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Rock-Shells (*Thais clavigera*) as an Indicator of As, Cu, and Zn Contamination on the Putai Coast of the Black-Foot Disease Area in Taiwan

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Abstract. This study presents the distribution of arsenic (As), copper (Cu), and zinc (Zn) in various seafoods—oysters (*Crassostrea gigas*), false fusus (*Hemifuscus tuba*), venus clams (*Cyclina sineasis*), green mussels (*Perna viridis*), blood clams (*Arca granosa*), flounders (*Psettodes erumei*), and rock-shells (*Thais clavigera*) collected from the Putai coast of the black-foot disease (BFD) area in Taiwan. Special attention is paid to evaluate the relationships among As, Cu, and Zn and effect of body size on metal concentration in *Thais clavigera.* Maximum Zn and Cu geometric mean (GM) concentrations (GM = 615 and 376 µg/g, dry wt, respectively) are found in oysters (*Crassostrea gigas*), and the values are much higher than those of the other organisms by about 1.7–208 and 1.8–375 times, respectively. Similarly, *Thais clavigera* has a high capacity for accumulating Cu and Zn collected from the same location. One interesting point is that relatively high As concentrations (GM = 65.7 µg/g, dry wt) in *Thais clavigera* are found as compared with those in other organisms (range from GM = 2.37 to 40.2 µg/g, dry wt). The As concentrations are significantly higher in *Thais clavigera* (1.62–27.7 times) than those in other organisms ($p < 0.05$), except for the false fusus (*Hamifuscus tuba*). A linear regression analysis shows a significant increase in Zn concentration with increasing Cu concentration in *Thais clavigera.* On the other hand, the As concentration is correlated with Cu and Zn concentrations ($r = 0.77$ and 0.77, respectively; $p < 0.05$) in *Thais clavigera.* Double logarithmic plots of metal content and concentration against dry-body weight and shell length show linear relationships. The result indicates that large individuals have higher contents of Cu, Zn and As, and have slopes of 1.58, 1.38, and 1.34, respectively. In addition, metal concentrations against shell length for all animal sizes also indicate that Cu, Zn and As have slopes of 1.92, 1.18, and 1.11, respectively. In conclusion, *Thais clavigera* has a high capactiy for accumulating As, Cu, and Zn and is a potential bioindicator for monitoring As, Cu and Zn.

Due to dense population and rapid industrial expansion, the estuarine and coastal environments of Taiwan are suffering an ever-increasing impact from human activities (Hung 1988; Han and Hung 1990; Han *et al.* 1994; Jeng and Han 1994).

We have monitored heavy metals in the coastal environment of Taiwan and found that heavy metal pollution is a serious problem and one of the most studied marine environmental problems in Taiwan. Most current health risks associated with seafood safety originate in the environment. For example, Han *et al.* (1994) reported the Cu intake and health threat by consuming seafood from Cu-contaminated coastal environments in Taiwan. The estimate indicates that the average Cu intake from the ‘‘green oyster’’ for female individuals is 14 times more than that of international limits.

Arsenic is a ubiquitous element widely distributed in the environment. It is transported mainly in the environment by water. Humans are exposed to inorganic and organic As through environmental, medicinal, and occupational exposures. Both inorganic and organic As are present in food in different amounts. For example, seafood contains a high concentration of organic As. However, organic As is much less toxic than inorganic As (Chiou *et al.* 1995).

Arsenic has been well documented as one of the major risk factors for black-foot disease (BFD) (Smith *et al.* 1992), a unique peripheral vascular disease identified in the endemic area of arseniasis located on the southwestern coast of Taiwan where residents had used high-As artesian well water for more than 50 years (Tseng 1968; Chen *et al.* 1980). In a series of studies in Taiwan, an increasing mortality from cancers of the lung, liver, and bladder has been documented among residents in the endemic area of BFD (Chen *et al.* 1985, 1986, 1988, 1992). A significant dose-response relationship between the As concentration in well water and the mortality from various cancers has also been reported (Wu *et al.* 1989).

