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Assessment of heavy metals in edible muscles of some marine organisms from the Arabian Gulf (in Saudi Arabia)

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ABSTRACT

Over the last decades, there has been growing interest in determining heavy metal levels in the marine environment and attention was drawn to the measurement of contamination levels in public food supplies, particularly seafood. The aim of this study was to determine the current levels of total copper, chromium, cadmium, lead, manganese ,magnesium, zinc and mercury in two crustacean species namely *Penaeus* sp. and *Portunus* sp. and one molluscan cephalopod species (*Sepia* sp.) which were caught from Saudi Arabia coastlines on the Arabian Gulf to ascertain whether these concentrations exceeded the permissible limits and to identify any potential public health risks that could be associated with dietary intakes of such seafood from the Arabian Gulf.

The results revealed that the average concentrations of these heavy metals, except for cadmium, in edible muscles of *Sepia* were higher than that of prawn and crab. The mean concentrations of these metals, except for chromium and lead, were lower in edible tissues of the prawn than that in the crab. Furthermore, it was recorded that mercury was not detected in edible muscles of both prawn and crab. Therefore, metals levels in tissues of these aquatic organisms were found below the maximum content recommended by health agencies and so they are considered safe for human consumption.

Key words: Arabian Gulf, heavy metals, muscles, *Sepia Penaeus*, Partunus.

INTRODUCTION

The term heavy metal refers to any metallic chemical element that has a relatively high density and is toxic or poisonous at high concentration. The definition may also include trace elements when considered in abnormally high toxic doses. They are natural components of the Earth’s crust and can not be degraded or destroyed (Kalay *et al.,* 1999). Heavy metals have long been recognized as serious pollutants of aquatic environment. They may affect organisms directly by transferring to next trophic level of the food chain. In the aquatic environment, heavy metals in dissolved form are easily taken up by aquatic organisms, where they are strongly bound with sulfhydril groups of proteins and accumulate in their tissues (Szeffer *et al*., 1999 and Shah, 2005). Metal bioaccumulation by marine organisms has been the subject of considerable interest in recent years because of serious concern that high levels of heavy metals may have determinable effects on marine organisms and may create problems in relation to their suitability as food for humans (Asharf, 2006 and Ahdy *et al*., 2007).

Aquatic environment is of the greatest importance for the Kingdom of Saudi Arabia, since seawater constitutes the unlimited source for potable water production through desalination processes. Furthermore, the Arabian Gulf as well as the Red Sea acquires an added importance as a reservoir of vastly abundant and relatively cheap seafood especially for shrimps. This seafood is considered as an important alternative source of livestock proteinaceous food. Moreover, the seafood is of value for both local consumption and export revenue. The shores of Arabian Gulf had witnessed phenomenal development activities during the past decades in the wake of massive oil explorations. Several industrial centers have cropped up in this region and the threat from metal pollution has been stated to be more real than ever before, besides consequences of the 1991-Gulf war (Sadia & Mc caine, 1993). In light of this concern and the pollution that is the Arabian Gulf facing, questions have been raised about the safety of eating seafood from the Arabian Gulf water. Therefore, the present study was carried out to measure the concentrations of heavy metals (copper, chromium, cadmium, lead, manganese, magnesium, zinc and mercury) in species of commercial importance; two crustacean species (*Penaeus* sp. and Portunus sp.) and one cephalopod species (*Sepia* sp.) which were caught from the Gulf in order to assess the seafood safety.

MATERIAL AND METHODS

Two crustacean species (*Penaeus* and *Portunus*) and one cephalopod species (*Sepia*) were collected from local fishermen (during October, 2007), muscles tissues were removed from each group and samples were kept frozen and prepared for analysis according to Agemian *et al.* (1980). About 5 g of each sample (dry weight) was placed into a digestion tube. Then 5 ml of conc. Nitric acid and then 5 ml of Sulphuric acid were added to the sample. When the reaction was slow, the tubes were placed in a hot block digestion apparatus and heated at low temperature (60°C) for 30 minutes. Then the tubes were allowed to cool, then 10 ml of Nitric acid were added and the tubes returned to digestion apparatus and heated slowly to 120°C then to 150°C. When the tubes with the samples became black, they were allowed to cool, and then 1ml of H_2O_2 was added. A vigorous reaction occurred. The tubes returned to the block with H_2O_2 additions until the samples were clear. The tubes were removed and filled up to 50 ml with deionized water. All the digested samples were analyzed by Perkin Elmer analyst HGA-800 atomic absorption (Micro analytical center, Faculty of Science, Cairo University).

