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Environmental PCBs in Guánica Bay, Puerto Rico: Implications for Community Health

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Abstract

Guánica Bay, located in southwestern Puerto Rico, has suffered oil spills and other pollution discharges since the 1960s. Previous research showed elevated concentrations of polychlorinated biphenyls (PCBs) in coral reef and sediment. This research examined PCB concentrations in sediment and fish. Sediment and fish sampling in the bay was facilitated by community members. This study identified the second highest reported PCB level (129,300 ng/g) in sediment in the United States. Fish samples also showed elevated concentrations (1,623ng/g to 3,768 ng/g), which were higher than the thresholds of safe levels of PCBs in fish for human consumption. The alarmingly high concentration of PCBs calls for proactive community engagement to bring awareness about contamination of the bay and more extensive sampling to test for the concentration PCBs in seafood and the people of Guánica. This study also underscores the value of the involvement of local communities during sampling design aimed at identifying hot spots of contaminants.

Keywords

PCB; fish; sediment; Guánica; community-based participatory approach; health risks; PCB exposure

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INTRODUCTION

The environmental contamination of polychlorinated biphenyls (PCBs) poses a threat to public health. PCBs can damage the immune, reproductive, nervous, and endocrine systems (Axelrad et al. 2009; EPA 2013c). PCB exposure is shown to cause cardio- and cerebro-vascular disorders, increased rates of autoimmunity, cognitive and behavioral problems, and hypothyroidism (Carro et al. 2013; Hennig et al. 2002). Recent evidence also implicates PCBs in obesity, type 2 diabetes, and hypertension (Everett et al. 2011). While the Environmental Protection Agency (EPA) classified PCBs as *probable human carcinogens* (EPA 2013b), International Agency for Research on Cancer classified PCBs as *carcinogenic to humans* (Lauby-Secretan et al. 2013). PCBs are persistent organic pollutants and remain in the environment for long periods of time (Grimm et al. 2015). Depending upon the number and nature of the chlorine substitutions, PCBs display different degrees of hydrophobicity and are thus prone to absorption and bioaccumulation in fatty tissues (Field and Sierra-Alvarez 2008; National Research Council 2001). The most common exposure route of PCB to humans is through food. The highest levels of PCBs have been found in people who regularly eat fish that have been contaminated with PCBs (Bjerregaard et al. 2013; Dallaire et al. 2014). The production of PCBs was banned in the US in 1976 because of their toxicity. Since then, the levels of PCBs in food chain (such as fish) and human blood have declined in the United States (EPA 2013c; Wenning et al. 1994; Xue et al. 2014). However, elevated concentrations of PCBs are still found in the environment and sites contaminated with PCBs are still a concern because of PCB resistance to natural degradation and bioaccumulation in the food chain (Abraham et al. 2002; CDC 2014; Ohtsubo et al. 2004). Therefore, environmental PCBs continue to pose threats to communities, especially for communities that lack resources to measure the contamination and to promote (and seek) clean-up efforts. The Guánica Municipality, located in the southern west part of Puerto Rico and the study area of this research, is an example of such a community.

Guánica Bay receives pollution discharges from many different sources including sugar mills, fertilizer plants, textile companies, and sugar and coffee plantations, which drain into the Rio Loco (Rodrigueq 2015; Sturm et al. 2012). This river discharges at the northeast corner of the bay (Morales 2009). Domestic and industrial waste and raw and partially treated sewage are also discharged into the bay (Morales 2009). Moreover, several oils spills have been reported in the bay, including three documented oil spills in and around the bay in 1962, 1994 and 2007 (CSA Group 2001; Diaz-Piferrer 1962; Golob 1994; Orlando Sentinel 1994; Robles 2007).

NOAA has focused on pollution damage to the coral reef ecosystems in Guánica Bay and just outside the bay, which provided an important base for this study (NRCS 2009). In 2005 and 2009, NOAA collected sediment and coral samples in the bay, and offshore in the Caribbean Sea (Figure 1). The NOAA data indicated that the average PCB concentration in the bay sediments was higher (214 ng/g; expressed as the sum of all congeners) than sediments outside the bay (mean ~ 6.69 ng/g; 95% confidence interval ± 6.10 ng/g). The maximum concentration found in the bay sediments during this earlier work was 1,918 ng/g (NCCOS 2014a; Whitall, Bauer, Sherman, Edwards, Mason, Pait, Caldow 2013). Such concentrations were elevated and motivated the present study objectives to: a) examine levels of PCBs in the bay and identify hot spots of PCBs, b) determine the levels of PCBs in fish, c) assess potential implications for community health, and d) increase community engagement in issues related to environmental health.

