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1 **Screening of seaweeds in the East China Sea as potential bio-monitors of heavy metals**

2

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22

23 **Abstract:** Seaweeds are good bio-monitors of heavy metal pollution and have been included in
24 European coastal monitoring programs. However, data for seaweed species in China are scarce or
25 missing. In this study, we explored the potential of seaweeds as bio-monitor by screening the natural
26 occurring seaweeds in the “Kingdom of seaweed and shellfish” at Dongtou Islands, the East China Sea.
27 Totally, 12 seaweed species were collected from six sites, with richness following the sequence of
28 Rhodophyta > Phaeophyta > Chlorophyta. The concentration of heavy metals (Cu, Cr, Ni, Zn, Pb, Cd,
29 As) in the seaweeds were determined, and the bioaccumulation coefficient was calculated. A
30 combination of four seaweeds, *Pachydictyon coriaceum*, *Gelidium divaricatum*, *Sargassum thunbergii*,
31 and *Pterocladia capillacea*, were proposed as bio-monitors due to their high bioaccumulation
32 capabilities of specific heavy metals in the East China Sea, and hence hinted the importance of using
33 seaweed community for monitoring of pollution rather than single species. Our results provide first
34 hand data for the selection of bio-monitor species for heavy metals in the East China Sea and contribute
35 to selection of cosmopolitan bio-monitor communities over geographical large area, which will benefit
36 the establishment of monitoring programs for coastal heavy metal contamination.

37 **Keyword:** seaweed; heavy metal; East China Sea; richness; bio-monitor; bioaccumulation coefficient

38

39 1. Introduction

40 Heavy metals released from anthropogenic activities are affecting the coastal systems increasingly
41 (Elsikaily et al. 2011; Gao and Chen 2012; Tamilselvan et al. 2013). The fact that heavy metals in
42 coastal waters are persistent with no degradation opened the opportunity of identifying bio-monitors
43 that denotes an anthropogenic impact (Reis et al. 2011). Bio-monitor needs to fulfill certain required
44 characteristics including sedentary nature, widespread distribution with considerable biomass, easy to

45 identify etc., in addition to being pollutants accumulators (Rainbow 1995). Seaweeds are therefore
46 considered to be good agents for bio-monitoring of heavy metals in seawater (Akcali and Kucuksezgin
47 2011; Chaudhuri et al. 2007; Murphy et al. 2007). By now, in Europe, seaweeds have been included as
48 key organisms to classify and monitor the ecological status of coastal ecosystems with decades of
49 studies (Reis et al. 2014; Viana et al. 2010), as documented in the European Water Framework
50 Directive (Ballesteros et al. 2007; Juanes et al. 2008). For instance, the seaweed *Alaria esculenta* in
51 Irish coast was suggested to be used in the European Real-Time Environmental Monitoring Networks,
52 because of its accumulation efficiency of heavy metal contamination (As, Cd, Cu, Fe, Hg, Pb, and Zn)
53 (Reis et al. 2016).

54 Compared to progress made on Western world seaweed species, much less is known for their
55 Eastern relatives, despite the need to identify cosmopolitan bio-monitor to allow intra-specific
56 comparisons of bio-availabilities between geographical areas (Rainbow 1995). The bio-monitoring
57 potential of seaweed species from the Eastern world include a few case-studies from China (Caixue et
58 al. 2010; Zhang et al. 2014), India (Chakraborty et al. 2014; Kumar et al. 2009), and Russia
59 (Khristoforova and Kozhenkova 2002). For instance, brown algae (*Padina gymnospora* and *Dictyota*
60 *bartayresiana*) and green algae (*Ulva lactuca* and *Enteromorpha intestinalis*) in the Gulf of Kutch
61 (India) were suggested as bio-monitors for Cd, Ni, Cr, Cu, Zn and Pb (Chakraborty et al. 2014), as well
62 as *Gracilaria confervuoides* in the Liusha Bay of South China Sea for As, Cu, Pb, Cr and Fe (Caixue et
63 al. 2010), *Cladophoropsis zollingeri* for Fe in the mangrove area of South China Sea (Zhang et al.
64 2014). The lack of investigation of seaweed potential as bio-monitor for heavy metal in China is
65 surprising, taking into account that 47% of the world's seaweed production is derived from China
66 (FAO 2017) representing a high biodiversity (Ding et al. 2011). Indeed, there is a need for screening

67 the native seaweed species for potential use as bio-monitors in future.

68 Furthermore, large-scale seaweed aquaculture has been proved to be an important way of nutrient
69 reduction in coastal water in China due to high nitrogen and phosphorus removal capacity (Xiao et al.
70 2017). Investigating the heavy metal concentrations of seaweed tissues will benefit the potential
71 application of seaweeds as bio-monitor of coastal heavy metal contaminations. In addition, seaweed
72 composition could also reflect and monitor the ecological status of coastal environments (Diez et al.
73 2009; Paraskevi et al. 2015; Pinedo et al. 2007). For instance, in the Northwestern Mediterranean,
74 healthy coastal ecosystems were dominated by dense *Cystoseira* forests, comparing to those comprised
75 by *Ulva* spp. and other green algae (Pinedo et al. 2007). In Hainan Island of China, seaweed species
76 richness and composition were found closely related to a pollution gradient (Li et al. 2016).

77 Therefore, we conducted a survey in Dongtou Islands of the East China Sea, the so-called
78 “kingdom of seaweed and shellfish” of China for its rich seaweed biodiversity. The objectives of this
79 study are to (i) investigate the seaweed richness, (ii) quantify the seaweed tissue heavy metal
80 concentrations, and (iii) calculate the bioaccumulation coefficient for heavy metals for
81 cross-comparisons between seaweed species. Our study aims to fill the knowledge gap of biodiversity
82 and bio-monitoring potential of seaweeds in East China Sea, as well as to bring new insights into a
83 bio-monitoring program for heavy metal pollution in Chinese coastal waters.

