



# **VETERINARIA** RIVISTA DI SANITÀ PUBBLICA VETERINARIA **ITALIANA**

**Paper**



# Bioaccumulation of trace elements (Cd, Hg, Pb, Fe and Zn) in seven fish species, crustaceans and mussels from the Gulf of Skikda, Southern Mediterranean Sea

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*Veterinaria Italiana*, Vol. 62 No. 1 (2026) DOI: 10.12834/VetIt.3849.37542.2

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## Abstract

The study investigates the concentrations of five heavy metals in seven seafood species from the Gulf of Skikda, the southern Mediterranean Sea, Algeria. The aim is to assess contamination levels, and compliance with European food safety regulations. Between February and August 2018, muscle samples were collected from five fish species (*Sardina pilchardus*, *Sarda sarda*, *Mugil cephalus*, *Xiphias gladius*, *Thunnus thynnus*), the deep-water rose shrimp (*Parapenaeus longirostris*), and the Mediterranean mussel (*Mytilus galloprovincialis*). Samples were oven-dried, acid-digested, and analysed by inductively coupled plasma optical emission spectrometry (ICP-OES) for Cd, Pb, Fe and Zn; and by the flow injection hydride analysis system (FIAS) for Hg. Data were expressed in mg kg<sup>-1</sup> wet weight (ww) and statistically compared by one-way ANOVA ( $p \leq 0.05$ ). The results revealed significant interspecies differences ( $p < 0.001$ ) in metal accumulation patterns. Cd (0.139 mg·kg<sup>-1</sup> ww) and Hg (0.040 mg·kg<sup>-1</sup> ww) were significantly higher in grey mullet and tuna respectively ( $p < 0.001$ ); both exceeding permissible regulatory limits. The remaining of heavy metals complied with guidelines. Mussels showed the highest Cd and Pb levels but remained within food safety limits. Sardines demonstrated a favorable nutritional profile, with high essential elements and low toxic metals. The study data recommend avoiding consumption of grey mullet and avoiding regular consumption of tuna caught in the fishing areas of Skikda Bay. Regular monitoring of heavy metal contamination in Algerian seafood is necessary to ensure consumer safety.

## Keywords

heavy metals, bioaccumulation, muscle, seafood, Mediterranean Sea, Skikda

## Introduction

The Mediterranean Sea, being a semi-enclosed basin with limited water exchange, experiences evaporation rates that surpass precipitation and river runoff. Coupled with dense coastal populations, this configuration leads to significant pollutant accumulation in its sub-basins (Aytekin et al., 2019; Merhaby et al., 2019; Storelli et al., 2020; Sharma et al., 2021). Human activities including mining, industrial processes and agricultural practices are considered to be the primary sources of heavy metals, which have a detrimental impact on marine life. Heavy metals induce oxidative stress, disrupt metabolism, and impair growth, reproduction, and immunity in marine organisms. Their accumulation also causes population declines and food web imbalances through biomagnification (Griboff et al., 2020; Esposito et al., 2021; Zhang et al., 2023; Rahayu et al., 2024; Ebadi et al., 2025). Excessive amounts of certain metals present in marine ecosystems can accumulate in marine organisms to potentially toxic concentrations (Barone et al., 2018; Mol et al., 2018; Łuczyńska et al., 2018). Exposure to heavy metals can induce various adverse effects in humans,

including renal and hepatic pathologies, haematological and neurological disorders, homeostatic imbalances and cancers, making such exposure a major public health problem (Oehlenschläger, 2012; Korkmaz et al., 2019; Mao et al., 2019 ; Zhang et al., 2023).

The concentrations of heavy metals in fish and their bioaccumulation in muscle tissue vary between species. Physiological mechanisms play a key role, with some species showing a higher affinity for metal uptake and storage, while others are more efficient at excreting metals (Govind and Madhuri, 2014; Łuczyńska et al., 2018; Antović et al., 2019; Landrigan et al., 2020; Lall et al., 2021; Zhang et al., 2023). The trophic level is also a determining factor. Indeed, while bioaccumulation refers to the direct uptake of chemical contaminants from the surrounding environment by marine organisms, biomagnification occurs when these pollutants become increasingly concentrated at successive trophic levels due to food web interactions.

Only a few studies have focused on pollution along the Algerian coast (Ouali et al., 2008; Taleb et al., 2009; Mehoul et al., 2019; Silhadi et al., 2020, Mankou-Haddadi et al., 2021). Given the large number of pollutants present in the marine environment and the absence of official data on the conditions of Algerian coastal ecosystems, there is a pressing need for focused research to provide valuable ecotoxicological data.

The Algerian fisheries sector represents an emerging economic resource. According to ONS (2022), exports of fresh and frozen fish, crustaceans, and mollusks are mainly directed towards European and Asian markets, highlighting the sector's growing potential. Enhancing quality, traceability, and sanitary standards is crucial to strengthen international access and competitiveness.

