ELSEVIER

Environmental Pollution 110 (2000) 207–215

ENVIRONMENTAL
POLLUTION

www.elsevier.com/locate/envpol

Mussel Watch: a review of Cu and other metals in various marine organisms in Taiwan, 1991–98

Ming-Shiou Jeng[a], Woei-Lih Jeng[b], Tsu-Chang Hung[b,c], Ching-Ying Yeh[d], Rong-Jeng Tseng[e], Pei-Jie Meng[c], Bor-Cheng Han[d,*]

[a]*Institute of Zoology, Academia Sinica, Taipei, Taiwan*
[b]*Institute of Oceanography, National Taiwan University, Taipei, Taiwan*
[c]*Institute of Chemistry, Academia Sinica, Taipei, Taiwan*
[d]*School of Public Health, Taipei Medical College, 250 Wu-Hsing Street, Taipei, Taiwan*
[e]*Department of Biology, Chinese Cultural University, Taipei, Taiwan*

Received 19 October 1998; accepted 5 November 1999

"Capsule": *Mussels and oysters were especially useful in monitoring trace elements in the marine environment around Taiwan.*

Abstract

This study presents the distribution of Cu, Zn, Pb, Cd, Hg and As in various marine organisms collected along the western coast of Taiwan from 1991 to 1998, and also evaluates the time variation of Cu in oysters before (1980–85) and after (1986–98) the "green oyster" incident. The results show that relatively high geometric mean (GM) concentrations of Cu, Zn, Pb, Cd, As and Hg were generally found in *Crassostrea gigas* (Cu = 229 μg/g, Zn = 783 μg/g), *Gomphina aeguialtera* (Pb = 30.3 μg/g), *Tegillarca granosa* (Cd = 2.85 μg/g), *Thais clavigera* (As = 96.9 μg/g) and *Parapenaeopsis cornuta* (Hg = 1.35 μg/g), respectively. Especially, maximum Cu and Zn concentrations (GM = 229 and 783 μg/g, respectively) in oysters (*C. gigas*) from different culture areas were much higher than those of the other organisms by about 1.13–458 and 2.40–63.7 times, respectively. Similarly, rock-shells (*Thais clavigera*) had a high capacity for accumulating Cu (GM = 202 μg/g) and Zn (GM = 326 μg/g) under the same physico-chemical conditions. The highest GM Cu and Zn concentrations of 1108 (range from 113 to 2806) and 1567 (range from 303 to 3593) μg/g were obtained in oysters from the Hsiangshan coastal area, one of the most important oyster culture areas in Taiwan. However, the highest GM Cd and As concentrations of 6.82 and 19.3 μg/g were found in oysters from the Machu Islands. Mean Cu concentrations in the oysters from the Erhjin Chi estuary declined from 2194 ± 212 μg/g in 1986–90 to 545 μg/g (GM) in 1991–96. In the Hsiganshan area, GM Cu concentrations of 909 μg/g (1991–96) and 1351 μg/g (1997–98) in oysters were significantly higher than those of 201 μg/g (1980–85) and 682 μg/g (1986–90). The gradually increasing levels of Cu and Zn in the oysters from the Hsiangshan area have been observed year by year.

Keywords: Mussel Watch; Metal contamination; Indicator organisms

1. Introduction

Due to dense population and rapid industrial expansion, the estuarine and coastal environments of Taiwan are suffering an ever-increasing impact from human activity (Hung, 1988; Han and Hung, 1990; Han et al., 1994; Jeng and Han, 1994). In recent years, the pollution of various rivers and coastal areas by trace metals (Cu, Zn, Pb, Cd, Hg, As) and organohalogen compounds (polychlorinated biphenyls and polychlorinated dibenzo-*p*-dioxins) has been the subject of intense public concern in Taiwan (Ling et al., 1995; Han et al., 1996). Aquatic organisms may bioaccumulate anthropogenic organic compounds and trace metals in their tissues, both from water and sediment, and it is used as an aid in the monitoring of aquatic pollutants (Phillips, 1980). Bivalves (such as oysters) and especially mussels (such as green mussels) are widely used as sentinel organisms for monitoring the concentration of selected pollutants in coastal environments (Goldberg et al., 1978; Han and Hung, 1990; Hung et al., 1997). Heavy metals, organochlorine compounds and petroleum hydrocarbons have

* Corresponding author. Tel.: +886-2-2736-1661, ext. 666; fax: +886-2-2738-4831.
E-mail address: bchan@mail.tmc.edu.tw (B.-C. Han).

0269-7491/00/$ - see front matter
PII: S0269-7491(99)00304-8

long been recognized as the most deleterious contaminants to biota in the world's marine and estuarine waters (Martin and Richardson, 1991). During the past 20 years various biomonitoring strategies have been developed to monitor and evaluate the adverse impact of these compounds on marine ecosystems in Taiwan. One of the most successful efforts has involved the use of bivalve shellfish as sentinel organisms. Mussels (and oysters) have become widely used for monitoring contamination in coastal and estuarine ecosystems because they, as filter feeders, bioaccumulate contaminants. This approach has become popularly known as the "Mussel Watch" (UNEP, 1993). Goldberg et al. (1978) were the first to initiate the "Mussel Watch Program" in which the bivalves were used as a surveillance tool of coastal pollution. Recently, the Mussel Watch Program has been expanded into the Asia/Pacific region by International Mussel Watch Committee, under the support of the United Nations University (UNU, 1994). Scientists from 12 Asia/Pacific countries and territories including Taiwan were invited to join the program.

