

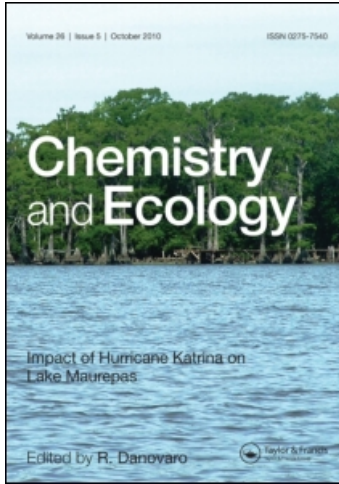
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RESTORATION OF THE MARINE ECOLOGICAL ENVIRONMENT ALONG THE CHARTING COASTAL AREA: CHEMICAL STUDIES

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The Charting coastal area is an important mariculture and fishery ground in Taiwan. Due to copper and organic pollution, mariculture has been prohibited by the Taiwan Government since the first case of green oysters appeared in this area in 1986. Growing algae in the polluted environment may improve water quality and re-establish the marine ecosystem. Since September, 1993, various algal species (such as *Ulva*, *Sargassum*, *Grateloupia*, *Halymenia*, *Galaxaura*, etc.) have been cultured in the Charting coastal area. Although some of the cultivated algae were damaged by a typhoon in August, 1994, the environmental and ecological effect of the cultivated algae is still evident. The non-biological and biological components surveyed include: salinity, temperature, dissolved oxygen, biochemical oxygen demand (BOD), particulate organic carbon (POC), nutrients, heavy metals and chlorophyll-a, adenosine triphosphate, primary productivity, species composition and intra-specific relationships of phyto- and zoo-plankton. The purpose of this paper is to evaluate the possible impact before and during culture of large algae on the heavy metals and organic pollution of water and sediments over the period August 1992 to May 1995. The results indicate that during algal culture, higher levels of dissolved oxygen ($6.1\text{--}7.6\text{ mg l}^{-1}$) with lower BOD ($0.4\text{--}2.3\text{ mg l}^{-1}$) and POC ($20.6\text{--}948\text{ }\mu\text{g l}^{-1}$) were found. The concentrations of copper ($0.02\text{--}4.74\text{ }\mu\text{g l}^{-1}$), zinc ($0.90\text{--}12.3\text{ }\mu\text{g l}^{-1}$), lead ($<0.05\text{--}1.47\text{ }\mu\text{g l}^{-1}$) and cadmium ($<0.02\text{--}0.14\text{ }\mu\text{g l}^{-1}$) also decreased. As a result of these positive findings, the cultivation area of algae was expanded in 1995. We expect that with extended algal cultivation, inorganic and organic pollutants will be minimized, and that the marine ecosystem and oyster mariculture will be restored.

KEY WORDS: Heavy metals, organic pollutants, mariculture, oysters, water quality, marine ecosystem recovery, Charting sea.

INTRODUCTION

The coast along Kaohsiung to Tainan, including the Charting coastal area, is a very important area for fishing and for oyster mariculture in Taiwan. In recent decades the dense population and expansion of heavy industry has resulted in domestic and industrial wastes, mostly without any treatment, being discharged via the Erhjin Chi river to the coastal environment (EPA/ROC, 1990). The first incident of green oysters (*Crassostrea gigas*) in the Charting area was observed in January 1986, and oyster mortality was reported three months later (Su *et al.*, 1986). Immediately after the incident, mariculture was prohibited by the Government to ensure the protection of human health.

The cause of green oysters was identified as copper combined with organic waste pollution (Hung *et al.*, 1987, 1989; Hung, 1988). The copper content in oysters was extremely high – 2100 $\mu\text{g g}^{-1}$ and 4400 $\mu\text{g g}^{-1}$ dry weight in January 1986 and January 1989 (Han and Hung, 1990a). The copper species present were characterized as complexes with inorganic and organic anions, labile and non-labile, polar and non-polar, and free ion, particulate and dissolved forms in the mariculture water. In sediments these were: exchangeable, carbonate, readily/moderately reducible, organic with sulphide and detritus with minerals. The extremely high concentrations of copper in water (739 $\mu\text{g l}^{-1}$) and sediments (2130 mg kg^{-1}) and of organic matter in water (15 mg l^{-1}) and sediment (81 mg g^{-1} , dry wt.) were found to play an important role in the greening and mortality of oysters (Hung *et al.*, 1989, 1993, 1995; Han and Hung, 1990a, 1990b; Hung and Tsai, 1992). Algae are able to accumulate various pollutants such as heavy metals and organic matter from sea water (Largo and Ohno, 1993; Aziz and Jern, 1994). Growing algae in a polluted environment may clean the water and recreate the marine ecosystem, protecting maricultural activity and the fishery resource.