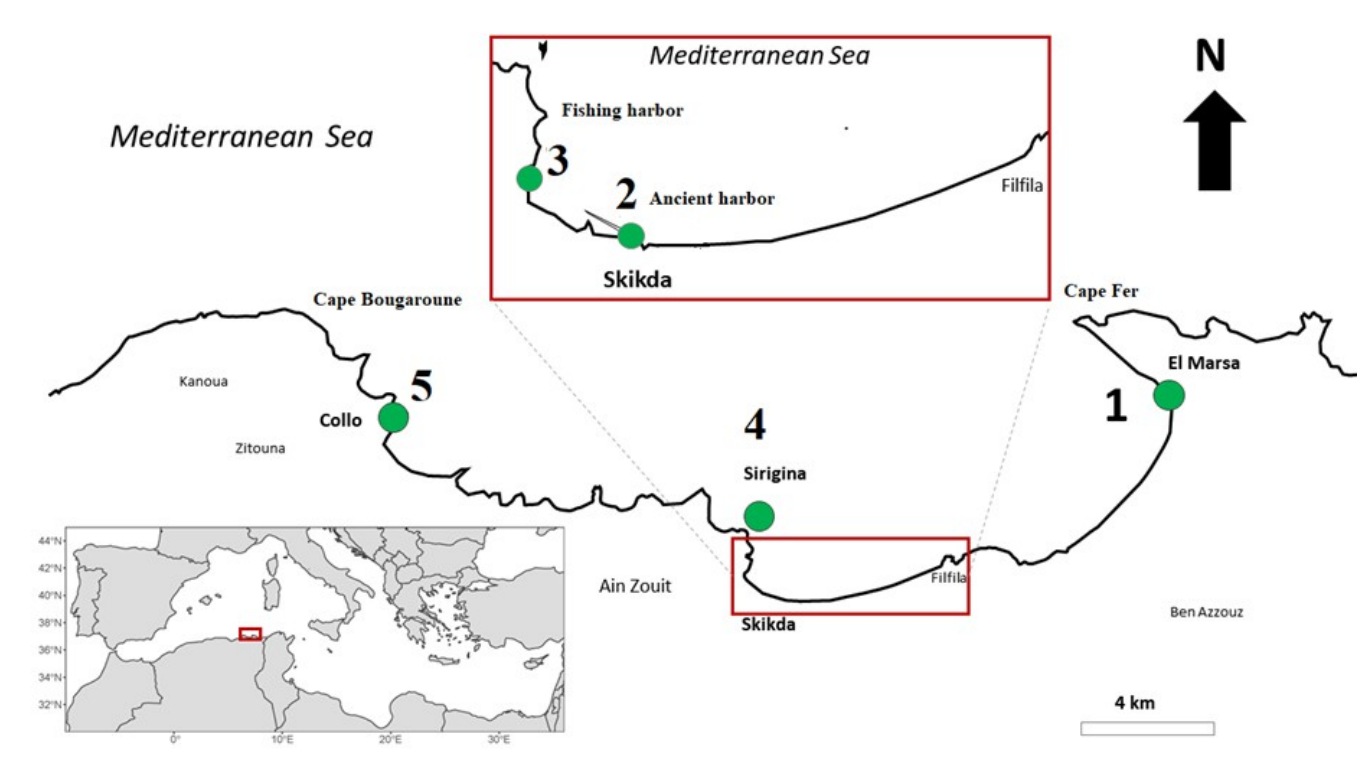
This study examines the heavy metal content of some of Algeria's most commercially important seafood species and assesses their compliance with European regulations on heavy metal concentrations. The concentrations of two essential elements (Fe and Zn) and three non-essential elements (Pb, Cd, and Hg) were measured in the muscle tissue of five fish species (*Sardina pilchardus*, *Sarda sarda*, *Mugil cephalus*, *Xiphias gladius*, *Thunnus thynnus*), as well as in shrimp (*Parapenaeus longirostris*) and mussels (*Mytilus galloprovincialis*), which were collected from the Skikda region on Algeria's eastern coast from February to August 2018.

## Materials and Methods

### Study area

The analyzed fish species were caught in the main fishing areas along the Mediterranean coast of the Skikda region, including Collo Bay, Elmarsa Bay, Skikda Bay and Sirigina Island.

The city of Skikda is located on the northeastern coast of Algeria, (latitudes 36°15'–36°75' N; longitudes 7°15'–7°30' E), and has a coastline of 160 km coastline and a fishing area of 3,068 km<sup>2</sup>, representing 4.69% of the total national fishing area (ISTPM, 1982). The Gulf of Skikda is bordered by Cape Bougaroune to the west and Cape de Fer to the east (between 06°27'10"E and 07°10'02"E; (Fig. 1), and is fed by three principal rivers: Ouadi Guebli, Ouadi El kebir and Ouadi Saf-Saf. Known for its fishing and trading port, the Gulf of Skikda also hosts a great petrochemical complex. In addition to its prominent fishing and trading port, the gulf is home to various industries, including a plastics complex, food processing companies, a cement factory and a marble production facility.



