

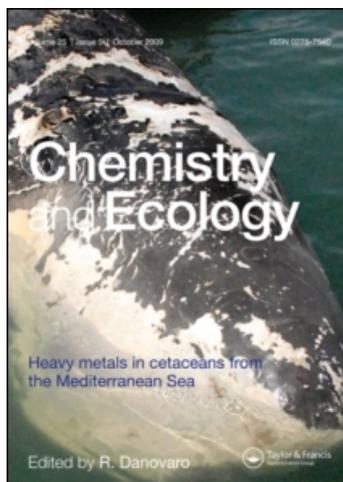
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SPATIAL VARIATIONS OF HEAVY METALS IN THE EAST CHINA SEA CONTINENTAL SHELF SURFACE SEDIMENTS

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Surface sediments collected from continental shelf of the East China Sea were analyzed for heavy metals (iron, manganese, copper, zinc, lead, cadmium), carbonate, organic carbon contents, and grain sizes. The range of concentrations observed were iron: 0.3–1.3 wt%, manganese: 2.3–14 $\mu\text{mole/g}$, copper: 7.1–184 $\mu\text{mole/g}$, zinc: 0.16–0.77 $\mu\text{mole/g}$, lead: 15–98 $\mu\text{mole/g}$, cadmium: 0.17–3.9 $\mu\text{mole/g}$, carbonate: 3.6–87 wt%, sand: 10–100%, silt: 0–70% and clay: 0–50%.

A zonal distribution pattern of the heavy metals was found in the East China Sea Continental shelf sediments. High concentrations of most heavy metals, organic carbon and fine-grained sediments were observed in the inner shelf zone, especially those near the discharge of the Yangtze River. Concentrations of these heavy metals decreased from the inner shelf to the shelf break region. High concentrations of metals were also found in sediments near Taiwan. Iron concentrations decreased north-east of the central shelf region. High concentrations of cadmium were found in the shelf break region where biogenic carbonate is predominant. This study showed that biogenic carbonate in the East China Sea shelf break region and the terrigenous sediments from the Yangtze River and island of Taiwan were the major sources of heavy metals. Heavy metal concentrations were strongly influenced by the content of the coarse-grained quartz sand present in the sediments.

Keywords: Spatial variation; heavy metals; reactive metals; sediments; East China Sea Continental shelf

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INTRODUCTION

Sediments are an ultimate sink for river-derived heavy metals discharged to the aquatic environment (Turekian, 1977). The spatial distribution of heavy metals in sediments reflects the extent of chemical, physical and sedimentary controls on their supply, deposit and removal from the ocean. Major controls include the composition of suspended particles delivered from the river, the biogeochemical cycling of the metals in the ocean, physical condition of the bottom sediment, and the deposition environment where significant diagenetic metal recycling and reprecipitation may occur. In addition, the anthropogenic source of the heavy metals may also play a major role in their distribution in coastal sediments. Investigation of heavy metals distribution in sediments, therefore, is of prime importance in identifying and interpreting the source and pathway of sedimentary heavy metals.

East China Sea is one of the world's largest continental shelf systems. Its dominating source of sediments include the Yangtze river (480×10^6 tons/year) and the Huanghe River (1100×10^6 tons/year) (Milliman and Meade, 1983). Major deposition areas for the fine-grained sediments originating from the Huanghe River are the central Yellow Sea, the Gulf of Bohai, and the area around Shandong Peninsula (Lee and Chough, 1989). For the Yangtze River, approximately 40% of the total suspended load were deposited in the Yangtze estuary (Milliman *et al.*, 1985). According to these studies, shoreline progression within the Yangtze estuary during the past 2000 to 3000 years has accounted for less than 5% of the river's load. Most sediment is temporarily deposited offshore and later resuspended and transported southward by subsequent winter storms (Milliman *et al.*, 1985; McKee *et al.*, 1983; DeMaster *et al.*, 1985). Our understanding of the influence of Yangtze River on the East China Sea Continental shelf is still limited in the region near the river mouth. Even among the few existing data, most information was derived mainly from either sediment texture or river discharge. Very few sediment data are available now for the broader East China Sea Continental shelf. The purposes of this study were to examine the distribution of heavy metals in the East China Sea Continental shelf, and to identify the major sources of sedimentary heavy metals in the study region based on their spatial characterization.

