The tropical African mercury anomaly: Lower than expected mercury concentrations in fish and human hair

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Mercury is a neurotoxin and global pollutant, and wetlands and newly flooded areas are known to be sites of enhanced production of monomethylmercury, the form of mercury that is readily biomagnified in aquatic food chains to potentially toxic levels. The Okavango Delta in Botswana, Southern Africa, is the largest inland delta in the world and a wetland ecosystem that experiences dramatic annual flooding of large tracts of seasonal floodplains. The Delta was, therefore, expected to be home to high mercury levels in fish and to be an area where local subsistence fishing communities would be at substantial risk of mercury toxicity from fish consumption. Total mercury concentrations measured in 27 species of fish from the Okavango Delta averaged (mean ± s.d., wet weight) 19 ± 19 ng g^{-1} in non-piscivorous fish, and 59 ± 53 ng g^{-1} in piscivorous fish. These mercury concentrations are similar to those reported for fish from lakes in other areas of tropical Africa, demonstrating that not all wetlands are sites of elevated mercury concentrations in biota. Even more intriguing is that concentrations of mercury in fish from across tropical Africa are systematically and substantially lower than those typically reported for fish from freshwater ecosystems elsewhere globally. The reasons for this apparent "African mercury anomaly" are unclear, but this finding poses a unique opportunity to improve our understanding of mercury's biogeochemical cycling in the environment. Mercury concentrations measured in human hair collected in subsistence fishing communities in the Okavango Delta were similarly low (0.21 ± 0.22 μg g^{-1} dry weight) despite high levels of fish consumption, and reflect the low mercury concentrations in the fish here.

1. Introduction

Mercury is a toxic heavy metal, and its biomagnification in aquatic food webs has become a global problem (Mergler et al., 2007; National Research Council, 2000) as a result of (1) its ability to be transported worldwide via the atmosphere and then be deposited far from its source (Fitzgerald et al., 1998), (2) its microbial methylation to monomethylmercury (MMHg) in aquatic sediment (Benoit et al., 2003), and (3) the subsequent bioaccumulation and biomagnification of that MMHg in aquatic food webs (Watras et al., 1998). This combination has resulted in elevated mercury concentrations in fish even in remote areas far from major sources of the contaminant and in regions with low mercury levels in surface waters and aquatic sediment (Evans et al., 2005; Håkanson et al., 1990). These elevated mercury levels in fish adversely affect the health of both wildlife (Scheuhammer et al., 2007) and humans (Mergler et al., 2007; National Research Council, 2000) that eat them, and have led to consumption advisories being issued for fish in areas of Europe, Canada, and all 50 states in the U.S. (e.g., U.S. EPA, 2007). Fish consumption is responsible for the vast majority of human exposure to mercury (National Research Council, 2000), and although fish consumption advisories are nearly nonexistent in Africa and developing countries elsewhere, this does not necessarily indicate a lack of a mercury problem. In fact, mercury emissions are estimated to be greater in Africa than in North America or Europe (Streets et al., 2009), but data on mercury bioaccumulation in African aquatic systems is limited.

Previous environmental studies of mercury in Africa have focused either on areas contaminated with mercury from mining activities (Donkor et al., 2006; Ikingura et al., 2006; Van Straaten, 2000), or on lakes in eastern Africa (Campbell et al., 2003a, 2003b, 2004, 2006, 2008; Desta et al., 2007; Ikingura and Akagi, 2003; Kidd et al., 2003, 2004; Machiwa, 2005; Ramlal et al., 2003). These reports have largely concluded that mercury levels in fish in the African lakes studied are not exceptionally high, and likely do not pose a substantial threat to local populations that consume fish from their waters. However, it is

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possible that the consistently low mercury concentrations generally reported for fish in Africa are the result of similarities among the lake ecosystems studied, rather than systematically lower fish mercury levels in Africa relative to other regions.

One aquatic ecosystem in Africa which we expected would have fish and other organisms with high mercury concentrations was the Okavango Delta of Botswana, an area also home to a substantial subsistence fishing community (Botswana Ministry of Agriculture, 2003; Mosepele, 2001). Although the Okavango Delta has only minor local point sources of mercury pollution, atmospheric deposition plays an important role in controlling mercury methylation and bioaccumulation in freshwater ecosystems (Hammerschmidt and Fitzgerald, 2006; Orihel et al., 2007; Paterson et al., 2006), and coal combustion and industrial activities in adjacent South Africa and biomass burning in the region are major sources of mercury to the atmosphere (Streets et al., 2009), so this region may experience elevated atmospheric mercury deposition, as suggested by some global models (Selin et al., 2008). In addition, wetlands are hotspots for the microbial production of monomethylmercury (MMHg) (Hurley et al., 1995; St. Louis et al., 1996), and MMHg is the form of mercury that is readily biomagnified up aquatic food chains and typically constitutes >80% of the mercury found in fish (Ikingura and Akagi, 2003; Watras et al., 1998). The Okavango Delta is the world's largest inland delta and is comprised of a variety of wetland types. Finally, newly flooded areas are locations of enhanced MMHg production and bioaccumulation (Belger and Forsberg, 2006; Roulet et al., 2001; St. Loius et al., 2004). The Okavango Delta is subject to a dramatic annual flood, and the system consists of permanent swamps and seasonal floodplains. These floodplains comprise an area of over 8000 km^2, and are utilized extensively by fish when flooded for foraging and reproduction (Høberg et al., 2002).

