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

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Occurrence in surface water, sediments and biota samples

3

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9 The Mediterranean Sea is the largest semi-enclosed sea and one of the worst affected regional seas with 10 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 11 12 first time, the MPs pollution of the Lebanese coast (Levantine Basin) as well as the most common 13 polymers found, and to assess the potential role of coastal landfills in this pollution. Two important 14 seafood species that are wholly consumed by the Lebanese community: the European anchovy Engraulis 15 encrasicolus and the spiny oysters Spondylus spinosus were sampled in three different sites englobing 16 the littoral (Tripoli, Beirut and Sidon). Sea water and sediment samples were also collected from the 17 same sites. Results showed different patterns of MPs concentration in the analyzed matrices: Sidon water sample was highly contaminated in MPs (6.7 MPs/m³) whilst Tripoli had the highest MPs in 18 sediments (4.68 MPs/g). The occurrence of MPs in the biota was high (83.4% and 86.3% in anchovies and 19 20 spiny oysters, respectively). Both anchovies and oysters from Beirut region had the highest ingested 21 MPs/ individual (2.9 ± 1.9 and 8.3 ± 4.4 MPs/individual, respectively). This study is the first that investigated microplastics ingestion by Spondylus spinosus while indicating the most common polymers 22 23 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 24 25 Mediterranean regions. In addition, the obtained results indicate a potential contribution of coastal 26 landfills to this pollution.

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28 Keywords: Microplastics, Levantine Basin, coastal landfill, filter-feeders, micro-Raman

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31 **1. Introduction**

The world plastic production is continuously increasing, reaching 350 million tons in 2017 32 (PlasticsEurope, 2018). Among the produced plastics, only 6–26% are recycled. Landfills receive 33 21-42% of wastes, whereas the rest are wildly released into the environment due to 34 35 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 36 2025, about 250 million tons of wastes are expected to enter the ocean water (Jambeck et al., 37 2015) leading to their accumulation in the marine environment (Thompson et al., 2009; UNEP, 38 2014). Plastic pollution in the marine environment is becoming a problem of global concern. 39 40 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 41 42 environments (Thompson et al., 2004; Andrady, 2011). They are either intentionally produced in 43 small sizes such as polystyrene beads for example, or they are the resultant of macroplastics 44 degradation (whether by Photochemical reaction, or mechanical actions.).

45 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 46 47 (Lebreton et al., 2012; Cincinelli et al., 2019). Mainly owing to the semi-enclosed situation and 48 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 49 50 Mediterranean Basin. Also, most studies on microplastics have been conducted in the 51 northwestern Mediterranean Sea (e.g. Collignon et al., 2014; Fossi et al., 2016; Ruiz-Orejón et al., 2016; Schmidt el al., 2018; Lefebvre et al., 2019). Results showed that their distribution and 52 composition are not homogeneous and geographical differences exist between Mediterranean 53 sub-basins (Suaria et al., 2016; Cincinelli et al., 2019). Such heterogenicity in MPs distribution 54

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 67 the Eastern part of the Mediterranean Basin (Van der Hal et al., 2017; Güven et al., 2017; 68 69 Gündoğdu and Çevik, 2017). In the Levantine basin, the Lebanese coast is highly affected by 70 numerous anthropogenic pressures such as harbor activities, fishing, domestic and industrial 71 wastewater discharges (generally without prior treatment). Waste production in Lebanon has 72 increased significantly during the last decade and the country has experienced waste crises. The 73 regulated dumping sites could no longer manage the tremendous quantity of municipal garbage generated and, therefore, numerous unregulated and open dumping sites were created 74 75 throughout the country mainly along the coast (Khalil et al., 2018). The average composition of 76 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). 77 Though the processes may be slow, landfills are affected by UV radiation and mechanical 78 79 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). 80

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, 84 sediments and marine biota) along the Lebanese coast. All suspected MPs were characterized 85 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; 86 Kazour et al., 2019), making them a potential vector of toxic chemicals through food chains and 87 into human diets. In the present study we analyzed two marine species that are wholly 88 consumed by the Lebanese community: one pelagic fish species: the European anchovy, 89 engraulis encrasicolus (Linneaus, 1758) and a bivalve species, the spiny oyster, Spondylus 90 91 spinosus (Schreibers, 1793).

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93 2. Materials and Methods

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2.1. Study and sampling area

95 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 96 97 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 98 fishing harbor, and the existence of a coastal landfill. This latter was constructed in the year 99 1999 and reached today 36 m height receiving about 400 tons of wastes/day. The second site 100 101 Beirut, located in the middle, is the country's capital. This site is near the most important harbor 102 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, 103 104 located in the south, is facing a river's mouth and is near a closed landfill operated between 105 1982 and 2013. With a height of 58 m, this landfill was the biggest in the country receiving 300 106 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 107 108 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). 109

110 **2.2. Contamination control**

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111 Several precautions were followed to minimize air-born contamination during sampling, 112 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 113 equipment were rinsed before usage with filtered ethanol 70% and distilled water. Laboratory 114 supplies were made of glass (bottles, Petri dishes, Erlenmeyers, filtration system) and they were 115 116 covered immediately after manipulation. All solutions (distilled water, Ethanol 70%, potassium 117 hydroxide KOH 10% and zinc chloride ZnCl₂) were filtered 3 times on glass fibers filter (GF/A 118 filters, 1.6 µm porosity, Whatman, France) to eliminate unwanted particles or fibers. Controls 119 were made to follow up the contamination occurring with every step of preparation and 120 observation.

121 **2.3. Water and sediments sampling and analyses**

122 Sea surface water and sediments samples were collected during spring 2018. Sea surface water was collected using a manta trawl net (52 μ m mesh size) with an opening of 0.63 m and a length 123 of 0.255 m equipped with a flowmeter. The net was trawled for 10 minutes parallel to the coast 124 125 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 m³ of sea water was sampled at each site (Supplementary Table 1). The net 126 127 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 µm and 128 129 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 130 131 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°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 ZnCl₂ (d= 1.8 g/cm³) (Imhof et al., 2013; Nuelle et al., 2014). Briefly, 50 mL of filtered ZnCl₂ 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 m³ in sea water samples.

