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CADMIUM, MANGANESE AND LEAD DISTRIBUTION IN THE SOUTH CHINA SEA OFF THE SOUTH TERENGGANU COAST, MALAYSIA DURING POST-MONSOON AND PRE-MONSOON

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Abstract: Concentrations of cadmium, manganese and lead were determined in the water column of the South China Sea off the South Terengganu coast in May 2007 and September 2007. Seawater samples were collected from the surface, middle and bottom layers, in a grid of 18 stations 25 km apart. The dissolved metals were extracted on-board ship and preconcentrated on-line using Chelex-100 packed in Teflon tubing after filtration of seawater. The particulate metals were digested under microwave heating. Trace metals were analysed using ICP-MS and validated against CASS-4 and MESS-3. The concentration of dissolved and particulate metals were in the range of 0.002-0.194 µg/L and 45-249 µg/g for Cd, 0.04-2.71 µg/L and 21-2885 µg/g for Mn and 0.03-0.49 µg/L and 10-4277 µg/g for Pb respectively. All metals were dominantly in the dissolved phase except Cd which was predominantly in the particulate phase during September 2007. The order of mean log K_D found was Cd>Pb>Mn. Positive correlation of log K_D with suspended particulate matter were found during both sampling periods except during September 2007, Cd was negatively correlated with suspended particulate matter. The present measurements were to provide baseline data for any future comparison of trace metals distribution in the South China Sea off Southern Terengganu coast.

KEYWORDS: *dissolved metals, particulate metals, Chelex-100, South China Sea*

Introduction

Trace metals exist in sea water in both dissolved and particulate forms. Their concentration depends on such as natural and anthropogenic sources, suspended particulate matter amount and composition, salinity, pH and redox (Hunter *et al*., 1997) whereas their bioavailability depends on the process of complexation with organic matter (Censi *et al*., 2006).

Generally metals released from natural and anthropogenic sources will interact with surrounding activities and lead to the metal complexation and speciation which ultimately influence accumulation or excretion in organisms (Nguyen *et al*., 2005). Biogeochemical cycling processes associated with gravitational settlement and decomposition strongly control the metal vertical distribution in the water column (Cuong *et al*., 2008). For example, the vertical distribution of dissolved cadmium is characterised by a surface depletion and deep-water enrichment, which corresponds to the pattern of nutrient concentrations in the water column.

Although a number of studies have been done on the distribution and fate of heavy metals in the marine environment (Norisuye *et al*., 2007; Scoullos *et al*., 2007; Manfra *et al*., 2005 & Sokolowski *et al*., 2001), available data for the South China Sea region is sparse. There have been a few studies providing data on trace metal concentrations in the marine environment of the South China Sea region but much of the work was focussed on sediments and marine organisms (Shazili *et al*., 2006). Research on dissolved and particulate metals in sea water is scarce due to their trace-level existence (Jiménez *et al*., 2002) and the need for special precautions from sample collection to the analysis process (Hunter *et*

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Table 1: Concentration of trace metals in seawater in the South China Sea off the South Terengganu coast. (Source Shamsuddin, 1996)

Sampling trip	**Element**	**Concentration**	
		Dissolved (ug/L)	**Particulate (ug/g)**
May 2007	Cadmium	0.02 – 0.19	50 - 249
	Manganese	0.05 – 2.71	21 - 531
	Lead	0.07 – 0.49	11 - 157
September 2007	Cadmium	0.002 – 0.06	45 - 126
	Manganese	0.04 – 0.59	73 - 2885
	Lead	0.03 – 0.27	10 - 4277

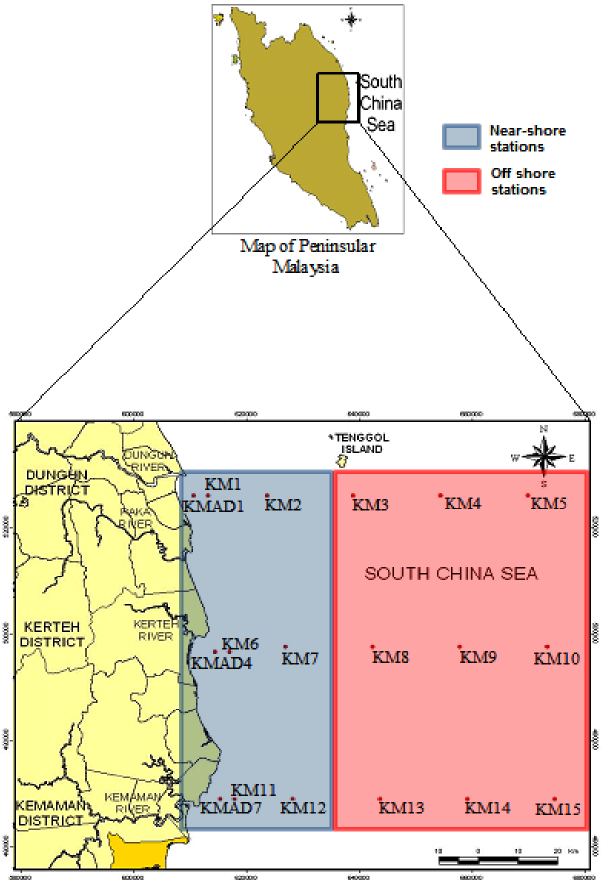
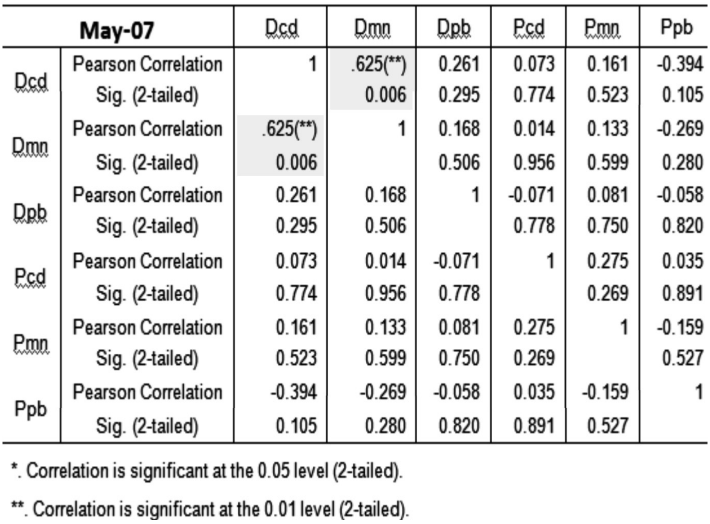