SAMPLING AND METHODS

Surface sediments (Fig. 1) were collected on board R/V *Vinogradov* using a Van-Veen type grab in July-August, 1992. Box core sediments were collected on board R/V Ocean Researcher I, cruise 293 (August, 1991) and 322 (July, 1992). Upon retrieving the core on deck, sediments were subsampled and/or subcored, sectioned into two centimetre sections with a plastic spatula and stored frozen in the polyethylene (PE) bottles. Sediments from 0 to 2 cm were used in this study and defined as the surficial sediment.

Sediments were freeze-dried using a Labconco freeze-dryer (Lyph-Lock, 6 l) for one week, and then ground to fine powder using an agate mortar and stored in PE vials. Well-mixed subsamples were analyzed for reactive metals (Huerta-Diaz and Morse, 1992), organic carbon, and inorganic carbon. Untreated sediments were sieved through a 63 μm standard sieve (Endecotts, England) to separate the sand size grain. The fraction smaller than that of 63 μm were determined by the standard pipette technique (Folk, 1974). The precision for clay (diameter $d < 63 \mu\text{m}$), silt ($4 > d > 63 \mu\text{m}$) and sand ($d > 4 \mu\text{m}$) is 0.37%, 0.96% and 1.56% respectively.

Total carbon and organic carbon were analyzed using a LECO C/S Analyzer (SC-444) equipped with a high temperature resistance furnace and a IR detector. Approximately 0.3 g of dry sediments was combusted at 1400 °C with high purity oxygen to determine the total carbon content. Organic carbon was determined with pre-acidified (~ 2 ml 6N HCl) and later hot-plate dried (~ 50 °C) sediments. Calcium carbonate content was calculated from the difference between the total carbon and organic carbon content assuming calcite and aragonite were the only carbonate phases. The precision for the total carbon and organic carbon is 1.2% and 3% respectively.

Sedimentary metals (Fe, Mn, Cu, Zn, Pb and Cd) were extracted using a method based on Huerta-Diaz (1990) with 1 N HCl (Fisher) for 16 hours. 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 analyses. Heavy metals (Fe, Mn, Zn: flame; Cu, Pb, Cd: graphite) were determined using a Hitachi 8100Z atomic absorption spectrometer. For metals high in concentrations, i.e., iron, manganese and zinc, the extracted solution was diluted