From August 1995 to July 1998, we carried out a 3-year Asia/Pacific Mussel Watch (APMW) project funded by the National Science Council, Republic of China. A series of studies in Taiwan have been carried out; e.g. Han et al. (1996) found that Cu concentrations in rock-shells (*Thais clavigera*) were 12–32 times higher than those in other benthic organisms. That capacity makes *Thais clavigera* a potential candidate for monitoring Cu in marine sediments. A strong multiple regression correlation ($P<0.05$, $r^2=0.7894$) also indicates that the Cu carbonates may dominate as the available form of Cu to *Hiatula diphos* from various environments in the Lukang coastal area of Taiwan under natural physicochemical conditions (Han et al., 1996). On the other hand, one interesting point is that relatively high As concentrations (65.7 μg/g dry wt.) in *Thais clavigera* are found as compared with those in other organisms (range from 2.37 to 40.2 μg/g dry wt.). For *Thais clavigera*, double logarithmic plots of metal content and concentration against dry body weight and shell length show linear relationships. In conclusion, *Thais clavigera* has a high capacity for accumulating As, Cu and Zn and is a potential bioindicator for monitoring As, Cu and Zn (Han et al., 1997).

The APMW project was implemented to measure concentrations of metals and organic chemicals in surface sediments, whole soft parts of oysters, other fish and shellfish collected from 17 coastal and estuarine sites of Taiwan. This study presents the distribution of Cu, Zn, Pb, Cd, Hg and As in various marine organisms collected along the western coast of Taiwan from 1991 to 1998, and also selects some potential bioindicator candidates for monitoring various metals in the marine environment. In addition, this paper also evaluates the time variation of Cu in oysters before (1980–85) and after (1986–98) the "green oyster" incident, and examines the concentrations of metals in organisms to see if they exceed the levels which would be hazardous to either human health or to the fish population.

2. Materials and methods

The APMW project is national in scale, and sampling sites should be representative of large areas rather than the small-scale patches of contamination commonly referred to as 'hot spots'. Therefore, our sampling locations were widely distributed along the Taiwan coast. Sampling locations are shown in Fig. 1. Since no single species of organisms is common to all coasts, it has been necessary to collect different ones. Thirty-three species of marine organisms such as oysters (*Crassostrea gigas*) were sampled from different sites on the western coast of Taiwan during the period from August 1991 to July 1998.

The samples were returned to the laboratory, individually scrubbed, and shucked, and the flesh was placed in weighed acid-washed Teflon beakers for reweighing to obtain individual wet weight values. All samples were digested using a microwave digester (Model MDS-2000) with a mixture of nitric and sulfuric acids (1:1, v/v) solution, and the supernatant analyzed for Cu and Zn by flame atomic absorption spectrophotometry (AAS; Han and Hung, 1990). Pb, Cd, As and Hg were measured by graphite atomic absorption spectrometry (GAAS) using Ni (for As) and TeO_2 (for Hg) as matrix modifiers, and standard addition procedures were used for the calculation of the analyte concentrations (Han et al., 1997). All concentration (μg/g) data on the basis of dry weight by using wet wt.≅dry wt.×5 (Han and Hung, 1990; Han et al., 1994).

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 (Han et al., 1996, 1997).

For statistical analyses, left-skewed data were normalized by logarithmic transformation. Accordingly, geometric mean (GM) values were reported. Student's *t*-test was used to study differences of various metal concentrations between different organisms (Han et al., 1997).

3. Results and discussion

3.1. Quality assurance of metals in various organisms

Replicate analysis of standard reference materials (Oyster tissue SRM 1566a; Tuna Fish IAEA-350) from

