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Heavy Metals in Frozen and Canned Marine Fish of Korea

M. M. Islam[1, 2], S. Bang[1], Kyoung-Woong Kim[1], M. K. Ahmed[2*], and M. Jannat[2]

[1]International Environmental Research Center (IERC), Gwangju Institute of Science and Technology (GIST), Gwangju, South Korea

[2]Department of Fisheries, University of Dhaka, Dhaka-1000, Bangladesh

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Abstract

Heavy metal contaminants in fish are of particular interest because of the potential risk to humans who consume them. The edible muscles of eight different species of fishes were analyzed by ICP-MS (Inductively Coupled Plasma Mass Spectroscopy) for heavy metals, collected from Market in Gwangju, Korea during April-May in 2008. The concentrations of Hg, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn varied between 0.24±0.007 - 0.01±0.001, 44.54±5.69 - 1.23±0.20, 0.13±0.05 – ND (not detected), 1.32±0.47 - 0.09± 0.02, 3.13±2.53 - 0.63±0.06, 107.17±28.02 - 11.27±1.56, 12.38±1.23 - 0.25±0.02, 1.025±1.41 - 0.12±0.09, 0.74±0.28 - 0.05±0.03 and 80.30±17.09 - 22.35±6.89 mg/kg, respectively. The concentrations of arsenic and nickel exceeded the maximum allowable intake level.

Keywords: Heavy metals; Marine fish; Korea.

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1. Introduction

Metals and other elements are present in water from natural sources such as the rocks of the sea bed and as a result of human activities such as emissions from industrial processes. These elements are taken up by marine organisms and many tend to be accumulated in organisms such as predatory fish which are higher up the food chain. As a result, the concentrations of many elements including mercury, arsenic, lead, and cadmium in fish can be relatively high compared to other foods. Many of these metals (for example, cobalt, copper, manganese, molybdenum, nickel and zinc) are essential trace elements for aquatic organisms and are involved in biochemical processes such as enzyme activation. However, although essential in small amounts, many are toxic at only slightly elevated free ion concentrations [1]. Others such as cadmium, lead and mercury have no known biological roles and are detrimental to essential life processes [2].

* *Corresponding author*: kawser_du@yahoo.com

Mercury occurs naturally as a mineral and is widely distributed throughout the environment as a result of natural and human activities [3]. Entering into water bodies the inorganic mercury is trapped in sediment particles where the sulphur-reducing anaerobic bacteria bio-transforms the mercury into methylmercury [4]. Due to its bio-accumulation and bio-magnification properties through the aquatic food chain, longer lived and larger fish than smaller fish accumulates the highest levels of methylmercury and consumption of these contaminated fish can lead to severe poisoning in both humans and wildlife [5]. Mercury can also have adverse effects on the fish populations themselves [6].

Arsenic has been considered as essential trace element for the normal growth and development of animals [7]. However, naturally occurring arsenic is found as a contaminant in drinking water. The accumulation and bio-magnification of arsenic in marine flora and fauna are a phenomenon that has generated a great deal of interest in the nutrition and trade industries in recent years. The notorious association of arsenic with poisoning has led to many studies on the possible risks associated with human exposure.

With a rather dramatic change in dietary patterns because of rapid economic development during the past three decades and the globalization of the food market, more Koreans have concerns about the safety of their diets. Excessive ingestion of some heavy metals may occur in people who have especially high intakes of certain contaminated foods. It is well known that ingestion of contaminants in excessive amounts can have detrimental effects on health [8].

According to the Codex Committee for Food Additives and Contaminants, dietary intakes of heavy metals with high public concern need to be monitored on a regular basis and rapidly updated to identify recent dietary intakes of heavy metals in developing countries. Korea, as one of the developing countries, definitely needs a monitoring system to ensure a safe food supply, especially because the average Korean diet includes an appreciable amount of fish and shellfish, which typically contain high levels of heavy metals. The Korea Food and Drug Administration (KFDA) has been responsible for monitoring the heavy metal content in raw foods for the past decade. However, there has been no well-planned study to estimate the heavy metal intake of the Korean population based on the analysis of “ready-to-eat” foods/dishes from nationally representative dietary intake data of the population. On the other hand, several studies have recently reported estimated intakes of food additives and preservatives in Korea [9-11].

