

Journal Pre-proof

Target gastropods for standardizing the monitoring of tar mat contamination in the Arabian Gulf

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PII: S2352-4855(22)00086-X
DOI: <https://doi.org/10.1016/j.rsma.2022.102328>
Reference: RSMA 102328

To appear in: *Regional Studies in Marine Science*

Received date: 1 November 2021
Revised date: 4 March 2022
Accepted date: 18 March 2022

Please cite this article as: B.W. Giraldes, J.A.K.H. Al-Thani, S. Dib et al., Target gastropods for standardizing the monitoring of tar mat contamination in the Arabian Gulf. *Regional Studies in Marine Science* (2022), doi: <https://doi.org/10.1016/j.rsma.2022.102328>.

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1 **Target gastropods for standardizing the monitoring of**
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4 **tar mat contamination in the Arabian Gulf**

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Abstract:

This study proposes a standardization process for the monitoring the level of tar mat contamination in the Arabian Gulf. In the study, we selected target gastropod species and evaluated the ecological and ecotoxicological effects of tar mats on their populations, the bioaccumulation of heavy metals, and these species' potential as bioindicator taxa for tar mat-associated heavy metals. The study was carried out at two sites on the Qatar coast on shoreline rocks, both of which have areas of tar mat coverage and areas without tar mats. The species selected as representative species for the ecoregion were the endemic *Clypeomorus bifasciata persica* (Houbrick, 1985) in the intertidal zone and the *Echinolittorina arabica* (El Assal, 1990) in the supratidal zone. Both are grazers and are niche restricted to hard substrates and daily scraping/grazing on the tar mat deposition zone of the shorelines. The key heavy metal indicators chosen were As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Ni and Zn, and the gastropod species were divided into classes according to their size. Tar mat presence was found to negatively affect both the overall population size of *Clypeomorus* and different size classes, and lead to high levels of assimilation of heavy metals. Tar mat presence also negatively affected different size classes of *Echinolittorina*, where large sizes were found to be most significantly affected. The results also demonstrated that Cu and Ni are the heavy metals most associated with tar mat contamination of this kind. Overall, our results confirmed that grazer gastropods are good bioindicators of tar mat-associated contaminants in shoreline ecosystems. Our study provides the database on the heavy metal contamination of the proposed target gastropods, and offers information that will be relevant for further monitoring and comparisons among threatened coastal areas in the Arabian Gulf.

Keywords: Tarmat, Biomonitoring, Heavy metals, *Echinolittorina arabica*, *Clypeomorus bifasciata persica*, Arabian Gulf

1. Introduction:

Among the human influences recorded in the Anthropocene, oil spills deserve particular

1 48 attention for their potential to contaminate coastal ecosystems in the long run (Darwish and
2 49 Mohtar, 2013; Sheppard et al., 2010; Smith and Zeder, 2013). In the Arabian Gulf, the 1991
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4 50 Gulf War oil spill represents the main environmental disaster affecting the entire marine
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6 51 ecosystem and associated biodiversity in this semi-enclosed sea (Gerges, 1993; Jones et al.,
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8 52 1998; Price, 1998; Vaughan et al., 2019). After the initial deleterious impact of a spill on natural
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10 53 biodiversity, oil begins to dehydrate and forms ‘tar mats’ on the shoreline of the affected region
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12 54 (Arekhi et al., 2020; Veerasingam et al., 2020). Given its physical and chemical properties, tar
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14 55 mat adheres to hard substrates (rocks) in the supratidal and intertidal zones of the shoreline
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16 56 (Rajendran et al., 2021). The exact toxicity of spilled oil (consisting of aliphatic and
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18 57 aromatic hydrocarbons) to marine organisms depends on the persistence and bioavailability of
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20 58 specific hydrocarbons. The formation and deposition of tar mats along the Qatar coast, caused
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22 59 by the 1991 oil spill (Rajendran et al., 2021), have been determined by weathering processes
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24 60 such as evaporation, emulsification, dissolution, oxidation, horizontal and vertical movement,
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26 61 and sedimentation.
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35 62 It is possible, therefore, that the toxic chemicals from the tar mats may have reduced over the
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37 63 past three decades, but may still be contaminating the environment and associated flora and
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39 64 fauna for a long time. The matter of ‘how long’ is a question that needs to be resolved.
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43 65 On shorelines around the world, different species have evolved to be able to inhabit specific
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45 66 coastal zones through evolutionary specialization, where isolated congeners are recorded in
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47 67 niche equivalence inhabiting the same zone, but within different ecoregions (Al-khayat and
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49 68 Giraldes, 2020; Spalding et al., 2007). Amongst coastal marine fauna, gastropod assemblages
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51 69 merit particular focus for their high diversity and niche restriction, with groups of species
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53 70 inhabiting specifically hard or soft substrates, and with assemblages living in supratidal,
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55 71 intertidal or subtidal zones (Bosch et al., 1995). Moreover, species which have physiologically
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57 72 and anatomically evolved to inhabit supratidal or intertidal sites may exhibit desiccation
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1 73 avoidance (Reid et al., 2010, 2007). The radula (mouth parts) are adapted to scrape the hard
2 74 substrate, allowing the animals to graze on the organic layer that covers the hard substrate in
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4 75 each tidal zone (Amini-Yekta et al., 2019; Houbrick, 1985; Ronald Janssen et al., n.d.). As a
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7 76 result, tar mat contamination can be expected to have more significant long-term impacts on
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10 77 grazers in niches restricted to hard substrates in supratidal and high-intertidal sites, because
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12 78 those species inhabit rocks with tar mats and feeding on it on a daily basis.
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15 79 Gastropods are recognized as valuable for their ability to serve as bioindicators of
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17 80 anthropogenic impacts. For this reason, they are commonly used in environmental assessment
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20 81 programmes and protocols (Smith, 2005), including monitoring of environmental pollution
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22 82 (Baroudi et al., 2020) and heavy metal accumulation (Astani et al., 2012; Krupnova et al.,
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24
25 83 2018). Assessments are usually based on ecological data and indices of abundance and
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27 84 diversity, in which a variation in abundance of target species may point to a specific
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30 85 anthropogenic pressure (Smith, 2005). Tar mats may affect gastropods by acting as a source of
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32 86 pollutants, which will accumulate in their bodies for a significant period. Heavy metals are
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35 87 considered to be major inorganic pollutants for marine ecosystems because of their persistence,
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37 88 bioaccumulation and natural bio-transfer from low to high trophic levels in the food chain (Bo
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40 89 et al., 2015; Krupnova et al., 2018). These metals pose a direct threat, and may negatively
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42 90 impact aquatic organisms and entire ecosystems with their high toxicity, carcinogenicity, non-
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45 91 biodegradability, persistence and bioaccumulation properties (Feng et al., 2017). Furthermore,
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47 92 tar mats from oil spills are major sources of bio-concentrated heavy metals (Amin et al., 2009;
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50 93 Humood A. Naser, 2013). These contaminants can enter the trophic chain in coastal
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52 94 environments through contaminated water, and pass into soft tissue (e.g. into the gills during
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55 95 respiration) or trophic fractions (e.g. sediment and substrates), contaminating basic trophic
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57 96 levels and transferring to upper trophic levels (Baroudi et al., 2020). As the last consumer in
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59 97 the food chain, the contaminants reach humans when they consume contaminated organisms
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98 which have ingested gastropods, either directly or indirectly, through such means as trophic
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 43 44 45 46 47 48 49 50 51 52 53 54 55 56 57 58 59 60 61 62 63 64 65

99 transfer to upper levels of the food web (Gawad, 2018; Yüzereroğlu et al., 2010a). It is therefore
100 safe to assume that grazer gastropods which scrape hard substrates on tar mat-contaminated
101 shorelines during feeding could be a practical target for monitoring heavy metal accumulation.
102 In the Arabian Gulf, multiple oil spills have been reported in recent decades (Al Maslamani et
103 al., 2018; Arekhi et al., 2020; Darwish and Mohtar, 2013; Sheppard et al., 2010). Consequently,
104 we can expect tar mats to be present along specific stretches of the Qatar coast, covering the
105 hard substrate of the supratidal and intertidal shorelines. In the present study, we evaluated the
106 ecological effect of tar mats on populations of common grazer gastropod species in intertidal
107 and subtidal shoreline zones, and assessed the heavy metal contaminants which had
108 accumulated in those species. The aim of this study is to provide a standardization process for
109 monitoring the level of tar mat contamination in the Arabian Gulf. Selecting target tar-
110 associated heavy metals and selecting dominant and abundant gastropod species in the Gulf
111 which leaves and feed on coastal areas where tar is deposited.