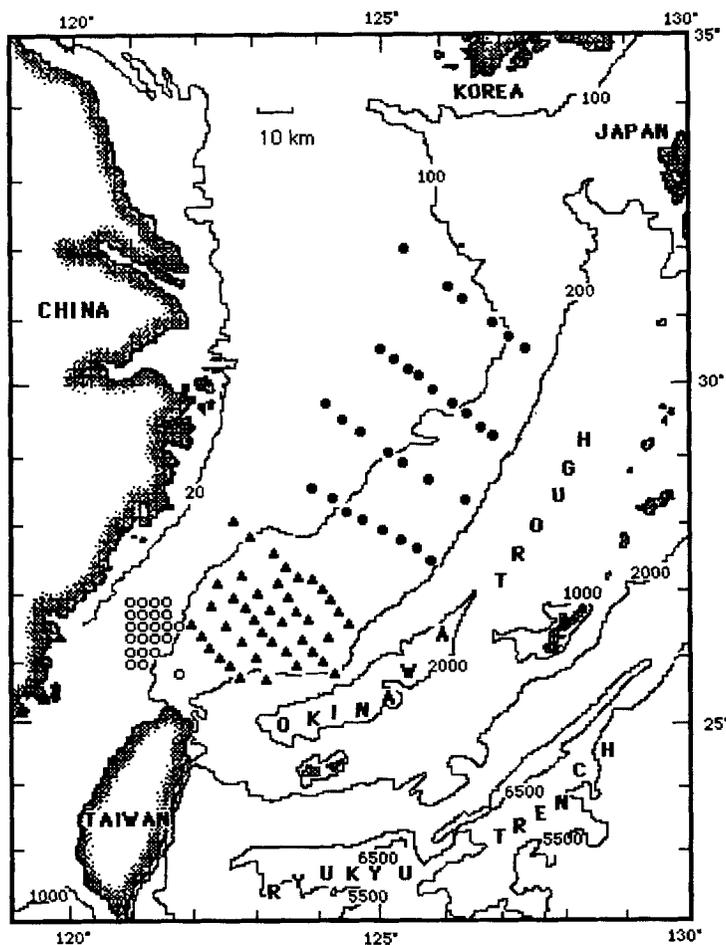


FIGURE 1 Study area and sampling stations in the East China Sea. Sampling cruises: ▲-R/V OR-91.293; ○-R/V OR-92.322; ●-R/V Vinogradov-1992. (Contours in metres).

(20 ~ 50 times) with deionized water (Milli-Q, ~ 18Ω) 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. 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 other than copper and zinc (12%) of the BCSS-1. The metals extracted represented 80–100% of total manganese, cadmium and approximately 25–60% of total iron, zinc, copper, lead in sediments (Chen, 1992). No significant variations were observed between total levels and this method in previous tests. As a result, this extraction scheme was chosen for its simplicity and less time consuming in conducting analyses.

RESULTS

Calcium Carbonate

Calcium carbonate was most abundant in the sediments of the outer continental shelf (Fig. 2). Values between 15 and 25% were common on the outer shelf; and values greater than 25% were observed mostly in areas directly under the route of the Kuroshio Current. The highest concentration, up to 90%, was found in the upwelling region north of Taiwan. The concentrations of calcium carbonate for other areas of the East China Sea Continental shelf were in the range of 5 and 15%, much less than those found in the South China Sea (Calvert *et al.*, 1993). The concentration and distribution pattern of our findings are similar to those observed by Niino and Emery (1961).

Organic Carbon

The distribution of organic carbon in the East China Sea Continental shelf sediments (Fig. 2) shows that the concentrations are higher in

TABLE I The analytical accuracy and precision of this study using the NIST and BCSS Standard Sediments

	NIST1646		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 ± 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 ± 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

