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Impact of Different Cooking Processes on Proximate Metals Composition of Fish and Shrimp

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Abstract: This study analyzed three cooked species of fish and one species of shrimps (grilled, fried, baked, marinated for fish and traditional cooked for shrimp) commonly consumed in Arabian countries for their proximate, mineral and heavy metal content. The results revealed that the highest concentration of metals was found in marinated and fried fish. The difference in Arsenic (As), Cadmium (Cd), Nickel (Ni), Lead (Pb) and Copper (Cu) between raw and cooked fish were decreased but not significantly after baking and grilling. The cooking process of shrimp increase significantly metals content in except of Pb which was present at the same extend. It is advisable to avoid excessive frying and use minimal spices. In general, the cooking process is only of a very limited value as a means of reducing metal concentrations.

Key words: Minerals, fish, shrimp, cooking processes, metal content

INTRODUCTION

The metals content of fishery products need to be well established as fishery products are widely consumed by humans. Since, fish is the last link in the aquatic food chain, the metal concentrations in many fish species have been determined in relation to the metal content of the aquatic environment. Industrialization has improved general technology as well as quality of life but has also resulted in an increase in metal concentrations in water (Tarley *et al*., 2001). These metals can be classified as potentially toxic (aluminium, arsenic, cadmium, lead, mercury, etc.), probably essential (nickel, vanadium, cobalt) and essential (copper, zinc, selenium) (Munoz-Olivas and Camara, 2001). The most important heavy metals from the point of view of water pollution are Zn, Cu, Pb, Cd, Hg, Ni and Cr. Some of these metals (Cu, Ni, Cr and Zn) are essential trace metals to living organisms but become toxic at higher concentrations. Others such as Pb and Cd have no known biological function but are toxic elements (Rashed, 2001a, b). Toxic elements can be very harmful even at low concentration when ingested over a long time period. Other elements which are also present in seafood are essential for human life at low concentration however, they can also be toxic at high concentrations (Oehlenschlager, 2002).

Concerns on the exposure of reared aquatic organisms to metals derive both from toxicity, i.e., the potential of deleterious effects on the reared population (Carbonell and Tarazona, 1993) and from bioaccumulation with possible health hazards for the human consumer (Vos and Hovens, 1986). Crustaceans have a high sensitivity to metals, demonstrated for several taxonomic groups including those with a high commercial value such as shrimps, prawns, lobster or crabs (Bambang *et al*., 1995). Toxicity is related to the concentration of the bioavailable form of the metal in the surrounding environment. For decapods crustaceans the free (hydrated) metal ion is commonly considered the available (toxic) form for uptake from solution (Rainbow, 1995). The available form is in equilibrium with the non-available form bound to complexing ligands. Bioavailability can result for the bioaccumulation of the metal. Any increase in total dissolved metal concentration would correspondingly increase the free metal ion concentration and thereby lead to an increase in metal uptake rate (Rainbow, 1995). If the rate of excretion does not follow a parallel increase, a net accumulation of metal occurs within the body. The accumulation of metals in aquatic invertebrates is generally greater than in fish as result of the differences in the evolutionary strategies adopted by the various phyla (Phillips and Rainbow, 1993).

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, 2009). 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) reduced risk of dementia and alzheimer disease in certain conditions (Huang *et al.*, 2005).

Dietary habits may include foods components and cooking processes. Fish is usually eaten after various cooking treatments such as boiling, grilling, baking or frying but sometimes it is eaten raw (in sushi meal). On the other hand, the use of the microwave oven for cooking has increased greatly during the past few decades (Garcia-Arias *et al.*, 2003). It is known that cooking may affect the amount and speciation of chemical pollutants in foods (Burger *et al.*, 2003; Ersoy and Ozeren, 2009; Ersoy *et al.*, 2006; Gokoglu *et al.*, 2004; He *et al.*, 2010; Perello *et al.*, 2008).

