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**Assessment of trace element contamination and bioaccumulation in algae (*Ulva lactuca*),  
bivalves (*Spondylus spinosus*) and shrimps (*Marsupenaeus japonicus*) from the Lebanese coast**

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## **Abstract**

In this study, levels of 20 trace elements (Al, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Ag, Cd, Sn, Sb, Ba, Hg, Pb and U) and the respective biota-sediment accumulation factors (BSAF) were assessed for three marine species from the Eastern Mediterranean (Lebanese coast) belonging to different trophic levels in the food chain (algae, bivalves and shrimps). Multivariate analyses showed significant differences in trace element accumulation between seasons, sites and species thus highlighting the importance of a multi-species and seasonal approach to monitor trace metals in coastal areas. Biota-sediment accumulation factors (BSAF) showed limited transfer from sediments to marine organisms suggesting that the bioaccumulation of trace elements by marine organisms should be assessed by considering multiple sources of contaminants. Lead (Pb) levels (0.07-4.44 mg.kg<sup>-1</sup>) measured in algae and Cd (0.54-7.74 mg.kg<sup>-1</sup>) and Zn (42.88-972 mg.kg<sup>-1</sup>) levels found in the oyster, were similar and sometimes higher than those found in other polluted areas indicating that the Lebanese coast is subject to environmental pollution.

**Keywords:** Bioaccumulation, trace elements, marine organisms, Lebanon, Eastern Mediterranean

## **1. Introduction**

The Mediterranean Sea is known for its high biodiversity; however, significant amounts of chemical contaminants mainly introduced in coastal areas via industrial, agricultural and mining activities, are threatening the marine ecosystem (Lafabrie et al., 2008; Ourgaud et al., 2018; Wang et al., 2010). Toxic trace elements, especially trace metals, which constitute an important part of these discharges, are important pollutants of aquatic environments mainly because of their persistence and tendency to concentrate in marine organisms (Afshan et al., 2014; Battuello et al., 2016).

Three marine compartments including water column, sediment and biota are usually used to monitor the spatial and temporal trends of marine pollution. Solubility, bioavailability and toxicity of trace elements in seawater are controlled by several physico-chemical parameters, such as pH, temperature, salinity, dissolved oxygen, suspended particles and dissolved organic matter (Chakraborty et al., 2014; Lim et al., 2012). Moreover, trace element concentrations in water and sediment are affected by a variety of environmental factors such as tides, winds, currents and fresh water input (Hatje, 2003; Strady et al., 2011). These processes create a stochastic pattern making it difficult to predict the potential adverse health effects on the surrounding organisms caused by accumulation, suspension and mobilization of sediments (Meng Chuan and Yunus, 2019; Rajeshkumar et al., 2018; Ribeiro et al., 2018). Sediments do not only act as sinks, but also as sources of chemical contaminants in the marine environment; therefore, they can be used as geo-markers for potential pollution sources and conditions (Meng Chuan and Yunus, 2019). Moreover, unlike water, sediments reflect long term contamination conditions (Thomann et al., 1995) but they do not necessarily reflect the biota contaminants (Meng Chuan and Yunus, 2019; Rainbow, 2007; Wang et al., 2010). Many marine species have been shown to accumulate trace elements, hence acting as suitable indicators for monitoring local trace elements pollution and their potential ecological and human risks (Campanella et al., 2001; Conti and Cecchetti, 2003). These species accumulate trace elements reaching levels that are many times higher than those in the water column or sediments (Conti and Cecchetti, 2003; Diop et al., 2016).

Accordingly, biomonitoring by using species that belong to different taxonomic groups and trophic levels provides a better assessment of the extent of any contamination in terms of presence and relative magnitude (Campanella et al., 2001; Pérez-Cadahía et al., 2004; Rainbow, 1995a). A suitable bioindicator should (i) be able to accumulate high levels of pollutants without death; (ii) have a sessile lifestyle to reflect local pollution; (iii) be relevant in the food chain and (iv) be widespread and abundant (Zhou et al., 2008). Additionally, ease of sampling and identification are criteria to take into consideration when targeting a bioindicator (Rainbow and Phillips, 1993). So far, mollusks, crustaceans and macrophytes have been shown to be valuable cosmopolitan bioindicators and have been used in several studies (Amiard et al., 2006; Bonsignore et al., 2018; Chakraborty et al., 2014; Chiarelli and Roccheri, 2014; Diop et al., 2016; Rouane-Hacene et al., 2015; Viarengo and Canesi, 1991). Understanding how trace elements are bioaccumulated in food webs is essential for elucidating their environmental behavior, ecotoxicological risks and the potential hazards to human health (Chen et al., 2000).

The substantial development of urban and industrial activities along the coast in Lebanon, as well as, the lack of wastewater treatment strategies, resulted in increased inputs of chemical contaminants to the marine environment (Fakhri, 2012). In addition, waste production in Lebanon has increased significantly during the last decade, and the country had experienced a waste crisis in 2015. The regulated dumpsites could no longer manage the tremendous quantity of municipal garbage generated; therefore, numerous illegal and open dumpsites were created throughout the country mainly along the coast (Khalil et al., 2018). Leachate effluents originating from these coastal landfills are an important source of trace elements contamination in the surrounding aquatic environment (Stefanakis et al., 2014; Youcai, 2018), and constitute a serious risk to biota (Brand et al., 2018; Toufexi et al., 2013).

Most environmental studies conducted along the Lebanese coast focused on the contamination of water and sediments (Abi-Ghanem, 2008; Abi-Ghanem et al., 2011; Merhaby et al., 2018). Few studies mostly dating back to the 1980s have focused on marine organisms (Kouyoumijian et al.,

1986; Kouyoumjian et al., 2001; Shiber, 1981; Shiber and Shatila, 1978; Nakhlé, 2003) while environmental pressure along the coast has been in continuous increase. To our knowledge, no studies have assessed the bioaccumulation of trace elements along the Lebanese coast, and particularly in organisms belonging to different ecological niches or trophic levels.

The aim of this study was to evaluate the contamination status related to 20 trace elements (Al, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Ag, Cd, Sn, Sb, Ba, Hg, Pb and U) of three benthic marine species from different trophic levels, representative of the Lebanese coast (Eastern Mediterranean). Samples were collected at three sites during dry and wet seasons in 2017. The species selected are the green algae *Ulva lactuca* that is widely used as a bioindicator of trace elements pollution, the spiny oyster *Spondylus spinosus* and the kuruma shrimp *Marsupenaneus japonicus* that are commonly consumed by the Lebanese population and studied for the first time to assess trace elements contamination along the Lebanese coast. Moreover, the results of trace elements in sediments were used to calculate the biota-sediment transfer factor (BSAF) from sediments to biota.

## **2. Materials and methods**

### *Study area*

Three sites representing the coastline with different levels and types of anthropogenic pressures were selected for this study (Fig. 1). Tripoli (site 1) is mainly known for its artisanal fishing and boat maintenance activities. Several sewage outfalls that discharge wastewater without prior treatment are identified along its coast. A controlled dumpsite north of its port is still receiving waste despite a decision of closure in 2012, thus, leading to increased waste height and uncontrolled leachate (El-Hoz, 2017). Beirut, the capital of Lebanon (site 2), is a densely populated (up to 3500 individuals per square meter) and an industrialized area (Fakhri, 2012). Several sewage outfalls discharge polluted water in the latter's coastal zone, in addition to the Beirut River that drains industrial effluents into the sea causing water quality degradation. The area comprises textiles, cement and metal fabrication industries, in addition to tanneries, slaughterhouses, as well as, wood

and furniture production (Fakhri, 2012). Moreover, there are three dumpsites present in this area with the biggest one in the Dora region. The latter dumpsite is responsible for more than half of Lebanon's trace elements discharges in the coastal area (Fakhri, 2012). Saida (site 3) is less impacted by anthropogenic pressures compared to Beirut despite being the third densest coastal city in Lebanon. A dumpsite was rehabilitated there in 2014, and a primary wastewater treatment plant has been recently established in its coastal area.

#### *Sample description and collection*

Three marine species belonging to different levels of the food chain were selected for this study. The green macroalgae, *Ulva lactuca* is a cosmopolitan species that is widely used as a bioindicator (Diop et al., 2016; Ho, 1990; Kaimoussi et al., 2004; Kamala-Kannan et al., 2008). The spiny oyster, *Spondylus spinosus* is a Lessepsian species that is very well-established in the Eastern Mediterranean, has gained commercial importance and is frequently consumed by the Lebanese population. This bivalve species lives attached to rocky substrates and feeds on particles from the water column (Aksit et al., 2013). The kuruma shrimp, *Marsupenaeus japonicus*, is also a Lessepsian species in the Mediterranean. This species inhabits sandy and mud bottoms and feeds on algae, small fish and bivalves (Galil, 2006; Quigley et al., 2013). These three species were chosen because they live at different depths, and can thus represent the contamination levels at different habitats along the coast.

