

METALS CONTENT IN THE MUSCLE AND HEAD OF COMMON FISH AND SHRIMP FROM RIYADH MARKET AND ASSESSMENT OF THE DAILY INTAKE

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Three highly consumed fish including Nile Tilapia (*Oreochromis niloticus*), Gray Mullet (*Mugil cephalus*) and California Sardine (*Sardinops sagax*) and black tiger shrimp (*Penaeus monodon*) in Riyadh market, Saudi Arabia were evaluated for their composition of arsenic, cadmium, chromium, nickel, lead, copper, zinc, and manganese metals. The concentration of metals was determined in the muscle and the head of fish and shrimp using atomic absorption spectrophotometry (AA). The maximum metals in the samples were found in the head of shrimp and sardine. The maximum As, Cd, Cr, Ni, Pb, Cu, Zn, and Mn concentrations in fish muscles were 0.033, 0.369, 0.007, 0.012, 0.005, 0.76, 6.28 and 0.26 µg/g respectively. Meanwhile, zinc and copper were the highest in shrimp muscle. The concentration of metals was below the maximum allowed daily intake limit by the Saudi and International Legislations for fish human consumption permissible limit. The calculated maximum daily intakes (MDI) were found to be 0.055, 0.613, 0.012, 0.020, 0.008, 1.262, 10.432 and 0.432 µg/day for As, Cd, Cr, Ni, Pb, Cu, Zn, and Mn in fish muscle respectively. While the MDIs' of those metals in shrimp muscle were 0.382, 0.728, 0.050, 0.033, 0.013, 3.588, 22.525 and 0.432 µg/day.

Key words: metals; Nile Tilapia; Gray Mullet; California Sardine; black tiger shrimp; daily intake; Saudi market

INTRODUCTION

Metals are natural components of the earth's crust, which are present in air, soils, and waters (Biegalski and Landsberger, 1999; Cook *et al.*, 2005; Massadeh *et al.*, 2007). However, certain elements can be also environmental pollutants, particularly in areas with high anthropogenic pressure (Zhang and Wong, 2007; Amiard *et al.*, 2008). Many chemical elements that are present in seafood are essential for human life at low concentrations, but can be toxic at high concentrations. Metals such as cobalt (Co), chromium (Cr), iron (Fe), manganese (Mn), or zinc (Zn), among others, are essential for humans, elements such as arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) have no beneficial effects, and there is no known homeostasis mechanism for them in humans (Chang, 1996). Although the adverse health effects of a specific metal depend upon the concentration in the media, chronic exposure to elements such as As, Cd, Hg, and Pb can cause toxic effects at relatively low levels (Chang, 1996). Metals can enter the human body mainly through inhalation and ingestion, with the diet being the main route of human exposure for non-occupationally exposed individuals (Bocio *et al.*, 2005; Leblanc *et al.*, 2005). It must be taken into account that, although metals can change their chemical form, they cannot be degraded or destroyed. Therefore, the assessment of the health risks of metals via dietary intake is an issue of special

interest (Ferre-Huguet *et al.*, 2008; Zukowska and Biziuk, 2008).

Therefore many consumers regard any presence of these elements in fish as a hazard to health. Trace metals are generally released in aquatic environments in different ways and accumulation of these metals is dependent on the concentration of the metal and the exposure period. Levels of heavy metals in fish have been widely reported (Romeo *et al.*, 1999; Edwards *et al.*, 2001; Gaspic *et al.*, 2002; Satarug *et al.*, 2003; Küçüksezgin *et al.*, 2006).

To the well-known toxic effects of As, Cd, Hg, and Pb in humans, it must also be added that the European Commission (EC) has recommended some restrictions concerning the consumption of marine species (especially predatory species, which accumulate over a long lifetime (CEC, 2006). This is particularly important for children, pregnant women, and breastfeeding mothers.

Fishes are notorious for their ability to concentrate heavy metals in their muscles and since they play important role in human nutrition, they need to be carefully screened to ensure that unnecessary high level of some toxic trace metals are not being transfer to man through fish consumption (Mertz, 1981; Adeniyi *et al.*, 2008; Olowu *et al.*, 2010).

Fish has long been a favorite meal of people living around the Arabian Gulf, even before the discovery of oils and natural gas (Al-Jedah and Robinson, 2001). During the past 20 years, there has been renewed interest in dietary

components such as fish, which are rich sources of omega-3 fatty acids, and might favorably improve lipid profiles and reduce risk of coronary heart disease (Stone, 1996, Tawfik, 2008). Additional reported benefits of fish consumption also include their hypolipidemic and/or antiatherogenic effects (Harris, 1989), decreased risk of prostate cancer (Terry *et al.*, 2001), reduced occurrence of renal cell carcinoma in women (Wolk *et al.*, 2006), and reduced risk of dementia and alzheimer disease in certain conditions (Huang *et al.*, 2005).

Heavy metals are the most hazardous pollutants due to the speed of their dissemination in biosphere and their accumulative concentration. They permeate the environment by various means, penetrate the circle of metabolism, become toxic and disturb physiological functions of organism. While regulating constituents of food products, the World Health Organization (WHO) as well as the Food and Agriculture Organization (FAO) suggest monitoring the concentrations of heavy metals (HMs) (Staniskiene *et al.*, 2009).

