

Research Note

Heavy Metals in Canned Tuna from Italian Markets

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MS 12-346: Received 6 August 2012/Accepted 30 September 2012

ABSTRACT

Fish is a good source of nutrients for humans but can pose a risk to human health because of the possible presence of some xenobiotics such as heavy metals and persistent organic contaminants. Constant monitoring is needed to minimize health risks and ensure product quality and consumer safety. The aim of the present study was to use atomic absorption spectrometry to determine the concentrations of some heavy metals (Hg, Pb, and Cd) in tuna packaged in different kinds of packages (cans or glass) in various countries (Italy and elsewhere). Concentrations of Cd and Hg were within the limits set by European Commission Regulation (EC) No 1881/2006 and in many samples were below the detection limit. Pb concentrations exceeded European limits in 9.8% of the analyzed samples. These results are reassuring in terms of food safety but highlighted the need to constantly monitor the concentrations of heavy metals in fish products that could endanger consumer health.

Fish products are important components of the human diet and are a significant source of proteins, polyunsaturated fatty acids, and micronutrients. However, consumers could be exposed to various contaminants through the consumption of fish that have accumulated these contaminants while living in polluted waters. Pollution in seawater is due to both natural sources and to human activities that lead (directly or indirectly) to the release into the aquatic environment of substances that can have harmful effects on living organisms and consequently on human health.

In addition to persistent organic compounds such as polychlorinated biphenyls and dioxins, some heavy metals such as lead (Pb), cadmium (Cd), and particularly mercury (Hg) can cause problems for consumers due to their bioaccumulation and biomagnification through the food chain (15), reaching the highest levels in predatory fish species at the highest trophic levels, such as tuna and swordfish (14, 46). Within these species, accumulation of contaminants, particularly Hg, is related to the age and size of the animal (6, 14, 27).

The presence of Pb, Cd, and Hg in fish products is one of the most serious chemical risks to food safety in the fishing industry. The gradual increase in pollution of the oceans, the globalization of markets, in terms of both production of raw materials and location of processing plants, and the increasing sensitivity of consumers to food safety issues make heavy metal pollution a critical factor for development and competitiveness in the fish industry. Human activities can contaminate the environment with

metals, although contamination may also derive from natural geological sources (35, 38). The effects of Pb, Cd, and Hg toxicity in humans occur primarily in kidney, liver, and nervous system. In particular, the development of the nervous system of the fetus is affected irreversibly by methylmercury through the exposure of women during pregnancy (13, 31). The transfer of metals from the mother to the fetus is possible through the placenta, and carryover from breast milk can produce some symptoms in early childhood such as neuropsychological disorders and urinary tract defects (1, 11, 12, 17, 40, 42). Problems more frequently associated with heavy metal exposure from food include long-term effects in several tissues, in particular mutagenicity (25, 39), carcinogenicity (3, 8), teratogenicity (43), immunotoxicity (36), and endocrine disruption (34).

Because humans are exposed to these metals mainly through consumption of fish products, it is particularly important to verify and/or minimize exposure to humans by controlling the levels of contamination in fish products. The maximum limits of Pb, Cd, and Hg in fish and methods for determination of the concentrations of these metals are set by European Union (EU) legislation (20, 21). The maximum limits of Hg, Pb, and Cd in fresh tuna muscle are 1.0, 0.3, and 0.1 mg/kg wet weight (w.w.), respectively.

In recent years, many surveys have been carried out on seawater and sediments and on marine organisms (7, 10, 16, 23, 41, 47, 48, 51). Nevertheless, few studies have been conducted on processed and fresh fish products. The aim of the present study was to determine the concentrations of the heavy metals Hg, Pb, and Cd in samples of marketed canned tuna in olive oil, which is one of the most important canned foods in Italian markets (29).

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TABLE 1. Concentrations of heavy metals (Pb, Cd, and Hg) in canned tuna^a

Analyte	Mean (mg/kg w.w.)	SD (mg/kg w.w.)	Minimum (mg/kg w.w.)	Maximum (mg/kg w.w.)
Pb	0.159	0.115	<LOQ	0.513
Cd	0.012	0.016	<LOQ	0.065
Hg	0.048	0.056	<LOQ	0.205

^a Mean, standard deviation (SD), minimum, and maximum values are indicated for each metal. LOQ, limits of quantification: 4.870 µg/kg w.w. for Pb, 0.550 µg/kg w.w. for Cd, and 0.455 µg/kg w.w. for Hg.

MATERIALS AND METHODS

Collection of samples. Samples of canned tuna ($n = 41$) in olive oil (packaged in either metal cans or glass jars) representing the most popular brands were obtained from Italian market during 2010 and 2011. These products was processed either in Italy or elsewhere.

Chemical and instrumental analysis. Glassware and laboratory equipments used for the preparation of the samples were decontaminated before use with diluted ultrapure nitric acid and rinsed with ultrapure distilled water.