Chen *et al.* (1994) reported that the total dissolved As concentration is 671 ± 149 µg/L with a range of 470–897 µg/L for all the well waters (a total of 54 samples) collected from the Putai area. The results are about 13 times greater than the maximum contaminant level (MCL) for As in drinking water. In addition, the predominant As species in the well waters of the BFD area is As^{3+} with an average As^{3+}/As^{5+} ratio of 2.6. At the same time, As concentrations in the well waters of Hsinchu, a city in the northwest of Taiwan, are less than 0.7 µg/L (Chen *et al.* 1994). Furthermore, As species in well waters and sediments were analyzed by a sequential leaching technique (Liaw 1995). The results indicate that the sum of phases I (exchangeable), II

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Table 1. Determination of Cu, Zn, and As in standard reference materials (SRM) by FAAS and GAAS

SRM	Metal	Method	Value Obtained in our Lab (μg/g)	Certified Value (μg/g)	Precision (%)	Accuracy (%)
Oyster (SRM1566a)	Cu	FAAS[a]	60.3 ± 3.9 (N = 7)	66.3 ± 4.3	6.47	−9.05
	Zn	FAAS[a]	850 ± 58 (N = 7)	830 ± 57	6.81	+2.41
	As	GAAS[b]	14.3 ± 2.1 (N = 8)	14.0 ± 1.2	14.4	+2.29
Tuna fish (IAEA-350)	Cu	FAAS[a]	2.70 ± 0.17 (N = 5)	2.83 (2.55–3.10)	6.44	−4.59
	Zn	FAAS[a]	19.6 ± 3.0 (N = 5)	17.4 (16.6–18.5)	15.2	+12.6
	As	GAAS[b]	5.04 ± 0.35 (N = 6)	5.28 (3.36–5.75)	7.03	−4.48

[a] FAAS, Flame atomic absorption spectrophotometry
[b] GAAS, Graphite atomic absorption spectrophotometry

(bound to carbonates) and III (iron and manganese oxides) accounted for 96.3% of total particulate As (1,641 μg/L) in well waters. The high mobility and bioavailability of As may be easily uptake for humans.

However, there is no information on the concentrations of As in different seafoods and bioaccumulation by the marine organisms from the coastal water of the BFD area in Taiwan. This information is of fundamental importance for understanding the cause of BFD and the potential hazards of As in the aquatic environment. In spite of the fact that aquatic organisms may bioconcentrate different metals in their tissues, both from water and sediment, as well as through the food chain, organisms are used as an aid in the monitoring of aquatic pollutants (Phillips 1980). In Taiwan, reliable correlations between the concentrations of various Cu species in the seawater and oysters (*Crassostrea gigas*) may prove useful for monitoring purposes (Han *et al.* 1990).

Rock-shells (*Thais clavigera*) are widely distributed throughout the Putai and Lukang areas of Taiwan, and *Thais clavigera* is the commonest intertidal muricids found on the Putai and Lukang rocky shores. Lin and Hsu (1979) reported that this carnivorous oyster drill (*Thais clavigera*) is one of the pests that prey on the oyster (*Crassostrea gigas*) in Taiwan, particularly in areas where stick or rack culture systems are practised. In addition, *Thais clavigera* may be particularly susceptible to metal contamination (such as Cu, Zn, and As) and may act as a good biological indicator of metal contamination in the marine environment (Han *et al.* 1996).

This study presents the distribution of As, Cu, and Zn in various seafoods and rock-shells (*Thais clavigera*) collected from the Putai coast of the BFD area in Taiwan. The purpose of this work is to assess *Thais clavigera* as a potential candidate for monitoring As in the marine environment. Special attention is paid to evaluate the relationships among As, Cu, and Zn and the effect of body size on metal concentration in rock-shells (*Thais clavigera*) from the Putai coast of the BFD area in Taiwan. The inter-relationships between As, Cu and Zn levels in the same organism may provide information on the similar metabolic pathways from metals (Cu and Zn) and metalloid (As).

