

Trends in concentrations of selected metalloid and metals in two bivalves from the coral reefs in the SW lagoon of New Caledonia

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Laetitia Hédouin, Paco Bustamante, Carine Churlaud, O. Pringault, Renaud Fichez, et al.. Trends in concentrations of selected metalloid and metals in two bivalves from the coral reefs in the SW lagoon of New Caledonia. Ecotoxicology and Environmental Safety, 2009, 72 (2), pp.372- 381. 10.1016/j.ecoenv.2008.04.004. hal-00336151

HAL Id: hal-00336151 https://hal.science/hal-00336151v1

Submitted on 2 Nov 2008

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23 Abstract

24 The concentrations of 9 elements (Ag, As, Cd, Co, Cr, Cu, Mn, Ni and Zn) were measured 25 in the oyster Isognomon isognomon and the edible clam Gafrarium tumidum from different 26 sites along the SW New Caledonian coast which is subjected to important chemical inputs due 27 to intense land-based mining activities (New Caledonia is the third world producer of nickel). 28 Results indicate that concentrations in the two organisms mirrored the geographical 29 differences in contamination levels as established through element analyses in sediment. On 30 the basis of organism analyses, two out of the seven investigated stations can be considered as 31 relative "reference" sites, except for As, for which very high levels were detected in clam and oyster tissues (up to 441 µg g⁻¹ dry wt for clams). Overall, our results indicate that both 32 33 tropical organisms investigated could be used as valuable bioindicator species for surveying 34 metal contamination in the coastal waters of New Caledonia with reasonable perspectives of 35 wider application to other coral reef environments.

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37 Keywords: Tropical Environment, Metals, Bioindicators, Mining activities

39 1. Introduction

40 Surrounded by a barrier reef of 1600 km, the New Caledonia lagoon is one of the largest 41 in the world (Labrosse et al., 2000). However, the lagoon of New Caledonia is subjected to 42 important anthropogenic inputs of metals mainly due to intense land-based Ni mining 43 activities but also to urban development and lack of efficient wastewater treatment. Open-cast 44 mining exploitation presently constitutes the major economical resource of the Territory and 45 results in important coastal discharges of metals, which constitute a threat to coral reef 46 ecosystems (Labrosse et al., 2000). Recently, more efficient extraction processes based on acidic extraction (viz lixiviation) have been developed (Mihaylov et al., 2000; Goro-Nickel, 47 48 2001), making the extraction from low Ni grade ores (limonite) possible. The acidic 49 extraction of metals is not Ni-selective and makes also soluble all other ore-contained by-50 product metals. Therefore, the lixiviation process will obviously lead to an increased multi-51 elemental contamination of the coastal marine environment.

Although mining activities are rising up in the island, studies reporting concentrations or behaviour of metals in marine organisms from New Caledonia are scarce (Monniot et al., 1994; Bustamante et al., 2000; Labrosse et al., 2000; Hédouin et al., 2007). In this context, acquisition of reliable and relevant data in the New Caledonian lagoon is a strong priority and the development and implementation of risk assessment studies and metal monitoring programme is expected by the local authorities.

Among the approaches used to assess environmental contamination, the usefulness of bioindicator species is now well established. Marine organisms provide valuable information on the geographical and temporal variations of the bioavailable metal concentrations in their environment (eg, Rainbow, 1995; Warnau et al., 1998). Ideally, selected bioindicators should display a simple relationship between metals accumulated in their tissues and the ambient

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63 metal concentrations. This should be true regardless of location and environmental conditions64 considered.

65 Molluses have been extensively used in temperate regions (eg, Goldberg et al., 1983; 66 Rainbow, 1995), whereas little attention has been paid to the identification of bioindicators 67 specifically adapted to tropical and sub-tropical regions (Phillips, 1991) despite the constant 68 increase in industrial and human activities. Some efforts were devoted to the extension of the 69 Mussel Watch to the Asia/Pacific and Latin America regions (see eg, UNU, 1994; IMWC, 70 1995), using bivalves such as Saccostrea spp., Crassostrea spp. and Perna spp. as 71 bioindicators. However, none of the above-cited species is present in sufficient abundance 72 along the New Caledonia coasts to be considered as a useful candidate to monitor local 73 contamination. Hence, other tropical organisms have to be selected. In this context, recent 74 studies screened metal concentrations in a variety of local marine organisms from different 75 areas of the New Caledonian lagoon with contrasting contamination status (Breau, 2003; 76 Hédouin, 2006; Hédouin et al., 2006; 2007). The latter studies showed that two bivalves, 77 namely the oyster Isognomon isognomon and the edible clam Gafrarium tumidum, are 78 satisfying the basic ecological and ecotoxicological requirements to be met by a bioindicator 79 species sensu Moore (1966) and Phillips (1990b). Among others, metal bioaccumulation and 80 retention capacity of G. tumidum and I. isognomon exposed via different pathways 81 (seaweater, food, and sediment) were characterized in controlled conditions, including the 82 relationships between metals concentrated in the bivalve tissues and the ambient metal 83 concentrations (Hédouin 2006; Hédouin et al., 2006; 2007; submitted). Results indicate that 84 both species are promising bioindicator candidates for tropical environments.

The aim of the present study was to further assess, in the field, the reliability of these two species as sentinel organisms and to provide information on the degree of contamination of selected elements of local concern (Ag, As, Cd, Co, Cr, Cu, Mn, Ni, Zn) in different locations along the SW coast of New Caledonia. Results presented in this paper also provide baselinedata for future monitoring programmes.

90 **2.** Materials and methods

91 2.1. Sampling sites

92 The sampling stations were selected according to supposedly contrasting contamination status (Fig. 1). Oysters were collected in the subtidal zone of Maa Bay, Koutio Bay, Boulari 93 94 Bay and Grande Rade (GR_s). Maa Bay is subjected to low anthropogenic and terrigenous 95 inputs and was considered as the relative "reference" station for oysters. Koutio Bay is 96 influenced by inputs of domestic wastes from Noumea City and by the occurrence of an 97 important rubbish dump. Boulari Bay is under the influence of La Coulée River that delivers 98 important inputs of lateritic materials to the lagoon due to soil erosion of closed mine sites. 99 Grande Rade (GR_s) is subjected to anthropogenic inputs from the Ducos industrial zone and 100 the metallurgic factory "Société Le Nickel" (SLN).

For clams, three intertidal sampling stations were selected: Ouano Beach, Dumbéa Bay and Grande Rade (GR_I). Ouano Beach is situated 100 km northward from Noumea, and is not influenced by industrial activities; it was considered as the "reference" station for clams. Grande Rade (GR_I) is subject to anthropogenic inputs from the SLN factory (scoria and waters), from the Shell Pacific factory (effluents) and from domestic discharges. Dumbéa Bay is an estuarine bay, influenced by waters from La Dumbéa River and subjected to terrigenous inputs.