The algae and the spiny oysters were collected by hand picking in the subtidal zone at depth varying between 0.5 and 1.0 m, as well as, 10 and 20 m, respectively. The kuruma shrimps were sampled with trammel nets at an average depth of 40 m. All species were collected at each site and during every season (wet: December-March and dry: June-September). *Ulva lactuca* was not found at Beirut and Saida during the dry season, and *M. japonicus* was not found at Beirut during the wet season and Saida during the dry season. The samples were taken immediately to the laboratory on ice. Each individual was measured (nearest 1 mm) and weighed (nearest 0.1 g) in order to standardize individual size between sites. Ten individuals of each species were selected for analysis.

The soft tissues of bivalves and shrimps were removed under a laminar flow hood using a stainless-steel scalpel. They were then stored in low-density polyethylene vials at -20 °C. Algae were rinsed with distilled water to remove substrate residues. Samples were then lyophilized at -100 °C until constant weight was achieved and stored at room temperature until analysis. Additionally, surficial sediment samples were collected (hand picking using a stainless-steel shovel) by diving at each site of the coastal area at depths between 5 and 20 m, during the dry season. They were then placed in plastic bags and taken to the laboratory.

### *Sample analysis*

Bivalves and shrimp soft tissues, as well as, algae were analyzed using an accredited method (French Accreditation Comity-COFRAC) described elsewhere (Chevallier et al., 2015). Samples were lyophilized and their moisture content was determined. Aliquots of 0.3 g to 0.5 g of each sample were weighed in quartz digestion vessels and then mixed with 3 mL of HNO<sub>3</sub> 67 % (m/m) (suprapur grade, VWR chemicals, Prolabo, France). Three mL of ultrapure water were added, and the mineralization procedure was carried out using a closed microwave system (Multiwave 3000 from Anton Paar, France). Sample digests were quantitatively transferred to polypropylene tubes and a solution containing a mixture of internal standards, scandium (Sc), indium (In), bismuth (Bi), rhenium (Re) and yttrium (Y) was added at a final concentration of 2 µg.L<sup>-1</sup>. The mixtures were then added up with ultrapure water to a final volume of 50 mL. In order to reduce the memory effect of mercury, a solution of gold (Au) was added at a final concentration of 20 µg.L<sup>-1</sup>. Samples were analyzed using an ICP-MS 7700 (Agilent Technologies, Courtaboeuf, France).

### *Sediment analysis procedure*

An aliquot of 0.5 g of dried sediment was totally digested in a heating block (HotBlock® SC100 Digestion System), with a mixture of concentrated acids containing HF, HNO<sub>3</sub> and HCl in 10:1:3 (v/v/v) ratio, based on the procedure described by Ouddane et al [1999]. The sample was weighed into PTFE digestion cups, when the sediments were totally dissolved at 120 °C, the solution was then diluted with ultrapure Milli-Q+ water (18.2 MΩ cm) and filtered through a 0.45 µm cellulose



acetate membrane. Trace element concentrations were determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES, Agilent 5110, dual view) for the major elements (Al and Fe) and by ICP-MS (Varian, 820) for the trace elements (Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, V and Zn). Procedural blanks and certified reference materials (CRMs): ERM-278K (National Research Council of Canada) were prepared using the same analytical procedure and analyzed for quality assurance/quality control. All results were >90% comprised in the confidence intervals.

### *Quality control*

In order to ensure the reliability of the results, several internal quality controls (IQCs) and associated criteria were used. Each run included a standard calibration, three blanks, three CRMs, two different spiked sample solutions, 33 samples including 2 samples analyzed in duplicate and a mid-range standard for every eight samples and at the end of the sequence. The set criteria were as follows: calibration ( $r^2 > 0.995$ ), blanks (values < limit of quantification (LOQ)), internal standards (values within 70 and 130% of the target value), mid-range standards (values within 80 and 120% of the target value), spiked standard solutions (spike recovery within 70 and 130% of the theoretical spiked standard value), CRMs (Z-score <  $\pm 2$ ) and duplicates (acceptable if the relative standard deviation (RSD)  $\leq 20\%$  when mean value  $\geq 5 \times \text{LOQ}$  or RSD  $\leq 40\%$  when mean value  $\geq \text{LOQ}$  and  $< 5 \times \text{LOQ}$ ). When acceptance criteria were not achieved, the results were discarded, and the samples were re-analyzed.

### *Data treatment*

Statistics were performed using RStudio Team (2016). The level of significance was set at  $\alpha=0.05$ . As the data was not normally distributed, Kruskal-Wallis non-parametric test and Mann Whitney U-test for pairwise comparison were used to evaluate spatial and inter-specific differences as well as seasonal variations in elemental concentrations. Principal component analysis (PCA) was used after  $\log(x+1)$  transformation of the data to explore relationships between sites, species and elemental concentrations for the two sampling seasons. Left censored data that are < LOQ and < LOD were

excluded from statistical analysis (23 % of the data). All results were reported in  $\text{mg.kg}^{-1}$  dry weight (dw).

The biota-sediment accumulation factor (BSAF) is defined as the ratio between the concentration of a given element in a marine organism ( $C_o$ ) and its concentration in sediment ( $C_s$ ) (both concentrations are expressed in  $\text{mg.kg}^{-1}$  dry weight). Bioaccumulation was confirmed when BSAF was  $> 1$  (Thomann et al., 1995).

### **3. Results and discussion**

The concentrations of 15 trace elements are presented in Table 1 and Table 5. Trace elements like Ag, Sn, Sb, Ba and U were not detected or showed very low detection frequencies ( $< 20\%$ ) in the tissues of the selected marine organisms; thus, they were not presented in this study.

#### ***3.1. Interspecific variability of trace element concentration***

For simplicity, the data were processed to reflect the interspecific variability by averaging all data for all sites regardless of the season. Al and Fe showed highest concentrations in algae ( $378\pm 413 \text{ mg.kg}^{-1} \text{ dw}$  and  $515\pm 473 \text{ mg.kg}^{-1} \text{ dw}$ , respectively), while Zn ( $124\pm 162 \text{ mg.kg}^{-1} \text{ dw}$ ) and As ( $35.2\pm 27.42 \text{ mg.kg}^{-1} \text{ dw}$ ) showed highest concentrations in bivalves, and Cu showed highest concentrations in shrimps ( $30.91\pm 11.96 \text{ mg.kg}^{-1} \text{ dw}$ ) regardless of the season or site (Table 1). High standard deviations were observed indicating a strong variability among the levels in the same species. Trace element concentrations showed significant differences between all species ( $p < 0.001$ ). Through multivariate analysis (not shown here), the elemental concentration explained 79% of the interspecific variation. Each species showed affinity to a certain group of trace elements. For instance, *U. lactuca* showed significantly higher levels of trace elements in the following order: Fe  $>$  Al  $>$  Ti  $>$  Mn  $>$  V  $>$  Pb  $>$  Cr ( $p < 0.001$ ), while the spiny oyster had the highest levels of Zn  $>$  As  $>$  Ni  $>$  Se  $>$  Cd  $>$  Co ( $p < 0.001$ ) and the kuruma shrimp had the highest levels of Cu and Hg ( $p < 0.001$ ). Similar concentration trends were also recorded in previous studies (Table 2) (Akcali and

Kucuksezgin, 2011; Circuncisão et al., 2018; Kaimoussi et al., 2004; Shams El-Din et al., 2014; Villares et al., 2002).

Marine organisms acquire trace elements from solution and/or particulate matter. The latter pathway consists of the ingestion of suspended detritus, food prey or even deposited organic matter (Amiard et al., 2006; Rainbow and Phillips, 1993). The bioaccumulation of trace elements in marine organisms is also related to several parameters contributing to inter- and intraspecific variability, namely biotic factors such as the size, age, sex, and metabolism in addition to the position of the organism in the food chain (Jakimska et al., 2011). For example, macrophytic algae essentially respond to dissolved trace element sources and are thus good bioindicators of water quality. They are also known for their exceptional capacity to accumulate essential and trace elements (Farias et al., 2018). That capacity is actually related to the cell walls of green macroalgae and particularly the *Ulva* genus. The cell walls are composed of ulvans which are sulphated heteropolysaccharides with ionic binding properties due to their sulphate groups (Alves et al., 2013; Cunha and Grenha, 2016). More specifically, *U. lactuca* is known to have a high capacity to accumulate trace elements with particular affinity to Mn, Fe, Cu, Zn, Pb and Cd (Ghoneim et al., 2014; Ho, 1990). It has been used in bioremediation for Cd, Pb and Hg removal from aqueous solutions (Ghoneim et al., 2014; Henriques et al., 2017). Studies dealing with *U. lactuca* usually report higher levels of Fe (Abdallah and Abdallah, 2008) that seems to be dominant in Chlorophyta in general, while Rhodophyta and Phaeophyta (red and brown algae, respectively) tend to accumulate higher levels of Mn (Circuncisão et al., 2018). When comparing to other studies conducted in the Mediterranean area (Table 2), levels of trace elements in the *Ulva* from our study were generally lower and even closer to values recorded in relatively “uncontaminated” areas; however, Fe was higher (Campanella et al., 2001; Conti and Cecchetti, 2003). Iron (Fe) is actually known to be an essential element for the normal growth of marine plants (Storelli and Marcotrigiano, 2001). Algal species are able to biomagnify Fe from the surrounding water while industrial effluents continuously increase Fe inputs in the marine environment, which may explain the higher levels observed in our study (Akcali and

Kucuksezgin, 2011). A decreasing trend of Pb in the marine environment has been reported, since a regulation limiting the use of leaded gasoline consumption was established in the year 1970 (Storelli, 2008). Indeed, levels of Pb in *U. lactuca* exhibited a decrease when compared to a study done in Beirut in 1977 (Shiber, 1980). However, the difference between the detection techniques used back then and the recent methods currently should be taken into consideration.