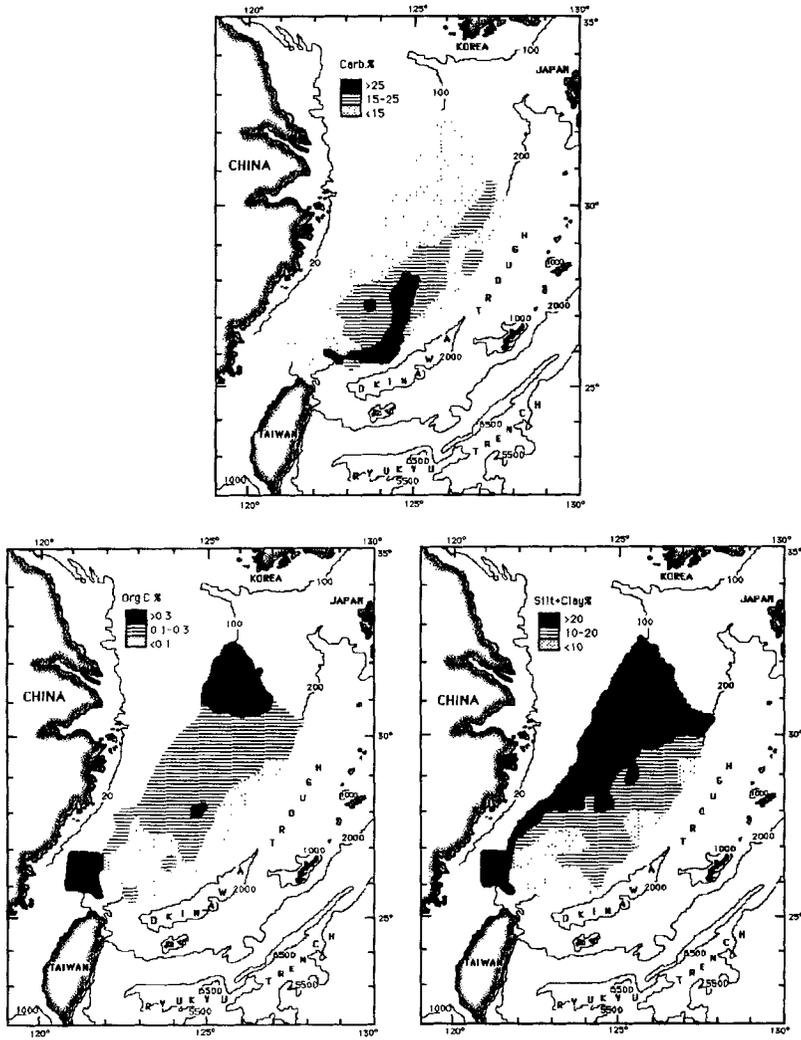


FIGURE 2 Sediment properties (calcium carbonate (wt%), organic carbon contents and grain size) of the East China Sea Continental shelf sediments. Only fine-grained sediments (%Silt + % Clay = diameter less than $63 \mu\text{m}$) were plotted in this figure.

samples from the northern and southern part of the sampling area where fine-grained sediments often exceed 40%. In general, the organic carbon concentration decreased seawards, reflecting a correspondent increase in the size of sediment. More than 0.5% of organic

carbon were found in areas where the fine-grained sediments exceeded 70%. For the majority of the East China Sea Continental shelf sediments, the organic carbon concentration was between 0.1–0.3%. In some cases less than 0.1% organic carbon was found in areas where approximately 90% of the sediments were composed of coarse grained quartz sand or carbonate shell debris.

Grain-sizes

The distribution of fine-grained sediments (diameter less than $63\mu\text{m}$) was presented in Figure 2. Fine-grained sediment was found in the area near the Yangtze River discharge, extending south to the inner shelf north of Taiwan, the mud belt of Niino and Emery (1961). Up to 90% of fine-grained sediment ($\sim 60\%$ silt and $\sim 30\%$ clay) was observed in the inner shelf region. Grain size appeared to be increasing seawards. Sand-size quartz was the predominant grain type in the outer continental shelf sediment. With the exception of the mud belt in the inner shelf, the average grain size was $77.8 \pm 20.1\%$ sand ($n = 85$).

Iron, Manganese and Zinc

The concentration of iron (Fig. 3) in the East China Sea Continental shelf sediments is in the range of 0.4–1.4 wt%. High concentrations of iron were found in the northern part of the East China Sea shelf region, mostly between 0.8 and 1%. Another area with high reactive iron concentration was observed in a region north of Taiwan with a concentration between 1 and 1.4%. The lowest iron was observed in an area where high concentration of carbonate was also observed (Figs. 2 and 3). Iron was as low as 0.3% with a carbonate content of more than 85%. For the majority of the East China Sea Continental shelf, iron content is within 0.4–0.8%. Reactive iron in the East China Sea is extremely high as compared to those in the South China Sea which are mostly composed of carbonaceous materials (Calvert *et al.*, 1993). In addition, iron is about 30 to 50% higher than those in the Gulf of Mexico (Huerta-Diaz and Morse, 1991).

The distribution of manganese (Figure 3) in the East China Sea shelf sediments is similar to the iron. High concentrations of manganese ($\sim 8\mu\text{mole/g}$) were found in the northern part of the study region

where carbonate contents were approximately 10 %. The majority of the shelf sediments were in the range of 2–5 $\mu\text{mole/g}$. Manganese concentrations in the region near Taiwan were slightly higher, $\sim 5\text{--}7$ $\mu\text{mole/g}$. Highest concentrations of manganese in this region were observed in sites (station 910936) with high carbonate contents (%Carb: 37%, Mn: $\mu\text{mole/g}$) and sites (station 920713) with low carbonate content (%Carb: 4.5%, Mn: 8.8 $\mu\text{mole/g}$).