112 **2. Materials and Methods:**

113 **2.1. Selection of Target Gastropod Species:**

114 In conducting our literature review, we sought suitable target gastropods which met the
115 following requirements: 1) ecological zoning on supratidal and high-intertidal levels, the
116 shoreline areas where tar mats are deposited; 2) inhabitants of hard-substrate, the main
117 substrate where tar mats stick; 3) grazer on hard substrates as feeding behaviour,
118 eating/scraping the micro-organic layer which covers the rocks and tar mats; 4) species which
119 are abundant, for easy location on shorelines; 5) species which are widespread in the Gulf and
120 north-eastern Indian Ocean, allowing for standardized use of the same species in the Arabian
121 Gulf.

2.2. Biological Field Sampling:

We selected the following two sampling sites on the Qatar coast: Al-Khor (25°40'02.8"N, 51°32'11.6"E), in the mangrove ecosystem and Al-Ruwais (25°08'50.7"N, 51°11'15.6"E), near a coral reef ecosystem (Figure 1A). Each sampling site contained hard substrates with tar mat (Figure 1D, E) and without tar mat (Figure 1F, G), in both the supratidal and intertidal zones. Highlighting that tar mats are irregularly speeded on the rocks and the areas with high concentration of tar mat was considered as the contaminated samples and the rocks without tar coverage considered as the control samples. In March 2020, 10 quadrats (each 0.5 m²) were placed randomly on the hard substrate in each zone (supratidal, intertidal) (10 on tar mat and 10 on the "clean" rocks) at the two studied sites during low tide (Figure 1B, C), resulting in a total of 40 quadrats. Areas with tar and without tar they were more than 50 meters apart avoiding the bias regarding the displacement of the grazer species. Photos were taken in each quadrat for evaluation of the tar mat coverage (Figure 1D-G). All gastropods of the selected target species within the quadrats were recorded, collected using plastic or Teflon forceps, and placed in plastic re-sealable Ziploc bags. All samples were kept in an icebox, and transported to the laboratory where they were stored in the freezer at -20 °C.

2.3. Ecological Data Analysis:

Sampled specimens were identified in the laboratory using taxonomic references (Bosch et al., 1995; Houbrick, 1985; Reid et al., 2007) to the lowest taxonomic level (i.e. species). Tar mat coverage was analysed by dividing the photo-quadrats into sub-quadrats using Illustrator software. The tar mat coverage in each sub-quadrat was then evaluated as a percentage (%) of quadrat area. Based on the percentage tar mat coverage in the sub-quadrats, each quadrat was categorized as being with tar mat (1-100%) or without tar mat (0%). All the specimens collected in each quadrat were divided into size classes (large, medium, and small), according

146 to species dimension (specific size per species within each size class is described below). The
147 dominant size class at each site and the percentage of tar mat in each quadrat were compared
148 for each species. The influence of tar mats on the population of each species was established
149 by comparing their abundance in quadrats with tar mat and quadrats without tar mat (total
150 population and population in different size classes).

151 Specimens collected from rocks with tar mat (or species which would presumably graze the
152 microorganism layer that covers the tar mat), and specimens collected on clean rocks without
153 tar mat (or species which would presumably graze the natural microorganism layer, unaffected
154 by tar mat deposits), were separated for the chemical analysis.

2.4. Sample Processing and Chemical Analysis:

156 The gastropod samples were rinsed in double-distilled deionized (DDI) water, in order to
157 remove any particles, and oven-dried at 55 °C for 24 hours. Samples for analysis were created
158 by measuring 1 g portions (dry weight) into Teflon tubes. The dry samples were subjected to
159 strong acid digestion by adding 5 mL of 16M trace-metal grade nitric acid (HNO₃) on a hot
160 block inside a fume hood, at 95 °C for 30 minutes. Initially, the hot block temperature was
161 gradually raised to 135 °C for 1 hour, then to 150 °C until the sample volume reduced to a few
162 drops. 3 mL of HNO₃ and 20 mL of DDI water were then added to all the samples, which were
163 then heated again until the digest volume was reduced to being particle-free, and a clear
164 solution was obtained. The totally digested samples were transferred to acid-cleaned
165 volumetric flasks, and the final sample volume brought up to 50 mL by adding DDI water at
166 room temperature. The digest solutions were transferred to trace element-certified 50 mL
167 sample vials in Ziploc bags, and kept at room temperature until analysis for the key heavy
168 metal indicators arsenic (As), barium (Ba), cadmium (Cd), cobalt (Co), chromium (Cr), copper
169 (Cu), iron (Fe), manganese (Mn), nickel (Ni) and zinc (Zn). These analyses were carried out at

170 the Environmental Science Center Laboratories of Qatar University, which are accredited and
171 certified for ISO17025 analysis by the American Association for Laboratory Accreditation
172 (A2LA). Heavy metal concentrations were determined by means of an inductively coupled
173 plasma-optical emission spectrometer (ICP-OES), using a Perkin Elmer Optima 5300 DV
174 device. Data analysis was performed using Syngistix-ICP software. Duplicate samples, field-
175 sampling replicates, spiked samples and certified reference materials (see below) were digested
176 and analysed twice, for quality control and assurance. Three samples were analysed in
177 duplicate during sample run-off, within the same batch. The correlation coefficient was always
178 greater than 0.9999 for a calibration curve plotted using freshly prepared certified standards of
179 each element. The precision was about 10%. In almost all cases, the average measured values
180 were within the 95% confidence limit of certified values, meaning the accuracy was
181 comparable to, or higher than, the precision. Limit of detection (LoD) and limit of quantitation
182 (LoQ) were also calculated for each element.

183 For quantification of trace metal content, and to ensure the accuracy of these measurements,
184 three certified reference materials (CRM) were included in the analyses: PACS-3, Nist 2976,
185 and Nist 1643f. PACS-3 is the marine sediment reference material for trace metals, collected
186 in Esquimalt by the National Research Council of Canada (NRC), while Nist 2976 is a freeze-
187 dried mussel tissue (trace elements and methylmercury) reference material, and Nist 1643f is
188 a reference material of trace elements in water. The average recovery of all CRM was 102.72,
189 with percentage error within 5% of the expected value for all elements. Heavy metal
190 concentrations in gastropods were reported using $\mu\text{g/g}$ for all samples.

