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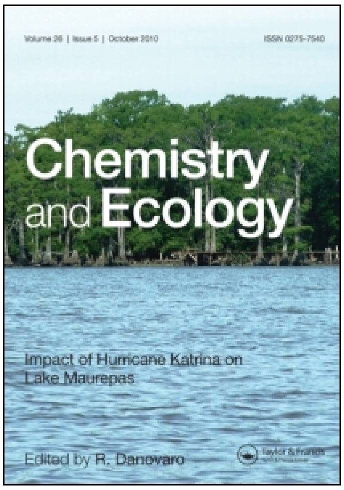

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Enrichments of Trace Metals in Particulate Matter From the Southern East China Sea, North of Taiwan

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ENRICHMENTS OF TRACE METALS IN PARTICULATE MATTER FROM THE SOUTHERN EAST CHINA SEA, NORTH OF TAIWAN

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This study investigates the distributions and enrichments of trace metals in suspended and sinking particulate matter from southern East China Sea (ECS) north of Taiwan during the period April 1992 to April 1993. According to these results, concentration of suspended particulate matter in the inner shelf of southern China Sea, the upwelling-influenced shelf break, and in Kuroshio water are 1.30 (surface)–4.2 (bottom) $mg\,l^{-1}$, ca. $0.4\,mg\,l^{-1}$ and $0.1-0.2\,mg\,l^{-1}$, respectively, reflecting various influences of terrestrial inputs. A benthic nepheloid layer (BNL), apparently owing to resuspension of local and/or remote bottom sediments, formed over the shelf region. Temporal variations in trace metal contents and enrichments in suspended matter from the shelf region reflect the variation of metal inputs from Chinese rivers, particularly from the Changjiang runoff. The enriched metals are more likely to be derived from anthropogenic input, rather than from biological accumulation. In addition, a decrease in metal contents and an increase in salinity confirm the transport of suspended particulate metals from the East China Sea shelf to the open ocean. The feature of metal plume in the intermediate layer (550–800 m) of Kuroshio water also verifies this occurrence. Moreover, the sinking particles collected from a sediment trap on the upper slope are relatively enriched in lithogenic matter and trace metals, suggesting the deposit of anthropogenic metals in the slope area.

Keywords: Southern East China Sea; trace metals; enrichment; particulate matter

* Corresponding author.

INTRODUCTION

The continental shelf of the East China Sea in the western North Pacific represents one of the largest continental marginal zones in the world. The shelf receives enormous amounts of fresh water, nutrients, sediments and pollutants from two of the largest rivers in the world, the Changjiang and Huanghe Rivers (Edmond *et al.*, 1985; Shen *et al.*, 1983; Milliman *et al.*, 1985a; Milliman, 1991). However, the zonal distribution patterns of mineralogy and texture of the shelf sediments (Chin, 1979; Milliman *et al.*, 1985b; Sternberg *et al.*, 1985) imply that the shelf circulation can influence heavily distribution and sedimentation of terrigenous materials in the shelf water. Meanwhile, a number of boundary conditions, including the Changjiang runoff, influence shelf circulation. The Changjiang runoff flows predominantly to the northeast as it leaves the estuary during the summer (Beardsley *et al.*, 1985; Chao, 1991), whereas the Changjiang Diluted Water flows southward along the China coast and turns cyclonically in the northern Taiwan Strait, as attributed to the blocking by a topographic high level as well the northward Kuroshio branching current (Chao, 1991; Jan, 1995). Previous studies on China Sea sediments, meanwhile, have suggested that the inputs of Changjiang sediments are largely deposited offshore temporarily, and later resuspended and transported southward by Changjiang water (Milliman *et al.*, 1985a; McKee *et al.*, 1983). This later mixes and exchanges with intruded and/or upwelled Kuroshio water on the shelf and slope of southern East China Sea off northern Taiwan (Liu *et al.*, 1992; Wong *et al.*, 1991).

Anthropogenic metals discharged to the East China Sea over the last two decades have to increase significantly owing to rapid industrial development and increasing population along the Changjiang and Huanghe drainage basins in China. The discharged metals can be transported primarily as a water mass for those nutrient-type or as suspended sediment for certain particle-reactive metals. Also the signals of anthropogenic metals can appear in the coastal zone off northern Taiwan if they were transported by the pathways of water mass and suspended sediment. However, evaluating the enrichments of trace metals in this variable and non-uniform environment on the basis of the distributions of dissolved trace metals can be difficult. In this study, we assess the origins and enrichments of trace metals in the

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southern East China Sea using the distributions of trace metals in suspended and sinking particulates during the geochemical exploration in the Kuroshio Edge Exchange Program.