RESULTS

Number of sample, scientific, common name, and feeding habits of each sample are summarized in Table (1).

Sample species	Number	Common name	Feeding habits
Penaeus sp.	20	Shrimp, prawn	Mussel and small crustaceans
Portunus sp.	20	Crab	Small fish and other animals
Sepia sp.	20	Cuttlefish	Small molluscs, shrimp, other crustacean

As shown in Table (2) and Fig (1), the highest mean levels of Copper (Cu) were recorded in edible muscles of *Sepia* (3.346ppm) and crab (2.433 ppm), while a lesser was observed in prawn (0.450 ppm). The total Chromium (Cr) concentration in prawn ranged from 0.124 ppm to 0.166ppm, while levels of this metal ranged from 0.110 to 0.155ppm and 0.130 to 0.168ppm in the crab and *Sepia* respectively. Furthermore, the total Cadmium (Cd) and Lead (Pb) concentrations in prawn were (0.055 and 0.701 ppm respectively); in crab (0.250 and 0.311 ppm respectively) and in *Sepia* (0.223 and 1.029 ppm respectively).

Table (2): Mean concentrations (ppm) ± standard error and ranges of heavy metals in studied marine organisms.

Sample species	Cu	Cr	Cd	Pb
Penaeus sp.	0.450±0.02 0.250-0.515	0.140±0.0023 0.124-0.166	0.055±0.0084 0.030-0.130	0.701±0.022 0.522-0.822
Portunus sp.	2.433±0.06 1.99-3.22	0.132±0.0026 0.110-0.155	0.250±0.021 0.090-0.410	0.311±0.013 0.199-0.400
Sepia sp.	3.346±0.19 1.99-4.55	0.151±0.0023 0.130-0.168	0.223±0.017 0.090-0.340	1.029±0.066 0.560-1.500

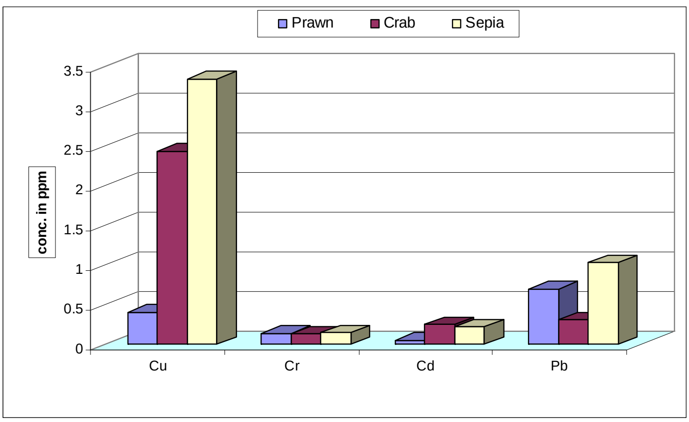

Fig. (1): Concentrations of Cu, Cr ,Cd and Pb in studied marine organisms.

With regard to concentration levels of Manganese (Mn), the present results in Table (3) and Fig. (2) pointed out that higher concentrations of Mn were seen in the cephalopod (0.297ppm) and the crab (0.267 ppm) than in prawn (0.204 ppm). Also, the highest concentrations of Magnesium (Mg) were recorded in edible muscles of *Sepia* (124.11 ppm) than in prawn (29.33ppm) and in crab (119.24ppm). Furthermore, from Table (3) and Fig. (2) ,it was observed that the concentration levels of Zinc (Zn) ranged from 1.89 to 3.66 ppm in prawn; from 4.88 to 8.22 ppm in crab and from 8.50 to 16.20 ppm in *Sepia*. However, Mercury (Hg) was not detected in edible muscles of prawn and crab, its mean concentration in *Sepia* was 0.380 ppm.

Table (3): Mean concentrations (ppm) ±standard error and ranges of heavy metals in studied marine organisms.

Sample species	Mn	Mg	Zn	Hg
Penaeus sp.	0.204±0.007 0.300-0.175	29.33±0.903 20.14-33.11	3.171±0.121 1.89-3.66	ND
Portunus sp.	0.267±0.006 0.200-0.300	119.24±1.08 110-122.8	6.641±0.201 4.88-8.22	ND
Sepia sp.	0.297±0.008 0.236-0.333	124.11±3.42 88-140	12.647±0.596 8.50-16.20	0.380±0.053 0.08-0.61

ND= not detected.

