Ecotoxicology and Environmental Safety 72 (2009) 372–381

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

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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ARTICLE INFO

Article history:
Received 28 January 2007
Received in revised form
31 March 2008
Accepted 7 April 2008
Available online 29 May 2008

Keywords:
Tropical environment
Metals
Bioindicators
Mining activities

ABSTRACT

The concentrations of nine 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 oyster tissues (up to 441 $\mu g\,g^{-1}$ 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.

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 (e.g., 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 (e.g., 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 e.g., 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

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0147-6513/$ - see front matter
doi:10.1016/j.ecoenv.2008.04.004

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 for publication). 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 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. 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 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.

2.2. Organisms

The clam *G. tumidum* was collected by handpicking in the intertidal stations. The oyster *I. isognomon* is associated with rocky substrata at depth ranging from 2 to 25 m and were collected by SCUBA diving. All the organisms ($n = 6$ per species per station) were collected from October to November 2004 (mean water temperature: $25.6 \pm 0.8\,^{\circ}C$) 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 (e.g., 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, 2003) were selected for analysis. Collected clams (mean ± S.D., $n = 18$) measured 38.1 ± 2.8 mm (width) and weighed 22.7 ± 4.9 g; oysters (mean ± S.D., $n = 24$) measured 98 ± 13 mm (length) and weighed 36.9 ± 13.2 g. Back to the laboratory, the bivalves were kept for 24 h 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.

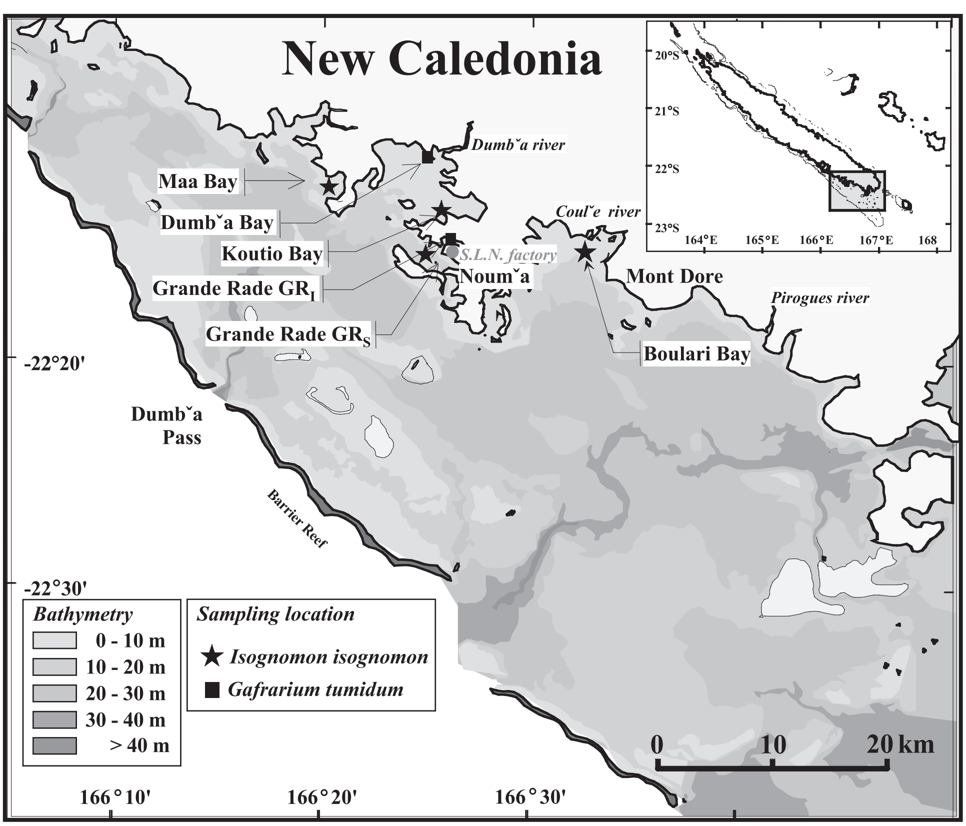

Fig. 1. Map of the sampling sites along the SW coast of New Caledonia (Ouano Beach is not represented on the map).

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.