Thus, there is a clear necessity to continuously monitor and to employ a surveillance system to ensure that the food supply and diet of the Korean population is safe. This is especially important because globalization and free-trade agreements affect the food supply and/or dietary patterns worldwide. A study was therefore carried out to determine the concentration of heavy metals in some fresh as well as processed fish of Korea. Thus the objectives of this study were to determine the concentrations of total Hg, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn in different fresh frozen and canned marine fish of Gwangju, Korea.

2. Materials and Methods

For the determination of heavy metals, several adult fresh, frozen as well as canned fishes were collected from market in Gwangju, Korea during April-May, 2008. The frozen fish species include *Thunnus thynnus* (bluefin tuna), *Salmo salar* (salmon/ Atlantic salmon), *Gadus macrocephalus (*pacific cod), *Theragra chalcogramma* (walleye Pollock/Alaska pollock), *Scomberomorus niphonius* (Japanese Spanish mackerel), Wooluck and *Pampus argenteus* (silver pomfret), while canned fish include *T. tonggol* (longtail tuna) and *T. thynnus* (bluefin tuna). The longtail tuna can that was preserved in water, imported from the USA, while bluefin tuna can which was preserved in oil, imported from Thailand as well as Korean own product.

For the quantitative analyses of total Hg, As, Pb, Cd, Ni, Cu, Cr, Al, Zn, Co, Fe and Mn, fish samples were digested. Only edible muscles from dorsal and ventral side of frozen fishes were collected after removing scales, skin and bones. Canned oil and water were removed and only fish muscles were taken for canned tuna samples. Approximately 2 g of wet sample and 9 ml of concentrated HNO_3 were taken in the tube of microwave digestion machine (Ethos TC) and digested according to the program of EPA3052. Blank digestion was also performed to quantify possible contamination during sample preparation and analysis. After digestion each sample was transferred to 50 ml volumetric flask and filled up to the mark by deionized water. The sample was filtered by syringe filter (0.2µm) and further diluted by four times to be analyzed by ICP-MS (Agilent 7500 CE, USA). The standard solutions were prepared by diluting the required amount of the solution from the stock solution, manufactured by Agilent, Germany.

The analytical quality of the work was cheeked by analysis (n = 9) of standard reference materials (SRM 1947, Lake Michigan Fish Tissue, NIST, USA). The recovery percentages for Hg, As, Cu and Mn were 103.75±5.23, 108.02±9.54, 107.53±9.89 and 129.81±11.78, respectively. All the data are represented as mean (x) ± standard deviation (SD) in the table.

3. Results and Discussion

Total mercury (Hg) and arsenic (As) concentration (mg/kg) in the muscle of different fresh frozen and canned fish are given in Table 1. The levels of Hg (wet wt. basis) in different types of marine fish samples were below the acceptable standard level given by Korea (0.5 mg/kg) (wet wt. basis) as well as WHO (0.5-1 mg/kg) (wet wt. basis) [12]. This means these fish is safe for human consumption in terms of Hg content. The highest (0.24±0.007 mg/kg) level of Hg was found in the frozen ventral muscle of bluefin tuna, while the lowest (0.01±0.001 mg/kg) was in the same type of muscle of silver pomfret. Several factors such as pH and redox potential of the water, species, age and size of the fish are responsible for variation of Hg content. One of the important reasons for higher concentration in bluefin tuna is the carnivore nature of tuna. Since mercury biomagnifies in the aquatic food web, fish on the higher food chain (or higher trophic level) tends to have higher levels of mercury. Hence, large predatory fishes, such as king mackerel, pike, shark, swordfish, walleye, barracuda, large tuna (as opposed to the small tuna usually used for canned tuna), scabbard and marlin, as well as seals and toothed whales, contain the highest concentrations [13]. Among the canned fish, longtail tuna, which was imported from the USA contained more Hg (1.07±0.04 mg/kg) than that of bluefin tuna which was imported from Thailand as well as Korean own product. The Hg level in Thailand canned tuna (0.09±0.01 mg/kg) was higher than that of Korean tuna (0.07±0.006 mg/kg).

