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Full Length Research Paper

Comparative study of mercury accumulation in some brackish water fishes in a tropical lagoon and its adjacent creek in south western Nigeria

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The Hg content of some brackish water fishes, water and sediment in Lagos lagoon and Abule - Agege creek were investigated from February to September, 2004. The pH of the lagoon was neutral however the creek was mildly acidic (6.75) in the dry season. The Hg content of the lagoon ranged from 0.27 - 0.40 mg/l for water and 0.46 - 0.56 µg/g dry wt for sediment lower than the creek range (0.41 - 0.42 mg/l and 0.71 - 0.72 µg/g) for the dry season. For the rainy season high Hg levels were recorded in lagoon water ranged from 0.41 - 0.48 mg/l more than the creek ranged from 0.42 - 0.44 mg/l. However, more mercury was recorded in the creek sediment 0.95 µg/g than that in lagoon sediment 0.71 - 0.81 µg/g. Surface water temperature for the creek was lower than the lagoon. Generally, higher mercury values were recorded in sediment than in the water in both the lagoon and the creek. For the fish species 0.72 ± 0.05 µg/g of Hg was detected in *Sarotherodon melanotheron,* more than in both *Tilapia guineensis* and *Hemichromis fasciatus,* this is related mainly to the mode of feeding. Hg content of the examined fish was below maximum permissible levels (0.5 - 1.5 µg/g) in the food chemical codex.

Key words: Biomagnifications, Hg, brackish water, fish, chemical codex.

INTRODUCTION

As a result of high level of urbanization and industrialization of Lagos and its surroundings, there is an inevitable generation of domestic wastes and industrial effluents (Ajao and Fagade 1990; Nwankwo 2004) which find their way into coastal ecosystems. These inputs have ecological consequences on the array of coastal aquatic ecosystem (Chukwu, 2002).

The literature on Hg contamination of fish is extensive. A bibliography by Taylor (1973) contains over a hundred references of Hg in fish. For instance Hg is a notable pollutant in Sweden (Johnels et al., 1967; Westoo, 1967; Westoo and Rydalv, 1969; Ackefors et al., 1970; Jernelov and Lann, 1971). Some alarm has also been raised concerning possible health implications of some heavy metals in seafood example, shellfish (Han et al., 1994; Clark et al., 1997). In Nigeria, recent studies by Asuquo et al. (1997) and Asuquo et al. (1999) revealed that the concentrations of heavy metals have increased in the activities. Ogri (2002) reported some heavy metal Cross River system and its environs as a result of man's concentration in fish and shellfish that may be harmful to man. The damages caused by Hg contaminants has been reported by Wood (1983) on Minamata Bay, Japan where local inhabitants had neurological ailments after consuming sea fish and shellfish contaminated with methyl mercury.

Unlike gold and silver, Hg is not considered rare because it is found in highly concentrated deposits and most of it occurs as cinnabar a mineral composed of mercury sulfide (Environment Canada, 2004). Anthropo-genic activities such as mining are known to release more Hg from rocks and minerals than natural weathering processes. Hg is of special concern because during pregnancy it can be passed from a pregnant mother to her unborn child through placenta (Blagojevich and Whitaker, 2006). The USEPA (2001) warned specifically pregnant women, women of child bearing age, nursing mothers and young children not to eat any fish known to carry high levels of Hg. USEPA (2001) reported further that the sources of mercury in soil include direct application of fertilizers, fungicides and disposal of waste, including

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batteries and thermometers to landfills.

When mercury enters the body as an organic form called methyl Hg, it moves into the blood stream and is carried to the liver, kidneys and brain (Blagojevich and Whitaker 2006). Hg is a neuro - toxicant, which means that it impairs the brain's normal function causing a variety of neurological symptoms (Environment Canada, 2004).

According to USEPA (2001) in most adult fish, 90 - 100% of the mercury present is in form of methyl-mercury. Methyl mercury is found primarily in the fish muscles (fillets) bound to proteins (Blagojevich and Whitaker, 2006). Skinning and trimming the fish has not been reported to significantly reduce the mercury concentration in the fillet, nor is it removed by cooking processes (USEPA, 2001).