84

85 2. Materials and methods

86 2.1 Study area

87 Dongtou Islands (120°59'45" E~121°15'58" E, 27°41'19" N~28°01'10" N), located in the East
88 China Sea (China), has subtropical marine monsoon climate. Annual seawater salinity in Dongtou

89 Islands ranges from 27 to 35 ‰. Annual average water temperature is 18.4 °C, where February has
90 minimum temperature with average monthly water temperature of 8.5 °C, and August has maximum
91 temperature with average monthly water temperature of 27.9 °C. The average tidal range is about 4.5 m.
92 The seaweed community in this area is diverse due to the unique regional geographical and
93 oceanographic conditions. This region is climatically influenced from the Taiwan warm ocean current,
94 and is affected by Oujiang River.

95 2.2 Sampling

96 Seaweeds were collected from six sites in the Dongtou Islands (Fig. 1) during low tide period of
97 May, 2015. Sites 1-4 were located nearshore on the main island, and sites 5-6 were located in open sea.
98 Site 1 and site 2 are close to fishery harbor and village, which are influenced by heavier pressure of
99 human activity compared to the other sites. Seaweeds were collected from the intertidal and subtidal
100 zone by free diving. The seaweed samples were carefully washed at the sampling site with seawater to
101 remove the epiphytic organisms and trapped sediment, stored in polyethylene bags containing seawater,
102 and then transferred to the laboratory in portable cooler at 4 °C.

103 2.3 Seaweed identification

104 In the lab, the samples were washed with tap water and rinsed with Milli-Q water (Millipore,
105 Milli-Q Advantage A10, France) and then wiped with dry towels. Sampled seaweeds were identified
106 and classified based on their morphological features including the size, shape and color.

107 2.4 Chemical analysis

108 After the identification, the samples were dried at 60 °C for 48 h. The concentrations of heavy
109 metals in seaweed samples were determined by inductively coupled plasma-mass spectrometry
110 (ICP-MS, Agilent, 7900, USA) for Cu, Cr, Ni, Zn, Pb, Cd and As (Caixue et al. 2010; La Rocha et al.

111 2009). For the detection of Cu, Cr, Ni, Zn, Pb, Cd and As, the dried samples were homogenized in an
112 agate mortar and reduced to a fine powder after sieving (200 μm) (Caixue et al. 2010), and then
113 digested by microwaving (LabTech, ETHOS UP, Italy) with 7 ml 65 % nitric acid and 1 ml 30 %
114 hydrogen peroxide; all the reagents used during procedure were of analytical grade (Caixue et al. 2010;
115 Kamalakannan et al. 2008). The digestion steps were as follows: (i) 0.2 MPa, 4 min; (ii) 0.5 MPa, 2
116 min; (iii) 0.8 MPa, 2 min; (iv) 1.0 MPa, 2 min; (v) 1.2 MPa, 2 min; (vi) 1.5 MPa, 5 min (Caixue et al.
117 2010). The solution was transferred into a 50 ml volumetric flask and diluted with Milli-Q waters
118 (Millipore, Milli-Q Advantage A10, France). Laver (35 g, Chinese National Standard Material No.
119 GBW 10023), from the Institute of Geophysical and Geochemical Exploration, Chinese Academy of
120 Geological Science, was used as certified reference material (Duan et al. 2015; Jiang et al. 2009; Wang
121 et al. 2014). Three replicate detections were performed on the certified reference material. Analytical
122 blanks were run in the similar way as that of the samples (Kamalakannan et al. 2008). The recovery of
123 the procedures was checked using the certified reference material (Giusti 2001). The detection results
124 of certified reference material (Laver) by ICP-MS method proved the reliability of the detections for
125 Cu, Cr, Ni, Zn, Pb, Cd and As (Table 1).

126 2.5 Bioaccumulation coefficient

127 The annual mean concentrations of heavy metals in seawater in Dongtou Islands in 2015 were
128 derived from the bulletin of coastal environment released by the Ministry of Environment Protection,
129 China (MEP 2015). The seawater quality monitoring frequency is two to three times a year in March to
130 May, July to August and October for in total 301 coastal water quality monitoring stations in China.
131 The seawater sampling and detection method followed the technical specification for offshore
132 environmental monitoring by Ministry of Environmental Protection (HJ442-2008, (MEP 2008)).The

133 seawater monitoring data from the monitoring site (NO. ZJ0304, E: 121.2100, N: 27.8144), which is
134 close to the study area, was used to calculate the bioaccumulation coefficient in the seaweeds. The
135 concentration of Zn in seawater was below the detection limit of Zn (6.4 µg/L) (MEP 2008). The
136 bioaccumulation coefficient was calculated as the ratio of the seaweed tissue concentration to the
137 concentration in the coastal waters.

138 2.6 Statistical analysis

139 Principal component analysis (PCA) of tissue heavy metal concentrations in seaweed was
140 performed in MATLAB (version 2016a). The concentrations of Cu, Cr, Ni, Zn, Pb, Cd and As were
141 used as discriminating variables. Cluster analysis of tissue heavy metal concentrations in seaweed was
142 performed by the Ward grouping method using SPSS (version 20). $p < 0.05$ was considered as
143 significant.