Table 1. Mercury (Hg) and arsenic (As) (total) concentration (x ± SD) (mg/kg) in the muscle of different fresh frozen and canned fish of Korea.

Group	Types of sample	Hg (dry wt. basis)	Hg (wet wt. basis)	As (dry wt. basis)	As (wet wt. basis)
Fresh frozen	Bluefin tuna (dorsal)	0.69 ± 0.04	0.18 ± 0.01	5.86 ± 0.18	1.56 ± 0.05
	Bluefin tuna (ventral)	1.07 ± 0.03	0.24 ± 0.007	7.83 ± 0.15	1.80 ± 0.03
	Japanese Spanish mackerel (dorsal)	0.15 ± 0.008	0.04 ± 0.002	5.81 ± 0.54	1.47 ± 0.14
	Japanese Spanish mackerel (ventral)	0.16 ± 0.01	0.04 ± 0.003	11.22 ± 0.91	2.71 ± 0.22
	Wooluck (dorsal)	0.27 ± 0.05	0.06 ± 0.01	6.08 ± 0.92	1.47 ± 0.22
	Wooluck (ventral)	0.24 ± 0.03	0.06 ± 0.008	5.87 ± 0.35	1.55 ± 0.09
	Silver pomfret (dorsal)	0.06 ± 0.009	0.01 ± 0.002	8.83 ± 0.26	1.82 ± 0.05
	Silver pomfret (ventral)	0.05 ± 0.005	0.01 ± 0.001	20.70 ±14.10	5.42 ± 3.69
	Walleye pollock (dorsal)	0.18 ± 0.009	0.03 ± 0.002	7.88 ± 0.40	1.40 ± 0.07
	Walleye pollock (ventral)	0.17 ± 0.008	0.03 ± 0.002	7.42 ± 0.01	1.44 ± 0.004
	Salmon (mixed)	0.03 ± 0.006	0.01 ± 0.002	1.23 ± 0.20	0.42 ± 0.07
	Cod (mixed)	0.53 ± 0.03	0.08 ± 0.004	44.54 ± 5.69	7.14 ± 0.91
	Walleye Pollock (mixed)	0.04 ± 0.001	0.005 ±0.0001	9.56 ± 0.81	1.39 ± 0.12
Canned	Longtail tuna (USA)	1.07 ± 0.04	0.22 ± 0.009	5.04 ± 0.20	1.03 ± 0.04
	Bluefin tuna (Thailand)	0.26 ± 0.03	0.09 ± 0.01	4.09 ± 0.22	1.41 ± 0.08
	Bluefin tuna (Korea)	0.24 ± 0.02	0.07 ± 0.006	3.46 ± 0.29	1.06 ± 0.09

The maximum (7.14±0.91 mg/kg) (wet wt. basis) level of As was found in cod which is a demersal fish, while the minimum (0.42±0.07 mg/kg) was in salmon. In general, the canned fishes contained less As than those of fresh frozen fish, however, most of the concentrations exceeded the maximum standard limit of 1.4 mg/kg (wet wt. basis) for human consumption [14]. Levels of arsenic in marine organisms can range from < 1 mg/kg up to more than 100 mg/kg [13] [15] [16]. Studies in laboratory animals have demonstrated that the toxicity of arsenic is dependent on its form and its oxidation state. It is generally recognized that the soluble inorganic arsenicals are more toxic than the organic ones, and the trivalent forms [As(III)] are more toxic than the pentavalent ones

[As(V)]. There are multiple end-points, with several different organ systems being affected, including the skin and the respiratory, cardiovascular, immune, genitourinary, reproductive, gastrointestinal and nervous systems. However, accurate determination of the chemical speciation of arsenic is an important concern in studies involving assessment of arsenic toxicity.