MATERIALS AND METHODS

Distribution of trace metals in suspended and sinking materials were studied in the southern East China Sea north of Taiwan. The suspended materials were sampled on board the *R/V Ocean Researcher* 1 during a number of cruises, OR-314 (April 16–22, 1992), OR-331B (October 2–8, 1992), OR-338 (December 3–8, 1992) and OR-352B (April 22–28, 1993). Figure 1 depicts the each cruise's sampling station. According to this figure, the sinking materials were collected with PP3 time-series sediment trap deployed on Mien-Hua Submarine Canyon during the period 27 October to 1 December, 1992.

For analyses of particulate trace metals, sea water samples were retrieved with cleaned 20 l Go-flo PE bottles mounted on a CTD/Rosette which recorded the temperature and salinity profiles as well. After transferring the retrieved sea water to acid-cleaned PE containers,

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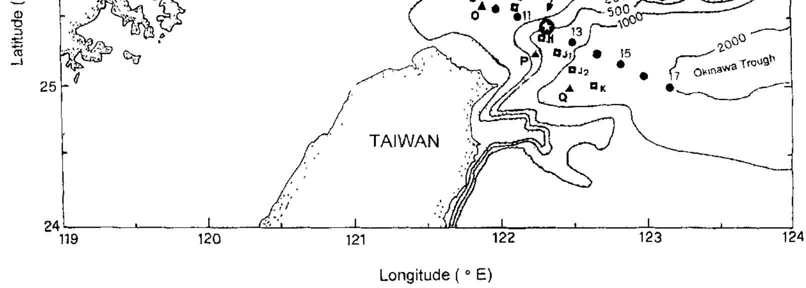

FIGURE 1 Sampling and location of study area.

the water sample was filtered through an acid-cleaned and pre-weighed 142 mm membrane filter (Nucleopore PC, 0.4 μm) driven by a peristaltic pump and a silicon tube connected to a Teflon filter holder. The total volume of filtered sea water ranged from 15 to 40 litres, depending on the content of suspended particulate matter. The filter's residue was washed with 500 ml distilled deionized water to remove any remaining sea salt. Next, the filters were sealed in acid-cleaned dishes and stored in 4°C during transfer to the laboratory. The filters were then dried at 60°C and then re-weighed with a microbalance (Mettler AT20) to determine suspended particulate matter in sea water. Finally, particulate metals were analyzed by digesting the filtered residue with mixed super-pure acids (HNO_3:HCl:HF = 3:3:4) heated by a microwave (CEM 2000), followed by dilution with distilled water.

The sinking materials recovered from a sediment trap were divided into several portions. One of these materials was rinsed with distilled water to remove sea salt and then dried at 60°C. The dried sinking materials were ground to powder using a mortar and pestle, and then weighed exactly (ca. 0.2 g) into the microwave Teflon bottle for digestion with these procedures. Another portion of the sinking materials was used to analyse particle size and organic carbon.

Trace metals in the digested solution were determined either with a flame or a flameless atomic absorption spectrophotometer (Perkin-Elmer 5100 PC, HGA 600). Matrix modifiers and Zeeman background corrector were applied throughout the flameless measurement. The analysis preciseness generally exceeded 90% reproduction (Hung, 1988, 1995). Next, particulate organic carbon in specific particulate matter and sinking materials was determined with a C/N/S analyzer (Fisons NCS 1500) after removing the inorganic carbon with diluted hydrochloric acid (2 M). Finally, size distributions of sinking materials were determined with a particle size counter (Counter LS Particle Size Analyzer).

RESULTS AND DISCUSSION

Distribution of Suspended Particulate Matter

Temperature, salinity and concentration of suspended particulate matter were measured in three transects on the southern eastern China