191 **2.5. Statistical Data Analysis:**

192 All chemical data were analyzed using IBM SPSS statistics software (version 28). In order to
193 evaluate the variability of the heavy metals in the selected gastropod species with the existence

194 of tar and with location. Normality of data was investigated using Shapiro-Wilk test, and the
195 heavy metal data did not follow normal distribution. Therefore, data were log transformed
196 before conducting the multivariate analysis (MANOVA). Tukey post-hoc test was performed
197 to analyze the interaction of heavy metals in the gastropod species with location and tar mat
198 existence. We also performed Pearson correlation to compare the significant correlations of the
199 various heavy metals in gastropods with other metals in order to further investigate the trends
200 within the heavy metals and their influence on gastropod population abundance.

201 The tar mat coverage of the rock substrate was presented in percentage illustrated on box plots.
202 Population analysis was performed correlating the abundance of each species (per size classes)
203 with their presence on tar mat. Statistical analyses based on F and T-tests were performed to
204 compare the species in quadrats with tar mat and quadrats on clean rock (without tar), with the
205 significance level set at $p < 0.05$. Significant results were then plotted in boxplots. Secondary
206 data analysis was performed for Cu, Ni and Zn, which are known contaminants in gastropods
207 after oil spills (Bu-Olayan and Subrahmanyam, 1997), and their concentrations evaluated for
208 the entire population of each species. Aiming illustrate the target contaminants effects on the
209 target gastropod species.

210 3. Results

211 3.1. Target species

212 Previous studies have demonstrated the gastropod biodiversity that inhabits coastal zones in
213 the marine ecosystems in the entire Arabian Gulf (Al-Maslamani et al., 2015; Bosch et al.,
214 1995; Houbrick, 1985; Reid et al., 2007). From these, we identified two target species that met
215 the specified requirements. The first was *Echinolittorina arabica* (El Assal, 1990), a species
216 from the *Littorinidae* family, which exhibit high diversity and physiological adaptation for
217 living and grazing on supratidal hard substrates (Reid et al., 2007). The second was

218 *Clypeomorus bifasciata persica* (Houbrick, 1985), from the genus *Clypeomorus* in the family
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2 219 *Cerithiidae* (Houbrick 1983), which are adapted for living and grazing on intertidal hard
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4
5 220 substrates.
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7
8 221 In the study, taxonomic analysis confirmed that the primary species collected in the supratidal
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10 222 zone at the study sites was the endemic *E. arabica* (Figure 2A, B). At the intertidal sites, the hard
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12 223 substrate was completely dominated by *C. bifasciata persica* (Figure 2 C-E). The sizes for *E.*
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14 224 *arabica* in the selected categories were large (>5 mm), medium (2-5 mm) and small (<2 mm);
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16 225 and for *C. bifasciata persica* they were large (>16 mm), medium (9-16 mm) and small (<9 mm).
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21 226 **3.2. Ecological Influence of Tar mats**

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23 227 At both sites (Al-Khor and Al-Ruwais), the supratidal zone had higher coverage of tar mats
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25 228 than the corresponding intertidal zone (figure 3). Quadrats with tar mat had an average of 70%
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27 229 tar mat coverage in the supratidal zone at both sites, and only 5% and 38% tar mat coverage in
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29 230 the intertidal zone at Al-Khor and Al-Ruwais, respectively. Therefore, the influence of tar mat
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31 231 was expected to be more significant for the gastropods in the supratidal zone, which is also
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33 232 more exposed to the atmosphere, to solar radiation, and to heat.
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38 233 A total of 3,856 specimens of *E. arabica* were recorded in quadrats in the supratidal zone, with
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40 234 an average of 120 specimens per m². Regarding the influence of tar mat deposition on *E.*
41
42 235 *arabica*, a total of 1,619 specimens were found in quadrats with tar mat and 2,227 specimens
43
44 236 in quadrats without tar mat, with an average of 101 and 140 specimens per m², respectively
45
46 237 (p=0.369) (Figure 4A). Comparing the proportion of specimens withing each size classes
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48 238 (Figure 4B) the areas with tar mats have more smaller specimens and less larger specimens.
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50 239 Suggesting a recent occurrence of massive recruitment in the populations of this species.
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52 240 Differences between the populations of *E. arabica* in quadrats with and without tar (rock) were
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54 241 mainly found in the larger size classes (Figure 4C), with an average of 4 and 8.4 specimens in
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56 242 quadrats with and without tar mat, respectively (p=0.0515; F=5.14); medium and small
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243 specimens were also more abundant on rocks, but those differences were not significant
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 2 244 ($p=0.102$; $F=1.99$) and ($p=0.499$; $F=1.8$) respectively (Figure 4D,E).

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 5 245 In the intertidal zone 1,347 specimens of *C. bifasciata persica* were recorded, with an average
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 8 246 of 33.7 specimens per m^2 . In comparing *C. bifasciata persica* on tar mat and on rocks without
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 10 247 tar, we found a statistically significant difference ($p=0.0298$; $F=1.8$), with an average of 31
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 12 248 specimens per m^2 tar mat and 83 specimens per m^2 in quadrats without tar (Figure 4F).
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 14 249 Regarding sizes, on rocks with tar we found fewer large and small specimens, with higher
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 16 250 dominance of medium size animals (Figure 4G). In large animals there was an average of 6.2
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 18 251 specimens per m^2 on rocks without tar and 2.3 on tar (Figure 4H) ($p=0.506$; $F=2.5$); on medium
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 20 252 an average of 69.3 specimens per m^2 on rocks without tar and 30.2 on tar (Figure 4I) ($p=0.0592$;
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 22
 23 253 $F=7.2$); and on small sizes, an average of 10.5 specimens per m^2 on rocks without tar and 2.4
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 25
 26 254 on tar (Figure 4J) ($p=0.098$; $F=7.9$).

31 255 **3.3. Heavy Metal Contamination**

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 34 256 For heavy metal contamination analysis, we used and processed a total of 1,339 specimens of
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 36 257 *Echinolittorina* and 262 specimens of *Clypeomorus*. All representative specimens were found
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 38 258 within the 40 quadrats. The concentrations of certain heavy metals in the two target gastropod
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 40 259 species were found to increase with the presence of tar mat in their habitat (Fig. 4), with mean
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 42 260 accumulated metal concentrations in samples decreasing in the order $Zn > Cu > Mn > Ni > Cd$
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 44 261 $> Cr$. In Al-Khor, the mean concentrations of these heavy metals in the gastropod *C. bifasciata*
 45
 46 262 *persica* in tar mat-contaminated areas followed the order $Fe > Zn > Mn > Cu > As > Ni > Ba$
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 48 263 $> Cr > Cd > Co$, while for *E. arabica*, it followed the order $Fe > Zn > Cu > Mn > As > Ba >$
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 50 264 $Ni > Cd > Cr > Co$. In Al-Ruwais, meanwhile, the mean concentrations of the heavy metals in
 51
 52 265 tar mat-contaminated areas followed the order $Fe > Zn > Cu > Mn > As > Ni > Ba > Cr > Cd$
 53
 54 266 $> Co$ for *C. bifasciata persica*, and $Fe > Zn > Cu > As > Mn > Ni > Ba > Cr > Cd > Co$ for *E.*