A three year programme of mitigation of the polluted marine environment along in western (Kaohsiung–Tainan) mariculture area was initiated in August 1992. In the first year, the background components such as salinity, temperature, nutrients, heavy metals, organic matter were monitored to find the most suitable season and locations for growing large algae (such as *Sargassum* spp., *Grateloupia flicina*, *Halymenia microcarpa*, *Padia arborescens*, *Pterocladia capillacea*, *Ulva lactuca*, *U. arasaki*) were first cultured in the laboratory and then transferred to the field study area. By September 1993, about 10 kg of various algal species had been cultured in the Shinda area (22°49' 33"N, 122°12' 15"E; stations X1–X4, Fig. 1). Although some of the cultured algae were damaged by a typhoon in August 1994, environmental and ecological studies have been continued. The purpose of this paper is to evaluate the possible effect of the algal cultivation on the heavy metal and organic pollution in water and sediments of the area before and during algal culture, from August 1992 to May 1995.

EXPERIMENTAL METHODS

Water and sediment samples were collected from the Erhjin Chi and its estuary, as well as the Charting coastal area, in twelve cruises during the period September 1992 to May 1995. The sampling locations and dates are shown in Table I and Figure 1. Water samples were taken at different depths (0, 3, 10 and 20 metres) in non-metallic Niskin bottles aboard the research vessel "Oceanic Research 1" or fishing boats. Surface sediment was collected by dredging and then acidified to $\text{pH} < 2$ with nitric acid for analysis of total concentrations of heavy metals. Immediately after collection, water samples were measured for temperature, salinity and pH, and analyzed for chemical components: dissolved oxygen, biochemical oxygen demand (BOD), heavy metals (copper, zinc, lead, cadmium) and nutrients (nitrite, nitrate, phosphate, silicate) as well as ecological parameters (chlorophyll-a, adenosine triphosphate, particulate organic carbon and primary productivity). The methods of analysis, except for heavy metals, have been described previously by Hung *et al.* (1982). The analysis of total

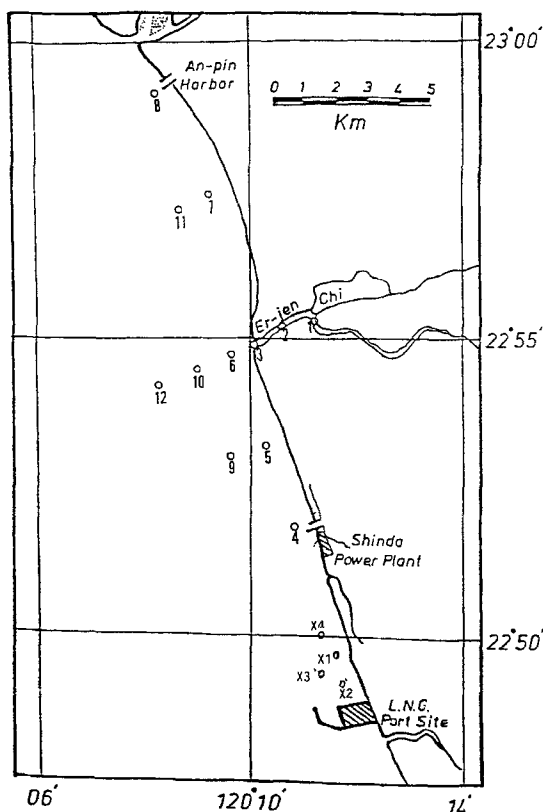


Figure 1 Sampling stations along the Erhjin Chi river (1–2), estuary (3), the Charting coastal (4–12) and the algal cultural areas (X1–X4) in Taiwan (L.N.G.: Liquid Natural Gas).

Table I Research cruises for hydrographical, chemical and ecological studies along the Erhjin Chi (River) and estuary, and in the Charting coastal areas.

Cruise No.	Date Period	Cruise No.	Date Period
1	September 17 to 18, 1992	7	March 3 to 4, 1994
2	December 17 to 18, 1992	8	June 15 to 16, 1994
3	March 17 to 18, 1993	9	September 9 to 10, 1994
4	June 22 to 23, 1993	10	December 29 to 31, 1994
5	September 8 to 9, 1993	11	March 8 to 10, 1995
6	December 30 to 31, 1993	12	May 24 to 25, 1995

concentrations of heavy metals in water and sediments followed the methods described by Su *et al.* (1995) and Hung and Meng (1992). In this paper we discuss only the parameters relevant to heavy metals and organic pollution of the water and sediments.

HYDROGRAPHICAL AND CHEMICAL ANALYSIS OF THE WATER

Along the Charting coastal area, including the Erhjin Chi river and its estuary, temperature varied with season and location. Highest temperature (30.2 to 34.9°C) were generally observed in the river water (stations 1–3) from May to September, and lowest temperatures were found in the coastal sea water (19.6 to 20.3°C) from December to March (Fig. 2A and 2B). However, salinity varied only with location, as expected. High average values for salinity (29.438–34.658‰) were found at coastal stations, and low values (as low as 1.405‰) at river stations (Fig. 3A and 3B). No significant differences in temperature and salinity distributions were found before and during algal culture.

