

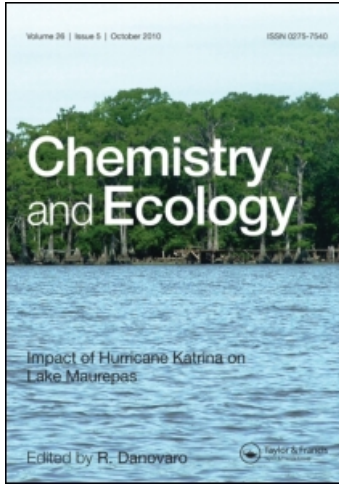
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METAL CONCENTRATION IN OYSTER, *CRASSOSTREA GIGAS*, AND SEDIMENT IN ANN-PING MARICULTURE GROUND, TAIWAN

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This study was to assess the metal contamination in oyster tissue grown in the Ann-ping mariculture ground in Taiwan. The information generated from this work also revealed general metal pollution problem for Taiwan's oyster farmers. Oysters, *Crassostrea gigas*, and surficial sediments collected from ten locations in Ann-ping mariculture ground in Taiwan for metals concentration (Cu, Zn, Pd, Cd, Fe and Mn) were performed. Analytical results indicated that the yearly averaged oyster copper concentrations ($\mu\text{g g}^{-1}$, wet weight) in oyster soft parts from Ann-ping increased from 21.3 ± 4.1 in 1993; 24.1 ± 6.8 in 1994; 36.8 ± 11.9 in 1995 to $43.9 \pm 23.1 \mu\text{g g}^{-1}$, wet weight, in the 1996 raising season. The mean oyster copper concentration reached a level of $50 \mu\text{g g}^{-1}$, wet weight, in December 1996. This increasing trend of metal concentration in oyster tissue indicates a potential pollution source which may pose a potential disaster as green oyster incidence, which occurred on the Charting coast in 1986, in Taiwan. Sediment samples in Ann-ping mariculture ground were also collected and examined. The seasonal variation of the copper concentration in surficial sediment from Ann-ping did not show an increasing trend as observed in oyster tissue.

Keywords: Metals; bio-monitoring; oyster; sediment; mariculture

INTRODUCTION

As it is well known, oyster accumulates heavy metals, particularly copper and zinc, from their aquatic environment (Han and Hung,

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1990; Lee *et al.*, 1996; Marcintic *et al.*, 1986; Mo and Nielsm, 1991). In general, the copper concentration in oysters reaches to approximately $100 \mu\text{g g}^{-1}$ level (wet weight), the oysters may become green and mortality may occur (Hung, 1990; Hung and Huang, 1990). A previous investigation reported the notorious green oyster incident, which occurred on the Charting coast of Taiwan (south of the Ann-ping mariculture ground) in 1986, and was due to the high copper concentration (up to $880 \mu\text{g g}^{-1}$ wet weight) in those oysters (Han and Hung, 1990). Several physiological variables such as temperature, size and reproductive state may affect the bioaccumulation of trace metals (van Haren *et al.*, 1994). Various researchers have examined the correlation between oyster metal concentration and the ambient water quality (Han and Hung, 1990; Lee *et al.*, 1996; Marcintic *et al.*, 1986; Mo and Nielsm, 1991). The studies concluded that the oyster copper concentration is related to the copper concentration in the aquatic environment. The studies (Zumuda and Sunda, 1982) showed that the particulate copper in the aquatic environment could be one of the most important environmental parameters affecting oyster copper accumulation. A recent work (Lee *et al.*, 1996) demonstrated that significant correlations between the copper and zinc concentrations in oysters as well as suspended particulates sampled in a raising area were in good agreement. Suspended particulate, as the important food source of the oyster, may play an important role to transport and bioaccumulate metal to oysters (Bewers and Yeats, 1989; Everaarts, 1989; Hung, 1990; Hung and Huang, 1990; Juracic *et al.*, 1987; Windom *et al.*, 1983). Suspended particulate has a tendency to sink to the bottom sediment and resuspend to the water column. These suspended particles may act as a metal source for oyster uptake. In this study, both oyster and sediment were monitored to examine the health of Ann-ping mariculture ground.

