a vital ecological habitat to the area. (Laboy et al., 2002). Lugo et al. (2007) determined that under ideal conditions Jobos Bay mangroves are capable of high production rates and fast circulation of nutrients. Mangroves in Puerto Rico produce leaf litter year-round, continually adding organic matter to the system (Mejias et al., 2013).

Jobos Bay is an ecologically significant estuary in Puerto Rico providing a habitat for endangered species. This field site has been impacted by agricultural practices adjacent to its northern boundary and receives high inputs of nutrients and pollutants (Martínez-Colón et al., 2009; Whitall et al, 2011). Nearby urbanization and industrial activities also have an impact on the health of the bay. Located within Jobos Bay estuary is a landfill, an old sugar mill, a thermoelectric plant, agricultural lands, urban centers (the largest being Guayama and Salinas), metal manufacturing, pharmaceutical plants, a superfund site, petrochemical plants, and a chlorinated underwater pipe. Chevron Phillips, the Aguirre power plant, and the Ball Metal

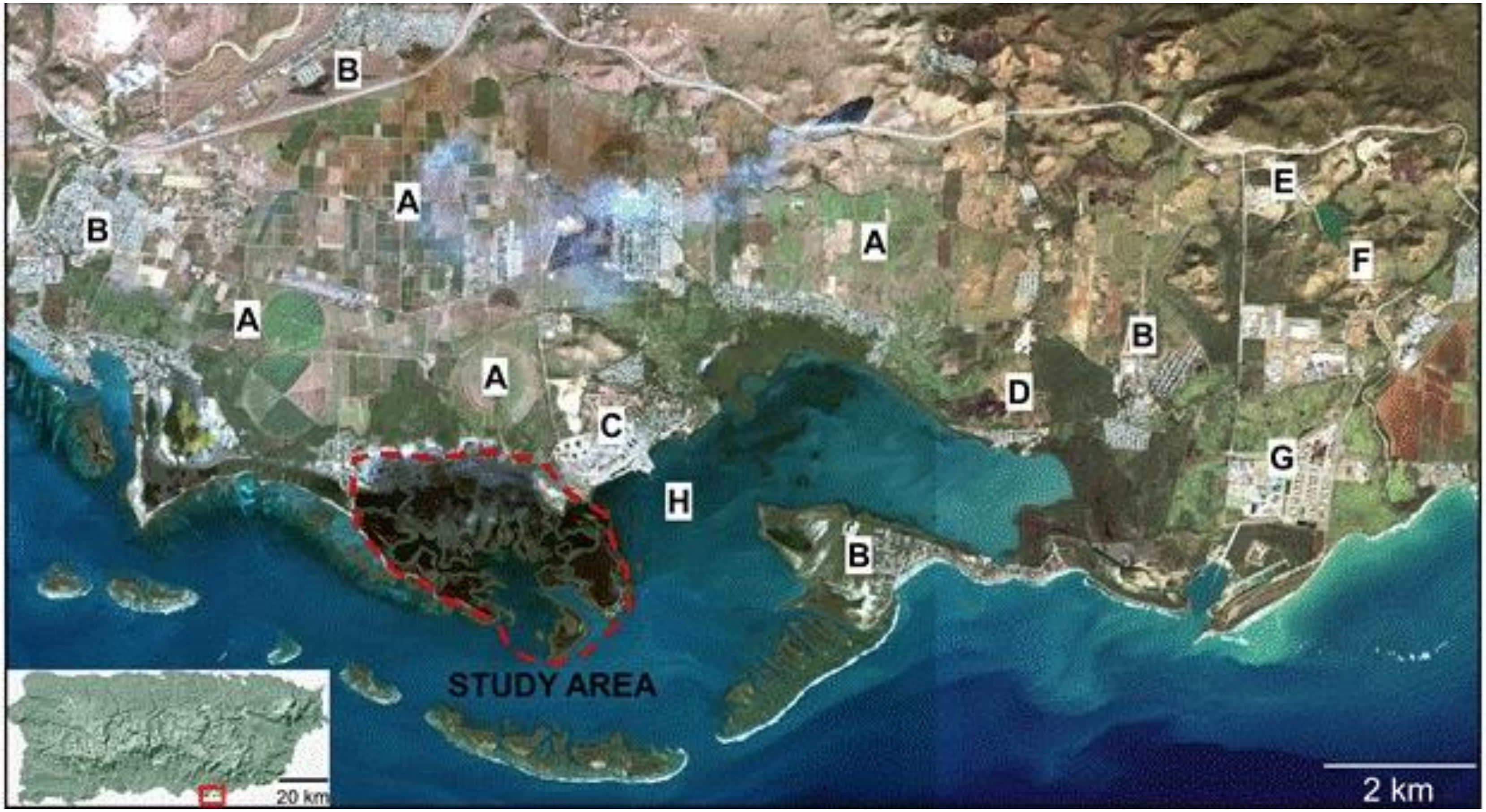


Figure 2. Study Area. (Legend: A- agriculture, B- urban, C- thermoelectric plant, D- metal manufacturing, E- pharmaceutical, F- superfund site, G- petrochemical, H- chlorinated underwater pipe.)

Beverage Container are point sources that discharge into Jobos Bay (Whitall et al, 2011). Historically, the land around Jobos Bay was mainly used for agricultural purposes from Spanish Colonial times until the decline of sugar production in the 1960s. Currently, any remaining sugarcane lands have been converted into fruit and vegetable cultivation or removed completely. Only 11% of Jobos Bay's total land cover is used for agricultural practices. The remaining land has been allowed to revert back to its natural state, making up over 70% of vegetated lands in Jobos Bay (Laboy et al., 2002). However, even though visually Jobos Bay may appear to be healthy and pristine, the sugarcane cultivation of the past has had severe and lasting effects on this ecosystem (Whitall et al., 2011). In addition, urbanization and the many other industrial activities in the surrounding area have added to the adverse impacts.

Several studies have addressed the vitality and health of mangrove forests within Jobos Bay. These efforts concentrated on conducting surveys and spatial distribution of the mangrove forests (Martinuzzi et al., 2009; Cintron et al., 1978); identifying sources of stress (Fuentes & Sastre, 1995; Lugo et al., 1974; Kuniansky et al., 2010); macrofaunal ecology (Demopoulos, 2004); photosynthesis (Thomas, 2009); mangrove succession (Cintron et al., 1978); and ecophysiology (Lugo et al., 2007). A recent study documented the historical decline and recovery of mangrove forests in Puerto Rico (including Jobos Bay). This study found a 32% net island-wide decline between the years 1800-2002 associated with sugar cane agriculture and mangrove deforestation (Dominguez-Cristobal, 2008; Martinuzzi et al., 2009). At Jobos Bay, *L. racemosa* and

A. germinans were least affected by changes in hydrological conditions but more susceptible to habitat encroachment due to urbanization than *R. mangle* (Martinuzzi et al., 2009). However, Kuniansky and Rodriguez (2010) concluded that the *A. germinans* mass mortality of 75 acres since 1990 resulted from changes in irrigation and agricultural practices leading to a 25 foot drawdown of the water table and the alteration of the salinities of the basin mangroves (Kuniansky & Rodriguez., 2010).

Environmental studies concerning pollutants conducted at Jobos Bay to date have focused mainly on the quantification of anthropogenic input of agrochemicals and other pollutants (Norat et al., 2005; Apeti et al., 2011). A handful of studies have addressed the presence of POPs in organisms such as mussels (Cantillo et al., 1999; Kimbrough, 2008) and amphibians (Barrios-Santiago, 2000). Sediment and water samples have been found to be contaminated with several types of POPs, including OC pesticides, PCBs, polycyclic aromatic hydrocarbons (PAHs), and atrazine (Potter et al., 2013; Aldarondo-Torres et al., 2010; Fuentes & Sastre, 1995; Whitall et al., 2011). No studies have tried to look at the temporal distribution of POPs and their uptake by mangroves.

GOALS AND OBJECTIVES

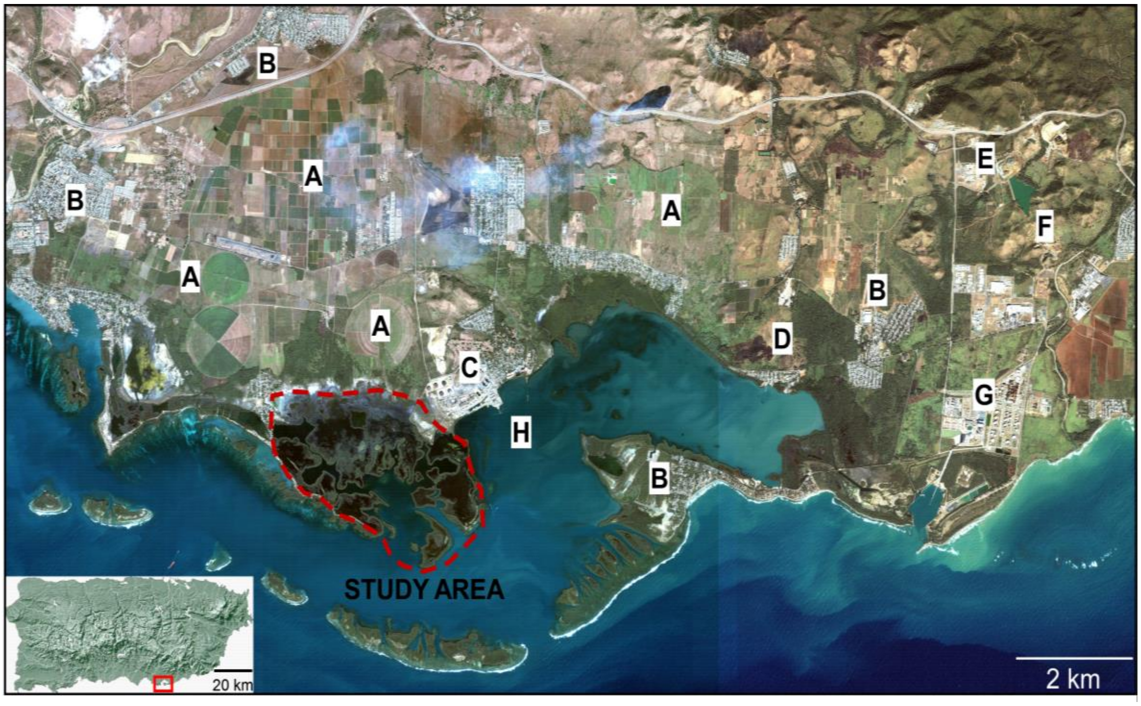
The aim of this work is to assess historical patterns and potential sources of PCBs in sediments in Jobos Bay, and the potential for their bioaccumulation in mangroves. This will help to better understand the fate and transport of these chemicals in the study. The overall goal will be achieved by: (i) measuring levels of PCBs in a core taken from the study area; (ii) measuring levels of PCBs in mangrove tissue from the study area; and (iii) measuring levels of PCBs in surface sediments from the study area.

MATERIALS AND METHODS

Sampling Collection

Marine surface sediment samples were collected at 18 sites in Jobos Bay (Figure 3B). Surface sediment samples were collected using a ponar grab sampler. Wherever possible, the root and tissue samples from *R. mangle*, *L. racemosa*, and *A. germinans* mangal were collected at the same sampling sites where the sediment samples were taken following the protocols of Shete et al. (2009) (Fig. 3B). A push core was used to collect a 1 m sediment core for temporal POPs analysis at site 2 (Fig. 3B). Sediment samples were split, with one portion placed in plastic jars for metals analysis (not reported here) and one portion in pre-cleaned glass jars for POPs analysis. Mangrove root and tissue samples for POPs analysis were wrapped in pre-cleaned aluminum foil and placed in Ziploc bags. All jars were placed in an ice cooler and then stored in a freezer until extraction. The sediment core was placed on a table and for dating purposes sections were sampled at 0.5–1 cm for the first 5 cm and at 2 cm intervals thereafter. Aliquots of each slice were placed in pre-cleaned glass jars for POPs analysis and into propylene jars for dating, then stored in a freezer until analysis.

3A.



3B.

Figure 3 A. Study Area. (Legend: A- agriculture, B- urban, C- thermoelectric plant, D- metal manufacturing, E- pharmaceutical, F- superfund site, G- petrochemical, H- chlorinated underwater pipe.) B. Sample collection sites. The orange circle signifies where the core was taken.

Chemical Analysis

Sediment and tissue samples were freeze-dried. Ten grams of sediment subsamples were mixed with 10 g of anhydrous sodium sulfate (to ensure complete dryness), placed in a glass extraction thimble and spiked with 100 μL of a surrogate solution consisting of [$^{13}C_{12}$]-labeled PCBs (IUPAC 9, 15, 28, 32, 52, 101,138, 153, 180, and 209). The samples were then Soxhlet-extracted for 16 hours using 250 mL of dichloromethane. To remove sulfur that was present in the sediment samples, active copper granules were added to the round bottom flask during the Soxhlet-extraction process. The extracts were filtered to remove the copper, then concentrated and solvent-exchanged into isooctane via rotary evaporation followed by a gentle stream of nitrogen.

Twenty grams (20 g) of freeze-dried mangal tissue samples were chopped using a stainless-steel vegetable chopper into the smallest pieces possible, pulverized in a mortar and pestle, and weighed for extraction. The tissue samples were placed in a 250 mL beaker and spiked with 100 μL of a surrogate solution (described previously). After an equilibration period of 30 min, 200 mL of a 1:1 mixture of dichloromethane:hexane was added, and the beaker was then covered with aluminum foil and left undisturbed for 24 hours. The sample was then ultrasonicated for 60 minutes. Once ultrasonication was complete the extract was decanted and the tissue subjected to a second round of ultrasonication. The extracts were combined, dried over anhydrous sodium sulfate and concentrated and solvent-exchanged into isooctane via rotary evaporation followed by a gentle stream of nitrogen.

All extracts were cleaned by column chromatography using Florisil. A 1-in diameter glass column was prepared by adding 8 g of Florisil (2% deactivated with HPLC-grade water) topped with 1 cm anhydrous sodium sulfate. The column was pre-eluted with 50 mL of dichloromethane followed by 50 mL of hexane. The sample was added onto the top of the column and eluted with 50 mL of 1:1 dichloromethane:hexane. The extract was concentrated, solvent-exchanged into isooctane at a final volume of 1 mL, and $^{13}C_{12}$-PCB105 was added as internal standard.

All samples were analyzed for 50 PCB congeners by gas chromatography - mass spectrometry (GC-MS) in the electron impact (EI) mode using selected ion monitoring on an Agilent 6890 GC – 5973 MSD. Analysis was done on a 60-m DB-5 column (0.25 mm i.d., 0.25 µm film, J&W Scientific) with He carrier gas at 1.2 mL min^{-1}. Sample volumes of 2 µL were injected in the splitless mode (split opened after 2 min). Inlet and transfer line temperatures were both 280 °C. The GC oven for temperature program was: 100 °C (1 min hold), ramped to 250 °C at 5 °C min^{-1} (10 min hold), then to 310 ^{o}C at 25 °C min^{-1} (5 min hold). Ion source and quadrupole temperatures were 230 °C and 150 °C. Instrumental detection limits (IDLs) were estimated by injecting low concentrations of target analytes until a small peak at ~3:1 signal:noise ratio was obtained. These IDLs are expressed in pg/g sediment. The latter calculation was done assuming extract volumes of 1.0 mL and 10 g of sediment of 20 g of mangrove tissue. Method detection limits (MDL) in sediment samples were defined as the average concentration of relevant congener in blank samples plus 3xSD (Standard deviation). If no peak was observed for a sample constituent (<IDL), the concentration was considered zero (nd = not detected).

Target congeners were not detected in blank samples. Calibration plots were made from 7 dilutions. Samples were quantified vs. a [$^{13}C_{12}$]-PCB-105 internal standard using the linear regression algorithm provided by MSD Chemstation software.

Several quality control steps were taken. Solvent blanks and anhydrous sodium sulfate blanks (anhydrous sulfate treated as a sediment sample) were run. Standard solutions of known concentrations were run every 10 sample runs. A mixture of $^{13}C_{12}$PCB-28, -52, -101, -138, -153, -180 and 209 (5 ng each) was added to samples and blanks as surrogate compounds. The recovery efficiencies (%) of the surrogate PCBs were found 89.5±15.0, 100.3±17.6, 90.2±14.0, 87.7±14.9, 104.6±19.9, 96.2±14.9 and 92.6±14.5 for [13C12]-PCB-28, -52, -101, -153, -138, -180 and -209, respectively. The mean RSD of analyses of unlabeled spikes was calculated to be 4.5 %. All solvents used were pesticide-grade, and anhydrous sodium sulfate and all glassware used was baked at 450 ^{o}C.

Core dating

Age dating was performed on sediment core samples following the protocols of Brooks et al. (2015). Samples were isotopically analyzed for ^{137}Cs (half-life = 30.2 years) and ^{210}Pb (half-life= 22.3 years) to determine annual- to decadal-scale timing of events and sediment accumulation rates over the past ~100 years (Brooks et al., 2015). Samples were analyzed on a GWL Series HPGe (high-purity germanium) coaxial well or planar photon detector for total ^{210}Pb (46.5 keV), ^{214}Pb (295keV and 351 keV), ^{214}Bi (609 keV), ^{137}Cs (661 keV), and ^{7}Be (447 keV) activities.

The constant rate of supply model (CRS) was applied to assign dates for each data point (Binford, 1990). This widely used model assumes a constant supply of unsupported ^{210}Pb (originating from atmospheric and water column ^{222}Rn decay) (Sánchez-Cabeza et al., 2000). Caveats of the CRS model could be related to self-adsorption as a function of sample density (Cochran et al., 1998) which varies according to sample composition, grain size and sample size. Samples from Jobos Bay are a combination of organic, carbonate, and soil material that can change due to tidal action, weather-related events (e.g., hurricanes) and anthropogenic input (e.g., dredging, boating). Such conditions have an impact on the moisture content and sample density (Ithier-Guzmán, 2010). The CRS model is generally used where sediment accumulation rates are variable (Binford, 1990).

Sedimentology

Sedimentology analysis was performed on sediment core samples following the protocols of Brooks et al. (2015). A subsample of about 20 grams of the original freeze-dried samples were used. In samples where less than 20 grams existed, the whole amount was used. To get an adequate representation of the sample, all of the samples were split using the cone and quarter procedure. Samples were coned by pouring the subsample from one piece of paper onto another piece of paper. While pouring the sample onto the paper the bottom paper is spun to create a conical mound of sediment. The subsample was then quartered by dividing the mound sediment into four representative samples. Two of the quarters were combined and archived into a labeled Ziploc bag. The other two quarters were coned and split again so that one contained approximately 1-2 grams of sediment and the other contained more than four grams of sediment.

Calcium carbonate content was determined using the acid leaching method (Milliman, 1972). The 1-2 gram representative sample obtained from the cone and quartered procedure was used for calcium carbonate analysis. The samples were each added to a preweighed 250 mL beaker and reweighed to obtain an accurate amount of sample used. To dissolve the calcium carbonate, 10% HCl was slowly added to the samples. After the reaction was complete, the acid was decanted off. The samples were then washed four times with DI water. The samples were dried in the oven at a temperature of 50°C. Once dried, the samples were stored in a desiccator until they equilibrated with the atmosphere, after which their weights were recorded. The Carbonate percent was then obtained from the difference of the pre HCl sediment weight and the sediment weight post HCl divided by the pre sediment weight and multiplied by 100.

Total organic matter (TOM) was determined by Loss on Ignition (LOI) at 550ºC (Dean, 1974). For TOM analysis the representative sample that already contained calcium carbonate removed was used. Using a scoopula sample was mixed in the beaker and 1 gram of representative sample was extracted out and placed into a preweighed crucible. To get the pre sediment weight, the weight of the crucible plus sediment was subtracted from the weight of the crucible. The crucible was then placed into a muffled furnace at a temperature of 550°C for two and a half hours. Once the sample had equilibrated to room temperature, the crucible and sample were weighed to obtain a post sediment weight. To calculate the percent TOM samples were corrected for carbonate by multiplying the sediment weight difference by the post calcium carbonate sediment weight and dividing by the pre sediment weight, this value was then divided by the pre calcium carbonate sediment weight. The TOM percentage was then calculated by multiplying the carbonate correction value by 100.