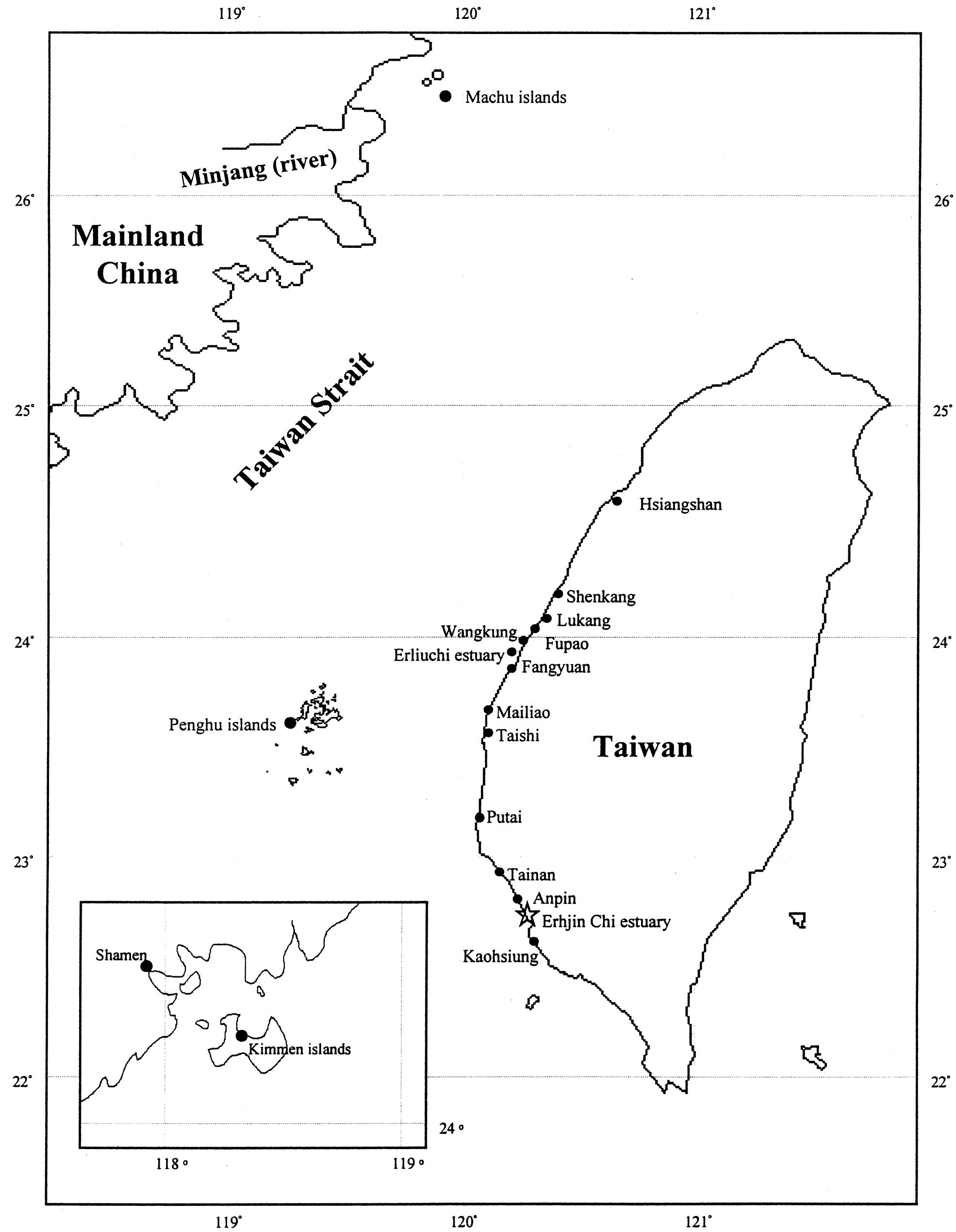

Fig. 1. Sampling locations (●) of various organisms collected from different coastal areas in Taiwan.

the US National Institute of Standards and Technology and International Atomic Energy Agency showed good accuracy (<20%) 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 850±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.

3.2. Metal concentrations in various organisms

In Taiwan, most industrial areas are located on the western coast, where the main oyster mariculture areas are also located. Most pollutants from factories are discharged on the coast or into rivers of western Taiwan, causing serious pollution (Han and Hung, 1990; Han et al., 1996; Jeng and Han, 1996; Meng, 1998). The Environmental Protection Administration (EPA), Republic of China, reported that nine major and minor rivers of 297 km total length in western Taiwan appeared to be heavily polluted (EPA, 1997). The results of various metals in oysters (from 1991 to 1998) and other organisms (25 species, from 1991 to 1996) collected along the western coast of Taiwan are listed in Table 2. The GM metal concentrations in organisms varied with species and location. For example, relatively high concentrations of Cu, Zn, Pb, Cd, As and Hg were

Table 1
Determination of metals in standard reference materials (SRM) by flame atomic absorption spectrophotometry (FAAS) and graphite atomic absorption spectrophotometry (GAAS)[a]

SRM	Metal	Method	Value obtained in our lab. (μg/g)	Certified value (μg/g)	Precision (%)	Accuracy (%)
Oyster (SRM1566a)	Cu	FAAS	60.3 ± 3.9 ($n=7$)	66.3 ± 4.3	6.47	−9.05
	Pb	GAAS	0.428 ± 0.029 ($n=7$)	0.371 ± 0.014	6.78	+15.4
	Zn	FAAS	850 ± 58 ($n=7$)	830 ± 57	6.81	+2.41
	Cd	GAAS	4.32 ± 0.44 ($n=7$)	4.15 ± 0.38	10.2	+4.10
	As	GAAS	14.3 ± 2.1 ($n=8$)	14.0 ± 1.2	14.4	+2.29
	Hg	GAAS	0.057 ± 0.009 ($n=8$)	0.064 ± 0.007	15.8	−10.9
Tuna Fish (IAEA-350)	Cu	FAAS	2.70 ± 0.17 ($n=5$)	2.83 (2.55–3.10)	6.44	−4.59
	Zn	FAAS	19.6 ± 3.0 ($n=5$)	17.4 (16.6–18.5)	15.2	+12.6
	Cd	GAAS	0.024 ± 0.002 ($n=6$)	0.020 (0.014–0.026)	8.33	+20.0
	As	GAAS	5.04 ± 0.35 ($n=6$)	5.28 (3.36–5.75)	7.03	−4.48
	Hg	GAAS	4.46 ± 0.52 ($n=6$)	4.68 (4.36–4.91)	11.7	−4.70

[a] Pb certified value not found for Tuna Fish (IAEA-305).

generally found in *C. gigas* (Cu = 229, Zn = 783 μg/g), *Gomphina aeguilatera* (Pb = 30.3 μg/g), *Tegillarca granosa* (Cd = 2.85 μg/g), *Thais clavigera* (As = 96.9 μg/g) and *Parapenaeopsis cornuta* (Hg = 1.35 μg/g), respectively. In addition, these data indicate that Zn in various organisms was higher than those of other metals. Especially, maximum Cu and Zn concentrations (GM = 229 and 783 μg/g, respectively) in oysters (*C. gigas*) were much higher than those of the other organisms by about 1.13–458 and 2.40–63.7 times, respectively. It can be seen that the ranges of Cu and Zn concentrations in the oysters differed significantly ($P < 0.05$) from those of the other organisms. The result indicates that the ability of oysters to concentrate Cu and Zn is much stronger than that of other organisms. Therefore, the potential risk of consuming oysters is relatively higher than that of other seafood (such as *H. diphos*, etc.) due to the high bioaccumulation of oysters. These high concentrations of Cu and Zn may not affect oysters directly (Han and Hung, 1990), but high concentrations of Cu, Zn, and As may transfer toxicity to humans through the food chain.

By arranging the GM metal concentrations in various organisms (Table 2) in decreasing order, Table 3 shows the top four or five potential bioindicator candidates for monitoring various metals in the marine environment. These organisms have a large capacity for the accumulation of various metals, and have therefore commonly been suggested as biomonitors of metal contamination (Phillips, 1990). It was noted by Bryan et al. (1985) that indicator organisms should be good accumulators of metals and their body metal concentrations must reflect differences in metal bioavailability. For example, oysters 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.

