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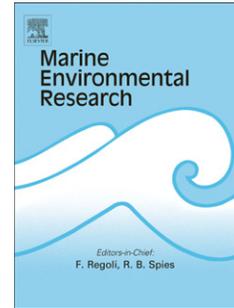
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1 **Ecotoxicological approach for assessing the contamination of a Hawaiian**
2 **coral reef ecosystem (Honolua Bay, Maui) by metals and a metalloid**

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16 **Abstract**

17 The goal of this study was to assess the contamination of Honolulu Bay using an
18 ecotoxicological approach. First, the concentrations of 9 contaminants (metals and metalloid)
19 were assessed in sediments and tropical marine organisms (alga *Halimeda kanaloana*, goatfish
20 *Parupeneus multifasciatus* and urchin *Tripneustes gratilla*) sampled from Honolulu and
21 surrounding Bays. Then, the ecological parameters characterizing coral health (e.g. coral cover)
22 were evaluated in Honolulu Bay in the context of these contaminants. High concentrations of Co,
23 Cr, Mn, Ni, and V in sediments from Honolulu and Honokohau Bay were measured, but these
24 concentrations were not mirrored in the organisms examined, except for Mn, suggesting that the
25 metals are generally bound in chemically inert forms in these sediments. Moreover, few
26 anthropogenic activities impact these bays and so the elevated Co, Cr, Mn, Ni and V
27 concentrations in sediments appear to stem from their high natural background in Honolulu and
28 Honokohau watersheds. An analysis of the relationship between the ecological parameters and
29 metal concentrations in Honolulu Bay revealed a significant correlation between coral cover and
30 Co, Cr, Mn, Ni, V, Zn concentrations in sediments, with coral cover decreasing with increasing
31 metal concentration. Collectively, however, the data suggest that a complex mixture of land-
32 based stressors (e.g. sediment, metals, nutrients) affect the coral health in Honolulu Bay, rather
33 than metal stress alone.

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37 **Keywords:** Hawaii, Ecology, Metals, Coral Reefs, Urchin, Algae

38 **1. INTRODUCTION**

39 Coral reefs are one of the most diverse and biologically complex marine ecosystems on
40 earth. However there is now deep concern whether coral reefs will survive shifts in the marine
41 environment associated with global changes and local stresses (e.g. Hoegh-Guldberg et al. 2007,
42 Wilkinson 1999). Bryant et al (1998) report that 58 percent of the world's reefs are at risk from
43 human activities, and that coastal development poses one of the most serious among these
44 threats. Indeed, the expansion of human populations in coastal areas and the accompanying
45 urbanization and agricultural activities, have increased the discharge of industrial wastes and
46 untreated sewages onto the adjacent coral reefs. Such local activities have resulted in high metal
47 concentrations in seawater, sediments, corals and tissues of other reef organisms sampled from a
48 number of tropical ecosystems (e.g. Brown and Holley 1982, Hanna and Muir 1990). The
49 adverse effects of metal contaminants on coral reefs have led to reduced growth and
50 reproduction, coral death and loss of biodiversity (e.g. Negri et al. 2002, Richmond 1993).
51 Metals have also been recognized as an environmental trigger that can induce bleaching of corals
52 (e.g. David 2003, Scott and Davies 1997). As such, metal contamination in urban aquatic
53 ecosystems is a major concern. In the Hawaiian Islands, a combination of basaltic rocks and
54 large human populations with high traffic densities have led to elevated concentrations of copper,
55 chromium, lead and zinc in streambed sediments (Andrews and Sutherland 2004, McMurtry et
56 al. 1995), which in some cases, exceed the aquatic-life guidelines (De Carlo et al. 2005). While
57 metal concentrations in streambed sediments from the Hawaiian Islands are relatively well
58 known, with the exception of two studies, one in Kane'ohu Bay (Hunter et al. 1995) and one in
59 French Frigate Shoals in the North Western Hawaiian Islands (Miao et al. 2001), concentrations
60 in sediments and biota from coastal reef areas are not well characterized. To address this

61 knowledge gap, the Coral Reef Land Based Pollution Local Action Strategy (CRLBP-LAS)
62 identified three watersheds in the main Hawaiian Islands as focal areas for studies to better
63 understand the threats that land-based contaminants pose to the near shore coral reef ecosystems.
64 Within this framework, an assessment of metal and metalloid concentrations in three reef sites
65 revealed very high concentrations of Co, Cr, Ni and Mn in sediments from Honolulu Bay, Maui
66 ($<464 \mu\text{g Ni g}^{-1} \text{dwt}$) as compared to the Southern fringing reef of Moloka'i and Kane'ohe Bay,
67 Oahu ($<27 \mu\text{g Ni g}^{-1} \text{dwt}$; Hédouin et al. 2009). Ni concentrations in Honolulu Bay were 9 times
68 higher than the Effective Range-Median for Sediment Quality Criteria (Long et al. 1995, NOAA
69 1999), which is in the range observed in reef areas impacted by Ni mining activities such as in
70 New Caledonia (e.g. 797 to 900 $\text{g Ni g}^{-1} \text{dwt}$, Hédouin et al. 2008a), and definitely high enough
71 to elicit adverse biological effects.

72 The difference between contamination and pollution is sometimes misunderstood and thus
73 the terms are often misused. All pollutants are contaminants, but not all contaminants are
74 pollutants (Chapman 2007, Warnau 1996). Elements or compounds are considered as
75 contaminants when they are in concentrations over natural background whereas contaminants are
76 considered as pollutants when they have an anthropogenic origin and negatively influence the
77 environment, organisms, human health or human activities (GESAMP 1984). Metals and
78 metalloid are naturally present in earth (Salomons and Forstner 1984), so their presence in
79 sediments and biota is not always indicative of pollution. For this reason, we use the term
80 contamination and contaminant to refer to the metals and metalloid in this paper.

81 The aims of the present study were to: 1) evaluate whether contaminant concentrations
82 measured in Honolulu Bay are unique to this bay by providing a comprehensive baseline of
83 Arsenic (As), cobalt (Co), chromium (Cr), copper (Cu), manganese (Mn), nickel (Ni), lead (Pb),

84 vanadium (V) and zinc (Zn) in reef sediments from Honolua Bay and adjacent bays in the West
85 Maui region; 2) assess the bioavailability of metals and metalloid to the reef biota (the alga
86 *Halimeda kanaloana*, the goatfish *Parupeneus multifasciatus* and the urchin *Tripneustes*
87 *gratilla*); and 3) explore the relationship between coral ecology and the gradient of metal and
88 metalloid contamination in Honolua Bay.

89 **2. MATERIALS AND METHODS**

90 **2.1. Description of Honolua Bay, West Maui, Hawai'i**

91 Honolua Bay has been a Marine Life Conservation District since 1978 and is divided into 3
92 broad regions (Fig. 1), that include (1) a fringing reef that extends into the bay and borders the
93 north and south shorelines with a coral cover averaging 10.3 % on the north side and 23.3 % on
94 the south side (CRAMP 2004), (2) a deeper sandy bottom area localized between the reefs, and
95 (3) an inner bay composed of silt, sand, boulder bottom with corals present but reefs absent. This
96 last area is the closest to the Honolua Stream and reflects this proximity by in its higher turbidity,
97 lower salinity and higher dissolved nutrients (e.g. NO_3^-) (Marine Research Consultants Inc
98 2007). Honolua Stream is an interrupted perennial stream with an average stream flow of 19
99 million liters d^{-1} in the upper elevations. A detailed description of coastal circulation and
100 sediment dynamics of the study area has been conducted by the United States Geological Survey
101 (USGS) (e.g. Storlazzi and Jaffe 2003, Storlazzi et al. 2004, Storlazzi and Presto 2005).

102 **2.2. Sample collection for metal and metalloid analyses**

103 Four stations were selected in Honolua Bay for this study, two in the inner bay (Honolua1 =
104 Hon1, Honolua3 = Hon3) and two on the reef bed offshore (Honolua2 = Hon2, Honolua4 =
105 Hon4; Fig. 1). To better understand the degree of contamination in West Maui relative to
106 Honolua Bay, other sampling sites were selected North of Honolua Bay (Honokohau Bay and

107 Lipoa Point) and South of Honolua Bay (Mokulei‘a Bay, Kapalua Bay and Honokeana Cove).
108 Reef sediments and organisms (depending on their availability) were collected from each of
109 these sites (Fig. 1). Honolua Bay is a Marine Life Conservation District and sampling of marine
110 organisms requires a permit from the Division of Aquatic Resources (DAR). Permits were issued
111 for collections of the alga *Halimeda kanaloana*, the goatfish *Parupeneus multifasciatus* and the
112 urchin *Tripneustes gratilla*, but not for corals. Indeed these organisms are proposed as potential
113 bioindicator species since they respond to most of the criteria that should be met by a
114 bioindicator species. They are widely distributed in the West coast of Maui, available all year
115 long and easy to collect (e.g. Moore 1966, Phillips 1990, Warnau et al. 1998). In addition since
116 the selected organisms represent different trophic levels, the combination of data from these
117 different organisms allow for a broader assessment of the bioavailability of metals and metalloid
118 present in the marine environment.

119 2.2.1. Sampling of marine organisms and sediments

120 All organisms were collected between September 12 - 23, 2007. *H. kanaloana* was collected
121 from Honolua Bay and Honokeana Cove. Epiphytes and sediments were removed from the algae
122 by gentle scrubbing and thorough rinsing with clean 5- μ m filtered seawater. *P. multifasciatus* (n
123 = 6; mean \pm S.D.: length: 24.2 \pm 1.7 cm) were collected in Honolua Bay by spear fishing and
124 samples of liver and muscle removed by dissection. *T. gratilla* between 79.7 and 84.2 mm in size
125 were collected on snorkel from Honolua Bay, Kapalua Bay, Lipoa Point and Honokeana Cove (n
126 = 7 per location, except for Lipoa Point n = 4). Urchins were kept alive for 24 h in plastic boxes
127 containing 30 L of 5- μ m filtered seawater (from the same sampling stations) to allow the gut
128 contents and particulates to be excreted and shed. Sampled animals were dissected to separate
129 gonads, intestine and body wall (including spines). The tissues sampled from each organism
130 were weighed (wet wt), dried at 60 °C until the weight was constant, and then re-weighed (dry

131 wt). Dried biological samples were hermetically sealed in PET containers until analyzed for
132 metal and metalloid contents.