**Figure 1.** Geographic location of the Gulf of Skikda along the Algerian coast, Southern Mediterranean Sea.

## Sampling and sample preparation

Between February and August 2018, a total of 72 fish (*Sardina pilchardus*, *Sarda sarda*, *Mugil cephalus*, *Xiphias gladius*, *Thunnus thynnus*), 40 shrimp (*Parapenaeus longirostris*) and 30 mussels (*Mytilus galloprovincialis*) were collected. Upon collection, the samples were promptly transported to the laboratory in an icebox. Edible portions were meticulously extracted, and the total weight of each individual specimen was recorded. For smaller species (mussels, shrimp and sardines), the edible portions were pooled to create composite samples. The tissues were then homogenized and frozen at -20°C until analysis.

## Chemical preparation and instrumental analysis

Muscle tissues were oven-dried at 40°C for 48 hours to determine their dry weight (dw). Sample digestion was performed in a closed system to prevent the loss of the volatile elements particularly mercury. Approximately 0.8 g of homogenized dry tissue was digested in 100 mL Teflon digestion vessels containing 10 mL of concentrated nitric acid (HNO<sub>3</sub> 65%, Biochem Chemopharma). Following an initial one-hour period with the vessels left open to facilitate CO<sub>2</sub> release, they were subsequently sealed and incubated in a Binder FED 53 oven at 110 ± 5°C for two hours. The digested samples were then transferred to graduated polypropylene autosampler tubes and brought to a final volume of 50 mL using 0.5% HNO<sub>3</sub>. This protocol ensured complete digestion producing clear solutions with turbidity levels below 1.5 NTU, confirming complete digestion and eliminating the need for filtration. For mercury analysis, digestion was carried out using a mixture of 8 mL HNO<sub>3</sub> (65%) and 2 mL hydrogen peroxide H<sub>2</sub>O<sub>2</sub> (30%), thus eliminating the need for filtration. Blanks underwent the same digestion procedure as the samples to verify the absence of contamination, and all results were below detection limits.

Reagent blanks, quality control samples, and all test samples were processed identically with each analysis performed in triplicate to check the reproducibility of the results.

## ICP-OES analysis

Concentrations of Cd, Pb, Hg, Zn, and Fe were quantified using an ICP-OES spectrometer (PerkinElmer OPTIMA 8000, Waltham, MA, USA), performed with the instrument equipped with an autosampler (PerkinElmer S10 autosampler, Waltham, MA, USA) for standard and sample introduction. The instrument was operated using WinLab32 Software under the following conditions: plasma argon gas flow rate of 15 L/min, auxiliary argon gas flow rate of 0.2L/min and nebulizer argon gas flow rate of 0.8 L/min, radio frequency power of 1300 W, and a peristaltic pump flow rate sample of 1.8 mL/min, with axial plasma viewing.

Mercury concentrations were determined using a flow injection analysis system (FIAS-100, PerkinElmer, Waltham, MA, USA) coupled to the ICP-OES. Calibration curves for Cd, Pb, Hg, Fe, and Zn were constructed from individual certified standard solutions (1000 µg/mL) in 2-5% nitric acid purchased from VWR Chemicals company. The concentrations of the analyses were performed in accordance with ISO11885 (2007) standard method, using linear regression of the emitted spectral intensity versus element concentration. The calibration curves showed correlation coefficients ( $R^2$ ) greater than 0.999 for all metals analysed. The concentration of metals was expressed as in mg kg<sup>-1</sup> wet weight (ww).

## Method validation and quality assurance

The method's accuracy was validated using the IAEA-407 certified reference material (fish tissue), with recovery rates ranging from 97.7% to 103.3% (Table I). The limits of quantification (LOQ) were as follow: Cd: 0.10, Pb: 0.035, Hg: 0.018, Fe: 1.05, and Zn: 1.58 mg/kg-1 ww, and the limits of detection (LOD) estimated at approximately one-third of these values (LOQ  $\approx$  3  $\times$  LOD): Cd: 0.0333, Pb: 0.0117, Hg: 0.006, Zn: 0.526, Fe: 0.350 mg/kg-1 ww. Emission wavelengths for Cd, Pb, Hg, Fe, and Zn were 228.802 nm, 220.353 nm, 253.652 nm, 238.204 nm, and 206.200 nm, respectively. Measurement uncertainty was estimated according to repeatability, calibration, recovery rates, and precision of sample preparation steps (pipetting, weighing, and dilution). Analytical performance was assessed following ISO/IEC 17025 and EURACHEM/CITAC (2012) guidelines. The measurement uncertainty ( $U = k \cdot s$ , with  $k = 2$  for 95% confidence level) remained below 50% of the measured concentrations, confirming the method's reliability. Reproducibility was assessed using the HORRAT index (acceptable range: 0.3–2.0) derived from the Horwitz equation, with all values within this range. Precision was evaluated based on the relative standard deviation (RSD) of triplicate measurements, with all RSD values remaining below 10%, thereby demonstrating acceptable repeatability in accordance with the specified performance criteria.

Metals	Cd	Pb	Hg	Zn	Fe
<b>Certified values (mg kg<sup>-1</sup>)</b>	0.189±0.009	0.120±0.06	0.222±0.024	67.1±3.8	146±14
<b>Observed values (mg kg<sup>-1</sup>)</b>	0.186±0.005	0.124±0.002	0.225 ±0.002	66.66±1,52	142.66±2,51
<b>Recovery (%)</b>	98.4	103.3	101.3	99.3	97.7
<b>RSD (%)</b>	2.69	1.61	0.89	2.28	1.76

**Table I.** Metal concentrations measured in Certified Reference Material (IAEA-407) (All data are presented as means  $\pm$  standard deviations, in mg kg<sup>-1</sup> ww), Recovery (%) and Relative Standard Deviation (RSD %).