While high levels of mercury in fish are undesirable wherever they occur, they are of particular concern in locations, such as the Okavango Delta, where both wildlife and humans rely heavily upon fish for food and their livelihood. To determine if mercury in fish is a health concern for local communities or ecosystems in the Okavango Delta we measured total mercury concentrations in fish and surficial sediment, focusing on locations with large fishing communities. We also measured mercury concentrations in scalp hair, a commonly used biomarker of mercury exposure in humans (Björnberg et al., 2003; Díez et al., 2008; Knobeloch et al., 2005; McDowell et al., 2004).

2. Materials and methods

2.1. Study location

The 1500 km long Okavango River flows southeast from its source in the Angolan highlands into the Kalahari Desert in northern Botswana, where it fans out into three primary tributary channel systems, creating the world's largest inland delta (Fig. 1). Due to the lack of large scale agriculture or industry in its catchment, the Okavango River is one of the least polluted or disturbed rivers on the African continent. The Delta's area of inundation and standing wetland coverage exhibits marked seasonal and annual variability, and ranges from roughly 4000 km^2 at base flow to 12,000 km^2 at the height of the annual flood (McCarthy et al., 2000). While rainfall near the headwaters of the Okavango River in central Angola occurs primarily from December to March, the basin's low topographic gradient results in a 5 to 6 month lag before the annual flood reaches its peak at the southern end of the Delta in August (Fig. 2). Rainfall within the lower Delta, on the other hand, occurs primarily between October and May, averages ~500 mm/year, and represents a volume similar to the annual flood inflow (McCarthy et al., 2000). More than 95% of the total water inputs to the system are lost via evapotranspiration.

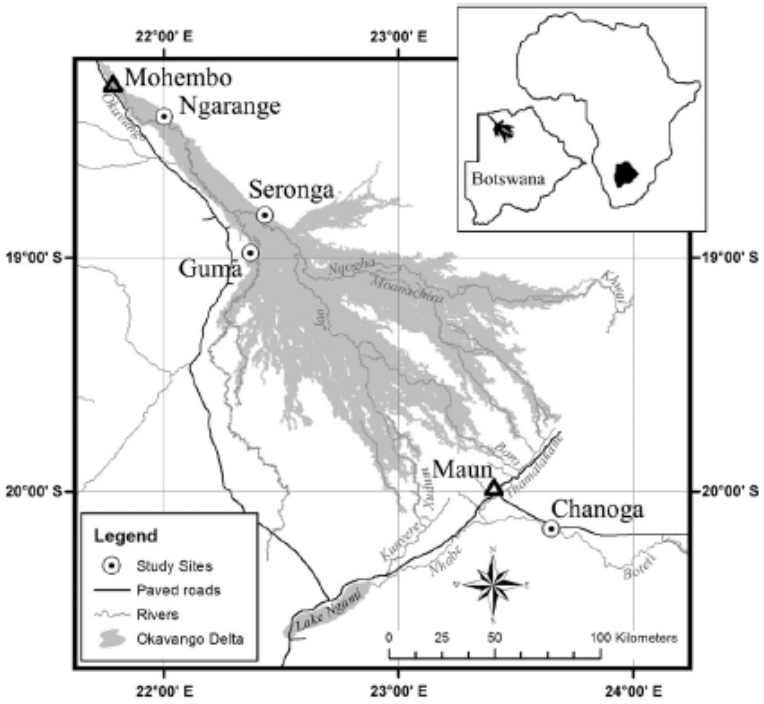

Fig. 1. Location of sample sites (circles) and gauging stations (triangles) in the Okavango Delta, Botswana.

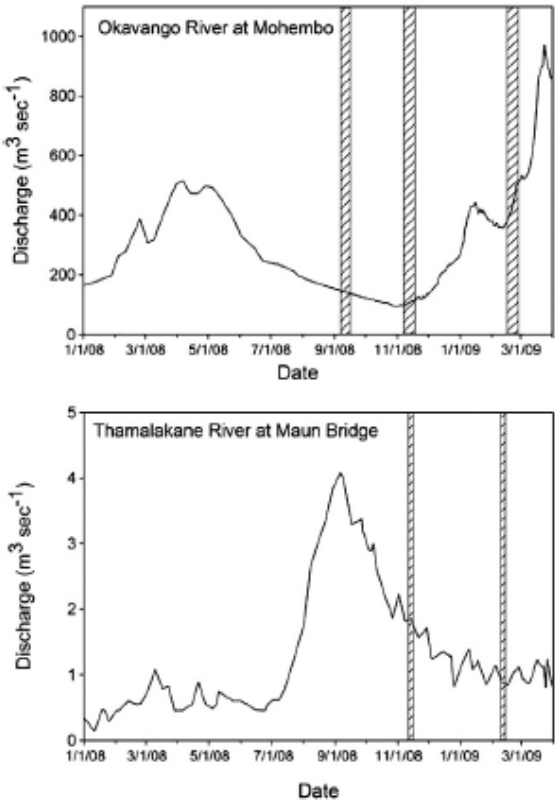

Fig. 2. Hydrographs in the Okavango Delta during the study period. Locations of the Mohembo (upper Delta) and Maun Bridge (lower Delta) gauging stations are shown in Fig. 1. Vertical bars represent periods of fish collection.

