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Heavy metals monitoring using commercially important crustaceans and mollusks collected from Egyptian and Saudi Arabia coasts

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Abstract: The objectives of this study was to determine and compare the concentration levels of five selective essential trace metals (Fe, Cu, Zn, Co and Se) and three non-essential metals (Hg, Pb and Cd) in the edible muscle tissues of commercially important species of male and female crustaceans (*Erugosquilla massavensis*, *Penaeus semisulcatus*, *Metapenaeus monoceros*, *Portunus pelagicus*) and mollusks (*Sepia spp*. And *Cardium edule*). It was carried out also to evaluate the bioaccumulation process of the elements based on the Metal Pollution Index (MPI) as an attempt to use these organisms as bioindicators of pollution of Mediterranean Sea, Red Sea and the Arabian gulf and to ensure the seafood safety from these regions. The obtained results revealed that significant variations of Fe, Cu, Zn, Co, Hg, Pb and Cd levels in edible muscles of crustacean and mollusk organisms were observed at different localities surveyed, as well as between sex of each species and environmental areas. The present data also, show that MPI factor of essential metals were higher than that of non-essential heavy metals in all studied organisms. Moreover, MPI values suggested that mantis shrimps *E. massavensis* and bivalve mollusks *C. edule* have a greater capacity for metal bioaccumulation than shrimps, crab and cephalopod mollusks, so *E. massavensis* and C. edule are more vulnerable to metal pollution than the other studied species. Therefore, it is suggested that *E. massavensis* and *C. edule* can be used as bioindicators of metal pollution.

Keywords: Essential, Metal Pollution Index, Non-Essential, Heavy Metals, Bioaccumulation, Edible Muscles, Crustaceans, Mollusks

1. Introduction

Coastal pollution has been increasing significantly over the recent years and found expanding environmental problems in many developing significant amount of trace metals into the marine environment, causing permanent disturbances in marine ecosystems, leading to environmental and ecological degradation and constitute a risk to a number of flora and fauna species including humans, through food chains [1]. Marine organisms require varying amounts of essential trace metals "heavy metals" and acquire them from ambient water such as: Fe which is the most abundant transition element and probably the most well known metal in biologic systems [2]. Zn and Cu are present within the aquatic environment at low level and are essential for some metabolism in living organisms at particular concentration [3]. Furthermore, Cu required by crustaceans species as an essential part of their oxygen-carrying pigment haemocyanin. Zn is vital component of many enzymes such as carbonic anhydrase, carboxy-peptidase and several dehydrogenases. Additionally, Co is readily absorbed from the gastro-intestinal tract and the surrounding water by fish and crustacean and other aquatic organisms. It is an essential trace mineral that is a constituent of vitamin B_{12} [4]. Moreover, Se is an essential trace element. It is an antioxidant which prevents damage of cells and inhibits chromosome damage and mutations. It is also known to participate in several important metabolic interactions with a variety of hazardous elements such as Hg, and Cd [5]. However, their importance for living organisms, the essential metals can also produce toxic effects when they accumulate and their level is excessively elevated in aquatic organisms. Furthermore, these toxic effects tend to be more complicated than that of the non-essential heavy elements [6]. These non-essential metals are exemplified by the Hg, Pb and Cd which are considered toxic elements even at relatively low concentrations that have no known vital or beneficial effect on organisms and their accumulation over time in the bodies of organisms can cause serious illness due to their competition with the essential metals for binding sites [7]. Hence, their concentration in the environment is of major importance, since the accumulation of such metals in the water and aquatic organisms has both direct and indirect consequences on both aquatic and human life [8, 9].

In aquatic ecosystem, heavy metals are considered as the most important pollutants, since they are present throughout the ecosystem [10, 11]. Many marine organisms have the potential to accumulate high levels of metals from their environment [12, 13], Pollution enters fin and shell fishes through five main routes: via food or non-food particles, gills, oral consumption of water and the skin [14, 15] i.e., 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 [16]. Bioaccumulation means an increase in metals concentration in biological organisms compared to their concentration in the environment [17]. Metals accumulation in living things anytime they are taken up and stored faster than they are broken down (metabolized or excreted). Bioaccumulation varies among organisms based on uptake, detoxification and outside environment. Moreover, bioaccumulation indicates the pollution level in organisms which live in polluted environments [18]. Hence, bioaccumulation of heavy metals in commercial marine organisms is of global importance. Continuous monitoring of aquatic animals is very important in polluted environment to evaluate the possible risk of human consumption [19]. Furthermore, the bioaccumulation studies led to adoption of the bioindicators concept. Seafood is widely used as bioindicators of marine pollution by metals [20, 21]. The use of marine organisms as bioindicators of metal pollution of aquatic environments and suitability for human use from toxicological point has been documented [9, 13-15, 22, 23] .

Over the last decades, there is an increasing concern regarding the roles and fates of trace metals in aquatic ecosystem of Mediterranean Sea, Red Sea and the Arabian gulf. Much of this concern arises from the low level of available information on the concentration of these metals within the environment and in marine organisms. So, the present paper aims to highlight the level of five selective essential trace metals (Fe, Cu, Zn, Co and Se) and three non-essential metals (Hg, Pb and Cd) in the edible muscle tissue of commercially important species of crustaceans (mantis shrimps, shrimps and crabs) and mollusks (cephalopods and bivalves) caught from these regions. According to many researchers, some shellfishes by virtue of their mobile nature are not fair indicator of aquatic contamination, but their regular consumption by human beings makes it absolutely necessary to monitor their different organs, particularly the muscles. The present study is therefore important not only from the safety point of view of human health, but also from the quality point of view as many of these shellfish species have high export value.

2. Material and Methods

2.1. Collection of Samples

Six crustaceans species (2 species of mantis shrimps species, 2 species of shrimps, 2 species of crabs and four mollusks species (2 species of cephalopods and 2 species of bivalves) were selected in this study and collected from :

A- Egyptian coasts (fishing ports of the Suez and Ismailia regions) that included males and females marine mantis shrimp *Erugosquilla massavensis*.

B- Saudi Arabia coasts that included the following species:

i- Crustaceans species: Both sexes of *Penaeus semisulcatus* (shrimps), and *Portunus pelagicus* (crabs). Additionally to mollusks species; cephalopod mollusks; *Sepia* spp. and bivalve mussel *Cardium edule* that were obtained from local fishermen at Jeddah as source of Red Sea samples.

ii- Both sexes of of *Metapenaeus monoceros* (shrimps), *P. pelagicus*, and mollusks (*Sepia* spp. and *C. edule*) were collected from local fishermen at Dammam that included the Arabian gulf strains .

The analyses were carried out on composite samples of 5 specimens of each species (in each crustaceans species, 5 males and 5 females were included in each analysis) having uniform size. This is a measure to reduce possible variations in metal concentrations due to size and age.

2.2. Separation of Muscle away from Exoskeleton

- Fresh whole bodies of all samples of crustaceans were stored at – 20 ° C to facilitate peeling process after thawing when needed as most crustaceans. While, internal shell of *Sepia* and the carapace of *C. edule* were removed from their edible muscles.

2.3. Edible Muscles Analysis for Estimation of Heavy Metal Contents

The tissues samples were dried in an oven for 6 hours at 105ºC and then burn in a muffle furnace for 16 hours at 550ºC till reduced to ash [24].

- The obtained white ash was digested by 1 ml of mixture of concentrated nitric acid (2/3) and concentrated perchloric acid (1/3; Merck). It was then dissolved in 1ml of 1:1000 dilute nitric acid and analyzed for heavy metals concentrations (Fe, Cu, Zn, Co, Se, Hg, Pb and Cd) by using Perkin Elmer analyst 100 atomic absorption Spectrophotometer.