2.4. Sample preparation and analysis

Aliquots of the biological samples (0.1–0.5 g) and of the dried sediment samples (0.5 g) were digested using a 3:1 (v/v) 65% HNO_3–30% HCl mixture (Merck, suprapur quality). Acidic digestion of the samples was carried out overnight at room temperature, then using a 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 (e.g., Coteur et al., 2003; Dalto et al., 2006). Each sample was eventually diluted to 30–50 ml with milli-Q 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) 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) and 106 (Zn). The detection limits ($\mu g\, g^{-1}$ 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 concentrations are given on a dry weight basis ($\mu g\, g^{-1}$ dry wt).

2.5. Statistical analysis

Comparisons of the data were performed using one- or two-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 (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 element concentrations among stations. Boulari Bay and Grande Rade (GR_I) displayed the highest concentrations for all elements. Concentrations in all elements were always significantly higher in Grande Rade (GR_I) than in Ouano Beach ($p_{Tukey} < 0.0001$). Concentrations in sediment from Boulari Bay were significantly higher (up to one 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 16–70% 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 14–44% 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 the body compartments of *I. isognomon* (two-way ANOVA, $p_{sampling\ locations} < 0.0001$), except for 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 in Maa Bay ($p_{Tukey} \leqslant 0.001$; Table 2; Fig. 2A). Oysters from Grande Rade (GR_S) displayed significantly higher Ag and Cu concentrations than those measured in Maa Bay ($p_{Tukey} \leqslant 0.01$) whereas the highest Cr concentrations were measured in oysters from Koutio Bay ($p_{Tukey} \leqslant 0.05$). 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 one-way ANOVA and Tukey test. Results were similar to

Table 1
Element concentrations in sediment (mean ± SD; $\mu g\, g^{-1}$ dry wt, $n = 3$)

Sampling stations	Ag	As	Cd	Co	Cr	Cu	Mn	Ni	Zn
Sediment									
Ouano Beach[a]	<0.1[c]	2.9±1.2a	<0.3[c]	0.8±0.4a	7.2±2.7a	0.5±1.0a	41.7±15.7a	5.1±3.2a	3.3±1.9a
Grande Rade GR_I[a]	0.4±0.1	8.5±1.6b	2.4±0.3	46.2±9.1b	292±56b	26.9±8.8b	288±40b	797±149b	141±18b
Maa Bay[b]	<0.1[c]	6.4±0.3a	1.0±0.2b	4.6±2.3a, b	44.1±7.9a	10.7±3.5b	132±7.8b	64.2±13.5a	15.2±3.1a
Koutio Bay[b]	<0.1[c]	9.9±0.7b	0.5±0.01a	6.2±1.1b	38.6±2.7a	1.1±0.3a	81.6±3.5a	82.0±5.1a	9.4±0.6a
Boulari Bay[b]	<0.1[c]	46.9±1.5c	5.1±0.5c	61.2±16.2c	662±50b	4.6±1.6b	565±15c	900±78c	33.1±3.4b

Significant differences among the mean concentrations in sediments from the different sampling stations are indicated by letters (a, b, c); means sharing the same letters 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.

[a] Stations where clams were collected.
[b] Stations where oysters were collected.
[c] Concentrations <detection limit.

Table 2
Element concentrations in the oyster *Isognomon isognomon* (mean±SD; μg g^{-1} dry wt, $n=6$)

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	3983±2555
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	<1.0[a]	1356±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±5953
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	6346±4224
WSP		14.5±7.1a	21.6±2.4a	1.23±0.40a	0.69±0.20a	9.0±1.6c	3.1±0.9a	20.4±8.3a	3.6±1.1a	3832±1529b
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±5904
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[a]	5781±3299
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±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±8103
WSP		1.47±1.09b	76.6±9.3b	1.80±1.4a	0.45±0.16a	3.5±0.5a, b	6.8±0.5a, b	22.3±14.6a	2.2±0.5a	13,817±6621a
Grande Rade (GRS)										
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	8188±2757
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[a]	1958±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±3503
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±3724
WSP		32.8±6.5c	38.2±4.3c	1.18±0.44a	0.67±0.09b	2.7±0.3b	17.3±5.3c	34.7±11.5a	4.4±0.8a	7873±2087a
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	1741±2175
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
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	4437±1880
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	2017±1427
WSP		16.5±4.0a	51.7±10.8d	1.28±0.68a	1.60±0.49a	5.7±2.9a	9.8±2.1a	30.8±16.0a	16.0±3.7b	1718±1290b

Body compartments: VM (visceral mass), M (adductor muscle), G (gills), R (remaining soft parts), WSP (whole soft parts; reconstructed values). Differences among the concentrations in WSP from the four locations are indicated by letters (a, b, c); means sharing the same letters are not significantly different among sampling stations ($p_{\text{Tukey}}>0.05$).
[a] Concentration <detection limit.