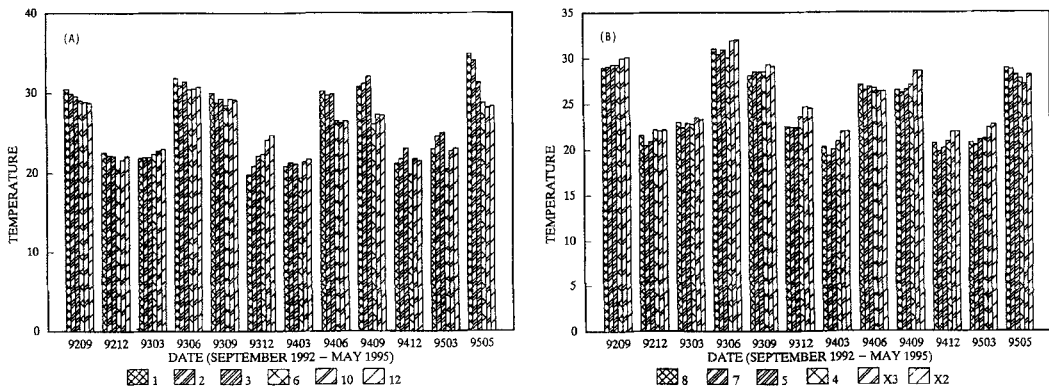


Figure 2 Distribution of temperature (°C) in water collected from the Erhjin Chi (river, 1–2; estuary, 3), Charting coastal (4–12) and the algal cultural (X2–X3) areas (A, Stations 1, 2, 3, 7, 10 and 12; B, Stations 8, 7, 5, 4, X3 and X2).

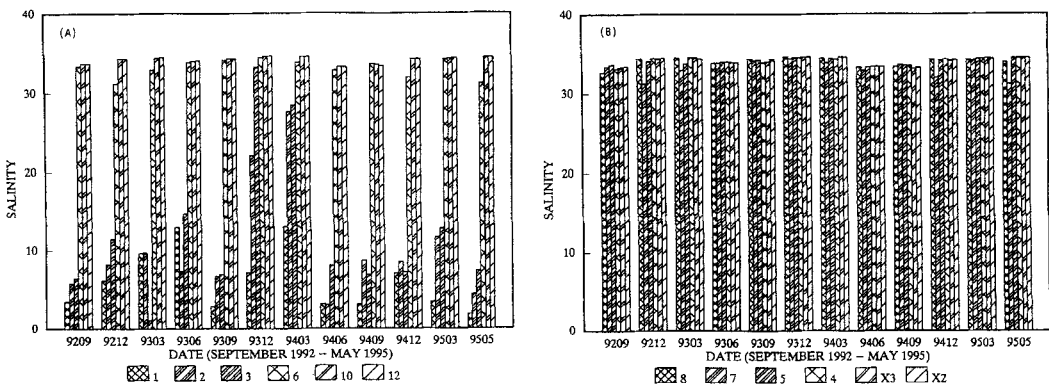


Figure 3 Distribution of salinity (‰) in water collected from the Erhjin Chi (river, 1–2; estuary, 3), Charting coastal (4–12) and the algal cultural (X2–X3) area (A, Stations 1, 2, 3, 6, 10 and 12; B, Stations 8, 7, 5, 4, X3 and X2).

Organic matter pollution is indicated by the levels of dissolved oxygen, biochemical oxygen demand and particulate organic carbon in water. In general, low dissolved oxygen ($0.0\text{--}6.4\text{ mg l}^{-1}$) (Fig. 4A) associated with high BOD ($4.1\text{--}120\text{ mg l}^{-1}$) (Fig. 5A) and POC ($367\text{--}21600\text{ }\mu\text{g l}^{-1}$) (Fig. 6A) were observed at river stations; high values of dissolved oxygen ($2.0\text{--}7.5\text{ mg l}^{-1}$) (Fig. 4B), with low values of BOD ($0.5\text{--}9\text{ mg l}^{-1}$) (Fig. 5B) and POC ($49.6\text{--}1740\text{ }\mu\text{g l}^{-1}$) (Fig. 6B) were found at the Charting coastal stations. Relatively lower dissolved oxygen (Fig. 4C) with higher BOD (Fig. 5C) and POC (Fig. 6C) was found generally in the surface water samples. Relatively lower dissolved oxygen (Fig. 4C with higher BOD (Fig. 5C) and POC (Fig. 6C) were found generally in the surface water samples. However, if we compare these data with those for the stations (X2 and X3) in the algal cultivation area, it is clear that in the latter, higher dissolved oxygen ($6.1\text{--}7.6\text{ mg l}^{-1}$), lower BOD ($0.4\text{--}2.3\text{ mg l}^{-1}$) and POC ($20.6\text{--}948\text{ }\mu\text{g l}^{-1}$) in surface or whole column water were observed since September 1993 during algal culture. This demonstrates that growing algae in the organic polluted area can reduce pollution.

