

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

Laetitia Hédouin, Paco Bustamante, Carine Churlaud, O. Pringault, Renaud

Fichez, Michel Warnau

To cite this version:

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 /i.ecoenv.2008.04.004. hal-00336151

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

Submitted on 2 Nov 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

```
* Manuscript
```


Abstract

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

Keywords: Tropical Environment, Metals, Bioindicators, Mining activities

1. Introduction

 Surrounded by a barrier reef of 1600 km, the New Caledonia lagoon is one of the largest in the world (Labrosse et al., 2000). However, the lagoon of New Caledonia is subjected to important anthropogenic inputs of metals mainly due to intense land-based Ni mining activities but also to urban development and lack of efficient wastewater treatment. Open-cast mining exploitation presently constitutes the major economical resource of the Territory and results in important coastal discharges of metals, which constitute a threat to coral reef 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, 2001), making the extraction from low Ni grade ores (limonite) possible. The acidic extraction of metals is not Ni-selective and makes also soluble all other ore-contained by- product metals. Therefore, the lixiviation process will obviously lead to an increased multi-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

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

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

2. Materials and methods

2.1. Sampling sites

 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 94 Bay and Grande Rade (GR_S). Maa Bay is subjected to low anthropogenic and terrigenous inputs and was considered as the relative "reference" station for oysters. Koutio Bay is influenced by inputs of domestic wastes from Noumea City and by the occurrence of an important rubbish dump. Boulari Bay is under the influence of La Coulée River that delivers 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 the metallurgic factory "Société Le Nickel" (SLN).

 For clams, three intertidal sampling stations were selected: Ouano Beach, Dumbéa Bay 102 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. 104 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.

2.2. Organisms

 The clam *Gafrarium tumidum* was collected by handpicking in the intertidal stations. The 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 reduce as much as possible the variability of element concentrations due to season or sexual cycle. Body size is well-known to affect metal concentrations in organisms (eg, Boyden, 1977; Warnau et al., 1995); therefore only samples with shell width longer than 35 mm for *G. 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 ± 4.9 g; oysters (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 in 30 l of seawater from the same sampling station to allow for depurating gut contents and particulate material present in the mantle cavity. Three body compartments for the clams (digestive gland, gills and remaining soft parts) and four body compartments for the oysters (visceral mass, gills, adductor muscle and remaining soft parts) were removed from the shells. The separated body compartments were weighed (wet wt), dried at 60°C until constant weight, and weighed again (dry wt). They were then stored in acid-washed, hermetically sealed PET containers until analysis for their metal contents.

2.3. Sediments

 In parallel to organisms, superficial sediments (top 3-cm layer) were analysed in all the 129 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.

2.4. Sample preparation and analysis

 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 138 MARS[®] V microwave (30 min with constantly increasing temperature up to 100° C for sediment and up to 115°C for biota, then 15 min at this maximal temperature). These conditions allowed for a complete digestion of the biological matrices and a strong although not total (highly refractory humic acids may resist) leaching of the sediment (eg, Coteur et al., 2003; Dalto et al., 2006). Each sample was eventually diluted to 30 to 50 ml with milli-Q water according to the amount of sample digested.

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

2.5. Statistical analysis

 Comparisons of the data were performed using 1- or 2-way analysis of variance (ANOVA) followed by the multiple comparison test of Tukey (Zar, 1996). Two-way ANOVA was used with sampling location and body compartment as fixed factors. The 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 $\alpha =$ 0.05.

3. Results

3.1. Sediments

 Table 1 shows the element concentrations measured in the sediment collected in the different stations. Except for Ag, for which comparison among stations was not possible due 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 highest concentrations for all elements. Concentrations in all elements were always 168 significantly higher in Grande Rade (GR_I) than in Ouano Beach ($p_{Tukev} < 0.0001$). Concentrations in sediment from Boulari Bay were significantly higher (up to 1 order of magnitude) than those measured in the other stations where oysters were also sampled.

3.2. The oyster Isognomon isognomon

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

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

3.2.1. Geographical variation

 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 comparison tests on the mean concentrations indicated that one sampling location displayed generally the highest concentrations for one or several elements, whereas the three other locations did not show significant difference in element concentrations, except for Mn (no significant difference among none of the sampling stations) and As (all stations significantly different from each others).

 Concentrations of Co and Ni in the oysters were significantly higher in Boulari Bay than 194 in Maa Bay ($p_{Tukey} \le 0.001$; Table 2, Fig. 2A). Oysters from Grande Rade (GR_S) displayed 195 significantly higher Ag and Cu concentrations than those measured in Maa Bay ($p_{\text{Tukev}} \le 0.01$) 196 whereas the highest Cr concentrations were measured in oysters from Koutio Bay ($p_{Tukey} \leq$ 0.05). In contrast to all other elements, As and Zn concentrations were higher in oysters from Maa Bay.