133 Superficial sediments (50 - 100 g, n = 5 replicates) were collected from all 6 sampling
134 locations along the West coast of Maui (Fig. 1). These were placed in acid-washed PET bags and
135 frozen (-20°C) until analyzed. Each sample was dried at 60 °C until the weight was constant, (5
136 d), sieved (4-mm mesh size) to eliminate stones and fragment of corals, ground using a mortar
137 and pestle and stored in acid-washed and hermetically sealed in PET containers until analyzed
138 for metal and metalloid contents.

139 2.2.2. *Analyses*

140 As, Co, Cr, Cu, Mn, Ni, Pb, V and Zn were analyzed by the Agricultural Diagnostic Service
141 Center, at the University of Hawai'i. Aliquots of biological samples (0.5 g) and sediment
142 samples (0.5 g) were digested with 5 ml of 65 % HNO₃ (Merck, suprapur quality).
143 Concentrations of metals were determined using Atom San 16 ICP (Thermo Jarrell Ash
144 Corporation, Franklin, MA). Two control samples (one Certified Reference Materials -CRM-
145 lobster hepatopancreas TORT-2 (NRCC) and one blank) treated and analyzed in the same way as
146 the samples were included in each analytical batch. The results for CRM indicated recoveries
147 ranging from 86 % (Cu) to 115 % (Pb). Mean element concentrations were given on a dry weight
148 basis ($\mu\text{g g}^{-1}$ dwt).

149 2.2.3. *Statistical analyses*

150 Comparisons of the data were performed using one- or two-way analysis of variance
151 (ANOVA) followed by the multiple comparison test of Tukey (Zar 1996). Two-way ANOVA
152 was used with sampling location and body compartment as fixed factors. The variability
153 explained by each factor and their interactions were derived from the sum of squares (Zar 1996).
154 The correlation and statistical significance of the relationship between the metal concentrations

155 in sediments and those in different body compartments of the urchin *P. lividus* were evaluated
156 using Pearson's correlation coefficient with the software R (Hornik 2010). The level of
157 significance for statistical analyses was set at $\alpha = 0.05$.

158 **2.3. Coral ecology survey**

159 Ecological surveys were performed at the four stations in Honolulu Bay (depth ranging from
160 1.5 to 2.5 m) to evaluate whether any traits in the coral ecology were correlated with the gradient
161 of metal and metalloid concentrations observed in sediments, (Fig. 1). Six 20 m long transects
162 were deployed randomly at each station and sampled using a line-intercept method (Hill and
163 Wilkinson 2004). Percent cover was estimated by dividing the substrate into 6 categories:
164 boulder bottom, reef bed, sand, alga, dead coral, and live coral. Individual coral colonies under
165 each transect were identified to species and the size of the colony approximated by measuring
166 maximum length, width and height. The condition of the colony was assessed for partial
167 mortality as defined as percentage of the surface area of the colony that was dead. Colonies were
168 also assigned to a category based on color. The color categories used were healthy, bleached,
169 partially bleached and pale. Corals classified as healthy showed no signs of tissues discoloration;
170 bleached coral colonies were totally white; partially bleached corals had patches of fully white
171 tissues interspersed within the darker colored healthy tissues; and pale corals were globally paler
172 in color than healthy colonies of the same species. Hobo® data loggers were deployed at each
173 station (1.5 to 2.5 m depth) to record temperature and light over 6 d at a sampling interval of 15
174 min. Seawater samples (1500 ml, n = 3 per station) were collected on September 17th, 18th and
175 19th, 2007 and filtered through GF/C filter to estimate the turbidity of seawater (mg l^{-1}).

176 The coral cover was compared among stations using one-way ANOVA and appropriate post-
177 hoc multiple range tests. Relationships between ecological parameters, environmental variables
178 and sediment associated metal and metalloid concentrations were examined using correlation

179 (Spearman) and linear regression analysis using average values for whole station reefs using
180 SPSS® statistical software 16.0 (SPSS Inc., Chicago, IL, USA).

181 **3. RESULTS**

182 **3.1. Sediments**

183 *3.1.1. Honolua Bay*

184 The four stations in Honolua Bay can be ranked according to their degree of contamination
185 Hon1 > Hon3 > Hon2 > Hon4 with Hon1 being the most contaminated by all elements except Cu
186 and Fe, and Hon4 the least (Table 1). Statistical analyses of the data are presented in Figure 2-
187 A1. Results indicate that the concentrations of Co, Mn, Ni, V and Zn in sediments were
188 significantly different among all stations. Concentrations of Cr and Cu were similar at Hon1 and
189 Hon3, and at Hon2 and Hon4, respectively. No significant differences were observed for the Fe
190 concentrations in sediments at Hon1, Hon2 and Hon3. There was no significant difference in the
191 concentration of Pb measured in sediments at the four stations. For As, no significant difference
192 was observed in sediments between station Hon1 and Hon3, and Hon1 and Hon2.

193 *3.1.2. Honolua Bay and other adjacent Bays*

194 One-way ANOVA was performed on element concentration measured in sediments collected
195 from the different bays in the West Maui. For Honolua Bay, Hon1 and Hon4 were chosen for the
196 statistical analyses because they have the broadest range of contamination (Fig. 2-A1).
197 Sediments collected at Honokohau Bay were more contaminated than those collected from the
198 other bays surrounding Honolua Bay and the concentrations of contaminants were in the range of
199 those measured in the most contaminated stations of Honolua Bay (Hon1 and Hon3, Table 1).
200 For example, Ni concentrations were 374 $\mu\text{g g}^{-1}$ dwt and 303 - 481 $\mu\text{g g}^{-1}$ dwt in Honokohau Bay
201 and Honolua Bay (Hon1 and Hon3), respectively. In addition, statistical analyses (Fig. 2-A2)

202 indicate that the concentrations of most contaminants measured in sediments from Hon1 and
203 Honokohau Bay were generally higher than those measured in sediments from other bays, and
204 that concentrations of Co, Fe, Mn, V and Zn measured in sediments from Hon1 and Honokohau
205 Bay were not significantly different from one another. In contrast, metal and metalloid
206 concentrations measured in sediments from Hon4, Lipoa Point, Kapalua Bay and Honokeana
207 Cove were similar and lower than Honokohau Bay and Honolulu sites Hon1 and Hon3. No
208 significant difference was observed for Co, Cr, Cu, Ni, Pb, V and Zn among sediments from
209 Hon4, Lipoa Point and Kapalua Bay.

210 3.1.3. Sediment Quality Guidelines

211 Element concentrations in sediments from all stations were compared to the NOAA
212 Sediment Quality Guidelines (Long et al. 1995, NOAA 1999) that were developed to define the
213 concentrations of metals and metalloid that have adverse effects on biological organisms. Within
214 this framework, the Threshold Effect Level (TEL) represents the concentration below which
215 adverse effects rarely occur; the Effect Range-Low (ERL) corresponds to concentration above
216 which negative effects are more common, and the Effect Range-Median (ERM) corresponds to
217 concentrations at or above which negative effects frequently occur. Results of the comparison are
218 presented in Table 1.

219 3.2. Reef biota

220 Except for Fe ($p_{\text{Tukey}} < 0.05$) and Mn ($p_{\text{Tukey}} < 0.01$) there was no significant difference in
221 the element concentrations measured in the alga *H. kanaloana* collected from Honokeana Cove
222 and Honolulu Bay (Table 2).

223 Concentrations of Co, Cu, Fe, Mn, and Zn measured in the livers of the goatfish *P.*
224 *multifasciatus* were one to two orders of magnitude higher than in the muscle (Table 2). Very
225 high Fe concentrations were recorded in goatfish livers reaching 4,400 $\mu\text{g g}^{-1}$ dwt compared to

226 $16 \mu\text{g g}^{-1}$ dwt in muscle. Since individuals of *P. multifasciatus* were all collected from Honolua
227 Bay, it was not possible to compare concentrations measured with other geographical areas.

228 The element concentrations measured in the gonads, intestines and body wall of the
229 urchin *T. gratilla* collected from stations inside and outside Honolua Bay are presented in Table
230 2. Statistical analyses performed on the element concentrations in urchins collected from stations
231 inside Honolua Bay (Hon1 and Hon4) and in Honokeana Cove, Lipoa Point and Kapalua Bay
232 indicated that body compartment explained the majority of the variability for As, Cu, Fe, V, and
233 Zn (accounting for 52 to 89 % of the global variance). Sampling station explained 40 % of the
234 variability for Mn and 23 to 30 % of the variability observed for Cr and Cu, respectively. For Co,
235 Cr, Cu, Ni, Pb, and Zn, an important part of the variation was associated with the residual term,
236 ranging from 28 to 49 %, indicating that other, non-investigated factors (biological and/or
237 environmental factors) were also influencing element concentrations in these urchin tissues.
238 Multiple comparison tests on the mean concentrations of elements in each sampling location (all
239 body compartments together) indicated that the concentrations of As, Co, and Mn were
240 significantly higher ($p_{\text{Tukey}} < 0.05$) in urchins from station Hon1 compared to those from the four
241 other stations (Fig. 2-B1). Cr and Cu concentrations in urchins from station Hon4 were
242 significantly higher ($p_{\text{Tukey}} < 0.05$) than those measured in urchins from Honokeana Cove. Ni
243 concentrations in urchins from Hon1 and Hon4 were not significantly different, but significantly
244 higher than those from urchins collected at Honokeana Cove and Kapalua Bay ($p_{\text{Tukey}} < 0.05$).
245 No significant difference was observed for Zn concentrations in urchins from the five stations.
246 Multiple comparison tests on the mean concentrations in each body compartment (all sampling
247 locations together) indicated that the concentrations of As, Co, Cu, Fe, Mn and Ni were
248 significantly higher ($p_{\text{Tukey}} < 0.01$, except for Co $p_{\text{Tukey}} = 0.05$) in the intestine than in the other

249 body compartments (Fig. 2-B2). Zn concentrations in intestine and gonads were not significantly
250 different, but significantly higher ($p_{\text{Tukey}} < 0.0001$) than those in body wall. Cr, Pb and V
251 concentrations were significantly higher ($p_{\text{Tukey}} < 0.0001$) in the body wall compared to the other
252 body compartments.