Similarly, rock-shells (*Thais clavigera*) have a high capacity for accumulating Cu (GM = 202 μg/g) and Zn (GM = 326 μg/g) under the same physico-chemical conditions. This capacity makes rock-shells a potential candidate except oysters for monitoring Cu and Zn in coastal environments. Bryan et al. (1985) reported that indicator organisms should be good accumulators of metals and their tissue concentrations must reflect metal availability. For this reason, oysters and rock-shells are clearly suitable. In addition, the results of Table 2 reveal that relatively high As concentrations (GM = 96.9 μg/g) in *Thais clavigera* are found as compared with those in other organisms (GM range from ND to 45.9 μg/g). Han et al. (1996, 1997) also indicate that *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. In addition, GM As concentrations (40.2 and 45.9 μg/g) in two different species were higher than the ‘high’ concentrations (17 μg/g) of As by O’Connor (1992). Two general features about the ‘high’ concentrations are worth noting. First, the ‘high’ concentrations can often be attributed to human activities because they are found at places where human population is high. Second, there are many instances showing places where trace elements are at ‘high’ concentrations for purely natural reasons and are not evidence of contamination (O’Connor and Beliaeff, 1995) such as black-foot disease in Taiwan caused by high As concentrations in well-water (Chiou et al., 1995). In general, we know that all species are not the same with regard to accumulation of all trace elements. From a previous

Table 2
Geometric mean metal concentrations (μg/g dry wt.) in various organisms collected from several coastal areas of Taiwan[a]

Organism	Location	Cu	Zn	Pb	Cd	As	Hg
Crustacea							
Sword prawn (*Parapenaeopsis cornuta*)	Mailiao	20.3	83.4	ND	0.090	0.120	1.35
Sword prawn (*Parapenaeopsis hardwickii*)	Mailiao	16.3	67.8	ND	0.030	0.030	0.240
Swimming crab (*Charybdis japonica*)	Kaohsiung	19.2	134	0.090	0.01	11.6	0.270
Swimming crab (*Charybdis natator*)	Kaohsiung	10.8	103	0.720	0.060	4.32	ND
Swimming crab (*Charybdis* sp.)	Kaohsiung	28.1	102	0.060	0.210	14.5	ND
Swimming crab (*Portunus hastatoides*)	Mailiao	13.8	13.8	0.810	0.120	0.810	0.150
Smooth pebble crab (*Philyra pisum*)	Mailiao	30.0	93.6	2.58	0.480	0.540	1.05
Mollusca							
Rock-shells (*Thais clavigera*)	Tanshui, Lukang, Putai Chiku, Machu Islands	202	326	1.22	1.15	96.9	0.845
Lined moon shell (*Natica lineata*)	Mailiao	62.7	101	2.49	0.600	13.7	0.180
Bladder moon shell (*Polinices didyma*)	Chiangchun	194	191	0.300	1.23	9.54	0.150
Ear shell (*Sinum* sp.)	Mailiao	110	73.5	1.26	0.120	0.090	0.120
Granular ark clam (*Tegillarca granosa*)	Putai	1.17	19.1	ND	2.85	2.37	0.150
Variegate venus clam (*Ruditapes variegatus*)	Shamen (Mainland China)	5.46	21.2	0.279	0.390	8.16	0.175
Equilateral venus clam (*Gomphina aeguilatera*)	Lukang	6.30	21.5	30.3	0.240	ND	ND
Pocker-chip venus clam (*Meretrix lusoria*)	Lukang	2.43	29.3	11.0	0.090	ND	ND
Purple clam (*Hiatula diphos*)	Lukang	4.98	31.2	26.6	1.26	ND	ND
False fusus (*Hemifuscus tuba*)	Putai	12.8	76.5	ND	0.420	40.2	0.210
Corbula clam (*Corbula formosensis*)	Mailiao	24.2	25.8	5.97	0.090	1.71	ND
Mactra clam (*Mactra* sp.)	Kaohsiung	8.58	85.8	16.2	1.62	1.65	ND
Dosinia clam (*Dosinia sinensis*)	Putai	6.27	48.0	0.180	0.003	5.37	0.090
Jackknife clam (*Solen strietus*)	Chiku	6.00	55.5	ND	0.540	4.17	0.120
Green mussel (*Mytilus smarrangdinus*)	Machu Islands	13.7	47.7	ND	1.21	2.21	0.056
Green mussel (*Perna viridis*)	Putai	3.69	19.8	0.090	0.390	3.42	0.210
Oyster[b] (*Crassostrea gigas*)	Hsiangshan, Shenkang, Fupao, Wangkung, Erliuchi estuary, Fangyuan, Taishi, Putai, Anpin, Penghu Islands, Machu Islands, Kimmen Islands	229	783	0.446	1.76	10.8	0.270
Fish							
Anchovy (*Thryssa dussumieri*)	Kaohsiung	5.52	45.9	8.91	0.300	4.56	ND
Flounder (*Psettodes erumei*)	Putai, Chiangchun, Kaohsiung	ND	12.3	ND	0.030	45.9	0.319

[a] All data are 6-175 determinations for various organisms. ND: Cu < 0.5; Cd < 0.003; As < 0.03; Hg < 0.05; Pb < 0.05 (μg/g).
[b] Oysters collected from 1991 to 1998, other organisms from 1991 to 1996.