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Seas shelf, slope and Okinawa Trough during the cruises (OR-314, 331B and 338). Figures 2–4 confirm that the distribution of suspended particulates is related to hydrodynamic processes. The weather and oceanographic parameters observed during the three cruises were spring for OR-314, later summer OR-331B, and winter OR-338. The OR-314 stations were not monitored frequently during the OR-331B and OR-338 cruises in lieu of any major biological purpose and rough sea conditions during these cruises. In the OR-314 cruise, surface temperature increased from 17°C at station 1 to 24°C at station 17; meanwhile, salinity increased from 32.0 to 34.6 psu (Fig. 2). Such increases verify the existence of various types of water masses. The transect of OR-314 cruise contained at least four types of water masses, China Coastal Water (stations 1–3), China Seas Mixing Water (stations 4–7), Upwelled Water (stations 8–12) and Kuroshio Water (stations 15–17). Low temperature and salinity, as well as a heavy specific particulate, characterize the China coastal water influenced by Changjiang runoff. The Kuroshio water is oligotrophic, characterized by high temperature and salinity but a low specific particulates China Sea water and Kuroshio water generally range between the China Sea and Kuroshio. Specific particulates concentration generally decrease seaward, but dramatically increase downward in the shelf and upper slope zones (Fig. 2). The seaward decrease of specific particulate suggests a horizontally advective transport, while the downward increase of particulates in the shelf region is attributed primarily to the resuspension of bottom sediment. The resuspension diminishes as the site shifts away from the lower slope due to more quiescent condition of the deeper bottom. Apparently, particulate matter was transported laterally from the China coast to the southern Okinawa Trough overlaid with Kuroshio Water. The apparent flux of sinking particles on the slope confirm this lateral transport, in which the flux was considerably higher in the bottom layer than in the upper layer. The other two transects, OR-331B and OR-338, have a relatively short distance. The stations were largely occupied by Kuroshio and China Seas. Although the locations of the two transects are not identical to those of the transect OR-314, the concentrations of particulate matter in three transects are comparable on the basis of similar distances away from the coast. This finding suggests that the inputs of sources from mainland China heavily influence the

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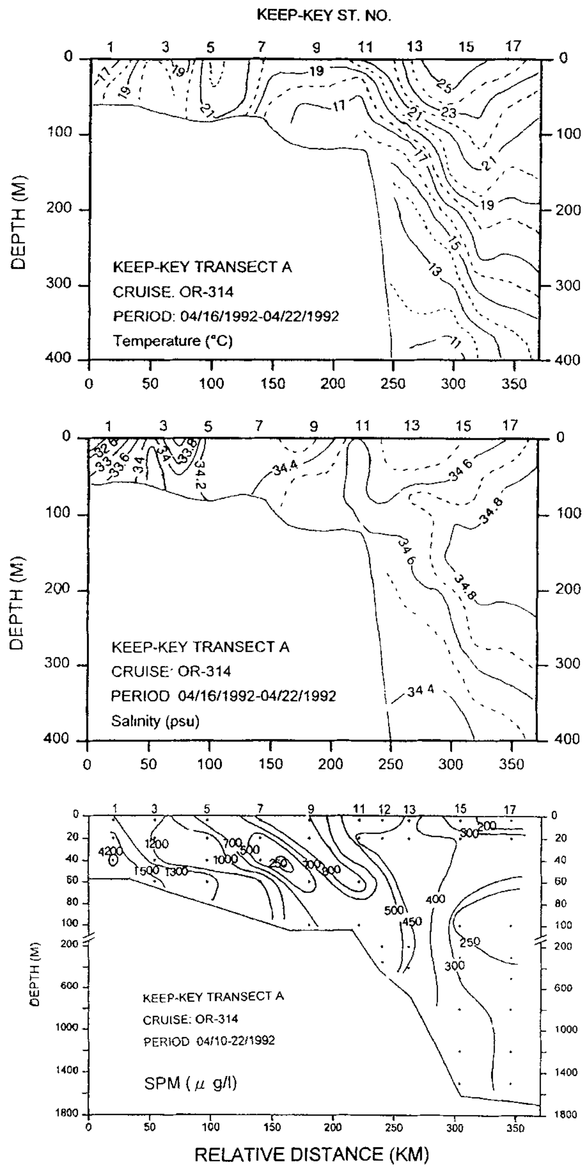

FIGURE 2 Temperature, salinity and SPM distributions in the OR-314 cruise.