253 Concentrations of As, Co, Fe and Mn in intestine, showed significant positive
254 correlations with their respective concentrations in sediments (Pearson coefficient from 0.51 to
255 0.80, see Fig. 3). Significant correlations were also observed for Co, Mn, Ni and Zn between
256 concentration in urchin gonads and sediments (Pearson coefficient from 0.45 to 0.78, Fig. 3). Mn
257 concentrations in urchin carapaces were also significantly correlated with Mn sediment
258 concentrations (Pearson coefficient of 0.87, Fig. 3).

259 3.3. Ecological survey

260 In Honolulu Bay, total live coral cover was 10.9 and 13.4 % at the two inner stations (Hon1
261 and Hon3, respectively) and 17.4 and 26.2 % on the two offshore stations (Hon2 and Hon4,
262 respectively) (Fig. 4). Live coral cover was significantly lower at station Hon1 compared to
263 Hon4 ($p_{\text{Tukey}} = 0.017$), while no significant difference was observed among the other stations.
264 Data from the ecological survey clearly indicated the presence of two types of stations in the bay.
265 Stations Hon1 and Hon3 were characterized by a high percentage of boulder bottom (from 64 to
266 78 %), whereas the stations offshore were mainly composed of reef bed structure (38 to 81 %)
267 (Fig. 4). The major species encountered in order of abundance were *Porites lobata* (percent of
268 total number of colonies = 36.3 %), *Montipora patula* (17.7 %), *Montipora capitata* (14.6 %),
269 *Pocillopora meandrina* (14.2 %), and *Porites compressa* (6.6 %) (Fig. 4). These five species
270 were present at all four stations in Honolulu Bay, except *P. compressa*, which was absent from
271 Hon1. Other coral species were present but at low abundances (< 5 %).

272 Based on the color categories defined in § 2.3, corals present at station Hon2 were mainly
273 healthy (78 %) compared to the other three stations where corals showed more pronounced signs
274 of bleaching (56 - 77 %, Fig. 4). Statistical analyses (one-way ANOVA) were performed on the
275 diameter of colony, partial mortality and the proportion of the colony that was bleached (Pale,
276 Bleach and Partially Bleach) (after $\arcsin(\sqrt{x})$ transformation) for the five most abundant coral
277 species present in Honolua Bay (*P. lobata*, *M. patula*, *M. capitata*, *P. meandrina* and *P.*
278 *compressa*). Results indicated that there was no significant difference in the colony diameters
279 and partial mortality among stations except for *P. lobata*. Colonies of *P. lobata* had a
280 significantly higher diameter at station Hon4 as compared to the three other stations (Hon4 >
281 Hon2 = Hon3 = Hon1; $p_{\text{Tukey}} < 0.002$), and a higher percentage of partial mortality (Hon4 =
282 Hon2 > Hon3 = Hon1; $p_{\text{Tukey}} < 0.05$).

283 Results of the correlation matrices performed on the different ecological parameters and
284 environmental conditions (element concentrations) indicated that total live coral cover was
285 negatively correlated with Co, Cr, Mn, Ni, V, and Zn concentration in sediments (-0.955 to -
286 0.979, $p_{\text{Pearson}} < 0.05$, Table 3), but significant linear regressions were only observed for Co and
287 V (Fig. 5). In addition, for the coral *P. compressa*, partial mortality was correlated positively to
288 the diameter of the colony (0.568, $p_{\text{Pearson}} < 0.05$).

289 The turbidity measured in Honolua Bay ranged from 14.5 (Hon4) to 68.2 (Hon1) mg l^{-1} . No
290 significant difference was observed among the four stations in Honolua Bay on September 17
291 and 19, 2007, whereas the turbidity at Hon1 on September 18, 2007 was significantly higher by a
292 factor of 3.8 compared to the other three stations in the bay (Fig. 6). Seawater temperature in
293 Honolua Bay ranged from 26.2 to 28.5 °C and no difference was observed among the four
294 selected stations. The salinity of seawater measured at the surface and 1.50 m depth ranged from

295 36.2 to 36.9 parts per thousand (ppt) except for September 18, 2007, when a rain event occurred.
296 At the latter date, salinity of seawater decreased to 18.2 ppt at the Hon1, and a slight decrease of
297 salinity was also observed at the three other stations (32.1 to 35.6 ppt) compared to the other
298 sampling times (September 17 and 19, 2007).

299 **4. DISCUSSION**

300 Sediments are sinks for contaminants (Salomons et al. 1987), therefore the measurement of
301 metal and metalloid concentrations in reef sediments provides basic and essential information on
302 the degree of contamination of the reef environment. Element concentrations measured in
303 sediments collected from Honolulu Bay displayed a very wide range of concentrations, from 2.71
304 $\mu\text{g g}^{-1}$ dwt for Co up to 7,023 $\mu\text{g g}^{-1}$ dwt for Fe. Most interestingly, some elements such as Co,
305 Cr, Mn, Ni and V reached very high concentration (e.g. up to 481 $\mu\text{g g}^{-1}$ dwt for Ni). A recent
306 study has confirmed that the concentration of Co, Cr, Mn, Ni and V in reef sediments from
307 Honolulu Bay are elevated when compared to two other watersheds in Hawaii (SE Moloka'i and
308 Kane'ohe Bay) (Hédouin et al. 2009). In addition, among the four stations investigated, a clear
309 gradient of element concentrations was observed, suggesting that the degree of contamination
310 present in Honolulu Bay is related to the discharge of sediments from Honolulu Stream. To better
311 understand the source of the elevated concentrations of metal and metalloid contamination in
312 Honolulu Bay, we investigated element concentrations in sediments from bays surrounding
313 Honolulu Bay. Concentrations of all elements measured in sediments collected at Honokohau
314 Bay, located to the North of Honolulu Bay, were very similar to the concentrations of
315 contaminants measured in the most contaminated stations in Honolulu Bay, Hon1 and Hon3, (e.g.
316 376 - 594 vs. 492 $\mu\text{g Mn g}^{-1}$ dwt, 303 - 481 vs. 374 $\mu\text{g Ni g}^{-1}$ dwt, for Honolulu Bay - Hon1 and
317 Hon3- and Honokohau Bay, respectively). Comparison of As, Cr, Cu, Ni, Pb concentrations in

318 sediments from Honolulu Bay and/or Honokohau Bay with Sediment Quality Guidelines (NOAA
319 1999) indicate that Cr, Ni, and Pb are in high enough concentration to elicit negative effects on
320 biota. The similarity observed in element concentrations of sediments from both bays suggests
321 that these bays may be contaminated by the same source. Interestingly, a ditch (Honokohau
322 ditch) was built in 1904 to divert the flow of the Honokohau stream for off-stream uses (Fontaine
323 2003). The Honokohau ditch is 2.9 km long and links Honolulu Bay and Honokohau Bay. This
324 connection may explain why the metal concentrations observed in sediments of the two bays are
325 so similar. Honolulu and Honokohau watersheds are mainly forested (69 and 83 %, respectively;
326 Parham et al. 2008), and support few anthropogenic activities. This indicates that the elevated
327 concentrations of Co, Cr, Mn, Ni, V and Zn reflect the geology of the area and stem from high
328 natural background in both Honolulu and Honokohau watersheds as suggested by Hédouin et al.
329 (2009) for Honolulu Bay.

330 Regardless of the origin of the high concentrations of metals observed in sediments of
331 Honolulu Bay, the question is whether the contaminants are bioavailable and are adversely
332 affecting the health of reef biota. Ni bound in the lattice of naturally occurring silicate minerals
333 (e.g., olivine or pyroxenes) is less available for uptake by marine organisms as compared to
334 water-soluble forms (CEPA 1994). That said, it is extremely difficult to predict how sediment-
335 bound metals bioaccumulate in aquatic organisms in any given location because their
336 bioavailability depends on their mineralogical origin (Kabata-Pendias 2001), characteristics of
337 the sediments such as pH, oxidation-reduction potential, ionic strength, type, and concentration
338 of organic and inorganic ligands (e.g. Callahan et al. 1979, Snodgrass 1980), and biological and
339 physiological traits of the organism (e.g. species, age, sexual cycle, Boyden 1977, Cossa et al.
340 1979, Eisler 1981).

341 In this context, the measurement of elements in reef organisms from different trophic levels
342 (alga, fish, urchins) collected from Honolua Bay and surrounding bays provides a snapshot view
343 of the bioavailability of metals and a metalloid from the various sampling stations. The alga *H.*
344 *kanaloana* was chosen to investigate the metal bioaccumulation in Honolua Bay because it is
345 abundant in these areas and *Halimeda* species are generally efficient bioaccumulators of metals
346 (Al-Shwafi and Rushdi 2008). Comparison of metal and metalloid concentrations measured in
347 alga from Honolua Bay and Honokeana Cove indicated that while the sediments in these bays
348 are characterized by very different degrees of contamination, with the exception for Fe and Mn,
349 element concentrations in alga were low and similar between sites, and in the same range of
350 concentrations reported in the literature for *Halimeda* species (see Table 4). These results suggest
351 that either there are low concentrations of dissolved metals in Honolua Bay (with the exception
352 of Mn and Fe) or this species of *Halimeda* does not bioaccumulate metals efficiently. The
353 concentrations of contaminants in the goatfish *P. multifasciatus* from Honolua Bay are also low
354 and similar to those measured in other non-contaminated areas (e.g. Mauritania coast, Manila,
355 French Frigate Shoals -Hawai'i- see Table 4), supporting the conclusion that the contaminants in
356 sediments from Honolua Bay are not bioavailable to this fish species.