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

Factors	Explained variability (%)								
	Ag	As	Cd	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 × 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	11.1
Residual	41.1	9.2	47.1	40.7	35.3	9.8	43.9	35.0	60.6

those from the two-way ANOVA performed on body-compartment specific concentrations, except for Cd and Zn. For these two latter elements, no significant difference was observed 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 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 two-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%).

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 23–84% 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, 35–61% 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 (e.g., nature of exploited soils in different areas) or indirectly (e.g., 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 (two-way ANOVA, $p_{\text{sampling locations}}$ always $\leqslant 0.002$) (Tables 3 and 4; Fig. 2B).

A

A-1

Metal	Compartment ranking			
Ag	G	VM	R	M
As	M	G	VM	R
Cd	R	G	VM	M
Co	VM	G	R	M
Cr	VM	G	R	M
Cu	VM	G	R	M
Mn	R	VM	G	M
Ni	G	VM	R	M
Zn	G	R	VM	M

A-2

Metal	Location ranking			
Ag	GR_S	KOU	BOU	MAA
As	MAA	BOU	GR_S	KOU
Cd	MAA	KOU	BOU	GR_S
Co	BOU	GR_S	KOU	MAA
Cr	KOU	BOU	MAA	GR_S
Cu	GR_S	BOU	MAA	KOU
Mn	GR_S	BOU	MAA	KOU
Ni	BOU	GR_S	KOU	MAA
Zn	MAA	GR_S	KOU	BOU

B

B-1

Metal	Compartment ranking		
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

Metal	Location ranking		
Ag	GR_I	DUM	OUA
As	OUA	GR_I	DUM
Cd	GR_I	OUA	DUM
Co	DUM	GR_I	OUA
Cr	GR_I	DUM	OUA
Cu	GR_I	DUM	OUA
Mn	GR_I	DUM	OUA
Ni	GR_I	DUM	OUA
Zn	GR_I	DUM	OUA

Fig. 2. Comparisons of element concentrations in bivalves, using multiple comparison test of Tukey performed after two-way ANOVA in (A) the oyster *I. isognomon* and (B) the clam *Gafrarium tumidum*. Mean concentrations are ranked from the left to the right by decreasing order. Concentrations in underlined body compartments or locations are not significantly different ($\alpha = 0.05$). Body compartments: DG (digestive gland), G (gills), M (adductor muscle), VM (visceral mass) and R (remaining soft parts). Sampling locations: OUA (Ouano beach), GR_S (Grande Rade, subtidal), GR_I (Grande Rade, intertidal), DUM (Dumbéa Bay), and KOU (Koutio Bay).

Table 4
Element concentrations in the clam *Gafrarium tumidum* (mean ± SD; $\mu g\,g^{-1}$ dry wt, $n = 6$)

Compartments	% Weight	Ag	As	Cd	Co	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±1.1a	37.4±7.4a	0.17±0.03a	3.8±0.7a	4.8±1.3a	7.9±1.3a	35.9±43.5a	30.2±6.0a	62.7±10.2a
Ouano Beach										
DG	14±3	<1.4[a]	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[a]	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[a]	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] a	441±84b	0.19±0.04a	1.1±0.2b	3.2±2.2a	5.6±1.0a	5.5±1.5a	8.1±1.5b	55.6±7.8a
Grande Rade (GR_I)										
DG	29±6	51.5±33.6	67.9±14.6	1.30±0.88	2.4±1.4	10.7±.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±13.4b	55.0±15.1a	0.74±0.25b	2.2±1.0c	8.0±1.7b	77.3±17.5b	139±104b	52.3±11.9c	154±102b

Differences among the concentrations in WSP from the four locations are indicated by letters (a, b, c); means sharing the same letters are not significantly different among sampling stations ($p_{Tukey} > 0.05$). Body compartments: DG (digestive gland), G (gills), R (remaining soft parts), WSP (whole soft parts; reconstructed values).