MATERIALS AND METHODS

There is an increasing interest in involving community members in environmental monitoring as they are familiar with recent and historical environmental events, such as oil spills and other disasters, in their community (Azaroff et al. 2011; Holloman and Newman 2010; Kondo et al. 2014). Utilizing this framework, the University of Miami team has established a trustworthy partnership with Guánica Municipality for the past three and a half years. Sampling efforts capitalized on this partnership for developing and implementing sampling strategy. The local knowledge of community members about the known oil spills served as a critical input in identifying sites along two sampling transects: a) northwest to southeast, and b) west to east. Although this sampling strategy is not completely probabilistic, it captures spatial contamination levels as perceived by the community. Three rounds of sediment sampling were administered, and sampling covered areas (along these transects) that community members suspected were contaminated. A total of 23 sediment samples were collected: a) twelve samples in April 2013, b) eight in December 2013 and c) three in March 2014 (Figure 1; Supplement Table – S1). In the first round, samples were collected along a northwest to southeast transect, suspecting a flow of pollution from the Rio Loco in the northern part of the bay. In December 2013, sediment was sampled along a new west to east transect. Community members informed us of several oil spills, including the used oil spill from a storage tank that flowed through a dry stream into the northeast side of the bay. Despite these spills, the eastern part of the bay had not been sampled earlier by NOAA and others. However, community members suggested eastern part of the bay as the most contaminated areas. Since PCB levels were highest near the dry stream bed during the first round of sampling in December 2013, sampling was repeated in this area (in March 2014) to confirm the consistency of the results.

The University of Miami team and community members participated in the sampling. While Mr. Rodriguez, a local community member, navigated the boat to the sample sites drawn along two transects, UM team members (Kumar and Ortiz-Ramirez) collected the sediment samples. The boat was anchored and surface sediment was dredged. Since sediment on the anchor was lifted through the water column, the top one inch of dredged sediment was removed, and the eight-ounce, wide-mouth clear glass jars (w/70–400 PTFE Lined Cap, Certified), was filled with the remaining sediment on the anchor. Upon collection, samples were placed in an ice cooler and were flown back to the University of Miami for analysis. Fish were caught using a drift net deployed in the center of the bay (latitude ~ 17.96395° N; longitude ~ 66.90685° W) for three hours (starting from 9:00AM) on September 11, 2014 (Figure 1). The five fish trapped in the net were measured, labeled, weighed and wrapped individually in aluminum foil, placed in zip plastic bags and positioned under dry ice in coolers (EPA 2000a). Of these five, two randomly selected fish were analyzed for PCB concentration.

Sediment samples were extracted and processed according to standard protocols (U.S. EPA 3550B (for Aroclors) and 3545A (EPA 2007; EPA 2013d) (for PCB congeners)). The samples, collected in April 2013, December 2013 and March 2014 were analyzed for PCBs as Aroclors (specifically for Aroclor 1016, 1221, 1232, 1242, 1248, 1254 and 1260) by Florida Spectrum Environmental Services, Fort Lauderdale, FL. A subset of three samples (collected in March 2014) and two fish samples (collected in September) were analyzed for 80 PCB congeners by TDI-BI/B&B Labs, College Station, TX (see online supplementary material (OSM) for details on the sampling method; analysis of field blank and duplicate analysis); summaries of PCB concentrations in sediment and fish samples are presented in Table 1 and 2, respectively.

To analyze data, descriptive statistical methods and geographic information systems (GIS) were used to describe and visualize results ArcGIS 10.1 (ESRI 2014). PCB data in sediment samples were also acquired from NOAA (NCCOS 2014a) and superimposed on the data collected by the University of Miami team. T-tests (ttest function in Stata Ver 11) was employed to examine differences between NOAA’s data and those collected by our team. Although these data were skewed, the analysis was performed on the log-transformed (normalized) data.

