

Determination and Comparison of Non-essential and Essential Elements in Different Species of Fish Available in Omani Markets by Using Inductively Coupled Plasma-optical Emission Spectrometry

Issa Al-Amri^{1*}, Isam Kadim¹, Abdulaziz AlKindi¹, Ahmed Al-Harrasi¹, Khadija Al-Hosni¹, Aiysha Al-Kiyimi¹, Anwar Al-Amri¹, Muzna Al-Salehi¹, Fazal Mabood¹, Ahmed Hamaed¹, Saud M Al Jufaili³, Samera Khalaf² and Mohamed Al-Omairi²

¹Department of Biological Sciences and Chemistry, College of Arts and Sciences, University of Nizwa, Sultanate of Oman, Oman

²Natural and Medical Sciences Research Center, University of Nizwa, Sultanate of Oman, Oman

³Department of Marine Science and Fisheries, College of Agricultural and Marine Sciences, Sultan Qaboos University, Muscat, Sultanate of Oman, Oman

*Corresponding Author: Issa Al-Amri, Department of Biological Sciences and Chemistry, College of Arts and Sciences, University of Nizwa, Sultanate of Oman.

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Abstract

In this study an ICP-OES (inductively coupled plasma-optical emission spectrometry) was used to investigate the concentration levels of both essential and non-essential elements in 70 edible portion of fish samples representing 11 different species of fish collected from Omani markets. Sample preparation via microwave digestion were analyzed, using ICP-OES for seven non-essential elements [Arsenic (As), cadmium (Cd), nickel (Ni), lead (Pb), titanium (Ti), chromium (Cr) and mercury (Hg)] and seven essential elements [iron (Fe), copper (Cu), zinc (Zn), phosphorus (P), calcium (Ca), selenium (Se), and magnesium (Mg)]. A comparison of the results for the concentration of Cd, Ni, Pb, Ti, Cu and Fe elements between the species of fish showed that there was significant ($P < 0.05$) difference in concentrations. Tissue samples from *Seriola dumerili* and *Xiphias gladius* tissues contained significantly ($P < 0.05$) higher levels of Cd, while *Lutjanus argentimaculatus* and *Parastromateus niger* samples had significantly ($P < 0.05$) higher levels of Ni than those from other fish species. The levels Pb found in the tissues of *Merluccius bilinearis*, and *Lethrinus nebulosus* fish were significantly ($P < 0.05$) higher than other fish species. Further, *Lethrinus nebulosus* and *Seriola dumerili* tissue samples contained significantly ($P < 0.05$) higher concentrations of Ti. The concentrations of Ni and Pb were found higher than the maximum permissible limits in certain fish species samples for human consumption. The results indicated that the edible portion contained significantly ($P < 0.05$) higher levels of Cu in *Seriola dumerili* species and Fe elements in *Ocyurus chrysurus* and *Pagellus affinis* than other fish species. This study concluded that Cd, Ni, Pb, Ti, Cu and Fe elements were species-dependent.

Keywords: Non-essential Element; Essential-element; Tissue; Inductively Coupled Plasma-optical Emission Spectrometry; Heavy Metals; Fish; Oman

Introduction

Arsenic (As), cadmium (Cd), nickel (Ni), lead (Pb), titanium (Ti), chromium (Cr) and mercury (Hg) elements are considered as non-essential elements because they do not have a biological role,

and are toxic for the health of humans. On the other hand, iron (Fe), copper (Cu), zinc (Zn), phosphorus (P), calcium (Ca), selenium (Se), and magnesium (Mg) are considered as essential elements due to their important roles in the physiological functions of the human body [1-4]. Deficiency of essential elements may cause inappropri-

ate metabolic functions of enzymes and can result in physiological body failures, and long-lasting diseases [5]. Fish like other organisms need these elements, but if they exceed the metabolic requirement they become accumulated in their tissues, because most of these elements are non-biodegradable [6]. The heavy metals are known as environmental pollutants and characterized by their atomic weights [7,8] and their affinity with biological human tissues [9,10]. Non-essential elements including As, Cd, Ni, Pb, Ti, Cr, and Hg due to its toxicity although at low levels is hazardous for human health because of bioaccumulation in some food-stuffs [11-14]. The heavy elements may be taken up by various fish species and accumulate in their body organs from food, water, or sediments for their body metabolism [15]. Even at low levels of concentration, non-essential elements can be dangerous for fish consumers' health and long-term intake may result in the accumulation of toxic heavy metals in human body [16]. It has also been reported that health hazardous effects of trace elements occur when the biological system in humans are no longer able to counter uptake, which affects the physiological and histopathological functions of body organs [17-20].

Although, fish products are among the most popular of the human diet [21], their products might be contaminated by toxic elements and may be dangerous for human health due to bioaccumulation in the human body. Fish species have the ability to bio-accumulate heavy metals in their tissues through adsorption and absorption via gill surface [22-26]. Areas with higher traffic and agricultural activities have reported high levels of heavy metals [25,27]. Therefore, contaminated fish are unfit for human consumption due to high concentrations of heavy metals in their products [3,28-32]. The toxic trace elements may cause kidney failure, liver damage, and cardiovascular diseases, therefore, to protect people's health, levels of heavy metals should be assessed by using chemical biomonitoring as early indicators of biological effects [25]. It has been reported that certain fish species may be used as a bio-indicator of specific trace elements compared to others [33-36]. Heavy metal contamination in fish may also be due to sediment from bottom dwelling and bottom feeding fish. Many other factors like reproductive cycle, sex, size, age, swimming patterns, living environment and feeding behavior may affect contamination of heavy metals in fish [37-41]. It has been stated that harmful trace elements are linked to increased consumption of fish products with the growing concern of their nutritional and therapeutic benefits [26].