Grain Size Analysis

A <63 µm wet sieve was set up in a large funnel with a 1000mL graduated cylinder placed underneath it. In a preweighed 50mL beaker, 10mL of calgon dispersant was pipetted into the 4-gram representative subsample. The addition of calgon is necessary to inhibit flocculation of clay particles and to disperse the aggregates in the soil. To prepare the calgon solution, 10 grams of calgon was weighed out and mixed into one liter of DI water. 10 mL of the solution was then dried in an oven at 50°C to determine the weight of the dispersant. This weight was then subtracted from the samples after pipetting. The sample was then poured onto the <63 µm sieve, making sure that the beaker was rinsed completely clean with DI water. Using a rubber policeman and DI water, the sand and silt was rinsed through the screen. Sieving was stopped once the water that went through the funnel ran clear. The graduated cylinder was then filled to 1000mL with DI water. The sediment that did not pass through the sieve was collected and placed back into the original 50mL beaker. This sediment that did not pass through is referred to as the sand size fraction and large fraction. This beaker was then placed into an oven at a temperature of 50°C until dry. The sample was then reweighed and kept to use for grain size analysis.

Grain size analysis was performed on the sediment samples that were too large to pass through the <63µm sieve. The sample was poured onto a sieve and shake/pushed through the container. The sediment lithofacies associations (LFA) sieve was then weighed with the gravel sample that remained on the top and then weighed by itself. The LFA or gravel was determined by subtracting the sieve-sediment weight from the empty sieve. The sand weight was classified by subtracting the weight of the empty beaker and the LFA from the beaker-sediment weight.

The percent silt and percent clay fractions of the sediment samples were determined by pipette analysis (Folk, 1974). The 1000mL graduated cylinder with the wet sieved mud fraction already in it was used for this analysis. For each sample 2 beakers were used, one representing 4 phi and the other 8 phi. These 50mL beakers were pre-weighed and recorded. Using a stirring rod, the sample was stirred for exactly 60 seconds. After 20 seconds passed, 20mL of water at a depth of 20cm from the meniscus was extracted through the use of a pipette. This was then transferred into the 4 phi beaker. To get a representation of the clay-size fraction, another 20 mL of sample was extracted from the graduated cylinder at a depth of 10 cm from the meniscus an hour and 51 minutes after the completion of the last stirring. This was then pipetted into the 8 phi beaker. Both the 4 phi and 8 phi beakers were then placed into the oven at ~50°C to dry with a paper towel covering the samples. Once the samples were completely dry and had equilibrated to room temperature, they were weighed and recorded. The silt weight was determined by the subtracting sediment weight of the 8 phi sample from the sediment weight of the 4 phi sample, then multiplying by 50. The clay weight was determined by multiplying the sediment weight of the 8 phi sample by 50 and subtracting the calgon.

Creating Maps

Maps were created using Esri ArcGis 10.4 for Desktop. The basemap, World Imagery, was provided from ArcGIS online. The geographic coordinate system used was the World Geodetic System 1984 (GCS_WGS_1984). To show where the sample sites were on the map the coordinate data for the sample sites were added as a layer. Data gathered from chemical analysis and sedimentology were added to the map. The layer properties were set to adequately represent the intended data.

Hierarchical Cluster Analysis

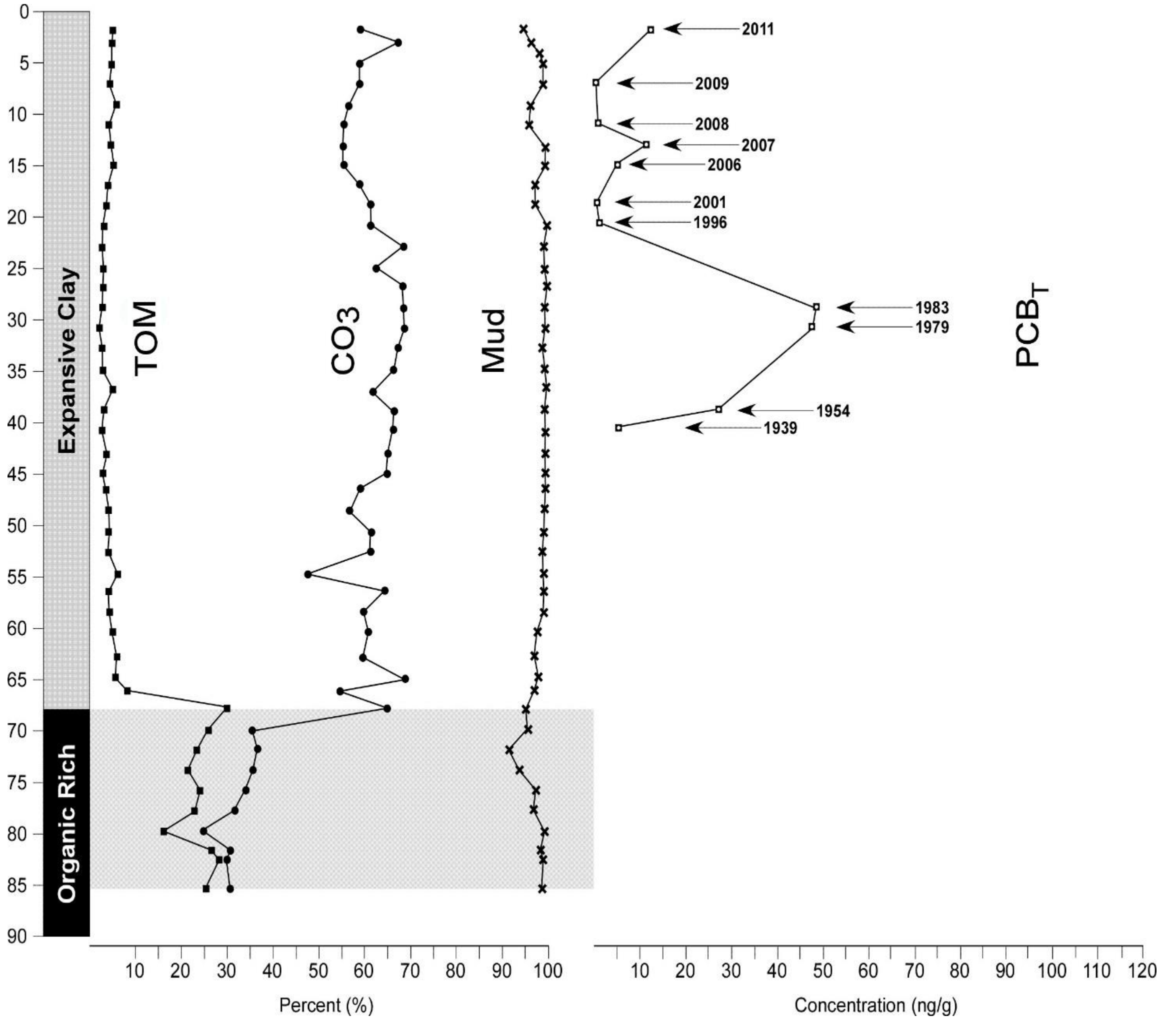
Cluster analysis was performed on the percent of light versus heavy PCBs within the surface sediments and on the total PCBs, percent TOM and percent mud (silt and clay fraction) in the study site. Prior to cluster analysis, the data was fourth root transformed in Excel. The statistical program PAST 3.14 was used for statistical analysis. The Wards method with Euclidean distances was used to generate a cluster dendrogram (Mourier et al., 2014).

RESULTS AND DISCUSSION

Temporal trends of PCBs

Table A1 in Appendix A presents the concentrations of PCBs in each segment of the sediment core taken at Jobos Bay. Figure 4 shows the temporal trends of PCBs in the core. When total PCBs are plotted against depth, a general pattern is seen with an increase toward the surface until a subsurface maximum, followed by a decrease to the sediment-water interface. Core dating could be done confidently until around the late 1930s (mean error +/-5 years), beyond which the percent error becomes unacceptable. Figure 4 shows that the historical trend in concentrations is in agreement with the known history of use of PCBs. PCBs were introduced in 1929 and saw increased sales and use through subsequent decades in the United States (including Puerto Rico), peaking around 1970, with bans in use and production in most countries beginning in the 1970s, culminating in their inclusion in the Stockholm Convention on POPs in 2001. Figure 4 shows a general trend of increasing levels from the late 1930s peaking in the late 1970s-early 1980s and subsequently a decrease, coinciding with the bans in production and use, but with a couple of smaller spikes around 2007 and at the time of sampling. Mourier et al. (2014) reported similar spikes years after PCB levels in sediment cores in France had been declining. These spikes may be the result of more recent increased inputs from surrounding sources as detailed previously. Therefore, the PCB sedimentary profile observed in this study is likely a fair representation of the history of local pollution, albeit taking into consideration potential confounding factors. Lebeuf and Nunes (2005), for example, have indicated that in some locations the effect of sedimentation rates and mixing may significantly impact POPs sedimentation profiles. This, along with differing degradation rates as well as the effect of atmospheric deposition (a factor not measured in this study), may explain deviations from the general trend. 2007 was a very active hurricane/tropical storm year in the Caribbean. Hurricane Noel, in particular, affected the Caribbean, including Puerto Rico. It has been noted that severe storms often lead to pulses of sediments and soils being washed into coastal areas due to intense rainfall events. This may explain the spike in PCBs concentrations in 2007 as PCB-contaminated soils and sediments may have been washed into the study site from the surrounding area. The reason(s) for the spike in PCBs concentration in 2011 are unclear.

Figure 4 indicates that total organic matter associated with sediments (TOM) is fairly constant over time. Previous studies have shown that normally concentrations of POPs increase with an increase in organic content of sediments and soils (Bayen, 2012; Faroon & Olson, 2000). Due to their hydrophobicity, these chemicals adsorb onto the organic phase. However, TOM is not the sole determining factor in PCB adsorption. Estuaries have variable hydrodynamic conditions, which can also affect the uptake of PCBs. Several studies have demonstrated the lack of a correlation between TOM and PCBs concentrations in the sediments of estuarine environments, indicating that the concentrations of organic pollutants cannot be explained just by their affinity for the organic matter (Piérard *et al*., 1996; Herrmann and Thomas, 1984; Zhao et al., 2010). The fact that PCB concentrations in the core fluctuated despite the TOM remaining quite constant may indicate that in some locations the input magnitudes may mask any relationship between those two factors. Additionally, other factors including localized parameters (including different profiles in microorganisms that degrade PCBs) may result in different degradation rates which mask any relationship between TOM and concentrations.



TOM = total organic matter, CO_3 = calcium carbonate

Figure 4. Temporal trends in total PCBs (PCB_T) in core

Figure 5 shows the results when comparing congener profiles of total PCBs measured in each core segment in terms of light (defined as sum of tri-, tetra-, and penta-chlorinated PCBs) versus heavy (defined as sum of hexa-, hepta-, and octa-chlorinated PCBs) PCBs. Overall, in the majority of segments (84%) the lighter PCBs were predominant, although the percentage of heavy PCBs increases going up to the surface (more recent times). In general, the presence of lighter PCBs is suggestive of increased atmospheric contribution (since these congeners are more susceptible to long range transport). Heavier PCBs suggest a larger contribution from direct emissions from local sources. The dominance of lighter PCBs can also be attributed to the anaerobic conditions deeper down the core. Anaerobic conditions allow for PCB dechlorination of heavier PCBs to occur, thereby leading to an increase in lighter PCBs (Liang et al., 2014). The presence of heavy PCBs closer to the surface suggests that local sources (e.g. power plants, oil refinery, solid waste landfill) have become more significant sources to the PCB load to the study area in more recent times. The use of PCB-containing materials lagged in Puerto Rico compared to the mainland United States so it is expected that input from local sources should be more important more recently.

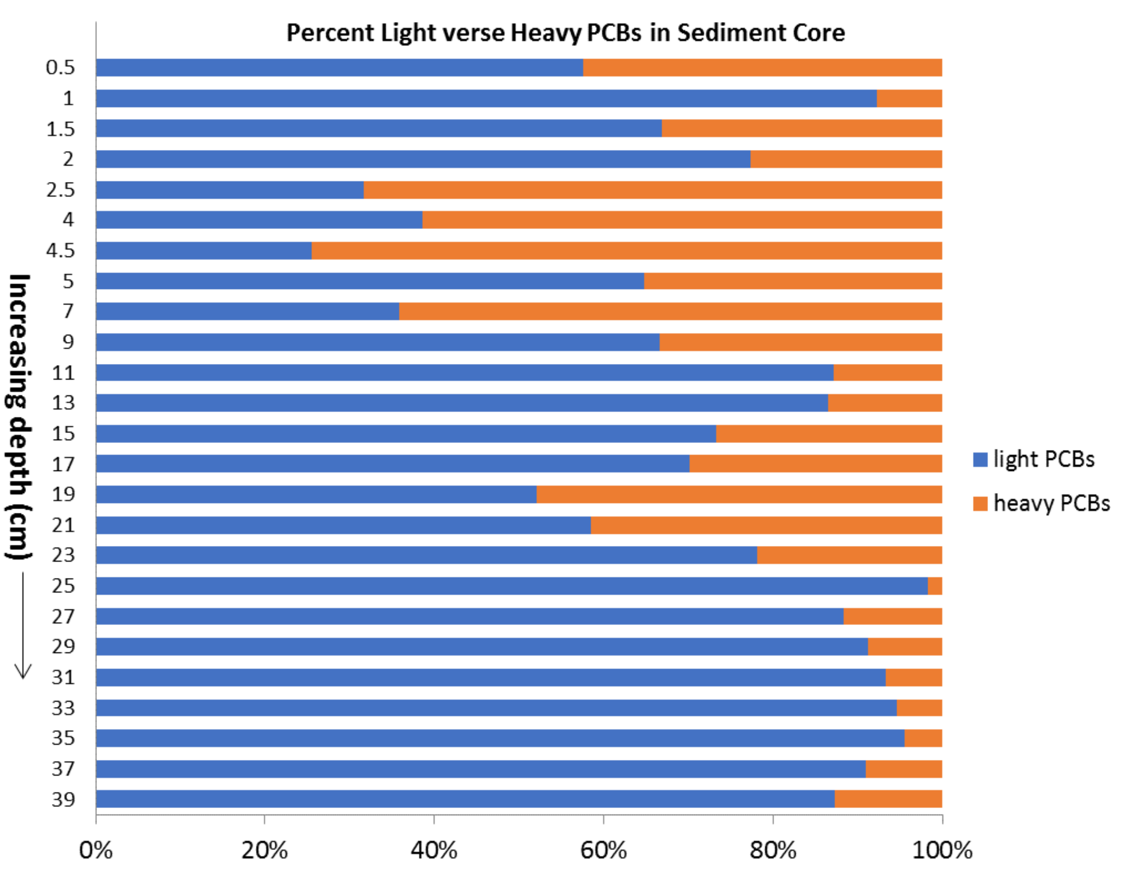


Figure 5. Light vs heavy PCB congeners by core segment (depth in cm)

PCB levels in surface sediments

Table A2 in Appendix A presents the concentrations of individual congeners of PCBs measured in the surface sediments collected in the study area. Table 1 and Figure 6 show total PCBs (ng g^{-1} dw) measured at the 18 stations. Levels ranged from 0.42 ng g^{-1} at site 15 to 1232 ng g^{-1} at site 2. As noted previously, caution must be exercised in interpreting results since factors such as resuspension, mixing, and local movement patterns of waters can affect the ultimate deposition of PCBs in sediments and therefore levels at different sites. In addition, atmospheric deposition may affect different sites differently even in a relatively small study area. There is no information on atmospheric deposition of PCBs in the study area and this study did not have as an objective the determination of this input mechanism. Finally, different rates of degradation at different sites due to different local conditions (environmental, chemical, etc.) may result in different total concentrations. Nonetheless, higher levels were generally seen at sites closer to shore and potential sources of PCBs. For example, sites 1, 2, and 4 are located close to Salinas (population > 30,000) and sites 11 and 14 close to Central Aguirre (a neighborhood of Guayama that is site of a petrochemical plant and a sugar mill that operated from the late 1800s until 1990) while lower levels were seen at sites farther removed from these direct sources (e.g. sites 15 – 17).

Salinity is another factor that can play a role in POPs distribution. Salinity can facilitate sediment redispersal within estuaries and enhance the sorption potential of orgnic contaminants (Bayen, 2012; Li et al., 2016). However, as indicated by Figure 7, salinities at the different sites were quite uniform (with a mean +/- standard deviation of 31.22 +/- 0.87). Therefore, the differences in PCB concentrations seen between sites cannot be explained by differences in salinities. Figure 8 shows the lack of correlation between salinity and total PCBs. A rain event occured in Jobos Bay and the surrounding area the day before sampling was conducted. This input of fresh water could have flushed the area, thereby affecting the Bay’s salinity. That coupled with the water movement within the Bay (this factor was not examined in this study) and tidal exchange could have contributed to the homogenization of salinities within the area. Thus, any differences in salinities that may have helped explained the variation in PCB concentrations may have been masked by this rain event.

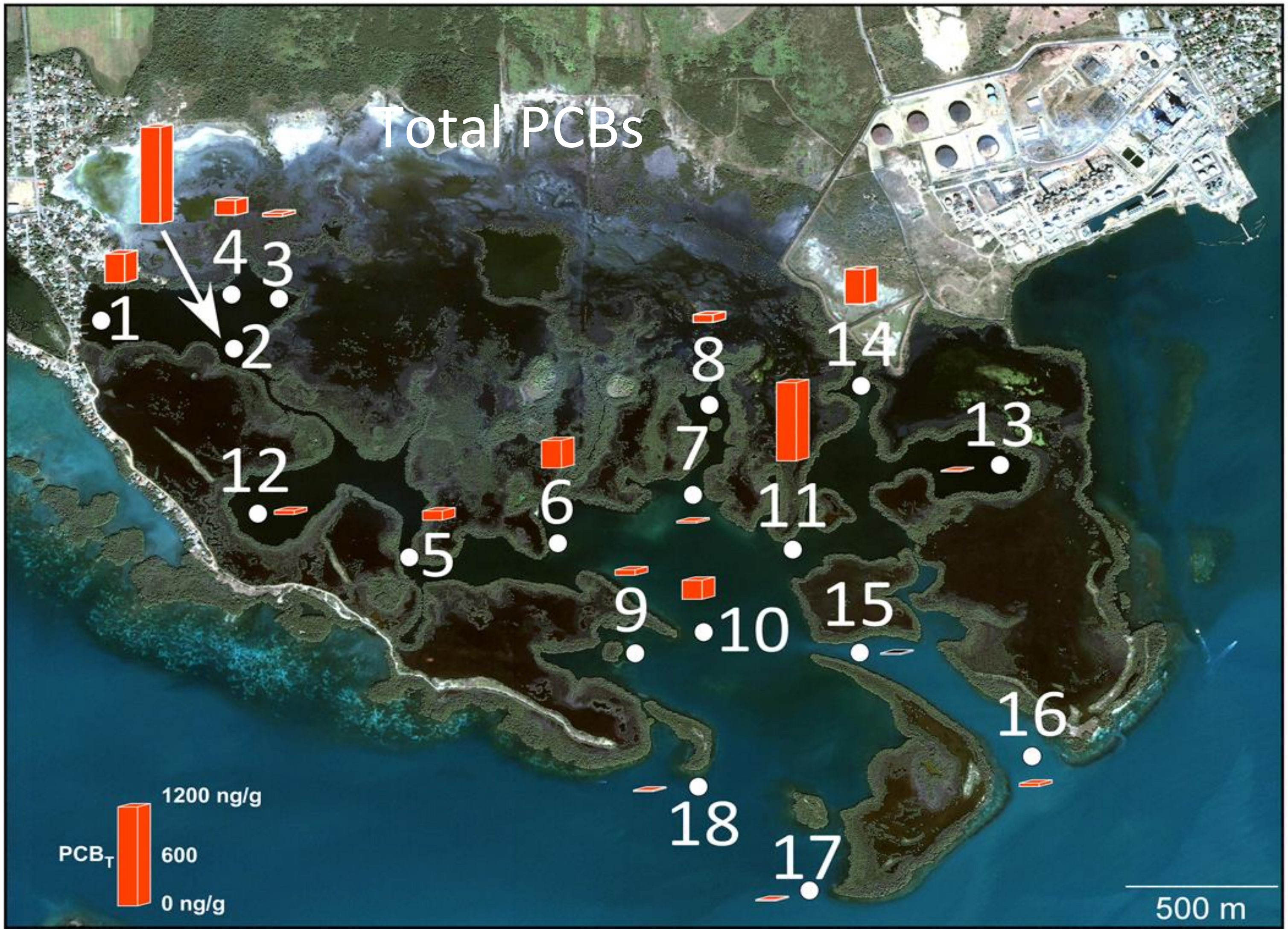


Figure 6. Total PCBs (ng g^{-1} dw) measured at the sampling sites.