Both the World Health Organization and the Food and Agriculture Organization of the United Nations state that monitoring of eight elements in fish Hg, Cd, Pb, As, Cu, Zn, Fe, Sn is obligatory and monitoring of the others (Sb, Ni, Cr, Al, F, J) is suggested (European Commission, 2000).

Several organizations such as Food and Agriculture Organisation (FAO) and the World Health Organisation (WHO) provide guidelines on the intake of trace elements by humans. The provisional tolerable weekly intake (PTWI) recommended by the Joint FAO/WHO Expert. Heavy metal concentrations in organic samples such as fish products are mostly assayed using atomic absorption spectrometry (Sures *et al.*, 1995; Tüzen, 2003; Henry *et al.*, 2004; Mendil *et al.*, 2005; Türkmen *et al.*, 2005).

The United Nations World Health Organization (WHO) has also given recommendations for maximum minerals content in food and water (WHO, 1995, CAC/FAO, 1999). The Joint FAO/WHO Expert Committee on Food Additives (JECFA, 2000) has established a provisional tolerable weekly intake (PTWI) for minerals. PTWI is an estimate of the amount of a contaminant that can be ingested over a lifetime without appreciable risk. An intake above the PTWI does not automatically mean that health is at risk. Transient excursion above the PTWI would have no health consequences provided that the average intake over long period is not exceeded as the emphasis of PTWI is a lifetime exposure (Al-Othman, 2010).

The Arabian Gulf has been subject to inputs of trace metals from a variety of sources, and it has been estimated that oil pollution in the Gulf represents 4.7% of the total oil pollution in the world (National Research Council, 1985). This figure has increased even more after the Gulf war where, approximate 11 million oil barrels were discharged into the Gulf (Price and Sheppard, 1991). Bu-Olayan and

Subrahmanyam (1997) investigated the contribution of the 1991 oil spill to heavy metal contamination in marine environment of the Gulf in Kuwait. Significant increase in copper, zinc and nickel concentrations was found in the 1994 snail and oyster samples compare to samples collected in 1990. Refineries and petrochemical industry wastes contribute significantly to metal pollution of the Arabian Gulf marine environment (Sadiq and Zaidi, 1985) where lead, cadmium and nickel were found in the sediments from the coastal region of Saudi Arabia.

Therefore, the main purpose of the present study were to determine the concentration of As, Cd, Cr, Ni, Pb, Cu, Zn, and Mn in most consumable types of fish (Nile Tilapia, Gray Mullet, and California Sardine) and Shrimp in the muscle and head and to determine their potential detrimental effects via calculation of the daily intake for normal daily consumption of these fishes and shrimp, for adult male and female gender.

MATERIALS AND METHODS

Sample preparation: Local, common and scientific names of fishes and shrimp used through this study are presented in Table 1. Fresh fish and shrimps were purchased from the central market in Riyadh city, the capital of Saudi Arabia. They were kept in cold iced boxes and transported to the laboratory within 2h. On arrival at the laboratory, fresh fish were washed with tap water several times to remove adhering blood and slime; whole fish were gutted and removed the internal organs. Raw samples (muscle and head) were homogenized in a stainless-steel meat mincer and blender and 50 g test portions were stored at -20°C and each group was analyzed in the same way.

Table 1. Local, common and scientific names of fish species included in the study

Local name	Common name	Scientific name
Boulty	Nile Tilapia	<i>Oreochromis niloticus</i>
Boury	Gray Mullet	<i>Mugil cephalus</i>
Sardine	California Sardine	<i>Sardinops sagax</i>
Shrimp	Black tiger shrimp	<i>Penaeus monodon</i>

Reagents: De-ionized water was used to prepare all aqueous solutions. All mineral acids and oxidants used were of the highest quality (Merck, Germany). All the plastic and glassware was cleaned by soaking overnight in a 10% (w/v) nitric acid solution and then rinsed with deionized water.

Apparatus: Unicomp Analytical System Model 919, Cambridge, UK atomic absorption spectrophotometer was used in this study.

Chemical analyses: Duplicate of ten grams of homogenate tissue (muscle and head) was weighted into a 150 ml air-tight quick flask with glass stopper. Five ml of conc. HNO₃ and 3 ml of 60% perchloric acid were added to each sample

and digested in a temperature controlled waterbath at 85°C. After digestion, the sample volumes made up to 100 ml with 0.5% HNO₃ and were separately filtered using an ashless filter paper, then used for the determination of heavy metals (Eboh *et al.*, 2006).

Assessment of daily intake: To assess public health impact of metals in fish, it is essential to calculate the daily intake of metals by humans expressed as µg/day. This can be obtained by multiplying the average quantity of fish consumed per Saudi family per day (9.967g) by the concentration of metal in studied fish (Al-Nozha *et al.*, 1991) and divided by 6 (the average Saudi family members).

Statistical analysis: Analysis of variance was used to evaluate the analysis data, and significant differences among means were determined by one-way analysis of variance (ANOVA) and Duncan's multiple range test ($p \leq 0.05$) (SPSS 10.0 for windows).

RESULTS AND DISCUSSION

Metals content in fish: The average concentrations of arsenic, cadmium, chromium, nickel, lead, copper, zinc, and manganese in each fish species in the muscle and in the head are illustrated in Table 2. In general, the metal content was higher in Sardine compared to Boury and Boulty. Regarding to the distribution of investigated metals inside fish, the results indicated that the concentrations of arsenic, cadmium, chromium, nickel, lead, copper, zinc, and manganese in the three fish species were higher in heads than in muscles. The highest metal concentrations were found to be Zn and Cu, whereas the lowest were Cr and Pb.

