

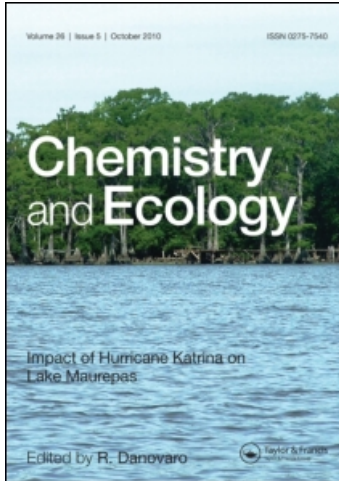
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## Chemistry and Ecology

Publication details, including instructions for authors and subscription information:

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**To cite this Article** Hung, Tsu-Chang and Tsai, Charley C. H.(1992) 'Relationships Between Copper Species and Forms, Hydrographic and Biomass Parameters in the Taiwan Erhjin Chi Coastal Area', *Chemistry and Ecology*, 6: 1, 1 – 17

**To link to this Article:** DOI: 10.1080/02757549208035259

**URL:** <http://dx.doi.org/10.1080/02757549208035259>

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# RELATIONSHIPS BETWEEN COPPER SPECIES AND FORMS, HYDROGRAPHIC AND BIOMASS PARAMETERS IN THE TAIWAN ERHJIN CHI COASTAL AREA

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(20 June 1991)

The first incident of green oysters (*Crassostrea gigas*) was reported in Taiwan in the Charting coastal area in January, 1986 and mortality was reported three months later. The cause of the greened oysters was identified as copper pollution. The copper content of the green oysters was extremely high – 2100 ppm, 2225 ppm, and 4400 ppm dry weight, in January, 1986, February 1987, and January 1989, respectively. In this paper we summarize the seasonal and regional distributions of copper species (complexed by inorganic and organic ligands, labile and non-labile, polar and non-polar) and forms (dissolved and particulate) and the hydrographic and biomass parameters (mainly particulate organic carbon, chlorophyll  $\alpha$ , adenosine triphosphate and primary production) in sea water in the Erhjin Chi coastal area. In general, high concentrations of particulate material (0.24 to 724 ppb) and non-labile organic copper (0.03 to 21.5 ppb) were observed. Low values of polar organic copper ( $<0.02$  to 16.5 ppb) indicated that non-polar organic complexes (0.3 to 20.4 ppb) from man-made organic pollutants were the major complexes in the area studied. On the basis of this data, the cause of greening and mortality in oysters is evaluated in relation to the bioavailable copper (sum of particulate and labile copper) and copper assimilative capacity (detoxicant). Finally, the correlations between the species and forms of copper, hydrographic and biomass parameters are also discussed.

KEY WORDS: Copper species/forms, green oysters, *Crassostrea gigas*, Taiwan coastal sea

## INTRODUCTION

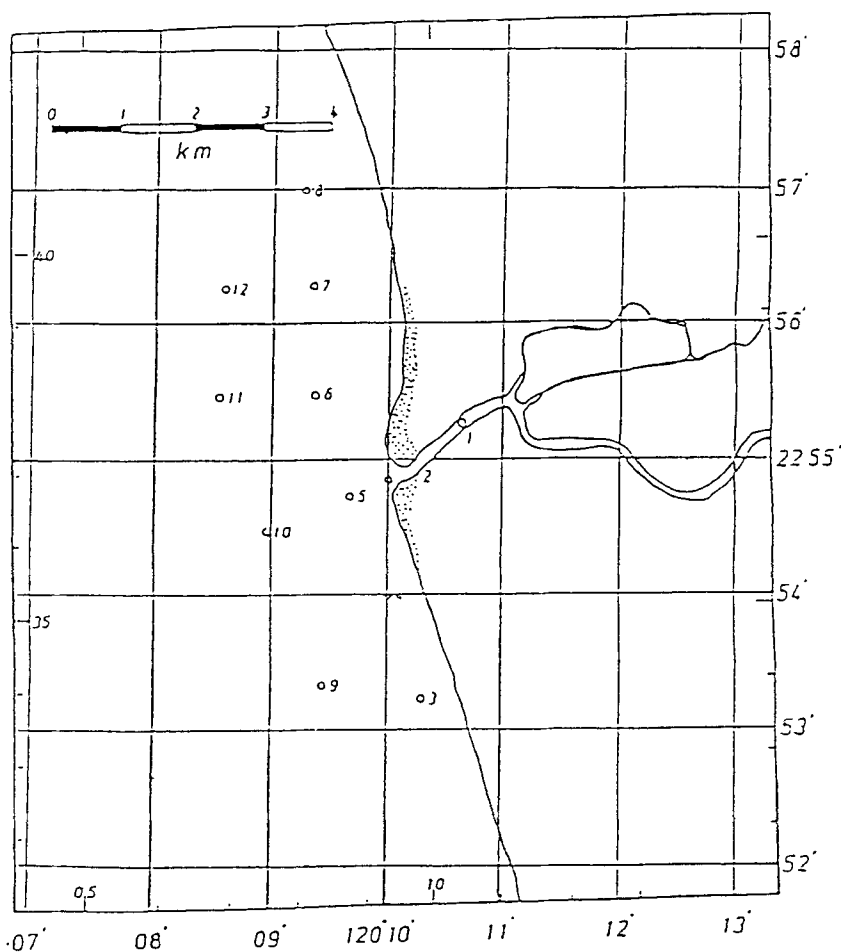
In January 1986, the first incident of green oysters (*Crassostrea gigas*) was reported, and reports of oyster mortality appeared three months later. The cause of greened oysters was immediately identified as due to the copper pollution discharged from the local copper recycling practice (Hung *et al.*, 1987). During the development of green oysters, a high adenylate energy charge (AEC) (value  $0.84 \pm 0.01$ ) was observed, and a low AEC value ( $0.21 \pm 0.01$ ) was found during mortality (Hung, 1988). Chen *et al.* (1988, 1989) found a high percentage (to 98%) of dissolved copper was involved in organic complexes in the Erhjin Chi Estuary, with organic copper concentration  $0.2$  to  $4.5 \mu\text{g l}^{-1}$  (9.2 to 87.6% of total dissolved copper) in the western coastal water, including the Erhjin Chi coastal water; these values were much higher than those off the eastern coast of Taiwan ( $0.2$  to  $0.5 \mu\text{g l}^{-1}$ , 5.5 to 14% of total dissolved copper). Hung *et al.* (1989) also found that high concentrations of particulate organic copper ( $>50\%$  of total copper) and nonlabile organic copper ( $>70\%$  of total dissolved copper) indicated a high value (as high as  $10.10 \mu\text{g l}^{-1}$  of copper assimilative capacity). Based on these data, the cause of greening and mortality of oysters in relation to bioavailable copper and assimilative capacity was

also evaluated (Hung and Han 1990; Han and Hung, 1990).

The purpose of this paper is to summarize seasonal distribution profiles of hydrographic conditions (temperature and salinity), chemical (BOD, nutrients, copper species and forms including total copper) and biomass parameters (mainly particulate organic carbon, chlorophyll- $\alpha$ , ATP and primary production) observed since November 1985. Finally, the paper evaluates the copper assimilative capacity and the possible relationships among copper species and forms and the biomass and hydrographic parameters in sea water in the Erhjin Chi coastal area.

## EXPERIMENTAL METHODS

Water samples, sediments and oysters have been collected seasonally from the Erhjin Chi area since November 1985. The sampling locations are shown in Figure 1. Water samples at different depths (0, 3, 10 and 25 metres) were taken with metal-free Van Dorn bottles on board either the research vessel "Ocean Research 1" or



**Figure 1** Sampling stations along the Erhjin Chi (River) coastal area.

fishing boats. Immediately after collection, water samples were filtered through 0.45 $\mu\text{m}$  fibre filters and analyzed for hydrographical, chemical and ecological variables. The procedures for analysis are shown in Figure 2. The methods of analysis, except the species and forms of copper as well as the classes of organic compounds, were carried out by standard methods (Hung *et al.* 1987, 1982). The analysis of copper species (inorganic and organic, polar and non-polar, labile and non-labile) and the forms (dissolved and particulate) were performed by the differential pulse anodic stripping voltammetric (DPASV) technique and the C-18 bound Sep-Pak cartridge method (Chen *et al.*, 1989). The classes of organic compounds such as lipids, acid-mobile and base-mobile compounds, isolated from sea water by the C-18 Sep-Pak technique, were further separated by TLC with chloroform, chloroform-methanol-ethylamine (10:15:1), and methanol-formic acid-water (5:5:1:1) solutions, respectively. Extra-pure phytol, n-octylamine and suberic acid obtained from Sigma chemical company was used as the standards of lipids, acid-mobile and base-mobile compounds. However, in this paper we only summarize and discuss the seasonal distributions of hydrographical, chemical and biomass parameters, and finally, evaluate the copper assimilative capacity of the oysters and possible relationships between the species and forms of copper and biomass parameters in sea water in the Erhjin Chi coastal area.