As the largest wetland of its type on earth and as one of the world's largest Ramsar sites (wetlands of international importance), the Okavango Delta is a unique natural environment which provides a substantial range of ecological and economic services. The Okavango Delta provides critical habitat for a multitude of plants and animals, and the resulting rich biodiversity and scenic beauty provide the basis for a growing tourism industry. An estimated 120,000 people within the Okavango basin rely economically on the resources of this aquatic system; roughly 3300 of these identify themselves as fishermen, and 40–60% of the population in the panhandle depend on fishing as a source of livelihood (Botswana Ministry of Agriculture, 2003; Mosepele, 2001). Approximately 80% of households in the study region have subsistence diets, with fish comprising half of this diet.

2.2. Sample collection

Samples were collected from four sites (Ngarange, Seronga, Guma Lagoon, and Chanoga) in the Okavango Delta of northern Botswana (Fig. 1). Sample collection was conducted at these areas because of the large subsistence fishing communities here. Fish were collected in September and November, 2008 (during the latter half of the dry season in the panhandle of the upper Delta when subsistence fishing is most intensive), and in February, 2009 (following the unset of the annual flood in the panhandle) (Fig. 2). Fish were caught using gill nets of various mesh sizes to facilitate the collection of fish of a range of sizes and species. When fish diversity or numbers caught were deemed insufficient, additional fresh fish were purchased from fisherman in the same area. A total of 290 fish samples were collected, representing 27 different fish species (Table 1). After species identification and total length measurements, a small sample of boneless, skinless axial muscle tissue was collected from behind the gills using a stainless steel fillet knife that was cleaned and rinsed with DI water between samples. Fish fillets were placed in a zipper top bag and frozen immediately in the field or kept on ice until frozen within 48 h back in the laboratory.

A total of 101 human scalp hair samples were collected in November, 2008 in the fishing villages of Seronga and Chanoga (Fig. 1). Each participant (or participant guardian) completed a survey of age, sex, use of hair products or hair treatment, frequency of fish consumption, and types of fish consumed. Information on hair sample collection, hair cleaning procedures, and participant demographics can be found in the Supplementary Information.

Surficial (upper 4 cm) aquatic sediment and floodplain soil grab samples were collected in duplicate from the three upper delta sites in February, 2009, transferred to acid cleaned borosilicate glass bottles, transported to the laboratory on ice where frozen until digested, with sub-samples dried to measure water content.

2.3. Sample digestion and total mercury analysis

Fish and sediment samples were digested after homogenization by weighing approximately 1.2 g (wet weight) of sample into an acid-cleaned Teflon® digestion bomb and adding 10 mL of a 2:1 mixture of HNO_3 and H_2SO_4 (trace metal grade). Prior to acid digestion, 0.5–0.9 g (dry weight) of hair was weighted and cleaned to remove exogenous mercury (see Supplementary Information). Samples were digested by heating for 4 h at ~90 °C, and then transferred to acid-cleaned borosilicate bottles. Samples were then preserved by amendment to 1% BrCl. The digestion of procedural blanks, analytical triplicates, and the certified reference material DORM-2 (dogfish muscle; National Research Council Canada) accompanied and comprised greater than 10% of all sample digestions.

Total mercury concentrations were determined by oxidation with BrCl, reduction with $SnCl_2$, gold trap amalgamation, and quantification by cold vapor atomic fluorescence spectrometry (CVAFS) using established methods (U.S. EPA, 2002). The average Hg_T detection limit, based on three times the standard deviation of digestion blanks ($n = 18$) was 0.23 ng g^{-1} for fish and sediment samples. The average relative standard deviation of fish samples ($n = 6$) digested and analyzed for Hg_T in triplicate was 12%. The concentration (mean ± standard deviation) of Hg_T measured for the certified reference material DORM-2 was 4.27 ± 0.52 μg g^{-1} ($n = 14$), which represents an average recovery of 92% given its certified value of 4.64 ± 0.26 μg g^{-1} (National Research Council Canada).

3. Results and discussion

3.1. Mercury concentrations in fish in the Okavango Delta

Total mercury concentrations measured in muscle tissue from fish collected in the Okavango Delta, grouped by fish species, are presented in Table 1 and Fig. 3. Mercury concentrations (mean ± s.d., wet weight) in non-piscivorous fish (19 ± 19 ng g^{-1}) were both lower ($p < 0.001$, t-test) and less variable than those in piscivorous fish (59 ± 53 ng g^{-1}). Only 1% of all fish had mercury concentrations exceeding 0.2 μg g^{-1}, the recommended lower guideline set by the World Health Organization to protect groups vulnerable to mercury toxicity (FAO/WHO, 2004).