141 **2.4. Biota sampling and analyses**

Anchovies and spiny oysters were also collected during spring 2018. Anchovies were caught 142 143 using purse seine nets. Ten individuals of approximately the same size (9 cm \pm 0.9) were chosen from each sampling site and were immediately individually conserved in aluminum foil and 144 145 conserved at -4°C. In the laboratory, their mormophetric characteristics (total length and 146 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 147 collected by fishermen from each sampling site. In the laboratory, the whole individual was 148 149 removed from its shell and was conserved in aluminum foil. The net weight of the animal 150 ranged between 4.8 g to 38.2 g -YY with an average of 20.2 g \pm 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.

156 **2.5. Stereomicroscope and micro-Raman spectroscopy**

157 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 158 159 (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 160 items: (1) absence of cellular or organic structures; (2) a homogenous thickness across the 161 162 particles; and, (3) homogenous colors (Hidalgo-Ruz et al. 2012). Suspected microplastics were 163 divided into 6 size classes: <200 µm, 200-400 µm, 400-600 µm, 600-800 µm, 800-1000µm, and 164 > 1000 µm.

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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 168 169 anchovies and five filters of spiny oysters from each site containing the highest number of 170 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 171 172 wavelength of 532 nm and 785 nm with a range of 200-3400 cm⁻¹. Two objectives (Olympus, Rungis, France) were used: x10 and x100. Filters were either analyzed manually or using 173 174 ParticleFinder module for LabSpec 6. Each particle spectrum is compared to a database polymer 175 identification software (KnowItAll, BioRad®) and a personal library made with specific polymers 176 obtained from Goodfellow (France).

177 **2.6. Data and Statistical analysis**

178 Anchovy Fulton's K condition index was calculated as an indicator of the fish general well-being: K= (Body weight/total length³)*100. The feeding intensity for the fish was calculated using the 179 Gastro Somatic Index (GaSI) (Desai 1970): GasI = stomach weight/ (body weight – stomach 180 weight). The mean (±SD) abundances of microplastics were calculated for anchovies (MPs/ind) 181 and spiny oysters (MPs/g). After a Shapiro-Wilk test, data distribution of ingested MPs were non 182 normal and a non-parametric tests (Kruskal-Wallis and Mann-Whitney U tests) were used to 183 184 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 185 186 was used. Spearman's rank correlation was used to test any correlation between the number of 187 ingested MPs and the fish condition index or GasI. Non-metric Multidimensional Scaling (n-MDS) 188 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 189 190 contribution of each observed polymers in sea water, sediments and biota to the similarity and 191 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).

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196 **3. Results and Discussion**

197 **3.1.** Microplastics concentrations in sea surface waters and sediments

Microplastics found along the Lebanese coastal waters consisted mainly of fragments (77.5%), 198 fibers (15.25%) and microbeads (7.25%) (Figure 2- A). For all sites, blue and red colors were the 199 most abundant representing 70% of all MPs. Concentration in the surface waters (4.3 MPs/m³) 200 201 were high compared to other Mediterranean regions and particularly the western part where MPs concentrations vary between 0.17 and 0.62 MPs/m³ (Table 1). In the Eastern Mediterranean, the 202 MPs concentration fluctuates importantly across regions, ranging from 0.7 items/m³ in Northeast 203 204 Levantine coast of Turkey (Güven et al., 2017) to 7.68 ± 2.38 MP/m³ in the Israeli surface water (van der Hal et al., 2017). 205

206 The heterogenicity 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 207 microplastics caught during sampling. The high concentration of MPs found in the present study 208 209 maybe due to the low mesh size manta trawl net used (52 μ m) compared to other studies (generally between 200 and 500 µm). Also, the different methodological analyses and the lack 210 of a standard protocol could also be the cause of that difference. In fact, the water samples in 211 this study were filtered on GF/A filters and all suspected items were observed; whereas other 212 213 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 214 215 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 216 217 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 218 219 al., 2016). On the other hand, the influence of hydrodynamic features such as currents, up and 220 down-welling, gyres and fronts could also be responsible for this heterogeneity in MPs

221 concentrations. The Eastern Mediterranean has been reported for its high contamination of 222 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 223 along the Levantine Basin coast from Egypt, Lebanon, Syria and Turkey (Constantino et al., 224 225 2019). Differences between the two parts of the Mediterranean Sea can be explained by the 226 different circulation patterns: water currents in the Western part promote circulation; whereas 227 currents in the Eastern part are somewhat attractors leading to the accumulation of floating 228 debris (Mansui et al., 2015). In addition, the complexity of circulation with the existence of 229 mesoscale and sub-basin scale eddies in the Levantine basin (most specifically Shikmona anti-230 cyclonic eddy) may also explain the high number of MPs found on the Lebanese coast (Menna et 231 al., 2012).

232 High concentrations of MPs in sediments were also found along the Lebanese coast (2433 ± 233 2000 MPs/Kg d.w.). These concentrations were higher than those found in others 234 Mediterranean regions (Supplementary Table 2). This important difference can be either caused 235 by the different density separation solution used (ZnCl₂ used in our study is of higher density than NaCl used in other studies and more microplastics can be identified) or by the mismanaged 236 237 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. 238 239 Lebanon is a potential landfall of plastics along with other Eastern Mediterranean regions 240 (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 241 242 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 243 244 (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 245 246 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 247 248 al., 2013; Turner and Holmes, 2011). The first signaled pellets on the Lebanese coast were due 249 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).

255 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/m³ (Figure 2-256 257 A). This site is characterized by a high presence of microbeads with 1.16 microbeads/m³. Beirut had the lowest concentration of MPs with 2.35 MPs/m³ consisting mainly (90%) of fragments. 258 259 The high number of MPs and pellets observed in the south of Lebanon can be explained by site's 260 localization near the biggest landfill. Although this landfill is closed since 2013, results of our 261 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-262 263 term process. Microplastics inside leachates from active and closed landfills have been 264 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 265 microplastic is not listed as pollutant in the any country's regulations of landfill. This area is also 266 267 characterized by the presence of the mouth of a major river, Al-Awali River. During sampling, we 268 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 269 to the non-existence of sewage treatment system. These microplastics are prone to be washed 270 271 from surface waters onto beaches and, therefore, integrate within the sediments (Barnes et al., 272 2009; Poeta et al., 2014). The general circulation along the coast of Lebanon is dominant 273 northwards and the water movements are strongly related to surface currents and seasonal 274 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, 275 respectively) (Figure 2- B). The high levels of MPs found in the surface water in Sidon and Beirut 276 277 might have been carried by surface circulation ending up washed on Tripoli beaches.