144 3. Result and discussion

145 3.1 Richness of natural seaweed species

146 In this study, we investigated the seaweed species distributed in Dongtou Islands in summer.
147 Totally 12 seaweed species were found at the sampling sites (Table 2), belonging to three phyla -
148 Phaeophyta (brown algae), Rhodophyta (red algae) and Chlorophyta (green algae). Two additional
149 samples of *Gracilaria lemaneiformis* and *Ulva pertusa* were collected from floating seaweed
150 cultivation raft. Rhodophyta showed highest richness in this survey, accounting for 59% of all the
151 species, followed by Phaeophyta (33%) and Chlorophyta (8%). *Sargassum fusiforme*, *Sargassum*
152 *thunbergii* and *Chondracanthus intermedius* were found in both intertidal and sublittoral zone, from
153 -1.0 m to 1 m (Fig. 2). *Sargassum vachellianum*, *Pachydictyon coriaceum*, *Ahnfeltiopsis flabelliformis*,
154 *Laurencia tropica* and *Pterocladia capillacea* were confined to the sublittoral zone, which was no

155 more than 2.0 m deep (Fig. 2). *Polyopes polyideoides* and *Gelidium divaricatum* were restricted to the
156 intertidal zone (Fig. 2).

157 Seaweed species richness increased significantly from offshore to the sea (Table 2, Fig. 1), which
158 is similar to patterns reported from the South China Sea (Li et al. 2016). However, the seaweed species
159 richness in our study was higher in site 3 and 4 located on the main island comparing to the uninhabited
160 islands (site 5 and 6), which contradicts to the SW Atlantic, where coastal urbanization was found to
161 lead to seaweed species loss and shifts in community structure (Schermer et al. 2013). A possible
162 explanation is that nutrient inputs (i.e. domestic sewage) stimulated the growth of seaweeds on the
163 coasts and increased the complexity of seaweed community composition. Several seaweed species
164 appeared in a list of economic important seaweeds in the East China Sea about half a century ago,
165 including *Sargassum fusiforme*, *Sargassum thunbergii*, *Gelidium divaricatum* and *Ulva pertusa*
166 suggesting that these species have been present in historic times (Tseng and Chang 1959). Due to lack
167 of earlier documents in the region, it's difficult to evaluate potential changes of seaweed richness and
168 composition over time emphasizing the need for baseline studies to be used for future research on
169 regional marine ecosystem in response to global changes and human activities.

170

171 3.2 Heavy metal comparison on the phylum level

172 3.2.1 Phaeophyta

173 Phaeophyta has the highest concentrations of Cd (4.59 mg/kg DW) and As (44.02 mg/kg DW)
174 among all seaweeds, and equal concentrations of Cr (2.25 ± 0.10 mg/kg DW) and Pb (1.95 ± 0.10
175 mg/kg DW) compared to Rhodophyta and Chlorophyta (Fig. 3). Similar to our study, Phaeophyta was
176 found to absorb or accumulate significantly higher concentrations of Cd (Mata et al. 2009; Schintu et al.

177 2010) and As (Malea and Kevrekidis 2014; Ryan et al. 2012) compared to Rhodophyta and
178 Chlorophyta from seawater. In general, brown seaweeds are resistant to marine pollutions (Costa et al.
179 2016) and at the same time, brown seaweeds have been proven to be effective and predictable in the
180 biosorption of Pb from aqueous solutions (Davis et al. 2003). More specifically, *Sargassum filipendula*
181 (Luna et al. 2010) and *Macrocystis pyrifera* (Cazon et al. 2012) showed high efficiency in biosorbing
182 Cd. Similarly, *Lessonia nigrescens* - a brown seaweed in the Chilean coast - absorbed both Cd and As
183 from sea water (Gutierrez et al. 2015; Hansen et al. 2006), and *Fucus vesiculosus* had been found to
184 bioaccumulate heavy metals in estuaries and coastal waters throughout northwest Europe (Varma et al.
185 2011).

186 Our results suggest that in the East China Sea, Phaeophyta can be used as bio-monitor of Cd and
187 As pollution. Indeed, various brown algae have been proposed as bio-monitors, including *Fucus*
188 *spiralis* for Cd contamination in the northwest coast of Portugal (Reis et al. 2014), *Padina pavonica*
189 (Schintu et al. 2010) and *Cystoseira* (Kravtsova et al. 2015) for both Cd and Pb contamination in the
190 Mediterranean Sea and the Black Sea, respectively, and *Cystoseira barbata* for Fe, Mn and Zn in
191 Aegean Sea (Aydın-Önen and Öztürk 2017).

192 3.2.2 Rhodophyta

193 Concentrations of Cu (20.66 mg/kg DW) and Zn (60.89 mg/kg DW) in Rhodophyta were
194 significantly higher than those of Phaeophyta and Chlorophyta (Fig. 3) suggesting potential use of
195 Rhodophyta as bio-monitor of Cu and Zn pollutions in the East China Sea. Similar to this study,
196 Rhodophyta was an effective adsorbent or accumulator for Cu and Zn, i.e. *Pterocladia capillacea* for
197 Cu (Elsikaily et al. 2011), *Polysiphonia lanosa* for Zn (Ryan et al. 2012), and *Gymnogongrus torulosus*
198 for Cu and Zn (Areco and Dos 2010). Interestingly, Rhodophyta in the Gulf of Kutch were found to

199 accumulate less Cu and Zn compared to Phaeophyta and Chlorophyta (Chakraborty et al. 2014),
200 indicating that heavy metal accumulation in seaweeds is complex and regional scale study is needed
201 before using the seaweed as bio-monitor in a specific area (Fig. 3, Table 3). Furthermore, Rhodophyta
202 was found that it had effective biosorption capacity of Pb (Ibrahim 2011), i.e. *Chondracanthus*
203 *chamissoi* (Yipmantin et al. 2011), *Gracilaria corticata var cartecala* and *Grateloupia lithophila*
204 (Tamilselvan et al. 2013).