#### DISTRIBUTION OF HYDROGRAPHICAL, CHEMICAL AND BIOMASS PARAMETERS

Water temperature (19.6 to 32.4°C) and salinity (0.98 to 34.98 ppt) of the Erhjin Chi coastal area varies with season and location. For example, low salinity (Figure 3A) was generally found during the rainy season. In southern Taiwan, the rainy season (mainly contributed by storm rainfall) usually begins from March/April and the dry season in September/October. Streamflow in winter and spring is much less than that in summer and autumn. Shortage of water usually occurs from January through to March. During the dry season, the river-bed (mainly mud) absorbs inorganic and organic wastes discharged freely from domestic sources, industry and agriculture without any control. Immediately after heavy rain, the copper, as well as other inorganic and organic pollutants contained in the river sediments, is released to the river water and then discharged to the estuarine and coastal areas. After heavy rain, copper with other pollutants continues to accumulate in the sediments as the input from polluting sources continues. Figures 3B-3E show that high values of BOD (as high as 10.4 mg l<sup>-1</sup>), nitrate (as high as 17  $\mu\text{M}$ ), phosphate (as high as 210  $\mu\text{M}$ ) and total copper (as high as 370  $\mu\text{g l}^{-1}$ ) were found in the Erhjin Chi estuarine (Station 2) and coastal waters (Station 5), and were generally found during June/July 1987 immediately after heavy rain. Unfortunately, we had no particulate organic carbon (POC) and chlorophyll- $\alpha$  measurements during this period; however, similar conditions with an extremely high POC value (2.55 mg l<sup>-1</sup>, Figure 3F) with low chlorophyll- $\alpha$  (0.50  $\mu\text{g l}^{-1}$ , Figure 3G) was observed in March 1989, immediately after heavy rain. After heavy rain, the concentrations of pollutants were decreased and the chlorophyll- $\alpha$  values were increased.

Regional variations (Figures 3B-3G) with extremely high values of total copper (as high as 739  $\mu\text{g l}^{-1}$ , Table 1), as well as other pollutants (Tsai *et al.*, 1988) were generally to be found in river water (Station 1) compared with those in estuarine water (to 540  $\mu\text{g l}^{-1}$ , Figure 3E, Station 2) and in nearshore coastal water (to 370  $\mu\text{g l}^{-1}$ , Figure 3E,

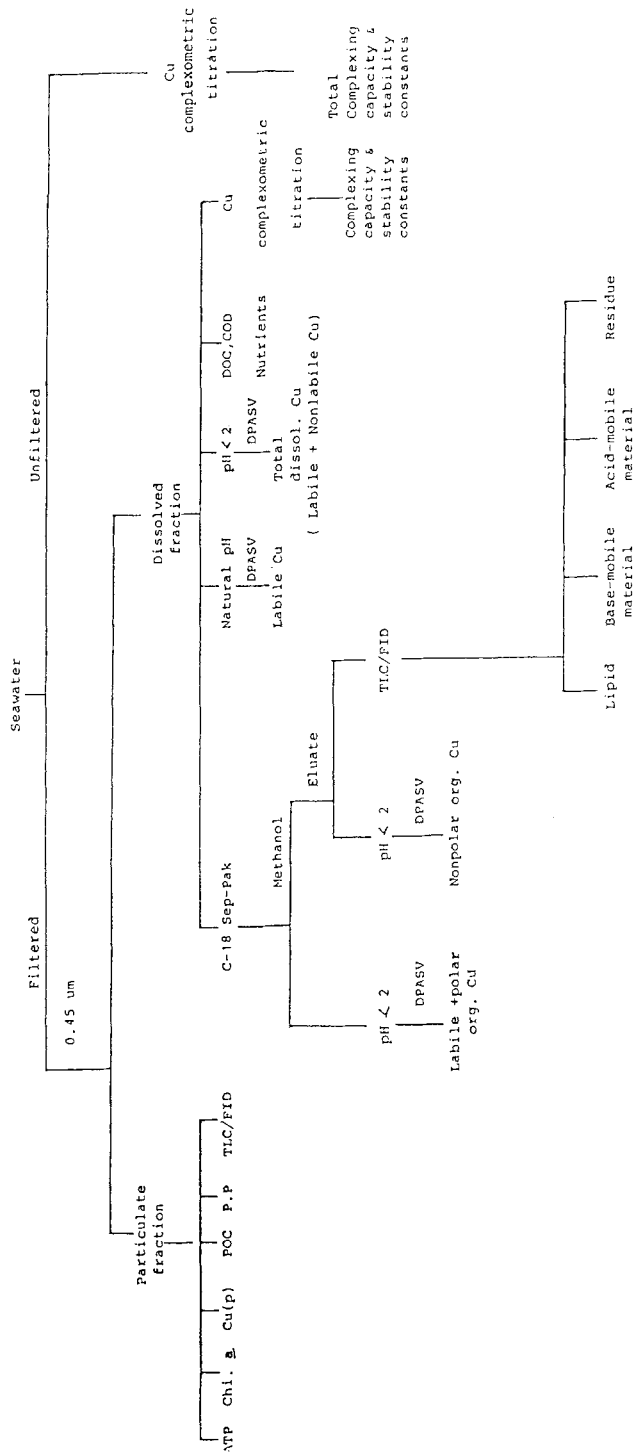


Figure 2 Scheme for the classification of copper forms and species in sea water.

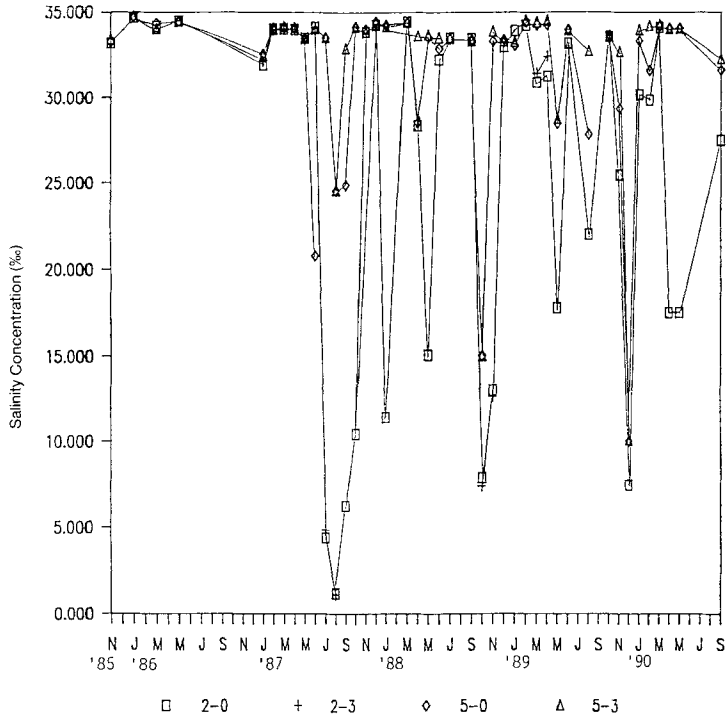


Figure 3A Seasonal variation of salinity in the waters of Stations 2 and 5.

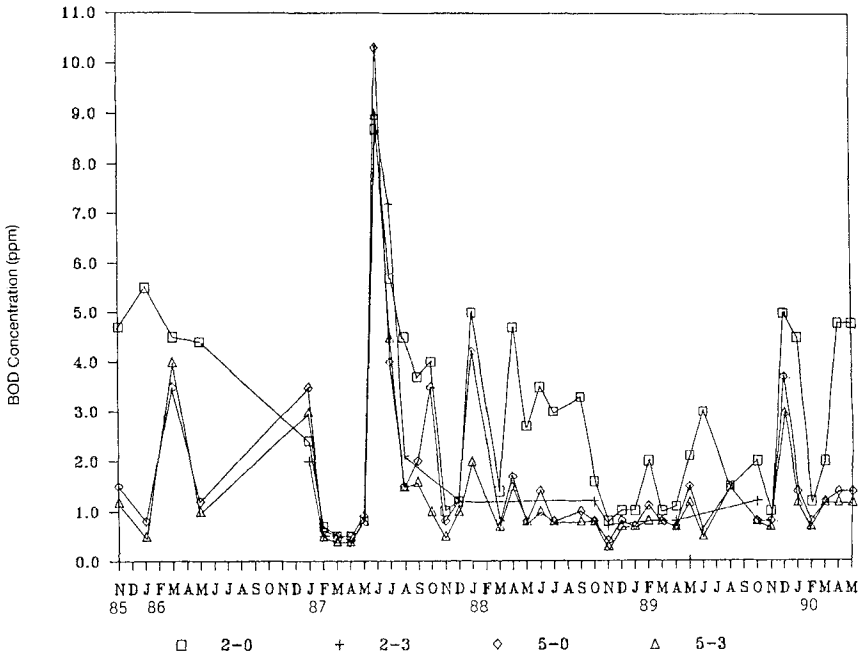


Figure 3B Seasonal variation of biochemical oxygen demand in the waters of Stations 2 and 5.