Aim of the Study

Due to hazardous health effects because of the contamination of heavy metals in fish meats, this study was aimed to characterize heavy metal concentration in different fish species available in local markets of Oman. Concentration of seven non-essential elements including As, Cd, Ni, Pb, Tl, Cr and Hg and seven essential elements including Cu, Fe, Zn, P, Ca, Se, and Mg were carried out in 70 fish samples using ICP-OES.

Materials and Methods

Fish species samples

A total of 42 fish samples representing different species were purchased from local markets from the Nizwa city in Sultanate of Oman in the month of March 2020. Samples details are described in Table 1. The 14 fish species vary naturally in size. Silver Hake, Aquamarine and Premium fillets were frozen and imported fillets, while the remaining species were fresh. After collection the samples were put in polyethylene bags and transported to the University of Nizwa laboratory in an ice box for investigation. Then clean forceps and scissors were used to dissect the fish samples after they had been classified into demersal or pelagic fish.

Preparation of samples

Approximately 300 g of flesh from each species of fish sample was cut in small pieces with a clean knives, and dried using Memmert UF10m, Germany oven at 80°C until a constant weight was obtained. The dried fish samples were allowed to cool and then ground (Grinder Panasonic, brand MX- AC210S, India) into fine powder, kept in labeled containers and stored in desiccators in the dark until digestion. Mineralization was accomplished using ultra-microwave-assisted acid digestion system (Single Reaction Chamber Microwave Digestion System, Ultrawave, Milestone, USA). Briefly, 2 mL of H₂O₂ (30% v/v) and 6 mL of HNO₃ (68% v/v) (all analytical grade from company MRS Scientific Limited, UK) was added to 0.8 g of each fish sample and the vessels were placed into the microwave digestion system. Fish samples digestion was achieved by heating up to 120°C for 25 min, then increasing the temperature to 190°C for 35 min and then cooled down for 30 min to reach room temperature. The solution of each sample was then transferred into 50 mL volumetric flasks and were diluted to the mark by adding ultrapure water for subsequent analysis by ICP-OES. Each sample

Number	Commercial name	Scientific name	Geolocation
1	Silver Hake (Imported)	<i>Merluccius bilinearis</i>	Demersal
2	Naiser fish	<i>Ocyurus chrysurus</i>	reef-associated
3	Arabian Pandora	<i>Pagellus affinis</i> (Boulenger, 1887)	Demersal
4	Aqumarine fillet	<i>Merluccius bilinearis</i>	Demersal
5	Brownspeckled Grouper	<i>Epinephelus chlorostigma</i>	reef-associated
6	Premium fillet	<i>Merluccius bilinearis</i>	Demersal
7	Skipjack Tuna	<i>Katsuwonus pelamis</i> (Linnaeus, 1758)	Pelagic-oceanic
8	Sword fish	<i>Xiphias gladius</i> (Linnaeus, 1758)	Pelagic-oceanic
9	Mangrove Snapper	<i>Lutjanus argentimaculatus</i> (Forsskal, 1775)	Reef-associated
10	Black pomfret	<i>Parastromateus niger</i> (Bloch, 1795)	Reef-associated
11	Local Emperor	<i>Lethrinus nebulosus</i> (Forsskal, 1775)	Reef-associated
12	Spangled Emperor	<i>Lethrinus nebulosus</i> (Forsskal, 1775)	Reef-associated
13	Greater Amberjack	<i>Seriola dumerili</i> (Risso, 1810)	Reef-associated
14	Indian Mackerel	<i>Rastrelliger kanagurta</i> (Cuvier, 1817)	Pelagic-neritic

Table 1: Commercial and scientific names and geolocations of fish species samples collected from the local markets in Oman.

was analyzed in triplicates using the Optima™ 8000 ICP-OES by PerkinElmer, USA. Elemental standard solutions of Hg, Pb, Cd, As, Ni, Ti, Cr, Cu, Zn, Fe, P, Ca, Se and Mg) (1000 mg L⁻¹) were supplied by MRS Scientific Limited, UK and were used for building the standard calibration curve. The samples were analyzed by ICP-OES and the results were compared to a multi-element standard calibration curve to quantify the amount of each heavy metal analyzed in the digested solution of fish sample.

Statistical analysis

For all the data analysis the Microsoft Excel 365 and SPSS 16.0 were used to find the independent T-test. Differences at the P < 0.05 level was considered significant.

Results and Discussion

Fish is considered by many consumers as a highly nutritious food, therefore, many people consume fish due to its availability, flavor and palatability. The overall mean and range results from ICP-OES analysis for 14 elements (7 non-essential- and 7 essential elements) in different species of fish are shown in Tables 2 to 5. All the heavy metal elements were almost detected in all edible portions of fish species with the exception of very few samples.

Non-essential elements

Analyses of all the fish species samples examined revealed the highest level of Ni, followed by Pb as a non-essential element, with range values from 0.00 to 6.188 µ/g and from 0.00 to 13.2 µ/g, respectively (Table 2). Overall pattern of distribution of non-essential elements as Ni > Pb > Ti, > Cd > Cr > As > Hg for all the fish species. It was interesting to note that the *Xiphias gladius* and *Seriola dumerili* species had among the highest levels of Cd, while *Lutjanus argentimaculatus*, and *Parastromateus niger* had the highest levels of Ni. The *Merluccius bilinearis*, *Lethrinus nebulosus* and *Lethrinus nebulosus* had the highest levels of Pb. *Seriola dumerili*, *Lethrinus nebulosus*, and *Lutjanus argentimaculatus* had the highest levels of Ti. Therefore, certain species of fish have more potential for bioaccumulation of several trace elements in the ecosystem. It can be assumed from the above that accumulation of the trace elements is more species-related. It was also noted that there was no relationship between the concentration patterns of different trace elements in the different fish species. The results indicated that accumulation of the trace elements (overall mean) analyzed in the sampled fish species was of the following trend: *Seriola dumerili* > *Lethrinus nebulosus* > *Merluccius bilinearis* > *Xiphias gladius* and the pattern of distribution was Ni > Pb > Ti > Cd > for all the fish species. Similar findings were reported by Özden., *et al.* [5].