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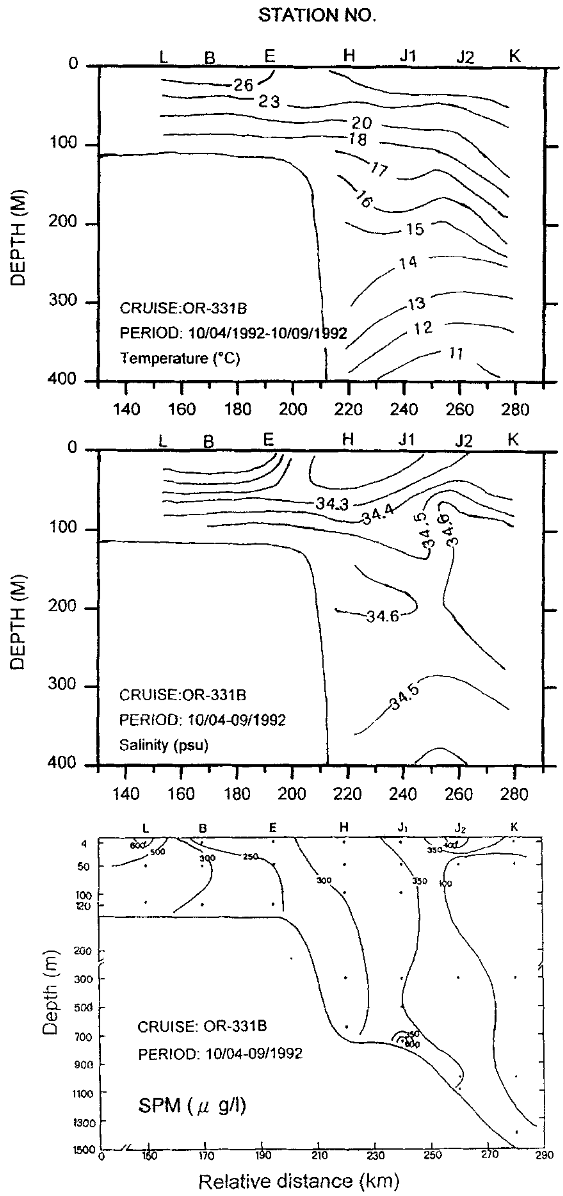

FIGURE 3 Temperature, salinity and SPM distributions in the OR-331B cruise.

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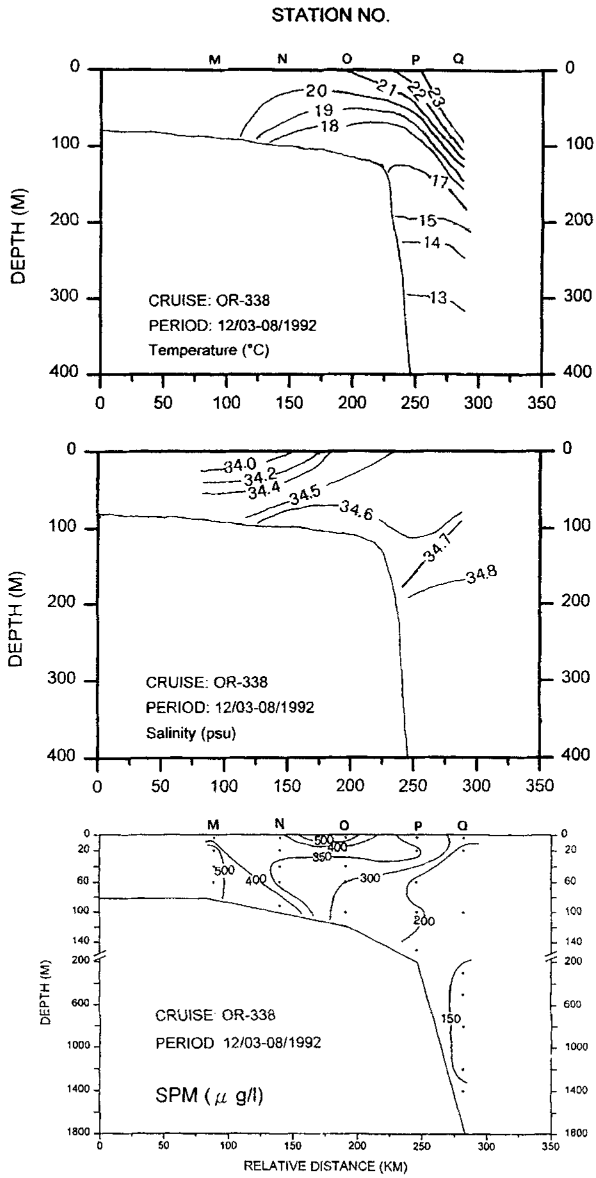

FIGURE 4 Temperature, salinity and SPM distributions in the OR-338 cruise.

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particulate distribution in southern East China Seas. Meanwhile, the abundance of particulates, particularly for the surface concentration, varied between the different cruises, reflecting the influence of biological activity.