## Statistical Analysis

An analysis of variance (ANOVA) was conducted using MedCalc version 22.016 to evaluate variations in contaminant accumulation levels and to determine whether accumulation patterns differed between the various seafood groups. Statistically significant differences were considered at  $p \leq 0.05$ .

## Results

Table II summarizes the mean recorded concentrations of essential and non-essential elements, namely Fe, Zn, Cd, Pb, and Hg, in muscle samples of seven commonly consumed marine species (fish, mussels, and shrimp).

Trace metal concentrations in the different species are reported based on their expanded uncertainties ( $k=2$ ). Uncertainties ranged from 1.4% to 88.6%, with most values below 50%. The few higher values corresponded to concentrations near the detection limits. HORRAT values (0.4 – 1.5) remained within the acceptable range ( $<2.0$ ), indicating that the method was precise, repeatable, and compliant with Horwitz and ISO 5725 performance criteria.

Species	Cd	Pb	Hg	Fe	Zn
Bonito	0.048±0.006 (0.05)	0.102±0.017 (0.3)	0.249±0.035 (1)	21.375±6.598	5.983±1.829 (50)
Grey Mullet	0.139±0.001 (0.05)	0.053±0.001 (0.3)	0.056±0.001 (1)	16.616±1	4,466±0.5 (50)
Sardina Pilchardus	0.079±0.035 (0.25)	0.102±0.017 (0.3)	0.059±0.015 (0.3)	35.330±4.546	19.338±4.326 (50)
Tuna	0.050±0.005 (0.1)	0.153±0.002 (0.3)	1.040±0.015 (1)	10.220±0.26	4.13±0.07 (50)
Swordfish	0.053±0.02 (0.25)	0.076±0.015 (0.3)	0.350±0.132 (1)	5.340±0.085	9±0.5 (50)
Shrimp	0.438±0.128 (0.5)	0.079±0.013 (0.5)	0.123±0.006 (0.5)	25.116±1.568	12.133±0.403 (50)
Mussels	0.571±0.291 (1)	1.332±0.061 (1.5)	0.382±0.23 (0.5)	50.416±1.71 (40 – 45)	41.446±5.155 (50)
Legal limits references	Official Journal of the European Union, (2014)	Official Journal of the European Union, (2015)	Official Journal of the European Union, (2022)	Food and Nutrition Board of the Institute of Medicine (IOM, 2003)	Dietary Reference Intake (DRI), Institute of Medicine, (IOM, 2006)

**Table II.** Estimated concentrations (mg kg<sup>-1</sup> ww) of essential and non-essential elements in the muscles of the analyzed fish, mussels and shrimp (mean ± standard deviation) and their compliance with legal limits for human consumption (values in parentheses). Concentrations exceeding the regulatory thresholds are highlighted in bold

Analysis of essential and non-essential elements in muscle samples from the seven marine species reveals significant inter-species variations ( $p < 0.05$ ) and different compliance levels with legal limits.

The concentration trends of the five contaminants varied according to the species tested. For tuna and bonito, the order was Fe > Zn > Hg > Pb > Cd, while mussels and sardines exhibited the sequence Fe > Zn > Pb > Cd > Hg. Swordfish showed the following order: Zn > Fe > Hg > Pb > Cd. Meanwhile, grey mullet and shrimp displayed the sequence Fe > Zn > Cd > Hg > Pb. Notably, tuna, bonito, and swordfish showed similar trends for non-essential elements.

Of the seven marine species tested, mussels stood out as having the highest Cd (0.571 mg kg<sup>-1</sup>) and Pb (1.332 mg kg<sup>-1</sup>) levels ( $P < 0.05$ ), both within the permissible limits for human consumption (1 mg kg<sup>-1</sup> for Cd and 1.5 mg kg<sup>-1</sup> for Pb). Only shrimp had a Cd concentration close to that of mussels ( $P > 0.05$ ) at 0.44 mg kg<sup>-1</sup> vs 0.57 mg kg<sup>-1</sup>. The highest levels of essential elements were also found in mussels ( $P < 0.05$ ), with the Fe concentration (50.416 mg kg<sup>-1</sup>) slightly exceeding the upper range of the legal limit (40–45 mg kg<sup>-1</sup>). The highest Hg concentration (1.040 mg kg<sup>-1</sup>) was found in tuna ( $P < 0.05$ ), which was just above the permissible limit (1 mg kg<sup>-1</sup>). The Cd level in the bonito muscle sample was close to that of the legal limit (0.048 mg kg<sup>-1</sup> vs 0.05 mg kg<sup>-1</sup>).

