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Polychlorinated biphenyl (PCB) contamination in Galveston Bay, Texas: Comparing concentrations and profiles in sediments, passive samplers, and fish[☆]

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ABSTRACT

The industrialized portion of the Houston Ship Channel (HSC) is heavily contaminated with anthropogenic contaminants, most prominent of which are the polychlorinated biphenyls (PCBs). This contamination has driven adaptive evolution in a keystone species for Galveston Bay, the Gulf killifish (*Fundulus grandis*). We investigated the geographical extent of PCB impacts by sampling 12 sites, ranging from the heavily industrialized upper portion of the HSC to Galveston Island. At each site, PCB concentrations and profiles were determined in three environmental compartments: sediment, water (polyethylene passive samplers), and fish tissue (resident Gulf killifish). We observed a steep gradient of PCB contamination, ranging from 4.00 to 100,000 ng/g organic carbon in sediment, 290–110,000 ng/g lipid in fish, and 4.5–2300 ng/g polyethylene in passive samplers. The PCB congener profiles in Gulf killifish at the most heavily contaminated sites were shifted toward the higher chlorinated PCBs and were highly similar to the sediment contamination profiles. In addition, while magnitude of total PCB concentrations in sediment and total fish contamination levels were highly correlated between sites, the relative PCB congener profiles in fish and passive samplers were more alike. This strong correlation, along with a lack of dependency of biota-sediment accumulation factors with total contamination rates, confirm the likely non-migratory nature of Gulf killifish and suggest their contamination levels are a good site-specific indicator of contamination in the Galveston Bay area. The spatial gradient of PCB contamination in Galveston Bay was evident in all three matrices studied and was observed effectively using Gulf killifish contamination as an environmentally relevant bioindicator of localized contamination in this environment.

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1. Introduction

The Houston Ship Channel (HSC) in Texas is a heavily industrialized estuary (Fig. 1) and is part of the greater Galveston Bay. Because of the high levels of industrial activity in the HSC, the channel and the greater bay have been extensively studied for

contamination of polychlorinated biphenyls (PCBs), and other persistent pollutants (Aguilar et al., 2013; Santschi et al., 2001; Subedi and Usenko, 2012). Resident fish have been exposed to contaminants in the HSC since the 1940's (Yeager et al., 2007). Some of the efforts to remediate this contamination include dredging as part of the Environmental Protection Agency (EPA) Superfund program that occurred in several locations in the HSC, and its efficacy has been monitored in sediment samples (Yeager et al., 2010). The distribution of contaminants for each site differed based on the class and matrix in which contamination is distributed (Howell et al., 2011). PCBs have shown a pattern of locally elevated concentrations in the HSC, likely stemming from

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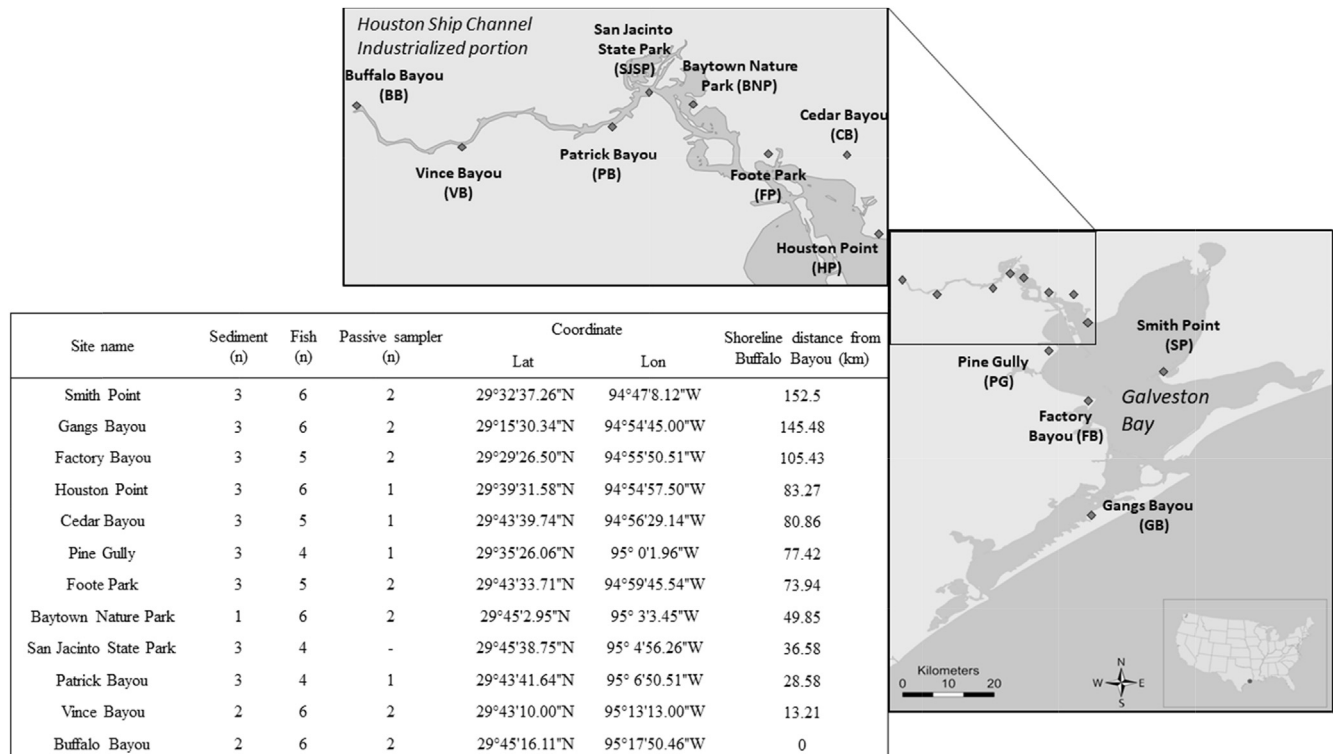