Ann-ping mariculture ground is located on the southwest coast of Taiwan Strait between Tsen-wen Chi (estuary) and Erh-jen Chi (Fig. 1). This area is surrounded by a domestic waste water treatment plant, a coal fired power plant, fishing boat industry, and many small and medium sizes of electronic and paper industry. Also, the nearby riverine fluxes of copper (78.2 ton y^{-1}) and zinc (421 ton y^{-1}) transported via suspended particles from the Tsen-wen Chi to the area was previously reported (Hung and Huang, 1990). Recently an industrial

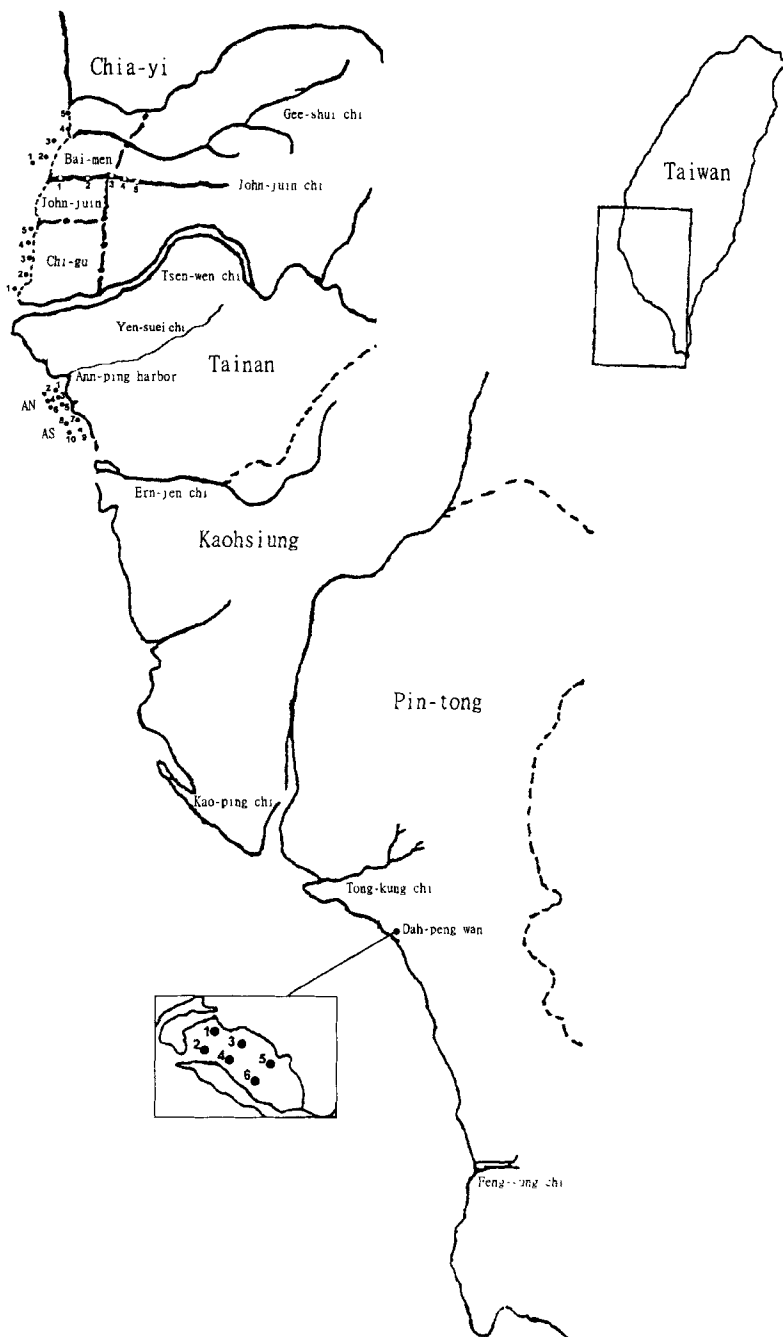


FIGURE 1 Location of sampling sites (AN: Northern Ann-ping, 1-6; AS: Southern Ann-ping, 7-10), and elsewhere.

complex development housing petrochemical and steel plants are just located about five miles north of the Tsen-wen Chi estuary. Both domestic and industrial waste effluent and the new industrial complex development could pose a potential problem for Ann-ping mariculture ground. This study was established to monitor both oyster and sediment in the area to protect the health of aquaculture industry and to assess the contamination problem along the coastal area. This study also provides a general background information to assess the impact of the new industrial complexes to the coastal environment in Taiwan (His *et al.*, 1997). The study was conducted from April 1994 to December 1996. The results of background information was reported in this paper. Comparisons were also made with levels of metal in oysters and sediments in other raising ground in southwestern Taiwan.

MATERIAL AND METHODS

Ten sampling stations are located in Ann-ping mariculture ground as depicted in Figure 1. Here, the surface temperature, pH and salinity were measured at each station using a portable pH/thermometer (JEMCO Model 6007) and a salinometer (ATAGO S/Mill-E). The average depth of those stations were about 6 metres and the oysters were raised by strings (about 2.4 metres) hung to floating bamboo complexes. Since those oysters were raised by different fishermen, the ages of oysters sampled for analysis were unknown. However, their weights and sizes were recorded and summarized in Table I. At least twenty five individuals of oyster (*Crassostrea gigas*) were sampled and sediments were collected by a surface dredge in each sampling station guided by a portable Global Position System (Trimble Navigation, Model 17319). Samples of oysters and sediments were kept cool and brought back to laboratory. The samples were immediately frozen until they were ready for analysis.