Distribution of Suspended Particulate Trace Metals

Table I lists the distributions of particulate trace metals in the samples taken from three cruises. Aluminium, an indicator of lithogenic source, not only decreases in concentration (based on weight) from China coastal water to Kuroshio, but is also accompanied by an increasing POC from China coastal water (ca. 5%) to Kuroshio (ca. 20%) in the OR-314 cruise. This trend indicates the increase of biological accumulation in controlling particulate trace metals in upwelling and Kuroshio waters. Particulate iron, similar to particulate aluminium, decreases in concentration from China coast to Kuroshio, indicating the decreasing influence of lithogenic materials. An increasing particulate aluminium and iron with depth also reflects the resuspension of bottom sediments. Significant correlations ($p < 0.001$) also exist among particulate matter, particulate aluminium and iron. Hence, the distribution profiles (based on volume) closely correspond to the distribution pattern of particulates (Fig. 4); all of the above profiles can be indicators for the transport of terrigenous materials. Meanwhile, particulate aluminium and iron are

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TABLE I Distribution of particulate metals in the study area

	Org. C (%)	*Al* (%)	*Fe* (%)	*Mn* $\mu g\, g^{-1}$	*Zn* $\mu g\, g^{-1}$	*Cu* $\mu g\, g^{-1}$	*Pb* $\mu g\, g^{-1}$	*Ni* $\mu g\, g^{-1}$	*Cd* $\mu g\, g^{-1}$
OR-314 cruise									
Shelf zone	5	5.22	3.62	1800	367	85.5	43.8	52.4	1.5
Upwelling zone	15	2.66	2.62	2671	484	120	83.8	73.4	2.0
Kuroshio water	20	3.77	2.67	2720	818	142	177	84.1	2.8
OR-331B cruise	17	3.39	3.72	3201	900	186	235	175	6.3
OR-338 cruise	20	3.62	4.48	4036	357	325	311	218	3.6
Changjiang river[a]	–	8.4	4.7	972	182	48	34	–	0.33
World's rivers[b]	–	9.4	4.8	1050	350	100	150	90	1.0
Mean crust[c]	0.0002	8.23	5.63	950	70	55	12.5	75	0.2

[a] Qu and Yan (1990).
[b] Martin and Meybeck (1979).
[c] Taylor (1964).

lower in the southern China Seas than in Changjiang river, average of world rivers and mean crust (Tab. I). The lower concentration is largely attributed to the dilution of particulate by biological materials in the China Seas. In contrast, the concentrations (based on weight) of manganese, lead and biophilic trace metals (Cu, Zn, Cd and Ni) increase seaward, from China coast to Kuroshio (Tab. I); these concentrations exceed those in Changjiang river, global rivers and mean crust. Apparently, these trace metals were enriched progressively seaward in particulate material by biological activity. On the other hand, the abundance of these particulate metals (using lead as an example, ng/l) still corresponds to the distribution trend of particulate, and decreases generally with an increasing distance from the coast. Notably, the abundance of particulate metals in the OR-331B and OR-338 cruises (not shown here) generally decrease with offshore distance. However, the concentrations by weight are invariable with location, owing to the lesser influence of terrestrial inputs. Apparently, the different concentrations of particulate metals among the three cruises are attributed primarily to seasonal variation.

The distributions of particulates and particulate manganese in the water columns of station 17, OR-314 and OR-352B cruises (Fig. 6) reveal an unusual plume (maximum concentration) at the intermediate layer (550–800 m). Wei *et al.* (1991) also observed the plume of dissolved manganese at the same depth near station 17. This plume appears to exist quite frequently. From its existence, we can infer than particulates and trace metals can be transported from the shelf to the Kuroshio through this plume. However, its formation remains unclear; we speculate that the slope sediment was somehow resuspended or slumped followed by lateral advection away from the continental slope. Nevertheless, the actual mechanism requires more studies.

Enrichment of Particulate Trace Metals in Southern China Sea

According to Table II, the aluminium concentration normalizes the distribution of particulate trace metals listed in Table I. The particulate metals were significantly more enriched than those obtained from global background data, either in the riverine or in the crustal composition. As this table reveals, the Me/Al ratio

