

LEAD, CADMIUM, ZINC, AND COPPER CONCENTRATIONS IN DIFFERENT ARABIAN GULF FISH SPECIES

BY

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ABSTRACT

Background: While fish is well-known for its nutritional importance due to the omega-3 fatty acids, vitamins, and high-quality proteins, it might pose dangers such as heavy metals that may present a serious threat to fish consumption. Numerous heavy metals are believed to contribute to various physiological abnormalities, including renal failure, and when consumed above legal limits, they may cause morbidities or mortality. Increased concentrations of metals in aquatic species result from atmospheric pollution and industrial effluents continuously draining into nearby water bodies. When metals surpass their allowed limits, a severe health risk might ensue. **Aim of the Work:** The assessment of four metals' profiles, namely; Cadmium (Cd), Copper (Cu), lead (Pb), and Zinc (Zn), in some selected commonly consumed fish species caught from Qatif, Dammam, and Jubail in the Eastern province of Saudi Arabia. **Material and Methods:** Several marine fish species from the Arabian Gulf area were analyzed using inductively coupled plasma-mass spectrometry (ICP-MS). Quantitative evaluation of Copper (Cu), lead (Pb), Cadmium (Cd), and Zinc (Zn) was done to estimate the potential toxicity of four metals to humans via consumption of fish caught from the three largest zones of the Saudi Arabian Eastern Province (Qatif, Dammam, and Jubail). **Results:** The investigated fish tissues in Dammam, Qatif, and Jubail had Cu levels between (1.13-2.38), (1.01-2.12), and (1.17-2.97) simultaneously. Pb ranged from 0.03-0.87, 0.01-0.19, and 0.01-0.30 for the same locations and marine species. Zn and Cd were (0.05-0.85) and (0.01-0.18) in Dammam, (0.03-0.92), (0.02-0.24) in Qatif, and (0.06-0.99), (0.01-0.67) in Jubail. Jubail recorded significantly higher metal concentrations than the other two regions. There was a statistically significant difference between the Cd concentrations in marine species and tissues in Jubail and those in Dammam and Qatif for the same species and tissues. The highest recorded levels of Cd were found in the eggs of mullet > shrimp > summer flounder > cuttlefish in the Jubail region. **Conclusions:** The current findings imply the necessity for toxicological evaluations of human populations in the Jubail region who consume substantial quantities of fish for indicators of heavy metal toxicity. It is recommended that more research be conducted on the hazardous effects of metal contamination through fish consumption in various Eastern Province regions. Establishing relevant pollution criteria requires realistically evaluating trace metals as environmental toxicants.

Keywords: Toxic Heavy metals; lead; Copper; Zinc; Cadmium; Analytical Toxicology; Saudi Arabia.

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INTRODUCTION

Eastern Saudi Arabia borders the Arabian Gulf. Water pollution has recently increased due to high water evaporation, protracted drilling, and oil extraction. These activities dump large amounts of effluent into the shallow, semi-closed Gulf, disrupting the coastal environment. Heavy metals in natural streams are problematic. Toxic metals can spread from aquatic ecosystems to humans

through many biological networks (*Zyadah, and Almoteiry, 2012*). Sediments are the main conduit for metal pollution and fish heavy metal uptake. Fish that consume organic matter and seafloor invertebrates absorb heavy metals in sediments (*Rahman, et al., 2012*).

Coastal residents eat fish, crustaceans, and molluscs for protein. It is an inexpensive source of vitamins and minerals with low fat

and calories. Micronutrients like Copper, zinc, iron, and selenium and macronutrients such as carbohydrates, proteins, fatty acids, and polyunsaturated fatty acids can all be obtained from fish-containing meals. Human health requires micro and macronutrients. Unfortunately, elevated heavy metal concentrations in aquatic systems can chemically contaminate fish (Arul-Kumar, et al., 2017; Liu, et al. 2018). Technology, industrial development, rising residential waste due to population growth, and industrial waste are drained into water bodies and accumulated in marine creatures worldwide (Ahmed, et al. 2016; Yap and Al-Mutairi, 2022) Thus, vulnerable populations like infants and pregnant women should be constantly assessed for the health hazards of increased fish consumption due to seafood's chemical pollutants (Martorell et al. 2011; Alharbi, et al. 2017) Heavy metals like cadmium and lead accumulate and may be hazardous when they exceed the acceptable daily limits since consumers' bodies cannot remove them. Heavy metal buildup in fish tissues and organs depends on species, metal type, environmental concentration, age, ambient temperature, salinity, and pH. Thus, heavy metal concentrations in marine animals from diverse coastal locations and fish sections are a global priority for periodic risk evaluations (Velusamy, et al. 2014; Sarkar, et al. 2019).

Iron (Fe), Copper (Cu), and zinc (Zn) are essential to the metabolism of fish. In contrast, other metals, including cadmium (Cd), mercury (Hg), and lead (Pb), serve unknown functions in their biological system (Arul-kumar et al., 2017). Heavy metals such as Cd, Cu, Pb, and Zn are the most common contaminants in fish and fish products. According to Sivaperumal et al. (2007), necessary metals can cause toxicity when they are above their permissible levels. Food contamination with (Pb) causes lethargy, irritation, cancer, and myalgia. Lead toxicities affect the kidneys, liver, and brain. Cadmium toxicity caused Japanese Itai-Itai sickness, bone disease, anosmia, and tooth discoloration (Selda, and Nursah, 2012; Bosch, et al. 2016). Although the micronutrients Zn and Cu are required for

healthy biological functions and must therefore be consumed by humans, they can be dangerous when consumed in large quantities (Mielcarek, et al. 2020). Various analytical methods have been developed to determine heavy metals in fish. The most common and widely used technique is inductively coupled plasma-mass spectrometry (Selda, and Nursah, 2012; Raknuzzaman, et al. 2016).

