Bioaccumulation of selected trace elements in some aquatic organisms from the proximity of Qeshm Island ecosystems: Human health perspective

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Abstract

In this study selected marine species from north Persian Gulf ecosystems were collected to investigate the concentration of 15 trace elements (Al, As, Co, Cr, Cu, Fe, Li, Mo, Ni, Pb, Se, Sr, V, Zn and Hg) in muscle and liver tissues for the purpose of evaluating potential health risks for human consumers. The results indicated that Fe, Zn, Sr, Cu and As are the most abundant TEs in the tissues of the species. The concentration of Cu in *P. semisulcatus* and As in most investigated species pose the highest risk of exposure. The carcinogenic risk values indicate that As and Ni concentrations in the species are above the acceptable lifetime risk for adults and children in most of the species. The margin of exposure risk approach indicated that the risk of detrimental effects due to dietary Pb intake for age groups is low, except for consumers of *T. tonggol*.

Keywords: Seafood safety; Trace element; Fish; Prawn; Persian Gulf.

1. Introduction

Marine ecosystems, as a vital and fragile environment, are endangered due to anthropogenic influences, including inputs of a variety of pollutants, especially in areas affected by industrial activities and unregulated discharge of polluted/waste-water (Cánovas et al., 2020). Most pollutants, including both organic and inorganic, interrupt directly or indirectly the ecological balance of the aquatic environments (Maurya et al., 2019). Since the last several decades have witnessed increased concerns of pollutants such as potentially toxic trace elements (TEs) exposure and their implications for human health through consuming seafood with elevated levels of TEs (Parsai & Kumar, 2020). Because of their persistence and bioaccumulative nature, trace elements remain in the environment for long periods and accumulate in human through the food chain and hence can pose serious hazards and potential toxic effect to ecosystems (Ni et al., 2021; M. Zhang et al., 2019). Chronic

exposure to some TEs is also linked to serious threats to normal biological functions in human organs including renal toxicity, hepatic damage, neurological and cardiovascular diseases, and immunotoxicity (Genchi et al., 2017; Lin et al., 2021; Ramos-Miras et al., 2019; Rehman et al., 2018). However, seafood is a highly recommended nutrient and protein source worldwide, because it is a rich source of long chain omega-3 polyunsaturated fatty acids (PUFAs), proteins and essential minerals content along with vitamins (Nøstbakken et al., 2018). Global marine species ingestion per capita rate in 2018 was estimated to be 20.5 kg per year (56.2 g day^{-1}) (FAO, 2020). The TEs accumulation in tissues of marine species is affected by several biological factors, such as feeding habit, position in the trophic chain, size, age and ecological needs of the species (Di Bella et al., 2020). A typical example is compartments located at higher trophic levels, where TEs accumulation is affected by the external environment pollution (Briand et al., 2018). In an aquatic system, TEs fate in marine organisms depends on environmental and physico-chemical conditions including oxidation/reduction, exposure duration, aggregation, dissolution, pH, temperature, geographical location and water quality (Di Bella et al., 2020; Łuczyńska et al., 2018; Ternengo et al., 2018; van der Oost et al., 2016). Aquatic species from the Persian Gulf are also affected by a wide array of environmental stressors embracing both global and local pollution including sewage discharges, toxic chemicals, petrochemical, agricultural and municipal activities and oil and gas production (Abbasi et al., 2019). Many studies have already been conducted to assess the levels of trace elements in the Persian Gulf marine environment. Zarezadeh et al. (2017) have reported concentrations of trace elements in sediments of the northern part of mangrove in Hara biosphere reserve of Qeshm Island (Hormozgan Province, Persian Gulf). Soltani et al. (2021) have studied the distribution, accumulation and implications of 12 trace elements for human health in muscle and liver tissues of six commercially important marine species from North of the Persian Gulf. Due to widespread intake and environmental dynamics of Persian Gulf, seafood plays an important role in the dietary exposure to trace elements. Therefore, to assure food safety, steady monitoring of hazardous substances such as TEs contamination is essential. The present work is an attempt to highlight trace elements content including Al, As, Cr, Hg, Li, Ni, Pb, Se, Sr, V (potentially toxic) and Co, Cu, Fe, Mo, and Zn (essential elements) in the muscle and liver tissues of selected commercially important marine species. Furthermore, potential health risks associated with fish and prawn species consumption is investigated.

2. Materials and methods

2.1. Study area and sample collection

The study area, Qeshm Island, is located in a subtropical region north of the Persian Gulf (Vaezi et al., 2015). Qeshm Island is the largest island in the region with an area of 1491 km^2, which is close the Strait of Hormuz (Mortazavi & Sharifian, 2012). The semi-enclosed Persian Gulf system with mean salinities exceeding 40 psu and surface temperature fluctuations between winter and summer values as much as 20°C, represents an important ecological resource (Khatir et al., 2020). It provides regionally important sea grass habitats, mangroves and coral reefs (Naser, 2014) used as natural home to many biota species including dugongs, birds, crustaceans, turtles, fish and bivalves. It (Qeshm Island) stretches along the coast of a commercial-industrial free zone of Iran and is also close to the largest southern port of Iran i.e., Bandar Abbas (Nowrouzi et al., 2012). Despite active development of oil and gas resources in the Persian Gulf and the ensuing rapid industrialization and urbanization, the region faces many environmental pressures and threats (Sheppard et al., 2010). Presently, the Persian Gulf ecosystem is affected by several environmental challenges including limited circulation, poor flushing characteristics (3 to 5 years) and low turnover. The semi-enclosed nature of the gulf makes it prone to the accumulation of anthropogenic pollutants by physical and chemical pathways (Khatir et al., 2020).