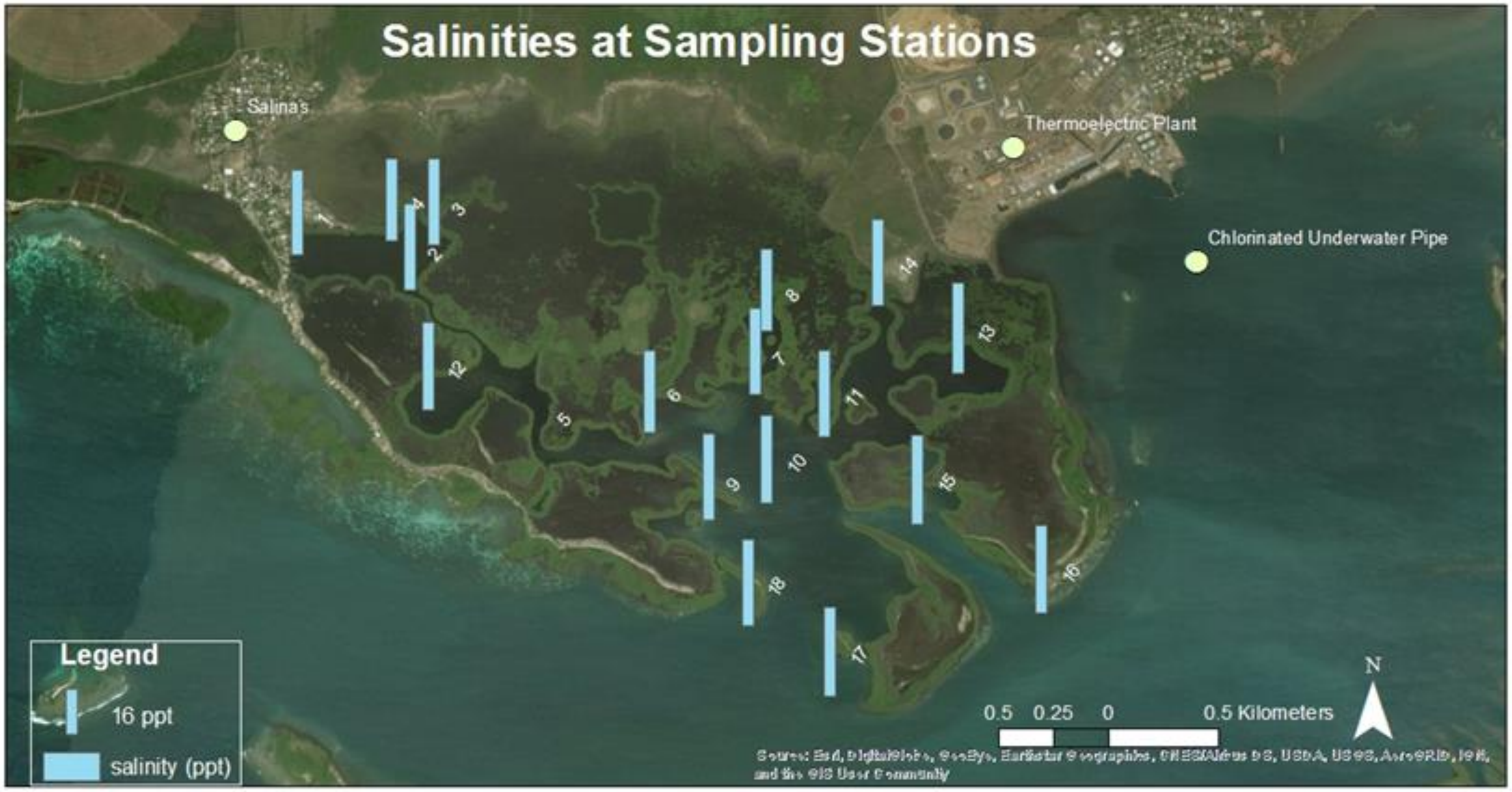


Figure 7. Salinity (ppt) at the sample sites. There was no data for sample site 5.

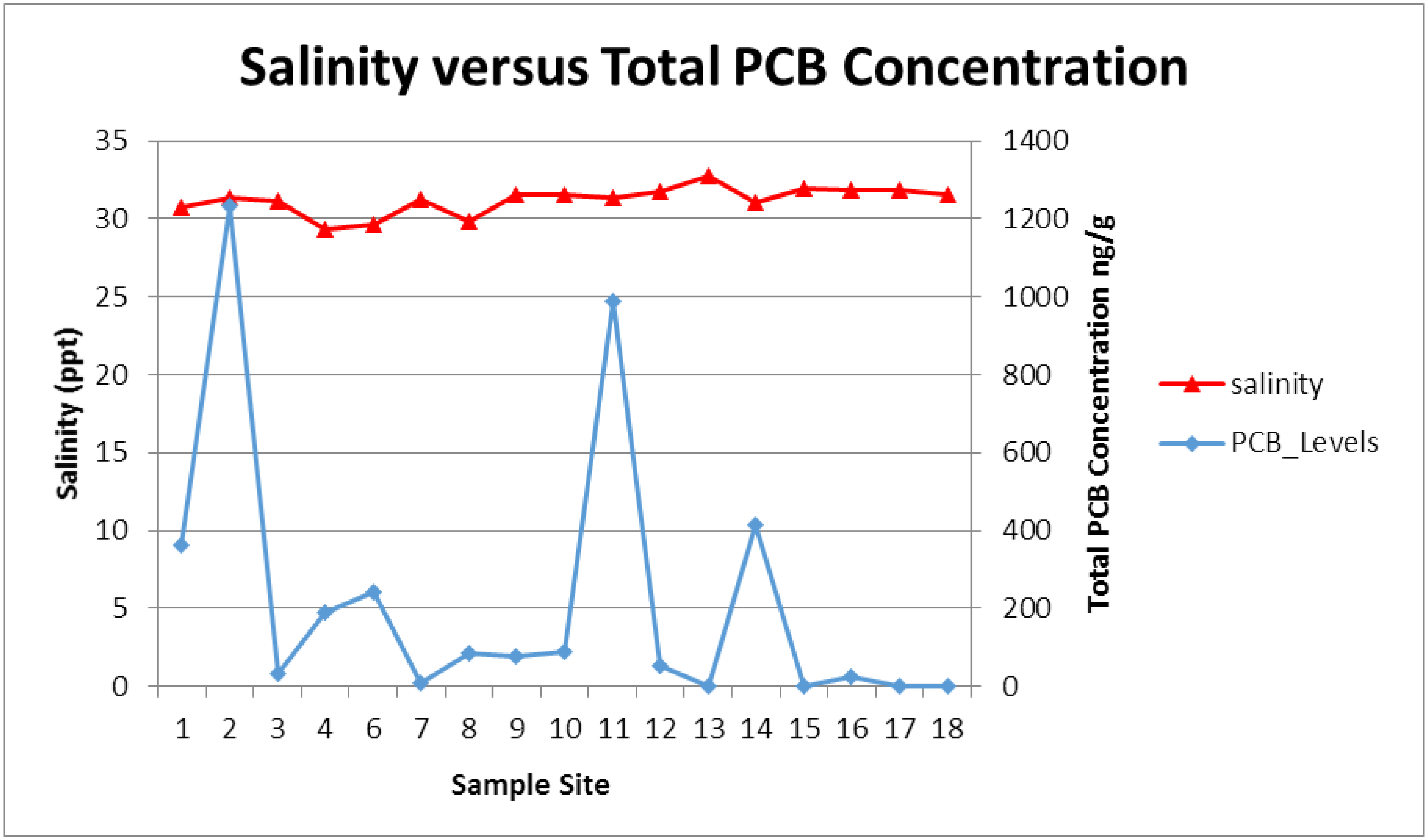
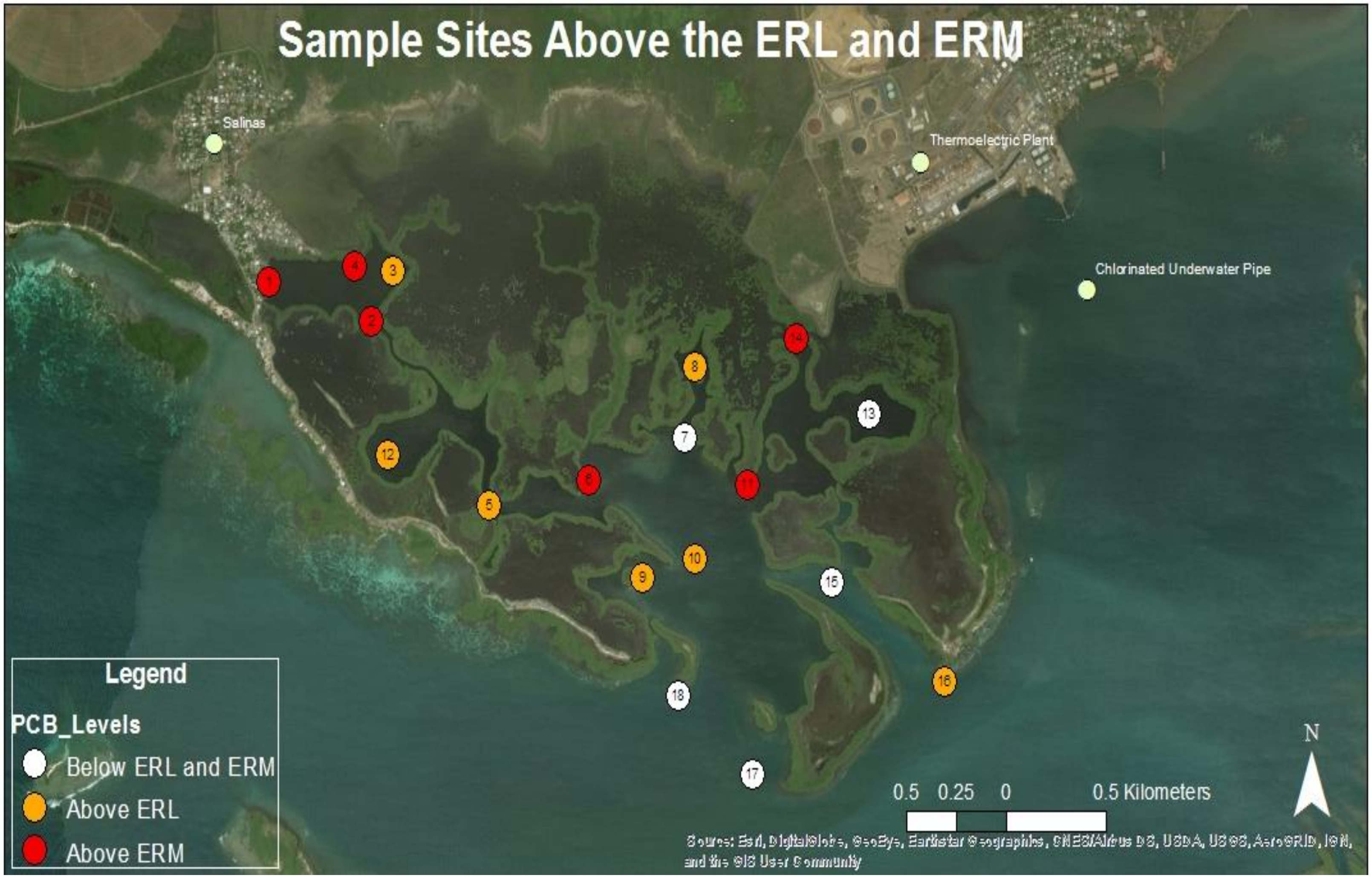


Figure 8: Salinity compared to the total PCB concentration in the sample sites.

The levels measured in this study are generally higher than those reported for the same study site by Aldarondo-Torres et al. (2010) for samples taken in 2003. In that study the authors reported levels of total PCBs from below detection to 11.21 ng g^{-1} dry weight, although it targeted 14 rather than 50 PCB congeners as in this study so any comparison must be interpreted with caution. That study reported total PCBs consistently being dominated mainly by two congeners, PCB 138 and PCB 153, with the others being below detection. In our study, the situation was similar, with total PCBs being dominated by a small number of congeners, although the dominant ones were not consistently the same in each sample. Unlike the 2010 study, only in 4 stations (13, 15, 17 and 18) were most congeners below the quantification limits. In the other stations, although a few congeners dominated total PCBs, most of the other congeners were quantifiable but at far lower concentrations. Compared with reports for other parts of Puerto Rico, the levels in this study are similar to those reported for sediments in Guanica Bay (Whitall et al., 2014) and higher than values reported for Vieques (Pait et al., 2010). In Guanica Bay, 53 congeners were targeted (33 common to the 50 targeted in this study) so the comparison is reasonable. In the Vieques study, however, only 17 congeners were targeted so more caution is necessary in making a comparison.

The National Oceanic and Atmospheric Administration (NOAA) has set sediment water quality guidelines (SQG) for total PCBs of 22.7 ng g^{-1} as the ERL (effects range – low) and 180 ng g^{-1} as the ERM (effects range – median) (Long et al., 1995). These values represent statistically-derived levels of contamination above which toxic effects would be expected to be observed in benthic organisms with at least a 50 % frequency (ERM) and below which effects were rarely (< 10 %) expected (ERL). Six sites (sites 1, 2, 4, 6, 11 and 14) showed levels above the ERM and seven (sites 3, 5, 8, 9, 10, 12, 16) showed levels above the ERL (Figure 9). Based on these results, the study area can be described as moderately to highly polluted.

Total PCBs in our study are also higher than values reported for the Rio de La Plata estuary in Argentina of <0.1 – 100 ng g^{-1} dw based on 42 congeners (Colombo et al., 1990), Cienfuegos Bay in Cuba of 3.1 – 15.5 ng g^{-1} based on 11 congeners (Tolosa et al., 2010), Gulf of Batabano, Cuba of 0.12 – 0.28 ng g^{-1} based on 25 congeners (Alonso-Hernandez et al., 2014), and and 11 coastal sites in the Dominican Republic of 0.46 – 41.9 ng g^{-1} based on 23 congeners (Sbriz et al., 1998). They are lower than values reported for the Lenga estuary, Chile of 20 – 10,000 ng g^{-1} based on 36 congeners (Pozo et al., 2014).



Source: Esri, DigitalGlobe, GeoEye, Earthstar Geographics, CNES/Airbus DS, USDA, USGS, AeroGRID, IGN, and the GIS User Community

Figure 9. Sample sites above the ERL and ERM.

Table 1 and Figure 10 show the breakdown of PCBs by homolog type (tri-chlorinated through octa-chlorinated PCBs) at each of the 18 sites. Figure 10 indicates that there is no clear, consistent pattern in terms of homolog composition at the sites. This suggests varied sources of the PCBs measured, including atmospheric deposition as a potentially significant source. When congeners are divided between light (tri- through penta-chlorinated) versus heavy (hexa- through octa-chlorinated) PCBs, lighter PCBs were dominant in 12 sites and heavy PCBs were dominant in 6 sites (Figure 11). There is no clear correlation between type of PCBs (light or heavy) and total concentrations. For example, of the six sites where heavy PCBs were dominant, two (sites 2 and 6) contained very high levels of total PCBs while two sites (sites 13 and 18) contained very low levels. There is also no clearly obvious correspondence between the locations of the sites (close to shore versus more offshore) and the predominance of light verse heavy PCBs. Therefore, it is difficult to make conclusions. The results may indicate different sources along with other factors such as current directions, mixing of surface sediments, etc.

Table 1. Total PCBs (ng g-1) and congener profiles (%) of surface sediments.

Station =	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**	**15**	**16**	**17**	**18**
ΣPCBs	361	1232	34	189	117	241	8.5	83	77	89	988	54	1.2	415	0.42	25	0.80	0.70
triPCBs	6.0	3.7	3.9	87.1	2.7	14.4	6.8	43.1	2.6	50.4	0.9	9.9	5.2	44.3	0.0	49.9	0.0	1.5
tetraPCBs	22.0	2.7	23.2	2.2	9.3	1.1	2.8	19.5	2.7	2.0	0.7	15.8	22.0	7.4	48.7	3.8	23.5	13.1
pentaPCBs	41.0	9.0	57.7	7.5	68.9	27.8	9.5	31.3	35.6	15.5	56.7	46.7	0.0	37.5	10.6	13.5	51.7	13.9
hexaPCBs	10.0	5.9	9.5	1.7	6.6	7.1	0.9	2.6	2.7	3.0	0.3	21.5	53.9	6.9	40.8	3.5	22.5	71.5
heptaPCBs	6.0	35.3	3.8	0.3	5.9	21.0	73.2	1.9	28.4	2.5	3.2	1.6	19.0	2.5	0.0	2.1	2.4	0.0
octaPCBs	15.0	43.4	2.0	1.3	6.6	28.7	6.9	1.6	28.0	26.6	38.3	4.5	0.0	1.4	0.0	27.2	0.0	0.0

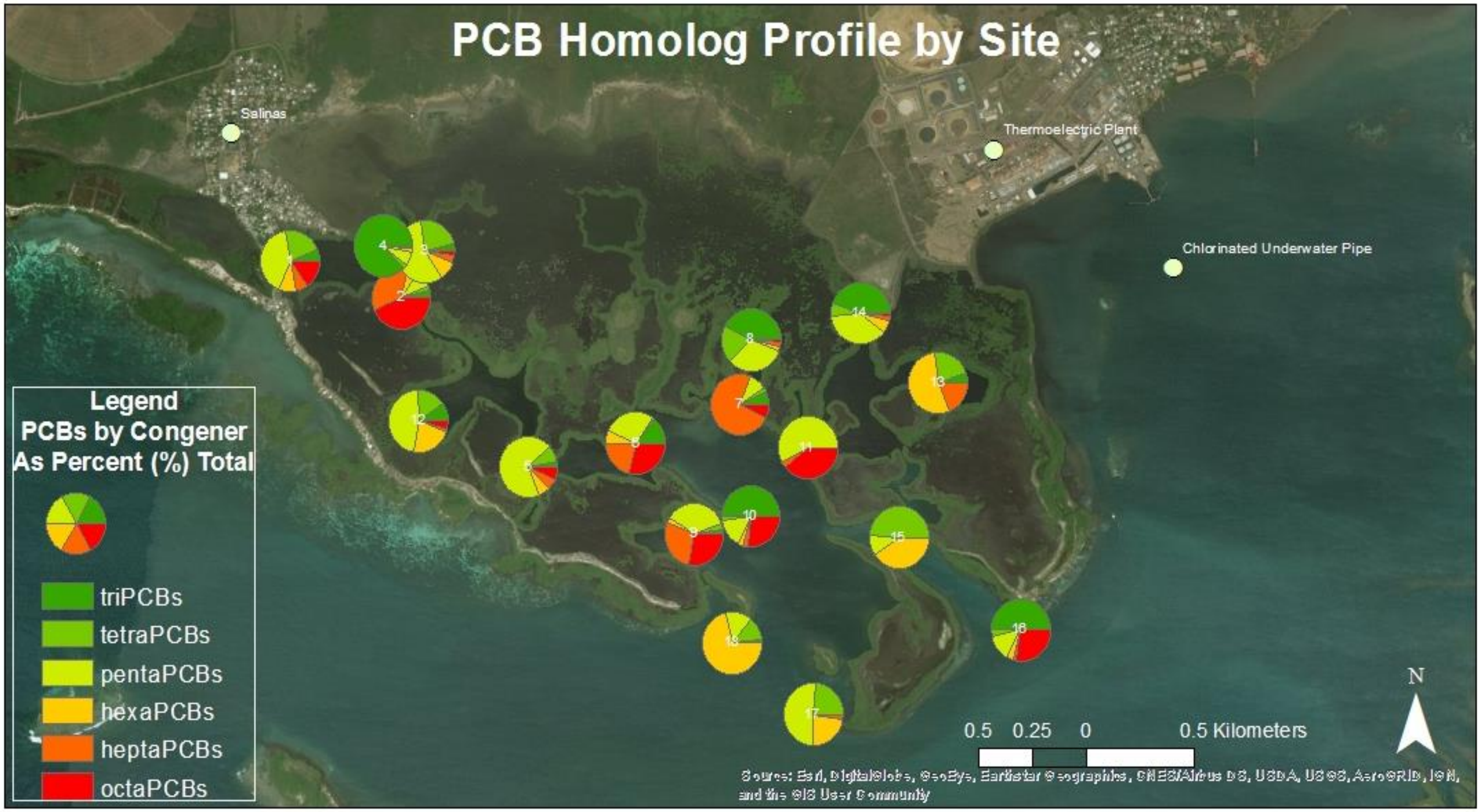


Figure 10. PCB homolog profile by site.