THE AIM OF THE WORK

This study examined the concentrations of four metals— Cadmium (Cd), Copper (Cu), lead (Pb), and Zinc (Zn)—in some of the most popularly consumed fish species from Qatif, Dammam, and Jubail in the Eastern Province of Saudi Arabia. Summer flounder, Striped-red mullet, Marbled spinefoot, Gilt-head bream, Bluefish, Mullet, Drummer blackfish, crustaceans (shrimp and crabs), and squid (cuttlefish) were examined to assess their content of the prementioned metal levels in their muscles, eggs, and viscera.

MATERIAL AND METHODS

Seven of the most consumed types of fish species in the Eastern region (Summer flounder, Striped-red mullet, Marbled spinefoot, Gilt-head bream, Bluefish, Mullet, Drummer blackfish), two crustaceans (shrimps and Crabs), and cuttlefish samples (3-4 specimens of each type) were purchased from the three local fish markets of the Eastern Province Governorates (Dammam, Qatif, and Jubail) under study as seen in figure (1).



Figure (1): The three study sites of the Eastern Region of Saudi Arabia (Dammam, Qatif, and Jubail) are marked with arrows on Google Maps.

After their purchase, all samples were rapidly transferred to the laboratory in ice boxes. Fish tissues degrade over time, affecting metal concentrations in different tissues. Enzymatic activity and microbial breakdown can change metal concentrations in fish tissues. Fish metabolism and environmental circumstances can also affect metal buildup. This highlights the importance of properly handling and preserving fish samples to reduce the impact of sample freshness on metal assay results. This involves optimal storage temperature, minimizing sample collection and analysis delays, and avoiding sample contamination (Bawuro, *et al.* 2018).

Each sample's muscles, eggs, and viscera were cut into fine pieces with a stainless-steel scalpel. A two-gram specimen was accurately weighed and transferred to a clean digestion vessel from each sample of the studied fish type. The samples were initially digested with 5 mL of 65% nitric acid in digestion vessels sealed with glass funnels to permit nitric acid reflux during the digestion. They were retained in the microwave digester at 120°C for five hours. The resulting digested sample was diluted to 25 ml with ultrapure water and filtered to remove insoluble materials such as tissue debris or lipids. All digested samples were kept at four degrees Celsius until the analysis time. For the simultaneous assay of Cd, Cu, Pb, and Zn, an Agilent Technologies Inductively Coupled Plasma Mass Spectrometer (ICP-MS) equipped with an auto-sampler, nebulizer, spray chamber, and re-circulating refrigeration was utilized. The concentration of all metals in each sample was evaluated in triplicate. Before use, every piece of glassware and plastic was cleansed for 15 minutes in a nitric acid solution and then washed with deionized water. The analysis used analytical grade reagents (Merck; Germany) and deionized water. The analytical technique-certified reference material (CRM 320, Merck KGaA, Darmstadt, Germany) was utilized for validation and precision. Using the proper analytical method, the recovery rate of the selected metals ranged between 79% and 107%. In addition, contamination during the analytical process was abolished, and the relative standard deviation (RSD) for each

test was 10%. The calibration curves for the four elements under study were developed at seven concentration levels. To assure linearity, the coefficient determination of the calibration curves was ≥ 0.99 for all examined metals. The standard solutions and reagents utilized in this research were bought from Merck Company (Germany). All standards adopted were of reagent grade. Nitric acid of trace metal quality was bought from Sigma-Aldrich (United States). A Milli-Q water purification system (Millipore, USA) provided the ultrapure water used during the laboratory work.

STATISTICAL ANALYSIS

The differences between the heavy metal concentration levels (Cd, Cu, Pb, and Zn) were considered statistically significant at the 5% level with a 95% confidence interval ($P < 0.05$) based on the analysis of variance (ANOVA) performed on the study's data. Data were fed to the computer using IBM SPSS software package version 24.0. Quantitative data were described using mean and standard deviation for normally distributed data. Chi-Square was employed to determine the relationship between qualitative nominal variables performed mainly on frequencies. Significance test results are quoted as two-tailed probabilities.

RESULTS

The relevant criteria, weight, length, habitat, and feeding habits of all studied marine species are presented in **table (1)**. A comparison between the three studied sites regarding metal concentration in $\mu\text{g/g}$ (ppm) in all the different types and parts of the studied fish is displayed in **table (2)** and **figure (2)**. There was a significant increase in the four studied metal concentrations in Jubail compared to the other two regions.

The concentrations of copper in the various fish species from the three study sites (Dammam, Qatif, and Jubail) are shown in **table (3)**. Copper concentrations in Jubail were significantly high in muscles, eggs, and viscera of all studied fish species except crustaceans muscles and cuttlefish eggs. Cu levels were the highest in the eggs of crabs > shrimps > cuttlefish > Summer flounder fish compared to the other species in the same region, as seen in **figure (3)**.

Lead concentrations in the different types of fish species from Dammam, Qatif and Jubail, as shown in **table (4)**, showed that Jubail had significantly high Pb levels in muscles, eggs, and viscera in all the studied fish species. Pb levels were the highest in the viscera of the Summer flounder fish species from Dammam compared to all other species, followed by the eggs of crab, Summer Flounder, and cuttlefish, as seen in **figure (4)**.

A significant difference existed in the Zn levels in all the studied marine species and tissues in the Jubail area compared to the same species and tissues in Dammam and Qatif, except in eggs of crustaceans and cuttlefish and the muscles of the crustaceans as seen in **table (5)**. It was the highest in the muscles of Gilt-head bream > Shrimps > marbled spinefoot caught from the Jubail coasts, as seen in **figure (5)**.