RESULTS

Distribution of Aroclors in Guánica Bay

Results showed detectable levels of Aroclor 1260 1232, and 1016 only. The average concentration (of the sum of Aroclors 1260, 1232 and 1016) was 1,388 ng/g (95% confidence interval ~1,245 ng/g; n=23) (Figure 2; Supplement Table S1). The highest concentration of 14,080 ng/g (or ~14 ppm) was found in the eastern part of the bay; the second highest concentration (4,150 ng/g) was found at an adjacent site (Figure 2). Aroclor 1260 was detected 21 of the 23 samples (Table S1). Its average concentration within the bay was 865 ng/g (95% CI interval ~678; n = 22; the confidence interval was wide due to a skewed distribution, ranging from 36 ng/g to 7,190 ng/g, as a result of extremely high values at several sites in eastern parts of the bay (Figure 2)). The levels of Aroclor 1232 were as high as 6,890 ng/g. The highest concentrations of all three Aroclors were found in the eastern part of the bay near the town of Guánica. We found elevated concentrations of Aroclors 1260 and 1016 (> 200 ng/g) in the central part of the bay as well (Figure 2). The spatial distribution of PCB congeners (Figure S3) indicates that hexachlorobiphenyls are most dominant (22.5% of the total PCBs) across all sites, followed by hepta-, penta-, tetra- and tri-chlorobiphenyls (with 20, 18, 17 and 12%, respectively) (Figure S3).

Distribution of PCB congeners

We conducted the comparison on a congener basis, recognizing that the sum of Aroclors may not be comparable to the sum of individual congeners of PCBs for two reasons. First, some Aroclors share some of the same congeners. Aroclor 1260 consists of more chlorinated PCB (5 to 9 chlorine atoms), whereas Aroclors 1016 and 1232 consist of congeners that have only 1 to 4 chlorine atoms. Thus, Aroclor 1260 does not have congeners that overlap with either Aroclor 1016 or 1232. For Guánica bay samples, when more than one Aroclor was detected, it was Aroclor 1260 in combination with either Aroclor 1016 or Aroclor 1232 (Figure S2). Thus, there was no overlap in congeners among these sets of Aroclors, and adding them together can provide estimates of total PCB provided other Aroclors do not exist (EPA 2013a; Fieldler 1997). However, for samples where both Aroclors 1016 and 1232 were detected, the sum of the Aroclors would overestimate the value on a congener basis. Second, it is difficult to compare the sum of individual PCB congeners to Aroclor measures is that there is a possible differential in degradation among the various congeners that make up a specific Aroclor. It is likely that some Aroclors (that consist of a combination of different PCB congeners in certain ratio; e.g. in Aroclor 1260 percentages of congeners with chlorine atoms 5, 6 7, 8 and 9 are 12,38,41,8 and 1, respectively) may not be even detected if (as a result of degradation) the signature chromatogram of the original Aroclor is missing (Narquis et al. 2007). Therefore, in comparing our data with those of NOAA, a subset of three samples was analyzed for individual congeners of PCBs. Two of the three samples (sample IDs: 21 and 22) were chosen because they had very high concentrations of Aroclors 1260 and 1232. The third sample (sample ID: 24) was chosen because it was adjacent to one of NOAA's sites (Figure 3; see Supplement Table S3). From these data, several important findings emerged. First, total PCB levels expressed as the sum of all congeners were substantially greater than the sum of Aroclors. Although the samples were analyzed for eight different Aroclors using the US EPA method 3550B, only two were detected in the three samples analyzed for PCB congeners. It is important to note that Aroclor analysis is less robust and a preliminary screening tool. Thus, congener specific analysis is recommended for the total concentration of PCBs (Narquis, Hyatt, Prignano 2007), because of poor Aroclor detection caused by degradation of individual congeners and shifting of signature chromatograms.

The PCB concentration (at the site of highest concentration of Aroclors) was 129,284 ng/g (or 129 ppm), which is the highest level ever reported in the United States NOAA database and is 67-fold higher than previously found in the bay (1,918 ng/g) (Table 1) (NCCOS 2014b). Two other sites, 22 and 24, showed very high concentrations as well: 44,150 ng/g and 4,213 ng/g respectively. Second, the total PCB concentration at the third site (4,213 ng/g) was 15-fold higher than the concentration found at an adjacent site by NOAA (Figure 3).