108 2.2. Organisms

109 The clam *Gafrarium tumidum* was collected by handpicking in the intertidal stations. The 110 oyster *Isognomon isognomon* is associated with rocky substrata at depth ranging from 2 to 25 111 m and were collected by SCUBA diving. All the organisms (n = 6 per species per station) 112 were collected from October to November 2004 (mean water temperature: 25.6 ± 0.8 °C) to 113 reduce as much as possible the variability of element concentrations due to season or sexual 114 cycle. Body size is well-known to affect metal concentrations in organisms (eg, Boyden, 115 1977; Warnau et al., 1995); therefore only samples with shell width longer than 35 mm for G. 116 tumidum (Hédouin et al., 2006) and shell length longer than 70 mm for I. isognomon (Metian, 117 2003) were selected for analysis. Collected clams (mean \pm SD, n = 18) measured 38.1 \pm 2.8 118 mm (width) and weighed 22.7 \pm 4.9 g; ovsters (mean \pm SD, n = 24) measured 98 \pm 13 mm 119 (length) and weighed 36.9 ± 13.2 g. Back to the laboratory, the bivalves were kept for 24 hrs 120 in 30 l of seawater from the same sampling station to allow for depurating gut contents and 121 particulate material present in the mantle cavity. Three body compartments for the clams 122 (digestive gland, gills and remaining soft parts) and four body compartments for the oysters 123 (visceral mass, gills, adductor muscle and remaining soft parts) were removed from the shells. 124 The separated body compartments were weighed (wet wt), dried at 60°C until constant 125 weight, and weighed again (dry wt). They were then stored in acid-washed, hermetically 126 sealed PET containers until analysis for their metal contents.

127 2.3. Sediments

In parallel to organisms, superficial sediments (top 3-cm layer) were analysed in all the sampling stations (Fig. 1), except in Dumbéa Bay and in Grande Rade (GR_S) (samples lost during diving). Sediments were stored in acid-washed PET bags until return to the laboratory; they were then dried at 60°C until constant weight (5 days). In order to eliminate heterogeneous materials (stones, fragment of corals), sediments were sieved (1-mm mesh size) prior to analysis for their metal contents.

134 2.4. Sample preparation and analysis

135 Aliquots of the biological samples (0.1 to 0.5 g) and of the dried sediment samples (0.5 g)136 were digested using a 3:1 (v:v) 65 % HNO₃ - 30 % HCl mixture (Merck, suprapur quality). Acidic digestion of the samples was carried out overnight at room temperature, then using a 137 MARS[®] V microwave (30 min with constantly increasing temperature up to 100°C for 138 139 sediment and up to 115°C for biota, then 15 min at this maximal temperature). These 140 conditions allowed for a complete digestion of the biological matrices and a strong although 141 not total (highly refractory humic acids may resist) leaching of the sediment (eg, Coteur et al., 142 2003; Dalto et al., 2006). Each sample was eventually diluted to 30 to 50 ml with milli-Q 143 water according to the amount of sample digested.

Elements were analysed using a Varian[®] Vista-Pro ICP-OES (As, Cr, Cu, Mn, Ni, and Zn) 144 145 or a Varian[®] ICP-MS Ultra Mass 700 (Ag, Cd and Co). Three control samples (two Certified 146 Reference Materials -CRMs- and one blank) treated and analysed in the same way as the 147 samples were included in each analytical batch. CRMs were dogfish liver DOLT-3 (NRCC) 148 and lobster hepatopancreas TORT-2 (NRCC). The results were in good agreement with the 149 certified values given for the CRMs and indicated the following recoveries (in %): 103 (Ag), 150 98 (As), 103 (Cd), 112 (Co), 79 (Cr), 95 (Cu), 84 (Mn), 113 (Ni), and 106 (Zn). The detection limits (µg g⁻¹ dry wt) were 10.1 (As), 0.8 (Cr), 0.5 (Cu), 0.04 (Mn), 1.1 (Ni) and 0.7 151 152 (Zn) for ICP-OES and 0.1 (Ag), 0.3 (Cd) and 0.03 (Co) for ICP-MS. Mean element concentrations are given on a dry weight basis ($\mu g g^{-1} dry wt$). 153

154 2.5. Statistical analysis

155 Comparisons of the data were performed using 1- or 2-way analysis of variance 156 (ANOVA) followed by the multiple comparison test of Tukey (Zar, 1996). Two-way 157 ANOVA was used with sampling location and body compartment as fixed factors. The 158 variability explained by each factor and their interaction was derived from the sum of squares 159 (Warnau et al., 1998). The level of significance for statistical analyses was always set at $\alpha =$ 160 0.05.

161 **3. Results**

162 3.1. Sediments

163 Table 1 shows the element concentrations measured in the sediment collected in the 164 different stations. Except for Ag, for which comparison among stations was not possible due 165 to concentrations always under the detection limit, statistical analyses indicated contrasting 166 element concentrations among stations. Boulari Bay and Grande Rade (GR_I) displayed the 167 highest concentrations for all elements. Concentrations in all elements were always significantly higher in Grande Rade (GR_I) than in Ouano Beach ($p_{Tukey} < 0.0001$). 168 169 Concentrations in sediment from Boulari Bay were significantly higher (up to 1 order of 170 magnitude) than those measured in the other stations where oysters were also sampled.

171 3.2. The oyster Isognomon isognomon

172 Among the two factors considered (body compartment and sampling location) and their 173 interaction, the sampling location explained the major part of the variability observed for As, 174 Cd, Cr and Ni (accounting for 16 to 70 % of the global variance) (Tables 2 and 3, Fig. 2A) 175 whereas the body compartment was the predominant factor explaining the variability observed for Cu (29 %), Mn (49 %) and Zn (36 %). Significant interaction between body 176 177 compartment and sampling location factors was detected for all elements except Mn, and 178 accounted for 14 to 44 % of the global variance, indicating that geographical variation of 179 measured concentrations was dependent upon the body compartment considered. For all 180 elements, an important part of the variation was associated to the residual term, ranging from 181 12 to 58 %, indicating that other, non-investigated factors (biological and/or environmental

182 factors) were also influencing metal concentrations in the oyster soft tissues (see also section183 3.3).

184

3.2.1. Geographical variation

185 The sampling location significantly affected the concentrations of all studied elements in 186 the body compartments of *I. isognomon* (2-way ANOVA, p_{sampling locations} < 0.0001), except for 187 Mn for which calculated probability was borderline (p = 0.054) (Table 3, Fig. 2A). Multiple 188 comparison tests on the mean concentrations indicated that one sampling location displayed 189 generally the highest concentrations for one or several elements, whereas the three other 190 locations did not show significant difference in element concentrations, except for Mn (no 191 significant difference among none of the sampling stations) and As (all stations significantly 192 different from each others).