In general, concentrations of heavy metals were extremely high at river stations (maximum values copper $313\text{ }\mu\text{g l}^{-1}$, zinc $803\text{ }\mu\text{g l}^{-1}$, lead $62.4\text{ }\mu\text{g l}^{-1}$, cadmium $9.94\text{ }\mu\text{g l}^{-1}$) compared with those in the coastal stations (copper $0.56\text{--}28.5\text{ }\mu\text{g l}^{-1}$, zinc

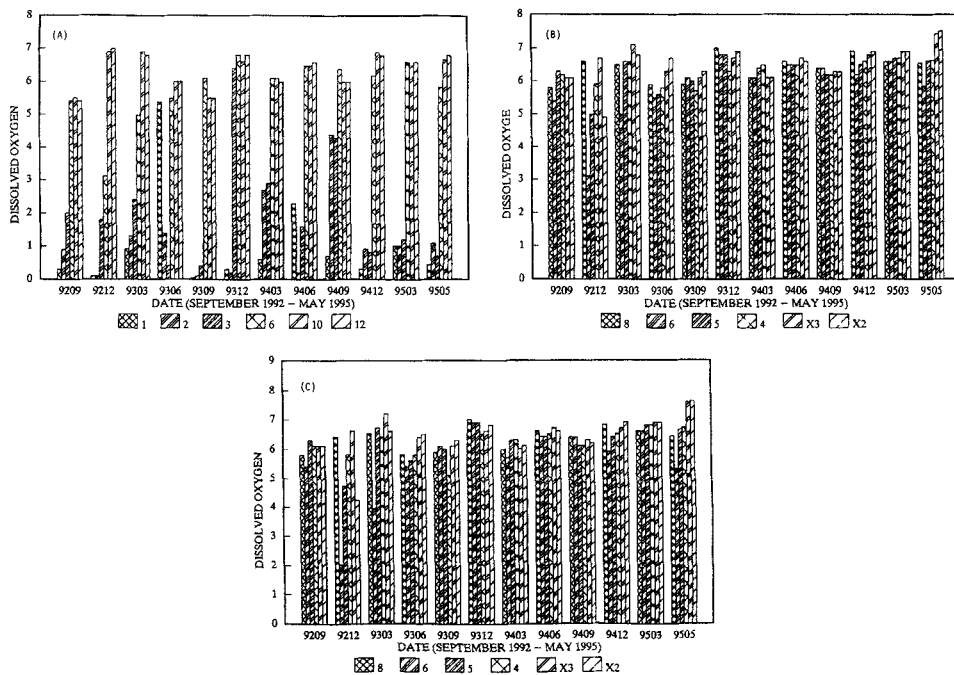


Figure 4 Distribution of dissolved oxygen (mg l^{-1}) in water collected from the Erhjin Chi (river, 1–2; estuary 3), Charting coastal (4–12) and the algal cultural (X2–X3) areas (A and B, Average data; C, Surface data).

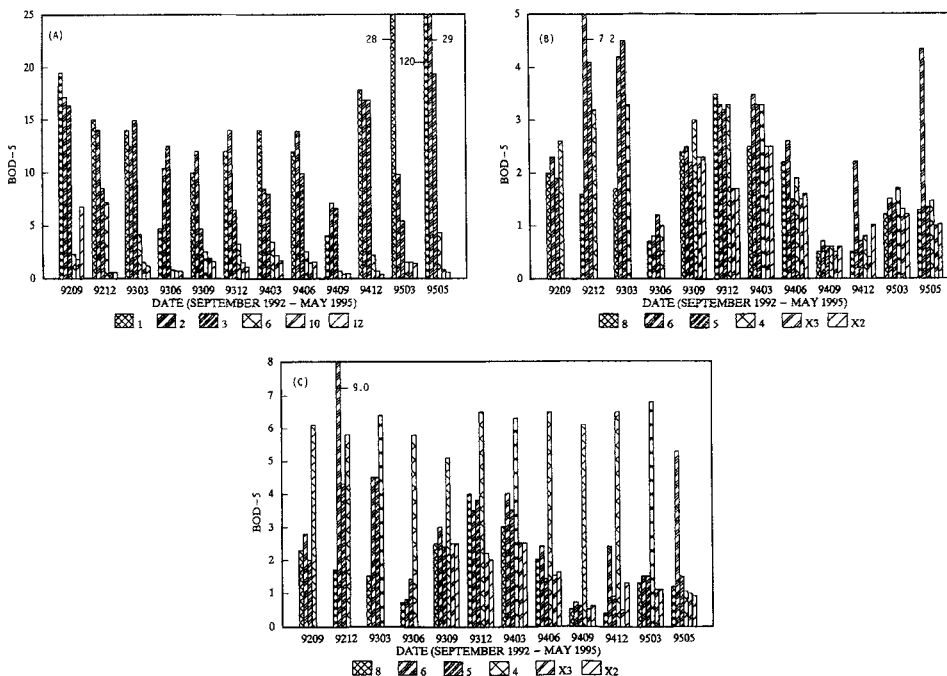


Figure 5 Distribution of biological oxygen demand (BOD, mg l^{-1}) in water collected from the Erhjin Chi (river, 1–2; estuary, 3), Charting coastal (4–12) and the algal cultural (X2–X3) areas (A and B, Average data; C, Surface data).

