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Trace metals in fish of economic interest from the west of Alexandria, Egypt

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Trace metals in fish of economic interest from the west of Alexandria, Egypt

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The concentration of some heavy metals (Cd, Cu, Fe, Mn, Ni, Pb, and Zn) in the muscle, liver, and gills in eight fish species, *Caranx crysos, Euthynnus alleferatus, Scomberomorus commerson, Sphyraena viridensis, Sargus sargus, Siganus rivulatus, Mugil* species, and *Sardinella aurita* were collected seasonally from the Mediterranean Sea in the region of Alexandria. The highest concentrations of Cd, Cu, Fe, and Zn were measured in liver tissue, while gill tissue yielded the highest concentrations of Mn, Ni, and Pb. Muscle is the organ of poor accumulation factor for all metals under investigation. Concentration of cadmium in muscle in *Mugil* species exceeds the permissible limit in summer, while *Siganus rivulatus* exceeds it in the summer and autumn seasons. On the other hand, copper, nickel, lead, and zinc are still much lower than the permissible levels. The metal pollution index (MPI) for metals was studied, revealing that *Siganus rivulatus, Mugil* species and *Sardinella aurita* had the highest MPI. Provisional tolerable daily intake (PTDI) indicates that the concentration levels of Cd, Cu, Fe, Mn, Ni, Pb, and Zn in the muscle of all fish species under investigation are much lower than recommended PTDI values, and accordingly there is no risk for the human consumption of these fish species.

Keywords: heavy metals; fish; metal pollution index (MPI); El-Mex Bay, Alexandria, Egypt

1. Introduction

There has been growing interest in determining heavy metal levels in the marine environment, and attention has been drawn to the measurement of contamination levels in public food supplies, particularly fish [1–3]. Rapid industrialisation and the discharge of potentially toxic trace metals into aquatic ecosystems have become a serious threat because of their toxicity, long persistence, bioaccumulation, and bio-magnification in the food chain [4–6]. Fish, as consumers, accumulate trace metals from the environment and therefore have been extensively used in marine pollution monitoring programmes [7]. Trace metals can be accumulated by fish, both through the food chain and water [8]. Heavy metals like copper, zinc and iron are essential for fish metabolism, while some others such as mercury, cadmium and lead have no known role in biological systems. For the normal fish metabolism, essential metals must be taken up from water, sediment or food. However, similar to the route of essential metals, non essential ones are also taken up by fish and accumulate in their tissues [9].

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Alexandria is the second largest city in Egypt and the main summer resort lying on the southern coast of the Mediterranean Sea, between $29^{\circ}49'$ and $30^{\circ}03'$ east longitude, and $31^{\circ}09'$ and $31^{\circ}20'$ north latitude. The area of study, which extended from El-Mex Bay to Marbilla, is a multi-polluted area (Figure 1). The bay has a mean depth of 10 m and extends about 15 km; its surface area is about 19.4 km^2 , and its volume $190.3 \times 10^6 \text{ m}^3$. The shoreline of El-Mex Bay is rocky, with narrow sandy beaches. The currents along the Egyptian Mediterranean coast follow the North African Current from west to east. The long-shore current is also eastward. The cross-shore current has no uniform regime and is controlled by the shore configuration. The current regime inside El-Mex Bay and in front of El-Nobaria Drain is controlled by the discharge from the drains. The wind system over that area is N to NW winds for most of the year.

The investigated area receives a heavy load of wastewater both directly from industrial outfalls and indirectly from Lake Mariut via the El-Mex Pumping Station. It lies about 1 km upstream of El-Umum Drain canal, pumping about 2.6 million m³/year [11]. This is mainly agricultural drainage water collected by El-Umum Drain, and comprises the overflow from Lake Mariut. Lake Mariut receives wastewater from the four sources in its eastern section, consisting of domestic, industrial and agricultural wastes. This liquid waste fills the lake and overflows into the El-Umum Drain and is discharged into the sea via the El-Mex Pumping Station. The El-Mex district is an industrial zone west of Alexandria city. As a consequence of growing heavy industries (chlor-alkali plant, petrochemicals, pulp, metal plating, industrial dyes, and textiles) and uncontrolled disposal of resulting wastes, the coastal waters of El-Mex Bay also receive huge amounts of untreated industrial waste.

The Mediterranean Sea, at kilometre 21 of the Alexandria-Matrouh Highway, receives drainage water, industrial and domestic wastes through the west Naubaria Canal. At the west of kilometre 21, there are three tourist villages on the coast, which may directly affect the environment with the discharge of domestic wastes and the construction of buildings in the main areas along the shoreline of the area, which consists of fine sandy beaches [11].

Because the metal pollution in aquatic environments can be harmful to human health, it is necessary to understand and control the hazard levels of pollution in seafood. Therefore, this study aimed to determine the levels of Cd, Cu, Fe, Mn, Ni, Pb, and Zn in the muscle, liver and gill tissues of different fish species from the western part of Alexandria along the Mediterranean Sea



Figure 1. Map of Alexandria, Egypt.

from spring 2005 to winter 2006, and to assess the public health risks associated with consuming fish harvested from this area by estimating daily and weekly intakes and by comparing them with the provisional tolerable weekly intake (PTWI) and provisional tolerable daily intake (PTDI) recommended by various authorities.

2. Materials and methods

The available fish species in each season were collected from the same area of study by professional fishermen. *Caranx crysos, Euthynnus alleferatus, Scomberomorus commerson, Sphyraena viridensis, Sargus sargus* (spring, 2005); *Siganus rivulatus, Sargus sargus, Mugil* species (summer, 2005); *Siganus rivulatus, Sargus sargus, Mugil* species, *Sardinella aurita* (autumn, 2005); *Siganus rivulatus, Sargus sargus, Mugil* species, *Sphyraena viridensis* (winter, 2006).