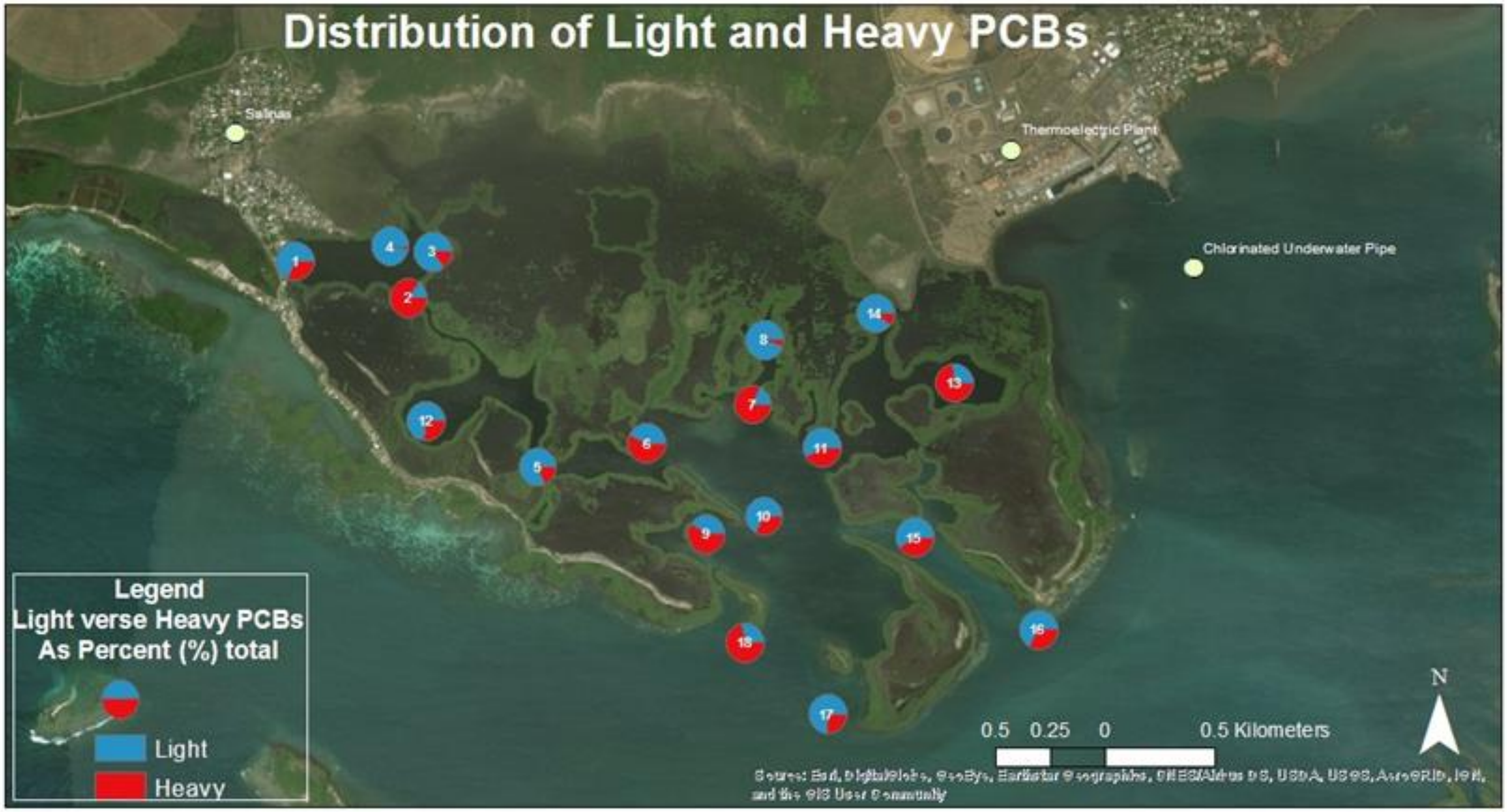


Figure 11. Comparison of light versus heavy PCBs by site.

Cluster analysis revealed 3 different clusters (Figure 12). The green outlined cluster contains stations 6, 9, 2, 7, 13, 18. The blue outlined cluster contains stations 11, 15, 12, 17, 1, 10, 16, and the orange outlined cluster contained 4, 8, 14, 3, 5. From the green to blue to orange clusters the total PCBs are increasingly dominated by the lighter PCBs. There is, however, no correlation between the chlorination of PCBs to their location in the study area (i.e. there are no spatial clusters with similar profiles). Other sources, like atmospheric deposition, dechlorination, and point sources could be driving the distribution of light and heavy PCBs.

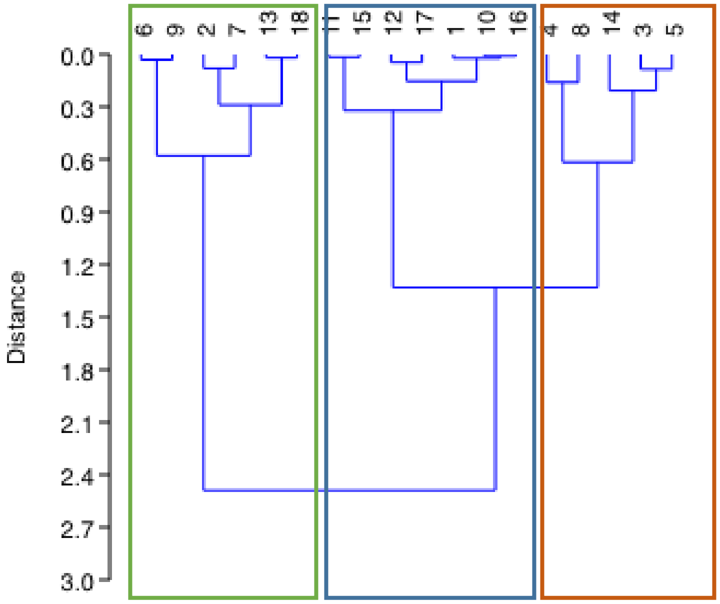


Figure 12. Dendrogram showing the distribution of light verse heavy PCBs

PCBs were sold as mixtures, called Aroclors in the United States, which differed in the percent chlorination and thus had different homolog profiles by weight. For example, triPCBs was the dominant homolog in Aroclor 1016 and Aroclor 1262, pentaPCBs were the dominant homolog in Aroclor 1248 and Aroclor 1252, and hexaPCBs and heptaPCBs were the dominant homologs in Aroclor 1260 and Aroclor

1262 (information available at: ttps://www.atsdr.cdc.gov/toxprofiles/tp17-c4.pdf). Table 1 indicates that triPCBs are the dominant homolog at stations 4, 8, 10 and 16; pentaPCBs were dominant at stations 1, 3, 5, 11, 12, and 17; and hexaPCBs and heptaPCBs were dominant at stations 7, 13 and 18. Aroclor 1016 was used mainly in capacitors; Aroclor 1262 was used mainly as plasticizers; Aroclor 1248 was used in a variety of applications including in hydraulic fluids, vacuum pumps, in plasticizers and adhesives; Aroclor 1260 saw use in transformers, hydraulic fluids, synthetic resins and dedusting agents; and Aroclor 1262 was used mainly in synthetic resins. Thus, the sources of PCBs to different sites may be different materials containing different types of Aroclors. At stations 4, 8, 10 and 16 materials containing Aroclors 1016 and 1262 may be the major sources; stations 1, 3, 5, 11, 12, and 17 materials containing Aroclors 1248 and 1252 may be the dominant sources; and at stations 7, 13 and 18 materials using mainly Aroclors 1260 and 1262 are dominant. It must be stressed that caution must be used in making such interpretations since additional factors such as degradation of the heavier PCBs to the lighter ones may change the original PCB profiles at given sites. Differences in degradation rates of heavier PCBs at different sites may help explain why there are no spatial clusters – that is, groups of sites with similar PCB profiles in clustered geographical locations. Instead, the sites with similar profiles are dispersed throughout the study area, all the way from the stations closer to the urban center to the stations closer to the petrochemical plant, for example. More comprehensive studies would be needed to elucidate these details.

PCB levels in mangrove tissue

In the Caribbean, including Puerto Rico, limited studies have focused mainly on the effects of agricultural POPs on sediment-water quality (Potter et al., 2013; Rawlins et al., 1998). There are no reports documenting the effect of POPs on plants including mangroves. Previous studies in other areas have shown that POPs, such as pesticides, can bioaccumulate in vegetation (Shete et al, 2009; Hellstrom, 2004; Patterson et al., 1990; Walsh et al., 1974; Walsh 1973). Walsh et al. (1974) reported the bioaccumulation of some POPs, including PCBs, in *Rhizophora mangle L* (red mangrove). They reported translocation of different POPs from soils to various parts including roots, hypocotyls, stems and leaves. In the more recent report by Shete et al. (2009) the presence of OC pesticides was documented in the roots and leaves of *Avicennia marina* (grey mangrove) in India as well as the sediments associated with this mangal species. Levels were highest in the roots and the authors concluded uptake of these organic pollutants by *A. marina*.

In this study, levels of PCBs were determined in mangrove tissues collected when possible at the sediment sampling sites. Due to analytical difficulties, values are not available for all samples or all types of mangroves. Data was more complete for samples of *R. mangle* (red mangrove). Therefore, Table 2 shows the values for total PCBs in the roots, leaves and seeds of *R. mangle*. Figure 13 shows the representation of PCB levels in the sediment and the mangrove tissues. The results indicate that PCBs are bioaccumulated in mangroves in Jobos Bay, likely from the sediment with which they are associated. In five of the nine sites (sites 3, 6, 8, 12 and 17) the concentrations in leaves are actually higher than that found in the nearby sediments (although it should be cautioned that lower PCB concentrations in sediments may be due to faster degradation of PCBs in sediments at different sites due to localized conditions, which will also affect any calculations of bioconcentration factors).

Table 2. Total PCBs in R. mangle tissue (ng g-1 dw)

Station =	**2**	**3**	**4**	**5**	**6**	**8**	**11**	**12**	**17**
ΣPCBs-seed	NM	16.0	75.2	NM	NM	NM	24.3	NM	93.4
ΣPCBs-leaves	18.6	154	108	14.1a	392	322	216	747	270
ΣPCBs-root	4.60	NM	18.7	58.5	120	5.16	NM	0.26	8.36
ΣPCBs-sediment	1232	34.0	189	117	241	83.0	988	54.0	0.80

NM = not measured (due to analytical difficulties), [a]*L. racemosa*.

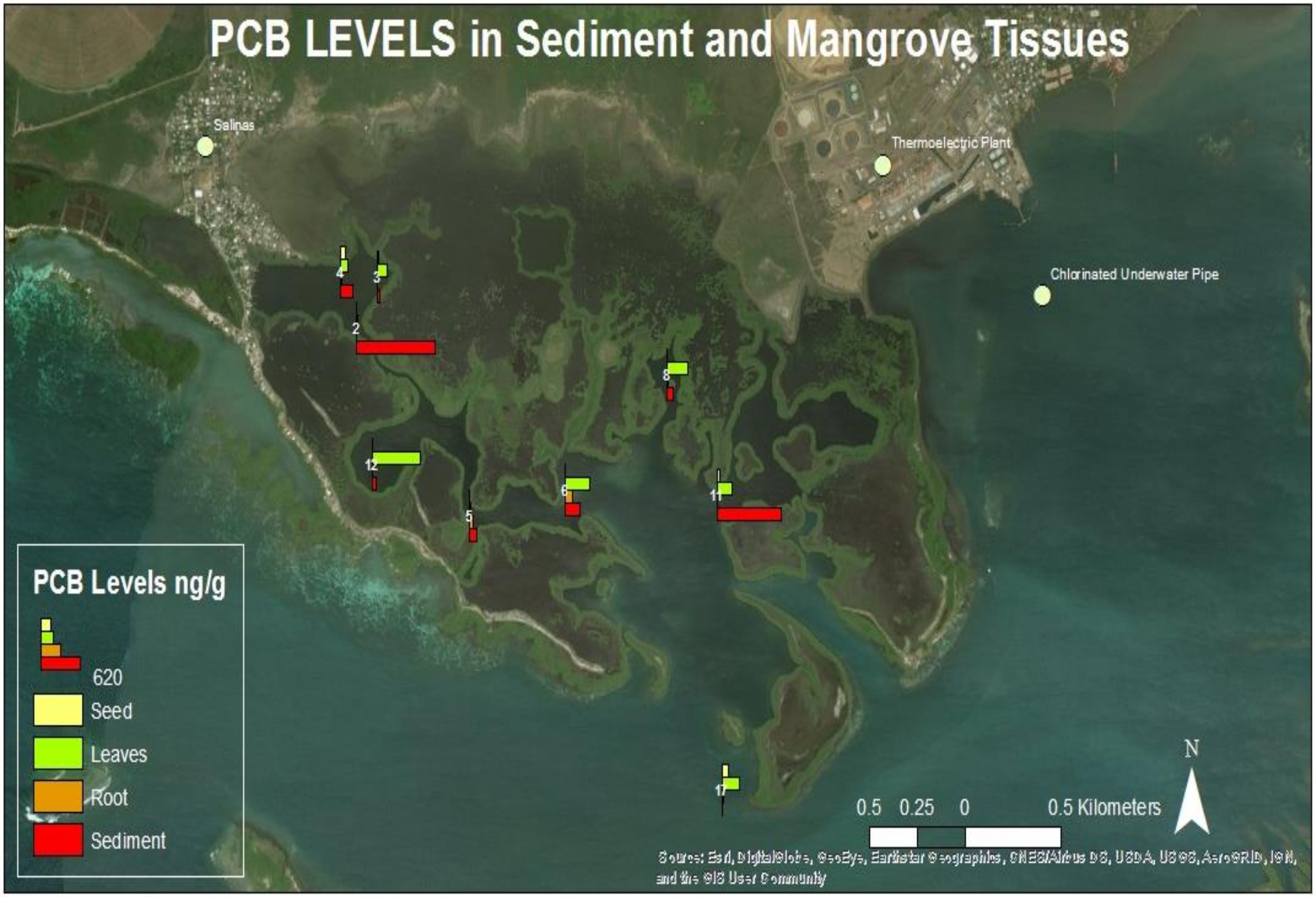


Figure 13. PCB levels in the sediment and mangrove tissues in relation to sampling site.

To assess if bioconcentration between the mangrove sediments and the mangrove plant were occurring, the bioconcentration factor was computed. The bioconcentration factor (BCF) was calculated as the ratio of total PCB concentrations in the mangrove leaf versus the total PCB concentration in the associated sediment. Where BCF = $C_{leaf}/C_{sediment}$. C_{leaf} is the total PCB concentration in the mangrove leaf and $C_{sediment}$ is the total PCB concentration in the mangroves associated sediment (MacFarlane et al., 2007). Despite the limitations inherent when dealing with a small number of samples, bioconcentration factors (BCF = $\text{concentration}_{\text{mangrove leaf}}/\text{concentration}_{\text{sediment}}$) calculated for these sites are: site 3 (4.5), site 6 (1.6), site 8 (3.9), site 12 (13.8) and site 17 (338) (Figure. 14). BCFs greater than 1 indicate that bioaccumulation is likely occurring in mangroves in the area.

At sites where values are available for roots and leaves (station 2,4,6,8,12,17), PCB levels were higher in leaves, suggesting translocation from roots to leaves (the exception is at station 5, where the concentration in leaves was not available for red mangrove; the value shown in Table 2 is for white mangrove, suggesting differences between mangrove species may be possible). The translocation factor (TF) was calculated to determine PCBs mobility within the mangrove plant. TF was calculated by TF= C_{leaf}/C_{root}. C_{leaf} is the total PCB concentration in the mangrove leaf and C_{root} is the total PCB concentration in the mangroves root (Lotfinasabasl et al., 2013). The TF calculations for station 2 is 4.0, station 4 is 5.8, station 6 is 3.3, station 8 is 62.4, station 12 is 2873, and station 17 is 32.3. A TF value greater than one (1) signifies that the mangrove plant is translocating PCBs effectively from the root to the leaves. There was minimal data for PCB levels in mangrove seeds. However, in the available samples seedlings showed uptake of PCBs. This translocation of PCBs in the mangrove plant suggests that PCBs are capable of bioconcentrating throughout trophic levels. Patterson et al. (1990) indicated that POPs may enter roots in solution from soil or directly from water in aquatic plants. They suggested that uptake into plant roots is directly proportional to the substance’s octanol/water partition coefficient (Kow) and that transfer to shoots is more efficient for substances of intermediate solubility such as PCBs. This probably explains the bioaccumulation of PCBs seen in this study.

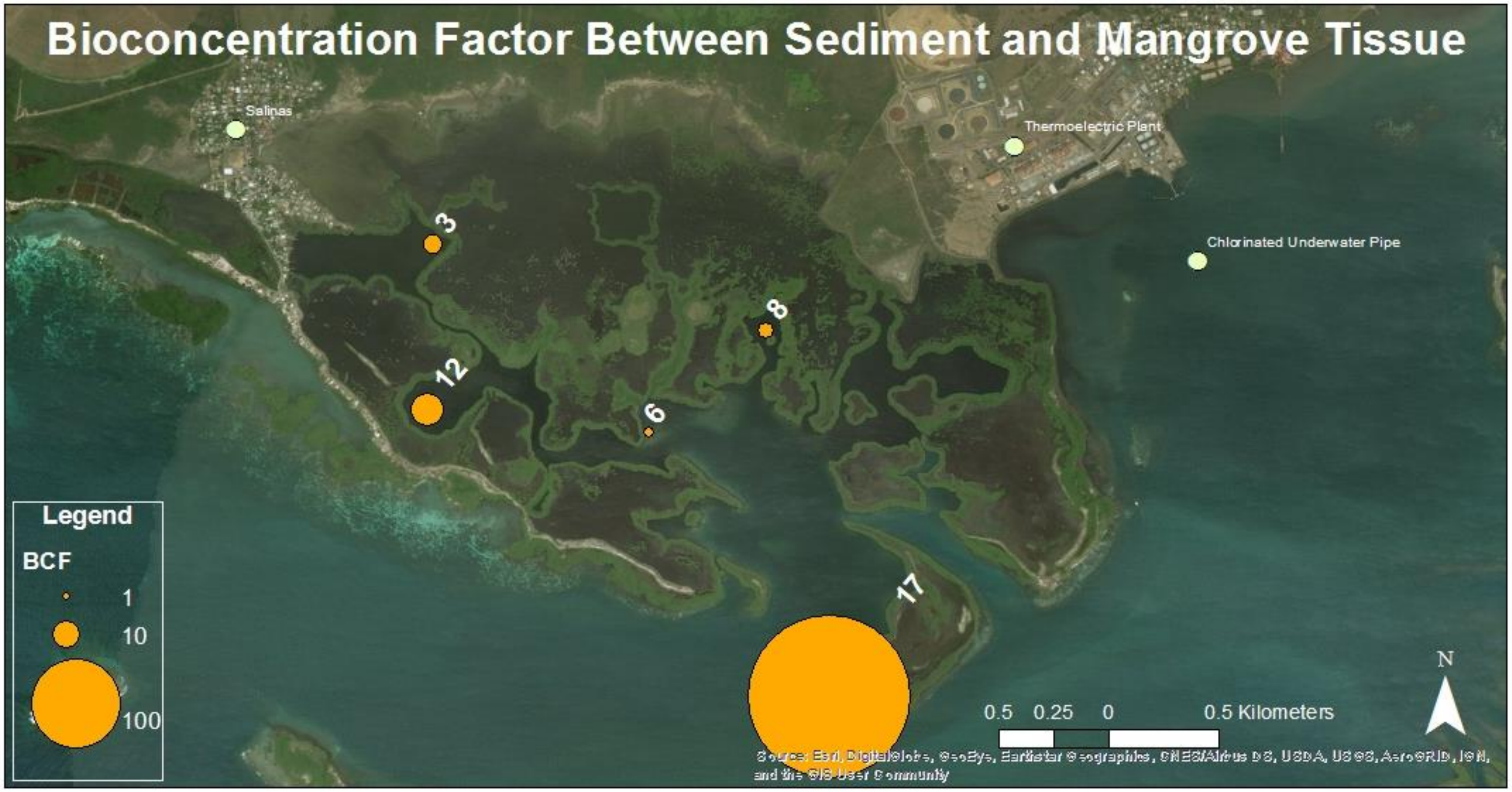


Figure 14. Representation of bioconcentration factor from the sediment to the leaves in reference to sample site.