357 The urchin *T. gratilla* collected in Honolua Bay had higher concentrations of As, Co, Cr, Cu,
358 Mn and Ni than those from less contaminated stations along the West Coast of Maui (e.g.
359 Honokeana Cove, Kapalua Bay). However, while a clear gradient of As, Co, Cr, Cu and Ni
360 contamination was observed in sediments, the trend was less pronounced in urchin tissues (and
361 non-existent for algae) and significant correlations between metal concentrations in sediments
362 and those in the urchins *T. gratilla* were only observed for some metals (e.g. Co, Mn, Ni) and for
363 certain urchin body compartments. For example, Ni concentration in sediments collected inside

364 Honolua Bay were up to 48 times higher than those measured in sediments outside the bay,
365 whereas Ni concentration measured in urchin intestines from the same stations only differed by a
366 factor of 4. However, compared to the elevated concentrations of Ni reported in sediments from
367 Honolua Bay (up to $481 \mu \text{g g}^{-1}$ dry wt at Hon1), the Ni concentrations reported in urchin tissues (<
368 $2.9 \mu \text{g g}^{-1}$ dwt), and alga (< $3.9 \mu \text{g g}^{-1}$ dwt) were low. These concentrations are also low
369 compared to concentrations of Ni measured in reef organisms living in areas highly impacted by
370 Ni mining. For example, in the New Caledonia lagoon, concentrations of Ni reached $99.7 \mu \text{g Ni}$
371 g^{-1} dwt in the digestive gland of the clam *Gafrarium tumidum* in a site with $797 \mu \text{g g}^{-1}$ dwt of Ni
372 in sediments (Hédouin et al. 2008a) and $125 \mu \text{g Ni g}^{-1}$ dwt in the alga *Lobophora variegata*
373 living in a site with $900 \mu \text{g g}^{-1}$ dwt of Ni in sediments (Hédouin et al. 2008a, Hédouin et al.
374 2008b). These studies demonstrate that high Ni concentrations are measurable in marine tropical
375 organisms when Ni is present in a bioavailable form. As such, data presented for the reef
376 organisms in Honolua Bay collectively suggest that despite high concentration of Ni in
377 sediments of Honolua Bay, Ni is bound in the sediments in a chemically inert form that is not
378 bioavailable for the selected reef organisms in the present study.

379 In contrast to Ni, Mn concentrations were found in relatively equal proportions in the reef
380 sediments and urchins, as confirmed by the significant correlations observed between Mn
381 concentrations in sediments and the three body compartments of the urchins *T. gratilla*. For
382 example, Mn concentrations were 7 times higher in sediments sampled at Hon1 than at Hon4,
383 and 3.8 (tissues) to 7 (body wall) times higher in urchins from Hon1 compared to Hon4. This
384 suggests that Mn is bioavailable in Honolua Bay. The higher concentration of Mn in algae from
385 Honolua Bay as compared to Honokeana Cove also supports the presence of bioavailable Mn in
386 Honolua Bay. Mn is an essential metal involved in many metabolic processes (Simkiss and

387 Taylor 1981) and it is highly bioaccumulated in marine organisms (e.g. Howe et al. 2004). Mn is
388 currently not included in the Sediment Quality Guidelines, probably because it is thought to have
389 low toxicity for marine species of phytoplankton, invertebrates and fish (e.g. ranging from 1.5
390 mg l^{-1} (5-day EC_{50}) for the marine diatom *Ditylum brightwellii* based on growth inhibition to 300
391 mg l^{-1} (168-h LC_{50}) for the softshell clam *Mya arenaria*, Howe et al. 2004). However, after 7-
392 days exposure to $> 0.01 \text{ mg l}^{-1}$ Mn, embryos of the yellow crab *Cancer anthonyi* exhibited
393 reduced survival and hatching (Macdonald et al. 1988), suggesting that early life stages of
394 organisms are sensitive to Mn. This metal has also been reported to induce iron deficiency in
395 some algae, leading to inhibition of chlorophyll synthesis (e.g. blue green alga *Anacystis*
396 *nidulans*, Csatorday et al. 1984; green algae *Ulothrix minuta* and *U. fimbriata*, Rousch and
397 Sommerfeld 1999). Given that corals rely on the photosynthetic activities of endosymbiotic
398 dinoflagellates for nutrition, the impact of Mn on their biology could have a very negative effect.
399 Further studies are needed that focus on the sensitivity of different life stages of corals to Mn to
400 resolve the potential impact.

401 With the exception of Mn, the contaminants measured here displayed low bioavailability for
402 reef biota. However, the gradient of metal concentrations observed in sediments from the four
403 stations in Honolua Bay correlated with concentrations of sediment discharge into Honolua Bay
404 from Honolua Stream. This provided the ecological context to examine how different levels of
405 land-based contaminants influences the coral ecology. Interestingly, while stations located in the
406 inner part of the bay were mainly composed of boulder bottom (64 - 78 %), corals were present
407 at all sites, albeit with lower coral cover (10.9 and 13.4 % vs. 17.4 and 26.2 % for inshore and
408 offshore stations, respectively), and lower species diversity (5 vs. 9 species for inshore vs.
409 offshore stations). In addition, a shift in distribution of abundant species was also observed with

410 the coral *Porites compressa* being more abundant in offshore than inshore stations, and
411 *Montipora patula* more abundant in inshore than offshore stations. Although the inshore stations
412 are rarely included in ecological survey of Honolua Bay, our results are consistent with Torricer
413 et al. (1979), who surveyed corals in close proximity to Honolua Stream and recorded values of
414 1.3 to 9.5 %. Previous surveys in Honolua Bay have reported differences in coral community
415 structure between North and South Bay. For example, *Porites* sp. dominate the north reef and
416 sediment resistant *Montipora* sp. dominate the southern reef (Brown 2004). Interestingly, in our
417 study, *P. lobata* was the most abundant species (32 - 39 %) in all four stations investigated. This
418 is surprising given that *P. lobata* is reported to have a lower active sediment rejection rate than
419 other species (Stafford-Smith 1993), and in Hawaiian Islands *P. lobata* is mainly observed in
420 regions of moderate wave actions (Jokiel et al. 2004). These preferences are clearly reflected in
421 the ecology of corals along the South-East fringing reef of Moloka'i, where *P. lobata* is present
422 in stations close to the reef crest (> 700m from shore) where sediments are mainly composed of
423 gravel and coarse sand (38 to 93 %), but generally absent in near shore stations where sediments
424 are fine and muddy (up to 44 % of mud) (Hédouin et al. Personal Communication, Rodgers et al.
425 2005). That said, *P. lobata* is sometimes found in areas with high sedimentation and/or turbidity
426 (Australia, Ayling and Ayling 1987, Bull 1982; Pandora Reef, Palm Islands, Potts et al. 1985).
427 Piniak (2007) recently identified a possible reason for these inconsistencies in documenting a
428 greater negative impact on the fluorescence yield of *P. lobata* from a short-term sedimentation
429 event (< 30 h) of 1,500 to 2,800 mg cm⁻² composed of terrigenous sediments than the same
430 sedimentation event with carbonate sand. These data indicate that *P. lobata*, is more sensitive to
431 terrigenous fine particle of sediments than to coarse sand. This may explain why *P. lobata* is
432 observed in the inner sites at Honolua Bay (Hon1 and 3), where coarse sand sediments are

433 present (48 – 64 % of coarse sand, Hédouin et al. 2009). Finally, the coral cover in the four
434 stations selected in Honolua Bay reflected the gradient of contaminant concentration observed in
435 the Bay, with significant correlation observed for Co, Cr, Mn, Ni, V and Zn. However, since
436 most of these contaminants (with the exception of Mn) are not bioavailable for the marine
437 organisms assessed in this study, these correlations likely reflect the complex mixture of land-
438 based contamination stressors (e.g. increasing turbidity, decreasing salinity) discharged from
439 Honolua Stream during rainfall events (as during the event recorded on September 18th, 2008)
440 rather than metal stress alone. In the context of the low metal bioavailability in sediments for the
441 reef organisms, metal concentrations in sediments appears more useful as a proxy of the land-
442 based contamination present at each site.

443 **5. CONCLUSION**

444 The sediments in the inshore stations in Honolua Bay displayed very high concentrations of
445 metals and a metalloid, but the concentrations of most of these compounds measured in reef
446 biota (alga, goatfish and urchins) are comparable to concentrations observed in non-
447 contaminated areas (Tables 4, 5). This suggests that with the exception of Mn, the contaminants
448 present in the marine environment of Honolua Bay exhibit low bioavailability for reef biota. The
449 data demonstrate that the bioavailability of metals and metalloid in Honolua Bay is different
450 depending on the element under consideration. For example, Mn is more bioavailable than Ni
451 even though the gradients of metal concentrations observed in sediments for these two elements
452 were relatively similar. Nevertheless, given that the bioavailability of metals in marine organisms
453 is influenced by physical and chemical condition such as pH and temperature, the bioavailability
454 of these contaminants may change in the context of global warming in the future. In addition,
455 further works should focus on assessing the bioaccumulation and effects of contaminants on

456 Scleractinian corals present in Honolua Bay, since the sensitivity of corals may be different from
457 the other organisms selected in this study. The high concentration of sediment associated metal
458 and metalloid concentrations highlight the influence of Honolua Stream in discharging sediments
459 into Honolua Bay. Other chemical materials associated with agriculture, such as pesticides, that
460 were not measured here are also likely to be associated with these sediments and to contribute to
461 the complex mixture of threats facing the corals of Honolua Bay. Alleviating local stress level by
462 reducing sediment discharges into Honolua Bay is a feasible and direct management action that
463 has the potential to help coral reefs better face global stressors that are less easy to ameliorate.
464 Such activity has thus the potential to contribute significantly to the conservation of coral reef
465 ecosystem of Honolua Bay.

466

467

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Figure 1. Selected sampling locations for reef sediments, the alga *Halimeda kanaloana*, the goatfish *Parupeneus multifasciatus* and the urchin *Tripneustes gratilla*, along the West coast of Maui, Hawaii

Figure 2. Comparisons of element concentrations in (A) sediment collected (A1) inside and (A2) inside and outside Honolua Bay, using multiple comparison test of Tukey performed after one-way ANOVA, and (B) the urchin *Tripneustes gratilla* using multiple comparison test of Tukey performed after two-way ANOVA (B1, Geographical variation; B2, Body compartment). Mean concentrations are ranked from the left to the right by decreasing order. Concentrations in underlined body compartments or stations are not significantly different ($\alpha = 0.05$).

