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Effects of northeast monsoon on trace metal distribution in the South China Sea off Peninsular Malaysia

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Abstract Concentrations of trace metals in the South China Sea (SCS) were determined off the coast of Terengganu during the months of May and November 2007. The concentrations of dissolved and particulate metals were in the range of 0.019–0.194 μg/L and 50–365 μg/g, respectively, for cadmium (Cd), 0.05–0.45 μg/L and 38–3,570 μg/g for chromium (Cr), 0.05–3.54 μg/L and 21–1,947 μg/g for manganese (Mn), and 0.03–0.49 μg/L and 2–56,982 μg/g for lead (Pb). The order of mean log K_D found was Cd>Cr>Pb>Mn. The study suggests that the primary sources of these metals are discharges from the rivers which drain into the SCS, in particular the Dungun River, which flows in close proximity to agricultural areas and petrochemical industries. During the northeast monsoon, levels of particulate metals in the bottom water samples near the shore were found to be much higher than during the dry season, the probable result of re-suspension of the metals from the bottom sediments.

Keywords Dissolved metals · Particulate metals · Chelex-100 · South China Sea · Distribution coefficient · Northeast monsoon

Introduction

Trace metals exist in seawater in both dissolved and particulate phases, and their bioavailability depends on the complexation process with organic matter (Censi et al. 2006). The concentration of trace metals present in the water column varies due to changing inputs and seasonal effects on biological, geochemical, and physical properties (Valdés et al. 2008). In some cases, nutrient-typed trace metals being are depleted in the surface water and enriched with depth as a result of gravitational settlement and the decomposition of particulate matter (Cuong et al. 2007). Metals return to the water column through the decomposition of organisms, as well as via the dissolution of sinking fecal pellets and mineralization.

Studies on metal concentrations in seawater (Table 1) indicate that the metal concentrations within Southeast Asia regions are comparable, except in areas polluted by specific metals due to agriculture, industrial run-off, effluent discharge, and domestic activities.

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Table 1 Range of metal concentrations in the coastal waters around Peninsular Malaysia (micrograms per liter for dissolved and micrograms per gram for particulate metal)

Location	Cd	Cr	Mn	Pb	Dissolved/ particulate	Reference
Kranji and Pulau Tekong, Singapore	0.013–0.109 0.16–0.73	0.072–0.35 –	– –	0.009–0.062 1.10–6.08	Dissolved Particulate	Cuong et al. 2007
Chukai river estuary, Kemaman	0.13–0.21	–	–	1.20–1.84	Dissolved	Kamaruzzaman et al. 2002
Kemaman coast, Terengganu	0.01–0.06 <100–400	0.21–1.23 7,200–16,000	– –	0.01–0.29 700–1,100	Dissolved Particulate	Shamsuddin 1996
Southern coast of Terengganu	0.019–0.194 0–365	0.05–0.45 38–3,570	0.05–3.54 21–1,947	0.03–0.49 2–56,982	Dissolved Particulate	This study, 2007

Cd cadmium, *Cr* chromium, *Mn* manganese, *Pb* lead

Cuong et al. (2007), in 2004, monitored the levels of dissolved and suspended particulate metal in the marine water column and surface sediments near Singapore and found that heavy metal concentrations were very low when compared with the Criterion Continuous Concentrations given by the National Recommended Water Quality Criteria, United States Environmental Protection Agency. Surface water had depleted metals while deep water had enriched metals, as indicated by the trace metal profile, in both dissolved and particulate fractions.

Shamsuddin (1996) studied the distribution of trace metals in seawater and surface sediments off the coast of Kemaman. Possible anthropogenic inputs of metals to the marine environment of the study area include shipping activities at the Kemaman port, input from the titanium oxide processing plant nearby, quarry activities, as well as fishing and shipyard activities. An earlier study by Kamaruzzaman et al. (2002) in the region found that natural processes influenced on the concentrations of dissolved trace metal in the water column indicating conservative behavior of the metals in the environment. However, at certain sampling location, concentrations of some metals were relatively high due to ongoing construction at the Chukai River area but were not enough to pollute the Chukai River estuary system.

Data on heavy metals in the South China Sea (SCS) being have been published elsewhere; however, according to database collected by www.scopus.com, since 1968 to 2012, less than 10 % of studies on heavy metals have been focused at the southern part of SCS. Nevertheless, much of the studies primarily focus on sediments and marine organisms (Shazili et al. 2006) as little research on dissolved and particulate metals in seawater due to their trace level existence (Jiménez et al. 2002) and the need for special precautions during sample collection and sample analysis (Hunter and Boyd 1997). This study provides a baseline data for future comparisons of trace metal distribution in the water column as well as variations of trace metals due the northeast (NE) monsoon in southern SCS. It also determines to what degree anthropogenic activity influences trace metal concentrations in the SCS off the southern coast of Terengganu.

Materials and methods

Sampling

Dungun, Kerteh, and Kemaman are located at the southern part of the state of Terengganu, on the east coast of Peninsular Malaysia. The Dungun, Paka, Kerteh, and Kemaman Rivers flow into the SCS. Agricultural and urban development dominates the catchments of the Dungun and Kemaman Rivers, whereas Kerteh and Kemaman, along the coastline, are in the vicinity of petrochemical industries. An oil and gas facility landing is located in the Kerteh industrial complex while Bukit Besi (a rural region of Dungun) had previous iron ore mining activities.

Sampling was carried out in May and November 2007. May represents the dry season with monthly average and total precipitation of 2.09 and 64.9 mm, respectively. November signifies the NE monsoon season with monthly average and total precipitation of 11.56 and 346.8 mm, correspondingly. Seawater samples were collected from the surface, middle, and bottom layers, in a grid of 18 stations covering the coast of Dungun, Kerteh, and Kemaman (Fig. 1). The study area covers an area of about 3,289 km^2, with the most coastal station approximately 0.9 km from the coastline and the furthest offshore about 61 km.

Seawater samples were collected using a 1 L Teflon Mercos sampler at 3 depths: surface layer (1 m), middle layer (10 m), and bottom layer (1 m off the seabed). Water depths varied from 11 to 60 m.