 Geographical variation of the element concentrations in the whole-soft parts of the oysters (reconstructed data) were tested using 1-way ANOVA and Tukey test. Results were similar to those from the 2-way ANOVA performed on body-compartment specific concentrations, 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) for Zn (Table 2). The particular opposite pattern of As and Zn displaying highest 205 concentrations in Maa Bay (up to 77 µg As g^{-1} dry wt and 13,817 µg Zn g^{-1} dry wt) was confirmed in whole soft parts data treatment.

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 216 (43 %) in the gills of oysters from Grande Rade (GR_S) compared to those from the other stations (5 - 24 %).

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 223 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 227 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 232 to the sampling locations (2-way ANOVA, $p_{\text{sampling locations}}$ 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 235 Grande Rade (GR_I) . In contrast the concentrations of As were significantly higher in Ouano 236 Beach compared to all the other locations ($p_{\text{Tukev}} \le 0.001$; 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 body- compartment specific concentrations, except for Co which showed significant differences 241 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 243 up to 441 μ g g⁻¹ dry wt.

3.3.2. Body distribution

 The mean concentrations of all elements investigated differed according to the body 246 compartments (2-way ANOVA, $p_{body\,\,comment}$ always ≤ 0.003). Multiple comparison tests of 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 concentrations were similar in the digestive gland and the gills. No major difference was found when considering body distribution in clams collected from Ouano Beach and Dumbéa 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 (GR_I) , the elements were similarly distributed between the remaining soft parts and the digestive gland.

4. Discussion

 Sediments are a sink for marine contaminants (eg, Salomons et al., 1987) and their 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_I) may be considered as highly contaminated stations compared to Ouano Beach and Maa Bay. In turn, the two latter ones may be defined as relatively non-contaminated stations (see Table 1). However, it is now well known that sediment-associated concentrations are not necessarily representative of the contaminant fraction that is bioavailable, viz the fraction of "direct ecotoxicological relevance" for marine organisms (Phillips and Rainbow, 1993). Therefore, the present study was carried out to assess the usefulness of the oyster *Isognomon isognomon* and the clam *Gafrarium tumidum* as sentinel species over sediment for Ag, As, Cd, Co, Cr, 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).

 The elevated concentrations of Co and Ni measured in oysters from Boulari Bay strongly 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 further confirmed by the high concentrations of Co, Cr, Mn and Ni measured in the sediment from Boulari Bay. However, element analysis in oyster tissues showed that other stations, not 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 concentrations recorded in oysters suggest that Maa Bay would be subjected to agrochemical 279 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 283 Ouano Beach were always lower than in those from Grande Rade (GR_I) . 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 287 elements considered, except for As. In contrast, Grande Rade (GR_I) can be defined as a highly contaminated station for Ag, Cr, Cu, Mn, Ni and Zn.