Many studies have been published on the determination of heavy metals in fish (Topcuoglu *et al.*, 2002) but these studies are inadequate for estimating the intake of these heavy metals by humans since they were carried out on raw fish. However, in the literature, there are some studies on cooked fish (Steiner-Asiedua *et al.*, 1991; Atta *et al.*, 1997; Devesa *et al.*, 2001a, b; Dugo *et al.*, 2006; Gokoglu *et al.*, 2004; Hoffman *et al.*, 1994; Khansari *et al.*, 2005). Jorhem *et al.* (1994) have reported that some changes occurred during cooking in the Cd, Ni, Co, Pb, Cu and Mn concentrations of crayfish, varying according to the metal and the organ considered. It is known that cooking may, in certain conditions, change the amount of chemical pollutants in foods (Bayen *et al.*, 2005; Burger *et al.*, 2003; Hori *et al.*, 2005; Schecter *et al.*, 1998, Schecte *et al.*, 2006; Tsutsumi *et al.*, 2002; Voorspoels *et al.*, 2007; Zabik *et al.*, 1995). With respect to metals, it has been reported that although cooking may reduce their contents, some foods can also absorb metals if the cooking water is contaminated (Morgan, 1999). Atta *et al.* (1997) observed a decrease of Cd, Cu, Pb and Zn concentrations after steaming or baking. During thermal processing, the application of heat hastens protein degradation and loss of weight and water and therefore, chemical contaminants may also be affected by the heat applied (Lind *et al.*, 1995; Burger *et al.*, 2004; Cabanero *et al.*, 2004).

To evaluate the possible effect of cooking on the contents of metals, the concentrations obtained in the cooked products were compared with the concentrations found in the same products in the raw state (Munoz *et al.*, 2000).

Therefore, the main purpose of the present study was to assess metals composition in three famous types of fish (Boulty, Boury and Sardine) and metals composition in shrimp and to evaluate the possible effect of cooking on the concentration of these metals.

MATERIALS AND METHODS

Sample preparation and cooking: Fresh samples of four species included Nile Tilapia (*Oreochromis niloticus*), Gray Mullet (*Mugil cephalus*), California Sardine (*Sardinops sagax*) and black tiger shrimp (*Penaeus monodon*) which known in the market as Boulty, Boury, Sardine and Big Shrimp, respectively used in this study. Local, common and scientific names of these fishes and shrimps are presented in Table 1. All fish and shrimp 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 2 h. 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. They were washed with water and were subsequently cooked.

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

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

Table 2: Ingredients and methods of preparation of fish and shrimp

Common names	Ingredients	Method of preparation
Fried fish	Fish, sunflower oil and salt	Place fish in salt for 1 h. Wash with water, fry in hot oil at 180°C for 4 min until brown
Grilled fish	Fish and salt	Place fish in salt for 1h. Wash with water and grill at 180°C for 20 min till done
Baked fish	Fish, butter and salt	Place fish in a greased baking pan. Brush with melted butter and season with salt. Bake in center of preheated oven at 180°C for 20 min
Marinated fish	Fish, onions, garlic, green coriander, corn oil, salt, mixed spices, dried lemon and water	Marinate fish in salt and spices for 1 h. Wash with water, brown onions and garlic in oil. Add fish and other ingredients and cook under low heat till done
Cooked shrimp	Shrimp, onions, garlic, corn oil, butter, salt, mixed spices, ground cardamom, dried lemon, chopped pepper and water	Place shrimp in salt for 1 h. Wash with water, brown onions and garlic, add shrimp and rest of ingredients. Place shrimp with rest of the ingredients and cook till done

The fish were divided into five groups. The first group was uncooked. The other four groups were fried, grilled, baked and marinated. Shrimp were cooked according to traditional method (Table 2). The skin of the fish was removed after the cooking process. Raw and cooked samples (muscle) 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.

Total raw and cooked fish individuals were analyzed for arsenic, cadmium, chromium, nickel, lead, copper, zinc and manganese. Metal contents were expressed as μg/g wet wt. of fresh fish.

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

Chemical analyses: The 10 g of tissue homogenate were weighted into a 150 mL air-tight quick flask with glass stopper. About 5 mL of conc. HNO_3 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 samples were separately filtered using an ashless filter paper and the volumes made up to 100 mL with 0.5% HNO_3 then used for the determination of metals (Eboh *et al*., 2006).

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 = 0.05) (SPSS 10.0 for windows).

RESULTS AND DISCUSSION

The concentrations of metals in raw, fried, grilled, baked and marinated fish (boulty, boury and sardine) are given in Table 3-5. There were no significant differences in Pb concentrations between the raw, grilled and backed fish. However, a slight decrease in Pb concentration was observed in baked sardine. Results did not indicate much variation for lead in backed boulty and boury. The increase in Pb concentration was significant ($p<0.05$) for fried and marinated cooking methods when compared to the raw control. This increase in lead could be due to the ingredients used in marinated cooking and due to loss of moisture during frying. Earlier study indicate that spices could contain high levels of lead due to contamination (Woolf and Woolf, 2005). The concentrations of Pb in sardine and tuna were lower in grilled samples where the Pb level was increased in frying sardine (Musaiger and D'Souza, 2008). Comparison between different values for the *Lethrinus nebulosus* indicated an increase in lead values in the curried

