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## **SOURCE AND EFFECT OF HEAVY METAL POLLUTION IN THE COASTAL TAIWAN SEDIMENTS**

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Coastal sediments and oyster farming is severely affected by the industrial effluents in the coastal region near LuGong, Taiwan. Surficial sediments and oyster samples were collected from a coastal area near a major electro-plating industry. Spatial variation in heavy metal contents in sediments was studied in relationship to the main pathway of contamination and geochemical association with natural constituents of the sediments.

Spatial trends of heavy metals in the sediment reflected inputs of pollutants from the poorly-treated electro-plating effluents in the surrounding county and transported downstream by the Yang-tse-tru River. Effluent related heavy metal (Zn, Cu, Cd) is extremely high in the study region sediments. The highest concentrations were observed near shore, especially upstream in the Yang-tse-tru River sediments. Heavy metal concentrations decreased rapidly in a seaward direction. Heavy metal concentrations in near shore sediments were as high as some highly industrialized harbour sediments in the USA (e.g. Boston and San Diego).

Significant differences and high concentrations of heavy metals were also observed in oysters from the study region. Heavy metals from the study region were two to five times higher than in oysters from other pristine areas in Taiwan.

KEY WORDS: Oysters *(Crassostrea* gigas), zinc, copper, cadmium, sediments.

#### INTRODUCTION

Heavy metals in the surface sediments often represent a combination effect of chemical, biological and physical processes occurring in the coastal area. Surface sediments integrates through time these chemical and physical changes that occur in the water column and acting both as a repository and sources of suspended materials. Spatial variation of heavy metal in the surface sediments and their vertical changes are the results of these processes. Any changes act upon the chemical, physical and biological processes may, therefore, record its signature in the sediments. Accumulation of heavy metal in surface sediment from nearby related industry effluents discharged into the river without much prior cleaning will easily be identified through sediment heavy metal study.

Taiwan is currently undergoing a period of rapid development and industrialization. Unfortunately, most urban sewage and industrial effluents are discharged directly into the river illegally and subsequently into the coastal area. Furthermore, a number of urban waste dumps are situated on the beach front. The coastal area and estuaries are at a high risk of being polluted. However, since very few efforts were given to study their potential impact on the coastal Taiwan, the extent to which this potential hazard might have on the environment is not clearly understood. This study investigates the spatial variation of sedimentary heavy metals near the coastal city of LuGong. Through a close examination of heavy metals in the sediment and oysters from the study region and other pristine coastal area, the input of pollutant and its effect on the nearby marine biota is discussed.

#### STUDY AREA AND METHODS

LuGong, a small city situated in the central Taiwan with a population of only  $\sim$  80,000, and its adjacent area has a large metal plating industry in Taiwan (IDB, 1994). Oyster farming is a major business in the local community prior to the recent development.

Surface sediments were collected using a Van-Veen type grab (Fig. 1). Sediments were stored in the polyethylene (PE) bottles at  $4^{\circ}$ C in the field and frozen in the laboratory until drying. Sediments were freeze-dried using a Labconco freeze-dryer (Lyph-Lock, 6 1) for one week, and then ground to fine powder using an agate mortar and stored in PE vials. Untreated sediments were sieved through a 63  $\mu$ m Standard Sieve (Endecotts, England) to separate the sand size grain. The fraction smaller than the 63  $\mu$ m were determined by the standard pipette technique (Folk, 1974). The precision for the sand (diameter  $d > 63 \text{ µm}$ ), silt  $(4 < d < 63 \text{ µm})$  and clay  $(d < 4 \mu m)$  is 0.37%, 0.96%, and 1.56% respectively.

Oyster (Crassostrea gigas) samples were collected directly above each sediment samples from the oyster farming rack. **As** a comparison, another set of oyster samples were collected at the same season in an other coastal areas. Sediments adhere to the oyster shell were washed thoroughly in the field. Samples were then packed in an ice-chest, maintained at  $4^{\circ}$ C, shipped back to laboratory and shucked. Each individual oyster was washed with deionized water (Milli-Q,  $\sim$  18  $\Omega$ ), dried and stored frozen in the pre-cleaned glass bottle until digestion.