Fish samples were stored in pre-washed polyethylene bags and brought to the laboratory on ice on the same day of fish capture (4°C). Total length and weight of the samples were measured to the nearest millimetre and gram before dissection (Table 1). Composite samples from dorsal fillets, liver, and gills of the fish were taken, weighed, packed in polyethylene bags and stored in a refrigerator until chemical analysis. The minimum number for a composite sample was ten fish, selected from one size class over the area of investigation (Figure 1). The collected fish were selected according to the recommendation given by Butler et al. (1971) and Haug et al. (1974) [12,13].

Tissues were homogenised and an exact weight of dry sample (0.2–0.3 g) was placed in Teflon vessels and 4 ml of nitric acid (analar) was added to soft organs, while gills were digested in concentrated nitric and perchloric acids. The samples predigested at room temperature overnight. Samples were digested on a hot plate at 100 °C for 2 h, and were then cooled at room temperature. If the solution was not clear, it was reheated for another 1 h at 100 °C. The samples were transferred to 25 ml volumetric flasks. Before analysis, the samples were filtered. Sample blanks were prepared in the laboratory in a similar manner to the field samples. All samples were analysed three times for Cd, Cu, Fe, Mn, Ni, Pb, and Zn by flame atomic absorption spectrophotometer (SpectrAA-10Plus, Varian) and all the concentrations were expressed in terms of wet weight as microgram per gram [14]. All glassware and plastic devices used in the manipulation of samples were completely acid-washed according to Moody and Lindstrom (1977) and reagents of analytical grade were utilised for blanks and calibration curves [15]. The performance of the method was evaluated by analysing a reference material mussel homogenate (MA-A-2/TM) provided by the International Atomic Energy Agency (IAEA). Recoveries between 93% and 104% were accepted to validate the calibration (Table 2). The results showed good agreement between the certified

Table 1. Range of total length and weight of fish species collected from the western part of Alexandria from spring 2005 to winter 2006.

Fish species			
Latin name	Common name	Total length range (cm)	Weight range (g)
Caranx crysos	Mira	20.5 - 23.0	105.0 - 130.0
Euthynnus alleferatus	Kabrit	64.0 - 68.5	2920.0 - 3400.0
Mugil species	Bouri	31.5 - 34.0	185.0 - 200.0
Sardinella aurita	Sardine	14.5 - 16.0	34.0 - 40.0
Sargus sargus	Sharaghish	19.5 - 21.0	100.0 - 155.0
Scomberomorus commerson	Scomber	33.0 - 37.5	200.0 - 270.0
Siganus rivulatus	Batata	17.5 - 20.0	75.0 - 90.0
Sphyraena viridensis	Maghazil	26.0 - 28.5	140.0 - 155.0

			Reference material				
Element	Wavelength (nm)	Detection limit	Certified value	Experimental value	% Recovery		
Cd	228.8	0.001	0.07	0.067 ± 0.002	95.714		
Cu	324.8	0.010	4.00	3.834 ± 0.023	95.850		
Fe	238.2	0.004	54.00	55.824 ± 0.242	103.370		
Mn	276.5	0.002	0.81	0.775 ± 0.027	95.679		
Ni	232.0	0.015	1.10	1.053 ± 0.012	95.727		
Pb	217.0	0.003	0.58	0.557 ± 0.062	96.034		
Zn	213.9	0.006	33.00	31.686 ± 0.158	96.081		

Table 2. The absorption wavelengths, limits of detection, and summary of certified and observed values of reference material (MA-A-2/TM).

and analytical values, the recovery of the elements being partially complete for most of them. Statistical analysis (T-test) in the present study was performed by SPSS Version 15.

3. Results and discussion

Cadmium concentrations in muscles were found to be low and showed little variability among species (Tables 3-6). T-test for Cd concentration in muscle tissues on the period of study for each pair of fish species: Siganus rivulatus and Sargus sargus; Siganus rivulatus and Mugil sp.; and Sargus sargus and Mugil sp. was performed and recorded (p > 0.05) as illustrated in Table 7, revealing that there is no relation between Cd concentration in muscles and the food habits of fish species, where Siganus rivulatus is herbivorous, Sargus sargus is carnivorous and Mugil sp. is omnivorous [21,22]. This agrees with Moore and Ramamoorthy (1981), who reported that there is no correlation between Cd concentration and food habits [23]. Compared to muscle, the liver presented higher Cd concentrations in the period of study and varied among the fish species, whereas Siganus rivulatus (herbivorous) and Sardinella aurita (filter feeder) recorded the highest concentrations. Gill tissues recorded comparable values with that in the liver for Sargus sargus, Mugil species and Sphyraena viridensis, which indicated that the uptake of Cd could occur through the gills as part of the food chain. The concentration levels of Cd in muscle of Mugil species of the studied area is lower than that reported by França et al. (2005): $(0.94-1.6 \,\mu g/g)$ from the Tagus Estuary, Portugal, and from that observed by Canli and Atli (2003): $(0.66 \,\mu g/g)$ caught from the northeast Mediterranean Sea, but comparable with that recorded by Tüzen (2003) in fish samples of the middle Black Sea (Turkey) [9,24,25]. The concentration of Cd in the edible part of the fish species under investigation in the four seasons were generally lower than the levels issued by FAO (1983), NHMRC (1987), and Turkish legislation (2007), where the National and Medical Research Council and the Turkish legislation have established maximum levels for Cd $(1.0 \,\mu g/g)$ wet weight), while the Food and Agricultural Organisation limit for cadmium is $0.5 \,\mu g/g$ wet weight [26–28]. On the other hand, the concentration level in muscle of Siganus rivulatus (in summer 0.216 ± 0.003 , autumn $0.231 \pm 0.002 \,\mu g \,\text{Cd/g}$) as well as Mugil species in the summer season $(0.274 \pm 0.001 \,\mu g \,\text{Cd/g})$, which is slightly higher than that proposed by the Australian National Health and Medical Research Council $(0.2 \mu g Cd/g)$ [29].