Zn level was minimum in the muscle of Boulty (*Oreochromis niloticus*) while the maximum zinc level found in the head of sardine. In the study of Mendil and Uluozlu (2007) zinc values varied from 13.9 to 48.6 µg/g in *C. tinca* and *C. carpio* respectively. The range of Zn levels recorded in fish in this study was low when compared to that levels recorded in some other fishes of the river. Frozen anchovy showed the highest zinc content (566 µg/g) and in canned anchovy fillet the zinc content was 33.8 µg/g (Mendil *et al.*, 2005). Tüzen (2003) reported the zinc value

of anchovy as 17.4 µg/g (dry weight).

Ni concentration range was found to be between 0.009-0.012 µg/g and 0.019- 0.060 µg/g in the muscle and head of fish. Such traces could be the result of oil spills and/or transportation in the region, as oil contains small amounts of nickel and vanadium (Sadiq and Zaidi, 1985). The range of Ni levels in the fish recorded in this study was low in comparison to WHO and FEPA maximum recommended limits of 0.5 - 0.6 µg/g. Further, these values are lower than the literature value of Mendil *et al.* (2005) who determine trace metal in fish species from lakes in Turkey, and Al-Saleh and Shinwari (2002) who determine elements in four fish species from the Arabian Gulf of Saudi Arabia. While it were similar to Ashraf *et al.* (1991) who determine trace metals in fish from three freshwater reservoirs on the Indus river and Rashed (2001) who monitor heavy metals in fish from Nasser lake, Egypt.

Mn levels (0.11 – 0.47 µg/g) in fish samples were low when compared to the 0.5 µg/g WHO (1985) and FEPA (2003) recommended standards except of sardine head. Significant difference was found between manganese concentration in the muscle and head of Boulty (*Oreochromis niloticus*), Boury (*Mugil cephalus*), and sardine (*Sardinops sagax*). In this study manganese value was lower than other reports (Tariq *et al.*, 1993; Mendil *et al.*, 2005; Mendil and Uluozlu, 2007). In Turkey, Mn accumulated in fish species at a level of 11.1–72.9 µg/g (Mendil *et al.*, 2005).

Cr levels were found to be (0.03 – 0.041 µg/g). These values were low when compared to WHO and FEPA limits of 0.05 - 0.15 µg/g in food fish. These values are also below to the literature value of Mendil and Uluozlu (2007), Mendil *et al.* (2005) and similar to other reported values (Ashraf *et al.*, 1991; Rashed, 2001).

Mn and Cr are recognised as essential trace elements for humans, and several of their metabolic roles have been determined. These include Mn-containing enzyme systems (Tinggi *et al.*, 1997) and Cr involvement in insulin function. However, for neither of the elements, have human requirements or levels of absorption from the diet been clearly determined (Offenbacher and Pi-Sunyer, 1988; Tinggi *et al.*, 1997).

Table 2. Mean values (µg /g) for metal concentrations in the muscle and head of the fish samples

Sample	Location	Arsenic (As)	Cadmium (Cd)	Chromium (Cr)	Nickel (Ni)	Lead (Pb)	Copper (Cu)	Zinc (Zn)	Manganese (Mn)
Boulty	Head	0.022 ^d	0.031 ^e	0.009 ^c	0.019 ^b	0.004 ^b	0.53 ^d	4.77 ^d	0.35 ^b
	Muscle	0.010 ^e	0.016 ^f	0.006 ^c	0.012 ^c	0.002 ^c	0.37 ^e	4.18 ^d	0.26 ^c
Boury	Head	0.075 ^b	0.398 ^b	0.016 ^b	0.022 ^b	0.005 ^b	0.83 ^b	6.97 ^b	0.24 ^c
	Muscle	0.014 ^e	0.168 ^d	0.007 ^c	0.010 ^c	0.002 ^c	0.76 ^c	5.02 ^c	0.11 ^d
Sardine	Head	0.181 ^a	0.488 ^a	0.041 ^a	0.060 ^a	0.016 ^a	1.02 ^a	21.48 ^a	0.47 ^a
	Muscle	0.033 ^c	0.369 ^c	0.003 ^d	0.009 ^c	0.005 ^b	0.56 ^d	6.28 ^b	0.25 ^c

Within the column values with different letters are significantly different ($P < 0.05$).

Values are shown as means of duplicates

Cd and Pb concentrations were measured in fish samples (Table 2) varied from 0.016 to 0.369 µg/g in the muscle and from 0.031 to 0.488 µg/g in the head for Cd, and 0.002 to 0.005 µg/g in the muscle and from 0.004- 0.016 µg/g in the head for Pb. The maximum lead and cadmium level were found in the muscle and head of sardine (*Sardinops sagax*). Heavy metal concentrations found in this study were in comparable with previous reports. Eboh *et al.* (2006) reported lower level of Pb in the muscle, gills and liver tissue of fish at a range of 0.001-0.002 mg kg⁻¹ while salmon and mackerel species were free from any heavy metals. At the same manner, the cadmium concentrations in tuna fish samples from the gulf area of Iran were lower as 0.0046-0.0720 µg/g with a mean of 0.0223 µg/g (Khansari *et al.*, 2005). Higher results of Cd (0.37-0.79 µg/g) and Pb (4.27-6.12 µg/g) reported by Canli and Atli (2005) in muscle tissues of six different fish species. Furthermore, the result of Cd and Pb are similar with those of Mendil *et al.* (2005), who reported the range 0.1- 1.2 µg/g for Cd whereas higher (0.7-2.4 µg/g) for Pb in seven fish species obtained from some lakes in Tokat, Turkey. These differences in the results of studies may be due to direct contamination of water by the metals concerned, the geochemical structure of different marine environments, differences in fish species, analytical methodology and other factors. The Pb and Cd levels found in this study do not constitute a risk factor for human health and were consistently below the Saudi acceptable limits.