Total cadmium (Cd), chromium (Cr), copper (Cu) and iron (Fe) concentrations (mg/kg) in the muscle of different fresh frozen and canned fish are presented in Table 2. In most of the samples, Cd was not detectable, but the maximum (0.13 ± 0.05 mg/kg) level of Cd was found in the ventral muscle of Japanese Spanish mackerel, which is much lower than the maximum consumption limit of 0.3 mg/kg (wet wt. basis) [15]. In animals, cadmium concentrates in the internal organs like kidney and liver rather than in muscle or fat. Cadmium levels usually increase with age [17]. The acute toxicity of cadmium to aquatic organisms is variable, even between closely related species, and is related to the free ionic concentration of the metal. Cadmium interacts with the calcium metabolism, and in fish it causes abnormally low calcium levels (hypocalcaemia), probably by inhibiting calcium uptake from the water. However, high calcium concentrations in the water protect fish from cadmium uptake by competing at uptake sites. Effects of long-term exposure can include larval mortality and temporary reduction in growth [18].

Table 2. Cd, Cr, Cu and Fe total concentration (x ± SD) (mg/kg) (dry wt. basis) in the muscle of different fresh frozen and canned fish of Korea.

Group	Types of sample	Cd	Cr	Cu	Fe
Fresh frozen	Bluefin tuna (dorsal)	*ND	0.2 ± 0.11	0.97 ± 0.04	58.02 ±47.38
	Bluefin tuna (ventral)	0.06 ± 0.01	0.14 ± 0.04	1.20 ± 0.10	44.99 ±17.25
	Japanese Spanish mackerel (dorsal)	*ND	0.34 ± 0.03	1.83 ± 0.55	21.54 ± 1.89
	Japanese Spanish mackerel (ventral)	0.13 ± 0.05	0.46 ± 0.27	1.64 ± 0.21	17.03 ± 2.58
	Wooluck (dorsal)	*ND	0.29 ± 0.22	0.69 ± 0.20	23.09 ±21.10
	Wooluck (ventral)	*ND	1.03 ± 0.37	1.41 ± 0.11	35.44 ±31.96
	Silver pomfret (dorsal)	*ND	0.44 ± 0.28	0.67 ± 0.08	65.87 ±80.39
	Silver pomfret (ventral)	*ND	0.17 ± 0.04	0.63 ± 0.06	28.36 ±23.56
	Walleye pollock (dorsal)	*ND	0.24 ± 0.01	1.56 ± 0.05	11.27 ±1.56
	Walleye pollock (ventral)	0.04 ± 0.01	0.31 ± 0.06	2.81 ± 0.66	16.63 ±5.53
	Salmon (mixed)	*ND	0.10 ± 0.03	1.02 ± 0.09	14.82 ±7.67
	Cod (mixed)	*ND	0.21 ± 0.04	1.08 ± 0.05	23.92 ±13.59
	Walleye pollock (mixed)	*ND	0.36 ± 0.06	2.77 ± 0.99	22.34 ±11.60
Canned	Longtail tuna (USA)	*ND	1.32 ± 0.47	0.93 ± 0.11	107.17±28.02
	Bluefin tuna (Thailand)	0.13 ± 0.04	0.12 ± 0.04	1.86 ± 0.18	35.66 ±4.51
	Bluefin tuna (Korea)	0.053±0.005	0.09 ± 0.02	3.13 ± 2.53	31.07 ± 0.41

* ND= Not detected

Higher (1.32 ± 0.47 mg/kg) level of Cr was found in the longtail canned tuna, while lower (0.09 ± 0.02 mg/kg) level was found in bluefin canned tuna of Korea. Both of these levels are much lower than the maximum Cr consumption limit of 8 mg/kg of fish given by USEPA [3].

The highest (3.13±2.53 mg/kg) level of Cu was found in the bluefin canned tuna of Korea while the lowest (0.63±0.06 mg/kg) level was in the ventral muscle of silver pomfret. Both of these levels are much lower than the maximum Cu consumption limit of 120 mg/kg of fish given by USEPA [3]. In case of Fe maximum (107.17±28.02 mg/kg) level was found in the longtail canned tuna, while lower (11.27±1.56 mg/kg) level was in the dorsal muscle of walleye pollock.

