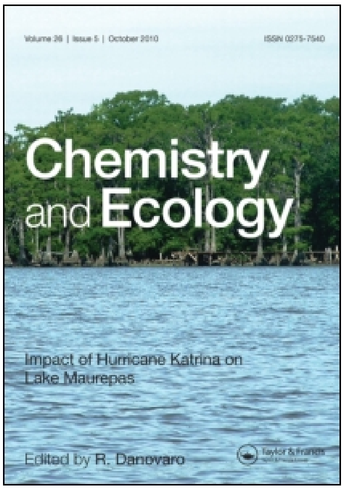

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HEAVY METALS IN FISH TISSUES AND DIFFERENT SPECIES OF FISH FROM THE SOUTHERN COAST OF TAIWAN

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Heavy metal contents varied with different species of demersal fishes (such as *Megalops cyprinoides*, *Pseudorhombus dupliciocellatus*, *Sillago sihama*, *Acanthopagrus australis*, *Terapon jarbua*, *Acanthopagrus latus*, *Cephalopholis miniata*, *Lutjanus bohar*, *Scarus forsteni*, and *Siganus guttotus*) and pelagic fishes (*Sphyraena jello* and *Sphyraena forsteri*) caught from the southwestern (Erhjin Chi, a seriously polluted area) and southeastern (Green Island, a relatively unpolluted area) coast of Taiwan in August of 1997 and February of 1998. The orders of heavy metal concentrations (dry weight) in demersal fishes were: copper (over $100\,\mu g\,g^{-1}$), *T. jarbua* ≫ *A. australis* > *M. cyprinoides* > *P. dupliciocellatus*; zinc (over $500\,\mu g\,g^{-1}$), *L. bohar* > *T. jarbua* > *P. dupliciocellatus* > *S. sihama*; lead (over $4\,\mu g\,g^{-1}$), *P. dupliciocellatus* > *S. guttotus*; nickel (over $5\,\mu g\,g^{-1}$), *S. guttotus* > *S. sihama*; chromium (over $20\,\mu g\,g^{-1}$), *P. dupliciocellatus* > *L. bohar* > *T. jarbua*; arsenic (over $1\,\mu g\,g^{-1}$), *A. australis* > *M. cyprinoides*; tin (over $3\,\mu g\,g^{-1}$), *L. bohar*. For the pelagic fishes, high values of nickel and chromium were found in *S. jello*. Higher concentrations of heavy metals were generally observed in livers compared with those in gills, guts, eggs and flesh of demersal fish. Significant correlations were also obtained between heavy metal contents in fish and fish livers. The seasonal and regional variations as well as their impact on sedimentary heavy metal concentrations are evaluated and discussed.

Keywords: Heavy metals; demersal and pelagic fish; seasonal and regional variations; correlation coefficients; Taiwan

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INTRODUCTION

During the past few decades, industrialization and urbanization have taken place in Taiwan, accompanied by an increase in heavy metal pollution associated with organic pollution in the coastal environment, especially near estuaries (Hung *et al.*, 1998a, 1998b, 1996a, 1993; Han *et al.*, 1998; Hung, 1989). For instance, the high contents of cadmium (as high as 1,320 $\mu g\, g^{-1}$ with an average of $378 \pm 216\, \mu g\, g^{-1}$) and lead (as high as 12,700 $\mu g\, g^{-1}$ with an average of $3{,}150 \pm 369\, \mu g\, g^{-1}$), discharged from industries, were found in the surface soils (0 – 10 cm) of agricultural environments (Hong *et al.*, 1983). The extremely high concentrations of lead ($1{,}420 \pm 709\, \mu g\, l^{-1}$) were usually observed in the industrial waste water, and the lead concentrations (with an average of $273 \pm 494\, \mu g\, l^{-1}$) in river water from downstream were also high (Han *et al.*, 1992). The copper pollution caused green oysters (*Crassostrea gigas*) (Hung and Chuang, 1994) and mortalities were observed three months later in both mariculture and wild oysters off the Erhjin Chi coast of Taiwan (Hung, 1989; Han and Hung, 1990; Hung and Han, 1992; Goldberg, 1993; Hung and Chuang, 1994).