205 3.2.3 Chlorophyta

206 Chlorophyta has the highest concentration of Ni (7.14 mg/kg DW) (Fig. 3), which suggests
207 potential use as bio-monitor for Ni pollution in the East China Sea, consistent with investigations of
208 *Chaetomorpha aerea* in the Arabian Gulf (Alhomaïdan 2008) and *Ulva reticulata* in India
209 (Vijayaraghavan et al. 2005). Similar to this study, Chlorophyta were good bio-monitors of Pb, i.e.
210 *Ulva lactuca* (Figueira et al. 2016) and *Caulerpa lentillifera* (Pavasant et al. 2006). Furthermore, the
211 capacity of biosorption or bioaccumulation of Cu and Zn was highlighted for Chlorophyta, i.e.
212 *Helimeda gracilis* (Jayakumar et al. 2015), *Chaetomorpha linum* (Ajjabi and Chouba 2009), and *Ulva*
213 sp. and *Enteromorpha* sp. (Akcali and Kucuksezgin 2011).

214 The uptake behaviors of heavy metals are also different through the three seaweed phyla, which
215 will help to explain why these seaweed phyla showed dramatically different pattern of heavy metal
216 accumulation in addition to the species specific favor of metals of seaweeds (He and Chen 2014;
217 Sawidis et al. 2001). Similar to our suspicion, a study on the distribution of rare earth elements (REE)
218 and silicon in seaweed showed that the uptake behavior of these elements dramatically differed in
219 green, red, and brown algae. In details, the REE patterns similar to that of suspended particles for the
220 green and red algae, but patterns closer to that of seawater solutions and relatively lower Si

221 concentration for the brown algae (Fu et al. 2000).

222 3.3 Analysis on the species level

223 3.3.1 Principal component analysis (PCA) and cluster analysis

224 PCA analysis based on concentrations of heavy metals in the seaweed species showed that about
225 79% of the total variance was explained by three PCs (Fig. 4). PC1 is mostly associated with Cu, Cr,
226 and Pb, and explains 41 % of the total variance. *Gelidium divaricatum* shows the highest
227 concentrations of Cr (6.83 mg/kg DW) and Pb (4.21 mg/kg DW) (Table 3). In addition to having the
228 highest Cu concentration (47.62 mg/kg DW) (Table 3), *Pachydictyon coriaceum* also has relative high
229 concentrations of Cr (5.9 mg/kg DW) and Pb (3.34 mg/kg DW) (Table 3). This suggests that these two
230 seaweed species are more sensitive to the bioaccumulation of Cu, Cr and Pb than other species,
231 respectively. PC2 is mainly associated with Zn and Cd, explaining 23 % of the total variance. *Gelidium*
232 *divaricatum* has the highest Zn concentration (134.50 mg/kg DW) (Table 3), *Sargassum vachellianum*
233 has the highest Cd concentration (8.48 mg/kg DW) (Table 3), indicating that these two seaweed species
234 are the most sensitive to the bioaccumulation of Zn and Cd, respectively. PC3 is clearly featured by As
235 and explains 15 % of the total variance. *Sargassum fusiforme* shows the highest As concentration
236 (57.71 mg/kg DW) (Table 3), followed by *Sargassum thunbergii* (49.08 mg/kg DW) (Table 3),
237 reflecting that these two species are more sensitive to accumulate As compared with other seaweed
238 species. According to the PCA, the pattern of heavy metal concentrations in seaweeds is determined in
239 the order PC1 (Cu, Cr and Pb) > PC2 (Zn and Cd) > PC3 (As), and the concentrations of heavy metals
240 in the seaweed species are more closely related to species rather than location (sampling site). Cluster
241 analysis reflects the similarity in distribution of heavy metals and hence the comprehensive
242 bioaccumulation ability of seaweed species, which were divided into five groups (Fig. 5). For instance,

243 *Gelidium divaricatum* and *Pachydietyon coriaceum* are divided into the same group, showing that they
244 have similar elements distribution and bioaccumulation ability. This is consistent with the PCA result,
245 which demonstrates that both of them are sensitive to accumulate Cr and Pb. Similar finding is
246 presented in the group of *Sargassum fusiforme* and *Sargassum thunbergii* for the bioaccumulation of
247 As.

248 3.3.2 Bioaccumulation coefficient of heavy metal in seaweed species

249 The bioaccumulation coefficient showed that *Pachydietyon coriaceum* is the strongest
250 accumulator of Cu, *Gelidium divaricatum* of Cr and Pb, *Sargassum vachellianum* of Cd, *Sargassum*
251 *fusiforme* of As, and *Laurencia tropica* of Ni (Table 4). Thus, our data reveal seaweed species with the
252 strongest accumulation of particular elements. For instance, our findings that *Sargassum* has a high
253 ability to accumulate As are consistent to other studies, i.e. of *Sargassum* spp. in the Todos os Santos
254 Bay, Brazil (Brito et al. 2012). Moreover, *Sargassum* sp. has high potential biosorption capacity of Pb,
255 Zn, Cu and Ni (Sheng et al. 2004), in addition to our findings of its high accumulation capacity of Cd
256 and As. Therefore, our data suggest the identification of potential cosmopolitan bio-monitors for
257 several heavy metal elements. Similarly, in addition to Cu, *Pachydietyon coriaceum* has high
258 accumulation capacities for Cr and Pb. Other good potential cosmopolitan bio-monitors consist of (1)
259 *Sargassum thunbergii* for Cd and As, and (2) *Pterocladia capillacea* for Cu and Ni.

260 Interspecific differences of bioaccumulation coefficient for heavy metals in seaweeds have
261 usually been attributed to the binding sites for elements (Stengel et al. 2004) and biochemical
262 composition (Malea and Kevrekidis 2014), which is determined by the species phylogeny (Hoek CVD
263 et al. 1995). However, even in the same seaweed species, the bioaccumulation coefficient is often
264 affected by the heavy metals concentrations in the coastal waters (Caixue et al. 2010; Ryan et al. 2012;

265 Zhang et al. 2014). Similarly, it seems as the Cu concentration in seawater influenced the
266 bioaccumulation coefficient, i.e. in the Liusha Bay, the Cu concentration of seawater was 12.3µg/L and
267 the bioaccumulation coefficient of *Sargassum thunbergii* was 1788 (Caixue et al. 2010), while in this
268 study, the Cu concentration in seawater was 0.064 µg/L, and the corresponding bioaccumulation
269 coefficient of *Sargassum thunbergii* was 20,016. Therefore, research efforts are needed for elements
270 and seaweed species where data are scarce or missing (Malea and Kevrekidis 2014), emphasizing the
271 importance of the current study in the East China Sea. Moreover, our data will also contribute to the
272 selection of bio-monitor species and the design of bio-monitoring programs over large geographical
273 areas, as well as the interpretation of bio-monitoring data for coastal environment protection and
274 management.