The NOAA data from two rounds of sampling in 2005 and 2009 showed an average level of PCBs in Guánica bay sediments as 352 ng/g (95% CI ~ 206.45; n = 17). The gradient of PCB concentration declined towards the outside of the bay. The average concentration outside the bay in the Caribbean Sea was 6.69 ng/g (95% CI ~ ±6.10; n = 10), more than 50-fold lower than that found in the bay. There was significant spatial variation in the distribution of PCBs even in the bay. NOAA's samples were spread across the northwest to southeast transect, along the main shipping channel. Variation in the deposition rate, the depth of the sediment core and fate and transport processes of sediment and contaminants could result in such heterogeneity. For example, the NOAA data showed one relatively high concentration (765 ng/g) of PCB in the northern part of the bay, but relatively low concentrations at adjacent sites (Figure 4).

To further compare the data of this study with NOAA data the analysis: a) restricted the comparison of Aroclor 1260 to the sum of penta- to nona-chlorobiphenyls, which are dominant in Aroclor 1260, and b) excluded the data from all four eastern locations of the highest PCB concentrations, where NOAA did not sample. The average concentration of Aroclor 1260 (235 ng/g; 95% confidence interval ~ 101.5; n = 17) did not vary significantly (difference ~7.01 ng/g; p ~0.93) from the average concentration of the sum of penta- to nona-chlorobiphenyls (241 ng/g; 95 confidence interval ~ 134 ng/g; n=17) (Whitall, Bauer, Sherman, Edwards, Mason, Pait, Caldow 2013). While the statistical analysis did not show any significant variations across these two datasets, their spatial distribution did not completely overlap, as shown in Figure 3. This difference in spatial distribution is likely due to heterogeneity of the sediment samples. Nonetheless, both datasets showed elevated levels of PCBs in Guánica Bay, with average values among the comparable portions of the datasets within 3 percent of one another.

PCB Concentrations in Fish

Elevated concentration in sediment led to the assessment of PCB concentration in fish. The PCB concentrations (sum of 68 congeners) in the *Lutjanus analis* and *Eucinostomus gula* were 1,620 ng/g and 3,770 ng/g, respectively (Table 2). Aroclors 1248 and 1260 were detected in the fish samples. The concentration of Aroclor 1248 in *Lutjanus analis* was 568 ng/g and in *Eucinostomus gula* was 2,640 ng/g. The concentration of Aroclor 1260 in *Lutjanus analis* was 1,060 ng/g and in *Eucinostomus gula* was 1,130 ng/g (Table 2).

DISCUSSION

Environmental concentrations of PCBs in the US have declined since PCBs were banned in the US in 1976 (Wenning, Bonnevie, Huntley 1994). But elevated concentrations are still being observed at and around the designated U.S. contamination sites known as Superfund sites (ATSDR 2000), and other places which have been subjected to pollution discharges. The concentrations of PCBs detected in the sediment of Guánica Bay are much higher than those found at and around many Superfund Sites, including the LCP Chemicals site in Georgia (Kannan et al. 1997), at Lake Hartwell in South Carolina (Magar et al. 2005a; Magar et al. 2005b), along the Hudson River (Baker et al. 2006; Bopp et al. 1998), around San Francisco Bay (Venkatesan et al. 1999), and around Tampa and Galveston Bays (Santschi et al. 2001). At all of these sites, the PCB levels were lower than the highest values reported in the U.S. prior to this study of 17 mg/kg, reported in Boston Harbor (Robertson 1988). The highest concentration of PCBs ever reported was in the St. Lawrence River in Canada (i.e., 5,700 mg/kg) (ATSDR 1995). The 129 mg/kg concentration of PCBs in Guánica Bay is to date the second highest ever recorded worldwide.

