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## Determination of cadmium, lead and mercury residual levels in meat of canned light tuna (*Katsuwonus pelamis* and *Thunnus albacares*) and fresh little tunny (*Euthynnus alletteratus*) in Libya

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### Abstract

Surveillance for mercury (Hg), lead (Pb) and cadmium (Cd) contamination in tuna products is crucial for consumer food safety. Hg, Pb and Cd contaminants were monitored in a total of 60 specimens of fresh little tunny (*Euthynnus alletteratus*) and popular brands of skipjack and yellowfin (*Katsuwonus pelamis* and *Thunnus albacares*) canned tuna commercially available in Tripoli, Libya. Direct Mercury Analyzer (DMA-80) was implemented for determination of total Hg level and graphite furnace atomic absorption spectrometry (GFAAS) was employed for determination of Cd and Pb concentrations. The results indicated that Hg had the highest concentration level and Cd had the lowest concentration level either in tested canned tuna or fresh little tunny samples. The average concentration of Hg in fresh little tunny samples was  $1.185 \pm 0.968$  mg kg<sup>-1</sup> wet weight (ww) and often exceeded the standard permissible limit. In addition, canned yellowfin tuna had the lowest levels of Cd ( $0.027 \pm 0.026$  mg kg<sup>-1</sup> ww), Pb ( $0.075 \pm 0.071$ ) and Hg ( $0.163 \pm 0.122$  mg kg<sup>-1</sup> ww). Results of the current surveillance indicated that canned skipjack and yellowfin tuna sold in Tripoli markets show contaminant levels well under the European thresholds adopted for Cd, Pb and Hg. However, consumption of large quantities of Mediterranean little tunny products significantly increases human exposure to the risk of Hg toxicity.

**Keywords:** Cadmium, Canned tuna, Lead, Little tunny, Mercury.

### Introduction

Marine organisms could bioaccumulate numerous inorganic pollutants, with mercury (Hg), lead (Pb) and cadmium (Cd) contaminants being the most commonly studied elements. These elements are characterized by bioaccumulation, biomagnification in the food chain and tendency to persist in the environment (Morgano *et al.*, 2014). Since fish is the last link in the aquatic food chain, the trace elements concentrations in many fish species have been determined in relation to the metal content of the aquatic environment (Çelik and Oehlenschläger, 2007).

Tuna species are large pelagic predator fish that have high performance with very high metabolism rates and, thus, high food intake rates, a property that elevates the exposure to trace elements bioaccumulation (Kojadinovic *et al.*, 2007). Little tunny (*Euthynnus alletteratus*) and Atlantic bluefin tuna (*Thunnus thynnus*) constitute an important component of the total pelagic fishing activities in Libya (Tawil, 2002). During late spring and early summer, large numbers of tuna migrate to Libya's Gulf of Sidra, which constitutes one of world's most critical spawning areas for *T. thynnus* tuna in the Mediterranean Sea.

Little tunny (*E. alletteratus*, Rafinesque, 1810) is the only species belonging to the genus *Euthynnus* that occurs in Mediterranean Sea along the Tunisian, Libyan and Egyptian coasts. Large catches of little tunny are recorded in May, June and July. Spawning period runs from June to September (Tawil, 2002; Hajjej *et al.*, 2010; El-Haweet *et al.*, 2013).

On the other hand, canning stays to be a particularly important form of commercial food preservation, and canned tuna constitutes a source of nutritious and relatively inexpensive healthy food, which has long shelf life at ambient temperatures and suitable for worldwide distribution (Bratt, 2010). According to the color designations and Munsell value, canned tuna meat is classified into white, light, dark and blended (Lassen, 1965). The most popular types of canned tuna include white tuna obtained from Albacore (*Thunnus alalunga*) and light tuna obtained from skipjack (*Katsuwonus pelamis*) or yellowfin (*Thunnus albacares*) (Bratt, 2010).