The oyster samples were shucked with a stainless steel knife. Whole soft parts of the oysters were dried for at least 12 hours in an oven at 105°C. Loss of metals due to volatilization was not found during this drying procedure. The dried samples were ground and homogenized with a ceramic pestle and mortar. About 1.5 grams of dried oyster tissue composited from twenty five individual oyster samples, were

TABLE I Mean concentrations (mg kg^{-1} , weight) of trace metal in oysters collected from the Ann-ping mariculture ground (1994 - 1996)

Station	n	Cu	Zn	Pb	Cd	Fe	Mn	weight(g)	Water content %
1	9	32.2 ± 14.5	177.6 ± 45.1	0.38 ± 0.32	0.12 ± 0.07	208.1 ± 118.0	8.2 ± 3.2	2.85 ± 1.01	75.6 ± 9.57
2	12	27.7 ± 13.7	193.0 ± 40.8	0.38 ± 0.34	0.18 ± 0.11	166.1 ± 57.3	6.4 ± 1.1	3.01 ± 1.30	78.1 ± 8.49
3	9	28.9 ± 12.7	176.7 ± 42.6	0.37 ± 0.21	0.14 ± 0.09	162.1 ± 80.0	7.2 ± 1.0	3.09 ± 1.41	74.3 ± 9.60
4	9	38.1 ± 29.5	170.5 ± 32.0	0.38 ± 0.28	0.23 ± 0.17	128.3 ± 161.1	6.5 ± 2.2	2.95 ± 1.56	75.6 ± 7.66
5	9	33.3 ± 7.8	206.6 ± 48.5	0.39 ± 0.24	0.23 ± 0.15	121.6 ± 39.3	5.9 ± 1.7	3.07 ± 1.46	76.2 ± 8.76
6	9	30.3 ± 7.2	186.5 ± 32.5	0.33 ± 0.32	0.22 ± 0.23	164.9 ± 48.2	5.5 ± 1.3	2.74 ± 1.54	76.6 ± 7.28
7	9	32.2 ± 18.1	168.8 ± 33.6	0.32 ± 0.22	0.17 ± 0.11	153.7 ± 50.2	4.9 ± 1.4	2.65 ± 1.27	78.1 ± 4.65
8	9	29.5 ± 9.7	189.4 ± 52.2	0.38 ± 0.36	0.17 ± 0.10	140.6 ± 57.5	5.9 ± 1.8	3.09 ± 1.33	79.6 ± 5.93
9	9	27.8 ± 7.9	187.6 ± 41.9	0.33 ± 0.32	0.23 ± 0.16	131.1 ± 39.4	6.1 ± 1.0	3.27 ± 1.48	78.4 ± 7.49
10	5	36.1 ± 18.4	200.4 ± 125.5	0.18 ± 0.07	0.13 ± 0.07	162.7 ± 73.5	6.0 ± 3.1	1.90 ± 0.78	73.7 ± 8.50

microwave digested (Milestone, MLS-1200) with concentrated nitric acid (Merk, GR grade). The samples were then filtered and measured by Atomic Absorption Spectrometry (Hitachi Z8000 with a graphite furnace) (Lee *et al.*, 1996). Due to high sensitivity of instrument, the digested samples were diluted with 5 times of ultra pure (metal free) water for copper, lead, cadmium, iron, manganese analysis and 50 times of dilution for zinc. Standard oyster tissue (SRM 1566a) obtained from National Institute of Standards and Technology, USA, was analyzed along with unknown samples. This practice was used as a quality control measure for sample analysis. The range of mean recoveries of trace metal analysis for SRM 1566a was 88.9 to 102.8% (Tab. II). Sediment samples were homogenized by a glass rod and wet sieved through 1000 μm (Teflon, GilsonTM) before metal analysis. About half of the samples collected were wet sieved into two grain size fractions, < 1000 μm (total) and < 63 μm (silt and clay). Samples were then dried, 80°C for at least 12 hours, microwave digested with mixture of acids (HNO_3 , HF and H_2O_2). Samples were filtered (0.4 μm , 4.7 mm; Poretice) and metal concentrations were measured by AAS (Lee *et al.*, 1998). The dilution factors of sediment analysis were five-fold for copper, zinc, lead, cadmium, manganese and five hundred-fold for iron. Standard sediment materials (SRM 1646a obtained from National Institute of Standards and Technology, USA and MESS-2 obtained from National Research Council of Canada) were used for quality control shown in Table II. The ranges of mean recoveries of trace metal analysis for MESS-2 and SRM 1646a were 93.6 to 108.3% and 87.8 to 108.1%, respectively. Volatile fractions of sediments were analyzed by ignition, at 550°C, with weight loss measurement method (LOI, loss on ignition). Water was purified using Milli RO Plus / Milli Q water purification device.

