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Levels of Mercury in Some Commonly Consumed Fish in Ghana and Their Potential Health Risk to Consumers

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Abstract:A total of seventy five (75) marine fishes comprising eight (8) species were collected from local wholesale markets at Kaneshie and Abeka, Accra, Ghana. The samples obtained from Kaneshie market were *Salmon salar* (Salmon), *Thunnus obesus* (Tuna), *Scomber scombrus*(Atlantic mackerel) and *Clupea harengus* (Atlantic herring) and those obtained from Abeka market were *Centroberyx affinis* (Red fish), *Merluccus paradoxas*(Hake fish), *Scomber trachurus* (Atlantic horse mackerel) and *Melanogrammus aeglefinus*(Haddock). Mercury concentrations in the muscle and liver tissues were determined by cold vapour atomic absorption spectrophotometry using a semi-automated mercury analyzer. Mean mercury concentrations in the muscle ranged from 0.06 to 0.33 µg g^{-1} wet weight, with *Thunnus obesus* having the highest followed by *Merluccus paradoxas* which are all predatory fishes. Mean mercury concentration in the liver tissue ranged from 0.06 to 0.34 µg g^{-1} wet weight, with *Thunnus obesus* having the highest followed by *Melanogrammus aeglefinus*. There was no significant difference between mercury concentrations in the muscle and liver tissues for any of the samples. Low levels of mercury were found in both tissues for all the samples. Mercury concentrations were relatively greater in the tissues of higher trophic level fish such as *Thunnus obesus*, *Merluccus paradoxas* and *Melanogrammus aeglefinus* whereas low trophic level fishes recorded low mercury concentrations. The results obtained for total mercury concentration in the muscles analyzed in this study were below the WHO/FAO threshold limit of 0.5µg/g wet weight, which suggest that the exposure of the general public to mercury through fish consumption can be considered negligible.
Keywords: Mercury, Fish, muscle, liver, Abeka, Kaneshie.

INTRODUCTION

The main supply of protein to the world's population is fish. In Ghana fish is the main protein in diet. It provides high quality proteins when compared with those obtain from milk, beans, meat and eggs [1]. Although fish provides us with good health benefits, several reports show that fish from different environments have been polluted with chemicals. The anthropogenic compounds that are known to be main pollutants on fisheries to date are undoubtedly organochlorine compounds, including pesticides such as dieldrin, dichlorodiphenyltrichloroethane (DDT), and industrial materials such as the polychlorinated biphenyl (PCB) and methyl mercury [2].Beside organic pollutants, heavy metals are also persistent in the aquatic eco-systems. Even though mercury occurs freely in nature [3], it is present in aquatic systems through anthropogenic activities [4].

Mercury exists in fish as methyl mercury, even though it exists in different states within the environment. These therefore put humans at risk to mercury when they consume mercury-contaminated fish [5]. The occurrence of mercury in fish is a common issue for human health risk assessment especially when it comes to humans' food and drug administration (FDA). The joint FAO/WHO professional committees on food additives in provisional allowable weekly intake endorse that in the meal of an adult individual of weight 60 kg, the level of total weekly mercury intake must not exceed 5µg / kg of total mercury and 1.5 µg/kg of methyl mercury [6]. In recent times, mercury contamination in the world over has attracted attention from both scientists and policy makers due to its persistence in the environment and its health effects on humans.

Mercury is a toxic metal and is emitted into the atmosphere naturally from volcanoes and the weathering of rocks. Activities of humans resulting in burning, mining and municipal or medical waste, forest fires, and soils also release mercury in the environment. When mercury is discharged into the atmosphere, it is transported and deposited on land and in water bodies; which accumulate in fish. If higher tropic organisms such as humans consume fish become contaminated

with mercury, it is capable of inducing central nervous system damages by migrating into the brain and crossing the Blood Brain Barrier (BBB).Toxicity therefore results, leading to brain damages, deformities, death etc. Therefore, there is the need to monitor mercury levels in fish from time to time. In this study, mercury concentrations in the muscle and liver tissue will be determined to ascertain the level of toxicity. The liver tissue is used because of its function of cleansing (detoxification) xenobiotic substance in biological organisms. Moreover, the state of the liver in an organism is an indication of its health status [7].