2.4. Statistical Analysis

The results of the present work were analyzed statistically by using the Statistical Package for Social Science (SPSS version 15 package software). Data were expressed as Mean ± S.E Two way analysis of variance (ANOVA) was applied to illustrate the effect of region, sex and their interaction on the studied biochemical parameters. Means with the same letter for each parameters are not significantly different, otherwise they do (P<0.05). In order to compare the total content of heavy metals at different locations, the metal pollution index (MPI) was used [25, 26].

MPI = ($M_1 \times M_2 \times M_3 \ldots . \times M_n)^{1/n}$, where M_n is the concentration expressed in mg/kg of any investigated metals in the sample.

3. Results

3.1. Essential Heavy Metals Analysis

The mean concentrations of essential trace metals Fe, Cu, Zn , Co and Se in muscle tissues of commercially important crustaceans and mollusks species as mg/ kg are presented in Figures 1, 2, 3, 4, 5. In the present study, significant differences were observed in all determined essential heavy metal concentrations between sexes of crustaceans as well as among all studied species (p > 0.0001) except Se levels, where non-significant variations were recorded. The range of Fe varied from 44.33 to 158.57 mg/ kg, while that of Cu, Zn , Co and Se were from 0.98 to 188.58 mg/kg, 31.29 to 61.50 mg/kg, 0.09 to 16.70 mg/kg and 0.05 to 0.26 mg/kg respectively.

In crustaceans species, both sexes of *E. massavensis* accumulated the higher levels of Fe, Cu, Co and Se compared to that of males and females of crabs and shrimps species. Additionally, *E. massavensis* males had the highest levels of Zn, while their females accumulated the lowest level (29.70 mg/kg , 28.76 mg/kg from Suez and Ismailia respectively). Furthermore, the present results detected that all edible muscles of shrimps species accumulated higher concentrations of Fe, Cu, Co and Se except for shrimps species from the Arabian gulf that had lower mean values of Fe (48. 30 mg/kg , 57 mg/kg in males and females respectively) in comparison with crab species from two studied regions. On the other hand, in crab species, both sexes had higher Zn contents than in shrimps species.

Comparing heavy metal contents of the same crustaceans species with one another from different regions, the present data recorded that in mantis shrimps, both sexes from Suez region had higher levels of Fe and Co than that from Ismailia region. Furthermore, Cu accumulation had the higher concentration in edible muscles of females mantis shrimps from Suez (188.58 mg/ kg) followed by females from Ismailia (182.15 mg/kg), while lower level was observed in males from Suez (104.31 mg/ kg). While, no differences in mean values of Zn was detected between same sex from two studied regions. Regarding, Se contents, this essential heavy metal had the higher concentration in males mantis shrimps from Ismailia and females from Suez (o.25 and o.26 mg/kg respectively) but this increase was non-significant.

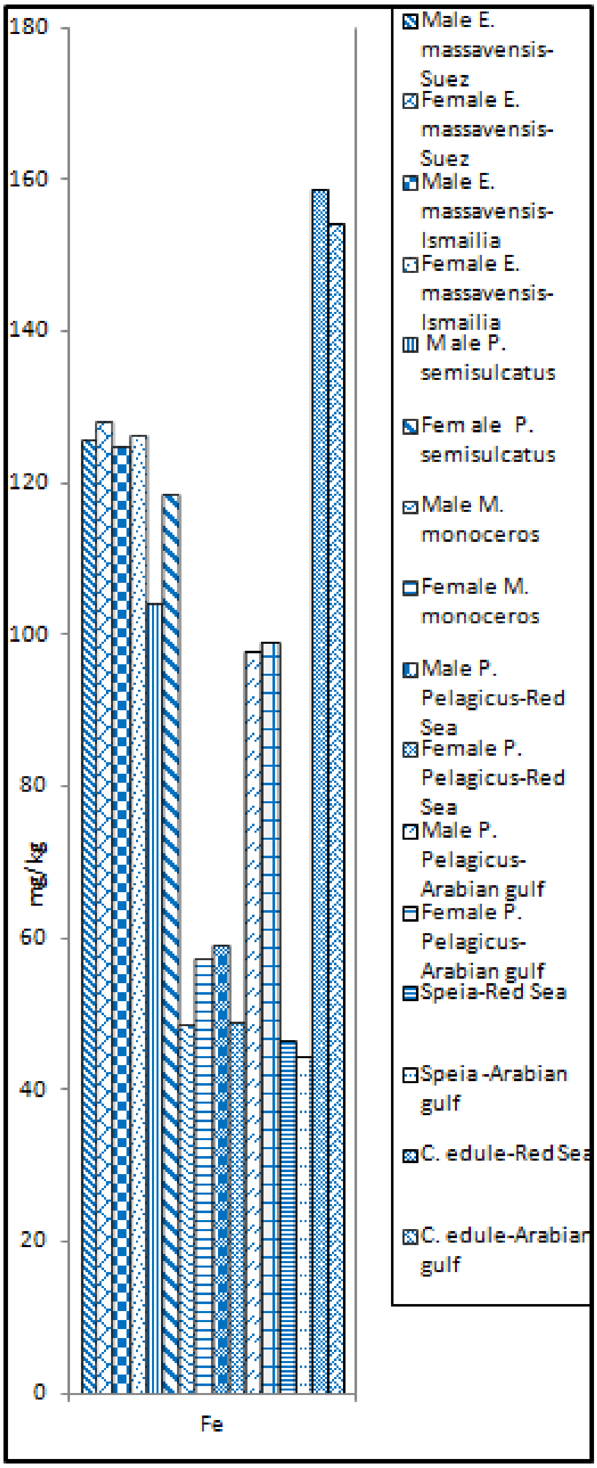

Fig 1. *Fe concentration in muscles of crustaceans and mollusks species.*

In shrimps species, both sexes of *P. semisulcatus* accumulated higher levels of Fe, Cu and Zn compared to *M. monoceros*. While, Co accumulation, the higher concentration was detected in males *M. monoceros* (8.15 mg/kg), however their females had lower level (3.83 mg/kg). Furthermore, Se showed higher concentrations in males *P. semisulcatus* and females *M. monoceros* and lower level was observed in males *M. monoceros*, however these differences statistically were non-significant. In crab species, both sexes of Arabian gulf strain had higher levels of Fe, while higher concentration of Cu was recorded in females from Red Sea and Arabian gulf (2.65 mg/kg and 2.35 mg / kg respectively) while lower level of Cu was observed in males from Red Sea (1.03 mg/kg). On the other hand, both sexes of crabs from Red Sea had the higher concentrations of Se, but this increase was non-significant as in shrimps species.