Mugil species living near the bottom ingest sediment and detritus and *Siganus rivulatus* which is a pelagic herbivorous feeding on algae and seaweeds both recorded the highest concentration of copper in the liver (Figure 2). Meanwhile *Sardinella aurita*, which filter feeds, accumulates copper in the gills and liver. On the other hand, *Caranx crysos, Euthynnus alleferatus, Scomberomorus commerson, Sphyraena viridensis*, and *Sargus sargus*, which are mainly carnivorous, recorded relatively low concentrations of copper [21,22]. The concentration of copper decreases in the

			Organ	
Metal	Species	Muscle	Liver	Gills
Cd	Caranx crysos Euthynnus alleferatus Scomberomorus commerson Sphyraena viridensis Sargus sargus	$\begin{array}{c} 0.176 \pm 0.002 \\ 0.113 \pm 0.008 \\ 0.114 \pm 0.001 \\ 0.090 \pm 0.003 \\ 0.133 \pm 0.006 \end{array}$	$\begin{array}{c} 0.605 \pm 0.009 \\ 0.502 \pm 0.002 \\ 0.657 \pm 0.003 \\ 0.686 \pm 0.005 \\ 0.702 \pm 0.000 \end{array}$	$\begin{array}{c} 0.499 \pm 0.001 \\ 0.377 \pm 0.009 \\ 0.404 \pm 0.002 \\ 0.563 \pm 0.006 \\ 0.536 \pm 0.007 \end{array}$
Cu	Caranx crysos Euthynnus alleferatus Scomberomorus commerson Sphyraena viridensis Sargus sargus	$\begin{array}{c} 1.354 \pm 0.034 \\ 1.052 \pm 0.026 \\ 1.208 \pm 0.031 \\ 1.365 \pm 0.024 \\ 1.165 \pm 0.034 \end{array}$	$\begin{array}{c} 5.373 \pm 0.025 \\ 4.397 \pm 0.034 \\ 9.630 \pm 0.028 \\ 6.798 \pm 0.027 \\ 7.471 \pm 0.019 \end{array}$	$\begin{array}{c} 2.816 \pm 0.024 \\ 2.491 \pm 0.038 \\ 2.894 \pm 0.012 \\ 3.847 \pm 0.031 \\ 1.999 \pm 0.011 \end{array}$
Fe	Caranx crysos Euthynnus alleferatus Scomberomorus commerson Sphyraena viridensis Sargus sargus	$\begin{array}{c} 28.514 \pm 0.324 \\ 31.944 \pm 0.183 \\ 20.370 \pm 0.327 \\ 23.037 \pm 0.184 \\ 15.183 \pm 0.223 \end{array}$	$\begin{array}{c} 211.313 \pm 0.317 \\ 83.897 \pm 0.367 \\ 58.668 \pm 0.271 \\ 243.911 \pm 0.281 \\ 92.113 \pm 0.421 \end{array}$	$\begin{array}{c} 55.458 \pm 0.201 \\ 77.759 \pm 0.327 \\ 45.943 \pm 0.381 \\ 126.436 \pm 0.521 \\ 69.081 \pm 0.325 \end{array}$
Mn	Caranx crysos Euthynnus alleferatus Scomberomorus commerson Sphyraena viridensis Sargus sargus	$\begin{array}{c} 1.289 \pm 0.062 \\ 1.153 \pm 0.281 \\ 0.654 \pm 0.103 \\ 0.976 \pm 0.0627 \\ 0.686 \pm 0.023 \end{array}$	$\begin{array}{c} 2.438 \pm 0.019 \\ 1.959 \pm 0.327 \\ 2.584 \pm 0.034 \\ 3.480 \pm 0.0351 \\ 2.433 \pm 0.0374 \end{array}$	$\begin{array}{c} 3.036 \pm 0.021 \\ 4.805 \pm 0.038 \\ 4.047 \pm 0.022 \\ 3.359 \pm 0.032 \\ 4.391 \pm 0.153 \end{array}$
Ni	Caranx crysos Euthynnus alleferatus Scomberomorus commerson Sphyraena viridensis Sargus sargus	$\begin{array}{c} 0.184 \pm 0.023 \\ 0.450 \pm 0.016 \\ 0.270 \pm 0.017 \\ 0.533 \pm 0.063 \\ 0.665 \pm 0.026 \end{array}$	$\begin{array}{c} 1.547 \pm 0.013 \\ 0.998 \pm 0.026 \\ 1.204 \pm 0.031 \\ 1.592 \pm 0.034 \\ 1.490 \pm 0.013 \end{array}$	$\begin{array}{c} 2.223 \pm 0.013 \\ 1.470 \pm 0.023 \\ 1.157 \pm 0.022 \\ 3.107 \pm 0.037 \\ 2.093 \pm 0.038 \end{array}$
Pb	Caranx crysos Euthynnus alleferatus Scomberomorus commerson Sphyraena viridensis Sargus sargus	$\begin{array}{c} 1.041 \pm 0.012 \\ 0.857 \pm 0.032 \\ 0.764 \pm 0.008 \\ 1.529 \pm 0.036 \\ 0.954 \pm 0.031 \end{array}$	$\begin{array}{c} 4.313 \pm 0.021 \\ 2.995 \pm 0.011 \\ 3.698 \pm 0.039 \\ 5.223 \pm 0.022 \\ 3.234 \pm 0.013 \end{array}$	$\begin{array}{c} 6.281 \pm 0.021 \\ 6.261 \pm 0.012 \\ 7.196 \pm 0.036 \\ 7.657 \pm 0.063 \\ 5.577 \pm 0.041 \end{array}$
Zn	Caranx crysos Euthynnus alleferatus Scomberomorus commerson Sphyraena viridensis Sargus sargus	$\begin{array}{c} 8.167 \pm 0.056 \\ 8.230 \pm 0.083 \\ 2.843 \pm 0.091 \\ 9.417 \pm 0.082 \\ 6.745 \pm 0.092 \end{array}$	$\begin{array}{c} 58.991 \pm 0.073 \\ 85.612 \pm 0.039 \\ 42.433 \pm 0.63 \\ 52.372 \pm 0.161 \\ 49.775 \pm 0.082 \end{array}$	$\begin{array}{c} 18.959 \pm 0.028 \\ 26.480 \pm 0.063 \\ 27.877 \pm 0.193 \\ 32.010 \pm 0.082 \\ 30.876 \pm 0.352 \end{array}$