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3.2. Microplastics in anchovies and spiny oysters

280 Microplastics can be ingested by a large variety of marine species and more specifically by filter-281 feeders (Van Cauwenberghe and Janssen, 2014; Catarino et al., 2018; Collard et al., 2015) raising 282 the alarm for commercial species that are highly consumed by humans. In our samples, 83.3% of 283 European anchovies analyzed had ingested MPs. Such high level of ingestion is in agreement 284 with what was described for anchovy from the Adriatic Sea (90%) (Renzi et al., 2019a). However, 285 in the Western Mediterranean Sea, lower occurrences of MPs were described in anchovy 286 digestive tracts (11 to 40 %) (Collard et al., 2015; Compa et al., 2018; Lefebvre et al., 2019) 287 (Table 2). In the present study, anchovies ingested an average of 2.5 ± 0.3 MPs/individual which 288 was higher than that found in anchovies collected on the Northwestern Mediterranean coasts 289 such as Spanish coasts (0.18 ± 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 290 291 collected in the Adriatic Sea (6.62 ± 3.67 items/individual) (Renzi et al., 2019a). Anchovies 292 caught in the Western Mediterranean Sea had lower ingested MPs than those caught in the 293 Eastern and Central Mediterranean confirming the higher pollution of these regions (Table 2). 294 For instance, Japanese anchovies, Engraulis japonicus from the heavy polluted area of Tokyo 295 bay displayed the same MPs ingestion (2.30 ± 2.50 MP/ind Tanaka and Takada, 2016) as in our 296 study.

Three shapes of plastics were found: fragments, films and fibers (Figure 2 – C; Figure 3). There 297 was a slight dominance of fragments (1.3 \pm 0.36 fragments/fish) over of fibers (1.1 \pm 0.17 298 fragment/fish), whereas films had the lowest occurrence (0.1 ± 0.1 films/fish). The items had 299 300 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 301 302 was the dominant color. This predominance of blue plastic items has been previously reported 303 in fish and bivalves (Romeo et al., 2015; Güven et al., 2017; Compa et al., 2018; Digka et al., 304 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 308 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 309 described in the western Mediterranean (Compa et al., 2018; Lefebvre et al., 2019). As a pelagic 310 species, anchovy are highly moving fish so they potentially did not ingest MPs at the site they 311 were collected, making correlations harder to underline. Gastro-Somatic Index (GaSI) and Fulton 312 313 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> 314 315 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 316 studies also revealed that anchovies' MPs ingestion was not related to the body condition 317 318 (Compa et al. 2018; Lefebvre et al., 2019).

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Spiny oyster is a Lessepsian species of high commercial value in Lebanon. The collected 320 321 individuals had an average of 7.2 ± 1.4 items/individual and 0.45 ± 0.3 items/g ww. To date, this is the first study done on MPs ingestion in S. spinosus in the Mediterranean and worldwide. 322 323 Therefore, our results can only be compared to other oysters or bivalves species. Oysters from 324 the present study had the same average of items found in *Crassostrea gigas* collected from the 325 Atlantic Ocean with an average of 0.47 ± 0.16 particles/g ww (Van Cauwenberghe and Janssen, 2014), yet lower than that found in wild mussels (Mytilus galloprovincialis) collected from the 326 central Tyrrhenian Sea (Mediterranean) with a an average of 7.2 items/g ww (Renzi et al., 327 328 2018a) and from the Adriatic Sea with an average of 1.06-1.3 items/g ww (Gomiero et al., 329 2019). Our samples were less contaminated that those collected from the Mediterranean which is probably due to the difference in the analyzed species and not the geographical MPs 330 331 concentration in the environment. Oysters tend to have more flesh than mussels explaining the 332 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 ± 4.4 items/individual (or 0.78 ± 0.62 items/g) out of which fibers constituted more than 50%. Sidon had the highest number of fragments/individual reaching 4.3 ± 4.4 fragments/individual. Tripoli had the lowest number of items/g with 0.22 ± 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.

342 **3.3. Size class distribution and polymers characterization**

343 In the water samples, the size class < 200 µm dominated in Tripoli (48.78%) and Sidon (51.8%) 344 whereas for Beirut, the size class > 1000 μ m dominated (31.7%) (Figure 3 – A). Suspected items < 1000 µm represented a percentage of 88%, 69.3% and 87% at Tripoli, Beirut and Sidon, 345 respectively. All sediment samples had a high abundance of the size class > 1000 μ m 346 347 representing more than 70% of the samples (Supplementary Figure 1). For the biota, the 348 smallest size class < 200 μ m was the most abundant in all sites (Figure 3 - B). For anchovies, the ingested size class 200-400 μ m was the second most dominant in Tripoli and Beirut with a 349 percentage of 34%. The size class 600-800 µm was only present in Sidon's anchovies, whereas 350 351 the size class > 1000 μ 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 <2 352 mm (Bacha and Amara, 2009). No relationship between the size of the anchovies and the size of 353 354 the ingested items was observed. Although Beirut's anchovies were significantly (P < 0.05) bigger 355 than those from the other two sites, they ingested items of a similar size than those ingested in 356 Tripoli and Sidon. This indicates that the size of ingested items may be related to the available 357 particles during feeding rather than the morphometric characteristics of organisms (i.e. mouth 358 opening size). Plastics of all size classes were present inside the oyster's body soft tissue. The 359 smallest size class (0-200 μ m) was the most abundant for all sites. The two size classes: 200-400 μ m and > 1000 μ m were the second most abundant between the ingested MPs. Even though 360 361 there was a significant difference in spiny oyster's net weight (P<0.05) between sites, all MPs 362 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 363 364 surrounding water.

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366 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 367 (water, sediments and biota): polypropylene (PP), polyethylene (PE), polystyrene (PS), 368 polyamide (PA), polyethylene terephthalate (PET) and polyurethane (PUR). In addition to these 369 370 polymers, polyvinyl chloride (PVC) was present in water and sediments; polylactic acid (PLA) was 371 only present in the water whereas acrylonitrile butadiene styrene (ABS) was only present in 372 biota. Each of the three matrices had a specific polymer that dominated over the others: PE was 373 the most abundant in water samples (70.83%, 50% and 47.37% at Tripoli, Beirut and Sidon, 374 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 375 376 plastic bags and bottles (Suaria et al., 2016). It is well documented that plastics as PE float in the 377 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 378 379 denser than seawater have been already reported (Suaria et al., 2016; Enders et al., 2015) with 380 no clear mechanism leading to their existence. During sampling, there was strong waves and the 381 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 382 383 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 384 biota specimens (having a percentage equal to or above 25% for anchovies and spiny oysters). 385 386 Polypropylene was observed in all anchovies and spiny oysters of all sites except in the 387 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 388 their polymer type was of PE, PA, and PP. The same for fragments, different colors were related 389 390 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.