 In this work, the distribution of the considered elements in bivalve tissues was also investigated in order to possibly identify some organs that could be more sensitive than the 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 compartments of the clam *G. tumidum*, the digestive gland displayed the highest bioconcentration capacity. In addition, the concentrations measured in this organ easily allowed discriminating the stations according to their contamination levels. Hence, this organ could be proposed as a target for future biomonitoring programmes. In the oyster *I. isognomon*, no clear trends could be observed in bioaccumulation and geographical discrimination ability among the different body compartments. Consequently, in a future biomonitoring programme, consideration of the whole soft parts of oysters could be 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 305 this metal. Indeed, Ni concentrations in clams from Grande Rade (GR_I) were higher (52 \pm 12 μ 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 312 subtropical bivalves are generally lower than 30 μ g g⁻¹ dry wt (< 10 μ g g⁻¹ dry wt if one 313 considers clams and oysters; see Table 5). However, two studies on sub-tropical areas 314 indicated elevated As concentrations in *Isognomon* spp from Florida (37.3 \pm 6.9 µg g⁻¹ dry 315 wt) (Valette-Silver et al., 1999) and in the clam *Circentia callipyga* from the Gulf of Oman $(156 \text{ µg g}^{-1} \text{ dry wt})$ (de Mora et al., 2004). In the present study, As was found to reach 317 extremely high concentrations in the clams from Ouano Beach (441 \pm 84 µg g⁻¹ dry wt) 318 compared to those observed in Grande Rade (GR_I) (55 \pm 15 µg g⁻¹ dry wt) and in the oysters 319 from Maa Bay (77 \pm 9 µg g⁻¹ dry wt). To the best of our knowledge, such high body 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 322 as in the cirratulid polychaete *Tharyx marioni* which displays extremely high body 323 concentrations of total As $(2000 \mu g g^{-1}$ dry wt; Gibbs et al., 1983), the Mediterranean fan 324 worm *Sabella spallanzanii* which shows As concentrations higher than 1000 μ g g⁻¹ dry wt in 325 its branchial crown (Fattorini and Regoli, 2004) or the very high arsenic concentrations 326 monitored in muscles of edible fish (500 μ g g⁻¹ dry wt) from the Bay of Cienfuegos, Cuba, a 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 concentrations in *G. tumidum* tissues is not clear. Some authors have reported that As concentrations in organisms were related to the sediment concentrations (such as in *Scrobicularia plana*; Langston, 1980). However no similar correlation was observed here. In addition, laboratory experiments have shown that bivalves generally displayed a limited capacity in accumulating As from seawater (eg, Ünlü and Fowler, 1979; Hédouin, 2006; Gómez-Batista et al., 2007). Thus, the elevated As concentrations reported in this study would 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 proposed as the main route of uptake for As in bivalves, suggesting that food of both oysters and clams are enriched in As in Maa Bay and Ouano Beach, compared to the other sampling locations. Whereas some agricultural activities are carried out in Maa Bay, Ouano Beach is rather subjected to waste discharges from shrimp aquaculture. Hence the important discharges of N-enriched products (due to terrestrial leaching of fertilisers used for local agriculture or to release of aquaculture food excesses) could locally modify the N:P ratio. In environments with phosphate deficit relative to nitrogen, phytoplankton metabolises As much more easily (Benson and Summons, 1981; Phillips, 1990a). This in turn may lead to enhanced trophic transfer of As to filter-feeders and enhanced As accumulation in the tissues of the bivalves (Warnau et al., 2007). Although further investigations are needed to validate such a hypothesis in Maa Bay and Ouano Beach, the extremely high As levels measured in clam tissues are of considerable interest because (1) *G. tumidum* is a seafood product in New Caledonia and (2) little is known about the speciation of As in the tissues of this species (Francesconi et al., 1999) which determines its potential toxicity to consumers (see eg, Kaise and Fukui, 1992; Warnau et al., 2007).

 I. isognomon displayed also very high Zn concentrations in Maa Bay and in Grande Rade 353 (GRs), viz 13,817 \pm 6,621 and 7,873 \pm 2,087 μ g g⁻¹ dry wt, respectively. Elevated

354 concentrations of Zn have been reported for *I. alatus*, reaching 4,010 μ g Zn g⁻¹ dry wt in 355 individuals collected in the Dominican Republic and 12,163 μ g g⁻¹ dry wt in the Guadeloupe (see Table 5). Although, Zn is well known to be essential to organisms, acting for example as a co-factor in numerous metalloenzymes (eg, Vallee and Falchuk, 1993), the amounts accumulated are clearly far above the physiological needs of the bivalve. *I. isognomon* must therefore possess a natural capacity to accumulate Zn up to very high levels while avoiding 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). Indeed, in many bivalves and especially in oysters, granules may contain up to 60 % of the total body load of Zn (Eisler, 1981).

 Ag is well known as a highly toxic metal (eg, Warnau et al, 1996a; Ratte, 1999) and the scarcity of data concerning Ag levels in tropical and subtropical organisms in general, and in particular in clams and oysters, is therefore quite surprising (see Table 5). In this way, the concentrations measured in the two investigated species (see Tables 2 and 4) can be considered as baseline data for the New Caledonia lagoon as well as for other tropical environments. Clams and oysters collected from Grande Rade displayed quite elevated Ag 370 concentrations (33 \pm 13 and 33 \pm 7 µg g⁻¹ dry wt, respectively), which are one to two orders 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 373 areas (< 1 μ g g⁻¹ dry wt in mussels; Klumpp and Burdon-Jones, 1982) and temperate areas (< 6μ g g⁻¹ dry wt in clams and oysters; Cohen et al., 2001). Various bivalves are able to accumulate Ag up to very high concentrations by trapping it as Ag2S, a stable and non-toxic compound (eg, Berthet et al., 1992; Bustamante and Miramand, 2005). The occurrence of a 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

 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.

5. Conclusions

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

Acknowledgements

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

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

References

- Benson, A.A., Summons, R.B., 1981. Arsenic accumulation in Great Barrier reef invertebrates. Science 211, 482-483.
- Berthet, B., Amiard, J.C., Amiard-Triquet, C., Martoja, R., Jeantet, A.Y., 1992.

Bioaccumulation toxicity and physico-chemical speciation of silver in bivalve molluscs:

ecotoxicological and health consequences. Sci. Total Environ. 125, 97-122.