Table 3. Mean concentration of heavy metals ($\mu g/g$ wet weight, mean \pm SD) in fish species collected in spring 2005.

order of liver > gills > muscle tissues. Compared to soft tissues (liver and muscle tissues), gill tissues seem to be independent of fish species, as shown in Tables 3–6. It was also observed that the hard tissues accumulate less copper than the liver. The concentration of copper in the muscle, liver, and gill in *Mugil* species in the current study is lower than that reported by Canli and Atli (2003) for the same species [9]. The recommended FAO allowable concentration of copper for human consumption is $30 \mu g/g$ wet weight, while Spanish legislation establishes a maximum level of $20 \mu g/g$ wet weight [9,26,29,30]. The concentration of Cu in muscle in all fish species under investigation over the four seasons was lower than the permissible limit and, therefore the muscle of all the fish analysed was good enough for human consumption in Egypt.

Iron is the most abundant transition element and plays a vital role in the enzymatic and respiratory processes of aquatic animals [31]. In the present study, the liver showed higher iron concentrations, followed by gill and muscle. The considerably higher Fe concentrations in liver relative to muscle tissue in all fish species are expected, due to the physiological role of liver in

			Organ			
Metal	Species	Muscle	Liver	Gills		
Cd	Siganus rivulatus Sargus sargus Mugil species	$\begin{array}{c} 0.216 \pm 0.003 \\ 0.126 \pm 0.006 \\ 0.274 \pm 0.001 \end{array}$	$\begin{array}{c} 1.492 \pm 0.002 \\ 0.647 \pm 0.007 \\ 0.647 \pm 0.008 \end{array}$	$\begin{array}{c} 0.402 \pm 0.007 \\ 0.624 \pm 0.002 \\ 0.647 \pm 0.004 \end{array}$		
Cu	Siganus rivulatus Sargus sargus Mugil species	$\begin{array}{c} 2.562 \pm 0.024 \\ 1.966 \pm 0.037 \\ 2.196 \pm 0.012 \end{array}$	$\begin{array}{c} 65.968 \pm 0.027 \\ 12.926 \pm 0.032 \\ 42.280 \pm 0.011 \end{array}$	$\begin{array}{c} 3.922 \pm 0.028 \\ 4.711 \pm 0.033 \\ 5.898 \pm 0.027 \end{array}$		
Fe	Siganus rivulatus Sargus sargus Mugil speies	$\begin{array}{c} 69.563 \pm 0.307 \\ 30.569 \pm 0.382 \\ 65.565 \pm 0.192 \end{array}$	$\begin{array}{c} 376.374 \pm 0.716 \\ 119.308 \pm 0.382 \\ 149.915 \pm 0.217 \end{array}$	$\begin{array}{c} 204.141 \pm 0.302 \\ 111.965 \pm 0.392 \\ 96.439 \pm 0.218 \end{array}$		
Mn	Siganus rivulatus Sargus sargus Mugil species	$\begin{array}{c} 2.562 \pm 0.016 \\ 0.888 \pm 0.032 \\ 1.629 \pm 0.106 \end{array}$	$\begin{array}{c} 4.935 \pm 0.016 \\ 3.913 \pm 0.037 \\ 3.205 \pm 0.031 \end{array}$	$\begin{array}{c} 9.588 \pm 0.018 \\ 5.414 \pm 0.116 \\ 8.183 \pm 0.037 \end{array}$		
Ni	Siganus rivulatus Sargus sargus Mugil species	$\begin{array}{c} 1.144 \pm 0.023 \\ 0.883 \pm 0.009 \\ 1.518 \pm 0.023 \end{array}$	$\begin{array}{c} 1.157 \pm 0.016 \\ 1.950 \pm 0.029 \\ 1.521 \pm 0.039 \end{array}$	$\begin{array}{c} 2.775 \pm 0.018 \\ 3.270 \pm 0.026 \\ 2.760 \pm 0.037 \end{array}$		
Pb	Siganus rivulatus Sargus sargus Mugil species	$\begin{array}{c} 1.204 \pm 0.009 \\ 1.059 \pm 0.023 \\ 1.047 \pm 0.006 \end{array}$	$\begin{array}{c} 6.184 \pm 0.036 \\ 5.448 \pm 0.012 \\ 4.660 \pm 0.063 \end{array}$	$\begin{array}{c} 7.957 \pm 0.025 \\ 9.296 \pm 0.036 \\ 10.610 \pm 0.017 \end{array}$		
Zn	Siganus rivulatus Sargus sargus Mugil species	$\begin{array}{c} 16.387 \pm 0.082 \\ 9.740 \pm 0.963 \\ 10.800 \pm 0.224 \end{array}$	$\begin{array}{c} 246.145 \pm 0.236 \\ 74.777 \pm 0.182 \\ 98.903 \pm 0.082 \end{array}$	$\begin{array}{c} 27.858 \pm 0.325 \\ 39.060 \pm 0.236 \\ 36.222 \pm 0.192 \end{array}$		

Table 4. Mean concentration of heavy metals ($\mu g/g$ wet weight, mean \pm SD) in fish species collected in summer 2005.

blood synthesis [32]. *Sphyraena viridensis* and *Caranx crysos* recorded high liver concentrations respective to *Euthynnus alleferatus*, *Sargus sargus* and *Scomberomorus commerson* in spring. In the other three seasons, the liver of *Siganus rivulatus* recorded the highest concentration compared to the other investigated fish species, which could be attributed to the food habit of this species (herbivorous). The concentration of iron in the present study for *Mugil* species is lower than that reported by Canli and Atli (2003) and from that obtained by Yilmaz (2003) from Iskenderun Bay, Turkey. Generally the level of Fe in fish species under investigation is lower than that reported by Tüzen (2003) in fish samples from the middle Black Sea (Turkey) and that recorded by Tuzen and Soylak (2007) in canned fish sold in Turkey [9,25,33–35].