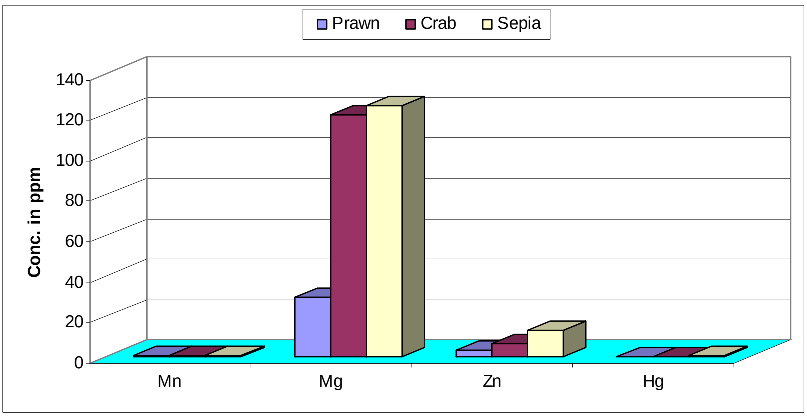

Fig. (2): Concentrations of Mn, Mg, Zn and Hg in studied marine organisms.

DISCUSSION

Many marine organisms have the potential to accumulate high levels of metals from their environment (Szeffer *et al*., 1999). Bioaccumulation means an increase in metals concentration in biological organisms compared to their concentration in the environment. Metals accumulation in living things anytime they are taken up and stored faster than they are broken down (metabolized or excreted). Because of the persistence toxicity of heavy metals and their bioaccumulation in the biota, several organizations have pointed out the need for monitoring heavy metals in the environment (UNEP/ FAO, 1996).

Metal pollution of the sea is less visible and direct than other types of marine pollution, but its effects on marine ecosystems are intensive and very extensive (Ahdy *et al.*,2007). As indirect measure of the abundance and availability of metals in the marine environment, the bioaccumulation of metals by the tissues of marine organisms had been studied. The bioaccumulation studies led to adoption of the bio-indicators concept. Seafood is widely used as bio-indicators of marine pollution by metals (Evans *et al.,* 1993). Therefore, the present study was carried out to measure mean concentration of eight heavy metals (Cu, Cr, Cd, Pb, Mg, Mn, Zn and Hg) in edible muscles of three commercially important seafood organisms for human which are represented by prawn, crab and *Sepia*.

Cu occurs in foods in many chemical forms and has important role in the physiological activities of living bodies (Abou Arab *et al*., 1996). However, Cu is the most toxic metal after mercury and silver (Clark, 1989) and considered as public health hazard if abnormal high level of Cu ingested. This essential heavy metal may cause Mediterranean anemia, haemochromatosis and liver cirrhosis (Underwood, 1977). Furthermore, Yamuna *et al.* (2002) reported that high concentrations of Cu have inhibitory effects on immune function of the haemocytes of aquatic organisms. The present study revealed that the highest concentration level was recorded in edible muscles of *Sepia*. This result is in agreement with the study of Ahdy *et al*., (2007), who observed that edible muscles of *Sepia* had a higher concentration for Cu than that of prawn and crab from Alexandria region. The estimation of Cu concentration in different tissues of different species of prawn, crab and *Sepia* in different regions of the world were recorded by Hashmi *et al*.(2002);Inzunza& Osuna(2004); Raimundo *et al*.(2005); Celik *et al*.(2006) and Panutrakul *et al*.(2007). They reported that the concentrations of Cu in edible parts of these marine samples were found to be below the limits proposed by health agencies. In the present study, Cu concentration in studied samples is not in concern according to the Saudi Arabian standards organizations (SASO, 1977) which suggested maximum allowable limits (MALS) for Cu in crustaceans and molluscs species as 20 ppm.