The greater mercury concentrations measured in fish occupying higher trophic levels is consistent with mercury's biomagnification in aquatic feed webs. The positive relationship between mercury concentration and total fish length (Supplementary Information Fig. 1) observed for the majority of fish species in this study is consistent with previous studies reporting increasing mercury concentrations with age, and is due to rates of MMHg depuration

Table 1
Compiled measurements of fish length and total mercury concentration in muscle tissue of fish collected from the Okavango Delta.

Fish species		n	Fish length (cm)					[HgT] (μg g^{-1} wet weight)				
Scientific name	Common name		Median	Mean	Stdev.	Min.	Max.	Median	Mean	Stdev.	Min.	Max.
Barbus poechii	Dashtail barb	6	11	11.5	1.0	11	13	14.7	28.9	32.5	8.8	77.3
Brycinus lateralis	Astriped robber	8	13	12.3	1.2	11	14	24.3	27.4	11.5	16.2	50.3
Clarias gariepinus	Sharp tooth catfish	18	53	52.4	12.5	16	74	73.9	102.9	96.0	5.7	216.6
Clarias ngamensis	Blunt tooth catfish	14	39	40.5	5.1	34	52	38.5	36.3	27.4	2.7	91.9
Hepsetus odoe	African pike	18	31	30.6	4.3	20	38	45.1	45.7	18.2	5.6	73.3
Hydrocynus vittatus	Tigerfish	21	32	33.2	11.7	18	57	62.4	76.9	46.5	25.1	168.8
Marcusenius macrolepidotus	Bulldog	24	23	22.1	2.3	18	26	20.1	25.5	22.6	3.7	109.2
Mormyrus lacerda	Western bottlenose	8	40	38.5	3.9	32	43	24.8	25.2	8.2	14.4	40.5
Oreochromis andersonii	Three spot tilapia	10	28	28.1	3.3	23	34	2.2	5.1	4.3	1.3	11.1
Oreochromis macrochir	Green head tilapia	8	21	21.6	4.9	16	28	7.6	9.1	7.2	2.7	23.2
Pharyngochromis acuticeps	Zambezi River bream	1	15	15.0	–	–	–	12.8	12.8	–	–	–
Sargochromis carlottae	Rainbow bream	4	11	11.5	1.0	11	13	9.0	9.7	3.0	6.9	14.0
Sargochromis condringtonni	Green bream	8	24	21.5	4.4	15	26	10.8	12.6	5.8	7.6	25.9
Sargochromis giardi	Pink bream	2	23	23.0	14.1	13	33	89.6	89.6	56.9	49.4	129.8
Schilbe intermedius	Silver catfish	31	26	26.4	2.6	22	31	52.8	59.7	46.5	8.2	206.2
Serranochromis altus	Humpback largemouth	2	37	36.5	3.5	34	39	83.0	83.0	54.8	44.2	121.7
Serranochromis angusticeps	Thin face largemouth	8	30	29.4	2.9	23	32	35.4	46.8	39.5	10.8	123.5
Serranochromis macrocephalus	Purple face largemouth	19	27	26.1	4.9	16	33	33.2	29.4	18.0	1.8	70.7
Serranochromis thumbergi	Brown spot largemouth	4	18	17.8	3.3	14	21	15.4	14.3	6.1	5.8	20.4
Synodontis nigromaculatus	Spotted squeaker	10	21	20.4	2.3	17	25	22.8	37.0	31.8	10.3	114.1
Synodontis spp.*	Squeaker	33	19	18.3	2.5	14	23	19.8	24.6	19.3	0.2	98.5
Tilapia rendalli	Red breast tilapia	11	25	23.9	6.5	14	33	3.5	4.8	3.9	1.4	15.4
Tilapia sparrmanii	Banded tilapia	22	16	17.0	6.4	12	44	5.9	7.3	4.7	1.2	18.8
Non piscivores		176	20	21.4	6.8	11	44	15.4	19.4	19.0	0.2	114.1
Piscivores		114	31	34.7	11.7	16	74	49.8	59.4	53.3	2.7	216.6

**leopardinus, woosnami, vanderwaali, thamalakanensis,* and *macrostigma.*

being much slower than rates of accumulation, and because of shifts to higher trophic levels during development for some fish (Driscoll et al., 1995; Wang and Wong, 2003; Watras et al., 1998).