- Boyden, C.R., 1977. Effect of size upon metal content of shellfish. J. Mar. Biol. Assoc. U. K. 57, 675-714.
- Breau, L. 2003. Etude de la bioaccumulation des métaux dans quelques espèces marines

tropicales: recherche de bioindicateurs de contamination et application à la surveillance de

l'environnement côtier dans le lagon sud-ouest de la Nouvelle-Calédonie. Ph.D. Thesis,

- Université de La Rochelle, France.
- Brown, B.E., Holley, M.C., 1982. Metal levels associated with tin dredging and smelting and their effect upon interdital reef flats at Ko Phuket, Thailand. Coral Reefs 1, 131-137.
- Bustamante, P., Grigioni, S., Boucher-Rodoni, R., Caurant, F., Miramand, P., 2000.
- Bioaccumulation of 12 trace elements in the tissues of the *Nautilus macromphalus* from New Caledonia. Mar. Pollut. Bull. 40, 688-696.
- Bustamante, P., Miramand, P., 2005. Subcellular and body distributions of 17 trace elements
- in the variegated scallop *Chlamys varia* from the French coast of the Bay of Biscay. Sci.
- Total Environ. 337, 59-73.
- Campos, N.H., 1988. Selected bivalves for monitoring of heavy metal contamination in the
- Colombian Caribbean. In: Seeliger, U., De Lacerda, L.D., Patchineelam, S.R. (Eds),
- Metals in coastal environments of Latin America. Springer-Verlag, pp. 270-275.
- Cheung, Y.H., Wong, M.H., 1997. Depuration and bioaccumulation of heavy metals by clams from Tolo Harbour, Hong Kong. Toxicol. Environ. Chem. 58, 103-116.
- Cohen, T., Que Hee, S.S., Ambrose, R.F., 2001. Trace metals in fish and invertebrates of
- three California coastal wetlands. Mar. Pollut. Bull. 42, 224-232.
- Corrêa Junior, J.D., Allodi, S., Amado Filho, G.M., Farina, M., 2000. Zn accumulation in
- phosphate granules of *Ucides cordatus* hepatopancreas. Braz. J. Med. Biol. Res. 33, 217- 221.
- Coteur, G., Gosselin, P., Wantier, P., Chambost-Manciet, Y., Danis, B., Pernet, P., Warnau,
- M., Dubois, P., 2003. Echinoderms as bioindicators, bioassays, and impact assessment tools of sediment-associated metals and PCBs in the North Sea. Archives of Environ.
- Contam. Toxicol. 45, 190-202.
- Dalto, A.G., Grémare, A., Dinet, A., Fichet, D., 2006. Muddy-bottom meiofauna responses to
- metal concentrations and organic enrichment in New Caledonia South-West Lagoon. Estuar. Coast. Shelf Sci. 64, 629-644.
- de Mora, S., Fowler, S.W., Wyse, E., Azemard, S., 2004. Distribution of heavy metals in marine bivalves, fish and coastal sediments in the Gulf of Oman. Mar. Pollut. Bull. 49, 410-424.
- Dougherty, G., 1988. Heavy metal concentrations in bivalves from Fiji's coastal waters. Mar. Pollut. Bull. 19, 81-84.
- Eisler, R., 1981. Trace metal concentrations in marine organisms. Pergamon Press, New York.
- Fattorini, D., Alonso Hernandez, C.M., Diaz Asencio, M., Munoz Caravaca, A., Pannacciulli,
- F., Tangherlini, M., Regoli, F., 2004. Chemical speciation of arsenic in different marine organisms: importance in monitoring studies. Mar. Environ. Res. 58, 845-850.
- Fattorini, D., Regoli, F., 2004. Arsenic speciation in tissues of the Mediterranean polychaete *Sabella spallanzanii*. Environ. Toxicol. Chem. 23, 1881-1887.
- Francesconi, K.A., Gailer, J., Edmonds, J.S., Goessler, W., Irgolic, K.J., 1999. Uptake of
- arseno-betaines by the mussel *Mytilus edulis*. Comp. Biochem. Physiol. C Comp. Pharmacol. 122, 131-137.
- Gibbs, P.E., Langston, W.J., Burt, G.R., Pascoe, P.L., 1983. *Tharyx marioni* (Polychaeta): a