Concentrations of Co and Ni in the oysters were significantly higher in Boulari Bay than in Maa Bay ($p_{Tukey} \le 0.001$; Table 2, Fig. 2A). Oysters from Grande Rade (GR_S) displayed significantly higher Ag and Cu concentrations than those measured in Maa Bay ($p_{Tukey} \le 0.01$) whereas the highest Cr concentrations were measured in oysters from Koutio Bay ($p_{Tukey} \le$ 0.05). In contrast to all other elements, As and Zn concentrations were higher in oysters from Maa Bay.

199 Geographical variation of the element concentrations in the whole-soft parts of the oysters 200 (reconstructed data) were tested using 1-way ANOVA and Tukey test. Results were similar to 201 those from the 2-way ANOVA performed on body-compartment specific concentrations, 202 except for Cd and Zn. For these two latter elements, no significant difference was observed 203 among whole soft parts in the four sites for Cd and between Maa Bay and Grande Rade (GR_s) 204 for Zn (Table 2). The particular opposite pattern of As and Zn displaying highest concentrations in Maa Bay (up to 77 µg As g⁻¹ dry wt and 13,817 µg Zn g⁻¹ dry wt) was 205 206 confirmed in whole soft parts data treatment.

207 **3.2.2. Body distribution**

Multiple comparison tests performed after 2-way ANOVA on the mean concentrations in each body compartment (all sampling locations together) indicated that the concentrations of all elements were lower in the adductor muscle than in the other body compartments (Fig. 2A). Generally, concentrations in gills and visceral mass were not significantly different, but significantly higher than in the other body compartments.

In terms of distribution of total element load among body compartments, visceral mass and remaining soft parts contained the highest proportion of the elements. Body distribution did not differ among sampling locations, except for Ag which occurred in higher proportion (43 %) in the gills of oysters from Grande Rade (GR_S) compared to those from the other stations (5 - 24 %).

218 3.3. The clam Gafrarium tumidum

The two-way ANOVA performed on the whole set of data indicated that, with the exception of Mn and Zn, the sampling location was the predominant factor affecting element concentrations, accounting for 23 to 84 % of the global variance (Tables 3 and 4, Fig. 2B). The ranking of sampling stations by order of decreasing concentration depended on the considered clam body compartment. In the case of Ag, Cd, Co, Cr, Mn, Ni and Zn, 35 to 61 % of the element concentration variability was due to undetermined factor(s) (residual term).

It is noteworthy that the elements for which the residual terms are the highest (Cd, Co, Cr, Mn) both in clams and oysters are those that are co-occurring in Ni-ores. Therefore, it is most plausible that the undetermined factor(s) are related to mining activities, either directly (eg, nature of exploited soils in different areas) or indirectly (eg, climatic factors such as rains temporarily enhancing soil-erosion and riverine inputs).

3.3.1. Geographical variation

The mean concentrations of all elements measured in clams varied significantly according to the sampling locations (2-way ANOVA, $p_{sampling locations}$ always ≤ 0.002) (Tables 3 and 4, Fig. 2B). Results showed significant differences between Ouano Beach and Grande Rade (GR_I) for Ag, Cd, Cr, Cu, Mn, Ni and Zn, with the highest concentrations always found in Grande Rade (GR_I). In contrast the concentrations of As were significantly higher in Ouano Beach compared to all the other locations ($p_{Tukey} \leq 0.001$; Table 4 and Fig. 2B).

Geographical variations were tested using 1-way ANOVA and Tukey test for the reconstructed element concentrations in the whole-soft parts of the clams (Table 4). Results were similar to those previously obtained with 2-way ANOVA performed on bodycompartment specific concentrations, except for Co which showed significant differences among whole soft parts in the three sampling locations ($p_{Tukey} < 0.05$). Similarly to oysters, As levels in clams were highest in the "reference" station (Ouano Beach), reaching mean values up to 441 µg g⁻¹ dry wt.

244

3.3.2. Body distribution

245 The mean concentrations of all elements investigated differed according to the body 246 compartments (2-way ANOVA, $p_{body compartment}$ always ≤ 0.003). Multiple comparison tests of 247 Tukey indicated that the concentrations of Cd, Cu, Cr, Mn and Zn were significantly higher in 248 the digestive gland than in the other tissues (p < 0.05; Fig. 2B). Ag, As, Co and Ni 249 concentrations were similar in the digestive gland and the gills. No major difference was 250 found when considering body distribution in clams collected from Ouano Beach and Dumbéa 251 Bay. In these two stations, the remaining soft parts contained the main fraction (55 to 77 %) 252 of the total body burden for all elements. In contrast, in Grande Rade (GR₁), the elements 253 were similarly distributed between the remaining soft parts and the digestive gland.

254 **4.** Discussion

255 Sediments are a sink for marine contaminants (eg, Salomons et al., 1987) and their 256 element concentrations are often used to assess and monitor the contamination status of the 257 marine environment. According to this concept, Boulari Bay and Grande Rade (GR_1) may be 258 considered as highly contaminated stations compared to Ouano Beach and Maa Bay. In turn, 259 the two latter ones may be defined as relatively non-contaminated stations (see Table 1). 260 However, it is now well known that sediment-associated concentrations are not necessarily 261 representative of the contaminant fraction that is bioavailable, viz the fraction of "direct 262 ecotoxicological relevance" for marine organisms (Phillips and Rainbow, 1993). Therefore, 263 the present study was carried out to assess the usefulness of the ovster *Isognomon isognomon* 264 and the clam Gafrarium tumidum as sentinel species over sediment for Ag, As, Cd, Co, Cr, 265 Cu, Mn, Ni and Zn contamination in the SW lagoon of New Caledonia.

In agreement with sediment analyses, Maa Bay can also be considered as a relative reference site when considering element measurements in the oyster *I. isognomon* for all elements, except As and Zn. The low element concentrations reported in the oysters from this bay are in the same range as those reported in the literature for *Isognomon* spp. as well as in other oyster genera from clean areas (see Table 5).

271 The elevated concentrations of Co and Ni measured in oysters from Boulari Bay strongly 272 suggest that a high degree of mining-related contamination occurs in this area, most probably due to releases from surrounding mines and mining-enhanced erosion of the soils. This was 273 274 further confirmed by the high concentrations of Co, Cr, Mn and Ni measured in the sediment 275 from Boulari Bay. However, element analysis in oyster tissues showed that other stations, not 276 identified through sediment analysis, are also highly contaminated for some elements, 277 especially Maa Bay for As and Zn and Grande Rade (GR_s) for Ag. The elevated 278 concentrations recorded in oysters suggest that Maa Bay would be subjected to agrochemical inputs (eg, Francesconi et al., 1999; Warnau et al., 2007) and Grande Rade (GR_S) to important
domestic wastewater discharges (eg, Martin et al., 1988; Sañudo-Willhelmy and Flegal,
1992).