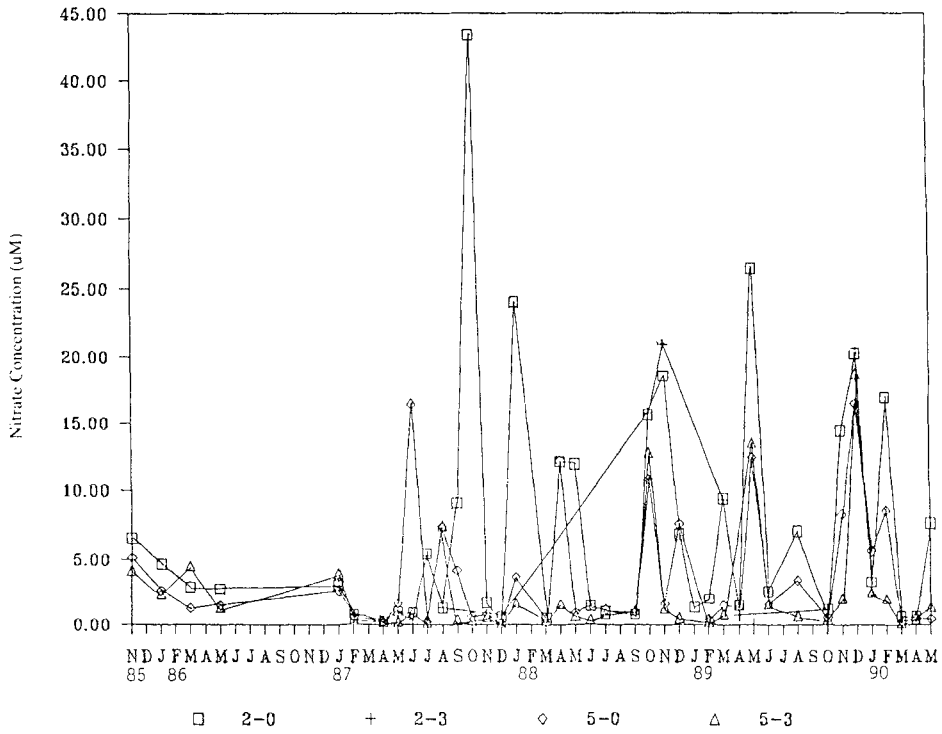


Figure 3C Seasonal variation of nitrate in the waters of stations 2 and 5.

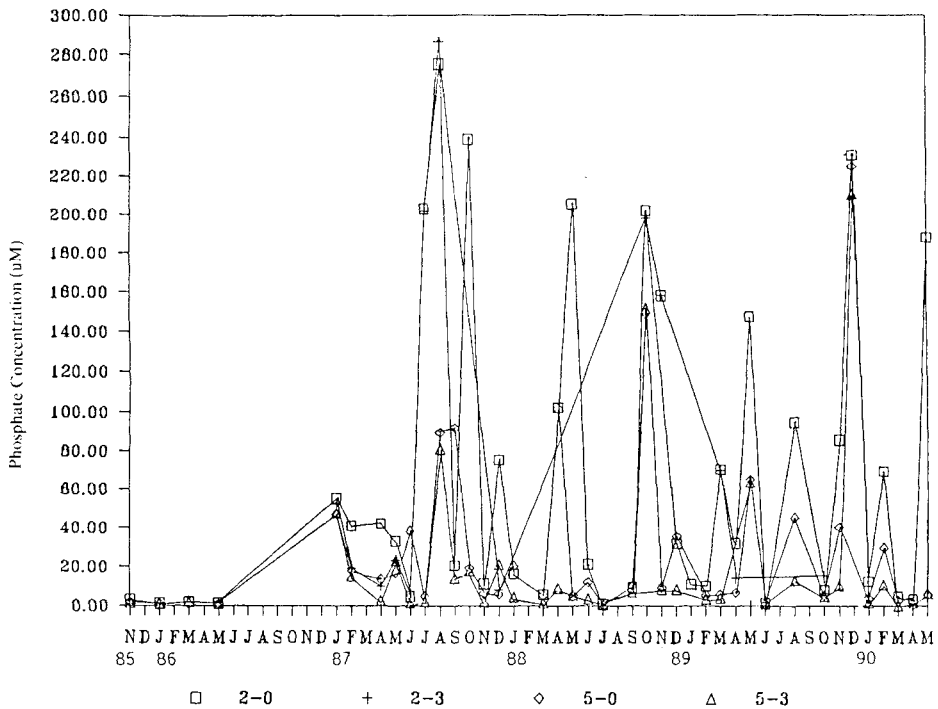


Figure 3D Seasonal variation of phosphate in the waters of stations 2 and 5.

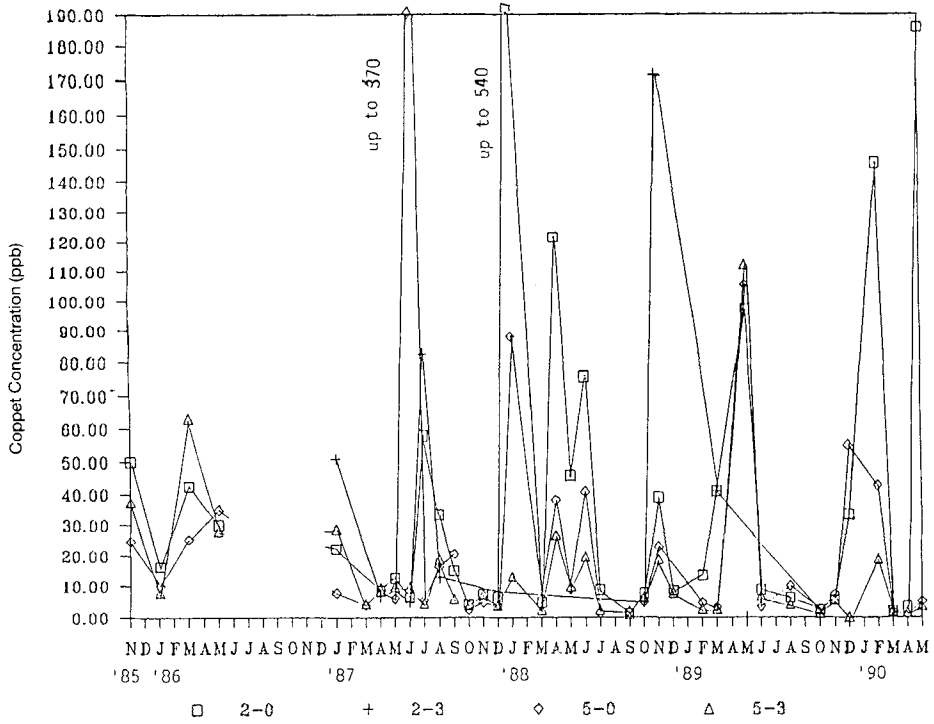


Figure 3E Seasonal variation of total copper concentration in the waters of Stations 2 and 5.