- remarkable accumulator of arsenic. J. Mar. Biol. Assoc. UK 63, 313-325.
- Goldberg, E.D., Koide, M., Hodge, V., Flegal, A.R., Martin, J.H., 1983. U.S. Mussel Watch:
- 1977-1978 results on trace metals and radionuclides. Estuar. Coast. Shelf Sci. 16, 69-93.
- Gómez-Batista, M., Metian, M., Teyssié, J.L., Alonso-Hernández, C., Warnau, M., 2007.
- Bioaccumulation of dissolved arsenic in the oyster *Crassostrea virginica*: a radiotracer study. Environ. Bioindic. 2, 237-244.
- Goro-Nickel, 2001. Projet Goro Nickel. Evaluation environnementale.
- Hédouin, L., 2006. Caractérisation d'espèces bioindicatrices pour la surveillance des activités
- minières et la gestion de l'environnement en milieu récifal et lagonaire: application au lagon de Nouvelle-Calédonie. Ph.D. Thesis, Université de La Rochelle, France.
- Hédouin, L. Metian, M., Teyssié, J.L., Fichez, R., Warnau., M., submitted. Delineation of
- heavy metal contamination pathways (seawater, food and sediment) in tropical oysters
- from New Caledonia using radiotracer techniques. submitted to Mar. Pollut. Bull.
- Hédouin, L., Metian, M., Teyssié, J.L., Fowler, S.W., Fichez, R., Warnau, M., 2006.
- Allometric relationships in the bioconcentration of heavy metals by the edible tropical
- clam *Gafrarium tumidum*. Sci. Total Environ. 366, 154-163.
- Hédouin, L., Pringault, O., Metian, M., Bustamante, P., Warnau, M., 2007. Nickel bioaccumulation in bivalves from the New Caledonia lagoon: Seawater and food exposure. Chemosphere 66, 1449-1457.
- Hung, T.-C., Meng, P.-J., Han, B.-C., Chuang, A., Huang, C.-C., 2001. Trace metals in different species of mollusca, water and sediments from Taiwan coastal area. Chemosphere 44, 833-841.
- IMWC (International Mussel Watch Committee), 1995. International Mussel Watch Project -
- Initial Implementation Phase, Final Report. In: Farrington, J.W., Tripp, B.W. (Eds),
- NOAA Technical Memorandum NOS ORCA 95. NOAA Office of Ocean Resources Conservation and Assessment, Rockville, MD.
- Jaffe, R., Leal, I., Alvarado, J., Gardinali, P.R., Sericano, J.L., 1998. Baseline study on the levels of organic pollutants and heavy metals in bivalves from the Morrocoy National park, Venezuela. Mar. Pollut. Bull. 36, 925-929.
- Jones, G.B., 1992. The effects of *Trichodesmium* blooms on water quality in the Great Barrier
- Reef Lagoon. In: Carpenter, E.J., Capone, D.G., Rueter, J.G. (Eds), Marine pelagic
- cyanobacteria *Trichodesmium* and other diazotrophs. Kluwer Academic Press, Boston, pp. 273-287.
- Jones, G.B., Mercurio, P., Olivier, F., 2000. Zinc in fish, crabs, oysters, and mangrove flora and fauna from Cleveland bay. Mar. Pollut. Bull. 41, 345-352.
- Kaise, T., Fukui, S., 1992. The chemical form and acute toxicity of arsenic compounds in marine organisms. Appl. Organomet. Chem. 6, 155-160.
- Klumpp, D.W., Burdon-Jones, C., 1982. Investigations of the potential of bivalve molluscs as indicators of heavy metals in tropical marine waters. Aust. J. Mar. Freshw. Res. 33, 285-
- 300.
- Labrosse, P., Fichez, R., Farman, R., Adams, T., 2000. New Caledonia. In: Sheppard, C.R.C.
- (Ed.), Seas at the Millenium: An environmental evaluation. Pergamon, Amsterdam, pp. 723-736.
- Langston, W.J., 1980. Arsenic in U.K. estuarine sediments and its availability to benthic organisms. J. Mar. Biol. Assoc. U. K. 60, 869-881.