Body compartments: *Intestinal tissues* (I), *Gonad* (G), *Body wall* (B)

Sampling locations: *Honolua 1* (Hon1), *Honolua 2* (Hon2), *Honolua 3* (Hon3), *Honolua 4* (Hon4), *Honokeana Cove* (HC), *Lipoa Point* (LP), *Kapalua Beach* (KB), *Honokohau Bay* (HB) and *Mokulei'a Bay* (MB)

Figure 3. Element concentrations in three body compartments of the urchin *Tripneustes gratilla* (intestine, gonads and carapace) as a function of their respective element concentrations in sediments. (“*” indicate that the significant correlation was observed between metal concentration in urchin body compartments and those in sediments; * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$, **** $p < 0.001$)

Figure 4. Ecological parameters of four stations in Honolua Bay. (A) Percent distribution of various bottom types (Live coral, dead coral, boulder bottom, alga, sand and reef bed); (B)

Percentage (Distribution frequency) of coral colonies measured at each station; (C) Percent distribution of coral colors (Healthy; Pale, Partially bleached, Bleached); (D) Average colony diameters for the five most abundant coral species (Mean \pm S.D.); (E) Percentage of mortality of the five most abundant coral species.

Species codes: *Porites lobata* (PLOB), *Porites compressa* (PCOM), *Montipora capitata* (MCAP), *Montipora patula* (MPAT), *Pocillopora meandrina* (PMEA)

Station codes: *Honolua 1* (Hon1), *Honolua 2* (Hon2), *Honolua 3* (Hon3), *Honolua 4* (Hon4)

Figure 5. Linear regressions of mean live coral cover versus metal concentration. Mean total coral cover vs. (A) Co and (B) V concentration in sediments, all stations. Coefficient of determination and significance shown only for significant regressions.

Figure 6. Turbidity (Mean \pm S.D.) (A) and temperature (B) of seawater at the four stations in Honolua Bay over time. A rain event occurred on September 18th, 2007.

Station codes: *Honolua 1* (Hon1), *Honolua 2* (Hon2), *Honolua 3* (Hon3), *Honolua 4* (Hon4)

Metal and metalloid levels in sediments and tropical organisms from West Maui.

Ecological surveys of corals in Honolua Bay.

Elevated levels of Co, Cr, Mn, Ni and V in reef sediments.

Low bioavailability of contaminants for organisms.

Correlations between coral cover and Co, Cr, Mn, Ni, V, Zn levels in sediments.

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Sites	As m ± S.D.	Co m ± S.D.	Cr m ± S.D.	Cu m ± S.D.	Fe m ± S.D.	Mn m ± S.D.	Ni m ± S.D.	Pb m ± S.D.	V m ± S.D.	Zn m ± S.D.
Honolua 1	39.2 ± 4.8 ^b	48.4 ± 7.3	95.9 ± 12.0 ^b	13.9 ± 2.5	7013 ± 175	594.4 ± 49.8	481.3 ± 62.1 ^c	36.2 ± 4.6 ^a	346.1 ± 48.3	54.9 ± 4.4
Honolua 2	34.3 ± 2.3 ^b	16.9 ± 2.5	40.7 ± 5.4	7.08 ± 1.34	7023 ± 171	249.8 ± 26.4	151.4 ± 34.8 ^c	19.0 ± 3.3	179.5 ± 26.3	20. ± 2.0
Honolua 3	42.5 ± 1.5 ^b	36.6 ± 1.2	93.5 ± 4.3 ^b	17.1 ± 1.3	7018 ± 172	375.8 ± 12.2	302.8 ± 16.5 ^c	34.7 ± 4.5 ^a	269.7 ± 12.9	47.8 ± 1.8
Honolua 4	24.0 ± 0.3 ^b	2.71 ± 0.4	13.9 ± 1.0	5.25 ± 0.88	3253 ± 247	84.6 ± 10.3	9.93 ± 1.35	14.6 ± 16.7	89.4 ± 10.0	6.52 ± 0.67
Hokuanui bay	34.8 ± 3.2 ^b	3.61 ± 0.61	16.7 ± 0.4	6.00 ± 0.24	5131 ± 962	135.4 ± 16.8	17.7 ± 3.7 ^a	17.6 ± 0.6	108.3 ± 7.6	10.7 ± 2.2
Honokeana cove	27.01 ± 0.4 ^b	5.41 ± 0.5	21.3 ± 1.0	7.54 ± 0.24	6828 ± 5	168.4 ± 8.9	22.6 ± 2.4 ^b	20.7 ± 0.7	103.2 ± 4.4	16.7 ± 2.5
Lipoa point	25.7 ± 0.5 ^b	2.65 ± 0.2	14.7 ± 0.9	6.09 ± 0.45	3259 ± 251	91.1 ± 11.6	10.31 ± 1.71	15.9 ± 0.8	94.4 ± 3.8	6.7 ± 0.61
Honokohau bay	56.2 ± 1.4 ^b	45.5 ± 3.2	73.1 ± 5.1 ^a	17.1 ± 1.0	7016 ± 169	491.7 ± 21.0	373.9 ± 43.9 ^c	35.3 ± 1.7 ^a	343.0 ± 27.0	51.2 ± 2.1
Mokulei'a bay	48.1 ± 1.4 ^b	19.9 ± 2.1	47.3 ± 5.7	9.39 ± 0.66	6836 ± 8	296.5 ± 15.3	144.6 ± 28.4 ^c	26.1 ± 0.9	190.6 ± 13.1	25.1 ± 2.4
TEL	7.24		52.3	18.7			15.9	30.24		124.0
ERL	8.2		81	34			20.9	46.7		150.0
ERM	70		370	270			51.6	218		410.0

Table 1. Metal and metalloid concentrations in superficial reef sediments collected from 9 sites in West Maui, and values of the Sediment Quality Guidelines. TEL: Threshold Effect Level; ERL: Effect-Range Low; ERM: Effect-Range Median based on NOAA sediment quality guideline (Long et al. 1995) and NOAA Screening Quick Reference Tables (NOAA 1999).

^a indicated that the element concentration is superior to the TEL, ^b to the ERL, and ^c to the ERM.

<i>Species</i>	<i>Sites</i>	As m ± S.D.	Co m ± S.D.	Cr m ± S.D.	Cu m ± S.D.	Fe m ± S.D.	Mn m ± S.D.	Ni m ± S.D.	Pb m ± S.D.	V m ± S.D.	Zn m ± S.D.	
<i>T. gratilla</i>		Gonads										
	Honolua 1	17.6 ± 4.7	1.41 ± 0.29	5.05 ± 1.93	3.34 ± 0.63	239 ± 121	14.2 ± 5.4	2.47 ± 1.35	6.06 ± 0.91	37.8 ± 8.3	52.8 ± 24.3	
	Honolua 4	14.8 ± 4.7	1.05 ± 0.65	7.99 ± 1.88	4.53 ± 0.86	202 ± 131	3.64 ± 2.16	2.23 ± 0.57	9.16 ± 2.10	47.0 ± 15.1	25.4 ± 9.1	
	Kapalua	15.1 ± 2.4	0.53 ± 0.26	2.38 ± 0.95	2.14 ± 0.83	65.4 ± 24.1	2.83 ± 2.57	0.87 ± 0.25	2.93 ± 1.95	48.0 ± 24.2	28.0 ± 23.6	
	Honokeana	9.74 ± 3.43	0.70 ± 0.84	1.75 ± 0.71	2.26 ± 0.65	42.0 ± 18.0	3.89 ± 2.03	0.58 ± 0.19	3.21 ± 1.35	24.2 ± 5.2	30.7 ± 12.2	
	Lipoa Point	14.6 ± 2.8	0.94 ± 0.06	2.95 ± 0.60	3.19 ± 1.11	52.9 ± 34.6	4.26 ± 2.58	0.98 ± 0.09	5.04 ± 0.33	36.1 ± 15.0	28.3 ± 12.1	
			Intestine									
	Honolua 1	80.3 ± 17.8	2.33 ± 0.54	3.36 ± 0.86	7.66 ± 1.05	946 ± 163	28.9 ± 9.3	2.82 ± 0.92	6.83 ± 0.77	26.0 ± 6.1	36.3 ± 5.1	
	Honolua 4	37.1 ± 5.3	1.35 ± 0.57	4.04 ± 1.77	7.30 ± 1.23	625 ± 82.4	7.54 ± 1.58	2.80 ± 0.88	7.20 ± 1.45	27.7 ± 3.9	43.2 ± 11.4	
	Kapalua	38.7 ± 11.5	1.06 ± 0.08	2.50 ± 0.70	4.94 ± 1.75	738 ± 210	8.80 ± 1.00	1.60 ± 1.67	5.65 ± 0.46	35.1 ± 8.0	38.0 ± 8.7	
	Honokeana	40.0 ± 6.9	0.55 ± 0.45	1.14 ± 1.17	5.96 ± 0.85	1096 ± 413	16.0 ± 5.7	1.22 ± 0.44	5.02 ± 0.59	31.7 ± 5.8	31.8 ± 3.9	
	Lipoa Point	59.6 ± 19.8	1.33 ± 0.02	2.97 ± 0.34	8.86 ± 1.90	672 ± 219	12.2 ± 3.2	2.42 ± 0.93	6.91 ± 0.56	32.2 ± 2.3	43.7 ± 10.7	
			Body Wall									
	Honolua 1	14.3 ± 2.8	0.85 ± 0.29	4.97 ± 2.14	3.48 ± 2.02	62.7 ± 24.7	9.03 ± 3.56	1.28 ± 0.63	6.61 ± 4.14	106.7 ± 7.7	3.28 ± 1.58	
	Honolua 4	15.0 ± 3.1	0.89 ± 0.29	5.14 ± 1.46	3.37 ± 1.39	22.6 ± 11.0	1.24 ± 0.54	1.18 ± 0.53	7.86 ± 3.71	103.4 ± 9.0	2.93 ± 0.70	
Kapalua	17.5 ± 2.3	1.23 ± 0.10	6.04 ± 1.11	4.93 ± 1.20	29.9 ± 6.6	2.06 ± 0.68	1.66 ± 0.68	13.1 ± 0.6	111.9 ± 7.0	2.56 ± 0.27		
Honokeana	14.0 ± 3.2	1.09 ± 0.16	4.13 ± 1.82	2.98 ± 1.84	13.4 ± 4.3	2.57 ± .76	1.05 ± 0.31	6.05 ± 5.21	98.5 ± 7.9	2.72 ± 0.43		
Lipoa Point	14.2 ± 3.1	0.69 ± 0.76	4.05 ± 1.51	2.69 ± 1.87	10.0 ± 3.8	2.22 ± 0.73	0.80 ± 0.83	5.93 ± 4.75	104.2 ± 8.3	2.47 ± 0.65		
<i>P. multifasciatus</i>		Muscle										
	Honolua	39.9 ± 10.7	0.17 ± 0.11	1.40 ± 0.47	0.87 ± 0.24	14.8 ± 6.5	0.56 ± 0.34	0.69 ± 0.46	1.45 ± 0.44	3.85 ± 0.86	10.3 ± 1.9	
		Liver										
	Honolua	54.8 ± 27.0	1.98 ± 0.79	1.44 ± 1.05	15.6 ± 3.2	4400 ± 931	7.42 ± 1.76	0.53 ± 0.38	2.89 ± .89	5.11 ± 2.38	112 ± 10.3	
<i>H. kanaloana</i>	Honolua 5	15.8 ± 5.6	0.92 ± 0.43	6.24 ± 2.77	4.02 ± 1.52	531 ± 155	12.8 ± 2.3	3.81 ± 1.88	8.74 ± 3.10	23.4 ± 3.4	2.63 ± 0.61	
	Honokeana	16.0 ± 7.5	1.33 ± 0.68	7.32 ± 2.82	4.96 ± 2.24	374 ± 74.5	9.86 ± 0.89	1.97 ± 1.27	8.52 ± 5.22	23.2 ± 15.0	3.09 ± 1.95	