It is well known that manganese is an essential element that activates many enzyme systems [36–38]. The present study revealed that concentration of Mn decreases in the order gill > liver > muscle. The high manganese level in the gills of all fish species in the current study illustrated that uptake of manganese occurs through the gills as well as through the food chain. The liver of *Siganus rivulatus, Mugil* species and *Sardinella aurita* which are herbivorous, omnivorous, and filter feeding, respectively, recorded high manganese levels relative to the other carnivorous species in the present study. Similar results have been reported previously for *Carassius auratus* (deterivorous), $9.7 \pm 1.6 \,\mu$ g/g, and *Esox lucius* (carnivorous), $5.3 \pm 1.3 \,\mu$ g/g dry weight, as well as *Siganus rivulatus* (herbivorous), which recorded a higher manganese level than *Sargus sargus* (carnivorous) in El-Mex and Eastern Harbour, Egypt [39,40]. The mean manganese concentration for muscle in fish species in the present work fluctuated between 0.654 and 2.322 μ g/g wet weight (Tables 3–6), which is lower than those reported by many authors but within the range (1.33–3.76 μ g/g) recorded by Tuzen (2003) in some fish collected from the Black Sea (Turkey) [25,41–43].

			Organ					
Metal	Species	Muscle	Liver	Gills				
Cd	Siganus rivulatus Sargus sargus Mugil species Sardinella aurita	$\begin{array}{c} 0.231 \pm 0.002 \\ 0.107 \pm 0.008 \\ 0.175 \pm 0.001 \\ 0.176 \pm 0.006 \end{array}$	$\begin{array}{c} 0.710 \pm 0.003 \\ 0.689 \pm 0.008 \\ 0.596 \pm 0.002 \\ 0.821 \pm 0.004 \end{array}$	$\begin{array}{c} 0.546 \pm 0.008 \\ 0.425 \pm 0.003 \\ 0.510 \pm 0.007 \\ 0.658 \pm 0.006 \end{array}$				
Cu	Siganus rivulatus Sargus sargus Mugil species Sardinella aurita	$\begin{array}{c} 2.113 \pm 0.013 \\ 1.069 \pm 0.037 \\ 1.804 \pm 0.024 \\ 2.111 \pm 0.018 \end{array}$	$\begin{array}{c} 27.146 \pm 0.031 \\ 7.488 \pm 0.018 \\ 34.650 \pm 0.012 \\ 5.571 \pm 0.027 \end{array}$	$\begin{array}{c} 4.665 \pm 0.025 \\ 5.205 \pm 0.032 \\ 4.102 \pm 0.016 \\ 4.099 \pm 0.032 \end{array}$				
Fe	Siganus rivulatus Sargus sargus Mugil species Sardinella aurita	$55.751 \pm 0.281 \\ 24.822 \pm 0.382 \\ 39.065 \pm 0.281 \\ 40.329 \pm 0.165$	$\begin{array}{c} 279.172 \pm 0.382 \\ 120.672 \pm 0.165 \\ 91.736 \pm 0.281 \\ 243.701 \pm 0.382 \end{array}$	$\begin{array}{c} 191.328 \pm 0.135 \\ 120.359 \pm 0.165 \\ 85.665 \pm 0.432 \\ 97.915 \pm 0.327 \end{array}$				
Mn	Siganus rivulatus Sargus sargus Mugil species Sardinella aurita	$\begin{array}{c} 2.322 \pm 0.025 \\ 0.871 \pm 0.037 \\ 1.406 \pm 0.031 \\ 1.529 \pm 0.026 \end{array}$	$\begin{array}{c} 4.846 \pm 0.037 \\ 2.786 \pm 0.023 \\ 1.603 \pm 0.019 \\ 6.091 \pm 0.027 \end{array}$	$\begin{array}{c} 8.623 \pm 0.038 \\ 5.562 \pm 0.027 \\ 5.498 \pm 0.021 \\ 13.150 \pm 0.082 \end{array}$				
Ni	Siganus rivulatus Sargus sargus Mugil species Sardinella aurita	$\begin{array}{c} 1.043 \pm 0.016 \\ 0.931 \pm 0.009 \\ 0.788 \pm 0.034 \\ 0.646 \pm 0.021 \end{array}$	$\begin{array}{c} 2.911 \pm 0.023 \\ 0.610 \pm 0.016 \\ 0.458 \pm 0.008 \\ 2.267 \pm 0.016 \end{array}$	$\begin{array}{c} 2.640 \pm 0.032 \\ 4.772 \pm 0.012 \\ 2.321 \pm 0.036 \\ 2.784 \pm 0.029 \end{array}$				
Pb	Siganus rivulatus Sargus sargus Mugil species Sardinella aurita	$\begin{array}{c} 1.241 \pm 0.053 \\ 1.085 \pm 0.025 \\ 1.255 \pm 0.008 \\ 1.090 \pm 0.042 \end{array}$	$\begin{array}{c} 4.581 \pm 0.028 \\ 3.748 \pm 0.039 \\ 4.542 \pm 0.017 \\ 4.542 \pm 0.062 \end{array}$	$\begin{array}{c} 6.319 \pm 0.016 \\ 8.067 \pm 0.021 \\ 7.757 \pm 0.023 \\ 8.755 \pm 0.019 \end{array}$				
Zn	Siganus rivulatus Sargus sargus Mugil species Sardinella aurita	$\begin{array}{c} 13.638 \pm 0.126 \\ 7.097 \pm 0.084 \\ 10.457 \pm 0.392 \\ 9.704 \pm 0.226 \end{array}$	$\begin{array}{c} 134.143 \pm 0.392 \\ 49.920 \pm 0.285 \\ 52.235 \pm 0.382 \\ 33.765 \pm 0.662 \end{array}$	$\begin{array}{c} 27.806 \pm 0.063 \\ 39.591 \pm 0.039 \\ 21.666 \pm 0.028 \\ 31.299 \pm 0.106 \end{array}$				

