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# Microplastics pollution along the Lebanese coast (Eastern Mediterranean Basin):

## Occurrence in surface water, sediments and biota samples

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The Mediterranean Sea is the largest semi-enclosed sea and one of the worst affected regional seas with sub-basin scale heterogeneity in plastics concentration. Few studies on microplastics (MPs) pollution have been conducted in the Eastern part of the Mediterranean basin. This study aims to evaluate, for the first time, the MPs pollution of the Lebanese coast (Levantine Basin) as well as the most common polymers found, and to assess the potential role of coastal landfills in this pollution. Two important seafood species that are wholly consumed by the Lebanese community: the European anchovy *Engraulis encrasicolus* and the spiny oysters *Spondylus spinosus* were sampled in three different sites englobing the littoral (Tripoli, Beirut and Sidon). Sea water and sediment samples were also collected from the same sites. Results showed different patterns of MPs concentration in the analyzed matrices: Sidon water sample was highly contaminated in MPs (6.7 MPs/m<sup>3</sup>) whilst Tripoli had the highest MPs in sediments (4.68 MPs/g). The occurrence of MPs in the biota was high (83.4% and 86.3% in anchovies and spiny oysters, respectively). Both anchovies and oysters from Beirut region had the highest ingested MPs/ individual ( $2.9 \pm 1.9$  and  $8.3 \pm 4.4$  MPs/individual, respectively). This study is the first that investigated microplastics ingestion by *Spondylus spinosus* while indicating the most common polymers found in the three matrices (water, sediments and biota) in the Eastern Mediterranean. These results highlighted the high MPs pollution found in the Levantine Basin in comparison to other Western Mediterranean regions. In addition, the obtained results indicate a potential contribution of coastal landfills to this pollution.

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## **1. Introduction**

The world plastic production is continuously increasing, reaching 350 million tons in 2017 (PlasticsEurope, 2018). Among the produced plastics, only 6–26% are recycled. Landfills receive 21–42% of wastes, whereas the rest are wildly released into the environment due to mismanagement through a variety of pathways (Ellen MacArthur Foundation, 2016; O'Connor et al., 2016). Most of the manufactured plastics will eventually end up in the ocean. By the year 2025, about 250 million tons of wastes are expected to enter the ocean water (Jambeck et al., 2015) leading to their accumulation in the marine environment (Thompson et al., 2009; UNEP, 2014). Plastic pollution in the marine environment is becoming a problem of global concern. These plastics are capable of transforming into dangerous small particles known as microplastics (MPs) existing as particles of a size below 5 mm ubiquitously available in the aquatic environments (Thompson et al., 2004; Andrady, 2011). They are either intentionally produced in small sizes such as polystyrene beads for example, or they are the resultant of macroplastics degradation (whether by Photochemical reaction, or mechanical actions.).

The Mediterranean Sea is prone to several anthropic pressures and has recently been described as one of the most affected marine environments with regards to marine litter and microplastics (Lebreton et al., 2012; Cincinelli et al., 2019). Mainly owing to the semi-enclosed situation and limited outflow of surface waters, a densely populated coastline and intensive fishing, shipping, touristic and industrial activities, substantial amounts of marine litter are accumulating in the Mediterranean Basin. Also, most studies on microplastics have been conducted in the northwestern Mediterranean Sea (e.g. Collignon et al., 2014; Fossi et al., 2016; Ruiz-Orejón et al., 2016; Schmidt et al., 2018; Lefebvre et al., 2019). Results showed that their distribution and composition are not homogeneous and geographical differences exist between Mediterranean sub-basins (Suaria et al., 2016; Cincinelli et al., 2019). Such heterogenicity in MPs distribution

and of its concentration levels are likely reflecting a complex interplay between pollution sources, hydrodynamic features such as currents, up and down-welling, gyres, fronts and the shape of the coastline (Suaria et al., 2016; Schmidt et al., 2018; Cincinelli et al., 2019). In addition, the absence of a standardized sampling protocol for the identification and quantification levels of MPs could also be responsible for this heterogeneity in results.

Plastics in general, and microplastics specifically, are prone to be ingested by a wide range of organisms (Lusher et al., 2016) and about one hundred species in the Mediterranean have been affected either via ingestion or entanglement (Deudero and Alomar, 2015). Microplastics were already found in several Mediterranean fish species such as *Boops boops*, *Sardina pilchardus*, *Mullus barbatus* and *Merluccius merluccius* (Giani et al., 2019; Bellas et al., 2016; Rios-Fuster et al., 2019) and in wild mussels (*Mytilus galloprovincialis*) caught off the Adriatic Sea and central Tyrrhenian Sea (Gomiero et al., 2019; Renzi et al., 2018a).

Although it is suggested that it is the most polluted region, few studies have been conducted in the Eastern part of the Mediterranean Basin (Van der Hal et al., 2017; Güven et al., 2017; Gündoğdu and Çevik, 2017). In the Levantine basin, the Lebanese coast is highly affected by numerous anthropogenic pressures such as harbor activities, fishing, domestic and industrial wastewater discharges (generally without prior treatment). Waste production in Lebanon has increased significantly during the last decade and the country has experienced waste crises. The regulated dumping sites could no longer manage the tremendous quantity of municipal garbage generated and, therefore, numerous unregulated and open dumping sites were created throughout the country mainly along the coast (Khalil et al., 2018). The average composition of municipal solid waste generated shows an important percentage of plastics (15%) which is two to three times higher than that found in Sweden, France and the United States (El-Hoz, 2019). Though the processes may be slow, landfills are affected by UV radiation and mechanical abrasions leading to the fragmentation of plastics to microplastics, and small plastic debris would find their way into the aquatic environment via leachates discharges (Alimi et al., 2018).

Despite these concerns, there have been no studies specifically examining the occurrence and abundance of microplastics along the Lebanese coastal environment. The aim of this study was to investigate the quantity and types of microplastics found in three matrices (sea water,

sediments and marine biota) along the Lebanese coast. All suspected MPs were characterized by size, color and identified according to their polymer nature using micro-Raman spectroscopy. Fish and bivalves are known to ingest microplastics (Collard et al., 2015; Phuong et al., 2018; Kazour et al., 2019), making them a potential vector of toxic chemicals through food chains and into human diets. In the present study we analyzed two marine species that are wholly consumed by the Lebanese community: one pelagic fish species: the European anchovy, *engraulis encrasicolus* (Linnaeus, 1758) and a bivalve species, the spiny oyster, *Spondylus spinosus* (Schreibers, 1793).