$0.68\text{--}31.9 \mu\text{g l}^{-1}$, lead $<0.05\text{--}10.6 \mu\text{g l}^{-1}$, cadmium $<0.02\text{--}0.30 \mu\text{g l}^{-1}$). Immediately after the start of algal culture in September 1993, concentrations decreased (copper $0.02\text{--}4.74 \mu\text{g l}^{-1}$, zinc $0.90\text{--}12.3 \mu\text{g l}^{-1}$, lead $<0.05\text{--}1.47 \mu\text{g l}^{-1}$, cadmium $<0.02\text{--}0.14 \mu\text{g l}^{-1}$).

Although ecological data, such as nutrients (nitrite, nitrate, phosphate and silicate), chlorophyll-a, adenosine triphosphate (ATP) and primary productivity are not presented and discussed here, there are significant correlations between environmental and ecological parameters in the Erhjin Chi river as well as in the Charting coastal area. There are good correlations ($r > 0.22$) between primary productivity and chlorophyll-a, phosphate, POC, salinity, dissolved oxygen, silicate, zinc, copper, lead, ATP and pH, and between chlorophyll-a and BOD, phosphate, copper, dissolved oxygen, silicate, zinc, lead, salinity, temperature and POC using averaged data for all depths ($n = 320$) and surface water data only ($n = 136$) (Tab. II). From these results, it may be concluded, in accordance with the findings of Largo and Ohno (1993) and Aziz and Jern (1994) that the growing of algae in a heavy metals and organic polluted area may clean the water and recreate the marine ecosystem, as well as maintaining marine production.

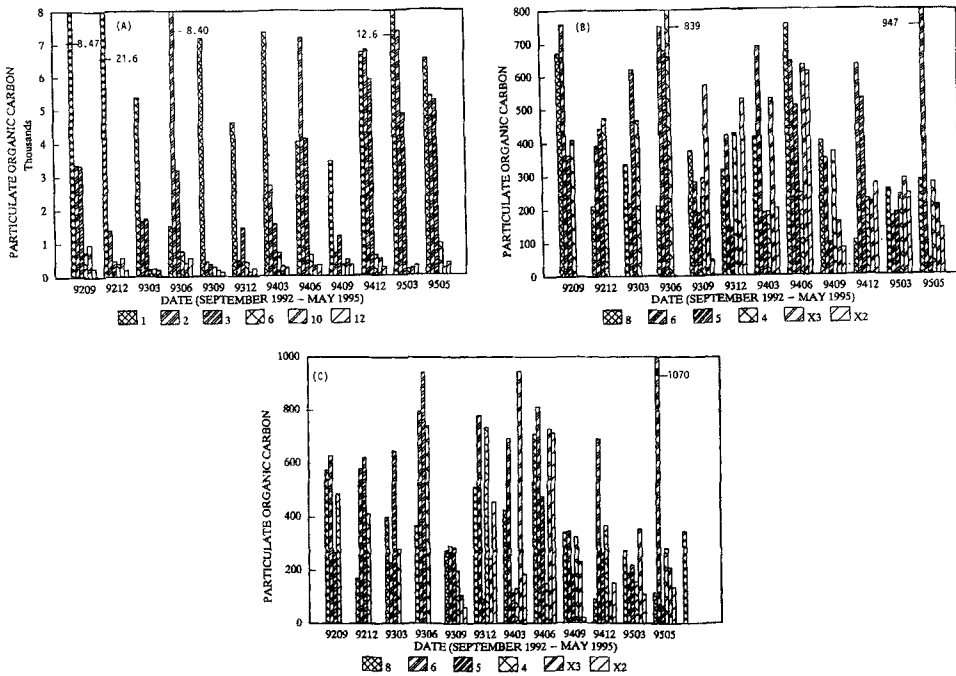


Figure 6 Distribution of particulate organic carbon ($\mu\text{g l}^{-1}$) in water collected from the Erhjin Chi (river, 1–2; estuary, 3), Charting coastal (4–12) and the algal cultural (X2–X3) areas (A and B, Average data; C, Surface water data).