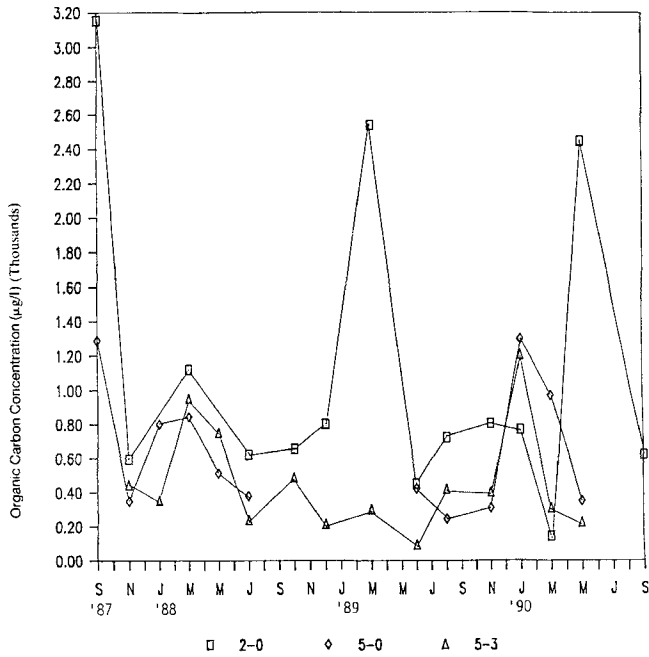
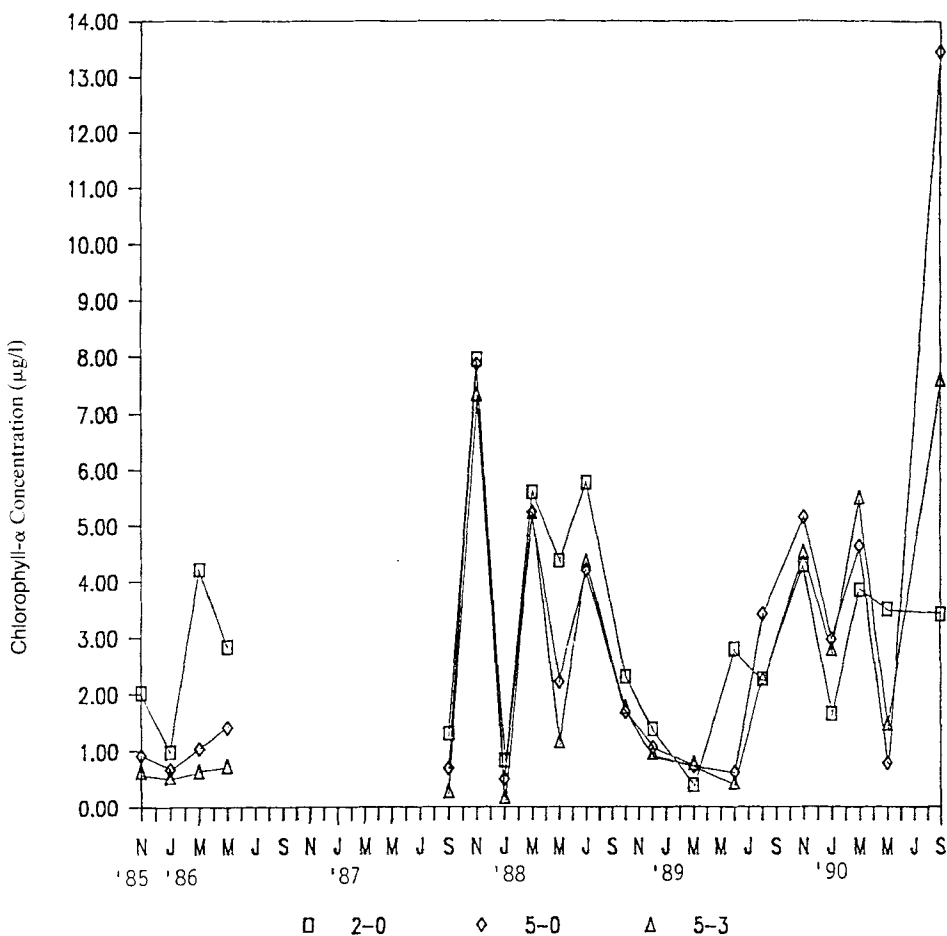


Figure 3F Seasonal variation of particulate organic carbon in the waters of stations 2 and 5.





**Figure 3G** Seasonal variation of chlorophyll  $\alpha$  in the waters of stations 2 and 5.

**Table 1** Concentration of copper species and forms (ppb) along the Erhjin Chi river, estuarine and coastal waters.

| Station | Sampling Date | Total | Particulate | Total | Dissolved in Water |      |           |       |          |
|---------|---------------|-------|-------------|-------|--------------------|------|-----------|-------|----------|
|         |               |       |             |       | Labile             |      | Nonlabile |       |          |
|         |               |       |             |       | Inorganic          | Free | Total     | Polar | NonPolar |
| 1       | 10/11/88      | 739   | 723         | 15.5  | --                 | --   | 15.5      | --    | --       |
| 2       | 10/11/88      | 9.97  | 3.25        | 6.72  | 1.81               | 0.03 | 4.88      | 3.39  | 1.49     |
| 4       | 10/11/88      | 10.3  | 4.26        | 6.00  | 2.62               | 0.05 | 3.33      | 0.18  | 3.12     |
| 6       | 10/11/88      | 17.9  | 6.90        | 11.0  | 5.20               | 0.09 | 5.73      | 3.71  | 2.02     |
| 7       | 10/11/88      | 16.4  | 5.61        | 10.7  | 5.21               | 0.09 | 5.45      | 3.25  | 2.20     |
| 8       | 10/11/88      | 21.2  | 4.84        | 16.4  | 8.42               | 0.15 | 7.79      | 6.25  | 1.54     |
| 9       | 10/11/88      | 8.11  | 3.09        | 5.02  | 1.44               | 0.02 | 3.56      | 3.44  | 0.12     |
| 10      | 10/11/88      | 12.1  | 4.25        | 7.82  | 4.99               | 0.09 | 2.74      | 1.09  | 1.65     |
| 11      | 10/11/88      | 11.5  | 3.11        | 8.40  | 4.75               | 0.08 | 3.57      | 0.44  | 3.13     |
| 12      | 10/11/88      | 10.5  | 4.04        | 6.51  | 3.17               | 0.05 | 3.29      | 0.58  | 2.71     |
| 5       | 10/11/88      | 14.1  | 4.13        | 10.0  | 2.35               | 0.04 | 7.62      | 1.38  | 6.24     |