Field sampling was designed to allow us to test for both spatial and temporal (hydrologic regime) differences in fish mercury concentrations (Figs. 1 and 2). However, the effect of the annual flood on mercury concentrations could not be fully evaluated because we were unable to collect sufficient fish numbers in the panhandle of the upper Delta after the onset of the annual flood. These low catches reflect a "dilution effect" typical of floodplain fisheries as fish migrate to newly

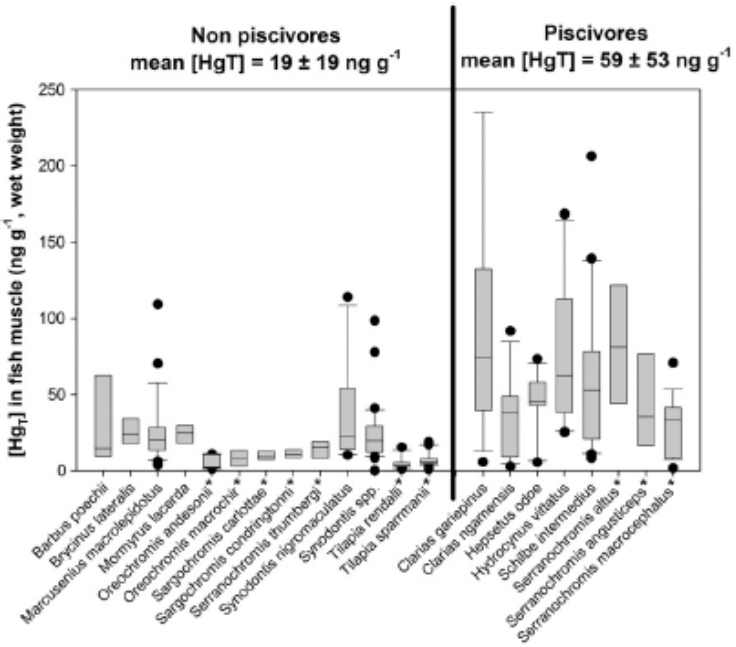

Fig. 3. Box plots of total mercury concentrations in fish in the Okavango Delta of Botswana. The median is represented by the middle line of each box, hinges represent the 25% and 75% quartiles, whiskers represent the 10 and 90 percentiles, and points are outliers. Asterisks denote species referred to locally as "bream", which was reported as the fish consumed most frequently.

inundated floodplains. The much lower catch per unit effort during the flood season results in a substantial reduction in fishing activities by the local community during this time of year, so our low catches could not be supplemented by purchasing fresh fish locally, an alternative which was further hampered by a seasonal moratorium on commercial fishing.

An ANCOVA statistical approach was used to test for spatial differences in fish mercury concentrations while accounting for differences between fish species and variations in fish size. To satisfy the requirements of ANCOVA, this analysis used only those 6 species of fish that were caught at all 4 sites, and for which the positive relationship between fish length and mercury concentration was evaluated and found to be similar (Supplementary Information Fig. 1). There was no measurable difference in mercury concentrations in fish between the three panhandle sites, but mercury concentrations were significantly higher ($p \ll 0.001$; ANCOVA) in fish from the upper Delta (3 panhandle sites combined) compared to those just outside the lower Delta on the Boteti River at Chanoga (Supplementary Information Fig. 2).

3.2. Estimated mercury exposure to local subsistence fishing communities

Surveys of human participants during hair collection indicated that fish consumption is particularly high in local communities during the low water/intensive fishing season (roughly April–December in the panhandle). When surveyed in November, participants ($n = 101$) reported consuming fish 3.4 ± 2.2 meals per week, with 22% reporting consuming fish ≥ 7 meals per week.

Estimating mercury exposure to local subsistence communities in the Okavango Delta is somewhat problematic because most participants reported "bream" as the fish they consume most often. Bream is a common term which can refer to as many as 10 of the fish species sampled in this study (Fig. 3), including both piscivores and non-piscivores, and thus having a range of mercury concentrations. A more detailed, but limited, survey of the local community was used to estimate a mean total mercury concentration in fish consumed of 0.033 μg g^{-1}, when weighted according to reported consumption. Using the average reported diet of 3.4 ± 2.2 meals of fish per week and assuming a fish portion of 200 g, the average mercury exposure in this community would be 22 ± 15 μg per week. For a 70 kg individual this would represent a mercury dose of 0.046 ± 0.030 μg kg^{-1} body weight d^{-1}, which is well below both the US EPA reference dose for methylmercury of 0.1 μg kg^{-1} d^{-1} (U.S. EPA, 2001), and the World Health Organization's tolerable intake for methylmercury of 0.2 μg kg^{-1} d^{-1} (FAO/WHO, 2004). We estimate that a 70 kg individual could consume up to 7 meals of fish from this area per week and still not exceed that US EPA reference dose. Thus, mercury toxicity would not appear to be a major concern for most members of this community. However, considering the very high fish consumption reported by some individuals during the fishing intensive low water season and differences in the average food intake and masses of males (70 kg), females (60 kg), and children (50 kg for ages 11–15, 35 kg for ages <11), we estimate that 13% of the females and 2% of males (but no children) in this community exceed the mercury reference dose of 0.1 μg kg^{-1} d^{-1} due to fish consumption during part of the year.