While the grey mullet had lower concentrations of Pb and Hg than mussels and tuna ( $P < 0.05$ ), its Cd level was almost three times higher than the authorized limit of 0.05 mg kg<sup>-1</sup> (Figure 2). Regarding the concentrations of essential elements, *Sardina pilchardus* ranked second with 35.33 mg kg<sup>-1</sup> for Fe and 19.33 mg kg<sup>-1</sup> for Zn. This species showed significantly higher levels of these nutrients compared to bonito, grey mullet, tuna, swordfish, and shrimp ( $p < 0.05$ ), while maintaining lower levels of heavy metals. This indicates a favorable profile for human consumption. Finally, shrimp and swordfish maintained acceptable levels of all the tested elements.



## Discussion

The present study assesses the heavy metal content in seven widely consumed marine species in Algeria, emphasizing the need for monitoring contamination levels. Although muscle tissue exhibits lower bioaccumulation compared to other tissues (Bervoets et al., 2001; Zhuang et al., 2013; Chan et al., 2021), it is the primary edible part and was therefore the focus of this analysis.

Although a few measurements exhibited higher uncertainties, these were associated with concentration levels close to the limits of detection, which is typical in trace analysis (Worsfold et al., 2019). At such low concentrations, relative uncertainty naturally increases, but this does not compromise the overall validity of the dataset.

Given the confirmed reliability and repeatability of the measurements, the observed differences in trace metal concentrations among species can be interpreted and compared with regulatory limits and literature values.

All muscle samples exhibited detectable concentrations of heavy metals, with no single element predominating overall, despite Hg's known affinity for muscle tissue (Jezierska and Witeska, 2006). Notable exceptions include i) markedly elevated Hg levels in tuna muscle and ii) significantly higher Pb concentrations in mussels. The excessive Hg accumulation in tuna is due to three key processes. Firstly, methylmercury (MeHg) binds strongly to the protein thiol groups of muscles (Scheuhammer et al., 2007), which greatly reduces its excretion such that large long-lived predatory fish like tuna consistently exhibit high concentrations relative to the surrounding waters (Thera et al., 2019; Ajsuvakova et al., 2020). Secondly, MeHg biomagnifies approximately eightfold at each trophic level, meaning that each predator concentrates far higher levels than its prey (Lavoie et al., 2013). Thirdly, with assimilation efficiencies exceeding 95% at each feeding stage (Ebadi et al., 2025), long-lived top predators such as tuna inevitably exceed regulatory limits for muscle Hg concentrations (Boudou and Ribeyre, 1997).

On the other hand, significantly higher Pb concentrations in mussels compared to pelagic predators such as tuna and swordfish ( $p < 0.05$ ) are consistent with the strong negative relationship between Pb concentration and trophic level for pelagic and benthic species (Leeves, 2011). Generally, Pb concentrations in organisms decrease with increasing trophic level in both benthic and pelagic food chains, indicating that Pb is not biomagnified, but is instead progressively diluted along the food web (Barwick and Maher, 2003). Half-life data for Pb retention in muscle tissues are not explicitly provided in the literature, but the downward trend of Pb with increasing trophic level implies that pelagic fish do not extensively accumulate or retain Pb, which is consistent with a short biological half-life or efficient excretion in these species. Cadmium concentrations in mussels were also significantly higher than those in predatory fish species such as tuna, swordfish, and bonito ( $p < 0.05$ ), suggesting that there is no evidence of cadmium biomagnification (Griboff et al., 2020; Ordiano-Flores et al., 2020), as reported by previous studies.

Overall, the study findings highlight significant interspecies variability in the concentrations of essential and non-essential elements.

Of the seven species studied, tuna exhibited the highest concentration of Hg in muscle tissue, highlighting the pronounced biomagnification of MeHg in apex pelagic predators (Barone et al., 2018; Chan et al., 2018; Taslima et al., 2022; Rahayu et al., 2024). In the present study, Hg levels in tuna muscle slightly exceeded the regulatory limit of  $1 \text{ mg kg}^{-1}$ . Similarly, Storelli et al. (2001) reported that all tuna specimens weighing over 40 kg (43% of the analysed sample) surpassed this threshold. Supporting this finding, a global survey of 5854 seafood samples reported that 3.11% of tuna and 11.30% of swordfish exceeded EU Hg limits (Garofalo et al., 2025), with even higher non-compliance rates observed in Italy (17.3%; Esposito et al., 2018) and Spain (37%; Torres et al., 2010). Given Hg's neurotoxic and teratogenic potential (Counter et al., 2004), even modest exceedances pose considerable public health risks. Nevertheless, Hg risk assessment should systematically take into account the molar ratio between Hg and selenium (Se). Tuna have consistently been reported to have higher Se:Hg molar ratios (2.89) according to Barone et al. (2021), suggesting enhanced detoxification capacity via formation of inert Hg-Se complexes.

Cadmium levels in our tuna samples were lower than those reported for fresh Mediterranean tuna (Storelli et al., 2020), and Pb concentrations remained below those measured by Hussein et al. (2024) in canned tuna. However, our tuna showed higher levels of all three toxic metals (Cd, Pb, and Hg) than those reported by Mol et al. (2018), Miedico et al. (2020), Lee et al. (2016), Jinadasa et al. (2019) and Novakov et al. (2017), highlighting geographic and methodological variability.