Marine organisms are capable of accumulating numerous pollutants from natural water by many orders of magnitude. Since Goldberg *et al.* (1978) first initiated the "Mussel Watch" programme in which molluscs are used as sentinels of coastal chemical pollution, many investigators (Addison, 1996; Livingstone *et al.*, 1988; Lack and Johnson, 1985; Hung *et al.*, 1998a, 1998b, 1985, 1983, 1981; McIntyre and Pearce, 1980) studied the biological effects of mussels and/or bivalves for monitoring in studies of marine pollution. Monitoring environmental impact through the bioaccumulation of pollutants by nearshore fishes was also applied (Hung and Yang, 1975; Hall *et al.*, 1995; Evan *et al.*, 1993; Hornung and Kress, 1991; Fowler, 1986; Greig *et al.*, 1976). However, few papers concerned the effects of bioaccumulation of heavy metals on different fish, species especially along the southwestern (Erhjin Chi) coast, a seriously polluted area in Taiwan. This paper presents data on the concentrations of heavy metals (Cu, Zn, Cd, Pb, Cr, Ni, As and Sn) in ten species of demersal fishes: *Megalops cyprinoides*, *Pseudorhombus dupliciocellatus*, *Sillago sihama*, *Acanthopagrus australis*, *Terapon jarbua*, *Acanthopagrus latus*, *Cephalopholis miniata*, *Lutjanus bohar*, *Scarus forsteni*, and *Siganus guttotus* as well as two species of pelagic fish, *Sphyraena jello* and *Sphyraena forsteri*, along the Erhjin Chi coastal area. Some species collected from off the Taiwan southeastern coast (Green Island, a relatively unpolluted area) are also analyzed for comparison. The choice of species analysed was based on relative abundance of the regions.

MATERIALS AND EXPERIMENTAL METHODS

Sampling Techniques

Ten species of demersal fish (*M. cyprinoides*, *P. dupliciocellatus*, *S. sihama*, *A. australis*, *T. jarbua*, *A. latus*, *C. miniata*, *L. bohar*, *S. forsteni*, and *S. guttotus*) and two species of pelagic fish (*S. jello*, *S. forsteri*) were collected along the southwestern coast (a seriously polluted Erhjin Chi area) and the southeastern coast of Taiwan (Green Island, a relatively unpolluted area) during the periods August 24–31, 1997 and February 17–22, 1998. The sampling locations are shown in Figure 1. Immediately after the collection (see UNEP, 1991), the samples, kept frozen

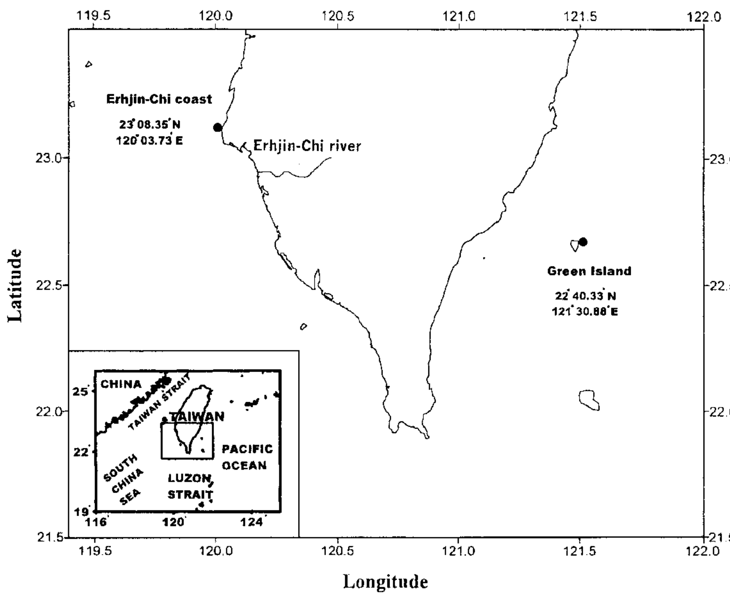

FIGURE 1 Sampling locations.

at $-20°C$, were separated into different parts (gills, guts, livers, eggs and flesh). Each part of fish tissues was washed with sea water, blotted dry with cheese-cloth, and then treated for heavy metal analysis (NOAA, 1989).

For investigating the environments in which the fishes occur, surface sediments were also collected. Surface sediments, collected by dredging, were stored in plastic bags and kept in an icebox until analysis.