3.3. Mercury concentrations in hair from local subsistence fishing communities

Hair is a convenient, non-invasive, and commonly used biomarker of human exposure to mercury, and mercury levels in hair generally reflect levels of MMHg in the blood (National Research Council, 2000). Concentrations of total mercury in scalp hair from subsistence fishing communities in the Okavango Delta ranged from 0.003 to 0.97 μg g^{-1}, with a median of 0.10 μg g^{-1} and a mean of 0.21 ± 0.22 μg g^{-1}. There were no hair samples in this study which exceeded the US National Research Council recommended level for mercury in hair of 1 μg g^{-1} (National Research Council, 2000). This observation supports the above conclusion, based on estimates of mercury exposure from fish rich diets, that mercury toxicity does not represent a significant risk for most of this community. Mercury hair concentrations and fish consumption data stratified by sex and age are presented in Table 2, respectively, while additional plots of mercury hair concentrations and demographic information can be found in the Supplementary Information Figs. 3–6.

Higher levels of fish consumption were associated with higher mercury concentrations in hair (Fig. 4). This significant ($p < 0.001$, ANOVA) effect of diet on mercury levels in hair is consistent with the majority of human exposure to mercury occurring via fish consumption (National Research Council, 2000). While the positive relationship between fish consumption and mercury levels in hair was not a simple linear one across all groups, the general trend is consistent with previous studies (Björnberg et al., 2003; Díez et al., 2008; Knobeloch et al., 2005; McDowell et al., 2004). When normalized to fish consumption, the mercury concentrations we measured in hair in the Okavango Delta are somewhat lower than those measured elsewhere (Fig. 4). This difference reflects the lower mercury levels in the fish consumed by subjects in our study compared to fish elsewhere, although other factors (hair growth rate, nutritional status, toxicokinetics, exogenous contamination, etc.) can also be responsible for inter-individual and between group differences in hair mercury levels (Canuel et al., 2006; Knobeloch et al., 2005; McDowell et al., 2004).

3.4. Fish mercury concentrations in the Okavango Delta compared to other regions globally

Concentrations of mercury in freshwater fish in the Okavango Delta are similar to those reported for aquatic ecosystems in other areas of tropical Africa (Fig. 5). This similarity is rather surprising given that most of those data are from lakes, whereas the fish from this study were from a large wetland complex which also experiences annual flooding, both of which favor the production of MMHg, the form of mercury that is biomagnified in aquatic food webs. Even more surprising is that levels of mercury in fish from the Okavango Delta and other areas in tropical Africa are substantially lower than those in freshwater fish from comparable regions globally.

Fig. 6 presents a comparison of mercury concentrations in fish from this study to those from large ($n > 250$), robust data sets from other regions where, like the Okavango Delta, mercury inputs are dominated by atmospheric deposition rather than local point sources. While the necessary data are not reported in many of these studies to normalize fish mercury concentrations or use an ANCOVA approach to fully account for the effects of fish species, size, diet, or other factors, the large number of fish ($n > 250$) of different species, ages, and trophic levels analyzed from each area limits possible sampling bias and allows for a meaningful comparison. Fig. 6 shows that despite the many factors contributing to the spatial, temporal, and interspecies variability in mercury concentrations in fish within a given geographic region, the clear difference that remains indicates that mercury concentrations in fish from the Okavango Delta and the rest of tropical Africa (Fig. 5) are systematically and substantially lower than those from freshwater systems in temperate North America, temperate Europe, or tropical South America.

This difference does not appear to be due to mercury abundance or inputs. The concentrations of total mercury we measured in surficial aquatic sediment and floodplain soils in the Okavango panhandle were relatively low (0.035 ± 0.010 μg g^{-1} dry weight, $n = 6$), but much higher concentrations are found in other areas of tropical Africa (Campbell et al., 2003a; Ramlal et al., 2003). The relatively low sediment mercury concentrations in the Okavango are typical of unpolluted areas elsewhere and similar to other regions that still have fish with much

Table 2
Concentrations of mercury in human hair and fish consumption data stratified by sex and age for individuals participating in the study of hair mercury levels in the upper Okavango Delta.

Meals fish reported eaten per month	n	Sex		Age					Hair [HgT] (μg g^{-1} dry weight)				
		Males	Females	Median	Mean	Stdev.	Min.	Max.	Median	Mean	Stdev.	Min.	Max.
0–4	8	1	7	22	33	25	8	64	0.08	0.08	0.04	0.030	0.14
5–12	60	22	38	23	26	14	4	70	0.09	0.16	0.19	0.003	0.82
13–20	11	7	4	35	35	18	10	68	0.38	0.41	0.32	0.021	0.92
>20	22	11	11	31	34	10	21	55	0.24	0.29	0.23	0.040	0.97
All	101	41	60	26	29	15	4	70	0.10	0.21	0.23	0.003	0.97

Sex	n	Age					Meals of fish reported eaten per week					Hair [HgT] (μg g^{-1} dry weight)				
		Median	Mean	Stdev.	Min.	Max.	Median	Mean	Stdev.	Min.	Max.	Median	Mean	Stdev.	Min.	Max.
Female	60	26	29	16	5	70	2	3.1	2.1	0	7	0.08	0.17	0.20	0.003	0.87
Male	41	26	28	13	4	60	3	4.0	2.3	1	10	0.19	0.27	0.25	0.019	0.97
All	101	26	29	15	4	70	2	3.4	2.2	0	10	0.10	0.21	0.23	0.003	0.97