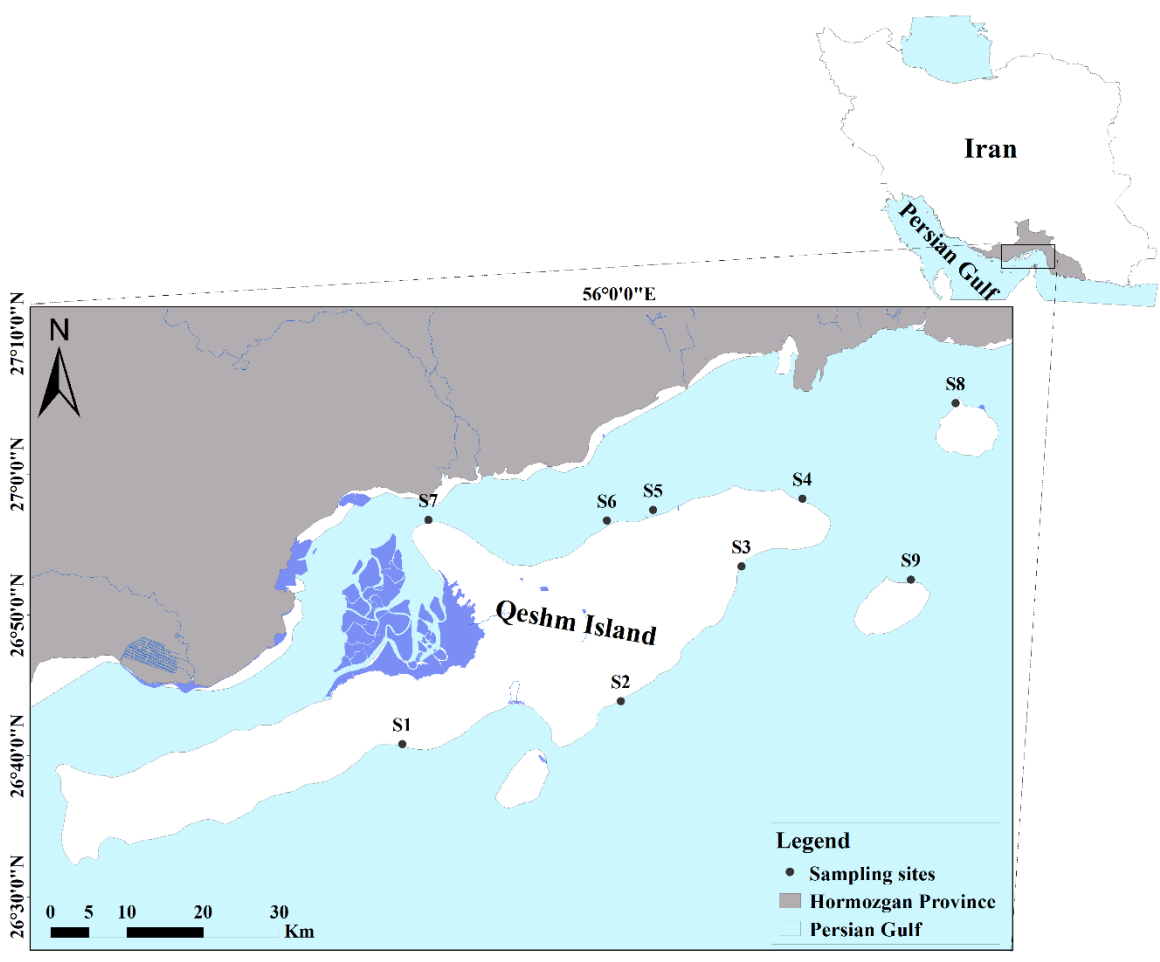

Fig 1. Location of the species sampling sites in the Persian Gulf (S1: Salakh fishing port, S2: Messen fishing pier, S3: Ramchah, S4: Qeshm, S5: Dargahan, S6: Kaveh Port, S7: Laft, S8: Port of Hormuz and S9: Lark).

The data presented in this work is based on analysis of 368 individual samples (16 composite samples) from three commonly consumable marine fish species including Tuna Longtail (*Thunnus tonggol*), Mangrove red snapper (*Lutjanus argentimaculat*) and Klunzinger's mullet (*Liza klunzingeri*), and a species of prawn i.e., green tiger (*Penaeus semisulcatus*). The samples were collected randomly from various fishery zones along the northern and southern zones of Qeshm Island (Fig. 1). The collected species are commonly used as seafood by local population. Targeted species were sampled during Jan 2019 in compliance with operational procedures using several sampling gears including gill net, fishhook, gargoor and trap net (Moshta in local language). Following collection, the samples were promptly put into an icebox and transferred to the laboratory and they were preserved frozen at -20 ˚C until analysis.

2.2. Sample preparation and chemical analysis

Prior to biometric measurements, in the laboratory, the samples were rinsed in deionized water to remove surface adherents. Composite samples of 15–25 g of liver and muscle from each species were prepared using a deionized water-cleaned stainless-steel scalpel. Whenever possible the dorsal musculature of fish (Lozano-Bilbao et al., 2021) was used. For TEs analysis, the samples were first lyophilized in a freeze dryer (Model Alpha 2-4 LD Plus, Christ Germany) at −50 °C and 0.001 mbar for 48 hours. The dried samples were then ground and homogenized into fine powder using ceramic pestle and mortar before sending to Zarazma Minerals Studies Company (Iran) for analysis. Analyses of 14 trace elements (Al, As, Co, Cr, Cu, Fe, Li, Mo, Ni, Pb, Se, Sr, V and Zn) were analyzed using inductively coupled plasma mass spectrometry (ICP-MS). Total Hg (T-Hg) content was measured by atomic absorption spectrometry and cold vapor flow injection technique (FIMS). The sample digestion was implemented according to Erdemir & Gucer (2013) and Soultani et al. (2019) with minor modification. The samples were digested as follows. Briefly, aliquots of 250 mg of each freeze-dried, homogenized sample were digested in a mixture 3 mL of nitric acid (HNO_3) and 1 ml of hydrogen peroxide (H_2O_2) in a closed microwave digestion system for 10 min and kept for another 20 min. The final digested sample solution transferred into a flask and diluted to 25 mL with deionized water, and analyzed for TEs (except for Hg). For the Hg vapor release from the liquid mixture and reductant solution, argon was bubbled to liberate and transport the Hg atoms into an absorption cell.

2.3. Quality control and quality assurance

Several quality assurance protocols were implemented to ensure the reliability of the preparation methods, analytical and precision and accuracy. Briefly, quality control was carried out by using certified reference standard solutions (1000 ppm) for each analyzed trace element, replicate control and replicated digestion and analysis. The detection limit of the methodology calculated using average dry weight of samples in mg kg^{-1}. The results are as follows: AL = Cu = Fe = Zn =1 mg kg^{-1}; Co = Cr = Hg = Li = Mo = Ni = Pb = Se = Sr = V = 0.1 mg kg^{-1}. The mean recoveries of trace elements for the above digestion method were within the acceptable range (80 % – 120 %) from certified reference materials.

2.4. Data analysis

Statistical analyses were carried out using SPSS software (version 22). The normality of the variables was checked using the Shapiro-Wilk test at the 95% confidence level prior to analysis. The data was not normally distributed. Thus, Kruskal-Wallis test was conducted to compare the differences in average concentrations of TEs in the tissues (muscle and liver) of the aquatic species investigated. The variations among trace element concentrations between muscles and liver tissue samples were examined using the Mann-Whitney U test.

2.5. Estimation of dietary intake and human health risk assessment

The health risk was evaluated using several approaches to assess the potential risks associated with consumption of the marine species and their chemical quality. Comparison of trace element concentrations in fish tissues and prawn with maximum permissible limit (MPL) as defined by international organizations for the TEs on a wet weight (ww) basis was assessed. Metal pollution index (MPI) used by Hao et al., (2013) was also calculated. To assess the risk of a potentially detrimental intake of individual elements, target hazard quotient (THQ) and hazard index (HI) were used (USEPA, 2002, 2010). Carcinogenic risk (CR) was also considered to evaluate the incremental probability of an individual to develop cancer by chronic daily intake of contaminated marine organisms by carcinogenic Ni, Pb and inorganic As (USEPA, 1989). THQ, HI and CR values were compared to reference and benchmark integrated USEPA risk analysis values (USEPA, 2002). In order to assess the health risks of dietary Pb exposure, margin of exposure (MOE) approach was carried out using the lower bound of the benchmark dose of extra risk (BMDL) (EFSA, 2005, 2010). All equations used in health risk assessment are presented in the Supplementary Information (SI). All stages of the research work were performed following guidelines of the ethical committee of the Shiraz University, Iran.