## **2. Materials and Methods**

### **2.1. Study and sampling area**

Lebanon is located in the Eastern Mediterranean (Levantine Basin) with a coastal length of 220 km (Figure 1). Three sampling sites were chosen along the coast according to their degree of anthropogenic pressures making them pollution hotspots (Shaban, 2008). The first site, Tripoli, located in the north, is facing the town's public beaches, characterized by the presence of a fishing harbor, and the existence of a coastal landfill. This latter was constructed in the year 1999 and reached today 36 m height receiving about 400 tons of wastes/day. The second site Beirut, located in the middle, is the country's capital. This site is near the most important harbor in Lebanon and surrounded by three coastal landfills: One closed and two active landfills that reached their maximum capacities (55 meters of height) in July 2019. The third site Sidon, located in the south, is facing a river's mouth and is near a closed landfill operated between 1982 and 2013. With a height of 58 m, this landfill was the biggest in the country receiving 300 tons of wastes/day. Actually, its transformation to a public park led to the burying of all wastes underground; traces of these wastes can always be observed underwater. These landfills lack an appropriate leachates treatment or it is not operational, and leachates are directly dumped to the sea. The sampling for all three matrices was done in May 2018 (spring).

### **2.2. Contamination control**

Several precautions were followed to minimize air-born contamination during sampling, samples preparation and analysis. Cotton lab coats were worn and different procedures (filtration, measurements and digestion) were done under a laminar flow hood. All surfaces and equipment were rinsed before usage with filtered ethanol 70% and distilled water. Laboratory supplies were made of glass (bottles, Petri dishes, Erlenmeyers, filtration system) and they were covered immediately after manipulation. All solutions (distilled water, Ethanol 70%, potassium hydroxide KOH 10% and zinc chloride  $\text{ZnCl}_2$ ) were filtered 3 times on glass fibers filter (GF/A filters, 1.6  $\mu\text{m}$  porosity, Whatman, France) to eliminate unwanted particles or fibers. Controls were made to follow up the contamination occurring with every step of preparation and observation.

### **2.3. Water and sediments sampling and analyses**

Sea surface water and sediments samples were collected during spring 2018. Sea surface water was collected using a manta trawl net (52  $\mu\text{m}$  mesh size) with an opening of 0.63 m and a length of 0.255 m equipped with a flowmeter. The net was trawled for 10 minutes parallel to the coast at about 500 m from the coast with a small artisanal vessel (3 m of length) at a speed of 2 knots. On average, 100  $\text{m}^3$  of sea water was sampled at each site (Supplementary Table 1). The net was washed with filtered distilled water and all collected residues were rinsed inside clean glass bottles. The collected water was filtered on a stainless-steel sieve of a mesh size of 850  $\mu\text{m}$  and the recovered water was vacuum filtered on GF/A filters. The suspected MPs present on the sieve were sorted visually under the stereomicroscope, fixed on a microscopic slide and conserved inside a clean glass Petri dish.

The first two centimeters of sublittoral sediments were sampled using a steel ring with two stainless-steel plates, rinsed with filtered distilled water before usage. The samples were put inside clean glass jars and stored at  $-4^\circ\text{C}$ . Samples were thawed at room temperature 12 hours prior to analysis. Then particles with a size range of 1 – 5 mm were visually sorted and the remaining sediments underwent a density separation using filtered  $\text{ZnCl}_2$  ( $d = 1.8 \text{ g/cm}^3$ ) (Imhof et al., 2013; Nuelle et al., 2014). Briefly, 50 mL of filtered  $\text{ZnCl}_2$  was added to the sample; the solution was then shaken for 5 minutes twice (once every 4 hours) and let rest for 16 hours. The

supernatant was then recovered and filtered on GF/A filters. Microplastics found were reported per gram of sediments sample and per m<sup>3</sup> in sea water samples.

#### **2.4. Biota sampling and analyses**

Anchovies and spiny oysters were also collected during spring 2018. Anchovies were caught using purse seine nets. Ten individuals of approximately the same size (9 cm ± 0.9) were chosen from each sampling site and were immediately individually conserved in aluminum foil and conserved at -4°C. In the laboratory, their morphometric characteristics (total length and weight, digestive tube weight) were measured and their digestive tracts were dissected, conserved in aluminum foil and frozen at -4°C. For the spiny oysters, ten individuals were collected by fishermen from each sampling site. In the laboratory, the whole individual was removed from its shell and was conserved in aluminum foil. The net weight of the animal ranged between 4.8 g to 38.2 g with an average of 20.2 g ± 9.4.

All samples were defrosted 12 hours prior to digestion. Fish digestive tracts and oyster soft tissues were digested using potassium hydroxide 10% KOH (Dehaut et al., 2016). In short, the organic material of each individual was put inside separate Erlenmeyer containing solution of KOH 10% and then they were incubated at 60°C for 24 hours. Then, the digestat was recovered and vacuum filtered on GF/A filters and put inside clean glass Petri dishes.

#### **2.5. Stereomicroscope and micro-Raman spectroscopy**

All samples were first observed under a Leica M165 C Stereomicroscope with a 120x magnification. Photos of items were captured using a Leica M170 HD camera connected to LAS (Leica Application Suite) software. They were listed, measured, and their morphology, color and shape were noted. Several criteria were taken into consideration while counting the suspected items: (1) absence of cellular or organic structures; (2) a homogenous thickness across the particles; and, (3) homogenous colors (Hidalgo-Ruz et al. 2012). Suspected microplastics were divided into 6 size classes: <200 µm, 200-400 µm, 400-600 µm, 600-800 µm, 800-1000µm, and > 1000 µm.

Suspected microplastics were analyzed using Micro-Raman Xplora Plus (HORIBA Scientific® France). This machine is equipped with a closing chamber that isolates the filter from its surrounding, hence minimizing any airborne contamination.

Due to the high amount of time it takes when analyzing filters under micro-Raman, five filters of anchovies and five filters of spiny oysters from each site containing the highest number of potential MPs were identified. Subsamples of sediments and water were analyzed in a way that 25% of particles were analyzed. For MPs polymers identification, two lasers were used with a wavelength of 532 nm and 785 nm with a range of 200-3400 cm<sup>-1</sup>. Two objectives (Olympus, Rungis, France) were used: x10 and x100. Filters were either analyzed manually or using ParticleFinder module for LabSpec 6. Each particle spectrum is compared to a database polymer identification software (KnowItAll, BioRad®) and a personal library made with specific polymers obtained from Goodfellow (France).