Grain Size Analysis on Sediment Core

Figure 15 shows the sediment composition of the core sample in terms of percentage of gravel, sand, silt, and clay at each sample interval. The most widespread grain size in the sediment core were clay and silt (<63 μm). These fine grain particles accounted for an average of 97.8% of the sediment core samples. Silt was the dominate sediment size having an average 59.3%, clay had an average percentage of 38.5%. Gravel sediment was only found on the surface of the core and at 72.5 cm. Sand sediment was found in low amounts throughout the core, having an average percentage of 1.51 percent. A higher percentage of sand was found at the ends of the core sample.

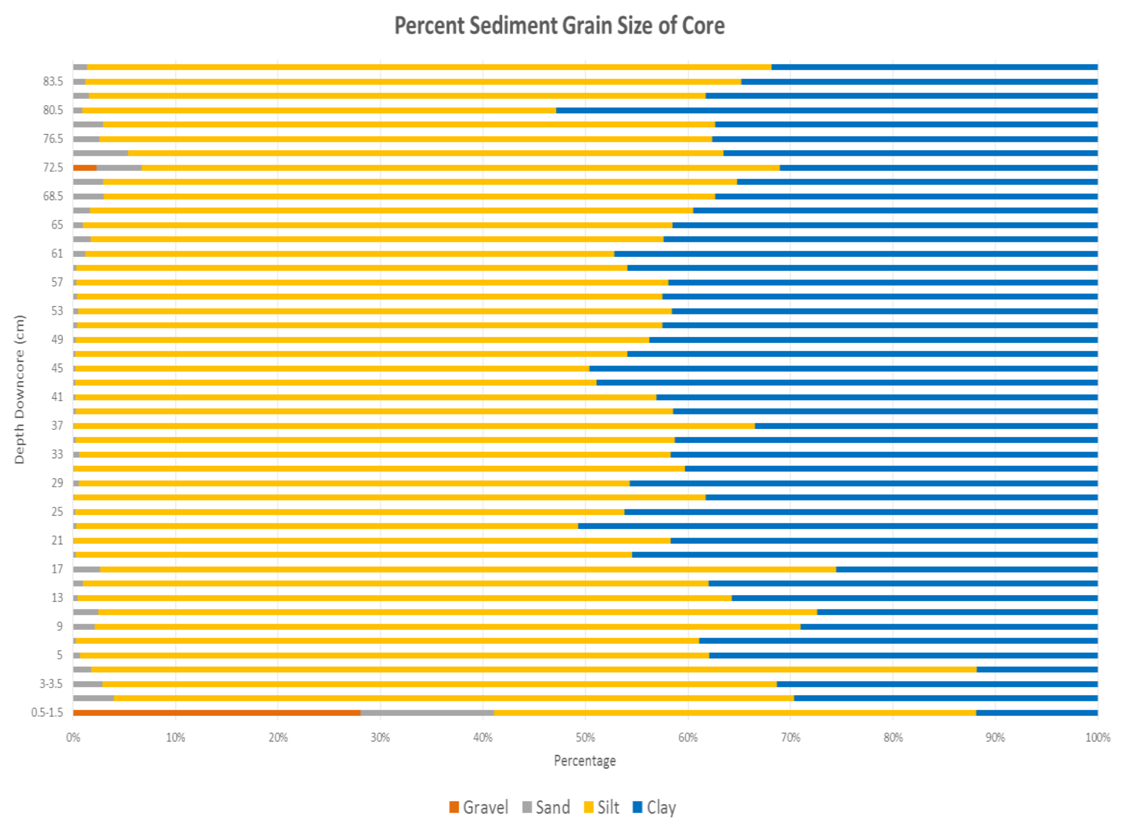


Figure 15. Grain size distribution in the sediment core samples (%)

Grain Size Analysis on Surface Samples

The sediment composition of the surface samples is reflective of the profile of Jobos Bay, sandy mud, silty clay, and carbonate sediments (Lugo et al., 2007). Figure 16 demonstrates the varying composition of the surface sediments at each of the sample sites. This varying composition is likely to affect the adsorption of PCBs because PCBs adhere more readily to sediments with large amounts of clay and organic matter than sand or gravel sediments.

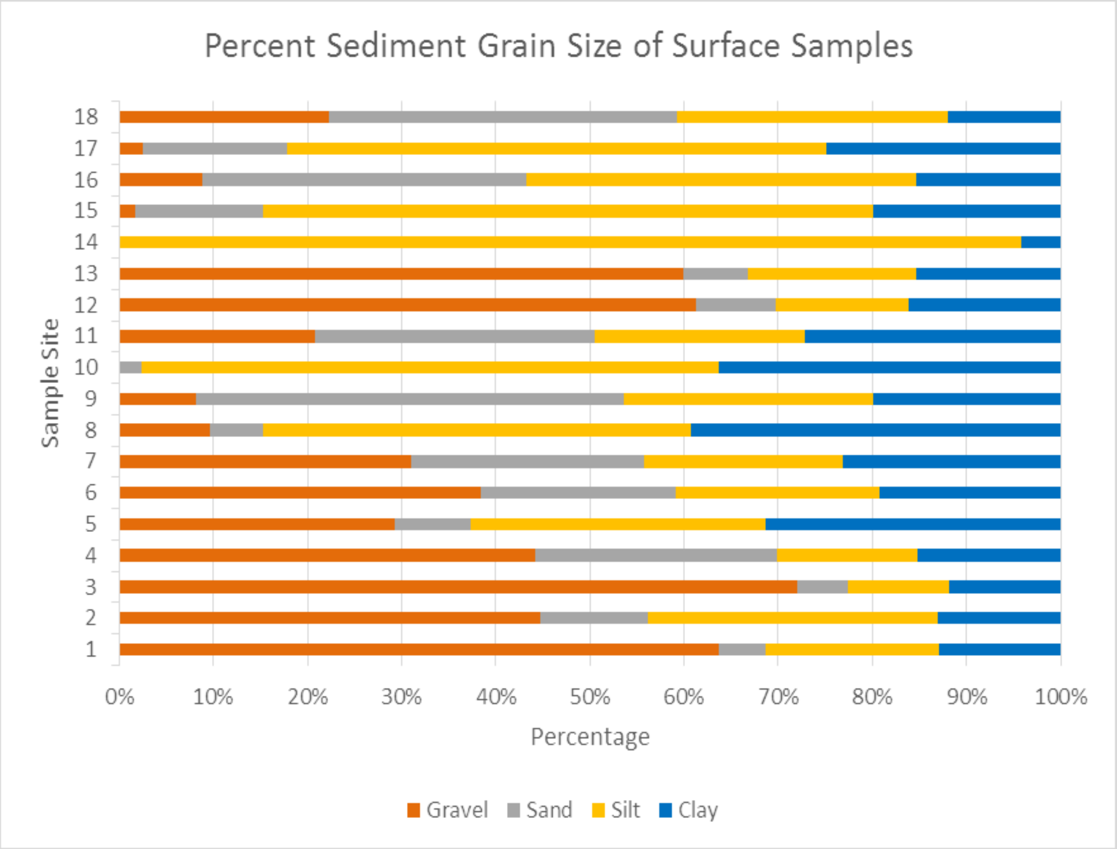


Figure 16. Grain size distribution in the surface samples (%)

Hierarchical cluster analysis was used to determine the correlation between total PCB levels, percent TOM, and percent mud. This cluster analysis grouped the sample sites into four clusters (figure 17). The green outlined cluster contains stations 13, 18, 15, and 17. The blue outlined cluster contains stations 14, 2, and 11, the orange outlined cluster contained 7, 16, 9, 3, and 12, and the red cluster contained 8, 5, 10, 4, 5, 6. There is no correlation between the total concentration of PCBs to their location in the study area. There are many factors that can influence the uptake of PCBs such as tidal exchange, grain size, and the hydrodynamic conditions within the estuary (Zhoa et al., 2012). This study examined grain size and TOM. While there is no strong correlation with location percent mud and percent TOM appear to have some effect on the samples, thereby generating the four (4) different clusters.

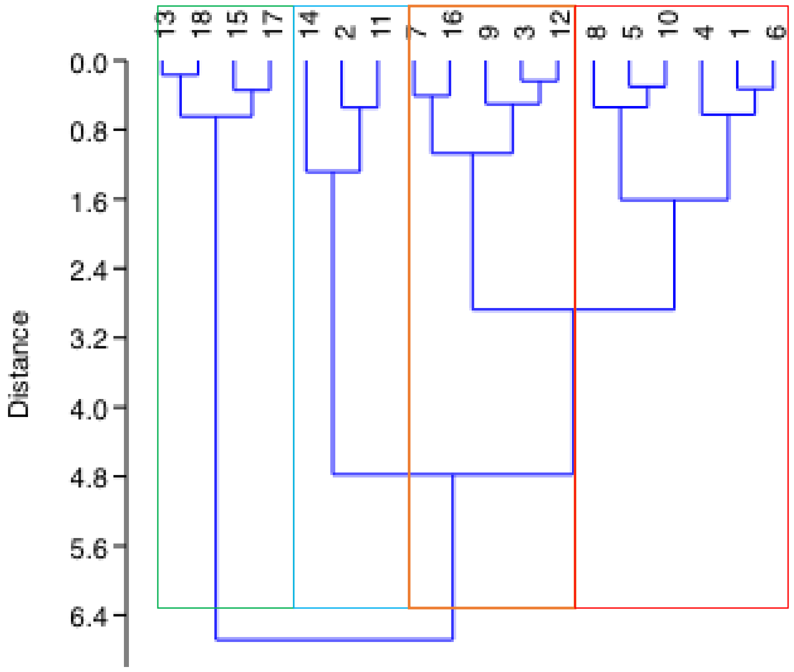


Figure 17. Dendrogram showing the distribution of total PCB, total organic matter, and the percent mud fraction.

CONCLUSIONS

The purpose of this study was to assess historical patterns and potential sources of PCBs in sediments in Jobos Bay, and the potential for their bioaccumulation in mangroves. This will help to better understand the fate and transport of PCBs within this National Estuarine Research Reserve. It is the first study to document the historical record of PCBs in the study area and the first to document bioaccumulation of these pollutants in sensitive mangrove ecosystems that provide valuable services.

The results of this research show; (i) levels of PCBs in a core taken from the study area; (ii) levels of PCBs in mangrove tissue from the study area; and, (iii) levels of PCBs in surface sediments from the study area.

Temporal data from the sediment core confirms that sediments are a good way to determine historical fluctuations of pollutants in the area. This can be applied to other pollutants in this area as well as other areas in Puerto Rico. The historical profile obtained from the core suggests that input of PCBs into the area has generally mirrored trends in use with fluctuations in PCBs over time correlating well with historical usage patterns. Results show that PCB concentrations rose steadily until around the early 1980s. It was during this time that the EPA banned the use of PCBs and other POPs. Results following this time frame show a gradual decrease in PCB levels. There are, however, spikes in PCB concentration which may be correlated to various anthropogenic sources like the leaching of the nearby landfill or pulsed inputs of contaminated sediments due to storm events. This study also demonstrated that there was not a strong correlation between the TOM of the sediments and PCB levels, with spikes in PCB concentrations when TOM remained relatively constant. Since salinity within the Bay had little fluctuation it is also likely not a contributing factor in PCB levels. However, it must be cautioned that due to a rain event immediately before sampling may have masked any true differences in salinities. Therefore, other factors must have played a part in PCB deposition and/or PCB movement and degradation at the different stations sampled. The fluctuations of PCB over time, coupled with no correlation between total PCBs and TOM, supports the use of sediments as a proxy in pollution profiles for the area.

Even though Jobos Bay appears to be a healthy estuary visually, the results of this study would result in a classification of the sediments as moderately to highly polluted. There was a general trend of higher total concentrations of PCBs closer to the shoreline and potential sources of PCBs. However, there was no apparent correlation between the concentrations of PCBs and congener profiles, nor was there a correlation between heavy and light PCBs with location. This supports the notion that multiple factors play a role in PCB distribution within the bay. One of those factors is the Bay's sedimentology. The sediment characteristics gathered from the grain size analysis and TOM content demonstrate the heterogeneous composition of the surface samples causing different adsorption rates. Atmospheric conditions may also affect the rate of PCB dispersal. Future research that focuses on the atmospheric deposition of PCBs in the area would aid in determining the fate of PCBs within this system.

Mangroves are a unique and vital coastal ecosystem. PCB concentrations in *R. mangle* tissue (roots, leaves and seeds) indicate bioaccumulation in these sensitive coastal plants. PCB input into the sediment is likely contributing to the levels of PCBs in the mangrove tissue. Several sites demonstrated a higher concentration of PCBs than the surrounding sediment, potentially demonstrating the mangrove's ability to uptake pollutants from surrounding sediments. Higher concentrations of PCBs were found in the leaves of the mangrove samples, suggesting translocation between mangrove roots and leaves. The plants' physiology may account for this difference within their tissues. Due to limitations on this study it is difficult to determine the effects that PCBs have on the mangrove plant itself. There are many contributing factors that affect the occurrence and speciation of this plant species. It would be beneficial to investigate how the input of PCBs and other pollutants affect the mangrove plant in a controlled laboratory setting.

The results of this research highlight the need for further investigations into other POPs, especially newer ones, and to determine their impact on the marine food web of Jobos Bay. Continual monitoring of POP pollution is also suggested for future research. These results are an important contribution to the data regarding levels of POPs in the Caribbean region and their potential impact on mangroves.

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APPENDIX A

Table A1. The concentrations of PCBs in each segment of the sediment core taken at Jobos Bay.

PCBs in sediment core														
	Core Segment =	**0.5**	**1.0**	**1.5**	**2.0**	**2.5**	**3.0**	**4.0**	**4.5**	**5.0**	**7.0**	**9.0**	**11.0**	**13.0**
2,4,6-TrCB	PCB30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.03	0.00
2,2',5-TrCB1	PCB18	0.00	1.70	1.34	0.61	0.40	0.00	1.23	0.40	0.49	0.22	0.09	0.14	0.49
2',3,5-TrCB	PCB34	0.79	0.00	0.00	0.00	0.00	0.00	0.06	0.00	0.04	0.00	0.00	0.14	0.03
2,4',5-TrCB	PCB31	0.00	0.00	0.00	0.00	0.00	0.00	0.11	0.00	0.20	0.00	0.00	0.04	0.00
2,4,4'-TrCB1	PCB28	0.88	0.00	0.00	3.27	0.25	0.00	1.44	0.00	0.22	0.00	0.00	0.04	0.06
2,3,4'-TrCB	PCB22	1.21	0.00	0.91	2.45	0.00	0.00	0.00	0.03	0.00	0.00	0.00	0.09	1.35
2,2',6,6'-TeCB	PCB54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.01
2,2',5,5'-TeCB1	PCB52	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.00	0.03	0.00	0.00	0.00	0.68
2,2',4,5'-TeCB	PCB49	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.65
2,3,4,6-TeCB	PCB62	0.57	0.00	0.12	0.00	0.00	0.00	0.00	0.00	0.07	0.00	0.01	0.01	0.58
2,2',3,5'-TeCB1	PCB44	0.76	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.07	0.00	0.00	0.03	1.56
2,2',3,4-TeCB/2,3,4',6-TeCB	PCB41/64	1.02	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.50	0.00	0.00	0.01	0.00
2,4,4',5-TeCB	PCB74	5.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.58	0.00	4.04
2,3',4',5-TeCB	PCB70	0.00	0.00	1.03	2.06	2.18	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.23
2,3,4,4'-TeCB	PCB60	0.49	0.60	0.00	0.00	1.04	0.00	0.00	0.00	0.00	0.00	0.25	0.00	0.06
2,3,3',4'-TeCB	PCB56	0.15	0.00	0.00	0.00	0.01	0.00	0.00	0.17	0.00	0.00	0.02	0.00	0.06
2,2',4,6,6'-PeCB	PCB104	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2,2',3,5',6-PeCB	PCB95	0.29	0.00	0.00	0.18	0.00	0.00	0.00	0.13	0.01	0.00	0.00	0.05	0.02
2,2',3,4',5-PeCB/2,2',4,5,5'-PeC	PCB90/101	0.20	0.02	0.22	0.23	0.01	0.00	0.24	0.06	0.00	0.00	0.06	0.03	0.14
2,2',4,4',5-PeCB	PCB99	0.04	0.00	0.00	0.00	0.17	0.00	0.00	0.15	0.00	0.00	0.00	0.02	0.01
2,2',3,4,5'-PeCB	PCB87	1.23	0.00	0.00	0.00	0.00	0.00	0.00	1.06	0.05	0.00	0.00	0.00	0.00
2,3,3',4',6-PeCB	PCB110	0.96	0.14	0.00	0.00	0.00	0.00	0.00	0.28	0.00	0.00	0.00	0.00	0.04
2,3,3',4',5-PeCB	PCB107	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2',3,4,4',5-PeCB2	PCB123	0.00	0.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.00

PCBs in sediment core														
	Core Segment =	**0.5**	**1.0**	**1.5**	**2.0**	**2.5**	**3.0**	**4.0**	**4.5**	**5.0**	**7.0**	**9.0**	**11.0**	**13.0**
2,3',4,4',5-PeCB1,2	PCB118	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2,3,4,4',5-PeCB1,2	PCB114	0.00	9.25	0.00	0.00	0.00	0.00	0.00	0.06	0.00	0.00	0.00	0.00	0.00
2,3,3'4,4'-PeCB1,2	PCB105	0.00	0.00	0.00	0.88	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.34	0.00
2,2',4,4',6,6'-HxCB	PCB155	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02
2,2',3,5,5',6-HxCB	PCB151	0.14	0.00	0.00	0.00	0.00	0.00	0.46	0.31	0.00	0.00	0.03	0.00	0.02
2,2',3,4',5',6-HxCB	PCB149	0.00	0.00	0.31	0.00	0.63	0.00	0.00	0.16	0.00	0.00	0.01	0.03	0.13
2,2',4,4',5,5'-HxCB1	PCB153	0.03	0.00	0.22	0.00	2.06	0.00	0.00	1.99	0.07	0.16	0.07	0.00	0.02
2,2',3,3',4,6'-HxCB	PCB132	0.71	0.00	0.00	0.00	0.77	0.00	0.00	2.18	0.00	0.00	0.08	0.00	0.00
2,2',3,4,5,5'-HxCB	PCB141	0.69	0.00	0.00	0.00	0.90	0.00	1.03	0.00	0.00	0.00	0.00	0.00	0.00
2,2',3,4,4',5'-HxCB1	PCB138	2.03	0.00	1.00	1.39	1.08	0.00	1.00	0.00	0.31	0.00	0.11	0.00	0.00
2,3,3',4,4',6-HxCB	PCB158	1.22	0.79	0.00	0.00	0.84	0.00	0.60	1.43	0.20	0.19	0.00	0.01	0.00
2,3,4,4',5,5'-HxCB2	PCB167	0.02	0.00	0.00	1.44	1.57	0.00	1.86	0.00	0.00	0.00	0.00	0.04	0.03
2,3,3',4,4',5-HxCB2	PCB156	4.48	0.15	0.00	0.00	0.88	0.00	0.00	1.15	0.32	0.00	0.09	0.01	0.68
2,3,3',4,4',5'-HxCB2	PCB157	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.00	0.00	0.08	0.01	0.33
2,3,3',4,4',5'-HxCB2	PCB188	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.02	0.00	0.03
2,2',3,4,5,5',6-HpCB1	PCB187	0.13	0.06	0.17	0.00	0.00	0.00	0.08	0.00	0.00	0.00	0.00	0.01	0.09
2,2',3,4,4',5',6-HpCB	PCB183	0.13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.04	0.00	0.04	0.08
2,2',3,3',4,5,6'-HpCB	pcb174	0.61	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2,2',3,4,4',5,5'-HpCB1	PCB180	0.18	0.00	0.00	0.00	0.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10
2,2',3,3',4,4',5-HpCB1	PCB170	0.00	0.00	0.09	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2,2',3,4',5,6,6'-HpCB	PCB189	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2,2',3,3',4,5,5',6'-OcCB	PCB199	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2,2',3,4,4',5,6,6'-OcCB	PCB204	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2,2',3,3',4,5,5',6-OcCB	PCB198	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2,2',3,4,4',5,5',6-OcCB	PCB203	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.03
2,2',3,3',4,4',5,5'-OcCB	PCB194	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00

Table A1. Cont.