3. Results and discussion

3.1. Trace element concentrations in marine fish and prawn species

The concentration of the 15 selective TEs in muscle and liver tissues of the four analyzed marine species on a wet weight basis (mg kg^{-1} ww) are presented in Table 1. These TEs were selected based on their local occurrence, potential toxicity and nutritional value. The results revealed contamination by trace elements in the investigated marine species related to polluted ecosystem (Rahmanpour et al., 2014; Bastami et al., 2015; Zarezadeh et al., 2017). The Mann-Whitney analysis revealed that Al, Co, Fe, Hg, Mo and Se concentrations display significant differences ($p < 0.05$) between liver and muscle tissues of the investigated species (Table S2). The mean concentrations of trace elements including Cr, Fe, Mo, Pb, Se, and Zn were higher in the liver of *T. tonggol* than in the muscle tissues of targeted species. This result is consistent with those of Eastern Pacific Ocean fish species, where liver had the strongest enrichment of Pb and Zn (Moreno-Sierra et al., 2016). The results also revealed unequal accumulation of TEs in muscle and liver tissues from the marine species studied (Fig. 2). The highest Fe content was 92.77 mg kg^{-1} ww (mean of 72.66 mg kg^{-1} ww) in the liver tissues of *T. tonggol*. Also, in this species, Fe was 3–14 times greater than muscle tissue concentration compared to other studied species. Molybdenum was determined only in liver of *T. tonggol* (mean of 0.03 mg kg^{-1} ww). Molybdenum is probably involved in molybdopterin cofactor of certain enzymes and catalyse redox reactions (EFSA, 2013). Aluminium concentration varied in the muscle tissues between 0.16 mg kg^{-1} ww (*L. argentimaculatus*) to 11.44 mg kg^{-1} ww (*L. klunzingeri*), while it was not detected in *T. tonggol* (Table 1). Pb concentration varies between 0.021 and 0.71 mg kg^{-1} w.w in the liver tissue of *T. tonggol* compared to muscle tissues of other species, and is higher than in previous studies from the study area (Sobhanardakani et al., 2011). Similar results have been reported by other studies in different type of fish (Keshavarzi et al., 2018; Rajeshkumar & Li, 2018). Varol & Sünbül. (2020) measured higher Pb concentration in the liver tissues of trout barb compared to muscle tissues due to industrial and domestic wastewater discharges. It is important to note that the differences in the bioaccumulation of trace elements are partly due to differential TEs binding affinities in certain organs (Rainbow, 2007). Furthermore, the reason for the high accumulation of TEs in different tissues might be due to metallothionein induction. In aquatic environments, an excess amount of TEs increases production metallothionein which bind to the elements in order to detoxify them, and hence controls the tolerance capacity of aquatic species (Javed et al., 2015).

The most Cu accumulation occurs in the muscle tissues where *P.semisulcatus* recorded higher concentrations compared to other species (Table 1). According to Kruskal-Wallis analysis, there is no significant variations ($p > 0.05$) in trace elements and their concentration in the analyzed muscle, except for Cr, Li, Mo and Ni (Table S3). Li concentration ranges from 0.04 to 1.46 mg kg^{-1} ww (Table 1). As the oceanic Li content is homogeneous at 0.18 µg mL^{-1}, Thibon et al. (2021) believe that the observed range of Li concentrations in marine species must be related to biotic and abiotic processes in differing habitats (benthic, demersal, and pelagic) occupied by various species. Chassard-Bouchaud et al. (1984) showed that Li retention by aquatic species is not related to geography, though this could change due to wastes of the battery industry. The mean As content is the highest in muscle of *P.semisulcatus* followed by liver of *T. tonggol* (2.6 and 2.05 mg kg^{-1} respectively), while the lowest content occurs in muscle of *L. argentimaculatus* (0.18 mg kg^{-1}). These results are in good agreement with those previously reported in literature and similar to those reported in 10 species of fish from markets of the Shandong province, China (0.852 – 2.26 mg kg^{-1}) (Yang et al., 2021). Ruttens et al. (2012) reported a higher range of As levels (0.93 – 5.96 mg kg^{-1}) in marine fish species. In a recent study on As concentration in 19 species of wild marine organisms collected from Chinese Daya Bay (Zhang et al., 2018), the highest As content was found in crabs, followed by shrimps, benthic fish, and pelagic fish. Literature survey indicates different reasons including human activity, sediment pollution, sex, size, trophic level, geography of the sampling areas and ingestion of benthic prey may contribute to high As content in marine organisms. Demersal organisms are more susceptible to direct ingestion of As released from the sediment with the consumption of the benthic preys (Gusso-Choueri et al., 2018; Rahman et al., 2019; W. Zhang & Wang, 2012). In this study, the highest As content was found in muscle of *P.semisulcatus*, is attributed to this species benthic habitat (Table S1). Trace elements may also find their way into the Persian Gulfs' ecosystems through oil and gas refining industries, industrial zones and petrochemical industries, severe marine transportation in Strait of Hormuz, coastal zone power plants, cement factory, water desalination units, industrial and domestic discharge, and aluminum, lead and zinc facilities in Qeshm Island (Khatir et al., 2020; Rezaei et al., 2021; Zarezadeh et al., 2017). Response to demand for fresh water, some desalination plants are also installed in Qeshm Island. As a result, discharges from these plants during the desalination process and also brines in the area of pumping to the surrounding waters, and in time create a “salty desert” in the vicinity of the brine outfalls (Einav & Lokiec, 2003). Changes in overlying water salinity most likely influence the concentration and fate of TEs such as As in aquatic

environments (Azizur Rahman et al., 2012). Larsen & Francesconi, (2003) reported a positive correlation between salinity and arsenic burdens of marine fish species from the Baltic and the North Sea.

Muscular and liver Hg concentrations of the investigated species ranged from 0.029 mg kg^{-1} ww in *T. tonggol* muscle tissues to 0.24 mg kg^{-1} ww in tissues of muscle *L. argentimaculatus.* (Table 1). The difference in mercury concentration probably reflects different feeding behavior and habitat as *L. argentimaculatus* is a carnivorous species in the tropical regions, and feeds on crab, prawn and some fish and is commonly found in muddy and sheltered areas of mangrove ecosystems. A study by Chouvelon et al. (2009) in New Caledonia reported that total Hg content in *L. argentimaculatusus* varied between 0.403 and 0.994 mg kg^{-1} dw. Bosch et al. (2016) believe that the accumulation and effects of individual elements are not always independent of each other and may facilitate or decrease the uptake of other elements. Recently, Sobolev et al. (2019) have investigated Cu and Se detoxifying effect on Hg. Furthermore, some researchers have found Hg to be more easily enriched in muscle proteins (Ynalvez et al., 2016). In contrast, other studies (e.g. Kojadinovic et al., 2007; Storelli et al., 2005) reported that hepatic levels of Hg are higher than muscular ones. These contradictory views, indicate that bioaccumulation of trace elements in aquatic organisms differ, depending upon many factors including the physiological state of the marine species, the specific TE and its concentration in the water (Gedik et al., 2017). Olsvik et al., (2021) state that possible consequence of Hg bioaccumulation in hepatic tissue of *Brosme brosme* may be disruption of processes involved in fatty acid metabolism and osteogenesis. Marine species take up mercury mostly as dietary MeHg and it bioaccumulates in their muscles (Sakamoto et al., 2015). Aquatic organisms use different mechanisms of homeostasis and biotransformation to reduce the accumulation and effects of TEs (Cáceres-Saez et al., 2018). In this regard Se as an essential element can protect marine species from Hg toxicity through antioxidant defense mechanisms, competing for binding sites and formation of nontoxic complexes (Cuvin-Aralar & Furness, 1991). The –SeH group of selenocysteine or monomethylselenide can react with the –SH or –SeH group of a protein in an oxidation process and preventing $CH3Hg^{+}$ from binding to these groups (Yang et al., 2008). Liver is generally considered to be metabolically more active than muscle and plays an important role in metabolism of all substances that come via the blood. Liver is involved in different physiological processes involving absorption, accumulation, elimination, detoxification and storage of trace elements. Besides, liver commonly participates in the synthesis of metallothioneins (Amiard et al., 2006; Varol et al., 2020). The tissue-specific concentration of TEs in liver is mainly related to metallothioneins, proteins rich in cysteine (thiol (−SH) group) with natural capacity of elemental detoxification. These proteins of low molecular