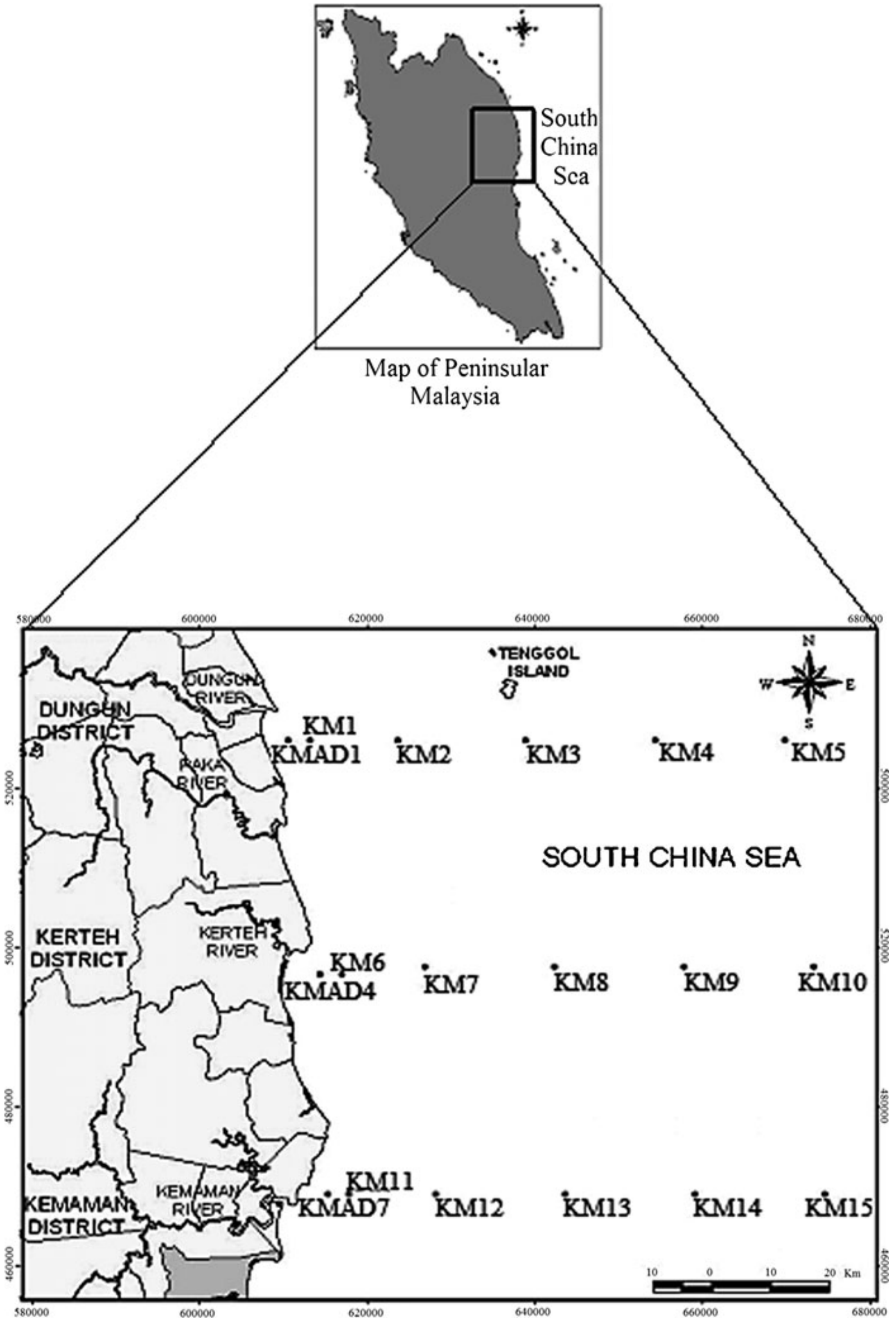


Fig. 1 Sampling stations off the southern coast of Terengganu, South China Sea

Method and analysis

Laboratory wares were cleaned by soaking in diluted nitric acid for 1 week followed by thoroughly rinsing with deionized water to remove all the metal contaminants. The wares were then dried under a laminar flow hood and kept in clean polyethylene plastic bags. Water samples were prepared under a class-100 laminar flow hood to avoid contamination. Metal concentrations were measured using a Perkin Elmer Elan 9000 inductively coupled plasma mass spectrometry (ICP-MS) in a class-100 clean room.

For dissolved metal analysis, 200 ml duplicate samples of seawater were immediately filtered upon sampling using a 0.45 μm polytetrafluoroethylene (PTFE) membrane filter fitted online with a peristaltic pump, forcing the seawater from the bottle samplers through PTFE tubing packed with Chelex-100. The suspended particulate matter (SPM) retained on the PTFE membrane filters were analyzed to determine the particulate metal concentrations. The Chelex-100 columns and PTFE membrane filters (containing the suspended particulate matter) were placed in a clean transport container for subsequent laboratory analysis. All on-board sample preparations were conducted under a class-100 laminar flow hood.

Trace metals were eluted from each Chelex-100 column using 10 ml of 2 M nitric acid, after washing the column with 25 ml of ammonium acetate to remove the entire saline matrix (Scoullos et al. 2007). The eluted metals were placed in 15 ml centrifuge tubes and kept at 4 °C.

The PTFE membrane filters (containing suspended particulate matter) were dried under a laminar flow hood for several days until constant weight. The filters were then digested using 2 ml of 48 % HF, 3 ml of 37 % HCl, and 4 ml of 65 % HNO_3 in a closed Teflon vessel under microwave heating at 210 °C for 30 min. The HF was neutralized by adjusting the sample volume to 10 ml with saturated boric acid and finally made up to 25 ml with Milli-Q water. Sample blanks were also prepared to check for contamination. The digested samples were kept in the centrifuge tubes at 4 °C before analysis.

Table 3 Detection limit for each trace metal

Element	Dissolved[a] (ug/L)	Particulate[a] (mg/kg)
Cadmium	0.0081	0.018
Chromium	0.054	0.009
Manganese	0.0285	0.12
Lead	0.003	0.024

[a] Detection limit calculated as three of the standard deviation of the procedural blanks (*n*=7)

As a QA/QC procedure, Nearshore Seawater Reference Material for Trace Metals (CASS-4) as well as Marine Sediment Reference Material for Trace Elements and Other Constituents (MESS-3) from the National Research Council Canada (NRCC) were analyzed for their trace metal contents (Table 2). The analyses were carried out using an ICP-MS in a clean room. Table 3 shows the detection limit of each trace metal as the sample results were verified ±20 % accurate.

Result and discussion

Table 4 shows a summary of the range of trace metals concentration in the seawater in the SCS off the southern Terengganu coast.

High concentrations of particulate trace metals were found during November 2007, when there was strong current velocity. In May 2007 (Fig. 2), strong current movement caused high re-suspension of surface sediment into the water column. Re-suspension of sediment particles explains metal remobilization into the water column (Charriau et al. 2011) as high amounts of

Table 2 Analytical quality control data for dissolved and particulate metals

Element	CASS-4 certified value (ug/L)	CASS-4 certified measured value (ug/L)	MESS-3 certified value (mg/L)	MESS-3 measured value (mg/kg)
Cadmium	0.026+0.003	0.034+0.005	0.24+0.01	0.20+0.06
Chromium	0.144±0.029	0.11±0.03	105±4	89±5
Manganese	2.78±0.19	2.20±0.27	324±12	259±9
Lead	0.0098±0.0036	0.0123±0.0028	21.1±0.7	19.99±0.98

Table 4 Concentrations of trace metals recorded in waters around the south coast of Terengganu, South China Sea

Element	May 2007		November 2007	
	Dissolved (μg/L)	Particulate (μg/g)	Dissolved (μg/L)	Particulate (μg/g)
Cadmium	0.019–0.194	50–245	0.019–0.118	72–365
Chromium	0.097–0.45	63–511	0.05–0.32	38–3,570
Manganese	0.05–2.71	21–531	0.14–3.54	59–1,947
Lead	0.07–0.49	11–157	0.03–0.16	2.0–56,982

metals are bound to the sediment particles, especially the clay fraction (Helios-Rybicka and Kyziol 1991).