Methods of Analysis

Heavy metals in fish tissues and sediments were determined by both methods of graphite atomic absorption spectrophotometry (GAAS, Hitachi Z-8000) and differential pulse anodic stripping voltammetry (DPASV, EG and G 384B) under the clean-room conditions. The calibration curves and the method detection limits (Hg, $< 0.05\,\mu g\,l^{-1}$; Zn, $< 0.50\,\mu g\,l^{-1}$; Cu, $< 0.51\,\mu g\,l^{-1}$; Cd, $< 0.01\,\mu g\,l^{-1}$; Pb, $< 0.15\,\mu g\,l^{-1}$; Ni, $< 2.0\,\mu g\,l^{-1}$; Sn, $< 5.3\,ng\,l^{-1}$) for both GAAS and DPASV analysis, as well as the control charts for these metals, have been carried out. The difference for analysis of samples between GAAS and DPASV was less than $\pm 5\%$. Surface sediments were dissolved with a mixture of ultrapure hydrofluoric acid and nitric acid in Teflon beakers, and the resulting solution analyzed for copper, zinc, cadmium, lead, chromium, nickel, arsenic, tin. Fish samples were digested with a mixture of nitric acid and sulphuric acid (v/v, 1 : 1) solution (AOAC, 1975), and the supernatant solution was taken for analysis of trace metals.

The analysis of heavy metal contents in the Standard Reference Material samples such as bovine liver (NBS-SRM-1577), orchard leaf (NBS-SRM-1571) and oysters (*Crassostrea gigas*, prepared by this laboratory) were intercalibrated with the Trace Metal Laboratory, Department of Chemistry, University of Idaho and Silver Valley Laboratory, Inc. (USEPA-core laboratory), Kellogy, Idaho (USA). The results indicated that the accuracies of copper ($\pm 8.11\%$), zinc ($\pm 0.80\%$), lead ($\pm 3.10\%$), and nickel ($\pm 6.83\%$) for sediments and copper ($\pm 3.54\%$), zinc ($\pm 3.75\%$), lead ($\pm 5.56\%$) and nickel ($\pm 17.2\%$) for organisms among these laboratories were satisfactory (Hung *et al.*, 1996b). The method detection limits (dry weight) for organisms and sediments were $1.2\,\mu g\,g^{-1}$ and $0.39\,\mu g\,g^{-1}$ for copper, $0.30\,\mu g\,g^{-1}$ and $0.33\,\mu g\,g^{-1}$ for zinc, $0.003\,\mu g\,g^{-1}$ and $0.01\,\mu g\,g^{-1}$ for cadmium, $0.004\,\mu g\,g^{-1}$ and $0.33\,\mu g\,g^{-1}$ for lead, $0.006\,\mu g\,g^{-1}$ and $0.36\,\mu g\,g^{-1}$ for nickel, 0.006 μg g^{-1} and 0.27 μg g^{-1} for chromium, 0.005 μg g^{-1} and 0.45 μg g^{-1} for arsenic and 0.53 μg g^{-1} and 0.10 μg g^{-1} for tin.

RESULTS AND DISCUSSION

Figure 2 shows that the contents (dry weight) of trace metals [Cu (14.6 – 365.0 μg g^{-1}), Zn (42.3 – 1464 μg g^{-1}), Cd (< 0.003 – 7.40 μg g^{-1}), Pb (< 0.004 – 12.26 μg g^{-1}), Ni (< 0.006 – 123.8 μg g^{-1}), Cr (4.9 – 39.0 μg g^{-1}), As (< 0.005 – 5.95 μg g^{-1}) and Sn (< 0.53 – 3.60 μg g^{-1})] in eleven species of fish collected from the southwestern coast (Erhjin Chi area) in August of 1997 vary with the species of fish and habits of feeders. In general, higher contents of heavy metals were observed in demersal fishes (*Megalops cyprinoides*, *Pseudorhombus dupliciocellatus*, *Sillago sihama*, *Acanthopagrus australis*, *Terapon jarbua*, *Acanthopagrus latus*, *Cephalopholis miniata*, *Lutjanus bohar*, *Scarus forsteni*, and *Siganus guttotus*) compared with those values in pelagic fishes (*Sphyraena jello* and *Sphyraena forsteri*). Among demersal fishes, the detritus and benthic organism feeders containing heavy metals were generally higher than those in the detritus and herbivorous feeders. The concentration order for copper (over 100 μg g^{-1}) was *T. jarbua* (365 μg g^{-1}) > *A. australis* (120 μg g^{-1}) > *M. cyprinoides* (115 μg g^{-1}) > *P. dupliciocellatus* (110 μg g^{-1}); that for zinc (over 500 μg g^{-1}) was *L. bohar* (1464 μg g^{-1}) > *T. jarbua* (830 μg g^{-1}) > *P. dupliciocellatus* (578 μg g^{-1}) > *S. sihama* (550 μg g^{-1}); that for lead (over 4 μg g^{-1}) was *P. dupliciocellatus* (8.40 μg g^{-1}) > *S. guttotus* (4.45 μg g^{-1}); that for nickel (over 5 μg g^{-1}) was *S. guttotus* (58.4 μg g^{-1}) > *S. sihama* (5.0 μg g^{-1}); that for chromium (over 20 μg g^{-1}) was *P. dupliciocellatus* (27.5 μg g^{-1}) > *L. bohar* (24.8 μg g^{-1}) > *T. jarbua* (23.1 μg g^{-1}); that for arsenic (over 1 μg g^{-1}) was *A. australis* (2.15 μg g^{-1}) > *M. cyprinoides* (1.15 μg g^{-1}); and that for tin (over 3 μg g^{-1}) was *L. bohar* (3.60 μg g^{-1}). For the pelagic fishes, high values of nickel (44.3 μg g^{-1}) and chromium (35.7 μg g^{-1}) were found in *S. jello*.