weight have a high tendency to bind to Cd and Zn ions along with other ions such as Cu, Fe, and Ag (Klaassen et al., 1999). This high affinity is well reflected in the TEs considered in this study. The liver mainly has a high capability for Fe assimilation followed by Zn, reflecting their roles in blood cells and hemoglobin synthesis and bile secretion respectively (Görür et al., 2012). The mean concentration of Fe in liver of *T. tonggol* was 4.6, 12, 3.5 and was 15 times higher than muscle tissues of the analyzed species (Table 1, Fig. 2). The results exhibited significant differences in different analyzed species in terms of their muscle TEs accumulation potential. *L. argentimaculatus* showed strong bioaccumulation capability for Hg, and Cr; *L. klunzingeri* for Fe, Al, Li and V; *T. tonggol* for Pb and Se, and *P.semisulcatus* for As, Co, Cu, Sr, Ni and Zn. The analyzed *L. klunzingeri* in this research had lower Pb and Cu content than previously reported by Bastami et al. (2015). Saei-Dehkordi et al. (2010) detected Hg and As concentration in *T. tonggol* muscle from Bandar Abbas (Hg: 0.527 ± 0.190 and As: 0.220 ± 0.118 mg kg^{-1} ww), whereas in this study it was considerably lower. The reported TEs contents for *P. semisulcatus* (Akhbarizadeh et al., 2019; Heidarieh et al., 2013) are much higher than those found in this study. Anandkumar et al. (2019) suggest that trace elements accumulation in aquatic organisms directly occurs through the bioavailability of TEs from en route water and food chain, through gills during respiration or gastrointestinal tracts during feeding. Ruiz et al. (2018) believed that the pathways for trace elements uptake are related to the ecological lifestyle, life cycle, habitat and diet of the marine organism. Moreover, TEs bioaccumulation is influenced by some key factors including excretion and detoxification mechanisms and environmental factors such as salinity, temperature, dissolved organic matter and contaminant concentrations in the aquatic environment (Ho et al., 2021; Pilote et al., 2018; Yuan et al., 2020).

Table 1. Mean ± SD and ranges (min – max) of trace elements concentration in mg kg^{-1} ww in different marine species (< DL: below detection limit).

Species		TEs														
		Al	As	Co	Cr	Cu	Fe	Hg	Li	Mo	Ni	Pb	Se	Sr	V	Zn
T. tonggol	Muscle	< DL	1.45 ± 0.05	0.006± 0.002	0.13 ± 0.02	2.22 ± 0.24	15.81 ± 2.7	0.03 ± 0.001	0.10 ± 0.07	< DL	0.06 ± 0.03	0.32 ± 0.19	0.08 ± 0.01	0.42 ± 0.21	0.009 ± 0.002	4.96 ± 0.17
		< DL	1.40 - 1.49	0.004 - 0.008	0.11 - 0.15	2.08 - 2.5	14.14 - 18.9	0.029 - 0.03	0.02 - 0.15	< DL	0.04 - 0.1	0.15 - 0.53	0.07 - 0.09	0.21 - 0.62	0.006 - 0.01	4.82 - 5.16
	Liver	< DL	2.05 ± 0.31	0.03 ± 0.01	0.2 ± 0.06	2.91 ± 0.83	72.66 ± 17.73	0.029 ± 0.001	0.104 ± 0.102	0.03 ± 0.01	0.08 ± 0.01	0.27 ± 0.38	0.26 ± 0.01	4.23 ± 2.34	0.019 ± 0.004	24.27 ± 4.56
		< DL	1.72 - 2.33	0.02 - 0.04	0.14 - 0.26	2.08 - 3.74	59.28 - 92.77	0.027 - 0.029	0.011 - 0.21	0.02 - 0.04	0.07 - 0.09	0.05 - 0.71	0.25 - 0.27	1.66 - 6.24	0.015 - 0.022	21.64 - 29.54
L. argentimaculatus	Muscle	0.51 ± 0.5	0.18 ± 0.08	0.003 ± 0.002	0.15 ± 0.06	1.25 ± 0.3	6.03 ± 2.9	0.112 ± 0.09	0.04 ± 0.06	< DL	0.07 ± 0.01	0.022 ± 0.025	0.05 ± 0.005	1.3 ± 0.80	0.007 ± 0.003	6.9 ± 2.8
		0.16 - 1.25	0.11 - 0.28	0.002 - 0.01	0.13 - 0.16	1.04 - 1.66	1.66 - 7.70	0.06 - 0.24	0.002 - 0.123	< DL	0.06 - 0.09	0.021 - 0.03	0.05 - 0.062	0.42 - 2.1	0.004 - 0.01	3.97 - 9.8
L. klunzingeri	Muscle	6.17 ± 4.57	0.93 ± 0.11	0.009 ± 0.003	0.16 ± 0.022	1.73 ± 0.4	20.8 ± 9.05	0.038 ± 0.005	1.46 ± 0.47	< DL	0.08 ± 0.02	0.07 ± 0.06	0.03 ± 0.008	3.95 ± 0.72	0.021 ± 0.005	14.43 ± 1.4
		3.33 - 11.44	0.81 - 1.03	0.006 - 0.012	0.14 - 0.18	1.25 - 2.1	14.77 - 31.2	0.03 - 0.044	0.99 - 1.93	< DL	0.06 - 0.1	0.02 - 0.13	0.023 - 0.04	3.12 - 4.37	0.02 - 0.03	13.13 - 15.9
P.semisulcatus	Muscle	1.87 ± 0.62	2.6 ± 0.67	0.08 ± 0.008	0.1 4 ± 0.02	9.43 ± 4.6	4.85 ± 0.84	0.06 ± 0.02	0.05 ± 0.03	< DL	0.11 ± 0.01	0.11 ± 0.06	0.06 ± 0.02	13.94 ± 5.77	0.014 ± 0.001	23.14 ± 0.94
		1.25 - 2.5	1.98 - 3.31	0.008 - 0.02	0.12 - 0.16	4.2 - 12.5	4.37 - 5.82	0.052 - 0.08	0.02 - 0.09	< DL	0.10 - 0.12	0.06 - 0.18	0.05 - 0.09	10.4 - 20.6	0.012 - 0.015	22.13 - 23.97