Arsenic (As)

The mean of As concentration level was determined at a value of 0.033 µ/g (Table 2). According to WHO [42], and European Commission [43], the contamination of food with As level is measured as hazardous at concentrations higher than 0.100 µ/g, therefore, the present level is not harmful for people consuming fish species available in local markets. In the study of Toppe., *et al.* [44] who found that the concentration levels of As in nine fish species

Non-essential ¹	Range (µg/g)	Average ± SD (µg/g)
As	0.012-0.076	0.033 ± 0.049
Cd	0.000-1.052	0.256 ± 0.085
Ni	0.000-6.188	1.673 ± 0.447
Pb	0.000-13.2	1.493 ± 0.906
Ti	0.061-1.471	0.427 ± 0.114
Cr	0.059-0.099	0.084 ± 0.003
Hg	0.012-0.049	0.030 ± 0.004

Table 2: Non-essential elements in 70 samples (14 species x 5 replicates) of fish with statistical descriptors: range, average, and standard deviation (SD).

¹As: Arsenic, Cd: Cadmium, Ni: Nickel, Pb: Lead, Ti: Titanium, Cr: Chromium, Hg: Mercury.

ranged from 0.30 to 3.8 µg, which are much higher than values in the present study. For the human diet, the FAO/WHO [45] has set the maximum tolerable weekly As intake at 15 µ/kg body weight. The low levels of As detected in the present study may not release into human organs and may not cause the risk of human pulmonary disease [46]. There was no significant differences in concentration levels of As among the 14 species of fish tested in the current study. While, the variation of the As levels in four fish species (*O. niloticus*, *S. Galilaeus*, *C. gariepinus* and *H. nilotics*) were greater than those values reported in the present study [47]. The highest As levels determined 0.335 µg/g for *Merlangius merlangus*, 0.400 µg/g for *Solea solea*, and 0.966 µg/g for *Mullus surmuletus* [5].

Cadmium (Cd)

Fish species	Non-essential elements ¹						
	As	Cd	Ni	Pb	Ti	Cr	Hg
Fish Species²							
EFFF	0.050 ± 0.001	0.186 ± 0.033 ^b	0.123 ± 0.032 ^b	0.680 ± 0.053 ^b	0.186 ± 0.024 ^a	0.088 ± 0.004	0.049 ± 0.002
NF	0.036 ± 0.002	0.125 ± 0.022 ^b	0.498 ± 0.041 ^b	0.374 ± 0.035 ^b	0.062 ± 0.002 ^a	0.089 ± 0.003	0.013 ± 0.001
SB	0.024 ± 0.004	0.187 ± 0.034 ^b	0.249 ± 0.024 ^b	0.623 ± 0.054 ^b	0.685 ± 0.045 ^a	0.088 ± 0.006	0.022 ± 0.002
AWFF	0.053 ± 0.003	0.245 ± 0.043 ^b	0.368 ± 0.023 ^b	13.23 ± 1.234 ^d	0.674 ± 0.055 ^a	0.095 ± 0.007	0.048 ± 0.003
GF	0.018 ± 0.004	0.061 ± 0.003 ^b	0.306 ± 0.025 ^b	0.000 ± 0.000 ^a	0.061 ± 0.001 ^a	0.059 ± 0.004	0.033 ± 0.001
PWFF	0.048 ± 0.003	0.245 ± 0.033 ^b	0.123 ± 0.016 ^b	0.675 ± 0.076 ^b	0.061 ± 0.001 ^a	0.099 ± 0.006	0.052 ± 0.002
TF	0.037 ± 0.005	0.000 ± 0.000 ^a	0.248 ± 0.036 ^b	0.931 ± 0.096	0.124 ± 0.02 ^a	0.098 ± 0.007	0.043 ± 0.001
SF	0.028 ± 0.004	0.920 ± 0.081 ^c	0.123 ± 0.023 ^b	0.184 ± 0.021 ^b	0.491 ± 0.039 ^a	0.066 ± 0.004	0.014 ± 0.001
RSF	0.012 ± 0.003	0.000 ± 0.000 ^a	6.188 ± 0.092 ^c	0.743 ± 0.054 ^b	0.743 ± 0.071 ^a	0.079 ± 0.007	0.023 ± 0.002
BPF	0.076 ± 0.004	0.123 ± 0.024 ^a	2.771 ± 0.073 ^c	0.431 ± 0.032 ^b	0.185 ± 0.022 ^a	0.086 ± 0.006	0.012 ± 0.001
LEF	0.019 ± 0.001	0.131 ± 0.011 ^a	0.248 ± 0.025 ^b	1.055 ± 0.092 ^c	0.434 ± 0.024 ^a	0.067 ± 0.004	0.014 ± 0.001
EF	0.017 ± 0.005	0.123 ± 0.023 ^a	0.551 ± 0.043 ^b	0.919 ± 0.087 ^c	1.471 ± 0.095 ^b	0.075 ± 0.006	0.024 ± 0.002
AF	0.022 ± 0.002	1.052 ± 0.092 ^c	0.557 ± 0.044 ^b	0.681 ± 0.065 ^b	1.114 ± 0.085 ^b	0.086 ± 0.007	0.023 ± 0.001
MF	0.019 ± 0.004	0.185 ± 0.014 ^b	0.000 ± 0.000 ^a	0.370 ± 0.031 ^b	0.370 ± 0.032 ^a	0.094 ± 0.0083	0.034 ± 0.002
Significant	NS	*	*	*	*	NS	NS

Table 3: Mean (µg/g) and standard error of mean of non-essential elements in 14 fish samples selected from local markets.