Consequently, it is known that fish and other seafood products are important sources of methyl mercury in man's diet.

This study was aimed to assess Hg levels in selected brackish water fish species (*Sarotherodon melanotheron, Hemichromis fasciatus, Caranx hippos, Bathygobius soporator and Tilapia guineensis*) in relation to their environment. In addition the bio–concentration factor was studied to investigate the effects of seasonal changes in the mercury levels in the species and the sediment.

MATERIALS AND METHODS

Description of the area of investigation

The Lagos lagoon is a large expanse of shallow water covering an area of about 208 km^2 (Emmanuel and Kusemiju, 2005). It serves as an important seaport, nursery ground for fisheries, and a source of food supply and for recreational purposes. The adjacent creek (Abule Agege) is situated near the faculty of science and directly behind the computer science department building where it extends to the Lagos lagoon. The vegetation around the creek was characterized by several types of plants, including *Paspalum vaginatum, Acostichum aureum, Phoenix reclineta, Rhizophora racemosa, Avicenia nitida, Drepanocarpus lunatus* and *Cyperus articulatus*.

Water Sampling

Surface water samples were collected about 10cm below the water surface into clean bottles (2 L plastic) and were labeled to indicate the sample location, date and time. These were preserved in a refrigerator prior the analysis. pH was measured in the field using a Lovibond comparator and cross-checked in the laboratory with a Griffin pH meter model 80, the dissolved oxygen (mg/l) was measured by the Winkler method (Barnes, 1980) and water temperature (°C) was estimated with an ordinary Hg thermometer.

Fish sampling

Fish samples were collected from February - May, 2004 for the dry season and June - September, 2004 for the rainy season with a cast net designed and constructed with polyamide material (0.24 mm diameter) with 31 mm stretched mesh size. The sampling was carried out at the early hours of each day, thrice a week for the period of sampling. Twelve replicates of samples for both wet and dry season were used for analysis in this study.

The fish species sampled were *Sarotherodon melanotheron* (Dumeril) (Cichlidae), *Hemichromis fasciatus* (Peters) (Cichlidae), *Bathygobius soporator* (Valenciennes) (Gobiidae), *Caranx hippos* (Linnaeus) (Carangidae) and *Tilapia guineensis* (Bleeker) (Cichlidae).

Sediment sampling

2 - 3 cm of surface sediment was collected with a grab at the 4 sampling stations. The sediment was sieved and then the fraction which passed through a 180 µm nylon sieve was retained so as to include all fume particles containing heavy metals of toxicological interest. The sediment slurry was kept in an acid glass stopper jar and placed in ice for transportation to the laboratory. Samples were refrigerated prior to analysis.

Procedures for mercury analysis

The method employed is the same as that used by the British Ministry of Agriculture, Fisheries and Food (MAFF) for routine monitoring up to the year 1977 (ICESS, 1974). 2.5 ml nitric acid (HNO_3) and 0.4 g Potassium persulphate ($K_2S_2O_8$) were added to 100ml water sample in a 250 ml conical flask and the mixture stirred to allow complete dissolution. 1 g of potassium permanganate was added to the sample and continuously stirred until it dissolved. The conical flask was covered with a watch glass. The mixture was then heated to 90°C for 2 h and then cooled to room temperature. The purple colour was discharged with addition of diluted solution of hydroxylamine hydrochloride.

The digested sample solution was transferred into cold vapour washing bottle. The Hg absorber containing 8 ml of Hg (tetraoxosulphate VI acid solution) was connected to 100 ml Erlenmeyer flask and this was in turn connected to the vacuum pump for 5 min. The Erlenmeyer flask was replaced with 10 ml distilling receiver while 2 ml of sodium hypochlorite was pipette into the Hg absorber column which was connected to the gas washing bottle that contain the digested sample.