LITERATURE REVIEW

Mercury

According to Schoellhamer [8] mercury happens to be part of the naturally occurring poisonous weighty metallic elements located in soil, air, rocks, living things and water. At room temperature, mercury is identified as the only metal that is liquid. When mercury is in its pure form, which is normally called metallic or elemental, it is an odourless, glistening, silver-white liquid. It has the tendency to evaporate into poisonous and colourless gaseous state that is odourless to people. Historically, mercury has been found to be in existence before 500 BCE. According to Calvert [9], an amalgam with other metals was made from metals by 500BCE. Ancient Chinese and Hindus were among the earlier people who came into contact with mercury.

Environmental cycling of mercury

According to [10], mercury species are required to hover between the soil, vegetation media and water when released.

Atmospheric cycling of mercury

Panel [11], reported that on the local, regional and global scale, mercury is widely seen as a problem to the environment during atmospheric dispersion. The physical and the chemical forms of the Hg and the extent of inter-version among species determine the spatial scale. According to [12], Hg (II) is capable of travelling a very long distance. It may go as far as tens of thousands of kilometers. The Hg (p) is however likely to be dropped at intermediate distances depending on their mass or the diameter of their aerosol.

Atmospheric reactions of mercury

A laboratory research by [13] revealed that under the relevant conditions of the atmosphere, mercury will have only a few chemical reactions. Hg involving the ozone layer has been identified as in the gas phase through the process of oxidation. This according to [14] has also been identified in the aqueous phase. The overall Hg atmospheric cycling needs both phases. Till now, only the aqueous-phase reduction of Hg (II) by Sulphur (IV) has been identified as one of such process. This is in accordance with [15].The essence of this particular reaction perhaps is limited on a worldwide scale with regard to it importance. However, extra reduction procedures may occur (e.g., photo-chemically started reduction of mercury (II) ion).

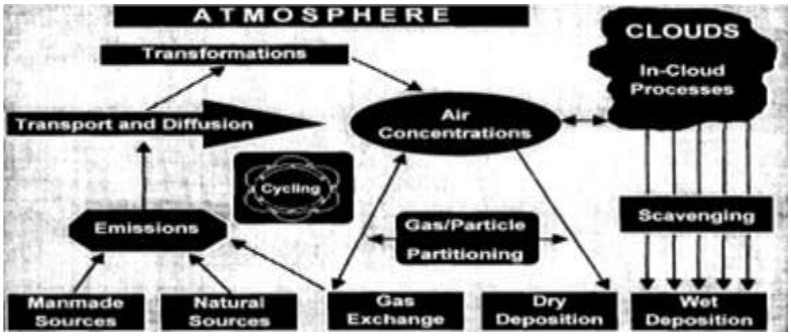

Fig-1: Conceptual framework of the atmospheric emissions-to-deposition cycle for mercury [35] EARTH'S SURFACE (water, soil, vegetation)

Air-surface exchange of mercury

One of the differentiating factors of mercury when mercury is compared to heavy metals is its ability to reoccur in the environment or the atmosphere. According to [11], an estimation of 2 x $l0^5$ tons mercury has been deposited in the terrestrial since 1890. A significant change in the atmosphere globally may be due to re-emission of Hg. processes.