## **2.6. Data and Statistical analysis**

Anchovy Fulton's K condition index was calculated as an indicator of the fish general well-being:  $K = (\text{Body weight} / \text{total length}^3) * 100$ . The feeding intensity for the fish was calculated using the Gastro Somatic Index (GaSI) (Desai 1970):  $\text{GaSI} = \text{stomach weight} / (\text{body weight} - \text{stomach weight})$ . The mean ( $\pm$ SD) abundances of microplastics were calculated for anchovies (MPs/ind) and spiny oysters (MPs/g). After a Shapiro-Wilk test, data distribution of ingested MPs were non normal and a non-parametric tests (Kruskal-Wallis and Mann-Whitney U tests) were used to determine differences in microplastic numbers among species and sites. GaSI values followed a normal distribution and a one way analysis of variance (ANOVA) test with a post-hoc Tukey test was used. Spearman's rank correlation was used to test any correlation between the number of ingested MPs and the fish condition index or GaSI. Non-metric Multidimensional Scaling (n-MDS) was performed on a Bray-Curtis similarity matrix created from transformed data (square root). This test was used as an ordination method alongside ANOSIM and SIMPER tests to compare the contribution of each observed polymers in sea water, sediments and biota to the similarity and dissimilarity between sites. Significance level was set at  $p < 0.05$ . All statistical tests were done



using SPSS software (IBM SPSS STATISTICS 20) and PRIMER v7.0 (Primer-E Ltd., Plymouth Marine Laboratory, UK).

### **3. Results and Discussion**

#### **3.1. Microplastics concentrations in sea surface waters and sediments**

Microplastics found along the Lebanese coastal waters consisted mainly of fragments (77.5%), fibers (15.25%) and microbeads (7.25%) (Figure 2- A). For all sites, blue and red colors were the most abundant representing 70% of all MPs. Concentration in the surface waters (4.3 MPs/m<sup>3</sup>) were high compared to other Mediterranean regions and particularly the western part where MPs concentrations vary between 0.17 and 0.62 MPs/m<sup>3</sup> (Table 1). In the Eastern Mediterranean, the MPs concentration fluctuates importantly across regions, ranging from 0.7 items/m<sup>3</sup> in Northeast Levantine coast of Turkey (Güven et al., 2017) to  $7.68 \pm 2.38$  MP/m<sup>3</sup> in the Israeli surface water (van der Hal et al., 2017).

The heterogeneity of the MPs distribution and of its concentration levels could be related to several factors. The mesh size of the net used could play an important role on the number of microplastics caught during sampling. The high concentration of MPs found in the present study maybe due to the low mesh size manta trawl net used (52 µm) compared to other studies (generally between 200 and 500 µm). Also, the different methodological analyses and the lack of a standard protocol could also be the cause of that difference. In fact, the water samples in this study were filtered on GF/A filters and all suspected items were observed; whereas other studies used organic material digestion (Gündoğdu and Çevik, 2017; Güven et al., 2017) or just visual observation (Fossi et al., 2016; Lefebvre et al., 2019) which can lead to an underestimation and a bias in microplastics counting. In addition, the distance of the sampling location from the coast may also influence the concentration of MPs found. Our samples were collected in the coastal waters (about 500 m from the shore). MPs concentration might have differed if the collected samples came from the open sea (Van der Hal et al., 2017; Pedrotti et al., 2016). On the other hand, the influence of hydrodynamic features such as currents, up and down-welling, gyres and fronts could also be responsible for this heterogeneity in MPs

concentrations. The Eastern Mediterranean has been reported for its high contamination of floating plastic debris with a dominance of small fragments < 2.5 cm in comparison to the Western part (Cózar et al., 2015). High fluxes of plastic debris (10-30 kg/km.day) were observed along the Levantine Basin coast from Egypt, Lebanon, Syria and Turkey (Constantino et al., 2019). Differences between the two parts of the Mediterranean Sea can be explained by the different circulation patterns: water currents in the Western part promote circulation; whereas currents in the Eastern part are somewhat attractors leading to the accumulation of floating debris (Mansui et al., 2015). In addition, the complexity of circulation with the existence of mesoscale and sub-basin scale eddies in the Levantine basin (most specifically Shikmona anti-cyclonic eddy) may also explain the high number of MPs found on the Lebanese coast (Menna et al., 2012).

High concentrations of MPs in sediments were also found along the Lebanese coast ( $2433 \pm 2000$  MPs/Kg d.w.). These concentrations were higher than those found in others Mediterranean regions (Supplementary Table 2). This important difference can be either caused by the different density separation solution used ( $\text{ZnCl}_2$  used in our study is of higher density than NaCl used in other studies and more microplastics can be identified) or by the mismanaged waste along the Lebanese coast where high plastic items were visually observed during sampling. Both micro and macroplastics (MaPs) were present in all collected sediments samples. Lebanon is a potential landfall of plastics along with other Eastern Mediterranean regions (Duncan et al., 2018) explaining the high abundance of macroplastics found in our samples (consisting of half of the observed anthropogenic materials found). All kind of shapes were found: square, rectangular, spherical and cylindrical (Figure 3). White was the dominant color in Sidon and Tripoli (46.3% and 36.8%, respectively); blue color was the most abundant in Beirut (23.7%). Pellets were highly abundant in two of our sampling sites (Tripoli and Sidon) which explain the high abundance of white color in these two locations. Several studies investigating MPs abundance in the Mediterranean sediments also reported the high presence of plastic pellets among their samples (Karkanorachaki et al., 2018; Fanini and Bozzeda, 2018; Kaberi et al., 2013; Turner and Holmes, 2011). The first signaled pellets on the Lebanese coast were due to direct waste discharge of plastic factories on the beaches or in the rivers (Shiber, 1979). To

this day, and with the lack of an appropriate treatment, these industries are still releasing their wastes into the aquatic system and are susceptible to unintentional losses leading to this high accumulation of plastic pellets. All of which indicate their persistence whether in the Eastern or Western Mediterranean and they (pellets and fragments) consist potential harm to the benthic organisms inhabiting sandy beaches (Carson et al., 2011; Nelms et al., 2016).