In many developed countries, limits of toxic metal concentrations, Pb and Cd in fish have been set in order to safeguard public health. The fact that toxic metals are present in high concentrations in fish is of particular importance in relation to the FAO/WHO (1976) standards for Pb and Cd as toxic metals. Saudi Arabia has set a maximum limit of contaminants for Pb and Cd in fish and shellfish at 2 and 0.5 µg/g respectively (SASO, 1997). The European Commission (EC) has proposed limits for cadmium of 0.05 and 0.5 µg/g for lead in fish (Commission of the European Communities, 1997). WHO (1985) and FEPA (2003) have proposed the maximum recommended limits of 2.0 µg/g for Pb in fish. The regulations of the European Union permit a limit for cadmium of 0.1 µg/g wet weight for bonito, common two-banded seabream, eel, European anchovy, grey mullet, horse mackerel or scud, louvar or luvar, sardine, sardinops, tuna and wedge sole; and 0.05 µg/g wet weight for all others. EU regulations specifies

the maximum heavy metal limits in fish muscle as 0.2 µg/g for Pb and 0.05 µg/g for Cd, (Anonymous, 2005).

As levels were found to be (0.010-0.181 µg/g), Saudi Arabia has set a maximum limit of contaminant for arsenic in fish and shellfish of 1 µg/g (SASO, 1997). Similar results were found by Al-Saleh and Shinwari (2002).

Cu concentration in the muscle and head was found to be 0.37-0.76 and 0.53-1.02 µg/g respectively. These values are below to the literature value of Mendil and Uluozlu (2007). Cu levels recorded were lower than the 3.0 µg/g maximum recommended standards in food (WHO, 1985, FEPA, 2003). The highest copper value belonged to frozen anchovy (45.7 µg/g) and the lowest to canned anchovy fillet (7.06 mg/kg) (Mendil *et al.*, 2005). The copper content of anchovy found as 1.94 mg/kg (dry weight) by Tüzen (2003).

Out of the obtained results, sardine had the highest metal concentration in these three species of fish (Table 2). In previous study (Ashraf *et al.*, 2006) the levels of metals in canned sardine consumed in Kingdom of Saudi Arabia varied from 0.13 to 1.97 µg/g with a mean of 0.84 µg/g for Pb; from 0.01 to 0.69 µg/g with a mean of 0.18 µg/g for Cd; from 0.79 to 2.13 µg/g with a mean of 1.33 µg/g for Ni; from 8.92 to 23.90 µg/g with a mean of 16.15 µg/g for Zn; from 0.63 to 4.25 µg/g with a mean of 2.26 µg/g for Cu; from 0.02 to 0.89 µg/g with a mean of 0.31 µg/g for Cr.

Metals content in shrimp: Mean metal concentrations in shrimp (*Penaeus monodon*) sample are presented in Table 3. The levels of all metal were relatively high in the head than in the muscle. Similar levels of Mn in shrimp samples were noted (25.43 ppm) in gulf area (Heidarieh *et al.*, 2013). The most abundant elements were Zn and Cu. Pérez-Osuna and Tron-Mayer (1995) showed that muscle contains the highest load of both metals. There was a significant positive correlation between Zn and Cu in the muscle tissue. Magliette *et al.* (1995) reported that white shrimps were less susceptible to the toxic effect of Zn than *Daphnia magna* and *Ceriodaphnia dubia*. In previous study, essential metals, Cu and Zn were detected in shrimps from the Gulf and found to be at similar levels detected in the present study (Vos and Hovens, 1986). No problem is likely to occur with the consumption of shrimps reared in the Gulf area (Mason and Simkiss, 1983). For all studied metals, levels in the shrimps must be considered independent of the metal level in the sediments. The low metal levels and the absence of significant correlations between sediment and shrimp metal

Table 3. Mean values (µg /g) for metal concentrations in the muscle and head of the shrimp

Sample	Location	Arsenic (As)	Cadmium (Cd)	Chromium (Cr)	Nickel (Ni)	Lead (Pb)	Copper (Cu)	Zinc (Zn)	Manganese (Mn)
Shrimp	Head	0.427 ^a	0.553 ^a	0.042 ^a	0.274 ^a	0.008 ^a	3.17 ^a	25.0 ^a	0.47 ^a
	Muscle	0.230 ^b	0.438 ^b	0.030 ^b	0.020 ^b	0.008 ^a	2.16 ^b	13.56 ^b	0.26 ^b

Within the column values with different letters are significantly different (P < 0.05).

Values are shown as means of duplicates

concentration suggest that the degree of contamination by heavy metals in the shrimp culture area (Vos and Hovens, 1986; Mason and Simkiss, 1983).

