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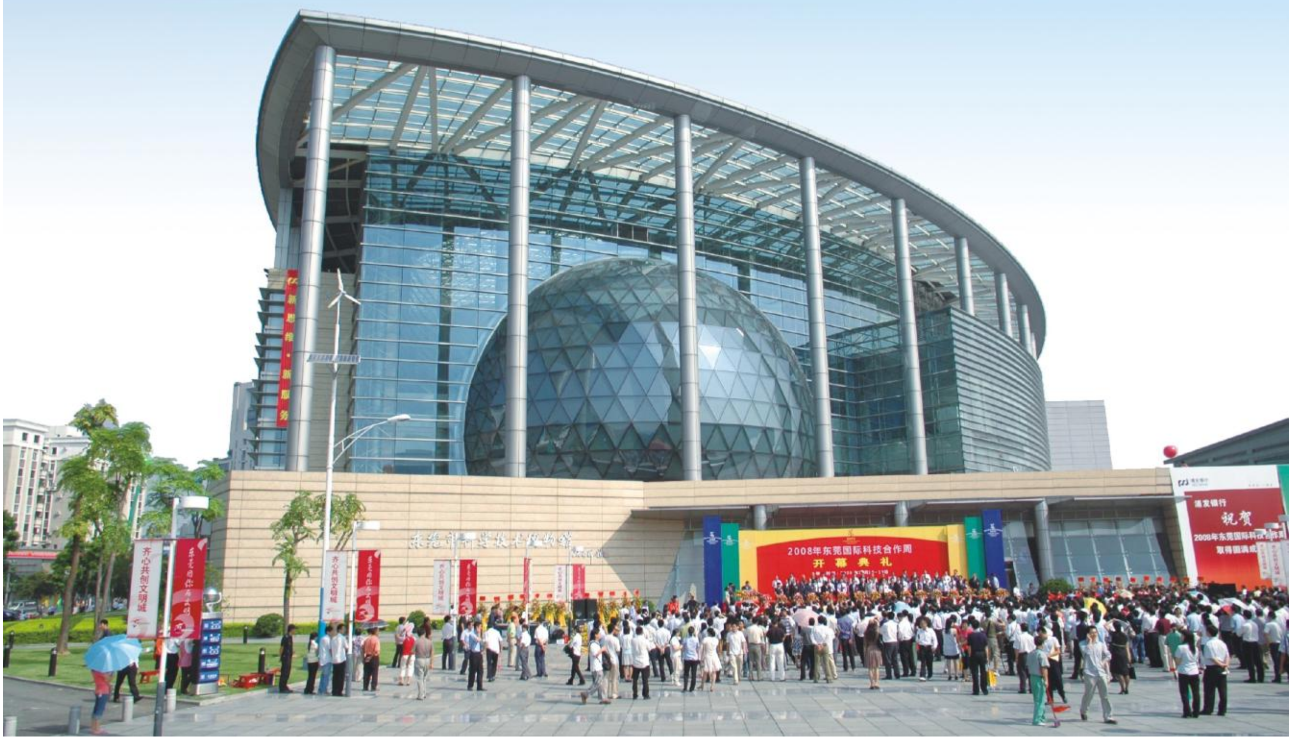

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Tracking Down an Unusual Source of Mercury Enrichment in Fish from Saipan Lagoon, Saipan, Commonwealth of the Northern Mariana Islands

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Abstract: In 2004-2005, several species of fish were collected for mercury analysis from Saipan Lagoon. The highest overall levels, while not excessive, were found in representatives collected from *Hafa Adai Beach,* located some distance from known sources of mercury contamination. A follow-up investigation, aimed at identifying land-based sources of mercury in this particular area, was launched in 2007. To this end, sediment and soil samples were respectively collected from the beach and all potential drainage pathways to the coast. Bivalve mollusks and two types of fish with limited foraging ranges were also collected from the focus area and from other sites to the north and south. Marginally elevated mercury levels were detected in sediments at the mouth of a storm drain at the southern end of *Hafa Adai Beach* and prompted additional soil sampling further inland along a drainage canal. Mercury concentrations in these samples generally increased from around 30 ng/g (parts per billion dry weight) at the mouth of the storm drain to approximately 200 ng/g at the *Middle Road* entrance to Saipan's only public hospital, about 1 km away. Additional soil sampling along a small drainage ditch within the hospital grounds revealed mercury levels in excess of 1000 ng/g a few meters down gradient of an old incinerator site. The incinerator was operated by the hospital for approximately 20 years before it was finally dismantled in early 2006. Bivalves collected from the intertidal zone and nearshore waters down gradient of the impacted storm drain contained mercury levels that were unremarkable. Fish captured from nearshore seagrass beds in the same area and from coral patch reefs further offshore generally revealed lower mercury levels than their counterparts sampled three years earlier. Marine sediments taken from each of these habitats yield mercury values within normal ranges. The implications of the data are discussed both from an environmental and human health perspective.