The levels of trace elements in the spiny oyster from the present study were found to be mostly lower than concentrations found in bivalves of other countries around the Mediterranean (Table 2). For instance, trace elements in the spiny oyster were lower than the ones reported from the Iskenderun Bay, which is considered a polluted area in Turkey (Türkmen and Türkmen, 2005) with few exceptions for Fe, Cu and Zn that are essential elements. Despite the fact that the spiny oyster satisfies all the criteria for a good bioindicator (Aksit et al., 2013), it has not been commonly used in biomonitoring studies in the Mediterranean. In fact, this species lives attached to rocky substrates in the subtidal zone at a depth of more than 10 m; therefore, its sampling is difficult. Species that are easy to sample and identify such as *Patella caerulea* and *Brachidontes variabilis*, have been the subject of several studies around the Mediterranean (Table 2). As a filter feeder, living a little further from the coast compared to other bivalve candidates, *S. spinosus* would provide information about the contamination status of the surrounding water in the subtidal zone. Levels of Cd found in the spiny oyster were similar to the ones found in *P. caerulea* and *B. variabilis* collected previously along the Lebanese coast (Table 2; Nakhlé et al., 2006). The concentrations of Cu, Cr, Cd and Pb were lower than the ones found in *M. galloprovincialis* and *P. Caerulea* from the Tyrrhenian Sea, Italy and concentrations of Fe, Cu, Cd, Zn and Mn were lower compared to concentrations found in *Donax trunculus* (bivalve) from the Egyptian coast (Table 2). Bivalves are known for their ability to accumulate trace elements in their tissues (Zuykov et al., 2013). They are filter-feeding organisms with low biotransformation capacities that accumulate the bioavailable fraction of contaminants from water at higher levels than those in the ambient environment. Therefore, they can be used as sentinel species to monitor pollution in coastal waters (Storelli and Marcotrigiano, 2001; Türkmen

and Türkmen, 2005). High levels of Zn and Cu in mollusks are often attributed to their essentiality. In general, ranges of trace elements in the spiny oyster of this study fall between the levels observed in contaminated and relatively uncontaminated areas. This suggests a relatively moderate level of trace element contamination in this species when compared to oysters, mussels and limpets collected from other regions around the Mediterranean (Table 2). However, caution must be taken as these species neither share the same habitat of *S. spinosus* nor the same feeding habits. *Patella caerulea* also known as the Mediterranean limpet, is a gastropod that inhabits the medio littoral zone and grazes on phytoplankton and macroscopic algae. *Brachidontes variabilis*, also known as the variable mussel, inhabits shallow waters (at sea level or just below) and lives attached to rocky substrates (Nakhlé et al., 2006).

The kuruma shrimp had the highest levels of Cu compared to ulva and spondylus. Crustaceans are known to have hemocyanin, which is an oxygen carrying molecule containing a copper binding site also found in mollusks and arthropods, thus, explaining higher levels of these molecules in crustaceans and bivalves compared to algae (Downs et al., 2016; Rainbow and Luoma, 2011). Also known for being deposit feeders, crustaceans are more exposed to sediment related contaminants such as As which is more abundant in sediments than in the water column (Kalantzi et al., 2017). Few studies have measured trace element concentrations in the shrimp *M. japonicus*, particularly in the Mediterranean Sea (Ayas, 2016). More studies were conducted on the caramote prawn, *Penaeus kerathurus* (Table 2). Levels of trace elements in our study lie in the same range as the ones found in other studies except for Cu and Zn that were around two-fold higher than the ones found in species collected from other sites (Table 2). These two elements are essential as mentioned above however, anthropogenic inputs may increase their levels.

### **3.2. Seasonal and spatial variations**

Trace element levels showed seasonal and spatial variation patterns in all species (Table 3 and Table 5). The sites chosen in this study have a different pluviometric pattern during the year. North

Lebanon receives more rain than its southern region. During the wet season surface runoff and riverine inputs increase with rainfall and snow melting, all of which affect the physico-chemical properties of water. Parameters such as pH, salinity, water hardness and oxygen concentration revealed significant variation according to seasons (Abboud-Abi Saab et al., 2004; Fakhri and Romano, 2005). These conditions are known to govern the bioavailability of trace elements in the marine environment and their bioaccumulation by marine organisms (Rainbow, 1995a).

Levels of Al, V, Cr, Mn, Fe, Co, Ni, As, Se and Pb were significantly higher in algae during the dry season ( $p < 0.05$ ) (Table 3). Such a pattern was previously observed in *U. lactuca*, with increasing trace element concentrations from winter to summer (Kaimoussi et al., 2004). Since the intake of trace elements in algae depends on their concentration in the surrounding water (Ghoneim et al., 2014; Rybak et al., 2012), a higher concentration during summer could be attributed to the increased evaporation and decreased rainfall especially for species living in shallow waters like ulva (Rajeshkumar and Li, 2018; Wang et al., 2012). Higher trace element accumulation during summer can also be explained by the existence of a higher metabolic rate during this season (Villares et al., 2002). Finally concerning the spatial variation, levels of Al ( $779 \pm 611 \text{ mg.kg}^{-1}$ ), As ( $7.55 \pm 2.15 \text{ mg.kg}^{-1}$ ), Cr ( $1.09 \pm 0.13 \text{ mg.kg}^{-1}$ ) and Pb ( $1.42 \pm 0.23 \text{ mg.kg}^{-1}$ ) were significantly higher in the algae collected from Tripoli compared to Beirut and Saida ( $p < 0.05$ ) (Table 5). Indeed, the accumulation pattern of trace elements by algae depend on the developmental stage and the physiology of algae (Farias et al., 2018). It has been reported that higher trace element levels were found during the growth period of algae (Kaimoussi et al., 2004; Villares et al., 2002). Trace elements showed significant spatial variations in algae ( $p < 0.05$ ). For most elements, the highest levels were found in Tripoli. More particularly, levels of Al ( $779 \pm 611 \text{ mg.kg}^{-1}$ ), As ( $7.55 \pm 2.15 \text{ mg.kg}^{-1}$ ), Cr ( $1.09 \pm 0.13 \text{ mg.kg}^{-1}$ ) and Pb ( $1.42 \pm 0.23 \text{ mg.kg}^{-1}$ ) were significantly higher at this site. Lead (Pb) levels are close to the ones obtained in relatively polluted areas (Table 2) (Akcali and Kucuksezgin, 2011; Malea et al., 2015; Malea and Kevrekidis, 2014). These levels can be attributed to the leachate from the

dumpsite in Tripoli, as high levels of these elements were detected in the leachate (Khalil et al., 2018).

The kuruma shrimp showed seasonal variation for Ti, Co, As, and Se with higher levels during the wet season. For the rest of the elements, no seasonal changes were recorded, and values were almost the same during both seasons. *Penaeus japonicus* mainly spawns during the spring and summer months (May- July) at temperatures ranging from 24 to 28 °C in the Eastern Mediterranean (Pourang et al., 2004; Yilmaz and Yilmaz, 2007). In addition, the increase in body tissue may contribute to the lower levels of elements observed during the dry season. Significant spatial variation ( $p < 0.05$ ) was only observed for As, Co, Cd, Cu, Pb, Se and V. The shrimps showed less seasonal and spatial variations compared to the alga and oyster. This could be due to the fact that the shrimp lives further from the coastal zone and may be less subjected to the dynamics of the coastal area (Stehr et al., 2004). In fact, the kuruma shrimp was caught at an average depth of 40 m but is known to live down to 90 m depth (Quigley et al., 2013).

The spiny oyster was chosen for the multivariate analysis as it was available at all sites and during all seasons, thus, providing a clearer idea about the spatial variations related to each season. For each season, the oyster showed a different spatial accumulation pattern (Fig. 2). Elemental concentration explained 65% of spatial variability during the wet season (Fig. 2A) and 73 % during the dry season (Fig. 2B). Significant seasonal differences in the spiny oyster were found for Al, Ti, Fe, Zn and As ( $p < 0.05$ ) with higher levels found during the wet season. Similar findings with higher values during the winter season were found in the blue mussel *M. galloprovincialis* from the Moroccan coast (Azizi et al., 2018). Higher values recorded during the wet seasons are often associated with increased inputs of trace elements from land-based sources and their accumulation in the water due to increased rain (Zuluaga Rodríguez et al., 2015). Moreover, the spiny oyster undergoes sexual maturation and spawns in the period between June and August (Shabtay et al., 2015). Therefore, an increase in body tissues is probably contributing to the lower levels of elements observed during the dry season (Shabtay et al., 2015).