|    |          |       |       |       |      |      |      |      |      |
|----|----------|-------|-------|-------|------|------|------|------|------|
| 5  | 12/25/88 | 25.9  | 9.57  | 16.3  | 7.79 | 0.13 | 8.43 | 5.23 | 3.20 |
| 5  | 01/11/89 | 27.1  | 3.52  | 23.6  | 2.04 | 0.03 | 21.5 | 7.82 | 13.7 |
| 5  | 01/25/89 | 23.9  | 2.11  | 21.8  | 13.1 | 0.23 | 8.48 | 0.80 | 7.68 |
| 5  | 02/16/89 | 15.3  | 3.98  | 11.3  | 2.32 | 0.04 | 8.91 | 2.39 | 6.52 |
| 5  | 03/15/89 | 10.6  | 5.51  | 5.12  | 1.70 | 0.03 | 3.39 | 2.83 | 0.56 |
| 3  | 10/11/88 | 23.5  | 5.33  | 18.2  | 10.7 | 0.19 | 7.28 | 1.81 | 5.47 |
| 3  | 12/25/88 | 25.2  | 3.13  | 22.1  | 12.6 | 0.22 | 9.24 | 8.14 | 1.10 |
| 3  | 01/11/89 | 9.11  | 1.25  | 7.86  | 3.24 | 0.06 | 4.62 | 2.03 | 2.59 |
| 3  | 01/25/89 | 10.1  | 5.16  | 4.99  | 2.12 | 0.04 | 2.87 | 1.09 | 1.78 |
| 3  | 02/16/89 | 10.2  | 2.49  | 7.71  | 2.15 | 0.04 | 5.56 | 3.81 | 1.75 |
| 3  | 03/15/89 | 9.22  | 3.19  | 6.03  | 2.50 | 0.04 | 3.83 | 2.64 | 1.19 |
| 3  | 12/25/88 | 10.1  | 2.08  | 8.07  | --   | --   | --   | --   | --   |
| 3  | 01/11/89 | 11.4  | 1.09  | 10.3  | 2.76 | 0.05 | 7.55 | 2.81 | 4.74 |
| 3  | 01/25/89 | 9.34  | 3.10  | 6.24  | 2.87 | 0.05 | 3.37 | 1.59 | 1.78 |
| 3  | 02/16/89 | 6.51  | 1.54  | 4.97  | 3.03 | 0.05 | 1.94 | 1.59 | 0.35 |
| 3  | 03/15/89 | 9.68  | 3.06  | 6.62  | 2.42 | 0.04 | 4.20 | 0.73 | 3.47 |
| 1  | 12/15/88 | 186   | 165.2 | 20.7  | ud   | ud   | 20.7 | 9.33 | 11.4 |
| 2  | 12/15/88 | 30.5  | 11.46 | 19.0  | 8.48 | 0.15 | 10.4 | 8.21 | 2.19 |
| 4  | 12/15/88 | 15.9  | 5.93  | 9.97  | 6.62 | 0.11 | 3.24 | 1.41 | 1.83 |
| 6  | 12/15/88 | 11.0  | 2.54  | 8.46  | 3.38 | 0.06 | 5.02 | 2.15 | 2.87 |
| 7  | 12/15/88 | 4.78  | 0.75  | 4.03  | 1.01 | 0.02 | 3.00 | 0.48 | 2.52 |
| 8  | 12/15/88 | 29.5  | 0.90  | 28.6  | 8.77 | 0.15 | 19.6 | 5.71 | 13.9 |
| 9  | 12/15/88 | 6.55  | 1.42  | 5.13  | ud   | ud   | 5.13 | 3.24 | 1.89 |
| 10 | 12/15/88 | 11.0  | 1.07  | 9.93  | 2.23 | 0.06 | 6.64 | 4.12 | 2.52 |
| 11 | 12/15/88 | 18.1  | 5.29  | 12.87 | 5.29 | 0.09 | 7.45 | 4.07 | 3.38 |
| 12 | 12/15/88 | 9.38  | 2.45  | 6.93  | 3.17 | 0.05 | 3.71 | 1.54 | 2.17 |
| 2  | 06/15/88 | --    | --    | 11.0  | 1.86 | 0.03 | 9.08 | 2.70 | 6.38 |
| 3  | 06/15/89 | --    | --    | 8.24  | 1.40 | 0.02 | 6.82 | 0.07 | 6.75 |
| 4  | 06/15/89 | --    | --    | 6.34  | 2.75 | 0.05 | 3.54 | ud   | 3.54 |
| 5  | 06/15/89 | --    | --    | 8.45  | 1.84 | 0.03 | 6.58 | 0.36 | 6.22 |
| 6  | 06/15/89 | --    | --    | 7.56  | 4.06 | 0.07 | 3.43 | ud   | 3.43 |
| 7  | 06/15/89 | --    | --    | 4.43  | 0.96 | 0.02 | 3.45 | 2.00 | 1.45 |
| 8  | 06/15/89 | --    | --    | 6.29  | 2.85 | 0.05 | 3.39 | 3.39 | 3.22 |
| 9  | 06/15/89 | --    | --    | 11.4  | 5.93 | 0.10 | 5.36 | ud   | 5.36 |
| 10 | 06/15/89 | --    | --    | 11.0  | 2.65 | 0.05 | 8.34 | 0.27 | 8.07 |
| 11 | 06/15/89 | --    | --    | 5.41  | 2.75 | 0.05 | 2.61 | 0.05 | 2.56 |
| 12 | 06/15/89 | --    | --    | 5.93  | 4.59 | 0.08 | 1.26 | ud   | 1.26 |
| 2  | 08/22/89 | 22.9  | 11.9  | 11.0  | 3.94 | 0.07 | 6.98 | 3.06 | 3.92 |
| 3  | 08/22/89 | 7.31  | 2.51  | 4.80  | 3.46 | 0.09 | 1.25 | ud   | 1.25 |
| 4  | 08/22/89 | 16.51 | 3.24  | 13.27 | 3.80 | 0.07 | 9.40 | 0.02 | 9.38 |
| 5  | 08/22/89 | 9.94  | 3.24  | 6.70  | 3.23 | 0.06 | 3.41 | 1.44 | 1.97 |
| 6  | 08/22/89 | 8.86  | 2.28  | 6.58  | 4.40 | 0.08 | 2.10 | 1.43 | 0.67 |
| 7  | 08/22/89 | 14.9  | 1.89  | 13.0  | 4.87 | 0.08 | 8.03 | 0.02 | 8.01 |
| 8  | 08/22/89 | 5.98  | 0.86  | 5.12  | 1.98 | 0.03 | 3.11 | 0.96 | 2.15 |
| 9  | 08/22/89 | 8.17  | 1.89  | 6.28  | 4.51 | 0.08 | 1.70 | 0.01 | 1.70 |
| 10 | 08/22/89 | 5.13  | 0.84  | 4.29  | 3.60 | 0.06 | 0.60 | 0.01 | 0.60 |
| 11 | 08/22/89 | 5.02  | 1.11  | 3.91  | 2.50 | 0.04 | 1.37 | 0.36 | 1.01 |
| 2  | 11/26/89 | 7.02  | 3.55  | 3.52  | 3.17 | 0.05 | 0.30 | 0.00 | 0.30 |
| 3  | 11/26/89 | 12.9  | 2.33  | 10.6  | 3.53 | 0.06 | 6.96 | 0.22 | 6.74 |
| 4  | 11/26/89 | 11.2  | 2.73  | 8.47  | 4.69 | 0.08 | 3.74 | 0.19 | 3.55 |
| 5  | 11/26/89 | 10.5  | 3.37  | 7.08  | 6.34 | 0.11 | 0.60 | 0.00 | 0.60 |
| 6  | 11/26/89 | 6.58  | 1.63  | 4.95  | 3.16 | 0.05 | 1.70 | 0.00 | 1.70 |
| 7  | 11/26/89 | 7.28  | 1.82  | 5.28  | 4.21 | 0.07 | 1.00 | 0.00 | 1.00 |
| 8  | 11/26/89 | 6.81  | 1.82  | 4.99  | 3.11 | 0.05 | 1.80 | 0.00 | 1.80 |
| 9  | 11/26/89 | 6.44  | 1.65  | 4.79  | 3.33 | 0.06 | 1.40 | 0.00 | 1.40 |
| 10 | 11/26/89 | 9.60  | 1.67  | 7.93  | 3.16 | 0.05 | 4.70 | 0.00 | 4.70 |
| 11 | 11/26/89 | 8.10  | 2.05  | 6.05  | 2.64 | 0.05 | 3.40 | 0.00 | 3.40 |
| 12 | 11/26/89 | 5.78  | 0.68  | 5.10  | 3.16 | 0.05 | 1.90 | 0.00 | 1.90 |
| 2  | 01/19/90 | 34.1  | 13.5  | 20.7  | 2.60 | 0.04 | 18.0 | 8.00 | 10.0 |
| 3  | 01/19/90 | 20.7  | 5.18  | 15.5  | 6.88 | 0.12 | 8.47 | 0.24 | 8.23 |
| 4  | 01/19/90 | 30.18 | 6.42  | 23.8  | 9.54 | 0.16 | 14.1 | 0.02 | 14.1 |