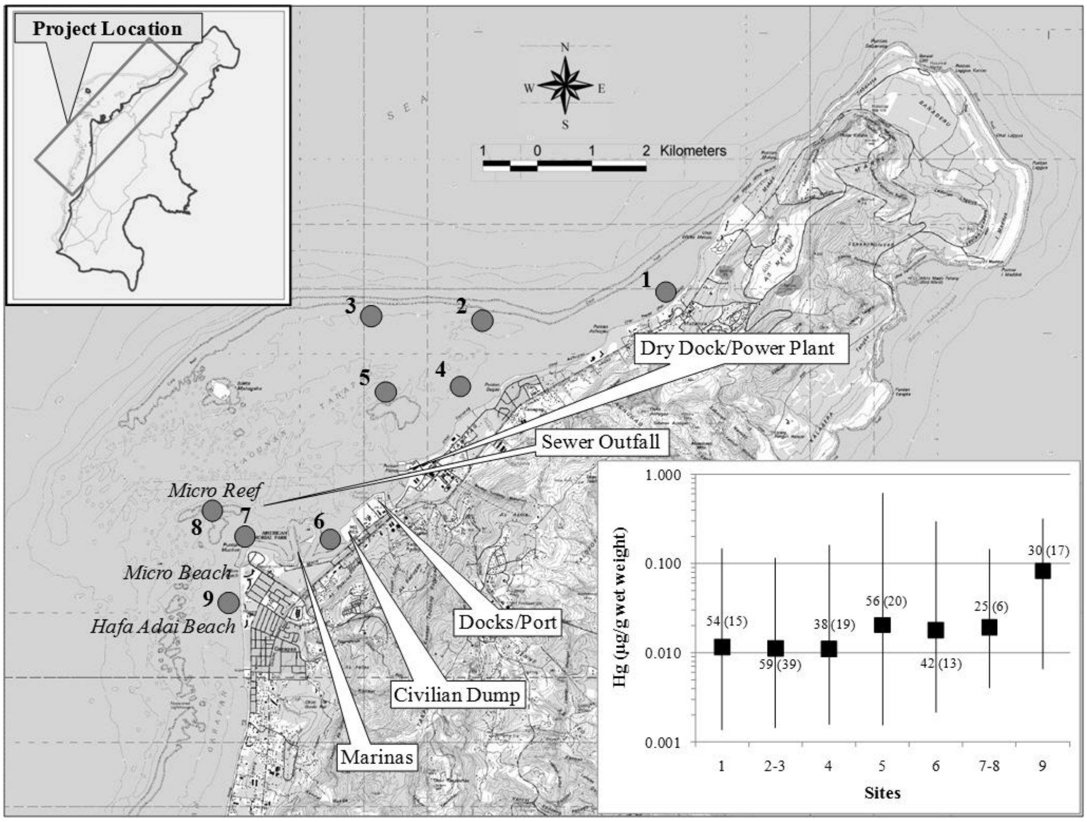
Keywords: Saipan; mercury; fish; sources and status

1 Introduction

Saipan Island (15°18'0.93"N, 145°75'59.97"E) is located in the Western Pacific Ocean, approximately 200 km north of Guam, within the Mariana Archipelago. The island is about 20 km long and 9 km wide at its widest point. It has a total land mass area of approximately 115 sq km, and is the second most densely populated island in Micronesia. While the eastern side of Saipan is composed primarily of rugged rocky cliffs, a barrier coral reef system on the western side creates a large lagoon that extends almost the entire length of the island. This stretch of shallow water is locally referred to as Saipan Lagoon and contains large expanses of patch reef interspersed with sand and rubble. This in turn provides for a diversity of shallow water habitats that harbor rich assemblages of flora and fauna[1-2]. Aside from Saipan Lagoon's ecological significance, it also supports a variety of recreational activities and local residents traditionally harvest many of its fisheries resources at a subsistence level. Protecting and preserving this environment and its resources for future generations is, therefore, of considerable importance to the people of Saipan.

Prior to the last world war, Saipan was a small, rural community. Apart from extensive land clearing to make way for sugar cane plantations during the Japanese occupation and the traditional harvesting of nearshore fisheries resources for food, anthropogenic impacts on the island were relatively minor. Sources of pollution were predominantly associated with the disposal of domestic wastes from small settlements located around the coast. As a result, Saipan's coastal waters were relatively pristine from a water quality standpoint. Today, things are somewhat different, particularly on the western side of the island where the bulk of the population now exists. This area has undergone considerable urban growth and economic expansion in recent years. Such development has greatly added to the waste disposal, urban runoff, chemical pollution and environmental management problems that the island currently has to deal with. The shoreline running along the northern half of Saipan Lagoon, for example, is replete with pollution sources that have significantly impacted water quality over the years. These include a major sea port, two small boat marinas, bulk fuel holding facilities, two sewer outfalls, the largest power plant on island, several large garment factories (all now closed), auto and boat repair shops, junk yards, government vehicle maintenance yards and storage areas for old lead-acid batteries, PCB-laden electrical transformers and waste oil, and a municipal dump (also now closed) that is rumored to contain a plethora of hazardous chemicals of both military and civilian origin[3].

Only recently have we started to gather fundamental data describing the abundance and distribution of persistent and potentially toxic pollutants within Saipan Lagoon. Contaminant assessments of surface sediments and dominant ecological representatives within the northern half of the lagoon were initiated in 2000 and 2003, respectively[4-7]. These studies identified areas of heavy metal enrichment in both environmental compartments around the power station, port and municipal dump. In 2004-2005, over 300 popular food fish (65 different species) were collected for mercury analysis from nine sites within this region (Fig. 1). While values in fish axial muscle were marginally elevated in representatives from around the municipal dump (Site 6), concentrations, though not excessive, were generally higher in specimens taken some distance to the south at Site 9[8]. This particular stretch of beautiful Saipan coastline is known as *Hafa Adai Beach* and partially boarders the village of Garapan in the tourist center of the island. It conjoins picturesque *Micro Beach* a half km or so to the north and is a popular fishing, relaxation and recreational area for tourists and local residents alike. The relatively high mercury values found in fish from this area were of interest considering the aesthetic beauty of the location and the relatively large distance separating it from obvious pollution sources. Subsequently, a preliminary investigation was undertaken to search for significant land-based sources of mercury in the area and determine their impact on adjacent fisheries resources. To this end, levels of mercury were determined in beach sediments from *Hafa Adai Beach* to the small boat marinas further north (Fig. 1). Sediments and soil samples were also collected from all major surface water drainage pathways to the coast. Additionally, biotic representatives popularly harvested for food by island residents were collected for analysis from *Hafa Adai Beach* and several other sites to the north and south. The findings of the study are presented here and are discussed from an environmental and human health perspective.