It is interesting to note from Figure 2 that, except tin, the contents of copper (126 μg g^{-1}), zinc (469 μg g^{-1}), cadmium (1.36 μg g^{-1}), lead (12.3 μg g^{-1}), nickel (58.4 μg g^{-1}), chromium (11.8 μg g^{-1}) and arsenic (3.87 μg g^{-1}) in *S. guttotus* collected in February 1998 were higher than those contents (Cu, 33.8 μg g^{-1}; Zn, 321 μg g^{-1}; Cd, 0.15 μg g^{-1}; Pb, 4.45 μg g^{-1} Ni, 4.3 μg g^{-1}; Cr, 7.5 μg g^{-1}; and As, < 0.005 μg g^{-1};

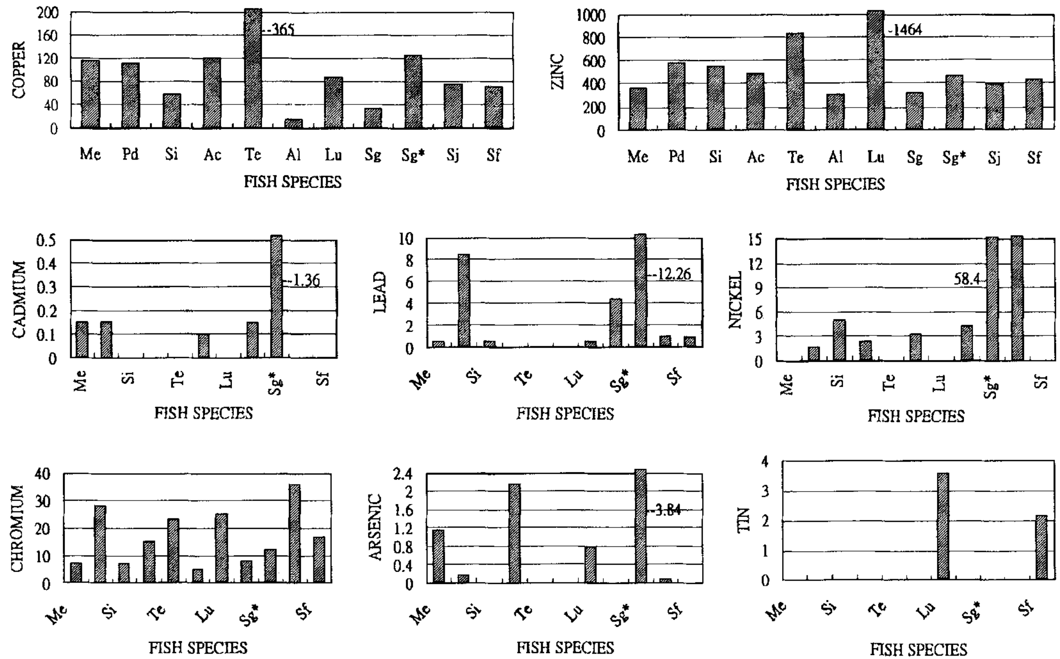

FIGURE 2 Heavy metal concentrations ($\mu g\, g^{-1}$, dry weight) in different species of fish collected from the southwestern coast (Erhjin Chi area) of Taiwan on August 24–31, 1997 [**Demersal fish**: detritus feeder, Me = *Megalops cyprinoides*, Pd = *Pseudorhombus dupliciocellatus*; detritus and benthic organism feeder: Si = *Sillago sihama*, Ac = *Acanthopahrus australis*, Te = *Terapon jarbua*, Al = *Acanthopagrus latus*; benthic organisms feeder: Ce = *Cephalopholis miniata*, Lu = *Lutjanus bohar*; herbivorous feeder: Sg = *Siganus guttotus* (* Collected on February 17–20, 1998). ***Pelagic fish***: Sj = *Sphyraena jello*, Sf = *Sphyraena forsteri*].

and Sn, < 0.53 μg g^{-1}) in *S. guttotus* collected in the same area in August 1997. The tin concentrations in *S. guttotus* collected in August 1997 and February 1998 were under the detection limit (< 5.3 μg g^{-1}). The higher concentrations of heavy metals in fishes collected in February might be due to the rainy season that dominates the distributions of trace metals. The rainy season (mainly contributed by storm rainfall) usually begins from March/April; and the dry season begins from September/October. Stream flow in the dry season is much less than that in the rainy season. Shortage of water usually occurs from January through March. During the dry season, the river bed (mainly mud) absorbs trace metals discharged freely from domestic and industrial