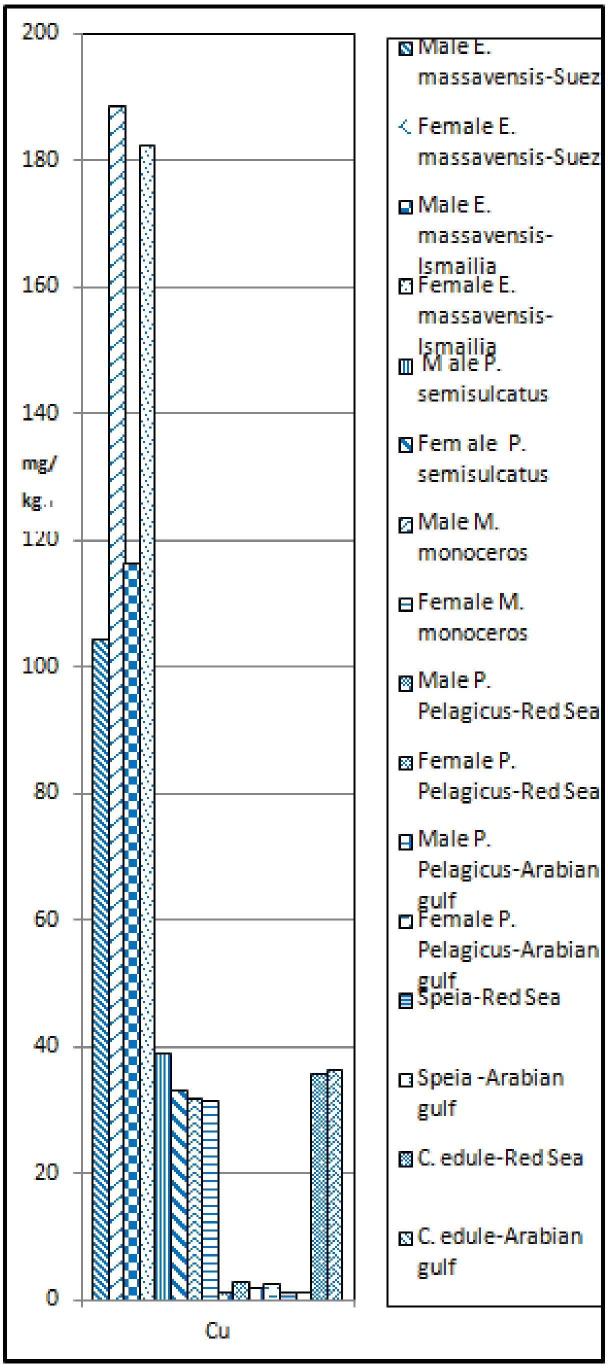

Fig 2. *Cu concentration in muscles of crustaceans and mollusks species.*

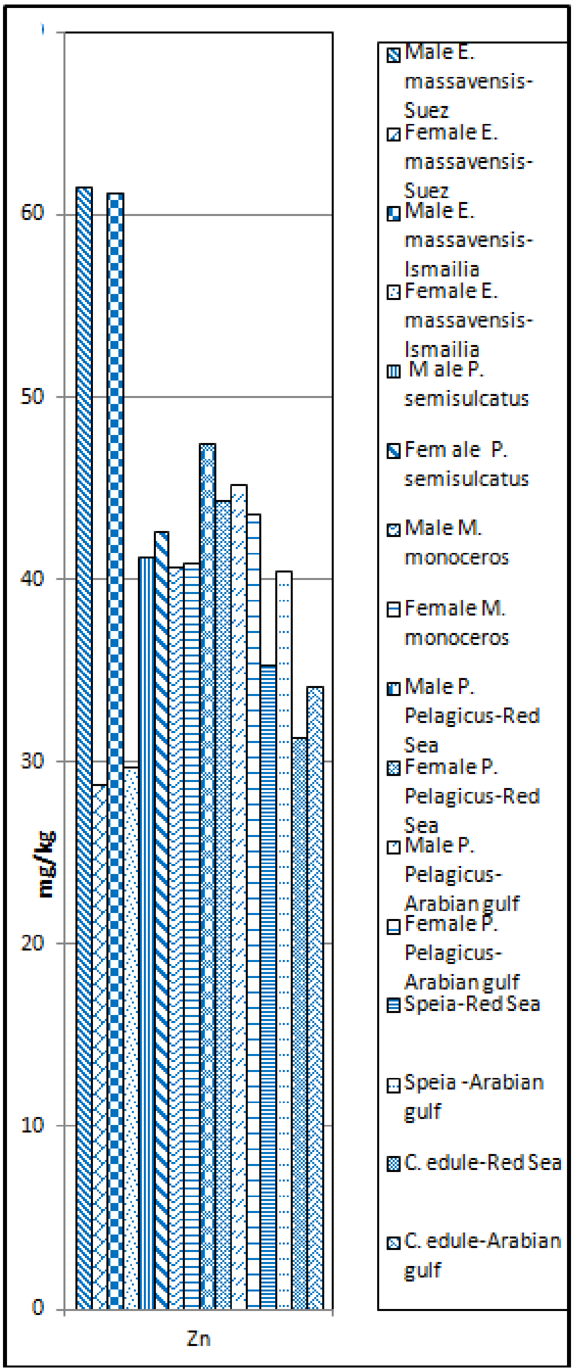

Fig 3. *Zn concentration in muscles of crustaceans and mollusks species.*

In mollusks species as shown in Figs. 1, 2, 3, 4, 5, edible muscle of cephalopods (*Sepia* spp.) had the lower levels of Fe, Cu, Co and Se than in bivalve mollusks (*C. edule*). On the other hand, cephalopods accumulated the higher level of Zn (35.26 mg/kg, 40.43 mg/kg from Red Sea and Arabian gulf respectively). As regards, in cephalopods species, *Sepia* from Red Sea had higher levels of Fe, Cu and Se and lower concentrations of Zn and Co in comparison with the same species from Arabian gulf. While, in bivalves species, edible muscles of *C. edule* from Red Sea accumulated higher levels of Fe and Co and lower mean values of Cu, Zn and Se than those from Arabian gulf.