Along the Lebanese coast, inter-site differences in MPs concentrations were observed. Surface water samples showed that Sidon was the most contaminated site with 6.7 MPs/m<sup>3</sup> (Figure 2-A). This site is characterized by a high presence of microbeads with 1.16 microbeads/m<sup>3</sup>. Beirut had the lowest concentration of MPs with 2.35 MPs/m<sup>3</sup> consisting mainly (90%) of fragments. The high number of MPs and pellets observed in the south of Lebanon can be explained by site's localization near the biggest landfill. Although this landfill is closed since 2013, results of our study provide preliminary evidence that landfill isn't the final sink of plastics, but a potential source of microplastics (He et al., 2019). Fragmentation and release of microplastics is a long-term process. Microplastics inside leachates from active and closed landfills have been identified (He et al., 2019). After the stop of post-closure care, microplastics in leachate would be discharged directly into coastal waters. This can pose serious environmental problems since microplastic is not listed as pollutant in the any country's regulations of landfill. This area is also characterized by the presence of the mouth of a major river, Al-Awali River. During sampling, we observed fragments and other wastes being brought into the coastal waters via the river runoff. This river extends for 48 km of length and collects all the urban water from nearby villages due to the non-existence of sewage treatment system. These microplastics are prone to be washed from surface waters onto beaches and, therefore, integrate within the sediments (Barnes et al., 2009; Poeta et al., 2014). The general circulation along the coast of Lebanon is dominant northwards and the water movements are strongly related to surface currents and seasonal metrological factors (Goedicke, 1972). This northwards movement might explain why MPs and MaPs were the most abundant in sediments samples of Tripoli (4.68 MPs/g and 4.08 MaPs/g, respectively) (Figure 2- B). The high levels of MPs found in the surface water in Sidon and Beirut might have been carried by surface circulation ending up washed on Tripoli beaches.

### 3.2. Microplastics in anchovies and spiny oysters

Microplastics can be ingested by a large variety of marine species and more specifically by filter-feeders (Van Cauwenberghe and Janssen, 2014; Catarino et al., 2018; Collard et al., 2015) raising the alarm for commercial species that are highly consumed by humans. In our samples, 83.3% of European anchovies analyzed had ingested MPs. Such high level of ingestion is in agreement with what was described for anchovy from the Adriatic Sea (90%) (Renzi et al., 2019a). However, in the Western Mediterranean Sea, lower occurrences of MPs were described in anchovy digestive tracts (11 to 40 %) (Collard et al., 2015; Compa et al., 2018; Lefebvre et al., 2019) (Table 2). In the present study, anchovies ingested an average of  $2.5 \pm 0.3$  MPs/individual which was higher than that found in anchovies collected on the Northwestern Mediterranean coasts such as Spanish coasts ( $0.18 \pm 0.20$  items/individual) (Compa et al., 2018) and Gulf of Lions (0.11 to 0.85 MPs/individual) (Collard et al., 2015; Lefebvre et al., 2019) but lower than those collected in the Adriatic Sea ( $6.62 \pm 3.67$  items/individual) (Renzi et al., 2019a). Anchovies caught in the Western Mediterranean Sea had lower ingested MPs than those caught in the Eastern and Central Mediterranean confirming the higher pollution of these regions (Table 2). For instance, Japanese anchovies, *Engraulis japonicus* from the heavy polluted area of Tokyo bay displayed the same MPs ingestion ( $2.30 \pm 2.50$  MP/ind Tanaka and Takada, 2016) as in our study.

Three shapes of plastics were found: fragments, films and fibers (Figure 2 – C; Figure 3). There was a slight dominance of fragments ( $1.3 \pm 0.36$  fragments/fish) over of fibers ( $1.1 \pm 0.17$  fragment/fish), whereas films had the lowest occurrence ( $0.1 \pm 0.1$  films/fish). The items had different colors: black, blue, red, white, transparent, and green. For the three sites, blue fragments were the most abundant representing more than 50%; whereas for the fibers, black was the dominant color. This predominance of blue plastic items has been previously reported in fish and bivalves (Romeo et al., 2015; Güven et al., 2017; Compa et al., 2018; Digka et al., 2018).

Along the Lebanese coast, anchovies from Beirut had ingested the highest number of MPs items ( $2.9 \pm 1.9$  MPs/individual) and those from Tripoli the lowest number ( $2.3 \pm 1.6$  items/individual) (Supplementary Table 3). However, there were no significant differences between the numbers

of ingested items at the three analyzed sites ( $P > 0.05$ ). No relationship was highlighted between the concentration of MPs in anchovy digestive tracts and in the surface water as it has been described in the western Mediterranean (Compa et al., 2018; Lefebvre et al., 2019). As a pelagic species, anchovy are highly moving fish so they potentially did not ingest MPs at the site they were collected, making correlations harder to underline. Gastro-Somatic Index (GaSI) and Fulton condition index were higher in Beirut yet not significantly different (Supplementary Table 3). There was no correlation between GaSI and the number of ingested items ( $r = 0.067$ ;  $P > 0.05$ ). Fulton condition index and the number of ingested items showed a weak but not significant correlation ( $r = 0.316$ ;  $P = 0.089 > 0.05$ ). In the western Mediterranean Sea, recent studies also revealed that anchovies' MPs ingestion was not related to the body condition (Compa et al. 2018; Lefebvre et al., 2019).

Spiny oyster is a Lessepsian species of high commercial value in Lebanon. The collected individuals had an average of  $7.2 \pm 1.4$  items/individual and  $0.45 \pm 0.3$  items/g ww. To date, this is the first study done on MPs ingestion in *S. spinosus* in the Mediterranean and worldwide. Therefore, our results can only be compared to other oysters or bivalves species. Oysters from the present study had the same average of items found in *Crassostrea gigas* collected from the Atlantic Ocean with an average of  $0.47 \pm 0.16$  particles/g ww (Van Cauwenberghe and Janssen, 2014), yet lower than that found in wild mussels (*Mytilus galloprovincialis*) collected from the central Tyrrhenian Sea (Mediterranean) with an average of 7.2 items/g ww (Renzi et al., 2018a) and from the Adriatic Sea with an average of 1.06–1.3 items/g ww (Gomiero et al., 2019). Our samples were less contaminated than those collected from the Mediterranean which is probably due to the difference in the analyzed species and not the geographical MPs concentration in the environment. Oysters tend to have more flesh than mussels explaining the difference in MPs ingestion per gram of ww.

Oysters from all sites had the presence of fragments, films and fibers with 86.3% of analyzed individuals having suspected MPs in their soft tissue (Figure 2- C). Due to a difference in individual total body soft tissue weight, the number of items was also reported to the number of items per gram. Oysters from Beirut had the highest items concentration with  $8.3 \pm 4.4$

items/individual (or  $0.78 \pm 0.62$  items/g) out of which fibers constituted more than 50%. Sidon had the highest number of fragments/individual reaching  $4.3 \pm 4.4$  fragments/individual. Tripoli had the lowest number of items/g with  $0.22 \pm 0.13$  items/g. Beirut was significantly different when comparing to the other sites ( $P < 0.05$ ). The observed items had six different colors similar to those found in the anchovies; with blue color being dominant in the oysters' samples.