Table 3
Potential candidates for monitoring various metals in the marine environment

Metal	Potential bioindicator
Cu	*Crassostrea gigas, Thais clavigera, Polinices didyma, Sinum* sp.
Zn	*Crassostrea gigas, Thais clavigera, Polinices didyma, Charybdis japonica*
Pb	*Gomphina aeguilatera, Hiatula diphos, Mactra* sp., *Meretvix lusoria, Thryssa dussunieri*
Cd	*Tegillarca granosa, Crassostrea gigas, Mactra* sp., *Hiatula diphos*
As	*Thais clavigera, Psettodes erumei, Hemifuscus tuba, Charybdis* sp.

report (Han and Hung, 1990), the accumulation of metals by aquatic organisms depends not only on the physico-chemical properties of the elements themselves, but also on the organism's biological strategies which have evolved to detoxify metals or to otherwise reduce the exposure of species to high concentrations of metals in their tissues (Phillips, 1995).

3.3. Time and location variation of Cu and Zn in oysters

Furthermore, Figs. 2 and 3 show the GM metal concentrations in oysters collected from different sites on the western coast of Taiwan during 1991–98. Metal concentrations in oysters varied with different sampling

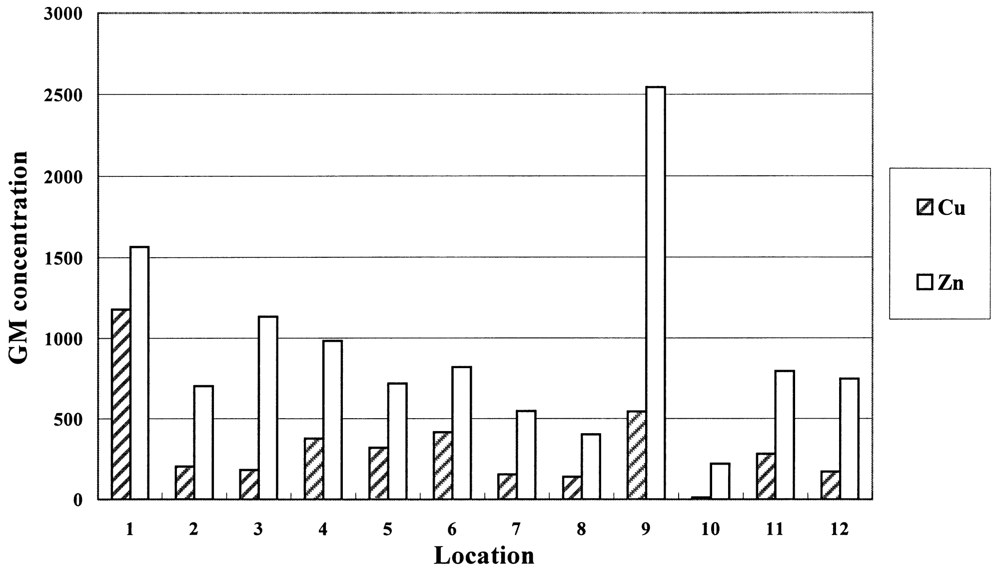

Fig. 2. Geometric mean (GM) Cu and Zn concentrations (μg/g dry wt.) in oysters (*Crassostrea gigas*) collected from different coastal areas of Taiwan during 1991–98. 1, Hsiangshan; 2, Shenkang; 3, Fupao; 4, Wangkung; 5, Erliuchi estuary; 6, Fangyuan; 7, Taishi; 8, Putai; 9, Anpin; 10, Penghu Islands; 11, Machu Islands; 12, Kimmen Islands.

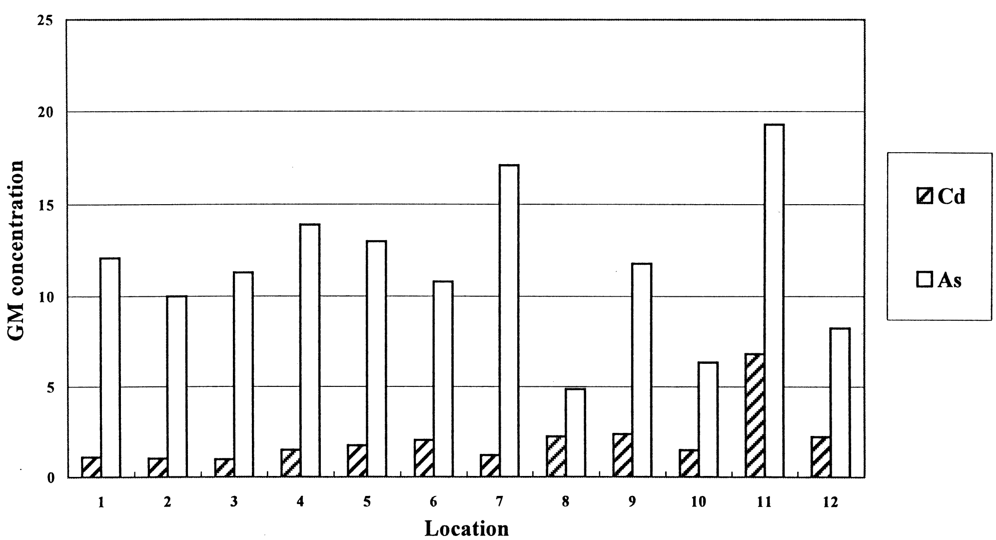

Fig. 3. Geometric mean (GM) Cd and As concentrations (μg/g dry wt.) in oysters (*Crassostrea gigas*) collected from different coastal areas of Taiwan during 1991–98. 1, Hsiangshan; 2, Shenkang; 3, Fupao; 4, Wangkung; 5, Erliuchi estuary; 6, Fangyuan; 7, Taishi; 8, Putai; 9, Anpin; 10, Penghu Islands; 11, Machu Islands; 12, Kimmen Islands.