A decade earlier, U.S. Food and Drug Administration (FDA) and U. S. Environmental Protection Agency (USEPA) warned that certain species of tuna are specially known to accumulate Hg more than any other,

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and people who consume large amounts of certain tuna may increase the risk of being affected by Hg toxicity (Morgano *et al.*, 2014).

Increasing ingestion of Hg-contaminated food surpassing the permissible limit established by World Health Organization (WHO) was associated with frequent intake of large size fish of tuna species. Canned tuna consists of large species of tuna (e.g. albacore and yellowfin tuna), contains moderate amounts of Hg, whereas, canned tuna consists of smaller species (e.g. skipjack), contains around one-third the Hg concentrations of albacore and yellowfin tuna (Bratt, 2010).

Interestingly, Hg, Pb and Cd contaminants were investigated in fresh bluefin tuna (*T. thynnus*) and canned tuna commercially available in Italy. None of the tested samples exceeded the European control limits fixed for Cd and Pb, however 8.9% of the canned tuna and 20% of fresh tuna samples surpassed permissible limit of Hg (Storelli *et al.*, 2010). In addition, tuna caught off Libya and canned in Misurata cannery was considered of low concentration of Hg, Pb and Cd trace elements, well below the permissible levels recommended by the FAO/WHO (Voegborlo *et al.*, 1999).

Trace elements content of tuna fish needs to be well documented as tuna products are widely consumed by humans. As a part of the national monitoring programme in Libya, Cd, Pb and Hg are frequently investigated in imported canned tuna by the National Centre for Food and Drug Control (FAO/WHO, 2005). However, published articles on the levels of Cd, Pb and Hg trace elements in fresh tuna (*E. alletteratus*) from offshore waters of the west Libyan coast are lacking. Also for commercially available canned tuna distributed in Tripoli, publications on this topic are rather limited. Therefore, the objective of this investigation was to estimate the levels of Cd, Pb and Hg trace elements to determine probable toxicological relations and to warrant agreement with reference values established by European legislations (European-Commission, 2006).

#### Materials and Methods

##### Collection of samples

A total of 60 random tuna samples consisted of 20 chunk meat samples (weight approximately 100 g) of fresh little tunny and 40 canned light tuna samples (20 skipjack and 20 yellowfin) of different brand origin and country of manufacture (Italy, Libya, Spain and Thailand) were collected from retail markets at Tripoli city (Libya) with a valid shelf life. The fresh little tunny samples were carefully picked with a clean stainless steel scalpel from the axial muscle anterior to the origin of the first dorsal fin (Adams, 2004). Fresh samples were immediately transferred to sterile polyethylene bags and kept at -20 °C until analyzed.

##### Analytical procedures for estimation of total Hg

Direct thermal decomposition amalgamation atomic absorption spectrometry (TDA/AAS) was implemented

for the determination of Hg using DMA-80 (Milestone Srl, Sorisole (BG) - Italy). DMA-80 is fully compliant with USEPA test method 7473 (USEPA, 2007).

Canned tuna were opened; oil and brine were drained off carefully. The chunk meat samples were homogenized thoroughly in a food blender with stainless steel cutters (Voegborlo *et al.*, 1999). Approximately 125 mg of each undigested sample homogenate was loaded directly into a Quartz sample boat, and then the sample boat was delivered into a decomposition furnace. In brief, the working principles in operation with the DMA-80 system are as follows: (1) Samples drying and thermal decomposing in an oxygen-rich stream at 750°C with the consequent sublimation of Hg; (2) Catalytic reduction of Hg; (3) Hg is selectively trapped by gold amalgamation; and (4) The amalgamation furnace is heated and Hg is quickly introduced into multiple measuring cells situated along the optical path of the spectrophotometer, then Hg is quantitatively determined by atomic absorption spectrometry using a low-pressure Hg lamp at a wavelength of 253.65 nm.