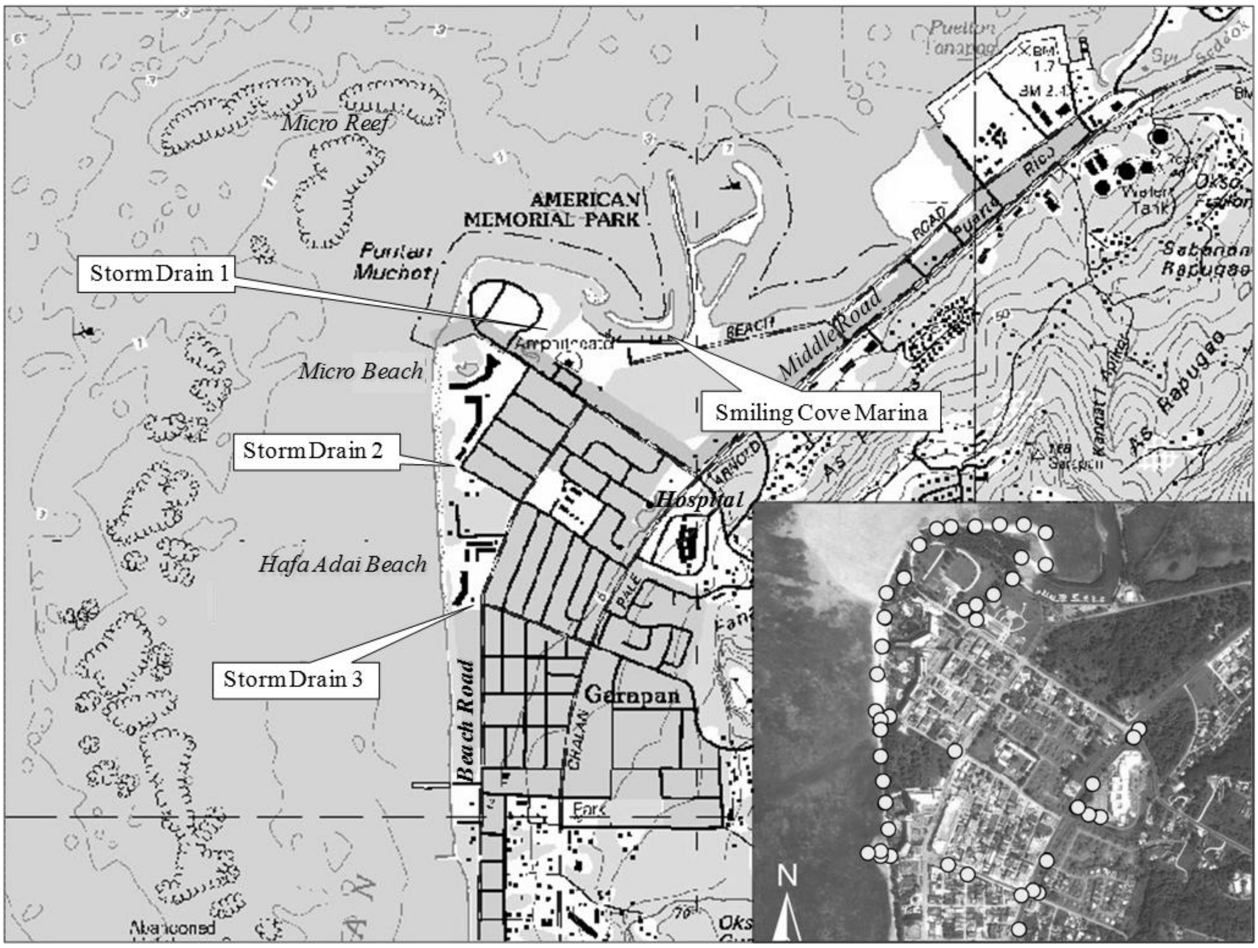


Bottom inset shows geometric means and ranges of mercury concentrations determined in fish from each site. Numerical values are total numbers of fish (and species) analyzed from each site. Potential sources of mercury pollution known to exist at the time of the investigation are also shown

Fig. 1 Portion of a 1:25,000 United States Geological Survey (USGS) Topographical Map of Saipan showing fish sampling sites 1-9 in the northern half of Saipan Lagoon

2 Materials and Methods

In April 2007, beach sands from the lower intertidal region were collected for mercury analysis at ~100 m intervals along the coast, from *Hafa Adai Beach* to the western entrance of *Smiling Cove Marina*, approximately 2 km to the northeast (Fig. 2). Samples from all sites were predominantly composed of clean, biogenic carbonates of marine origin with minimal terrigenous material. Surface deposits were also taken from three storm water drains in the area, including two that discharged at the northern and southern ends of *Hafa Adai Beach* (Fig. 2). Samples from these locations were visibly higher in lithogenic material and organic matter. Over the next four months, soil samples were taken further upstream in stormwater catchments that warranted further investigation. Sample characteristics here ranged from fine muddy sands to coarse textured loams primarily of volcanic origin. All samples were scooped directly into acid-washed, polyethylene vials using a clean, stainless steel trowel and placed in chilled containers for transportation to the laboratory. Site locations were fixed using GPS and later transferred to satellite imagery maps using GIS technology. All sampling site locations are clearly shown in Fig. 2 (inset).



Inset shows sampling sites superimposed upon a satellite imagery map of the area

Fig.2 USGS topographic map of Garapan village, Saipan, showing storm drain locations and known potential sources of mercury pollution relative to *Hafa Adai Beach*

In the laboratory, all soil and sediment samples were air dried at ~30^{o}C and sieved through a 1-mm nylon screen prior to digestion in hot nitric acid for 3-h. They were then analyzed for total mercury by flameless atomic absorption spectroscopy (AAS) using the syringe technique described by Stainton[9]. All calibration standards (5-20 ng/L) for mercury were made up in 10% nitric acid containing 0.05% potassium dichromate as a preservative[10].

The biotic representatives examined included three species of bivalves (*Atactodea striata*, *Ctenna bella,* and *Gafrarium pectinatum*) and two types of fish (the thumbprint emperor, *Lethrinus harak,* and the squirrel fish, *Myripristis* spp.). Of the bivalves, *Atactodea striata* were found in relatively clean, medium coarse sand in the lower intertidal region of the lagoon, while *Ctenna bella* and *Gafrarium pectinatum* generally preferred the muddier substrates of nearshore seagrass beds (*Enhalus acoroids*). All three species were collected between July and August, 2008, from the *Hafa Adai Beach* area, and from a number of other sites adjacent to storm drain discharge points further south. They were held in clean seawater for 48 hours to allow them to purge themselves of their gut contents and were then stored at -20^{o}C prior to mercury analysis of their bulk soft tissues.

The two fish types examined were selected for study because of their restricted foraging ranges and their popularity among local fishermen. Although they are both relatively abundant throughout the study area, they occupy very different niches and have very different food preference and feeding habits. *Myripristis,* for example, are highly territorial, nocturnal planktivores with limited home ranges and are exclusively associated with coral reefs. *Lethrinus harak*, on the other hand, is a roving carnivore, although it also feeds only at night. Members of this species migrate from nearshore to offshore waters during the day and return to the same patch of seagrass at night in search of invertebrates and small fish upon which they feed.