Table 5. Mean concentration of heavy metals ($\mu g/g$ wet weight, mean \pm SD) in fish species collected in autumn 2005.

Nickel recorded its highest concentration in gill tissues, followed by liver, while its lowest concentration was observed in muscle tissues in the investigated fish species (Tables 3–6). Liver of *Siganus rivulatus* recorded the lowest value compared to that observed in liver of *Sargus sargus* in summer, and it also recorded a lower nickel level compared to that recorded for *Mugil* species and *Sphyraena viridensis* in winter, indicating that nickel did not accumulate through the food chain only. This is in agreement with Moore and Ramamoorthy (1981), who reported that the levels of nickel in the kidneys of carnivorous fish were substantially higher than those of omnivorous fish [23]. The recorded nickel concentration in muscle of *Mugil* species is comparable to that previously reported level by Yilmaz (2003) in Turkey [33]. The concentrations of nickel in muscle of all species under investigation are about five times lower than the legislated level given by the Western Australian Food and Drug Regulation (5.5 μ g/g) [44].

According to the present study, the gills are the organ which show the highest accumulation of lead (Figure 2). High content of lead in gills is approved by the NRCC (1973); this finding is attributed to the possibility of particulate or organic lead adsorbed in gills of fish, as well as lower pH at the gill surface due to respired CO_2 , which may dissolve lead to a soluble form that could diffuse into the gill tissue [45]. The concentration of lead in liver is considerably higher than that in the corresponding muscle tissue of the fish under investigation. This is in agreement with some authors, though they the specific mechanism for the greatly elevated Pb levels in liver remains unknown [46–48]. In contrast to the metals copper, zinc and cadmium, there is no evidence that lead produces the metal-binding protein metallothionein (MT) [48]. Muscle is the organ which

		Organ				
Metal	Species	Muscle	Liver	Gills		
Cd	Siganus rivulatus	0.186 ± 0.006	0.603 ± 0.009	0.331 ± 0.002		
	Sargus sargus	0.140 ± 0.003	0.455 ± 0.001	0.399 ± 0.008		
	Mugil species	0.192 ± 0.011	0.486 ± 0.003	0.430 ± 0.005		
	Sphyraena viridensis	0.132 ± 0.001	0.301 ± 0.007	0.267 ± 0.004		
Cu	Siganus rivulatus	1.706 ± 0.023	58.283 ± 0.027	5.109 ± 0.032		
	Sargus sargus	1.299 ± 0.026	10.819 ± 0.023	3.820 ± 0.022		
	Mugil species	1.183 ± 0.017	39.908 ± 0.009	5.459 ± 0.017		
	Sphyraena viridensis	2.491 ± 0.032	5.230 ± 0.021	4.585 ± 0.033		
Fe	Siganus rivulatus	32.172 ± 0.167	228.802 ± 0.164	75.906 ± 0.352		
	Sargus sargus	14.710 ± 0.371	109.981 ± 0.285	76.833 ± 0.327		
	Mugil species	28.030 ± 0.293	95.959 ± 0.192	76.129 ± 0.327		
	Sphyraena viridensis	38.797 ± 0.167	98.498 ± 0.371	84.325 ± 0.036		
Mn	Siganus rivulatus	1.455 ± 0.033	1.934 ± 0.082	7.552 ± 0.022		
	Sargus sargus	1.161 ± 0.017	3.986 ± 0.031	5.266 ± 0.037		
	Mugil species	1.725 ± 0.038	1.842 ± 0.021	9.251 ± 0.024		
	Sphyraena viridensis	2.163 ± 0.019	1.030 ± 0.034	6.638 ± 0.023		
Ni	Siganus rivulatus	0.938 ± 0.026	1.466 ± 0.034	2.100 ± 0.011		
	Sargus sargus	0.633 ± 0.021	1.686 ± 0.024	4.751 ± 0.008		
	Mugil species	1.311 ± 0.039	2.017 ± 0.012	3.233 ± 0.012		
	Sphyraena viridensis	1.096 ± 0.028	1.903 ± 0.031	3.911 ± 0.033		
Pb	Siganus rivulatus	1.035 ± 0.007	5.485 ± 0.016	6.496 ± 0.027		
	Sargus sargus	1.033 ± 0.034	3.823 ± 0.037	5.212 ± 0.025		
	Mugil species	0.984 ± 0.052	4.957 ± 0.038	7.166 ± 0.022		
	Sphyraena viridensis	0.999 ± 0.012	4.940 ± 0.027	6.466 ± 0.013		
Zn	Siganus rivulatus	13.030 ± 0.283	114.910 ± 0.026	23.492 ± 0.028		
	Sargus sargus	6.860 ± 0.635	47.001 ± 0.093	30.967 ± 0.163		
	Mugil species	8.027 ± 0.052	51.423 ± 0.107	28.730 ± 0.019		
	Sphyraena viridensis	10.123 ± 0.082	37.835 ± 0.032	27.017 ± 0.293		

Table 6. Mean concentration of heavy metals ($\mu g/g$ wet weight, mean \pm SD) in fish species collected in winter 2006.