Concerning spatial variation, *S. spinosus* samples collected in Tripoli were characterized by higher levels for most trace elements during the wet season, while samples from Saida displayed highest concentrations during the dry season. This is mainly due to the differences in rainfall patterns and physico-chemical parameter variation of water between northern and the southern sites as explained above, and also to the sources involved in trace element input at each site.

More precisely, Abou Ali river drains effluents into the coastal area in Tripoli (Fig. 1). A wastewater treatment plant was established in the area but has not been working properly. In addition, a dumpsite in Tripoli is still receiving waste until present. This is leading to increased waste load and a difficulty to control leachate that is drained to the sea without treatment (El-Hoz, 2017). A study carried out on the leachate produced by dumpsites in Lebanon showed increased levels of trace elements of Mn, Cr, Ni and Cd due to lack of efficient waste management (Khalil et al., 2018). For most contaminants, concentrations in dumpsite leachate in Lebanon were higher than the toxicity reference values (TRVs) that are used for ecological risk assessment (Khalil et al., 2018). They reached 12 times the TRV for Pb and 27 folds for Cd. *Spondylus spinosus* showed relatively high levels of Cd ( $p < 0.05$ ) mainly recorded in Tripoli (Table 5) for both seasons ( $4.21 \pm 0.77$  and  $3.80 \pm 0.01$  mg.kg<sup>-1</sup> for the wet and dry season, respectively), with higher levels ( $p < 0.05$ ) for the wet season. High levels of Cd were also recorded by Nakhlé (2006) in mussel tissues sampled in Selaata (South of Tripoli), (0.6-8.7 and 1.08-4.03 mg.kg<sup>-1</sup> in *Patella* and *Brachidontes*, respectively, Table 2) and a decreasing trend was reported southward as the influence of a phosphogypsum plant southern of Tripoli (Fig. 1) decreased, highlighting the influence of the plant on Cd levels in the area. A study on the effects of the plant on the marine coastal zone in that area (Fakhri and Romano, 2005) has shown that wind regime along the Lebanese coast played a substantial role in spreading various contaminants over a wide area. In addition, the impact of that plant is likely to affect the neighboring northern marine environment especially with a dominant northern marine current along the Lebanese coast (Fakhri and Romano, 2011). Accumulation of Cu, Cr, Ni, Zn, Ti and Cd are more likely to be influenced by anthropogenic inputs due to the presence of industries



such as tanneries and electroplating industries, especially in Beirut, releasing those contaminants in the environment (Fakhri, 2012; Jaishankar et al., 2014; Oliveira, 2012) and studies conducted on the leachate are supportive of this hypothesis.

### ***Biota-sediment accumulation factor (BSAF)***

The concentrations of different elements in the sediment are presented in Table 1 and the calculated biota-sediment accumulation factors (BSAFs) are provided in Table 4. The BSAFs displayed in Table 4 represent the average of BSAFs calculated per species and site for the dry season.

For *U. lactuca* all the elements had a  $BSAF < 1$  suggesting no bioaccumulation from the sediment. The same has been observed in a study done along the Senegalese coast. However, they found a higher TF (Transfer Factor) from water to the green algae suggesting higher accumulation from water rather than from sediments (Diop et al., 2016). Several studies reported significant correlation between the levels of trace elements accumulated by *U. lactuca* and those in the water and sediments (Akcali and Kucuksezgin, 2011; Bonanno et al., 2019; Chakraborty et al., 2014) while other researchers found no significant correlation between trace element concentrations in algae and those in water and sediment (Kamala-Kannan et al., 2008; Malea and Haritonidis, 2000). Although the uptake by seaweed is mainly from solution, levels of trace elements in sediments reflect the degree of pollution to a certain extent in the water column, as concentrations in water are indirectly related to the sediment through processes of resuspension and interstitial diffusion of trace elements (Bonanno et al., 2019; Thomann et al., 1995).

The spiny oyster accumulated Cu, Zn and Cd with the highest BSAF recorded for Zn and Cd in Tripoli compared to the shrimp (Table 4). Bivalves are known to accumulate substantial amounts of trace elements from water (Zuykov et al., 2013) and are able to accumulate trace elements at higher concentrations than those in water and sediments (Swaleh et al., 2016). BSAF for Zn was the highest for the oyster compared to the alga and shrimp. Bivalves are high accumulators of Zn as

they absorb it readily from water because it is an essential trace element for their metabolism as is Cu for shrimps. In fact, Cu, Zn and Cd induce the metal binding protein, metallothionein; thus, they are metabolically active having a high assimilation efficiency and low depuration rates (Thomann et al., 1995). However, the levels of essential trace elements accumulated by marine organisms do not always reflect a metabolic requirement as marine invertebrates may often accumulate trace elements due to their high ambient level (Rainbow and Luoma, 2011).

The kuruma shrimp had BSAFs >1 for Cu and Zn with higher values for Cu (Table 4). Copper (Cu) is an essential element in shrimps as it is associated to hemocyanin. Crustaceans take up trace elements from food and water in proportion to the dissolved concentration of the element (Rainbow, 1995b). In some cases, an increase of uptake is reflected by an elevated concentration, such as, the case of Cd in decapod crustaceans. However even if the uptake rates of trace elements by crustaceans respond to changes in the total dissolved trace element concentration, not all contaminants are in an available form for uptake (Rainbow, 1995b). Concerning Hg, the BSAF was >1 only in Tripoli. Sediments in Tripoli had a higher level of Hg compared to Beirut and Saida (Table 1). The shrimp is known to live close to the bottom and thus may be more exposed to sediment-related pollutants than algae and bivalves (Pinedo-Hernández et al., 2015).

Finally, trace elements found in marine organisms may be the result of uptake from different sources and not only from sediment. Bioaccumulation is the result of several factors including bioavailability, multiple routes of exposure and metabolism that can widely vary among species (Jakimska et al., 2011). Each species uptakes trace elements differently; hence, it is important to consider different taxa and even tissues in the same organism when establishing monitoring programs and assessing bioaccumulation (Yüzereroğlu et al., 2010).

#### **4. Conclusion**

The present study provided levels for 20 trace elements in three marine species from the Lebanese coast in relation to spatio-temporal variation and biota-sediment transfer. Significant differences

were observed in trace element accumulation between species revealing the importance of using multiple bioindicator organisms to monitor coastal pollution by trace elements. Our results generally ranged in value between those previously recorded in relatively uncontaminated areas and those recorded in polluted ones. This suggested an overall moderate level of contamination in the studied sites with highest levels mostly found in Tripoli. This study reported for the first time trace element levels in the spiny oyster from the Lebanese coast. Seasonal and spatial variations were observed and reflected differently for each species, highlighting the importance of seasonality when it comes to monitoring programs. The sediment-biota transfer study showed limited transfer of trace elements between these environmental compartments. Ecological risks from trace elements are not simple to document, as marine organisms accumulate trace elements through different routes and responses may vary widely among species. Thus, understanding toxicity requires more than just total element accumulated in tissues especially when considering the complexity of environmental factors. However, bioindicators remain good integrative markers of chemical exposure to contaminants in a polluted ecosystem, which helps to identify research needs as a first step towards further investigation.