Materials and Methods

Biological samples were collected from the Putai coast of the BFD endemic district, Taiwan. Six species of shellfish and one species of fish including oysters (*Crassostreagigas*), false fusus (*Hemifuscus tuba*), rock-shells (*Thais clavigera*), venus clams (*Cyclina sineasis*), green mussels (*Perna viridis*), blood clams (*Arca granosa*), and flounders (*Psettodes erumei*) were sampled from the same coastal area of Putai during the period from January 1995 to July 1995. Oysters, venus clams, green mussels and blood clams are filter feeders; false fusus are infaunal deposit feeders; and rock-shells and flounders are carnivorous feeders.

Field biological samples were returned to the laboratory, individually scrubbed, shucked, and the flesh placed in tared acid-washed Teflon beakers for reweighing to obtain individual wet weight values. All biological samples were digested with a microwave digester (Model MDS-2000) with a mixture of nitric and sulfuric acid (1/1, v/v) solution, and the supernatant fractions were analyzed for Cu and Zn by flame atomic absorption spectrophotometry (Han and Hung 1990). Arsenic was measured by graphite atomic absorption spectrometry (GAAS), using nickel as a matrix modifier, and a standard addition procedure was used for the calculation of the analyte concentrations. The detection limit was 0.06 μg/g when using a sample size of 0.25 g dry material.

Determination of metals was performed with a Hitachi Zeeman AAS (model Z-8000 with an autosampler). The results generated were, in most cases, in good agreement with certified values. Standard reference materials (SRM 1566a Oyster Tissue and IAEA 350 Tuna Fish Tissue) were analyzed at regular intervals.

For statistical analyses, left skewed data were normalized by logarithmic transformation. Accordingly, geometric mean (GM) values were reported. Analysis of variance (ANOVA) and Scheffe's test were used to study differences of various metal concentrations between different organisms. In addition, various linear regressions were used to test the possible correlation between metal concentrations and contents and the body size of the rock-shells (*Thais clavigera*).

Results and Discussion

Quality Assurance of As, Cu and Zn in Various Organisms

Replicate analysis of standard reference materials (Oyster tissue SRM 1566a; Tuna Fish IAEA-350) from the U.S. National Institute of Standards and Technology, and International Atomic Energy Agency showed good accuracy (<13%) with all results comparable with certified values. For example, 850 ± 58 μg/g Zn was found as compared to the SRM 1566a for oyster reference value of 830 ± 57 μg/g, with good precision and accuracy of 6.81 and +2.41%, respectively. Precision and accuracy of various metals from the two biological reference materials are reported in Table 1.

Table 2. Geometric mean metal concentrations (μg/g, dry wt) in various organisms collected from the Putai coast of the BFD area in Taiwan

Organism	No. of samples	Metals in organisms		
		Cu	Zn	As
Oysters (*Crassostrea gigas*)	20	376 (217–582)	615 (417–823)	18.7 (12.3–21.4)
False fusus (*Hemifuscus tuba*)	10	12.8 (8.17–22.7)	76.5 (49.4–131)	40.2 (29.8–68.3)
Rock-shells (*Thais clavigera*)	14	213 (142–370)	359 (238–564)	65.7 (47.6–106)
Venus clams (*Cyclina sineasis*)	16	6.27 (4.07–15.2)	48.0 (21.2–66.7)	5.37 (2.71–8.45)
Green mussels (*Perna viridis*)	8	3.69 (1.78–5.41)	19.8 (14.4–25.7)	3.42 (1.17–4.87)
Blood clams (*Arca granosa*)	16	1.17 (0.99–3.71)	19.1 (12.7–24.3)	2.37 (0.98–4.47)
Flounders (*Psettodes erumei*)	16	0.05 (0.03–0.14)	2.96 (1.17–4.43)	14.7 (7.88–19.75)