PCBs in sediment core														
	Core Segment =	**15.0**	**17.0**	**19.0**	**21.0**	**23.0**	**25.0**	**27.0**	**29.0**	**31.0**	**33.0**	**35.0**	**37.0**	**39.0**
2,4,6-TrCB	PCB30	0.00	0.00	0.00	0.00	0.00	0.34	0.06	0.08	0.17	0.03	0.05	0.18	0.06
2,2',5-TrCB1	PCB18	0.03	0.02	0.04	0.17	0.03	0.45	40.17	9.96	0.31	7.42	0.26	0.67	0.14
2',3,5-TrCB	PCB34	0.00	0.00	0.00	0.00	0.00	3.23	0.84	0.33	0.57	0.12	0.41	0.89	0.23
2,4',5-TrCB	PCB31	0.00	0.00	0.00	0.00	0.00	3.95	1.44	0.23	0.85	0.06	0.45	2.09	0.13
2,4,4'-TrCB1	PCB28	0.00	0.07	0.00	0.01	0.00	3.88	1.60	0.14	0.30	0.05	0.50	0.71	0.19
2,3,4'-TrCB	PCB22	0.39	0.00	0.06	0.32	0.02	0.58	1.40	0.38	0.36	0.20	0.62	1.16	0.35
2,2',6,6'-TeCB	PCB54	0.01	0.00	0.00	0.00	0.00	0.01	0.02	0.15	0.16	0.00	0.15	1.59	0.02
2,2',5,5'-TeCB1	PCB52	0.11	0.00	0.00	0.00	0.00	0.13	0.08	0.21	0.19	0.02	0.18	0.39	0.78
2,2',4,5'-TeCB	PCB49	0.10	0.03	0.00	0.00	0.00	0.06	0.15	0.20	0.88	0.04	0.20	0.59	0.56
2,3,4,6-TeCB	PCB62	0.04	0.03	0.00	0.00	0.00	0.06	0.09	1.13	1.58	0.02	0.07	0.18	0.18
2,2',3,5'-TeCB1	PCB44	0.33	0.03	0.01	0.00	0.01	0.07	0.10	0.61	0.43	0.03	0.34	1.51	0.19
2,2',3,4-TeCB/2,3,4',6-TeCB	PCB41/64	0.03	0.00	0.00	0.00	0.00	0.13	0.29	0.56	0.69	0.07	0.48	0.87	0.33
2,4,4',5-TeCB	PCB74	1.84	0.00	0.00	0.00	0.00	0.35	0.65	0.85	0.53	0.04	0.46	1.61	0.38
2,3',4',5-TeCB	PCB70	0.00	0.00	0.00	0.00	0.16	0.30	0.38	0.63	0.40	0.04	0.47	0.97	0.41
2,3,4,4'-TeCB	PCB60	0.77	0.30	0.00	0.00	0.19	0.92	0.47	2.87	0.70	0.08	1.51	2.46	1.07
2,3,3',4'-TeCB	PCB56	0.00	0.00	0.02	0.00	0.04	0.15	0.18	0.84	0.95	0.07	0.28	0.75	0.44
2,2',4,6,6'-PeCB	PCB104	0.00	0.00	0.00	0.00	0.00	0.02	0.01	0.03	0.03	0.00	0.02	0.05	0.01
2,2',3,5',6-PeCB	PCB95	0.01	0.00	0.00	0.00	0.00	0.04	0.05	0.08	0.09	0.02	0.03	0.15	0.09
2,2',3,4',5-PeCB/2,2',4,5,5'-PeC	PCB90/101	0.00	0.00	0.00	0.00	0.01	0.04	0.07	0.06	0.07	0.03	0.26	0.12	0.05
2,2',4,4',5-PeCB	PCB99	0.13	0.00	0.00	0.00	0.08	0.16	0.14	0.64	0.38	0.08	0.55	0.77	0.09
2,2',3,4,5'-PeCB	PCB87	0.00	0.00	0.00	0.03	0.00	0.12	1.08	3.01	4.88	0.34	0.51	3.16	2.85
2,3,3',4',6-PeCB	PCB110	0.00	0.03	0.00	0.00	0.00	35.63	0.22	0.55	0.71	0.17	0.30	1.62	0.37
2,3,3',4',5-PeCB	PCB107	0.00	0.00	0.00	0.00	0.00	17.97	0.23	0.76	1.11	0.12	1.17	1.12	0.52
2',3,4,4',5-PeCB2	PCB123	0.00	0.00	0.00	0.38	0.02	0.26	1.79	5.24	8.35	0.17	1.65	2.91	4.62

PCBs in sediment core														
	Core Segment =	**15.0**	**17.0**	**19.0**	**21.0**	**23.0**	**25.0**	**27.0**	**29.0**	**31.0**	**33.0**	**35.0**	**37.0**	**39.0**
2,3',4,4',5-PeCB1,2	PCB118	0.00	0.00	0.00	0.00	0.00	32.94	1.94	1.63	1.47	0.18	1.73	1.25	1.22
2,3,4,4',5-PeCB1,2	PCB114	0.00	0.00	0.36	0.00	0.00	8.39	3.98	8.45	10.52	0.19	64.75	58.33	5.94
2,3,3'4,4'-PeCB1,2	PCB105	0.00	0.00	0.10	0.00	0.00	0.87	1.77	5.05	8.29	0.38	2.06	4.12	2.46
2,2',4,4',6,6'-HxCB	PCB155	0.00	0.01	0.01	0.00	0.00	0.01	0.02	0.01	0.00	0.00	0.01	0.01	0.01
2,2',3,5,5',6-HxCB	PCB151	0.00	0.00	0.06	0.08	0.00	0.03	0.18	0.70	0.18	0.01	0.04	0.73	0.10
2,2',3,4',5',6-HxCB	PCB149	0.00	0.00	0.03	0.13	0.00	0.09	0.07	0.20	0.25	0.04	0.39	0.22	0.12
2,2',4,4',5,5'-HxCB1	PCB153	0.20	0.04	0.12	0.00	0.03	0.08	0.10	0.06	0.11	0.02	0.13	0.20	0.45
2,2',3,3',4,6'-HxCB	PCB132	0.00	0.05	0.02	0.01	0.00	0.10	0.03	0.19	0.28	0.01	0.14	0.37	0.16
2,2',3,4,5,5'-HxCB	PCB141	0.00	0.00	0.00	0.00	0.00	0.03	0.04	0.21	0.29	0.01	0.22	0.42	0.14
2,2',3,4,4',5'-HxCB1	PCB138	0.32	0.00	0.15	0.13	0.04	0.28	0.12	0.38	0.45	0.06	0.25	1.83	0.30
2,3,3',4,4',6-HxCB	PCB158	0.38	0.00	0.00	0.09	0.00	0.16	0.06	0.10	0.22	0.03	0.07	0.49	0.16
2,3,4,4',5,5'-HxCB2	PCB167	0.28	0.00	0.00	0.00	0.00	0.11	0.17	0.38	0.48	0.02	0.12	0.68	0.31
2,3,3',4,4',5-HxCB2	PCB156	0.10	0.00	0.00	0.16	0.08	0.14	0.12	0.26	0.29	0.02	0.21	0.95	0.13
2,3,3',4,4',5'-HxCB2	PCB157	0.00	0.00	0.00	0.00	0.00	0.10	0.69	0.12	0.25	0.00	0.26	0.28	0.15
2,3,3',4,4',5'-HxCB2	PCB188	0.01	0.02	0.02	0.00	0.00	0.02	0.02	0.01	0.00	0.01	0.02	0.02	0.03
2,2',3,4,5,5',6-HpCB1	PCB187	0.03	0.04	0.06	0.02	0.01	0.02	0.09	0.03	0.04	0.01	0.02	0.02	0.04
2,2',3,4,4',5',6-HpCB	PCB183	0.01	0.00	0.03	0.02	0.00	0.03	0.10	0.03	0.02	0.01	0.02	0.02	0.07
2,2',3,3',4,5,6'-HpCB	pcb174	0.00	0.00	0.00	0.00	0.00	0.04	0.03	1.20	0.01	0.00	0.71	1.13	0.12
2,2',3,4,4',5,5'-HpCB1	PCB180	0.05	0.05	0.07	0.00	0.00	0.02	0.08	0.02	0.02	0.01	0.03	0.07	0.02
2,2',3,3',4,4',5-HpCB1	PCB170	0.00	0.00	0.00	0.00	0.00	0.04	0.02	0.04	0.02	0.01	0.10	0.21	0.16
2,2',3,4',5,6,6'-HpCB	PCB189	0.00	0.00	0.00	0.00	0.00	0.01	0.03	0.02	0.03	0.00	0.09	0.10	0.03
2,2',3,3',4,5,5',6'-OcCB	PCB199	0.00	0.00	0.00	0.00	0.00	0.45	0.31	0.28	0.25	0.23	0.69	0.54	0.78
2,2',3,4,4',5,6,6'-OcCB	PCB204	0.00	0.00	0.00	0.00	0.00	0.02	5.13	0.01	0.00	0.03	0.03	0.02	0.02
2,2',3,3',4,5,5',6-OcCB	PCB198	0.00	0.00	0.00	0.00	0.00	0.08	0.02	0.01	0.01	0.00	0.04	0.02	0.08
2,2',3,4,4',5,5',6-OcCB	PCB203	0.00	0.00	0.00	0.00	0.00	0.02	0.37	0.01	0.01	0.00	0.04	0.03	0.03
2,2',3,3',4,4',5,5'-OcCB	PCB194	0.00	0.00	0.00	0.00	0.00	0.03	0.04	0.02	0.01	0.01	0.09	0.58	0.08

Table A1. Cont.

PCBs in sediment core														
	Core Segment =	**41.0**	**43.0**	**45.0**	**47.0**	**49.0**	**51.0**	**53.0**	**55.0**	**57.0**	**59.0**	**61.0**	**63.0**	**65.0**
2,4,6-TrCB	PCB30	0.03	0.13	0.05	0.03	0.02	0.03	0.02	0.03	0.20	0.26	0.09	0.03	0.05
2,2',5-TrCB1	PCB18	0.30	10.86	6.24	3.33	0.06	0.04	0.35	4.62	1.09	0.19	13.12	15.62	0.09
2',3,5-TrCB	PCB34	0.21	0.53	0.11	0.06	0.11	0.05	0.39	0.16	1.82	0.56	0.45	0.13	0.30
2,4',5-TrCB	PCB31	0.17	0.73	0.57	0.22	0.01	0.11	0.21	0.43	4.84	0.28	0.98	0.12	0.23
2,4,4'-TrCB1	PCB28	0.37	0.82	0.64	0.24	0.04	0.03	0.24	0.47	5.35	0.31	1.09	0.13	0.13
2,3,4'-TrCB	PCB22	0.17	0.78	0.41	0.03	0.07	0.04	0.38	0.23	6.00	0.49	0.38	0.38	0.39
2,2',6,6'-TeCB	PCB54	0.00	0.03	0.09	0.01	0.01	0.01	0.02	0.01	0.02	0.03	0.04	0.01	0.04
2,2',5,5'-TeCB1	PCB52	0.03	0.13	0.59	0.06	0.02	0.05	0.39	0.08	7.71	0.20	0.20	0.09	0.08
2,2',4,5'-TeCB	PCB49	0.01	0.12	0.45	0.04	0.02	0.03	0.28	0.04	0.13	0.17	0.21	0.09	0.09
2,3,4,6-TeCB	PCB62	0.03	0.26	0.79	0.04	0.02	0.07	0.06	0.13	0.77	0.16	0.17	0.08	0.13
2,2',3,5'-TeCB1	PCB44	0.04	0.35	0.21	0.09	0.05	0.07	0.11	0.09	0.65	0.69	0.17	0.11	0.24
2,2',3,4-TeCB/2,3,4',6-TeCB	PCB41/64	0.02	0.48	0.33	0.18	0.05	0.16	0.25	0.21	0.38	0.62	0.27	0.22	0.28
2,4,4',5-TeCB	PCB74	0.03	0.37	0.18	0.16	0.07	0.12	1.18	0.95	1.86	0.67	1.06	0.32	0.54
2,3',4',5-TeCB	PCB70	0.18	0.48	0.33	0.09	0.05	0.14	0.18	0.86	1.88	0.91	1.07	0.18	0.26
2,3,4,4'-TeCB	PCB60	0.23	2.27	0.35	0.12	0.09	0.23	1.09	0.80	1.74	0.59	0.44	0.34	0.58
2,3,3',4'-TeCB	PCB56	0.07	0.56	0.44	0.18	0.02	0.12	0.29	0.23	0.45	0.85	0.24	0.13	0.24
2,2',4,6,6'-PeCB	PCB104	0.00	0.02	0.02	0.01	0.01	0.01	0.00	0.00	0.01	0.03	0.02	0.02	0.01
2,2',3,5',6-PeCB	PCB95	0.01	0.06	0.06	0.07	0.04	0.08	0.05	0.05	0.08	0.16	0.04	0.07	0.04
2,2',3,4',5-PeCB/2,2',4,5,5'-PeC	PCB90/101	0.01	0.06	0.05	0.01	0.03	0.04	0.06	0.02	0.05	0.05	0.04	0.04	0.03
2,2',4,4',5-PeCB	PCB99	0.06	0.59	0.37	0.13	0.05	0.15	0.19	0.25	0.23	0.74	0.27	0.04	0.22
2,2',3,4,5'-PeCB	PCB87	0.13	0.52	1.18	0.12	0.19	0.61	1.63	1.02	2.18	3.87	0.77	0.06	1.09
2,3,3',4',6-PeCB	PCB110	0.32	0.56	0.43	0.12	0.13	1.07	0.20	0.57	1.24	0.69	0.26	0.10	0.22
2,3,3',4',5-PeCB	PCB107	0.11	0.48	0.68	0.16	0.08	0.15	0.49	0.13	0.38	1.08	0.38	0.11	0.17
2',3,4,4',5-PeCB2	PCB123	0.47	4.80	3.75	1.14	0.13	1.20	0.45	1.47	3.65	6.25	2.03	0.09	1.74

PCBs in sediment core

	Core Segment =	41.0	43.0	45.0	47.0	49.0	51.0	53.0	55.0	57.0	59.0	61.0	63.0	65.0
2,3',4,4',5-PeCB1,2	PCB118	0.52	5.14	4.01	1.32	0.17	1.30	0.47	1.58	0.70	1.05	2.25	0.16	1.93
2,3,4,4',5-PeCB1,2	PCB114	0.93	6.01	4.02	1.75	2.05	1.71	3.21	2.19	6.27	15.04	7.15	6.70	2.57
2,3,3'4,4'-PeCB1,2	PCB105	0.47	2.62	2.81	0.63	0.20	1.34	2.51	0.87	2.13	3.70	1.19	0.51	1.69
2,2',4,4',6,6'-HxCB	PCB155	0.00	0.01	0.01	0.00	0.01	0.01	0.01	0.00	0.01	0.01	0.01	0.01	0.01
2,2',3,5,5',6-HxCB	PCB151	0.06	0.55	0.13	0.03	0.03	0.02	0.30	0.05	0.40	0.89	0.07	0.05	0.03
2,2',3,4',5',6-HxCB	PCB149	0.02	0.13	0.13	0.02	0.02	0.06	0.04	0.04	0.10	0.19	0.05	0.11	0.10
2,2',4,4',5,5'-HxCB1	PCB153	0.03	1.05	0.32	0.22	0.05	0.03	0.08	0.06	0.13	0.28	0.09	0.09	0.07
2,2',3,3',4,6'-HxCB	PCB132	0.01	0.16	0.39	0.03	0.02	0.03	0.04	0.03	0.07	0.22	0.04	0.04	0.08
2,2',3,4,5,5'-HxCB	PCB141	0.03	0.23	0.11	0.11	0.03	0.07	0.13	0.03	0.07	0.19	0.08	0.02	0.07
2,2',3,4,4',5'-HxCB1	PCB138	0.04	0.26	0.26	0.06	0.07	0.31	0.24	0.16	0.48	0.42	0.31	0.14	0.15
2,3,3',4,4',6-HxCB	PCB158	0.03	0.20	0.14	0.05	0.02	0.05	0.11	0.08	0.19	0.14	0.09	0.03	0.07
2,3,4,4',5,5'-HxCB2	PCB167	0.02	0.32	0.28	0.05	0.03	0.08	0.17	0.12	0.24	0.16	0.09	0.06	0.09
2,3,3',4,4',5-HxCB2	PCB156	0.06	0.10	0.21	0.04	0.03	0.09	0.03	0.03	0.21	0.38	0.14	0.01	0.03
2,3,3',4,4',5'-HxCB2	PCB157	0.02	0.11	0.10	0.04	0.01	0.02	0.13	0.03	0.13	0.13	0.07	0.02	0.05
2,3,3',4,4',5'-HxCB2	PCB188	0.01	0.01	0.01	0.01	0.02	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01
2,2',3,4,5,5',6-HpCB1	PCB187	0.01	0.02	0.02	0.02	0.01	0.02	0.02	0.02	0.01	0.03	0.02	0.04	0.03
2,2',3,4,4',5',6-HpCB	PCB183	0.02	0.02	0.03	0.02	0.01	0.02	0.02	0.01	0.02	0.03	0.02	0.04	0.60
2,2',3,3',4,5,6'-HpCB	pcb174	0.24	0.05	0.04	0.02	0.03	0.01	0.01	0.01	0.02	0.39	0.03	0.05	0.02
2,2',3,4,4',5,5'-HpCB1	PCB180	0.01	0.02	0.02	0.03	0.03	0.05	0.02	0.01	0.02	0.05	0.05	0.10	0.04
2,2',3,3',4,4',5-HpCB1	PCB170	0.02	0.02	0.03	0.02	0.02	0.02	0.01	0.02	0.01	0.06	0.06	0.02	0.32
2,2',3,4',5,6,6'-HpCB	PCB189	0.02	0.02	0.03	0.01	0.01	0.01	0.01	0.01	0.05	0.02	0.04	0.01	0.02
2,2',3,3',4,5,5',6'-OcCB	PCB199	0.23	0.23	0.59	0.40	0.57	0.41	0.14	0.15	0.39	0.29	0.56	0.43	0.45
2,2',3,4,4',5,6,6'-OcCB	PCB204	0.00	0.01	0.57	0.08	0.24	0.01	0.12	0.02	1.02	0.01	0.01	4.44	24.09
2,2',3,3',4,5,5',6-OcCB	PCB198	0.01	0.01	0.01	0.01	0.00	0.03	0.01	0.01	0.01	0.01	0.02	0.02	0.02
2,2',3,4,4',5,5',6-OcCB	PCB203	0.01	0.02	0.01	0.01	0.04	0.01	0.01	0.00	0.02	0.02	0.02	0.07	0.53
2,2',3,3',4,4',5,5'-OcCB	PCB194	0.01	0.02	0.03	0.02	0.02	0.02	0.01	0.01	0.02	0.07	0.03	0.03	0.40

Table A1. Cont.