The sources of the PCB contamination in Guánica bay are unknown. The Rio Loco and the shipping channel within the Bay do not seem to be the primary source, as PCB levels along the northern-west to southern-east transect were lower than those along the east-west transect (Figure 2), and PCB levels near the discharge point of Rio Loco was substantially lower than the levels of PCBs found in eastern parts of the bay (Figure 3). However, it is recognized, that sediments and contaminants could have been redistributed as a result of dynamic transport processes. Documented literature suggests that PCBs may have also come from oil spilled into the bay from an used storage tank on the grounds of a former sugar mill on the eastern shore of the bay (CSA Group 2001; Golob 1994; Orlando Sentinel 1994). However, nothing has been reported (or proven) to date to implicate the sugar mill complex.

Suggested rephrasing this paragraph: “Findings from this study indicate high levels of PCBs in sediments and fish in the Guánica Bay, and suggest high potential risk of exposure to community members. Potential exposure could occur through ingestion of contaminated fish, dermal exposure, and inhalation of PCB vapors above the water column. PCBs in sediment can desorb into the pore water and water column, and then enter the atmosphere in a gas phase (ATSDR 2000; Hu et al. 2010; Zhao et al. 2010). Consequently, elevated concentrations of PCBs in sediment can increase the likelihood of PCBs exposure through air and water just as they do through the consumption of contaminated seafood, albeit at a lower rate. However, the level of exposure is unknown and any inference to exposure should be made with caution for several reasons. First, true sediment concentrations could have been disturbed during the sampling processes. Second, PCB levels in the water column were not measured. Thus, the cause effect relationship between PCB levels in fish and PCB levels in sediment cannot be established. Third, elevated concentrations of PCBs in sediment and fish do not directly translate to human exposure to PCBs, unless community members have been consuming contaminated fish and utilizing the bay for recreational purposes. Nonetheless, our field observations and informal discussion with the community members suggest that community members are not aware of PCB contamination of the bay and continue to fish in the bay (see OSM Figure S5).

The average half-life of PCBs in river sediment has been reported to be 9.5 (±2.2) years (ATSDR 2000). Once PCBs are released in the environment, they gradually biodegrade and move across space and time due to their complex interactions with physical, chemical, biological, and ecological elements (Totten et al. 2006). Because PCBs are lipophilic and because marine organisms are constantly exposed to PCBs, PCBs bioaccumulate in marine organisms. A review of two studies provides insight into the association between PCB concentrations in sediment and in fish. First, in the eastern Georgia estuary, the average PCB concentration in sediment was 7.8 ng/g as compared to 0.48 ng/g (±0.24 ng/g 95% confidence interval CI) in spotted sea trout and 0.78 ng/g (±0.21ng/g 95% CI) in striped mullet (Maruya and Lee 1998). Second, in the Saginaw River, Lake Huron, the total PCB concentrations ranged from 33 to 280 ng/g, dry weight, in sediments as compared to 46 to 290 ng/g, wet weight, in caged fish (Echols et al. 2000). At a minimum, these finding indicate that where there are some PCBs in sediment, there is likely to be some level of PCBs in fish. Elevated concentrations we found in fish sample further substantiate this literature.

PCBs in sediment have been shown to have adverse effects on marine environments and on human health. According to NOAA’s sediment quality guidelines (SQG) “effect range-low (ERL)” and “effect range-median (ERM)” thresholds for the sum of congeners are ≤22.7 ng/g and ≤180ng/g, respectively (Long 1992; Long et al. 1995; Long and Morgan 1990). The EPA defines the ERL and ERM as the concentrations where toxicity in benthic organisms would occur at 10% and 50% frequencies, respectively. PCB concentrations observed throughout Guánica Bay are consistently greater than the ERM, suggesting a potential adverse impact on benthic organisms.