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

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405 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 406 way into the sea due to mismanagement and lack of proper treatments. Some of the polymers 407 408 found in our samples such as polyethylene, polypropylene and polystyrene were observed in 409 transparent, white and different colors. They are widely used in packaging, consumer products, 410 pipes and fitting, and are the common polymers found in landfills and their leachates (Kazour et 411 al., 2019; He et al., 2019; Praagh et al., 2018). The high abundance of these types of polymers in 412 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 413 the Eastern Mediterranean (Duncan et al., 2018), it should also be considered as an important 414 415 microplastics source in the Mediterranean basin. In most studies, whether in the surface water, 416 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 417 418 more than 77%, 90% and 50% of items found in surface water, sediments and biota samples, 419 respectively. The same pattern was found in a study done near an abandoned coastal landfill, 420 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 421 landfills in fragments input into the Mediterranean Basin. Other factors can also lead to this 422 423 severe microplastics contamination and, most precisely, on fragments abundance on the

424 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 425 Lebanese beaches are affected by riverine runoff as well as a significant anthropogenic pressure 426 leading to a major microplastics pollution along the beaches (100 times higher than other 427 428 reported studies, see Supplementary Table 3). These microplastics are prone to re-enter the 429 surface water with waves hitting the shoreline (directed by wind speed and direction) increasing 430 their chance to end up in nearby regions via the northward general water circulation pattern. 431 The above stated factors increase microplastics availability to marine organisms leading to 432 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 433 434 average of ingested MPs/g in oysters to the number of seafood ingested by a Lebanese person, we would have approximatively 31, 500 MPs ingested per person (4500 MPs/kg of oysters). This 435 is similar to that obtained by Catarino et al. (2018) (13,731 and 68,415 particles/y/person) yet 436 437 lower than that obtained by Van Cauwenberghe & Janssen (2014) (11, 000 MPs/y/person). 438 European anchovies are eaten wholly by the Lebanese population as *Bizree* (juvenile pelagic fish 439 caught by purse seiners) as well as spiny oysters that are served in various Lebanese restaurants 440 raising the concern on the human consumption of these contaminated seafood.

441

442 4. Conclusion

443 These results should be considered as a preliminary analysis on MPs levels recorded off the 444 Lebanese coast in three different matrices. This is the first contribution to characterize the area 445 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 446 MPs found were alarmingly high in comparison to other Mediterranean regions. The high 447 number of ingested items by both anchovies and oysters could suggest a potential health 448 concern since these two species are wholly consumed (without being degutted). This study 449 450 highlighted the role of landfills in MPs entry into the coastal marine environment. Coastal 451 landfills and mismanaged wastes are important issues existing in 80% of the Mediterranean

452 Basin explaining the elevated number of MPs found in this semi-enclosed basin. The whole 453 Mediterranean should be regarded as an important case study for microplastics pollution where 454 more initiative should be taken to insure an appropriate waste management and, hence, 455 decreasing plastics pollution.

456

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473 **References**

Abidli, S., Antunes, J.C., Ferreira, J.L., Lahbib, Y., Sobral, P., Trigui El Menif, N., 2018.
Microplastics in sediments from the littoral zone of the north Tunisian coast (Mediterranean
Sea). Estuar. Coast. Shelf Sci. 205, 1–9. https://doi.org/10.1016/j.ecss.2018.03.006

- Alimi, O. S., Farner Budarz, J., Hernandez, L. M., Tufenkji, N., 2018. Microplastics and
 nanoplastics in aquatic environments: aggregation, deposition, and enhanced contaminant
 transport. Environ. Sci. Technol. 52(4), 1704-1724. https://doi.org/10.1021/acs.est.7b05559
- Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. 62, 1596–1605.
 https://doi.org/10.1016/j.marpolbul.2011.05.030
- Bacha, M., Amara, R., 2009. Spatial, temporal and ontogenetic variation in diet of anchovy
 (Engraulis encrasicolus) on the Algerian coast (SW Mediterranean). Estuar. Coast. Shelf Sci. 85,
 257–264. https://doi.org/10.1016/j.ecss.2009.08.009
- Baini, M., Fossi, M.C., Galli, M., Caliani, I., Campani, T., Finoia, M.G., Panti, C., 2018. Abundance
 and characterization of microplastics in the coastal waters of Tuscany (Italy): The application of
 the MSFD monitoring protocol in the Mediterranean Sea. Mar. Pollut. Bull. 133, 543–552.
 https://doi.org/10.1016/j.marpolbul.2018.06.016
- Barnes, D.K., Galgani, F., Thompson, R.C., Barlaz, M., 2009. Accumulation and fragmentation of
 plastic debris in global environments. Philos. Trans. R. Soc. London. Ser. B, 364, 1985-1998.
 https://doi.org/10.1098/rstb.2008.0205
- Bellas, J., Martínez-Armental, J., Martínez-Cámara, A., Besada, V., Martínez-Gómez, C., 2016.
 Ingestion of microplastics by demersal fish from the Spanish Atlantic and Mediterranean coasts.
 Mar. Pollut. Bull. 109, 55–60. https://doi.org/10.1016/j.marpolbul.2016.06.026
- Blašković, A., Fastelli, P., Čižmek, H., Guerranti, C., Renzi, M., 2017. Plastic litter in sediments
 from the Croatian marine protected area of the natural park of TelašČica bay (Adriatic Sea).
 Mar. Pollut. Bull. 114, 583–586. https://doi.org/10.1016/j.marpolbul.2016.09.018
- Cannas, S., Fastelli, P., Guerranti, C., Renzi, M., 2017. Plastic litter in sediments from the coasts
 of south Tuscany (Tyrrhenian Sea). Mar. Pollut. Bull. 119, 372–375.
 https://doi.org/10.1016/j.marpolbul.2017.04.008
- Carson, H.S., Colbert, S.L., Kaylor, M.J., McDermid, K.J., 2011. Small plastic debris changes water
 movement and heat transfer through beach sediments. Mar. Pollut. Bull. 62, 1708–1713.
 https://doi.org/10.1016/j.marpolbul.2011.05.032

Catarino, A.I., Macchia, V., Sanderson, W.G., Thompson, R.C., Henry, T.B., 2018. Low levels of
microplastics (MP) in wild mussels indicate that MP ingestion by humans is minimal compared
to exposure via household fibres fallout during a meal. Environ. Pollut. 237, 675–684.
https://doi.org/10.1016/j.envpol.2018.02.069