##### Analytical procedures for estimation of Cd and Pb

Approximately 1 g of homogenized samples was transferred to high pressure 100 mL TFM vessels (SK-10, Milestone Inc.) that were carefully cleaned and soaked for 24 h in 15% HNO<sub>3</sub>, then thoroughly rinsed with Milli-Q® water and dried at 80°C for about 2 h before use. The samples were subjected for microwave-assisted digestion in closed quartz vessels (ETHOS One, Milestone Inc.) with 3 mL of H<sub>2</sub>O<sub>2</sub> (≥30%, TraceSELECT®), 6 mL of HNO<sub>3</sub> (≥69.0%, TraceSELECT®) and 1 mL of HCl (≥37%, TraceSELECT®) for 30 min (Olmedo *et al.*, 2013). The digested solution was then transferred to a sterile polyethylene vials for later analysis.

Graphite furnace atomic absorption spectrometry (GFAAS) was used for determination of Cd, Pb levels using Varian 240Z Zeeman atomic absorption spectrophotometer (Varian, Inc., Walnut Creek, CA) equipped with a Varian GTA-120 graphite tube atomizer and a programmable sample dispenser (PSD-120).

Cadmium standard for AAS (1000 mg/L, TraceCERT®) was diluted in 0.2% HNO<sub>3</sub> (TraceSELECT®) to prepare a calibration curve (0, 0.1, 0.5, 1 and 2 µg/L) and samples were also diluted 1:4 in 0.2% HNO<sub>3</sub>. Aliquots of 20 µL of digested samples were introduced directly into graphite tube atomizer and a Zeeman background correction system (Varian GTA-120/240Z AA/PSD-120) with an equal volume of matrix modifier (a mixture of 3.3% Palladium, Pd and 0.03% Magnesium, Mg as nitrates in 0.2% HNO<sub>3</sub>).

Lead standard for AAS (1000 mg/L, TraceCERT®) was diluted in 0.2% HNO<sub>3</sub> (TraceSELECT®) to prepare a calibration curve (0, 0.1, 0.5, 1 and 2 µg/L) and samples were also diluted 14 in 0.2% HNO<sub>3</sub>. Aliquots of 20 µL of digested samples were introduced directly into Varian

GTA-120/240Z AA/PSD-120 with an equal volume of matrix modifier (10 g/L of  $\text{NH}_4\text{H}_2\text{PO}_4$  dissolved in 0.2%  $\text{HNO}_3$  and 0.1% Triton X-100). A hollow cathode was operated at 228.8 nm with a current of 4 mA and a slit width of 0.5 nm for Cd and at 283.3 nm, 10 mA and 0.5 nm for Pb.

#### Quality control

Quality control for each analytical group contained a sample spiked with 2 mg  $\text{kg}^{-1}$  of each trace element. The quality control specimens were prepared according to the procedure used in the digestion of tuna samples for the Cd and Pb analysis. Recovery rates were  $88 \pm 15\%$ ,  $127 \pm 33\%$  and  $101 \pm 2\%$  for Cd, Pb and Hg, respectively.

#### Statistical analysis

One-way Analysis of Variance (ANOVA) was implemented using SPSS version 16 software to test the significance of contaminating trace elements level variations among the tuna samples. The statistical significance level was set at  $p < 0.05$  for all tests. Tukey's procedure was used as a *post hoc* test.

### Results

#### Determination of Cd level

The average value of Cd in canned yellowfin samples ( $0.027 \pm 0.026$  mg  $\text{kg}^{-1}$  wet weight (ww)) was significantly lower than canned skipjack samples ( $0.079 \pm 0.058$  mg  $\text{kg}^{-1}$  ww) and fresh little tunny samples ( $0.072 \pm 0.027$  mg  $\text{kg}^{-1}$  ww) (Table 1). However, no significant difference was found between mean values of Cd in canned skipjack and fresh little tunny samples (Fig. 1).