The lowest reactive zinc found in the East China Sea Continental shelf sediments (Fig. 3) was in an area with the highest carbonate content. High concentrations of zinc were observed both in the northern and southern shelf region with high reactive iron. In most shelf sediments, the concentrations of zinc was in the range of 0.3 and 0.5 $\mu\text{mole/g}$, with an overall average of 0.42 $\mu\text{mole/g}$.

Copper and Lead

The concentrations of copper and lead decreased seawards (Fig. 3) from the inner shelf to the shelf break. High concentrations of copper were found in stations where fine-grained sediments were the major components. In areas where carbonaceous sediments are the major components, copper and lead were generally low.

Cadmium

Unlike other metals in this study, cadmium in the East China Sea Continental shelf sediments showed a unique distribution pattern (Fig. 3). The highest concentration of cadmium was found in the area northeast of Taiwan where a high concentrations of carbonate was also observed. Cadmium concentration decreased in northern areas toward the coast. A moderate high concentration of cadmium was also observed in the northern part of the study region.

DISCUSSION

Metal concentrations varied greatly in sediments from various geographic locations in the East China Sea Continental shelf region. The spatial distribution of heavy metals in the study region shelf sediments

revealed a number of distinct patterns: 1) the predominantly fine-grained terrigenous sediments with higher concentrations of heavy metals (Fe, Mn, Cu, Zn, Pb), and organic carbon; 2) the sandy terrigenous sediments with lower concentrations of heavy metals; 3) the biogenous coarse-grained carbonate sand and shell debris with higher concentrations of cadmium and lower iron, manganese, copper, zinc and lead; and 4) the fine-grained terrigenous sediments with high concentrations of iron north of Taiwan.

Heavy metals sources

High concentrations of heavy metals observed near the Yangtze River indicated that the primary source of heavy metals entering the East China Sea Continental shelf is provided by the Yangtze River sediments. With its high average suspended sediment concentration of 540 mg l^{-1} (Shen *et al.*, 1983) and an extremely high average water discharge of $\sim 30,000 \text{ m}^3\text{s}^{-1}$ (Cai, 1982), the Yangtze River is undoubtedly the only dominating source of heavy metal on the shelf. These sediments were deposited near the river mouth and later redistributed along the mud belt along the inner shelf, as indicated by the large differences between the long and short term sedimentation rate (DeMaster *et al.*, 1985). Sediments near the Yangtze River discharge in the study area were mostly composed of fine-grained terrigenous material with high concentrations of heavy metals (e.g., Fe: 1.0%, Mn: $7.79 \mu\text{mole/g}$, Cu: $81.0 \mu\text{mole/g}$, Zn: $0.581 \mu\text{mole/g}$, Pb: $58.7 \mu\text{mole/g}$). With the exception of zinc, most metals analyzed were about 10–100 times higher than in the South China Sea sediment where biogenic carbonate was predominant (Calvert *et al.*, 1993). The heavy metal distribution indicated unquestionably that the main source of the East China Sea shelf sediments is terrigenous in origin.

The unusually high concentration of iron near Taiwan and high concentration of cadmium in the carbonaceous sediments showed that detrital sediments from Taiwan and the biogenic carbonate present produced in the water column were also the major sources of heavy metals in the East China Sea shelf sediments. The annual sediment load for the combined rivers from Taiwan island is 300×10^6 ton/year, similar to the whole United States (Milliman and Meade, 1983). The sedimentation rate near the island, however, was lower. Only 0.37 cm y^{-1} was observed at one site in the largest river

mouth (Lee *et al.*, 1993). Most fine-grained sediments may have been transported by the prevailing northward current to the north (Boggs *et al.*, 1979).

