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Contaminants in tropical island streams and their biota

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ABSTRACT

Environmental contamination is problematic for tropical islands due to their typically dense human populations and competing land and water uses. The Caribbean island of Puerto Rico (USA) has a long history of anthropogenic chemical use, and its human population density is among the highest globally, providing a model environment to study contaminant impacts on tropical island stream ecosystems. Polycyclic Aromatic Hydrocarbons, historic-use chlorinated pesticides, current-use pesticides, Polychlorinated Biphenyls (PCBs), and metals (mercury, cadmium, copper, lead, nickel, zinc, and selenium) were quantified in the habitat and biota of Puerto Rico streams and assessed in relation to land-use patterns and toxicological thresholds. Water, sediment, and native fish and shrimp species were sampled in 13 rivers spanning broad watershed land-use characteristics during 2009–2010. Contrary to expectations, freshwater stream ecosystems in Puerto Rico were not severely polluted, likely due to frequent flushing flows and reduced deposition associated with recurring flood events. Notable exceptions of contamination were nickel in sediment within three agricultural watersheds (range 123–336 ppm dry weight) and organic contaminants (PCBs, organochlorine pesticides) and mercury in urban landscapes. At an urban site, PCBs in several fish species (Mountain Mullet *Agonostomus monticola* [range 0.019–0.030 ppm wet weight] and American Eel *Anguilla rostrata* [0.019–0.031 ppm wet weight]) may pose human health hazards, with concentrations exceeding the U.S. Environmental Protection Agency (EPA) consumption limit for 1 meal/month. American Eel at the urban site also contained dieldrin (range < detection-0.024 ppm wet weight) that exceeded the EPA maximum allowable consumption limit. The Bigmouth Sleeper *Gobiomorous dormitor*, an important piscivorus sport fish, accumulated low levels of organic contaminants in edible muscle tissue (due to its low lipid content) and may be most suitable for human consumption island-wide; only mercury at one site (an urban location) exceeded EPA's consumption limit of 3 meals/month for this species. These results comprise the first comprehensive island-wide contaminant assessment of Puerto Rico streams and biota and provide natural resource and public health agencies here and in similar tropical islands elsewhere with information needed to guide ecosystem and fisheries conservation and management and human health risk assessment.

1. Introduction

Environmental pollution is a problem for many tropical islands, especially those with dense human populations and competing land uses (Hunter and Arbona, 1995). Few contaminant studies have been conducted in the tropical Caribbean islands (Rodríguez and Pérez de González, 1981; Neal et al., 2005), and there have been no studies to date on the occurrence and effects of contaminants in freshwater stream ecosystems in this region. More knowledge is needed on the occurrence and patterns of contaminants in Caribbean freshwater stream ecosystems to inform fisheries and natural resource conservation and management and human health risk.

The Caribbean island of Puerto Rico is densely populated, supporting nearly 440 people per square kilometer, which provides an appropriate model to study human-influenced aquatic contaminants (Martinuzzi et al., 2007; Neal et al., 2009; Kwak et al., 2016). During

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the past century, rapid industrialization and the subsequent human population growth have strained the limited natural resources of the island (Hunter and Arbona, 1995). Most of the rivers have been transformed by dam construction or other structures that are conducive for water collection (Cooney and Kwak, 2010, 2013; Kwak et al., 2016). However, the residents of Puerto Rico are becoming increasingly aware of the benefits associated with conserving stream ecosystems. González-Cabán and Loomis (1997) demonstrated that citizens would be willing to pay a total of $11.33 million to prevent dam construction on Río Mameyes, the last remaining free-flowing river in Puerto Rico.

The streams of Puerto Rico provide many services for local populations, including water for drinking, recreation, irrigation, and as a source of fish and crustaceans for consumption. Good water quality is necessary to protect human health as well as ecological integrity. However, Puerto Rico has experienced an era of rapid and fluctuating human population growth leading to deteriorated water quality (Hunter and Arbona, 1995; Fitzpatrick and Keegan, 2007). The streams have a history of die-offs of fish, shellfish, and shrimp, resulting from contamination by industrial, agricultural, and municipal wastes, and epidemiological evidence also suggests that water contamination has jeopardized human health (Hunter and Arbona, 1995; Colón et al., 2000). Additionally, some human populations in Puerto Rico have high marine fish consumption rates, potentially exposing them to high levels of contaminants such as metals (Mansilla-Rivera and Rodríguez-Sierra, 2011). Yet, there is a notable lack of research and available information on the degree and effects of water contamination in Puerto Rico and associated human risks from consumption of native freshwater fish, and no Commonwealth agency is mandated with developing fish and shellfish consumption advisories.

Rivers and streams are influenced by their surrounding landscapes (Vannote et al., 1980; Allen, 2004). Direct correlations have been clearly demonstrated between land use and water quality (Lenat and Crawford, 1994; Bolstad and Swank, 1997; Fisher et al., 2000; Tong and Chen, 2002). Surface runoff, especially after a drought, is a major contributor to non-point source pollution because it transports sediment and associated chemicals into aquatic ecosystems. Runoff from varying types of land use is enriched with different contaminants; for example, runoff from urban areas may be enriched with rubber fragments and heavy metals from vehicles, whereas runoff from agricultural lands may be enriched with fertilizers and pesticides (Lenat, 1984; Osborne and Wiley, 1988; Cooper, 1993; Johnson et al., 1997; Tong and Chen, 2002). Further, vegetation modifies land surface characteristics, water balance, and the hydrologic cycle through evapotranspiration, interception, infiltration, percolation, and absorption (Tong and Chen, 2002). Human-altered land use also transforms the hydrological system by changing runoff patterns and composition and quality of receiving water bodies (Changnon and Demissie, 1996; Mander et al., 1998; Warne et al., 2005).