Myripristis were taken from patch reef areas within the lagoon using Hawaiian sling and spear-gun between May and December 2007. The lethrinids were captured over one year from February 2007 to February 2008, and almost all of them were taken by hook and line from nearshore seagrass beds at night. Both species were sampled from the *Hafa Adai Beach* area and several other sites to the north and south. All specimens were immediately placed in chilled containers for transportation purposes. In the laboratory, the fish were weighed and measured (fork length), and axial muscle samples were removed for mercury analysis from immediately below the dorsal fin on the left-hand side of the body. All dissections were performed with high quality stainless steel instruments and all tissues were stored at -20^{o}C until required for analysis.

The analytical procedure involved digesting ~1 g of wet muscle tissue in 10 ml of 2:1 concentrated nitric and sulfuric acids in 80-ml polypropylene tubes. The charged tubes were loosely capped with Teflon stoppers and allowed to cold digest overnight before refluxing at 100^{o}C for 3 hours. Upon cooling, the digests were topped up to 50 ml with distilled water and analyzed by flameless AAS as previously described. Mean recoveries of total mercury from a standard reference material (RM 50: Albacore Tuna) by this method were greater than 95%.

3 Results and Discussion

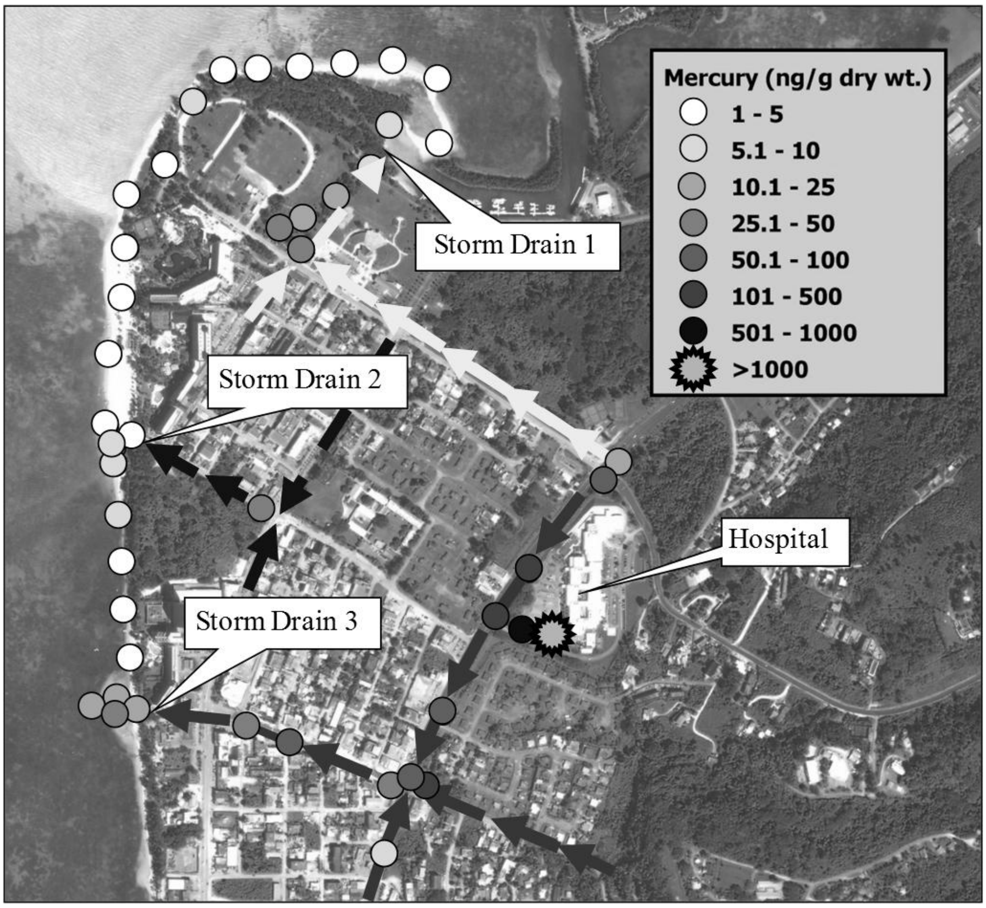
3.1 Abiotic Components

Total mercury levels found in the sediments and soil samples are summarized in Fig. 3. Levels found in beach sand were low and rarely exceeded 6 ng/g (parts per billion dry weight). Such values are typical of clean, bioclastic sediments[4,11]. Watershed soils collected further inland were more geologically enriched with mercury and yielded levels of 30-60 ng/g. Mercury concentrations in uncontaminated soils are usually less than 100 ng/g and typically ranging from 10-60 ng/g[12]. The sandy deposits at the mouths of storm drains 1 and 2 were unremarkable at 10 ng/g and 4 ng/g mercury respectively. In contrast, levels determined at the mouth of storm drain 3 at the southern end of *Hafa Adai Beach*, averaged 33 ng/g and were sufficiently elevated to warrant additional analysis of deposits further upstream in the drainage basin.

The outcome of this part of the study generally revealed increasingly higher soil mercury concentrations along an open drainage channel (grassy swale) in the direction of the Commonwealth Health Center, Saipan's only public hospital. Levels peaked at around 200 ng/g at the entrance to this facility and attenuated rapidly further north. Soil samples from a small drainage ditch inside the hospital grounds yielded even higher levels that exceeded 1000 ng/g a few meters down gradient of an old incinerator site at the southern end of the main building (Fig. 3). Apparently, the incinerator was used for the destruction of medical waste from the hospital and other medical clinics on island. It was closed down by the U.S. Environmental Protection Agency (USEPA) in 2006 for multiple violations of the Clean Air Act[13]. Up until that time, the incinerator had been operating for approximately 20 years.

Waste streams from hospital and health care facilities are typically high in mercury from dental wastes, broken thermometers and other medical devices, personal care products and medicinal compounds. Mercury amalgams used in tooth restoration, for example, are 50% mercury, and the organic mercury compound, *Thimerosal* ($C_9H_9HgO_2SNa$), has been used extensively as a preservative in a number of medicines, and is commonly found in waste streams from hospitals, clinical laboratories and pharmaceutical industries[14].