RESULTS AND DISCUSSION

The *p* value derived from statistical analysis for pH (0.60); temperature (0.86); salinity (0.37), indicates that there were no significant differences between stations in the north and south of Ann-ping. The means of these environmental parameters are 8.18 ± 0.12 , $27.1 \pm 3.4^\circ\text{C}$ and $3.11 \pm 0.2\%$ for pH, temperature and salinity,

TABLE II Analysis results of reference materials of sediment and oyster

Reference material	Values	n	Cu	Zn	Pb	Cd	Mn	Fe	Hg
Sediment	Certified		39.0 ± 2.0	172.0 ± 16.0	21.9 ± 1.2	0.24 ± 0.10	365.0 ± 21.0		0.092 ± 0.009
MESS-2	Measured	18	38.2 ± 1.2	169.6 ± 10.0	20.5 ± 0.95	0.26 ± 0.06	363.7 ± 4.6	504.7 ± 16.5	0.093 ± 0.007
Sediment	Certified		10.01 ± 0.34	48.9 ± 1.6	11.7 ± 1.2	0.148 ± 0.007	234.5 ± 2.8	2.008 ± 0.039	0.04
SRM 1646a	Measured	27	10.8 ± 1.0	46.9 ± 2.1	11.9 ± 1.4	0.16 ± 0.02	205.8 ± 19.9	2.00 ± 0.08	0.039 ± 0.004
Oyster	Certified		66.3 ± 4.3	830 ± 57	0.371 ± 0.014	4.15 ± 0.38	12.3 ± 1.5	539 ± 1.5	0.0642 ± 0.067
SRM 1566a	Measured	38	65.8 ± 2.5	844.5 ± 33.9	0.376 ± 0.113	3.69 ± 0.69	11.8 ± 2.2	537.8 ± 15.4	0.066 ± 0.006

respectively. Table I lists the average metal concentrations found in oysters collected from the Ann-ping mariculture ground. The mean oyster concentrations are not significantly different between stations in the north and south of Ann-ping, with p -value (equal) of 0.28 using the Mann-Whitney test (Hollander and Wolfe, 1973). The mean copper and zinc concentrations in Ann-ping were 29.6 and 185.2 $\mu\text{g g}^{-1}$ wet weight. Compared with the concentration of copper in green oyster samples detected in 1986 incidence (with a peak concentration at 880 $\mu\text{g g}^{-1}$, wet weight in Charting coast), the oyster copper concentrations found in Ann-ping were relatively lower. Nevertheless, our recent analysis (Lee *et al.*, 1996) demonstrated that the average copper concentration of adult oysters collected from the Charting coastal area was around 60 $\mu\text{g g}^{-1}$ wet weight. Our results shown in Figure 2 indicated that the monthly averaged oyster copper concentrations had an increasing trend, and a Kendall test (Hollander and Wolfe, 1973) with α value less than 0.01 and 0.05 for stations in the north and south of Ann-ping, from 20 $\mu\text{g g}^{-1}$ on October in 1994 to 50 $\mu\text{g g}^{-1}$ on December, 1996. This increasing trend showed that a possible pollution source could provide a potential problem in the mariculture ground.

In order to compare the metal concentrations collected from Ann-ping area with other raising grounds, Table III lists the mean oyster copper and zinc concentrations from all the major oyster raising grounds such as Bai-men, John-juin, Chi-gu and Dah-peng Wan, which are currently active in the southwestern Taiwan. Oysters from Ann-ping had approximately the same concentration of copper and zinc concentrations as oysters from the adjacent raising grounds, Bai-men, John-juin and Chi-gu. The Dah-peng-wan (a bay in Ping-tung county), produced oysters with the lowest copper concentrations detected which averaged as low as $22.2 \pm 14.9 \mu\text{g g}^{-1}$ wet weight.

The results of sediment analysis in Ann-ping ground during the study period are shown in Table IV. Two fractions of sediments with grain sizes of $< 63 \mu\text{m}$ and $< 1000 \mu\text{m}$ were analyzed. There was no significant difference observed in their copper concentrations between those two fractions. Mann-Whitney test (Hollander and Wolfe, 1973) result showed that p value was 0.92 for these two fractions of sediments. In order to estimate the anthropogenic enrichment of metals in sediment, Daskalakis's method (1995) using iron concentration