locations. The highest GM Cu and Zn concentrations of 1108 and 1567 μg/g were obtained in oysters from the Hsiangshan coastal area (Fig. 2), one of the most important oyster mariculture areas in Taiwan. In general, when the Cu concentration in oysters was over 500 μg/g dry wt., the color of the oysters became green (Han and Hung, 1990). The results indicate that local inputs of Cu and Zn are the major cause of "green oysters" in this area. This suggests that the Hsiangshan coastal environment is influenced by the local anthropogenic inputs of chemicals, especially Cu and Zn (Tseng and Han, 1998). There were low Cu (GM = 11.4 μg/g) and Zn (GM = 220 μg/g) concentrations in the oysters collected from the Penghu Islands (Fig. 2), probably because their location is far from pollution sources. However, the highest GM Cd and As concentrations of 6.82 and 19.3 μg/g were found in oysters from the Machu Islands (Fig. 3) which are very close to the Minjang estuary in the Fu-Chien province of southeastern China. Hung et al. (1997) reported that the average heavy metal concentrations in mud (< 63 μm), sand and seawater collected from the Minjang estuary were high compared with those concentrations observed in other areas of Taiwan. This may be due to the fact that the heavy metal pollutant discharges from the Minjang (river), which was near the urban areas of

Fu-Chou City, People's Republic of China. In other words, the generally high Cu, Zn, Cd and As concentrations in oysters indicate that local and regional inputs of Cu, Zn, Cd and As are the major pollution sources in the Hsiangshan coastal area and Machu Islands, respectively (Hung et al., 1997; Tseng and Han, 1998). In general, bivalves are a hazard to health because they accumulate and concentrate toxins by continuously filter feeding minute particles from water. Similarly, intake of high Cd and As concentrations through consumption of oysters collected from the Machu Islands could be dangerous, especially for some high-risk Machu residents. Pb and Hg concentrations in oysters from different areas were generally low (range ND–1.22 and 0.100–0.582 μg/g, respectively). It should be pointed out that, although Pb and Hg were not enriched in the organisms, Zn displayed a similar behavior to Cu. The correlation coefficient between these two metals in the oysters was 0.85 ($P<0.05$) (Tseng and Han, 1998). Whether we are concerned with Cu pollution or Zn pollution or some which may be combination of the two is not yet clear (Goldberg, 1992).

Table 4 shows that the GM concentrations of Cu, Zn and As (45.8, 157 and 2.16 μg/g wet wt., respectively) in Taiwan's oysters were beyond the upper limits of those reported for legal limits of fish and shellfish products from different countries in the world. A comparison with the other country's food standard for acceptable levels: Cu, 10–100; Zn, 30–100 and As, 0.1–10 μg/g wet wt. in fish and shellfish products shows that there is metal contamination in the food chain for Taiwan's oysters. The average concentrations of As, Cu and Zn are higher than the legal limits of some countries. It is worthy of note that these three metals have wider concentration ranges. Particularly, their high extremes are elevated by a factor of about 5 for Cu for instance. The relatively high values found for Cu, Zn and As are alarming because they are toxic through consuming oysters from different areas in Taiwan.

Table 4
Geometric mean metal concentrations in oysters of Taiwan compared to legal limits of metals in fish and shellfish products from different countries[a]

Country	Metals (μg/g wet wt.)		
	As	Cu	Zn
Australia	1.5	10–70	40–1000
Canada	3.5		
Chile	0.12, 1.0	10	100
Ecuador	1.0	10	
Finland	5.0		
Hong Kong	1.4–10		
India	1.0	10	50
New Zealand	1.0	30	40
Philippines	3.0		
Poland	4.0	10–30	30–50
Thailand	2.0	20	
United Kingdom	1.0	20	50
Venezuela	0.1	10	
Zambia	3.5–5.0	100	100
Range			
Minimum	0.1	10	30
Maximum	10	100	1000
Taiwan (this study)	2.16 (0.97–3.86)	45.8 (2.28–236)	157 (44.0–509)

[a] Data from USFDA (1983).

Fig. 4 shows the time and location variation of average Cu concentrations in the oysters collected from four areas from 1980 to 1998. The highest average concentrations of Cu (2194 ± 212 μg/g) in the oysters from the Erhjin Chi estuary were observed from 1986 to 1990, after the "green oyster" incident. The green oysters collected from the Erhjin Chi estuary on 26 January 1986, gave the highest Cu concentration of 4401 ± 79 μg/g (Han and Hung, 1990). At the same time, besides the Erhjin Chi estuary, Hsiangshan also produced green oysters from time to time. Comparing the Cu concentrations in the oysters collected from the Erhjin Chi estuary to those from Hsiangshan, Putai and the Penghu Islands, all except Hsiangshan displayed declined levels of Cu in the oysters year by year. Generally, a review of data shows that the Erhjin Chi estuary had the highest average concentrations of Cu in the oysters (3075 ± 826 μg/g) during 1988, but gradually decreasing levels have been observed since then because a program has been implemented by the EPA to reduce the pollution in this area. Hundreds of millions of dollars were lost because a major sector of aquaculture in this region had to be closed. Closure of the culture area caused Cu levels to be reduced. In addition, the government of Taiwan has allocated over $200 billion (NT dollars) in controlling the pollution of various rivers, showing its resolution to improve the polluted streams in Taiwan (Han et al., 1994). Mean Cu concentrations in the oysters from the Erhjin Chi estuary declined from 2194 ± 212 μg/g in 1986–90 to 545 μg/g (GM) in 1991–96 (Fig. 4). Lee et al. (1996) also reported that Cu concentrations in oysters significantly decreased in the Charting and Anpin areas between 1987 and 1993. In addition, Putai and Penghu islands also displayed declining levels of Zn in the oysters year by year (Fig. 5).