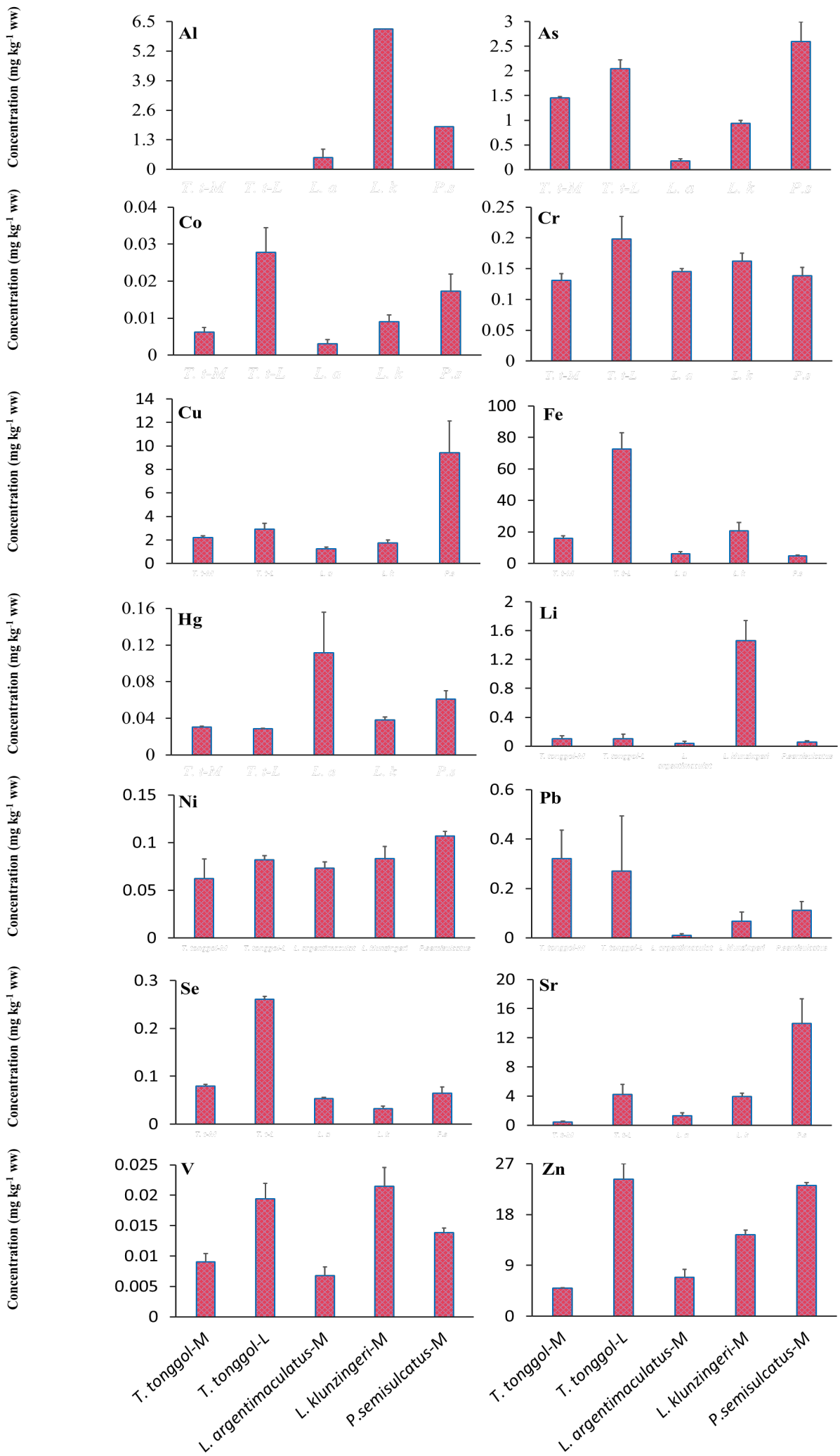

Fig 2. Comparison of trace element concentrations (mean ± standard error, mg kg^{-1} wet weight) in the investigated species from Persian Gulf ("M and L" abbreviations used to muscle and liver respectively).

Table 2. Metal pollution index (MPI), target hazard quotient (THQ) and hazard index (HI) of TEs (µg kg^{-1} bw. day^{-1}) for adults and children (in the examined species, based on TE concentrations on wet weight basis). Bold highlighted values exceed recommendations (<DL: below detection limit).

TEs	Species									
	T. tonggol-M *		*T. tonggol*-L*		*L. argentimaculat*us-M*		*L. klunzingeri*-M*		*P.semisulcatus*-M*	
	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children
Al	< DL	< DL	< DL	< DL	3.59E-04	6.34E-04	4.37E-03	7.71E-03	7.07E-04	1.21E-03
As [a]	3.42E-01	6.04E-01	4.83E-01	8.53E-01	4.18E-02	7.37E-02	2.21E-01	3.89E-01	3.27E-01	5.58E-01
Co	1.47E-02	2.60E-02	6.55E-02	1.16E-01	7.37E-03	1.30E-02	2.13E-02	3.76E-02	2.18E-02	3.72E-02
Cr	3.10E-02	5.46E-02	4.68E-02	8.26E-02	3.43E-02	6.05E-02	3.83E-02	6.76E-02	1.75E-02	2.98E-02
Cu	3.93E-02	6.93E-02	5.16E-02	9.10E-02	2.21E-02	3.90E-02	3.07E-02	5.42E-02	8.90E-02	1.52E-01
Fe	1.60E-02	2.82E-02	7.36E-02	1.30E-01	6.11E-03	1.08E-02	2.11E-02	3.71E-02	2.62E-03	4.47E-03
Hg	1.35E-01	2.38E-01	1.26E-01	2.22E-01	4.95E-01	8.73E-01	1.69E-01	2.98E-01	1.44E-01	2.46E-01
Li	3.59E-02	6.33E-02	3.68E-02	6.50E-02	1.47E-02	2.60E-02	5.18E-01	9.13E-01	1.02E-02	1.74E-02
Mo	< DL	< DL	3.73E-03	6.59E-03	< DL	< DL	< DL	< DL	< DL	< DL
Ni	2.21E-03	3.90E-03	2.90E-03	5.11E-03	2.56E-03	4.52E-03	2.95E-03	5.20E-03	2.02E-03	3.44E-03
Pb	6.33E-02	1.12E-01	5.32E-02	9.39E-02	4.40E-03	7.76E-03	1.31E-02	2.31E-02	1.16E-02	1.99E-02
Se	1.13E-02	1.99E-02	3.69E-02	6.52E-02	7.96E-03	1.40E-02	4.62E-03	8.15E-03	4.87E-03	8.31E-03
Sr	4.91E-04	8.67E-04	4.99E-03	8.81E-03	3.01E-03	5.31E-03	4.67E-03	8.23E-03	8.77E-03	1.50E-02
V	1.28E-03	2.25E-03	2.75E-03	4.85E-03	1.33E-03	2.34E-03	3.05E-03	5.37E-03	1.05E-03	1.79E-03
Zn	1.17E-02	2.07E-02	5.73E-02	1.01E-01	1.46E-02	2.58E-02	3.41E-02	6.02E-02	2.91E-02	4.97E-02
MPI	1.03		1.65		0.66		1.84		1.92	
HI	0.71	**1.24**	**1.05**	**1.84**	0.66	**1.16**	**1.09**	**1.92**	0.67	**1.14**

[a] 10% of total As concentration (mg kg^{-1} ww) was assumed as inorganic As used for THQ and HI assessment.