275 3.3.3 Selection of seaweed species for bio-monitoring

276 The phylogeny of seaweeds usually determine the biochemical composition and binding sites for
277 elements (Malea and Kevrekidis 2014) and thus contribute to the bioaccumulation coefficient of heavy
278 metals (Figs. 4 and 5, Table 4). Our result revealed that some species are better bio-monitors for certain
279 heavy metals. However, in coastal waters, the heavy metals are often present as multi-pollutions, rather
280 than a single element. Nevertheless, bioaccumulation studies focus mostly on the accumulation of
281 heavy metals from single metal solutions (Luna et al. 2010). Therefore, a better strategy of ecological
282 monitoring and accumulation would be to combine several seaweed species and measure several heavy
283 metals. The concept of using seaweed communities as bio-monitor has been applied in several
284 European countries, such as Spain, Poland, Germany, and Denmark, for early detection of increased
285 anthropogenic contaminations (Viana et al. 2010). Our results of the selective bioaccumulations of
286 certain heavy metals by local seaweeds, and the concept of using a suite of bio-monitors will allow

287 recognition of the presence and relative magnitude of heavy metal sources, and thus benefit the
288 establishment of novel monitoring program for heavy metal contaminated marine environment.

289 4. Conclusion

290 1. The seaweed richness of this survey follows the sequence of Rhodophyta > Phaeophyta >
291 Chlorophyta.

292 2. In the East China Sea, Phaeophyta are promising bio-monitor of Cd and As, as well as
293 Rhodophyta for Cu and Zn and Chlorophyta for Ni.

294 3. PCA and cluster analysis, together with the bioaccumulation coefficient, suggest that
295 *Pachydictyon coriaceum*, *Gelidium divaricatum*, *Sargassum thunbergii* and *Pterocladia capillacea*
296 have good potential to act as bio-monitors of heavy metals.

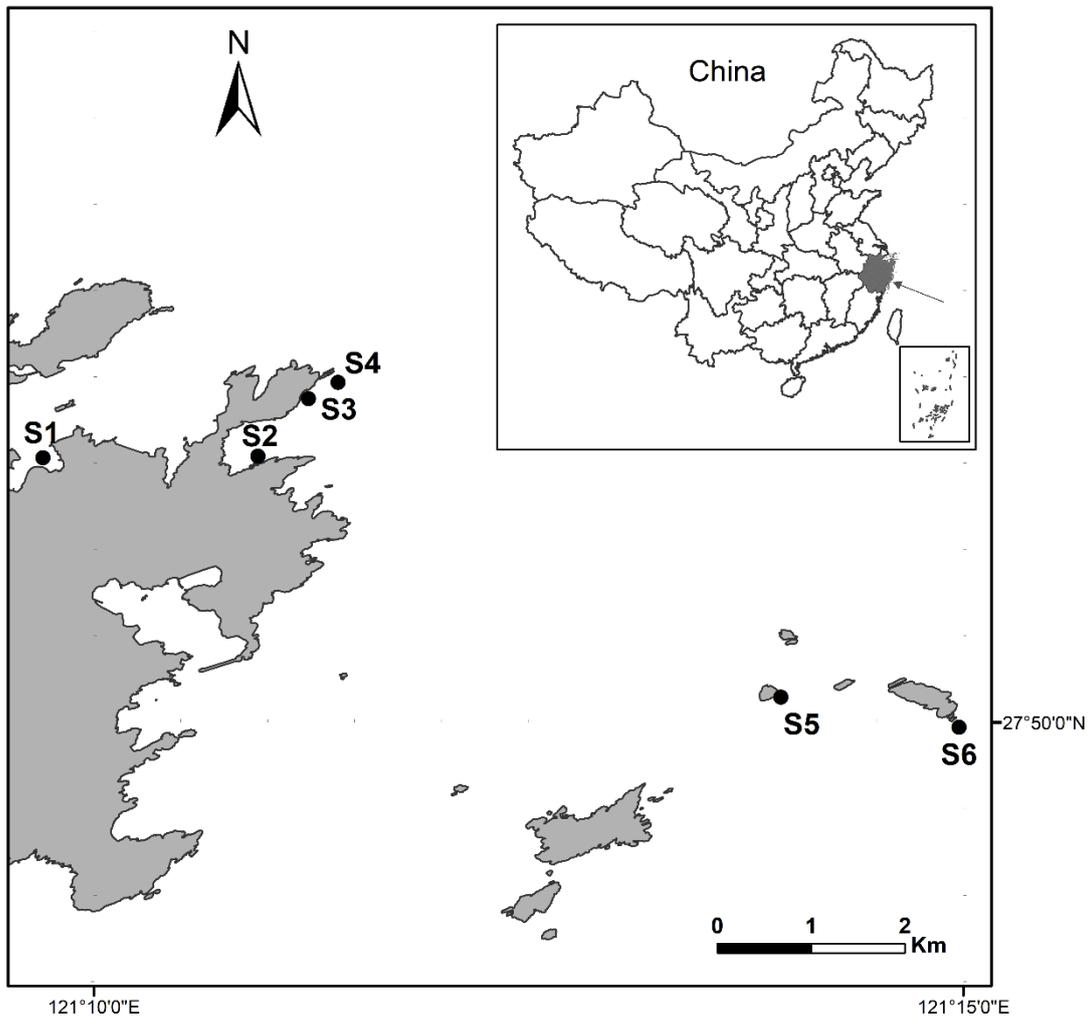
297 4. A single seaweed species does not reflect the comprehensive heavy metal pollutants in coastal
298 environment, and combining seaweed species posing different bioaccumulation capacities will benefit
299 the bio-monitoring of heavy metals.