Table 2. Element concentrations (mean ± S.D.; $\mu\text{g g}^{-1}$ dwt) in the urchin *Tripneustes gratilla* (n = 7 per station, except for Lipoa Point n = 4), the goatfish *Parupeneus multifasciatus* (n = 6) and the alga *Halimeda kanaloana* (n = 7) collected in the West Maui

A) GENERAL			
Factor 1		Factor 2	Pearson values
Coral cover		Co	-0.972 *
Coral cover		Cr	-0.955 *
Coral cover		Mn	-0.955 *
Coral cover		Ni	-0.960 *
Coral cover		V	-0.979 *
Coral cover		Zn	-0.958 *

B) BY CORAL SPECIES			
Species	Factor 1	Factor 2	Pearson values
PLOB	Diameter	As	-0.964 *
PLOB	Diameter	Fe	-0.976 *
MPAT	Diameter	Cu	-0.976 *
PCOM	Diameter	Partial mortality	+0.992 **
PCOM	Diameter	Turbidity	-0.975 *

Table 3. Results of the Pearson Correlation performed on the different reef health parameters and the element concentration in sediment for A) all species combined and B) for each coral species. Only significant correlations (<0.05) are presented (*: < 0.05; **: < 0.01)

Species codes: *Porites lobata* (PLOB), *Porites compressa* (PCOM), *Montipora patula* (MPAT)

	Species	Location	As	Co	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn
GOATFISH	<i>Mulloidichthys vanicolensis</i> ^{1,A}	French Frigate Shoals	121		7.5	30				20		273
	<i>Pseudupeneus prayensis</i> ^{2,B}	Mauritania coast				0.9 ± 0.2 - 1.6 ± 0.6						11 ± 1 - 16 ± 4
	<i>P. prayensis</i> ^{2,C}	Mauritania coast				1.6 ± 0.3 - 3.1 ± 1.3						79 ± 4 - 97 ± 18
	<i>P. prayensis</i> ^{2,D}	Mauritania coast				13.8 ± 3.1 - 18 ± 1.0						103 ± 13 - 134 ± 5
	<i>Upeneus moluccensis</i> ^{3,A}	Manila bay		0.108		2.12		8.32		0.0951		66.1
ALGA	<i>Halimeda opuntia</i> ⁴	Red Sea (Saudi Arabia)		0.14	0.49				0.64	2.52	0.54	
	<i>H. tuna</i> ⁵	Suez	14.5 ± 1.4	13.4 ± 3.3	7.4 ± 1.7	27.7 ± 1.7	16.7 ± 2.9	49.8 ± 4.1				37.3 ± 3.7
	<i>H. tuna</i> ⁵	Mars Alam	4.1 ± 1.3	3.8 ± 1.8	3.8 ± 1.0	7.3 ± 2.1	5.7 ± 1.4	8.9 ± 1.4				6.3 ± 2.1
	<i>H. tuna</i> ⁶	Chalkidiki, Aegean Sea			4.5	71.3	21.9	0.02				14.6
	<i>H. tuna</i> ⁷	Gulf of Aden, Yemen	0.3	3.4	8.31	60.2	3.5	4.7	3.2	0.3		2.53
	<i>H. tuna</i> ⁸	Sea Area off Beirut			32.1	476.0		42.0	51.9			
	<i>H. incrassata</i> ⁹	La Curieuse Island			< 1.2	< 0.6	18.98	6.28	< 1.2	< 2		7.29
	<i>H. Taenicola</i> ¹⁰	Suvorov Atoll				2.86	27.62	6.43				5.95
	<i>H. micronesica</i> ¹⁰	Suvorov Atoll				2.97	25.99	7.43				6.93
	<i>H. discoidea</i> ¹⁰	Bio Island				3.01	45.37	6.94		3.01		10.88
<i>H. simulaus</i> ¹⁰	Bio Island				3.20	41.63	6.90		2.46		9.85	

¹ Miao et al. 2001; ² Roméo et al. 1999; ³ Prudente et al. 1997; ⁴ El-Naggar and Al-Amoudi 1989; ⁵ Abdallah et al. 2005; ⁶ Sawidis et al. 2001; ⁷ Al-Shwafi and Rushdi 2008; ⁸ Shiber 1980; ⁹

Dolgushina et al. 1995; ¹⁰ Khristoforova and Bogdanova 1980

^A Whole, ^B Muscle, ^C Gills, ^D Liver

Table 4. Element concentrations (mean ± S.D. or range; µg g⁻¹ dwt) in goatfishes and the algae *Halimeda* sp. from various geographical areas

<i>Urchin species</i>	Location	Organs	As	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn
<i>Diadema setosum</i> ¹	Singapore reef	Gonads		1.39 ± 0.99 - 4.83 ± 3.57	2.91 ± 0.45 - 7.73 ± 7.18	88 ± 77 - 285 ± 229			0.78 ± 0.29 - 1.86 ± 0.65		129 ± 162 - 307 ± 369
<i>D. setosum</i> ¹	Singapore reef	Body wall		0.48 ± 0.15 - 1.11 ± 0.95	0.31 ± 0.22 - 1.65 ± 1.28	25 ± 16 - 81 ± 46			2.48 ± 0.67 - 4.24 ± 1.3		12.33 ± 1.17 - 15.14 ± 2.18
<i>D. setosum</i> ¹	Singapore reef	Skeleton		1.18 ± 0.76 - 2.22 ± 0.71	0.35 ± 0.12 - 1.01 ± 0.36	8 ± 3 - 34 ± 26			4.69 ± 0.85 - 7.4 ± 2.05		4.82 ± 0.6 - 6.36 ± 1.63
<i>Echinocardium cordatum</i> ²	Isle of Arran, Scotland	Gonads			7	184			4.4		239
<i>E. cordatum</i> ²	Isle of Arran, Scotland	Gut			11	3480			5.8		74
<i>Echinus esculentus</i> ³	Irish Sea, UK	Oral body wall			1.8	6.9			<0.62		110
<i>E. esculentus</i> ³	Irish Sea, UK	Aboral body wall			0.9	2.5			<0.62		12
<i>E. esculentus</i> ³	Irish Sea, UK	Gonads			16	15			<0.68		110
<i>Echinometra lucunter</i> ⁴	Cuba	Gonads			2.57 - 6.09	45.8 - 180					230 - 436
<i>Paracentrotus lividus</i>	Marseille (France)	Body wall			1 - 5.33				0.3 - 1.19		
<i>P. lividus</i> ⁵	Marseille (France)	Gonads			4.02 - 6.34				0.3 - 3.14		
<i>P. lividus</i> ⁵	NW Mediterranean	Gonads		0.88 - 1.59	2.68 - 3.18	35 - 101			1.38 - 2.02		124 - 161
<i>P. lividus</i> ⁵	Marseille (France)	Gonads			6.34				3.14		
<i>P. lividus</i> ⁵	Marseille (France)	Gut			22.9				28.1		
<i>P. lividus</i> ⁶	NW Mediterranean	Digestive wall		0.86 ± 0.17 - 1.23 ± 0.75	22 ± 6 - 27 ± 7	120 ± 58 - 287 ± 159			1.93 ± 0.56 - 2.12 ± 1.32		70 ± 14 - 80 ± 12
<i>P. lividus</i> ⁶	NW Mediterranean	Gonads		0.88 ± 0.34 - 1.59 ± 0.92	2.68 ± 0.94 - 3.18 ± 0.90	35 ± 17 - 101 ± 62			1.38 ± 0.70 - 2.02 ± 1.06		124 ± 111 - 161 ± 192
<i>P. lividus</i> ⁶	NW Mediterranean	Body wall		0.73 ± 0.64 - 0.89 ± 0.77	0.49 ± 0.18 - 0.65 ± 0.28	4.37 ± 2.01 - 13.7 ± 8.7			3.68 ± 2.94 - 5.39 ± 3.50		3.84 ± 0.78 - 5.21 ± 1.34
<i>P. lividus</i> ⁷	NW Mediterranean	Digestive wall		0.78 ± 0.12 - 1.74 ± 1.54	14.1 ± 4.7 - 31.1 ± 7.3	66 ± 7 - 320 ± 166			1.24 ± 0.51 - 4.90 ± 0.72		54 ± 10 - 88 ± 16
<i>P. lividus</i> ⁷	NW Mediterranean	Gonads		0.67 ± 0.20 - 2.16 ± 1.13	1.03 ± 0.57 - 3.83 ± 0.59	23 ± 9 - 119 ± 66			0.74 ± 0.17 - 3.02 ± 1.31		53 ± 51 - 383 ± 253
<i>P. lividus</i> ⁷	NW Mediterranean	Body wall		0.24 ± 0.05 - 2.05 ± 0.15	0.19 ± 0.13 - 0.86 ± 0.12	3.28 ± 1.30 - 17.9 ± 14.3			1.48 ± 0.55 - 7.76 ± 0.83		3.08 ± 0.85 - 6.28 ± 1.45
<i>P. lividus</i> ⁸	Balearic Islands	Full body tissues	0.7-4.1	0.4-3.2	1.6-7.5			0.8-2.7	0.8-6.7		5.7-74.5
<i>P. lividus</i> ⁹	Southern Adriatic Sea	Gonads			5.19 ± 2.68	184 ± 166			0.86 ± 0.67		157.13 ± 47.91
<i>Strongylocentrotus intermedius and nudus</i> ¹⁰	Amursky bay (Russia)	Gonads			0.8	147					28.5
<i>S. droebachiensis</i> ¹¹	Arctic bay	Whole		3.5 ± 0.6 - 3.9 ± 0.9	2.9 ± 0.5 - 3.2 ± 0.5	495 ± 300 - 1310 ± 570					34 ± 5.8 - 57 ± 17
<i>S. droebachiensis</i> ¹¹	Arctic bay	Gonads		11 ± 4.9	5.2 ± 3.3	217 ± 58					153 ± 50
<i>Sphrerechinus granularis</i> ¹²	Brest Bay	Gonads			3.7 - 5	189 - 320			0.79 - 1.14		476.4 - 706.3
<i>S. granularis</i> ¹²	Brest Bay	Gut			9.95 - 12.9	4057 - 4601			3.2 - 3.6		128.4 - 220.2
<i>Sterechinus neumayeri</i> ¹³	King George Islands	Body wall	0.04 ± 0.01	21.0 ± 11	7.0 ± 4.0	5400 ± 3200	100 ± 50	4.0 ± 2.0	10.0 ± 6.0	19 ± 12	260 ± 60
<i>S. neumayeri</i> ¹³	King George Islands	Digestive tract	0.05 ± 0.05	23.0 ± 12.0	1.0 ± 0.2	300 ± 100	9 ± 2	2.0 ± 1.0	1.0 ± 0.4	9 ± 1	100 ± 20
<i>S. neumayeri</i> ¹³	Deception Island	Body wall	1.0 ± 1.0	100 ± 53	0.2 ± 0.1	2400 ± 1000	100 ± 30	4.0 ± 2.0	13.0 ± 5.0	10 ± 4	620.0 ± 136.0