|    |          |      |      |      |      |      |      |      |      |
|----|----------|------|------|------|------|------|------|------|------|
| 5  | 01/19/90 | 16.8 | 3.19 | 12.6 | 7.67 | 0.13 | 5.84 | 0.39 | 5.45 |
| 6  | 01/19/90 | 5.38 | 1.42 | 3.96 | 2.75 | 0.05 | 1.20 | 0.00 | 1.20 |
| 7  | 01/19/90 | 5.49 | 0.64 | 4.88 | 2.93 | 0.05 | 1.90 | 0.90 | 1.00 |
| 8  | 01/19/90 | 4.87 | 0.61 | 4.26 | 2.20 | 0.03 | 2.00 | 0.00 | 2.00 |
| 9  | 01/19/20 | 12.5 | 1.74 | 10.8 | 5.52 | 0.09 | 5.20 | 0.00 | 5.20 |
| 10 | 01/19/90 | 13.1 | 4.22 | 8.91 | 6.30 | 0.11 | 2.50 | 0.00 | 2.50 |
| 11 | 01/19/90 | 7.93 | 0.93 | 7.00 | 2.42 | 0.04 | 4.54 | 0.95 | 3.59 |
| 12 | 01/19/90 | 5.48 | 0.24 | 5.24 | 2.27 | 0.04 | 2.93 | 1.72 | 1.21 |
| 2  | 03/19/90 | 7.07 | 1.06 | 6.00 | 4.88 | 0.08 | 1.10 | --   | --   |
| 3  | 03/21/90 | 6.90 | 0.69 | 6.21 | 5.11 | 0.09 | 1.00 | --   | --   |
| 4  | 03/21/90 | 11.2 | 1.45 | 9.70 | 4.52 | 0.08 | 5.10 | --   | --   |
| 5  | 03/21/90 | 6.82 | 0.63 | 6.19 | 3.34 | 0.06 | 2.80 | --   | --   |
| 6  | 03/21/90 | 8.71 | 1.93 | 6.78 | 4.03 | 0.07 | 2.70 | --   | --   |
| 7  | 03/21/90 | 11.0 | 0.66 | 10.3 | 4.81 | 0.08 | 5.40 | --   | --   |
| 8  | 03/21/90 | 5.36 | 0.31 | 5.05 | 4.00 | 0.07 | 1.00 | --   | --   |
| 9  | 03/21/90 | 7.82 | 0.39 | 7.43 | 6.30 | 0.11 | 1.00 | --   | --   |
| 10 | 03/21/90 | 10.2 | 0.75 | 9.48 | 2.87 | 0.05 | 6.60 | --   | --   |
| 11 | 03/21/90 | 8.55 | 1.03 | 7.52 | 4.78 | 0.08 | 2.70 | --   | --   |
| 12 | 03/21/90 | 10.9 | 1.25 | 9.46 | 2.44 | 0.04 | 7.20 | --   | --   |
| 1  | 05/24/90 | 299  | 273  | 26.0 | ud   | ud   | 26.0 | 5.62 | 20.4 |
| 2  | 05/24/90 | 234  | 197  | 36.5 | ud   | ud   | 36.5 | 6.50 | 30.0 |
| 3  | 05/24/90 | 14.1 | 1.68 | 12.4 | 2.58 | 0.06 | 8.78 | ud   | 8.78 |
| 4  | 05/19/90 | 11.8 | 2.04 | 9.80 | 3.13 | 0.05 | 6.60 | 1.02 | 5.58 |
| 5  | 05/24/90 | 7.59 | 1.33 | 6.26 | 4.31 | 0.07 | 1.88 | 0.88 | 1.00 |
| 6  | 05/24/90 | 13.8 | 2.00 | 11.8 | 5.24 | 0.09 | 6.43 | 3.27 | 3.16 |
| 7  | 05/24/90 | 8.42 | 2.00 | 6.40 | 5.34 | 0.09 | 0.99 | ud   | 0.99 |
| 08 | 05/24/90 | 9.44 | 1.04 | 8.40 | 4.89 | 0.08 | 3.43 | 0.79 | 2.64 |
| 9  | 05/24/90 | 9.04 | 0.83 | 8.21 | 4.42 | 0.07 | 3.72 | 0.13 | 3.59 |
| 10 | 05/24/90 | 11.9 | 2.02 | 9.90 | 4.69 | 0.08 | 5.13 | 0.53 | 4.60 |
| 11 | 05/24/90 | 15.0 | 3.39 | 11.6 | 5.40 | 0.09 | 6.13 | 1.99 | 4.14 |
| 12 | 05/24/90 | 7.75 | 0.71 | 7.04 | 2.27 | 0.04 | 4.73 | 4.39 | 0.34 |

ud: Undetectable; lower limits for labile: <1.0 ppb; for free ion and polar: <0.02 ppb.

--: Not detected.

Station 5). The EPA/ROC ocean water limits for BOD in class A, B and C waters are less than 2, 3 and 6  $\text{mg l}^{-1}$ , respectively; for total copper content in all classes (A, B, and C) less than 20  $\mu\text{g l}^{-1}$ . From Figures 3B and 2E, there is evidence that the Erhjin Chi estuarine and coastal waters have already been contaminated with organic wastes and copper. River water carries these pollutants, discharged from domestic and industrial sources, particularly from copper recycling operations, into the estuarine and coastal waters where the most important mariculture beds for oysters are located. Extremely high copper contents of 2100, 2225 and 4400 ppm dry weight in January, 1986, February 1988 and January, 1989, respectively, were found in cultured oysters (Hung *et al.*, 1990).

The fraction of living POC (mainly phytoplankton cells) and non-living POC (organic detritus including organic pollutants) can be estimated from the analytical results of POC and chlorophyll- $\alpha$  (Hung *et al.*, 1982). In this study, the proportions of living POC and non-living POC were, respectively, 0.009 to 1.40  $\text{mg l}^{-1}$  (from 0.99 to 73.5%) and from 0.008 to 13.4  $\text{mg l}^{-1}$  (from 26.5 to 99%), based on the POC concentration (0.10 to 2.30  $\text{mg l}^{-1}$ , Figure 3F) and chlorophyll- $\alpha$  (0.03 to 13.5  $\mu\text{g l}^{-1}$ , Figure 3G).

## COPPER ASSIMILATIVE CAPACITY

The copper assimilative capacity of water is estimated by the DPASV method, the

intercept on the abscissa of a plot of peak current in nA on the Y-axis with the concentration of copper added on the X-axis (Chen *et al.*, 1988). For sea water in the Erhjin Chi coastal area, the copper assimilative capacity ( $0.29$  to  $0.88 \mu\text{gl}^{-1}$ , Table 2) varies with season and location. High values of copper assimilative capacity were found in December, 1988, at Station 2, in the Erhjin Chi estuary, and a low value was found in October, 1988, at Station 9, offshore. Organic compounds are considered important binding components. Hung *et al.* (1989) indicated that the higher concentrations of organic compounds (lipids, base-mobile, acid-mobile compounds) and dissolved organic compounds (DOC) were to be found in the Erhjin Chi estuary (Station 2) water. The correlation coefficients in the nearshore and offshore stations were, respectively, 0.40 and 0.78.

## RELATIONSHIP BETWEEN SPECIES AND FORMS OF COPPER AND BIOMASS PARAMETERS

To investigate the influence of bioaccumulation of copper in oysters, the species and forms of copper in mariculture water were analyzed. The results are shown in Table 1 indicating that the particulate ( $0.24$  to  $723.4 \mu\text{gl}^{-1}$ , or 5.8 to 97.9% of total) and dissolved ( $3.52$  to  $36.5 \mu\text{gl}^{-1}$ ) copper concentrations vary with season and location. For instance, extremely high concentrations of particulate copper ( $165.3$  to  $723.4 \mu\text{gl}^{-1}$ , or 88.9 to 97.9% of total) were observed at the river station, and relatively low concentrations of particulate copper ( $0.24$  to  $197.1 \mu\text{gl}^{-1}$ ) were to be found in the estuarine and coastal waters. However, for dissolved copper, values as high as  $36.5 \mu\text{gl}^{-1}$  were found in the estuarine water. In the river and coastal waters, the ranges of dissolved copper concentrations were  $15.5$  to  $26.0 \mu\text{gl}^{-1}$  and  $3.5$  to  $28.6 \mu\text{gl}^{-1}$ , respectively.

For dissolved copper, both high concentrations of labile ( $<1.0 \mu\text{gl}^{-1}$  to  $13.3 \mu\text{gl}^{-1}$ ) and non-labile forms ( $0.3$  to  $21.5 \mu\text{gl}^{-1}$ ) were generally found at Station 5. The free cupric ion, estimated according to the method of Hanson *et al.* (1988), ranged from  $<0.02$  to  $0.23 \mu\text{gl}^{-1}$ . For the non-labile organic copper, polar and non-polar organic copper fractions were analyzed. High concentrations of non-polar organic copper ( $0.34$  to  $20.4 \mu\text{gl}^{-1}$ ) were generally found in river water compared with the concentrations found in estuarine and coastal waters. However, the high levels of polar organic copper (as high as  $16.5 \mu\text{gl}^{-1}$ , 96.6% of total non-labile organic copper) were usually observed in coastal water, and low values (as low as  $<0.02 \mu\text{gl}^{-1}$ , only

**Table 2** Average values of copper assimilative capacity ( $\mu\text{gl}^{-1}$ ) in waters along the Erhjin Chi coastal area.

| Station No. | Date      | Assimilative Capacity | Date      | Assimilative Capacity |
|-------------|-----------|-----------------------|-----------|-----------------------|
| 2           | Oct. 1988 | 0.57                  | Dec. 1988 | 0.88                  |
| 3           |           | 0.40                  |           | --                    |
| 4           |           | 0.61                  |           | --                    |
| 5           |           | 0.37                  |           | 0.54                  |
| 6           |           | 0.48                  |           | --                    |
| 7           |           | 0.33                  |           | --                    |
| 8           |           | 0.40                  |           | --                    |
| 9           |           | 0.29                  |           | --                    |
| 10          |           | 0.37                  |           | 0.45                  |
| 11          |           | 0.57                  |           | --                    |
| 12          |           | 0.50                  |           | --                    |

about 27.5% of total organic copper) in river water. Thus, non-polar organic copper, mainly from man-made pollution, originates from the river water.

