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Mercury Content in Yellowfin Tuna (*Thunnus albacares*) and Swordfish (*Xiphias gladius*) and Estimation of Mercury Intake

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Abstract Mercury (Hg) is a pollutant of global importance that adversely impacts ecological and human health. A total of 140 *Thunnus albacores* (yellowfin tuna) and 176 of *Xiphias gladius* (swordfish) samples collected from the Indian Ocean around Sri Lanka in the period of July 2009 to March 2010 were examined for total mercury content and used to evaluate the mercury intake of Sri Lankan population. Total mercury concentrations of muscular tissues of fish were determined using cold vapor atomic absorption spectrometry. The results indicated that the highest mercury concentration were noted in swordfish 0.90±0.52 mg/kg, wet weight and followed by yellowfin tuna (0.30±0.18 mg/kg, wet weight). The projected intake values of mercury through human consumption were calculated (µg kg^{-1} body wt. $weekly^{-1}$) and were compared with those of provisional tolerable weekly intake (PTWI) per kg body weight as stipulated by the Food and Agriculture Organization/World Health Organization (FAO/WHO) and Joint Expert Committee on Food Additives (JECFA).The estimated PTWI were lower than for yellowfin tuna and swordfish stipulated guidelines and therefore, was not considered to pose adverse effects to the humans with the present fish consumption volume.

Keywords: *mercury concentration, fish, food safety, PTWI*

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1. Introduction

Sri Lanka is a small tropical island in the Indian Ocean off the southern tip of India and having an exclusive economic zonal (EEZ) area of 517,000 km^2. Capture fisheries produced 385,270 MT and total fisheries production including aquaculture was 444,830 MT in 2011. This was an increase in seafood production by 15.9% compared to the previous year(MFAR, 2012).The Sri Lankan fisheries sector is an important contributor to export value; and fish is the main source of animal protein for the Sri Lankan population. Surrounded by the ocean, Sri Lanka is abundant in seafood, which is easy to acquire as the source of protein in diet.

Some metal like copper (Cu) and zinc (Zn) are essential for fish metabolism while other metals like mercury (Hg), lead (Pb) and cadmium (Cd) have no known biological role. Mercury is one of the ancient metals which has been used by many cultures for a variety of symbolic and useful purposes, such as in good luck charms, to ward off evil, as material for ceremony objects (Egyptians), as colorant and as cosmetics [5,14].

Mercury is a natural element in the environment, which has been used for thousands of years and it is released into the atmosphere naturally by degassing from earth crust, ocean and through human activities, primarily from burning household and industrial waste and fossil fuels. Once released, mercury undergoes a series of complex transformations and cycles between atmosphere, ocean and land. The three chemical forms of mercury are elemental or metallicmercury (Hg^0), inorganic mercury [mercurous (Hg^{2+}) and mercuric (Hg) cations] and organic mercury. Trace amounts of Hg are soluble in bodies of water, where bacteria can cause chemical changes that transform Hg to methyl Hg, that is a more toxic form and almost all of the Hg in fish meat is in this form. Fish absorb methyl Hg from water as they passes over their gills and as they feed on aquatic organisms [6]. Larger predator fish like swordfish, yellowfin tuna and sharks are at the top of the food web are exposed to higher levels of methyl Hg from their prey [1,16]. Further carnivorous fish species especially those in hierarchy of the food web can bio-accumulate trace metals, and contain high levels of trace metals that can be harmful to consumers' health. The amount of mercury in a particular species varies greatly, depending on the size of the fish, its position in the food chain, and where the place of caught.

The World Trade Organization (WTO) agreement, to which Sri Lanka is a signatory, requires the export or import country to certify that the product is of good quality and safe before it is consumed [15]. In addition, there are many international, regional and national regulations regarding the seafood safety. Sri Lanka has yet to implement appropriate procedures to ascertain the quality of seafood before it is exported or put on to the local market. The export regulation of seafood products from Sri Lanka is based mainly on EU regulations. The maximum acceptable concentration of trace metals differs from one seafood to another based on risk assessment. EU has established maximum permitted levels for three trace metals i.e. cadmium, lead and mercury in seafood and Sri Lanka follows this regulation. According to the EU regulation 2073/2005, 1881/2006, 629/2008 and Sri Lanka export regulation (No 1528/7) the maximum permissible level of contaminants in target fish species in this study are 1mg/kg, wet weight basis.

Food consumption is the main source for Hg exposure in general public. In Spain, Finland and Japan, the studies revealed that fishermen and seafood-lovers who consumed more seafood had higher mercury intake and higher mercury level in their tissues than the general public and vegetarians [2,11]. Thus indicates that seafood consumption is contributed to mercury intake. The definition of PTWI (Provisional Tolerable Weekly Intake), as set by the Food and Agriculture Organization/World Health Organization, Joint Expert Committee on Food Additives (JECFA), is the maximum amount of a contaminant to which a person can be exposed per week over a lifetime without an unacceptable risk of health effects. Accordingly test the food safety, the Hg intakes via dietary consumption of these fish are calculated and compared with the PTWI of 5 $\mu g/kg^{-1}$ body weight, set by the JECFA in 2003.