¹As: Arsenic, Cd: Cadmium, Ni: Nickel, Pb: Lead, Ti: Titanium, Cr: Chromium, Hg: Mercury.

²Fish species: EFFF: Eastco White Fish Fillet, NF: Naiser Fish, SB: Sea Bream (Red Koffer), AWFF, Aquamarine White Fish Fillet, GF: Grouper Fish (Hammour), PWFF: Premium White Fish Fillet, TF: Tuna Fish, SF: Sword Fish, RSF: Red snapper Fish, BPF: Black Pomfret Fish, LEF: Local Emperor Fish, EF: Emperor Fish, AF: Amberjack Fish, MF: Mackerel Fish.

Cd is considered one of the most harmful trace elements for human health. The overall concentration of Cd was ranged from 0.00 to 1.05 µg in all fish species samples analyzed with overall mean value of 0.256 µg/g. Considering the maximum allowed limit for Cd is 0.5 µg/g for fish [43], the concentration levels of Cd found in *Xiplias gladius* and *Seriola dumerili* were 0.920 and 1.052 µg/g respectively which are particularly toxic for humans. Consump-

tion of high quantities of *Xiplias gladius* and *Seriola dumerili* species may lead to acute renal failure in humans due to sever chronic Cd poisoning [48]. The mean Cd concentration levels in 10 fresh and frozen species of commercially important fish in Oman was in the range of 0.01 to 1.0 µg/g [49] and concluded that the results showed small variations in Cd levels between fish species tested. Similar differences between fish species were found by Mansour

and Sidky [50], who reported a range from 0.00 to 0.79 µg/g between *Mugil Sp.* And *Solea aegyptiaca* and concluded that the *Mugil sp* species had larger variations (0.019-0.790 µg/g) than samples from the *Solea aegyptiaca* species (0.00-0.001 µg/g). Higher level of Cd (7.72-11.58 µg/g) than the present results were reported by Hamilton., *et al.* [51] in different fish species. The accumulation of Cd in the human body may cause prostate cancer and breast cancer in humans [52]. In the present study, edible portions from the 12 fish species contained Cd levels below of 0.5 µg/g and does not cause particular food safety concerns, with the exception of some allergic reactions. Mean Cd concentration levels found in this study were lower than those reported by Ahmed., *et al.* [53] and Akoto., *et al.* [54] who reported Cd mean concentrations of 0.96 ± 0.15 and 0.275 ± 0.47 µg/g, respectively in fish tissues. Fawole., *et al.* [47] found the concentration level of Cd in four species of fish ranged from 100 to 200µg/g. Keskin., *et al.* [55] reported Cd concentrations of 0.054 µg/g in *Merlangius merlangus*; 0.012 µg/g in *Mullus barbatus*; and 0.022 µg/g in *Solea solea*.

Nickle (Ni)

The range of concentration levels of Ni in 14 fish species were found from 0.000 to 6.188 µg/g with an overall mean of 1.673 µg/g (Table 3). The maximum allowed limits [56] for Ni in fish is 70-80 µg/g. The concentration levels of Ni in all the fish samples were below the maximum limit, which will not cause health hazards but it may cause some allergic reaction [57]. Similar concentrations of Ni (5.95-14.45 µg/g) were reported by Hamilton., *et al.* [51]. If the concentrations of Ni are above the set limit it may cause cancer of nasal cavity and the lungs [58]. The concentration values of Ni in this study were higher than those reported by Idodo-Umeh [59] who recorded 1.64 - 3.58 µg/g from fish from the Olomoro water. Higher contaminated fish with Ni than the present study was reported by De Vive., *et al.* [60] who found high levels of Ni in fish and attributed the contamination to suspend particles and plants in water with high percentage of Ni. In the present study, the tissue samples of 2 of 14 fish species contained levels of Ni in excess of 2 µg/g (Table 3). For example, the levels of Ni in *L.campechannus* and *parastromateus niger* samples showed a significant ($P < 0.05$) elevated Ni concentration (6.188 µg/g) and 2.771 µg/g, respectively than other fish samples. The present results are in agreement with Murtala., *et al.* [61], who found that the concentration of Ni varied between *H. forskahlii*, *H. bebe occidentalis* and *C. gariepinus*. The *H. littorale* exhibited mean levels of 13.5 µg/g Ni element [62] that is higher than the mean value in the present study. Furthermore, Mansour and Sidky [50] found that samples from *Mugil sp* contained higher level of Ni (0.012-4.540 µg/g) than those from *Solea aegyptiaca* (0.00-0.00 µg/g). However, lower level (0.24 and 0.36 µg/g in *Mormyrops deliciosus* and *Mormyrus macrophthalmus* has been reported by Oronsaye., *et al.* [63] from the Ikpoba river

dam. The edible portion of 4 of 14 fish species were contaminated with Ni above the maximum recommended limits of 0.5 - 0.6 µg/g (FEPA, 2003) in fish.