PCBs in sediment core												
	Core Segment =	**66.5**	**68.5**	**70.5**	**72.5**	**74.5**	**76.5**	**78.5**	**80.5**	**82.5**	**83.5**	**85.5**
2,4,6-TrCB	PCB30	0.01	0.20	0.09	0.00	0.00	0.00	0.22	0.66	0.14	0.15	0.27
2,2',5-TrCB1	PCB18	0.03	1.16	2.28	0.00	0.00	0.00	1.36	1.16	1.89	0.72	3.43
2',3,5-TrCB	PCB34	0.06	0.28	0.46	0.00	0.00	0.00	0.71	11.77	2.27	4.41	1.22
2,4',5-TrCB	PCB31	0.04	0.35	0.57	0.00	0.00	0.00	0.66	6.25	1.28	0.54	0.94
2,4,4'-TrCB1	PCB28	0.05	0.20	0.48	0.00	0.00	0.00	0.68	3.48	1.32	1.23	0.86
2,3,4'-TrCB	PCB22	0.06	0.43	1.40	0.00	0.00	0.00	0.84	0.87	0.71	0.66	1.57
2,2',6,6'-TeCB	PCB54	0.01	0.07	0.03	0.00	0.00	0.00	0.02	0.09	0.04	0.02	0.05
2,2',5,5'-TeCB1	PCB52	0.25	0.51	0.71	0.00	0.00	0.00	0.19	0.28	0.85	0.23	0.79
2,2',4,5'-TeCB	PCB49	0.21	0.21	0.68	0.00	0.00	0.00	0.08	0.32	0.09	0.30	0.21
2,3,4,6-TeCB	PCB62	0.18	0.35	0.58	0.00	0.00	0.00	0.27	0.32	1.56	0.58	1.73
2,2',3,5'-TeCB1	PCB44	0.08	0.50	0.10	0.00	0.00	0.00	0.18	1.19	0.19	0.21	0.68
2,2',3,4-TeCB/2,3,4',6-TeCB	PCB41/64	0.12	2.34	0.37	0.00	0.00	0.00	0.70	0.63	0.09	0.94	0.40
2,4,4',5-TeCB	PCB74	0.21	9.08	0.80	0.00	0.00	0.00	4.91	1.74	0.82	7.86	2.56
2,3',4',5-TeCB	PCB70	0.19	7.78	0.33	0.00	0.00	0.00	1.05	1.53	0.76	2.54	1.24
2,3,4,4'-TeCB	PCB60	0.33	0.53	1.58	0.00	0.00	0.00	1.24	1.22	1.00	2.20	0.47
2,3,3',4'-TeCB	PCB56	0.06	0.42	0.44	0.00	0.00	0.00	0.15	0.95	0.45	0.70	0.52
2,2',4,6,6'-PeCB	PCB104	0.01	0.04	0.01	0.00	0.00	0.00	0.02	0.11	0.00	0.01	0.01
2,2',3,5',6-PeCB	PCB95	0.04	0.23	0.32	0.00	0.00	0.00	0.07	0.04	0.18	0.11	0.17
2,2',3,4',5-PeCB/2,2',4,5,5'-PeC	PCB90/101	0.04	0.31	0.52	0.00	0.00	0.00	0.10	0.09	0.27	0.36	0.09
2,2',4,4',5-PeCB	PCB99	0.08	0.61	0.51	0.00	0.00	0.00	0.33	0.21	0.15	0.51	0.17
2,2',3,4,5'-PeCB	PCB87	0.05	4.29	0.68	0.00	0.00	0.00	0.35	1.47	1.29	0.32	0.88
2,3,3',4',6-PeCB	PCB110	0.07	1.26	0.80	0.00	0.00	0.00	13.81	6.82	0.27	0.30	0.36
2,3,3',4',5-PeCB	PCB107	0.09	1.86	0.25	0.00	0.00	0.00	0.71	0.36	0.42	0.30	0.29
2',3,4,4',5-PeCB2	PCB123	0.09	4.31	0.45	0.00	0.00	0.00	0.76	2.40	3.07	0.32	0.34

PCBs in sediment core												
	Core Segment =	**66.5**	**68.5**	**70.5**	**72.5**	**74.5**	**76.5**	**78.5**	**80.5**	**82.5**	**83.5**	**85.5**
2,3',4,4',5-PeCB1,2	PCB118	0.10	3.96	1.24	0.00	0.00	0.00	0.78	2.61	3.34	1.34	0.56
2,3,4,4',5-PeCB1,2	PCB114	2.72	4.04	20.98	0.00	0.00	0.00	5.35	6.34	2.53	2.53	10.67
2,3,3'4,4'-PeCB1,2	PCB105	2.51	3.14	3.69	0.00	0.00	0.00	0.62	3.36	2.28	1.39	2.21
2,2',4,4',6,6'-HxCB	PCB155	0.01	0.03	0.13	0.00	0.00	0.00	0.02	0.01	0.28	0.00	0.01
2,2',3,5,5',6-HxCB	PCB151	0.04	0.18	0.31	0.00	0.00	0.00	0.15	0.34	0.90	18.62	0.06
2,2',3,4',5',6-HxCB	PCB149	0.03	6.03	0.09	0.00	0.00	0.00	0.05	0.82	0.54	75.20	0.30
2,2',4,4',5,5'-HxCB1	PCB153	0.07	4.99	1.30	0.00	0.00	0.00	0.04	0.20	0.33	0.41	0.22
2,2',3,3',4,6'-HxCB	PCB132	0.08	0.22	1.63	0.00	0.00	0.00	0.08	0.34	0.42	0.25	0.23
2,2',3,4,5,5'-HxCB	PCB141	0.01	0.23	0.22	0.00	0.00	0.00	0.05	0.19	1.10	0.09	0.27
2,2',3,4,4',5'-HxCB1	PCB138	0.09	1.06	1.42	0.00	0.00	0.00	0.38	1.46	1.70	1.30	0.33
2,3,3',4,4',6-HxCB	PCB158	0.07	0.16	0.99	0.00	0.00	0.00	0.03	0.54	0.13	0.12	0.22
2,3,4,4',5,5'-HxCB2	PCB167	0.05	0.39	0.17	0.00	0.00	0.00	0.16	0.14	0.73	0.23	0.10
2,3,3',4,4',5-HxCB2	PCB156	0.10	0.28	0.24	0.00	0.00	0.00	0.18	0.31	0.44	0.82	0.08
2,3,3',4,4',5'-HxCB2	PCB157	0.02	1.46	0.51	0.00	0.00	0.00	0.21	0.20	0.41	1.47	0.07
2,3,3',4,4',5'-HxCB2	PCB188	0.01	104.72	0.02	0.00	0.00	0.00	0.02	43.63	0.01	0.66	0.04
2,2',3,4,5,5',6-HpCB1	PCB187	0.04	0.27	0.30	0.00	0.00	0.00	0.04	0.24	0.11	3.17	0.08
2,2',3,4,4',5',6-HpCB	PCB183	0.47	5.10	0.31	0.00	0.00	0.00	0.04	0.19	0.12	4.11	0.04
2,2',3,3',4,5,6'-HpCB	pcb174	0.03	7.73	0.14	0.00	0.00	0.00	0.03	0.04	0.13	0.06	0.11
2,2',3,4,4',5,5'-HpCB1	PCB180	0.06	0.33	0.48	0.00	0.00	0.00	0.10	0.80	0.36	3.23	0.15
2,2',3,3',4,4',5-HpCB1	PCB170	0.07	28.44	0.25	0.00	0.00	0.00	0.08	30.28	0.10	17.43	0.05
2,2',3,4',5,6,6'-HpCB	PCB189	0.13	81.01	0.03	0.00	0.00	0.00	0.02	2222.24	0.16	611.34	0.06
2,2',3,3',4,5,5',6'-OcCB	PCB199	0.47	0.85	0.30	0.00	0.00	0.00	0.46	0.68	0.16	0.98	0.44
2,2',3,4,4',5,6,6'-OcCB	PCB204	20.46	26.21	2.08	0.00	0.00	0.00	0.02	0.03	0.54	136.83	0.02
2,2',3,3',4,5,5',6-OcCB	PCB198	0.02	1.07	0.14	0.00	0.00	0.00	0.02	0.50	0.00	66.22	0.03
2,2',3,4,4',5,5',6-OcCB	PCB203	0.19	0.85	0.11	0.00	0.00	0.00	0.02	1.23	0.06	38.44	0.03
2,2',3,3',4,4',5,5'-OcCB	PCB194	0.04	3.83	0.65	0.00	0.00	0.00	0.03	26.76	0.27	85.73	0.21

Table A2. **T**he concentrations of individual congeners of PCBs measured in the surface sediments collected in Jobos Bay

PCBs in surface sediments											
	Station =	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**
2,4,6-TrCB	PCB30	2.36	2.38	0.05	0.63	0.09	0.97	0.01	0.25	0.13	0.15
2,2',5-TrCB1	PCB18	1.66	9.55	0.12	0.80	0.61	24.13	0.17	26.68	0.77	13.52
2',3,5-TrCB	PCB34	6.45	11.01	0.31	103.03	0.87	0.66	0.04	0.89	0.23	26.84
2,4',5-TrCB	PCB31	2.98	2.19	0.36	1.45	0.55	3.61	0.15	1.82	0.27	1.52
2,4,4'-TrCB1	PCB28	3.07	2.26	0.15	2.24	0.47	3.77	0.17	1.98	0.34	1.32
2,3,4'-TrCB	PCB22	4.67	18.43	0.33	56.81	0.59	1.46	0.04	4.05	0.29	1.55
2,2',6,6'-TeCB	PCB54	0.63	1.75	0.01	0.03	0.43	0.03	0.00	0.02	0.02	0.03
2,2',5,5'-TeCB1	PCB52	9.08	5.45	0.85	0.12	0.32	0.17	0.02	1.20	0.04	0.08
2,2',4,5'-TeCB	PCB49	10.25	5.25	0.12	0.37	0.40	0.10	0.01	3.09	0.14	0.12
2,3,4,6-TeCB	PCB62	13.71	1.85	0.20	0.15	0.46	0.58	0.00	0.92	0.17	0.09
2,2',3,5'-TeCB1	PCB44	1.81	3.41	0.13	0.12	0.49	0.21	0.01	0.30	0.13	0.08
2,2',3,4-TeCB/2,3,4',6-TeCB	PCB41/64	3.19	2.62	0.39	0.54	0.41	0.31	0.00	0.69	0.18	0.23
2,4,4',5-TeCB	PCB74	11.16	0.96	3.50	0.90	2.62	0.26	0.04	2.03	0.24	0.29
2,3',4',5-TeCB	PCB70	9.98	0.86	1.47	0.79	1.96	0.35	0.02	2.37	0.18	0.14
2,3,4,4'-TeCB	PCB60	14.20	9.93	1.09	0.87	3.04	0.36	0.12	4.31	0.77	0.47
2,3,3',4'-TeCB	PCB56	5.54	1.17	0.13	0.27	0.78	0.25	0.01	1.16	0.20	0.24
2,2',4,6,6'-PeCB	PCB104	0.08	2.15	0.01	0.02	0.01	8.68	0.01	0.02	0.02	0.01
2,2',3,5',6-PeCB	PCB95	9.23	7.14	0.44	0.09	0.22	0.16	0.01	0.16	0.09	0.09
2,2',3,4',5-PeCB/2,2',4,5,5'-PeCB1	PCB90/101	4.44	7.47	0.30	0.10	0.31	5.52	0.00	0.24	0.31	0.10
2,2',4,4',5-PeCB	PCB99	8.50	3.29	0.24	0.36	0.32	10.40	0.01	0.24	0.30	0.10
2,2',3,4,5'-PeCB	PCB87	9.98	14.22	0.66	1.84	0.92	2.84	0.02	0.29	0.42	0.27
2,3,3',4',6-PeCB	PCB110	12.53	5.33	1.61	1.02	0.70	0.39	0.14	0.35	0.69	0.62
2,3,3',4',5-PeCB	PCB107	5.11	21.94	0.70	0.46	0.74	8.55	0.04	0.56	1.16	1.87
2',3,4,4',5-PeCB2	PCB123	1.95	8.92	0.67	1.21	0.98	7.88	0.04	0.49	0.82	1.72
2,3',4,4',5-PeCB1,2	PCB118	7.42	9.92	3.37	3.90	1.37	12.10	0.01	1.10	0.89	1.60
2,3,4,4',5-PeCB1,2	PCB114	35.30	24.03	5.74	3.83	66.40	6.27	0.48	11.42	21.76	5.38
2,3,3'4,4'-PeCB1,2	PCB105	53.03	6.69	5.85	1.30	8.83	4.09	0.02	11.05	1.00	2.05
2,2',4,4',6,6'-HxCB	PCB155	0.03	1.57	0.02	0.01	0.01	5.14	0.00	0.01	0.05	0.04
2,2',3,5,5',6-HxCB	PCB151	2.75	2.06	0.25	0.06	0.12	0.04	0.01	0.06	0.08	0.09
2,2',3,4',5',6-HxCB	PCB149	1.54	9.02	0.44	0.16	0.54	5.26	0.01	0.05	0.12	0.14
2,2',4,4',5,5'-HxCB1	PCB153	7.76	1.84	0.12	0.16	0.35	0.18	0.00	0.26	0.09	0.13
2,2',3,3',4,6'-HxCB	PCB132	9.97	3.47	0.14	0.06	0.49	0.15	0.01	0.27	0.07	0.19
2,2',3,4,5,5'-HxCB	PCB141	2.13	2.89	0.13	0.13	0.57	0.43	0.01	0.12	0.26	0.06
2,2',3,4,4',5'-HxCB1	PCB138	0.93	5.13	0.97	0.39	1.37	0.41	0.00	0.77	0.61	0.80
2,3,3',4,4',6-HxCB	PCB158	6.09	1.69	0.64	0.13	0.93	0.10	0.00	0.22	0.09	0.56
2,3,4,4',5,5'-HxCB2	PCB167	0.86	2.65	0.11	0.06	0.47	4.32	0.00	0.15	0.18	0.06
2,3,3',4,4',5-HxCB2	PCB156	1.14	3.00	0.20	0.24	0.47	0.24	0.01	0.08	0.20	0.12
2,3,3',4,4',5'-HxCB2	PCB157	0.51	2.33	0.08	1.76	2.37	0.55	0.00	0.06	0.17	0.28
2,3,3',4,4',5'-HxCB2	PCB188	2.36	36.39	0.11	0.06	0.11	0.19	0.02	0.08	0.13	0.20
2,2',3,4,5,5',6-HpCB1	PCB187	2.37	102.89	0.18	0.06	2.54	0.31	0.02	0.15	0.07	0.10
2,2',3,4,4',5',6-HpCB	PCB183	0.97	109.30	0.22	0.13	0.22	0.44	0.02	0.41	0.38	0.32
2,2',3,3',4,5,6'-HpCB	pcb174	7.02	142.86	0.16	0.16	0.40	0.35	0.03	0.36	0.07	1.64
2,2',3,4,4',5,5'-HpCB1	PCB180	4.29	35.77	0.24	0.07	0.32	0.11	0.02	0.34	0.28	0.06
2,2',3,3',4,4',5-HpCB1	PCB170	3.53	43.15	0.46	0.13	0.57	0.12	6.01	0.14	18.40	0.05
2,2',3,4',5,6,6'-HpCB	PCB189	3.23	1.76	0.05	0.08	2.85	49.31	0.01	0.19	2.73	0.03
2,2',3,3',4,5,5',6'-OcCB	PCB199	3.46	79.38	0.53	0.38	0.36	0.81	0.13	0.39	0.84	1.76
2,2',3,4,4',5,6,6'-OcCB	PCB204	3.91	1.99	0.01	1.78	6.94	14.75	0.01	0.00	0.02	1.20
2,2',3,3',4,5,5',6-OcCB	PCB198	46.48	3.38	0.02	0.01	0.02	34.29	0.01	0.07	2.62	0.52
2,2',3,4,4',5,5',6-OcCB	PCB203	1.00	0.38	0.07	0.02	0.03	17.96	0.40	0.81	14.83	0.50
2,2',3,3',4,4',5,5'-OcCB	PCB194	0.78	449.09	0.04	0.20	0.44	1.29	0.01	0.07	3.27	19.78
	ΣPCBs	**361.37**	**1232.13**	**33.96**	**189.46**	**117.32**	**240.84**	**8.34**	**82.70**	**77.13**	**89.09**

Table A2 Cont.

PCBs in surface sediments									
	Station =	**11**	**12**	**13**	**14**	**15**	**16**	**17**	**18**
2,4,6-TrCB	PCB30	1.13	0.25	0.00	3.60	0.00	0.03	0.00	0.00
2,2',5-TrCB1	PCB18	0.49	0.41	0.03	12.91	0.00	11.49	0.00	0.00
2',3,5-TrCB	PCB34	0.47	1.06	0.02	53.40	0.00	0.17	0.00	0.00
2,4',5-TrCB	PCB31	2.66	1.28	0.00	7.96	0.00	0.17	0.00	0.00
2,4,4'-TrCB1	PCB28	2.75	0.59	0.00	7.67	0.00	0.14	0.00	0.01
2,3,4'-TrCB	PCB22	1.33	1.76	0.02	98.65	0.00	0.42	0.00	0.00
2,2',6,6'-TeCB	PCB54	0.10	0.19	0.00	0.57	0.00	0.01	0.00	0.00
2,2',5,5'-TeCB1	PCB52	0.30	0.22	0.00	1.17	0.00	0.02	0.00	0.00
2,2',4,5'-TeCB	PCB49	0.22	0.20	0.00	0.89	0.00	0.05	0.00	0.00
2,3,4,6-TeCB	PCB62	0.13	1.56	0.00	1.72	0.00	0.08	0.00	0.00
2,2',3,5'-TeCB1	PCB44	0.22	0.32	0.00	2.02	0.00	0.09	0.00	0.00
2,2',3,4-TeCB/2,3,4',6-TeCB	PCB41/64	0.07	0.69	0.00	2.75	0.20	0.05	0.00	0.00
2,4,4',5-TeCB	PCB74	1.90	1.61	0.26	5.60	0.00	0.19	0.00	0.00
2,3',4',5-TeCB	PCB70	1.73	1.26	0.00	3.10	0.00	0.16	0.09	0.00
2,3,4,4'-TeCB	PCB60	1.42	1.72	0.00	8.35	0.00	0.25	0.09	0.00
2,3,3',4'-TeCB	PCB56	0.38	0.72	0.00	4.65	0.00	0.05	0.01	0.09
2,2',4,6,6'-PeCB	PCB104	0.72	0.46	0.00	8.31	0.00	0.01	0.00	0.00
2,2',3,5',6-PeCB	PCB95	0.24	0.20	0.00	0.85	0.00	0.02	0.04	0.00
2,2',3,4',5-PeCB/2,2',4,5,5'-PeCB1	PCB90/101	3.04	0.56	0.00	1.53	0.00	0.06	0.00	0.02
2,2',4,4',5-PeCB	PCB99	0.43	1.01	0.00	2.28	0.00	0.07	0.04	0.00
2,2',3,4,5'-PeCB	PCB87	2.09	5.45	0.00	13.84	0.02	0.15	0.29	0.01
2,3,3',4',6-PeCB	PCB110	0.62	0.60	0.00	2.46	0.00	0.16	0.00	0.00
2,3,3',4',5-PeCB	PCB107	95.14	1.63	0.00	6.33	0.00	0.11	0.00	0.00
2',3,4,4',5-PeCB2	PCB123	106.52	8.52	0.00	6.09	0.00	0.12	0.04	0.03
2,3',4,4',5-PeCB1,2	PCB118	142.02	1.49	0.00	6.52	0.03	0.10	0.00	0.03
2,3,4,4',5-PeCB1,2	PCB114	104.96	3.26	0.00	60.84	0.00	2.35	0.00	0.00
2,3,3'4,4'-PeCB1,2	PCB105	104.27	2.00	0.00	46.83	0.00	0.24	0.00	0.00
2,2',4,4',6,6'-HxCB	PCB155	0.22	0.28	0.00	0.98	0.00	0.01	0.00	0.00
2,2',3,5,5',6-HxCB	PCB151	0.23	2.11	0.09	2.32	0.00	0.14	0.00	0.00
2,2',3,4',5',6-HxCB	PCB149	0.14	0.61	0.00	2.08	0.04	0.02	0.06	0.02
2,2',4,4',5,5'-HxCB1	PCB153	0.29	1.25	0.00	2.72	0.00	0.13	0.09	0.15
2,2',3,3',4,6'-HxCB	PCB132	0.14	1.52	0.00	1.93	0.00	0.15	0.00	0.00
2,2',3,4,5,5'-HxCB	PCB141	0.17	0.42	0.06	2.68	0.00	0.03	0.00	0.00
2,2',3,4,4',5'-HxCB1	PCB138	0.11	0.97	0.33	6.44	0.00	0.15	0.04	0.00
2,3,3',4,4',6-HxCB	PCB158	0.07	0.35	0.00	4.56	0.13	0.10	0.00	0.18
2,3,4,4',5,5'-HxCB2	PCB167	0.10	0.82	0.00	1.19	0.00	0.08	0.00	0.00
2,3,3',4,4',5-HxCB2	PCB156	0.19	0.72	0.18	1.14	0.00	0.02	0.00	0.12
2,3,3',4,4',5'-HxCB2	PCB157	0.13	2.54	0.00	2.17	0.00	0.03	0.00	0.00
2,3,3',4,4',5'-HxCB2	PCB188	1.05	0.03	0.00	0.35	0.00	0.02	0.00	0.00
2,2',3,4,5,5',6-HpCB1	PCB187	0.38	0.21	0.06	0.66	0.00	0.04	0.00	0.00
2,2',3,4,4',5',6-HpCB	PCB183	5.94	0.16	0.06	0.88	0.00	0.04	0.02	0.00
2,2',3,3',4,5,6'-HpCB	pcb174	8.70	0.09	0.00	1.98	0.00	0.05	0.00	0.00
2,2',3,4,4',5,5'-HpCB1	PCB180	1.43	0.17	0.10	1.41	0.00	0.08	0.00	0.00
2,2',3,3',4,4',5-HpCB1	PCB170	5.71	0.16	0.00	2.18	0.00	0.29	0.00	0.00
2,2',3,4',5,6,6'-HpCB	PCB189	9.52	0.06	0.00	3.15	0.00	0.02	0.00	0.00
2,2',3,3',4,5,5',6'-OcCB	PCB199	19.09	0.23	0.00	1.56	0.00	0.58	0.00	0.00
2,2',3,4,4',5,6,6'-OcCB	PCB204	0.21	1.84	0.00	3.70	0.00	0.01	0.00	0.00
2,2',3,3',4,5,5',6-OcCB	PCB198	0.72	0.03	0.00	0.09	0.00	0.06	0.00	0.00
2,2',3,4,4',5,5',6-OcCB	PCB203	356.99	0.02	0.00	0.14	0.00	1.49	0.00	0.00
2,2',3,3',4,4',5,5'-OcCB	PCB194	1.18	0.33	0.00	0.33	0.00	4.63	0.00	0.00
	ΣPCBs	**988.07**	**53.89**	**1.20**	**415.49**	**0.42**	**24.90**	**0.80**	**0.65**