Overall, the concentrations of toxic elements recorded in swordfish were lower than those previously reported in the Mediterranean Sea (Olmedo et al., 2013; Gobert et al., 2017; Barone et al., 2018; Mehoul et al., 2019; Storelli et al., 2020).

Noteworthy, swordfish showed Hg values similar to those of mussels (0.350 vs 0.382 mg kg<sup>-1</sup>), a benthic filter-feeding species. This finding challenges the expectation of systematic Hg biomagnification in top predators, but aligns with literature reporting inconsistencies in Hg bioamplification among marine top predators. These variations are mainly attributable to factors such as differences in growth rates, physiological detoxification or elimination mechanisms, habitat depth affecting prey mercury load, and diet composition.

Despite their shared status as top predators, the results of the present study revealed significantly higher Hg levels in tuna than in swordfish ( $P < 0.05$ ). Similar findings were reported by Storelli et al. (2001) in specimens from the Italian coast (1.020 vs 0.490 mg.kg<sup>-1</sup>). This difference may be attributed to tuna's predominantly piscivorous diet, characterised by the consumption of larger teleost fish, which contributes to higher mercury accumulation compared to swordfish, whose diet includes smaller prey such as crustaceans and cephalopods which tend to concentrate more Cd than Hg (Storelli et al., 1999). Moreover, tuna tend to frequent coastal areas, increasing their exposure to local pollutants. In contrast, swordfish are migratory oceanic pelagic species that spend less time in contaminated coastal waters such as those around Skikda.

Bonito, a highly migratory top predator, exhibited greater concentrations of Cd, Hg, and Pb compared to previous findings by Kuplulu et al. (2018) in Marmara Sea, Chahid et al. (2014) in the Atlantic Ocean, and Makedonski et al. (2015) in the Black Sea. The Pb concentration we observed in bonito was also higher than that reported by Verép and Mutlu (2022) in the Black Sea.

The grey mullet, a pelagic species inhabiting regions near sediments at lower trophic levels, showed heavy metal content in muscle similar to Hg and Pb levels reported by Stancheva et al. (2014) in the Black Sea, though Cd levels were lower. Cd levels in our samples were considerably higher than those reported by Fazio et al. (2020) in the Ionian Sea (0.001 mg kg<sup>-1</sup>), and in the Black Sea (0.039 mg kg<sup>-1</sup>). Conversely, Pb concentrations were similar to the value observed in the Ionian Sea (0.057 mg kg<sup>-1</sup>) but substantially lower than those measured in the Black Sea (0.191 mg kg<sup>-1</sup>).

In this study, the concentrations of Cd were found to be below the maximum acceptable limit established by the European Commission for human consumption (OJEU, 2014) in all species, except for mullet, which had a mean concentration of 0.139 mg kg<sup>-1</sup>, i.e. almost three times higher than the permitted limit (0.05 mg kg<sup>-1</sup>). Not far from the coast of Skikda, grey mullet caught in the Gulf of Annaba (Algeria) contained over nine times the permitted limit, i.e. 4.6 mg/kg<sup>-1</sup>. These findings highlight a potential health risk for consumers regularly ingesting grey mullet from contaminated coastal areas in Algeria.

Due to its abundance, the sardine (*Sardina pilchardus*) is one of the most commonly consumed fish species in Algeria and the Mediterranean region. Overall, the average concentrations of heavy metals measured in sardine muscle were lower than those reported in previous Algerian studies, particularly those conducted by Mehoul et al. (2019) along the Algerian coast (Cd: 0.550 mg kg<sup>-1</sup>, Hg: 0.620 mg kg<sup>-1</sup>, Pb: 2.130 mg kg<sup>-1</sup>) and by Aissioui et al. (2021) in Boumerdes Bay (Cd: 0.100 mg kg<sup>-1</sup>, Hg: 0.070 mg kg<sup>-1</sup>, Pb: 0.170 mg kg<sup>-1</sup>). However, sardines collected from the same bay by Hamida et al. (2018) were less contaminated, with lower Pb (0.055 mg kg<sup>-1</sup>) and Cd (0.031 mg kg<sup>-1</sup>) levels. Elsewhere in the Mediterranean, Storelli et al. (2020) observed lower levels of all the tested toxic metals, whereas the lead concentrations reported by El-Sherbiny and Sallam (2021) were comparable to those found in our study.

Beyond the Algerian coast, Novakov et al. (2017), El-Sherbiny and Sallam (2021), and Hussein et al. (2024) reported higher levels of Pb and Hg in canned sardines, whereas Korkmaz et al. (2017) found lower Pb concentrations.

Sardine and anchovies are used as bioindicators in marine heavy metal pollution monitoring. With its high levels of essential elements and relatively low concentrations of heavy metals, *Sardina pilchardus* emerges as a favorable candidate for consumption, offering a beneficial nutrient profile with minimal contamination risks.

In this study, *Parapenaeus longirostris* commonly known as the deep-water rose shrimp, was found to have higher levels of Cd and Hg than those reported by Kuplulu et al. (2018) in shrimp from the Aegean Sea. Additionally, our findings exceeded the Cd and Pb concentrations found by Aytekin et al. (2019); and the Cd and Hg levels observed by Barone et al. (2015) and Nyarko et al. (2023) in shrimp from the Mediterranean Sea.