### **3.3. Size class distribution and polymers characterization**

In the water samples, the size class  $< 200 \mu\text{m}$  dominated in Tripoli (48.78%) and Sidon (51.8%) whereas for Beirut, the size class  $> 1000 \mu\text{m}$  dominated (31.7%) (Figure 3 – A). Suspected items  $< 1000 \mu\text{m}$  represented a percentage of 88%, 69.3% and 87% at Tripoli, Beirut and Sidon, respectively. All sediment samples had a high abundance of the size class  $> 1000 \mu\text{m}$  representing more than 70% of the samples (Supplementary Figure 1). For the biota, the smallest size class  $< 200 \mu\text{m}$  was the most abundant in all sites (Figure3 - B). For anchovies, the ingested size class 200-400  $\mu\text{m}$  was the second most dominant in Tripoli and Beirut with a percentage of 34%. The size class 600-800  $\mu\text{m}$  was only present in Sidon's anchovies, whereas the size class  $> 1000 \mu\text{m}$  was absent in Tripoli. These results agree with the feeding ecology of the European anchovy which is a filter-feeder that feeds on planktonic organisms generally  $< 2$  mm (Bacha and Amara, 2009). No relationship between the size of the anchovies and the size of the ingested items was observed. Although Beirut's anchovies were significantly ( $P < 0.05$ ) bigger than those from the other two sites, they ingested items of a similar size than those ingested in Tripoli and Sidon. This indicates that the size of ingested items may be related to the available particles during feeding rather than the morphometric characteristics of organisms (i.e. mouth opening size). Plastics of all size classes were present inside the oyster's body soft tissue. The smallest size class (0-200  $\mu\text{m}$ ) was the most abundant for all sites. The two size classes: 200-400  $\mu\text{m}$  and  $> 1000 \mu\text{m}$  were the second most abundant between the ingested MPs. Even though there was a significant difference in spiny oyster's net weight ( $P < 0.05$ ) between sites, all MPs items size classes were present in all collected individuals. This confirms that the size of ingested items is not related to the individual weight, but to the available particles existing in the surrounding water.

Among the suspected MPs analyzed, 94% were identified as microplastics. Nine types of polymers were identified in our samples (Figure 4). Six types were common to all three matrices (water, sediments and biota): polypropylene (PP), polyethylene (PE), polystyrene (PS), polyamide (PA), polyethylene terephthalate (PET) and polyurethane (PUR). In addition to these polymers, polyvinyl chloride (PVC) was present in water and sediments; polylactic acid (PLA) was only present in the water whereas acrylonitrile butadiene styrene (ABS) was only present in biota. Each of the three matrices had a specific polymer that dominated over the others: PE was the most abundant in water samples (70.83%, 50% and 47.37% at Tripoli, Beirut and Sidon, respectively). Polyethylene is the most common plastic on a global scale and consequently the most dominant plastic debris in the Mediterranean Sea and worldwide, deriving mainly from plastic bags and bottles (Suaria et al., 2016). It is well documented that plastics as PE float in the water surface, as its density is lower than this of the water. Polymers with a density higher than that of water (PVC, for example) were observed in the surface water. The existence of polymers denser than seawater have been already reported (Suaria et al., 2016; Enders et al., 2015) with no clear mechanism leading to their existence. During sampling, there was strong waves and the water was agitated which might explain their presence due to the high velocity and turbulent mixing which increases the chances of denser polymers to persist on the sea surface (Moritomi et al., 1982; Suaria et al., 2016). Polypropylene was the most abundant in sediments (48.11%, 55.88% and 43.33% at Tripoli, Beirut and Sidon, respectively), and polystyrene dominated in the biota specimens (having a percentage equal to or above 25% for anchovies and spiny oysters). Polypropylene was observed in all anchovies and spiny oysters of all sites except in the anchovies of Beirut where PE was observed instead. Polymers distribution didn't have a pattern with shapes or colors. For example, pellets found were mostly white (several were colored) yet their polymer type was of PE, PA, and PP. The same for fragments, different colors were related to different polymers with no similar pattern.

Comparison of the polymers composition identified in water, sediments and oysters indicated a dissimilarity between groups (ANOSIM,  $R=0.796$ , significance level = 0.2%). Cluster dendrogram led to the existence of two distinct groups (a and b) regrouping on the one hand the water and sediment samples and on the other hand the biota (anchovies and oysters) (Supplementary

Figure 2). Non-metric multidimensional scaling (n-MDS) showed the dimensional distance separating these groups (a and b) based on their polymer composition. Water and sediments formed 'group a' and were characterized by the high presence of PP and PE. Whereas 'group b' composed by biota samples, and were characterized by the high abundance of PS. Similarity of Percentages (SIMPER) indicated that PE, PP, PA and PUR contributed to 72.12% of dissimilarity between these two groups.

#### **3.4. Microplastics potential entry into the Mediterranean Basin via coastal landfills, and their fate and distribution in the coastal environment**

This study showed a high microplastics pollution in areas located in the vicinity of coastal landfills. Even though some of them are no longer active, but their waste continues to find its way into the sea due to mismanagement and lack of proper treatments. Some of the polymers found in our samples such as polyethylene, polypropylene and polystyrene were observed in transparent, white and different colors. They are widely used in packaging, consumer products, pipes and fitting, and are the common polymers found in landfills and their leachates (Kazour et al., 2019; He et al., 2019; Praagh et al., 2018). The high abundance of these types of polymers in sea surface water and sediment samples can be traced back to mismanaged landfills along with accidental garbage disposal. Even though Lebanon is considered as a potential sink of plastics in the Eastern Mediterranean (Duncan et al., 2018), it should also be considered as an important microplastics source in the Mediterranean basin. In most studies, whether in the surface water, sediments or biota, fibers were generally the dominant microplastic shape found (Lefebvre et al., 2019; Renzi et al., 2019b; Compa et al., 2018). In the present study, fragments constituted more than 77%, 90% and 50% of items found in surface water, sediments and biota samples, respectively. The same pattern was found in a study done near an abandoned coastal landfill, where fragments constituted the majority of found items (Kazour et al., 2019). Such dominance of fragments in the analyzed samples highlights on the potential role of Lebanese coastal landfills in fragments input into the Mediterranean Basin. Other factors can also lead to this severe microplastics contamination and, most precisely, on fragments abundance on the



Lebanese beaches. Munari et al. (2017) indicated a high number of fragments (approximately 90% of the samples) was observed near beaches affected by riverine run-off. All of the sampled Lebanese beaches are affected by riverine runoff as well as a significant anthropogenic pressure leading to a major microplastics pollution along the beaches (100 times higher than other reported studies, see Supplementary Table 3). These microplastics are prone to re-enter the surface water with waves hitting the shoreline (directed by wind speed and direction) increasing their chance to end up in nearby regions via the northward general water circulation pattern. The above stated factors increase microplastics availability to marine organisms leading to potential human health risks. In Lebanon, an average of 19.7g of fish and seafood per person is consumed (Nasreddine et al., 2006) making it around 7 kg per caput per annum. Converting the average of ingested MPs/g in oysters to the number of seafood ingested by a Lebanese person, we would have approximately 31, 500 MPs ingested per person (4500 MPs/kg of oysters). This is similar to that obtained by Catarino et al. (2018) (13,731 and 68,415 particles/y/person) yet lower than that obtained by Van Cauwenberghe & Janssen (2014) (11, 000 MPs/y/person). European anchovies are eaten wholly by the Lebanese population as *Bizree* (juvenile pelagic fish caught by purse seiners) as well as spiny oysters that are served in various Lebanese restaurants raising the concern on the human consumption of these contaminated seafood.