sources without any treatment. Immediately after heavy rain, trace metals in the river sediments are released into the river water and then discharged to the estuarine and coastal environments. After heavy rain, trace metals continue to accumulate in the sediments as the input from pollution sources continues. Therefore, the higher the precipitation, the lower the concentration of trace metals was observed in sediments. For example, along the Erhjin Chi coastal area, the precipitation of 425.1 mm in July and 464.9 mm in August of 1997 were much higher than those of 15.5 mm in December of 1997, 44.6 mm in January and 100.3 mm in February of 1998 (Fig. 3). The concentrations (dry weight) of copper (152 μg g^{-1}), zinc (387 μg g^{-1}), cadmium (0.60 μg g^{-1}), lead (110 μg g^{-1}), nickel (308 μg g^{-1}), chromium (244 μg g^{-1}), arsenic (7.07 μg g^{-1}) and tin (1.85 μg g^{-1}) in sediments collected in February of 1998 were much higher than those values (Cu, 45.1 μg g^{-1}; Zn, 61.5 μg g^{-1}; Cd, 0.04 μg g^{-1}; Pb, 1.2 μg g^{-1}; Ni, 60.5 μg g^{-1}; Cr, 11.8 μg g^{-1}; As, 4.31 μg g^{-1}; Sn, 1.30 μg g^{-1}) obtained in August of 1997. The higher the concentrations of heavy metals in sediments, the higher the contents of heavy metals in demersal fishes were also observed. Therefore, it was quite reasonable that the concentrations of heavy metals in demersal fishes caught in February of 1998 were higher than those in August of 1997.

Figure 3 also indicates that the concentrations of heavy metals in southeastern (Green Island, a relatively unpolluted area) coastal sediments were much lower than those in the southwestern (Erhjin Chi, a seriously polluted area) coastal sediments in August of 1997. Unfortunately, no data was available for the heavy metal contents in the same fish species collected from both polluted and less polluted areas. Only two species of demersal fishes, *C. miniata* (a benthic organism