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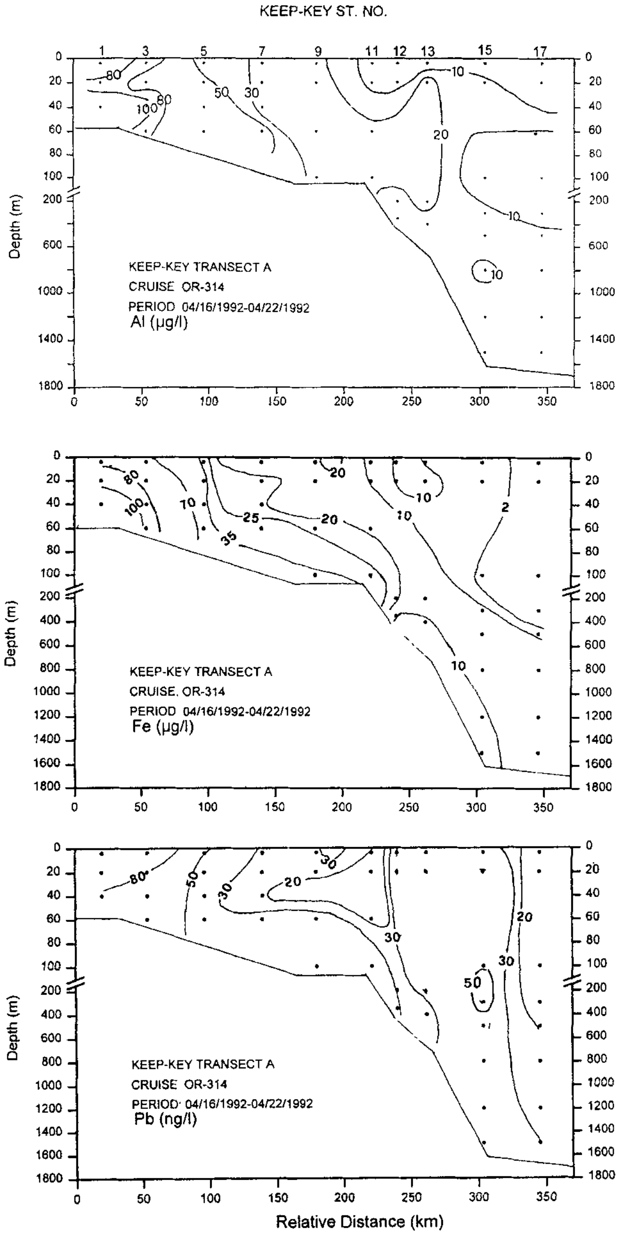

FIGURE 5 Distribution profiles of particulate Al, Fe and Pb in the OR-314 cruise.

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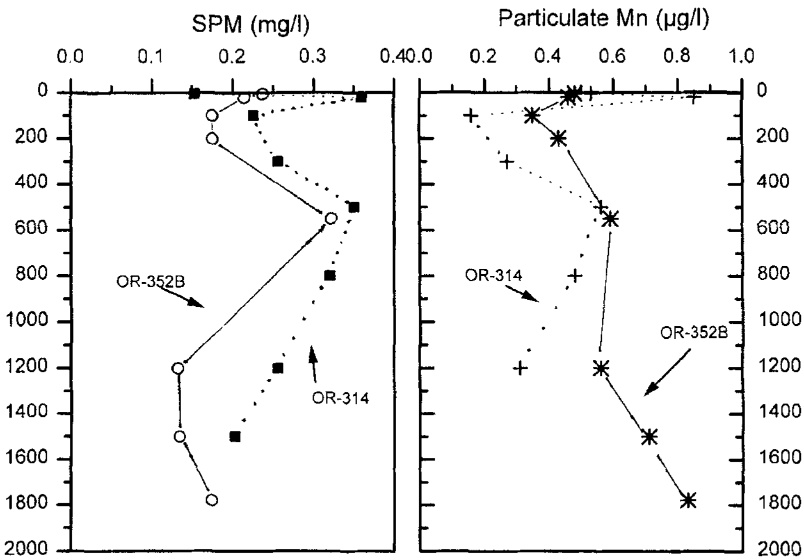

FIGURE 6 Distributions of SPM and particulate Mn in the water column of station 17.

TABLE II Normalization of particulate metals to aluminum

	Fe/Al	Mn/Al (*1000)	Zn/Al (*1000)	Cu/Al (*1000)	Pb/Al (*1000)	Ni/Al (*1000)	Cd/Al (*1000)
OR-314 cruise							
Shelf zone	0.68	33.7	7.10	2.00	0.83	1.00	0.03
Upwelling zone	1.06	96.0	23.9	6.26	4.08	3.58	0.11
Kuroshio water	0.72	94.0	33.2	6.32	5.76	3.59	0.15
OR-331B cruise	1.29	96.3	26.5	5.93	7.35	5.60	0.22
OR-338 cruise	1.32	114	9.8	10.1	9.12	6.51	0.12
Changjiang river[a]	0.56	11.6	2.17	0.57	0.40	–	0.004
World's rivers[b]	0.51	11.2	3.72	1.06	1.60	0.96	0.011
Mean crust[c]	0.68	11.5	0.85	0.67	0.15	0.91	0.002