MATERIALS AND METHODS

The fish species were bought from Kaneshie and Abeka markets in the Greater Accra region depending on the species available for sale. The samples obtained from Kaneshie market were *Salmonsalar* (salmon), *Thunnus obesus* (tuna), *Scomber scombrus* (Atlantic mackerel) and *Clupea harengus* (Atlantic herring) and those obtained from Abeka market were *Centroberyx affinis* (Red fish), *Merluccus paradoxas*(Hake fish), *Scomber trachurus*(Atlantic

horse mackerel) and *Melanogrammus aeglefinus*(haddock). Samples obtained were therefore reflective of species meant for consumption. A total of seventy- five (75) fishes covering eight (8) species were obtained. The samples were then transported to the laboratory, identified and kept in a freezer at -20^{o}C prior to preparation for chemical analysis. The samples were defrosted, washed with distilled water and dried on tissue paper. The total length and body weight of each fish was taken. A portion of the edible muscle tissue was removed from the dorsal part of each fish. The liver of each sample was obtained after careful dissection with the aid of stainless steel knife which has been cleaned thoroughly using distilled water. The tissues obtained were then homogenized and used for digestion.

Digestion

The fish tissues were digested for total mercury determination by an open tube procedure [16]. In the digestion method, 0.5 g of the homogenized tissue was weighed into a 50 ml digestion flask and 1ml of distilled water added. Mixture of 2 ml HNO_3-$HClO_3$ (1:1) and 5 ml of H_2SO4 were added in turns. The mixture was then heated at a temperature of 200^{0}C, for 30 minutes. The sample solution was then cooled to room temperature and diluted to the 50 ml mark with double distilled water. *Chart 1,* below provides a summary of the digestion process. A blank and standard solution digests using 25 and 50 of 1 mg ml^{-1} standard Hg solution were subjected to the same treatment.

Sample (0.5 g in 50 ml digestion tube)
H_2O, 1ml
HNO_3: $HClO_4$ (1:1), 2 ml
H_2SO_4, 5 ml
Heat at 200^{0}C for 30 minutes
Cool to room temperature
↓
Digested sample
↓
H_2O, make up to the mark
Sample Solution, 5ml
↓
10 % $SnCl_2$, 0.5ml
↓
AAS (Analyzer)

Chart 1 Summary of Analytical procedures for total mercury in tissue samples

Determination of Mercury

Determination of mercury in all the digests was carried out by cold vapour atomic absorption spectrophotometer using a semi-automatic Mercury Analyzer Model Hg-5000 (Sanso Seisakusho Co., Ltd., Japan) developed at the National Institute of Minamata Disease (NIMD).The analyzer is an instrument designed specifically for the measurement of mercury using cold vapour technique. The analyzer consists of an air circulation pump, a reaction vessel, tin (II) chloride dispenser, an acidic gas trap and a four- way stop-work with tygon tubes to which is attached a ball valve. The operation of the ball valve and the air circulation pump are controlled by the microprocessor.

During the determination, a known volume of the sample solution i.e. 5 ml was introduced into the reaction vessel using a micropipette (1-5ml). The reaction vessel was immediately closed tightly and 0.5 ml of 10% (w/v) tin (II) chloride in 1M HCl was added from a dispenser for the reduction reaction.

During this time, air was circulated through the four-way stop-cork to allow the mercury vapour to come to equilibrium and the acidic gases produced by the reaction also swept into the sodium hydroxide solution. After 30 seconds, the four-way stop-cork was rotated through 90^0 and the mercury vapour was swept into the absorption cell. Standard solution used for the calibration of the analyzer includes solution containing 25, 50 and 100ng Hg.

STATISTICAL ANALYSIS

The descriptive statistics (mean, standard deviation, range) and one-way analysis of variance (ANOVA) were conducted using Microsoft excel from windows Microsoft Office 2010. A one-way ANOVA statistical procedure was employed in the assessment of variation in mercury concentrations among fish species. A *p*-value of less than 0.05 was considered to indicate statistical significance. Two sample *t*-tests were used to compare mercury level between the two tissues. Linear regression analysis was conducted to determine the strength of association between mercury concentration in the liver, muscle and total (length & weight) of fish samples.