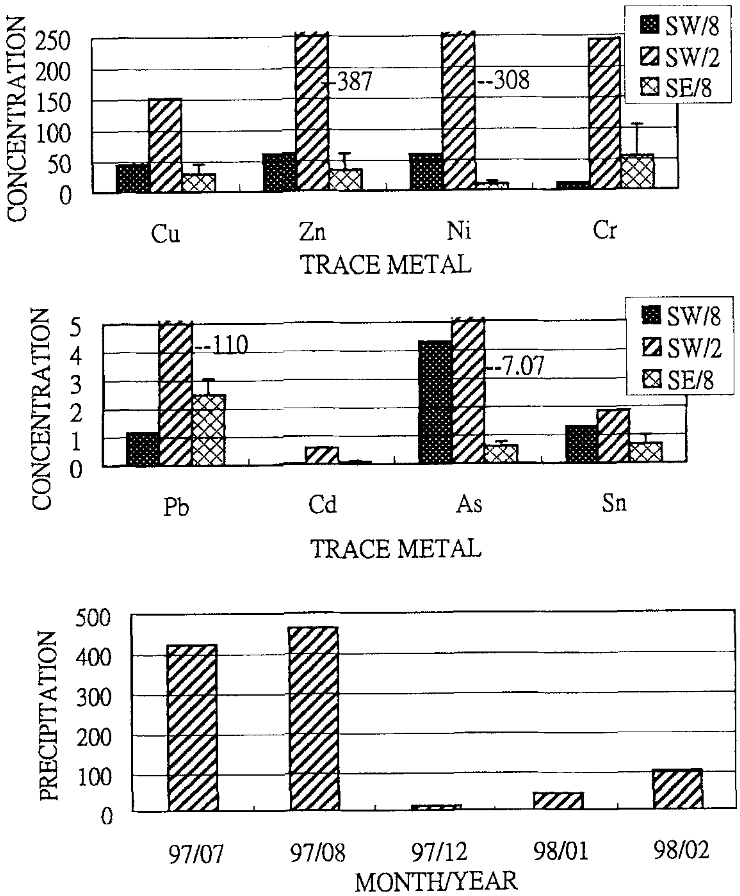

FIGURE 3 Concentrations ($\mu g\, g^{-1}$, dry weight) of trace metals in sediments from the southwestern (Erhjin Chi area) southeastern (Green Island area) coasts of Taiwan and precipitation (mm) in August of 1997 and February of 1998. Precipitation in Erhjin Chi is shown below.

feeder) and *S. forsteni* (a herbivorous feeder), were caught along the coast of Green Island. Figure 4 shows the results that the concentrations of copper (42.8 $\mu g\, g^{-1}$), zinc (569 $\mu g\, g^{-1}$), cadmium (7.4 $\mu g\, g^{-1}$) and arsenic (5.95 $\mu g\, g^{-1}$) in *C. miniata* were higher than those concentrations (Cu, 29.4 $\mu g\, g^{-1}$; Zn, 329 $\mu g\, g^{-1}$; Cd, 4.95 $\mu g\, g^{-1}$; and As, 5.2 $\mu g\, g^{-1}$) in *S. forsteni*. However, the concentrations of lead, nickel, chromium and tin in *C. miniata* were lower compared with those values in *S. forsteni*.