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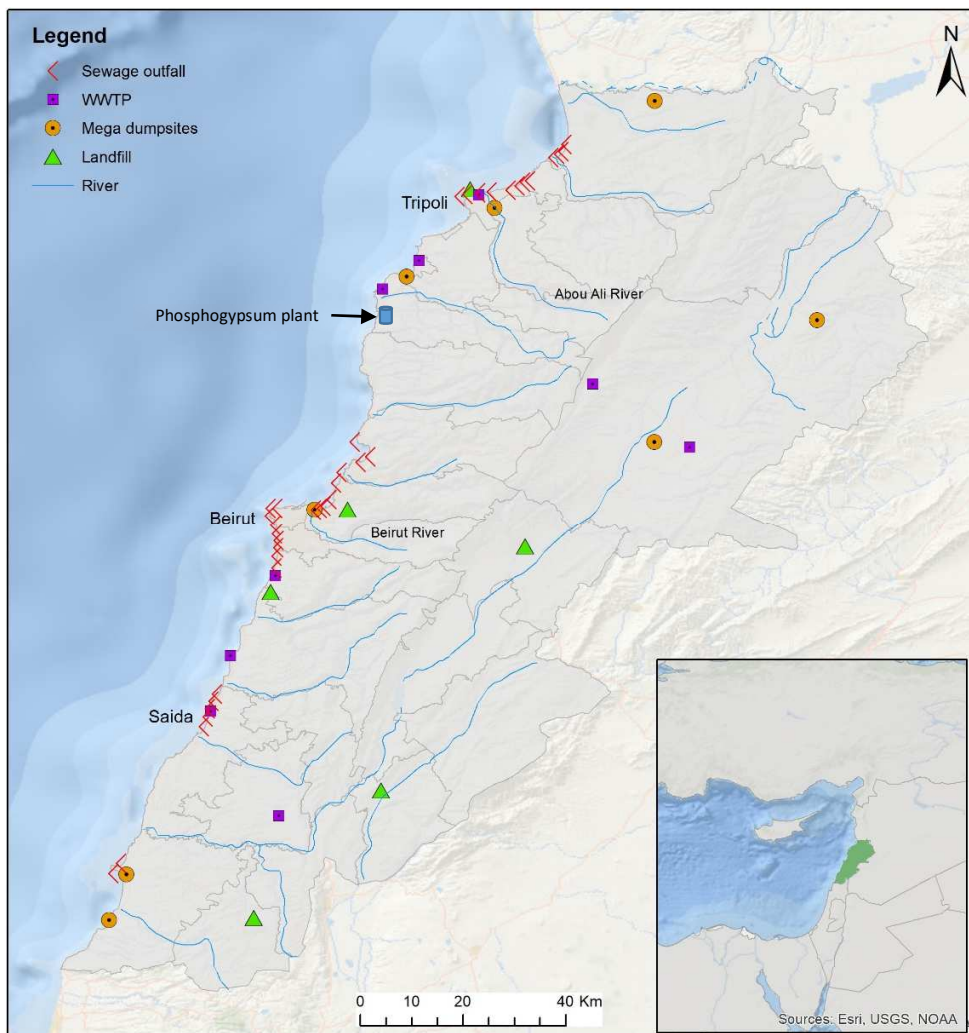
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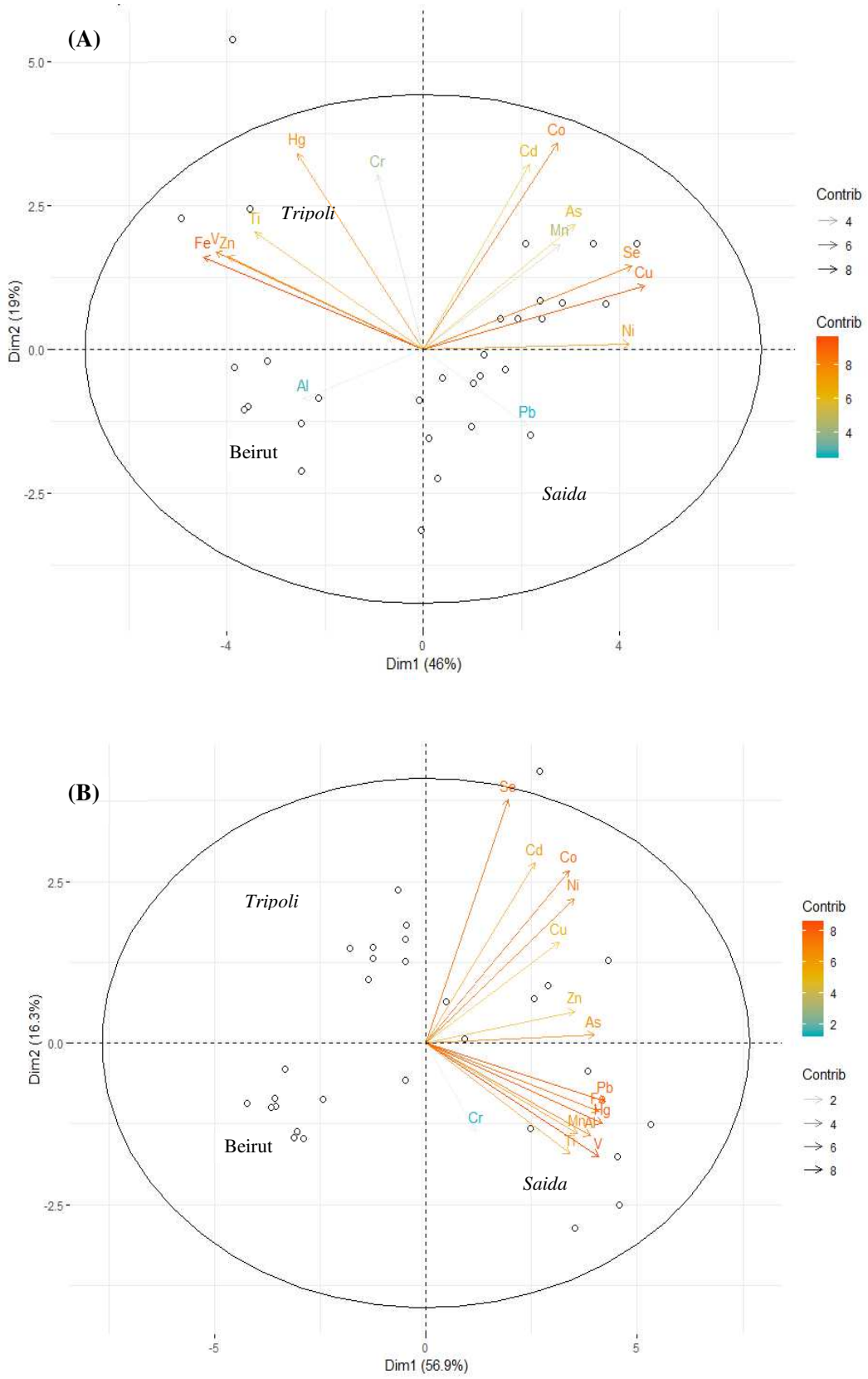
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**Figure 1** Map showing the sampling sites along the Lebanese coast (WWTP: waste water treatment plant)



**Figure 2** PCA showing the relation between sites and trace element concentrations in *S. spinosus* during the wet (A) and the dry (B) season

**Table 1** Mean concentration  $\pm$  standard deviation (SD) of trace elements (mg.kg<sup>-1</sup> dw) in sampled marine organisms and sediments. n: number of samples.

Species	n	Al	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Se	Cd	Hg	Pb
<i>U. lactuca</i>	40	378 $\pm$ 413	19.4 $\pm$ 12.7	3.53 $\pm$ 2.77	1.08 $\pm$ 0.90	29.4 $\pm$ 37.8	516 $\pm$ 473	0.34 $\pm$ 0.38	2.13 $\pm$ 0.63	2.30 $\pm$ 1.49	5.12 $\pm$ 2.29	4.78 $\pm$ 2.60	0.34 $\pm$ 0.15	0.05 $\pm$ 0.03	0.04 $\pm$ 0.02	1.04 $\pm$ 1.03
<i>S. spinosus</i>	60	11.1 $\pm$ 13.4	1.43 $\pm$ 1.54	0.21 $\pm$ 0.18	0.54 $\pm$ 0.14	4.51 $\pm$ 2.98	27.6 $\pm$ 24.5	0.86 $\pm$ 0.46	6.47 $\pm$ 3.27	8.50 $\pm$ 6.45	124 $\pm$ 162	35.2 $\pm$ 27.4	3.00 $\pm$ 0.94	3.00 $\pm$ 1.33	0.07 $\pm$ 0.04	0.89 $\pm$ 0.87
<i>M. japonicus</i>	30	2.75 $\pm$ 2.73	0.68 $\pm$ 0.69	0.05 $\pm$ 0.09	0.06 $\pm$ 0.04	0.64 $\pm$ 0.38	7.77 $\pm$ 7.06	0.05 $\pm$ 0.06	0.33 $\pm$ 0.40	30.9 $\pm$ 12.0	61.2 $\pm$ 9.8	28.7 $\pm$ 23.6	1.77 $\pm$ 0.49	0.02 $\pm$ 0.02	0.15 $\pm$ 0.15	0.04 $\pm$ 0.04
	1 (Tripoli)	3403	-	26.6	29.7	228	13287	5.19	12.0	6.29	21.5	-	-	0.32	0.120	17.3
Sediments <sup>a</sup>	1 (Beirut)	4305	-	38.2	22.9	228	22921	7.13	15.1	3.85	24.3	-	-	0.53	0.07	7.25
	1 (Saida)	2592	-	4.75	5.04	175	1550	0.97	3.58	2.02	4.99	-	-	0.13	0.07	3.35

<sup>a</sup> SD for sediment analysis is not available (n=1).

**Table 2** Comparison of trace element concentrations (mg.kg<sup>-1</sup> dw) measured in marine organisms from the Eastern Mediterranean (minimum and maximum levels in each group are marked in bold).