1 267 *arabica*. The mean concentrations (and standard deviations) of As, Ba, Cd, Co, Cr, Cu, Fe, Mn,
2 268 Ni and Zn are shown in the supplementary material.
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5 269 There were some differences in the concentrations of heavy metals, both between the sampling
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7 270 sites and between quadrats with and without tar mat. The concentrations of Ba, Cd, Cr, Cu, Fe,
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9 271 Mn, Ni and Zn differed according to whether tar mat was present (Figure 5 and supplementary
10
11 272 material). In the gastropod samples, we observed pronounced metal enrichment in the order Zn
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13 273 > Cu > Mn > Ni > Cd > Cr. We also observed a marked difference in concentrations between
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15 274 species in Al-Khor and Al-Ruwais, indicating differential accumulation in the species at the
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17 275 two sites. There were also differences in heavy metal bioaccumulation patterns between the
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19 276 species and sites. Some heavy metals were found in higher concentrations in Al-Khor, and
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21 277 others in Al-Ruwais. Cd, Cr and Zn, for example, were found in higher concentrations in *C.*
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23 278 *bifasciata persica* at Al-Khor, particularly in areas contaminated by tar mat. In tar mat-
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25 279 contaminated areas at Al-Ruwais, Ni was found in higher concentrations in both species.
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33 280 The intertidal *C. bifasciata persica*, in general was found to be the species of gastropods with
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35 281 the most enriched heavy metals found in their bodies, especially in Al-Khor; with high
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37 282 concentrations of As, Cd, Cr, Cu and Zn in their bodies in Al-Khor when they were exposed to
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39 283 tar mats, while they were found to have high concentrations of Cd, Cr, Cu, and Ni in Al-Ruwais
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41 284 (Supplementary material).
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46 285 Correlation analysis was performed on the concentrations of the heavy metals, seeking trends
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48 286 in the concentrations of metals and their association with other metals and the abundance of
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50 287 the gastropod populations, in gastropods which inhabit the tar mat areas. Table 2 shows the
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52 288 ensuing correlation matrix (Pearson) for tar mat-contaminated areas in Al-Khor. Pearson
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54 289 correlation analysis revealed strong and significant relationships between some metals, with
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56 290 different trends at the two sites. At Al-Khor, the metals were mainly positively correlated with
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291 each other. As and Cu ($R^2 = 0.96$), for example, showed a strong correlation ($p < 0.01$) in areas
292 with tar mats, as did Ba, Cd and Fe ($R^2 = 0.97, 0.90, 0.98$). Additionally, the presence of heavy
293 metals in the gastropod bodies was found to have some influence on the gastropod population.
294 This was confirmed by the correlation analysis of heavy metals vs. abundance of the population
295 (Table 3). While only two metals were shown to significantly correlate ($R^2 = 0.9$; $p < 0.05$),
296 which were Cu and Fe, with the population abundances in Al-Khor, none were found in Al-
297 Ruwais. In figure 5 it is highlighted the higher concentration of tar-associated heavy metals Cu
298 and Ni on the target gastropods recorded inhabiting the rocks covered by tar mat.

4. Discussion:

4.1. Heavy Metal Contamination:

302 The sediment and food sources of the targeted grazer gastropods were certainly contaminated
303 with heavy metals due to the presence of petroleum hydrocarbons from tar, which are known
304 to be major sources of specific tar-associated heavy metals (El-Sorogy and Youssef, 2015;
305 Humood A. Naser, 2013; Veerasingam et al., 2020). Previous studies have found Zn, Cu and
306 Mn to be the most dominant and highly concentrated metals in the bodies of gastropods (Astani
307 et al., 2012; Bu-Olayan and Thomas, 2001; Yüzereroğlu et al., 2010a), and the present study
308 also has recorded comparable results for the oil-contaminated shores of the Arabian Gulf. With
309 the proposed target gastropod shown a significant bioaccumulation of tar-associated heavy
310 metals (As, Cu, Ba, Cd, Cr, Zn).

311 On the other side, the studied gastropods display higher concentration of some other
312 metals even without the presence of tar. Suggesting an introductory pathway from other
313 contaminant sources. For instance, Cu and Fe were much higher in Al-Khor, a bay area near a
314 small fishing port located close to the largest urban centre at Qatar, a known polluted marine

1 315 urban zone (Liu et al., 2021). Receiving other anthropogenic inputs of heavy metals. Other
2 316 example is the high concentrations of Mn that is naturally incorporated into the shell of
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4 317 gastropods, replacing the calcium in CaCO₃, and is was previously detected in high
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7 318 concentrations in molluscs in the Arabian Gulf region (El-Sorogy and Youssef, 2015). Even in
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9 319 areas without tar, Fe was also recorded in high concentrations in the target gastropods.
10
11 320 Highlighting that Iron can originate from the dust storms which occur frequently in the Arabian
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13 321 Gulf (Yigiterhan et al., 2020), and enter sediments by deposition of dust. This could lead to Fe
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15 322 accumulation in the gastropods' food sources (e.g., macroalgae), and subsequently to Fe
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17 323 accumulation in the gastropods themselves, after grazing on their food sources.
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21 324 Regarding the correlation of size classes and metal contamination smaller gastropods
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23 325 had displayed higher bioaccumulated concentrations of heavy metals in this study.
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25 326 Contradicting previous study that recorded a positive correlation between body size and tissue
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27 327 metal concentrations (Al-Ansari et al., 2017; Elsayed et al., 2020). However, within the
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29 328 population of a same species, the enrichment of heavy metals in the body of smaller gastropods
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31 329 could be due to differences in metabolic rate, physiological response and exposure time, given
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33 330 that smaller gastropods engage in less movement than larger ones (Catsiki et al., 1994).
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35 331 However, considering that Potentially Toxic Elements (PTEs) as the mentioned heavy metal
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37 332 accumulation on specimens grazing on tar and that PTEs in high concentrations can cause
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39 333 visible physical deformities of the calcite shell (Amao et al., 2019), it is possible that species
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41 334 grazing on tarmat simply die before reaching large sizes or that the contamination interfere in
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43 335 the optimal growing of this gastropods.
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51 336 Despite the multiple sources of contaminants, recent studies show that most tar mats had
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53 337 originate with the 1991 Gulf War oil spill (Arekhi et al., 2020) and as recorded here, the main
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55 338 heavy metals accumulated in molluscs after the spill were Ni, Cu, and Zn (Bu-Olayan and
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57 339 Subrahmanyam, 1997). Corroboration with the hypothesis that these tar-associated heavy
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340 metals can be used for monitoring the contamination level of the entire marine environment
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2 341 using the proposed target gastropods. The results also suggest that old tar mats are still present
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5 342 along the coast of Qatar (Arekhi et al., 2020), and certainly will continue to contaminate
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7 343 ecosystems and result in bioaccumulation in marine flora and fauna.
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10 344 **4.2. Influence of Tar mats on Gastropod Ecology**