With the exception of As, element concentrations in the clams *G. tumidum* collected from Ouano Beach were always lower than in those from Grande Rade (GR₁). This is in agreement with the results obtained from sediment analysis. Concentrations measured in the clams from Ouano Beach were in the same range as those reported for clean areas from other tropical zones (see Table 5). Ouano Beach may thus be considered as a relatively clean station for all elements considered, except for As. In contrast, Grande Rade (GR₁) can be defined as a highly contaminated station for Ag, Cr, Cu, Mn, Ni and Zn.

289 In this work, the distribution of the considered elements in bivalve tissues was also 290 investigated in order to possibly identify some organs that could be more sensitive than the 291 use of the whole soft parts and able to respond more rapidly to changes in element contamination in the environment (eg, Warnau et al., 1996b; 1998; 1999). Among the body 292 293 compartments of the clam G. tumidum, the digestive gland displayed the highest 294 bioconcentration capacity. In addition, the concentrations measured in this organ easily 295 allowed discriminating the stations according to their contamination levels. Hence, this organ 296 could be proposed as a target for future biomonitoring programmes. In the oyster I. 297 isognomon, no clear trends could be observed in bioaccumulation and geographical 298 discrimination ability among the different body compartments. Consequently, in a future 299 biomonitoring programme, consideration of the whole soft parts of oysters could be 300 recommended.

The two investigated species accumulated some elements up to very high concentrations compared to the concentrations generally reported in the literature (Table 5). These particularities are discussed below. Ni concentrations measured in clams bear out the capacities of this species to accumulate this metal. Indeed, Ni concentrations in clams from Grande Rade (GR_I) were higher (52 ± 12 $\mu g g^{-1} dry wt$) by one order of magnitude than those usually reported in the literature for other tropical clams (see Table 5). The high levels that we measured for Cr and Ni in sediment and clams from Grande Rade (GR_I) are obviously due to mining activities (presence of SLN industry, which discharges wastes into the Rade) associated to mining-enhanced erosion of lateritic soils, which are enriched in Cr and Ni (Labrosse et al., 2000).

311 Although scarcely available, As concentrations reported in the literature for tropical and subtropical bivalves are generally lower than 30 μ g g⁻¹ dry wt (< 10 μ g g⁻¹ dry wt if one 312 313 considers clams and oysters; see Table 5). However, two studies on sub-tropical areas indicated elevated As concentrations in *Isognomon* spp from Florida $(37.3 \pm 6.9 \ \mu g \ g^{-1} \ dry$ 314 wt) (Valette-Silver et al., 1999) and in the clam Circentia callipyga from the Gulf of Oman 315 (156 μ g g⁻¹ dry wt) (de Mora et al., 2004). In the present study, As was found to reach 316 extremely high concentrations in the clams from Ouano Beach (441 \pm 84 µg g⁻¹ dry wt) 317 compared to those observed in Grande Rade (GR_I) (55 \pm 15 μ g g⁻¹ dry wt) and in the oysters 318 from Maa Bay (77 \pm 9 µg g⁻¹ dry wt). To the best of our knowledge, such high body 319 320 concentrations of As have never been reported in other clams. The Ouano Beach values were 321 in fact on the same order of magnitude than the highest As concentrations ever reported, such as in the cirratulid polychaete Tharyx marioni which displays extremely high body 322 concentrations of total As (2000 µg g⁻¹ dry wt; Gibbs et al., 1983), the Mediterranean fan 323 worm *Sabella spallanzanii* which shows As concentrations higher than 1000 μ g g⁻¹ dry wt in 324 325 its branchial crown (Fattorini and Regoli, 2004) or the very high arsenic concentrations monitored in muscles of edible fish (500 µg g⁻¹dry wt) from the Bay of Cienfuegos, Cuba, a 326 327 few weeks after an accidental release of arsenate oxides from a local nitrogen fertilizer factory 328 in Dec 2001 (Fattorini et al., 2004; Warnau et al., 2007). However the reason for so high As

329 concentrations in G. tumidum tissues is not clear. Some authors have reported that As 330 concentrations in organisms were related to the sediment concentrations (such as in 331 Scrobicularia plana; Langston, 1980). However no similar correlation was observed here. In 332 addition, laboratory experiments have shown that bivalves generally displayed a limited 333 capacity in accumulating As from seawater (eg, Ünlü and Fowler, 1979; Hédouin, 2006; 334 Gómez-Batista et al., 2007). Thus, the elevated As concentrations reported in this study would 335 be accumulated most probably from the diet of the organisms (Sanders et al., 1989; Gómez-Batista and Warnau, unpubl. results). Accordingly, transfer along the food chain could be 336 337 proposed as the main route of uptake for As in bivalves, suggesting that food of both oysters 338 and clams are enriched in As in Maa Bay and Ouano Beach, compared to the other sampling 339 locations. Whereas some agricultural activities are carried out in Maa Bay, Ouano Beach is 340 rather subjected to waste discharges from shrimp aquaculture. Hence the important discharges 341 of N-enriched products (due to terrestrial leaching of fertilisers used for local agriculture or to 342 release of aquaculture food excesses) could locally modify the N:P ratio. In environments 343 with phosphate deficit relative to nitrogen, phytoplankton metabolises As much more easily 344 (Benson and Summons, 1981; Phillips, 1990a). This in turn may lead to enhanced trophic 345 transfer of As to filter-feeders and enhanced As accumulation in the tissues of the bivalves 346 (Warnau et al., 2007). Although further investigations are needed to validate such a 347 hypothesis in Maa Bay and Ouano Beach, the extremely high As levels measured in clam 348 tissues are of considerable interest because (1) G. tumidum is a seafood product in New 349 Caledonia and (2) little is known about the speciation of As in the tissues of this species 350 (Francesconi et al., 1999) which determines its potential toxicity to consumers (see eg, Kaise 351 and Fukui, 1992; Warnau et al., 2007).