To the best of our knowledge, there have been a very few publications on Hg content of fish from the Indian Ocean around Sri Lanka. In this study, we emphasized on the level of Hg in the muscle tissues of yellowfin tuna and swordfish collected from Sri Lanka. Further, the determined levels of Hg are compared to the PTWI for toxic trace metals as set by the JECFA.

2. Materials and Methods

2.1. Sampling

One hundred and forty yellowfin tuna samples and one hundred and seventy six sward fish samples were obtained from Galle, Mutwal (Colombo), Negombo and Trincomallee areas of Sri Lanka during July 2009 to March 2010. The length and weight of fish were measured. Then approximately 250 g of edible portion of the belly area was obtained and packed in ice. Samples were transported in an insulated container to the analytical chemistry laboratory/NARA and stored at -20 °C until further analysis.

2.2. Reagents

Ultra-pure water (18.2 MΩcm) from Barnsted, Esay pure LF system was used for the preparation of all standards and reagents. All chemical used in the present study were analytical regent grade or better. The standard solutions of Hg at 1000 mg/L were obtained from Fluka, Switzerland. All glassware used were first soaked overnight in liquid detergent and then thoroughly rinsed with tap water. Then the glassware soaked in 10% (v/v) HNO_3 overnight and subsequent rinsing was performed using de-ionized water. Then all glassware were oven dried and plastic ware were air dried prior to use.

2.3. Mercury Analysis

Approximately 1 g of fish sample was weighed in to microwave digestion tube and weight was taken accurately to four decimal places. Then 10 mL of Conc.HNO_3 acid (65%,'AR'-Sigma) was added to sample and allowed for 15 minutes in fume hood for pre digestion. Each sample was analyzed in duplicate. After pre digestion, samples were digested under pressure in a closed vessel heated by microwaves using a microwave-accelerated system (CEM-Mass XP-1500+).The digest were allowed to cool to room temperature and pressure was released carefully by opening the valve. Then the digested fish samples were transferred into 50mL volumetric flask and interior of digestion tube was washed down 3 times and made up to 50 mL with deionized water as diluents. The determination of total mercury was performed by using a flow injection system (Varian VGA 77) on a Varian 240 FS atomic absorption spectrophotometer where mercury cold vapor was generated.

2.4. Quality Control

Quality control for each analytical batch consisted of standard quality control materials (canned fish muscle, T/0774, from Fapas, UK), spiked sample (spiked with 10 ppb) and ten reagent blanks. The quality control samples were digested according to the procedure used in the preparation of fish samples for the Hg analysis.

2.5. Risk Assessment

Risk of mercury intake was estimated based on calculating PTWI and comparing with, 5µg/kg body weight/week, set by WHO. PTWI of Hg was calculated by assuming the average adults weight as 55 kg.

2.6. Statistical Analysis

One-way Analysis of Variance (one-way ANOVA)was performed to test the significance of variation among the seafood. The statistical significance level was set at $p < 0.05$ for all tests.

3. Results and Discussion

The suitability of the method was evaluated in terms of their limit of detection (LOD) and recovery levels using spiked samples and certified quality control materials. The detection limit was calculated as the 3-fold of the standard deviation of ten blanks. In this study, the recovery obtained for T/0774 was 92% (18.31 ± 1.3µg/kg, n = 7). LOD was determined as a value of 0.0028 mg/kg. The results obtained for both certified quality control samples and spiked fish samples indicated that the method used was validated for the determination of Hg content in the samples.

The length, weight and total Hg content of the samples of yellowfin tuna (n=140) and swordfish (n=176) in the Indian Ocean around Sri Lanka are shown in Table 1. The mean Hg levels of two fish species were significantly different (p<0.05, Anova).