( ) = two extreme values

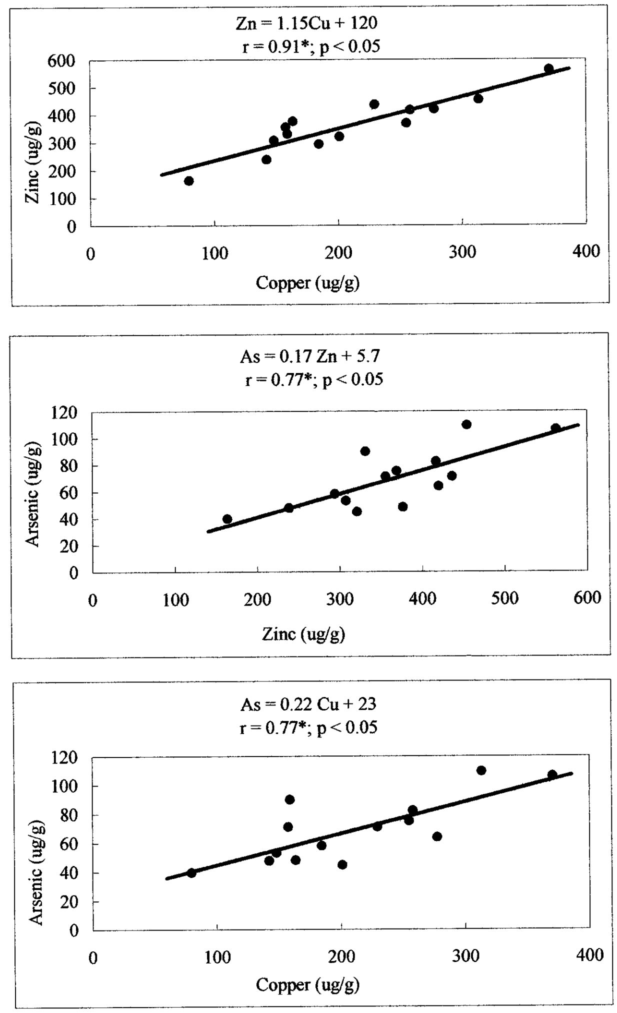

Fig. 1. Plots of the correlation between metal concentration pairs in rock-shells (*Thais clavigera*) from the Putai coastal area of Taiwan

Metal Concentrations in Various Organisms

Table 2 shows the geometric mean (GM) concentrations of As, Cu, and Zn in various organisms (soft) collected for this study. For example, maximum Zn and Cu concentrations (GM = 615 and 376 μg/g, dry wt, respectively) in oysters (*Crassostrea gigas*) are much higher than those of the other organisms by about 1.7–208 and 1.8–375 times, respectively. At the same time, it can be seen that the range of Cu and Zn concentrations in the oysters and rock-shells differ significantly ($p < 0.05$) from the other organisms. This suggests that the ability of oysters and rock-shells to concentrate Cu and Zn are much stronger than that of other species although they do grow under the same physico-chemical conditions. Indicator organisms should be good accumulators of metals, and their tissue concentrations must reflect differences in metal bioavailability. For this reason, organisms having an ability to regulate metals are clearly unsuitable (Bryan *et al.* 1985). The present results show that false fusus, venus clams, green mussels, blood clams and flounders are able to regulate (*i.e.,* low efficiency in accumulation) their internal Cu and Zn concentrations. Conversely, *Thais clavigera* has a high capacity for accumulating Cu and Zn under the sample physico-chemical conditions. This capacity makes *Thais clavigera* a potential candidate for monitoring Cu and Zn in coastal environments.