It is not clear why USEPA overlooked the potential cumulative impacts of the incinerator to soil in the area and marine water quality at the coast. Quite possibly mercury residues in the soil were considered insignificant when weighed against conventional remediation standards for this element in residential soils (typically 10-20 µg/g). Likewise, inland distance of the facility from the coast (~1 km) plus the fact that runoff from the hospital grounds flows into the grassy swale and percolates into the ground, may have tempered surface water quality concerns. Be that as it may, when soil infiltration rates are exceeded during heavy storm conditions, the excess runoff flows south along the swale into a storm drain network that eventually discharges highly turbid waters into the ocean at the southern end of *Hafa Adai Beach* (Fig. 3). The prevailing coastal currents in this area carry the discharge plumes northwards towards *Micro Beach* and seawards towards *Micro Reef*. Under normal climatic conditions, the plumes have little visual impact on coastal water quality much beyond 100 m south of the drain.



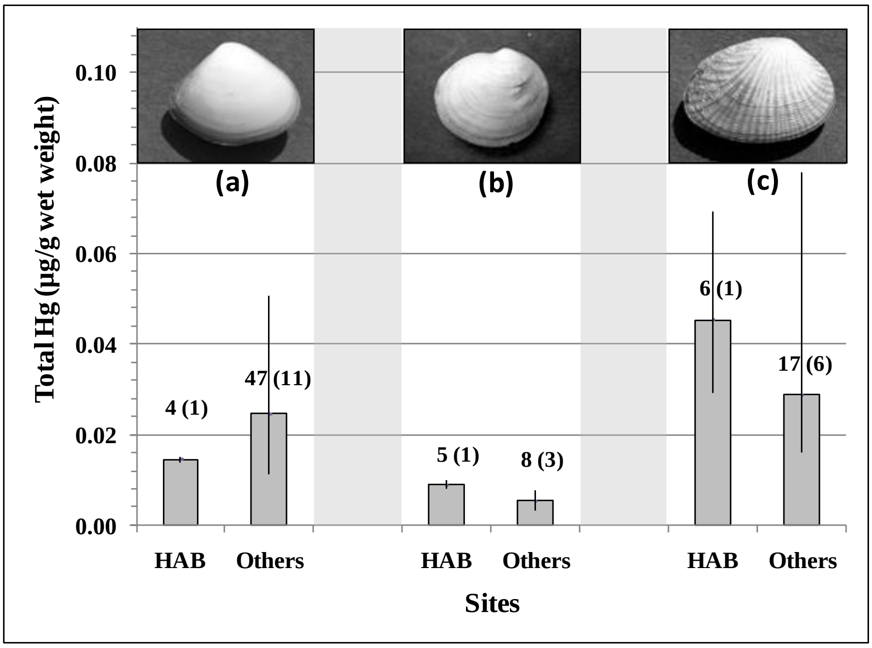
Arrows indicate direction of surface water drainage pathways within each catchment area

Fig. 3 Satellite imagery map of Garapan village, Saipan, and surrounding area showing mercury levels in beach sand, storm drain sediments and catchment soils

3.2 Biotic Components

Mercury levels in the bivalves examined are summarized in Fig. 4. Surprisingly, no statistically significant differences (P<0.05) emerged between representatives taken from *Hafa Adai Beach* and sites elsewhere in the lagoon. This strongly suggests that ambient mercury levels available to biotic components in the *Hafa Adai Beach* area have attenuated markedly since the incinerator was dismantled in 2006. Mercury turnover rates in bivalves are known to be relatively rapid with estimated half-lives reported in the literature ranging from 16 days to approximately 3 months[15-16]. Thus, mercury levels in bivalves from *Hafa Adai Beach* would essentially have returned to normal between the time the hospital ceased operating the incinerator in January 2006 and the time the bivalves were collected in August 2008.

While bivalve mollusks are certainly known to be excellent accumulators of mercury, levels in representatives from clean environments rarely exceed 0.100 µg/g (parts per million wet weight) and are usually well below this value[7,17-19]. The fact that the great majority of bivalves analyzed during the present study gave values lower than 0.050 µg/g, while none exceeded 0.080 µg/g, strongly suggests that mercury is no longer a contaminant of potential concern in this part of Saipan Lagoon.



(a) *Atactodea striata,* (b) *Ctenna bella,* and (c) *Gafrarium pectinatum* from *Hafa Adai Beach* (HAB) and other Saipan Lagoon sites (Others) to the south. Data are geometric means (bars) and ranges (whiskers). Total numbers of bivalves analyzed (and collection sites) also shown.