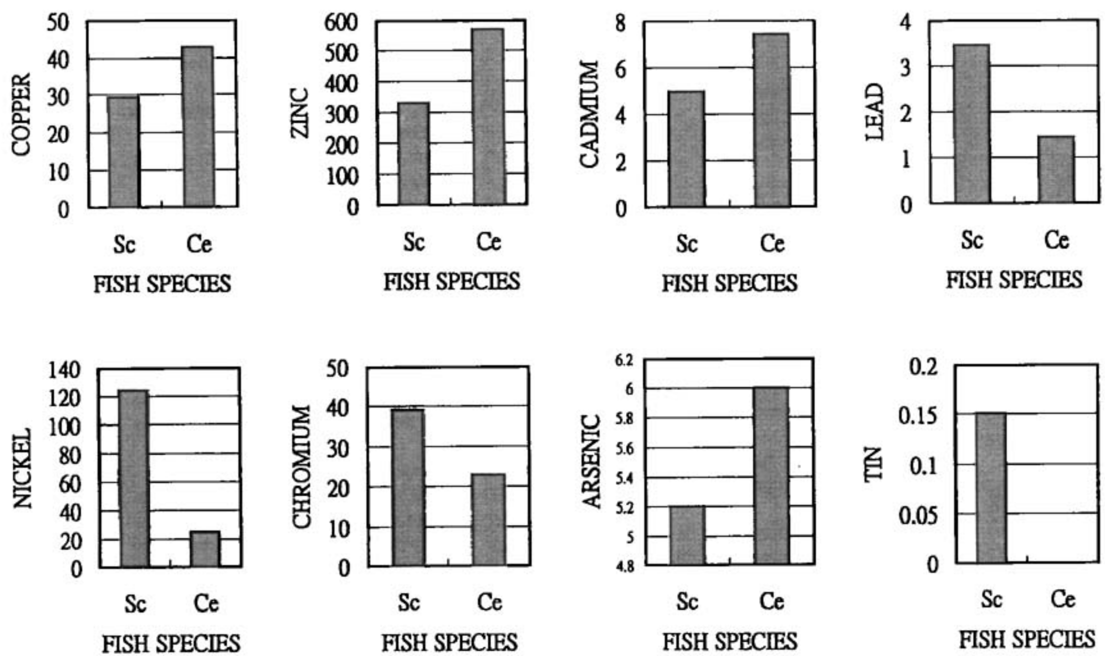

FIGURE 4 Heavy metal concentrations ($\mu g\, g^{-1}$, dry weight) in two species of demersal fish collected from the southeastern coast (Green Island area) of Taiwan in August of 1997: Sc = *Scarus forsteni*, a herbivorous feeder; Ce = *Cephalopholis miniata*, a benthic organism feeder.

Trace metal contents in different fish tissues such as gills, guts, livers, eggs and flesh have been analyzed. The results (Fig. 5) indicate that the concentrations of metals vary with the organ tissues of different fish species. For example, cadmium (100%), copper (82%) and lead (73%) are mainly contained in the liver of *M. cyprinoides*; cadmium (100%), nickel (100%) and arsenic (100%) in the flesh and lead (95%), copper (92%) and chromium (81%) in the liver of *P. dupliciocellarus*; lead (100%) in liver and nickel (100%) in the eggs of *S. sihama*; lead (100%) in the liver and nickel (100%) in the flesh of *A. australis*; copper (99%) in

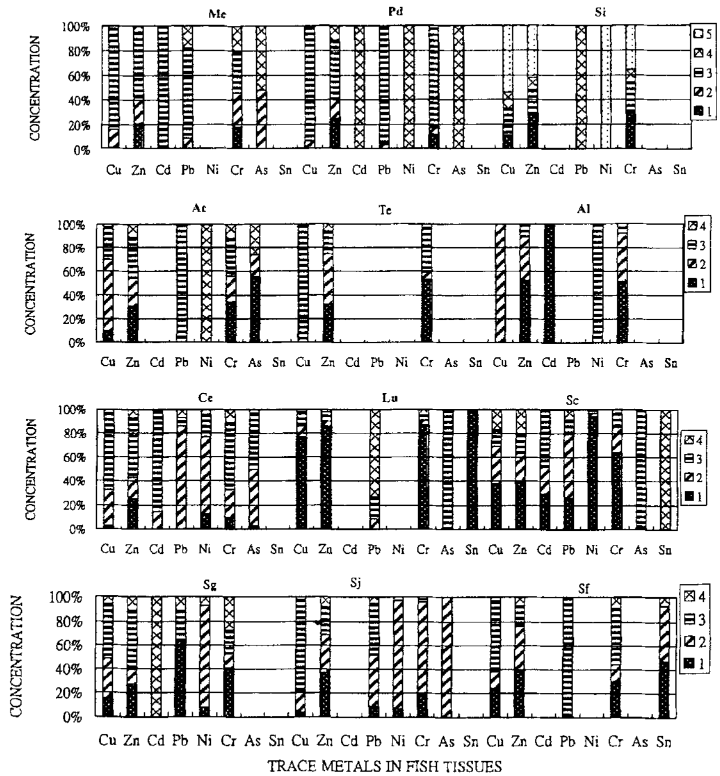

FIGURE 5 Relative proportions (expressed relative to the total concentration of one metal found in all tissues that is normalized to 100%) of heavy metals in tissues of different fish species collected from the southwestern coast (Erhjin Chi area) of Taiwan in August of 1997 **Demersal fish**: detritus feeder: Me = *Megalops cyprinoides*, Pd = *Pseudorhombus dupliciocellatus*; detritus and benthic organisms feeder, Si = *Sillago sihama*, Ac = *Acanthopagrus australis*, Te = *Terapon jarbua*, Al = *Acanthopagrus latus*; benthic organism feeder: Ce = *Cephalopholis miniata*, Lu = *Lutjanus bohar*; herbivorous feeder: Sg = *Siganus guttotus*; **Pelagic fish**: Sj = *Sphyraena jello*, Sf = *Sphyraena forsteri*; 1: Gills; 2: Guts; 3: Livers; 4: Flesh; and 5: Eggs.