Han and Hung (1990) report that Cu concentrations in oysters are significantly correlated ($p < 0.05$) with the concentration of particulate Cu. In other words, the food pathway from surrounding water may predominate in accumulating Cu by oysters. However, the relative importance of these process is quantitatively uncertain in rock-shells. Based on laboratory and field experiments, oysters appear to be versatile indicators for a wide range of metals although As is a notable exception (Bryan *et al.* 1985). Table 2 also shows As concentrations in various organisms collected from the same coastal area. The results reveal that relatively high As concentrations (GM = 65.7 μg/g, dry wt) in *Thais clavigera* are found as compared with those in other organisms (range from GM = 2.37 to 40.2 μg/g, dry wt). The data also show that As concentrations in *Thais clavigera* are 1.62–27.7 times higher than those in other organisms ($p < 0.05$), except for the false fusus. The generally high As concentrations in *Thais clavigera* indicate that local and regional inputs of As are the major cause of BFD in this area.

On the whole, relatively low As, Cu, and Zn concentrations are obtained for venus clams, green mussels, blood clams and flounders (Table 2). This may be due to the fact that they are able to regulate their internal concentrations. As, Zn, and Cu concentrations are not affected by these metal concentrations in sediment and seawater. Any reasonable monitoring program should involve analyses of different species such as a suspension feeder and a deposit feeder to try and assess contamination in different forms (Bryan *et al.* 1985). Therefore, the analysis of carnivorous species (*Thais clavigera*) is most likely to reveal examples of food-chain biomagnification of As.

In order to investigate the role of various biological processes in the metal dynamics of *Thais clavigera*, the correlations between Cu, Zn, and As concentrations are calculated and illustrated (Figure 1). A linear regression analysis shows a significant increase in Zn concentration with increasing Cu concentration in *Thais clavigera*. The results are in agreement with those of Hung *et al.* (1982) and Han *et al.* (1990), who

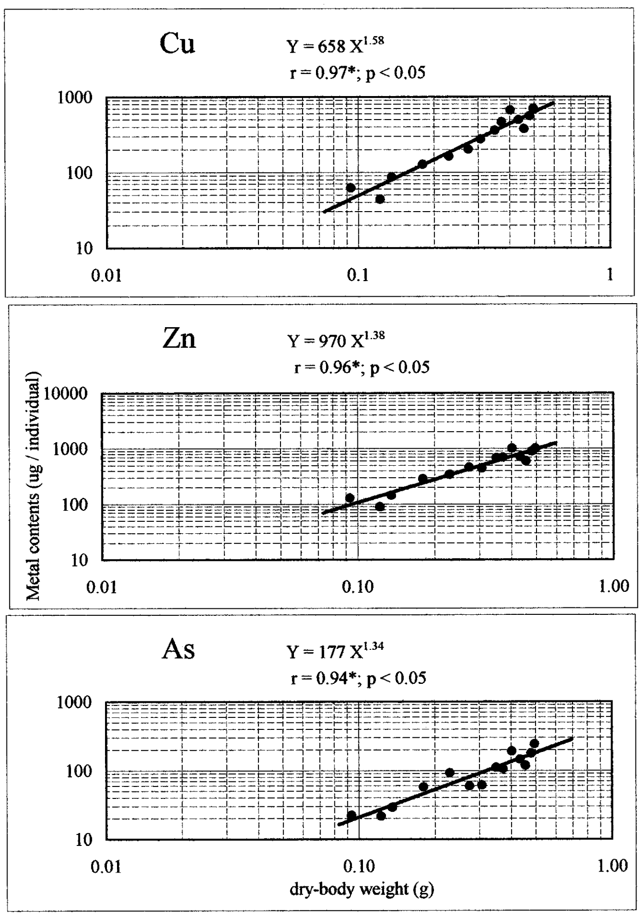

Fig. 2. Relationship between metal contents (µg/individual) and dry-body weight (g) of rock-shells (*Thais clavigera*) from the Putai coastal area of Taiwan

showed that Cu is significantly correlated with Zn in oysters. For instance, we obtain Zn = 1.15 Cu + 120, with a linear correlation coefficient of 0.91 ($p < 0.05$), indicating a significant correlation (Figure 1). It has been suggested that Cu and Zn are selectively localized in the tissue of *Thais clavigera,* for Zn the highest concentration being reached in hepatopancreas and gills and for Cu in the liver and haemocyanin in blood (Ahsanullah *et al.* 1981). Such selectivity could explain the pattern of Cu and Zn accumulation in the present study although specific tissues were not analyzed.