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14 345 The size classes observed in this study indicate the potential for population plasticity in the
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16 346 target species. As discussed above, heavy metal contamination was found to be higher in
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19 347 smaller specimens and in an ecological point of view this might be relate to their displacement,
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21 348 their recruitment, and their growing rates. In a given marine gastropod population, recruitment
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23 349 is usually represented by large numbers of small specimens appearing seasonally in the
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26 350 population, while larger specimens are usually older representatives of the same population
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28 351 (Richardson et al., 2005). On this basis, and given the fact that environmental influences and
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31 352 harsh conditions (pollutants) can influence the growth of molluscs (Zdelar et al., 2018), the
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33 353 larger number of small specimens recorded here may being just reflecting a recent recruitment
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36 354 or a large mortality of older specimens due the contamination. The displacement potential is a
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38 355 relevant factor considering that larger animals have better fitness for avoiding contaminated
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41 356 rocks. However, considering that we found during the long low tide period many animals
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43 357 resting on the tar mat, we can assume that those gastropods are adapted to feed and live with
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45 358 tar mats covering their natural niche. Whatever the reason, our ecological and contaminant
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48 359 results indicate an influence of tar mat on the abundance (and the size classes) of each species,
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50 360 providing a foundation for future monitoring research on the studied area.
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54 361 As expected, both species assimilated heavy metal contaminants when grazing on tar mat and
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56 362 in areas surrounded by tar mat (as discussed above). In addition, the species more exposed to
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58 363 water exhibited higher contamination rates, suggesting that tar mats continue to contaminate
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1 364 the environment (sediment, substrate, and water) for a long time, thereby affecting the
2 365 surrounding organisms. One unexpected finding, based on the recorded abundance of species
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4 366 inside and outside tar mat areas, was that tar mat had a more significant impact on the
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7 367 population of *C. bifasciata persica* than on the population of *E. arabica*, even though the tar
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9 368 mat concentration was much higher in the supratidal zone, the domain of *E. arabica*. In the
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12 369 intertidal zone, the domain of *C. bifasciata persica*, tidal oscillations are constantly washing
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14 370 the hard substrate, and the organisms are constantly exposed to water. The abundance results,
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17 371 which found significantly more *C. bifasciata persica* specimens in areas without tar mat and
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19 372 uncovered no significant difference for *E. arabica*, indicate that contaminants in water can
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22 373 directly enter the soft tissue of *C. bifasciata persica* (Baroudi et al., 2020; Bu-Olayan and
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24 374 Thomas, 2001; Yüzereroğlu et al., 2010b). Contradicting the previous hypothesis (discussed
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26
27 375 below) and suggesting that the leaching of tar by seawater and the exposed time to this
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29 376 contaminated coastal water is an important introductory pathway for tar-associated heavy
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32 377 metals, and not only the ingestion of tar particles during the grazing/feeding behaviour and the
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34 378 contamination of their food sources as was previously expected.

37 379 **4.3. Efficiency of Gastropods as Bioindicators of Tar mat Contamination**

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39 380 The results obtained suggest that the intertidal gastropod species studied can indeed be used as
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42 381 bioindicators of heavy metal contamination, as has been previously suggested (Cardoso et al.,
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44 382 2013; Sáez et al., 2012). By combining population ecology studies with a contamination
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47 383 assessment, we were able to better illustrate the ecological problem. Gastropods are already
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49 384 considered to be efficient bioindicators of several other contamination sources (Amin et al.,
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52 385 2009; Baroudi et al., 2020; Bu-Olayan and Subrahmanyam, 1997; Catsiki et al., 1994; El-
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54 386 Sorogy and Youssef, 2015; Krupnova et al., 2018; H. A. Naser, 2013), and this study
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56
57 387 demonstrated that this title can now be extended to include tar mat contaminants.

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60 388 Both target species (*E. arabica* and *C. bifasciata persica*) are highly abundant and widespread

1 389 in the Gulf region (Al-Maslamani et al., 2015; Bosch et al., 1995). Despite significant
2 390 accumulation of heavy metals and niche restriction in the supratidal zone, where tar mats are
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4 391 deposited, the endemic species *E. arabica* showed less contamination than the endemic
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7 392 subspecies *C. bifasciata persica*. This suggests that *Clypeomorus* species, abundant intertidal
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9 393 grazers with representatives in several ecoregions in the Indo-Pacific (Houbrick, 1985), would
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12 394 be better-suited to further studies and comparisons of tar mat contamination in different regions
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15 395 and ecoregions.

16 17 18 396 **5. Conclusion**

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20 397 The selected target species can be found in abundant quantities in all regions of the Gulf,
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22 398 occupying virtually all hard substrates in supratidal and intertidal zones. These zones are easy
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25 399 to access, facilitating the species' collection by scientists and their use for chemical analysis
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27 400 and pollution level assessment. This study provides a reliable database on the tar mat
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30 401 contamination of grazer gastropod species in intertidal and supratidal zones and offers
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32 402 information that will be relevant for further monitoring and comparisons among threatened
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35 403 coastal areas in the Arabian Gulf. Additionally, the study suggests more straightforward
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37 404 alternatives for standardizing information on long-term effects of oil spills. In addition, and in
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40 405 contrast to what had been hypothesized, the grazer gastropod which inhabits the supratidal zone
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42 406 with higher tar mat deposition was less contaminated than the gastropod from the intertidal
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44 407 zone. This suggests that the leaching of tar by seawater and the exposure to this water in the
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46
47 408 intertidal zone has a direct impact on heavy metal accumulation.

48 49 50 409 51 52 410 **Acknowledgments**

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54
55 411 The authors of this research would like to acknowledge and thank Prof. Hamad A. Al Saad, the
56
57 412 Director of the Environmental Science Center (ESC), Qatar University, for his encouragement
58
59
60 413 and continuous support. We also would like to thank the Technical Manager, Mrs. Hajer
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1
2 414 Ammar Al Naimi, and the technical staff for their dedicated work, especially, Mr. Ahmed
3
4 415 Helmy, Mr. Caesar Flonasca Sorino, Mrs. Marwa Mustafa Al-Azhari, Mr. Muhammed Rafeek
5
6 416 Punthiyakath, Mr. Mohammed AbdulKader and Mr. Muhammed Kunhi Pindath during
7
8 417 sampling, preparation, pre-treatment and analysis. This research was supported by the Qatar
9
10 418 Petroleum through the project of QU (QUEX-ESC-QP-TM-18/19).