Species	Location	Al	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Se	Cd	Hg	Pb	Reference	
<i>U. lactuca</i>	Ras Beirut (Lebanon)	-	-	-	< 7.5- <b>16.3</b>	-	258- 595	-	9.4- 43.1	24.4- <b>52.5</b>	41.3- <b>206</b>	-	-	< 0.8- 2.3	-	< 7.5- <b>37.5</b>	(Shiber, 1980)	
	Tyrrhenian Sea (Central Italy)	-	-	-	1.39- 2.06	-	-	-	-	4.9- 6.4	-	-	-	<b>0.13</b> - 0.21	-	1.67- 2.28	(Conti and Cecchetti, 2003)	
	Eastern Harbour (Egypt)	-	-	-	-	73.9± 4.8	515± 2	7.08± 1.3	-	14.5± 4.7	63.1± 3.2	-	-	1.84± 0.93	-	-	(Abdallah and Abdallah, 2008)	
	El Mex Bay (Egypt)	-	-	-	-	33.3± 3.9	709± 13	6.58 ±1.4	-	7.24± 2.30	27.4± 8.3	-	-	0.73± 0.24	-	-	(Yaich et al., 2011)	
	Monastir (Tunisia)	-	-	-	-	<b>13</b>	410	-	-	8	68	-	-	-	-	12.6	(Yaich et al., 2011)	
<i>U. fasciata</i>	Abou Qir Bay (Egypt)	-	-	-	-	16.0	65.2	1.12	3.64	2.96	<b>12.6</b>	-	-	1.27	0.06	2.84	(Shams El-Din et al., 2014)	
<i>Ulva sp.</i>	Gulf of Thessaloniki (Greece)	-	-	-	<b>16.9 ±</b> <b>0.69</b>	14.4± 3.72	132 ± 48	-	1.7± 0.12	9.25± 2.88	11.3± 1.02	116± 4.3	<b>0.59±</b> 0.41	2.89± 1.21	0.04± 0.01	-	6.27± 1.79	(Malea and Kevrekidis, 2014)
	Gulf of Thessaloniki (Greece)	-	-	-	15.8± 1.09	13.8± 2.95	37.3 ± 7.08	-	0.84± 0.17	7.41± 0.69	9.15± 1.73	122± 10.3	1.50± 0.47	<b>4.83±</b> <b>1.09</b>	0.034± 0.004	-	4.62± 1.19	(Malea et al., 2015)
<i>U. rigida</i>	Gulf of Thessaloniki (Greece)	-	-	-	16.24± 1.19	9.38± 1.49	37.9 ± 10.0	-	0.56± 0.13	6.44± 0.55	7.39± 0.89	122± 14	1.45± 0.25	2.71± 0.77	<b>0.03±</b> <b>0.003</b>	-	3.06± 0.67	(Malea et al., 2015)
	Çanakkale/City (Turkey)	-	-	-	<b>0.78±</b> <b>0.20</b>	-	198± 19	-	-	-	8.51± 0.86	50.1± 6.0	-	-	54.2± 13.7	53.2± 3.17	5.65± 0.87	(Akcali and Kucuksezgin, 2011)
<i>Ulva sp.</i>	Izmir/Foça (Turkey)	-	-	-	1.75± 0.42	-	130± 26.9	-	-	13.9± 3.11	67.8 ± 16.0	-	-	14.9± 1.89	48.4± 11.8	1.75± 0.42		
	Izmir/Bostanlı (Turkey)	-	-	-	1.95± 0.51	-	232± 28.6	-	-	8.29± 0.21	40.4± 6.1	-	-	18.7± 2.27	59.5± 14.4	1.95± 0.51		
	Izmir/Narlıdere (Turkey)	-	-	-	4.22± 1.53	-	308± 74	-	-	8.21± 1.21	61.3± 6.5	-	-	22 ± 6.24	199± 62	4.22± 1.53		
	Izmir/Urla (Turkey)	-	-	-	0.94± 0.12	-	146± 34	-	-	7.78± 0.35	81.0± 0.47	-	-	24.9± 7.88	73.0± 12.9	<b>0.94±</b> <b>0.12</b>		
<i>U. lactuca</i>	Lebanese Coast	66.5- 1535	2.28- 43.8	<b>0.25</b> - 9.54	0.21- 3.74	1.49- 128	53.6- <b>1714</b>	0.03- 1.31	1.36- 3.99	<b>0.24</b> - 5.64	1.20- 8.89	1.10- <b>10.9</b>	<b>0.17</b> - 0.72	0.01- 0.10	0.03- 0.06	0.07- 4.44	Present study	
	Sicilian Coast (Italy)	-	-	-	0.19- 0.46	-	-	-	-	1.21- 2.35	3.5- 14.6	-	-	3.30- 6.30	-	0.14- 1.52	(Campanella et al., 2001)	
<i>Patella Caerulea</i>	Sicilian Coast (Italy)	-	-	-	0.10- 1.01	-	-	-	-	0.47- 3.79	<b>2.2</b> - 19.1	-	-	1.7- 11.8	-	0.06- 2.18	(Cubadda et al., 2001)	
	Tyrrhenian Sea (Italy)	-	-	-	0.72- 0.96	-	-	-	-	10.2- 19.2	-	-	-	2.89- 4.06	-	0.51- 1.50	(Conti and Cecchetti, 2003)	
	Lebanese Coast	-	-	-	-	-	-	-	-	-	-	-	-	0.6- 8.7	<b>0.14</b> - 0.2	0.50- 2.6	(Nakhlé et al., 2006)	
	Iskenderun (Turkey)	-	-	-	-	-	61.2- 213	0.15- 0.33	<b>0.53</b> - 1.60	1.38- 5.58	6.50- 13.71	-	-	<b>0.41</b> - 0.68	-	0.13- 0.70	(Yüzereroğlu et al., 2010)	

	Yumurtalik (Turkey)	-	-	-	-	-	36.6-169	<b>0.05</b> -0.14	0.39-1.05	1.09-2.12	3.70-9.84	-	-	0.24-0.44	-	<b>0.05</b> -0.31	
	Tyrrhenian Sea, Central Italy	-	-	-	0.46-1.31	-	-	-	-	5.51-11.50	-	-	-	0.32-0.49	-	1.67-2.49	(Conti and Cecchetti, 2003)
<i>Mytilus galloprovincialis</i>	Cala Iris Al Hoceima, (Morocco)	-	-	-	-	-	340-673	N.D.	2.03-3.82	5.28-6.16	160-164	-	-	0.65-0.85	-	N.D.	(Azizi et al., 2018)
	Izmir Bay (Turkey)	-	-	-	-	1.23-2.0	-	-	1.09-1.84	<b>0.71</b> -1.72	18.7-30.0	-	-	-	-	-	(Kontas, 2012)
<i>Monodonta turbinata</i>	Sicilian Coast (Italy)	-	-	-	<b>0.13</b> -0.54	-	-	-	-	5.7-21.5	14-55	-	-	0.58-3.35	-	0.15-0.66	(Campanella et al., 2001)
<i>Brachidontes variabilis</i>	Lebanese Coast	-	-	-	-	-	-	-	-	-	-	-	-	1.08-4.0	<b>0.2</b> - <b>0.6</b>	1.0-2.5	(Nakhlé et al., 2006)
<i>Donax trunculus</i>	Eastern Harbour (Egypt)	-	-	-	-	21.2±0.23	570±8	-	-	12.57±0.22	74.0±4.5	-	-	3.93±0.20	-	-	(Abdallah and Abdallah, 2008)
<i>S. spinosus</i>	Iskenderun Bay, (Turkey)	66.0-113	-	-	9.78- <b>18.1</b>	24.8- <b>30.6</b>	64.4-251	3.47- <b>9.85</b>	0.86-	14.5-20.4	64.8-82.8	-	-	4.96-15.1	-	59.9- <b>132</b>	(Türkmen and Türkmen, 2005)
<i>S. spinosus</i>	Lebanese Coast	0.77-85.1	0.33-7.99	0.04-0.67	0.30-0.93	<b>0.75</b> -14.76	<b>6.78</b> - <b>136</b>	0.27-2.36	0.86- <b>13.9</b>	1.06- <b>31.03</b>	42.9- <b>972</b>	9.52- <b>186</b>	1.42-5.25	0.54- <b>7.74</b>	0.03-0.15	0.12-3.54	Present study
<i>Penaeus kerathurus</i>	Izmir Bay, Turkey (Eastern Mediterranean)	-	-	-	-	-	4.45	-	-	6.14	14.62	-	-	<b>0.20</b>	-	0.96	(Turkmen, 2012)
<i>P. kerathurus</i>	Izmir Bay, Turkey	-	-	-	-	0.57±0.06	6.70±3.63	-	2.11 ±0.50	<b>4.75±1.29</b>	<b>10.8 ±4.88</b>	-	-	-	-	-	(Kontas, 2012)
<i>P. semisulcatus</i>	Mersin Bay	-	-	-	2.81-5.80	-	-	-	-	-	-	-	-	1.05-1.40	-	5.01-10.8	(Ayas, 2016)
<i>P. semisulcatus</i>	Iskenderun Bay	-	-	-	40.75- <b>73.3</b>	-	12.6-28.5	-	-	21.9-57.5	21.5-36.6	-	-	10.0- <b>25.8</b>	-	-	(Firat et al., 2008)
<i>P. semisulcatus</i>	Iskenderun Bay	-	-	-	-	-	49.7-62.2	-	-	30.1-31.2	70.2-83.6	-	-	1.0-1.9	-	5.7-8.5	(Kargin et al., 2001)
<i>P. japonicus</i>	Turkish coast (North Eastern Mediterranean)	-	-	-	0.97-1.30	-	52.3- <b>113</b>	-	2.19- <b>3.37</b>	15.71-22.6	18.0-24.7	-	-	0.64-0.82	-	3.70-5.40	(Canli et al., 2001)
<i>Parapenaeus longirostris</i>	Mersin Bay	-	-	-	1.38-3.44	-	-	-	-	-	-	-	-	0.44-0.67	-	1.99-4.40	
<i>Metapenaeus monoceros</i>	Mersin Bay	-	-	-	2.38-4.38	-	-	-	-	-	-	-	-	0.55-0.97	-	7.29- <b>13.4</b>	(Ayas, 2016)
<i>M. japonicus</i>	Mersin Bay	-	-	-	2.40-5.37	-	-	-	-	-	-	-	-	0.80-1.10	-	5.55-8.30	
<i>M. japonicus</i>	Lebanese Coast	0.70-10.5	0.20-3.54	0.01-0.56	<b>0.02</b> -0.17	<b>0.34</b> - <b>2.57</b>	<b>2.95</b> -37.5	0.01-0.38	<b>0.11</b> -1.15	14.20- <b>80.4</b>	51.39- <b>116</b>	5.98-97.4	1.13-3.83	0.00-0.10	0.03-0.79	<b>0.01</b> -0.21	Present study

**Table 3** Mean concentration  $\pm$  standard deviation (mg.kg<sup>-1</sup> dw) of trace elements measured in marine organisms sampled during the wet (S1) and dry (S2) seasons.