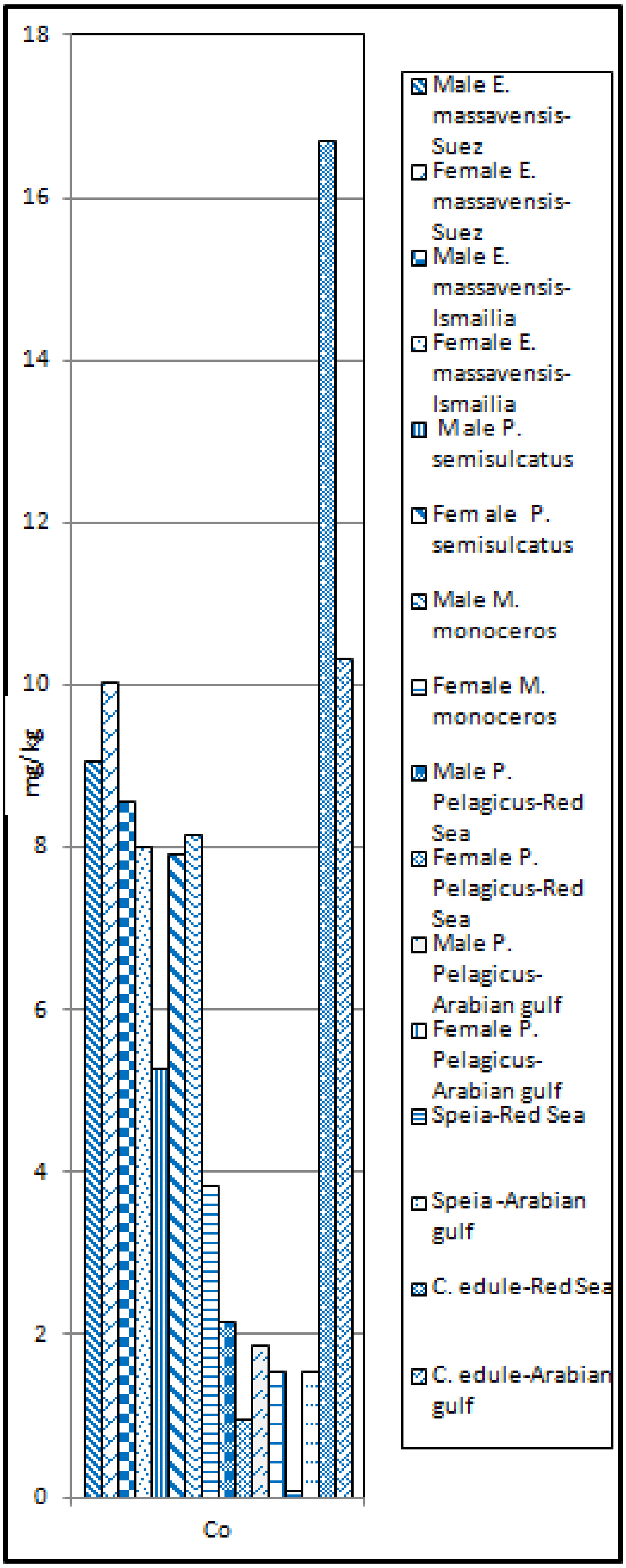

Fig 4. Co concentration in muscles of crustaceans and mollusks species.

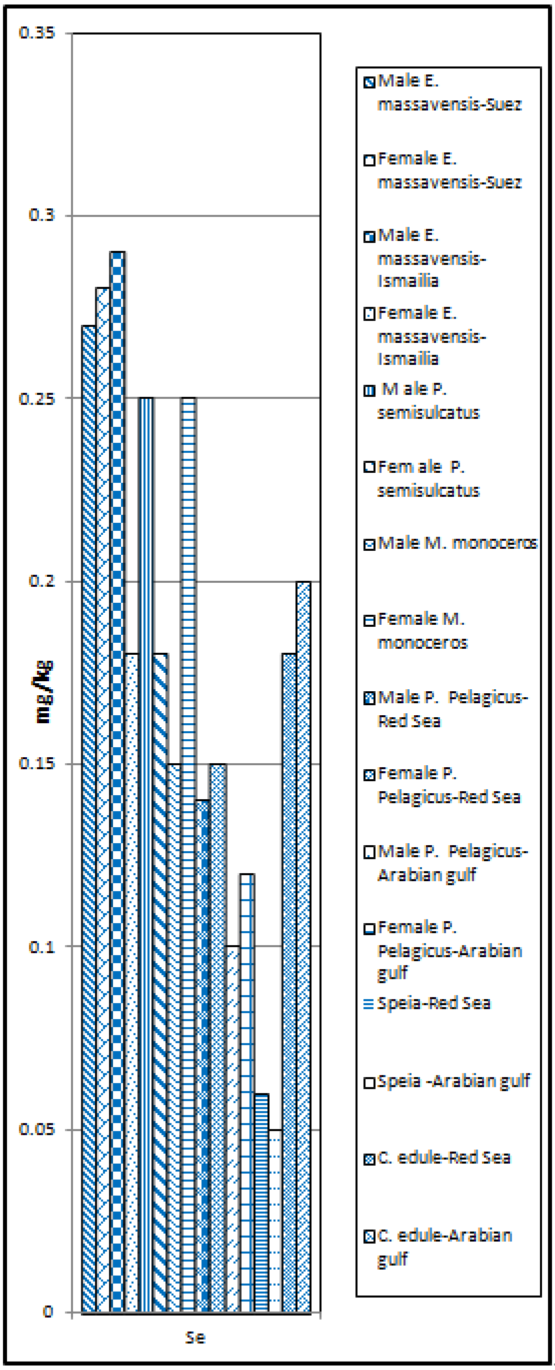

***Fig 5.** Se concentration in muscles of crustaceans and mollusks species.*

Comparing essential heavy metals accumulation in crustaceans and mollusks species in all studied regions, it was observed that Fe accumulated as per the order *C. edule* specie > both sexes of *E. massavensis* > both sexes of *P. semisulcatus* > both sexes of *P. pelagicus* > both sexes of *M. monoceros* > *Sepia* species **(**Fig 1)**.** Moreover, Cu accumulated in the highest concentrations in both sexes of *massavensis* > males *P. semisulcatus* > *C. edule* > females *P. semisulcatus* > both sexes of *M. monoceros* > both sexes of *P. pelagicus* > *Sepia* (Fig 2). In Fig 3 , Zn had this decreasing order males *E. massavensis* from two studied areas > both sexes of *P. pelagicus* > both sexes of *P. semisulcatus* > both sexes of *M. monoceros* > *Sepia* > *C. edule* > females *E. massavensis***.** Furthermore**,** Co accumulated as per the order *C. edule* > both sexes of studied species of *E. massavensis* > males *M. monoceros* > both sexes of *P. semisulcatus* > females *M. monoceros* > both sexes *P. pelagicus* > *Sepia* (Fig 4). Se had this order, where non-significant variation was detected both sexes of *E. massavensis* species > males of *P. semisulcatus* > females *M. monoceros* > females *P. semisulcatus* > *C. edule* species > both sexes of *P. pelagicus* > *Sepia* (Fig 5).

3.2. Non-essential Heavy Metals Analysis

Regarding non-essential heavy metals contents, Hg, Pb and Cd in muscle tissues of crustaceans and mollusks species are presented in Figs 6, 7, 8. The present results showed that the levels of non-essential heavy metals showed variations among species, as well as between sex of each species and environmental areas. The mean values of Hg ranged from 1.65 mg/kg to 4.91 mg/kg. Furthermore, that of Pb and Cd were 3.49 mg/kg to 11.49 mg/kg and 0.14 mg/kg to 0.87 mg/kg respectively. In crustaceans species, the concentrations of Hg and Pb in the abdominal muscles of both sexes of *E. massavensis* from two studied areas had the highest mean values comparable to their values in crabs and shrimps species. Furthermore, males mantis shrimps from Suez accumulated the highest Cd contents in comparison with all studied crustaceans (0.58 mg/kg).

While, the lowest mean values of Hg was detected in females *M. monoceros* (1.65 mg/kg). Also, both sexes of *M. monoceros* exhibited the minimum mean values of Pb (3.79 and 3.49 mg/ kg for males and females respectively). Furthermore, the present data recorded that edible muscles of all studied crab species accumulated Pb higher than that in shrimps species. Conversely, all shrimp samples had higher mean values of Cd than crab species except for females *P. semisulcatus* where lower Cd contents were recorded (0.270 mg/ kg). Moreover, the present data showed that all studied male crustaceans accumulated higher levels of determined non-essential trace metals in their muscles than their females except for crab species whose males had lower Cd than their females. According the studied localities, males and female mantis shrimps, from Suez region accumulated higher levels of studied non-essential heavy metals than those from Ismailia region. Additionally, both sexes of shrimps species from Red Sea had higher contents of Hg and Pb than those from Arabian gulf. Moreover, males shrimps from Red Sea accumulated more Cd than males and females of Arabian gulf, but, their females had lower content of Cd (0.230 mg/ kg). In crab species, both sexes from Red Sea had higher levels of Pb and lower level of Hg than those from Arabian gulf. While, non-significant variation was detected for Cd content between two studied region.