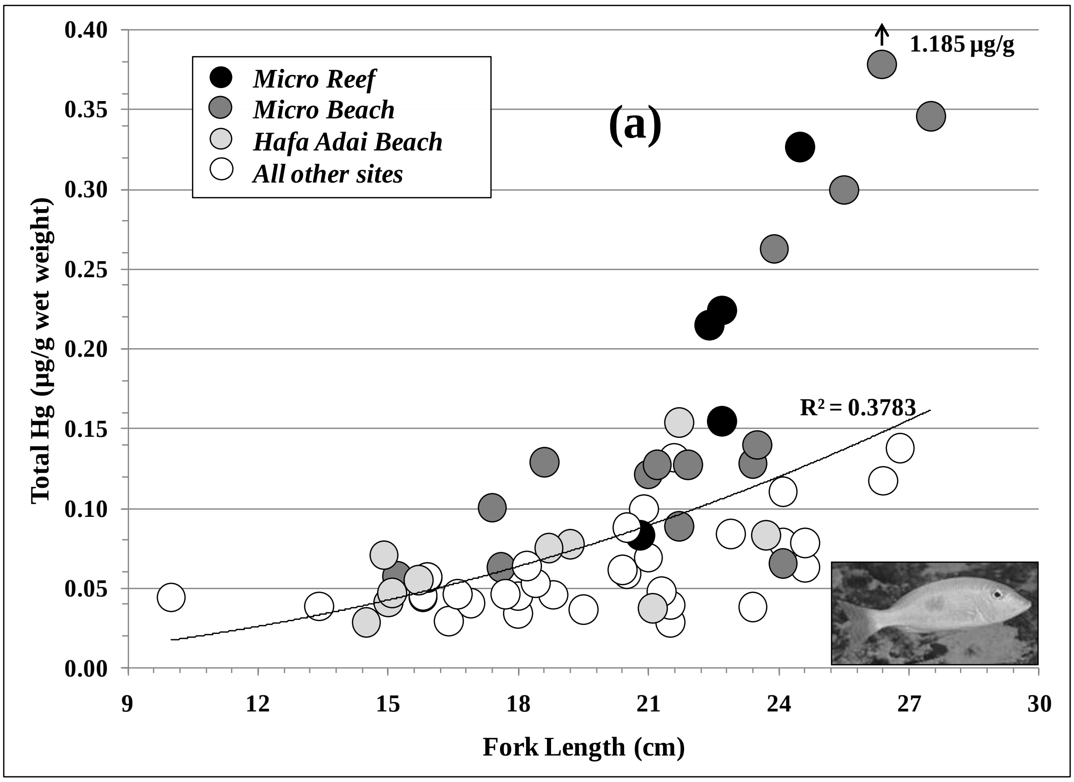
Fig. 4 Mercury levels in bivalves

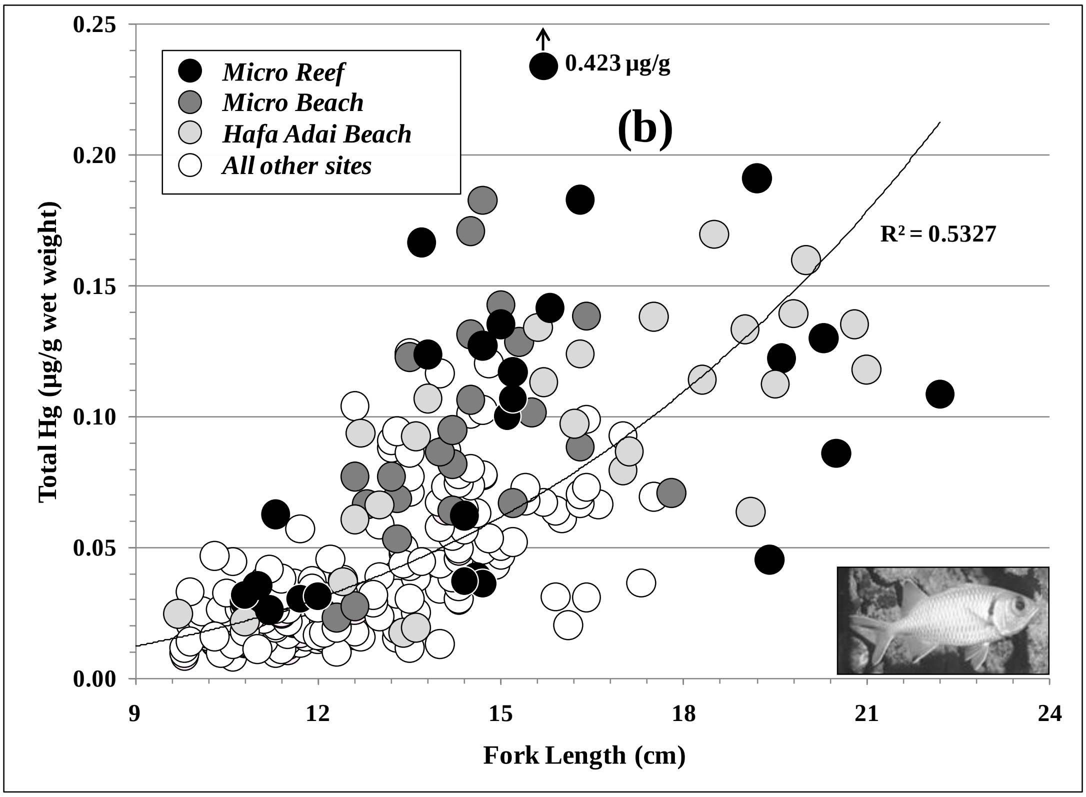
Scatter-plots of axial muscle mercury concentrations in the fish examined during the present study are given in Fig. 5. The preponderance of *L. harak* data points above the common regression line for samples from *Hafa Adai Beach, Micro Beach* and *Micro Reef,* implies fish were generally higher in mercury concentrations at these sites compared with equivalent sized individuals from elsewhere in the study area (Fig. 5a). Data point departures from the line also suggest the degree of enrichment was somewhat higher in specimens from the latter two sites. The *Myripristis* spp. data-sets generally show the same trend (Fig. 5b).

Mercury levels determined in fish from the *Hafa Adai Beach* area in 2004-5 and 2007-8 are compared in Fig. 6. The data again generally supports the contention that enrichment in biotic representatives from this part of the lagoon has declined since the hospital incinerator was shut down. This attenuation is more obvious in *Myripristis* spp. than *L. harak* and is thought to reflect trophic level difference between the two fish types. *Myripristis* spp. for example, derive their mercury load primarily from planktonic organisms that drift into their home ranges. Plankton, in turn, acquire their mercury loading from their aqueous surroundings. Any significant water quality changes occurring in a body of water will immediately be mirrored by plankton communities moving through it. Such changes will then be transmitted to organisms that consume them. *L. harak,* on the other hand, forages for invertebrates that live in seagrass bed sediments. Sediments generally serve as sinks for mercury and other pollutants. Any contaminant enrichment in this compartment is reflected to some degree by the resident biota. This may continue for some time after the source of contamination is removed and residual levels in the sediment slowly dissipate. Therefore, following the closure of the hospital incinerator, one might expect mercury levels at *Hafa Adai Beach* to attenuate far more rapidly in planktonic organisms in the area than in sediment dwelling biota. The bivalve and fish data presented here tend to support this argument.

The comparatively slow mercury clearance rates from fish compared with bivalves is to be expected. Long thought of as a first-order process, researchers are now of the opinion that mercury elimination in fish is biphasic, i.e., clearance occurs through fast and slow compartments. The elimination half-life for mercury ranges from days to months in the fast compartment and from months to years in the slow compartment. Chronically exposed fish (i.e., greater than 90 days) are thought to excrete mercury almost exclusively through the slow compartment at rates that are negatively correlated with size[20].