High concentrations of dissolved Cd (Fig. 3) and Mn (Fig. 4) were detected at stations that were in close proximity to the Dungun River estuary, located at the northern part of the sampling region. Dissolved Cr (Fig. 5) varied throughout the study area. High concentrations of particulate Cd, Cr, and Mn were detected

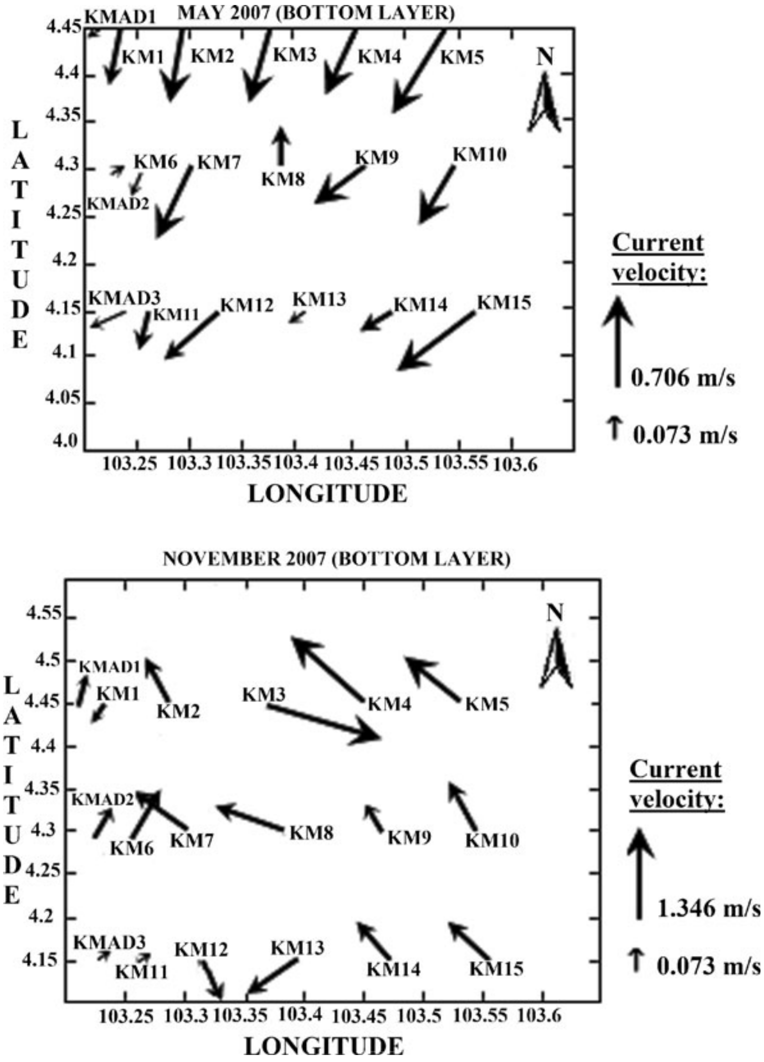


Fig. 2 Current direction and velocity of the Terengganu coast bottom waters

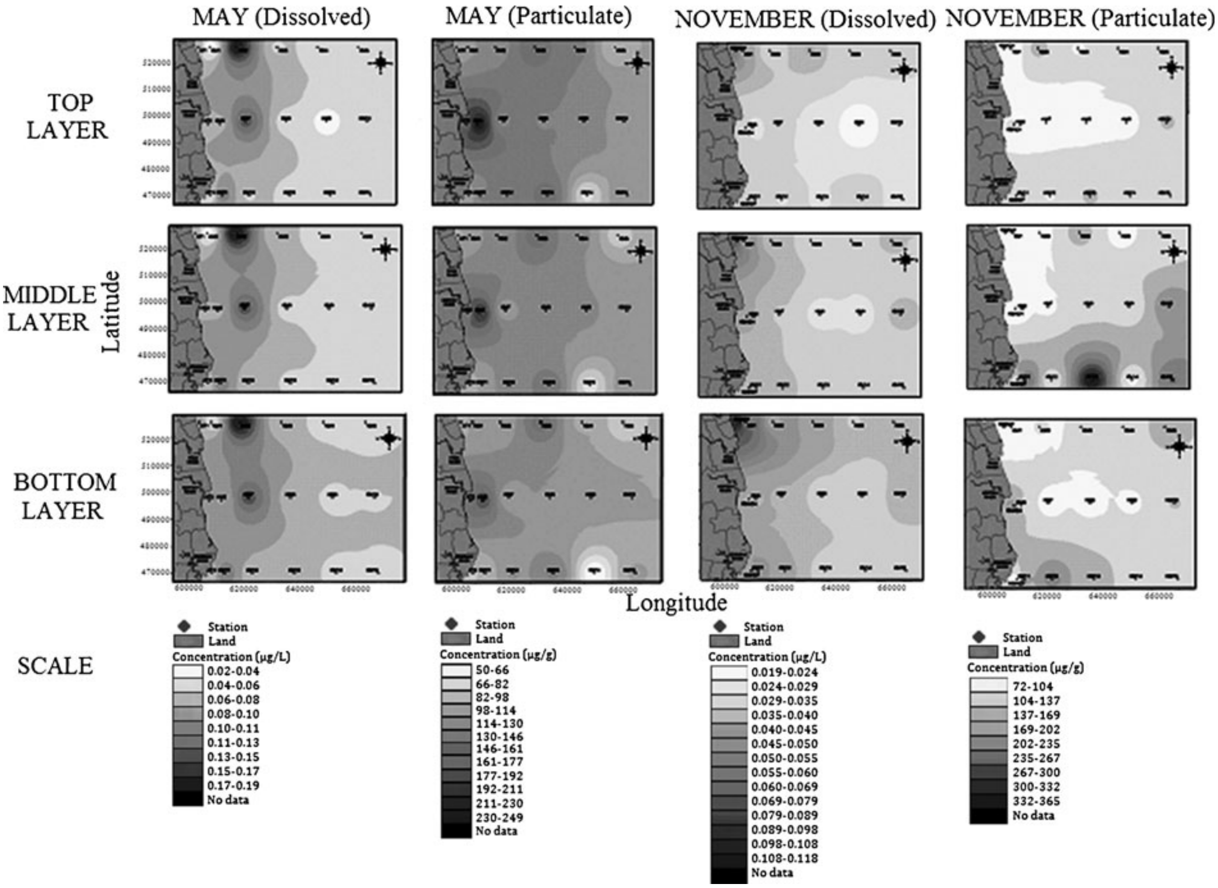


Fig. 3 Distribution of cadmium in the southern coast of Terengganu

at the near-shore stations. The high concentration of metals found in the northwest and at stations near the coastline suggests substantial anthropogenic inputs from the Dungun, Paka, Kerteh, and Kemaman rivers. Since the coastal areas in the region are fairly well populated, diffuse sources of metals also contribute. A similar trend was found by Guieu et al. (1996), showed that the Lena River transport Cu, Ni, Cd, Fe, Pb, and Zn toward the Laptev Sea in Russia. The results of this study suggest that high urban development, activities of the petrochemical industries, and agricultural activities along the coastline from Dungun to the Kemaman are the most likely contributing factors to the high metal content.