Age (years)	n	Meals of fish reported eaten per week					Hair [HgT] (μg g^{-1} dry weight)				
		Median	Mean	Stdev.	Min.	Max.	Median	Mean	Stdev.	Min.	Max.
0–9	6	2	1.7	0.5	1	2	0.08	0.08	0.03	0.037	0.12
10–19	20	2	2.3	1.0	0	5	0.09	0.13	0.12	0.003	0.48
20–29	37	3	3.6	2.1	0	7	0.08	0.16	0.18	0.012	0.73
30–39	13	3	4.2	2.4	2	7	0.25	0.29	0.29	0.022	0.97
40–49	13	5	5.3	2.5	2	10	0.36	0.36	0.26	0.019	0.92
50–59	6	4	4.3	2.3	2	7	0.27	0.27	0.20	0.032	0.51
≥60	6	1.5	1.8	1.2	1	4	0.13	0.34	0.40	0.030	0.87
All	101	2	3.4	2.2	0	10	0.10	0.21	0.23	0.003	0.97

higher mercury levels (Evans et al., 2005; Liu et al., 2008; Peterson et al., 2007; South Florida Water Management District, 2010). There is also no evidence that atmospheric mercury deposition in tropical Africa is significantly lower than most regions globally. Instead, global models estimate that atmospheric mercury deposition to most of sub-Saharan Africa is greater than the global mean for terrestrial systems (Selin et al., 2008), although regional emissions from South Africa were likely overestimated in that model (Dabrowski et al., 2008). These elevated atmospheric inputs of mercury to tropical Africa would be expected to result in higher mercury levels in fish because newly deposited atmospheric mercury is more labile and bioavailable to sediment microbes, and is thus more easily methylated and bioaccumulated, than mercury from other sources (Hammerschmidt and Fitzgerald, 2006; Orihel et al., 2007; Paterson et al., 2006). This makes the low mercury levels in fish in the Okavango Delta and the rest of tropical Africa even more surprising, and further indicates that differences in total mercury inputs or abundance are unable to explain these results.

Although differences in a number of other physical, chemical, or biological parameters affecting mercury methylation, demethylation, and bioaccumulation could be responsible for the low mercury concentrations in fish in tropical Africa, these parameters are highly variable across aquatic ecosystems in tropical Africa, and even if they were not, the vast majority would appear to be unable to account for the low mercury levels in fish here. For example, the Okavango Delta is characterized by long food chains, high mean temperatures, and substantial periodic remobilization of nutrients and labile organic

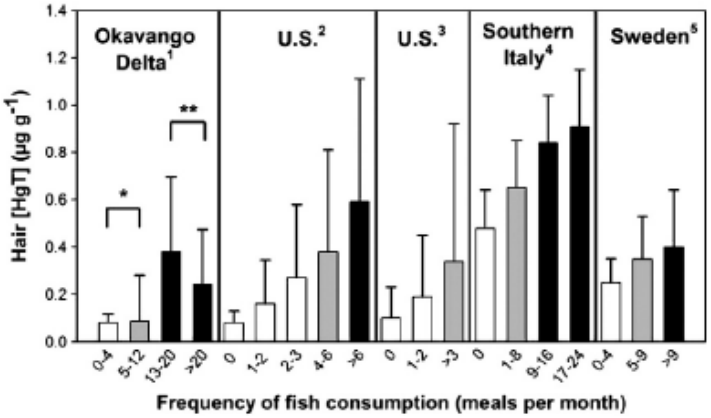

Fig. 4. Total mercury concentration in hair related to reported frequency of fish consumption. White bars represent low fish consumption, gray bars medium fish consumption, and black bars high fish consumption. Error bars represent the standard deviation about the median. Asterisks denote statistically significant differences at the $p<0.05$ level (Tukey post-hoc test). References are [1]this study, [2]Knobeloch et al. (2005), [3]McDowell et al. (2004), [4]Díez et al. (2008), and [5]Björnberg et al. (2003).