The absorbed Hg in the column was eluted with 8ml tetraoxosulphate VI acid solution, alkaline powder pillows and indication powder pillows were added to 10 ml eluate and mixed between additions. After that, 8 drops of sodium hydroxide (NaoH solution) were added, mixed and allowed to stand for 2 min. The complexion solution was added to the mixture. Hg level in the mixture was determined using Atomic absorption spectrometer (AAS).

The sediment slurry was dried out at 40°C in an oven for 24 h and then powdered using mortar and pestle. Wet ash and the resulting solution were pre-concentrated before they were analyzed for mercury using the Atom Absorption Spectrometer (AAS HACH 2010).

The fish were completely filleted; the gills and the skin were carefully removed from the fish. A blender was used to homogenize the fish fillets to a paste which was transferred into a tarred labeled bottle. 4g of homogenized sample were treated as mentioned for the sediment.

The result was analyzed using Microsoft Excel and STAT 7.0 statistical package (copyright 1984 – 2002.

RESULTS

The total Hg analysis in fish samples, water and sediments are shown in Tables 1, 2, 3 and 4. For the dry season, pH of 7.05 ± 0.11 and 7.05 ± 0.08 were observed in stations A and C while stations B and D recorded 7.00

Table 1. The mean (± SD) physical parameter and mercury analysis in water and sediment samples collected from the area of investigation during the dry season (Feb - May, 2004).

Sampling station	pH	D.O. (mg/l)	Temperature (°C)	Water (mg/l)	Sediment g/g Dry wt
A	7.05 ± 0.11	6.53 ± 0.02	28.30 ± 0.45	0.27 ± 0.02	0.46 ± 0.04
B	7.00 ± 0.16	6.55 ± 0.07	28.10 ± 0.29	0.33 ± 0.04	0.53 ± 0.04
C	7.05± 0.08	6.43 ± 0.02	28.50 ± 0.65	0.40 ± 0.04	0.56 ± 0.02
D	6.75 ± 0.04	7.33 ± 0.02	27.30 ± 0.62	0.41 ± 0.02	0.71 ± 0.03

A: Lagos lagoon, B: Lagos lagoon, C: Lagos lagoon, D: Abule – Agege, DO = dissolved oxygen, SD = standard deviation.

Table 2. The mean (±SD) Hg content (µg/g; wet wt) in tissues and gills of fish caught from Lagos lagoon and Abule Agege Creek (Feb. - May, 2004).

Species	Lagos Lagoon		Abule Agege Creek	
	Gills	Tissues	Gills	Tissues
Sarotherodon melanotheron	0.04 ± 0.01	0.34 ± 0.09	0.02 ± 0.01	0.43 ± 0.01
Hemichromis fasciatus	0.02 ± 0.01	0.39 ± 0.12	0.02 ± 0.01	0.39 ± 0.02
Bathygobius soporator	0.03 ± 0.01	0.34 ± 0.19	0.02 ± 0.01	0.23 ± 0.01
Caranx hippos	0.03 ± 0.02	0.24 ± 0.12	0.02 ± 0.01	0.35 ± 0.02
Tilapia guineensis	0.03 ± 0.01	0.44 ± 0.07	0.02 ± 0.01	0.41 ± 0.01

SD = standard deviation.

Table 3. The mean (± SD) physical parameter and Hg analysis in Water and Sediment samples collected from the area of investigation during rainy season (June – September, 2004).

Stations	pH	DO (mg/l)	Temp. (°C)	Hg concentration	
				Water (mg/l)	Sediment (µg/g)
A	7.65 ± 0.06	7.25 ± 0.04	23.5 ± 0.79	0.41 ± 0.04	0.71 ± 0.02
B	7.55 ± 0.07	7.50 ± 0.54	23.5 ± 0.36	0.48 ± 0.04	0.81 ± 0.06
C	7.50 ± 0.04	7.30 ± 0.41	24.5 ± 0.54	0.44 ± 0.05	0.76 ± 0.04
D	7.10 ± 0.02	7.75 ± 0.45	22.3 ± 0.22	0.42 ± 0.04	0.95 ± 0.11

A: Lagos lagoon, B: Lagos lagoon, C: Lagos lagoon, D: Abule – Agege, DO = dissolved oxygen, SD = standard deviation.