High concentrations of dissolved Pb (Fig. 6) were found at stations near the coastline and in the southern region of the study area. During May 2007, particulate Pb levels were found to be high at the offshore stations. The contrary was observed in November 2007 (NE monsoon period) where concentration of particulate Pb was found to be high in the bottom waters of the near-shore stations. Particulate Pb concentrations reached 56,982 μg/g (Table 4) in the coastal bottom waters. The petrochemical industries along the Paka and Kerteh coastline, as well as shipping and export terminal activities at the Kemaman estuary, are likely sources of the dissolved and particulate Pb found. A major road system runs along this coastline region, and the Pb emissions form transportation activities may contribute to the increase in Pb levels in the coastal waters as vehicle emissions and wastes from petrochemical-based industries contribute Pb (Soli et al. 2008). In November 2007, we found that particulate Cr and Mn in higher concentrations compared with the observed

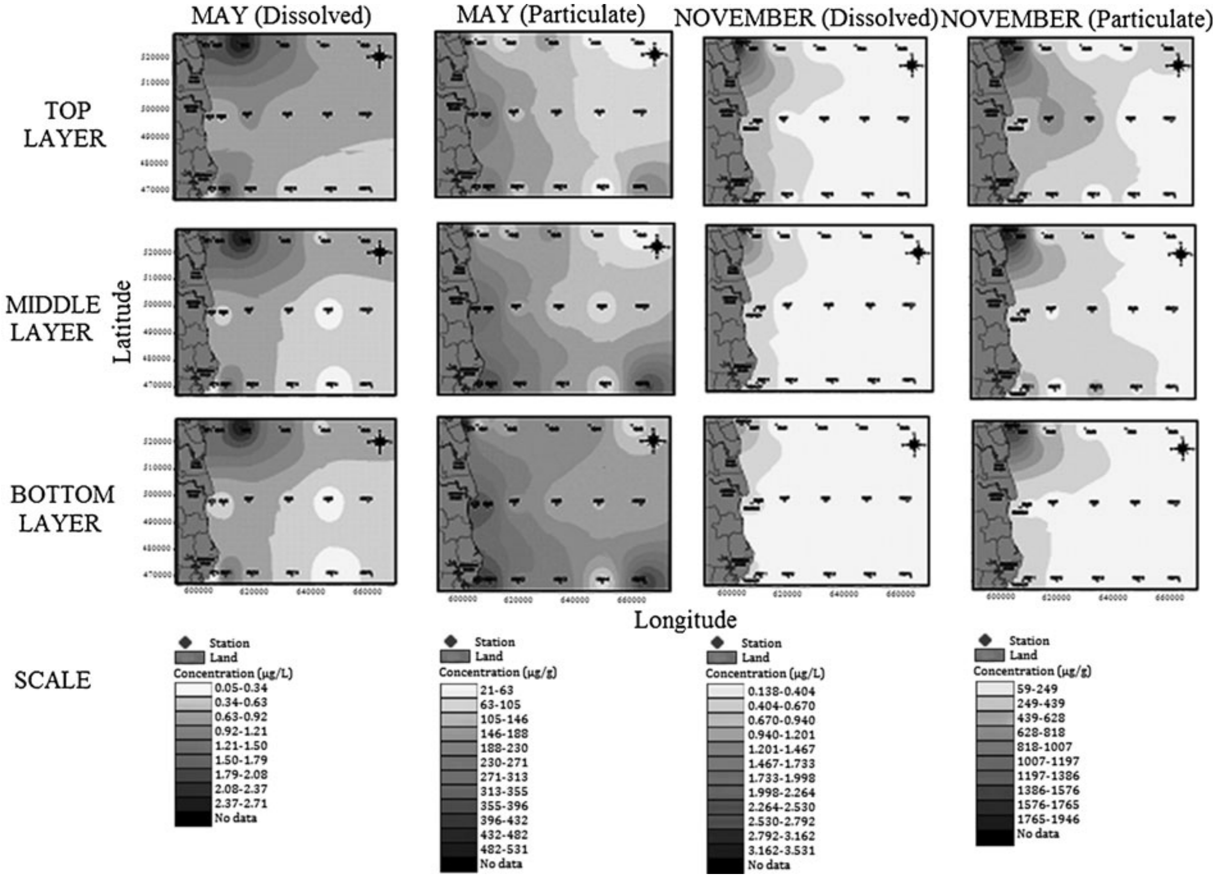


Fig. 4 Distribution of manganese in the southern coast of Terengganu

values in May 2007 are most likely contributed by the iron, steel, and petrochemical industries which is located along the coastline by which these industries residues were being run-off and flush-out into the southern SCS.

Based on the Pearson correlation ($p<0.05$, $r\geq 0.5$), more elements (Fig. 7) were correlated with each other during the November 2007 wet season, in contrast to the May 2007 dry season. This suggest that high current turbulence during the northeast monsoon season in November 2007 lead to re-suspension of surface sediment and the release of some metals from the SPM in the water column due to the strong current velocity (Fig. 2), as compared with May 2007. Owens and Balls (1997) showed that the removal of Cd, Cu, Mn, Ni, and Zn to the dissolved phase occurs in the turbidity maximum zone, and the release of the metals from the sediment system is high due to the higher re-suspension rate of particle sediment into the water column. Liu et al. (2003) documented that the average clay composition in the SCS region off the coast of Peninsular Malaysia is mainly made up of smectite and illite. These materials have a greater ability to exchange metal ions in the sediment than other clay varieties (Bedoui et al. 2008; Helios-Rybicka and Kyziol 1991). The high level of trace metals discharged into the water column and the higher SPM released from the surface sediment during strong current velocity are related.

Figure 8 shows the percentages of metals partitioning between the dissolved and particulate phase in the study area measured during May 2007 and November 2007. During May 2007, the metals were mainly found in the dissolved form, whereas during November 2007, more than 50 % of Cd was found in the particulate phase as effected by high re-suspension of surface sediment into the water column due to the strong current movement. The other elements were predominant in the dissolved phase.