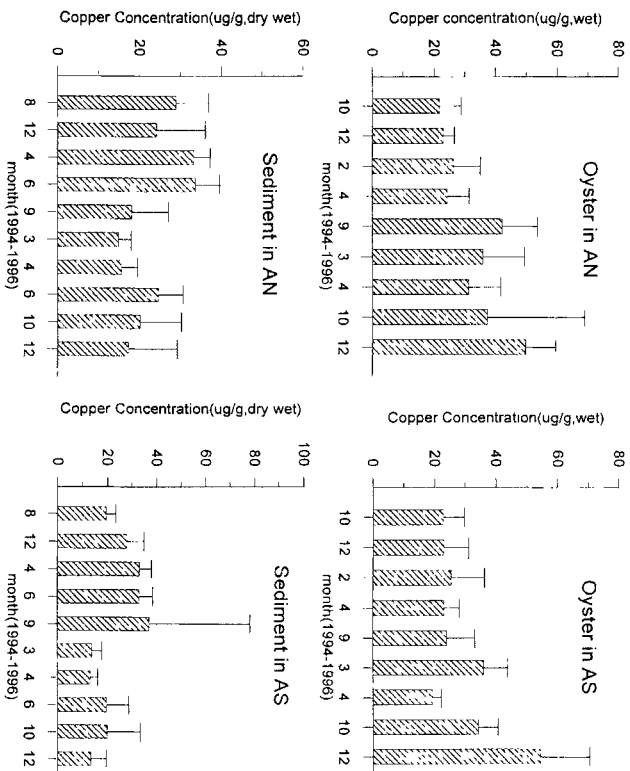


FIGURE 2 Seasonal variation of oyster and sediment copper concentration in Ann-ping mariculture ground.

TABLE III Mean concentrations (mg kg^{-1} , dry weight) of trace metal in sediments collected from the Ann-ping mariculture ground (1994 - 1996)

Station	n	Cu	Zn	Pb	Cd	Fe(%)	Mn	Median size(μm)
1	11	26.8 \pm 9.5 (38.7 \pm 6.0)*	104.2 \pm 42.5 (151.7 \pm 22.8)	18.8 \pm 9.0 (18.3 \pm 2.9)	0.28 \pm 0.29 (0.83 \pm 0.65)	1.86 \pm 0.89 (1.58 \pm 0.11)	287.5 \pm 68.1 (383.3 \pm 127.5)	159.9 \pm 70.5 (28.0 - 233.6)**
2	11	25.2 \pm 8.9 (32.3 \pm 13.1)	96.6 \pm 29.5 (136.3 \pm 22.8)	15.4 \pm 6.6 (20.3 \pm 0.6)	0.42 \pm 0.44 (0.63 \pm 0.40)	1.84 \pm 0.39 (1.69 \pm 0.29)	297.1 \pm 63.3 (331.7 \pm 49.0)	142.7 \pm 88.0 (6.7 - 299.6)
3	8	28.4 \pm 22.3 (28.3 \pm 5.5)	97.8 \pm 47.2 (119.5 \pm 28.0)	16.3 \pm 6.2 (19.7 \pm 1.5)	0.28 \pm 0.29 (1.03 \pm 0.45)	1.80 \pm 0.40 (1.37 \pm 0.05)	306.3 \pm 82.8 (284.7 \pm 14.0)	140.0 \pm 69.6 (14.6 - 221.5)
4	8	18.9 \pm 6.2 (28.5 \pm 5.6)	102.6 \pm 36.9 (115.3 \pm 15.2)	20.6 \pm 9.4 (17.3 \pm 6.4)	0.19 \pm 0.15 (0.90 \pm 0.30)	2.00 \pm 0.60 (1.57 \pm 0.48)	269.1 \pm 39.3 (319.3 \pm 85.7)	106.2 \pm 85.5 (6.6 - 270.8)
5	8	25.1 \pm 15.0 (32.7 \pm 2.5)	92.1 \pm 30.3 (129.7 \pm 29.4)	18.9 \pm 3.7 (15.3 \pm 5.0)	0.49 \pm 0.64 (1.13 \pm 0.32)	2.10 \pm 0.87 (1.93 \pm 0.20)	285.6 \pm 78.3 (212.7 \pm 55.9)	117.0 \pm 52.7 (20.6 - 212.6)
6	10	23.5 \pm 9.6 (28.9 \pm 8.6)	102.9 \pm 40.0 (136.3 \pm 128.9)	17.1 \pm 8.6 (15.0 \pm 5.0)	0.36 \pm 0.45 (2.00 \pm 0.15)	2.00 \pm 0.57 (1.95 \pm 0.21)	293.2 \pm 65.1 (209.2 \pm 26.6)	124.0 \pm 94.1 (9.7 - 320.0)
7	8	20.3 \pm 6.5 (34.7 \pm 8.4)	86.4 \pm 22.4 (145.3 \pm 29.1)	23.8 \pm 12.5 (16.3 \pm 4.0)	0.35 \pm 0.32 (0.97 \pm 0.57)	2.14 \pm 0.76 (1.61 \pm 0.13)	276.5 \pm 94.0 (250.3 \pm 88.6)	133.9 \pm 67.6 (6.9 - 241.2)
8	14	32.5 \pm 21.0 (32.2 \pm 7.7)	96.8 \pm 35.0 (124.8 \pm 25.8)	16.9 \pm 5.3 (19.1 \pm 4.1)	0.56 \pm 0.39 (1.04 \pm 0.37)	1.69 \pm 0.35 (1.48 \pm 0.40)	285.2 \pm 148.7 (278.6 \pm 65.6)	134.0 \pm 36.2 (85.5 - 219.5)
9	11	23.1 \pm 9.3 (40.3 \pm 7.5)	94.6 \pm 37.7 (149.2 \pm 4.8)	17.2 \pm 4.5 (23.7 \pm 5.5)	0.35 \pm 0.41 (1.20 \pm 1.20)	2.01 \pm 0.43 (1.48 \pm 0.38)	228.9 \pm 30.3 (241.1 \pm 54.2)	137.1 \pm 18.2 (117.4 - 175.4)
10	11	19.8 \pm 7.5 (32.3 \pm 9.8)	93.9 \pm 40.2 (106.1 \pm 17.4)	17.4 \pm 3.2 (20.7 \pm 1.2)	0.27 \pm 0.22 (1.23 \pm 0.68)	1.98 \pm 0.51 (1.53 \pm 0.34)	246.9 \pm 54.2 (345.3 \pm 159.4)	145.7 \pm 41.4 (80.0 - 211.1)
Ontario, Canada***								
Severe Effect level	110	820	250	10				
Lowest Effect level	16	120	31	0.6				
REF _M F _c	2.35 \pm 0.57	2.13 \pm 0.23	1.10 \pm 0.11	1.61 \pm 0.57				