Table (6) shows a statistically significant difference in the Cd levels in all the studied

marine species and tissues in the Jubail area compared to those in Dammam and Qatif. The highest recorded Cd levels were in the Jubail area in the eggs of the mullet > shrimps > summer flounder > cuttlefish, as seen in **figure (6)**.

Table (7) displays the correlation coefficients between heavy metals. Cadmium positively correlated to zinc and lead in fish and crustaceans. In contrast, it positively correlated to Copper in the different fish types and to lead in the cuttlefish as the species increased in weight. Lead negatively correlated to Copper only in cuttlefish as the species increased in weight and size. Cu positively correlated to Pb, Zn, and Cd in fish (0.393, 0.315, 0.475), while it was negatively correlated to lead in crustaceans (-0.374). The highest positive correlation was noted between Pb and Cd.

Table (1): Relevant criteria of the studied fish, crustaceans (shrimps and Crabs), and cuttlefish samples; weight, length, habitat, and feeding habits.

Local name	Common name	Scientific name	No.	Habitat	Weight (g) (Min-Max)	Length (cm) (Min-Max)	Feeding Habits
Flatfish	Summer flounder	Paralichthys dentatus	4	Pelagic	49-64	8.1-13.2	Carnivore/Omnivore
Sultan Ibrahim	Striped-Red mullets	Mullus surmuletus	4	Demersal	75-83	12.2-15.1	Omnivore
Forktail Rabbitfish	Stream-lined spinefoot	Siganus argenteus	4	Pelagic	96-104	8.1-11.3	Omnivore
Denees	Gilt-head bream	Sparus aurata	4	Pelagic	124-132	10.4-14.2	Carnivore
Meyas	Blue fish	Pomatomus saltatrix	4	Pelagic	522-751	28.0-39.2	Carnivore
Bouri	Flathead Mullet	Mugil cephalus	4	Pelagic	122-184	20.6-26.1	Omnivore
Pig	Drummer blackfish	Girella elevata	4	Demersal	487-492	35.3-45.2	Carnivore
Shrimps	Prawns	Penaeus monodon	4	Demersal	30.4-520.5	8.9-12.8	Omnivore
Crabs	Crustaceans	Liocarcinus vernalis	4	Demersal	78.4-120.5	9.7-18.9	Omnivore
Cuttlefish	Marine molluscs	Sepia officinalis	4	Demersal	80.5-142.7	12.4-24.3	Carnivore




 Fish  Crustaceans  Cuttlefish

Table (2): Comparison between different sites regarding metal concentration in ppm (ug/g wet weight) in all studied marine species types and tissues.

		DAMMAM	QATIF	JUBAIL	P-VALUE
Copper	Range	1.13-2.38	1.01-2.12	1.17-2.97	70.229 0.001*
	Mean	1.73	1.63	2.04	
	SD	0.27	0.27	0.29	
Lead	Range	0.03-0.87	0.01-0.19	0.01-0.30	84.882 0.001*
	Mean	0.07	0.04	0.16	
	SD	0.11	0.04	0.06	
Zinc	Range	0.05-0.85	0.03-0.92	0.06-0.99	97.737 0.001*
	Mean	0.23	0.31	0.69	
	SD	0.26	0.26	0.30	
Cadmium	Range	0.01-0.18	0.02-0.24	0.01-0.67	294.500 0.001*
	Mean	0.04	0.05	0.18	
	SD	0.03	0.04	0.07	

*P is significant at a 5% level ANOVA: Analysis of Variance

Table (3): Copper in ppm in all the studied marine species from the three studied sites (Dammam, Qatif and Jubail).

		Dammam	Qatif	Jubail	ANOVA	P-value
Fish	M	1.72±0.20	1.58±0.16	2.00±0.15*	4.12	0.036*
	E	1.65±0.28	1.74±0.22	2.08±0.16*	5.11	0.021*
	V	1.72±0.16	1.58±0.19	1.92±0.10*	3.98	0.046*
Crustaceans (Shrimps & Crabs)	M	1.72±0.20	1.62±0.13	1.85±0.22*	2.11	0.098
	E	2.06±0.27	1.81±0.30	2.64±0.26*	4.22	0.037*
	V	1.71±0.19	1.46±0.29	1.95±0.06*	6.02	0.011*
Cuttlefish	M	1.42±0.13	1.37±0.09	1.73±0.39*	4.11	0.035*
	E	2.09±0.12	1.98±0.19	2.56±0.35*	1.25	0.103
	V	1.65±0.16	1.39±0.16	1.90±0.06*	5.04	0.026*

Fish Crustaceans Cuttlefish

*P is significant at a 5% level (M: Muscles, E: Eggs & V: Viscera) ANOVA: Analysis of Variance

Table (4): Lead in ppm in all the studied marine species from the three studied sites (Dammam, Qatif and Jubail).

		Dammam	Qatif	Jubail	ANOVA	P-value
Fish	M	0.04±0.03	0.03±0.03	0.16±0.04*	6.01	0.011*
	E	0.09±0.10	0.05±0.04	0.17±0.05*	4.0	0.026*
	V	0.07±0.10	0.04±0.03	0.15±0.05*	5.22	0.016*
Crustaceans (Shrimps & Crabs)	M	0.06±0.04	0.04±0.03	0.15±0.03*	4.78	0.018*
	E	0.10±0.08	0.05±0.04	0.18±0.05*	3.25	0.045*
	V	0.06±0.06	0.06±0.04	0.17±0.08*	8.22	0.006*
Cuttlefish	M	0.083±0.077	0.033±0.013	0.157±0.031*	3.88	0.029*
	E	0.065±0.037	0.060±0.040	0.223±0.077*	11.25	0.001*
	V	0.060±0.070	0.061±0.068	0.174±0.049*	7.11	0.003*

Fish Crustaceans Cuttlefish

*P is significant at a 5% level (M: Muscles, E: Eggs & V: Viscera) ANOVA: Analysis of Variance

Table (5): Zinc in ppm in all the studied marine species from the three studied sites (Dammam, Qatif and Jubail).