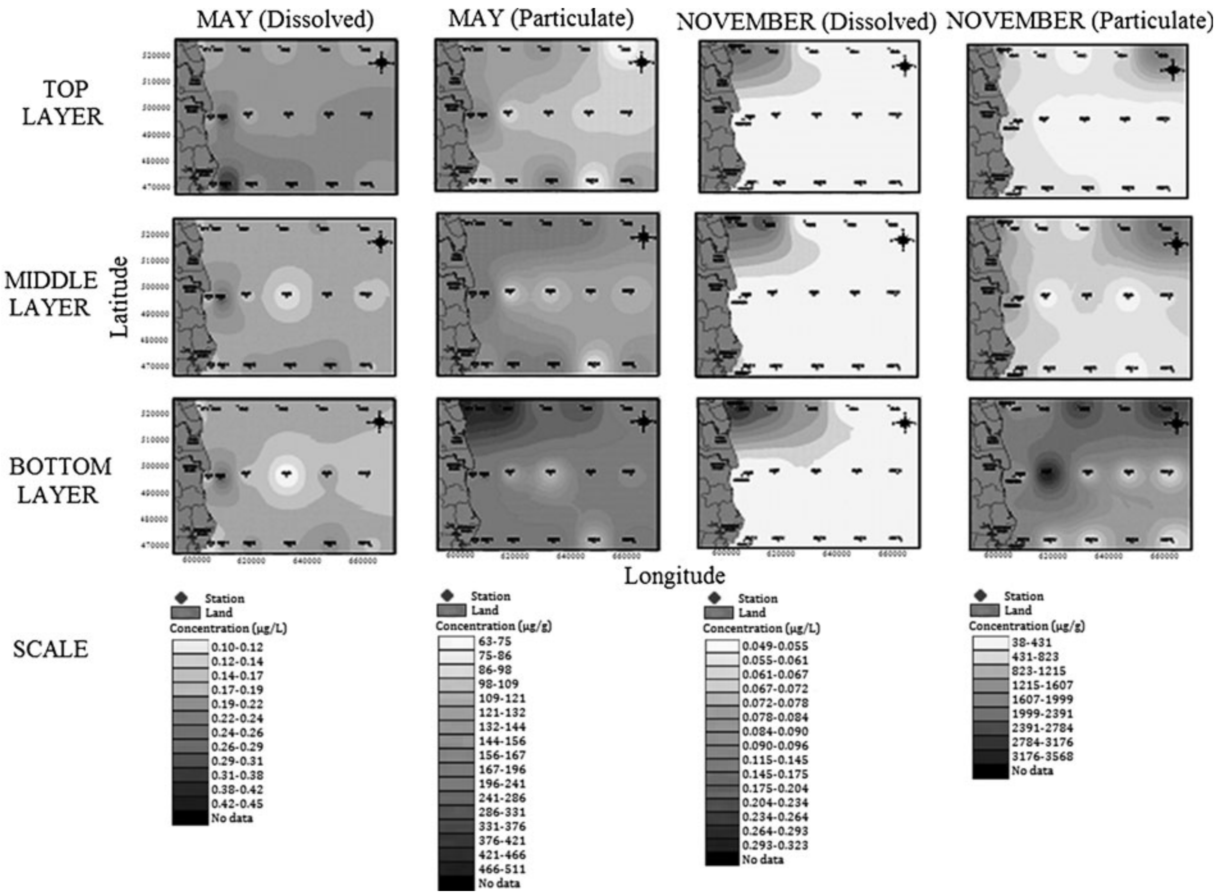


Fig. 5 Distribution of chromium in the southern coast of Terengganu

The distribution coefficient K_D commonly quantifies the partitioning of a metal between the particulate (>0.45 μm) and dissolved (<0.45 μm) phases (Benoit et al. 1994; Benoit and Rozan 1999; Munksgaard and Parry 2001; Nguyen et al. 2005):

$$K_D = \frac{[\text{particulate metal concentration}](\mu g/kg)}{[\text{dissolved metal concentration}](\mu g/L)}$$

K_D is a measure of the tendency of an element to be associated and transported with the particulate phase. High particle reactivity for a metal would tend to increase that metal's K_D value. Partitioning coefficient in the study area decreased following the sequence of Cd>Cr>Pb>Mn. This is similar to results obtained at the Terengganu River estuary by William (2008), who observed an order of Cr>Pb>Cu>Co>Zn. The highest mean log K_D values were for Cd and Cr, confirming that these metals have strong affinities for SPM while Pb and Mn, with the lowest log K_D, have the lowest affinities for SPM (Bibby and Webster-Brown 2006).

In May 2007, we found a positive correlation between log K_D and SPM levels for all trace metals in the southern coast of Terengganu (Fig. 9). For the November 2007 samples, log K_D of Cr and Mn were found to be negatively correlated with SPM whereas a positive correlation of log K_D with SPM was obtained for Pb and Cd. Similar positive correlations between log K_D of various metals and SPM concentrations were found in the Waikato and Kaipara rivers in New Zealand, the Sagami and Wakasa Bay in Japan, as well as the Danshuei River estuary in Taiwan (Bibby and Webster-Brown 2006; Jiann et al. 2005; Takata et al. 2010). The apparent positive correlation for the metals was likely due to the desorption reaction, where SPM concentrations were low, caused