Table 7. T-test for Cd in muscle of fish species among the different seasons.

				95% Confid of the d	lence interval lifference		
Pair	Mean	SD	SE mean	Lower	Upper	t	р
Sr-Ss Ss-Msp Ss-Msp	$0.087 \\ -0.027 \\ -0.093$	0.039 0.057 0.051	0.023 0.033 0.030	-0.010 -0.144 -0.217	0.184 0.139 0.038	$3.839 \\ -0.081 \\ -3.008$	0.062 0.943 0.095

Notes: p < 0.05 means the difference is significant. Sr, Siganus rivulatus; Ss, Sargus sargus; Msp, Mugil species.

accumulates the lowest concentration of lead in all fish species under investigation, as shown in Tables 3–6. This low level is explained by the relatively low binding to SH group, in addition to the low solubility of salts restricting movement across cell membranes. The concentration of lead in muscle for the investigated fish species are slightly higher than that reported by Tuzen (2003) for fish collected from the Black Sea (Turkey) [25]. Comparison with the National and Medical Research Council recommendations specify that the concentration of lead in the edible fish must not exceed $2.0 \,\mu$ g/g wet weight, and according to the present study, muscle tissues in all fish species recorded levels lower than the permissible limit, as shown in Tables 3–6 [27].



Figure 2. Mean concentration of different heavy metals in muscle, liver and gill tissues in fish species collected from West area of Alexandria, Egypt.

Notes: Cc: Caranx crysos, Ea: Euthynnus alleferatus, Sc: Scomberomorus commerson, Sv: Sphyraena viridensis, Ss: Sargus sargus, Sr: Siganus rivulatus, Ms: Mugil species, Sa: Sardinella aurita.

All samples of the species were characterised by high average zinc concentrations in the liver. Compared to other fish species, *Siganus rivulatus* and *Mugil* species accumulate high liver concentrations, attributed to the feeding type of these two species. The high levels of zinc in these two species confirms that the majority of zinc is obtained from dietary sources, rather than from the water [23,39]. *Sardinella aurita* recorded nearly the same level of zinc in liver and gill amongst all the species under investigation. Similar observations were previously recorded by Shakweer

and Abbas (1996) and Wong et al. (2001) due to the filter feeding of this species [49,50]. The concentration of zinc in gills has a different pattern than liver in the different species, indicating that the concentration of zinc in gills depends more on the habitat of the species and on the surrounding water than its dependence on the food habit. *Siganus rivulatus* recorded the highest zinc concentration in its muscle tissues, while *Scomberomorus commerson* recorded the lowest value. The highest concentration of zinc in flesh was recorded for *Siganus rivulatus* in summer (16.387 μ g/g wet weight). The concentration of zinc in muscle tissues for investigated fish species in the present study was lower than that reported by Tuzen (2003) in the Black Sea [25]. Comparison with the Canadian food standards (100 μ g/g), Hungarian standards (80 μ g/g), and Australian accepted limits (150 μ g/g), demonstrate that the concentration level of Zn in all studied fish are still much lower than the guidelines, and they are therefore safely within the limits for human consumption [51].

All tissue metal levels were higher in the summer season, compared to other seasons, as shown in Figures 3–5. T-test analyses for the concentration of the seven metals under investigation in muscle, liver and gill tissues for the studied fish species were performed to study the variation of metal concentrations within different seasons. Statistically significant T-tests (p < 0.05) were observed for both liver and muscle, while metal level difference between seasons for gill tissues was insignificant (Table 8). This observation indicates that concentrations of metals in gills reflects the concentrations of metals in waters, whereas the concentrations in the liver represent the storage of metals and metabolic activity of each organ [40,52,53]. Variation in metal concentrations with season has been well-documented in different studies from fresh and marine water, due to varying seasonal growth rate, reproductive cycle, water salinity and temperature [54–56].

The present data showed that metal concentrations in the liver and gill tissues were highest in all the species in every season. This finding is in agreement with previous studies [28,33,36,51,57]. Bioaccumulation in the liver was observed for most metals, since the liver is the major organ involved in xenobiotic metabolism in fish [58]. It is well known that the accumulation of metals in the liver could be due to the greater tendency of the elements to react with the oxygen carboxylate, amino group, nitrogen and/or sulphur of the mercapto group in the metallothionein protein, whose concentration is highest in the liver [59]. The gills are responsible for the water flow and are exposed to large water mass and thus are expected to have high metal concentration [60]. Concentrations of investigated metals are very low in muscle tissues, which may reflect the low levels of these binding proteins in muscle [57,61].

The overall metal contents of fish species under investigation were compared using the metal pollution index (MPI) calculated with the formula according to Usero et al. (1996, 1997) [62,63]:

$$MPI = (M_1 \times M_2 \times M_3 \times \dots \times M_n)^{1/n},$$

where M_n is the concentration of metal n expressed in $\mu g/g$ of wet weight.

MPI increases in the order of *Scomberomorus commerson* < *Caranx crysos* \leq *Euthynnus alleferatus* < *Sargus sargus* < *Sphyraena viridensis* < *Sardinella aurita* < *Mugil* species < *Siganus rivulatus*, as shown in Figure 6. This may be related to the differences in ecological needs, swimming, behaviour and metabolic activities among different fish species. Also, *Siganus rivulatus* is herbivorous, *Mugil* species is omnivorous (mainly diurnal, feeding on zooplankton, benthic organisms and detritus), and *Sardinella aurita* is a filter feeder. Similar observations were reported by Ney and Van Hassel (1983), Campbell (1994) and Dural et al. (2006) [64–66]. These data are comparable with Khaled (2004) but lower than that reported by El-Nemr (2003) for imported frozen fish in Egypt [40,67–68].