#### 4. Conclusion

These results should be considered as a preliminary analysis on MPs levels recorded off the Lebanese coast in three different matrices. This is the first contribution to characterize the area and to understand the degree of its MPs pollution. More studies are necessary to evaluate the ecological impacts of these MPs on the marine biodiversity in the Levantine Basin. The levels of MPs found were alarmingly high in comparison to other Mediterranean regions. The high number of ingested items by both anchovies and oysters could suggest a potential health concern since these two species are wholly consumed (without being degutted). This study highlighted the role of landfills in MPs entry into the coastal marine environment. Coastal landfills and mismanaged wastes are important issues existing in 80% of the Mediterranean

Basin explaining the elevated number of MPs found in this semi-enclosed basin. The whole Mediterranean should be regarded as an important case study for microplastics pollution where more initiative should be taken to insure an appropriate waste management and, hence, decreasing plastics pollution.

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Figure 1: Localization of the sampling sites (Tripoli, Beirut and Sidon) along the Lebanese coast.

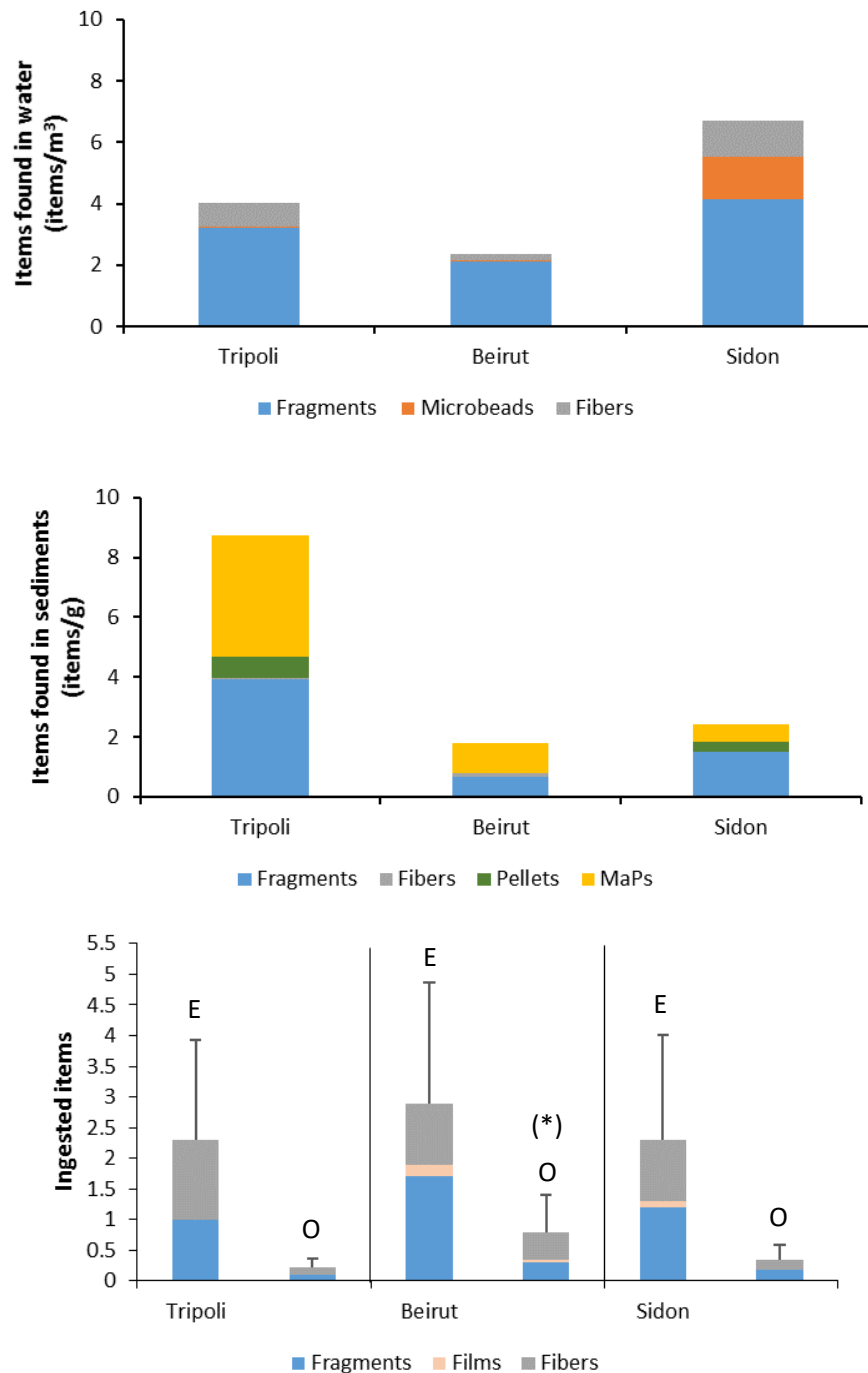
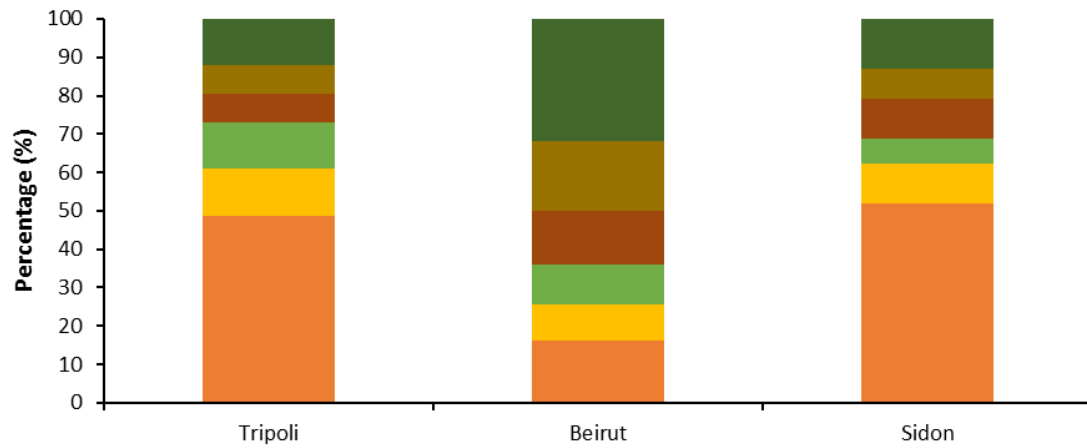