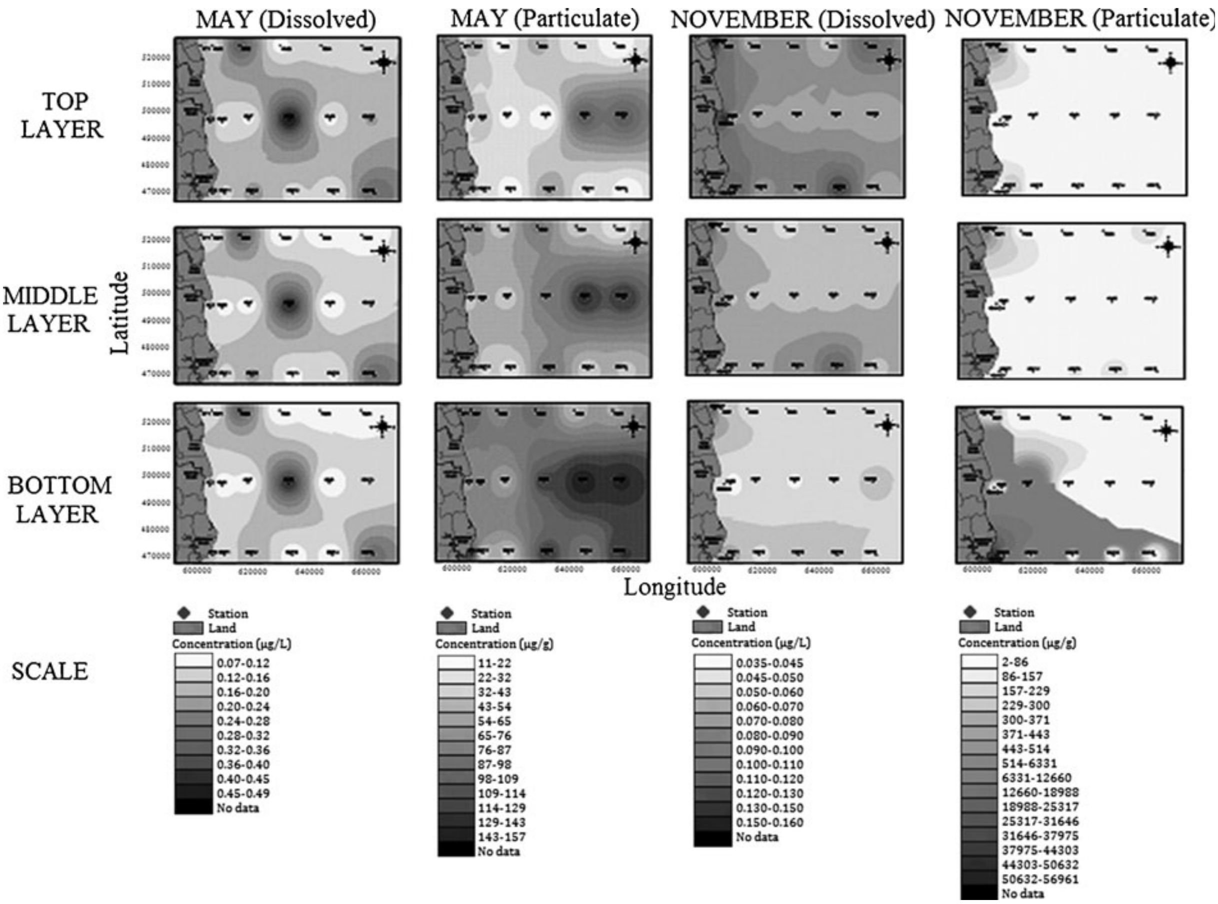


Fig. 6 Distribution of lead in the southern coast of Terengganu

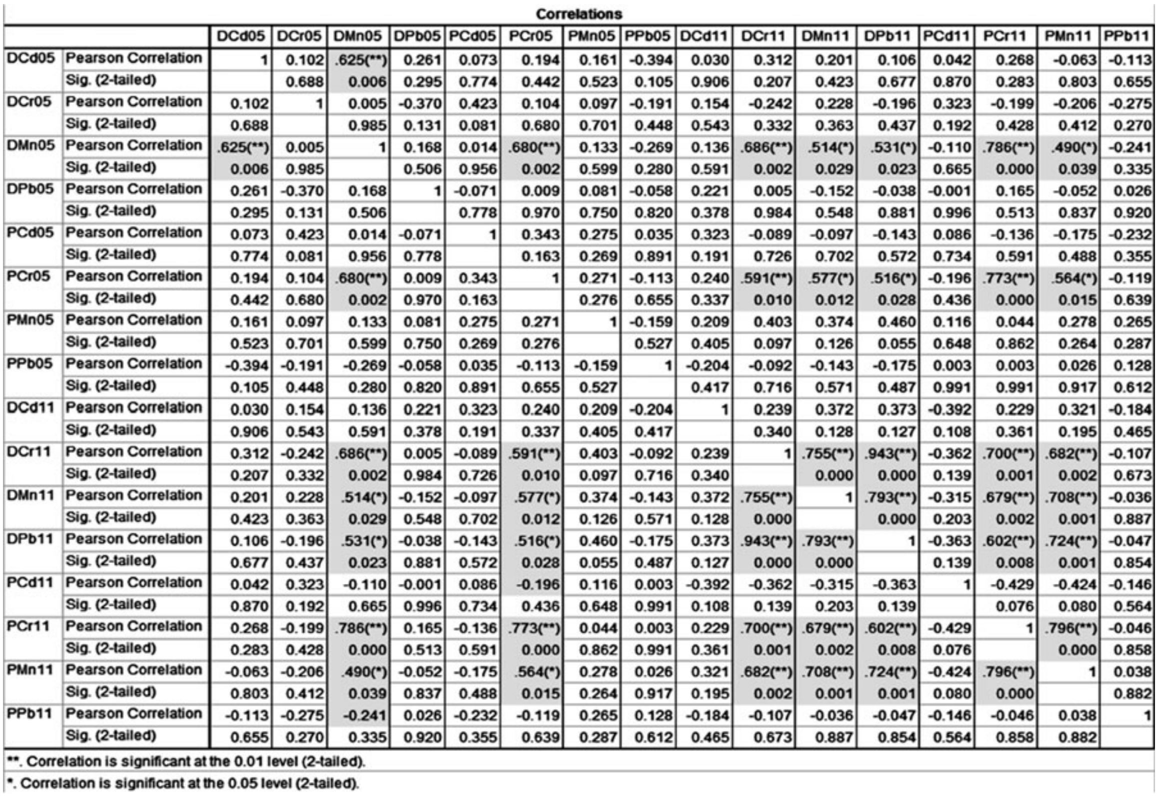
by the competition with Cl$^-$ ions, thus lowering K_D at low SPM concentrations.

Earlier studies have found the inverse relationship between log K_D and SPM levels at the North Australian coast (Munksgaard and Parry 2001), Lake Balaton, Hungary (Nguyen et al. 2005), and the Port Jackson estuary (Sydney Harbour), Australia (Hatje et al. 2003). We attribute the decrease in log K_D values with SPM concentrations, called the "particulate concentration effect" (PCE), to heterogeneity effects of particle size and composition, including the presence of colloidal organic matter (Benoit et al. 1994; Tang et al. 2002). It is possible to obtain PCE if the concentrations of colloids increase in proportion to the quantity of suspended macro-particles and then decrease in the apparent coefficient with increased SPM (Benoit et al. 1994).

A comparison of published data for this region suggests that the dissolved and particulate concentrations of Cd, Cr, Mn, and Pb found in the present study are generally low and within the values found in comparable waters.

Conclusion

The present study indicates that the metal concentrations observed in the study area which is closed by the coastline were highly influenced by anthropogenic activities. The climatic changes between NE and SW monsoon had a major influence on metal partitioning as well as affecting the metals distribution in the waters off the southern Terengganu coast.