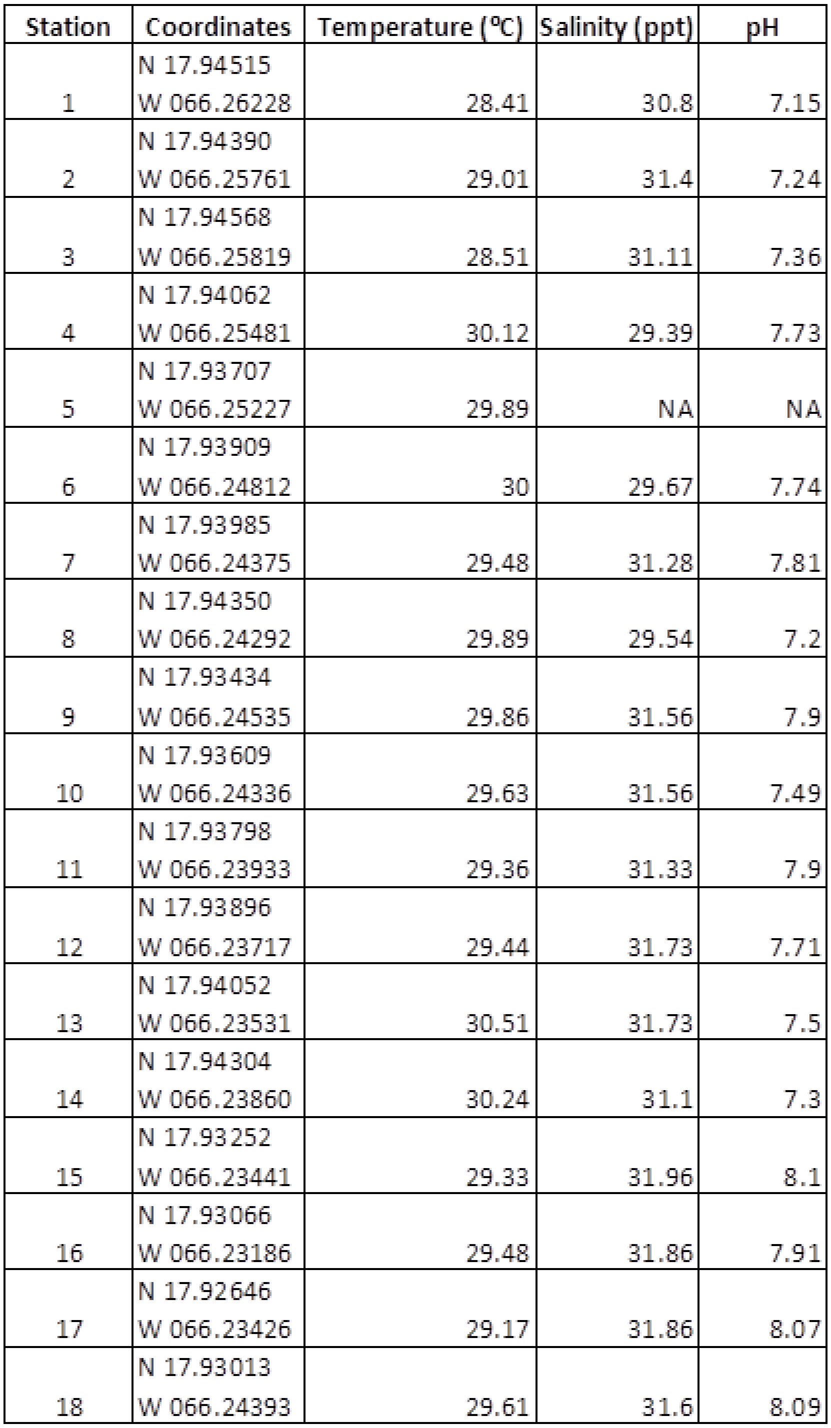
Table A2 Cont.

		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
Totals	ΣtriPCBs	21.19	45.82	1.32	164.96	3.18	34.60	0.57	35.66	2.03	44.90	8.83	5.35	0.06	184.19	0.00	12.41	0.00	0.01
	ΣtetraPCBs	79.55	33.24	7.89	4.16	10.90	2.62	0.23	16.09	2.06	1.76	6.47	8.49	0.26	30.82	0.20	0.96	0.19	0.09
	ΣpentaPCBs	147.57	111.08	19.58	14.11	80.78	66.88	0.79	25.90	27.47	13.81	560.03	25.18	0.00	155.88	0.04	3.37	0.41	0.09
	ΣhexaPCBs	36.06	72.03	3.22	3.22	7.80	17.00	0.07	2.13	2.05	2.67	2.85	11.59	0.65	28.55	0.17	0.87	0.18	0.47
	ΣheptaPCBs	21.40	435.74	1.30	0.62	6.88	50.64	6.10	1.58	21.94	2.18	31.70	0.84	0.23	10.26	0.00	0.52	0.02	0.00
	ΣoctaPCBs	55.62	534.21	0.67	2.40	7.78	69.10	0.57	1.34	21.59	23.75	378.19	2.45	0.00	5.81	0.00	6.77	0.00	0.00

		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
Percentages	StriPCBs	5.86	3.72	3.88	87.07	2.71	14.37	6.82	43.12	2.63	50.40	0.89	9.93	5.16	44.33	0.00	49.85	0.00	1.54
	StetraPCBs	22.01	2.70	23.22	2.19	9.29	1.09	2.76	19.46	2.66	1.98	0.66	15.75	21.96	7.42	48.68	3.84	23.47	13.08
	SpentaPCBs	40.83	9.02	57.65	7.45	68.86	27.77	9.51	31.32	35.62	15.50	56.68	46.72	0.00	37.52	10.55	13.54	51.69	13.85
	ShexaPCBs	9.98	5.85	9.49	1.70	6.65	7.06	0.84	2.58	2.66	3.00	0.29	21.50	54.17	6.87	40.48	3.49	22.50	72.31
	SheptaPCBs	5.92	35.36	3.82	0.33	5.87	21.02	73.21	1.91	28.44	2.45	3.21	1.55	18.97	2.47	0.00	2.10	2.37	0.00
	SoctaPCBs	15.39	43.36	1.96	1.26	6.63	28.69	6.85	1.62	27.99	26.66	38.28	4.54	0.00	1.40	0.00	27.18	0.00	0.00

APPENDIX B

Hydrographic Conditions for Surface Samples



Station	Coordinates	Temperature (ºC)	Salinity (ppt)	pH
1	N 17.94515 W 066.26228	28.41	30.8	7.15
2	N 17.94390 W 066.25761	29.01	31.4	7.24
3	N 17.94568 W 066.25819	28.51	31.11	7.36
4	N 17.94062 W 066.25481	30.12	29.39	7.73
5	N 17.93707 W 066.25227	29.89	NA	NA
6	N 17.93909 W 066.24812	30	29.67	7.74
7	N 17.93985 W 066.24375	29.48	31.28	7.81
8	N 17.94350 W 066.24292	29.89	29.54	7.2
9	N 17.93434 W 066.24535	29.86	31.56	7.9
10	N 17.93609 W 066.24336	29.63	31.56	7.49
11	N 17.93798 W 066.23933	29.36	31.33	7.9
12	N 17.93896 W 066.23717	29.44	31.73	7.71
13	N 17.94052 W 066.23531	30.51	31.73	7.5
14	N 17.94304 W 066.23860	30.24	31.1	7.3
15	N 17.93252 W 066.23441	29.33	31.96	8.1
16	N 17.93066 W 066.23186	29.48	31.86	7.91
17	N 17.92646 W 066.23426	29.17	31.86	8.07
18	N 17.93013 W 066.24393	29.61	31.6	8.09

APPENDIX C

Short-lived Radioisotope Results

Depth Interval	Average Depth	Dry Bulk Density	Total Pb-210	Total Pb-210 error	Background Pb-210	Background Pb-210 error	Excess Pb-210	Excess Pb-210 error	Cs-137	Cs-137 error	Be-7	Be-7 error
cm	cm	g/cm3	Activity (dpm/g)	Activity (dpm/g)	Activity (dpm/g)	Activity (dpm/g)	Activity (dpm/g)	Activity (dpm/g)	Activity (dpm/g)	Activity (dpm/g)	Activity (dpm/g)	Activity (dpm/g)
0-0.5	0.25	0.06	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
0.5-1	0.75	0.04	4.56	0.22	1.46	0.10	3.10	0.25	0.26	0.03	ND	ND
1-1.5	1.25	0.05	2.97	0.23	1.68	0.14	1.29	0.27	0.00	ND	ND	ND
1.5-2	1.75	0.07	2.23	0.17	0.95	0.09	1.28	0.19	0.00	ND	ND	ND
2-2.5	2.25	0.08	1.70	0.10	1.04	0.06	0.66	0.12	0.00	ND	ND	ND
2.5-3	2.75	0.08	2.69	0.19	0.83	0.08	1.86	0.20	0.00	ND	ND	ND
3-3.5	3.25	0.04	2.38	0.22	1.49	0.14	0.89	0.26	0.00	ND	ND	ND
4-4.5	4.25	0.05	1.14	0.09	1.13	0.07	0.01	0.12	0.00	ND	ND	ND
5-7	4.75	0.13	0.90	0.08	0.71	0.05	0.19	0.09	0.00	ND	ND	ND
7-9	6	0.07	0.75	0.06	0.59	0.04	0.17	0.07	0.00	ND	ND	ND
11-13	10	0.07	0.89	0.06	0.53	0.03	0.36	0.07	0.00	ND	0.00	ND
13-15	12	0.08	0.57	0.05	0.79	0.04	-0.22	0.07	0.00	ND	0.00	ND
19-21	18	0.08	3.19	0.13	1.22	0.06	1.97	0.15	0.00	ND	0.00	ND
29-31	28	0.10	1.32	0.07	0.60	0.03	0.72	0.07	0.00	ND	0.00	ND
39-41	38	0.10	1.55	0.07	0.55	0.03	1.00	0.08	0.00	ND	0.00	ND
49-51	48	0.11	0.58	0.04	0.84	0.04	-0.26	0.06	0.00	ND	0.00	ND
59-61	58	0.12	0.67	0.05	0.68	0.04	-0.01	0.06	0.00	ND	0.00	ND

APPENDIX D

Age Model with Pb-210 Profile

Depth Downcore		CRS Pb-210 Age	CRS Pb-210 Age	CRS Pb-210 Age	CRS Pb-210 Age	CRS Pb-210 Age	Mass Accumulation Rate (MAR)
Depth Interval (cm)	Average Depth (cm)	Top of Interval	Bottom of Interval	Average	Age Error	(cm/yr)	(g/cm2/yr)
0-0.5	0.25	2013.0	2012.1	2012.5	5.6	0.53	0.03
0.5-1	0.75	2012.1	2011.4	2011.7	5.6	0.63	0.03
1-1.5	1.25	2011.4	2011.1	2011.2	5.6	0.78	0.04
1.5-2	1.75	2011.1	2010.6	2010.9	5.6	0.85	0.06
2-2.5	2.25	2010.6	2010.4	2010.5	5.6	0.95	0.08
2.5-3	2.75	2010.4	2009.6	2010.0	5.6	0.89	0.07
3-3.5	3.25	2009.6	2009.4	2009.5	5.6	0.97	0.04
4-4.5	4.25	2009.2	2009.2	2009.2	5.5	1.19	0.06
5-7	4.75	2009.2	2009.1	2009.2	5.5	1.28	0.16
7-9	6	2009.1	2008.8	2009.0	5.4	1.68	0.12
11-13	10	2008.4	2007.9	2008.1	5.3	2.14	0.16
13-15	12	2007.9	2006.4	2007.1	5.3	1.98	0.16
19-21	18	2001.2	1996.5	1998.9	5.5	1.15	0.09
29-31	28	1982.7	1979.0	1980.8	6.2	0.85	0.08
39-41	38	1954.4	1939.8	1947.1	9.4	0.53	0.05

APPENDIX E

Percent Sediment Grain Size of Sediment Core

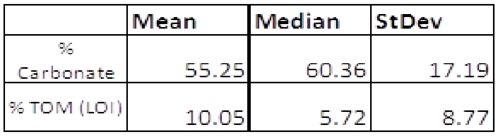
Sample Identification	% Gravel	% Sand	% Silt	% Clay	% Mud
0.5-1.5	28.07	12.99	47.06	11.88	58.94
2-2.5	0.00	3.98	66.37	29.65	96.02
3-3.5	0.00	2.87	65.79	31.35	97.13
4-4.5	0.00	1.76	86.39	11.85	98.24
5	0.00	0.68	61.42	37.90	99.32
7	0.00	0.28	60.79	38.93	99.72
9	0.00	2.11	68.87	29.02	97.89
11	0.00	2.49	70.11	27.40	97.51
13	0.00	0.47	63.82	35.71	99.53
15	0.00	0.97	61.06	37.97	99.03
17	0.00	2.66	71.77	25.57	97.34
19	0.00	0.26	54.29	45.45	99.74
21	0.00	0.12	58.19	41.68	99.88
23	0.00	0.35	48.96	50.69	99.65
25	0.00	0.21	53.62	46.18	99.79
27	0.00	0.08	61.68	38.24	99.92
29	0.00	0.56	53.78	45.66	99.44
31	0.00	0.13	59.56	40.31	99.87
33	0.00	0.65	57.65	41.69	99.35
35	0.00	0.30	58.41	41.29	99.70
37	0.00	0.11	66.39	33.49	99.88
39	0.00	0.28	58.25	41.46	99.72
41	0.00	0.21	56.69	43.10	99.79
43	0.00	0.22	50.83	48.94	99.78
45	0.00	0.21	50.19	49.61	99.79
47	0.00	0.24	53.87	45.89	99.76
49	0.00	0.30	55.92	43.77	99.70
51	0.00	0.40	57.13	42.47	99.60
53	0.00	0.53	57.91	41.56	99.47
55	0.00	0.38	57.11	42.52	99.62
57	0.00	0.35	57.74	41.91	99.65
59	0.00	0.32	53.76	45.92	99.68
61	0.00	1.18	51.62	47.20	98.82
63	0.00	1.74	55.90	42.37	98.26
65	0.00	0.99	57.51	41.50	99.01
66.5	0.00	1.66	58.84	39.50	98.34
68.5	0.00	2.97	59.66	37.37	97.03
70.5	0.00	2.92	61.85	35.23	97.08
72.5	2.28	4.44	62.26	31.03	93.29
74.5	0.00	5.37	58.11	36.51	94.63
76.5	0.00	2.59	59.78	37.62	97.41
78.5	0.00	2.95	59.69	37.37	97.05
80.5	0.00	0.89	46.24	52.87	99.11
82.5	0.00	1.58	60.13	38.29	98.42
83.5	0.00	1.17	64.02	34.80	98.83
85.5	0.00	1.37	66.77	31.86	98.63

Sediment (%)	Mean	Median	StDev
Gravel	0.66	0.00	4.14
Sand	1.51	0.68	2.16
Silt	59.30	58.41	7.01
Clay	38.53	39.50	8.42
Mud	97.83	99.32	6.04

APPENDIX F

Percent Total Organic Matter and Carbonate for Sediment Core

Sample Identification	% Carbonate	% TOM (LOI)
0.5-1.5	N/A	N/A
2-2.5	59.89	6.6
3-3.5	67.06	5.7
4-4.5	N/A	N/A
5	60.42	6.01
7	60.46	5.70
9	57.58	6.83
11	57.22	5.54
13	57.48	5.87
15	57.05	6.25
17	60.27	5.43
19	62.00	5.00
21	62.53	4.67
23	67.66	4.43
25	62.89	4.48
27	68.30	4.53
29	68.40	4.32
31	68.21	3.92
33	67.04	4.44
35	65.77	4.38
37	62.65	6.00
39	66.19	4.74
41	66.09	4.15
43	65.46	5.00
45	65.31	4.42
47	60.51	4.99
49	57.84	5.33
51	62.62	5.40
53	61.86	5.34
55	48.51	6.96
57	63.69	5.54
59	60.36	5.72
61	61.49	6.18
63	59.89	6.88
65	58.27	6.66
66.5	55.11	8.69
68.5	34.88	30.15
70.5	34.94	27.00
72.5	36.26	24.80
74.5	35.51	23.16
76.5	34.07	25.40
78.5	32.44	24.36
80.5	24.52	18.97
82.5	31.15	27.42
83.5	29.96	28.67
85.5	31.36	26.42



	Mean	Median	StDev
% Carbonate	55.25	60.36	17.19
% TOM (LOI)	10.05	5.72	8.77

APPENDIX G

Percent Grain Size on Surface Samples

Surface Samples	% Gravel	% Sand	% Silt	% Clay	% Mud
1	63.65	5.04	18.4	12.9	31.3
2	44.7	11.41	30.84	13.04	43.88
3	71.99	5.35	10.85	11.81	22.66
4	44.19	25.7	14.96	15.15	30.11
5	29.29	8.04	31.35	31.32	62.67
6	38.4	20.76	21.58	19.26	40.84
7	31.01	24.72	21.11	23.15	44.26
8	9.58	5.69	45.4	39.28	84.68
9	8.12	45.5	26.42	19.96	46.38
10	0	2.44	61.28	36.28	97.56
11	20.83	29.75	22.25	27.17	49.42
12	61.31	8.42	14.16	16.11	30.27
13	59.94	6.86	17.91	15.29	33.2
14	0	0	95.8	4.18	99.98
15	0.19	31.23	56.08	12.5	68.58
16	8.87	34.38	41.38	15.36	56.74
17	2.51	15.29	57.27	24.93	82.2
18	22.35	36.9	28.82	11.94	40.76

Grain Size (%)	Mean	Median	StDev
Gravel	28.71833	25.82	24.44838
Sand	17.63778	13.35	13.75425
Silt	34.21444	27.62	21.86878
Clay	19.42389	15.735	9.282787
Mud	53.63833	45.32	23.92367

APPENDIX H

Percent Total Organic Matter and Carbonate for Sediment Surface Samples

Surface Samples	% TOM (LOI)	% CO3
1	15.7	64.2
2	30.8	50.2
3	6.5	85.1
4	6.1	88.9
5	23.1	55.8
6	19.2	74.1
7	7	84.3
8	47.7	41.5
9	4.9	78.6
10	16.9	43.2
11	7.6	81.9
12	6.3	87.2
13	10.5	76.7
14	34.4	33.2
15	3.1	46.2
16	5	58.2
17	9.8	55.2
18	7.6	60.2

	Mean	Median	StDev
% TOM (LOI)	14.57	8.70	12.30
% CO3	64.71	62.20	17.81