I. isognomon displayed also very high Zn concentrations in Maa Bay and in Grande Rade (GRs), viz 13,817 \pm 6,621 and 7,873 \pm 2,087 µg g⁻¹ dry wt, respectively. Elevated

concentrations of Zn have been reported for *I. alatus*, reaching 4,010 µg Zn g⁻¹ dry wt in 354 individuals collected in the Dominican Republic and 12,163 μ g g⁻¹ dry wt in the Guadeloupe 355 (see Table 5). Although, Zn is well known to be essential to organisms, acting for example as 356 357 a co-factor in numerous metalloenzymes (eg, Vallee and Falchuk, 1993), the amounts 358 accumulated are clearly far above the physiological needs of the bivalve. I. isognomon must 359 therefore possess a natural capacity to accumulate Zn up to very high levels while avoiding 360 subsequent toxicity. Such a mechanism could be for example the immobilization of Zn under non-toxic forms in granules which are very slowly excreted (eg, Corrêa Junior et al., 2000). 361 Indeed, in many bivalves and especially in oysters, granules may contain up to 60 % of the 362 363 total body load of Zn (Eisler, 1981).

364 Ag is well known as a highly toxic metal (eg, Warnau et al, 1996a; Ratte, 1999) and the 365 scarcity of data concerning Ag levels in tropical and subtropical organisms in general, and in 366 particular in clams and oysters, is therefore quite surprising (see Table 5). In this way, the 367 concentrations measured in the two investigated species (see Tables 2 and 4) can be 368 considered as baseline data for the New Caledonia lagoon as well as for other tropical 369 environments. Clams and oysters collected from Grande Rade displayed quite elevated Ag concentrations $(33 \pm 13 \text{ and } 33 \pm 7 \mu \text{g g}^{-1} \text{ dry wt, respectively})$, which are one to two orders 370 371 of magnitude higher than those measured in bivalves from the "reference" stations (Ouano Beach or Maa Bay) and to the background concentrations generally considered for tropical 372 areas (< 1 μ g g⁻¹ dry wt in mussels; Klumpp and Burdon-Jones, 1982) and temperate areas (< 373 $6 \ \mu g \ g^{-1}$ dry wt in clams and oysters; Cohen et al., 2001). Various bivalves are able to 374 accumulate Ag up to very high concentrations by trapping it as Ag₂S, a stable and non-toxic 375 376 compound (eg, Berthet et al., 1992; Bustamante and Miramand, 2005). The occurrence of a 377 similar detoxification mechanism in G. tumidum and I. isognomon could explain the high Ag concentrations observed in their soft tissues. Natural sources of Ag are quite rare in the 378

environment (Luoma et al., 1995) and Ag is considered as a reliable proxy of anthropogenic
input s in coastal waters, such as sewage sludge and boating activities, (Martin et al., 1988;
Sanudo-Willhelmy and Flegal, 1992). Therefore the enrichment of Ag in bivalves from
Grande Rade would be most probably related to this kind of domestic inputs.

383 5. Conclusions

384 In New Caledonia, contaminants released in the lagoon are clearly a matter of concern, as 385 reflected by the elevated concentrations in some elements found in the marine organisms 386 investigated in the present work. The two bivalve species considered in this study merit 387 consideration as they appear to be bioindicator species of interest for surveying the 388 contamination status of the New Caledonian waters. Indeed, these species (1) are abundant 389 and widely distributed in New Caledonia (as well as in other tropical areas), (2) show elevated 390 bioaccumulation capacity, and (3) are able to reveal the differences in element concentrations 391 among different areas, even in complex environments (the locations examined here were 392 subjected to various contamination sources).

In a future biomonitoring programme in the SW lagoon of New Caledonia, element concentrations in organisms from Ouano Beach and Maa Bay could be considered as background concentrations for all elements, except for As and Zn. Furthermore, due to the very high levels of As measured in clams from Ouano Beach, the speciation of As in clam tissues should be determined in detail (particularly their inorganic As content) to assess whether their consumption could represent a potential hazard for local consumers (Warnau et al., 2007; Metian et al., 2008).

400

401 Acknowledgements

402 Authors thank M. Robert (CCA, Univ. La Rochelle) for his analytical advices. LH was
403 beneficiary of a CIFRE scholarship (ANRT, France) supported by the Goro-Nickel Company,

404 New Caledonia. MW is an Honorary Senior Research Associate of the National Fund for
405 Scientific Research (NFSR, Belgium). This work was supported by the IAEA, the French
406 PNEC Programme, the IRD and LIENSs. The IAEA is grateful for the support provided to its
407 Marine Environment Laboratories by the Government of the Principality of Monaco.

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599	Caption to Figures
600	
601	
602	Figure 1. Map of the sampling sites along the SW coast of New Caledonia (Ouano Beach is
603	not represented on the map).
604	
605	
606	Figure 2. Comparisons of element concentrations in bivalves, using multiple comparison test
607	of Tukey performed after 2-way ANOVA in (A) the oyster Isognomon isognomon and (B) the
608	clam Gafrarium tumidum.
609	Mean concentrations are ranked from the left to the right by decreasing order. Concentrations
610	in underlined body compartments or locations are not significantly different ($\alpha = 0.05$).
611	Body compartments: DG (digestive gland), G (gills), M (adductor muscle), VM (visceral
612	mass) and R (remaining soft parts).
613	Sampling locations: OUA (Ouano beach), GR _S (Grande Rade, subtidal), GR _I (Grande Rade,
614	intertidal), DUM (Dumbéa Bay), and KOU (Koutio Bay).
615	

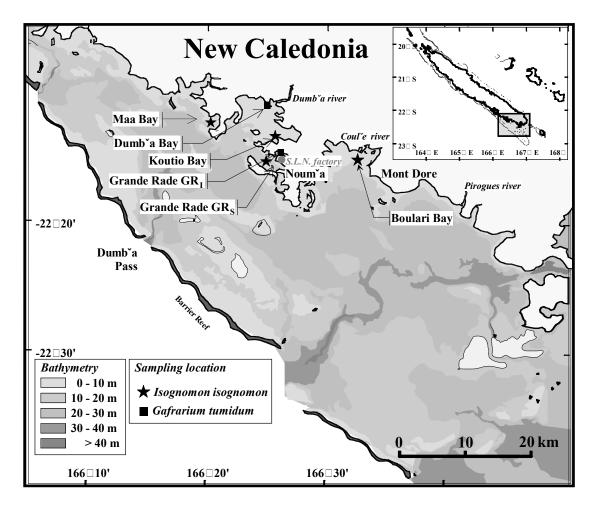


Figure 1.