* mean concentration of trace metal in < 63 μm grain size fraction sediment.

** range of the median size in samples collected.

*** bulk fraction (< 1000 μm) is used (Bonnievie *et al.*, 1994).

TABLE IV Mean concentrations of copper and zinc in oysters collected from raising areas in southwestern Taiwan ($\mu\text{g g}^{-1}$, weight)

Station	n	Cu	Zn
AN(3/94-12/96)	66	31.1 \pm 14.8(15.3-101.5)	184.9 \pm 38.3(99.5-265.6)
AS(10/94-12/96)	44	28.1 \pm 12.5(10.9-75.3)	185.5 \pm 57.3(103.4-417.9)
Bai-men* (10/95-12/96)	35	32.1 \pm 14.2(12.4-77.2)	156.7 \pm 63.7(59.1-343.8)
John-juin* (10/95-12/96)	35	30.5 \pm 11.9(15.0-65.4)	192.5 \pm 66.9(57.6-315.5)
Chi-gu* (10/95-1/97)	35	31.1 \pm 13.7(11.4-65.6)	140.5 \pm 57.9(40.8-293.8)
Dah-peng wan* (1/94-1/97)	16	22.2 \pm 14.9(6.3-63.2)	133.2 \pm 49.2(70.9-222.6)

* Lee, 1997.

to normalize the sediment contamination was employed, the relative enrichment factor was calculated (Lee *et al.*, 1998). Daskalakis and O'Connor (1995) formula used for the calculation is shown as follows.

$$REF_{Fe}^M = \frac{[M]_X}{[Fe]_X} \cdot \frac{[M]_{BS}}{[Fe]_{BS}}$$

where BS denotes the background station. According to our results, the REF_{Fe}^M for copper and zinc in sediments from Ann-ping is 2.35 and 2.13. The concentrations of copper, zinc, lead and cadmium in sediments from Ann-ping were close to or exceeded the Canadian Lowest Effect Levels concentration, which is 16, 120, 31 and $0.6 \mu\text{g g}^{-1}$, respectively (Bonnievie *et al.*, 1994).

Table V lists the average copper and zinc concentrations in sediments collected from other raising grounds along the southwestern Taiwan coast. The metal concentrations of sediments collected from Ann-ping and John-Juin are higher than those collected from Bai-men and Chi-gu.

Interestingly, elements of copper, zinc, iron and manganese were significantly correlated in oysters and sediment as shown in Tables VI and VII. The best correlation between elements in sediments and oysters was between copper and zinc (with correlation coefficients of 0.496 for sediment and 0.638 for oyster). The copper and zinc coefficients found in Charting coast (Lee *et al.*, 1996) were 0.69 for sediment and 0.91 for oyster. This similarity in metal accumulation pattern might have lead to the disaster, as in the 1986 green oyster incidence, to be repeated in the Ann-ping area if the pollution sources were not to be controlled.