[a] Concentration < detection limit.

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 ($p_{Tukey} \leqslant 0.001$; Table 4 and Fig. 2B).

Geographical variations were tested using one-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 two-way ANOVA performed on body-compartment 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 $\mu g\, g^{-1}$ dry wt.

3.3.2. Body distribution

The mean concentrations of all elements investigated differed according to the body compartments (two-way ANOVA, $p_{body\ compartment}$ always $\leqslant 0.003$). Multiple comparison tests of Tukey indicated that the concentrations of Cd, Cu, Cr, Mn and Zn were significantly higher in 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 (55–77%) 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 (e.g., Salomons et al., 1987) and their element concentrations are often used to assess and monitor the contamination status of the 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 *I. isognomon* and the clam *G. 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, 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 inputs (e.g., Francesconi et al., 1999; Warnau et al., 2007) and Grande Rade (GR_S) to important domestic wastewater discharges (e.g., 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 (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 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 (e.g., 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 in the following para.

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 ($52 \pm 12\, \mu g\, g^{-1}$ 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).

Although scarcely available, As concentrations reported in the literature for tropical and subtropical bivalves are generally lower than 30 $\mu g\, g^{-1}$ dry wt ($< 10\, \mu g\, g^{-1}$ dry wt if one 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 wt) (Valette-Silver et al., 1999) and in the clam *Circentia callipyga* from the Gulf of Oman (156 $\mu g\, g^{-1}$ dry wt) (de Mora et al., 2004). In the present study, As was found to reach extremely high concentrations in the clams from Ouano Beach ($441 \pm 84\, \mu g\, g^{-1}$ dry wt) compared to those observed in Grande Rade (GR_I) ($55 \pm 15\, \mu g\, g^{-1}$ dry wt) and in the oysters from Maa Bay ($77 \pm 9\, \mu g\, g^{-1}$ dry wt). To the best of our knowledge, such high body concentrations of As have never been reported in other clams. The Ouano Beach values were 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 concentrations of total As (2000 $\mu g\, g^{-1}$ dry wt; Gibbs et al., 1983), the Mediterranean fan worm *Sabella spallanzanii* which shows As concentrations higher than 1000 $\mu g\, g^{-1}$ dry wt in its branchial crown (Fattorini and Regoli, 2004) or

Table 5
Element concentrations (mean ± SD or range; $\mu g\,g^{-1}$ dry wt) in clams and oysters from tropical and subtropical areas

Species	Location	Ag	As	Cd	Co	Cr	Cu	Mn	Ni	Zn
Clams										
Gafrarium tumidum[a]	Hong-Kong					0.67	5.77		5.59	57.5
G. tumidum[b]	Fiji				1.0–2.8	1.0–1.6	4.2–11.0	28–45	1.7–4.5	
Anadara antiquate[b]	Fiji				0.9–2.5	0.8–1.8	4–13	32–50	2–4	
Chione subrugosa[c]	Tropical mangrove lagoon			0.72 ± 0.09–2.25 ± 0.5	0.13 ± 0.14–1.1 ± 0.17	1.48 ± 0.28–1.93 ± 0.53	20.8 ± 1.49–23.4 ± 1.43	4.08 ± 0.21–4.55 ± 0.08	2.32 ± 0.35–2.65 ± 0.46	51 ± 4–73 ± 11
Circe sinensis[a]	Hong-Kong					2.26	3.13		2.8	43.7
Codakia orbicularis[d]	Dominican Republic			3.8		1.66	3.08		1.57	22.9
Ruditapes philippinarum[a]	Hong-Kong					0.9	3.99		4.66	98
Tellina fausta[d]	Dominican Republic			0.04		4.15	14.1		4.91	51.4
Circentia callipyga[e]	Qatar	3.03	156	1.17	4.45	0.97	8.35	17.7	23.9	69.1
Oysters										
Isognomon isognomon [f]	Phuket, Thailand						< 150			900–2000
I. alatus[g]	Malaysia			0.47 ± 0.23–3.71 ± 0.12			11 ± 0.51–30.7 ± 0.8			23.8 ± 0.75–334.5 ± 12.5
I. alatus[h]	Venezuela			0.33–0.91		0.46–1.2	14–49.1		11–18	0.25–2.1
I. alatus[i]	Colombian Caribbean			0.8–15.6			0.42–52.3			
I. alatus[d]	Dominican Republic			0.24–0.26		2.38–4.96	7.58–19.7		1.25–2.90	4000–4010
I. alatus[j]	Guadeloupe					0.23–7.2	6.8–127			1060–12,160
I. alatus[j]	Martinique					0.32–1.75	5.4–248			2460–11,530
I. bicolor[i]	Colombian Caribbean			0.98–6.99			0.8–3.94			
I. legumen[k]	Taiwan						491 ± 29			
Isognomon sp.[l]	Biscayne Bay, Florida		37.3 ± 6.9							
Ostrea sandvicensis[m]	Hawaii						1400		20	
Saccostrea amasa[n]	North Queensland, Australia			1–12						673–20,906
S. echinata[o]	North Queensland, Australia			0.69–2.34						2080 ± 453
S. echinata[p]	North Queensland, Australia			0.198–4.63						325–4680
Crassostrea belcheri[q]	Merbok estuary, Malaysia						1–8.5			30–550
C. cucullata[r]	Goa, India		2.3–6.3				251–728	33.2–17.5		446–2800
C. echinata[o]	Cleveland bay, Australia									673–20,906
C. gryphoides[r]	Goa, India		3.2–5.8				175–210			325–550
C. iredalei[q]	Merbok estuary, Malaysia						4–8			80–550
C. gigas[19]	Derwent Estuary, Tasmania									38,700
C. virginica[k]	Taiwan						257 ± 196			1037 ± 778