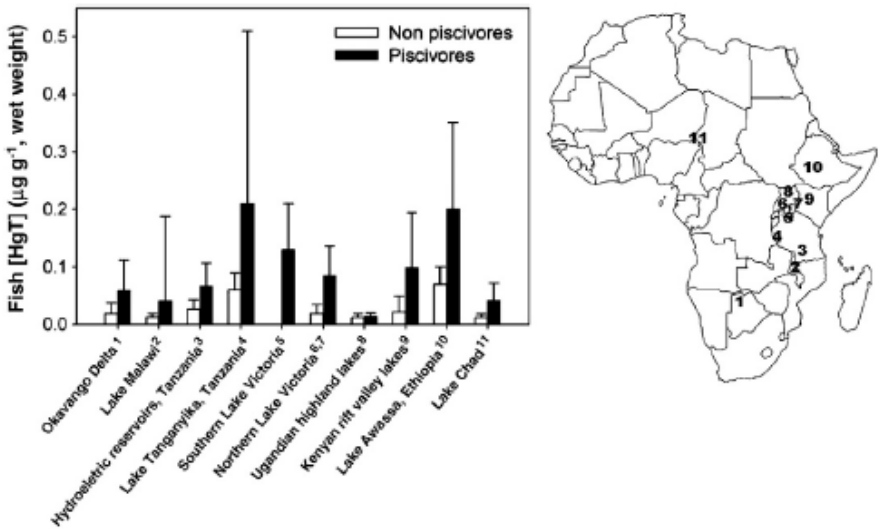

Fig. 5. Mercury concentrations measured in freshwater fish in the Okavango Delta and in other areas of Africa in previous studies. Error bars represent the standard deviation about the mean. Study locations are shown on map at right. References are [1]this study, [2]Kidd et al. (2003), [3]Ikingura and Akagi (2003), [4]Campbell et al. (2008), [5]Machiwa (2005), [6]Campbell et al. (2003a),[7]Ramlal et al. (2003), [8]Campbell et al. (2006), [9]Campbell et al. (2003b), [10]Desta et al. (2007), and [11]Kidd et al. (2004).

matter (Høberg et al., 2002), all of which favor MMHg production or bioaccumulation (Benoit et al., 2003; Bodaly et al., 1993; Cabana et al., 1994), so would be expected to result in high mercury levels in fish, which is the opposite of our findings. Microbial mercury methylation may be sulfate limited in systems like the Okavango Delta and Lake Victoria where surface water sulfate concentrations are low (generally <1 mg L^{-1}; Cronberg et al., 1996; Lehman and Branstrator, 1994), but much higher sulfate concentrations characterize other tropical African freshwaters. The higher growth rate (growth dilution) and shorter life span of fish in the tropics or differences in food web structure might result in lower mercury levels in fish here (Harris and Bodaly, 1998), while differences in phytoplankton and zooplankton communities and their bioaccumulation of mercury at the base of the food web can result in substantial differences in mercury biomagnification and concentrations in fish (Pickhardt and Fisher, 2007; Stewart et al., 2008). However, the nature of mercury biomagnification does not appear to differ substantially due to climate or other factors in tropical Africa compared to other regions, as suggested by previous studies that have documented similar relationships between stable nitrogen isotopic composition and mercury concentrations in fish at various trophic levels from lakes in temperate North American and tropical Africa (Campbell et al., 2004, 2003a; Kidd et al., 2003). And, although higher rates of MMHg photo-decomposition would be expected due to the higher annual photon flux at these low latitudes, other factors that influence this process, such as light attenuation and water column depth (Lehnherr and St. Louis, 2009), differ substantially among tropical African aquatic environments. Moreover, higher rates of MMHg photo-demethylation would also be expected at similar latitudes elsewhere, but mercury levels in fish from Lake Murray in Papua New Guinea (Bowles et al., 2001) and the Rio Negro in equatorial Brazil (Barbosa et al., 2003; Belger and Forsberg, 2006; Dórea and Barbosa, 2007) are much higher than those in tropical Africa (Fig. 6).

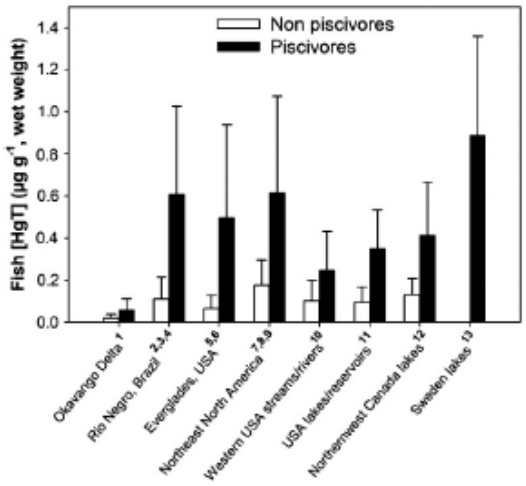

Fig. 6. Mercury concentrations measured in fish in the Okavango Delta compared to those in freshwater fish in areas of North America, South America, and Europe. Atmospheric deposition is believed to be the dominant external source of mercury in all sample areas, and the sample size was > 250 in all study regions. Error bars represent the standard deviation about the mean. References are [1]this study, [2]Barbosa et al. (2003), [3]Belger and Forsberg (2006), [4]Dórea and Barbosa (2007), [5]Liu et al. (2008), [6]South Florida Water Management District (2010) (2005–2009 data), [7]Rose et al. (1999), [8]Kamman et al. (2005), [9]Sveinsdottir and Mason (2005), [10]Peterson et al. (2007), [11]Stahl et al. (2009), [12]Evans et al. (2005), and [13]Håkanson et al. (1990).

4. Conclusions

Although there are a number of potential reasons for the lower than expected concentrations of mercury in fish in the wetlands comprising the Okavango Delta, and for the mercury levels in fish in tropical Africa being systemically lower than those elsewhere globally, the most important of these factors remains unclear. So while previous research on mercury in freshwater aquatic systems has been limited predominantly to (1) temperature regions in the northern hemisphere or (2) regions contaminated with local mercury pollution, the results from this study highlight the need for further research in tropical Africa and similar regions in order to improve our understanding of mercury's biogeochemical cycling in these environments.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.scitotenv.2010.11.027.

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