Cincinelli, A., Martellini, T., Guerranti, C., Scopetani, C., Chelazzi, D., Giarrizzo, T., 2019. A
potpourri of microplastics in the sea surface and water column of the Mediterranean Sea. TrAC Trends Anal. Chem. 110, 321–326. https://doi.org/10.1016/j.trac.2018.10.026

Collard, F., Gilbert, B., Eppe, G., Parmentier, E., Das, K., 2015. Detection of Anthropogenic
Particles in Fish Stomachs: An Isolation Method Adapted to Identification by Raman
Spectroscopy. Arch. Environ. Contam. Toxicol. 69, 331–339. https://doi.org/10.1007/s00244015-0221-0

- Collignon, A., Hecq, J.H., Galgani, F., Collard, F., Goffart, A., 2014. Annual variation in neustonic
 micro- and meso-plastic particles and zooplankton in the Bay of Calvi (Mediterranean-Corsica).
 Mar. Pollut. Bull. 79, 293–298. https://doi.org/10.1016/j.marpolbul.2013.11.023
- 518 Compa, M., Ventero, A., Iglesias, M., Deudero, S., 2018. Ingestion of microplastics and natural 519 fibres in Sardina pilchardus (Walbaum, 1792) and Engraulis encrasicolus (Linnaeus, 1758) along 520 the Spanish Mediterranean Mar. Pollut. Bull. 128, 89-96. coast. 521 https://doi.org/10.1016/j.marpolbul.2018.01.009
- Constant, M., Kerherve, P., Sola, J., Sanchez-Vidal, A., Canals, M., Heussner, S., 2018. Floating
 microplastics in the Northwestern Mediterranean Sea: temporal and spatial heterogeneities. In
 Proceedings of the International Conference on Microplastic Pollution in the Mediterranean Sea
 (p. 9-15). Springer, Cham. https://doi.org/10.1007/978-3-319-71279-6_2
- 526 Constant, M., Kerhervé, P., Mino-Vercellio-Verollet, M., Dumontier, M., Sànchez Vidal, A.,
- 527 Canals, M., Heussner, S., 2019. Beached microplastics in the Northwestern Mediterranean Sea.
- 528 Mar. Pollut. Bull. 142, 263–273. https://doi.org/10.1016/j.marpolbul.2019.03.032

- Constantino, E., Martins, I., Salazar Sierra, J.M., Bessa, F., 2019. Abundance and composition of
 floating marine macro litter on the eastern sector of the Mediterranean Sea. Mar. Pollut. Bull.
 138, 260–265. https://doi.org/10.1016/j.marpolbul.2018.11.008
- Cózar, A., Sanz-Martín, M., Martí, E., González-Gordillo, J.I., Ubeda, B., Á.gálvez, J., Irigoien, X.,
 Duarte, C.M., 2015. Plastic accumulation in the mediterranean sea. PLoS One 10, 1–12.
 https://doi.org/10.1371/journal.pone.0121762
- de Lucia, G.A., Caliani, I., Marra, S., Camedda, A., Coppa, S., Alcaro, L., Campani, T., Giannetti,
 M., Coppola, D., Cicero, A.M., Panti, C., Baini, M., Guerranti, C., Marsili, L., Massaro, G., Fossi,
 M.C., Matiddi, M., 2014. Amount and distribution of neustonic micro-plastic off the western
 Sardinian coast (Central-Western Mediterranean Sea). Mar. Environ. Res. 100, 10–16.
 https://doi.org/10.1016/j.marenvres.2014.03.017
- 540 Deudero, S., Alomar, C., 2015. Mediterranean marine biodiversity under threat: Reviewing 541 influence of marine litter Pollut. Bull. 98, 58-68. on species. Mar. https://doi.org/10.1016/j.marpolbul.2015.07.012 542
- Digka, N., Tsangaris, C., Torre, M., Anastasopoulou, A., Zeri, C., 2018. Microplastics in mussels
 and fish from the Northern Ionian Sea. Mar. Pollut. Bull. 135, 30–40.
 https://doi.org/10.1016/j.marpolbul.2018.06.063
- Duncan, E.M., Arrowsmith, J., Bain, C., Broderick, A.C., Lee, J., Metcalfe, K., Pikesley, S.K., Snape,
 R.T.E., van Sebille, E., Godley, B.J., 2018. The true depth of the Mediterranean plastic problem:
 Extreme microplastic pollution on marine turtle nesting beaches in Cyprus. Mar. Pollut. Bull.
 136, 334–340. https://doi.org/10.1016/j.marpolbul.2018.09.019
- El-Hoz, M., 2019. A Qualitative–Quantitative Methodological Approach for Sustainable
 Reclamation of Open Dumps: The Case of the Controlled Dump of Tripoli. In Waste Valorisation
 and Recycling (p. 105-118). Springer, Singapore. https://doi.org/10.1007/978-981-13-2784-1_10
- 553 Ellen MacArthur Foundation, 2016. The New Plastics Economy- Rethinking the Future of Plastics,
- p. 1-120. https://www.ellenmacarthurfoundation.org/publications/the-new-plastics-economy-
- 555 rethinking-the-future-of-plastics

Fanini, L., Bozzeda, F., 2018. Dynamics of plastic resin pellets deposition on a microtidal sandy
beach: Informative variables and potential integration into sandy beach studies. Ecol. Indic. 89,
309–316. https://doi.org/10.1016/j.ecolind.2018.02.027

Fastelli, P., Blašković, A., Bernardi, G., Romeo, T., Čižmek, H., Andaloro, F., Russo, G.F.,
Guerranti, C., Renzi, M., 2016. Plastic litter in sediments from a marine area likely to become
protected (Aeolian Archipelago's islands, Tyrrhenian sea). Mar. Pollut. Bull. 113, 526–529.
https://doi.org/10.1016/j.marpolbul.2016.08.054