The daily and weekly intakes were estimated for the investigated fish species consumed by adults in Egypt (Table 9). The average daily consumption in Egypt is 35 g per person; this is equivalent to 250 g per person per week. EWI and EDI were estimated using the recorded maximum



Figure 3. Concentration of Cd, Cu, Fe, Mn, Ni, Pb, and Zn in muscle tissues of fish species in different seasons from the western part of Alexandria, Egypt.

Notes: Cc: Caranx crysos, Ea: Euthynnus alleferatus, Sc: Scomberomorus commerson, Sv: Sphyraena viridensis, Ss: Sargus sargus, Sr: Siganus rivulatus, Msp: Mugil species, Sa: Sardinella aurita.



Figure 4. Concentration of Cd, Cu, Fe, Mn, Ni, Pb, and Zn in liver tissues of fish species in different seasons from the western part of Alexandria, Egypt.

Notes: Cc: Caranx crysos, Ea: Euthynnus alleferatus, Sc: Scomberomorus commerson, Sv: Sphyraena viridensis, Ss: Sargus sargus, Sr: Siganus rivulatus, Msp: Mugil species, Sa: Sardinella aurita.



Figure 5. Concentration of Cd, Cu, Fe, Mn, Ni, Pb, and Zn in gill tissues of fish species in different seasons from the western part of Alexandria, Egypt.

Notes: Cc: Caranx crysos, Ea: Euthynnus alleferatus, Sc: Scomberomorus commerson, Sv: Sphyraena viridensis, Ss: Sargus sargus, Sr: Siganus rivulatus, Msp: Mugil species, Sa: Sardinella aurita.

				95% Confidence of the differ	e interval rence		
Pair	Mean	SD	SE mean	Lower	Upper	t	р
				Muscle tissues			
Summer-Winter	4.957	11.343	2.475	-0.207	10.120	2.002	0.029
Summer-Autumn	2.601	6.326	1.381	-0.278	5.481	1.884	0.037
Winter-Autumn	-2.355	5.796	1.265	-4.994	0.283	-1.862	0.038
				Liver tissues			
Summer-Winter	20.787	42.446	9.263	1.466	40.109	2.244	0.018
Summer-Autumn	18.910	33.374	7.283	3.718	34.102	2.597	0.008
Winter-Autumn	-1.877	14.141	3.085	-8.314	4.560	-0.608	0.274
				Gill tissues			
Summer-Winter	10.148	28.382	6.194	-2.771	23.067	1.639	0.058
Summer-Autumn	1.825	5.099	1.113	-0.497	4.146	1.640	0.058
Winter-Autumn	-8.324	26.481	5.779	20.378	3.730	-1.440	0.085

Table 8. T-test for fish species among the different seasons in muscle, liver and gill tissues.

Note: p < 0.05 means the difference is significant.



Figure 6. MPI of fish species collected from the area of study. Notes: Cc: Caranx crysos, Ea: Euthynnus alleferatus, Sc: Scomberomorus commerson, Sv: Sphyraena viridensis, Ss: Sargus sargus, Sr: Siganus rivulatus, Ms: Mugil species, Sa: Sardinella aurita.

metal levels for each species (μ g/g) (Tables 3–6), multiplied by fish consumption (g/70 Kg body weight/week). As can be seen in Table 3, the estimated EWI and EDI values for the examined fish in the present study were far below the recommended values (Table 3) [69–71]. Accordingly, we concluded that consumption of these species from the area of study is not associated with enhanced metal intake, as the fish were safely within the limits for human consumption. These results are comparable with Emara et al. (1993), El-Moshelhy (1996), and Khaled (2004) along the Egyptian coast. Although there is not currently high levels of metals in fish muscles, this is a potential risk for the future, depending on the agricultural and industrial development, as seafood is not the only metal intake route [40,67,72,73].

Table 9. The estimated daily and weekly intakes for the eight fish species collected from the western part of Alexandria, Egypt.

Fish species	Cd	Cu	Fe	Mn	Ni	Pb	Zn
PTWI	490	245,000	392,000	68,600	2450	1750	490,000
PTDI	70	35000	56,000	9800	350	250	70,000
Caranx crysos EWI (EDI)	44.0 (6.2)	338.5 (47.4)	7128.5 (998.0)	322.3 (45.1)	46.0 (6.4)	260.3 (36.4)	2041.8 (285.8)
Euthynnus alleferatus EWI (EDI)	28.3 (4.0)	263.0 (37.0)	7986.0 (1118.0)	288.3 (40.4)	112.5 (15.8)	214.3 (30.0)	2057.5 (288.1)
Scomberomorus commerson EWI (EDI)	28.5 (4.0)	302.0 (42.3)	5092.5 (713.0)	163.5 (22.9)	67.5 (9.5)	191.0 (26.7)	710.8 (99.5)
Sphyraena viridensis EWI (EDI)	33.0 (4.6)	622.8 (87.2)	9699.3 (1357.9)	540.8 (75.7)	274.0 (38.4)	382.3 (53.5)	2530.8 (354.3)
Sargus sargus EWI (EDI)	35.0 (4.9)	491.5 (68.8)	7 642.3 (1070.0)	290.3 (40.6)	232.8 (32.6)	264.8 (37.1)	2435.0 (340.9)
Siganus rivulatus EWI (EDI)	57.8 (8.1)	640.5 (89.7)	17390.8 (2434.7)	640.5 (89.7)	286.0 (40.0)	310.3 (43.4)	4096.8 (573.5)
Mugil species EWI (EDI)	68.5 (9.6)	549.0 (76.9)	16391.3 (2294.8)	431.3 (60.4)	379.5 (53.1)	313.8 (43.9)	2700.0 (378.0)
Sardinella aurita EWI (EDI)	44.0 (6.2)	527.8 (73.9)	10082.3 (1411.5)	382.3 (53.5)	161.5 (22.6)	272.5 (38.2)	2426.0 (339.6)

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