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16 420 5. References

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573 **Figure 1.** Illustration of the study area and environment, highlighting (A) the location of Al-Khor and
1 574 Al-Ruwais; (B and C) the large intertidal area during low tide where the species were sampled; the
2 575 quadrats in areas (D) fully covered by tarmat, (E) partially covered by tarmat, (F) without tarmat, and
3 576 (G) without tarmat and with a microbial mat
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6 577 **Figure 2.** Details of *E.arabica* (A) on the tarmat layer that covers the rocks and (B) on the rock in the
7 578 supratidal zone; and *C. bifasciata persica* (C) on tarmat (D) close-up and (E) on rock (without
8 579 tarmat).
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10 580 **Figure 3.** Tar mat coverage (%) in the quadrats recorded in the intertidal and supratidal zone; the
11 581 quadrats considered as covered by tar on the present study.
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14 582 **Figure 4.** Abundance of *E.arabica* (A-E) and *C. bifasciata persica* (F-J) on tarmat and on rock
15 583 (without tarmat); considering the number of specimens on entire population (A, F), and on the
16 584 different size classes, Large (C, H), Medium (D, I) and Small (E, J), and the dominance (%) of each
17 585 size classes (B, G).
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20 586 **Figure 5.** Average (\pm standard error) of the heavy metals: As, Ba, Zn (A); Cd, Co, Cr (B); Cu, Mn, Ni
21 587 (C) in *C. bifasciata persica* and *E. arabica* gastropod species in Al-Khor and Al-Ruwais. The
22 588 averages of heavy metals are displayed according to tar existence in the aforementioned sampling
23 589 locations, without tar (top) and with tar (bottom).
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26 590 **Figure 6.** Concentrations of the heavy metals (A) Cu and (B) Ni [$\mu\text{g/g}$] in the populations of *C.*
27 591 *bifasciata persica* in Al-Ruwais, comparing quadrats with tarmat coverage and quadrats without. The
28 592 same comparison is then made for Cu [$\mu\text{g/g}$] concentrations in the populations of *E.arabica* in (C) Al-
29 593 Ruwais and (D) Al-Khor.
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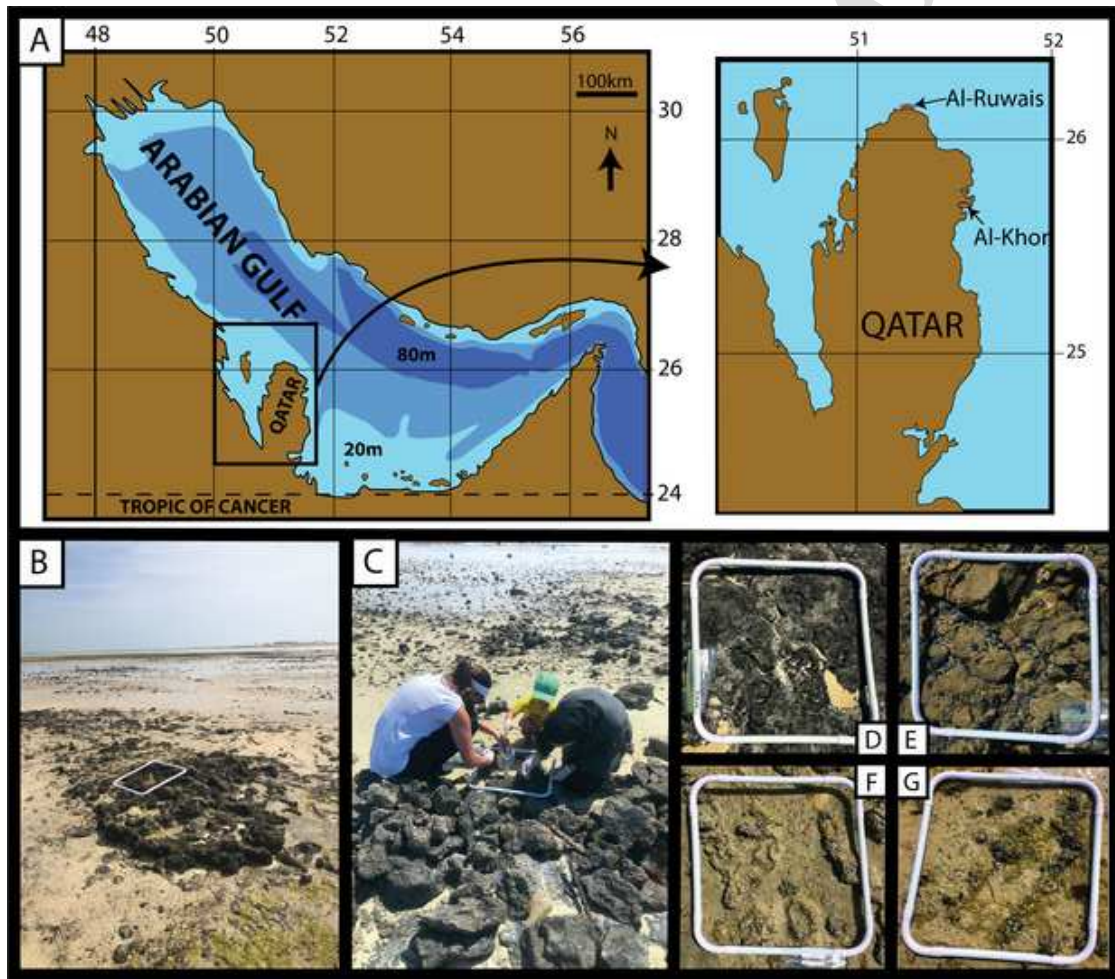
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Figure 1

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Figure 2

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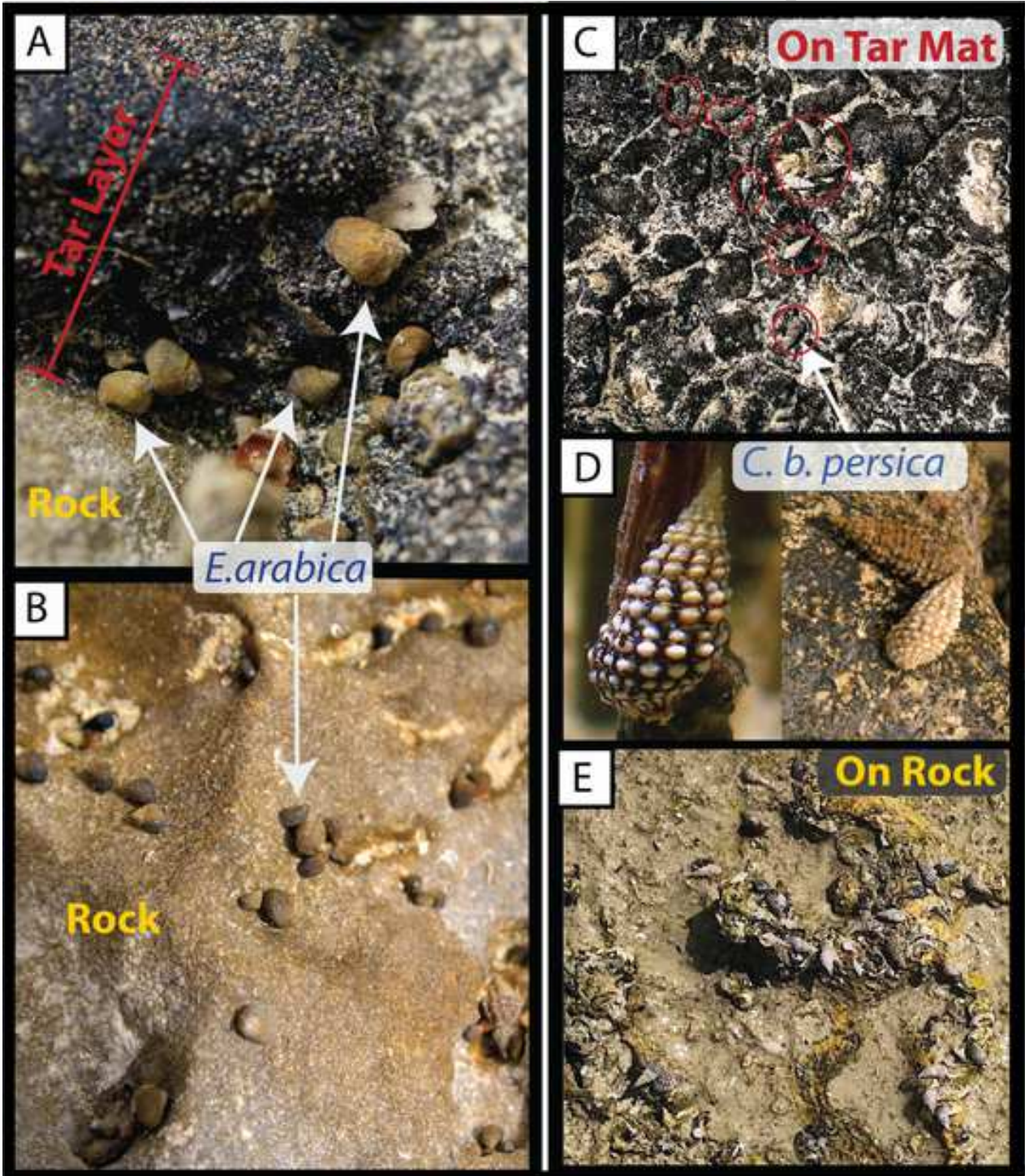