Toxic metal levels reported by Habte et al. (2015), in two shrimp species from the Western Pacific Ocean were lower than those observed in the current study. However, the Pb content we found in the analyzed muscle tissue was lower than that recorded by Kuplulu et al. (2018) in the Aegean Sea.

*Mytilus galloprovincialis*, commonly known as the Mediterranean mussel, is a sessile benthic filter-feeder that is in constant contact with metal-rich sediments. It has less efficient mechanisms for rapid excretion of heavy metals,



leading to significant bioaccumulation in its tissues.

The mussels collected by Kuplulu et al. (2018) from the Marmara Sea had Hg concentrations similar to those in our study. However, Barchiesi et al. (2020) found lower Hg, Cd, and Pb levels in mussels sampled between 2017 and 2019 than we did. Likewise, the Cd content found by Zhelyazkov et al. (2018) in mussels from the Black Sea was almost half of the level recorded in our study. Along the Italian coast, Esposito et al. (2021) found significantly lower levels of the three toxic elements, as did Novakov et al. (2021) in mussels consumed in Serbia.

Furthermore, the levels of toxic elements we recorded in mussels were higher than those reported by Kuplulu et al. (2018) in the Black Sea, Marmara Sea, Aegean Sea, and Mediterranean Sea, except for Hg in mussels from the Black Sea, which reached a value of  $0.405 \text{ mg kg}^{-1}$ . Notably, both Kuplulu's study and our own findings revealed that mussels exhibited the highest levels of Cd. This observation confirms their role as sedentary filter feeders and effective bioaccumulators of pollutants.

Levels of Fe and Zn consistently surpassed those of non-essential elements, confirming their biological importance. Mussels showed the highest concentrations of essential elements among the studied species, with Fe slightly exceeding the regulatory limit ( $50.41$  vs.  $45 \text{ mg kg}^{-1}$ ), suggesting both nutritional value and potential risk if consumed in excess. Zn concentrations did not exceed the permissible threshold ( $50 \text{ mg kg}^{-1}$ ) in any of the studied species. The obtained value of  $19.79 \text{ mg kg}^{-1}$  is consistent with that reported by Hamida et al. (2018) in sardines from Boumerdès Bay (Algeria).

The obtained values are within or below the ranges reported for fish from the Mediterranean, Black Sea, Atlantic, and subtropical coasts (Gobert et al., 2017; Barone et al., 2018; Antović et al., 2019; Aytakin et al., 2019; Korkmaz et al., 2019; Fazio et al., 2020; Storelli et al., 2020; Esposito et al., 2021;), though Fe concentrations in mussels were lower than those found by Ouali et al. (2018) in the Gulf of Annaba ( $147.73 \text{ mg kg}^{-1}$ ). Our results confirm Fe as the most accumulated essential element, in line with its physiological roles in haemoglobin synthesis, cellular metabolism, and gene regulation.

This study was conducted during spring and summer (February to August). While it is uncertain whether results would differ significantly in other seasons, previous studies have shown that heavy metal accumulation in fish tissues (particularly muscle) is influenced by seasonal variations affecting reproductive cycles, food availability, and diet composition (Rajeshkumar and Li, 2018; Aytakin et al., 2019; Mirzaei et al., 2019; Esposito et al., 2021; Sofoulaki et al., 2022; Monier et al., 2023). Zeghdoudi et al. (2018) reported that Cd and Pb concentrations in fish muscle from the Gulf of Skikda were higher during winter and summer, whereas their levels in sediments peaked in autumn and winter. In a study conducted along the Algerian coasts (Algiers and Bejaia), Aissioui et al. (2022) reported that Pb and Cd concentrations in sardines reached their highest levels in spring, whereas Hg concentrations peaked in autumn. Esposito et al. (2021) reported that Hg concentrations in Mediterranean mussels during summer were nearly twice those observed in spring and autumn.

Overall, these studies indicate that metal concentrations in aquatic organisms and seawater tend to be higher in spring and summer, underscoring the seasonal variability in heavy metal accumulation.