Future work will assess the level of PCBs in seafood will be assessed and the level of PCBs in the people of Guánica Municipality (through biomonitoring) will be determined. Given the levels of PCBs in sediment were very high, it is hypothesized that seafood contains elevated levels of PCB beyond the possibly safe levels, ranging from 1.1 mg/kg (or 1,100 ng/g) in whole fish to 25–70 mg/kg in adult fish liver (Meador et al. 2002). The Food and Drug Administration (FDA) established that total PCB level (i.e. sum of all congeners of PCBs) < 2000 ng/g for human consumption (FDA 1998). However, EPA's risk based approach set 2.45ng/g concentration for subsistence fish consumption (EPA 2000b). *Eucinostomus gula* (one of the two fish samples analyzed) showed PCBs at levels about two times higher than the FDA restricted value. These elevated PCB levels in fish (though only in two wild-caught fish) from Guánica bay are similar to the PCB levels found in several seafood/fish species from the Hudson River (Steinbacher and Baker 2002), Great Lakes (Straub and Sprafka 1983), St. Lawrence River (Straub and Sprafka 1983), Saginaw River/Bay (Echols et al. 1999; MDNR 1988), and higher than the concentrations found in Delaware Bay and Chesapeake Bay (Levinton and Waldman 2006). All of these sites have posted fish advisories. However, we did not find any fish advisories around the bay or in the community newsletters or local newspaper. As this is the first study to assess PCBs levels in the fish from Guánica Bay, further research is needed to assess the contamination of seafood in the bay, and body burden of PCBs in the people of Guánica. There are no existing sources of data (including biomonitoring by NHANES) to assess the levels of PCBs in serum in Puerto Rico, especially in Guánica Municipality.

CONCLUSION

This research found hazardous concentrations of PCBs in sediment, and elevated concentrations of PCBs in fish in Guánica Bay. The elevated concentrations of PCBs pose high risk of exposure to communities living near and/or using the bay for recreational and fishing purposes. Thus, it is critically important to: a) engage community members and other stakeholders in bringing community awareness of environmental PCBs in the bay, b) develop a base-line knowledge of the body burden of PCBs in the communities surrounding the bay and its associated adverse health effects, and c) engage community members in reducing their exposure to PCBs. The results of this research underscore the value of a community-centered approach. Without this approach, the extraordinarily high values of PCBs located in the eastern part of the bay may not have been discovered. Moreover, the community-centered approach also brought to light about the lack of awareness and fish advisories in the Guánica Municipality. This work shows that engaging the community in research activities and outcomes raises interest and awareness about the environmental risks posed by environmental contaminants.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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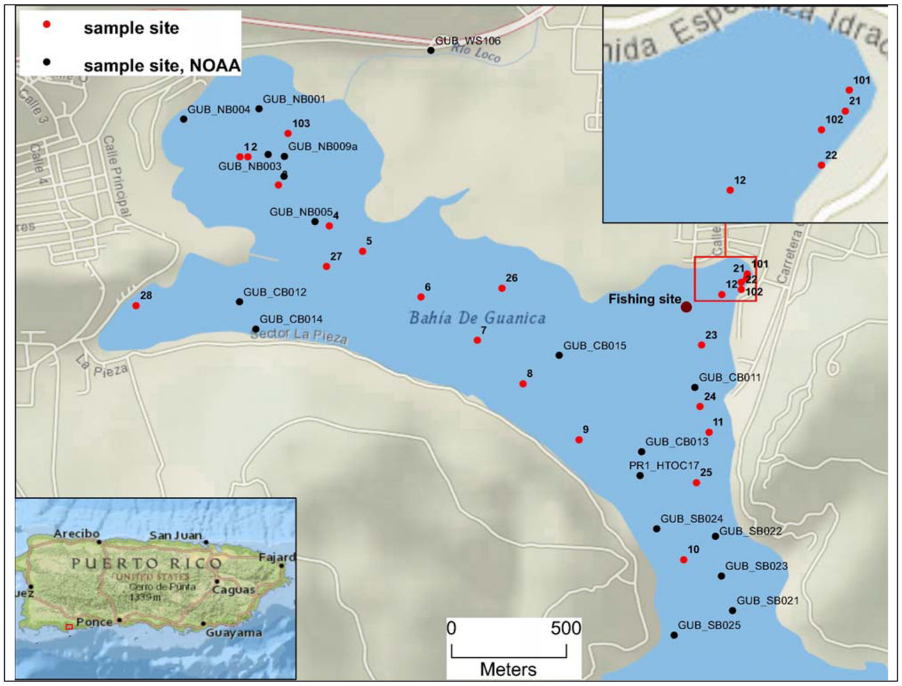

Figure 1.