The distribution of iron, in an elongated shape extending from Taiwan to the north shelf region (Fig. 3), indicated that Taiwan was the main source of iron in sediments. This was supported by Huang and Lin (1994) who found exceptionally high concentration of reactive iron (up to 2.2 wt%) in the coastal sediments near Taiwan. On the other hand, reactive iron in the area near the Yangtze River was relatively low, approximately only half of that observed in the coastal sediments near Taiwan. Given the fact that no other nearby source is capable of introducing such a considerable supply of sedimentary iron, it seems obvious that sediments from the rivers of Taiwan is also a main source of iron near Taiwan.

Sediments northeast of Taiwan are mostly composed of biogenic carbonate with very little detrital sediments. High concentrations of cadmium were observed in these sediments. Cadmium appeared to increase with increasing carbonate concentration (Fig. 3). Cadmium is a nutrient type metal being removed from surface water by plankton growth (Bruland, 1983). Hester and Boyle (1982) demonstrated that cadmium in the Foraminifera test reflect the chemical composition of the water they grow in. As a result, the upwelling of Kuroshio current in this region of the East China Sea Continental shelf not only increased the plankton production, but probably also sequestered cadmium away from the upwelled water and resulted in a cadmium enriched carbonaceous sediments

Dilution effects

Heavy metals as well as organic carbon were diluted by the coarse-grained quartz sand of the East China Sea Continental shelf sediments (Fig. 4). The well-sorted coarse-grained quartz sand was a relict from the last glacial phase (Niino and Emery, 1961). The composition of this relict sediment is mostly coarse-grained quartz sand with very low concentrations of metals and organic carbon. Heavy metals in the terrigenous sediments from both Yangtze River and Taiwan were transported to the shelf. The metal-poor relict quartz thus acted as a dilutant for these modern sediments.