- Filgueiras, A. V., Gago, J., Campillo, J. A., León, V. M., 2019. Microplastic distribution in surface
 sediments along the Spanish Mediterranean continental shelf. Environ. Sci. Pollut. Res. 1-10.
 https://doi.org/10.1007/s11356-019-05341-5
- Fossi, M.C., Panti, C., Guerranti, C., Coppola, D., Giannetti, M., Marsili, L., Minutoli, R., 2012. Are
 baleen whales exposed to the threat of microplastics? A case study of the Mediterranean fin
 whale (*Balaenoptera physalus*). Mar. Pollut. Bull. 64, 2374–2379.
 https://doi.org/10.1016/j.marpolbul.2012.08.013
- Fossi, M.C., Marsili, L., Baini, M., Giannetti, M., Coppola, D., Guerranti, C., Caliani, I., Minutoli, R.,
 Lauriano, G., Finoia, M.G., Rubegni, F., Panigada, S., Bérubé, M., Urbán Ramírez, J., Panti, C.,
 2016. Fin whales and microplastics: The Mediterranean Sea and the Sea of Cortez scenarios.
 Environ. Pollut. 209, 68–78. https://doi.org/10.1016/j.envpol.2015.11.022
- 574 Giani, D., Baini, M., Galli, M., Casini, S., Fossi, M.C., 2019. Microplastics occurrence in edible fish species (Mullus barbatus and Merluccius merluccius) collected in three different geographical 575 576 sub-areas of the Mediterranean Sea. Mar. Pollut. Bull. 140, 129-137. https://doi.org/10.1016/j.marpolbul.2019.01.005 577
- 578 GOEDICKE, T. R., 1972. Submarine canyons on the central continental shelf of Lebanon. The 579 Mediterranean Sea: Natural Sedimentation Laboratory, DJ Stanley Ed., Dowen, Hutchington & 580 Ross, Stroudsburg, PA, 800p.
- 581 Gomiero, A., Strafella, P., Øysæd, K.B., Fabi, G., 2019. First occurrence and composition 582 assessment of microplastics in native mussels collected from coastal and offshore areas of the

northern and central Adriatic Sea. Environ. Sci. Pollut. Res. https://doi.org/10.1007/s11356-01905693-y

- 585 Gündoğdu, S., 2017. High level of micro-plastic pollution in the Iskenderun Bay NE Levantine 586 coast of Turkey. Ege J. Fish. Aquat. Sci. 34, 401–408. 587 https://doi.org/10.12714/egejfas.2017.34.4.06
- 588 Gündoğdu, S., Çevik, C., 2017. Micro- and mesoplastics in Northeast Levantine coast of Turkey: 589 The preliminary results from surface samples. Mar. Pollut. Bull. 118, 341–347. 590 https://doi.org/10.1016/j.marpolbul.2017.03.002
- 591 Güven, O., Gökdağ, K., Jovanović, B., Kıdeyş, A.E., 2017. Microplastic litter composition of the 592 Turkish territorial waters of the Mediterranean Sea, and its occurrence in the gastrointestinal 593 tract of fish. Environ. Pollut. 223, 286–294. https://doi.org/10.1016/j.envpol.2017.01.025
- He, P., Chen, L., Shao, L., Zhang, H., Lü, F., 2019. Municipal solid waste (MSW) landfill: A source
 of microplastics? -Evidence of microplastics in landfill leachate. Water Res. 159, 38–45.
 https://doi.org/10.1016/j.watres.2019.04.060
- Imhof, H.K., Ivleva, N.P., Schmid, J., Niessner, R., Laforsch, C., 2013. Contamination of beach
 sediments of a subalpine lake with microplastic particles. Curr. Biol. 23, R867–R868.
 https://doi.org/10.1016/j.cub.2013.09.001
- Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law,
 K.L., 2015. Plastic waste inputs from land into the ocean. Science, 347, 768-771,
 https://doi.org/10.1126/science.1260352
- Kaberi, H., Tsangaris, C., Zeri, C., Mousdis, G., Papadopoulos, A., Streftaris, N., 2013.
 Microplastics along the shoreline of a Greek island (Kea isl., Aegean Sea): types and densities in
 relation to beach orientation, characteristics and proximity to sources. Proc. 4th Int. Conf.
 Environ. Manag. Eng. Plan. Econ. SECOTOX Conf. 197–202.
- Karkanorachaki, K., Kiparissis, S., Kalogerakis, G.C., Yiantzi, E., Psillakis, E., Kalogerakis, N., 2018.
 Plastic pellets, meso- and microplastics on the coastline of Northern Crete: Distribution and

609organicpollution.Mar.Pollut.Bull.133,578–589.610https://doi.org/10.1016/j.marpolbul.2018.06.011

Kazour, M., Terki, S., Rabhi, K., Jemaa, S., Khalaf, G., Amara, R., 2019. Sources of microplastics
pollution in the marine environment: Importance of wastewater treatment plant and coastal
landfill. Mar. Pollut. Bull. 146, 608–618. https://doi.org/10.1016/j.marpolbul.2019.06.066

Khalil, C., Al Hageh, C., Korfali, S., Khnayzer, R.S., 2018. Municipal leachates health risks:
Chemical and cytotoxicity assessment from regulated and unregulated municipal dumpsites in
Lebanon. Chemosphere 208, 1–13. https://doi.org/10.1016/j.chemosphere.2018.05.151

Laglbauer, B.J.L., Franco-Santos, R.M., Andreu-Cazenave, M., Brunelli, L., Papadatou, M.,
Palatinus, A., Grego, M., Deprez, T., 2014. Macrodebris and microplastics from beaches in
Slovenia. Mar. Pollut. Bull. 89, 356–366. https://doi.org/10.1016/j.marpolbul.2014.09.036

Lebreton, L.C.M., Greer, S.D., Borrero, J.C., 2012. Numerical modelling of floating debris in the world's oceans. Mar. Pollut. Bull. 64, 653–661. https://doi.org/10.1016/j.marpolbul.2011.10.027

Lefebvre, C., Saraux, C., Heitz, O., Nowaczyk, A., Bonnet, D., 2019. Microplastics FTIR 622 characterisation and distribution in the water column and digestive tracts of small pelagic fish in 623 624 the Gulf of Lions. Mar. Pollut. Bull. 142, 510-519. https://doi.org/10.1016/j.marpolbul.2019.03.025 625

Lots, F.A.E., Behrens, P., Vijver, M.G., Horton, A.A., Bosker, T., 2017. A large-scale investigation
of microplastic contamination: Abundance and characteristics of microplastics in European
beach sediment. Mar. Pollut. Bull. 123, 219–226.
https://doi.org/10.1016/j.marpolbul.2017.08.057

Lusher, A.L., O'Donnell, C., Officer, R., O'conner, I., 2016. Microplastic interactions with North
Atlantic mesopelagic fish. ICES J. Mar. Sci. 73, 1214–1225.
https://doi.org/10.1093/icesjms/fsv241