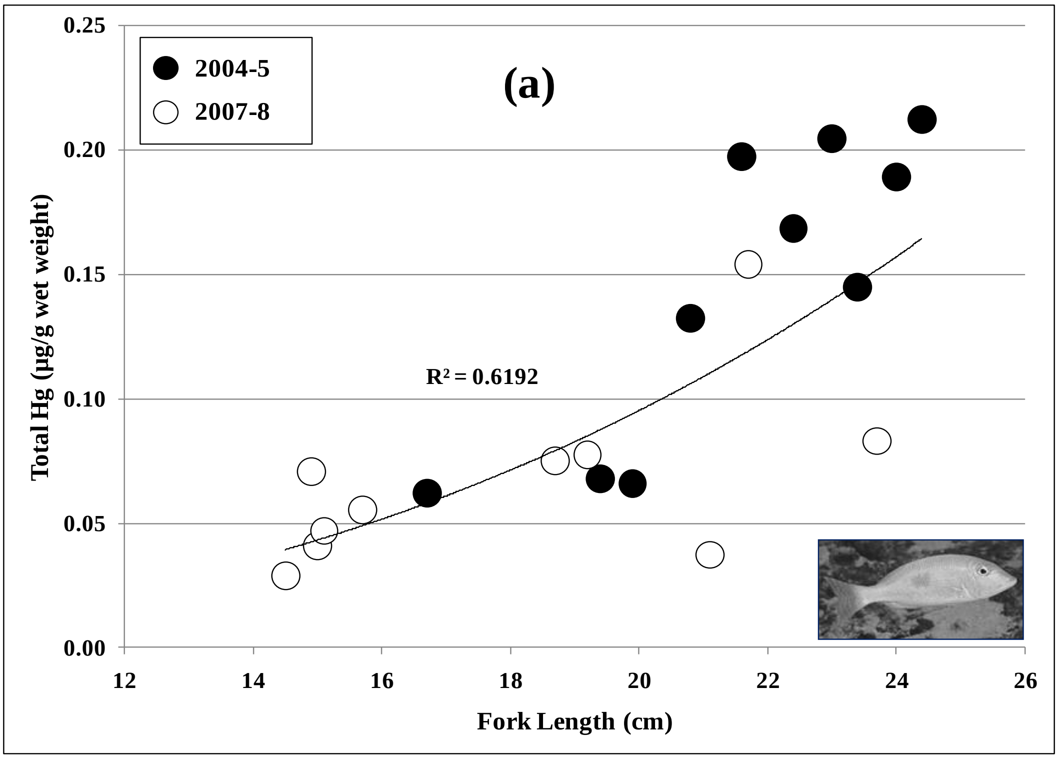
The generally higher mercury levels in both fish from *Micro Beach* and *Micro Reef* sites pose the question as to whether or not aquatic communities in these areas are being impacted by an additional mercury source (or sources). Significant mercury enrichment has previously been identified in bottom sediments adjacent to the dump and docks to the north[4,5]. The relatively close proximity of these facilities to the aforementioned sites certainly raises that possibility, particularly as the predominant currents in that part of the lagoon tend to move in a southerly direction.

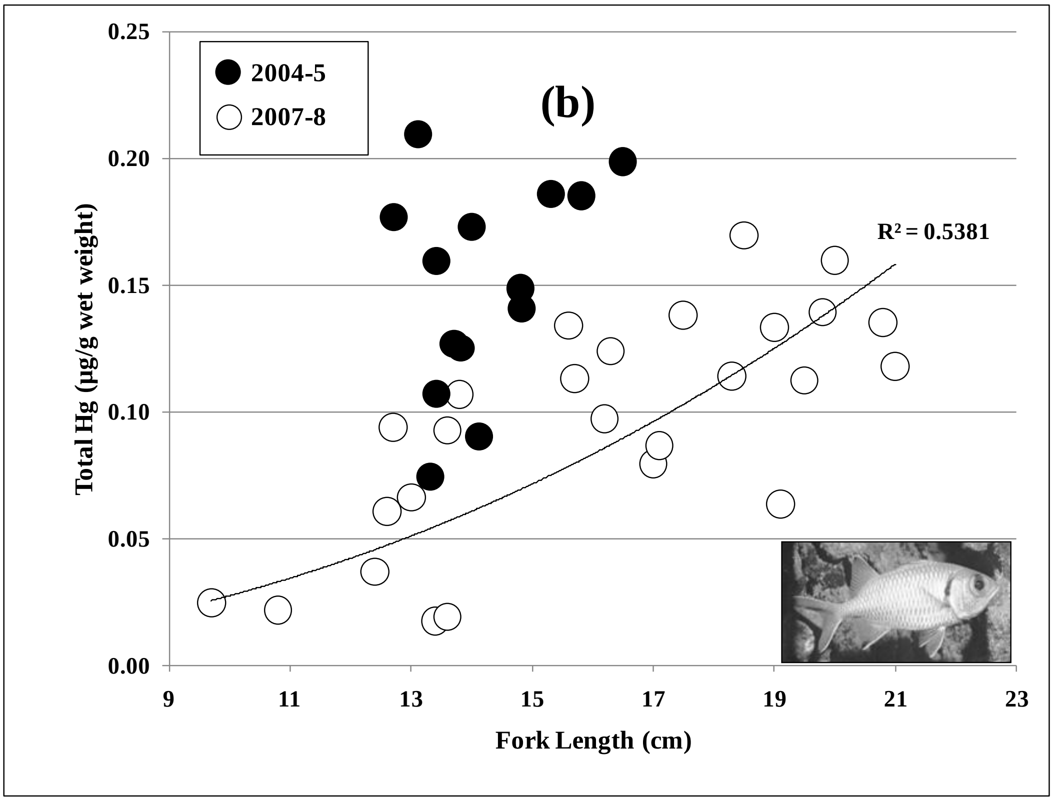




Power regression lines fitted to the pooled data-sets

Fig. 5 Scatter-plots of total mercury concentrations in axial muscle vs. body length of (a) *Lethrinus harak* and (b) *Myripristis* spp. from *Hafa Adai Beach* and other Saipan Lagoon sites to the north and south





Power regression lines were fitted to the pooled data sets in (a) and 2007-8 data-set in (b)

Fig. 6 Total mercury levels in axial muscle of (a) *Lethrinus harak* and (b) *Myripristis* spp. taken from *Hafa Adai Beach* a little over one year before and after the Saipan Commonwealth Health Center ceased operations of its medical waste incinerator

In 2007, a small number of surface sediment samples were taken from *Micro Reef* for mercury analysis and showed nothing unusual (all values less than 5 ng/g). Had the area been chronically impacted by a southerly drift of suspended sediments from the more mercury enriched areas further north, we almost certainly would have seen it in the analytical results. Of course, the fact that we did not does not preclude the possibility of mercury being imported into the area by planktonic organisms, as alluded to earlier.