- Lim, P.E., Lee, C.W., Din, Z., 1995. Accumulation of heavy metals by cultured oysters from
- Merbok estuary, Malaysia. Mar. Pollut. Bull. 31, 420-423.
- Luoma, S.N., Bo, Y.B., Bryan, G.W., 1995. Fate, bioavailability and toxicity of silver in estuarine environments. Mar. Pollut. Bull. 31, 44-54.
- Martin, M., Stephenson, M.D., Smith, D.R., Gutierrez-Galindo, E.A., Flores Munoz, G.,
- 1988. Use of silver in mussels as a tracer of domestic wastewater discharge. Mar. Pollut. Bull. 19, 512-520.
- Metian, M., 2003. Bioaccumulation des métaux lourds chez 4 espèces marines du lagon de
- Nouvelle Calédonie: Caractérisation de leur potentiel bioindicateur pour le monitoring des activités minières locales. Master Thesis, Université Libre de Bruxelles, Belgium.
- Metian, M., Bustamante, P., Hédouin, L., Warnau, M., 2008. Accumulation of trace elements
- in the tropical scallop *Comptopallium radula* from coral reefs in New Caledonia. Environ.
- Pollut. 152, 543-552.
- Mihaylov, I., Krause, E., Colton, D.F., Okita, Y., Duterque, J.-P., Perraud, J.-J., 2000. The
- development of a novel hydrometallurgical process for nickel and cobalt recovery from
- Goro laterite ore. Canadian Mining Metallurgical Bull. 93, 124-130.
- Monniot, F., Martoja, R., Monniot, C., 1994. Cellular sites of iron and nickel accumulation in
- ascidians related to the naturally and anthropic enriched New Caledonian environment.
- Ann. Inst. Oceanogr. 70, 205-216.
- Moore, N.W., 1966. A pesticide monitoring system with special reference to the selection of
- indicator. J. Appl. Ecol. 3, 261-269.
- O'Connor, T.P., 1989. A summary of data on tissue contamination from the first three years
- (1986-1988) of the Mussel Watch project. Technical Memorandum NOS OMA 49,
- National Oceanic and Atmospheric Admistration, USA, pp. 22.
- Olivier, F., Ridd, M., Klumpp, D., 2002. The use of transplanted cultured tropical oysters
- (*Saccostrea commercialis*) to monitor Cd levels in North Queensland coastal waters
- (Australia). Mar. Pollut. Bull. 44, 1051-1062.
- Phillips, D.J.H., 1990a. Arsenic in aquatic organisms: a review, emphasizing chemical speciation. Aquat. Toxicol. 16, 151-186.
- Phillips, D.J.H., 1990b. Use of macroalgae and invertebrates as monitors of metal levels in estuaries and coastal waters. In: Furness, R.W., Rainbow, P.S. (eds.), Heavy metals in the
- marine environment. CRC Press, Boca Raton, pp. 81-99.
- Phillips, D.J.H., 1991. Selected trace elements and the use of biomonitors in subtropical and tropical marine ecosystems. Rev. Environ. Contam. Toxicol. 120, 105-128.
- Phillips, D.J.H., Rainbow, P.S., 1993. Biomonitoring of trace aquatic contaminants, London.
- Rainbow, P.S., 1995. Biomonitoring of heavy metal availability in the marine environment.
- Mar. Pollut. Bull. 31, 183-192.
- Ratte, H.T., 1999. Bioaccumulation and toxicity of silver compounds: a review. Environ.
- Toxicol. Chem. 18, 89-108.
- RNO-Antilles, Réseau National d'Observation de la Qualité du Milieu Marin-Antilles. Ifremer
- et Ministère de l'Ecologie et du Développement Durable, unpublished work.
- Saed, K., Ismail, A., Omar, H., Kusnan, M., 2001. Accumulation of heavy metals (Zn, Cu, Pb,
- Cd) in flat-tree oysters *Isognomon alatus* exposed to pig farm effluent. Toxicol. Environ.
- Chem. 82, 45-58.
- Salomons, W., de Rooij, N.M., Derdijk, H., Bril, J., 1987. Sediments as a source for contaminants? Hydrobiologia 149, 13-30.