Unfortunately, Figs. 4 and 5 show the Hsiganshan area of GM Cu and Zn concentrations of 909 and 1346 μg/g (1991–96), 1351 and 1744 μg/g (1997–98) in oysters were significantly higher than those of 201 and 1186 μg/g (1980–85), 1271 and 682 μg/g (1986–90), respectively. The gradually increasing levels of Cu and Zn in the oysters have been observed in the Hsiangshan area year by year. In other words, our results show a statistically significant dominance of Cu and Zn increases in Hsiangshan oysters between 1980 and 1998. An extremely high concentration of Cu (2806 μg/g) with a

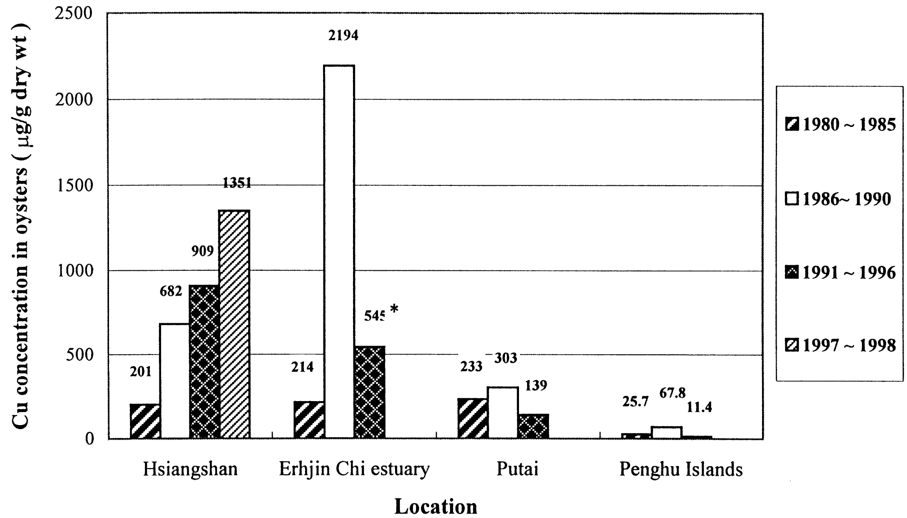

Fig. 4. Time and location variation of average Cu concentration in oysters (*Crassostrea gigas*) collected from four areas from 1980 to 1998. An asterisk denotes Anpin which is close to the Erhjin Chi estuary.

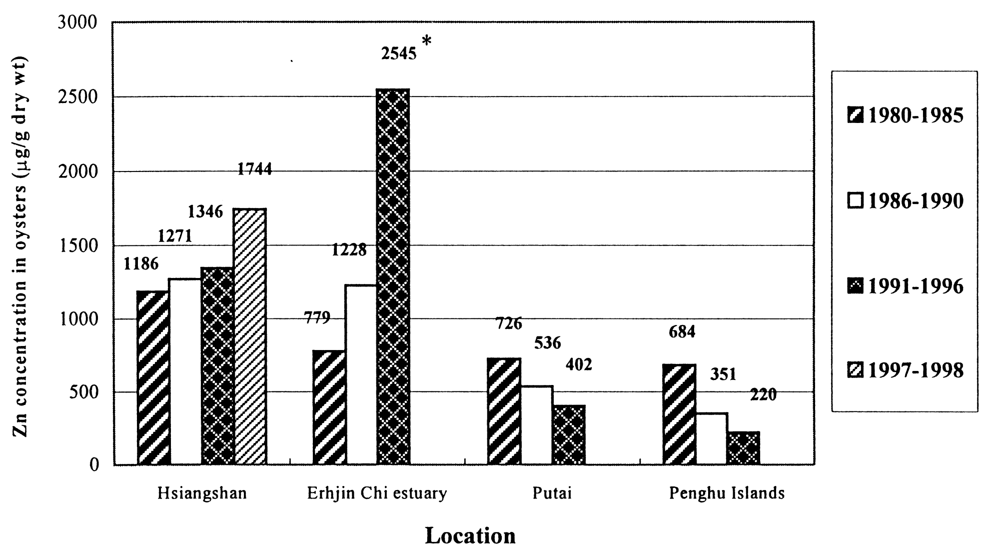

Fig. 5. Time and location variation of average Zn concentration in oysters (*Crassostrea gigas*) collected from four areas from 1980 to 1998. An asterisk denotes Anpin which is close to the Erhjin Chi estuary.

relatively high concentration of Zn (3593 µg/g) was found in the oysters of Hsiangshan. The high Cu and Zn concentrations found in the Hsiangshan oysters are attributed to local input. So, green oysters still were occasionally observed in the Hsianghsan coastal area since 1986. However, the results indicate Penghu Islands oysters had the lowest Cu levels in 1980–98 (11.4–67.8 µg/g), probably because of its remote location (i.e. away from pollution sources).

In general, the concentrations of Cu in the oysters collected from each location were below 100 µg/g before 1980. As industries grew and their environmental impact got strong in Taiwan from 1980 to 1990, the Cu and Zn concentration in the oysters for each location gradually increased year by year, especially for Cu. For instance, the Cu concentrations of oysters collected from the Erhjin Chi estuary and Hsiangshan from 1988 to 1990 were, respectively, 61 and 29 times higher than those of 10 years ago (Han et al., 1994). Obviously, metal pollution became worse over time without a sign of alleviation (Han et al., 1994). Stringent measures currently being taken by the government, including removing the sediment of some portions of rivers, should be able to bring the Cu and Zn concentrations in oysters down to the levels similar to those observed in legal limits of other countries.

On the whole, biomonitoring can show temporal changes and the spatial distribution of local metal contamination. Oysters with higher concentrations of Cu and Zn expressed as tissue load were found in contaminated areas, especially the Hsiangshan area. All the above several metals can be acutely or chronically toxic to marine life and to humans under some conditions. On the other hand, while the elements As, Cu and Zn can be toxic at high concentrations, they are also essential in small quantities to the maintenance of life (O'Connor and Beliaeff, 1995).