HEAVY METALS IN SEDIMENTS

Further evidence is obtained from the analysis of heavy metals in sediments (Fig. 7–10) in the Erhjin Chi river. Table III shows the extremely high values for concentrations (dry weights) of copper ($240\text{--}3100\ \mu\text{g g}^{-1}$), zinc ($390\text{--}9200\ \mu\text{g g}^{-1}$), lead ($150\text{--}1800\ \mu\text{g g}^{-1}$) and cadmium ($0.6\text{--}13\ \mu\text{g g}^{-1}$) compared with those found in coastal stations. Average values of copper, zinc, lead and cadmium were $9.2 \pm 3.4\ \mu\text{g g}^{-1}$, $65 \pm 7\ \mu\text{g g}^{-1}$, $15 \pm 4\ \mu\text{g g}^{-1}$, and $0.2 \pm 0.2\ \mu\text{g g}^{-1}$, respectively, in coastal sediments. In contrast, in the algal culture area, the average values were very low compared with those of the Charting coast: copper $8.8 \pm 1.0\ \mu\text{g g}^{-1}$, zinc $61 \pm 3\ \mu\text{g g}^{-1}$, cadmium $0.2 \pm 0.0\ \mu\text{g g}^{-1}$. Lead is the exception, $16.5 \pm 5\ \mu\text{g g}^{-1}$, possibly due to oil pollution at station 4 near the Shinda harbour (where there is an oil-fired power plant). The low concentrations in both water and sediments of the algal growing area may be the result of bio-accumulation of heavy metals in the algae. Huang (1993) found that zinc concentrations as high as $3120\ \mu\text{g g}^{-1}$ dry weight were found in the alga *Gracilaria tenuistipitata* var. *liui* cultured for 48 hours in various concentrations of zinc (0.3 to $2.0\ \text{mg l}^{-1}$).

As a result of these positive observations, more algal species have been cultured in the area during the period April 29 to May 25, 1995. We expect that organic and inorganic

Table II Correlation coefficients among the environmental parameters of Erhjin Chai and Charting coastal areas collected from September 1992 to May 1995 (Upper, average data, $n = 320$; Down, surface data, $n = 136$).

St.	Sal	Temp	pH	DO	BOD5	NO2	NO3	PO4	SiO4	Zn	Cd	Pb	Cu	CHL _a	PP	POC	ATP
Sal	1.0000	0.2613**	0.7094**	0.9133**	0.5804**	0.3781**	0.0539	0.6713**	0.8056**	0.5580**	0.3446**	0.4870**	0.4361**	0.3649**	0.4005**	0.8110**	0.6512**
Temp	0.2351**	1.0000	0.1060*	0.1877**	0.2062**	0.1600**	0.0525	0.0209	0.0603	0.0778	0.1137*	0.1250*	0.0492	0.1760**	0.0113	0.1196*	0.0472
pH	0.7383**	0.1179	1.0000	0.7609**	0.3292**	0.5283**	0.0629	0.4489**	0.6449**	0.4228**	0.3740**	0.4402**	0.4745**	0.2072**	0.2768**	0.4411**	0.6108**
DO	0.9145**	0.1653	0.7881**	1.0000	0.5636**	0.4930**	0.0974	0.6810**	0.7967**	0.4828**	0.2188**	0.3592**	0.3890**	0.2458**	0.4071**	0.8021**	0.6364**
BOD5	0.5631**	0.1851*	0.3283**	0.5463**	1.0000	0.2161**	0.3261**	0.5218**	0.4324**	0.2638**	0.0646	0.1558**	0.1074*	0.4620**	0.0904	0.5707**	0.3839**
NO2	0.3714**	0.1733*	0.5426**	0.5060**	0.2079**	1.0000	0.4906**	0.2138**	0.3364**	0.2016**	0.1873**	0.1430**	0.3298**	0.0307	0.0914	0.3362**	0.3659**
NO3	0.0336	0.0729	0.0815	0.0871	0.3069**	0.4796**	1.0000	0.0477	0.0046	0.3670**	0.0766	0.0067	0.1568**	0.0020	0.0963	0.0742	0.1172*
PO4	0.6650**	0.0067	0.4663**	0.6723**	0.5080**	0.2063*	0.0332	1.0000	0.5326**	0.6415**	0.1728**	0.3760**	0.3082**	0.3084**	0.4993**	0.6940**	0.3800**
SiO4	0.7974**	0.0328	0.6667**	0.7933**	0.4129**	0.3309**	0.0149	0.5223**	1.0000	0.3554**	0.1453**	0.2423**	0.2767**	0.2090**	0.3567**	0.7230**	0.7771**
Zn	0.5661**	0.0653	0.4387**	0.4829**	0.2530**	0.1977**	0.0291	0.6390**	0.3490**	1.0000	0.6229**	0.7174**	0.5662**	0.2180**	0.3261**	0.4791**	0.2542**
Cd	0.3459**	0.1112	0.3857**	0.2220**	0.0583	0.1880*	0.0712	0.1721*	0.1440	0.6233**	1.0000	0.9333**	0.6426**	0.1613**	0.1774**	0.1022	0.0630
Pb	0.5043**	0.1460	0.4400**	0.3615**	0.1517	0.1462	0.0101	0.3869**	0.2506**	0.7258**	0.9421**	1.0000	0.6807**	0.2708**	0.2990**	0.2776**	0.1212*
Cu	0.4393**	0.0503	0.4823**	0.3894**	0.0991	0.3340**	0.1529	0.3088**	0.2760**	0.5680**	0.6446**	0.6835**	1.0000	0.2970**	0.3195**	0.3556**	0.3850**
Chl _a	0.2950**	0.2494**	0.1654	0.2277**	0.0328	0.0358	0.0426	0.3306**	0.2276**	0.2258**	0.1610	0.2332**	0.2812**	1.0000	0.7397**	0.2300**	0.1646**
PP	0.4048**	0.0221	0.2341**	0.3885**	0.0727	0.1078	0.0307	0.5113**	0.3599**	0.3250**	0.1712*	0.2738**	0.3037**	0.7139**	1.0000	0.4045**	0.2439**
POC	0.8065**	0.0920	0.4613**	0.7994**	0.5570**	0.3309**	0.0579	0.6885**	0.7152**	0.4758**	0.1017	0.2880**	0.3566**	0.2428**	0.406**	1.0000	0.5689**
ATP	0.6194**	0.1014	0.6307**	0.6055**	0.3517**	0.3613**	0.0880	0.3657**	0.7398**	0.2409**	0.0571	0.1262	0.3943**	0.1896*	0.2515**	1.0000	0.5373**