Correlations

		DCd05	DCr05	DMn05	DPb05	PCd05	PCr05	PMn05	PPb05	DCd11	DCr11	DMn11	DPb11	PCd11	PCr11	PMn11	PPb11
DCd05	Pearson Correlation	1	0.102	.625(**)	0.261	0.073	0.194	0.161	-0.394	0.030	0.312	0.201	0.106	0.042	0.268	-0.063	-0.113
	Sig. (2-tailed)		0.688	0.006	0.295	0.774	0.442	0.523	0.105	0.906	0.207	0.423	0.677	0.870	0.283	0.803	0.655
DCr05	Pearson Correlation	0.102	1	0.005	-0.370	0.423	0.104	0.097	-0.191	0.154	-0.242	0.228	-0.196	0.323	-0.199	-0.206	-0.275
	Sig. (2-tailed)	0.688		0.985	0.131	0.081	0.680	0.701	0.448	0.543	0.332	0.363	0.437	0.192	0.428	0.412	0.270
DMn05	Pearson Correlation	.625(**)	0.005	1	0.168	0.014	.680(**)	0.133	-0.269	0.136	.686(**)	.514(*)	.531(*)	-0.110	.786(**)	.490(*)	-0.241
	Sig. (2-tailed)	0.006	0.985		0.506	0.956	0.002	0.599	0.280	0.591	0.002	0.029	0.023	0.665	0.000	0.039	0.335
DPb05	Pearson Correlation	0.261	-0.370	0.168	1	-0.071	0.009	0.081	-0.058	0.221	0.005	-0.152	-0.038	-0.001	0.165	-0.052	0.026
	Sig. (2-tailed)	0.295	0.131	0.506		0.778	0.970	0.750	0.820	0.378	0.984	0.548	0.881	0.996	0.513	0.837	0.920
PCd05	Pearson Correlation	0.073	0.423	0.014	-0.071	1	0.343	0.275	0.035	0.323	-0.089	-0.097	-0.143	0.086	-0.136	-0.175	-0.232
	Sig. (2-tailed)	0.774	0.081	0.956	0.778		0.163	0.269	0.891	0.191	0.726	0.702	0.572	0.734	0.591	0.488	0.355
PCr05	Pearson Correlation	0.194	0.104	.680(**)	0.009	0.343	1	0.271	-0.113	0.240	.591(**)	.577(*)	.516(*)	-0.196	.773(**)	.564(*)	-0.119
	Sig. (2-tailed)	0.442	0.680	0.002	0.970	0.163		0.276	0.655	0.337	0.010	0.012	0.028	0.436	0.000	0.015	0.639
PMn05	Pearson Correlation	0.161	0.097	0.133	0.081	0.275	0.271	1	-0.159	0.209	0.403	0.374	0.460	0.116	0.044	0.278	0.265
	Sig. (2-tailed)	0.523	0.701	0.599	0.750	0.269	0.276		0.527	0.405	0.097	0.126	0.055	0.648	0.862	0.264	0.287
PPb05	Pearson Correlation	-0.394	-0.191	-0.269	-0.058	0.035	-0.113	-0.159	1	-0.204	-0.092	-0.143	-0.175	0.003	0.003	0.026	0.128
	Sig. (2-tailed)	0.105	0.448	0.280	0.820	0.891	0.655	0.527		0.417	0.716	0.571	0.487	0.991	0.991	0.917	0.612
DCd11	Pearson Correlation	0.030	0.154	0.136	0.221	0.323	0.240	0.209	-0.204	1	0.239	0.372	0.373	-0.392	0.229	0.321	-0.184
	Sig. (2-tailed)	0.906	0.543	0.591	0.378	0.191	0.337	0.405	0.417		0.340	0.128	0.127	0.108	0.361	0.195	0.465
DCr11	Pearson Correlation	0.312	-0.242	.686(**)	0.005	-0.089	.591(**)	0.403	-0.092	0.239	1	.755(**)	.943(**)	-0.362	.700(**)	.682(**)	-0.107
	Sig. (2-tailed)	0.207	0.332	0.002	0.984	0.726	0.010	0.097	0.716	0.340		0.000	0.000	0.139	0.001	0.002	0.673
DMn11	Pearson Correlation	0.201	0.228	.514(*)	-0.152	-0.097	.577(*)	0.374	-0.143	0.372	.755(**)	1	.793(**)	-0.315	.679(**)	.708(**)	-0.036
	Sig. (2-tailed)	0.423	0.363	0.029	0.548	0.702	0.012	0.126	0.571	0.128	0.000		0.000	0.203	0.002	0.001	0.887
DPb11	Pearson Correlation	0.106	-0.196	.531(*)	-0.038	-0.143	.516(*)	0.460	-0.175	0.373	.943(**)	.793(**)	1	-0.363	.602(**)	.724(**)	-0.047
	Sig. (2-tailed)	0.677	0.437	0.023	0.881	0.572	0.028	0.055	0.487	0.127	0.000	0.000		0.139	0.008	0.001	0.854
PCd11	Pearson Correlation	0.042	0.323	-0.110	-0.001	0.086	-0.196	0.116	0.003	-0.392	-0.362	-0.315	-0.363	1	-0.429	-0.424	-0.146
	Sig. (2-tailed)	0.870	0.192	0.665	0.996	0.734	0.436	0.648	0.991	0.108	0.139	0.203	0.139		0.076	0.080	0.564
PCr11	Pearson Correlation	0.268	-0.199	.786(**)	0.165	-0.136	.773(**)	0.044	0.003	0.229	.700(**)	.679(**)	.602(**)	-0.429	1	.796(**)	-0.046
	Sig. (2-tailed)	0.283	0.428	0.000	0.513	0.591	0.000	0.862	0.991	0.361	0.001	0.002	0.008	0.076		0.000	0.858
PMn11	Pearson Correlation	-0.063	-0.206	.490(*)	-0.052	-0.175	.564(*)	0.278	0.026	0.321	.682(**)	.708(**)	.724(**)	-0.424	.796(**)	1	0.038
	Sig. (2-tailed)	0.803	0.412	0.039	0.837	0.488	0.015	0.264	0.917	0.195	0.002	0.001	0.001	0.080	0.000		0.882
PPb11	Pearson Correlation	-0.113	-0.275	-0.241	0.026	-0.232	-0.119	0.265	0.128	-0.184	-0.107	-0.036	-0.047	-0.146	-0.046	0.038	1
	Sig. (2-tailed)	0.655	0.270	0.335	0.920	0.355	0.639	0.287	0.612	0.465	0.673	0.887	0.854	0.564	0.858	0.882	

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

Fig. 7 Correlation between elements during May 2007 and November 2007

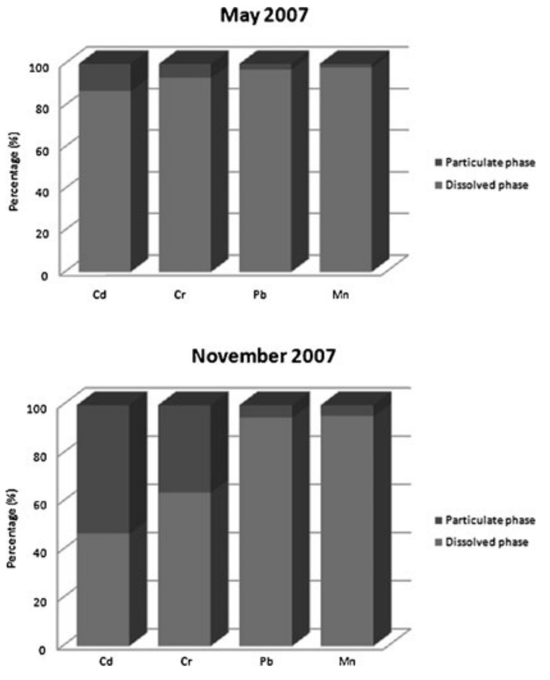


Fig. 8 Percentage of dissolved and particulate components of trace metals recorded during the study

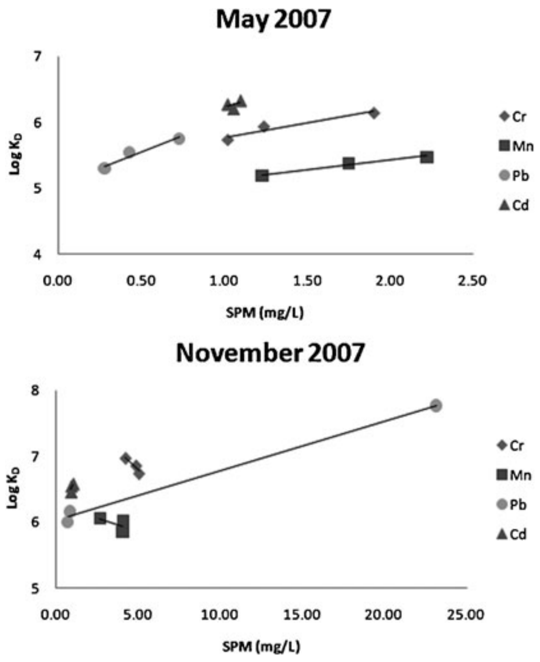


Fig. 9 Partitioning coefficients K_D of trace metals as a function of SPM concentrations in the Terengganu coastal waters

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References

Bedoui, K., Bekri-Abbes, I., & Srasra, E. (2008). Removal of cadmium (II) from aqueous solution using pure smectite and Lewatite S 100: The effect of time and metal concentration. *Desalination, 223*, 269–273.