Acknowledgements

This study was supported by grants from the National Science Council, Republic of China (NSC-86-2621-B-038-001-Z and NSC-85-2621-B-038-001-ZY).

References

Bryan, G.W., Langston, W.J., Hummerstone, L.G., Burt, G.R., 1985. A Guide of the Assessment of Heavy-metal Contamination in Estuaries Using Biological Indicators (Occasional Publication No. 4). Marine Biology Association, Plymouth, UK.

Chiou, H.Y., Hsueh, Y.M., Liaw, K.F., Horng, S.F., Chiang, M.H., Pu, Y.S., Lin, J.S.N., Huang, C.H., Chen, C.J., 1995. Incidence of internal cancers and ingested inorganic As: a seven-year follow-up study in Taiwan. Cancer Research 55, 1296–1300.

EPA, 1997. Year Book of Environmental Statistics. Office of Statistics, Environmental Protection Administration, Taipei, Taiwan, ROC.

Goldberg, E.D., 1992. Marine metal pollutants a small set. Marine Pollution Bulletin 25, 45–47.

Goldberg, E.D., Bowen, V.Y., Farrington, J.W., Harvey, G., Martin, J.H., Paker, P.L., Risebrough, R.W., William-Robertson, M.A., Schneider, E., Gamber, E., 1978. The mussel watch. Environmental Conservation 5, 101–125.

Han, B.C., Hung, T.C., 1990. Green oysters caused by copper pollution on the Taiwan coast. Environmental Pollution 65, 347–362.

Han, B.C., Jeng, W.L., Hung, T.C., Jeng, M.S., 1994. Copper intake and health threat by consuming seafood from copper-contaminated coastal environments in Taiwan. Environmental Toxicology and Chemistry 13, 775–780.

Han, B.C., Jeng, W.L., Hung, T.C., Wen, M.Y., 1996. Relationship between copper speciation in sediments and bioaccumulation by marine bivalves of Taiwan. Environmental Pollution 91, 35–39.

Han, B.C., Jeng, W.L., Jeng, M.S., Kao, L.T., Meng, P.J., Huang, Y.L., 1997. 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. Archives of Environmental Contamination and Toxicology 32, 456–461.

Hung, T.C., 1988. Heavy metal pollution and marine ecosystem as a case study in Taiwan. In: Abbou, R., Antoni, F., Korte, F., Truhaut, R. (Eds.), Hazardous Wastes: Detection, Control, and Treatment. Elsevier, Amsterdam, Netherlands, pp. 869–879.

Hung, T.C., Ling, Y.C., Jeng, W.L., Huang, C.C., Han, B.C., 1997. Marine environmental monitoring and QA/QC system in Taiwan. Journal of Chinese Environmental Protection Society 20, 69–90.

Jeng, W.L., Han, B.C., 1994. Sedimentary coprostanol in Kaohsiung harbour and the Tan-Shui estuary, Taiwan. Marine Pollution Bulletin 28, 494–499.

Jeng, W.L., Han, B.C., 1996. Coprostanol in a sediment core from the anoxic Tan-Shui estuary, Taiwan. Estuarine, Coastal and Shelf Science 42, 727–735.

Lee, C.L., Chen, H.Y., Chuang, H.Y., 1996. Use of oyster, *Crassostrea gigas*, and ambient water to assess metal pollution status of the charting coastal area, Taiwan, after the 1986 green oyster incident. Chemosphere 33, 2505–2532.

Ling, Y.C., Soong, D.K., Lee, M.K., 1995. PCDD/DFS and coplanar PCBs in sediment and fish samples from the Erjen river in Taiwan. Chemosphere 31, 2863–2872.

Martin, M., Richardson, B.J., 1991. Long term contaminant biomonitoring: review from southern and northern hemisphere perspectives. Marine Pollution Bulletin 22, 533–537.

Meng, P.J., 1998. Characteristics of Copper Species and Complexing Capacity of Humic Substances from an Estuarine Environment. PhD thesis, Institute of Oceanography, National Taiwan University, Taiwan, ROC.

O'Connor, J.O., 1992. Mussel Watch: Recent Trends in Coastal Environmental Quality. NOAA, Rockville, MD, USA.

O'Connor, J.O., Beliaeff, B., 1995. Recent Trends in Coastal Environmental Quality: Results from the Mussel Watch Project. US Department of Commerce, National Oceanic and Atmospheric Administration, Silver Spring, MD, USA.

Phillips, D.J.H., 1980. Quantitative Aquatic Biological Indicators. Applied Science Publishers Ltd, London.

Phillips, D.J.H., 1990. 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 FL, Chapter 6.

Phillips, D.J.H., 1995. The chemistries and environmental fates of trace metals and organochlorines in aquatic ecosystem. Marine Pollution Bulletin 31, 193–200.

Tseng, R.J., Han, B.C., 1998. From the Relationship Between Concentration of Heavy Metals in Sediments and Oysters to Assess Aquaculture Environment on the West Coast of Taiwan (Special Publication). Council of Agriculture, Taipei, Taiwan, ROC.

UNEP, 1993. United National Environment Program: Environmental Data Report, 1993–1994. Blackwell Publishers, Oxford, UK.

UNU, 1994. Report of the UNU-IOC Workshop on Asia/Pacific Mussel Watch: Monitoring, Research and Training, 18–21 November 1994, Bali/Indonesia.

USFDA, 1983. Food Additives Permitted for Direct Addition to Food for Human Consumption (Section 172.804, Aspartame, Title 21). Code of Federal Regulations, Washington, DC.