Mansui, J., Molcard, A., Ourmières, Y., 2015. Modelling the transport and accumulation of
floating marine debris in the Mediterranean basin. Mar. Pollut. Bull. 91, 249–257.
https://doi.org/10.1016/j.marpolbul.2014.11.037

23

- Menna, M., Poulain, P.M., Zodiatis, G., Gertman, I., 2012. On the surface circulation of the
 Levantine sub-basin derived from Lagrangian drifters and satellite altimetry data. Deep. Res.
 Part I Oceanogr. Res. Pap. 65, 46–58. https://doi.org/10.1016/j.dsr.2012.02.008
- Moritomi, H., Iwase, T., Chiba, T., 1982. A comprehensive interpretation of solid layer inversion
 in liquid fluidised beds. Chem. Eng. Sci. 37, 1751–1757. https://doi.org/10.1016/00092509(82)80047-X
- Munari, C., Scoponi, M., Mistri, M., 2017. Plastic debris in the Mediterranean Sea: Types,
 occurrence and distribution along Adriatic shorelines. Waste Manag. 67, 385–391.
 https://doi.org/10.1016/j.wasman.2017.05.020
- Nasreddine, L., Hwalla, N., Sibai, A., Hamzé, M., Parent-Massin, D., 2006. Food consumption
 patterns in an adult urban population in Beirut, Lebanon. Public Health Nutr. 9, 194–203.
 https://doi.org/10.1079/PHN2005855
- Nelms, S.E., Duncan, E.M., Broderick, A.C., Galloway, T.S., Godfrey, M.H., Hamann, M., Lindeque,
- 649 P.K., Godley, B.J., 2016. Plastic and marine turtles: a review and call for research. ICES J. Mar.
- 650 Sci. 73, 165–181. https://doi.org/10.1093/icesjms/fsv165
- 651 O'Connor, I.A., Golsteijn, L., Hendriks, A.J., 2016. Review of the partitioning of chemicals into
- different plastics: Consequences for the risk assessment of marine plastic debris. Mar. Pollut.
- 653 Bull. 113, 17–24. https://doi.org/10.1016/j.marpolbul.2016.07.021
- Panti, C., Giannetti, M., Baini, M., Rubegni, F., Minutoli, R., Fossi, M. C., 2015. Occurrence,
 relative abundance and spatial distribution of microplastics and zooplankton NW of Sardinia in
 the Pelagos Sanctuary Protected Area, Mediterranean Sea. Environ. Chem. 12(5), 618-626.
 https://doi.org/10.1071/EN14234
- 658 Pedrotti, M.L., Petit, S., Elineau, A., Bruzaud, S., Crebassa, J.C., Dumontet, B., Martí, E., Gorsky,
- 659 G., Cózar, A., 2016. Changes in the floating plastic pollution of the mediterranean sea in relation
- to the distance to land. PLoS One 11, 1–14. https://doi.org/10.1371/journal.pone.0161581

- Piperagkas, O., Papageorgiou, N., Karakassis, I., 2019. Qualitative and quantitative assessment
 of microplastics in three sandy Mediterranean beaches, including different methodological
 approaches. Estuar. Coast. Shelf Sci. 219, 169–175. https://doi.org/10.1016/j.ecss.2019.02.016
- 664 Phuong, N.N., Zalouk-Vergnoux, A., Kamari, A., Mouneyrac, C., Amiard, F., Poirier, L., Lagarde, F.,
- 665 2018. Quantification and characterization of microplastics in blue mussels (*Mytilus edulis*):
- 666 protocol setup and preliminary data on the contamination of the French Atlantic coast. Environ.
- 667 Sci. Pollut. Res. 25, 6135–6144. https://doi.org/10.1007/s11356-017-8862-3
- PlasticsEurope, 2018. Plastics the Facts 2017, An Analysis of European Plastics Production,
 Demand and Waste Data, p. 16, (Brussels Belgium)
- Poeta, G., Battisti, C., Acosta, A.T.R., 2014. Marine litter in Mediterranean sandy littorals: Spatial
 distribution patterns along central Italy coastal dunes. Mar. Pollut. Bull. 89, 168–173.
 https://doi.org/10.1016/j.marpolbul.2014.10.011
- Praagh, M. van, Hartman, C., Brandmyr, E., 2018. Microplastics in Landfill Leachates in the
 Nordic Countries. https://doi.org/10.6027/TN2018-557
- a) Renzi, M., Guerranti, C., Blašković, A., 2018. Microplastic contents from maricultured and
 natural mussels. Mar. Pollut. Bull. 131, 248–251.
 https://doi.org/10.1016/j.marpolbul.2018.04.035
- b) Renzi, M., Blašković, A., Bernardi, G., Russo, G.F., 2018. Plastic litter transfer from sediments
- towards marine trophic webs: A case study on holothurians. Mar. Pollut. Bull. 135, 376–385.
- 680 https://doi.org/10.1016/j.marpolbul.2018.07.038
- c) Renzi, M., Blašković, A., Fastelli, P., Marcelli, M., Guerranti, C., Cannas, S., Barone, L., Massara,
 F., 2018. Is the microplastic selective according to the habitat? Records in amphioxus sands,
 Mäerl bed habitats and *Cymodocea nodosa* habitats. Mar. Pollut. Bull. 130, 179–183.
 https://doi.org/10.1016/j.marpolbul.2018.03.019
- a) Renzi, M., Specchiulli, A., Blašković, A., Manzo, C., Mancinelli, G., Cilenti, L., 2019. Marine
 litter in stomach content of small pelagic fishes from the Adriatic Sea: sardines (*Sardina*)