The possible correlations among copper species and forms, and the biomass and hydrographical conditions of the Erhjin Chi coastal area, were analyzed and results tabulated in Tables 3A-3D. In general, the correlation coefficients between the parameters vary with season. For instance, during the dry winter season (January), there are significant correlations ( $>0.70$ ) as show below:

between living phytoplankton and bioavailable copper (the sum of particulate and labile copper) (0.93), particulate copper (0.86), total copper (0.85), dissolved copper (0.82), and non-polar organic copper (0.71);

between ATP and bioavailable copper (0.88), particulate copper (0.84), and dissolved copper (0.72);

between chlorophyll- $\alpha$  and particulate copper (0.86), total copper (0.85), dissolved copper (0.82) and non-polar organic copper (0.71);

between primary productivity and particulate copper (0.85), total copper (0.82), and dissolved copper (0.79).

However, during the late summer (September) and almost to the end of rainy season, the only significant correlation coefficients were:

between living phytoplankton and bioavailable copper (0.79);

between POC and detritus (0.91);

between salinity and total copper (0.98), particulate copper (0.98), bioavailable copper (0.98), polar copper (0.95).

During spring (March to May), no significant correlation ( $<0.70$ ) was found among the living phytoplankton (primary production or ATP) and these copper species or forms. Based on these results, there is no doubt that the extremely high copper concentrations in oysters found in January, 1966, February, 1987, and January, 1989, which were characteristic of the winter season, were likely to be the cause of the greening and mortality in the following few months.

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**Table 3A** Correlation coefficients among the copper species and forms, the hydrographical and biomass parameters along the Erhjin Chi coastal area. (n=10)  
Jan. 1990

| St. 3-12             | Total  |        | Particulate |       | Dissolved/Labile |       | Non-labile |       | Non-polar |        | Polar  |       | Non-polar |      | POC |    | Chl. α |    | P.P. |    | Bioavailable |    | Living phyto-plankton |    | Detritus |    |    |  |
|----------------------|--------|--------|-------------|-------|------------------|-------|------------|-------|-----------|--------|--------|-------|-----------|------|-----|----|--------|----|------|----|--------------|----|-----------------------|----|----------|----|----|--|
|                      | Cu     | Cu     | Cu          | Cu    | Cu               | Cu    | Cu         | Cu    | Cu        | Cu     | Cu     | Cu    | Cu        | Cu   | Cu  | Cu | Cu     | Cu | Cu   | Cu | Cu           | Cu | Cu                    | Cu | Cu       | Cu | Cu |  |
| Total Cu             | 1.00   |        |             |       |                  |       |            |       |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Particulate Cu       | 0.94** | 1.00   |             |       |                  |       |            |       |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Dissolved Cu         | 0.99** | 0.90** | 1.00        |       |                  |       |            |       |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Labile Cu            | 0.67   | 0.65   | 0.66        | 1.00  |                  |       |            |       |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Nonlabile Cu         | 0.99** | 0.81*  | 0.97*       | 0.67  | 1.00             |       |            |       |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Polar Cu             | -0.40  | 0.51   | -0.35       | -0.63 | -0.24            | 1.00  |            |       |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Nonpolar Cu          | 0.96** | 0.85*  | 0.98**      | 0.73* | 0.99**           | 0.37  | 1.00       |       |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| POC                  | 0.55   | 0.56   | 0.54        | 0.06  | 0.39             | -0.07 | 0.38       | 1.00  |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Chl. α               | 0.85*  | 0.86*  | 0.82*       | 0.48  | 0.67             | -0.49 | 0.71*      | 0.71* | 1.00      |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| P.P.                 | 0.82*  | 0.85*  | 0.79*       | 0.43  | 0.64             | -0.45 | 0.68*      | 0.71* | 0.99**    | 1.00   |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Bioavailable Cu      | 0.96*  | 0.98*  | 0.93**      | 0.61  | 0.83*            | -0.50 | 0.86*      | 0.64  | 0.93**    | 0.91** | 1.00   |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| ATP                  | 0.77*  | 0.84*  | 0.72*       | 0.46  | 0.55             | -0.61 | 0.61       | 0.80  | 0.87**    | 0.84*  | 0.88** | 1.00  |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Living phytoplankton | 0.85*  | 0.86*  | 0.82*       | 0.48  | 0.67             | -0.49 | 0.71*      | 0.71* | 0.99**    | 0.99** | 0.93** | 0.87* | 1.00      |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
| Detritus             | 0.06   | -0.07  | 0.06        | 0.40  | -0.11            | 0.39  | -0.17      | 0.71  | 0.71      | 0.02   | -0.02  | 0.26  | 0.87*     | 1.00 |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |
|                      |        |        |             |       |                  |       |            |       |           |        |        |       |           |      |     |    |        |    |      |    |              |    |                       |    |          |    |    |  |

\*: Significant at  $p < 0.05$

\*\* : Significant at  $p < 0.01$

Part = Particulate; Diss. = Dissolved; Lab. = Labile; Nonla. = Nonlabile; Bioa. = Bioavailable; POC = Particulate Organic Carbon; Chl.α = Chlorophyll α; ATP = Adenosine Triphosphate; Prim. Prod. = Primary Production; Detr. = Detritus; Sal. = Salinity.

**Table 3B** Correlation coefficients among the copper species and forms, the hydrographical and biomass parameters along the Erhjin Chi coastal water.

| March       | Copper Species and Forms in Water |       |       |      |         |       |        |       |      |        | Prim. POC | Liv. Chl. a | ATP  | Prod. | Phyt. | Detr. | Sal. | Temp. | pH |  |
|-------------|-----------------------------------|-------|-------|------|---------|-------|--------|-------|------|--------|-----------|-------------|------|-------|-------|-------|------|-------|----|--|
|             | Total 15-17, 1990                 | Part. | Diss. | Lab. | Nonlab. | Bioa. | Nonla. | Bioa. | POC  | Chl. a |           |             |      |       |       |       |      |       |    |  |
| Total       | 1                                 |       |       |      |         |       |        |       |      |        |           |             |      |       |       |       |      |       |    |  |
| Part.       | 0.46                              | 1     |       |      |         |       |        |       |      |        |           |             |      |       |       |       |      |       |    |  |
| Dis.        | 0.97                              | 0.23  | 1     |      |         |       |        |       |      |        |           |             |      |       |       |       |      |       |    |  |
| Labile      | 0.28                              | 0.25  | 0.22  | 1    |         |       |        |       |      |        |           |             |      |       |       |       |      |       |    |  |
| Nonlab.     | 0.89                              | 0.30  | 0.89  | 0.65 | 1       |       |        |       |      |        |           |             |      |       |       |       |      |       |    |  |
| Bioav.      | 0.07                              | 0.19  | 0.12  | 0.90 | 0.53    | 1     |        |       |      |        |           |             |      |       |       |       |      |       |    |  |
| POC         | 0.20                              | 0.30  | 0.14  | 0.41 | 0.08    | 0.54  | 1      |       |      |        |           |             |      |       |       |       |      |       |    |  |
| Chl. a      | 0.25                              | 0.24  | 0.22  | 0.09 | 0.13    | 0.19  | 0.27   | 1     |      |        |           |             |      |       |       |       |      |       |    |  |
| ATP         | 0.40                              | 0.01  | 0.44  | 0.07 | 0.31    | 0.07  | 0.20   | 0.08  | 1    |        |           |             |      |       |       |       |      |       |    |  |
| Prim. Prod. | 0.05                              | 0.13  | 0.10  | 0.12 | 0.02    | 0.06  | 0.45   | 0.82  | 0.41 | 1      |           |             |      |       |       |       |      |       |    |  |
| Liv. Phyt.  | 0.27                              | 0.30  | 0.23  | 0.02 | 0.19    | 0.11  | 0.25   | 0.98  | 0.06 | 0.71   | 1         |             |      |       |       |       |      |       |    |  |
| Detritus    | 0.24                              | 0.13  | 0.26  | 0.23 | 0.09    | 0.29  | 0.84   | 0.42  | 0.77 | 0.60   | 0.35      | 1           |      |       |       |       |      |       |    |  |
| Salinity    | 0.12                              | 0.23  | 0.17  | 0.10 | 0.10    | 0.00  | 0.07   | 0.08  | 0.61 | 0.21   | 0.26      | 0.47        | 1    |       |       |       |      |       |    |  |
| Temperat.   | 0.24                              | 0.16  | 0.30  | 0.01 | 0.24    | 0.08  | 0.06   | 0.08  | 0.54 | 0.05   | 0.04      | 0.57        | 0.79 | 1     |       |       |      |       |    |  |
| pH          | 0.01                              | 0.00  | 0.02  | 0.27 | 0.11    | 0.28  | 0.10   | 0.42  | 0.06 | 0.24   | 0.59      | 0.50        | 0.25 | 0.30  | 1     |       |      |       |    |  |

Part = Particulate; Diss. = Dissolved; Lab. = Labile; Nonlab. = Nonlabile; Bioa. = Bioavailable; POC = Particulate Organic Carbon;  
 Chl. a = Chlorophyll a; ATP = Adenosine Triphosphate; Prim. Prod. = Primary Production; Detr. = Detritus; Sal. = Salinity.