Figure 1 also shows plots of As vs. Zn and Cu in *Thais clavigera.* The As concentration is correlated with Cu and Zn concentrations ($r = 0.77$ and 0.77, respectively; $p < 0.05$) in *Thais clavigera.* Figure 1 further shows that As increases linearly with Cu and Zn levels, across all levels of Cu and Zn, implying that *Thais clavigera* can regulate As more than Cu and Zn. It may suggest that Cu and Zn play a different role in the *Thais clavigera* metabolism than in that of As because Cu and Zn are essential trace metals for animal metabolism.

The correlation results indicate that the Cu, Zn, and As concentrations in *Thais clavigera* vary with metal selectivity and bioavailability. These correlations may be attributable to similar physico/chemical properties of the metals involved (Szefer *et al.* 1994), and may be indicative of similar biochemical pathways (Mason and Simkiss 1983). Using this specific

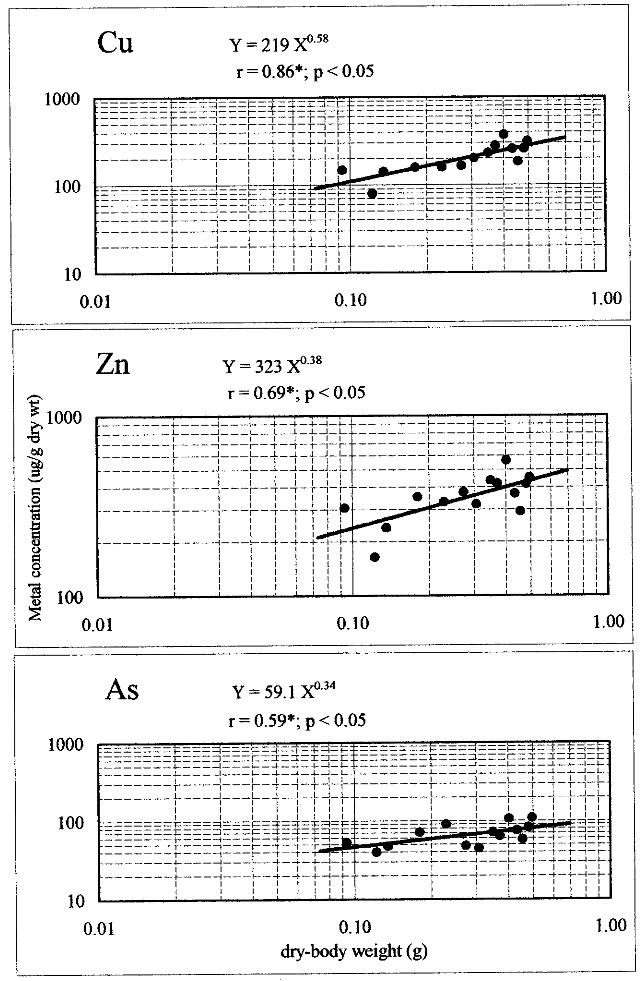

Fig. 3. Relationship between metal concentrations (µg/g dry wt) and dry-body weight (g) of rock-shells (*Thais clavigera*) from the Putai coastal area of Taiwan

character of *Thais clavigera* for accumulating Cu, Zn and As provides a simple and easy method of monitoring the environmental conditions.