Fig. 1. Three matrices were sampled from a total of 12 locations across Galveston Bay.

discharges from industrial production (Howell et al., 2008, 2011; Lakshmanan et al., 2010). They also have the highest Toxic Equivalency Quotients (TEQ), posing the strongest threat to local aquatic residents (Oziolor et al., 2014; Subedi and Usenko, 2012).

The high levels of persistent pollutants in the HSC are a cause for concern for the environmental health of aquatic environments. Evidence of biological availability is plentiful and includes rapid accumulation and slow depuration of PCBs in oysters transplanted in Galveston Bay (Sericano et al., 1996), elevated biomarkers in populations collected near the HSC (Willett et al., 1997), and elevated PCB levels in crab and channel catfish near the San Jacinto Waste Pits (Aguilar et al., 2013; Subedi and Usenko, 2012). More alarmingly, recent investigations from our group have identified evolutionary adaptation in Gulf killifish (*Fundulus grandis*) populations in the industrialized portion of the HSC (Oziolor et al., 2014, 2016b). This finding suggests that the chronic contamination in the HSC led to a population-wide selective sweep in *F. grandis* populations. Further, there is evidence that the level of adaptation may differ based on proximity to the HSC, with more tolerant populations being found closer to the HSC (Oziolor and Matson, 2015). This large gradient of resistance spanning Galveston Bay suggests divergent regional impacts on resident populations, stemming from the complex contamination mixtures throughout Galveston Bay (Howell et al., 2011; Oziolor and Matson, 2015). Thus, it is imperative to understand the distribution and availability of PCB contamination throughout Galveston Bay in resident populations of impacted aquatic biota.

To fully detail the spatial heterogeneity and bioavailability of PCBs we studied likely sources of contamination (areas from which main contamination may arise) and body burdens of contaminants in impacted populations. As PCBs adsorb strongly to sediment after initial release, the sediment is likely to deliver bioavailable PCBs to the water and porewater (Apell and Gschwend, 2016). To infer bioavailability, one can quantify the contamination in both

sediment and aquatic biota and obtain a biota-sediment accumulation factor (Howell et al., 2011; Ilyas et al., 2013; Niewiadowska et al., 2015; Oziolor et al., 2014; Xu et al., 2016). However, sediment concentrations of PCBs can be very heterogeneous and may not accurately reflect the overall exposure of fish due to the migratory nature of some species. Since water concentrations can be influenced by a larger spatial area than sediment, it may be a better indicator of exposure profiles in aquatic biota. Recently, the use of polyethylene (PE) passive samplers has been shown to accurately reflect freely dissolved water concentrations as well as mimic the uptake of hydrophobic contaminants, like PCBs, into biota in the environment (Apell and Gschwend, 2014; Fernandez and Gschwend, 2015; Joyce et al., 2015, 2016). We chose to study both sediment and water as possible matrices that harbor PCB contamination and relate them to the magnitude and composition of PCB body burdens in resident *F. grandis* populations.

The goal of this study was to examine PCB exposure and uptake in populations of *F. grandis* and to determine if these non-migratory fish (Nelson et al., 2014) are a good indicator of localized pollution profiles. Twelve sites across Galveston Bay were selected to represent an observed gradient of *F. grandis* adaptation. Site distribution varied from the industrialized portion of the HSC to coastal sites in the open Galveston Bay (Fig. 1). We aimed to understand the intensity and spatial distribution of PCB contamination across multiple matrices. Finally, this study was designed to test how effectively resident Gulf killifish could be used to represent local environmental PCB concentrations and profiles.

2. Methods

2.1. Chemicals

Chemicals were purchased from commercial vendors at reagent grade or higher and stored according to manufacturer

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