Figure 1: Sampling station off the South Terenggganu coast.

al., 1997). A better knowledge of trace-metal concentrations in sea water is needed in order to determine the seasonal changes in these trace-metal concentrations including the effects of the Northeast monsoon. A few studies (Cuong *et al.*, 2008; Fang *et al.*, 2006; Kamaruzzaman *et al.*, 2002 & Ahmad, 1996) indicated that the metal concentrations in sea water within this region are generally comparable except in certain areas that may be polluted by certain metals due to agricultural, industrial run-off, effluent discharge and domestic activities.

Shamsuddin (1996) reported that the trace metal concentrations in dissolved and particulate phase in seawater samples and sediments off the Kemaman coast were comparable to trace-metal

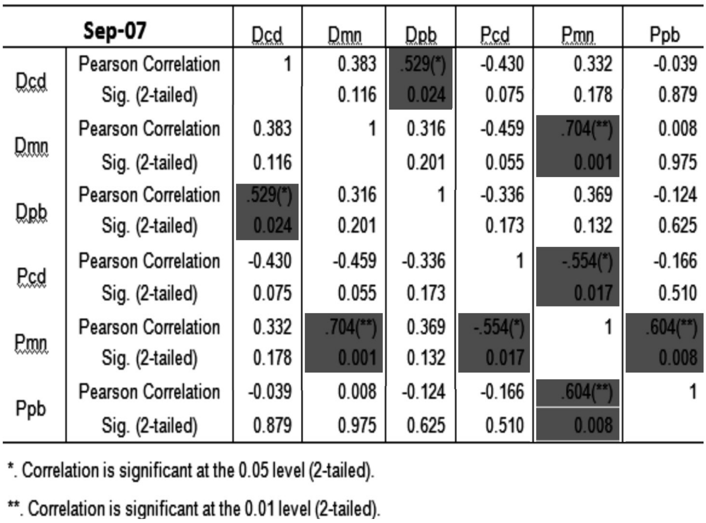
Table 2: Concentration between elements during May 2007 and September 2007.



May-07		Dcd	Dmn	Dpb	Pcd	Pmn	Ppb
Dcd	Pearson Correlation	1	.625(**)	0.261	0.073	0.161	-0.394
	Sig. (2-tailed)		0.006	0.295	0.774	0.523	0.105
Dmn	Pearson Correlation	.625(**)	1	0.168	0.014	0.133	-0.269
	Sig. (2-tailed)	0.006		0.506	0.956	0.599	0.280
Dpb	Pearson Correlation	0.261	0.168	1	-0.071	0.081	-0.058
	Sig. (2-tailed)	0.295	0.506		0.778	0.750	0.820
Pcd	Pearson Correlation	0.073	0.014	-0.071	1	0.275	0.035
	Sig. (2-tailed)	0.774	0.956	0.778		0.269	0.891
Pmn	Pearson Correlation	0.161	0.133	0.081	0.275	1	-0.159
	Sig. (2-tailed)	0.523	0.599	0.750	0.269		0.527
Ppb	Pearson Correlation	-0.394	-0.269	-0.058	0.035	-0.159	1
	Sig. (2-tailed)	0.105	0.280	0.820	0.891	0.527	