Location of sediment samples collected by the University of Miami Team and NOAA, and the location fishing site for collecting fish samples. NOAA sample sites begins with character “GUB”, and UM sites are numeric. UM Sample ID > 100 (namely 101, 102 and 103) were analyzed for both Aroclors and Congeners. Fishing net was deployed in the northeast part of the bay.

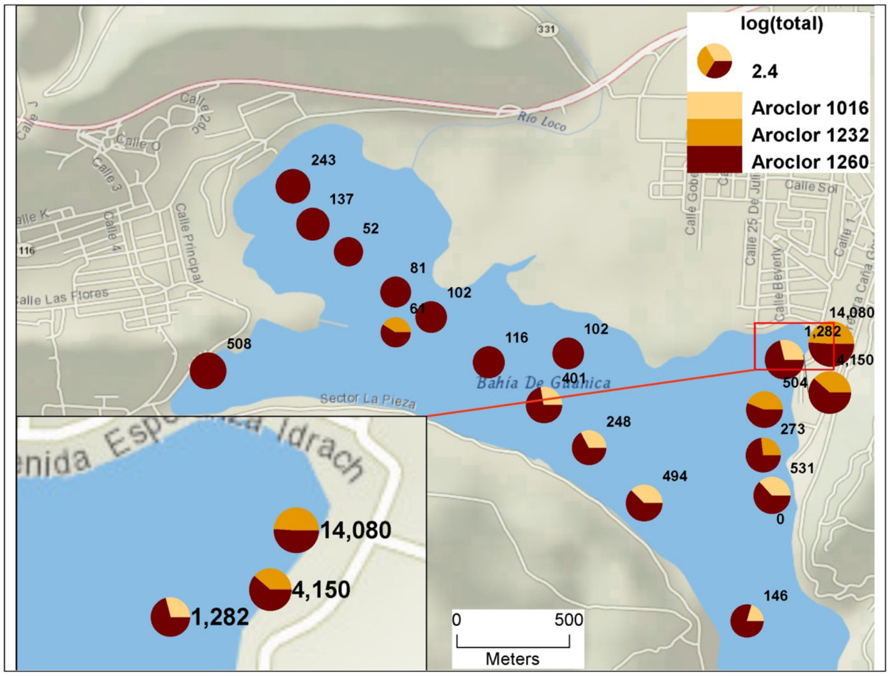

Figure 2.
Concentration of Aroclors in Guánica Bay; the sum of Aroclors for each site is written on its symbol, show concentration in ng/g, and the values shown in the legend are at log scale, i.e. ln(ng/g).

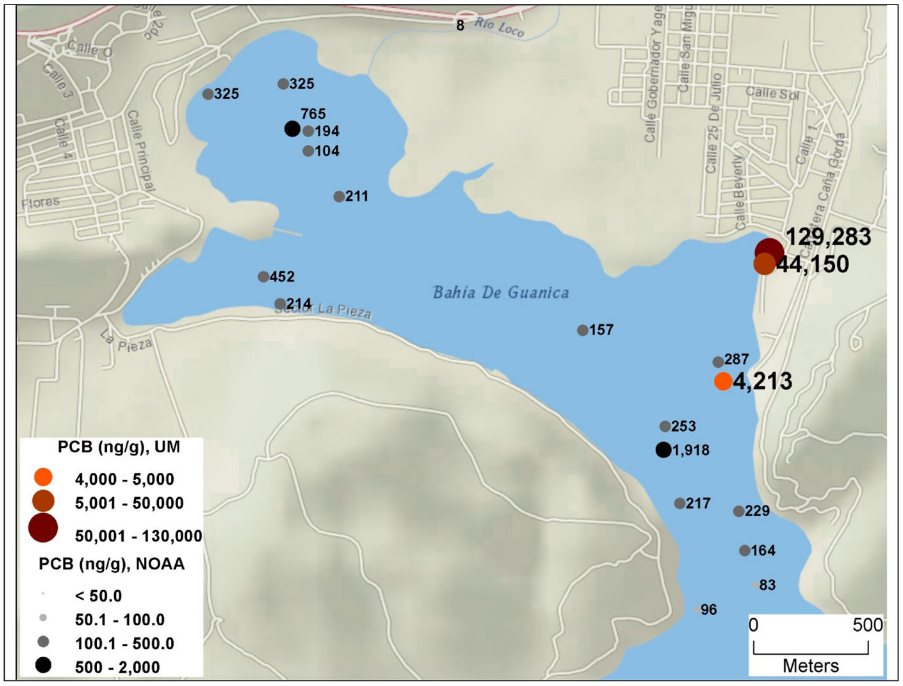

Figure 3.