Regression Between Metal Concentrations and Size

Boyden (1974, 1977) suggests that plotting metal content or concentration against body size on double logarithmic scales generally produces a linear relationship that can be easily defined by an equation according to:

$$Y = aX^b \qquad \text{hence} \quad \log Y = \log a + b \log X,$$

where Y represents the metal content (µg/individual), X represents the body weight of shellfish, log a is the intercept, and b is the slope. For example, Figure 2 shows the linear relationship between metal contents and dry body weight of *Thais clavigera.* Generally, the contents of Cu, Zn, and As are found to significantly increase with the body weight increase. The correlation coefficients and slopes of Cu, Zn, and As are 0.97, 0.96, and 0.94, and 1.58, 1.38, and 1.34, respectively. Generally, when $b = 1$, a linear relationship between the content of metals and the body weight of organisms is observed. When

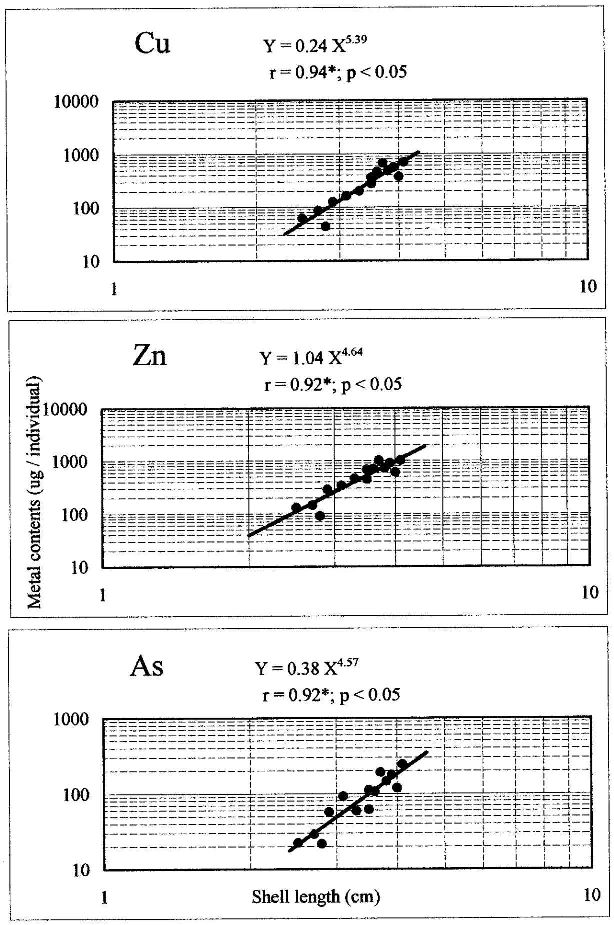

Fig. 4. Relationship between metal contents (μg/individual) and shell length (cm) of rock-shells (*Thais clavigera*) from the Putai coastal area of Taiwan

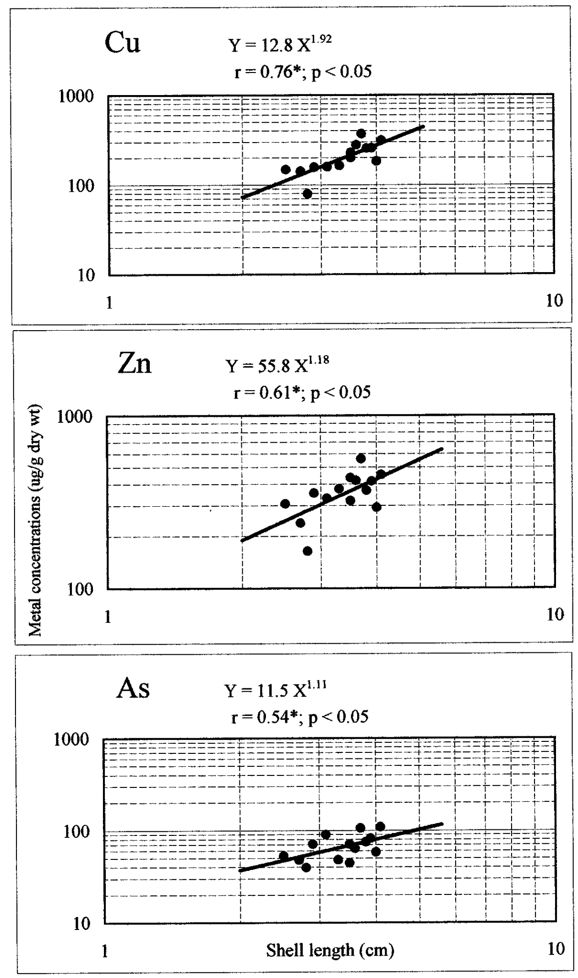

Fig. 5. Relationship between metal concentrations (μg/g dry wt) and shell length (cm) of rock-shells (*Thais clavigera*) from the Putai coastal area of Taiwan

