



Occurrence of toxic metals (Hg, Cd and Pb) in fresh and canned tuna: Public health implications

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ABSTRACT

Hg, Pb and Cd levels in fresh and canned tuna were determined and assessed by comparing element levels in these samples with maximum permissible limits set by European legislation. The estimated weekly intakes by human consuming both fresh and canned tuna were also evaluated for possible consumer health risks. Among tested metals, Hg had the highest concentrations, followed by Pb and Cd either in fresh tuna or canned tuna. None of the tested samples surpassed the European regulatory limits fixed for Cd and Pb, whereas 8.9% of the tuna cans and 20% of fresh tuna samples exceeded standard for Hg. The size of tuna was a determining factor of Hg burden. A high intake of Hg surpassing the toxicological reference value established by WHO, was associated with consumption of larger size tuna specimens. Also canned tuna consumption with Hg concentrations higher than $1 \mu\text{g kg}^{-1}$, strongly increased the consumer exposure. In contrast, Cd and Pb weekly intakes through consumption either of fresh tuna or canned tuna did not exceed the toxicological reference values established by WHO, and consequently there was no human health risk. A continuous surveillance system of Hg content in these fishery products is crucial for consumer protection.

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1. Introduction

An adequate human diet should satisfy the requirements for energy and nutritive components including essential polyunsaturated fatty acids, essential aminoacids, mineral components, vitamins and fat. Fish contains all these substances, but the most important feature of this food is an advantageous fatty acid profile, resulting from the consistent content of essential polyunsaturated fatty acids, such as eicosapentaenoic and docosahexaenoic acid, known to support good health (Usydus et al., 2009). It is estimated that the consumption of one portion of fatty fish, daily, delivers about 900 mg/day of *n*-3 acids, with consequent reduction of mortality in patients with coronary diseases (Kris-Etherton et al., 2002). However, the indisputable benefits deriving by fish consumption may be offset by the presence in their meat of toxic metals, such as Cd, Pb and especially Hg, which is present in many fish species often at levels exceeding the safety standards established by legislation of the various countries. In relation to this, it is important to remark that with the exception of occupational exposure, the main Hg exposure for human is the ingestion of contaminated fish, especially larger predators. Amongst those species recognised as potentially accu-

mulating elevated metal levels, tuna is one of the most frequently consumed and commercially available groups of fish worldwide (Burger et al., 2005). These pelagic organisms are high performance fish with very high metabolism rates and, thus, high food intake rates, a property that accentuates the exposure to trace elements (Kojadinovic et al., 2007). Consequently, adverse human health effects may occur if this fish is consumed too often or in large enough quantities. In this context it is also of interest to consider canned tuna, which amongst canned fishery products is doubtless the most largely and frequently consumed. Canned tuna is, in fact, well eaten in the developed world, especially in Europe because it is convenient and affordable for most working families. Europe is the world's largest market for canned tuna with 34% of the global consumption and Italy is, after Spain, the second country in canned tuna consumption (Food Market Exchange, 2003; Glitnir Seafood Team, 2007). Nevertheless, publications on the concentrations of toxic elements in canned tuna and dietary intakes of these elements via these fishery products in Italy are lacking. Also for fresh tuna, information on these topics is rather limited, although the few literature data confirms ability of these large pelagic predators to accumulate substantial levels of toxic metals, especially mercury (Storelli et al., 2002; Licata et al., 2005). In the light of what above reported the objectives of present investigation are the following:

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- 1) to determine the concentration levels of Hg, Pb, Cd either in the edible portion of tuna (*thunnus Thynnus*) from Mediterranean Sea (Tyrrhenian Sea), or in canned tuna (*thunnus albacares*) samples purchased from supermarkets located in different Italian cities
- 2) to ascertain whether the metal concentrations are below the maximum levels established by the European Legislation (Official Journal of the European Union, 2006; EFSA, 2009)
- 3) to estimated the weekly intake and compared it with the Provisional Tolerable Weekly Intake (PTWI) recommended by the Joint FAO/WHO Expert Committee on Food Additives (WHO, 2006; EFSA, 2009).