		Dammam	Qatif	Jubail	ANOVA	P-value
Fish	M	0.371±0.148	0.309±0.124	0.747±0.162*	5.98	0.014*
	E	0.538±0.152	0.453±0.186	0.636±0.232*	3.11	0.041*
	V	0.198±0.042	0.367±0.203	0.768±0.234*	7.22	0.002*
Crustaceans (Shrimps & Crabs)	M	0.558±0.272	0.631±0.131	0.681±0.248	1.25	0.311
	E	0.493±0.150	0.554±0.092	0.630±0.383	1.56	0.21
	V	0.246±0.086	0.265±0.035	0.719±0.218*	11.32	0.001*
Cuttlefish	M	0.405±0.007	0.770±0.212	0.775±0.092*	3.66	0.042*
	E	0.697±0.131	0.497±0.067	0.625±0.306	1.88	0.287
	V	0.165±0.021	0.345±0.228	0.885±0.118*	6.12	0.011*

Fish Crustaceans Cuttlefish

*P is significant at a 5% level (M: Muscles, E: Eggs & V: Viscera) ANOVA: Analysis of Variance

Table (6): Cadmium in ppm in all the studied marine species from the three studied sites (Dammam, Qatif and Jubail).

		Dammam	Qatif	Jubail	ANOVA	P-value
Fish	M	0.03±0.01	0.04±0.03	0.17±0.03*	12.65	0.001*
	E	0.05±0.03	0.07±0.05	0.20±0.06*	14.2	0.001*
	V	0.03±0.02	0.03±0.02	0.16±0.04*	13.52	0.001*
Crustaceans (Shrimps & Crabs)	M	0.03±0.02	0.05±0.04	0.13±0.04*	4.98	0.025*
	E	0.07±0.04	0.07±0.04	0.22±0.04*	8.21	0.006*
	V	0.06±0.04	0.03±0.03	0.19±0.07*	7.58	0.009*
Cuttlefish	M	0.03±0.02	0.03±0.02	0.15±0.04*	6.89	0.011*
	E	0.07±0.05	0.15±0.02	0.22±0.07*	3.21	0.039*
	V	0.04±0.02	0.05±0.05	0.19±0.07*	7.21	0.011*

 Fish  Crustaceans  Cuttlefish

*P is significant at a 5% level (M: Muscles, E: Eggs and V: Viscera) ANOVA: Analysis of Variance

Table (7): Coefficient Correlation between different measured metals in the different types of the studied seafood.

Fish		Copper	Lead	Zinc
lead	Pearson Correlation	0.393		
	P-value	0.0001*		
zinc	Pearson Correlation	0.315	0.388	
	P-value	0.0001*	0.0001*	
cadmium	Pearson Correlation	0.475	0.453	0.514
	P-value	0.0001*	0.0001*	0.0001*
Crustaceans (Shrimps & Crabs)		copper	lead	zinc
lead	Pearson Correlation	0.163		
	P-value	0.171		
zinc	Pearson Correlation	0.026	0.349	
	P-value	0.832	0.003*	
cadmium	Pearson Correlation	0.036	0.625	0.446
	P-value	0.762	0.0001*	0.0001*
Cuttlefish		copper	lead	zinc
lead	Pearson Correlation	-0.347		
	P-value	0.038		
zinc	Pearson Correlation	-0.040	0.208	
	P-value	0.817	0.224	
cadmium	Pearson Correlation	-0.053	0.630	0.328
	P-value	0.757	0.0001*	0.050

Fish

Table (8) Correlation coefficients between heavy metals concentrations and the studied marine species weight and length

	Cd	Pb	Cu	Zn	Weight	Length
Cd	1					
Pb	0.918**	1				
Cu	0.569	0.248	1			
Zn	0.794*	0.372	0.851**	1		
Weight	-0.028	-0.157	-0.037	0.256*	1	
Length	-0.227	-0.173	-0.061	0.058	0.051	1

*Correlation significant at $P < 5\%$.

**Correlation significant at $P < 1\%$.

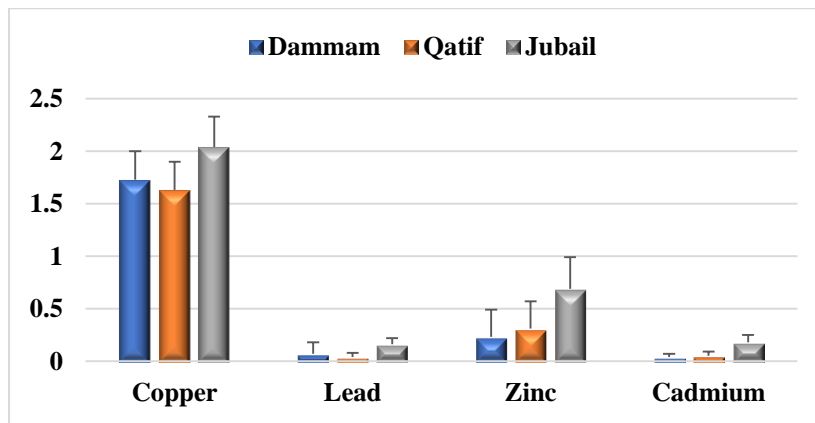


Figure (2): Comparison between different sites regarding heavy metal concentration in all studied fish types and parts.