- *pilchardus*) and anchovies (*Engraulis encrasicolus*). Environ. Sci. Pollut. Res. 26, 2771–2781.
 https://doi.org/10.1007/s11356-018-3762-8
- b) Renzi, M., Čižmek, H., Blašković, A., 2019. Marine litter in sediments related to ecological
- 690 features in impacted sites and marine protected areas (Croatia). Mar. Pollut. Bull. 138, 25–29.
- 691 https://doi.org/10.1016/j.marpolbul.2018.11.030
- Rios-Fuster, B., Alomar, C., Compa, M., Guijarro, B., Deudero, S., 2019. Anthropogenic particles
 ingestion in fish species from two areas of the western Mediterranean Sea. Mar. Pollut. Bull.
 144, 325–333. https://doi.org/10.1016/j.marpolbul.2019.04.064
- Romeo, T., Pietro, B., Pedà, C., Consoli, P., Andaloro, F., Fossi, M.C., 2015. First evidence of
 presence of plastic debris in stomach of large pelagic fish in the Mediterranean Sea. Mar. Pollut.
 Bull. 95, 358–361. https://doi.org/10.1016/j.marpolbul.2015.04.048
- Ruiz-Orejón, L.F., Sardá, R., Ramis-Pujol, J., 2016. Floating plastic debris in the Central and
 Western Mediterranean Sea. Mar. Environ. Res. 120, 136–144.
 https://doi.org/10.1016/j.marenvres.2016.08.001
- Schmidt, N., Thibault, D., Galgani, F., Paluselli, A., Sempéré, R., 2018. Occurrence of
 microplastics in surface waters of the Gulf of Lion (NW Mediterranean Sea). Prog. Oceanogr.
 163, 214–220. https://doi.org/10.1016/j.pocean.2017.11.010
- Shiber, J.G., 1979. Plastic pellets on the coast of Lebanon. Mar. Pollut. Bull. 10, 28–30.
 https://doi.org/10.1016/0025-326X(79)90321-7
- Suaria, G., Avio, C.G., Mineo, A., Lattin, G.L., Magaldi, M.G., Belmonte, G., Moore, C.J., Regoli, F.,
- Aliani, S., 2016. The Mediterranean Plastic Soup: Synthetic polymers in Mediterranean surface
 waters. Sci. Rep. 6, 1–10. https://doi.org/10.1038/srep37551
- Thompson, R.C., Olson, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., McGonigle, D.,
 Russell, A.E., 2004. Lost at Sea: Where Is All the Plastic? Science 304, 838.
 https://doi.org/10.1126/science.1094559

- Turner, A., Holmes, L., 2011. Occurrence, distribution and characteristics of beached plastic
 production pellets on the island of Malta (central Mediterranean). Mar. Pollut. Bull. 62, 377–
 381. https://doi.org/10.1016/j.marpolbul.2010.09.027
- UNEP (2014) UNEP year book 2014. Emerging issues update. Chapter 8: plastic debris in the
 ocean. ISBN: 978-92-807-3381-5
- Van Cauwenberghe, L., Janssen, C.R., 2014. Microplastics in bivalves cultured for human
 consumption. Environ. Pollut. 193, 65–70. https://doi.org/10.1016/j.envpol.2014.06.010
- van der Hal, N., Ariel, A., Angel, D.L., 2017. Exceptionally high abundances of microplastics in the
- 720 oligotrophic Israeli Mediterranean coastal waters. Mar. Pollut. Bull. 116, 151–155.
- 721 https://doi.org/10.1016/j.marpolbul.2016.12.052



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



Figure 2: A) Concentration of suspected microplastics (MPs/m3) 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 (±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.



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



■ PP ■ PE ■ PS ■ PA ■ PUR ■ ABS ■ PET ■ PVC ■ PLA

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.

A)

B)

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 m³ are presented.

Location	on Sampling nets		Identification method	MPs abundances	Reference	
NW Mediterranean (the Rhône	Manta traul not	222	Storeomieroscono /FTID	0.10.0.10 items (m ³	Constant et al.,	
River and the Têt River)	Manta trawi net	333	stereomicroscope/Filk	0.18- 0.19 items/m	(2018)	
NW Mediterranean Sea (Ligurian	WP2 standard net	200	Stereomicroscope	0.62 + 2 items/m ³	Fossi et al. (2012)	
Sea and Sardinian Sea)	Wi 2 Standard Het	200	Stereonneroscope	0.02 ± 2 ncm3/m	10331 Ct al., (2012)	
NW Mediterranean Sea (Gulf of	M/D2 mlambdam mat	200		0.23 ± 0.20	Lefebvre et al.,	
Lion)	WP2 plankton net		stereomicroscope/Filk	items/m ³	(2019)	
NW Mediterranean Sea (Gulf of	Nouston not	200	Stereomicroscope	0.31 ± 1.17	Fossi et al., (2016)	
Asinara and the Sardinian Sea)	Neusion net			items/m ³		
NW Mediterranean Sea (North	WP2 standard ring net	200	Stereomicroscope	0.17 ± 0.32	Panti et al., (2015)	

Western of Sardinia; Gulf of				items/m ³	
Asinara)					
NW/ Moditorrangan Soa (Tuscany					
in the international sea (ruscany	Manta trawl net	330	Stereomicroscope/FTIR	0.26/m ³	Baini et al., (2018)
coastal water)					
Central-Western Mediterranean		500	Binocular		De Lucia et al.,
Sea (Sardinian Sea)	Manta trawl net	500	stereomicroscope	0.15 items/m ³	(2014)
Eastern Mediterranean (Turkish			Microscopic		
Mediterranean (oast)	Manta trawl net	333	observation/FTIR on 24	0.7 items/m ³	Güven et al., (2017)
meanenanean cousty			items		
Eastern Mediterranean	Manta trawl net	333	Microscopic observation	7.26 items/m ³	Gündoğdu, (2017)
(Iskenderun Bay)					
Eastern Mediterranean Sea					Gündoğdu and
(Ickondorun Ray and Morein Pay)	Manta Trawl net	333	Microscopic observation	2.73 items /m ³	Cevik (2017)
(iskeliaeran bay and wersin bay)					ζCMN, (2017)

South Eastern Mediterranean Sea	Manta Travil nat	222	Stereomicroscope	7.68 ± 2.38	van der Hal et al.,
(Israeli surface waters)	Manta Trawi net	333		items/m ³	(2017)
Lebanese coast	Manta Trawl net	52	Stereomicroscope/Raman	4.3 ± 2.2 items/m ³	Present study

Table 2: Average (±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 Identificatio method	on Main polymer	Reference
Spanish Western Mediterranean coasts	None. Visual observation	0.07 (±0.35) to 0.33 (±0.26)	15%	FTIR	PET	Compa et al., (2018)
Western Mediterranean Sea (Iberian Peninsula coast and the Balearic Islands)	None Visual observation	0.07 ± 0.26	2.56%	None	-	Rios-Fuster et al., (2019)
North Western of the Mediterranean Sea (Gulf of Lions)	None. Visual observation	0.11 ± 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 ₂ O ₂	1.25	91%	μ-FT-IR	PVC	Renzi et al., (2019a)
Eastern Mediterranean Sea (Lebanese coast)	KOH 10%	2.5 ± 0.3	83.4%	Raman Spectroscopy	PS	Present study