A- Isognomon isognomon

A-1: Body compartment variation										
Metal	C	ompartm	ent rankir	ng						
Ag	G	VM	R	М						
As	М	G	VM	R						
Cd	R	G	VM	М						
Со	VM	G	R	М						
Cr	VM	G	R	М						
Cu	VM	G	R	М						
Mn	R	VM	G	М						
Ni	G	VM	R	М						
Zn	G	R	VM	М						

A-2: Geo	A-2: Geographical variation											
Metal		Location ranking										
Ag	GR _S	KOU	BOU	MAA								
As	MAA	BOU	GR_S	KOU								
Cd	MAA	KOU	BOU	GR _S								
Со	BOU	GR _s	KOU	MAA								
Cr	KOU	BOU	MAA	GR _S								
Cu	GR _s	BOU	MAA	KOU								
Mn	GRs	BOU	MAA	KOU								
Ni	BOU	GR _s	KOU	MAA								
Zn	MAA	GR _S	KOU	BOU								

B- Gafrarium tumidum

B-1: Body	compartment	variation
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Metal	Comp	artment r	anking
Ag	G	DG	R
As	DG	G	R
Cd	DG	R	G
Co	G	DG	R
Cr	DG	G	R
Cu	DG	G	R
Mn	DG	R	G
Ni	DG	G	R
Zn	DG	G	R

B-2: Geographical variation

Metal	Loc	ation ranl	king		
Ag	GRI	DUM	OUA		
As	OUA	GRI	DUM		
Cd	GR_I	OUA	DUM		
Со	DUM	GRI	OUA		
Cr	GR ₁	DUM	OUA		
Cu	GR _I	DUM	OUA		
Mn	GRI	DUM	OUA		
Ni	GR_{I}	DUM	OUA		
Zn	GRI	DUM	OUA		

Figure 2.

Sampling stations	Ag	As	Cd	Co	Cr	Cu	Mn	Ni	Zn			
	Sediment											
Ouano Beach ¹	< 0.1*	2.9 ± 1.2^{a}	< 0.3*	0.8 ± 0.4 ^a	7.2 ± 2.7^{a}	0.5 ± 1.0^{a}	41.7 ± 15.7^{a}	5.1 ± 3.2^{a}	3.3 ± 1.9^{a}			
Grande Rade GR I ¹	0.4 ± 0.1	8.5 ± 1.6^{b}	2.4 ± 0.3	46.2 ± 9.1^{b}	292 ± 56^{b}	26.9 ± 8.8^{b}	288 ± 40^{b}	797 ± 149^{b}	141 ± 18^{b}			
Maa Bay ²	< 0.1*	6.4 ± 0.3^{a}	1.0 ± 0.2^{b}	4.6 ± 2.3 ^{ab}	44.1 ± 7.9^{a}	10.7 ± 3.5^{b}	132 ± 7.8^{b}	64.2 ± 13.5^{a}	15.2 ± 3.1^{a}			
Koutio Bay ²	< 0.1*	9.9 ± 0.7^{b}	$0.5\pm0.01~^a$	6.2 ± 1.1^{b}	38.6 ± 2.7^{a}	1.1 ± 0.3^{a}	81.6 ± 3.5^{a}	$82.0\pm5.1^{\ a}$	9.4 ± 0.6^{a}			
Boulari Bay ²	< 0.1*	$46.9 \pm 1.5^{\circ}$	$5.1 \pm 0.5^{\circ}$	61.2 ± 16.2 ^c	662 ± 50^{b}	4.6 ± 1.6^{b}	$565 \pm 15^{\circ}$	$900\pm78^{\circ}$	33.1 ± 3.4^{b}			

Table 1. Element concentrations in sediment (mean \pm SD; μ g g⁻¹ dry wt, n = 3).

* Concentrations < detection limit.

^{1, 2} stations where clams and oysters were collected, respectively.

Significant differences among the mean concentrations in sediments from the different sampling stations are indicated by superscripts; means sharing the same superscript ($^{a, b, c}$) are not significantly different among sampling stations ($p_{Tukey} > 0.05$).

Comparisons among sediment concentrations were carried out separately among the stations where clams or oysters were collected.

Table 2. Element concentrations in the oyster *Isognomon isognomon* (mean \pm SD; μ g g⁻¹ dry wt, n = 6).

Body compartments: VM (visceral mass), M (adductor muscle), G (gills), R (remaining soft parts), WSP (whole-soft parts; reconstructed values).