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304 candidate in Zhejiang University for his assistant in drawing Fig. 4.

305 Author contribution

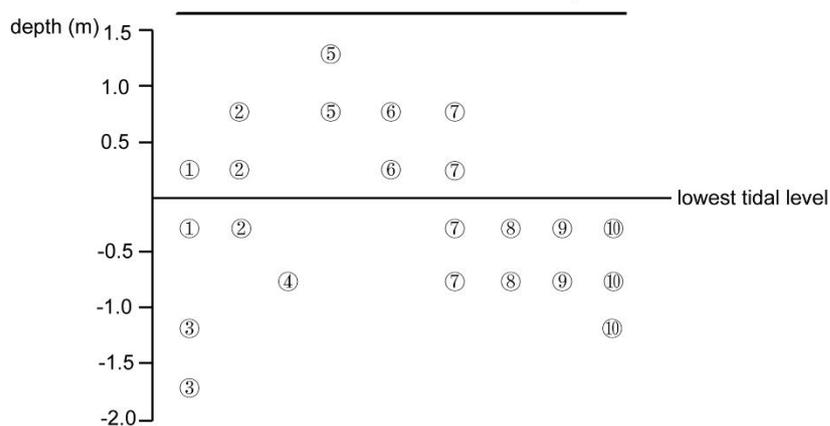
306 Conceived and designed the analysis: X.X., T.W., Y.P. Sampled and identified the seaweed: T.W.,
307 T.D., X.X., Y.P., K.L., F.L., J.W., Y.Y. Wrote the paper: Y.P., X.X., M.H., T.W. All authors analyzed,
308 interpreted the results, and edited the manuscript.



309

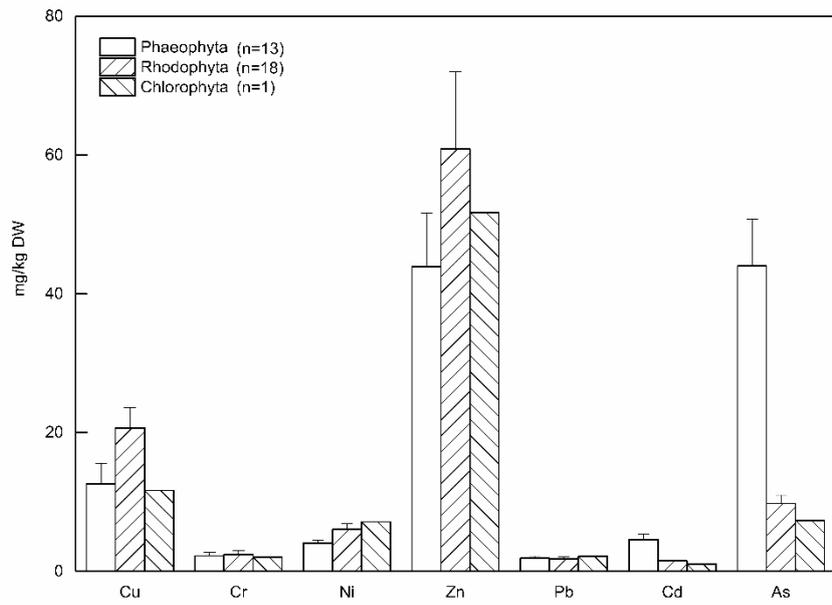
Fig. 1 Field sampling sites (S1-S6) in Dongtou Islands, East China Sea (i.e., S1 represents the sampling site 1)

No.	Name	Picture
1	<i>Sargassum fusiforme</i>	
2	<i>Sargassum thunbergii</i>	
3	<i>Sargassum vachellianum</i>	
4	<i>Pachydictyon coriaceum</i>	
5	<i>Polyopes polyideoides</i>	
6	<i>Gelidium divaricatum</i>	
7	<i>Chondracanthus intermedius</i>	
8	<i>Ahnfeltiopsis flabelliformis</i>	
9	<i>Laurencia tropica</i>	
10	<i>Pterocliadiella capillacea</i>	
11	<i>Gracilaria lemaneiformis</i>	
12	<i>Ulva pertusa</i>	



310

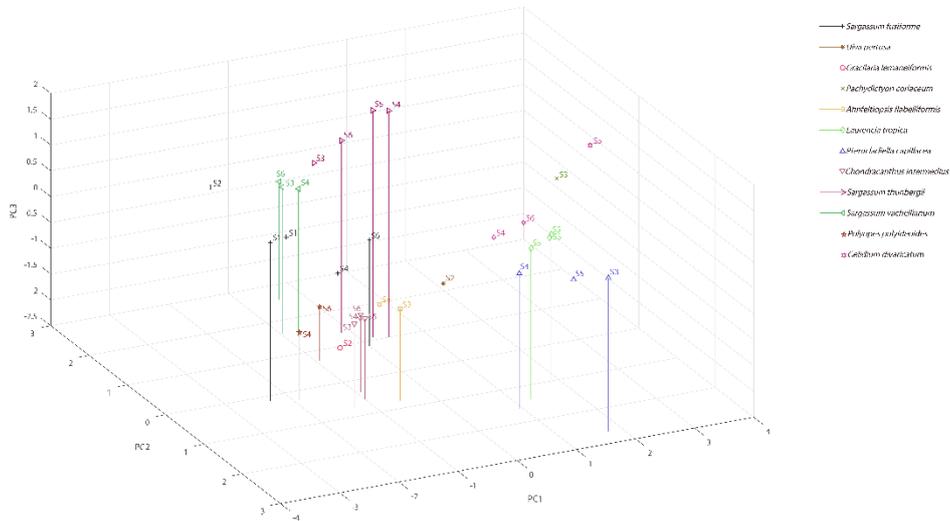
311 **Fig. 2** The seaweed species with specimen photos from field sampling, and their distributions in the
 312 tidal zones