[a] Qu and Yan (1990).
[b] Martin and Meybeck (1979).
[c] Taylor (1964).

obviously increases from China coastal seas to Kuroshio. The particulate Me/Al ratio in southern China Sea exceeds that in Changjiang, global rivers and mean crust. Apparently, biologically concentrated processes enhanced the enrichment. However, this enrichment cannot be attributed entirely to biological enhancement,

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and could largely result from the anthropogenic inputs. The metal 'excesses' (Me_{excess}), defined as the fractions of metals derived from the sources other than the crust (Brugmann *et al.*, 1992), are regarded generally as the available metals. These metals are calculated by subtracting the total contents from those lithogenic components. Therefore, the metal excesses in the particulate matter are calculated according to:

$$Me_{excess} = Me_{total} - (Al_{total} \times Me_{crust}/Al_{crust}).$$

The metal concentrations in the crust published by Taylor (1964) were used to calculate the metal excesses. Table III lists the calculated percentages of Me_{total} as Me_{excess}. Most trace metals have positive excess values, including iron and copper, which have negative values in Changjiang particulates. The greater positive value denotes the greater metal enriched in particulate material. The Me_{excess} values in China Sea exceed those in global rivers (including Changjiang). This finding suggests that dissolved trace metals can be adsorbed or uptaken by marine particles and, ultimately, contribute to total particulate metals after the riverine particulates enter the China Sea. The different values of Me_{excess} between Kuroshio and China coast seas than the lakes can be attributed to the concentrated effect of biota. However, the difference is relatively small for anthropogenic metals, implying that the Me_{excess} in China Sea may be largely attributed to the anthropogenic inputs. The

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TABLE III Percentage of (Me_{excess}/Me_{total}) in the particulate matter of study area

	Al	*Fe*	*Mn*	*Zn*	*Cu*	*Pb*	*Ni*	*Cd*
OR-314 cruise								
Shelf zone	−57.5	1.9	66.7	87.9	59.1	82.1	9.3	93.0
Upwelling zone	−209	30.9	88.5	95.3	85.1	95.2	67.0	97.3
Kuroshio water	−118	4.1	84.1	96.1	82.2	96.8	59.2	97.3
OR-331B cruise	−143	35.5	87.8	96.8	87.8	97.8	82.4	98.9
OR-338 cruise	−127	45.1	95.4	91.4	92.5	98.3	84.9	98
Changjiang river[a]	2.0	−21.2	0.62	60.8	−17.3	62.9	–	49.1
World's rivers[b]	12.4	−54.2	−2.95	77.2	37.0	90.6	5.0	81.2
Mean crust[c]	–	–	–	–	–	–	–	–
–								

[a] Qu and Yan (1990).
[b] Martin and Meybeck (1979).
[c] Taylor (1964).

negative excess values for aluminium in the China Sea are due to dilution of aluminium by biological materials; hence, more negative values are found upwelled water and Kuroshio than in the China coast seas. The relatively significant decrease of excess lead in vertical distribution (not shown here) implied that a large portion of metal excess represents the easily mobilizable fraction and originates possibly from the atmosphere. Other metals occur in high excess close to the bottom, owing to remobilization from the sediments and a new precipitation in the presence of oxygen.

Enrichments of Particulate Metals in Sinking Materials

Sinking materials were collected with time-series sediment traps from the station (25°27'N, 122°32'E) located in Mien-Hua Submarine Canyon (Fig. 1). Table IV lists the particulate metal concentrations and enrichment with respect to continental crust. According to this table, the mass fluxes were high anomalously and greater in the bottom layer than in the upper layer, implying a remarkably lateral transport of sediments. Apparently, the accumulated sinking materials do not represent the *in-situ* production, but may derive from the terrestrial inputs.

Interestingly, the concentration of organic carbon is lower in the bottom layer than in the upper layer, indicating the decomposition of organic matter with depth. Most particulate metals decrease in concentration with depth except for manganese. The decreasing concentration is attributed to decay of organic matter, whereas an increasing manganese may be attributed to the scavenging process. The major components of sinking materials were aluminosilicate (42–45%) and unidentified detritus (41–44%). From this composition, we can infer that sinking materials are mainly terrigenous and/or resuspended bottom sediments. This fact that individual metal correlated well ($p < 0.01$) with aluminium, organic carbon, mud verifies such an inference. Enrichment factor exceeds 1.0 for iron, manganese, cadmium, copper and > 2.0 for lead and zinc. Apparently, a significant portion of particulate metals may be derived from the anthropogenic inputs rather than from the biological enrichment as judged from their sources and enrichments.