Lead (Pb)

The average concentration value of Pb in all fish samples was 1.493 µg/g (0.00-13.2) as given in Table 3. With the exception of two species *Epinephalus chlorostigma* and *Xiphias gladius*, the remaining 12 fish species were above the maximum allowed limits of Pb (0.3 µg/g) in fish muscle [43]. Other findings have also reported similar values of Pb in fish tissues. Staniskiene., *et al.* [64] and Ako-to., *et al.*, [54] reported Pb concentrations of 6.82 and 3.13 µg/g in fish, respectively. Lower levels, of Pb (1.70-6.16 µg/g) to those in the present study were reported by Hamilton., *et al.* [51]. The present findings were higher compared to the findings 0.395 - 0.62 µg/g of Doherty., *et al.* [65], while lower than 9 µg/g [66], 9.69 µg/g [53], 6.82 µg/g [54] and 0.02-0.744 µg/g [49] of Pb in different species of fish. Daka., *et al.* [67] also obtained 0.01-0.06 µg/g in fish species from Nigeria. Liver damage and renal failure in humans may be due to extreme accumulation of heavy metals [68,69]. Therefore, The FAO, WHO, EU and other regulatory bodies of various countries have established the maximum permitted concentrations of heavy metals in foodstuffs [70,71]. This study indicated that tissue from *Epinephalus chlorostigma* and *Xiphias gladius* species had significantly ($P < 0.05$) less levels of Pb than the other species of fish. A similar conclusion was reported by Murtala., *et al.* [61], who found significant differences in concentration levels of Pb between *H. forskahlii*, *H. bebe occidentalis*, and *C. gariepinus* fish species. Farombi., *et al.* [72] also reported 0.73 to 4.12 µg/g difference in Pb levels in *C. gariepinus*, Obasohan., *et al.* [28] also reported 0.10 - 0.83 µg/g in fish species and Oronsaye., *et al.* [63] also gave 3.53 µg/g and 2.67 µg/g in *Mormyrops deliciosus* and *Mormyrus macrophthalmus*. Moreover, Oronsaye., *et al.* [63] found that contamination levels ranged from 3.53 µg/g to 2.67 µg/g in *Mormyrops deliciosus* and *Mormyrus macrophthalmus*. Doherty., *et al.* [65] reported 0.44 µg/g and 0.62 µg/g of lead in *C. nigrodigitatus* and *T. guineensis* respectively. Keskin., *et al.* [55] reported Pb concentrations of 0.207 µg/g in *Merlangius merlangus*, 0.035 µg/g in *Mullus barbatus*, and 0.133 µg/g in *Solea solea*. The *Mugil sp* species had lower concentrations (0.030-12.00 µg/g) of Pb compared to the *Solea aegyptiaca* samples (0.144-24.3 µg/g) [50].

Titanium (Ti)

The range concentration levels of Ti in 14 fish species samples was found to be between 0.061 and 1.471 µg/g with a mean value of 0.427 µg/g. The maximum acceptable weekly intake for Ti is 14 mg/ kg body weight [73]. This study showed substantial differences between the mean values of fish species. Muscle samples from

Lethrinus nebulosus (1.471 µg/g) and *Seriola dumerili* (1,114 µg/g) species had significantly ($P < 0.05$) higher Ti concentration levels than those from other species.

Chromium (Cr)

In the present study, the contamination level of Cr in all fish samples was 0.030 µg/g with a range from 0.012 to 0.063 µg/g as given in Table 3. The maximum allowed limit, 12-13 µg/g [43]. Similar to this study, the Cr levels in three species of fish (*Merluccius merluccius*, *Mugil sp.*, and *Mullus surmuletus*) ranged from 0.010 to 0.079 µg/g [74]. However, Uluozlu, et al. [75] found Cr concentrations of *Mullus surmuletus* at 1.63 µg/g, while Meche, et al. [62] found that tissue samples from *Hypostomus punctatus* and *Serrasalmus spilopleura* fish species contained more than 1µg/g Cr. High concentrations of Cr in fish reported by Jordao, et al. [76, 77]. The concentration of Cr in the tissue of eight of seventeen of fish species was found to be in excess of 1µg/g [62]. Variations in concentration levels of Cr in three different fish species (*H. forskahlii*, *H. bebe occidentalis* and *C. gariepinus*) were also reported by Murtala, et al. [61]. Concentrations of Cr level of 0.40 - 5.61 µg/g in *Parachanna obscura* was reported by Obasohan [29] and 29.8 - 31.6 µg/g from *T. zillii* and 28.1 - 32.2 µg/g from *C. gariepinus* reported by Ishaq, et al. [78]. Moreover, Nwani, et al., [79] found 1.19 µg/g Cr in *Chrysichthys nigrodigitatus* from freshwater while Odoemelan [80] found 1.86 µg/g in *A. nurse*. In the study of Mansour and Sidky [50], the *Mugil sp* species had higher concentrations (1.19-3.05 µg/g) of Cr compared to the *Solea aegyptiaca* samples (0.048-1.86 µg/g).

The results in the present study were lower than most of the previous studies. The levels of Cr presented in 14 fish species in the present study were less than most of published results and lower than maximum recommended limits of 0.15 - 1.0 µg/g [81] in fish.