The maximum range of Cd in the examined fresh little tunny and canned skipjack samples was 0.11 and 0.19 mg  $\text{kg}^{-1}$  ww, respectively (Table 1). Among tested samples, only 5% of fresh little tunny and 30% of canned skipjack specimens were barely surpassing the maximum level (0.1 mg  $\text{kg}^{-1}$  ww) adopted for Cd in tuna (European-Commission, 2006) (Fig. 2).

#### Determination of Pb level

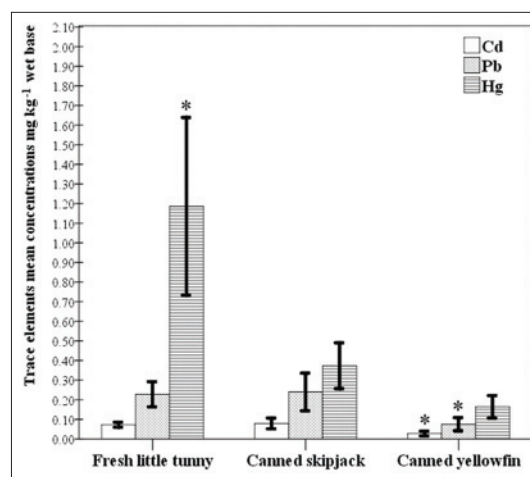
The average value of Pb concentration in canned yellowfin ( $0.075 \pm 0.071$  mg  $\text{kg}^{-1}$  ww) was significantly lower than Pb values in both fresh little tunny ( $0.228 \pm 0.136$  mg  $\text{kg}^{-1}$  ww) and canned skipjack ( $0.239 \pm 0.205$  mg  $\text{kg}^{-1}$  ww) (Table 2 and Fig. 1).

Furthermore, mean values of Pb estimated in current study were not exceeding the maximum level

(0.3 mg  $\text{kg}^{-1}$  ww) adopted for Pb in fish muscle (European-Commission, 2006), except 5 specimens out of 20 (25%) fresh little tunny samples and 7 out of 20 (35%) of canned skipjack tuna samples were exceeding the Pb maximum limit (Fig. 2).

#### Determination of Hg level

In the current study, 20 specimens each of fresh little tunny caught off Libya, canned skipjack and yellowfin tuna were analyzed for total Hg using TDA/AAS technology which is developed as a preferred direct technique for the analysis of total Hg in fish tissue because it is rapid (5 min per sample) and easy to use and requires no sample preparation (Lasrado *et al.*, 2005; Carbonell *et al.*, 2009). DMA-80 was employed to measure total Hg contamination in canned seafood. Canned tuna showed more frequent detection and higher content of total Hg than other canned fishery products (Yang *et al.*, 2015). Hg content in all tuna specimens was found at different measurable levels (Table 3), the average value of Hg in fresh little tunny ( $1.185 \pm 0.968$  mg  $\text{kg}^{-1}$  ww) was significantly higher than canned skipjack ( $0.373 \pm 0.249$  mg  $\text{kg}^{-1}$  ww) and canned yellowfin ( $0.163 \pm 0.122$  mg  $\text{kg}^{-1}$  ww) (Table 3 and Fig. 1). However, no significance difference was found between Hg mean levels in canned skipjack or yellowfin tuna (Fig.1).