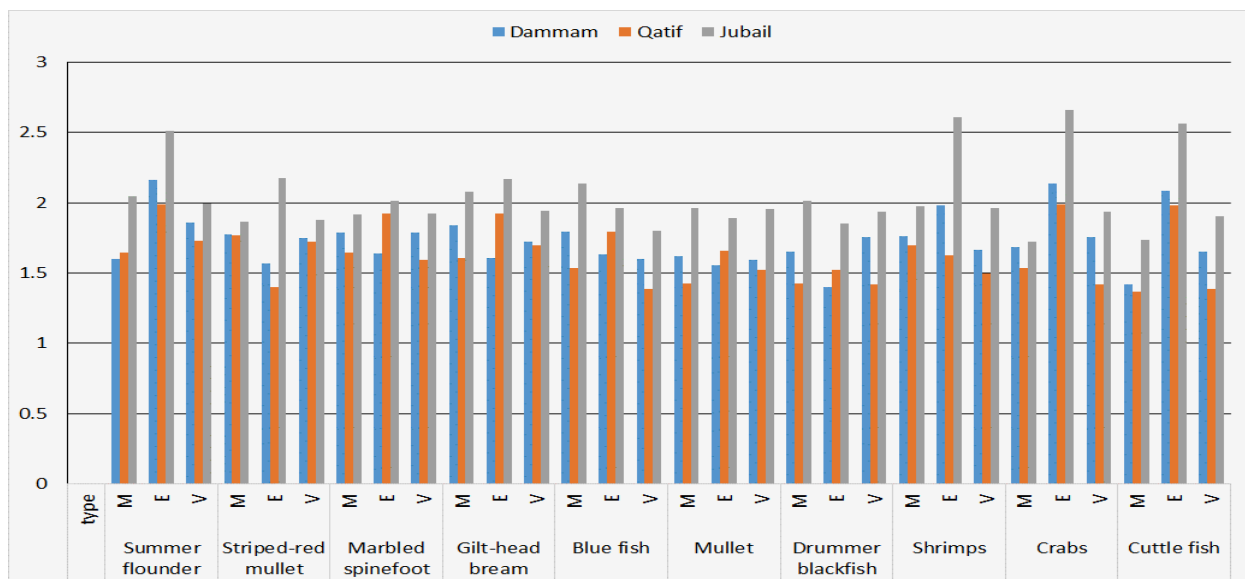


Fig. (3): Copper in ppm/mg/kg wet weight in different types of seafood from the three studied sites (Dammam, Qatif and Jubail).

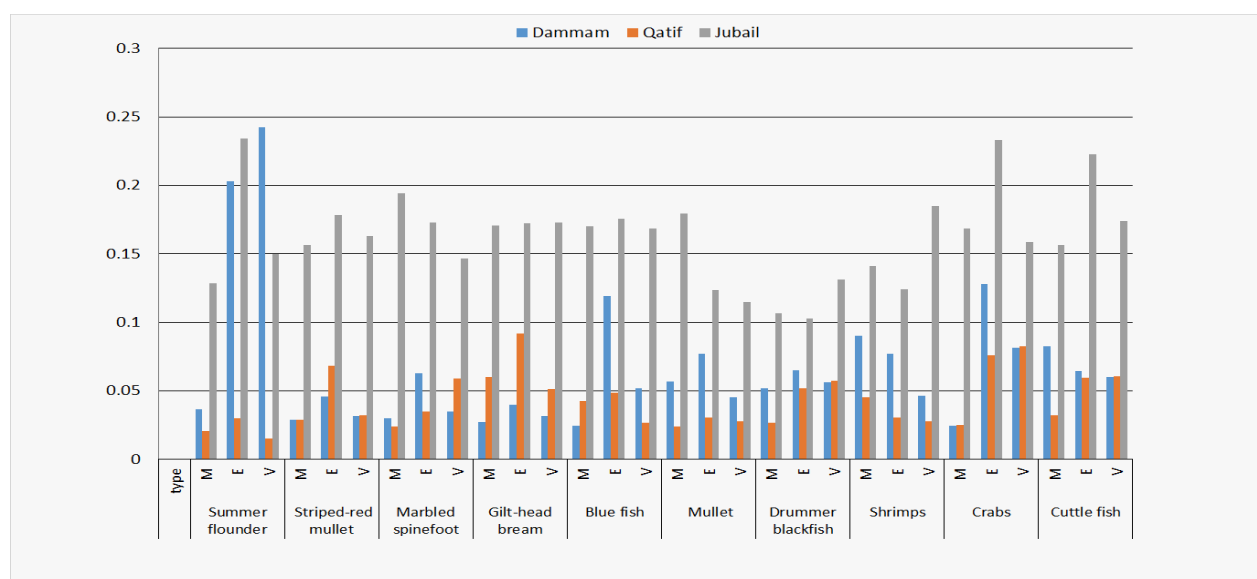


Fig. (4): Lead in ppm/mg/kg wet weight in different types of seafood from the three studied sites (Dammam, Qatif and Jubail)