Sedimentary metals (Fe, Mn, Cu, Zn, Pb, and Cd) were extracted with 1N HCl (Fisher) for 16 hours (Huerta-Diaz **el** *al.,* 1993; Di Tor0 *el a/.,* 1990). Clear supernatant was separated from the remaining residue with a centrifuge for 15 minutes at 4000 rpm and stored in acid clean PE vials for metal analysis. Oyster samples were digested with concentrated nitric acid (Fisher, ACS) in a water bath (90°C) for 4 hours. The remaining residue was filtered with 0.45 m Nucleopore filter. Clean extractant was stored in acid-cleaned PE vials for metal analyses.

Heavy metals (Fe, Mn, Zn: flame; Cu, Pb, Cd: graphite) were determined using a Hitachi 81002 atomic absorption spectrometer. For metals high in concentrations, i.e., iron, manganese and zinc, the extracted solution was diluted ( $20 \sim 50$  times) with deionized water and the concentrations were determined by calibration curves prepared from Merck Standard Metal Solution. For copper, lead and cadmium, the standard addition technique was employed in order to minimize the matrix effect.



**Figure 1** Study area and sampling sites.

The accuracy of the metal analysis was determined by total dissolution of the NIST-1646 and **BCSS-1** Standard Sediment (Tab. I). Relative errors, as compared to the Standard Sediments, were better than 6% for most metals except copper and zinc **(12%)** of the **BCSS-1.** The metals extracted represented 80-100% of total

	<b>NIST1646</b>		BCSS-1	
	Certified	This study ( $n = 8$ )	Certified	This study ( $n = 10$ )
$Al($ %)	$6.25 + 0.20$	$6.82 + 0.39$	$6.26 + 0.22$	$6.97 + 0.30$
Fe (%)	$3.35 + 0.05$	$3.45 + 0.23$	$3.29 + 0.1$	$3.43 \pm 0.14$
$Mn$ (ppm)	$375 + 20$	$354 + 8$	$229 + 15$	$232 + 9$
$Zn$ (ppm)	$138 + 6$	$130 + 8$	$119 + 12$	$114 + 6$
$Cu$ (ppm)	$18 + 3$	$16.9 \pm 2.5$	$18.5 + 2.7$	$16.3 + 1$
$Pb$ (ppm)	$28.2 + 1.8$	$28.6 + 1$	$22.7 + 3.4$	$23.4 + 1.1$
$Cd$ (ppm)	$0.36 + 0.07$	$0.38 + 0.03$	$0.25 + 0.04$	$0.28 + 0.02$

**Table I** The analytical accuracy and precision **of** this study **using** the NIST and **BCSS** Standard Sediments.

manganese and cadmium and approximately 25-60% of total iron, zinc, cadmium, lead in sediments (Chen, 1992). Similar method has been used in evaluating metal bioavailability in contaminated sediments (Ankley *ef ul.,* 1991, 1993).

#### RESULTS AND DISCUSSIONS

#### *Distribution of Heaziy Metals*

Heavy metal (Fe, Mn, Cu, Zn, Pb, Cd) concentrations varied greatly in sediments from various locations in the study area (Fig. 2). The highest concentrations were observed in sediments from the Yang-tse-tru River and the lowest at stations near the northwestern part (seaward) of the study area. For the rest of the study area, heavy metal concentrations decreased rapidly from the Yang-tse-tru River estuary seaward to the west, probably reflecting a dilution effect with other sediments. Low metal concentrations were occasionally observed in the centre of the study area where a patch of coarse grained sediment was also found.

#### *Heavy Metals and Grain Size*

Not all metals were grain-size controlled in the study region (Fig. 3). Even though grain-size was found to be one of the major factors controlling heavy metal distribution in coastal Taiwan (Huang and Lin, 1994) and other environments (e.g. Felipek and Owen, 1979; Gibbs, 1977; Oliver, 1973; and Goldberg, 1954), only manganese and lead were grain-size controlled. Huge deviaitons of grain-size/metal concentration relationship from the typical coastal Taiwan sediments were found on cadmium and copper. Zinc and iron differed slightly from the normal distribution (Fig. 3). This trend reflects a combination effects of a possible industry pollution with a natural constitutents of the sediments.