Investigations from 1983 (Marx and Brunner, 1998) on the trace element content of North Sea shrimp revealed an average Pb load of 0.052 µg/g in raw muscle tissue, which is higher than the present value. The low overall contamination with Pb might be due to the ability of decapods to actively release both Cu and Pb into the environment. This phenomenon was described for giant tiger prawn, *Penaeus monodon*, a species that is widespread in the Indopacific Ocean (Vogt and Qunitio, 1994). The principle of this mechanism is apocrine secretion of Pb into the thoracic extensions of the antennal gland followed by excretion in the urine. On the other hand, Pb content of prawns being higher than the recommended limits has been reported by Marx and Brunner (1998). Whereas the same study detected a lower Cd content (0.015 mg/kg) for muscle tissue of North Sea shrimp. Lower Cd content was reported by Pastor *et al.* (1994).

In comparison to squid (mollusc), known as an accumulator of Cd and containing up to (mean levels) 1.72 mg Cd/kg and 0.32 mg Cd per kg wet weight in its edible parts (Marx and Brunner, 1998; Falandysz, 1989), the mean Cd content of the investigated shrimp sample was significantly higher in the head. A very short period of food consumption in connection with a dramatic growth rate seems to be the main reason for the low Cd load of this species. Sufficient food supply provided, the shrimp grow to harvest size within several weeks. Even in a highly contaminated habitat, the Cd intake via plankton, algae and sludge is not sufficient for the accumulation of relevant levels of Cd in the tissues. Shrimp *Juvenile P. setiferus* was less sensitive to Cd than other shrimps such as *Crangon septemspinosa* and *Palaemonetes vulgaris* (Carbonell *et al.*, 1998).

Public health impact of metals in fish and shrimp: The estimated maximum total dietary intakes (MDI) of arsenic, cadmium, chromium, nickel, lead, copper, zinc, and manganese in fish muscle from these study were 0.055, 0.613, 0.012, 0.020, 0.008, 1.262, 10.432 and 0.432 µg/day respectively. The MDI in shrimp muscle were 0.382 µg/day, 0.728, 0.050, 0.033, 0.013, 3.588, 22.525 and 0.432 µg/day (Table 4). These estimates are below the Joint Expert

Committee on food additives of food and agriculture organization (JECFA) provisional maximum tolerable daily intake (PTDI) for cadmium, lead and arsenic of 0.06, 0.21 and 0.12 mg/day respectively for a 60 kg adult (WHO, 1989 and 1993). In addition, it is lower than the maximum acceptable daily and weekly intake of cadmium, lead and arsenic in Saudi Arabia (SASO, 1997). As per Saudi legislation, maximum acceptable weekly intake of cadmium and lead are 0.0067-0.083 mg/kg body weight respectively. Maximum acceptable arsenic daily intake is 0.002 mg/kg body weight.

The maximum permissible doses for an adult are 3 mg Pb and 0.5 mg Cd per week, but the recommended doses are only one-fifth of those quantities (FAO/WHO, 1976). Exposure to environmental contaminants can lead to immune suppression and increased susceptibility to disease in salmonids and other fish (Miller *et al.*, 2002). The estimated maximum total dietary intakes of cadmium, lead, and arsenic from the study of Al-Saleh and Shinwari (2002) were 0.07, 0.13, and 0.12 µg/day, respectively. The World Health Organization (WHO, 1993) has set a tolerable daily intake (TDI) of 0.3 mg/day for nickel for a 60 kg person. The estimated maximum total dietary intake of nickel from Al-Saleh and Shinwari (2002) survey was 0.73 µg/day. Using FAO/WHO (Anon, 2003), values the provisional tolerable weekly intake of Cd and Pb for a 60 kg adult was estimated to be 420 and 1500 µg person⁻¹ week⁻¹ respectively. Keeping in view the toxicity of Pb and Cd, the Joint Food and Agriculture Organization/World Health Organization (FAO/WHO) expert committee on food additives have suggested a tolerable intake of 400–500 µg Cd per week for man; for Pb a weekly intake of 3 mg can be tolerated (FAO/WHO, 1972). The maximum concentration of Pb which is permitted in prepared foods for babies and children is 200 µg/kg. According to the regulations made by the Commission of European Union, the permissible limit for Cd in sea bass is 0.1mg/kg wet weight (Anonymous, 1997). The recommended daily intakes of zinc and copper are 15 mg Zn for adult males and 12 mg Zn for adult females and 1.5–3.0 mg Cu (Anonymous, 1997).

Conclusions: Most metals concentrations in the head and muscle of selected fish and shrimp was Zn, Cu, Cd, Mn and

Table 4. The daily intake of metals (µg/day) in fish and shrimp muscles by humans in Saudi family per day

Sample	Arsenic (As)	Cadmium (Cd)	Chromium (Cr)	Nickel (Ni)	Lead (Pb)	Copper (Cu)	Zinc (Zn)	Manganese (Mn)
Boulty	0.017 ^c	0.027 ^d	0.010 ^b	0.020 ^b	0.003 ^c	0.615 ^d	6.944 ^d	0.432 ^a
Boury	0.023 ^c	0.279 ^c	0.012 ^b	0.017 ^b	0.003 ^c	1.262 ^b	8.339 ^c	0.183 ^b
Sardine	0.055 ^b	0.613 ^b	0.005 ^b	0.015 ^b	0.008 ^b	0.930 ^c	10.432 ^b	0.415 ^a
Shrimp	0.382 ^a	0.728 ^a	0.050 ^a	0.033 ^a	0.013 ^a	3.588 ^a	22.525 ^a	0.432 ^a

Within the column values with different letters are significantly different (P < 0.05).