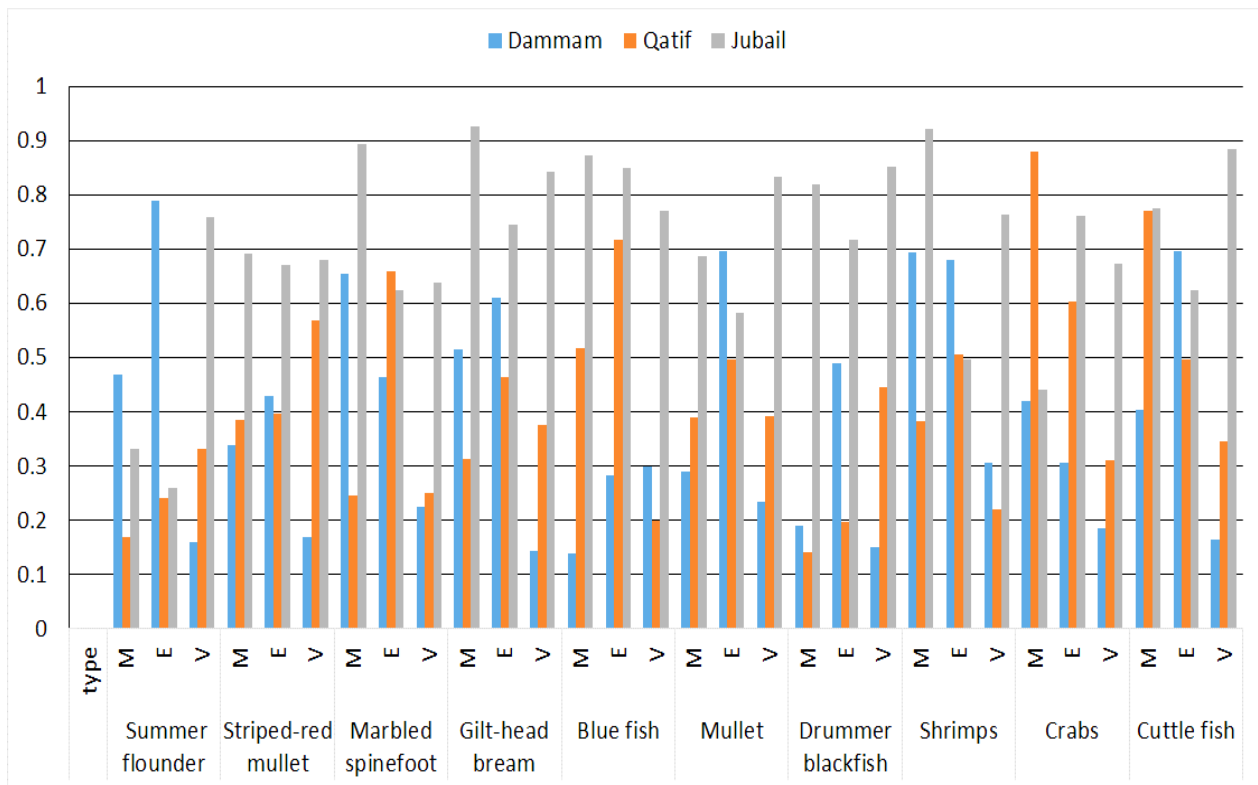


Fig. (5): Zinc in ppm/mg/kg wet weight in different types of seafood from the three studied sites (Dammam, Qatif and Jubail).

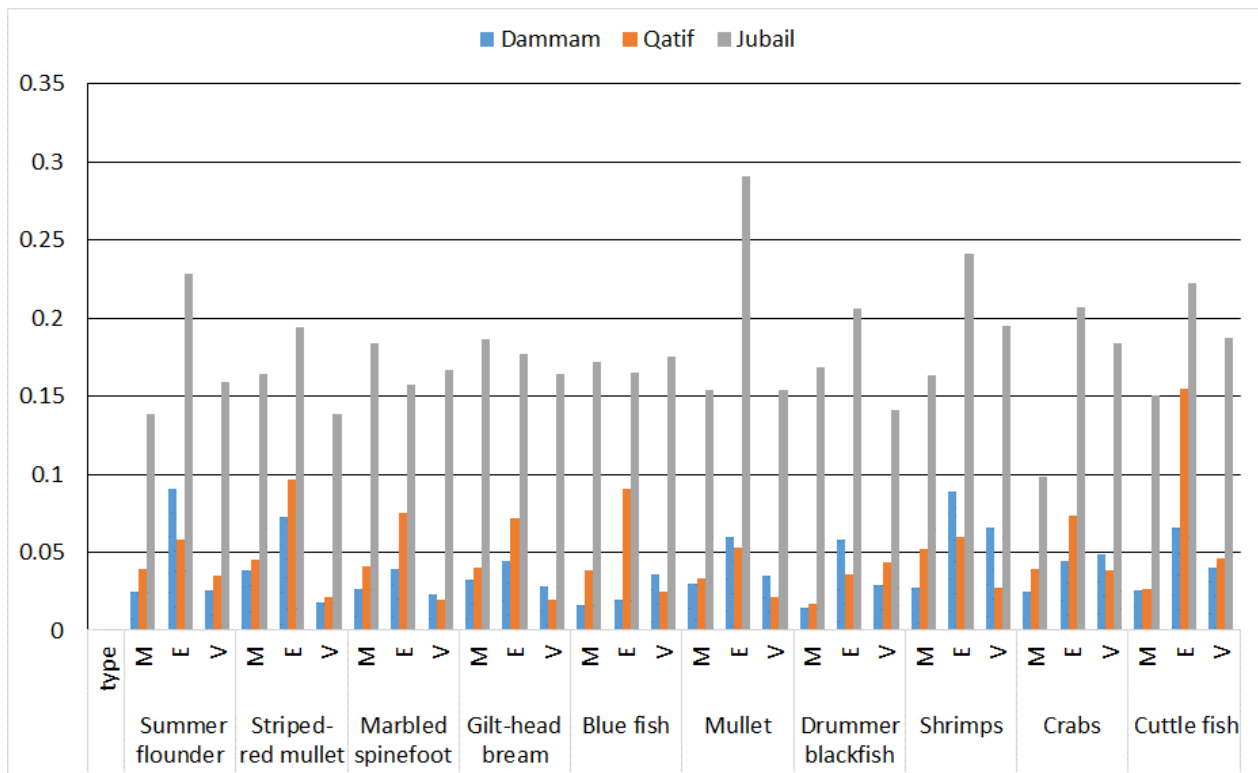


Fig. (6): Cadmium in ppm/mg/kg wet weight in different types of seafood from the three studied sites (Dammam, Qatif and Jubail).