**Table 3C** Correlation coefficients among the copper species and forms, the hydrographical and biomass parameters along the Erhjin Chi coastal area.

| St. 3-12      | Total |       | Dissol. |       | Labile |       | Nonlabile |       | Polar         |          | Cu   | POC  | Chl. $\alpha$ | P.P. | BCu  | ATP | Living |  |  |
|---------------|-------|-------|---------|-------|--------|-------|-----------|-------|---------------|----------|------|------|---------------|------|------|-----|--------|--|--|
|               | Cu    | Cu    | Cu      | Cu    | Cu     | Cu    | Cu        | Cu    | Phytoplankton | Detritus |      |      |               |      |      |     |        |  |  |
| Total Cu      | 1.00  |       |         |       |        |       |           |       |               |          |      |      |               |      |      |     |        |  |  |
| Particul. Cu  | 0.73  | 1.00  |         |       |        |       |           |       |               |          |      |      |               |      |      |     |        |  |  |
| Dissol. Cu    | 0.97  | 0.56  | 1.00    |       |        |       |           |       |               |          |      |      |               |      |      |     |        |  |  |
| Labile Cu     | 0.12  | 0.46  | 0.00    | 1.00  |        |       |           |       |               |          |      |      |               |      |      |     |        |  |  |
| Nonlabile Cu  | 0.82  | 0.31  | 0.90    | -0.43 | 1.00   |       |           |       |               |          |      |      |               |      |      |     |        |  |  |
| Polar Cu      | 0.05  | -0.05 | 0.05    | -0.21 | 0.20   | 1.00  |           |       |               |          |      |      |               |      |      |     |        |  |  |
| Nonpolar Cu   | 0.74  | 0.32  | 0.80    | -0.28 | 0.82   | -0.41 | 1.00      |       |               |          |      |      |               |      |      |     |        |  |  |
| POC           | 0.62  | 0.51  | 0.64    | 0.50  | 0.38   | -0.01 | 0.36      | 1.00  |               |          |      |      |               |      |      |     |        |  |  |
| Chl. $\alpha$ | -0.30 | -0.12 | -0.35   | 0.41  | -0.50  | -0.01 | -0.46     | 0.05  | 1.00          |          |      |      |               |      |      |     |        |  |  |
| P.P.          | -0.25 | -0.06 | -0.30   | 0.40  | -0.45  | 0.02  | -0.41     | -0.01 | 0.99          | 0.99     | 0.99 |      |               |      |      |     |        |  |  |
| Bioavail. Cu  | 0.53  | 0.82  | 0.38    | 0.86  | -0.04  | -0.23 | 0.10      | 0.60  | 0.19          | 0.21     | 1.00 |      |               |      |      |     |        |  |  |
| ATP           | 0.10  | -0.18 | 0.18    | 0.34  | 0.00   | 0.10  | -0.06     | 0.48  | 0.34          | 0.28     | 0.17 | 1.00 |               |      |      |     |        |  |  |
| Living        | -0.30 | -0.12 | -0.35   | 0.41  | -0.50  | -0.01 | -0.46     | -0.05 | 0.99          | 0.99     | 0.19 | 0.34 | 1.00          |      |      |     |        |  |  |
| Phytoplankton |       |       |         |       |        |       |           |       |               |          |      |      |               |      |      |     |        |  |  |
| Detritus      | 0.63  | 0.52  | 0.65    | 0.48  | 0.40   | -0.01 | 0.38      | 0.99  | -0.11         | -0.07    | 0.59 | 0.46 | -0.11         | 0.46 | 1.00 |     |        |  |  |

◇ : Significant at  $P < 0.05$

◇◇ : Significant at  $P < 0.01$

Part. = Particulate; Diss. = Dissolved; Lab. = Labile; Nonlab. = Nonlabile; Bioa. = Bioavailable; POC = Particulate Organic Carbon;

Chl.  $\alpha$  = Chlorophyll  $\alpha$ ; ATP = Adenosine Triphosphate; Prim. Prod. = Primary Production; Detr. = Detritus; Sal. = Salinity.



**Table 3D** Correlation coefficients among the copper species and forms, the hydrographical and biomass parameters along the Erhjin Chi coastal water.

|               | Total     |             | Copper Species and Forms |       |      |        |       | POC  | Chl. $\alpha$ | Prim. Prod. | Liv. Phyt. | Detr. | Sal. | Temp. | pH   |
|---------------|-----------|-------------|--------------------------|-------|------|--------|-------|------|---------------|-------------|------------|-------|------|-------|------|
|               | September | 20-22, 1990 | Part.                    | Diss. | Lab. | Nonla. | Polar |      |               |             |            |       |      |       |      |
| Total         |           | 1           |                          |       |      |        |       |      |               |             |            |       |      |       |      |
| Cu            | 0.99      |             |                          |       |      |        |       |      |               |             |            |       |      |       |      |
| Part.         | 0.98      | 1           |                          |       |      |        |       |      |               |             |            |       |      |       |      |
| Cu            | 0.87      | 0.95        | 1                        |       |      |        |       |      |               |             |            |       |      |       |      |
| Dis.          | 0.85      | 0.85        | 0.90                     | 1     |      |        |       |      |               |             |            |       |      |       |      |
| Labile        | 0.94      | 0.92        | 0.96                     | 0.74  | 1    |        |       |      |               |             |            |       |      |       |      |
| Cu            | 0.98      | 0.96        | 0.98                     | 0.85  | 0.96 | 1      |       |      |               |             |            |       |      |       |      |
| Nomlab.       | 0.61      | 0.57        | 0.66                     | 0.32  | 0.81 | 0.62   | 1     |      |               |             |            |       |      |       |      |
| Cu            | 0.99      | 0.99        | 0.96                     | 0.89  | 0.91 | 0.91   | 0.55  | 1    |               |             |            |       |      |       |      |
| Nonpol.       | 0.55      | 0.57        | 0.96                     | 0.48  | 0.39 | 0.53   | 0.01  | 0.58 | 1             |             |            |       |      |       |      |
| Cu            | 0.23      | 0.22        | 0.22                     | 0.09  | 0.29 | 0.55   | 0.50  | 0.70 | 0.72          | 1           |            |       |      |       |      |
| Bioav.        | 0.14      | 0.16        | 0.08                     | 0.24  | 0.03 | 0.02   | 0.13  | 0.57 | 0.11          | 0.74        | 1          |       |      |       |      |
| POC           | 0.19      | 0.52        | 0.25                     | 0.30  | 0.20 | 0.08   | 0.40  | 0.79 | 0.61          | 0.72        | 0.55       | 1     |      |       |      |
| Chl. $\alpha$ | 0.43      | 0.21        | 0.35                     | 0.48  | 0.23 | 0.44   | 0.25  | 0.47 | 0.91          | 0.62        | 0.08       | 0.45  | 1    |       |      |
| Pri. Prod.    | 0.98      | 0.98        | 0.93                     | 0.87  | 0.88 | 0.95   | 0.47  | 0.98 | 0.50          | 0.27        | 0.13       | 0.16  | 0.44 | 1     |      |
| Liv. Phyt.    | 0.35      | 0.37        | 0.50                     | 0.21  | 0.33 | 0.38   | 0.15  | 0.35 | 0.14          | 0.14        | 0.30       | 0.38  | 0.15 | 0.43  | 1    |
| Detritus      | 0.72      | 0.71        | 0.70                     | 0.70  | 0.63 | 0.76   | 0.18  | 0.72 | 0.77          | 0.47        | 0.18       | 0.35  | 0.77 | 0.10  | 0.54 |
| Salinity      |           |             |                          |       |      |        |       |      |               |             |            |       |      |       |      |
| Temperat.     |           |             |                          |       |      |        |       |      |               |             |            |       |      |       |      |
| pH            |           |             |                          |       |      |        |       |      |               |             |            |       |      |       |      |

Part. = Particulate; Diss. = Dissolved; Lab. = Labile; Nonla. = Nonlabile; Nonp. = Nonpolar; Bioa. = Bioavailable;  
 POC = Particulate Organic Carbon; Chl.  $\alpha$  = Chlorophyll  $\alpha$ ; Prim. Prod. = Primary Production; Liv. Phyt. = Living Phytoplankton;  
 Detr. = Detritus; Sal. = Salinity.

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