The jars or cans were opened, and the samples were drained for 2 min in a sieve to remove the oil and then homogenized with a laboratory mixer. Aliquots of each sample (0.50 ± 0.02 g) were digested in 5 ml of ultrapure 65% HNO₃ (Carlo Erba, Milan, Italy) and 2 ml of 30% (wt/wt) H₂O₂ in a microwave digestion system (Milestone, Bergamo, Italy). The final volume was obtained by adding ultrapure deionized water.

Metal concentrations in the digested samples were determined with an atomic absorption spectrometer (AAnalyst 600, Perkin-Elmer, Bonenseewerk, Germany) equipped with a graphite furnace and a L'vov platform for Pb and Cd. A flow injection analysis hydride system (FIAS 100, Perkin-Elmer) was used for the determination of the total Hg.

The equipment was calibrated with standard solutions (Perkin-Elmer), resulting in a calibration curve with three concentrations for Pb and Hg and four concentrations for Cd. Recovery of the metals was determined by adding known amounts to metal-free samples, which were then subjected to the same digestion procedure. The resulting solutions were analyzed for metal concentrations. Concentrations of each heavy metal were expressed as milligrams per kilogram wet weight taking into account a conversion factor (0.7 as established by Italian law (30)).

Quality assurance. Quality was monitored through analysis of procedural blanks, duplicate samples, and standard solutions. Standard solutions of analytes were prepared from certified stock solutions of Cd, Pb, and Hg with a relative matrix modifier (atomic spectroscopy standard, Perkin-Elmer). Concentrations for each set of samples were determined in the medium range of the calibration curve.

The performance of the method was assessed through participation in interlaboratory studies organized by FAPAS (Food Analysis Performance Assessment Scheme, Sand Hutton, UK). The FAPAS studies were conducted with fish tissue. The limit of detection and the limit of quantification were calculated by determining the standard deviation of 10 independent blanks spiked at 1, 2, 4, and 8 µg/kg for Cd and 25, 50, and 100 µg/kg for both Pb and Hg, with an external standardization curve.

Statistical analysis. All data are expressed as mean \pm standard deviation of at least three measurements. The STAT-GRAPHIC Centurion XV statistical package, version 15 (Stat Point, Warrenton, VA) was used to determine significant differences among means. The comparison was done with multiple range tests. Differences were considered to be significant at $P < 0.05$.

An initial analysis was carried out to compare samples packaged in metal cans or glass jars. Then, samples were separated into three groups according to the country of origin (i.e., the country where the fish were packaged, not the country where the fish were caught, which is largely unknown): Italy, Spain, and other countries. Multiple range tests were used to evaluate the differences among these groups.

RESULTS

Recovery of metals from spiked samples ranged from 85 to 120%. The limits of detection were 0.136 µg/kg w.w. for Hg, 0.165 µg/kg w.w. for Cd, and 1.461 µg/kg w.w. for Pb. The limits of quantification were 0.455 µg/kg w.w. for Hg, 0.550 µg/kg w.w. for Cd, and 4.870 µg/kg w.w. for Pb.

Table 1 displays mean concentrations of Pb, Cd, and Hg with associated standard deviations and minimum and maximum values for all analyzed samples. Samples were grouped according to the processing location (Italy, Spain, or other countries). At least one heavy metal was found in each analyzed sample; Pb was detected at the highest concentrations followed by Hg and Cd.

No significant differences ($P > 0.05$) in Pb concentrations were found between samples packaged in metal cans and those packaged in glass. The same results were found for all metals tested; therefore, the type of packaging had no significant effect on metal concentration in the tuna.

Significant differences ($P < 0.05$) in Pb concentrations were found between Italian (0.202 mg/kg w.w.) and Spanish (0.114 mg/kg w.w.) samples. Pb concentrations were highest in canned tuna processed in Italy. The mean Pb concentration found in samples processed in other countries was 0.124 mg/kg w.w.

For tuna processed in Italy, 14.6% of samples had Pb concentrations above 0.250 mg/kg w.w., and 9.8% of samples exceeded the legal limit for Pb (0.3 mg/kg w.w.). Pb concentrations were below the limit of quantification in 4.9% of the samples.

Cd concentrations were far below the limits set by EU legislation in all analyzed samples and never exceed 0.065 mg/kg w.w. No significant differences ($P > 0.05$) were found in Cd concentrations between the samples processed in different countries. However, the highest Cd concentrations, although still below the legal limit, were found in samples processed in other countries (mean, 0.017 mg/kg w.w.), compared with samples processed in Italian (mean, 0.011 mg/kg w.w.) and Spanish (mean, 0.010 mg/kg w.w.) factories. Cd concentrations were below the limit of quantification in 48.8% of the samples.