RESULTS AND DISCUSSION

Concentrations of mercury in liver and muscle tissue of fish species

A total of seventy- five fish samples covering eight (8) species namely; *Salmon salar* (Salmon), *Thunnus obesus*(Tuna), *Scomber scombrus* (Atlantic mackerel), *Clupea harengus* (Atlantic herring), *Centroberyx affinis* (Redfish), *Merluccus paradoxas* (Hake fish), *Scomber trachurus* (Atlantic horse mackerel) and *Melanogrammus aeglefinus*(Haddock) were analyzed for total mercury. Fish analyzed were grouped based on their trophic level;non-predatory andpredatory comprising planctivorous and benthophagous. Isa *et al*. (1998) provides the basis for establishing this grouping. Majority (66.7 %) of the fishes were non-predatory (i.e.40% planctivorous, 13.33% zooplankton and 13.33% benthophagous) and (33.3%) predatory. Feeding habit classification is important because it may represent the basis for accumulation of mercury in fishes. *Table 1* shows the classification of fish samples used in this study.

Table-1: Characteristics of fish samples

Scientific name	Common name	Sample size (n)	Total length(cm) mean(range)	Total weight(g) mean(range)	Feeding habit
Clupea harengus	Atlantic herring	10	24.1 (21.5 -25.7)	137.3 (124.9 - 159.9)	Zooplankton
Scomberscombre	Atlantic mackerel	10	25.3 (21.5 - 27.5)	150.3 (80.5 - 187.0)	Planctivorous
Salmonsalar	Salmon	10	33.7 (32.1- 35.4)	294.5(245.0 - 337.2)	Predatory
Thunnus obesus	Tuna	5	49.4(45.7 - 52.3)	1003.3 (849.7 - 1158.1)	Predatory
Centroberyx affinis	Red fish	10	22.6 (20.5 - 24.4)	168.6(112.8 - 203.6)	Planctivorous
Merluccus paradoxas	Hake fish	10	38.5(37.5 - 40.1)	410.2(396.5 - 421.2)	Predatory
Melanogrammus aeglefinus	Haddock	10	25.5 (23.1 - 27.2)	154.1 (129.1- 175.8)	Benthophang ous
Scomber trachurus	Atlantic horse mackerel	10	24.5 (23.5- 26.3)	104.1 (94.6 - 110.9)	Planctivorous

Total mercury concentration was estimated in the liver and muscle tissues based on an open tube digestion procedure, followed by CVAA - spectrophotometry. The methodology used was validated by carrying out recovery analysis and analysis of standard certified material.

Results obtained from the recovery analysis ranged between 91.84% and 97.62%, which is an indication that the methodology employed is accurate [17]. For precision, each sample was analyzed repeatedly and results obtained agreed to 95%.

Table 2 shows the results of the matrix spike recoveries of two fish samples *Centroberyx affinis* and *Merluccus paradoxas*.

Table-2: Recovery of mercury from *Centroberyx affinis* and *Merluccus paradoxas*

Sample	Hg added(ng)	Hg found (ng)	Hg recovered (ng)	% Recovered
Centroberyx affinis	0	154.57	-	-
0.50g	25	177.53	22.96	91.84
	50	202.98	48.41	96.82
Merluccus paradoxas	0	148.83	-	-
0.50g	25	167.43	23.6	94.4
	50	197.64	48.81	97.62

The three blank solutions on analysis gave a standard deviation of 0.00052. Analysis of the certified reference material gave mean mercury concentration of 0.22 µg/g wet wt.and standard deviation of 0.01. This agrees with the results of certified reference material as analyzed by IAEA from Austria. i.e. mean mercury concentration ranged from between 0.19 and 0.25µg/g wet wt.). This is an indication that the methodology used for the analysis of the samples obtained is valid. Table 3 below shows the results obtained for the analysis of the Certified Reference Material (CRM), fish Homogenate IAEA-407.