313

314 **Fig. 3** Average concentrations of heavy metal determined in three seaweed phyla - Rhodophyta,

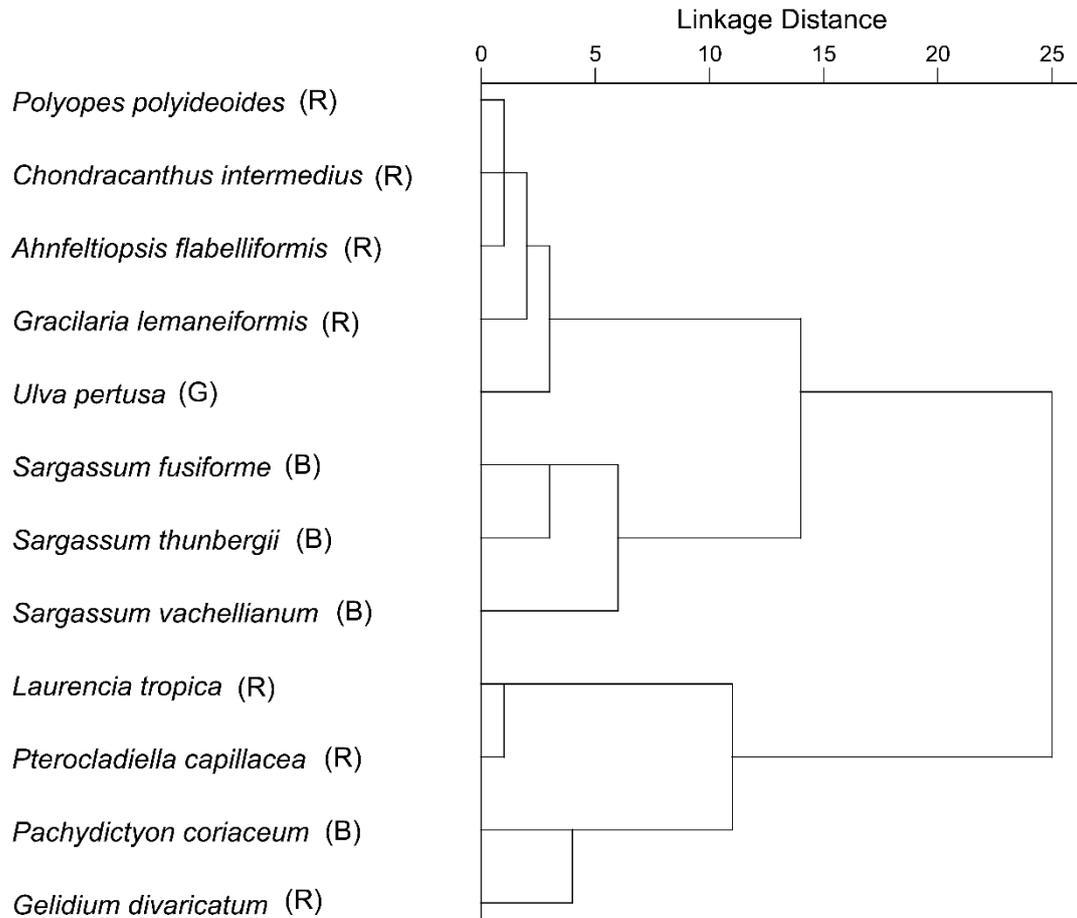
315 Phaeophyta, and Chlorophyta, with the error bars showing the standard errors



316

317 **Fig. 4** PCA of the heavy metal concentrations of seaweed species. S1-S6 represent the sampling site 1

318 to site 6



319

320

321 **Fig. 5** Cluster analysis on the heavy metal concentrations of seaweed species using Ward grouping
 322 method, B represents Phaeophyta (brown algae), R represents Rhodophyta (red algae), and G
 323 represents Chlorophyta (green algae)

324 **Table 1** Certified and detected tissue concentrations of heavy metals in the certified reference material
 325 Laver (GBW 10023)
 326

Element	Certified concentration (mg/kg DW)	Detected concentration (mg/kg DW)	Recovery (%)
Cu	12.200 ± 1.100 ^a	11.490 ± 0.020 ^b	94
Cr	2.400 ± 0.400	2.250 ± 0.030	94
Ni	2.250 ± 0.180	2.190 ± 0.040	97
Zn	28.000 ± 2.000	28.170 ± 0.020	101
Pb	2.050 ± 0.150	1.980 ± 0.040	97
Cd	0.570 ± 0.050	0.580 ± 0.010	102
As	27.000 ± 6.000	26.890 ± 0.060	100