Although Hg concentrations did not exceed the EU limit, the highest Hg concentrations were detected in samples produced by factories in both Italy (mean, 0.050 mg/kg w.w.) and other countries (mean, 0.057 mg/kg w.w.); the mean Hg concentration detected in samples from Spanish factories was

lower (mean, 0.039 mg/kg w.w.). Hg concentrations were below the limit of quantification in 24.4% of the samples.

DISCUSSION

Fishery products are important components of the human diet; however, in certain circumstances they may be a source of exposure to environmental contaminants, including heavy metals and persistent organic pollutants. The network of controls and surveys at both national and international levels are valuable and irreplaceable tools for preventing human exposure to xenobiotics that could pose a risk to public health. These controls are possible because of modern analytical instrumentation, which is increasingly sophisticated and sensitive for determining very low concentrations of chemicals. The reduction of the concentrations of a few contaminants such as heavy metals observed in recent years can certainly be attributed to European legislation that has excluded Pb from fuels (18) and limited the presence of Hg in many products of common use (19, 22), thereby reducing the environmental residues and subsequent presence in food fishes.

Concentrations of heavy metals, including Pb, Cd, and Hg, have been widely explored in tuna species, which are top-level predators. Biomagnification of large amounts of contaminants occur in these species, and these fishes are a dietary source of heavy metals in humans (6, 33, 37, 45, 48, 50). However, few studies have dealt with heavy metal contamination in marketed canned tuna. A study performed in United States to assess the potential dietary Hg exposure revealed that canned tuna is one of the most consumed seafoods and might play a significant role in exposure of consumers to this toxic metal (28). However, in general canned tuna had lower levels of Hg contamination than did fresh or frozen tuna (4, 14, 32).

In the present study, at least one heavy metal was found in all analyzed samples; Pb had the highest concentrations followed by Hg and Cd. Concentrations of Hg and Cd were generally in the range reported in the literature and often far below the permissible limits for human consumption set by European regulations (20), whereas Pb concentrations in 9.8% of the analyzed samples exceeded the limit.

Pb and Cd concentrations detected in the present study are in agreement with those reported previously in the literature for canned tuna. Ashraf et al. (2) detected Pb and Cd at 0.03 to 0.51 $\mu\text{g/g}$ w.w. and 0.07 to 0.64 $\mu\text{g/g}$ w.w., respectively, for canned tuna commercially available in Saudi Arabia. Pb and Cd average concentrations were 0.28 and 0.18 $\mu\text{g/g}$ w.w., respectively, in Libyan canned tuna (49) and 0.32 and 0.02 $\mu\text{g/g}$ w.w., respectively, in canned tuna from the United States (9).

Hg concentrations in the present study are generally in agreement with those found previously. Hg concentrations in canned tuna from Canada were considerably less than those found in swordfish, marlin, shark, and fresh tuna (24). Burger and Gochfeld (5) found Hg concentrations of 0.05 to 0.24 mg/kg w.w. in canned tuna purchased in Chicago, IL. Shim et al. (44) analyzed many samples of canned tuna sold in the United States and found mean Hg concentrations of 220 ppb in tuna canned in soy oil.

However, the results of the present survey do not agree with those of Storelli et al. (45), who found the highest concentrations of Hg (0.04 to 1.79 $\mu\text{g/g}$ w.w.) followed by Pb (0.02 to 0.16 $\mu\text{g/g}$ w.w.) and Cd (0.01 to 0.14 $\mu\text{g/g}$ w.w.) in samples of canned tuna collected from Italian markets.

Gerstenberger et al. (26) analyzed the Hg present in canned tuna purchased in Las Vegas, NV, and found that 5% of the canned tuna exceeded the action level established by the U.S. Food and Drug Administration (1.0 mg/kg). Yallouz et al. (52) found that 15% of the canned tuna in Brazilian markets also exceeded the 1.0 mg/kg guideline.

In conclusion, Cd and Hg concentrations in the canned tuna samples analyzed were much below the maximum limits established by international legislation and virtually absent (below the detection limit) in many samples. However, Pb concentrations exceeded the European standard in several samples (9.8%). For some xenobiotics, such as Pb, fish can become contaminated both from marine pollution and during processing. Technological processes and/or the materials may increase the metal content in food, although modern food technologies tend to minimize this risk. The food industry trend is toward the adoption of safer technologies, such as cans with lacquered walls and electric welding rather than Pb soldering of metal containers. However, the use of lead-based solder for cans is not currently prohibited, and in developing countries this risk to human health should not be disregarded.

Another issue is the traceability of raw materials. The label on tuna cans does not report the Food and Agriculture Organization catch area and tuna species; only the country where the product is processed must be listed. In the present study, samples were divided according to geographical location of the processing plant, which is the only possible territorial discrimination in the absence of compelling data on the source of the fish. In many cases, developed companies have relocated their production facilities to developing countries, raising questions about the characterization of the origin of the raw materials and the manufacturing processes.

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