Figure 2: A) Concentration of suspected microplastics (MPs/m<sup>3</sup>) according to their shapes found in the surface water; B) Suspected microplastics (MPs) and macroplastics (MaPs) concentration (items/g) found in one gram of sublittoral sediments; C) Average number ( $\pm$ SD) of ingested suspected microplastics by shape categories in the digestive tract of (E) European anchovies (MP/ind) and in the soft tissues of (O) spiny oysters (MP/gram) at the three sampling sites (\* indicate statistically significant difference).

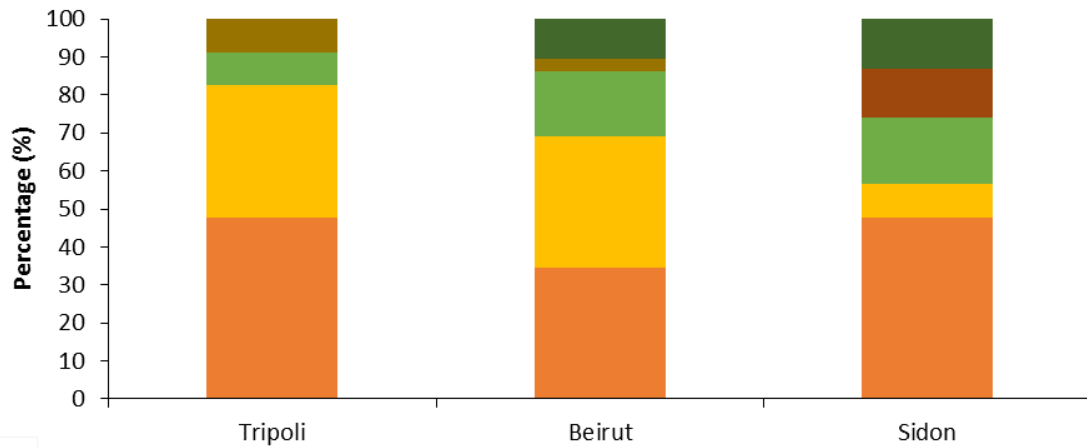


Figure 3: Microplastics of different shapes and colors collected along the Lebanese coast in three different matrices: e.g. A) blue microbead and B) green fiber found in the collected surface water, C) Macro and microplastics collected on the 2 mm sieve and D) white and black pellets collected in Tripoli sediments sample, E) blue fiber and F) blue fragment observed on biota filter.

A)



B)



C)

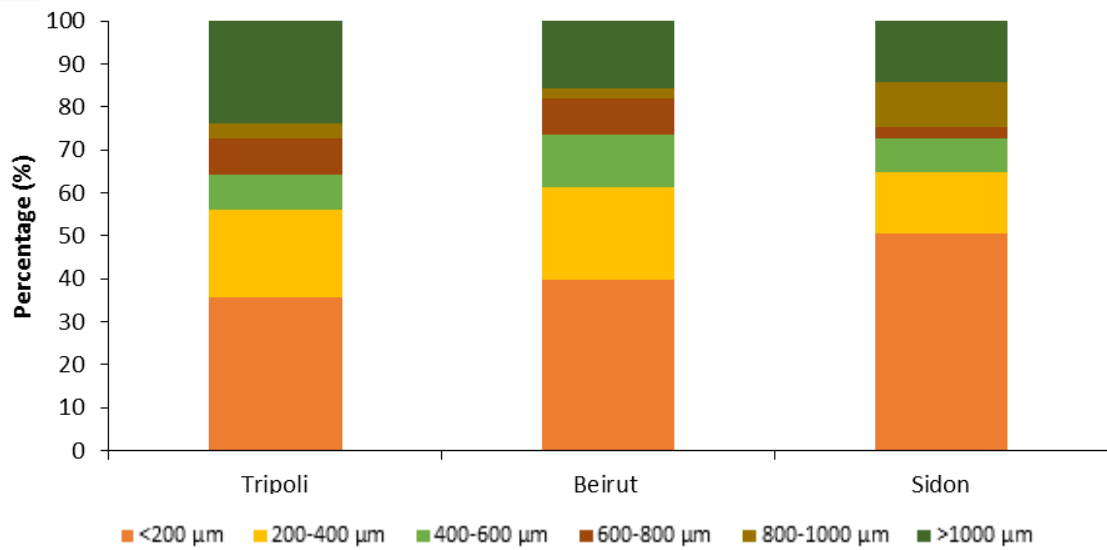


Figure 4: Proportion of size class composition of suspected microplastics in the A) Water surface, B) anchovies and C) spiny oysters from the different sampling sites