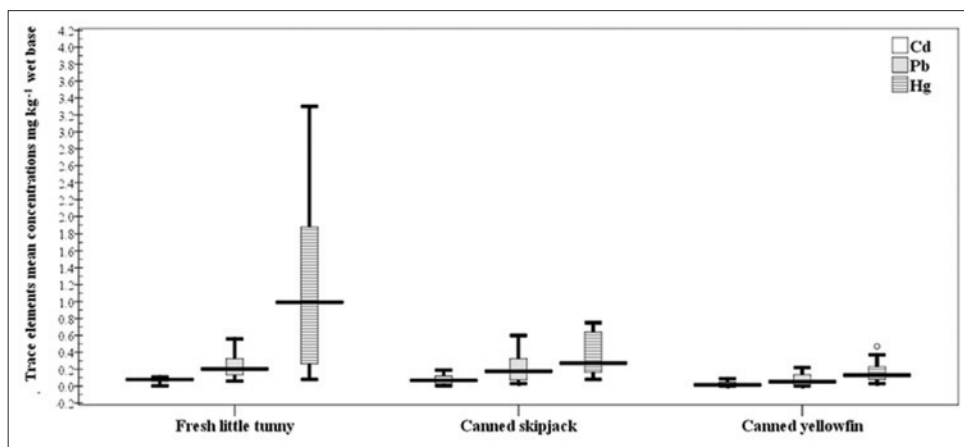


**Fig. 1.** Mean values of Cd, Pb and Hg in tested tuna samples. Error bars represent 95% confidence interval. Asterisks indicate statistical significance ( $p < 0.05$ ).

**Table 1.** Descriptive statistical analysis of estimated values of Cd contaminant level (mg  $\text{kg}^{-1}$  ww) in fresh and canned tuna samples (No.=20).

Type of tuna samples	Mean	Standard deviation	Standard error	Minimum	Maximum
Fresh little tunny	0.072	0.027	0.006	<LOQ	0.11
Canned skipjack	0.079	0.058	0.013	0.01	0.19
Canned yellowfin	0.027*	0.026	0.006	<LOQ	0.09

\*Statistical significance:  $p < 0.05$ . LOQ: Limit of Quantitation.



**Fig. 2.** Graphical representation of box blots show data sets distribution of Cd, Pb and Hg levels in tested tuna samples. Whiskers represent lower and upper quartiles; Cross band inside the box indicates median, the boxes represent the interquartile range, which covers the central 50% of the data. Open circle indicates outlier data. 95% confidence interval.

**Table 2.** Descriptive statistical analysis of estimated values of Pb contaminant level ( $\text{mg kg}^{-1}$  ww) in fresh and canned tuna samples (No.=20).

Type of tuna samples	Mean	Standard deviation	Standard error	Minimum	Maximum
Fresh little tunny	0.228	0.136	0.03	0.06	0.56
Canned skipjack	0.239	0.205	0.045	0.03	0.60
Canned yellowfin	0.075*	0.071	0.016	<LOQ	0.22

\*Statistical significance:  $p < 0.05$ . LOQ: Limit of Quantitation.

**Table 3.** Descriptive statistical analysis of estimated values of total Hg contaminant level ( $\text{mg kg}^{-1}$  ww) in fresh and canned tuna samples (No.=20).

Type of tuna samples	Mean	Standard deviation	Standard error	Minimum	Maximum
Fresh little tunny	1.185*	0.968	0.216	0.08	3.30
Canned skipjack	0.373	0.249	0.055	0.08	0.75
Canned yellowfin	0.163	0.122	0.027	0.03	0.47

\*Statistical significance:  $p < 0.05$ .

The mean value of Hg in fresh little tunny barely surpassed the maximum level ( $1 \text{ mg kg}^{-1}$  ww) adopted for Hg in tuna (European-Commission, 2006). Nevertheless, Hg levels surpassing the maximum limit were detected in 9 out of 20 (45%) of the analyzed samples, in addition box blot analysis of individual values in fresh little tunny samples revealed disparities in Hg content (Fig. 2).

### Discussion

According to Eisler (2010), whole finfishes in general contain Cd  $0.1 - 0.3 \text{ mg kg}^{-1}$  ww, whereas fish muscle usually contains Cd less than  $0.1 \text{ mg kg}^{-1}$  ww and fish liver Cd concentration is greater and reaches up to  $24.7 \text{ mg kg}^{-1}$  ww. High Cd concentration in the water sediments is reflected in Cd content of prey (Eisler, 2010). The trophic position of yellowfin ( $4.60 \pm 0.67$ ), is considered more elevated than in skipjack ( $3.94 \pm 1.06$ ) (Ruelas-Inzunza *et al.*, 2014).