DISCUSSION

In many developed countries, heavy metal concentration limits in fish have been set as a safety measure for public health. *Sivaperumal et al.*, during their research work in (2007), noted that human injury and toxicities can be inflicted by the increased levels of even the essential elements. Cadmium, Copper, lead, and zinc are the most prevalent contaminants in fish. Trace metals' toxicity and accumulation affect aquatic ecosystems. Lead, copper, cadmium, and zinc form poisonous "ligands" with organic molecules (*El-Badry, 2016*). Lead toxicities induce weakness, irritability, muscle pain, coma, and kidney, liver, and brain injury. Cadmium toxicity causes Japanese Itai-Itai disease, bone disease, anosmia, and tooth discoloration (*Sanders, et al. 2009; Sankar and Kumar, 2014; and Agumassie, 2018*).

Speedy urbanization and industrialization have increased heavy metal pollution across Saudi Arabia's Arabian Gulf shores. The typical sources of metal contamination are the oil industry, platforms, housing projects, industrial cities, oil terminals, offshore oil, ships, stainless steel, and cement industries, as well as power stations and many industries that expanded along the coastline and adjacent to nearby small cities since the 1970s, and have continued to date (*El-Sorogy et al., 2018*).

Due to limited dilution and slow dispersion rates, the Arabian Gulf region's arid climate, hot weather, and high evaporation rates result in amplified contamination and the persistence of metals in the ecosystem. Due to their bioaccumulation potential, metals in coastal waters and sediments impair the ecosystem and the biota, which raises health issues for humans (*Sharifuzzaman, et al. 2016*). There are many limitations to the current and similar works. The sample size of fish species analyzed in the study may be limited due to availability, access, and cost. A small sample size may not accurately represent the entire population of fish species or the variability within it. Selecting fish samples for analysis may introduce bias when certain species, sizes, or locations are overrepresented or underrepresented. This can impact the generalizability of the findings.

Metal concentrations in fish can vary spatially and temporally due to factors like water quality, habitat, migration patterns, and seasonal variations. A study in a specific location or period may not capture the full range of metal concentrations under investigation (*Darweesh et al. 2019*).

There was a significant increase in the four studied metal concentrations in Jubail compared to the other two regions. The buildup of heavy metals in marine organisms depends on many environmental conditions, seasonal variations, geographical location, habitat selection, tropic level, feeding behavioral patterns, ages, dimensions, the longevity of exposure to the metals, and physiological regulatory function (*Censi et al., 2006; Sankar et al., 2006 and Darweesh et al., 2019*). Heavy metals accumulate in the water and sediment columns, which enter the food web by feeding the pelagic species (*Burgos and Rainbow, 2001*). One key factor is the presence of heavy metals in the water column, which can be affected by seasonal variations and geographical location. In a study by *Darweesh et al. (2019)*, the authors reported that the concentration of heavy metals in the water column of the River Nile in Egypt varied significantly depending on the season and location.

Another critical factor is the habitat choice of the marine species in question. *Rajeshkumar, and Li*, during their work in (2018), reported that the accumulation of heavy metals in benthic organisms was higher in areas with high sedimentation rates, as the case along most of Arabian Gulf shorelines in Saudi Arabia's Eastern Province, as these organisms were more likely to come into contact with heavy metals in the sediment (*Avinash et al., 2016*).

In addition to these variables, the tropic level, feeding practices, age, dimensions, length of time exposed to the polluting elements, and metabolic activity of aquatic organisms may impact their accumulation of heavy metals. For example, a study by *Liao et al. (2021)* found that the accumulation of heavy metals in fish was influenced by their trophic level, with higher trophic level fish having higher tissue heavy metals concentrations. Overall, the accumulation of metals in marine

organisms is a complex and multifaceted phenomenon influenced by various environmental and biological factors (*Di Lena et al., 2018*).

Some heavy metals, including iron (Fe), copper (Cu), and zinc (Zn), are essential for the metabolism of fish. In contrast, there are no known biological activities of mercury (Hg), lead (Pb), cadmium (Cd), and other metals. The metabolic activities of marine species greatly influence their bioaccumulation ability of trace metals (*Velusamy et al., 2014*).

The accumulation pattern of the analyzed metals follows the order of $Cu > Zn > Cd > Pb$. The current results agreed with those reported by *Al-Yousuf et al. (2000)* during their work in Bahrain and *Zyadah and Almoteiry, (2012)* in the Eastern Province of Saudi Arabia.

The metal levels in the different fish species varied within the three studied sites; the highest recorded metal concentrations were in the Jubail area, followed by Qatif and Dammam. At Jubail, the highest recorded Copper, lead, zinc, and cadmium concentrations were 2.64 (eggs of crustaceans), 0.223 (eggs of cuttlefish), 0.885 (Viscera of cuttlefish), and 0.22 ppm (eggs of crustaceans and cuttlefish). This increase in heavy metal values in water at these sites may be attributed to the drainage of industrial wastewater. In contrast, Qatif showed the lowest Cu concentrations of the studied metals, recorded in the cuttlefish muscles (1.37 ppm), Pb (0.03 in muscles of fish), and Cd (0.02 eggs of crustaceans), simultaneously. The least Zn level recorded was 0.165 in the viscera of cuttlefish in Dammam. By examining the correlation between different metals, researchers can determine whether certain metals tend to co-occur as Cd to Pb and Zn and Cu to Pb, Zn, and Cd, as seen in the current work in the studied seafood species. This information can be useful in understanding potential sources of contamination and developing strategies for monitoring and managing metal pollution in aquatic environments (*Ajala et al., 2022*).