± 0.16 and 6.75 ± 0.04, respectively. The maximum dissolved oxygen was observed in station D with 7.33 ± 0.02 mgO_2 /l and the lowest was from station C with 6.43 ± 0.02 mgO_2/l. The Hg content of water at station D with 0.41 ± 0.02 mg/l was the maximum while that of the station A had the lowest Hg with 0.27 ± 0.02 mg/l. Station D recorded the maximum levels Hg with 0.71 ± 0.03 µg/g and the lowest value recorded was at station A with 0.46 ± 0.04 µg/g for sediment. The examination of the gill and the tissue for Hg showed that there were higher Hg levels in the tissues than in the gills for both the lagoon and the creek. More Hg was recorded for fish species in the creek than in the lagoon for the same species (Table 2). Likewise the maximum Hg was observed in the Cichlids; 0.43 ± 0.01, 0.41 ± 0.01 and 0.39 ± 0.02 µg/g wet wt for *S. melatheron, T. guineensis* and *H. fasciatus* respectively in the creek. While in the lagoon the highest levels were recorded in *T. guineensis* (0.44 ± 0.07 µg/g wet wt) and the lowest in *C. hippos* (0.24 ± 0.12 µg/g wet wt).

The total Hg analysis in fish samples, water and sediments are shown in Tables 1, 2, 3 and 4. For the dry season, pH of 7.05 ± 0.11 and 7.05 ± 0.08 were observed in stations A and C while stations B and D recorded 7.00 ± 0.16 and 6.75 ± 0.04, respectively. The maximum dissolved oxygen was observed in station D with 7.33 ± 0.02 mgO_2 /l and the lowest was from station C with 6.43 ± 0.02 mgO_2/l. The Hg content of water at station D with 0.41 ± 0.02 mg/l was the maximum while that of the station A had the lowest Hg with 0.27 ± 0.02 mg/l. Station D recorded the maximum levels Hg with 0.71 ± 0.03 µg/g and the lowest value recorded was at station A with 0.46 ± 0.04 µg/g for sediment. The examination of the gill and the tissue for Hg showed that there were higher Hg levels in the tissues than in the gills for both the lagoon and the creek. More Hg was recorded for fish species in the creek than in the lagoon for the same species (Table 2). Likewise the maximum Hg was observed in the Cichlids; 0.43 ± 0.01, 0.41 ± 0.01 and 0.39 ± 0.02 µg/g wet wt for

Table 4. The mean (± SD) Hg content (µg/g; wet wt) in tissues and gills of fish caught in Lagos lagoon and Abule Agege reek (June–September, 2004).

Fish species	Lagos Lagoon		Abule Agege Creek	
	Gills	Tissues	Gills	Tissues
Sarotherodon melanotheron	0.06 ± 0.02	0.72 ± 0.03	0.06 ± 0.02	0.72 ± 0.05
Hemichromis fasciatus	0.06 ± 0.01	0.64 ± 0.02	0.05 ± 0.02	0.53 ± 0.11
Bathygobius soporator	0.08 ± 0.02	0.60 ± 0.14	0.04 ± 0.02	0.49 ± 0.06
Caranx hippos	0.05 ± 0.01	0.56 ± 0.06	0.04 ± 0.02	0.58 ± 0.04
Tilapia guineensis	0.08 ± 0.01	0.70 ± 0.14	0.06 ± 0.01	0.60 ± 0.06

SD = standard deviation.

S. melatheron, T. guineensis and *H. fasciatus* respectively in the creek. While in the lagoon the highest levels were recorded in *T. guineensis* (0.44 ± 0.07 µg/g wet wt) and the lowest in *C. hippos* (0.24 ± 0.12 µg/g wet wt).