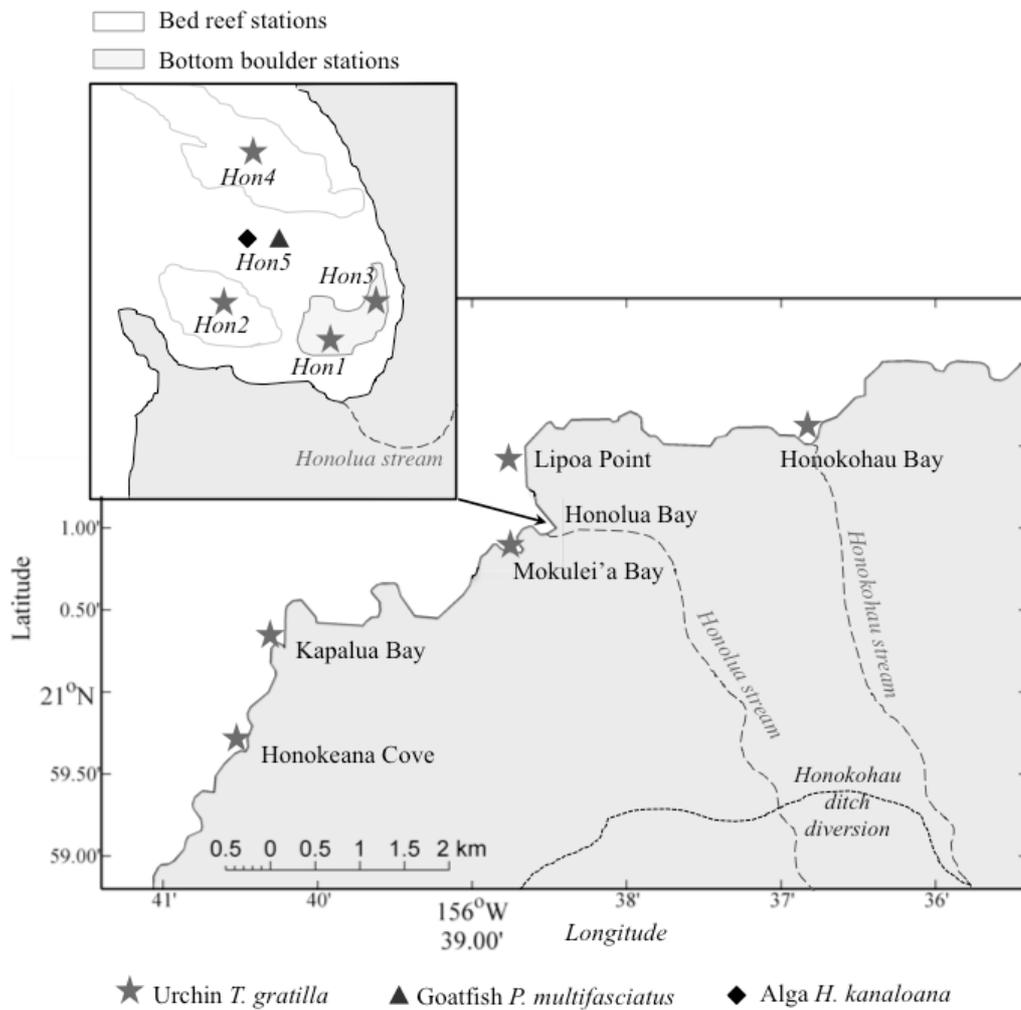
¹ Flammang et al. 1997; ² Buchanan et al. 1980; ³ Riley and Segar 1970; ⁴ Ablanedo et al. 1990; ⁵ Augier et al. 1989; ⁶ Warnau et al. 1995; ⁷ Warnau et al. 1998; ⁸ Deudero et al. 2007; ⁹ Storelli et

al. 2001; ¹⁰ Naidenko 1997; ¹¹ Bohn 1979; ¹² Guillou et al. 2000; ¹³ Deheyn et al. 2005;

Table 5. Element concentrations (mean \pm S.D. or range; $\mu\text{g g}^{-1}$ dwt) in urchins from various geographical areas

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Figure 1

**Geographical coordinates of the stations:**

Hon1 (N21°00.822, W156°38.332)

Hon2 (N21°00.835, W156°38.405)

Hon3 (N21°00.844, W156°38.277)

Hon4 (N21°00.913, W156°38.313)

Hon5 (N21°00.849, W156°38.297)

Kapalua Bay (N21°00.046, W156°40.057)

Lipoa Point (N21°01.046, W156°38.419)

Mokulei'a Bay (N21°00.747, W156°38.517)

Honokohau Bay (N21°01.324, W156°36.563)

Figure 2

A- Superficial sediments

A-1

Metal	Location ranking			
As	Hon3	<u>Hon1</u>	<u>Hon2</u>	Hon4
Cd	Hon1	Hon3	Hon2	Hon4
Co	Hon1	Hon3	Hon2	Hon4
Cr	<u>Hon1</u>	<u>Hon3</u>	Hon2	Hon4
Cu	Hon3	Hon1	<u>Hon2</u>	<u>Hon4</u>
Fe	<u>Hon2</u>	<u>Hon3</u>	<u>Hon2</u>	Hon4
Mn	Hon1	Hon3	Hon2	Hon4
Ni	Hon1	Hon3	Hon2	Hon4
Pb	<u>Hon1</u>	<u>Hon3</u>	<u>Hon2</u>	<u>Hon4</u>
V	Hon1	Hon3	Hon2	Hon4
Zn	Hon1	Hon3	Hon2	Hon4

A-2

Metal	Location ranking						
As	HB	MB	<u>Hon1</u>	<u>KB</u>	<u>HC</u>	LP	Hon4
Cd	<u>Hon1</u>	<u>HB</u>	MB	<u>HC</u>	<u>KB</u>	<u>LP</u>	Hon4
Co	<u>Hon1</u>	<u>HB</u>	MB	<u>HC</u>	KB	Hon4	LP
Cr	Hon1	HB	MB	<u>HC</u>	<u>KB</u>	<u>LP</u>	Hon4
Cu	HB	Hon1	MB	<u>HC</u>	<u>LP</u>	<u>KB</u>	Hon4
Fe	<u>HB</u>	Hon1	MB	<u>HC</u>	KB	<u>LP</u>	Hon4
Mn	Hon1	HB	MB	<u>HC</u>	<u>KB</u>	<u>LP</u>	Hon4
Ni	Hon1	HB	MB	<u>HC</u>	KB	<u>LP</u>	Hon4
Pb	<u>Hon1</u>	<u>HB</u>	MB	<u>HC</u>	<u>KB</u>	<u>LP</u>	Hon4
V	<u>Hon1</u>	<u>HB</u>	MB	KB	<u>HC</u>	<u>LP</u>	Hon4
Zn	<u>Hon1</u>	<u>HB</u>	MB	HC	<u>KB</u>	<u>LP</u>	Hon4

B- Urchin *T. gratilla*

B-1

Metal	Location ranking					
As	Hon1	<u>KB</u>	Hon4	LP	<u>HC</u>	
Cd	<u>LP</u>	<u>HC</u>	Hon4	Hon1	<u>KB</u>	
Co	Hon1	<u>Hon4</u>	<u>LP</u>	<u>HC</u>	<u>KB</u>	
Cr	Hon4	<u>Hon1</u>	<u>LP</u>	<u>KB</u>	<u>HC</u>	
Cu	Hon4	<u>Hon1</u>	<u>LP</u>	<u>HC</u>	<u>KB</u>	
Fe	<u>Hon1</u>	<u>Hon4</u>	<u>KB</u>	<u>LP</u>	<u>HC</u>	
Mn	Hon1	<u>LP</u>	<u>HC</u>	<u>KB</u>	<u>Hon4</u>	
Ni	<u>Hon1</u>	<u>Hon4</u>	<u>LP</u>	<u>KB</u>	<u>HC</u>	
Pb	Hon4	<u>Hon1</u>	<u>LP</u>	<u>HC</u>	<u>KB</u>	
V	<u>KB</u>	<u>Hon4</u>	<u>Hon1</u>	<u>LP</u>	<u>HC</u>	
Zn	<u>KB</u>	Hon4	Hon1	LP	HC	