$b < 1$, the metal accumulation rates in small size organisms are faster than in large size ones. When $b > 1$, the metal accumulation rates in small size organisms are slower than those in large size ones. Williamson (1980) suggests that the variation in metal content between smaller and larger mussels may be due to the difference in their metabolic activity, which may affect the metabolism of metals. The contents of Cu and Zn reach a plateau in the large size group which may indicate that these two essential metals are associated with growth, which also reaches a plateau in old individuals (Lobel and Wright 1982).

The existence of size-dependent metal accumulation by aquatic biota has been documented by several investigators (Boyden 1977; Rainbow 1989; Swaileh and Adelung 1994), although the information reported for *Thais clavigera* is limited in Taiwan. In this study, the relationships between metal concentration in *Thais clavigera* and body size are presented in Figure 3. For Cu, Zn, and As, the log-log function gives a similar correlation pattern. Positive relationships were observed for Cu, Zn and As concentrations in *Thais clavigera.* For Cu in *Thais clavigera*, the log-log function (power model: $Y = aX^b$) gives the best correlation ($r = 0.86$; $p < 0.05$). In other words, the power model provided the most accurate regression fit (Figure 3). Generally, the slope of the regression of metal concentrations versus size varies depending on the element. This may be determined by the life cycle and by environmental factors such as pH and organic matter (Paez-Osuna and Ruiz-Fernandez 1995). The present significant correlation found in various elements may be interpreted in terms of a certain disability to regulate internal concentrations shown by *Thais clavigera*, and body size and associated biochemical factors that have strong influence on the variability.

Our results also show that the concentrations of As, Cu, and Zn in *Thais clavigera* are significantly affected by body size, with large individuals having the greatest concentrations. The size effect may be a function of any one or several age dependent parameters. It may depend on the difference between surface/volume ratio, and on metabolic and feeding rates of larger (older) and small (younger) individuals (Phillips 1980). Differences might also reflect different types of diets between young and old individuals (Paez-Osuna *et al.* 1995), and unfortunatley this aspect is unknown in *Thais clavigera.* At the same time, concentrations of As, Cu and Zn are higher in large *Thais clavigera* than in small individuals. *Thais clavigera* is carnivorous and an oyster drill, and can therefore accumulate much higher As, Cu, and Zn from surrounding environments, especially oysters.

The metal contents in different shell lengths were also investigated. The shell lengths ranged from 2.6 to 4.1 cm.

Figure 4 indicates that the As, Cu, and Zn contents in *Thais clavigera* increase with shell length and have postive slopes of 4.57, 5.39, and 4.64, respectively. From the high slopes, it may be explained that the metals accumulation rates in large-sized *Thais clavigera* are faster than those in small-sized ones. These slopes are not significantly different from one another. The concentration/length plots (Figure 5) generally indicate a similar trend about metal behavior as content/length plots do. However, their correlation coefficients (0.76, 0.61, and 0.54, respectively) are much less than those obtained from content/length plots (0.94, 0.92, and 0.92, respectively) (Figure 4).

In summary, small (younger) individuals had lower concentrations of As, Cu, and Zn than large (older) individuals. This pattern and its variations are attributed to the metabolic activity for specific elements of the animals. The results indicate that biomonitoring programs using *Thais clavigera* as an indicator of As, Cu, and Zn contamination must take into account the *Thais clavigera* size as a source of metal concentration variations and that *Thais clavigera* may help to reduce monitoring requirements and provide early warning of potential contamination problems.

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