3.3 Toxicological Significance

Mercury in fish occurs predominantly as methylmercury and usually accounts for 80-100% of total mercury concentrations[21-24]. Methylmercury can induce toxic effects in several organ systems including liver, kidney and the reproductives, and it is particularly toxic to the nervous system[25]. The neurotoxic effects associated with excessive methylmercury exposure include neuronal loss, ataxia, visual disturbances, impaired hearing, paralysis and death. The developing brain is thought to be the most sensitive target organ[25]. High methylmercury intake by pregnant women has also been linked to adverse effects in neurological development in children[26]. It is noteworthy that mercury has caused more problems to consumers of fish than any other inorganic compound[27].

Levels of mercury found in fish from non-polluted environments generally range between 0.001-0.100 µg/g on a wet weight basis[21,28], although higher concentrations can be expected in long-lived, predatory species such as marlin, tuna, and sharks[29,30]. In highly contaminated waters, mercury levels accumulated by fish may be orders of magnitude higher again. Fujika, for example, reported mercury levels exceeding 300 µg/g in fish from Minimata Bay, in Japan[31]. Such high levels are well above those considered safe for human consumption.

The U.S. Food and Drug Authority’s enforceable standard for methylmercury in fish sold commercially currently stands at 1.0 µg/g wet weight[32]. In the present study, only one fish exceeded this value, assuming total mercury levels to be approximately equal to methylmercury concentrations. Mercury levels in the great majority of other fish analyzed were less than 0.100 µg/g and, therefore, suggestive of a relatively clean environment overall.

The USEPA has developed risk-based consumption guidelines for a number of contaminants in fish. The methylmercury guidelines for the general population (as opposed to sensitive sub-groups) are briefly discussed here. They are based on a chronic reference dose (safe level) for methylmercury of $3x10^{-4}$ mg/kg body weight/day and a standard body weight and fish meal size of 70 kg and 8 oz (227 g fresh weight) respectively. They also take into account the tissue concentrations of mercury (as µg/g wet weight) in the fish consumed, as well as the number of fish meals eaten per month[33]. Standard sized fish meals with average mercury levels at or below 0.088 µg/g may be eaten on an unrestricted (daily) basis, while those with values ranging from >0.088-0.13 µg/g may be eaten four times a week without any long-term, adverse health effects. The relationship between tissue concentrations of mercury in fish and meal frequencies per month is illustrated graphically below (Fig. 7).

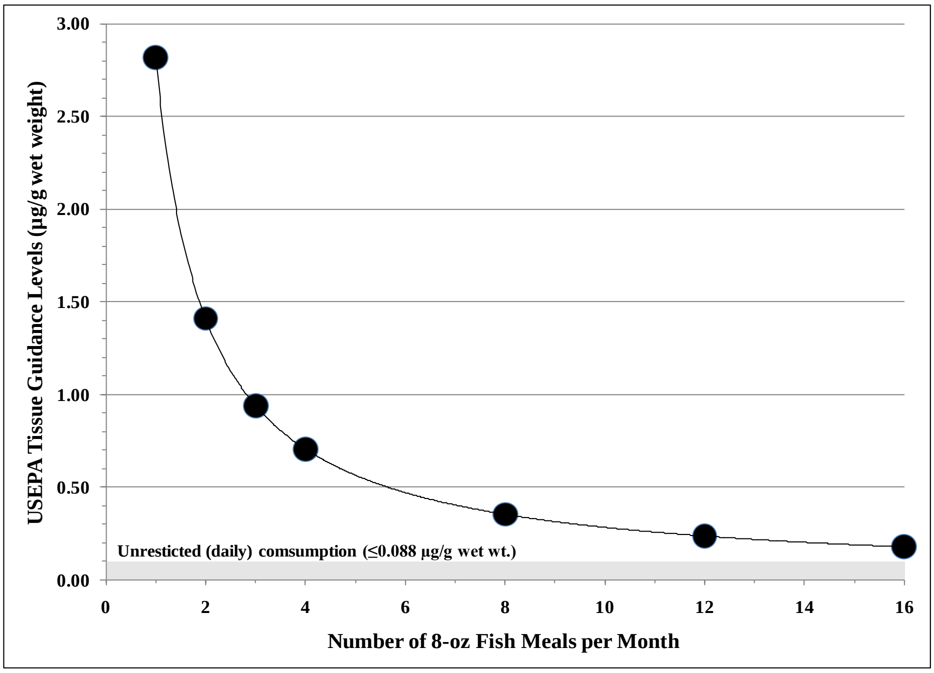


Fig.7 USEPA risk-based consumption guidelines for methylmercury in fish (USEPA 2000)

Using these USEPA guidelines, we conclude there is no significant health risk associated with consuming up to four 8-oz meals per week of *L. harak* or *Myripristis* spp. from anywhere within the study area, providing the size of individual fish that make up the meals are well mixed (i.e., composed of large and small specimens), and fishermen do not restrict their fishing activities to any of the higher risk areas mentioned here. For those who prefer fishing the *Hafa Adai Beach*, *Micro Beach* and *Micro Reef* areas, larger fish, especially carnivorous types like *L. harak,* should perhaps be consumed no more than three times per week as a precautionary measure for now.

4 Conclusions

The evidence presented here strongly suggests that the medical waste incinerator operated by Saipan's Commonwealth Health Center was responsible for the relatively high levels of mercury noted earlier in fish from *Hafa Adai Beach*[8]. Following closure of this waste disposal facility, levels transmitted to the coast in stormwater runoff from the hospital grounds appear to have attenuated rapidly resulting in a general cleansing of the lower reaches of the impacted drainage channel and adjacent nearshore environment. In 2007, for example, just 18 months after the incinerator ceased operating, we analyzed a limited number of surface sediment samples from seagrass beds down gradient of storm drain 3. Nothing unusual was found. Had there been any evidence of mercury enrichment in this particular environment during the life span of the incinerator, it had long since disappeared, buried, perhaps, under more recent sedimentary deposits, and/or dispersed and diluted by the prevailing winds, tides and ocean currents. The bivalve and fish data reported here lend support to this conclusion. The residual contamination noted in the grassy swale at the hospital entrance is thought to represent a mercury fraction that is tightly bound to the soil matrix and, therefore, unlikely to be exported to the coast unless the soil is in some way disturbed. The swale's well developed turfed covering greatly reduces the possibility of this happening, even during severe storm conditions. Mercury in this section of the drainage channel may, therefore, be of greater significance to terrestrial food chains with levels accumulating in birds and reptiles that feed on soil arthropods, earthworms and other community representatives in the area.

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