In the rainy season, the maximum pH was observed at station A (7.65 ± 0.06) while the lowest was recorded at station D (7.10 ± 0.02) in the Abule Agege Creek. The highest dissolved oxygen was observed in station D (7.75 ± 0.45) and the lowest at station A (7.25 ± 0.04). The lowest temperature was at station D while the highest was recorded at station C.

The maximum Hg in the water sample was observed in station B while the lowest was recorded for station D. The maximum mercury content in the sediment was recorded in at station D (0.95 ± 0.11 µg/g) while the lowest was recorded for station A (0.71 ± 0.02 µg/g).

The analysis of the fish tissues showed that higher levels of Hg were present in the tissues of fishes studied than in the gills for both of the lagoon and the creek. Specifically for the lagoon species, the maximum Hg levels were estimated for *B. soporator* and *T. guineensis* gills while the minimum was reported for *C. hippos* gill. Comparatively higher levels of Hg were also recorded for *S. melanotheron* tissues with a low level recorded for *C. hippos*. In the creek, the maximum mercury was observed in the gill of both *S. melanotheron* and *T. guineensis* (0.06 ± 0.14 mg/l) while the minimum was recorded in *B. soporator* and *C. hippos* with 0.04 ± 0.02 mg/l. The maximum Hg was recorded in *S. melanotheron* with 0.72 ± 0.05 mg/l while the lowest was recorded in *B. soporator* with 0.49 ± 0.06 mg/l. The summary of Hg concentration in tissues of fish species in lagoon and creek at different season (dry and wet) were presented in Tables 5 and 6.

Hg concentrations were significantly higher in fish tissues caught during the wet season than those caught during dry season. The maximum Hg was observed in *S. melanotheron* with 0.72 ± 0.03 µg/g; wet wt during wet season in Lagos lagoon, likewise in the creek with 0.72 ± 0.05 µg/g wet wt in the same species. While the minimum was recorded for *C. hippo* with 0.56 ± 0.06 µg/g wet wt in Lagos lagoon and *B. soporator* with 0.49 ± 0.06 µg/g wet wt in the creek, respectively.

lagoon is probably as a result of exudates released by the acadja set-up around the stations since fresh plants branches were the major components of the set-up. While lower Hg observed for station D (Abule Agege creek) was related to the buffering effect result from the inflow and outflow of water during high and low tide respectively.

Low surface water temperature of the creek is due to the inflow of cold water from inland during the period. A similar situation has been reported by Emmanuel and Kusemiju (2005) for the same area. The observed DO values fall within the WHO/FEPA allowable range (WHO, 1977).

Generally, more Hg was recorded in sediment than in water for both of the lagoon and creek. It is possible that the settling ability of heavy metal and the inability of the water to retain the metal due to its density allowed for this situation. Higher mercury levels recorded for the creek in

DISCUSSION

The pH of the lagoon was poorly alkaline where as the creek was poorly acidic throughout the whole study. This has been reported in other aquatic ecosystem and does not exceed the acceptable limits as reported by the WHO standard for natural waters (WHO, 1977). These levels has also been observed to have strong effect on the ultimate fate of mercury in an ecosystem (WHO, 1977).The pH of station B was neutral (7.0) while stations A and C were slightly alkaline(7.05 and 7.05) for the dry months probably indicating higher hydroxide ions than hydrogen ions. The high Hg content in water of the comparison with the lagoon sediment may be as a result of binding affinity of clay. The sediment in the creek as reported by Duke (1987) is essentially clay while that of the lagoon is known to be sandy (Oyenekan, 1988). Further more, this could be attributed also to exudates from the decaying plants from the mangrove flora around the creek and the preceding rain forests. This agrees with Hecky et al. (1991) where they reported that decaying plants in the Boreal lake in the northern hemisphere and the resulting increase in organic matter can lead to elevated methyl Hg in fish.

The accumulation of Hg in the tissue and the gills may be affiliated also to the ability of the fish to use the gills to filter water and the grazed food on the interface of the

Table 5. Summary of Hg concentration in tissues of fish species caught in Lagos lagoon at different seasons during 2004.