Table 3. Manganese (Mn), Nickel (Ni), Lead (Pb) and Zinc (Zn) (total) concentration (x ± SD) (mg/kg) (dry wt. basis) in the muscle of different fresh frozen and canned fish of Korea.

Group	Types of sample	Mn	Ni	Pb	Zn
Fresh frozen	Bluefin tuna (dorsal)	0.57 ± 0.42	0.26 ± 0.12	0.13 ± 0.08	33.76 ± 7.69
	Bluefin tuna (ventral)	0.44 ± 0.19	0.28 ± 0.29	0.07 ± 0.03	28.22 ± 16.22
	Japanese Spanish mackerel (dorsal)	0.44 ± 0.009	1.025 ± 1.41	0.06 ± 0.02	66.34 ± 14.50
	Japanese Spanish mackerel (ventral)	0.60 ± 0.11	0.24 ± 0.17	0.08 ± 0.04	60.84 ± 10.38
	Wooluck (dorsal)	0.77 ± 0.23	0.82 ± 1.03	0.05 ± 0.03	33.25 ± 16.07
	Wooluck (ventral)	0.93 ± 0.39	0.61 ± 0.23	0.12 ± 0.05	49.85 ± 16.94
	Silver pomfret (dorsal)	0.88 ± 0.72	0.28 ± 0.15	0.12 ± 0.02	74.14 ± 10.79
	Silver pomfret (ventral)	0.63 ± 0.35	0.19 ± 0.07	0.15 ± 0.02	52.42 ± 9.66
	Walleye pollock (dorsal)	0.45 ± 0.03	0.59 ± 0.05	0.12 ± 0.01	80.30 ± 17.09
	Walleye pollock (ventral)	0.60 ± 0.13	0.50 ± 0.25	0.12 ± 0.07	45.57 ± 25.30
	Salmon (mixed)	0.32 ± 0.06	0.12 ± 0.09	0.16 ± 0.04	22.35 ± 6.89
	Cod (mixed)	0.997 ± 0.09	0.83 ± 0.48	0.14 ± 0.10	58.94 ± 18.28
	Walleye pollock (mixed)	12.38 ± 1.23	0.39 ± 0.12	0.14 ± 0.04	79.16 ± 9.09
Canned	Longtail tuna (USA)	0.58 ± 0.06	0.36 ± 0.03	0.74 ± 0.28	41.38 ± 3.02
	Bluefin tuna (Thailand)	0.32 ± 0.002	0.26 ± 0.29	0.13 ± 0.05	57.60 ± 10.81
	Bluefin tuna (Korea)	0.25 ± 0.02	0.12 ± 0.17	0.12 ± 0.01	38.35 ±10.13

Total Manganese (Mn), Nickel (Ni), Lead (Pb) and Zinc (Zn) concentrations (mg/kg) in the muscle of different fresh frozen and canned fish are given in Table 3. Higher

(12.38±1.23 mg/kg) level of Mn was found in walleye pollock, while lower (0.25±0.02 mg/kg) level was found in the canned bluefin tuna of Korea. The maximum (1.025±1.41 mg/kg) level of Ni was found in the dorsal muscle of Japanese Spanish mackerel, while lower (0.12±0.09 mg/kg) level was found in salmon. The maximum level is slightly higher than the standard level (1 mg/kg) given by the USEPA [3].

Higher (0.74±0.28 mg/kg) level of Pb was found in canned longtail tuna, while lower (0.05 ± 0.03 mg/kg) level was found in wooluck. Both of them are much lower than the maximum allowable limit (2.0 mg/kg; wet wt. basis) in fish reported by [13]. According to the findings of [19] the cadmium and lead concentrations varied from 0.09 to 0.48 mg/g and 0.22 to 0.85 mg/g, respectively in fish samples of the middle Black Sea (Turkey), which are more or less similar to the present findings. Concentrations in marine organisms in Norway gave annual median concentration ranges of 0.3 to 77.8 ppm dry weight in blue mussel soft body tissue, 0.0075 to 0.138 ppm dry weight in cod liver tissue and 0.02 to 1.35 ppm dry weight in other fish species liver tissue [20]. The variations in the concentrations of Cd and Pb in fish samples ranging from 0.003 to 0.036 mg/kg with a mean of 0.01367 mg/kg for Cd, and 0.001 to 0.791 mg/kg with a mean of 0.17710 mg/kg for Pb were determined by [21]. Lead is a neurotoxin that causes behavioral deficits in vertebrates [22] and can cause decreases in survival, growth rates, learning, and metabolism [23]. Young stages of fish are more susceptible to lead than adults or eggs. Typical symptoms of lead toxicity include spinal deformity and blackening of the caudal region [24].