2. Materials and Methods

2.1. Collection of samples

In June–August 2007, 20 specimens of bluefin tuna (*T. thynnus*) (length: 105–218 cm, average 140.5 ± 27.1 cm; weight: 13–161 kg, average 56.4 ± 34.0 kg) from Tyrrhenian Sea were caught. Approximately 0.1–0.3 kg of muscle tissue was removed from the anterior portion of the carcass by a transverse dissection near the dorsal fin. The tissues were dissected with plastic materials that were washed with HNO₃ and rinsed with distilled and deionized water, in order to avoid metal contamination. Concerning canned tuna, 45 most popular brands (three lots of each brand) on the Italian market were purchased in large supermarkets and grocery stores. After opening each can, oil was drained off and the meat was manually minced and analyzed.

2.2. Chemical analysis and quality control

The extractive analytical procedure and the instrumental conditions for determine metal concentrations have been described in detail elsewhere (Storelli, 2008). Briefly for Pb and Cd, aliquots (about 1.0–2.0 g) of the homogenised samples were digested in a quartz Erlenmeyer flask with 11 ml of a mixture of HNO₃–HClO₄ (8:3) using a hotplate heated to 150 °C. Additional aliquots of nitric acid (maximum of 0.2 ml) were added until a completely colourless solution was obtained. After evaporation the residue was dissolved in 2 ml of water, and finally, the volume was made up to 25 ml with deionised water. For Hg, the sample was weighed into a conical flask and digested in 10 ml of H₂SO₄–HNO₃ (1:1). The flask was heated under reflux conditions until a completely colourless solution was obtained. After cooling, the resultant solution was diluted to a known volume (100 ml) with deionised water according to the method recommended by Official Italian Agencies (GURI, 1994). The content of trace elements was determined by atomic absorption spectroscopy (AAS) (Perkin Elmer Analyst 800). Cd and Pb was analyzed by graphite furnace technique (THGA-800 P.E.) and Hg by a hydride system (FIMS100) after reduction by SnCl₂. Reference tissue (TORT-2 Lobster Hepatopancreas) was treated and analysed in the same way as the samples. Results (Hg: 0.28 ± 0.03 ; Cd: 26.2 ± 2.4 ; Pb: 0.32 ± 0.18 $\mu\text{g g}^{-1}$ dry weight) were in good agreement with the certified values (Hg: 0.27 ± 0.06 ; Cd: 26.7 ± 0.60 ; Pb: 0.35 ± 0.13 $\mu\text{g g}^{-1}$ dry weight), and the standard deviations were low, proving good repeatability of the methods. The results for standard reference material displayed recoveries of the elements ranging from 91% to 104% ($n = 3$). The limit of detection (LOD) (Hg: 5; Cd: 0.10; Pb: 10 ng g^{-1} wet weight) is defined as the concentration corresponding to three times the standard deviation of blanks and the limits of quantification (LOQs) are the following: Hg: 13; Cd: 0.38; Pb: 40 ng g^{-1} wet weight. Samples with a concentration lower than LOQ were considered equal to 0. Two samples blanks were analysed together with each sample batch. Metal concentrations in blanks were below the detection limits in all the analyses. Blanks and calibration standard solutions were similarly analysed as the digested sample solution, and calibration curves constructed. Analyses were duplicated to check the reproducibility of the results. Recovery tests were performed for the investigated metals in selected samples by spiking analysed samples with aliquots of the metal standards and then carrying out digestion. All metal concentrations were determined on a $\mu\text{g g}^{-1}$ wet weight basis.

3. Results and discussion

3.1. Content of toxic metals in fresh tuna

Among the metals analysed, Hg (0.07–1.76, average: $0.61 \mu\text{g g}^{-1}$ wet wt) showed the highest concentration, followed by Pb (ND–0.33, average: $0.07 \mu\text{g g}^{-1}$ wet wt) and Cd (ND–0.03, average: $0.01 \mu\text{g g}^{-1}$ wet wt). Among these toxic metals only Hg was present in all tuna specimens, while Cd and Pb were detected

Table 1

Estimated weekly intake (EWI) of mercury, cadmium and lead ($\mu\text{g kg}^{-1}$ body weight) in fresh and canned tuna samples.