Benoit, G., Oktay-Marshall, S. D., Cantu, A., II, Hood, E. M., Coleman, C. H., Corapcioglu, M. O., & Santschi, P. H. (1994). Partitioning of Cu, Pb, Ag, Zn, Fe, Al and Mn between filter-retained particles, colloids and solution in six Texas estuaries. *Marine Chemistry, 45*, 307–336.

Benoit, G., & Rozan, T. F. (1999). The influence of size distribution on the particle concentration effect and trace metal partitioning in rivers. *Geochimica et Cosmochimica Acta, 63*, 113–127.

Bibby, R. L., & Webster-Brown, J. G. (2006). Trace metal adsorption onto urban stream suspended particulate matter (Auckland region, New Zealand). *Applied Geochemistry, 21*, 1135–1151.

Censi, P., Spoto, S. E., Sprovieri, M., Mazzola, S., Nardone, G., Di Geronimo, S. I., Punturo, R., & Ottonelo, D. (2006). Heavy metals in the coastal water systems. A case study from the northwestern Gulf of Thailand. *Chemosphere, 64*, 1167–1176.

Charriau, A., Ludovic, L., Gao, Y., Leermakers, M., Baeyens, W., Ouddane, B., & Billon, G. (2011). Trace metal behaviour in riverine sediments: Role of organic matter and sulphides. *Applied Geochemistry, 26*, 80–90.

Cuong, D. T., Karuppiah, S., & Obbard, J. P. (2007). Distribution of heavy metals in the dissolved and suspended phase of the sea-surface microlayer, seawater column and in sediments of Singapore's coastal environment. *Environment and Monitoring Assessment*. doi:10.1007/s10661-007-9795-y.

Guieu, C., Huang, W. W., Martin, J.-M., & Yong, Y. Y. (1996). Outflow of trace metals into Laptev Sea by the Lena River. *Marine Chemistry, 53*, 255–267.

Hatje, V., Apte, S. C., Hales, L. T., & Birch, G. F. (2003). Dissolved trace metal distributions in Port Jackson estuary (Sydney Harbour), Australia. *Marine Pollution Bulletin, 46*, 719–730.

Helios-Rybicka, E., & Kyziol, J. (1991). Clays and clay minerals as the natural barriers for heavy metals in pollution mechanisms—Illustrated by Polish rivers and soils. *Mitt.österr. geol. Ges, 83*, 163–176.

Hunter, K. A. and Boyd, P. (1997). *Has trace metal marine biogeochemistry come of age*? In R. B. Macaskill (ed.), Proceedings of the Trace Element Group of New Zealand, Waikato University, Hamilton, New Zealand.

Jiann, K.-T., Wen, L.-S., & Santschi, P. H. (2005). Trace metal (Cd, Cu, Ni and Pb partitioning, affinities and removal in the Danshuei River estuary, a macro-tidal, temporally anoxic estuary in Taiwan. *Marine Chemistry, 96*, 293–313.

Jiménez, M. S., Velarte, R., & Castillo, J. R. (2002). Performance of different preconcentration columns used in sequential injection analysis and inductively coupled plasma-mass spectrometry for multielement determination in seawater. *Spectrochimica Acta Part B, 57*, 391–402.

Kamaruzzaman, B. Y., Rosnan, Y., Shazili, N. A. M., & Nor Antonina, A. (2002). Physico-chemical characteristics and dissolved trace metals in the Chukai River estuary, Malaysia. *ACGC Chemical Research Communications, 15*, 41–50.

Liu, Z., Trentesaux, A., Clemens, S. C., Colin, C., Wang, P., Huang, B., & Boulay, S. (2003). Clay mineral assemblages in the northern South China Sea: Implications for the East Asian monsoon evolution over the past 2 million years. *Marine Geology, 201*, 133–146.

Munksgaard, N. C., & Parry, D. L. (2001). Trace metals, arsenic and lead isotopes in dissolved and particulate phase of North Australian coastal and estuarine seawater. *Marine Chemistry, 75*, 165–184.

Nguyen, H. L., Leermakers, M., Elskens, M., Ridder, F. D., Doan, T. H., & Baeyens, W. (2005). Correlations, partitioning and bioaccumulation of heavy metals between different compartments of Lake Balaton. *Science of the Total Environment, 341*, 211–226.

Owens, R. E., & Balls, P. W. (1997). Dissolved trace metals in the Tay Estuary. *Estuarine, Coastal and Shelf Science, 44*, 421–434.

Scoullos, M. J., Sakellari, A., Giannopoulou, K. P., & Dassenaki, M. (2007). Dissolved and particulate trace metal levels in the Saronikos Gulf, Greece, in, 2004. The impact of the primary wastewater treatment plant of Psittalia. *Desalination, 210*, 98–109.

Shamsuddin, A. A. (1996). *Distribution of some heavy metals in the Kemaman coast*. Terengganu: Universiti Pertanian Malaysia, Malaysia.

Shazili, N. A. M., Yunus, K., Ahmad, A. S., Abdullah, N., & Rashid, M. K. A. (2006). Heavy metal pollution status in the Malaysian aquatic environment. *Aquatic Ecosystem Health & Management, 9*(2), 137–145.

Soli, A. L., Stewart, A. I., & Byrne, R. H. (2008). The influence of temperature on $PbCO_3^0$ formation in seawater. *Marine Chemistry, 110*, 1–6.

Takata, H., Aono, T., Tagami, K., & Uchida, S. (2010). Processes controlling cobalt distribution in two temperate estuaries, Sagami Bay and Wakasa Bay, Japan. *Estuarine, Coastal and Shelf Science, 89*, 294–305.

Tang, D., Warnken, K. W., & Santschi, P. H. (2002). Distribution and partitioning of trace metals (Cd, Cu, Ni, Pb, Zn) in Galveston Bay waters. *Marine Chemistry, 78*, 29–45.

Valdés, J., Román, D., Alvarez, G., Ortlieb, L., & Guiñez, M. (2008). Metals content in surface waters of an upwelling system of the Northern Humboldt Current (Mejillones Bay, Chile). *Journal of Marine Systems, 71*, 18–30.

William, W. H. T. (2008). *Distribution of dissolved and particulate trace metals in Terengganu River estuary.* Malaysia: Universiti Malaysia Terengganu.