Figure 3

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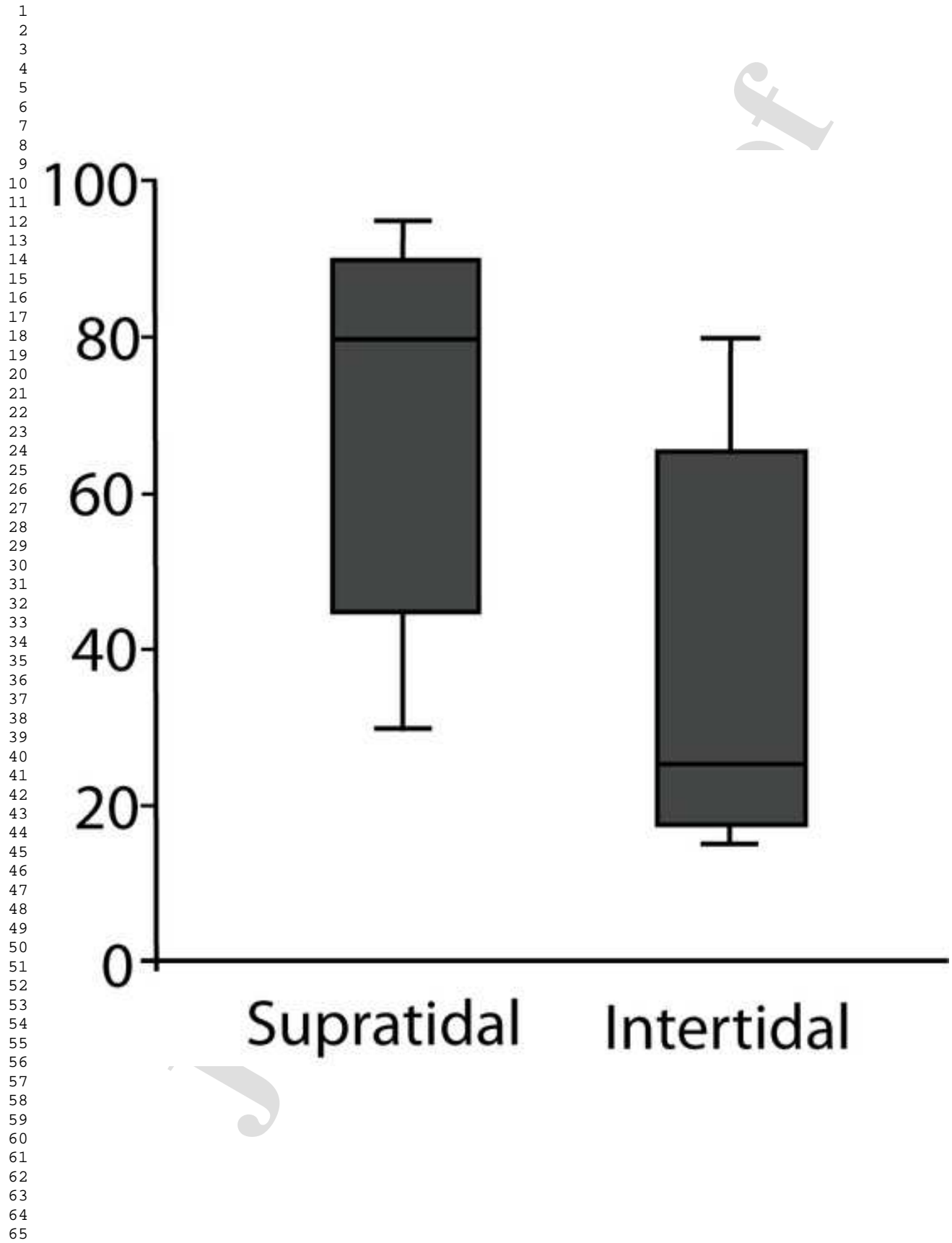
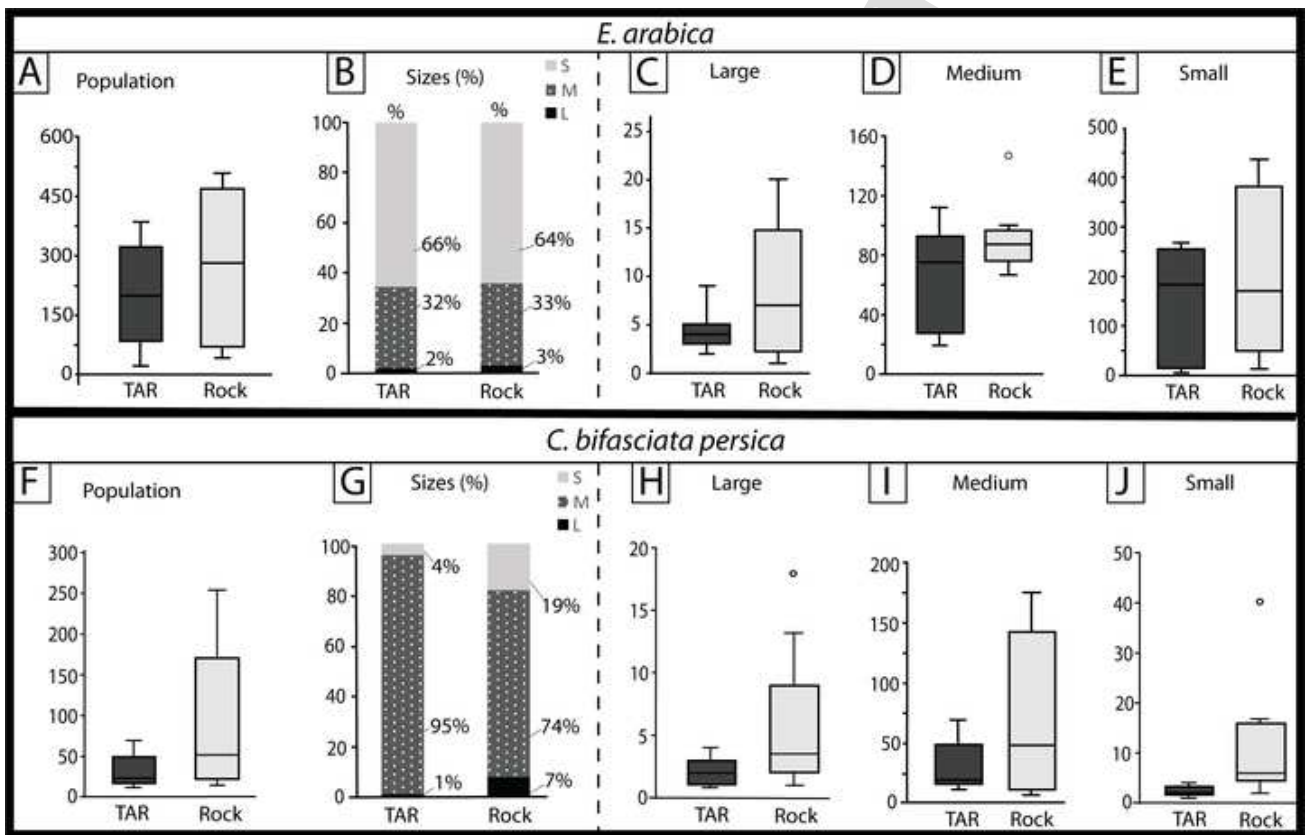