Higher (80.30 ± 17.09 mg/kg) level of Zn was found in the dorsal muscle of walleye pollock, while lower (22.35 ± 6.89 mg/kg) level was found in salmon. According to [14] the maximum consumption limit of Zn in fish is 50 mg/kg (wet wt. basis). Considering the average moisture content of 60-80%, this standard limit would be 150-200 mg/kg, which are well above the concentration of Zn found in the present investigation.

Fifty seven samples of canned tuna fish were studied by [25] and found the concentration of Pb, Cr, Cd, Cu, Ni, and Hg ranged between 0.14-0.82, 0.10-0.57, 0.08-0.66, 0.02-0.33, 0.09-0.48 and 0.18-0.86 mg/kg, respectively, which are more or less similar (except Cu) with the present findings. The arsenic, cadmium, lead, manganese, and mercury ranged from 0.23 to 3.3, 0.0001 to 0.01, 0.04–0.12, 0.1–1.0 and 0.05–0.6 ppm, respectively, in the marine fish and shrimp samples of New Jersey, USA were determined by [26], which are also more or less similar with the current findings. They found interspecific differences in levels of metals for all metals. However, the same fish metals did not have the highest values for more than two metals. They suggested that the differences were due to geography, trophic level, size, foraging method/location, and propensity of metals to undergo bio-magnification in the food chain. They concluded that the potential of harm from other metals suggested that people not only should eat smaller quantities of fish known to accumulate mercury but also should eat a diversity of fish to avoid consuming unhealthy quantities of other heavy metals.

Contaminants in fish can pose a health risk to the fish themselves, to their predators, and to humans who consume them [26]. While sampling by purchasing fish in supermarkets makes it difficult to compare among types and to interpret levels because the geographical sources of the fish are mostly unknown. From a public health perspective, people are faced with making choices in markets about what fish to buy based on available knowledge, which usually includes identification of species or at least type, and knowing which kinds of fish have low levels of contaminants.

The data in this paper suggest that some species have relatively low levels of contaminants of concern, such as mercury, cadmium, chromium, copper, lead and zinc. However, the same fish did not have either the highest levels of all metals or the lowest levels. Thus the greatest risk from different metals accumulated in different fish. Further, the species of fish with the highest levels of a given metal sometimes exceeded the guidance or standards for that metal. This result suggests that the risk information given to the public, does not present a complete picture. The potential of harm from other metals suggests that people not only should eat smaller quantities of fish known to accumulate mercury but also should eat a diversity of fish to avoid consuming unhealthy quantities of other heavy metals. Contaminant information on this broad range of metals in commercial fish is generally not available to the public. Thus, it is suggested that there is a need for more information on contaminant levels in fishes from specific regions of the world. Then data on contaminant levels in fishes from particular regions of the world will aid in understanding the risk from the contaminants.

This study found higher level of total arsenic and nickel in fishes in comparison to the standard consumption level. In general ventral part contained more heavy metals than dorsal part of fish. No significant difference was found between heavy metal concentrations in fresh frozen and canned fish. In most fishes, most of the heavy metals did not exceed the maximum consumption limit, however, if a person consumes more fish than normal intake, it would be detrimental to health. In the case of arsenic, in order to predict accurate toxicological risk, both organic and inorganic speciation studies are necessary. The present study mainly focused on the carnivorous fishes collected from markets. Future study should consider different trophic levels, feeding habits, age and size and geographical distribution of the species concern. Risk assessment should also be investigated based on the metal concentrations, consumption rate of fish, species and size of fish, weight of person etc.

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