Puerto Rico has undergone a number of anthropogenic alterations to its landscape as a result of agriculture, deforestation, stream channelization, industrial and municipal pollution, urbanization, and impoundment of rivers (Neal et al., 2009; Kwak et al., 2016). Historically, Puerto Rico's economy was predominantly agricultural, but in the early 1900s, global markets changed and the economy shifted toward industry and tourism (Hunter and Arbona, 1995). While rapid industrialization of Puerto Rico most likely led to an increased influx of a variety of contaminants into the environment, tourism relies upon clean waters, beaches, and other minimally disturbed areas, such as the El Yunque National Forest. Therefore, it is imperative that contaminants and water quality be assessed in Puerto Rico stream ecosystems to guide natural resource planning and economic development.

The aim of this study was to quantify contaminants in water, sediment, and biota in the stream ecosystems of Puerto Rico across a spectrum of watershed land-use patterns. This was accomplished in two phases; first by extensively sampling and analyzing habitat and biota island-wide for contaminants, and second by conducting focused, intensive studies in selected areas of management and human health importance.

2. Methods

This research began with an island-wide study of 13 of the 46 streams (hereafter, extensive study) encompassing a variety of land-use categories. Following the extensive sampling, more intensive sampling was conducted on one representative stream from each land-use classification (4 sites; hereafter, intensive study). Prior knowledge of target species distribution and abundance was provided by Kwak et al. (2007). Rivers were sampled during the summers of 2009 (extensive study) and 2010 (intensive study), and sites were categorized based on primary watershed land-use patterns or distinctive riparian features, including one reference site within a protected National Forest watershed [Río Mameyes (1R)], two industrial sites [Río Tallaboa (2I), Río Cañas (3I)], two urban sites [Río Piedras (4U), Río Bayamon (5U)], eight agricultural sites [Río Añasco (6A), Río Yauco (7A), Río La Plata (8A), Río Jacaguas (9A), Río Guanajibo (10A), Río Cartagena (11A), Río Arecibo (12A), and Río Fajardo (13A), a site with substantial recreational fishing activity] (Fig. 1). This study provided a 28% coverage of all river drainages in Puerto Rico, and because of its broad spatial coverage and sampling of its dominant land use types, it characterizes the potential pollution conditions from other riverine systems on the island.

Data from the initial extensive contaminant sampling (13 sites) were used to select sites and contaminants for additional intensive sampling, which included additional species and sample replicates. Four sites, among those sampled in the extensive contaminant survey, were selected for intensive contaminant investigation in 2010 to represent specific water quality or watershed land-use effects. These sites were 1R (reference), 7A (agricultural), 3I (industrial), and 4U (urban). Water, sediment, and biota were sampled at each site.

Physicochemical characteristics of water were measured at each site with a Yellow Springs Instrument (YSI) 556 multi-probe system and a Hach CEL/850 Portable Aquaculture Laboratory and included temperature, pH, alkalinity (mg/L $CaCO_3$), total hardness (mg/L $CaCO_3$), conductivity (μS/cm), nitrate concentration (μg/L NO_3^-), nitrite concentration (mg/L NO_2^-), and orthophosphorus concentration (mg/L PO_4) for both the extensive and intensive studies. Water was collected using a 1-L container, rinsed repeatedly with site water, and then was submersed 0.25–0.50 m beneath the water surface, filled, and stored on ice in a cooler.

2.1. Universal passive sampling devices (uPSDs)

Time-integrated contaminant concentrations in water were sampled using Universal Passive Sampling Devices (uPSDs) (Hirons, 2009). Passive sampling devices are an efficient method for sampling and measuring water contaminants (Heltsley et al., 2005). They estimate ecologically relevant contaminant exposure (Hirons, 2009) and bioconcentration for aquatic species (Heltsley et al., 2005). uPSDs offer advantages over traditional grab sampling because they represent exposure of the bioavailable portion and they collect transient contaminants at trace levels (Hirons, 2009).

Two types of uPSDs were deployed in this study; fiber passive sampling devices (fPSDs) and cartridge passive sampling devices (cPSDs). The fPSDs have a surface area of 5.8 cm^2, and cPSDs have an internal surface area of 6.2 cm^2 (Hirons, 2009). The fPSDs are hollow, polyethersulfone fibers filled with Waters Oasis HLB® sorbent, with a diameter of 1 mm and pore size of 0.2 μm. The cPSDs are incased in porous stainless steel and filled with the same polymeric sorbent, Oasis HLB®. Three fPSDs were deployed at each site during the extensive survey and six cPSDs were deployed at each site during the intensive study. They remained submersed in the water for 3–4 weeks. Each uPSD was wrapped in aluminum foil immediately upon retrieval and placed inside a plastic bag with a label, indicating the retrieval date and time,