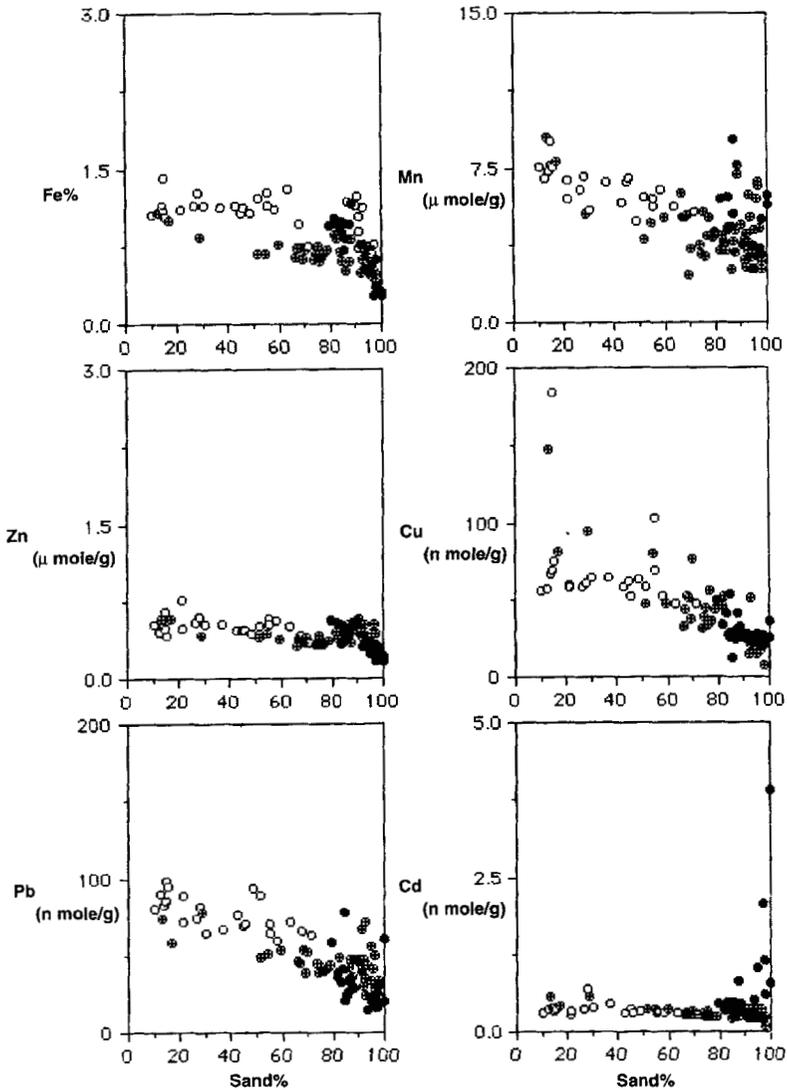


FIGURE 4 Metals correlated negatively with sand content. Carbonate rich sediments (>20% sand, ●); iron poor sediments (⊕), iron-rich sediment (○)

The correlation coefficients between metals, sand contents, organic carbon and carbonate concentrations are presented in Table II. In general, fewer heavy metals were observed in sandy sediments, as shown by the good correlation coefficients in Table II.

TABLE II Correlation coefficients between metals, %sand, and %organic carbon in the sediment

	<i>Fe</i>	<i>Mn</i>	<i>Zn</i>	<i>Cu</i>	<i>Pb</i>	<i>Cd</i>	% Sand	% Org.C
<i>Fe</i>								
<i>Mn</i>	0.587							
<i>Zn</i>	0.898	0.523						
<i>Cu</i>	0.508	0.543	0.447					
<i>Pb</i>	0.720	0.561	0.610	0.649				
<i>Cd</i>	-0.255	0.062	-0.270	-0.013	-0.054			
%Sand	-0.621	-0.610	-0.542	-0.766	-0.792	0.107		
%Org.C	0.606	0.590	0.494	0.747	0.819	-0.012	-0.914	
%Carb*	-0.300	-0.121	-0.286	-0.253	-0.350	0.775	-0.410	-0.324

%Carb*: Only sediments with greater than 20%wt carbonate were used.

The correlation coefficients of iron, manganese and zinc were slightly lower, likely a result of a combination diluting effect with heavy metals derived from a mixture of sediments from both Taiwan and the Yangtze River (Fig. 4).

The inter-element relationships between the studied heavy metals also indicated the importance of dilution effect and sources of sediments in the metal concentration. The concentrations of zinc, lead and, to a lesser extent, manganese and copper were all associated with iron and sand% ($r^2 = 0.51-0.90$) whereas cadmium was strongly associated with carbonate.

The metals/iron ratios can be extrapolated linearly, within the data scatter, through origin, and may represent a simple dilution relationship between metal-rich source sediments and metal-poor relict sand. The cadmium/carbonate relationship, on the other hand, represents a mixture of cadmium-rich carbonate and cadmium-poor sand. Both were related to the presence of a particular source(s) and a subsequent mixture between source sediments and quartz sand. Consequently, the source of heavy metals and dilution were the two primary factors controlling the heavy metals distribution in the East China Sea Continental shelf surface sediments.

CONCLUSION

Metals concentration varies greatly in sediments from various geographic locations in the East China Sea Continental shelf region.

Most variations in the surface sediments of the study region can be explained in terms of sources and a dilution effect from the metal-poor quartz sand.

Zonal distribution pattern of the heavy metals was observed in the East China Sea Continental shelf sediments. High concentrations of most heavy metals, organic carbon and fine-grained sediments were observed in the inner shelf zone, especially those near the Yangtze River. Metal concentrations decreased from inner shelf to the shelf break region. High concentration of metals were also observed in sediments near Taiwan. Concentrations of metals decreased north-east to the central shelf region. Major sources of heavy metals in the study region were the terrigenous sediments from the Yangtze River and the island of Taiwan. Biogenic carbonate sediments also contributed a significant amount of cadmium to the shelf break region. In addition to the source difference, the large observed variations in metal concentration was a result of quartz sand's dilution effect. The relict sediments from the last glacial period consisted of coarse-grained, metal-poor quartz sand. Heavy metals from different sources were diluted by this metal-poor quartz sand to a different degree.

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