Species	Season	Al	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Se	Cd	Hg	Pb
<i>U. lactuca</i>	S1	245 $\pm$ 214*	18.4 $\pm$ 14.1	3.05 $\pm$ 2.98*	0.63 $\pm$ 0.36*	8.66 $\pm$ 4.92*	359 $\pm$ 274*	0.14 $\pm$ 0.07*	2.35 $\pm$ 0.65*	2.47 $\pm$ 1.72	5.18 $\pm$ 2.65	3.86 $\pm$ 2.09*	0.27 $\pm$ 0.11*	0.06 $\pm$ 0.03	N.D	0.65 $\pm$ 0.60*
	S2	779 $\pm$ 610	22.3 $\pm$ 8.16	4.98 $\pm$ 1.51	2.43 $\pm$ 0.71	91.7 $\pm$ 23.2	2443 $\pm$ 1216	0.95 $\pm$ 0.24	1.72 $\pm$ 0.36	1.82 $\pm$ 0.25	4.92 $\pm$ 0.80	7.55 $\pm$ 2.15	0.52 $\pm$ 0.09	0.04 $\pm$ 0.01	0.04 $\pm$ 0.02	6.87 $\pm$ 12.2
<i>S. spinosus</i>	S1	14.9 $\pm$ 16.4*	2.00 $\pm$ 1.82*	0.23 $\pm$ 0.18	0.54 $\pm$ 0.17	4.72 $\pm$ 2.86	6.98 $\pm$ 5.44*	0.95 $\pm$ 0.48	5.59 $\pm$ 3.16	7.56 $\pm$ 6.75	186.80 $\pm$ 212*	40.98 $\pm$ 32.55*	3.19 $\pm$ 1.03	3.2 $\pm$ 1.51	0.14 $\pm$ 0.02	0.83 $\pm$ 0.72
	S2	7.94 $\pm$ 8.27	1.00 $\pm$ 1.06	0.21 $\pm$ 0.18	0.55 $\pm$ 0.11	4.51 $\pm$ 3.14	19.73 $\pm$ 10.37	0.77 $\pm$ 0.44	7.31 $\pm$ 3.19	9.45 $\pm$ 6.1	62.98 $\pm$ 20.9	31.00 $\pm$ 20.0	2.75 $\pm$ 0.80	2.74 $\pm$ 1.10	0.06 $\pm$ 0.03	1.02 $\pm$ 1.00
<i>M. japonicus</i>	S1	2.93 $\pm$ 2.85	1.27 $\pm$ 1.14*	0.08 $\pm$ 0.12	0.15 $\pm$ 0.03	0.68 $\pm$ 0.49	6.98 $\pm$ 5.44	0.08 $\pm$ 0.08*	N.D	32.72 $\pm$ 14.6	63.4 $\pm$ 12.73	36.0 $\pm$ 25.0*	1.92 $\pm$ 0.54*	0.03 $\pm$ 0.03	0.12 $\pm$ 0.05	0.03 $\pm$ 0.01
	S2	2.59 $\pm$ 9.31	0.48 $\pm$ 0.76	0.02 $\pm$ 0.03	0.05 $\pm$ 0.02	0.60 $\pm$ 0.41	8.55 $\pm$ 10.03	0.03 $\pm$ 0.02	0.17 $\pm$ 0.04	29.1 $\pm$ 9.39	58.9 $\pm$ 6.02	21.43 $\pm$ 17.84	1.61 $\pm$ 0.35	0.01 $\pm$ 0.09	0.17 $\pm$ 1.20	0.05 $\pm$ 0.15

<sup>a</sup> the symbol \*indicates significant differences between seasons p < 0.05

**Table 4** Biota-sediment transfer factors (BSAFs) in the sampled organisms along the Lebanese coast. Values in bold indicate a transfer of contaminants from sediment to biota (BSAFs > 1).

Species	Site	Al	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Cd	Hg	Pb
<i>U. lactuca</i>	Tripoli	0.229	0.081	0.082	0.402	0.184	0.183	0.143	0.430	0.228	0.113	0.072	0.397
	Beirut	0.041	0.015	0.011	0.011	0.009	0.009	0.000	0.067	0.086	0.033	0.000	0.018
	Saida	0.044	0.264	0.016	0.043	0.019	0.034	0.231	0.680	0.304	0.266	0.000	0.012
<i>S. spinosus</i>	Tripoli	0.001	0.003	0.017	0.012	0.001	0.182	0.695	<b>1.459</b>	<b>2.844</b>	<b>11.779</b>	0.399	0.032
	Beirut	0.001	0.003	0.019	0.011	0.001	0.075	0.329	0.974	<b>18.653</b>	<b>4.892</b>	0.316	0.015
	Saida	0.005	0.016	0.019	0.033	0.002	0.187	0.810	<b>2.064</b>	<b>3.540</b>	<b>9.222</b>	0.817	0.127
<i>M. japonicus</i>	Tripoli	0.001	0.001	0.001	0.002	0.000	0.007	0.004	<b>5.590</b>	<b>2.702</b>	0.071	<b>1.035</b>	0.005
	Beirut	0.001	0.001	0.002	0.003	0.000	0.005	0.005	<b>3.662</b>	<b>2.772</b>	0.011	0.507	0.001
	Saida	0.000	0.000	0.000	0.000	0.000	0.000	0.000	<b>6.496</b>	<b>3.044</b>	0.017	0.666	0.000



**Table 5** Mean concentration  $\pm$  SD of trace elements ( $\text{mg.kg}^{-1}$  dw) measured in marine organisms collected during the wet (S1) and dry (S2) seasons. Letters indicate statistically significant differences between sites per row (<sup>abc</sup> S1, <sup>def</sup> S2). Absence of letters indicates no significant differences between sites.