Table-3: Mercury concentration for certified reference material (CRM)

Sample Code	Mass(g)	Conc. (ng/g) wet wt.	Conc. µg/g wet wt.
CRM1	0.554	213.40	0.21
CRM2	0.506	205.61	0.21
CRM3	0.53	222.65	0.22
CRM4	0.521	226.49	0.23
CRM5	0.525	233.26	0.23

Two tissues namely liver and muscle obtained from the fish samples were analyzed for total mercuryby CV-AAS and the results of the mercury concentrations and mean standard deviations are shown in *Table 4*.

Table-4: Total mercury concentrations (µg/g wet wt.) in liver and muscle of fish species

Scientific name	Sample size	Portion	Total mercury (µg/g wet wt.) Mean ± SD	(Range)
Clupea harengus	10	muscle	0.04 ± 0.02	(0.01 - 0.06)
		liver	0.11 ± 0.17	(0.01-0.60)
Scomberscombre	10	muscle	0.06 ± 0.04	(0.01 -0.14)
		liver	0.06 ± 0.04	(0.02 -0.14)
Salmon salar	10	muscle	0.07 ± 0.03	(0.03 -0.12)
		liver	0.13 ± 0.07	(0.03 -0.16)
Thunnus obesus	5	muscle	0.33 ± 0.03	(0.30 -0.37)
		liver	0.34 ± 0.03	(0.30 - 0.37)
Centroberyx affinis	10	muscle	0.16 ± 0.14	(0.07 -0.55)
		liver	0.12 ± 0.05	(0.07 -0.24)
Merluccus paradoxas	10	muscle	0.25 ± 0.21	(0.08 -0.60)
		liver	0.10 ± 0.18	(0.01- 0.60)
Melanogrammus aeglefinus	10	muscle	0.06 ± 0.03	(0.02- 0.12)
		liver	0.15 ± 0.22	(0.02 -0.74)
Scomber trachurus	10	muscle	0.11 ± 0.06	(0.07- 0.21)
		liver	0.11 ± 0.12	(0.02 -0.55)

The total mercury concentrations measured in this study vary between 0.01 and 0.60 µg g^{-1} for the muscle and, 0.01 and 0.74 µg g^{-1} wet weights for the liver samples. Two samples of *Merluccus paradoxas* recorded the highest mercury concentration of 0.60 µgg^{-1} followed by a sample of *Centroberyx affinis* (0.55 µg g^{-1}) and *Thunnus obesus* (0.30-0.37 µg / g).

Even though higher concentrations of THg was found in the muscle of the two samples of *Merluccus paradoxas* and one sample of *Centroberyx affinis*, all other samples recorded total mercury concentrations which are far below the recommended limit (0.5 µg/g) given by FAO/WHO [18] which was accepted by most countries [19]. For the THg concentrations in the liver samples analyzed, one sample of *Clupea harengus* (0.60 µg/g), *Merluccus paradoxas* (0.60 µg/g), *Melanogrammusaglefinus* (0.74 µg/g) & *Scomber trachurus* (0.55 µg g^{-1}) recorded mercury concentrations higher than 0.5 µg/g as required by FAO/WHO (1972). Also from the results of this study, *Thunnus obesus* recorded the highest mean mercury concentrations for both muscle (0.33 ± 0.03) and liver (0.34 ± 0.03), followed by *Merluccus paradoxas* (0.25 ± 0.2) for muscle and *Melanogrammus aeglefinus* (0.15 ± 0.22) for liver.

*Clupea harengus*gave the lowest mean mercury concentration upon the analysis of the muscle tissues (0.04 ± 0.02), whose food is mostly water plant & zooplankton [20].