Table 3: Mineral composition of cooked boulty (μg g^{-1}, wet weight)

Samples	Arsenic (As)	Cadmium (Cd)	Chromium (Cr)	Nickel (Ni)	Lead (Pb)	Copper (Cu)	Zinc (Zn)	Manganese (Mn)
Raw	0.010^a	0.016^a	0.006^a	0.019^a	0.002^a	0.37^a	4.18^a	0.26^a
Fried	0.020^b	0.021^b	0.014^b	0.059^b	0.011^b	0.48^b	6.45^b	0.48^b
Grilled	0.009^a	0.012^a	0.007^a	0.020^a	0.002^a	0.38^a	4.22^a	0.27^a
Baked	0.009^a	0.014^a	0.008^a	0.019^a	0.002^a	0.39^a	4.98^a	0.31^a
Marinated	0.025^c	0.053^c	0.037^c	0.056^b	0.016^c	0.56^c	6.42^b	0.59^d

Table 4: Mineral composition of cooked boury (μg g^{-1}, wet weight)

Samples	Arsenic (As)	Cadmium (Cd)	Chromium (Cr)	Nickel (Ni)	Lead (Pb)	Copper (Cu)	Zinc (Zn)	Manganese (Mn)
Raw	0.014^a	0.168^a	0.007^a	0.022^a	0.002^a	0.76^a	5.02^a	0.11^a
Fried	0.027^b	0.360^b	0.015^b	0.033^b	0.011^b	0.89^b	8.97^b	0.44^b
Grilled	0.013^a	0.155^a	0.008^a	0.022^a	0.002^a	0.78^a	5.29^a	0.15^a
Baked	0.012^a	0.154^a	0.009^a	0.023^a	0.003^a	0.77^a	5.42^a	0.19^a
Marinated	0.033^c	0.395^b	0.039^c	0.032^b	0.016^b	0.88^b	8.61^b	0.94^c

Table 5: Mineral composition of cooked sardine (μg g^{-1}, wet weight)

Samples	Arsenic (As)	Cadmium (Cd)	Chromium (Cr)	Nickel (Ni)	Lead (Pb)	Copper (Cu)	Zinc (Zn)	Manganese (Mn)
Raw	0.033^a	0.369^a	0.003^a	0.009^a	0.005^a	0.56^a	6.28^a	0.25^a
Fried	0.062^b	0.821^b	0.010^b	0.015^b	0.022^b	1.13^b	13.86^b	0.82^b
Grilled	0.031^a	0.357^a	0.005^a	0.011^a	0.001^a	0.59^a	6.63^a	0.32^a
Baked	0.030^a	0.288^a	0.004^a	0.010^a	0.002^a	0.60^a	6.38^a	0.37^a
Marinated	0.067^b	0.636^b	0.014^b	0.014^b	0.026^b	0.94^b	13.46^b	1.21^c

Within the column values with different letters are significantly different ($p<0.05$). Values are shown as means of duplicates, n = 2

Lethrinus nebulosus and this increase in the lead level is due to loss of moisture. There was no difference in lead content in the curried *Scomberomorus commerson* and that cooked in rice but fried *Scomberomorus commerson* showed a slight increase (Morgan *et al*., 1997). Atta *et al*. (1997) have reported a decrease in the concentrations of Pb in *Tilapia nilotica* after baking. Earlier investigation indicated that grilling is the cooking process causing an increase on the concentration of Pb in food (Ersoy *et al*., 2006). There were no significant differences in Pb concentrations between the raw, grilled, fried fish. However, the decrease in Pb concentration was significant ($p<0.05$) for baked cooking method when compared to the raw control (Erosy *et al*., 2006). Perello *et al*. (2008) showed that the concentration of Pb in sardine was lower in grilled samples where the Pb level was increased in frying sardine.

The same trend in Cd concentrations of raw, baked, grilled, fried and marinated samples (Table 3-5). The modes of cooking (backing and grilling) had no significant effect on the cadmium content of fish and any amount of cadmium reported was from the original source which was the raw fish. The highest concentration of this element was found in sardine after fried and marinated. The increase of metal may be related to the changes in moisture concentration that occur during cooking. With regard to Cd, a number of earlier data show that the concentrations of this element in raw, roast, grilled and fried samples of white fish samples were under the detection limit (Falco *et al*., 2006; Ersoy *et al*., 2006). In the present study, Cd concentrations were similar to those previously found by other researchers (Llobet *et al*., 2003; Gonzalez-Weller *et al*., 2006). Alberti-Fidanza *et al*. (2002) reported that Cd content in raw and cooked foods showed slight differences after grilling and baking. Amiard *et al*. (2008) showed that bioaccessibility was significantly lowered for Cd after cooking a total of seven species of mollusks from France, UK and Hong Kong including clams, mussels, oysters, scallops and gastropods. Perello *et al*. (2008) showed that the highest concentration of this element in sardine was found after cooking.