Regarding Cr, the current results pointed out that all studied samples contained this heavy metal within the general guideline in food (MALS =2ppm). Moreover, the present data revealed higher concentration of Cr in *Sepia* and prawn than in crab. Panutrakul *et al*. (2007) observed that the concentration of Cr in cephalothorax of banana prawn is significantly higher than in its edible muscles. While, Al sulami *et al*. (2002) did not detect Cr in edible muscles of prawn from Arabian Gulf. On the other hand, the average value of Cr concentration in blue crab from Brazil exceeded the limit allowed for any type of health agencies (Virga *et al*., 2007). Moreover, Gagnten *et al.* (2008) recorded higher Cr concentration in the tissues of freshwater crab than in sediment. Cr is essential trace metal although its toxicity at high dose, it is used in metal alloys and pigments for paints, cement, rubber and other materials. Low level exposure can irrigate the skin and cause ulceration, while long term exposure cause kidney and liver damage (Burger & Gochfeld, 1995).

In contrast to results of Cr, the present results recorded that the mean highest level of Cd was observed in edible muscles of crab (0.250 ppm), while muscles of prawn contained low Cd concentration (0.055 ppm). Furthermore, the concentration of Cd in all studied samples was found to be safe for human consumption according to several agencies and organizations; European communities (2001) have setting maximum limits for Cd in molluscs at 1ppm. Moreover, Australia, New Zealand and Hong Kong have suggested MALS value for Cd in molluscs tissues as 2 ppm. The FDA (1993) action level for Cd in crustaceans is 3ppm and 4ppm for molluscan cephalopods. While MALS value for Cd level in tissues of crustacean and molluscan species is 0.5 ppm (SASO, 1977). The present results disagree with that of Al Sulami *et al.* (2002) who observed that prawns from the Arabian Gulf water had no Cd in their edible muscles. On the other hand, Tungare& Swant (2002) reported that significant elevation of Cd levels in three important shrimp species from India caused a threat for humans and their consumption should be avoided since it is a direct source of metal intoxication. Furthermore, Cd level recorded in the edible muscles of prawns in the current study was greater than that of small and large prawns (0.0013& 0.0004 ppm respectively) reported by Burger & Gochfeld (2005). While, the concentration of this metal in studied crab samples was below (30-50 ppm) that of edible crab (*Cancer pagurus*) as reported by Overnell (1996). Moreover, the recorded values of Cd concentration in all studied samples were below the results of Soliman (2006) who recorded (0.727, 0.844 and 0.769 ppm) of Cd in the edible muscles of prawn, crab and *Sepia* respectively which were assembled from a fishing harbor of Suez Canal at Port Said. Because, the body has no mechanism for the excretion of Cd, it accumulates in the tissues (Jarup *et al.,* 1998). Therefore, chronic exposure to low level of Cd has been associated with a number of pathologies such as renal failure, early onset of diabetic renal complications, osteoporosis, deranged blood pressure and increase a cancer risk (Satarug *et al*., 2000).

Moreover, Pb is neurotoxin that causes behavioral deficits in vertebrates (Weber & Dingel, 1997) and decrease in survival growth rates and metabolism (Burger & Gochfeld, 2000). SASO (1977) had suggested MALS for Pb concentration in crustaceans and molluscs at 2 ppm. Therefore, Pb concentration in all crustaceans samples analyzed was below the risk value. Othman & Azwa (2004); Cogun *et al*.(2005) ; Celik *et al*. (2006) ; Panutrakul *et al. (*2007) and Virga *et al* (2007), analyzed Pb in crustacean species (prawn and crab) from different regions of the world and they reported that these samples still fit with the human consumption.

Regarding to estimation of Mg and Mn, the present results revealed that the mean highest level of these heavy metals was observed in edible muscles of *Sepia* than that in prawn and crab. Al Sulami *et a*l. (2002) observed that Mg and Mn were found at fairly low or undetectable levels in prawn samples; however they were detected in plentiful amounts in sediments from Saudi Arabia coastlines on Arabian Gulf. While, Adhikari *et al.* (2007) reported the accumulation of Mn was minimum in edible muscles of prawn and maximum in their hepatopancreas. Mn is an essential trace element because it is an activator of several Mn-metalloenzymes; also it is a nonspecific activator of several other enzymes. Deficiency of this element has been included in several animal species by feeding diets low in Mn. Signs of deficiency in animals include impaired growth , skeletal defects, depressed reproductive functions and defects in metabolism(Keen & Zidenberg-Cherr, 1996). While, excess Mn may inhibit Iron absorption, depressed growth, decreased appetite and altered neurological function (Ingersoll *et al.,* 1995). Furthermore, Mg is an extremely important and valuable mineral, because it is critical in over 300 enzymatic reactions in the human body. Excess Mg or deficiency can affect virtually every organ in the body; symptoms involving impaired contraction of smooth muscles; muscle tension including constipation; urinary spasms; difficulty of swallowing; neck pain; back aches; tension headache; anxiety, hyperactivity and restlessness with constant movement. In the present study, Mg concentration levels in edible muscles of studied organisms were below legal limit of WHO (1982) (150 ppm). While, the Environmental protection agency (2004) suggested 10 ppm of Mn concentration as a safe oral intake.