Biomagnification of Cd in muscle of predator fish such as Eastern pacific yellowfin (average concentration  $0.18 \pm 0.15 \text{ mg kg}^{-1}$  dry base) and skipjack (average concentration  $0.23 \pm 0.22 \text{ mg kg}^{-1}$  dry base) is significantly depending on the function of the trophic position (Ruelas-Inzunza *et al.*, 2014).

Muscle tissue of *E. alletteratus* and skipjack obtained from fish landing sites in Tema, Ghana, West Africa, contained less than  $0.1 \text{ mg kg}^{-1}$  ww of Cd (Asmah and Biney, 2014). Meanwhile, North Atlantic *E. alletteratus* contained Cd  $0.2 \text{ mg kg}^{-1}$  on dry base (which corresponds to an estimated  $0.25 \text{ mg kg}^{-1}$  ww) (Windom *et al.*, 1973).

In Western Indian Ocean, the means of Cd content in yellowfin and skipjack tuna caught off Mozambique Channel and Reunion Island were  $0.06 \pm 0.05$  and  $0.18 \pm 0.11 \text{ mg kg}^{-1}$  ww, respectively (Kojadinovic *et al.*, 2007). On the other hand, mean Cd concentration

was magnified up to  $0.61 \pm 0.37 \text{ mg kg}^{-1}$  on dry base of skipjack (which corresponds to an estimated  $0.75 \text{ mg kg}^{-1} \text{ ww}$ ) caught off Reunion Island (Kojadinovic *et al.*, 2007). However, skipjack caught off Eastern Pacific Ocean and in front of the Baja California Peninsula had much less Cd ( $0.055 \text{ mg kg}^{-1} \text{ ww}$ ) (Ruelas-Inzunza *et al.*, 2012).

In yellowfin caught off New South Wales waters, Australia, the average Cd concentration in muscle was  $0.04 \text{ mg kg}^{-1} \text{ ww}$  (Bebbington *et al.*, 1977). However in yellowfin caught off Taiwan, Cd concentration is much lower ( $0.02 \text{ mg kg}^{-1} \text{ ww}$ ) (Han *et al.*, 1998). Much less concentrations of Cd ( $0.004 - 0.006 \text{ mg kg}^{-1} \text{ ww}$ ) were reported in yellowfin caught off Eastern Pacific and Atlantic Oceans (Besada *et al.*, 2006; Ruelas-Inzunza *et al.*, 2012).

To safeguard human consumers of tuna fish, the European Union recommends less than  $0.1 \text{ mg kg}^{-1} \text{ ww}$ ; the same level of protection is recommended in Turkey, and China (European-Commission, 2006; Eisler, 2010). Increased anthropogenic Pb concentrations in marine fish are reported in proximity to mining activities, metal finishing facilities, organic industries, areas where Pb arsenate pesticides are implemented and areas of Pb aerosol fallout (Eisler, 2010). The ability of tuna fish for bioaccumulation of Pb from seawater was reported elsewhere; concentration factors for whole tunas were about 100, being highest in liver and lowest in muscle (Eisler, 2010).

Apparently, yellowfin had a low tendency for Pb bioaccumulation, as the average muscular concentration of Pb in yellowfin caught off several waters worldwide (Atlantic Ocean, Western Indian Ocean, Eastern Pacific Ocean and Taiwan) ranged from less than 0.01 to  $0.06 \text{ mg kg}^{-1} \text{ ww}$  (Han *et al.*, 1998; Besada *et al.*, 2006; Kojadinovic *et al.*, 2007; Ruelas-Inzunza *et al.*, 2012, 2014).