Mercury (Hg)

The mean concentration value of Hg was detected in all fish samples with an average value of 0.033 µg/g (0.012-0.049 µg/g) as given in Table 3, which may not be significant because the maximum allowed limit for Hg is 0.05 µg/g for fish [43]. Therefore, Hg level in 14 fish species available in the local markets is not particularly toxic for humans at this level and does not cause particular food safety concern [57,82]. Higher level of Hg 11.41-17.03 µg/g [50], 0.2929-1.038 µg/g [5], and 0.01-570 µg/g [49], these range values are higher than the present results. Similar values were reported by Keskin, et al. [55] who found Hg concentrations of 0.0352 µg/g in *Merlangius merlangus*, 0.434 µg/g in *Mullus barbatus*, and 0.329 µg/g in *Solea solea*.

Essential elements

The overall concentrations of seven essential elements in the edible part of 14 species of fish are presented in Table 4. P, Ca, and Mg were mostly plentiful in the fish tissue analyzed in that order. For example, the *Ocyurus chrysurus*, *Pagellus affinis*, *Epinephelus coides* and *Seriola dumerili* had among the highest levels of certain elements tested. Se, Cu and Zn elements were detected in all fish samples at small to moderate levels. Ten of the 14 species are demersal fish that feed on detritus, while the other four species are pelagic fish, and thus the high levels of metals in these species cannot be explained by feeding behaviors. This study presented differences in the concentration levels of 14 elements between selected species of fish. This result was supported by the findings of Mansour and Sidky [50] and Ako and Salihu [84] who found variations in contamination levels of elements between species of fish and concluded that the differences may be due to chemical forms of trace elements and their levels in the water. Considering the remaining seven essential elements analyzed, there was no well-defined order of scale within and across the fish tissue samples analyzed. The variations in the concentration of essential elements in fish tissues studied could have been as a result of the rate in which they are available in the water and the ability of the fish to absorb these inorganic elements from their diets [85].

Copper (Cu)

Essential-elements ¹	Range (µg/g)	Average ±SD (µ/g)
Cu	0.434-6.314	2.487 ± 0.333
Fe	32.81-99.91	68.44 ± 19.12
Zn	4.512-6.421	5.102 ± 0.727
P	1999-2542	2210.3 ± 0155
Ca	299.9-345.6	326.1 ± 4.924
Se	0.212-0.312	0.246 ± 0.008
Mg	137.5-150.4	143.6 ± 1.526

Table 4: Essential elements in 70 samples of fish with statistical descriptors: range, average, and standard deviation (SD).

¹CU: Copper, F: Iron, Zn: Zinc, P: Phosphorus, Ca: Calcium, Se: Selenium, Mag: Magnesium.

The Cu concentrations as shown in Table 4 in the fish muscle is in the range 0.434-6.314 µg/g with an average value of 2.487 µg/g. The maximum allowed limits for Cu in fish was 30.0 µg/g for human health risk concerns [86]. The overall concentration level of Cu in 14 fish samples was below this value. Similar values for Cu in fish tissue was reported by Ozden, et al. [5]. In the present study, tissue samples from *Seriola du-*

merili contained significantly ($P < 0.05$) higher levels of Cu ($6.314 \mu\text{g/g}$) than those from other species, while samples from *Parastromateus niger* contained significantly lower ($P < 0.05$) lower concentrations of Cu ($0.434 \mu\text{g/g}$) compared to other fish species tissue (Table 5). Similar values were reported by Ozden., et al. [5] in *Solea solea*, *Mullus barbatus*, and *Merlangius merlangus* throughout the year. Mansour and Silky [50] also reported similar values to the present study for two fish species (*Mugil sp*: $4.07\text{-}6.76 \mu\text{g/g}$) and (*Solea aegyptiaca*: $0.142\text{-}7.72 \mu\text{g/g}$). Lower levels of Cu in fish species than the present values were reported by Çelik and Oehlschläger [87] who found mean Cu concentrations in three different species of 0.29 , 0.22 , and $0.12 \mu\text{g/g}$. While Keskin., et al. [55] reported Cu concentrations of 9.487 , 0.300 , $0.370 \mu\text{g/g}$ in *Merlangius merlangus*, *Mullus barbatus* and *Solea solea*. Although, Cu is considered an essential part of enzymes and is required for the synthesis of hemoglobin, it can cause harm at high concentrations [88]. The Recommended Dietary Allowances (RDA) for copper in normal healthy adults is 0.9 mg/day .

Iron (Fe)

Fe concentration levels in 14 species of fish tissue varied considerably with a range from 32.82 to $99.91 \mu\text{g/g}$ with a mean value of $68.44 \mu\text{g/g}$ (Table 4). *Ocyurus chrysurus*, *Pagellus affinis*, and *Epinephelus coi-*

des species contained significantly ($P < 0.05$) higher concentrations of Fe than those from other species (Table 5). Similarly, Bogard., et al. [89] found a great variation in Fe concentration levels among 30 fish species. The present findings showed a wide range in Fe concentration levels compared to values reported in the global FAO/INFOODS database on fish [90]. On the other hand, tissue samples from *Merluccius bilinearis*, and *R. kanagartha* species contained the lowest Fe levels than those from other species (Table 5). In the study of Suseno., et al. [82], the Fe concentration levels ranged from $0.4 \mu\text{g/g}$ in *Antigonia rubescens* to $0.8 \mu\text{g/g}$ in *Xenolepidichthys dalgleishi*. Similar Fe levels were reported as $163 \mu\text{g/g}$ for *mullus surmuletus*, $82.7 \mu\text{g/g}$ for *Mugil cephalus*, $104 \mu\text{g/g}$ for *Mullus surmuletus* [75], $5.40\text{-}8 \mu\text{g/g}$ for *Mugil cephalus* [91], and $70.28 \mu\text{g/g}$ for *Mugil cephalus* [92]. Mansour and Silky [50] found higher concentrations of Fe than the present study for *Mugil sp* ($37.0\text{-}372 \mu\text{g/g}$) and *Solea aegyptiaca* ($2.07\text{-}186 \mu\text{g/g}$). Fe is an essential element for life due to its presence in living cells and is necessary for the hemoglobin, myoglobin, and certain enzymes. Overall, the concentration levels of Fe in different species of fish available in local markets indicates that several species may contribute significantly to dietary Fe intake among local people. The Food and Drug Administration recommended daily allowance is 18 mg for iron [56].