The mean Cr concentration of the fillets determined in raw samples was 0.003-0.007 $\mu g\ g^{-1}$. The increase in Cr concentration were found for various cooking samples compared to the raw samples but it was significant ($p>0.05$) in fried and marinated samples. Cr concentration were higher in marinated fish than in the other cooked fish. Alberti-Fidanza *et al*. (2002) reported that Cd content in raw and cooked foods showed slight differences. Lind *et al*. (1995) and Amiard *et al*. (2008) showed that bioaccessibility was significantly lowered for Cd after cooking a total of seven species of mollusks from France, UK and Hong Kong including clams, mussels, oysters, scallops and gastropods. In different organs of fish, Atta *et al*. (1997) observed a decrease of Cd concentration after steaming or baking. Topcuoglu *et al*. (2002) found that the changes after cooking in Cr concentration were insignificant ($p>0.05$).

The As concentration of raw fish was found to be 0.010-0.033 $\mu g\ g^{-1}$. The highest value was found in the fried and marinated samples while the lowest value was in the grilled and baked samples (Table 3-5). The increase in As concentration was significant ($p<0.05$) for fried and marinated samples but the decrease in As concentration was not significant for grilled and backed samples. Sardine presented higher As concentrations when marinated than when fried (0.067 and 0.062 $\mu g\ g^{-1}$, respectively) with both values being higher than those observed in raw sardine (0.033 $\mu g\ g^{-1}$). Devasa *et al*. (2001b) who studied the effect of various cooking treatments were used (grilling, roasting, baking, stewing, boiling, steaming and microwaving) on total and inorganic arsenic concentrations have reported that after cooking, there was a significant increase in the concentration of inorganic. As for bivalves and squid but sardines, crustaceans, anchovies and Atlantic horse mackerel showed no significant differences among them. Lee *et al*. (2006) reported levels of 0.28 $\mu g\ g^{-1}$ of As in stir-fried anchovy. In earlier studies (Marti-Cid *et al*., 2007) As concentration in raw sample of sardine was similar to those found in the present study while the current As concentration in boulty was lower. A mechanism which would lead to an increase in the concentrations of inorganic arsenic after cooking might be attributed as in the case of total arsenic, to the weight reduction during cooking as a result of loss of water, volatiles and to a lesser extent the other gross sample constituents (lipids, carbohydrates and proteins) (Devesa *et al*., 2001b). With respect to the existence of losses of total arsenic, no data are available concerning their possible volatilization during the cooking of food products. However, several studies report that there is solubilization of organoarsenical species during the cooking process. This solubilization may be due to the lability of the bond (electrostatic link) between AB and the proteins in fish muscle. Solubilization of arsenic species (AB and DMA) in the accompanying liquid was shown in a earlier study carried out with canned fish (Velez *et al*., 1997). Ersoy *et al*. (2006) who studied effects of four cooking methods on the heavy metal concentrations of sea bass fillets (*Dicentrarchus labrax*) concluded that as concentrations of fried samples were significantly increased and the highest value found in the fried samples. Perello *et al*. (2008) showed that arsenic

Table 6: Mineral composition of cooked Shrimp (μg g^{-1}, wet weight)

Samples	Arsenic (As)	Cadmium (Cd)	Chromium (Cr)	Nickel (Ni)	Lead (Pb)	Copper (Cu)	Zinc (Zn)	Manganese (Mn)
Raw	0.230^a	0.438^a	0.030^a	0.020^a	0.008^a	2.16^a	13.56^a	0.26^a
Cooked	0.252^b	1.404^b	0.042^b	0.080^b	0.006^a	4.42^b	39.00^b	0.64^b

Within the column values with different letters are significantly different ($p<0.05$). Values are shown as means of duplicates, n = 2

concentrations in cooked fish were higher than those found in raw fish, sardine presented higher As concentrations when grilled than when fried (3.28 and 2.91 μg g^{-1}, respectively) with both values being higher than those observed in raw sardine (2.09 μg g^{-1}).

