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Mercury in sediment, water, and fish in a managed tropical wetland-lake ecosystem

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HIGHLIGHTS

- Hg and MeHg concentrations in municipal wastewater significantly >natural waters.
- Despite high Hg inputs, lake water and sediment Hg concentrations were very low.
- Hg in tilapia and carp were extremely low and did not exceed consumption advisories.
- Wetlands surrounding the lake reduced Hg inputs, likely through particle settling.

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ABSTRACT

Mercury pollution has not been well documented in the inland lakes or fishes of Mexico, despite the importance of freshwater fish as a source of protein in local diets. Total mercury and methylmercury in waters, sediments, and the commercial fish catch were investigated in Lake Zapotlán, Mexico. Concentrations of total and methylmercury were very high in runoff and wastewater inputs, but very low in sediments and surface waters of the open water area of the lake. Concentrations of total mercury in tilapia and carp were very low, consistent with the low concentrations in lake water and sediments. Particle settling, sorption, the biogeochemical environment, and/or bloom dilution are all plausible explanations for the significant reductions in both total mercury and methylmercury. Despite very high loading of mercury, this shallow tropical lake was not a mercury-impaired ecosystem, and these findings may translate across other shallow, alkaline tropical lakes. Importantly, the ecosystem services that seemed to be provided by peripheral wetlands in reducing mercury inputs highlight the potential for wetland conservation or restoration in Mexico.

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1. Introduction

Mercury (Hg) is an environmental pollutant of global concern with the exposure pathway for humans being largely through the consumption of fish (Dabeka et al., 2003; Hightower and Moore, 2003; Health Canada, 2007). In freshwater ecosystems, the vast majority of Hg research has been conducted in northern temperate catchments that are not impacted by point source discharges of Hg. Mercury pollution in freshwaters of the lower latitudes is less well documented with the exception of an important body of literature on Hg in Amazonia (e.g. Guimaraes et al., 2000; Roulet et al., 2001a,b; Berzas Nevado et al., 2010). There is little data on Hg concentrations in sediments, surface waters, and fish from tropical lakes and wetlands in Mexico. A report from the Commission for Environmental Cooperation (2013) provided a synthesis of THg in fish from Mexico, however only two datasets (one of which is this study) were for freshwater fish species commonly consumed by humans. Given that many of Mexico's waterways are heavily polluted with industrial and urban effluent (Sanchez, 1995; Soto-Galera et al., 1998; Avila-Pérez et al., 1999, 2006; Cortés et al., 2004; Fall et al., 2007), concern for Hg contamination and its associated human health risks is justified. The discharge of untreated municipal wastewater into adjacent waterways is an ongoing water quality issue in Mexico (Sanchez, 1995; Fall et al., 2007), and untreated municipal wastewater can have high concentrations of Hg and methylmercury (MeHg), largely associated with suspended solids (Balogh and Liang, 1995; Mugan, 1996). While a number of previous studies in the USA and Canada have investigated the effectiveness of Hg removal by wastewater treatment works, to our knowledge no investigations in Mexico have been made regarding Hg impacts to water quality and aquatic

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biota from untreated wastewater discharges. Certainly several studies of Hg concentrations in fish from coastal Mexico (Reimer and Reimer, 2005; Ruelas-Inzunza et al., 2007) and Lake Chapala (Ford et al., 2000; Trasande et al., 2010) report fish muscle tissue that exceeded consumption guidelines, however no direct links to Hg sources or transformations have been made.

The catch in Mexico's freshwater fisheries is dominated by introduced African tilapia (*Oreochromis* spp.), and Mexico is one of the largest producers in the Western Hemisphere (Fitzsimmons, 2000). The Common Carp (*Cyprinus carpio*) is also regularly stocked and fished from freshwater lakes in Mexico. Both tilapia and carp are some of the most important groups of farmed fish globally (Wang et al., 2010). While both tilapia and carp are generally regarded as lower risk fish for mercury exposure (Zahir et al., 2005; Levenson and Axelrad, 2006) studies from Lake Chapala in Mexico (Trasande et al., 2010) and Nicaragua (McCrary et al., 2006) have demonstrated that these species can accumulate Hg to advisory levels (>0.3 μg/g). While there are few data regarding Hg in commercially fished freshwater species in Mexico, widespread water quality impairments from municipal effluent discharges are conducive to Hg exposure risk through the consumption of fish.

The primary objective of this research was to assess the risk of MeHg exposure to human and other consumers of fish from a small shallow alkaline tropical wetland/lake in west-central Mexico. This lake (Lake Zapotlán, Jalisco) is impacted by urban runoff and wastewater discharges, sediment-laden surface flows from agricultural and forestry activities, and proximity to an active volcano which may be an additional source of geogenic Hg. We hypothesized that these factors would result in waters and sediments elevated in both total mercury (THg) and MeHg, and fish tissue to be elevated in THg (largely as MeHg). Further we hypothesized that the extensive marsh-type wetland fringe would contribute to further production of MeHg, exacerbating Hg exposure in an already Hg-contaminated ecosystem.

To test these hypotheses, THg and MeHg in water and sediments of the wastewater discharges, seasonal surface runoff, wetlands, and the open lake were measured spanning a complete hydrological year (wet season and dry season) over 2007–2008. THg in the muscle tissue of tilapia and carp from the local fishery were investigated to assess MeHg bioaccumulation in this system and the risk to consumers.