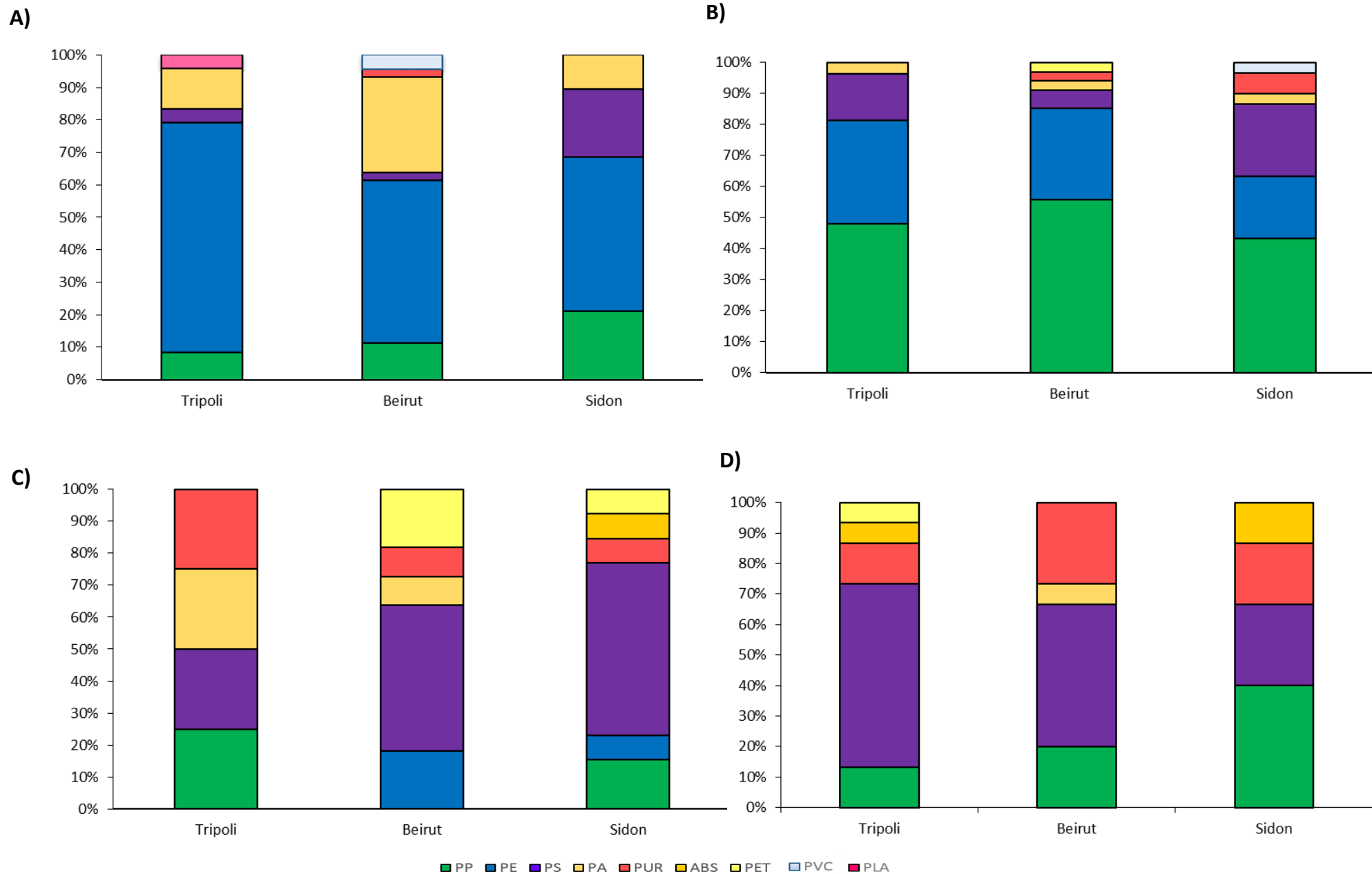


Figure 5: Proportion of different polymers identified by micro-Raman spectroscopy in the three analyzed compartments: A) water; B) Sediments, C) anchovies and (D) spiny oysters.

Table 1: Literature data on microplastics abundance in Mediterranean surface waters and instrumental analysis methods used for detection of MPs. To compare with our results, only studies expressing concentrations in m<sup>3</sup> are presented.

Location	Sampling nets	Mesh size ( $\mu\text{m}$ )	Identification method	MPs abundances	Reference
NW Mediterranean (the Rhône River and the Têt River)	Manta trawl net	333	Stereomicroscope/FTIR	0.18- 0.19 items/m <sup>3</sup>	Constant et al., (2018)
NW Mediterranean Sea (Ligurian Sea and Sardinian Sea)	WP2 standard net	200	Stereomicroscope	0.62 $\pm$ 2 items/m <sup>3</sup>	Fossi et al., (2012)
NW Mediterranean Sea (Gulf of Lion)	WP2 plankton net	200	Stereomicroscope/FTIR	0.23 $\pm$ 0.20 items/m <sup>3</sup>	Lefebvre et al., (2019)
NW Mediterranean Sea (Gulf of Asinara and the Sardinian Sea)	Neuston net	200	Stereomicroscope	0.31 $\pm$ 1.17 items/m <sup>3</sup>	Fossi et al., (2016)
NW Mediterranean Sea (North	WP2 standard ring net	200	Stereomicroscope	0.17 $\pm$ 0.32	Panti et al., (2015)

Western of Sardinia; Gulf of Asinara)				items/m <sup>3</sup>	
NW Mediterranean Sea (Tuscany coastal water)	Manta trawl net	330	Stereomicroscope/FTIR	0.26/m <sup>3</sup>	Baini et al., (2018)
Central-Western Mediterranean Sea (Sardinian Sea)	Manta trawl net	500	Binocular stereomicroscope	0.15 items/m <sup>3</sup>	De Lucia et al., (2014)
Eastern Mediterranean (Turkish Mediterranean Coast)	Manta trawl net	333	Microscopic observation/FTIR on 24 items	0.7 items/m <sup>3</sup>	Güven et al., (2017)
Eastern Mediterranean (Iskenderun Bay)	Manta trawl net	333	Microscopic observation	7.26 items/m <sup>3</sup>	Gündoğdu, (2017)
Eastern Mediterranean Sea (Iskenderun Bay and Mersin Bay)	Manta Trawl net	333	Microscopic observation	2.73 items /m <sup>3</sup>	Gündoğdu and Çevik, (2017)

South Eastern Mediterranean Sea (Israeli surface waters)	Manta Trawl net	333	Stereomicroscope	$7.68 \pm 2.38$ items/m <sup>3</sup>	van der Hal et al., (2017)
Lebanese coast	Manta Trawl net	52	Stereomicroscope/Raman	$4.3 \pm 2.2$ items/m <sup>3</sup>	Present study

Table 2: Average ( $\pm$ SD) of ingested items found in the digestive track of European anchovies (*Engraulis encrasicolus*) collected in different areas of the Mediterranean Sea.

Sites	Digestion method	Concentration (items/ind)	Occurrence	Polymers Identification method	Main polymer	Reference
Spanish Western Mediterranean coasts	None. Visual observation	0.07 ( $\pm$ 0.35) to 0.33 ( $\pm$ 0.26)	15%	FTIR	PET	Compa et al., (2018)
Western Mediterranean Sea (Iberian Peninsula coast and the Balearic Islands)	None Visual observation	0.07 $\pm$ 0.26	2.56%	None	-	Rios-Fuster et al., (2019)
North Western of the Mediterranean Sea (Gulf of Lions)	None. Visual observation	0.11 $\pm$ 0.31	11%	ATR-FTIR	PET	Lefebvre et al., (2019)
Northwestern Mediterranean Sea	NaClO	0.85	40%	Raman spectroscopy	PE, PP, and PET	Collard et al., (2015)
Northern Mediterranean (Adriatic Sea)	H <sub>2</sub> O <sub>2</sub>	1.25	91%	$\mu$ -FT-IR	PVC	Renzi et al., (2019a)
Eastern Mediterranean Sea (Lebanese coast)	KOH 10%	2.5 $\pm$ 0.3	83.4%	Raman Spectroscopy	PS	Present study