Zn is an essential trace element that is required for synthesis of DNA, RNA and protein and thus for cell division (Prasad, 1983). Furthermore, the Zn-Cd ratio is very important as Cd toxicity and storage are greatly increased with Zn deficiency. One illustration of the importance of Zn- Cd relationship is that the effects of Cd on several biological systems including the formation of tumors can be suppressed by the simultaneous injection of Zn (Abshire *et al*., 1996), but too much Zn is also harmful. Harmful effects generally begin at levels 10-15 times higher than the amount needed for good health. Large dose of Zn taken by mouth even for short time can cause stomach cramps, nausea and vomiting. Taken longer, it can cause anemia and decrease the levels of good cholesterol (ATSDR, 2005). The concentrations obtained for Zn in the current study are in harmony with the results of Soliman (2006) which recorded that higher concentrations of Zn were seen in *Sepia* (36.30ppm) and crab (29.912ppm) than in prawn samples (15.984ppm). However, the present study declared that the levels of Zn were below all these ranges. While, Ahdy *et al*. (2007) observed that Zn levels in edible muscles of *Sepia* and prawn are higher than that in crab from Alexandria region. Previously, Edema & Egborga (2001) measured Zn levels in prawn and crab from fresh water, where they observed bioaccumulation of this metal in their edible muscles. Moreover, Canli *et al.* (2001) recorded that concentration of Cu, Cd, Cr and Zn in edible muscles of prawns was relatively higher than fish from three stations on the Mediterranean Sea. Considering cephalopods, Miramand & Bently (1992) recorded Zn in *Sepia officcinalis* within general guideline limit for Zn in the food. The Zn concentrations found in the present study are not in concern; all studied samples contained Zn level below (50 ppm) recommended by several agencies and organizations (SASO, 1977; MAFF, 1995).

In the present study, it was observed that, the edible muscles of *Sepia* contained Hg, while it was not detected in the muscles of prawn and crab. Similar results were recorded by Al Sulami (2002) who did not detect Hg in any of the tissues of fish and prawn samples from Arabian Gulf. In contrast, Soliman (2006) recorded that the concentrations of Hg in edible muscles of prawn and crab collected from the Suez Canal were 0.01 and 0.54ppm respectively and its levels in *Sepia* and squid samples were 0.01 and 0.92 ppm respectively. Furthermore, Ahdy *et al.* (2007) observed the highest levels of Hg in prawn and crab than that in *Sepia* from Alexandria region. Hashmi *et al*. (2002); Hui *et al.* (2005); Celik *et al.* (2006) and Panutrakul *et al.*(2007) evaluated the accumulation of Hg in different species of prawn and crab in different regions, since Hg is one of the most important pollutants both because of its effect on marine organisms and it is potentially hazardous to humans. It is a toxic substance which has no known function in human biochemistry or physiology and does not occur naturally in living organisms. Methylmercury which is formed in aquatic sediments through the bacterial methylation of organic Hg, Methylmercury is toxic compound of Hg; nearly all the Hg in the muscles of aquatic organisms occurs as Methylmercury (Joiris *et al.*, 1999).This compound affects the kidneys and also the central nervous system particularly during development as it crosses the blood brain barrier and placenta (Clarkson, 2002). As the result of its toxicity, limits of total Hg levels have been established in various countries; the FAD has set a maximum level of Hg of 1ppm (US food, 2001). In Japan marine organisms containing total Hg concentration exceeding the Japanese maximum permitted limit of 0.3ppm are considered unsuitable for human consumption (Dickman & Leung, 1998). In Europe, the total Hg limit regulated by European commission is 0.5 ppm except for some species for which it is 1ppm (Anonymous, 1996). In Egypt, the total Hg limit regulated by Egyptian organization Standardization and Quality Control (1993) is 0.5ppm, while SASO (1977) suggested the guideline limit for Hg is 1ppm. On this basis, total Hg concentration in edible muscles of *Sepia* in the present study was below the regulatory limits.

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