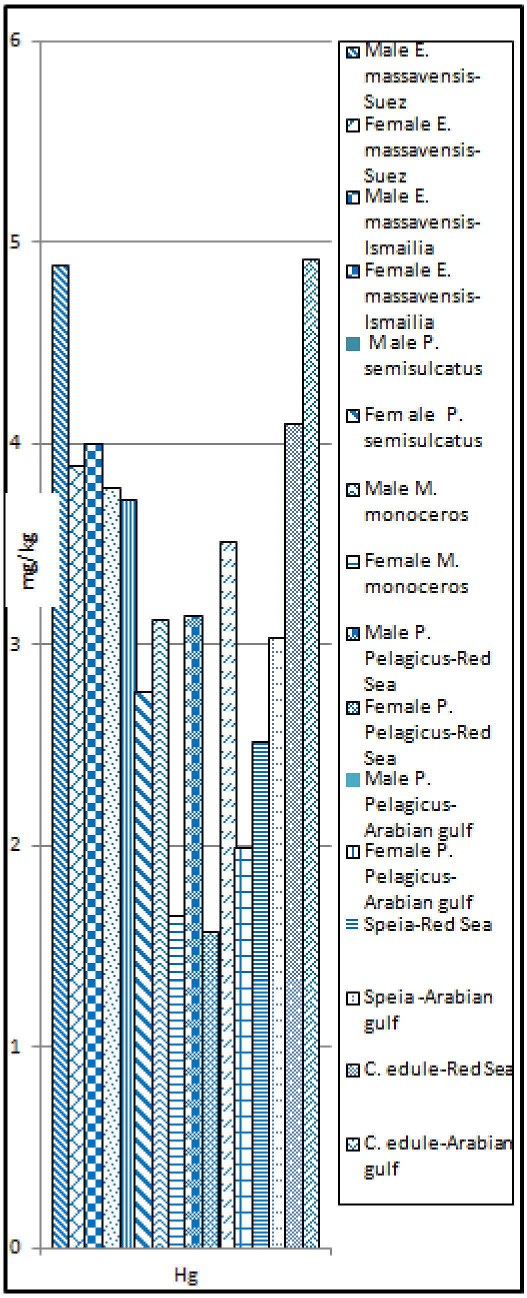

Fig 6. *Hg concentration in muscles of crustaceans and mollusks species.*

In mollusk organisms, the bivalves bioaccumulated the highest concentrations of non-essential trace metals as compared with their concentrations in cephalopod mollusks. Furthermore, in edible muscles of cephalopod species from Red Sea less Hg level was detected than Arabian gulf. However, non-significant differences were recorded in Pb and Cd contents between studied regions. Regarding, bivalves species, *C. edule* from Arabian gulf had higher contents of all determined non-essential heavy metals than that from Red Sea. Comparable to crustacean organisms, Fig 6 illustrated that Hg accumulation had this descending arrangement *C. edule* from Arabian gulf > males *E. massavensis* from Suez > *C. edule* from Red Sea> males *E. massavensis* from Ismailia > females *E. massavensis*>

males *P. semisulcatus* > males *P. pelagicus* > males *M. Monoceros* > *Sepia* from Arabian gulf > females *P. semisulcatus* > *Sepia* from Red Sea > females *P. pelagicus*> females *M. monoceros*. Moreover, Pb accumulation as per the order both females *P. pelagicus* > males *P. pelagicus* and females *E massavensis* from Ismailia> females *P. semisulcatus* > *Sepia*. > *M. monoceros* (Fig 7). While, Cd accumulated in all studied samples according to the following order: *C. edule* > males *E. massavensis* from Suez > males *P. semisulcatus* > males *E. massavensis* from Ismailia > females *E. massavensis* from Suez > both sexes *M. monoceros* > all females *P. pelagicus* > males *P. pelagicus* and females *E. massavensis* from Ismailia> females *P.semisculcatus* > *Sepia*