* $p < 0.05$ ** $p < 0.01$

Table III Heavy metal contents ($\mu\text{g g}^{-1}$) in sediments collected from the Erhjin Chi (river), estuarine and the Charting coastal areas.

Station No.	Cu	Zn	Pb	Cd
September 8, 1993				
1	3,100	9,200	1,800	13
2	610	2,400	410	1.4
3	240	390	150	0.6
4	6.9	72	18	0.1
5	5.7	64	11	0.1
6	17.0	81	17	0.1
7	8.1	66	15	0.2
8	8.7	66	13	0.3
9	9.9	69	7.4	0.1
10	8.1	60	20	0.2
11	9.3	60	15	0.2
12	6.3	53	9.3	0.1
Average	8.8 ± 3.1	66 ± 7	14 ± 4	0.2 ± 0.0
December 30, 1993				
1	1,200	2,900	920	5.3
2	680	2,600	460	1.8
3	240	420	180	1.0
4	7.2	60	28	0.2
5	6.8	65	17	0.2
6	20	80	26	0.3
7	9.1	58	15	0.3
8	9.4	71	14	0.3
9	10.0	64	8.6	0.2
10	7.9	62	19	0.2
11	9.6	63	15	0.1
12	6.6	58	10	0.2
Average	9.6 ± 3.9	64 ± 7	16 ± 5	0.2 ± 0.0
X1	9.6	65	19	0.2
X2	10	60	20	0.2
X3	7.4	58	16	0.2
X4	8.3	62	8.4	0.1
Average	8.8 ± 1.0	61 ± 3	16 ± 5	0.2 ± 0.0

River stations, 1–2; Estuary station, 3; Charting coastal stations, 4–12; Algal cultural stations, X1–X4.

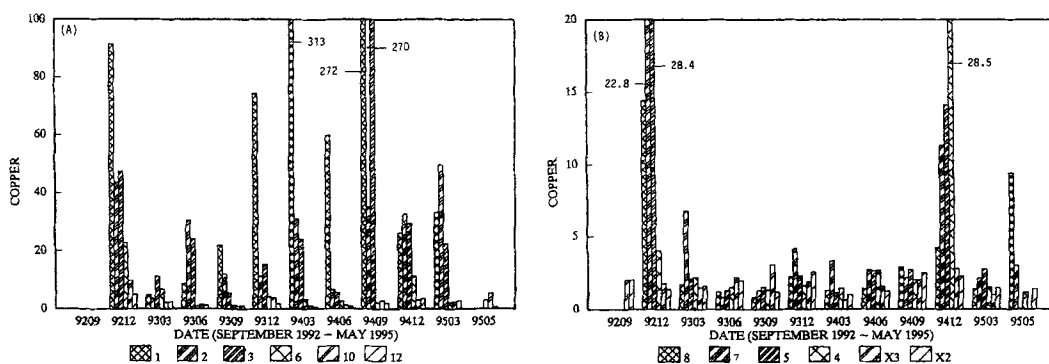


Figure 7 Distribution of copper concentration ($\mu\text{g l}^{-1}$) in water collected from the Erhjin Chi (river, 1–2; estuary, 3), Charting coastal (4–12) and the algal cultural (X2–X3) areas (A, Stations 1, 2, 3, 6, 10 and 12; B, Stations 8, 7, 5, 4, X3 and X2).