To minimize possible destruction of the bamboo complex used for hanging oyster strings during the typhoon season, the oyster raising season in this area was from September to May. As Figure 3 depicted, the yearly (from September to May) averaged copper concentration of oysters collected from the north of Ann-ping increased in the time span of study. However, there were no corresponding increasing trends detected in the sediment samples. Our results showed that the copper concentration ($\mu\text{g g}^{-1}$, wet weight) in oyster soft part was increased (Kendall test (Hollander *et al.*, 1973) with α value less than

TABLE V Mean concentration of copper and zinc in sediments collected from raising areas in southwestern Taiwan ($\mu\text{g g}^{-1}$, dry weight)

Station	n	Cu	Zn
AN(3/94-12/96)	83	25.0 \pm 11.9(3.8-76.0)	99.5 \pm 35.2(35.0-190.0)
AS(10/94-12/96)	56	24.7 \pm 13.6(7.5-98.8)	101.6 \pm 34.7(38.8-185.0)
Bai-men* (10/95-12/96)	35	14.7 \pm 5.9(2.5-27.5)	74.5 \pm 28.4(33.8-147.5)
John-juin* (10/95-12/96)	35	29.5 \pm 18.2(8.8-73.8)	136.6 \pm 122.6(33.8-687.5)
Chi-gu* (10/95-1/97)	35	17.9 \pm 9.7(3.8-47.5)	95.9 \pm 60.7(40.0-355.0)
Dah-peng wan* (1/94-1/97)	11	28.5 \pm 2.8(22.5-31.3)	113.6 \pm 9.5(97.5-125.8)

* Lee, 1997.

TABLE VI Correlation matrix of trace metals in sediments in Ann-ping area ($n = 143$)

	<i>Cu</i>	<i>Zn</i>	<i>Pb</i>	<i>Cd</i>	<i>Fe</i>	<i>Mn</i>	<i>r</i>	<i>LOI</i>
<i>Cu</i>	1							
<i>Zn</i>	0.58*	1						
<i>Pb</i>	0.22*	0.22*	1					
<i>Cd</i>	0.35*	0.44*	-0.01	1				
<i>Fe</i>	-0.08	-0.06	0.23*	-0.36	1			
<i>Mn</i>	0.28*	0.22*	0.34*	0.04	0.24*	1		
<i>Size</i>	-0.33*	-0.42	-0.13	-0.13	-0.10	-0.08	1	
<i>LOI</i>	0.27	0.38	0.23	0.20	0.05	0.22	-0.37	1

* Significance level, 0.01.

TABLE VII Correlation matrix of trace metals in oysters in Ann-ping area ($n = 130$)

	<i>Cu</i>	<i>Zn</i>	<i>Pb</i>	<i>Cd</i>	<i>Fe</i>	<i>Mn</i>
<i>Cu</i>	1					
<i>Zn</i>	0.64*	1				
<i>Pb</i>	-0.27*	-0.04	1			
<i>Cd</i>	-0.17	-0.09	0.59*	1		
<i>Fe</i>	0.56*	0.46*	-0.07	0.04	1	
<i>Mn</i>	0.26*	0.43*	0.28*	0.30*	0.45*	1

* Significance level, 0.01.

0.05) from 21.3 ± 4.1 in 1993; 24.1 ± 6.8 in 1994; 36.8 ± 11.9 in 1995 to $43.9 \pm 23.1 \mu\text{g g}^{-1}$, wet weight, in 1996 raising season. This phenomenon was also observed in the monthly averaged concentrations for both samples collected either in the north or south of Ann-ping mariculture ground as shown in Figure 2.

CONCLUSIONS

Results in this work demonstrate that the copper concentrations in oysters collected from Ann-ping mariculture area increased from 1994 and 1996. In addition, the level of oyster copper concentration in Ann-ping was even approaching that found in the Charting coast where the oyster raising is prohibited after the green oyster incidence in 1986. Further investigation is needed to determine the possible copper pollution sources which may cause the pollution problem in Ann-ping mariculture ground.