#### *Source of Heavy Metals*

A detailed examination of the spatial distribution of heavy metal in the study region revealed that Yang-tse-tru River may be the primary source of heavy metals. Concentrations of most metals decreased rapidly away from the river mouth towards the sea (Fig. 2). High concentrations of cadmium and copper were found upstream in the Yang-tse-tru River sediments. Cadmium and copper concentrations were much higher than sediments from other coastal areas with similar grain-size (Fig. 3). In fact, they are as high as those found in sediments near the major USA industrial region (e.g. Boston harbour, San Diego Bay) (NOAA, 1991).

The only possible source of this high concentrations of heavy metal is the poorlytreated effluents of the electroplating factories with copper, zinc and cadmium being the major constituents. There are more than 700 factories in LuGong and its surrounding county, compared to an average of approximately 50 for other counties in Taiwan (IDB, 1994). These metals are rapidly accumulating in sediments and may have profound effects on the surrounding biota.



**Figure 2** Spatial variations of sediment grain size  $(\%$  silt), %S, Cu, Cd, Zn, and Pb.



**Figure 3** Relationship between metal concentrations and % fine-grained sediments (grain size  $< 64 \, \mu m$ ). Fe, Mn and Pb generally followed the typical pattern observed in other coastal sediments in Taiwan. Cu, Cd and Zn deviate significantly from the typical distribution. (0: this study. Straight line is the typical grain-size controlled metal concentration from Huang and Lin (1994)).

#### *Heavy Metals in Oysters*

Heavy metal concentrations in the oysters of the study region are significantly higher than those found in other coastal areas (Fig. **4).** With the exception of lead, heavy metals are about 2 to 5 times higher than the oyster from other areas with similar weight. Zinc concentrations are as high as those in the northern Chesapeake Bay oysters which were influenced by the nearby Baltimore and Hampton Roads harbours (Sinex and Wright, 1988). High concentrations of heavy metals found in the study region sediments may be the cause of deterioration in local oyster production. A great number of oyster farms were already abandoned prior to our sampling (Fig. 1). The corresponding elevation of heavy metals (Cd, Cu and Zn) in oyster and sediments indicates that the industrial effluents from the nearby region are the primary source of pollutant input in the area.

**A** number of studies had indicated that metals/AVS ratio is a good normalization parameter in evaluating the possible toxicity of polluted sediments to the benthic organisms when acid-volatile sulphide is the predominating solid phase sulphide (Di Tor0 *et al.,* 1990, 1992; Ankley *et al.,* 1991, 1993). Pyrite is the predominant reducing sulphide found in the study region (Fig. 2). As a result, no direct toxic effect was indicated based on their method, with the calculated metals/AVS ratio less than one. However, Morse (1994) showed that pyrite related heavy metal is an important



**Figure4** Heavy metal concentrations (Zn, Cu, **Pb** and Cd) of the each individual oyster. Oyster weight is the wet weight prior to drying. (0: oyster **from** this study area, **x** : other pristine area.)

source of potential bioavailable metal. He indicated that a major portion of pyrite bound metal in the original anoxic sediments can be released within a day or less during resuspension. Our preliminary data also showed that heavy metals in resuspended particle and oysters increase rapidly during winter monsoon season. The exact relationship between heavy metal in biota and sediments is beyond the scope of this study. However, the fact that both the sediments and the oysters in the region showed unusually high concentrations of heavy metal indicate that the coastal area under investigation is severely polluted by the industrial effluent transported through the Yang-tse-tru River.

#### **CONCLUSION**

Concentrations of heavy metal in the coastal LuGong area and adjacent Yang-tsetru River sediments were unusually high, especially in upstream sediments. Copper, cadmium and zinc were about two- to five-folds higher than sediments from other pristine coastal areas in Taiwan. The poorly-treated electro-plating industry effluents discharged into the Yang-tse-tru River probably are the source of heavy metals in the study region.

Regional oyster farming is severely affected by the industrial effluent pollutant. Extremely high concentrations of metals (e.g. Zn, Cu, Cd) were observed in the oysters from this region, a sharp contrast to oysters from other pristine area in Taiwan which showed no signs of pollution. Concentrations of heavy metals from the study region were similar to those from areas near the highly industrial harbours such as Baltimore in the **USA.** Regulation in the quantity and quality of industrial effluents in the region is urgently needed.

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