PCB congener concentration (ng/g) in Guánica Bay. Gray color circles are PCB levels (i.e. the sum of congeners) detected by NOAA[20] and orange brown by the University of Miami Team.

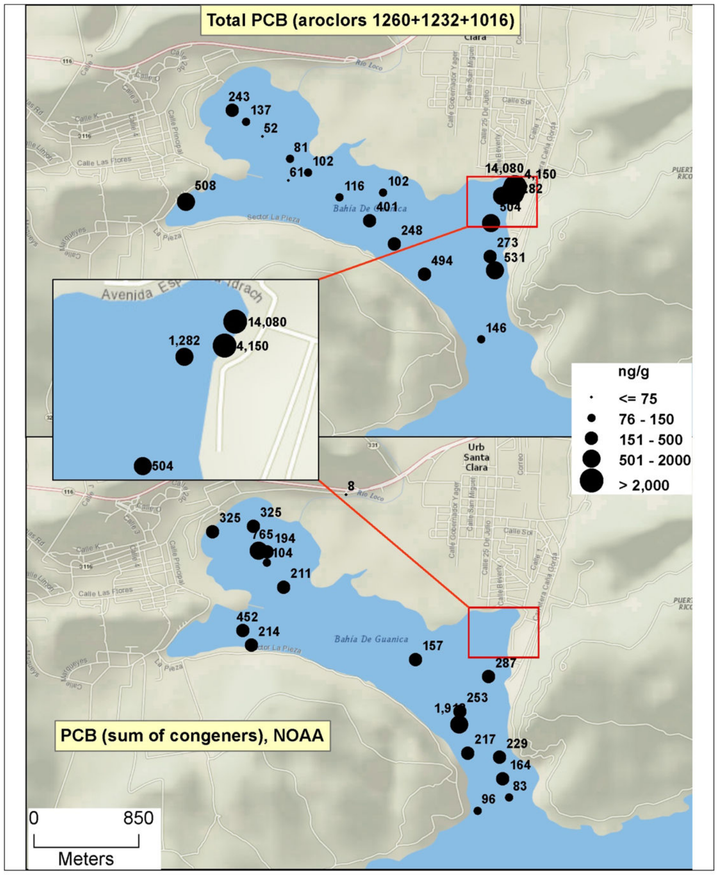

Figure 4.

a. Sum of the detected Aroclors in Guánica Bay (ng/g) sampled by the University of Miami team; b) the sum of congeners sampled by NOAA (ng/g).

Table 1

Concentration of PCB in sediment (ng/g) at three selected sites, located in the eastern part of Guánica Bay, March 2014.

PCB Group*	**Sample ID**		
	21	**22**	**24**
mono-	92	55	4
di-	2,752	1,773	152
tri-	34,878	15,558	1,027
tetra-	36,615	13,966	1,111
penta-	11,362	3,935	480
hexa-	25,887	4,779	829
hepta-	13,598	3,055	480
octo-	3,804	924	123
nona-	286	99	7
deca-	9	6	ND
Total	129,284	44,150	4,213

*see Supplement Table S2 for the concentration of individual congeners.

Table 2

PCB Concentrations in a *Lutjanus analis* and a *Eucinostomus gula* from Guánica Bay (see OSM Table S4 for congener specific concentrations in fish samples).

PCB Concentrations in Whole Fish (ng/g)							
Species	**Size (mm)**	**Fish weight (g)**	**Sample Wet Weight* (g)**	**Aroclor**			**PCB (sum of 68 Congeners)**
				Aroclor-1248	**Aroclor-1260**	**Total**	
Lutjanus analis	248.9	206.7	5.44	568	1,060	1,620	1,620
Eucinostomus gula	228.6	146.9	5.24	2,640	1,130	3,770	3,770

* Fish was homogenized and 5g sample of homogenized fish tissues were analyzed for PCB concentration.