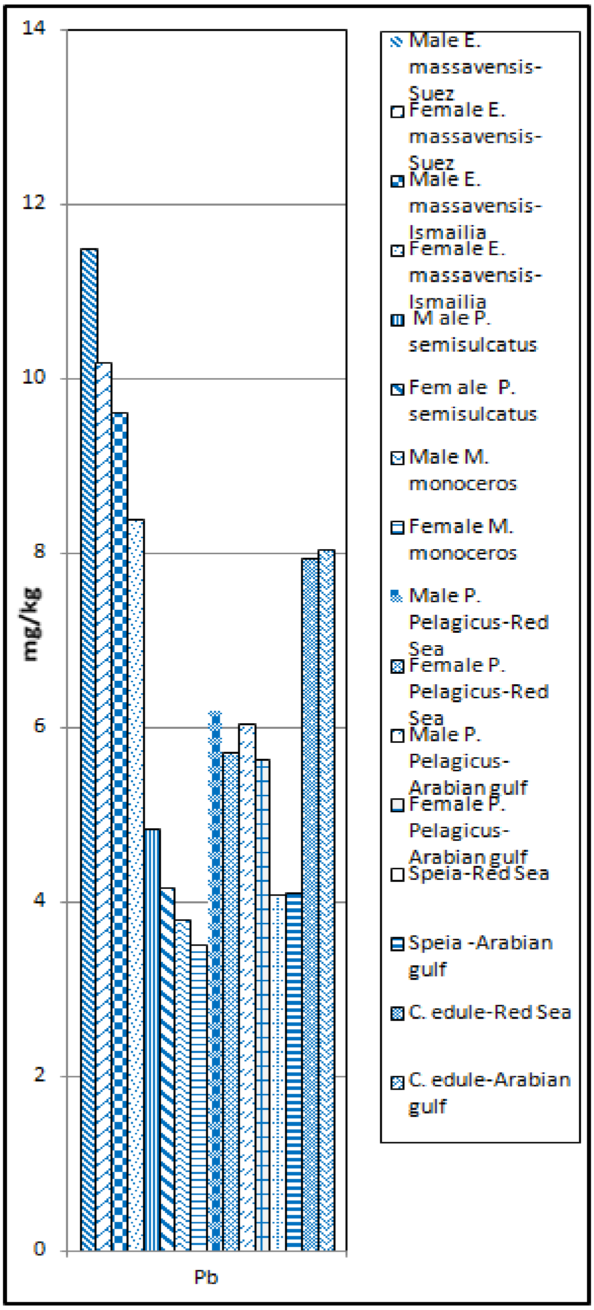

Fig 7. *Pb concentration in muscles of crustaceans and mollusks specie.*

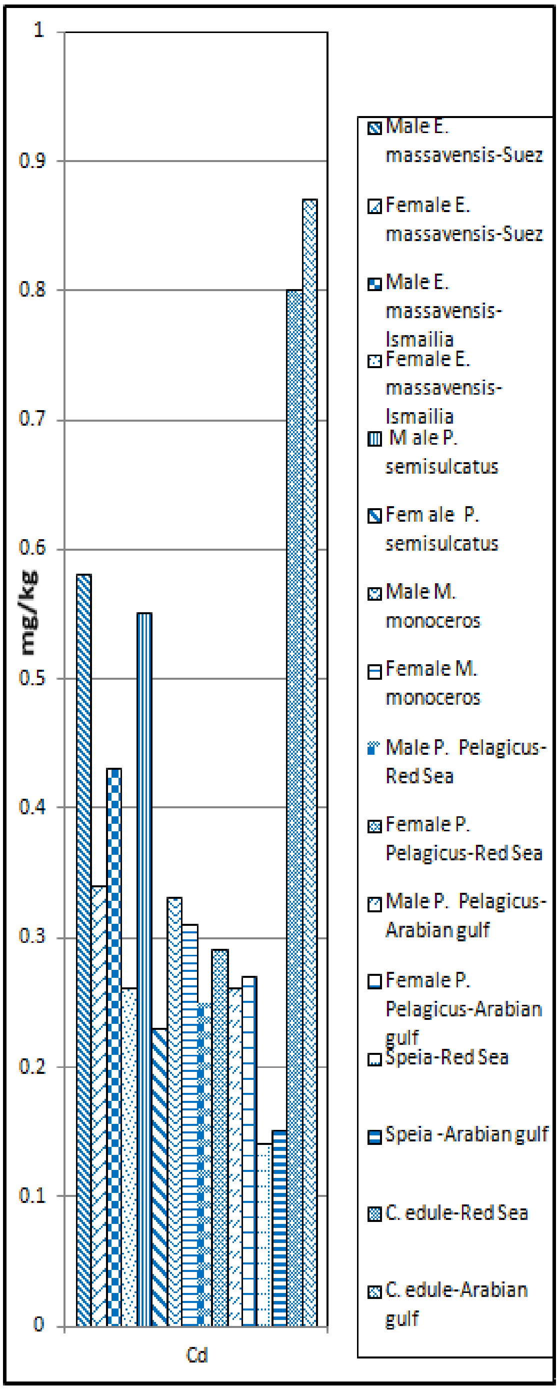

Fig 8. *Cd concentration in muscles of crustaceans and mollusks species.*

To compare the total metal content in the different sampling sites investigated in this study, the metal pollution index (MIP) was used. From Fig. 9 A & B, it is clear that MPI in essential heavy metals recorded higher level in all mantis shrimps followed by bivalve mollusks, followed by shrimps species and the lower content occurred in crabs species followed by cephalopod species. Furthermore, the present data showed that males mantis shrimps had higher MPI of essential and non-essential heavy metal than that of females. Regarding, shrimps species and bivalve mollusks from Red sea, they had higher MPI for all determined metals than in the same species from Arabian species.

While, crab species and cephalopod mollusks from Arabian gulf showed higher MPI than species from Red sea. Furthermore the data detected that, MPI of essential metals were higher than that of non-essential heavy metals in all studied organisms. Furthermore, MPI of non-essential heavy metals had this decreasing order bivalve mollusks > mantis shrimps > male shrimps from Red Sea > male crabs from studied regions> male shrimps from Arabian gulf > female crabs from studied localities > female shrimps from studied regions > cephalopod mollusks.

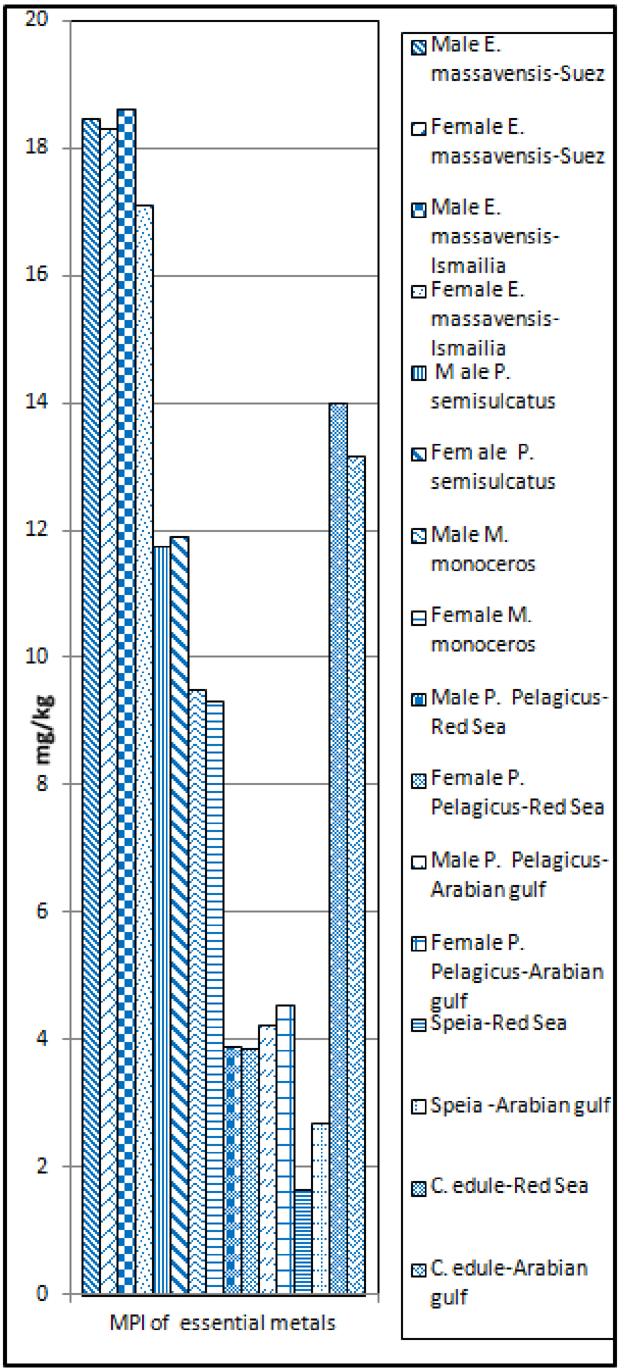

Fig 9. A. *Metal pollution index of heavy metals in studied organisms (MPI=(* $M_{1} \times M_{2} \times M_{3} \times \ldots \times M_{n})^{1/n}$ *,where* M_n *is the concentration expressed in mg/kg of any investigated metals [25, 26]. A- MPI of essential heavy metals.*

4. Discussion

Information regarding bioaccumulation levels of heavy metals in marine organisms is very important, with many implications in various domains, like environment protection, public health, control of standards compliance or risk assessment. Taking into account the ability of marine biota to accumulate metals from their environment (seawater, sediments, food), their utilization as marine pollution bioindicators has been confirmed by numerous examples [9, 13,15, 27]. Furthermore, several organizations have pointed out the need for monitoring heavy metals concentrations in the aquatic environment [28] because of their toxicity, persistence and accumulation in the biota. So, on this background the present study aims to investigate the accumulative capacity of important commercially crustaceans and mollusks with the heavy metals Fe, Cu, Zn, Co, Se , Hg, Pb and Cd. In particular, the present study is concerned with the determination of the residual heavy metal concentrations in the edible muscles of these organism to ensure their safety for human consumption. Additionally, to investigate the ability to use these marine organisms as bioindicators of water pollution especially, the selective marine organisms in this study caught in the Suez canal, Red Sea and Arabian gulf. Furthermore, the present study is therefore important not only from the safety point of view of human health, but also from the quality point of view as these selected organisms have high export value.

The present study has shown that significant spatial variations in metal contents among species and between sex of each species were observed. These variations are presumably due to individual samples being of different size categories, from different ecological niches, and from different trophic levels. Possibly, species also have different metabolic requirements for specific trace element [15, 29, 30].

An important aspect is the wide variety of factors that influence metal bioaccumulation, like type of food hydrochemistry conditions, metal bioavailability, genetic differences, physiological state [31]. These factors determine variations of metal concentration that can sometimes mask the organism responses to temporal or spatial gradient of pollution. Furthermore, ecological needs, sex, size, seasonal changes and moult of marine animals changes were also found to affect metal accumulation in their tissues, Moulting has often been considered as one of the main excretory mechanisms of crustaceans since large amounts of metals may be lost with the moulted carapace [22]. Moreover, many laboratory and field studies reported that metal accumulation in tissues of organisms depends on metal concentration in the water and also the exposure period, Physio-chemical parameters such as the temperature, pH, salinity and hardness of the water play a crucial role in heavy metal accumulation [9, 23]. Bioaccumulation varies among organisms based on uptake, detoxification and outside environment [18]. In the present study, the variation in metal bioaccumulation in studied crustacean and mollusk organisms might be due to variation of species, sex and species location.