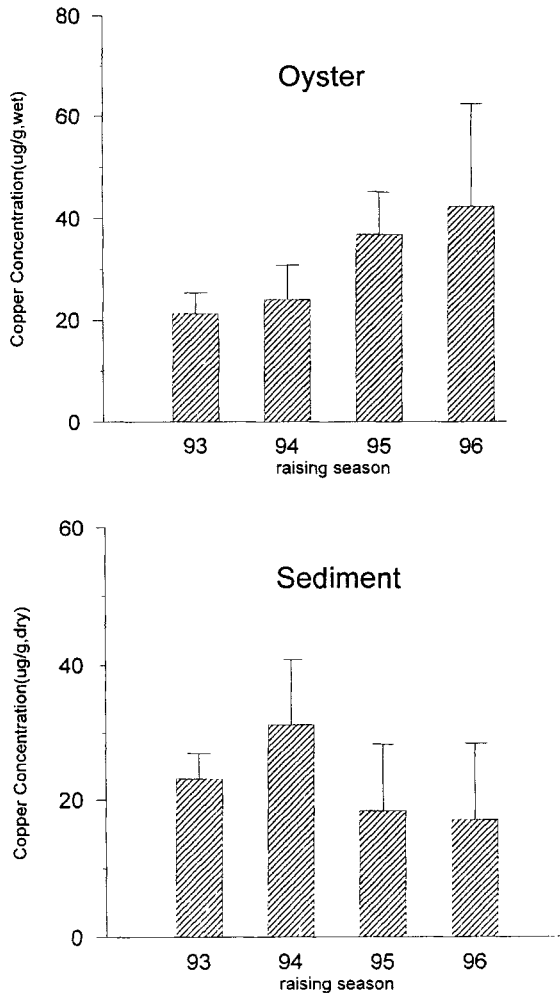


FIGURE 3 Comparison of raising season averaged copper concentration in Northern Ann-ping (AN) oyster and sediment raising area (raising season as defined in text).

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References

- Bewers, J. M. and Yeats, P. A. (1989) Transport of river-derived trace metals through the coastal zone. *Netherlands Journal of Sea Research*, **23**, 359–368.
- Bonnevie, N. L., Huntly, S. L., Found, B. W. and Wenning, R. J. (1994) Trace metal contamination in surficial sediments from Newark Bay, New Jersey. *The Science of Total Environment*, **144**, 1–16.
- Daskalakis, K. D. and O'Connor, T. P. (1995) Normalization and elemental sediment contamination in the coastal United States. *Environ. Sci. Technol.*, **29**, 470–477.
- Everaarts, J. M. (1989) Heavy metals (Cu, Zn, Cd, Pb) in sediment of the Java Sea, estuarine and coastal areas of east Java and some deep-sea areas. *Netherlands Journal of Sea Research*, **23**, 403–413.
- Han, B.-C. and Hung, T.-C. (1990) Green oyster caused by copper pollution on the Taiwan coast. *Environmental Pollution*, **65**, 347–362.
- His, E., Seaman, N. L. and Beiras, R. (1997) A simplification of the bivalve embryogenesis and larval development bioassay method for water quality assessment. *Wat. Res.*, **31**, 351–355.
- Hollander, M. and Wolfe, D. A. (1973) *Nonparametric Statistic Methods*, Wiley, New York.
- Hung, J.-J. (1990) Transport and behavior of trace metals in the Tsengwen river and estuary. *Terrestrial, Atmospheric and Oceanic*, **1**, 275–295.
- Hung, J.-J. and Huang, C.-C. (1990) Geochemical distribution and transport of trace metals in the Kaoping riverine, estuarine and coastal waters. *Proc. Natl. Sci. Council. ROC(A)*, **14**, 410–421.
- Juracic, M., Vitturi, L. M., Rabitti, S. and Rampazzo, G. (1987) The role of suspended matter in the biological cycles in the Adige River estuary (Northern Adriatic Sea). *Estuarine, Coastal and Shelf Science*, **24**, 349–362.
- Lee, C.-L., Chen, H.-L. and Chuang, M.-I. (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.
- Lee, C.-L. (1997) unpublished data.
- Lee, C.-L., Fang, M.-D. and Hsieh, M.-T. (1998) Characterization and distribution of metals in surficial sediments in southwestern Taiwan. *Mar. Pollution Bull.*, in press.
- Marcintic, D., Nurnberg, H. W. and Branica, M. (1986) Bioaccumulation of heavy metals by bivalves from Limski Kanal (North Adriatic Sea) II Copper distribution between oysters, *Ostrea edulis*, and ambient water. *Marine Chemistry*, **18**, 299–319.
- Mo, C. and Nielsm, B. (1991) Variability in measurements of zinc in oysters, *C. virginica*. *Mar. Poll. Bull.*, **22**, 522–525.
- van Haren, R. J. F., Schepers, H. E. and Kooijman, S. A. L. M. (1994) Dynamic energy budgets affect kinetics of xenobiotics in the marine mussel *Mytilus edulis*. *Chemosphere*, **29**, 163–189.
- Windom, H., Wallage, G., Smith, R., Dudek, N., Maeda, M., Dulmage, R. and Storti, F. (1983) Behavior of copper in southeastern United States Estuaries, *Marine Chemistry*, **12**, 183–193.
- Zamuda, C. D. and Sunda, W. G. (1982) Bioavailability of dissolved copper to the America oyster *Crassostrea virginica*. I. Importance of chemical speciation. *Marine Biology*, **66**, 77–82.