Values are shown as means of duplicates

their distribution in the head more than muscle. The maximum metals in the samples were found in the head of shrimp and sardine. Increasingly there is a need to assess the contaminant levels in fish as indicators of the health and well-being of both the fish and their consumers, including humans. The results obtained from this study showed that the daily intake of metals by humans from these fish species and shrimp had generally lower than are allowed by health authorities in the European Union (EU), World organization (WHO) and Saudi legislation.

Acknowledgement: This research project was supported by a grant from the research center of the center for female scientific and medical colleges in King Saud University.

REFERENCES

- Adeniyi, A.A., O.O. Yusuf and O.O. Okedeyi. 2008. Assessment of the exposure of two fish species to metals pollution in the Ogun river catchments, Ketu, Lagos, Nigeria. *Environ. Monit. Assess.* 137:451-458.
- Al-Nozha, M., A. Al-Kanhal, A. Al-Othaimeen, A. Al-Mohaeza, A. Osman, A. Al-Shammery and M. El-Shabrawy. 1991. Evaluation of the nutritional status of the people of Saudi Arabia. Final Report, King Abdulaziz City for Science and Technology (KACST), Saudi Arabia.
- Al Othman, Z.A. 2010. Lead contamination in selected foods from Riyadh City Market and estimation of the daily intake. *Molecules* 15:7482-7497.
- Al-Saleh, I. and N. Shinwari. 2002. Preliminary report on the levels of elements in four fish species from the Arabian Gulf of Saudi Arabia. *Chemosphere* 48:749-755.
- Al-Jedah, J.H. and R.K. Robinson. 2001. Aspects of the safety of the fish caught off the coast of Qatar. *Food Cont.* 12:549-552.
- Amiard, J.C., C. Amiard-Triquet, L. Charbonnier, A. Mesnil, P.S. Rainbow and W.X. Wang. 2008. Bioaccessibility of essential and non-essential metals in commercial shellfish from Western Europe and Asia. *Food Chem. Toxicol.* 46:2010-2022.
- Anonymous. 1997. Türk gıda kodeksi yönetmeliği. Dünya yayıncılık, İstanbul, Turkey.
- Anonymous. 2005. Commission Regulation (EC) No: 78/2005 of 16 January 2005 amending Regulation EC No: 466/2001 as regards heavy metals. *Official Journal L* 16/43:43-45.
- Ashraf, M., J. Tariq and M. Jaffar. 1991. Contents of trace metals in fish, sediment and water from three freshwater reservoirs on the Indus River. *Pak. Fish. Res.* 12:355-364.
- Ashraf, W., Z. Seddigi, A. Abulkibash and M. Khalid. 2006. Levels of selected metals in canned fish consumed in Kingdom of Saudi Arabia. *Environ. Monit. Assess.* 117: 271-279.
- Biegalski, S. and S. Landsberger. 1999. An evaluation of atmospheric deposition of trace elements into the Great Lakes. *Biol. Trace Elem. Res.* 71-72:247-256.
- Bocio, A., M. Nadal and J.L. Domingo. 2005. Human exposure to metals through the diet in Tarragona, Spain: Temporal trend. *Biol. Trace Elem. Res.* 104:193-201.
- Bu-Olayan, A.H. and M.N. Subrahmanyam. 1997. Accumulation of copper, nickel, lead and zinc by snail, *Lunella coronatus* and pearl oyster, *Pinctada radiata* from the Kuwait coast before and after the Gulf War oil spill. *Sci. Total Environ.* 97:161-165.
- CAC/FAO. 1999. Situation Analysis of Children and women in Kenya. Food Standards Programme, Codex Committee on Food Additives and Contaminants. 32nd Session. Draft Maximum Levels of Lead; GoK/UNICEF. UNICEF/GoK, Nairobi, Kenya.
- Canli, M. and G. Atli. 2005. The relationships between heavy metal (Cd, Cr, Cu, Fe, Pb, Zn) levels and the size of six Mediterranean fish species. *Environ. Pollut.* 121: 129-136.
- Carbonell, G., C. Ramos and J.V. Tarazona. 1998. Heavy metals in shrimp culture areas from the Gulf of Fonseca, Central America. II. Cultured Shrimps. *Bull Environ. Contam. Toxicol.* 60:260-265.
- Chang, L. 1996. *Toxicology of Metals*; CRC Lewis Publishers: Boca Raton, FL.
- Commission of the Europ. Comm. 1997. Draft commission regulation setting maximum limits for certain contaminants in foodstuffs. Doc III/5125/95/Rev 3.
- Commission of the Europ. Comm. (CEC). 2006. 2006/1881/EC setting maximum levels for certain contaminants in foodstuffs. *Off. J. Europ. Comm. (OJEC)*, 19 Dec, 2006, L364/5.
- Cook, A.G., P. Weinstein and J.A. Centeno. 2005. Health effects of natural dust: Role of trace elements and compounds. *Biol. Trace Elem. Res.* 103:1-15.
- Eboh, L., H.D. Mepba and M.B. Ekpo. 2006. Heavy metal contaminants and processing effects on the composition, storage stability and fatty acid profiles of five common commercially available fish species in Oron /local Government, Nigeria. *Food Chem.* 97:490-497.
- Edwards, J.W., K.S. Edyvane, V.A. Boxall, M. Hamann and K.L. Soole. 2001. Metal levels in seston and marine fish flesh near industrial and metropolitan centres in South Australia. *Mar. Pollut. Bull.* 42:389-396.
- European Commission. 2000. Amending Commission Regulation (EC), No 194/97. Brussels.
- FAO/WHO. 1976. List of maximum levels recommended for contaminants. Joint FAO/WHO Codex Alimentarius Commission. 2nd series. CAC/FAL 3: 1-8, Rome, Italy.