B-2

Metal	Compartment ranking		
As	I	<u>G</u>	<u>C</u>
Cd	I	<u>G</u>	<u>C</u>
Co	I	<u>G</u>	<u>C</u>
Cr	C	<u>G</u>	<u>I</u>
Cu	I	<u>G</u>	<u>C</u>
Fe	I	G	C
Mn	I	G	C
Ni	I	<u>G</u>	<u>C</u>
Pb	C	<u>I</u>	<u>G</u>
V	C	G	I
Zn	<u>I</u>	<u>G</u>	C

Figure 3

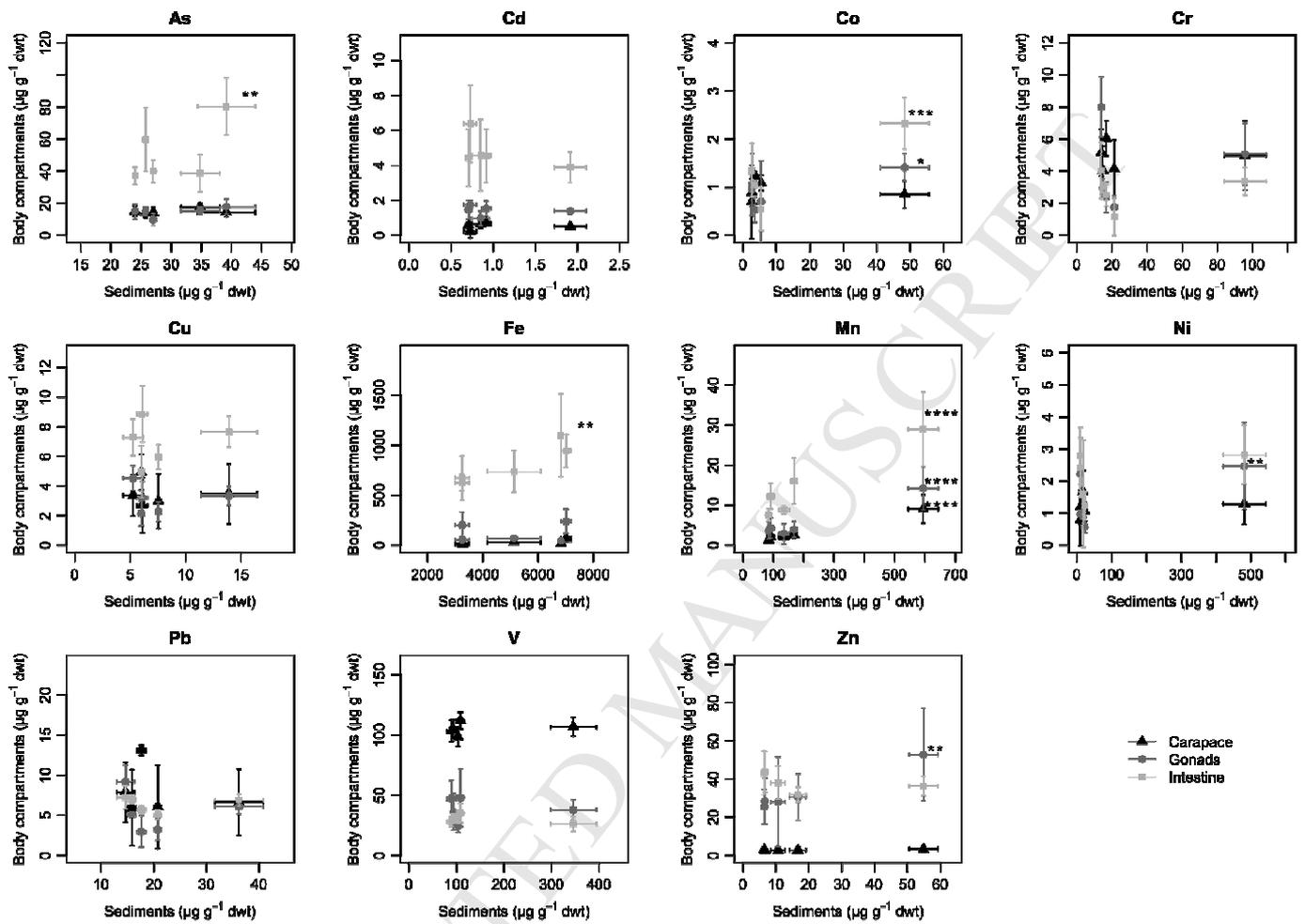


Figure 4

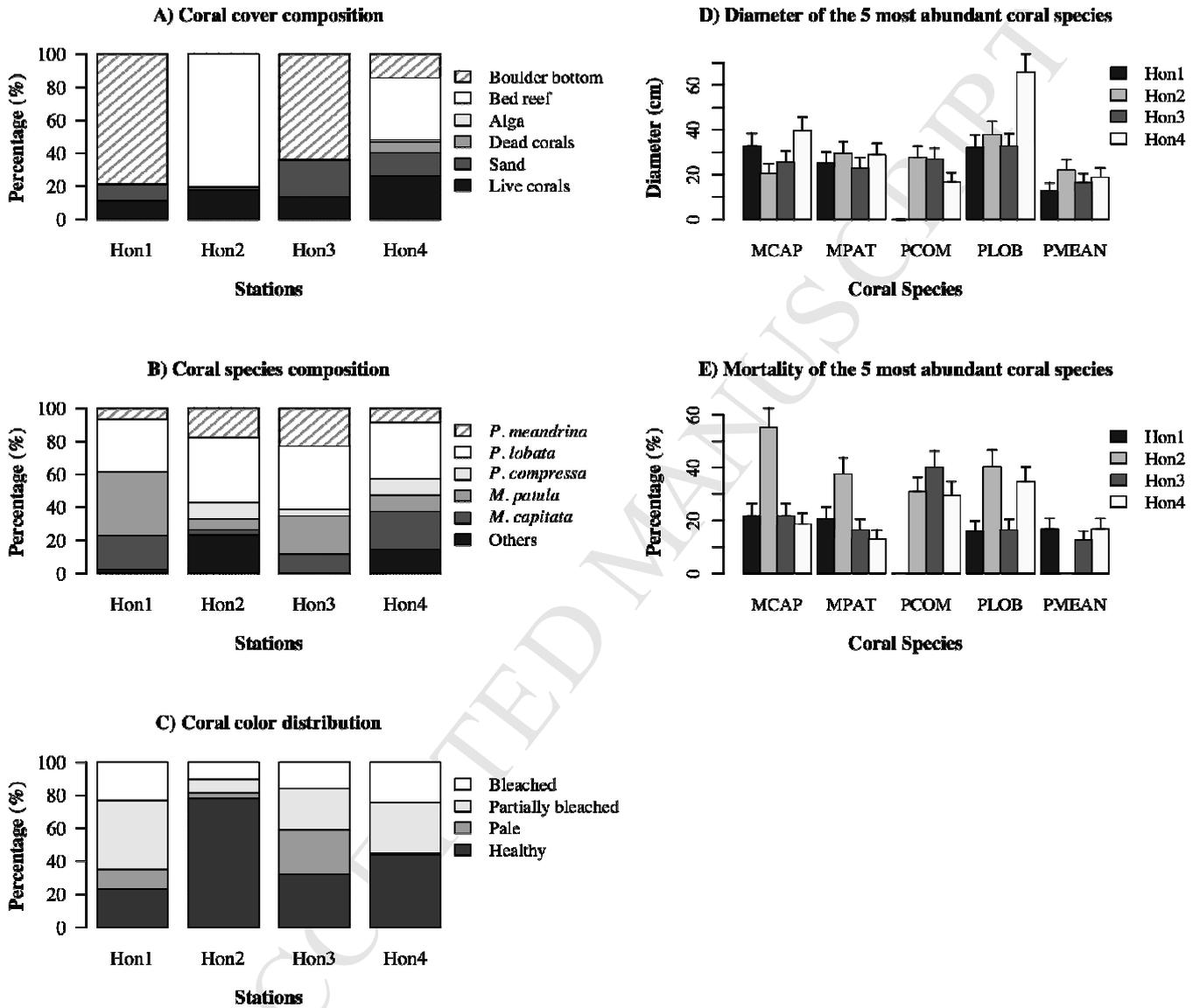


Figure 5

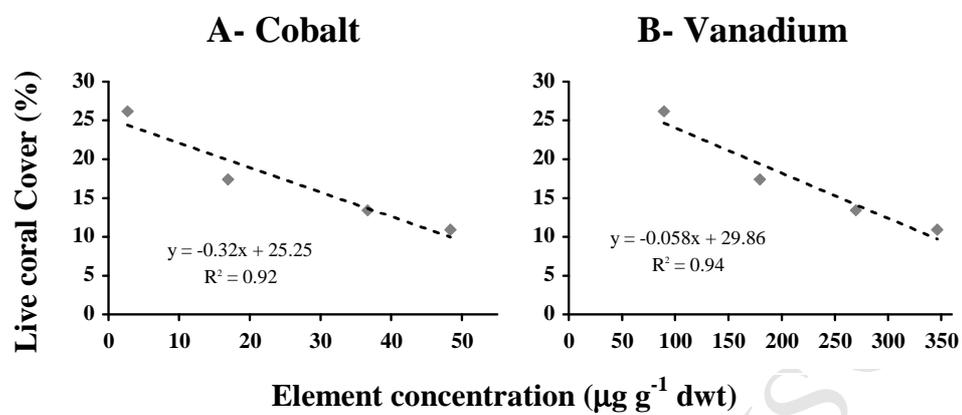


Figure 6