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TABLE IV Chemical composition of sinking particles at trap T1 station

Sample	clay	carb.*	Opal	O.M.#	Al	Fe	Mn	Cd	Cu	Ni	Pb	Zn
	%						ppm					
340 m-depth	49.6	11.4	0.68	4.38	3.97	2.55	437	0.09	29.9	33.9	25.9	93
430 m-depth	48.0	10.6	0.52	4.12	3.84	2.48	494	0.14	28.7	30.2	20.9	105
Underlying sediment (0–2 cm)	80	2.58	0.37	3.72	6.42	4.10	366	0.08	34.9	17.0	22.0	99.7

* carbonate.
# organic matter.

TABLE V Distributions of metal/Al ratios in sinking particles at trap T1 station

Sample	Fe/Al	Mn/Al (10^{-2})	Cd/Al (10^{-6})	Cu/Al (10^{-4})	Ni/Al (10^{-4})	Pb/Al (10^{-4})	Zn/Al (10^{-4})
340 m-depth	0.79	1.36	4.09	7.53	8.13	5.51	26.0
430 m-depth	0.86	1.58	4.02	8.72	8.67	6.33	27.3
Mean crust [a]	0.68	1.15	2.43	6.68	9.11	1.52	8.51
Underlying sediment (0–2 cm)	0.64	0.57	1.53	5.44	7.65	3.42	15.5

[a] Taylor (1964).

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Factor Analyses of Trace Metals in Three Cruises

Particulate trace metals measured from the OR-314 cruise (transects) and a sediment trap were analyzed and grouped by factor analysis. Factor analysis functions as a tool to analyze data so that the elemental or a real relationship among the distributed elements can be understood thoroughly. The Q-mode analysis did not reveal any significant clustering; therefore, the R-mode was used for data analysis. The fact that suspended and sinking particulates markedly differ with respect to the distributions of particulate metals accounts for the factor analyses of OR-314 cruise and sediment-trap are performed separately. Table VI lists the factor values. For the OR-314 cruise, three factors account for > 81% of the variance. The factor 1 is interpreted as representing the terrestrial origins; those elements associated with lithogenic materials have significant values. The inputs of terrestrial materials may largely determine the distributions of particulates, particulate aluminium, iron, manganese, cadmium, copper, zinc, calcium and magnesium. Factor 2 can be interpreted as

TABLE VI Factor matrix of particulate metals from samples of OR-314 cruise and T1 sediment traps

	OR-314			*Trap T1*		
Element	*Factor* 1	*Factor* 2	*Factor* 3	*Factor* 1	*Factor* 2	*Factor* 3
SPM	0.9167	0.2472	0.0719	–	–	–
Al	0.8503	0.4277	0.1046	0.7960	0.5401	0.0515
Fe	0.8774	0.2889	0.2639	0.9435	0.1566	0.1804
Mn	0.7105	0.4648	0.1722	0.7411	−0.2259	0.4913
Cd	0.5039	0.0097	−0.4028	0.7211	−0.5944	–
Cu	0.5878	0.0616	0.0747	0.8122	−0.5971	–
Pb	0.2361	0.0106	0.8636	0.8335	−0.2275	0.2217
Ni	0.2308	0.9092	−0.0745	0.7699	−0.4086	0.0753
Zn	0.8479	0.3373	0.2777	0.9107	−0.3097	–
Ca	0.7424	0.1899	−0.1689	0.4127	0.8709	−0.0456
Mg	0.8449	0.2807	−0.0994	0.6678	0.6511	0.0976
K	0.2448	0.9799	0.0754	0.5664	0.2434	−0.5501

the association of biological materials; the turnover of biota can largely determine particulate nickel and potassium. Factor 3, only significant for lead, can be interpreted as the association of aerosol deposition. For the factor analyses of trap particles, distributions of particulate metals (Al, Fe, Mn, Cd, Cu, Pb, Ni, Zn and K) may be constrained by the sources of terrestrial materials. However, particulate calcium and magnesium may be associated with biological carbonate. The major difference between two sorts of particulate metals, particularly for lead, nickel, calcium and magnesium, lies in the fact that, in this study, sinking particles were collected below the subsurface layer and may be largely derived from the resuspended sediments and/or sinking biological carbonate. The signal of aerosol influence diminishes, but biological carbonate becomes significant for sinking particles in the trap samples.

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