the liver of *T. jarbua*; nickel (100%) in the liver, copper (100%) in the gut and cadmium (100%) in the gills of *A. latus*; lead (86%) in the gut and cadmium (84%) in the liver of *C. miniata*; arsenic (100%) in the liver and tin (99%), chromium (87%), zinc (86%) and copper (77%) in the gills of *L. bohar*; tin (100%) in the flesh and arsenic (97%) in the liver and nickel (94%) in the gills of *S. forsteni*; and cadmium (100%) in the flesh and nickel (84%) in the gut of *S. guttotus*. In pelagic fish, higher concentrations of arsenic (100%) and nickel (90%) were found in the guts and copper (77%) in liver of *S. jello*; lead (100%) in the liver of *S. forsteri*. Regressions of trace metal concentration in fish and that in different tissues, such as gills, guts, livers, flesh and eggs, were made (Tab. I). In general, strong correlations of tin (0.9119, $p < 0.01$) and zinc

TABLE I Linear regressions between the total concentration of trace metals in fish (X) and the concentration of trace metals in different tissues (Y) of fish collected from the Taiwan southwestern coast in August of 1997

Tissue	*Metal*	*Regression*	R^2
Gills	Sn	$Y = 0.8630X - 0.07490$	0.9119($p < 0.01$)
	Zn	$Y = 0.8942X - 234.95$	0.8525($p < 0.01$)
	Cr	$Y = 0.2096X + 2.2389$	0.3053
	Cd	$Y = 0.0831X + 0.0593$	0.2785
	Pb	$Y = 0.1316X + 0.0700$	0.1360
	Ni	$Y = 0.0737X + 4.3152$	0.0179
	Cu	$Y = -0.020X + 12.537$	0.0103
	As	$Y = 0.0036X + 0.1527$	0.0008
Gut	As	$Y = 0.4012X + 0.031$	0.9067($p < 0.01$)
	Cd	$Y = 0.1312X + 0.0062$	0.8804($p < 0.01$)
	Ni	$Y = 0.3464X + 1.9368$	0.6301($p < 0.05$)
	Cr	$Y = 0.5125X - 3.9430$	0.5816($p < 0.05$)
	Pb	$Y = 0.2136X + 0.3900$	0.1953
	Sn	$Y = 0.1072X + 0.0819$	0.1568
	Zn	$Y = 0.0486X + 90.223$	0.0413
	Cu	$Y = -0.0095X + 15.893$	0.0023
Liver	Cu	$Y = 1.0184X - 28.577$	0.9228($p < 0.01$)
	Cd	$Y = 0.7154X + 0.0280$	0.9099($p < 0.01$)
	Pb	$Y = 0.804X - 0.5014$	0.7794($p < 0.01$)
	As	$Y = 0.5705X + 0.0888$	0.7745($p < 0.01$)
	Ni	$Y = 0.5053X - 1.4292$	0.5422($p < 0.05$)
	Zn	$Y = 0.0412X + 123.12$	0.0187
	Cr	$Y = 0.037X + 4.8061$	0.0072
Flesh	Ni	$Y = 0.0198X + 0.8900$	0.2185
	Sn	$Y = 0.0230X + 0.0162$	0.2125
	Pb	$Y = 0.0201X + 0.1176$	0.0688
	Cu	$Y = -0.003X + 2.7980$	0.0226
	Cr	$Y = 0.0067X + 1.3375$	0.0146
	As	$Y = 0.0055X + 0.1185$	0.0049
	Zn	$Y = 0.0022X + 42.203$	0.0012
	Cd	$Y = -0.0007X + 0.0301$	0.0009

(0.8525, $p < 0.01$) were found in gills; those of other metals (As (0.9067, $p < 0.01$), Cd (0.8804, $p < 0.01$), Ni (0.6301, $p < 0.05$) and Cr (0.5816, $p < 0.05$)) were in guts; and those other metals [Cu (0.9228, $p < 0.01$), Cd (0.9099, $p < 0.01$), Pb (0.7794, $p < 0.01$), As (0.7745, $p < 0.01$) and Ni (0.5422, $p < 0.05$)] were in livers. No significant correlation was found between total trace metal in fish and fish flesh. For egg analysis, unfortunately, only the data of *S. sihama* was available. The results indicated that 100% of nickel and 100% of lead were obtained in eggs and livers, respectively.

CONCLUSIONS

Concentrations of trace metals vary between metals in different species, depending on their diet. Some correlations exist between their zinc in gills. No significant effect between the total trace metals in fish.

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