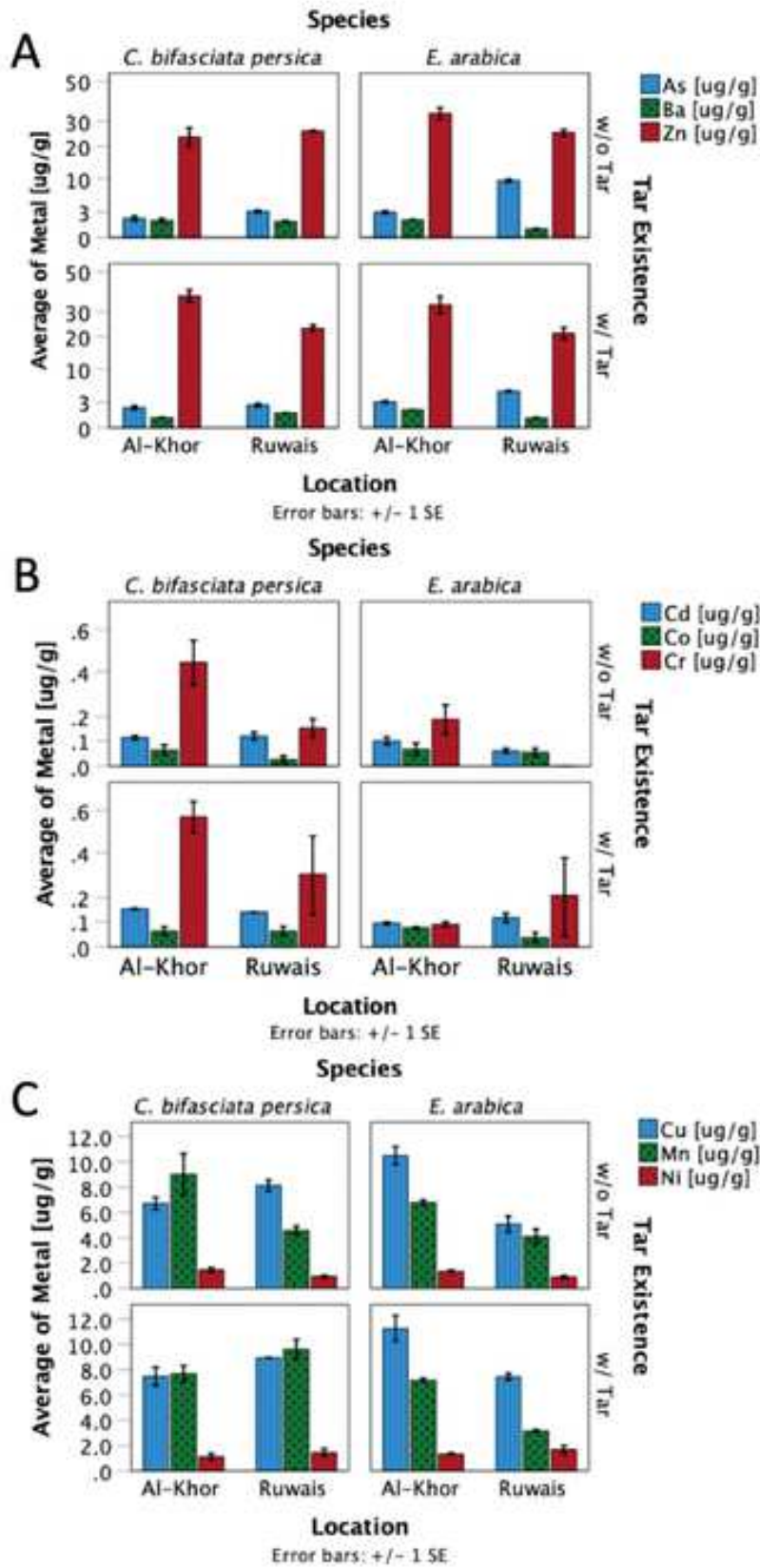
Figure 4

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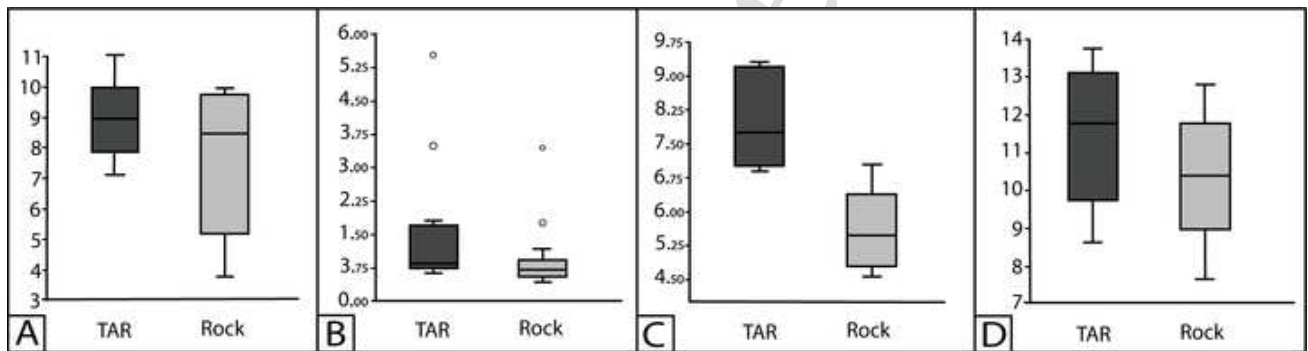
Figure 5

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Figure 6

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Highlights - Tarmat contaminants

- Shoreline gastropods can be used to standardize tarmat contamination monitoring
- Grazer snails are target for the bioaccumulation of tarmat-associated heavy metals
- *Clypeomorus* and *Echinolittorina* representatives are the bioindicators in the Gulf
- Intertidal grazer (*Clypeomorus*) was more contaminated than the supratidal species
- Cu, Ni were the target heavy metals associated to the tarmat contaminations

Table 1. Pearson Correlation matrix of the heavy metals As, Ba, Cd, Cr, Cu and Fe [$\mu\text{g/g}$] and their strong positive (black) and negative (red) correlations with each other ($R^2 > 0.80$, $p < 0.05$ and < 0.01) at the Al-Khor site with tarimat contamination. In the table, a dash represents weak or no strong relationship between the heavy metals

Metal	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Ni	Population Abundance
As	-	-	-	-	-	0.96^a	0.89 ^b	-	-	-
Ba	-	-	0.97^a	-	-0.91^b	0.90^a	0.98^a	-	-	-
Cd	-	0.97^a	-	-	0.97^a	-	-0.97^a	-	-	-
Co	-	-	-	-	-	-	-	-	-	-
Cr	-	-0.91^b	0.97^a	-	-	-	-0.89^b	-	-	-
Cu	0.96^a	0.90^a	-	-	-	-	0.95 ^b	-	-	0.90 ^b
Fe	0.90 ^b	0.98^a	-0.97^a	-	-0.89^b	0.95 ^b	-	-	-	0.93 ^b
Mn	-	-	-	-	-	-	-	-	-	-
Ni	-	-	-	-	-	-	-	-	-	-

^a Correlation is significant at the 0.01 level (values were highlighted as bold)

^b Correlation is significant at the 0.05 level

Table 2. Pearson Correlation matrix of the heavy metals As, Ba, Cd, Cr, Cu and Fe [$\mu\text{g/g}$] and their strong positive (black) or negative (red) correlations with each other ($R^2 > 0.80$, $p > 0.05$ and > 0.01) at the Al-Ruwais site with tarimat contamination. In the table, a dash represents weak or no strong relationship between the heavy metals

Metals	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Ni
As	–	-0.94^a	–	–	–	-0.92^a	–	-0.99^a	–
Ba	-0.94^a	–	–	–	–	0.95^a	–	0.90^a	–
Cd	–	–	–	–	–	–	-0.93^a	–	–
Co	–	–	–	–	0.89 ^b	–	–	–	–
Cr	–	–	–	0.89 ^b	–	–	–	–	0.83 ^b
Cu	-0.92^a	0.95^a	–	–	–	–	–	0.92^a	–
Fe	–	–	-0.93^a	–	–	–	–	–	–
Mn	-0.99^a	0.90 ^b	–	–	–	0.92^a	–	–	–
Ni	–	–	–	–	0.83 ^b	–	–	–	–

Manuscript Number: RSMA-D-21-01095

Target gastropods for standardizing the monitoring of marine life tarmat contamination in the Arabian Gulf

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- Samah Dib - Data curation; Formal analysis; Investigation; Methodology
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- Juha Mikael Alatalo - Supervision; Validation
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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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