The estimated mean values of Pb concentration in fresh little tunny and canned skipjack samples of current study were closely similar to Pb concentrations in *E. alletteratus* ( $0.20 \pm 0.03 \text{ mg kg}^{-1} \text{ ww}$ ) and skipjack ( $0.32 \pm 0.1 \text{ mg kg}^{-1} \text{ ww}$ ) obtained from fish landing sites in Tema, Ghana, West Africa (Asmah and Biney, 2014). Much lower Pb values were estimated in skipjack caught off Eastern Pacific and Western Indian Oceans ( $0.03 \text{ mg kg}^{-1} \text{ ww}$  and  $0.07 \pm 0.08 \text{ mg kg}^{-1}$  dry base, respectively) (Ruelas-Inzunza *et al.*, 2012). On the other hand, Pb content reached  $0.55 - 0.59 \text{ mg kg}^{-1}$  on dry base of skipjack (which corresponds to an estimated  $0.7 - 0.75 \text{ mg kg}^{-1} \text{ ww}$ ) specimens sold in Ibaraki Prefecture, Japan (Bassari, 1994).

The effects of canning on Pb content of tuna muscle have been misjudged for decades. This was supposedly due to analytical errors which resulted in reduced Pb concentrations in canned tuna by factors up to 10,000 times. However, the link between Pb content

in canned tuna and adverse impacts on human health remains unproven (Eisler, 2010). Canned tuna samples consumed in Libya contained Pb with a mean value of  $0.28 \text{ mg kg}^{-1} \text{ ww}$  (Voegborlo *et al.*, 1999). Likewise, samples of canned tuna purchased locally from Saudi Arabia contained Pb  $0.03 - 0.51 \text{ mg kg}^{-1} \text{ ww}$ , with a mean value of  $0.23 \text{ mg kg}^{-1} \text{ ww}$  (Ashraf *et al.*, 2006).

To safeguard human consumers of fish muscle in Turkey, the maximum allowable concentration of Pb is  $1.0 \text{ mg kg}^{-1} \text{ ww}$  and for China, it is  $0.5 \text{ mg kg}^{-1} \text{ ww}$  (Eisler, 2010). Regular monitoring the Hg contamination in fish and fishery products is essential to protect vulnerable population such as children (Yang *et al.*, 2015). The Hg body burden in fishes caught off Atlantic Ocean is higher than conspecific species of equal size caught off Mediterranean Sea. It is possible that the higher Hg body burdens were due to the greater natural geochemical Hg levels in the Mediterranean Sea (Eisler, 2010). Data on Hg accumulations in several types of tuna fish are particularly abundant; however there is lack of data reports on Hg levels in little tunny (*E. alletteratus*), especially in Libya.

The variation in Hg concentration in tuna represents a critical point during risk analysis for consumers (Storelli *et al.*, 2010). Elevated Hg levels can be harmful, especially if excess quantities of tuna are consumed by pregnant women and young children, as bioaccumulation of Hg can damage the developing brain of a fetus or a child (Bratt, 2010).

Geographical areas of concern where Hg concentrations in marine fish muscle exceed current regulations for human consumers include Italy, Spain, Taiwan, Florida, and Oregon (Eisler, 2010). *E. alletteratus* is one of the pelagic fish in several tropical and subtropical waters, including the Mediterranean Sea, Gulf of Guinea, Black Sea, Caribbean Sea and Gulf of Mexico. In North Atlantic Ocean, *E. alletteratus* caught off Sargasso Sea contained Hg  $1.4 \text{ mg kg}^{-1}$  on dry base (which corresponds to an estimated  $1.75 \text{ mg kg}^{-1} \text{ ww}$ ) (Windom *et al.*, 1973). Furthermore, total Hg in its muscle varied from 0.11 to 3.4 with a mean value of  $0.94 \pm 0.60 \text{ mg kg}^{-1} \text{ ww}$  in Florida (Adams, 2004), and from 2.1 to 2.3 with a mean value of  $2.2 \pm 0.14 \text{ mg kg}^{-1} \text{ ww}$  in Bermuda (Dewailly *et al.*, 2008). In Gulf of Mexico, *E. alletteratus* contained total Hg from 0.24 to 2.52 with an average value of  $1.08 \pm 0.72 \text{ mg kg}^{-1} \text{ ww}$  (Cai *et al.*, 2007).