Zinc (ZN)

Essential elements ¹							
Fish species ²	Cu	Fe	Zn	P	Ca	Se	Mg
EFFF	2.041 ± 0.454^b	32.81 ± 2.17^a	5.330 ± 0.776	2088 ± 5.66	328.3 ± 2.11	0.212 ± 0.041	139.8 ± 1.062
NF	2.242 ± 0.563^b	99.91 ± 6.77^c	4.984 ± 0.645	2064 ± 4.79	325.1 ± 1.56	0.312 ± 0.031	140.8 ± 1.112
SB	1.991 ± 0.231^b	90.63 ± 7.11^c	4.897 ± 0.599	2542 ± 6.04	313.2 ± 2.06	0.221 ± 0.032	143.9 ± 1.213
AWFF	2.391 ± 0.223^b	61.72 ± 4.76^{ab}	5.712 ± 0.666	2079 ± 3.77	334.7 ± 2.09	0.243 ± 0.043	137.5 ± 1.001
GF	2.382 ± 0.342^b	98.83 ± 6.89^c	5.112 ± 0.531	1999 ± 3.65	345.6 ± 3.08	0.224 ± 0.035	139.6 ± 0.958
PWFF	1.903 ± 0.214^b	72.24 ± 5.43^b	6.421 ± 0.554	2263 ± 4.22	343.6 ± 2.89	0.235 ± 0.041	150.1 ± 1.223
TF	2.674 ± 0.178^b	73.82 ± 4.77^b	6.254 ± 0.598	2380 ± 5.34	301.7 ± 1.78	0.253 ± 0.031	150.4 ± 1.312
SF	2.393 ± 0.243^b	56.93 ± 4.71^{ab}	4.134 ± 0.357	2285 ± 4.88	371.1 ± 3.01	0.312 ± 0.043	142.9 ± 1.154
RSF	2.592 ± 0.311^b	71.64 ± 5.87^b	3.798 ± 0.297	2008 ± 2.99	299.9 ± 1.65	0.233 ± 0.031	143.4 ± 1.231
BPF	0.434 ± 0.042^a	65.82 ± 4.99^{ab}	4.512 ± 0.421	2153 ± 3.31	321.7 ± 2.43	0.245 ± 0.044	140.9 ± 1.011
LEF	2.233 ± 0.265^b	58.92 ± 4.31^{ab}	5.412 ± 0.545	2222 ± 3.76	322.8 ± 2.33	0.244 ± 0.054	139.9 ± 0.875
EF	3.004 ± 0.311^b	57.73 ± 3.98^{ab}	4.652 ± 0.443	2354 ± 4.58	316.8 ± 1.32	0.222 ± 0.033	142.6 ± 0.989
AF	6.314 ± 0.897^c	73.64 ± 5.73^b	4.985 ± 0.525	2265 ± 3.33	319.8 ± 1.33	0.232 ± 0.032	143.9 ± 1.322
MF	2.223 ± 0.210^b	43.81 ± 3.45^a	5.231 ± 0.461	2234 ± 3.17	321.8 ± 2.15	0.254 ± 0.029	144.8 ± 1.411
Significant	*	*	NS	NS	NS	NS	NS

Table 5: Mean ($\mu\text{m/g}$) and standard error of mean of essential elements in 14 fish samples selected from the local markets.

¹Cu: Copper, F: Iron, Zn: Zinc, P: Phosphorus, Ca: Calcium, Se: Selenium, Mag: Magnesium.

²Fish species: EFFF: Eastco White Fish Fillet, NF: Naiser Fish, SB: Sea Bream (Red Koffer), AWFF, Aquamarine White Fish Fillet, GF: Grouper Fish (Hammour), PWFF: Premium White Fish Fillet, TF: Tuna Fish, SF: Sword Fish, RSF: Red snapper Fish, BPF: Black Pomfret Fish, LEF: Local Emperor Fish, EF: Emperor Fish, AF: Amberjack Fish, MF: Mackerel Fish.

The Zn concentration levels in 14 species of fish ranged from 4.512 to 6.421 µg/g with an overall mean level of 5.102 µg/g (Table 5). The variation between the fish species was small and non-significant. Similar results in different fish species were reported by Meche., *et al.* [62]. The amount ranging from 3.9 µg/g in *Rouleina guentheri* to 6.8 µg/g in *Alepocephalus bicolor* [82]. Higher values of Zn in fish species than the present study were reported by Mansour and Sidky [50] who found that the range of Zn in *Mugil sp* from 12.0-23.83 µg/g and from 2.37 to 48.7 µg/g in *Solea aegyptiaca*. Zn has important physiological immune functions, protein synthesis [93], wound healing [94], and DNA synthesis [95]. However, due to its high bioavailability [86], excessive Zn intake can cause a range of physiological problems [96]. Zn levels of *Merlangius merlangus* ranged from 3.0 to 0.31 µg/g [97] and the Zn levels of *Mullus barbatus*, *Merlangius merlangus*, and *Solea solea* were 5.03, 3.84, and 3.72 µg/g [87]. In the present study, the concentration levels of Zn in all fish samples was below the maximum allowed limit of 30 µg/g of Zn for safe human consumption [98]. Therefore, the amounts of Zn in the fish species available in local markets cannot cause health problems to consumers. Zn is an essential-element for humans and its toxicity is rare but, at concentrations of up to 40 µg/g, Zn may induce toxicity, characterized by symptoms of nausea, loss of appetite, irritability, pain and muscular stiffness [48]. The FDA (54) recommended daily allowance of 15 mg for zinc.