2. Methods

2.1. Study Site

The tropical endorheic basin (45,000 ha) of Lake Zapotlán, Mexico (19° 34′–19° 53′ N, 103° 24′–103° 39′ W) sits at an elevation of 1497 m and is home to a largely urban population of 150,000, the majority residing in Ciudad Guzmán (Fig. 1). The lake is shallow (mean 1.7 m), eutrophic, and alkaline (Ortiz-Jiménez et al., 2005, 2006; Greenberg et al., 2008), and surrounded by a wetland fringe dominated by *Typha* spp. At times, floating mats of the invasive Water Hyacinth (*Eichhornia crassipes*) are also present in the wetland fringe. Historically, these mats can become quite extensive; however, mechanical removal from open water areas has limited annual Hyacinth coverage to <5% (Ortiz-Jiménez et al., 2005). The construction of several municipal roads has physically segmented the lake into three regions, and caused changes in morphometry, hydrodynamics, aeration, and water quality of the lake (Fig. 1). The region has a distinct wet season from June to September with average annual rainfall of 813 mm and an average annual air temperature of 19.6 °C (Ortiz-Jiménez et al., 2005). Water inputs to the lake were modeled by Ortiz-Jiménez et al. (2005) and comprise ephemeral natural surface runoff (53%), direct precipitation (22%), groundwater input (14%) and discharges from wastewater treatment plants (11%). Despite dominating the hydrological inputs, flows in numerous natural ephemeral streams from the watershed typically occur only during the late wet season in August and September. The spatial distribution of these channels over the mountainous watershed, and the high temporal variability in flows make their quantification from a mass balance perspective challenging, and although there is recent work on hydrological processes governing runoff (Farrick and Branfireun, 2014), there are no empirical data on the total annual flows so we must rely on the estimates of water and sediment fluxes presented in Ortiz-Jiménez et al. (2005). Treated and untreated wastewater streams from the municipalities of C. Guzmán and San Sebastian del Sur are a smaller fraction of total volumetric flow, but are the only year-round surface runoff inputs. Equipment maintenance and operational issues resulted in the periodic but often frequent discharge of untreated urban wastewater making up most or all of this flow to the lake and surrounding wetlands (up to 10,300 m^3/d from 1982 to 2003; Ortiz-Jiménez et al., 2006).

The lake supports a local fishing economy for African tilapia (*Oreochromis* spp.) and the Asian common carp (*C. carpio*). Both species were introduced to Mexico in the 1960–80s as an inexpensive source of protein (Fitzsimmons, 2000; Tapia and Zambrano, 2003). Tilapia and carp fished from Lake Zapotlán are sold locally and regionally in Guadalajara, Michoacán, Toluca, and Puebla states. Tilapia and carp make up an estimated 68% and 32% of the total catch respectively (Ortiz-Jiménez et al., 2005).

2.2. Sampling and analyses

Sampling locations, the extent of the lake and wetlands, and locations of wastewater inputs to Lake Zapotlán are shown in Fig. 1. Samples were taken from a total of nine lake locations, three untreated wastewater streams, three wetland zones adjacent to wastewater outfalls and four ephemeral stream locations under stormflow conditions. The three wetland zones adjacent to the wastewater inputs were sampled intensively during the June/July 2008 event to assess potential gradients of sediment and water contamination.

2.2.1. Water sampling

Sampling campaigns were conducted in October 2007 (late wet season), February 2008 (mid dry season), and in June/July 2008 (early-mid wet season). All sampling was performed using ultra-clean protocols. Technicians were gloved in the field with powder-free nitrile gloves using what is commonly referred to as the clean hands, dirty hands method (U.S. EPA, 1996). The lake is considered well-mixed most of the year due to the combination of shallow water and wind (Ortiz-Jiménez and de Anda, 2007); therefore, only surface water was sampled from the lake. Surface water samples were taken in sterile and clean 250 mL PETG bottles either as a grab sample or using an acid-washed Teflon® sampling line and peristaltic pump. Separate samples were taken for both filtered and unfiltered water samples. Samples for dissolved Hg analysis were filtered using an acid-cleaned Teflon® filter apparatus (Savillex Inc.) and pre-muffled (500 °C) glass-fiber filters (Whatman GFF 0.7 μm). Water samples were acidified to 0.5% (by volume) using ultrapure trace metals grade hydrochloric acid.

Runoff waters from four sampling locations along natural ephemeral streams were sampled following two storm events in July 2008. Rainwater during these events was collected using a 0.25 m diameter acid-washed Teflon® funnel that delivered rainwater directly into a 250 mL PETG bottle. For both storm events, two samples of rainwater were collected. Runoff/rainwater samples were acidified to 0.5% (by volume) using ultrapure hydrochloric acid.

2.3. Sediment sampling

Lake and wetland sediments were collected by boat and on foot using a small (15 × 15 × 15 cm) Eckman bucket sampler and/or manual sediment coring by hand using clear acid-washed PVC tubing. The sample was returned to the surface, and the top 4 cm were subsampled (in duplicate) and transferred to a small leak-proof zip-closure bag using a

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