Table 1. The length (cm), weight (kg) and Hg concentration (mg/kg, wet weight basis) of yellowfin tuna and swordfish (Ranges are given in parenthesis)

	Length (cm)	Weight (kg)	Mercury (mg/kg; wet weight basis)
Yellowfin tuna	123.4 (64.0-173.0)	45.3 (18.0-83.5)	0.30 (ND-0.98)
Swordfish	136.5 (45.0-278.0)	44.9 (11.8-112.0)	0.90 (0.18-2.58)

Based on the results, 32% of swordfish samples were exceeded the maximum acceptable limits (1 mg/kg) of European legislation, but not any single sample of yellowfin tuna was not exceeded. The limits In generally Hg level of carnivorous fish species is higher than the herbivorous fish species [2]. According to the results, Hg concentration of swordfish was higher than yellowfin tuna. This can be explained by the trophic level and the characters of bioaccumulation and bio-magnification of Hg in the food chain. As compared with some other countries, the Hg level of swordfish in this study is lower than Canada (1.82 mg/kg), Fiji (1.81 mg/kg) and Reunion Island (1.24 mg/kg), but it is higher than Atlantic Ocean (0.62 mg/kg) and Mozambican Island (0.38 mg/kg) (Mendez *et al.*, 2011, Dabeka *et al.*, 2004, Kumar *et al.*, 2004, Jessica *et al.*, 2006). The Hg level of yellowfin tuna of this study is lower than Hawai (0.54 mg/kg) and Gulf of Guinea (0.42 mg/kg) and it is higher than Mozambican Island (0.13 mg/kg), Reunion Island (0.21 mg/kg) and Pacific Ocean (0.21 mg/kg) (Peterson *et al.*, 1973, Jessica *et al.*, 2006, Kraepiel *et al.*, 2003). These indicate that the Hg levels of large predatory fish, such as tuna and swordfish, captured around Indian Ocean, Sri Lanka are similar with other countries. The differences might be caused by different sample sizes, ages, season of captured and the characteristics of captured environment.

In this study, a reference adult, having a weight of 55 kg was taken to determine the amount of Hg consumed by an average Sri Lankan. The provisional tolerable weekly intake (PTWI) values for adult and children are given in Table 2. The estimated of the weekly fish consumption based on the average seafood consumption of Sri Lankan was 207 g and the contribution of large marine fish was 83 g (MOFAR, 2012).

Table 2. Estimated weekly intake of Hg from yellowfin tuna and swordfish

Species	Weekly consumption of studied fish without exceeding PTWI	Probable Weekly Intake (PWI), Hg (Sri Lankan consume 83 g, large pelagic fish/week)	% of PTWI
Yellowfin tuna	928 g	24.9 μg	9
Swordfish	305 g	74.7 μg	27

According to the Table 2, Hg intake from yellowfin tuna and swordfish of Sri Lankan was below the PTWI standards. As to the contribution of PTWI limits Hg intake from yellowfin tuna and swordfish by general public, the yellowfin tuna was accounted for 9% and swordfish accounted 27%. Therefore, owing to human health concerns and the necessity of reducing the risk of for Hg exposure, it is suggested that the average 55 kg adult in Sri Lanka can consume no more than six servings (50 g per serving)/week, or 305 g/week of swordfish. Furthermore, considering yellowfin tuna, same body size adult can consume 928 g/week without exceeding PTWI limits. Compared with other countries, Hg intake of Sri Lankan by the studied fish species quite similar of Taiwanese (30.4 μg/week), 1/10 of Canadians (4.9 μg/week), 1.25 fold of Spain (62.4 μg/week) and 24 fold of Japan (1190 μg/week) [4,9,11]. Furthermore, the fish consumption amount of Taiwanese is 2 times higher than Sri Lanka, 3 times higher than Spain and 6 times higher than Japan [2].

Recommendations suggest that 0.8g of protein/kg of healthy body weight per day [15], while other sources suggest that higher intakes of 1-1.4 g of protein per pound of body weight for enhanced athletes or 2 g/pound of body weight for those with or trying to attain a large muscle mass [13]. The amount of fish that should be consumed to fulfill recommended weekly protein intake was also calculated. According to that the average Asian adult (55 kg body weight) need 308 g of protein. Protein percentage of yellowfin tuna and swordfish were 23.6% and 18%, respectively (www.b2becuador.net, accessed on 01st July 2011). By assuming that the total protein requirement fulfilled by yellowfin tuna and swordfish alone required 1.31 kg yellowfin tuna and 1.71 kg swordfish. According to that the amount of protein requirement can't be achieved consumed only by swordfish or yellowfin tuna without exceeding recommended PTWI value for Hg.

4. Conclusion

In conclusion, the study recommended that the amount of fish consumption and the Hg levels of yellowfin tuna and swordfish are the two main factors that affect the level of the Hg intake for the general public. Therefore, the dietary reference regarding the fish consumption should be addressed. In general, the risk of excessive Hg intake is minimal based on the seafood consumption pattern in Sri Lanka. However, fishermen, fishmongers, their families, or seafood-lovers who consume more seafood can have higher risk of Hg intake. To those people, consumption of seafood with low Hg level or the reduction of seafood consumption is endorsed. Based on the PTWI set by the Joint FAO/WHO Expert Committee on Food Additives(JECFA) no more than 300 g/week of swordfish and 928 g/week of yellowfin tuna can be consumed by the general public, without exceeding the PTWI limits.

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