The Ni concentration of cooked samples was significantly increased in fried and marinated fish and not significantly change in grilled and baked fish. The highest value was detected in fried and marinated boulty (0.059 and 0.056 μg g^{-1}). Topcuoglu *et al.* (2002) have reported that Ni concentration of raw sea bass (*D. labrax*) was 0.06 μg g^{-1}. The highest value was detected in the microwave-cooked samples (0.93 mg kg^{-1}).

Zinc levels were higher in the muscles of raw fish (4.18 μg g^{-1} in boulty, 5.02 μg g^{-1} in boury and 6.28 μg g^{-1} in sardine) and among the cooked fish the highest levels were seen in the marinated and fried sardine (13.46 and 13.86 μg g^{-1}, respectively). On the other hand the grilled boulty had lower zinc content (4.22 μg g^{-1}). Zinc content was increased in all types of cooked fish but significantly increased in fried and marinated fish. The high content of zinc is important since zinc deficiency affecting the growth of the children and adolescents in the Arab Middle East countries including the Gulf States has been reported (Miladi, 1996).

The same trend in Cu and Mn concentrations of raw, fried, grilled, backed and marinated fish (Table 3-5). There was an increases in the content of Cu an Mn in different cooking types but it was increased significantly in fried and marinated fish and the highest content was observed in marinated fish. The metal levels were considerable in shrimps and most of the fish which is mainly due to the spices added to the diet to make it more palatable than the same diets without spices (Morgan, 1999). Copper were increased in all the varieties of cooked fish tested with not much of a variation observed in grilled and backed fish but were increased significantly in the various modes of cooking in sardine. Furthermore, increase in the metal content in the marinated fish compared to the other cooking variety can be attributed to the spices such as dried lemon and tamarind which contain considerable amount of metal and are used in the preparation of fish (Musaiger, 2006).

The metals content in raw and cooked shrimp is presented in Table 6. Lead content was generally lower (0.006 μg g^{-1}) in the muscle of cooked shrimp. There was no significant difference in the lead content between raw and cooked shrimp. Other metals content of cooked shrimp was increase significantly than the same raw. Cooked *Penaeus monodon* showed high levels of metal which could be a result of contribution from the spices commonly used in the Arabian Gulf region (Musaiger, 2006). There was a slight variation in the arsenic and chromium content in cooked shrimp (0.252, 0.042 μg g^{-1}). In all types of seafood observed by Devesa *et al.* (2001a, b), the concentrations of inorganic As varied between 0.008 and 1.08 μg g^{-1}. In hake, the highest As concentrations were noted when this species was grilled (1.38 versus 1.28, 1.06, 1.01 and 0.94 μg g^{-1} when fried, roasted, boiled or raw, respectively). The effect of the concentration of arsenic after cooking was indicated in a Canadian survey of arsenic in total diet food composites (Dabeka *et al*., 1993) this study reported a mean increase in the concentration of total arsenic in seafood after cooking (raw, 2466 ng g^{-1}, wet wt. cooked, 3048 ng g^{-1}, wet wt.) and the researchers indicated that this increase generally agreed closely with the decrease in weight. Cadmium content was higher in cooked shrimp. It is also observed that cooked shrimp had higher level of Ni, Cu, and Mn compared to raw shrimp. The variations in the concentration of metal in seafood products after cooking may be the result of the sum of two contrary effects: concentration of the metalloid due to the decrease in weight resulting from loss of water, volatiles and to a lesser extent the other gross sample constituents (lipids, carbohydrates and proteins) and loss of total metals as a result of volatilization or solubilization (Velez *et al*., 1997). No earlier studies were found on Ni concentration in cooked seafood.

CONCLUSION

The results of this study comparing the influence of several cooking processes on metal levels in various fish and shrimp can potentially be useful as a simple guide for the selection of food cooking for preparing a healthy diet with a low content of toxic metals. The different modes of cooking have an influence on metal composition of fish and shrimp. In the frying and marinating methods, the concentrations of metals increased significantly. The investigation indicates that the decrease were statistically insignificant for metal of baked and grilled fish. However, Zn, Mn, Cu and Cr concentration was insignificantly increased in baked and grilled samples. In general, sardine (marinated and fried) showed a clear tendency to increase metal concentrations after cooking. Anyhow, it is important to note that all of the differences between raw and cooked fish were not statistically significant ($p>0.05$) after baking and grilling. High concentrations of As, Zn, Cu and Mn was observed in fish with the highest Zn levels corresponding to sardine. Shrimp also showed increasing in the concentrations of metals after cooking. It can be recommended that grilling and baking are the most suitable methods for human consumption. Therefore, it is possible to reduce the metal in fish parts by choosing a suitable method of cooking and reduce using spices.

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