- Sanders, J.G., Osman, R.W., Riedel, G.F., 1989. Pathways of arsenic uptake and incorporation in estuarine phytoplankton and the filter-feeding invertebrates *Eurytemora affinis, Balanus improvisus* and *Crassostrea virginica*. Mar. Biol. 103, 319-325.
- Sañudo-Willhelmy, S., Flegal, R., 1992. Anthropogenic silver in the Southern California
- Bight: a new tracer of sewage in coastal waters. Environ. Sci. Technol. 26, 2147-2151.
- Sbriz, L., Aquino, M.R., Alberto de Rodriguez, N.M., Fowler, S.W., Sericano, J.L., 1998.
- Levels of chlorinated hydrocarbons and trace metals in bivalves and nearshore sediments
- from the Dominican Republic. Mar. Pollut. Bull. 36, 971-979.
- Szefer, P., Geldon, J., Ali, A.A., Paez-Osuna, F., Ruiz-Fernandes, A.C., Galvan, S.R.G.,
- 1998. Distribution and association of trace metals in soft tissue and byssus of *Mytella strigata* and other benthal organisms from Mazatlan harbour, mangrove lagoon of the northwest coast of Mexico. Environ. Int. 24, 359-374.
- Ünlü, M.Y., Fowler, S.W., 1979. Factors affecting the flux of arsenic through the mussel *Mytilus galloprovincialis*. Mar. Biol. 51, 209-219.
- UNU, 1994. Report of the UNU-IOC Workshop on Asia/Pacific Mussel Watch: Monitoring,
- Research and Training United Nations University, 18-21 November 1994, Bali, Indonesia
- Valette-Silver, N.J., Riedel, G.F., Crecelius, E.A., Windom, H., Smith, R.G., Dolvin, S.S.,
- 1999. Elevated arsenic concentrations in bivalves from the southeast coasts of the USA.
- Mar. Environ. Res. 48, 311-333.
- Vallee, B.L., Falchuk, K.H., 1993. The biochemical basis of zinc physiology. Physiol. Rev. 73, 79-118.
- Warnau, M., Biondo, R., Temara, A., Bouquegneau, J.M., Jangoux, M., Dubois, P., 1998.
- Distribution of heavy metals in the echinoid *Paracentrotus lividus* (Lmk) from the
- Mediterranean *Posidonia oceanica* ecosystem: seasonal and geographical variations. J. Sea Res. 39, 267-280.
- Warnau, M., Fowler, S.W., Teyssié, J.L., 1999. Biokinetics of radiocobalt in the asteroid *Asterias rubens* (Echinodermata): sea water and food exposures. Mar. Pollut. Bull. 39:
- 159-164.
- Warnau, M., Gómez-Batista, M., Alonso-Hernández, C., Regoli, F., 2007. Arsenic: Is it worth monitoring in the Mediterranean Sea? In: *Marine Sciences and Public Health - Some Major Issues.* CIESM Workshop Monographs n°31, Monaco, pp. 83-86.
- Warnau, M., Iaccarino, M., De Biase, A., Temara, A., Jangoux, M., Dubois, P., Pagano, G.,
- 1996a. Spermiotoxicity and embryotoxicity of heavy metals in the echinoid *Paracentrotus*
- *lividus.* Environ. Toxicol. Chem. 15, 1931-1936.
- Warnau, M., Ledent, G., Temara, A., Alva, V., Jangoux, M., Dubois, P., 1995. Allometry of
- heavy metal bioconcentration in the echinoid *Paracentrotus lividus* (Echinodermata).
- Arch. Environ. Contam. Toxicol. 29, 393-399.
- Warnau, M., Teyssié, J.L., Fowler, S.W., 1996b. Biokinetics of selected heavy metals and
- radionuclides in the common Mediterranean echinoid *Paracentrotus lividus*: seawater and
- food exposure. Mar. Ecol. Prog. Ser. 141, 83-94.
- Zar, J.H., 1996. Biostatistical analysis. Upper Saddle River, New Jersey.
- Zingde, M.D., Singbal, S.Y.S., Moraes, C.F., Reddy, C.V.G., 1976). Arsenic, copper, zinc
- and manganese in the marine flora and fauna of coastal and estuarine waters around Goa.
- Ind. J. Mar. Sci. 5, 212-217.