- Falandysz, J. 1989. Trace metal levels in the raw and tinned squid *Loligo patagonica*. Food Addit. Contam. 6:483-488.
- Food and Agriculture/World Health Organization (FAO/WHO). 1972. Evaluation of certain food additives and the contaminants mercury, cadmium and lead. WHO Techn. Rep. Ser. No. 505. Geneva.
- FEPA. 2003. Guidelines and Standards for Environmental Pollution Control in Nigeria, Federal Environmental Protection Agency, Nigeria. 238p.
- Ferre-Huguet, N., R. Marti-Cid, M. Schuhmacher and J.L. Domingo. 2008. Risk assessment of metals from consuming vegetables, fruits and rice grown on soils irrigated with waters of the Ebro River in Catalonia, Spain. Biol. Trace Elem. Res. 123:1-14.
- Gaspic, Z.K., T. Zvonaric, N. Vrgoc, N. Odzak and A. Baric. 2002. Cadmium and lead in selected tissues of two commercially important fish species from the Adriatic Sea. Water Res. 36:5023-5028.
- Harris, W.S. 1989. Fish oils and plasma lipid and lipoprotein metabolism in humans: a critical review. J. Lipid Res. 30:785-807.
- Heidarieh, M., M. Ghannadi, M. Shamami, M. Behgar, F. Ziaei and Z. Akbari. 2013. Evaluate of heavy metal concentration in shrimp (*Penaeus semisulcatus*) and crab (*Portunus pelagicus*) with INAA method. Springer Plus. 2:72-75.
- Henry, F., R. Amara, L. Courcot, D. Lacourte and M.L. Bertho. 2004. Heavy metals in four species from the French coast of the Eastern English Channel and Southern Bight of the North Sea. Environ. Int. 30:675-683.
- Huang, T.L., P.P. Zandi, K.L. Tucker, A.L. Fitzpatrick, L.H. Kuller, L.P. Fried, G.L. Burke and M.C. Carlson. 2005. Benefits of fatty fish on dementia risk are stronger for those without APOE 4. Neurol. 65:1409-1414.
- JECFA. 2000. Safety Evaluation of Certain Food Additives and Contaminants. 44 IPCS-International Programme on Chemical Safety Contaminants. Available online at: <http://www.inchem.org/documents/jecfa/jecmono/v44jeeI2.htm/>.
- Khansari, F.E., M.G. Khansari and M. Abdollahi. 2005. Heavy metals content of canned tuna fish. Food Chem. 93:293-296.
- Küçüksezgin, F., A. Kontas, O. Altay, E. Uluturhan and E. Darilmaz. 2006. Assessment of marine pollution in Izmir Bay: Nutrient, heavy metal and total hydrocarbon concentrations. Environ. Int. 32:41- 51.
- Leblanc, J.C., T. Gue'rin, L. Noel, G. Calamassi-Tran, J.L. Volatier and P. Verger. 2005. Dietary exposure estimates of 18 elements from the 1st French total diet study. Food Addit. Contam. 22:624-641.
- Magliette, R.J., F.G. Doherty, D. Mackinney and L. Venkataramani. 1995. Need for environmental quality guidelines based on ambient freshwater quality criteria in natural waters-case study "Zinc". Bull. Environ. Contam. Toxicol. 54:532-626.
- Marx, H. and B. Brunner. 1998. Heavy metal contamination of North Sea shrimp (*Crangon crangon* L.). Z. Lebensm. Unters. Forsch. A. 207:273-275.
- Mason, A.Z. and K. Simkis. 1983. Interaction between metals and their distribution in tissues of *Littorina littoria* (L) collected from clean and polluted sites. J. Mar. Biol. Ass. UK. 63:661-672.
- Massadeh, A., F. Al-Momani and A. Elbetieha. 2007. Assessment of heavy metals concentrations in soil samples from the vicinity of busy roads: Influence on *Drosophila melanogaster* life cycle. Biol. Trace Elem. Res. 122:1-8.
- Mendil, D., O.D. Uluözlu, E. Hasdemir, M. Tüzen, H. Sari and M. Suiçmez. 2005. Determination of trace metal levels in seven fish species in lakes in Tokat, Turkey. Food Chem. 90:175-179.
- Mendil, D. and O.D. Uluozlu. 2007. Determination of trace metal levels in sediment and five fish species from lakes in Tokat, Turkey. Food Chem. 101:739-745.
- Mertz, W. 1981. The Essential Trace Element. Science 213: 1332-1338.
- Miller, G.G., L.I. Sweet, J.V. Adams, G.M. Osmann, D.R. Passino-Reader and P.G. Meter. 2002. In vitro toxicity and interactions of environmental contaminants (Arochlor 1254 and mercury) and immunomodulatory agents (lipopolysaccharids and cortisol) on thymocytes from lake trout (*Salvelinus namaycusi*). Fish and Shellfish Immun. 13:11-26.
- National Research Council. 1985. Oil in the sea. Inputs fates and effects. National Academy Press, Washington, DC.
- Offenbacher, E.G. and F.X. Pi-Sunyer. 1988. Chromium in human nutrition. Ann. Rev. Nutr. 8:543-563.
- Olowu, R.A., O.O. Ayejuyo, G.O. Adewuyi, I.A. Adejoro, A.A.B. Denloye, A.O. Babatunde and A.L. Ogundajo. 2010. Determination of heavy metals in fish tissues, water and sediment from Epe and Badagry Lagoons, Lagos, Nigeria. E-J. Chem. 7:215-221.
- Pastor, A., F. Hernandez, M.A. Peris, J. Beltrán, J.V. Sancho and M.T. Castillo. 1994. Levels of heavy metals in some marine organisms from the Western Mediterranean Area (Spain) Mar. Pollut. Bull. 28:50-53.
- Price, A.R.G. and C.R.C. Sheppard. 1991. The Gulf: past, present, and possible future states. Marine Pollut. Bull., 22:222-227.
- Rashed, M.N. 2001. Monitoring of environmental heavy metals in fish from Nasser Lake. Environ. Int. 27:27-33.
- Romeo, M., Y. Siau, Z. Sidoumou and M. Gnassia-Barelli. 1999. Heavy metal distribution in different fish species from the Mauritania coast. Sci. Total Environ. 232:169-175.