The high levels of Cu encountered in the current work were likely a result of depositional sediments and antifouling paint

from ships due to the heavy fishing boat traffic in the studied areas (*Lattemann and Hopner, 2008*).

Another explanation for the detected high Cu levels is the hot weather-induced copper precipitation as CuS due to temperature rises (*Hegazy et al., 2016*). Zn is a prevalent pollutant in agriculture, food waste, pesticide manufacture, and antifouling coatings and paints (*Badr et al., 2009*).

In this research, the mean Copper concentration in fish was 1.63- 2.04 ppm, varying between 1.01- 2.97 ppm in all studied fish types. The highest copper concentration was detected in Jubail among crustaceans (2.64 ± 0.26 ppm), whereas the lowest concentration was found in Qatif in the cuttlefish (1.37 ± 0.09). The measured copper concentrations did not exceed both the national and international standards. The majority of international regulations limited Copper concentrations not to exceed thirty ppm. The concentrations of Cu recorded in the current work were below the international guidelines.

In Ghana's Asafo market, its levels ranged between 0.02-0.156 (*Kwaansa-Ansah, et al. 2019*), far less than the current recorded values. The range of Cu concentration in the fish from the Lower Meghna River and adjacent areas of Bangladesh was 36.438 ppm (*Hossain et al., 2022*), and from Pearl River, China, it was within 1.17- 6.72 (*Xie, et al., 2010*), and both were higher than the levels obtained in the current work. Copper is crucial for the body because it aids in synthesizing hemoglobin and other vital enzymes, but an excess can cause hepatic and renal injuries (*Travis, 2013; Vu et al., 2017*).

The range of Zn accumulation in fish in Dammam, Qatif, and Jubail was (0.05-0.85), (0.03-0.92), and (0.06-0.99), respectively. Some metabolic activities and coenzyme-catalyzed reactions involving zinc in the kidneys of marine species could account for these amounts. Hence, the largest concentrations were found in the viscera (*Freake, and Sankavaram, 2013*). Zinc also acts as a catalyst in metal biomolecules attached to amino acid side chains and/or sulfur donor ligands to produce tetrahedral

zinc metalloproteins and metalloenzymes in muscles (*Mohamad et al., 2023*).

The highest zinc concentration permissible for human consumption is 30 mg/kg. The Zn concentrations measured in all marine species studied in the current work did not exceed the recommended levels. The zinc fish levels were lower than that reported in the Bangshi River (*Rahman et al., 2012*).

However, all species' mean concentrations of Zn were higher than the other international reports, as seen in Table (9). Zn tends to accumulate in the fatty tissues of fish and other aquatic organisms, likely affecting their reproductive physiology. Besides, chronic exposure to Cu and Zn is reported to be associated with Parkinson's disease (*Ullah, et al., 2017*).

All estimated levels of Pb were higher than the values reported by *Zuluaga Rodríguez, J. et al. (2015)* in Colombia, Malaysia (*Hashim, et al., 2015*), and the Western Arabian Gulf coastal area in Egypt (*Amin and Almahasheer, 2022*). All Pb and Cd levels obtained in the current research were less than the values. *Kortei et al. (2020)*, reported in their study on fish samples from the Gulf of Guinea. The levels in Jubail were higher than that reported by *Dural et al. (2007)*, and were far above the reported levels by the *FAO (2011)*.

Regarding Pb, the current concentrations were below the levels reported by *Hossain et al.* during their work in (2022), which ranged between 0.202 to 0.68 ppm. Based on the *FAO and FAO/WHO* reports consecutively released in (2011) and (2009), the maximum permissible concentration for Pb is 0.5 ppm. On the contrary, the *USFDA in 2013*, reported the permissible Pb level to be 1.5 ppm. In all concentrations attained during the current work, the Pb concentrations in all studied fish species were below many previous levels. Current mean Pb concentrations are less than those reported by *Ahmed et al. (2019)*. Besides, *Staniskiene et al. (2006)*, and *Copat et al. (2012)*, obtained a comparable Pb concentration in their published work. By examining the correlation between different metals, researchers can determine whether certain metals tend to co-occur as Cd to Pb and Zn and Cu to Pb, Zn,

and Cd, as seen in the current work in the studied seafood species (*Ahmed et al., 2019*). As reported in the scientific literature, the Standard regulatory limits of the screened heavy metals in the fish samples are still below the permissible levels (*Dural et al., 2007; FAO/WHO, 2009; FAO, 2011; USFDA, 2013; CREC, 2008; CREC, 2014 and FSSAI, 2015*).

CONCLUSION

High heavy metal levels were recorded in the investigated fish types in Jubail. This finding highlights that aquatic pollution has started approaching hazardous levels for human health. The current data suggest the need for toxicological evaluation of human populations exceeding the permissible daily limits for signs of heavy metal toxicity, especially in the Jubail area.

RECOMMENDATIONS

The periodic performance of toxicological research is vital and recommended to guard against environmental and human toxicity. Further research concerning the study of toxic effects of heavy metals via fish consumption in different Eastern Province regions is recommended. Realistically assessing trace metals as toxicants in man's environment is essential to establishing meaningful pollution guidelines.

It's important to consider the consumption of higher trophic-level fish in terms of food safety and potential exposure to contaminants, as their position in the food chain can result in the bioaccumulation of some metals. Regulatory guidelines and advisories must exist for the existing fish species to help protect consumers from potential risks associated with consuming these fish. While studying metal concentrations in fish is essential for understanding ecosystem health, direct extrapolation to human health risks may be limited. Additional studies are needed to assess the potential impact on human consumers and establish safe consumption guidelines.

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