	EWI Hg	EWI Cd	EWI Pb
Fresh tuna	0.13–3.51 1.23 ± 0.95	ND–0.07 0.01 ± 0.02	ND–0.65 0.14 ± 0.15
Canned tuna	$0.02–1.19$ 0.27 ± 0.28	$0.01–0.10$ 0.03 ± 0.02	ND–0.10 0.04 ± 0.03

ND, not detected.

in 95% and 85% of the samples analyzed, respectively. From a more detailed analysis of results it appeared that Cd and Pb levels were rather low and did not exhibit a wide between-specimen variation, whereas the analysis of Hg content revealed strong differences in specimen contamination level, suggesting a trend in the tissue residues of this metal based on sample size ($R = 0.72$, $p < 0.001$). This finding is confirmed by literature data showing as larger, older fish have generally higher concentrations than smaller, younger fish. For example, Storelli et al. (2002) reported that size and mercury levels were highly correlated for bluefin tuna (*T. thynnus*) from the Mediterranean Sea, as well as other authors found a such correlation in yellowfin tuna (*T. albacares*) and skipjack tuna (*Katsuwonus pelamis*) from the Indian Ocean (Kojadinovic et al., 2007) and in different tuna species (*Thunnus obesus*, *T. albacares* and *Thunnus alalunga*) from the Atlantic (Besada et al., 2006) and South China Sea (Agusa et al., 2005). Many studies have also shown that mercury is bioamplified in the food chain with high-trophic-level predatory species, such tuna sharks and swordfish, having generally high mercury concentrations. For example, Licata et al. (2005) and Storelli et al. (2002) reported an average of $3.03 \mu\text{g g}^{-1}$ wet weight and $1.18 \mu\text{g g}^{-1}$ wet wt in bluefin tuna from Mediterranean Sea, respectively. As well as, Nakagawa et al. (1997) reported levels of mercury in tuna from Japan of $1.11 \mu\text{g g}^{-1}$ wet wt and Sivaperumal et al. (2007) in *Euthynnus affinis* reported concentrations equal to $2.31 \mu\text{g g}^{-1}$ wet wt. The propensity of this metal to undergo biomagnification in the food chain, has been well considered when the maximum permissible limits above which seafood would be unsuitable for human consumption were defined. It is no, in fact, coincidence that European legislation (Official Journal of the European Union, 2006), as most international regulations, have adopted an action limit of $0.5 \mu\text{g g}^{-1}$ wet wt for seafood, except for predatory fish as tuna, sharks etc. for which the allowed levels is $1 \mu\text{g g}^{-1}$ wet wt. According to this standard, Hg concentrations exceeding were observed in 20% of the samples analyzed, and obviously they were associated with larger size specimens. This large variation in Hg content constitutes a crucial point when addressing the assessment of potential health hazards for consumers. In our case, by using the mean mercury concentration (it was assumed that 100% of the total mercury was methylmercury), human body weight (60 kg) and a consumption of 120 g tuna/week (Catarci et al., 2007), the weekly intake (EWI) calculated ($1.2 \mu\text{g kg}^{-1}$ bw/week) (Table 1) was below the established PTWI of MeHg ($1.6 \mu\text{g kg}^{-1}$ bw/week) (WHO, 2006), but as the figures indicated (Fig. 1), specimen size was the key parameter which determined high exposure levels. Hg intakes surpassing PTWI were generally associated, with the consumption of the larger specimens, whereas the amount of Hg ingested remained within safe level with the consumption of smaller tunas. Concerning Cd and Pb, the concentrations recorded in this study were well below the legal limits (Cd: $0.10 \mu\text{g g}^{-1}$ wet wt, Pb: $0.30 \mu\text{g g}^{-1}$ wet wt) (Official Journal of the European Union, 2008), except in a case in which Pb concentration was close to the established threshold. However, compared to the safety standards set by (WHO, 2006; EFSA, 2009) for Cd ($2.5 \mu\text{g kg}^{-1}$ bw/week) and Pb ($25 \mu\text{g kg}^{-1}$ bw/week), the estimated intakes (Cd: $0.01 \mu\text{g kg}^{-1}$ bw/week; Pb: $0.14 \mu\text{g kg}^{-1}$

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