Compartments	% weight	Ag	As	Cd	Со	Cr	Cu	Mn	Ni	Zn	
Koutio Bay											
VM	51 ± 12	21.7 ± 24.3	21.7 ± 5.2	1.13 ± 0.42	1.05 ± 0.65	13.6 ± 2.0	3.0 ± 2.2	26.3 ± 9.8	4.6 ± 3.4	$3,983 \pm 2,555$	
М	26 ± 6	2.4 ± 2.1	20.5 ± 2.9	1.23 ± 0.56	0.15 ± 0.04	2.2 ± 1.7	1.1 ± 0.3	4.4 ± 1.7	< 1.0*	$1,356 \pm 876$	
G	6 ± 2	52.7 ± 23.5	24.2 ± 6.6	1.47 ± 0.63	1.04 ± 0.35	7.1 ± 2.0	8.7 ± 2.7	8.8 ± 3.6	6.9 ± 2.4	$11,357 \pm 5,953$	
R	17 ± 5	9.6 ± 10.7	25.2 ± 5.8	1.59 ± 0.74	0.62 ± 0.34	5.9 ± 1.6	5.7 ± 3.2	30.3 ± 25.3	5.1 ± 1.5	$6,346 \pm 4,224$	
WSP		14.5 ± 7.1^{a}	21.6 ± 2.4^{a}	1.23 ± 0.40^{a}	0.69 ± 0.20^{a}	$9.0\pm1.6^{\ c}$	3.1 ± 0.9^{a}	$20.4\pm8.3^{\ a}$	$3.6\pm1.1^{\ a}$	$3,832 \pm 1,529^{b}$	
Maa Bay											
VM	51 ± 4	2.04 ± 1.81	64.3 ± 9.9	1.97 ± 1.04	0.58 ± 0.26	3.1 ± 0.8	10.6 ± 1.6	17.1 ± 6.4	2.2 ± 1.1	$11,333 \pm 5,904$	
М	25 ± 1	0.12 ± 0.08	106 ± 13	0.81 ± 0.50	0.03 ± 0.03	2.7 ± 0.1	0.4 ± 0.3	4.7 ± 3.9	<0.2*	$5,781 \pm 3,299$	
G	4 ± 2	0.58 ± 0.31	91.5 ± 20.6	3.66 ± 1.79	0.53 ± 0.14	4.8 ± 0.2	14.0 ± 5.0	13.0 ± 5.3	3.8 ± 0.8	$41,790 \pm 14,629$	
R	20 ± 7	0.12 ± 0.08	57.9 ± 9.5	3.01 ± 2.81	0.50 ± 0.001	4.5 ± 0.6	6.6 ± 0.7	55.3 ± 29.6	4.0 ± 0.3	$17,694 \pm 8,103$	
WSP		1.47 ± 1.09^{b}	76.6 ± 9.3^{b}	1.80 ± 1.4^{a}	0.45 ± 0.16^{a}	$3.5\pm0.5^{\ ab}$	6.8 ± 0.5 ^{ab}	22.3 ± 14.6^{a}	$2.2\pm0.5^{\ a}$	$13,817 \pm 6,621$ ^a	
				G	rande Rade (G	R _s)					
VM	28 ± 2	37.1 ± 9.6	39.5 ± 4.5	1.21 ± 0.69	1.38 ± 0.25	3.8 ± 0.5	44.6 ± 17.3	42.8 ± 9.6	8.8 ± 2.0	$8,188 \pm 2,757$	
М	25 ± 3	3.7 ± 2.8	46.0 ± 4.9	0.81 ± 0.19	0.13 ± 0.04	0.6 ± 0.1	1.6 ± 0.3	5.0 ± 2.7	< 0.6*	$1,958 \pm 443$	
G	7 ± 2	217 ± 83	42.4 ± 8.8	0.93 ± 0.28	1.37 ± 0.44	6.3 ± 1.2	13.5 ± 3.5	8.7 ± 3.6	8.3 ± 2.7	$14,360 \pm 3,503$	
R	40 ± 3	18.3 ± 7.0	31.7 ± 4.1	1.46 ± 0.59	0.40 ± 0.14	2.5 ± 0.6	8.8 ± 2.7	52.0 ± 21.1	3.1 ± 0.5	$10,233 \pm 3,724$	
WSP		32.8 ± 6.5 ^c	38.2 ± 4.3 ^c	1.18 ± 0.44^{a}	0.67 ± 0.09^{b}	2.7 ± 0.3^{b}	$17.3 \pm 5.3^{\circ}$	34.7 ± 11.5^{a}	4.4 ± 0.8^{a}	$7,873 \pm 2,087^{a}$	
					Boulari Bay						
VM	28 ± 6	49.4 ± 8.4	59.2 ± 19.2	1.48 ± 0.88	3.29 ± 1.99	7.6 ± 2.9	24.8 ± 12.8	38.8 ± 21.8	26.0 ± 7.9	$1,741 \pm 2,175$	
М	21 ± 3	0.2 ± 0.2	56.5 ± 13.1	0.84 ± 0.37	0.24 ± 0.10	4.2 ± 0.5	1.0 ± 0.1	3.5 ± 1.4	1.7 ± 1.0	279 ± 94	
G	8 ± 2	9.0 ± 5.9	51.2 ± 9.1	0.96 ± 0.50	2.56 ± 0.48	5.3 ± 1.4	7.0 ± 0.6	9.3 ± 3.7	29.8 ± 5.1	$4,\!437 \pm 1,\!880$	
R	43 ± 3	4.3 ± 3.1	45.9 ± 9.6	1.43 ± 0.78	1.18 ± 0.41	5.5 ± 5.1	6.3 ± 1.1	41.8 ± 21.9	14.5 ± 5.2	$2,017 \pm 1,427$	
WSP		16.5 ± 4.0^{a}	51.7 ± 10.8^{d}	1.28 ± 0.68^{a}	1.60 ± 0.49^{a}	$5.7\pm2.9^{\ a}$	$9.8\pm2.1~^a$	30.8 ± 16.0^{a}	16.0 ± 3.7^{b}	$1,718 \pm 1,290^{b}$	

* Concentration < detection limit.

Differences among the concentrations in WSP from the four locations are indicated by superscripts; means sharing the same superscript are not significantly different among sampling stations ($p_{Tukey} > 0.05$)

Factors		Explained Variability (%)									
ractors	Ag	As	Cd	Со	Cr	Cu	Mn	Ni	Zn		
I. isognomon											
Body compartment	20.1	6.6	12.6	27.1	19.6	28.8	49.2	19.6	35.5		
Location	19.1	69.9	16.0	26.0	23.6	17.2	3.9	47.9	29.0		
Compartment × location	43.6	8.9	14.0	14.3	28.2	34.6	7.2	20.8	17.2		
Residual	17.1	14.7	57.5	32.6	28.6	19.3	39.7	11.7	18.4		
G. tumidum											
Body compartment	6.2	3.2	13.2	16.9	9.3	14.2	9.1	10.6	14.9		
Location	39.1	84.1	30.1	23.1	38.6	61.6	22.5	39.4	13.3		
Compartment × location	13.6	3.5	9.6	19.3	16.8	14.4	24.4	15.0	11.		
Residual	41.1	9.2	47.1	40.7	35.3	9.8	43.9	35.0	60.6		

 Table 3. Variability (%) in element concentrations measured in the oyster Isognomon

 isognomon and the clam Gafrarium tumidum explained by the factors considered (body

 compartment and sampling location) and their interaction

Compartments	% weight	Ag	As	Cd	Со	Cr	Cu	Mn	Ni	Zn		
Dumbéa Bay												
DG	14 ± 4	4.6 ± 6.0	70.3 ± 34.3	0.21 ± 0.11	4.6 ± 1.8	8.4 ± 4.3	22.0 ± 9.0	14.5 ± 9.9	33.9 ± 14.2	105 ± 42		
G	12 ± 3	0.49 ± 0.55	39.6 ± 13.1	0.20 ± 0.11	4.5 ± 2.0	2.5 ± 1.3	7.4 ± 3.0	25.2 ± 26.3	37.9 ± 13.5	76.7 ± 24.1		
R	75 ± 3	1.1 ± 0.9	32.8 ± 8.1	0.16 ± 0.02	3.6 ± 0.7	4.6 ± 1.6	5.9 ± 1.1	34.0 ± 36.5	29.0 ± 6.8	55.2 ± 9.8		
WSP		1.4 ± 1.1^{a}	$37.4\pm7.4^{\text{ a}}$	$0.17\pm0.03^{\ a}$	3.8 ± 0.7^{a}	4.8 ± 1.3^{a}	7.9 ± 1.3 ^a	$35.9\pm43.5^{\ a}$	$30.2\pm6.0^{\ a}$	62.7 ± 10.2^{a}		
	Ouano Beach											
DG	14 ± 3	< 1.4*	606 ± 135	0.33 ± 0.04	1.8 ± 0.2	3.5 ± 1.1	14.6 ± 2.7	5.0 ± 2.5	9.2 ± 1.7	78.3 ± 10.4		
G	11 ± 3	< 0.01*	516 ± 117	0.19 ± 0.09	1.8 ± 0.5	1.9 ± 1.5	6.4 ± 2.2	7.9 ± 2.9	14.1 ± 4.4	89.7 ± 27.9		
R	76 ± 5	< 0.02*	360 ± 121	0.16 ± 0.06	1.0 ± 0.3	3.1 ± 2.8	4.4 ± 1.1	5.9 ± 1.6	7.1 ± 1.5	50.7 ± 8.2		
WSP		$< 0.02^{*a}$	441 ± 84^{b}	0.19 ± 0.04^{a}	1.1 ± 0.2^{b}	3.2 ± 2.2^{a}	$5.6\pm1.0^{\text{ a}}$	$5.5\pm1.5^{\ a}$	8.1 ± 1.5 ^b	55.6 ± 7.8^{a}		
				Gra	nde Rade (GH	R _I)						
DG	29 ± 6	51.5 ± 33.6	67.9 ± 14.6	1.30 ± 0.88	2.4 ± 1.4	$10.7 \pm .4$	146 ± 37	324 ± 260	91.7 ± 45.8	282 ± 276		
G	12 ± 2	89.4 ± 75.6	63.2 ± 18.5	0.21 ± 0.09	5.6 ± 3.6	12.1 ± 3.4	119 ± 40	27.9 ± 20.3	49.0 ± 32.0	123 ± 65		
R	59 ± 7	16.3 ± 10.8	47.1 ± 16.2	0.52 ± 0.46	1.5 ± 1.0	5.8 ± 1.8	34.3 ± 17.5	93.4 ± 86.2	29.7 ± 9.6	74.5 ± 12.7		
WSP		33.1 ± 13.4^{b}	55.0 ± 15.1^{a}	$0.74 \pm 0.25^{\ b}$	$2.2 \pm 1.0^{\circ}$	$8.0\pm1.7^{\text{ b}}$	77.3 ± 17.5^{b}	139 ± 104^{b}	$52.3 \pm 11.9^{\circ}$	$154\pm102^{\text{ b}}$		