Species	Dry season		Wet season		Mean difference	Combined SD	t-test	P	P*
	Av	S D	Av	S D					
Sarotherodon melanotheron	0.3425	0.09432	0.7200	0.02828	0.38	0.0829	5.26	0.0063	0.0020
Hemichromis fasciatus	0.3875	0.11644	0.6350	0.02121	0.25	0.1014	2.82	0.0479	0.0207
Caranx hippos	0.2350	0.12077	0.5600	0.05657	0.33	0.1083	3.47	0.0259	0.0113
Tilapia guineensis	0.4350	0.06557	0.7000	0.14121	0.27	0.0906	3.38	0.0279	0.2022
Bathygobius soporator	0.3350	0.18947	0.5950	0.13730	0.26	0.1779	1.69	0.1667	0.1534

Av: average, SD: Standard Deviation, P: probability, P*: P-value when equality of variance is not assumed, Combined SD: Combined Standard Deviation.

Table 6. Summary of Hg concentration in tissues of fish species caught in creek at different seasons during 2004.

Species	Dry season		Wet season		Mean difference	Combined S.D	t-test	P	P*
	Av	S D	Av	S D					
Sarotherodon melanotheron	0.4700	0.07165	0.7150	0.04950	0.25	0.0668	4.23	0.0133	0.0155
Hemichromis fasciatus	0.3550	0.09913	0.5250	0.10607	0.17	0.1009	1.95	0.1236	0.203
Bathygobius soporator	0.2325	0.03500	0.4900	0.05657	0.26	0.0415	7.17	0.0020	0.0588
Caranx hippos	0.3500	0.08525	0.5800	0.04243	0.23	0.0768	3.46	0.0259	0.0125
Tilapia guineensis	0.4125	0.08770	0.6000	0.05657	0.19	0.0810	2.67	0.0557	0.0452

Av: average, SD: Standard Deviation, P: probability, P*: P-value when equality of variance is not assumed, Combined SD: Combined Standard Deviation.

sediment thereby retaining some quantities of metals along with the food. This is further bio - accumulated in gills and later in the tissue of the fish. This agrees with USEPA (2001) where significant high Hg concentration in bottom-feeding and predatory fish species were reported.

Higher Hg levels were estimated in *S. melanotheron, T. guineensis* and *H. fasciatus.* It is possible that Hg had bio-accumulated in the aquatic plant which they feed on. Consequently, the fish species bio-accumulated methyl Hg after feeding on the plants. This species are known as browsers (USEPA, 2001; Emmanuel and Kusemiju, 2005). This disagreed with USEPA (2001) which reported that, methyl mercury biomagnifies up the food chain. *T. guineensis* and *H. fasciatus* are detritus feeder and carnivores respectively, consequently they bio-accumulate Hg probably when they take in their food (biomagnifications) or via their gills (bio-concentration). This agree with More et al. (1998) who are of the view that nearly the entire Hg burden in fish is from their diets.

The concentration of Hg in *C. hippo* and *B. soporator* is probably as result of their predatory nature. This agreed with USEPA (2001) report that predatory fishes had significantly high tissue concentration than the mean tissue concentration in bottom feeders.

The Hg content of the fishes examined were below maximum pemissible levels in the food chemical codex as documented in Sadik (1990) for some local and imported smoked fishes. Levels recorded for this study were also less than those reported for fish from the Mediterranean Sea collected under the UNEP co-ordinated Mediterrenean Pollution Monitoring and Research Programme (UNEP/FAO/WHO 1980).It is suggested that since the fish used for this study were collected from the lagoon and the creek exposed to the influence of urban-runoffs and sewage effluent discharge (Akpata and Ekundayo, 1978) there is hence the need for constant monitoring.

Bearing in mind the standards of acceptable levels by WHO (1977), it is unlikely that any of the analyzed levels of the present study represent an acute risk to biota of the lagoon and the creek or to man as a consumer.

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