Phosphorous (P)

This study showed that the concentration of P ranged from 1999 to 2542 µg/g with a mean of 2210.3 µg/g (Table 5). Similar values were reported by FAO/INFOODS [90]. The present results indicate no significant differences in P concentration levels between the fish species. Edible portion of *Pagellus affinis* species had the highest while *Epinephalus chlorostigma* had the lowest content (Table 5). The P levels were between 1960 and 260 µg/g in *Polymixia sp* and 2300 and 820 µg/g in *Rouleina guenther* [82].

Calcium (Ca)

Ca level ranged from 299.9 to 345.6 µg/g (Table 5) with a mean content of 326.1 µg/g and similar values were reported by others [90]. The present values reveal that most of the fish species assessed would meet ≥30% of the recommended Ca intakes. Ca is an essential element for humans, and represents around 2% of body weight in the human body and is the main structure of bones and teeth. Adequate Ca intake during childhood may be linked to boosted bone mass, breast cancer, bone fragility, and hip fractures, and a reduction of osteoporosis [16]. The small variations in the values of Ca between the fish species might be due to an increase in the proportion of bone to flesh as the fish grow [47,99]. The Ca level ranged from 274.8 µg/g in *Setarches guentheri* to 400.2 µg/g

in *Rouleina guentheri* [82]. In two fish species (*H. niloticus* and *C. gariepinus*), Fawole., *et al.* [47] reported high concentration of Ca due to the benthic nature of these species and their relative preference for consumption of hard structure. Similarly [99], stated that the high Ca content found in *H. niloticus* and *C. angullaris* may probably be due to preferential accumulation and calcification of scales and hard tissues. The current study indicates that variety of fish species are a significant source of high bioavailable dietary Ca for Omani.

Selenium (Se)

The range of Se levels in all fish species ranged from 0.212 to 0.312 µg/g with overall mean of 0.246 µg/g (Table 5). The Se concentrations in species of fish analyzed in the present study showed small non-significant differences between them. The concentration levels of Se in the present study are consistent with values reported by FAO/INFOODS [90]. The findings of the present study showed that tissue samples from *Ocyurus chrysurus* contained the highest concentration of Se (0.312 µg/g) while *Merluccius bilinearis* contained the lowest Se level (0.12 µg/g) of other species. Similar values were reported by Suseno., *et al.* [82] who found Se levels between 0.2 µg/g in *Alepocephalus bicolor* and 0.4 µg/g in *Synagrops japonicas*. Se content of *Solea solea* ranged between 0.307 and 0.641 µg/g, while Se levels in *Merlangius merlangus* ranged from 0.331 to 0.478 µg/g.

Magnesium (Mg)

The range of Mg level in 14 species of fish ranged from 137.5 to 150.4 µg/g with overall mean of 143.6 µg/g (Table 5). In the present study, Mg concentrations in species of fish tested showed a small difference between the 14 species of fish. Fish samples from *Katsuwonus pelamis* contained the highest concentration of Mg (150.4 µg/g) while *Merluccius bilinearis* contained the lowest Mg level (137.5 µg/g). The present values were generally consistent with ranges for other fish species [90]. According to Suseno., *et al.* [82], the Mg levels were between 130.6 µg/g in *Alepocephalus bicolor* and 170 and 090 µg/g in *Setarches guentheri*. Mg is essential for muscle function and prevents cardiovascular disease, osteoporosis, and certain forms of cancer [5]. The daily recommended allowance for Mg is 400 mg [56].

Risk assessment

A general health risk assessments of the various heavy metals present in fish meats considered in this study indicates that consumers would have some experience of significant health risk if they only consume non-essential elements from certain species of fish available in the local markets. Among the non-essential elements assessed in this study, few fish species containing high con-

centration of Cd, Ti and Pb would have a relatively higher potential health risks, while the majority of the species have no health risks. Consuming more than one contaminant fish species may produce an additive effect on the body's physiological functions. Pb and Cd were the major health risk contributors in the study. The low levels of binding proteins in the fish muscle may account for their low concentrations of heavy metals [100]. It can therefore be concluded that levels of trace elements in fish muscle cannot necessarily represent the real impact of metal contamination. In order to assess possible contamination of fish with various elements, it is recommended that metal concentrations in other organs such as the liver, gills and kidney should be studied.

Conclusions

Essential and non-essential element composition showed small variations between 14 species of fish samples. Essential element levels of all fish species were sufficient for recommended daily allowance. The metal content is species-dependent, with some species showing high concentrations and some showing low concentrations. According to the results, mean concentrations of Pb in *Merluccius bilinearis*, *Lethrinus nebulosus*, *Lethrinus nebulosus* and Cd in *Xiphias gladius* and *Seriola dumerili* were above the maximum recommended values by the EU. High Ca, and Pb contents were found in edible portions of *Xiphias gladius*, *Seriola dumerili*, *Merluccius bilinearis*, *Lethrinus nebulosus* and *Lethrinus nebulosus*. As, Hg and Cr concentration levels were below the maximum allowed limits in all fish samples assessed. These toxic elements of fish species are recommendable periodically by monitoring studies because regular consumption of certain fish species could pose serious human health risks.

Declaration of Competing Interest

The authors declare that they have no conflict of interest.

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