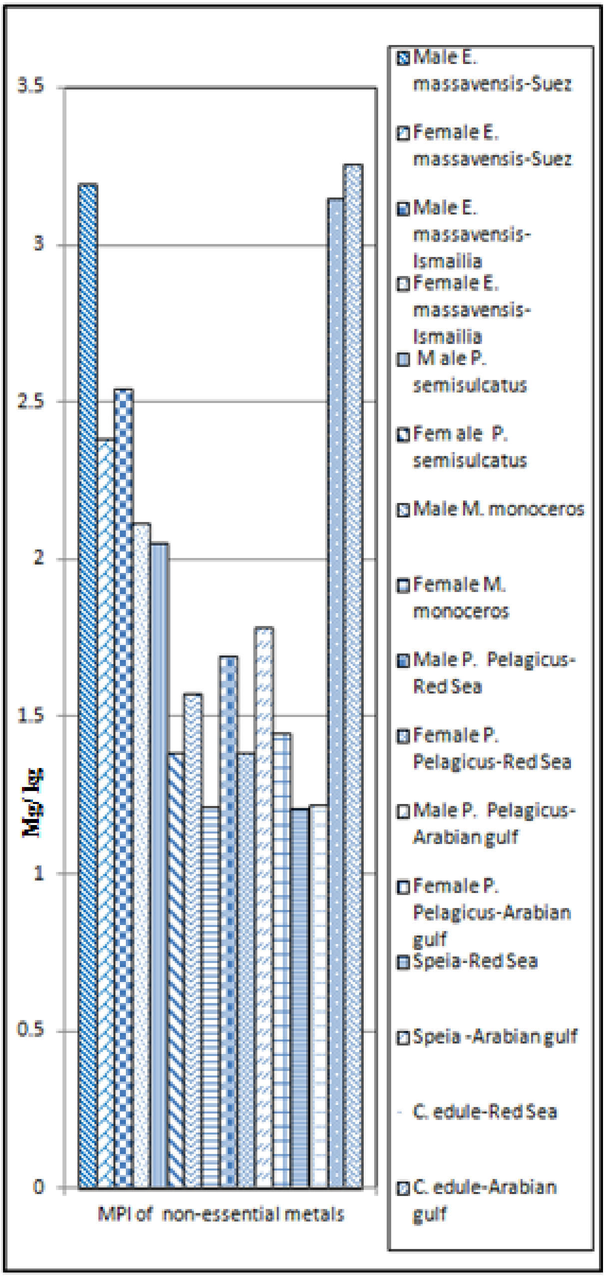

Fig 9. B. *Metal pollution index of non- essential heavy metals in studied organisms.*

Looking to the residual heavy metals in edible muscles of crustacean and mollusk organisms in the present work. It was clear that higher levels of Fe were found in bivalve mollusks followed by mantis shrimps followed by crabs and shrimps. While the lowest contents were detected in cephalopod mollusks. The present results coincides with the results of [27] which recorded that edible muscles of males *E. massavensis* accumulated the highest level of Fe followed by males *M. monoceros* and males *P. pelagicus* from the Egyptian Mediterranean coast off Port Said. However, the recorded values in their studies were higher than that of the present work. This might be attributed to variation in ecological niches where marine organisms found. Moreover, the present study is in agreement of study of [32] which recorded that bivalve mollusks accumulated higher level of Fe in their edible muscles. On the other hand, the present data are not in agreement with the results of [26] which reported this decreasing order of Fe level: cephalopod mollusks > bivalve mollusks > crab> shrimps.

Regarding to Cu, the present data show that *E. massavensis* accumulated the highest content of Cu in their edible muscles followed by *P. semisulcatus* shrimps, followed by *C. edule* and *M. monoceros* while the lowest levels were observed in crabs followed by *Sepia*. The present result are in agreement with the results of [15] which showed that *P. semisulcatus* accumulated the highest level of Cu compared to other shrimps species (*P. indicus, P. monodon, P. marguensis* and *M. brevicornis*). Similarly, the present data coincide with [9] who reported that *P. pelagicus* does not appear to accumulate excessively high concentrations of Cu in its edible tissues. On the other hand, [33] reported higher concentration of Cu in edible muscles of *P. pelagicus* from Kuwait coast. Moreover, the bioaccumulation of this trace metals exhibited the highest concentrations in muscles of *Sepia* (9.7 µg/g) followed by crabs (3.4 µg/g) followed by shrimps (3.1 µg/g) and lowest levels in bivalve mollusks (1.3 µg/g) as reported by [26].

Furthermore, from the present study, it is detected that all studied males of mantis shrimps had the highest Zn concentrations, while their females accumulated the lowest levels compared with other crustacean and mollusk organisms. It was observed that the mean values of Zn in males and females *E. massavensis* confirms the recorded values by [27]. Additionally, the present results detected that crabs species accumulated higher content of Zn followed by shrimps species followed by cephalopod and bivalve mollusks. The elevation of Zn in edible muscles of *P. pelagicus* wa recorded also by [9, 34]. Furthermore, the present study is in close agreement with [9] who recorded that Zn levels in muscle tissues of *P. pelagicus* were higher than Cu concentrations. Regarding Zn levels in other studied species in the present work, it was observed that in mantis shrimps and bivalve mollusks Cu pared to concentration of Zn, conversely, in shrimps and cephalopod mollusks Zn were relatively higher compared to concentration of Cu. This similar findings were also recorded in fishes [35, 36] and crustaceans [15, 36, 37] caught from other waters of the world. It is generally believed that marine organisms are actively regulate Zn concentrations in their tissues and that therefore, Zn tissue levels do not reflect the changes in Zn concentrations in the environment [38]. Previously, [39] stated that all the crustaceans require Cu and Zn for their biological functions and are capable of regulating internal concentrations up to thresholds above which regulations break down and net accumulation of the metals occurs. In general, these essential metals appear to diffuse passively probably as a soluble complex by the gradients created through adsorption of membrane surface [40].

Additionally, the present data show that the contents of Co had the following decreasing order bivalves > mantis shrimps > shrimps > crabs > cephalopods. These results confirm the findings of [40] who reported that crustaceans concentrated Co from the surrounding medium, but this ability varied among species. Moreover, [43] recorded higher amount of Co in edible muscles of bivalve mollusks and they attributed this increase to its richness in the surrounding niche. In contrast, the present study disagree with the results of [27] who demonstrated that Mediterranean Sea strain of females *P. pelagicus* crab had the higher content of Co in their muscles than *M. Monoceros* shrimps and *E. massavensis*. This variation may attributed to variation between species of crustaceans, mode of feeding of these aquatic organisms and percentages of Co in the surrounding medium.

Non-significant variations in Se concentrations among crustaceans and mollusks species were observed in the present study. In comparison to Se concentrations in marine organisms, [43] estimated concentrations of Se in small and large shrimp ranged from 0.14 - 0.16 and 0.19- 0.23 µg/ g wet weight respectively. These values were lower than that which recorded in this study. While, [26] recorded that the level of Se in *Penaeu*s *spp.* ranged from (1.0-2.7 µg/ g); (1.7- 4.3 µg/ g) in crab (*Portanus spp*.) and (0.9-1. 7 µg/ g) in *Sepia spp.* from Mediterranean Sea. Moreover, previously, [44] explained the lower content of Se in edible muscles of crustaceans and other marine organisms as the result of association of Se with free amino-acids or protein residues and was not present as characterizable inorganic Se species which indicate that Se is probably only incorporated into biota for specific biochemical purposes with any excess Se being excreted or eliminated. The elevation of essential heavy metals in some marine species is attributed to marine organisms accumulate some metals in direct proportion to the increase in the bioavailability from water and food chains. Many decapods can regulate their tissue and body burdens of heavy metals effectively [9].