* "M and L" abbreviations used to muscle and liver respectively.

3.2. Nutritional and Toxicological risk assessment of the trace elements

To accurately evaluate health risk arising from consumption of marine species in the study area, the mean concentrations of the trace elements in muscle and liver of the analyzed samples were compared to those given in international permissible criteria. The results indicate that accumulation of As and Pb in the liver of *T. tonggol*, which is consumed by locals, is higher than the maximum permissible levels specified by international guidelines (USEPA, 2015). Moreover, mean and range of As level found in the liver of *T. tonggol* and muscle tissues of all the investigated species with the exception of *L. argentimaculat*, and Cu in the prawn species exceed the recommended maximum levels established by international organizations (EC, 2006; FAO/WHO, 2008; USEPA, 2015). Although most TEs found in the sampled species are lower than the recommended legal limit for seafood (FAO/WHO, EC, and EPA standard), they may cause some health risks to humans through trophic biomagnification due to higher concentration (Zimmermann & Sures, 2018).

To examine the accumulation levels of the TEs in the sampled marine species tissues, the metal pollution index (MPI) was calculated (Table 2). MPI values in the muscle tissues is maximum in *P.semisulcatus* and minimum in *L. argentimaculatus*. MPI in liver tissue is higher than in muscle tissues in *T. tonggol*. MPI values higher than 2 indicates contamination, while MPI < 2 suggests not impacted (Jamil et al., 2014). In this study, MPI values in the muscle tissues were higher than those reported from fish in the Persian Gulf, and Turkey (Soltani et al., 2021; Töre et al., 2021). Considering the habitat, the order of MPI in the tested marine species was benthic prawn (mean: 1.92) > demersal fish (mean: 1.84) > pelagic fish (mean:1.65 in liver, 1.03 in muscle) > and reefal fish (mean: 0.66). This pattern probably reflects higher possibility of exposure to sediments-bound TEs and higher ingestion rate of contaminated benthic preys (Gu et al., 2015).

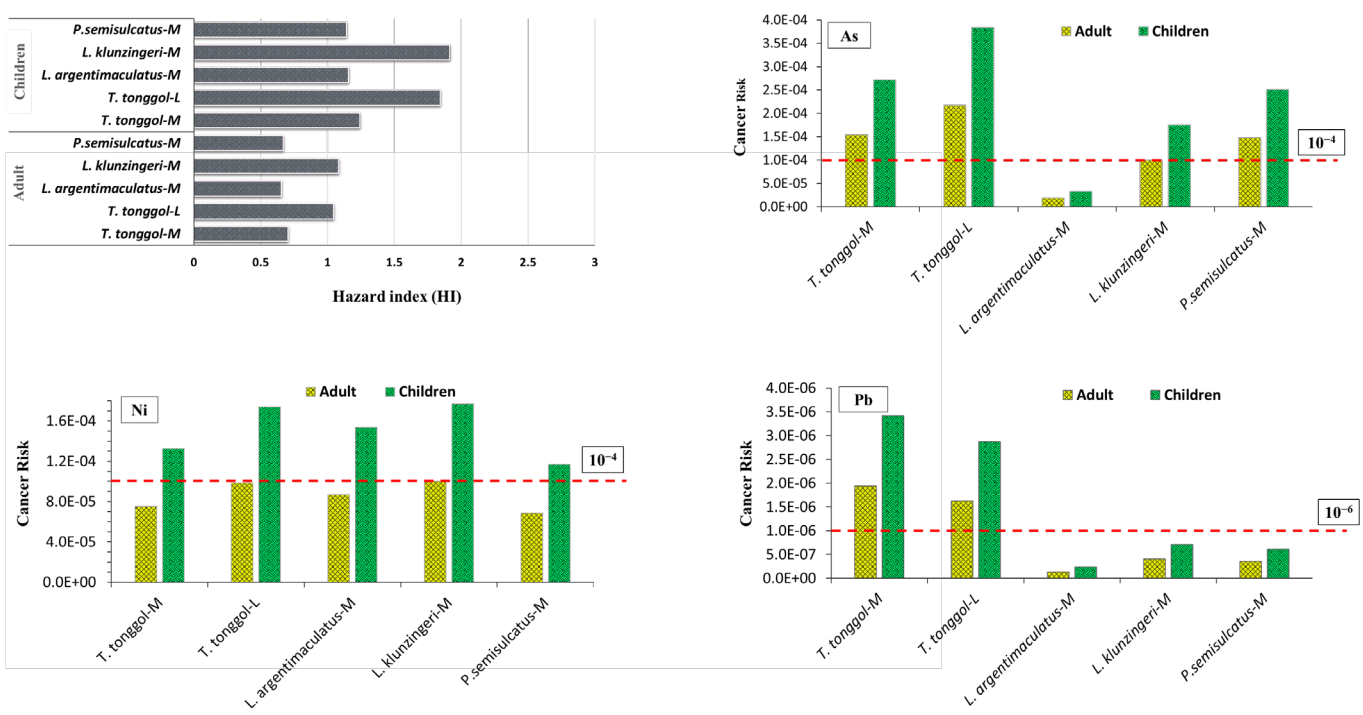

Fig 3. Carcinogenic risk (CR) of As, Ni, and Pb of targeted marine species in adults and children, arising from the consumption of marine species.

Target hazard quotient (THQ) is also estimated as a means of assessing the potential risk of adverse health effects arising from long-term exposure to trace elements (Table 2). The estimated THQ of all trace elements in tissues of the analyzed marine species for both considered groups, regardless of whether local population eat *T. tonggol* liver, is < 1, which suggest that TEs in all the collected marine species from sampling locations pose no chronic noncarcinogenic health hazard (Table 2). However, cumulative health risk arises when consumer population are exposed to multiple TEs from consuming marine species due to their combined effects. Therefore, the hazard index (HI) was determined as the sum of the effects of mixed trace elements and it ranged

from 0.66–1.09 for adult and 1.14–1.92 for children. This means that health risk faced by children is 1.7 times that of adults (Table 2, Fig. 3). Arsenic and Hg are the major risk contributors with maximum HI values in all the species considered in this study, except for *L. klunzingeri*.

Table 3. Estimated Margin of Exposure (MOE) using the lower confidence limit of the benchmark dose (BMDL) of an extra risk of 1% ($BMDL_{01}$) for adverse effect on developmental neurotoxicity and the BMDL for an extra risk of 10% ($BMDL_{10}$) for the nephrotoxicity) using either mean, median (P50) and 95 percentiles (P95) values of estimated daily intake (EDI) in adults and children. EDI and BMDL are in µg kg^{-1} bw day^{-1}; The MOE values lower than the threshold (MOE < 1) are in bold and underlined.