Species	Cd	Pb	Hg	Zn	Fe	References
Bonito	0.022	0.114	0.064	-	-	Chahid <i>et al.</i> , 2014
	0.015	0.060	0.013	10.00	-	Makedonski <i>et al.</i> , 2015
	0.005 0.008	0.060 0.070	0.028 0.064	- -	- -	Kuplulu <i>et al.</i> , 2018
	0.060	-	-	3.80-55.00	19.00	Korkmaz <i>et al.</i> , 2019
	-	0.250-0.330	-	8.40-16.60	10.41-18.42	Verep and Mutlu, 2022
Swordfish	-	-	0.490	-	-	Storelli <i>et al.</i> , 2001
	0.009	0.004	0.540	-	-	Olmedo <i>et al.</i> , 2013
	0.180	-	0.800	-	-	Barone <i>et al.</i> , 2015
	0.033	0.084	-	30.27	4.80	Gobert <i>et al.</i> , 2017
	0.160	0.110	0.770	8.34	-	Barone <i>et al.</i> , 2018
	0.570	3.900	0.560	-	-	Mehouel <i>et al.</i> , 2019
	0.170	0.130	0.590	12.00	-	Storelli <i>et al.</i> , 2020
Tuna	-	-	1.020	-	-	Storelli <i>et al.</i> , 2001
	-	-	0.760	-	-	Lee <i>et al.</i> , 2016
	0.030	0.150	0.180	22.00	20.36	Novakov <i>et al.</i> , 2017
	0.002	0.115	0.001	8.34	-	Mol <i>et al.</i> , 2017
	0.017	-	0.480	-	-	Jinadasa <i>et al.</i> , 2019
	0.110	0.130	0.470	10.25	-	Storelli <i>et al.</i> , 2020
	0.029	0.013	0.210	-	-	Miedico <i>et al.</i> , 2020
	-	0.307	0.650	-	-	Hussein <i>et al.</i> , 2024
Gray Mullet	0.012	0.050	0.050	5.20	2.20	Stancheva <i>et al.</i> , 2014
	0.475	1.210	-	108.00	147.73	Ouali <i>et al.</i> , 2018
	0.001 0.039	0.057 0.191	- -	19.00 4.50	5.90 11.00	Fazio <i>et al.</i> , 2020
Sardine	0.024	0.076	0.084	-	-	Chahid <i>et al.</i> , 2014
	0.020	0.150	0.130	18.00	21.98	Novakov <i>et al.</i> , 2017
	0.031	0.055	-	19.80	-	Hamida <i>et al.</i> , 2018
	0.550	2.130	0.620	-	-	Mehouel <i>et al.</i> , 2019
	0.047	0.194	0.272	-	-	El-Sherbiny and Sallam, 2021
	0.020	0.060	0.040	3.40	-	Storelli <i>et al.</i> , 2020
	0.100	0.170	0.070	-	-	Aissiou <i>et al.</i> , 2021
	0.050	0.477	0.673	-	-	Hussein <i>et al.</i> , 2024
Shrimp	0.002	0.004	0.034	3.40	18.00	Barone <i>et al.</i> , 2015
	0.156 0.013 0.034	0.054 0.039 0.452	0.010 0.003 0.007	- - -	- - -	Habte <i>et al.</i> , 2015
	0.037 0.051 0.091	0.173 0.015 0.068	0.061 0.094 0.014	- - -	- - -	Kuplulu <i>et al.</i> , 2018
	0.027	-	0.026	19.00	-	Nyarko <i>et al.</i> , 2023
	8.330 6.590	62.75 22.18	- -	61.42 37.43	24.23 22.50	Aytekin <i>et al.</i> , 2019
	0.097 0.087 0.143	0.375 0.267 0.405	0.405 0.341 0.079	- - -	- - -	Kuplulu <i>et al.</i> , 2018
	0.112	0.366	0.203	-	-	
	0.280	0.251	0.017	-	-	Zhelyazkov <i>et al.</i> , 2018
Mussel	0.099 0.105 0.108	0.128 0.180 0.215	0.038 0.032 0.017	- - -	- - -	Barchiesi <i>et al.</i> , 2020
	0.082 0.033 0.050	0.190 0.071 0.200	0.013 0.021 0.011	21.00 29.00 25.00	44.00 26.00 27.00	Esposito <i>et al.</i> , 2021
	0.160	0.320	0.060	-	-	Novakov <i>et al.</i> , 2021

**Table III.** Concentrations of Cd, Pb, Hg, Zn, and Fe (mg kg<sup>-1</sup> ww) in edible tissues of marine organisms reported in literature.

## Conclusion

This study demonstrates significant inter-species variability in both essential (Fe, Zn) and non-essential (Cd, Pb, Hg) element concentrations in muscle tissue of seven commercially important marine species from the Algerian coast. The results illuminate the dual role of seafood as both a nutrient source and a vector for toxicant exposure. While sardines and shrimp are a sustainable and risk-free source of nutrition, tuna and grey mullet consumption warrants caution. Tuna exhibited the highest Hg concentration, reflecting strong biomagnification; grey mullet had a Cd level nearly three times above the EU limit; and mussels accumulated the highest levels of Cd and Pb, yet remained within permissible limits.

Further investigations are needed to target other marine species in different locations and at various times of the year along the Algerian coast.

## Acknowledgments

In memory of Professor Smail MEHENNAOUI. We, the co-authors and his former students, remain eternally grateful for his guidance, generosity, and the enduring influence he exerted on our academic and professional paths.

## Conflict of interest

The authors declare that they have no conflict of interest.

## Author Contributions

Conceptualization: S.M and S.S; Methodology: Z.G, S.S, L.N and A.B; Formal analysis: A.B; Investigation: L.N, A.B and H.A; Writing original draft preparation: L.N, S.S and A.B; Writing, review and editing: S.S and A.B; Visualization: S.S, A.B and Z.G; Supervision: S.M and S.S; Project administration: L.N and H.A; Funding acquisition: L.N and Z.G; All authors have read and agreed to the published version of the manuscript.

## Data availability

All data generated or analysed during this study are available from the corresponding author upon request

## Fundings

This research was supported by the affiliated research laboratories

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