327 ^a The range of certified tissue concentrations of heavy metals

328 ^b Mean ± standard error from three replicate detections

329 **Table 2** Seaweed species collected in the Dongtou Islands, East China Sea

330

	Latitude (°N)	Longitude (°E)	Phylum	Species
SITE1	27.8588	121.1618	Phaeophyta	<i>Sargassum fusiforme</i>
SITE 2	27.8590	121.1824	Phaeophyta	<i>Sargassum fusiforme</i>
			Rhodophyta	<i>Gracilaria lemaneiformis</i>
			Chlorophyta	<i>Ulva pertusa</i>
SITE 3	27.8646	121.1872	Phaeophyta	<i>Pachydictyon coriaceum</i>
				<i>Sargassum thunbergii</i>
				<i>Sargassum vachellianum</i>
			Rhodophyta	<i>Ahnfeltiopsis flabelliformis</i>
				<i>Laurencia tropica</i>
				<i>Pterocladia capillacea</i>
				<i>Chondracanthus intermedius</i>
SITE 4	27.8661	121.1901	Phaeophyta	<i>Sargassum thunbergii</i>
				<i>Sargassum vachellianum</i>
				<i>Sargassum fusiforme</i>
			Rhodophyta	<i>Ahnfeltiopsis flabelliformis</i>
				<i>Polyopes polyideoides</i>
				<i>Gelidium divaricatum</i>
				<i>Pterocladia capillacea</i>
	<i>Chondracanthus intermedius</i>			
SITE 5	27.8357	121.2325	Phaeophyta	<i>Sargassum thunbergii</i>
			Rhodophyta	<i>Gelidium divaricatum</i>
				<i>Pterocladia capillacea</i>
				<i>Laurencia tropica</i>
				<i>Chondracanthus intermedius</i>
SITE 6	27.8329	121.2496	Phaeophyta	<i>Sargassum thunbergii</i>
				<i>Sargassum vachellianum</i>
				<i>Sargassum fusiforme</i>
			Rhodophyta	<i>Polyopes polyideoides</i>
				<i>Gelidium divaricatum</i>
	<i>Chondracanthus intermedius</i>			

331 **Table 3** Mean Concentration (\pm SE) of heavy metal in seaweed species collected in the Dongtou Islands, East China Sea

332

333

Phylum	Species	Heavy metal concentration (mg/kg DW)						
		Cu	Cr	Ni	Zn	Pb	Cd	As
Phaeophyta	<i>Sargassum fusiforme</i> (n=5)	7.93 \pm 0.89	0.85 \pm 0.11	2.87 \pm 0.38	20.87 \pm 6.69	1.50 \pm 0.62	1.71 \pm 0.17	57.71 \pm 13.44
	<i>Sargassum thunbergii</i> (n=4)	12.81 \pm 0.83	3.84 \pm 0.47	6.04 \pm 0.53	40.69 \pm 4.67	2.00 \pm 0.24	5.63 \pm 0.12	49.08 \pm 2.46
	<i>Sargassum vachellianum</i> (n=3)	8.59 \pm 0.23	1.21 \pm 0.12	3.11 \pm 0.38	81.03 \pm 12.90	1.90 \pm 0.39	8.48 \pm 0.29	23.77 \pm 3.88
	<i>Pachydictyon coriaceum</i> (n=1)	47.62	5.9	5.23	61.22	3.34	3.19	16.12
Rhodophyta	<i>Polyopes polyideoides</i> (n=2)	3.89 \pm 0.07	0.92 \pm 0.25	1.57 \pm 0.16	63.17 \pm 15.69	1.32 \pm 0.11	1.70 \pm 0.17	15.41 \pm 0.05
	<i>Gelidium divaricatum</i> (n=3)	23.76 \pm 0.40	6.83 \pm 1.11	6.58 \pm 0.34	134.50 \pm 40.88	4.21 \pm 0.64	1.56 \pm 0.30	8.89 \pm 1.07
	<i>Gracilaria lemaneiformis</i> (n=1)	12.32	0.86	0.8	97.84	0.9	1.11	8.31
	<i>Ahnfeltiopsis flabelliformis</i> (n=2)	11.47 \pm 0.82	1.16 \pm 0.03	4.85 \pm 0.18	32.25 \pm 2.27	1.09 \pm 0.09	1.93 \pm 0.11	4.81 \pm 0.74
	<i>Laurencia tropica</i> (n=3)	27.80 \pm 1.31	3.01 \pm 0.18	11.38 \pm 0.44	44.79 \pm 1.28	1.92 \pm 0.19	1.92 \pm 0.23	9.72 \pm 2.70
	<i>Pterocladia capillacea</i> (n=3)	41.87 \pm 5.09	1.89 \pm 0.23	9.68 \pm 0.53	30.51 \pm 2.04	1.58 \pm 0.04	1.41 \pm 0.03	3.35 \pm 0.23
	<i>Chondracanthus intermedius</i> (n=4)	12.13 \pm 0.26	1.04 \pm 0.07	3.06 \pm 0.09	44.48 \pm 2.57	0.77 \pm 0.12	1.10 \pm 0.06	15.41 \pm 0.25
Chlorophyta	<i>Ulva pertusa</i> (n=1)	11.68	2.05	7.14	51.74	2.18	1.09	7.35

334 **Table 4** Bioaccumulation coefficient of heavy metal in seaweed species collected in the Dongtou
 335 Islands, East China Sea
 336

Heavy metal	Cu	Cr	Ni	Pb	Cd	As
concentration in seawater ($\mu\text{g/L}$) ^a	0.640	0.700	0.670	0.032	0.036	1.430
<i>Sargassum fusiforme</i>	12391	1214	4284	46875	47500	40357
<i>Sargassum thunbergii</i>	20016	5486	9015	62500	156389	34322
<i>Sargassum vachellianum</i>	13422	1729	4642	59375	235556	16622
<i>Pachydictyon coriaceum</i>	74406	8429	7806	104375	88611	11273
<i>Polyopes polyideoides</i>	6078	1314	2343	41250	47222	10776
<i>Gelidium divaricatum</i>	37125	9757	9821	131563	43333	6217
<i>Gracilaria lemaneiformis</i>	19250	1229	1194	28125	30833	5811
<i>Ahnfeltiopsis flabelliformis</i>	17922	1657	7239	34063	53611	3364
<i>Laurencia tropica</i>	43438	4300	16985	60000	53333	6797
<i>Pterocladia capillacea</i>	65422	2700	14448	49375	39167	2343
<i>Chondracanthus intermedius</i>	18953	1486	4567	24063	30556	10776
<i>Ulva pertusa</i>	18250	2929	10657	68125	30278	5140

337 ^a The values are annual average value of heavy metal concentrations in seawater in 2015, from the
 338 bulletin of coastal environment quality, released by the Ministry of Environment Protection, China

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