Species		Age Group	Mean	P50	P95
T. tonggol-M	EDI	Adult	0.228	0.065	0.34
		Children	0.40	0.12	0.60
	MOE	Adult	2.76	9.69	1.85
		Children	1.24	4.35	**0.83**
T. tonggol-L	EDI	Adult	0.192	0.128	0.67
		Children	0.34	0.23	1.18
	MOE	Adult	3.28	4.92	**0.94**
		Children	1.48	2.22	**0.42**
L. argentimaculatus-M	EDI	Adult	0.016	0.007	0.028
		Children	0.03	0.013	0.05
	MOE	Adult	39.38	90.0	22.5
		Children	17.89	38.46	10.0
L. klunzingeri-M	EDI	Adult	0.047	0.022	0.114
		Children	0.08	0.039	0.20
	MOE	Adult	13.40	28.64	5.53
		Children	6.01	12.8	2.50
P.semisulcatus-M	EDI	Adult	0.042	0.021	0.108
		Children	0.07	0.038	0.19
	MOE	Adult	15.0	30.00	5.83
		Children	6.99	13.2	2.6

"M and L" abbreviations used to muscle and liver respectively.

Figure 3 displays the possibility of exposed consumers of the marine species developing cancer as a result of lifetime exposure to carcinogenic trace elements. It can be seen that cancer risk of inorganic As and Ni ranges from 1.88×10^{-5} to 3.84×10^{-4} and 6.85×10^{-5} to 1.77×10^{-4}, respectively, and both are above the acceptable lifetime risk in most species for adults and children. However, the CR values for Pb (1.35×10^{-7} to 3.42×10^{-6}) fell within the acceptable to negligible range, indicating that Pb concentration in marine species in the study area is insufficient to pose carcinogenic risk. The results are comparable to those reported by Storelli et al. (2020) and Yuan et al. (2020). Again, children are exposed to the highest CR from As through consumption of *T. tonggol* liver followed by the muscle of the same species. The results suggest that children who consume the examined marine species have disproportionately significant potential carcinogenic and non-carcinogenic risks from the exposure to trace elements than adults, despite the fact that both age groups are subjected to the same exposure pathways (Rauh & Margolis, 2016). In fact, rapid growth rate of children is accompanied by aspects of potential vulnerability and thus will affect the normal body functions. Previous studies also reported that TEs

can pose potential risks to humans arising from consumption of marine species with elevated TEs levels (Bosch et al., 2016; Varol & Sünbül, 2020). In this regard, arsenic is particularly important as it can cause spotted melanosis, infertility, lung cancer and cardiovascular diseases (Occupational Safety and Health Administration, 2004). In this study, Hg content in the marine species is high, thus, it presents a concern because the estimated magnitude of Hg exposure is very significant since the levels of THQ were only marginally lower than the threshold (>1). However, Hg exposure estimation is prone to uncertainties including whether total Hg is present in the marine species is in its toxic methylmercury (MeHg) form or seafood cooking can effectively mitigate this risk by its volatilization during cooking (Marques et al., 2011). Table 3 displays the estimated daily intake of lead for the investigated marine species, along with the calculated Margin of Exposure (MOE). Mean overall lead exposure via consumption of the studied marine species is 0.145 µg kg^{-1} bw (body weight) day^{-1}. The lowest daily intake of lead (0.016 µg kg^{-1} bw day^{-1}) is about 2.5% of the lower limit of the EFSA reference interval point (0.63 µg kg^{-1} per day) is evaluated by consumption of *L. argentimaculatus* in adults. The highest intake of lead (0.40 µg kg^{-1} bw day^{-1}) arises from consumption of *T. tonggol* muscle in children and corresponds to 80% of the lower limit of the EFSA reference point interval (0.5 µg kg^{-1} bw day^{-1}). The difference in the exposure to Pb from aquatic organisms depends on both the difference in individual consumption, and the concentration of Pb in the consumed species. Even though lead is normally stored in tissues such as bone, blood, kidney, and liver, but its distribution and metabolism in exposed organisms is commonly associated with calcium metabolism and it is not biomagnified in the food-chain (WHO, 1989). Despite international organizations' attempts to list health-based thresholds for tolerable dietary exposure to lead, the available evidence does not allow to set a level of exposure for critical lead-induced effects below that no harmful effects are expected (EFSA, 2012; WHO, 2016). The MOE values for both adverse effect of neurotoxicity development (Neu) in children and nephrotoxicity (Nep) in adults are well above the MOE threshold of 1 set by EFSA (EFSA, 2010) using either the mean (2.76-39.38 for Nep, 1.24-17.89 for Neu) or the median (P50) (4.92-90 for Nep, 2.22-38.46 for Neu) estimates of daily intake (EDI), suggesting low overall risk to human health from the intake of Pb from marine species in the proximity of Qeshm Island ecosystems, which is hardly surprising, given its low solubility. Furthermore, calculated MOE with the upper confidence limit (95%) of Pb, indicate that the lowest MOE values are for children (0.42) associated with liver of *T. tonggol*. The next lower MOEs was calculated for children (0.83) and in adults (0.94) for muscle tissues of *T. tonggol*. The other MOE values were > 1 in all species indicating a negligible health risk (Table 3). Additional sources of Pb exposure, such as dust inhalation, dermal contact and

consumption of non-marine foodstuffs suggest a higher degree of Pb exposure and bioaccumulation in human, and they should be considered. Additionally, several behavioral characteristics including outdoor activity, hand-to-mouth activities, insufficient attention to hygiene conditions and greater absorption of lead from gastrointestinal tract make can children vulnerable to lead poisoning (JECFA, 2000), hence their developing nervous system (central and peripheral) is more vulnerable to lead toxic effects (Dórea, 2019). However, the exposure level for chronic kidney diseases is much higher than those associated with the developmental of neurotoxicity adverse effects (EFSA, 2012; WHO, 2011).

4. Conclusion

Analyses of aquatic organisms from north Persian Gulf ecosystem and assessment of the potential risks, indicate that TEs concentrations vary considerably among the marine species investigated. Liver tissues of *T. tonggol* revealed the highest concentrations for most elements, followed by prawn species. The evaluated potential health risk of TEs in the four targeted species (*T. tonggol*, *L. argentimaculatus*, *L. klunzingeri*, and *P. semisulcatus*) including THQ, HI and CR suggest that the exposure doses of most elements for human consumption are safe for non-carcinogenic and carcinogenic risk, except for As and Ni with high CR values. Using a probabilistic approach for estimating human Pb exposure from marine species, revealed that the risk arising from Pb is the greatest health concern for those consuming large amounts of muscle and liver of *T. tonggol* species. This study shows that *T. tonggol* species can be used to study bioaccumulation and TEs toxicity in coastal ecosystems.

Author contributions

All authors discussed the results and contributed to the final manuscript.

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Supplementary Information

Bioaccumulation of selected trace elements in some aquatic organisms from the proximity of Qeshm Island ecosystems: Human health perspective

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Table S1. Morphometric measures, trophic level, and biological background information of the four investigated species.

Type	Scientific name	Common name	Tissue	n	Length (cm)		Weight (g)		Feeding habit	Habitat	TL*
					min-max	Mean	min–max	Mean			
Fish	*Thunnus tonggol*	Tuna Longtail	Muscle	(8[a], 3[b])	47-53	50.5	1668-2375	1992.9	Omnivorous	Pelagic	4.5
			Liver	(8, 3)							
	Lutjanus argentimaculatus	Mangrove red snapper	Muscle	(20, 4)	25-35	31.05	196.9-612	434.87	Carnivorous	Neritic	3.6
	Liza klunzingeri	Klunzinger's mullet	Muscle	(40, 3)	12.5-18.5	14.5	26.7-82.8	41.81	Omnivorous	Demersal	2.6
Prawn	*Penaeus semisulcatus*	Green tiger prawn	Muscle	(300, 3)	6-14.5	9.32	1.6-25.3	6.02	Carnivorous	Benthic	2.7

[a] Number of specimen of each species.