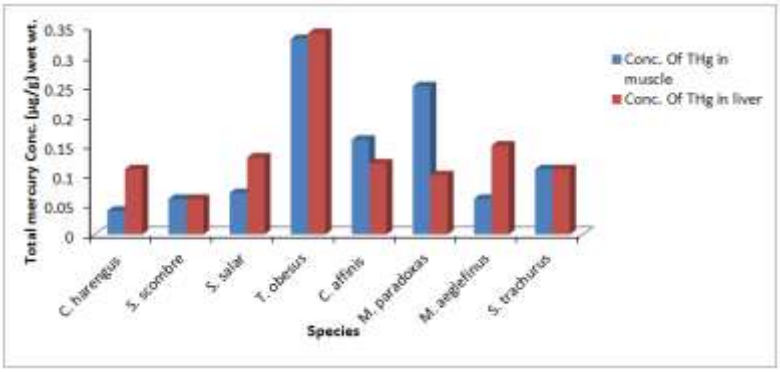

Fig-2: mean mercury concentration in muscle and liver tissues of fish samples

Many researchers have been able to establish that mercury levels in the liver far outweigh that found in the muscle [21]. With regard to results from this study, t-test was employed to check if significant difference in terms of mercury level exists between the two tissues. The results obtained from the t-test analysis show that no significant difference exists between the two tissues (p >0.05) in any of the samples. However, significant differences exist between the muscle and liver tissues among *Thunnus obesus*, *Merluccus paradoxas*, *Salmonsalar and Melanogrammus aeglefinus* which are predatory fishes.

Mercury concentrations in predatory fishes established from the samples analyzed, are in agreement with those obtain from Agusa*et al*, [21]. Voegborlo and Akagi[22] also reported mean THg levels ranging from 0.004 to 0.122 µg / g wet weight for thirteen different species; which values are lower or similar to those found in this current study. Generally low levels of mercury concentrations were obtained from this study and when these results are compared to most results from different areas of the globe, one can establish the fact that the fish samples obtained for this research are not from polluted environments. For instance research work on mercury in edible muscles of several fish samples bought in a supermarket in New Jersey in 2004 gave mercury levels ranging from 0.01 - 0.65 µg/g [23], which agree with the results of this study.

Comparing the results obtained for the analysis of the liver tissue in this study with other researchers [24, 25], the results are in complete agreement with their results. Some researchers have established that the liver in fish is the main organ that often gives high level of mercury [25, 26]. With regard to this study, the concentrations of mercury obtained are very low when compared to results obtained from less developed countries like Mozambique and Reunion Islands [19, 27]. This is also an indication that geographical position or location and factors such as metabolic difference determine the extent of toxicity in fish samples [28].

In the Gulf of Mexico, samples of commercial wahoo fish gave mercury concentrations ranging from 0.95 to 3.31 µg/g wet wt. Wahoo fish also obtained in the Bermuda region gave mercury concentrations ranging from 0.06µg/g to 1.0µg/g whereas Fijian wahoo had mean mercury concentration of 0.17 µg/g. Research on swordfish obtained from six different geological locations from grand banks to the Caribbean waters showed that, the mean mercury concentrations vary significantly from one location to the other. Predatory fishes accumulate more mercury than any other kind. The level of accumulation depends on a number of factors such as the size, its food and the extent of pollution with regard to its environment [29].The extent of mercury level in a fish species correlate positively with the extent of mercury in its environment and to its level within a food web [30]. Positive correlations between mercury concentration and size have been previously found in sharks [31] and freshwater eels [32].

Samples for this research work were purchased from two different market locations in the capital city of Ghana, Accra, and it is thus difficult to trace their origin. Looking at the various fishes bought, it is difficult to determine the source of pollution and its geographical location. Much information was not obtained from the market concerning the sources of the fishes. Since sampling was done based on purchasing the fish samples from the market, it is difficult to relate and interpret the source of mercury due to the lack of geological location of the fish samples.