Table 4. Element concentrations in the clam *Gafrarium tumidum* (mean \pm SD; μ g g⁻¹ dry wt, n = 6).

Body compartments: DG (digestive gland), G (gills), R (remaining soft parts), WSP (whole-soft parts; reconstructed values).

* concentration < detection limit.

Differences among the concentrations in WSP from the four locations are indicated by superscripts; means sharing the same superscript are not significantly different among sampling stations ($p_{Tukey} > 0.05$).

Species	Location	Ag	As	Cd	Со	Cr	Cu	Mn	Ni	Zn
Clams										
$Ga frarium tumidum$ 1	Hong-Kong					0.67	5.77		5.59	57.5
G. tumidum 2	Fiji				1.0 - 2.8	1.0 - 1.6	4.2 - 11.0	28 - 45	1.7 - 4.5	
Anadara antiquate ²	Fiji				0.9 - 2.5	0.8 - 1.8	4 - 13	32 - 50	2 - 4	
Chione subrugosa ³	Tropical mangrove lagoon			$0.72 \pm 0.09 - 2.25 \pm 0.5$	$0.13 \pm 0.14 - 1.1 \pm 0.17$	1.48 ± 0.28 - 1.93 ± 0.53	$20.8 \pm 1.49 - 23.4 \pm 1.43$	4.08 ± 0.21 - 4.55 ± 0.08	$2.32 \pm 0.35 - 2.65 \pm 0.46$	$51 \pm 4 - 73 \pm 11$
<i>Circe sinensis</i> ¹	Hong-Kong					2.26	3.13		2.8	43.7
Codakia orbicularis ⁴	Dominican Republic			3.8		1.66	3.08		1.57	22.9
Ruditapes philippinarum ¹	Hong-Kong					0.9	3.99		4.66	98
Tellina fausta ⁴	Dominican Republic			0.04		4.15	14.1		4.91	51.4
Circentia callipyga ⁵	Qatar	3.03	156	1.17	4.45	0.97	8.35	17.7	23.9	69.1
Oysters										
Isognomon isognomon ⁶	Phuket, Thailand						< 150			900 - 2,000
	-			0.47 ± 0.23 -			11 ± 0.51 -			23.8 ± 0.75 -
I. alatus ⁷	Malaysia			3.71 ± 0.12			30.7 ± 0.8			334.5 ± 12.5
I. alatus ⁸	Venezuela			0.33 - 0.91		0.46 - 1.2	14 - 49.1		11 - 18	0.25 - 2.1
I. alatus ⁹	Colombian Caribbean			0.8 - 15.6			0.42 - 52.3			
I. alatus ⁴	Dominican Republic			0.24 - 0.26		2.38 - 4.96	7.58 - 19.7		1.25 - 2.90	4,000 - 4,010
I. alatus ¹⁰	Guadeloupe					0.23 - 7.2	6.8 - 127			1,060 - 12,160
I. alatus ¹⁰	Martinique					0.32 - 1.75	5.4 - 248			2,460 - 11,530
I. bicolor ⁹	Colombian Caribbean			0.98 - 6.99			0.8 - 3.94			
I. legumen ¹¹	Taiwan						491 ± 29			
Isognomon sp. ¹²	Biscayne Bay, Florida		37.3 ± 6.9							
Ostrea sandvicensis ¹³	Hawaii						1,400		20	
Saccostrea amasa 14	North Queensland, Australia			1 - 12						673 – 20,906
S. echinata ¹⁵	North Queensland, Australia			0.69 - 2.34						$2,080 \pm 453$
S. echinata ¹⁶	North Queensland, Australia			0.198 - 4.63						325 - 4,680
Crassostrea belcheri ¹⁷	Merbok estuary, Malaysia						1 - 8.5			30 - 550
C. cucullata 18	Goa, India		2.3 - 6.3				251 - 728	33.2 - 17.5		446 - 2,800
<i>C. echinata</i> ¹⁴	Cleveland bay, Australia									673 – 20,906
C. gryphoides ¹⁸	Goa, India		3.2 - 5.8				175 - 210			325 - 550
C. iredalei ¹⁷	Merbok estuary, Malaysia						4 - 8			80 - 550
C. gigas ¹⁹	Derwent Estuary, Tasmania,									38,700
C. virginica ¹¹	Taiwan						257 ± 196			$1,037 \pm 778$

Table 5. Element concentrations (mean \pm SD or range; μ g g⁻¹ dry wt) in clams and oysters from tropical and subtropical areas.

 $\frac{257 \pm 196}{1,037 \pm 778}$ ¹ Cheung and Wong, 1997, ² Dougherty, 1988, ³ Szefer et al., 1998, ⁴ Sbriz et al., 1998, ⁵ de Mora et al., 2004, ⁶ Brown and Holley, 1982, ⁷ Saed et al., 2001, ⁸ Jaffe et al., 1998, ⁹ Campos, 1988, ¹⁰ RNO-Antilles, unpublished work, ¹¹ Hung et al., 2001, ¹² Valette-Silver et al., 1999, ¹³ O'Connor, 1989, ¹⁴ Jones, 1992, ¹⁵ Jones et al., 2000, ¹⁶ Olivier et al., 2002, ¹⁷ Lim et al., 1995, ¹⁸ Zingde et al., 1976