[b] Number of composite sample.

*Trophic level refers to (FishBase, 2021)

Table S2. Comparison of TE concentrations in the muscle and liver tissues of four analyzed organisms based on the Mann Whitney U test.

	AL	As	Co	Cr	Cu	Fe	Hg	Li	Mo	Ni	Pb	Se	Sr	V	Zn
Mann-Whitney U	4.5	7	2	7	11.5	0	1	18.5	0	17.5	17	0	16	7	6
Wilcoxon W	10.5	98	93	98	102.5	91	7	24.5	91	23.5	108	91	107	98	97
Z	-2.074	-1.682	-2.375	-1.684	-1.086	-2.627	-2.500	-0.135	-3.848	-0.270	-0.338	-2.625	-0.472	-1.701	-1.816
Asymp. Sig. (2-tailed)	0.038	0.093	0.018	0.092	0.278	0.009	0.012	0.893	0.000	0.787	0.736	0.009	0.637	0.089	0.069
Exact Sig. [2*(1-tailed Sig.)]	**0.039b**	0.111b	**0.014b**	0.111b	0.296b	**0.004b**	**0.007b**	0.900b	**0.004b**	0.800b	0.800b	**0.004b**	0.704b	0.111b	0.082b

Table S3. Comparison of TE concentrations in analyzed organisms muscle based on the Kruskal-Wallis test.

#	Al	As	Co	Cr	Cu	Fe	Hg	Li	Mo	Ni	Pb	Se	Sr	V	Zn
Chi-Square	11.208	11.275	8.864	3.457	10.423	9.674	10.293	7.713	0	6.171	8.852	8.331	9.346	8.921	9.89
df	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
Asymp. Sig.	**0.011**	**0.01**	**0.031**	0.326	**0.015**	**0.022**	**0.016**	0.052	1	0.104	**0.031**	**0.04**	**0.025**	**0.03**	**0.02**

Human health risk assessment of trace elements in organisms

Nutritional assessment of the trace elements:

The human health risk posed by TEs was determined based on approaches that consider:

– **Metal pollution index (MPI)**

To estimate the overall TE load for seafood muscle and liver in marine organisms, metal pollution index (MPI) was calculated based on the equation given by Sharma et al. (2008):

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

where M_n is the concentration of n trace element in the sample (mg kg^{-1} dry weight).

- Non-carcinogenic risk assessment

Target hazard quotient (THQ, µg kg^{-1} bw. day^{-1}) was used to calculate the risk of non-carcinogenic effects for humans from each TE via seafood consumption (Eq. (2)):

$$THQ = \frac{E_F \times E_D \times FIR \times C_m \times C_f}{BW \times A_{Tn} \times RfD} \times 10^{-3} \quad (2)$$

Hazard index (HI) is the sum of THQs obtained for all the trace elements assuming the dose addition of individual (USEPA, 2013) (Eq. (3)):

$$HI = HQ_1 + HQ_2 + \cdots + HQ_n \quad (3)$$

where C_m is TE concentration in seafood (mg kg^{-1}, wet weight); FIR (g day^{-1}) is the ingestion rate based on local consumption (adult, 49.6 g $capita^{-1}$ day^{-1} seafood per day; child, 20 g seafood per day based on the General Department of Fisheries of Hormozgan province (GDOFHP, 2010); E_F is the exposure frequency (365 days $year^{-1}$); C_f is the conversion factor to convert wet weight from dry weight considering 79% moisture content of the seafood muscle (Saha et al., 2016). E_D is the exposure duration (70 years for adults, 6 years for children); RfD is oral reference

dose (in mg kg^{-1} bw day^{-1}) (*see table below); BW is the body weight (70 kg for adults and 16 kg for children) (USEPA, 2013); and AT is average time and equal to the life expectancy over which cumulative exposures are averaged (365 days $year^{-1}$ × number of exposure years) (Griboff et al., 2018).

*Reference doses of trace elements in human beings.

TEs	Al	As	Co	Cr	Cu	Fe	Hg	Li	Mo	Ni	Pb	Se	Sr	V	Zn
R_fD	1	0.0003	0.0003	0.003	0.04	0.7	0.00016	0.002	0.005	0.02	0.0036	0.005	0.6	0.005	0.3

* Reference doses (RfD; mg kg^{-1} bw day^{-1}) of elements were established by the (USEPA, 2015).

If the THQ value is > 1, it means that the consumption of contaminated organisms by the single or combine toxic effects of TEs has a potential non-carcinogenic health risk to human health (Yuan et al., 2020). It is important to mention here that TE concentrations from a dry weight (dw) basis were multiplied by the moisture factor (0.208) to express them on wet weight (ww) basis (Baki et al., 2018). Furthermore, since most As content in seafood is considered in less harmful organic forms, all consumption limit calculations were made assuming 10% toxic inorganic As in total (Lorenzana et al., 2009).

– **Carcinogenic risk (CR)**

The incremental lifetime cancer risk indicates the probability of an individual developing cancer during one's lifetime as a result of a specific exposure to a carcinogenic compound (USEPA, 1989). Carcinogenic risk (CR, µg kg^{-1} bw. day^{-1}) was determined in the following way (Eq. (4)) (USEPA, 2006):

$$CR = EDI \times CPSo \quad (4)$$

USEPA recommends that oral carcinogenic slope factor values (CPSo) for inorganic As, Ni and Pb to be 1.5, 1.7 and 0.0085 mg kg^{-1} day^{-1}, respectively (USEPA, 2016). Estimated Daily Intake (EDI, µg kg^{-1} bw. day^{-1}) of each analyzed TE through seafood consumption was calculated using the following equation:

$$EDI = \frac{C_m \times FIR}{BW} \quad (5)$$

According to the US EPA method (USEPA, 2016), a CR value of less than 10^{-6} is considered as insignificant and to be negligible, above 10^{-4} is considered as harmful and the cancer risk is troublesome, while CR levels between 10^{-4} to 10^{-6} are considered to be acceptable (USEPA, 1989, 2010).

Toxicological risk assessment of the trace elements

The Pb risk of dietary exposure was performed by considering a margin of exposure (MOE) approach as recommended by the European Food Safety Authority (EFSA) to risk assessment of substances that are both genotoxic and carcinogenic. The MOE is determined as the ratio between a reference value, such as the lower confidence limit of the benchmark dose (BMDL), and an exposure level (EFSA Scientific Committee et al, 2017):

$$MOE = \frac{BMDL}{EDI} \quad (6)$$

where benchmark doses for dietary intake of Pb based on corresponding lower limits of a one-sided 95% confidence interval on the BMD ($BMDL_{01}$, $BMDL_{10}$) of 0.5 and 0.63 µg kg^{-1} body weight per day for developmental neurotoxicity in children and nephrotoxicity in adults, respectively. MOE value less than the unity (< 1) suggests a high health risk whereas MOE > 1 indicates an acceptable low risk to occur (EFSA, 2005). MOE and EDI were calculated using the mean, 50th and 95th percentiles values (abbreviated as P50 and P95, respectively) of TEs concentration in the targeted species.

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