Regarding to bioaccumulation of non-essential heavy metals, the present data detect that the distribution of non-essential metals in edible muscles of all studied species followed the order Pb > Hg > Cd. Furthermore, mantis shrimps accumulated the highest Pb in their muscles compared to that of all studied organisms followed by bivalve mollusks. The present results are in agreement with [27] in that *E. massavensis* are the most vulnerable to non-essential trace metals comparable to *M. monoceros* and *P. pelagicus*. Additionally, in the present work , it was found that bivalve mollusks had the highest levels of Hg and Cd. While, the lowest levels of Hg were recorded in females Red Sea *P. pelagicus* strain followed by females *M. monoceros* followed by *Sepia spp*.. This present findings coincide with the results of [45] who recorded that the concentrations of Hg in edible muscles of crab collected from the Suez Canal were higher than that in prawn and *Sepia* samples. However, [46] did not detect Hg in any of the tissues of fish and prawn samples from Arabian gulf. Moreover, in the present study both sexes of Arabian gulf shrimps *M. monoceros* exhibited the lowest level of Pb and *Sepia* had the lowest content of Cd in their edible muscles. These results are not in agreement of the results of [26] which showed that the bioaccumulation of these trace metals; non-essential heavy metals (Hg, Pb and Cd) exhibited this order cephalopods mollusks > bivalve mollusks > crabs > shrimps.

In comparison to the accumulative capacity of essential and non-essential trace metals in edible muscles of present studied species, it was observed that levels of Fe, Cu and Zn have been found to be higher than Hg, Pb and Cd in edible muscles of studied species .That could be explained because of these essential metals play a role in the enzymatic and respiratory processes of in aquatic animals [15, 26, 47] and relatively high level of these metals is necessary to carry out these biological functions. In addition to, these essential metals appear to diffuse passively probably as a soluble complex by the gradients created through adsorption of membrane surface [40]. In comparison with bioaccumulation of Fe, Cu, Co, Hg, Pb and Cd in carapaces of these studied species in the previous study [21] it was detected that carapaces of determined crustaceans and mollusks species had higher contents of essential and non-essential heavy metals than in their edible muscles. From these results, it is suggested that shell of marine organisms may be behave as an indicator of changes in environmental pollution, presenting a lower variability compared with soft tissue and providing a historical record of metal content in the body throughout its life cycle as attributed by [48]. This in turn suggest the tendency of shells of crustaceans and mollusks to detoxification mechanism of heavy metals as a mean of protection.

Comparing the present data with guidelines and limits as recommended by [49], the levels of essential trace metals Fe exceeds the maximum limit in all studied organisms. Similarly, it was found that Cu concentration in muscles of all studied samples were above the allowable level except in crab and cephalopods species. While, total Zn concentrations were above the regulatory limits in all males samples of mantis shrimps. Unlike, Fe, Zn and Cu metals, the present study show that the accumulation of Co ,Se and Hg elements in edible muscles of crustaceans and mollusks were below the maximum limit. On the other hand, the levels of Pb exceeded the permissible limit in both sexes of mantis shrimps and crabs as well as all studied bivalve mollusks. Similarly, both males of mantis shrimps and shrimps from Red Sea, besides all studied samples of bivalve mollusks had Cd levels in their edible muscles above the guide limits of Cd.

According to MPI (Fig 9) which compare the total metal content of essential and non-essential trace metals in the different sampling sites investigated in this study, it is clear that *E. massavensis* and *C. edule* had the highest MPI in essential heavy metals and non-essential heavy metals respectively, while the lowest values of MPI in essential and nonessential trace metals were recorded in all *Sepia* samples. Furthermore, it was observed that MPI of essential metals were higher than that of non-essential heavy metals this confirms the findings of [26]. However, the present results are not in agreement with the results of [26] in that higher MPI were recorded in cephalopod mollusks and followed by crustaceans (crabs and shrimps) followed by bivalve mollusks. The suggestion in the present study that *E. massavensis* and *C. edule* are more vulnerable to metal pollution than the other studied species may be attributed to their exposure to metals not only from the sediment or from surrounding water, but also through prey consumption which in turn bioaccumulation of heavy metals in their tissues [31]. Mantis shrimps are known to be obligate carnivores, feeding on live prey [50]. While mollusks as filter feeder organisms, are most frequently used to monitor the pollution of coastal water by metals [51]. Lying in the second trophic level in the aquatic ecosystem, mollusks have long been known to accumulate both essential and non-essential trace elements in aquatic ecosystems [52]. In addition to, mode of feeding, the present results may confirm that differences in ecological niches result in variations in metal bioaccumulation in edible muscles of marine organisms as recorded by the differences in accumulation levels of determined metals in *E. massavensis* in Suez and Ismailia regions, also in shrimps and crab species and mollusks species from Red Sea and Arabian gulf.

5. Conclusion

With regards to the objectives of this research and the results obtained, this current study has provides useful information and a baseline for future along with continuous studies on the heavy metals concentrations in crustaceans (mantis shrimps, shrimps and crab species) and mollusks (cephalopods and bivalve species). The present study is important not only from the human health point of view, but it also presents a comparative account of heavy metals in edible aquatic organisms from two different coasts (Egypt and Saudi Arabia) that are physico-chemically different. The knowledge of heavy metal concentrations in edible crustacean and mollusk organisms are very important with respect to nature management, human consumption of these species and to determine the most useful biomonitor species and the most polluted area.

The present study has shown that significant spatial variations in heavy metal contents among species and between sex of each species were observed these variations in metal bioaccumulation in studied crustaceans and mollusks organisms might be due to variation of species, sex and species location. Furthermore, the present data revealed that mantis shrimps and bivalve mollusks are more vulnerable to metal pollution in comparison to other studied species (shrimps, crabs and cephalopods) where higher MPI of essential and non-essential heavy metals were recorded in *E. massavensis* and *C. edule* respectively. On the other, *Sepia* (cephalopod mollusk) accumulated the lowest levels of essential and non-essential heavy metals. So, therefore, mantis shrimps and bivalve crustaceans can be used as bioindicator of water pollution. Moreover, the collection of such crustaceans and mollusks for human consumption should restricted to clean aquatic localities whether these are natural water bodies or aquaculture. The high concentrations of heavy metals in commercially important crustaceans and mollusks sampled from these coasts are a cause of concern, and requires regular monitoring of water quality in combination with the fact that commercially marine organisms consumption is the main source of heavy metal intake in people not occupationally exposed, amplifies the need for preventive measures to safeguard public health. Moreover, further studies on the mode of action and characterization of the active components in these crustaceans and mollusks species especially, crab species and *Sepia* spp. must be done to open the door in the future to use these species as specific health foods (functional supplements as antioxidant agents) and to play an important role in some pharmaceutical industries.

For future studies it is recommended that sampling of organisms should be done during different seasonal effects as to study on its potential on the heavy metals distribution in studied localities. Biological samples as well as water and sediment samples should be collected all together in an area that is polluted and other areas where the level of pollution is variable. This is to enable the observation and interpretation on absorption of metals and compare them with concentration of pollutants at different locations. Besides, the potential of these two biological samples as a pollution indicator can be evaluated.

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