The concentration of Hg in skipjack tuna caught off Eastern Pacific Ocean and in front of the Baja California Peninsula was  $0.104 \text{ mg kg}^{-1} \text{ ww}$  (Ruelas-Inzunza *et al.*, 2012). In addition canned tuna commercially available in California, USA and Genova, Italy contained  $0.16 - 0.27 \text{ mg kg}^{-1} \text{ ww}$  (Cugurra and Maura, 1976; Cappon and Smith, 1982).

Recently, low traces of Hg were recorded in yellowfin obtained from Eastern Pacific water, Brazil, Gulf

of California and Gulf of Guinea with a range 0.03 - 0.09 mg kg<sup>-1</sup> ww (Voegborlo *et al.*, 2006; García-Hernández *et al.*, 2007; Ruelas-Inzunza *et al.*, 2012; Schmidt *et al.*, 2015). Furthermore, the Hg range increased up to 0.3 mg kg<sup>-1</sup> ww in yellowfin caught off Atlantic and Pacific Oceans, Equatorial zone and Hawaii coast (Kraepiel *et al.*, 2003; Adams, 2004; Besada *et al.*, 2006; Kaneko and Ralston, 2007; Ordiano-Flores *et al.*, 2011). On the other hand, unusual high Hg level of 2.93 mg kg<sup>-1</sup> Hg ww was reported in yellowfin in Taiwan (Han *et al.*, 1998).

Frozen fillet of yellowfin tuna collected from different local markets in Brazil contained Hg 0.058 ± 0.001 mg kg<sup>-1</sup> ww. After different culinary treatments, no significant Hg loss was observed for uncooked and boiled fish. However, 4 to 25% Hg loss was observed in yellowfin tuna after roasting (175 °C for 20 min) or frying (180 °C for 5 min) (Schmidt *et al.*, 2015).

Canned tuna fish from the Mediterranean coast of Libya had Hg levels well below the permissible limits at range from 0.20 to 0.66 with an average value of 0.29 mg kg<sup>-1</sup> ww. (Voegborlo *et al.*, 1999). However, there is significant effect of canning on total Hg content in trawl-caught albacore tuna (*T. alalunga*); as the Hg concentration is significantly increased from 0.17 mg kg<sup>-1</sup> ww (range 0.09 - 0.24 mg kg<sup>-1</sup>) in the pre-canned samples to 0.21 mg kg<sup>-1</sup> ww (range 0.10 - 0.33 mg kg<sup>-1</sup> ww) in the post-canned samples (Rasmussen and Morrissey, 2007).

Current study revealed that Hg had the highest concentrations among tested trace elements, followed by Pb and Cd either in fresh little tunny or canned skipjack and yellowfin tuna. This order of measured trace elements concentrations is in agreement with occurrence of such toxic metals in Italian fresh and canned tuna (Storelli *et al.*, 2010).

The maximum level for tuna products (*Thunnus* species, *Euthynnus* species, skipjack) is set at 1 mg kg<sup>-1</sup> ww for Hg by the Regulation (EC) No 1881/2006 (European-Commission, 2006). Tuna intake with Hg levels more than 1 mg kg<sup>-1</sup> ww, exposes the consumer to Hg toxic effect (Storelli *et al.*, 2010). Unfortunately, about half the number of the tested fresh little tunny samples exceeded this limit. Therefore, a continuous monitoring of Hg concentration in little tunny products is essential for food safety. On the other hand, Cd and Pb concentrations either in fresh little tunny caught off Libya or canned tuna sold in Tripoli city did not surpass the toxicological standard levels (European-Commission, 2006), and accordingly there was minor food safety concerns of Cd or Pb toxicity from eating tuna therein.

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#### Conflict of interest

The authors hereby declare that there is no conflict of interest related to the present study.

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