- Sadiq, M. and T.H. Zaidi. 1985. Vanadium and nickel content of Nowruz spill tar flakes on the Saudi Arabian coastline and their probable environment impact. *Bull. Environ. Contam. Toxicol.* 32:635-639.
- SASO Saudi Arabian Standards Organization. 1997. Maximum limits of contaminating metallic elements in foods. Riyadh, Saudi Arabia.
- Satarug, S., J.R. Baker, S. Urbenjapol, M. Haswell-Elkins, P.E.B. Reilly, D.J. Williams and M.R. Moore. 2003. A global perspective on cadmium pollution and toxicity in non-occupationally exposed population. *Toxicol. Lett.* 137:65-83.
- Staniškienė, B., P. Matusėvičius and A. Urbonavičius. 2009. Distribution of heavy metals in muscles of fish: concentrations and change tendencies. *Environ. Res. Eng. Manag.* 48:35-41.
- Stone, N.J. 1996. Fish consumption, fish oil, lipids, and coronary heart disease. *Circul.* 94:2337-2340.
- Sures, B., H. Taraschewski and C. Haug. 1995. Determination of trace metals (Cd, Pb) in fish by electrothermal atomic absorption spectrometry after microwave digestion. *Anal. Chim. Acta.* 311:135-139.
- Tariq, J., M. Jaffar and M. Ashraf. 1993. Heavy Metal concentrations in fish, shrimp, seaweed, sediment and water from Arabian Sea, Pakistan. *Mar. Pollut. Bull.* 26: 644-647.
- Tawfik, M.S. 2008. Proximate composition and fatty acids profiles in most common available fish species in Saudi market. *Asian J. Clin. Nutr.* 1:50-57.
- Terry, P., P. Lichtenstein, M. Feychting, A. Ahlbom and A. Wolk. 2001. Fatty fish consumption and risk of prostate cancer. *Lancet.* 357:1764-1766.
- Tinggi, U., C. Reilly and C. Patterson. 1997. Determination of manganese and chromium in foods by atomic absorption spectrometry after wet digestion. *Food Chem.* 60:123-128.
- Türkmen, A., M. Türkmen, Y. Tepe and I. Akyurt. 2005. Heavy metals in three commercially valuable fish species from Iskenderun Bay, Northern East Mediterranean Sea, Turkey. *Food Chem.* 91:167-172.
- Tüzen, M. 2003. Determination of heavy metals in fish samples of the middle Black Sea (Turkey) by graphite furnace atomic absorption spectrometry. *Food Chem.* 80:119-123.
- Vogt, G. and E.T. Quinitio. 1994. Accumulation and excretion of metal granules in the prawn, *Penaeus monodon*, exposed to water-borne copper, lead, iron and calcium. *Aquat. Toxicol.* 28:223-241.
- Vos, G. and J.P.C. Hovens. 1986. Chromium, nickel, copper, zinc, arsenic, selenium, cadmium, mercury and lead in Dutch fishery products 1977-1984. *Sci. Total Environ.* 52:25-40.
- WHO (World Health Organization). 1985. Guidelines for the study of dietary intakes of chemical contaminants. World Health Organization, Geneva.
- WHO (World Health Organization). 1989. Evaluation of certain food additives and contaminants. In: 33rd Report of the Joint FAO/WHO Expert Committee on Food Additives. WHO: Geneva, Switzerland.
- WHO (World Health Organization). 1993. Evaluation of certain food additives and contaminants. In: 37rd Report of the Joint FAO/WHO Expert Committee on Food Additives. WHO: Geneva, Switzerland.
- WHO (World Health Organization). 1995. Trace elements in human nutrition and health, World Health Organization, Geneva, Switzerland.
- Wolk, A., S.C. Larsson, J.E. Johansson and P. Ekman. 2006. Long-term fatty fish consumption and renal cell carcinoma incidence in women. *JAMA.* 296:1371-1376.
- Zhang, L. and M.H. Wong. 2007. Environmental mercury contamination in China: Sources and impacts. *Environ. Int.* 33:108-121.
- Zukowska, J. and M. Biziuk. 2008. Methodological evaluation of method for dietary heavy metal intake. *J. Food Sci.* 73:R21-R29.