Analyte	Species	Tripoli		Beirut		Saida	
		S1	S2	S1	S2	S1	S2
Al	<i>U. lactuca</i>	444 $\pm$ 271 <sup>a</sup>	779 $\pm$ 611	140 $\pm$ 43 <sup>b</sup>	-	149 $\pm$ 71 <sup>b</sup>	-
	<i>S. spinosus</i>	7.38 $\pm$ 5.34 <sup>a</sup>	4.27 $\pm$ 2.83 <sup>d</sup>	28.1 $\pm$ 24.2 <sup>b</sup>	2.49 $\pm$ 1.50 <sup>d</sup>	10.8 $\pm$ 6.1 <sup>ab</sup>	15.5 $\pm$ 10.0
	<i>M. japonicus</i>	2.46 $\pm$ 2.17	2.35 $\pm$ 3.08	-	2.83 $\pm$ 1.50	3.36 $\pm$ 3.43	-
Ti	<i>U. Lactuca</i>	36.7 $\pm$ 5.3 <sup>a</sup>	22.3 $\pm$ 8.2	6.28 $\pm$ 3.15 <sup>b</sup>	-	12.1 $\pm$ 4.6 <sup>b</sup>	-
	<i>S. spinosus</i>	1.09 $\pm$ 0.36 <sup>a</sup>	0.64 $\pm$ 0.32 <sup>d</sup>	3.36 $\pm$ 2.53 <sup>b</sup>	0.45 $\pm$ 0.09 <sup>d</sup>	1.35 $\pm$ 0.42 <sup>ab</sup>	1.77 $\pm$ 1.57 <sup>e</sup>
	<i>M. japonicus</i>	2.00 $\pm$ 2.17	0.55 $\pm$ 0.32	-	0.40 $\pm$ 0.22	0.97 $\pm$ 0.62	-
V	<i>U. Lactuca</i>	1.72 $\pm$ 0.12 <sup>a</sup>	4.98 $\pm$ 1.51	0.39 $\pm$ 0.12 <sup>b</sup>	-	7.03 $\pm$ 1.11 <sup>c</sup>	-
	<i>S. spinosus</i>	0.13 $\pm$ 0.03 <sup>a</sup>	0.09 $\pm$ 0.02 <sup>d</sup>	0.45 $\pm$ 0.14 <sup>b</sup>	0.07 $\pm$ 0.02 <sup>d</sup>	0.11 $\pm$ 0.03 <sup>a</sup>	0.43 $\pm$ 0.14 <sup>e</sup>
	<i>M. japonicus</i>	0.13 $\pm$ 0.16 <sup>a</sup>	0.02 $\pm$ 0.01	-	0.02 $\pm$ 0.01	0.02 $\pm$ 0.02 <sup>b</sup>	-
Cr	<i>U. Lactuca</i>	1.09 $\pm$ 0.13 <sup>a</sup>	2.43 $\pm$ 0.71	0.34 $\pm$ 0.11 <sup>b</sup>	-	0.46 $\pm$ 0.18 <sup>b</sup>	-
	<i>S. spinosus</i>	0.54 $\pm$ 0.11	0.50 $\pm$ 0.09	0.58 $\pm$ 0.24	0.55 $\pm$ 0.09	0.49 $\pm$ 0.11	0.59 $\pm$ 0.14
	<i>M. japonicus</i>	N.D.	0.04 $\pm$ 0.01	-	0.06 $\pm$ 0.03	0.15 $\pm$ 0.03	-
Mn	<i>U. Lactuca</i>	13.6 $\pm$ 1.29 <sup>a</sup>	91.7 $\pm$ 23.2	2.58 $\pm$ 0.82 <sup>b</sup>	-	9.82 $\pm$ 2.47 <sup>a</sup>	-
	<i>S. spinosus</i>	6.74 $\pm$ 3.75 <sup>ab</sup>	2.76 $\pm$ 1.09 <sup>d</sup>	3.20 $\pm$ 1.16 <sup>a</sup>	2.54 $\pm$ 1.35 <sup>d</sup>	4.22 $\pm$ 1.88 <sup>b</sup>	7.62 $\pm$ 3.24 <sup>e</sup>
	<i>M. japonicus</i>	0.68 $\pm$ 0.67	0.46 $\pm$ 0.09 <sup>d</sup>	-	0.74 $\pm$ 0.24 <sup>e</sup>	0.67 $\pm$ 0.24	-
Fe	<i>U. Lactuca</i>	701 $\pm$ 132 <sup>a</sup>	2443 $\pm$ 1217	123 $\pm$ 67 <sup>b</sup>	-	254 $\pm$ 126 <sup>b</sup>	-
	<i>S. spinosus</i>	16.4 $\pm$ 4.74 <sup>a</sup>	15.61 $\pm$ 2.92 <sup>d</sup>	69.6 $\pm$ 33.8 <sup>b</sup>	11.1 $\pm$ 2.56 <sup>d</sup>	22.2 $\pm$ 7.3 <sup>ab</sup>	30.5 $\pm$ 9.8 <sup>e</sup>
	<i>M. japonicus</i>	5.50 $\pm$ 2.49	5.42 $\pm$ 5.92	-	11.7 $\pm$ 10.9	8.47 $\pm$ 7.17	-
Co	<i>U. Lactuca</i>	0.19 $\pm$ 0.02 <sup>a</sup>	0.95 $\pm$ 0.24	0.37 $\pm$ 0.02 <sup>b</sup>	-	0.22 $\pm$ 0.04 <sup>a</sup>	-
	<i>S. spinosus</i>	1.45 $\pm$ 0.35 <sup>a</sup>	0.95 $\pm$ 0.51 <sup>d</sup>	0.76 $\pm$ 0.37 <sup>b</sup>	0.39 $\pm$ 0.18 <sup>e</sup>	0.64 $\pm$ 0.22 <sup>b</sup>	0.97 $\pm$ 0.30 <sup>d</sup>
	<i>M. japonicus</i>	0.11 $\pm$ 0.10 <sup>a</sup>	0.04 $\pm$ 0.02	-	0.02 $\pm$ 0.01	0.05 $\pm$ 0.02 <sup>b</sup>	-
Ni	<i>U. Lactuca</i>	1.87 $\pm$ 0.14 <sup>a</sup>	1.72 $\pm$ 0.36	N.D.	-	2.78 $\pm$ 0.63 <sup>b</sup>	-
	<i>S. spinosus</i>	6.81 $\pm$ 1.41 <sup>a</sup>	8.36 $\pm$ 2.17 <sup>d</sup>	1.64 $\pm$ 0.79 <sup>b</sup>	3.96 $\pm$ 1.63 <sup>e</sup>	8.32 $\pm$ 1.56 <sup>a</sup>	9.75 $\pm$ 2.46 <sup>d</sup>
	<i>M. japonicus</i>	N.D.	0.15 $\pm$ 0.04	-	0.18 $\pm$ 0.06	N.D.	-
Cu	<i>U. Lactuca</i>	2.71 $\pm$ 0.35 <sup>a</sup>	1.82 $\pm$ 0.25	0.42 $\pm$ 0.13 <sup>b</sup>	-	4.28 $\pm$ 0.99 <sup>a</sup>	-
	<i>S. spinosus</i>	15.3 $\pm$ 5.5 <sup>a</sup>	9.18 $\pm$ 5.37 <sup>de</sup>	1.63 $\pm$ 0.57 <sup>b</sup>	6.13 $\pm$ 4.39 <sup>d</sup>	5.77 $\pm$ 2.72 <sup>a</sup>	13.0 $\pm$ 6.9 <sup>e</sup>
	<i>M. japonicus</i>	24.6 $\pm$ 6.2 <sup>a</sup>	35.2 $\pm$ 7.8 <sup>d</sup>	-	23.0 $\pm$ 3.5 <sup>e</sup>	40.9 $\pm$ 16.3 <sup>b</sup>	-

Zn	<i>U. Lactuca</i>	7.16±0.87 <sup>a</sup>	4.92± 0.80	1.85±1.36 <sup>b</sup>	-	6.55±1.11 <sup>a</sup>	-
	<i>S. spinosus</i>	74.9±10.6 <sup>a</sup>	61.19±13.47 <sup>de</sup>	401±261 <sup>b</sup>	50.3±9.2 <sup>d</sup>	84.2±20.0	76.2±28 <sup>e</sup>
	<i>M. japonicus</i>	61.3±2.5	58.14±3.86	-	59.6±6.1	65.5±18.1	-
As	<i>U. Lactuca</i>	4.14± 0.33 <sup>a</sup>	7.55±2.15	1.29±0.14 <sup>b</sup>	-	6.14±0.84 <sup>c</sup>	-
	<i>S. spinosus</i>	66.3±46.2 <sup>a</sup>	20.79±11.34 <sup>d</sup>	26.3±11.6 <sup>b</sup>	15.0±4.2 <sup>d</sup>	30.33±7.9 <sup>b</sup>	52.3±15.7 <sup>e</sup>
	<i>M. japonicus</i>	43.3±32.0	33.72±22.50 <sup>d</sup>	-	9.13±2.71	28.8±13.4 <sup>e</sup>	-
Se	<i>U. Lactuca</i>	0.21±0.06 <sup>a</sup>	0.52±0.09	0.27±0.04 <sup>ac</sup>	-	0.36±0.15 <sup>c</sup>	-
	<i>S. spinosus</i>	4.49±0.41 <sup>a</sup>	3.51±0.74 <sup>d</sup>	2.21±0.27 <sup>b</sup>	2.05±0.47 <sup>c</sup>	2.88 ± 0.37 <sup>b</sup>	2.85±0.45 <sup>d</sup>
	<i>M. japonicus</i>	1.93±0.34	1.88±0.34 <sup>d</sup>	-	1.34±0.17	1.90 ± 0.71 <sup>e</sup>	-
Cd	<i>U. Lactuca</i>	0.07 ±0.00 <sup>a</sup>	0.04 ± 0.01	0.02±0.00 <sup>b</sup>	-	0.09±0.01 <sup>a</sup>	-
	<i>S. spinosus</i>	4.21±0.77 <sup>a</sup>	3.80±0.01 <sup>d</sup>	2.66±2.16 <sup>b</sup>	1.58±0.86 <sup>e</sup>	2.76±0.66 <sup>b</sup>	2.97±0.68 <sup>d</sup>
	<i>M. japonicus</i>	0.04±0.03 <sup>a</sup>	0.02±0.01 <sup>d</sup>	-	0.004±0.003 <sup>b</sup>	0.01±0.01 <sup>e</sup>	-
Hg	<i>U. Lactuca</i>	N.D.	0.04±0.02	N.D.	-	N.D.	-
	<i>S. spinosus</i>	N.D.	0.048±0.01 <sup>d</sup>	0.14±0.02	0.038±0.01 <sup>d</sup>	N.D.	0.09±0.02 <sup>e</sup>
	<i>M. japonicus</i>	N.D.	0.28±0.23	-	0.06±0.02	N.D.	-
Pb	<i>U. Lactuca</i>	1.42±0.23 <sup>a</sup>	2.52±1.08	0.32±0.30 <sup>b</sup>	-	0.20±0.07 <sup>b</sup>	-
	<i>S. spinosus</i>	0.56±0.23 <sup>a</sup>	0.39±0.13 <sup>d</sup>	0.27±0.08 <sup>a</sup>	0.26±0.19 <sup>d</sup>	1.65±0.64 <sup>b</sup>	2.21±0.73 <sup>e</sup>
	<i>M. japonicus</i>	0.02±0.00	0.09±0.05 <sup>d</sup>	-	0.01±0.01	0.03±0.01 <sup>e</sup>	-