Figure 1.

A- *Isognomon isognomon*

B- *Gafrarium tumidum*

B-1: Body compartment variation B-2: Geographical variation

Figure 2.

| Sampling stations | Ag | As | C _d | C ₀ | \mathbf{C} r | Cu | Mn | Ni | Zn | | | |
|--|-------------|-----------------------|----------------|----------------|----------------|----|--|----|----|--|--|--|
| Sediment | | | | | | | | | | | | |
| Ouano Beach ¹ | $\leq 0.1*$ | $2.9 \pm 1.2^{\circ}$ | $0.3*$ | | | | $0.8 \pm 0.4^{\circ}$ $7.2 \pm 2.7^{\circ}$ $0.5 \pm 1.0^{\circ}$ $41.7 \pm 15.7^{\circ}$ $5.1 \pm 3.2^{\circ}$ $3.3 \pm 1.9^{\circ}$ | | | | | |
| 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 | | | | | | | $\leq 0.1^*$ 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 | | | | | | | $\leq 0.1^*$ $9.9 \pm 0.7^{\circ}$ $0.5 \pm 0.01^{\circ}$ $6.2 \pm 1.1^{\circ}$ $38.6 \pm 2.7^{\circ}$ $1.1 \pm 0.3^{\circ}$ $81.6 \pm 3.5^{\circ}$ $82.0 \pm 5.1^{\circ}$ $9.4 \pm 0.6^{\circ}$ | | | | | |
| Boulari Bay ² | | | | | | | $\leq 0.1^*$ 46.9 ± 1.5° 5.1 ± 0.5° 61.2 ± 16.2° 662 ± 50 ^b 4.6 ± 1.6 ^b 565 ± 15° 900 ± 78° 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 | Co | 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 | $3,983 \pm 2,555$ | | |
| $\mathbf M$ | 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 | ${}_{\leq 1.0^{*}}$ | $1,356 \pm 876$ | | |
| ${\bf G}$ | 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 | $21.6 \pm 2.4^{\text{a}}$ | $1.23 \pm 0.40^{\text{ a}}$ | 0.69 ± 0.20 ^a | $9.0 \pm 1.6^{\circ}$ | $3.1 \pm 0.9^{\text{a}}$ | $20.4 \pm 8.3^{\text{a}}$ | $3.6 \pm 1.1^{\text{a}}$ | $3,832 \pm 1,529^{\mathrm{b}}$ | | |
| Maa Bay | | | | | | | | | | | | |
| VM | 51 ± 4 | 2.04 ± 1.81 | 64.3 ± 9.9 | $\overline{1.97} \pm 1.04$ | 0.58 ± 0.26 | 3.1 ± 0.8 | 10.6 ± 1.6 | 17.1 ± 6.4 | 2.2 ± 1.1 | $11,333 \pm 5,904$ | | |
| M | 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 | $< 0.2*$ | $5,781 \pm 3,299$ | | |
| ${\bf G}$ | 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 | | $1.47 \pm 1.09^{\text{ b}}$ | $76.6 \pm 9.3^{\circ}$ | $1.80 \pm 1.4^{\text{a}}$ | 0.45 ± 0.16 ^a | 3.5 ± 0.5 ^{ab} | 6.8 ± 0.5 ^{ab} | $22.3 \pm 14.6^{\text{a}}$ | $2.2 \pm 0.5^{\text{a}}$ | $13,817 \pm 6,621$ ^a | | |
| Grande Rade (GR _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 | $8,188 \pm 2,757$ | | |
| M | 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 | $0.6*$ | $1,958 \pm 443$ | | |
| ${\bf G}$ | 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 | $14,360 \pm 3,503$ | | |
| $\mathbf R$ | 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 | $0.67 \pm 0.09^{\text{ b}}$ | $2.7 \pm 0.3^{\circ}$ | 17.3 ± 5.3 ^c | $34.7 \pm 11.5^{\text{a}}$ | 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 | $1,741 \pm 2,175$ | | |
| M | 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 | | |
| $\mathsf G$ | 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 | | $16.5 \pm 4.0^{\text{a}}$ | 51.7 ± 10.8 ^d | 1.28 ± 0.68 ^a | $1.60 \pm 0.49^{\text{ a}}$ | $5.7 \pm 2.9^{\text{a}}$ | $9.8 \pm 2.1^{\text{a}}$ | $30.8 \pm 16.0^{\text{ a}}$ | $16.0 \pm 3.7^{\mathrm{b}}$ | $1,718 \pm 1,290^{\mathrm{b}}$ | | |

* 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 (%) | | | | | | | | | |
|-------------------------------|----------------------------------|------|----------------|------|------|------|------|------|------|--|
| | Ag | As | C _d | Co | 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 | |
| $Compartment \times location$ | 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 | |
| $Compartment \times location$ | 13.6 | 3.5 | 9.6 | 19.3 | 16.8 | 14.4 | 24.4 | 15.0 | 11.1 | |
| 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

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 | Co | Cr | Cu | Mn | Ni | Zn |
|---|---------------------------------|------|----------------|-------------------|-------------------|-------------------------|--------------------------|-------------------|------------------------|----------------------------------|
| Clams | | | | | | | | | | |
| Gafrarium 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 ± 0.09 - | 0.13 ± 0.14 - | 1.48 ± 0.28 - | 20.8 ± 1.49 - | 4.08 ± 0.21 - | 2.32 ± 0.35 - | $51 \pm 4 -$ |
| Circe sinensis 1 | | | | 2.25 ± 0.5 | 1.1 ± 0.17 | 1.93 ± 0.53 2.26 | 23.4 ± 1.43 3.13 | 4.55 ± 0.08 | 2.65 ± 0.46 2.8 | 73 ± 11 43.7 |
| Codakia orbicularis ⁴ | Hong-Kong | | | 3.8 | | | 3.08 | | 1.57 | |
| | Dominican Republic | | | | | 1.66 0.9 | 3.99 | | 4.66 | 22.9 98 |
| Ruditapes philippinarum ¹ Tellina fausta ⁴ | Hong-Kong 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 | | | 0.47 ± 0.23 - | | | < 150 11 ± 0.51 - | | | $900 - 2,000$ 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>I.</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 | | $2.3 - 6.3$ | | | | $251 - 728$ | $33.2 - 17.5$ | | $446 - 2,800$ |
| C. echinata 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.

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 Z