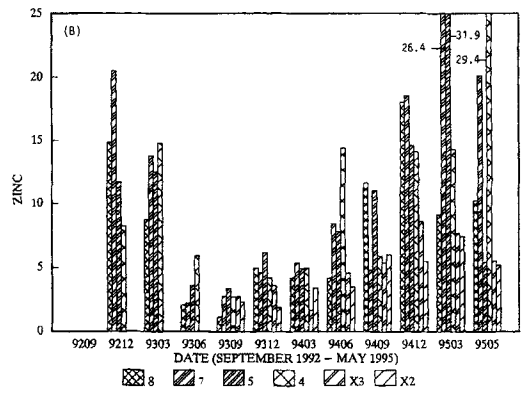
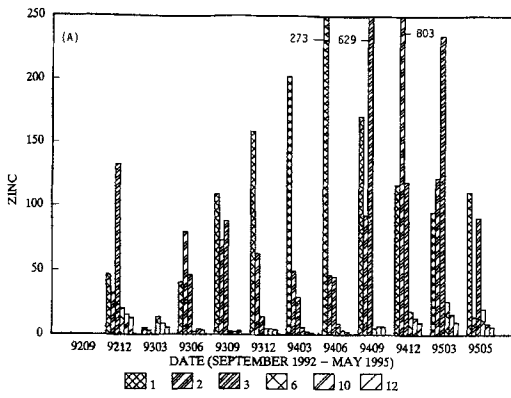


Figure 8 Distribution of zinc concentration ($\mu\text{g l}^{-1}$) in water collected from the Erhjin Chi (river, 1-2; estuary, 3), Charting coastal (4-12) and the algal cultural (X2-X3) areas (A, Stations 1, 2, 3, 6, 10 and 12; B, Stations 8, 7, 5, 4, X3 and X2).

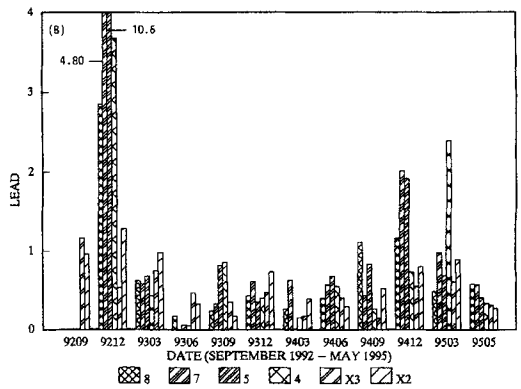
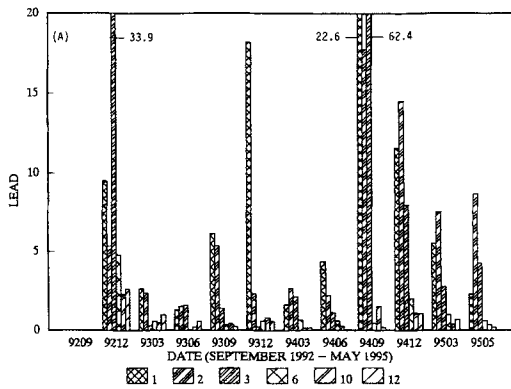


Figure 9 Distribution of lead concentration ($\mu\text{g l}^{-1}$) in water collected from the Erhjin Chi (river, 1-2; estuary, 3), Charting coastal (4-12) and the algal cultural (X2-X3) areas (A, Stations 1, 2, 3, 6, 10 and 12; B, Stations 8, 7, 5, 4, X3 and X2).

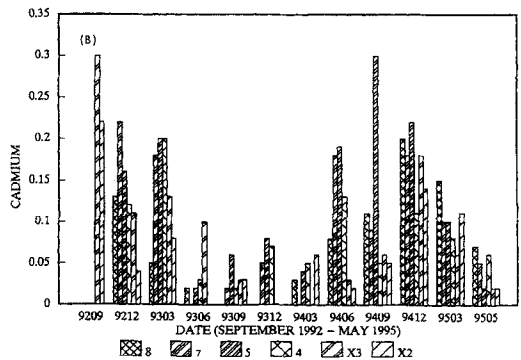
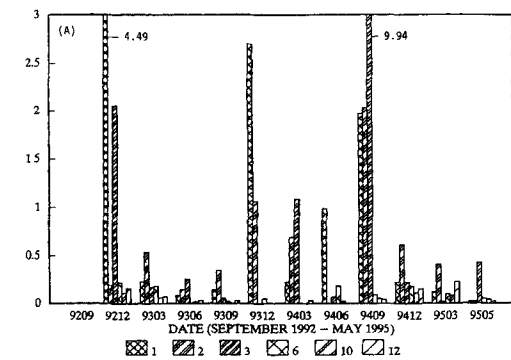


Figure 10 Distribution of cadmium concentration ($\mu\text{g l}^{-1}$) in water collected from the Erhjin Chi (river, 1-2; estuary, 3), Charting coastal (4-12) and the algal cultural (X2-X3) areas (A, Stations 1, 2, 3, 6, 10 and 12; B, Stations 8, 7, 5, 4, X3 and X2).

pollutants will be reduced in the new cultivation area, and that the marine ecosystem will be restored for oyster mariculture.

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