[a] Cheung and Wong (1997).
[b] Dougherty (1988).
[c] Szefer et al. (1998).
[d] Sbriz et al. (1998).
[e] de Mora et al. (2004).
[f] Brown and Holley (1982).
[g] Saed et al. (2001).
[h] Jaffe et al. (1998).
[i] Campos (1988).
[j] RNO-Antilles (unpublished work).
[k] Hung et al. (2001).
[l] Valette-Silver et al. (1999).
[m] O'Connor (1989).
[n] Jones (1992).
[o] Jones et al. (2000).
[p] Olivier et al. (2002).
[q] Lim et al. (1995).
[r] Zingde et al. (1976).

the very high arsenic concentrations monitored in muscles of edible fish (500 $\mu g\,g^{-1}$ dry wt) from the Bay of Cienfuegos, Cuba, a few weeks after an accidental release of arsenate oxides from a local nitrogen fertilizer factory in December 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 (e.g., 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; Warnau et al., 2007; Gómez-Batista and Warnau, unpublished 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 the area of Maa Bay, Ouano Beach is rather more directly 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; Gómez-Batista and Warnau, unpublished results). 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 e.g., Kaise and Fukui, 1992; Warnau et al., 2007).

I. isognomon also displayed very high Zn concentrations in Maa Bay and in Grande Rade (GR_S), viz., 13,817 $\pm$ 6621 and 7873 $\pm$ 2087 $\mu g\,g^{-1}$ dry wt, respectively. Elevated concentrations of Zn have been reported for *I. alatus*, reaching 4010 μg Zn g^{-1} dry wt in individuals collected in the Dominican Republic and 12,163 $\mu g\,g^{-1}$ 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 (e.g., 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 immobilisation of Zn under non-toxic forms in granules which are very slowly excreted (e.g., 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 (e.g., 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 concentrations (33 $\pm$ 13 and 33 $\pm$ 7 $\mu g\,g^{-1}$ 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 areas (< 1 $\mu g\,g^{-1}$ dry wt in mussels; Klumpp and Burdon-Jones, 1982) and temperate areas (< 6 $\mu g\,g^{-1}$ 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 Ag_2S, a stable and non-toxic compound (e.g., Berthet et al., 1992; Warnau et al., 1996b; 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; Sañudo-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).

Acknowledgments

Authors thank M. Robert (CCA, Université de La Rochelle) for his analytical advices. LH was beneficiary of a CIFRE scholarship (ANRT, France) supported by the Goro-Nickel Company, New Caledonia. M. W. is an Honorary Senior Research Associate of the National Fund for Scientific Research (NFSR, Belgium), and holds a 2008 Invited Expert position at LIENSs (Université de la Rochelle), supported by the Conseil Régional de Poitou-Charentes. 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.

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