In this study, linear regressions were used to determine correlations between fresh body weight, length of the fish samples and THg concentration in the muscle and liver. A total of eight species were subjected to regression studies. The correlations are indicated as regression lines in appendix II. There was positive correlation between THg concentration & fresh body mass and length of all fish samples. Significant positive correlation was observed between the fresh body mass & THg concentration within the flesh of *Thunnus obesus* and *Merluccus paradoxas* with regression coefficient r^2= 0.8822 and 0.616, respectively. This is in agreement with the suggestion that, for carnivorous fishes, good correlation normally exists [32, 31, 33]. For linear regression between the Hg concentration in flesh, liver and total length of fish samples, significant correlation coefficients were observed for *Thunnus obesus*(0.968) and *Scomber trachurus* (0.433).

Hazard assessment

In this study, risks to health from mercury in fish were assessed by equating estimations of food exposure with the Provisional Tolerable Weekly Intakes (PTWIs) suggested by the joint FAO/WHO Expert Committee on Food Additives (JECFA). For total mercury, the PTWI guideline of 5.0 µg/kg body wt / wk was used in this study [24].

According to recent surveys, the average Ghanaian consumes about 78 g of shell fish and fish in a day or 546 g/ week [28] which was used in the health-risk assessment. Human health risk in terms of hazard quotient (HQ) was determined by comparing the estimated weekly intake of mercury with the PTWI. The average body weight (BW) for an adult (60 kg) was used in the calculation and PTWI is the guideline value of the individual metal (µg/ kg body wt. / wk.). The average THg intakes from all the fish species in this study were much lower than the recommended value as shown in *Table 5*. The HQ for THg in each fish species was less than one, indicating no potential health risk to the consumers. The low % intakes (PTWI) of the different fish species indicate low exposure to THg. The mean % intake of PTWI for the fish species (7.28 – 60.06 %) was in agreement with the 1.5- 61% reported for the average consumer in Hong Kong [34].

Table-5: Mean dietary intake of total mercury

Scientific name	Mean Hg concentration	Weekly Ingestion Rate (g/person/wk)	Estimated weekly intake(µg/kg body weight/wk)	PTWI (µg/kg body weight/wk.)	Mean Intake % of PTWI	HQ
Clupea harengus	0.04	546	0.36	5	7.28	0.073
Scomberscombre	0.06	546	0.55	5	10.92	0.109
Salmon salar	0.07	546	0.64	5	12.74	0.127
Thunnus obesus	0.33	546	3.00	5	60.06	0.601
Centroberyx affinis	0.16	546	1.46	5	29.12	0.291
Merluccus paradoxas	0.25	546	2.28	5	45.50	0.455
Melanogrammus aeglefinus	0.06	546	0.55	5	10.92	0.109
Scomber trachurus	0.11	546	1.00	5	20.02	0.200

CONCLUSION

As per the study carried out, mercury concentrations obtained are of low levels. The THg concentrations in the edible fish muscles were generally below the FAO/WHO maximum permissible limit (0.5 μgg^{-1} wet weights). Generally, mercury levels in this study indicated that THg concentrations in the species increased with increasing body length and weight for predatory species. Also, Mean THg concentrations in the muscle and liver tissues of the fish analyzed ranged from 0.04 to 0.33 µg/g (for muscle tissue) and 0.06 to 0.34 (for liver tissue). *Thunnus obesus* (a predatory fish) recorded the highest mean THg concentration (for both tissues) among all the species analyzed. Again, Correlation between THg concentration in the muscle and body weight / total length was significantly positive for the predatory fishes, with *Thunnus obesus* recording the highest correlation coefficients. On the other hand Correlation between THg concentration in the liver and fresh body weight / total length was significantly positive in terms of predatory fishes with *Thunnus obesus* recording the highest correlation coefficients. Last but not the least, the survey revealed that hazard ratios obtained were all below one (1) i.e. ($\leq$ 1) which indicates that, no health risk is posed if the fishes used for this study were consumed. The highest hazard ratio obtained was 0.60 for *Thunnus obesus* which is a predatory fish.

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