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

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



Sep-07		Dcd	Dmn	Dpb	Pcd	Pmn	Ppb
Dcd	Pearson Correlation	1	0.383	.529(*)	-0.430	0.332	-0.039
	Sig. (2-tailed)		0.116	0.024	0.075	0.178	0.879
Dmn	Pearson Correlation	0.383	1	0.316	-0.459	.704(**)	0.008
	Sig. (2-tailed)	0.116		0.201	0.055	0.001	0.975
Dpb	Pearson Correlation	.529(*)	0.316	1	-0.336	0.369	-0.124
	Sig. (2-tailed)	0.024	0.201		0.173	0.132	0.625
Pcd	Pearson Correlation	-0.430	-0.459	-0.336	1	-.554(*)	-0.166
	Sig. (2-tailed)	0.075	0.055	0.173		0.017	0.510
Pmn	Pearson Correlation	0.332	.704(**)	0.369	-.554(*)	1	.604(**)
	Sig. (2-tailed)	0.178	0.001	0.132	0.017		0.008
Ppb	Pearson Correlation	-0.039	0.008	-0.124	-0.166	.604(**)	1
	Sig. (2-tailed)	0.879	0.975	0.625	0.510	0.008	

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

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

concentrations reported elsewhere (Table 1). Anthropogenic inputs are possibly sources that contribute to metal contamination in the marine environment off the Kemaman coast, including shipping activities at Kemaman port, titanium oxide processing plant, a quarry, fisheries activities and shipyards. A study reported by Kamaruzzaman *et al.* (2002) in the same area found that metal concentrations in the dissolved phase in the water column were much influenced by the natural processes indicating conservative behaviour of the metals in the environment. However, concentrations of some metals were relatively high due to rapid development at the Chukai River area but the activities present are not widespread enough as to cause widespread pollution of the Chukai River estuary system.

Utoomprurkporn and Snidvongs (1999) carried out an investigation in 1995 and 1996 to monitor the levels of dissolved and particulate heavy metals in the South China Sea off Sabah, Sarawak and Brunei Darussalam. High concentrations of Cd, Cr, Pb, Fe and Ni were found at the offshore area during July-August period due to the influence of the water current flowing northward as the wind blew from south to north.

The present measurements are to provide initial information for any future observations of the distribution of trace metals in the dissolved and particulate phase in the South China Sea off the Dungun to Kemaman coast.

Materials and method

Sampling

Dungun, Kerteh and Kemaman are located at the southern part of Terengganu, Malaysia. The Dungun, Paka, Kerteh and Kemaman rivers flow into the South China Sea. Land use within the catchments of Dungun and Kemaman are predominantly agricultural activities with urban development and petrochemical industries situated at the coastline, especially in Kerteh and Kemaman. An oil and gas landing facility is found in the industrial complex at Kerteh. Formerly, iron-ore mining activities were carried out at the rural part of Dungun at Bukit Besi.

Oceanographic cruises were carried out in May 2007 (post-monsoon) and September 2007 (pre-monsoon). Seawater samples were collected from the surface, middle and bottom layers, in a grid of 18 stations, 25 km apart, covering the coast of Southern Terengganu region (Figure 1). The area investigated covered approximately 3289 km^2 with the nearest station located 3.28 km from the coast and the furthest station at 62.34 km from the coast.

The sampling procedure involved the collection of seawater samples with a Mercos sampler from 3 depths, surface layer (1 meter), middle layer (10 meters) and bottom layer (depending on the depth). Water depths varied from 11 m to 60 m.

Analysis

All lab-wares were previously cleaned and soaked in diluted nitric acid (10%) and then dried under laminar flow hood and kept in clean polyethylene plastic bags. All works in the laboratory was carried out under class-100 laminar flow hood to avoid contamination. Metal concentrations were measured using Inductively Coupled Plasma Mass Spectrometer (ICP-MS - Perkin Elmer Elan 9000). ICPMS measurements were carried out in a class-100 clean room in the laboratory.

For dissolved metal analysis, 200 ml of sea water was immediately filtered through 0.45 µm teflon filter fitted on-line with a peristaltic pump forcing the sea water from the bottle samplers through the filter and through teflon tubing with Chelex-100 in a class-100 flow hood, on-board the ship. The Chelex-100 column was washed with ammonium acetate in order to remove the entire saline matrix (Scoullos *et al.*, 2007). The metals were then eluted with 2M nitric acid and kept in centrifuge tubes at 4^{0}C before analysis using ICP-MS.

For particulate trace-metal analysis, suspended particulate matter was filtered through 0.45 µm PTFE membrane filter paper on-board under class-100 laminar flow. Suspended particulate matter retained by 0.45 µm PTFE membrane filter paper was dried under laminar flow hood for several days until a constant weight was obtained. The PTFE membrane filter paper was then digested together with 48% HF, 37% HCl and 65% HNO_3 in closed teflon vessels under microwave heating for 30 minutes at 210^{0}C. The HF was then neutralised by adjusting with saturated boric acid and the sample volume made up to 25 mL with deionised water. Blanks were measured to check for contamination. Digested samples were kept in centrifuge tubes at 4^{0}C before analysis using ICPMS.

For quality assurance, CASS-4 and MESS-3 from NRCC were analysed for their trace-metal contents following all the procedures stated above as per seawater samples (Table 2). All trace analyses were carried out using ICPMS (Perkin Elmer Elan 9000) in a clean room.

Result and discussion

A summary of the results of trace-metal concentrations in the sea water in the South

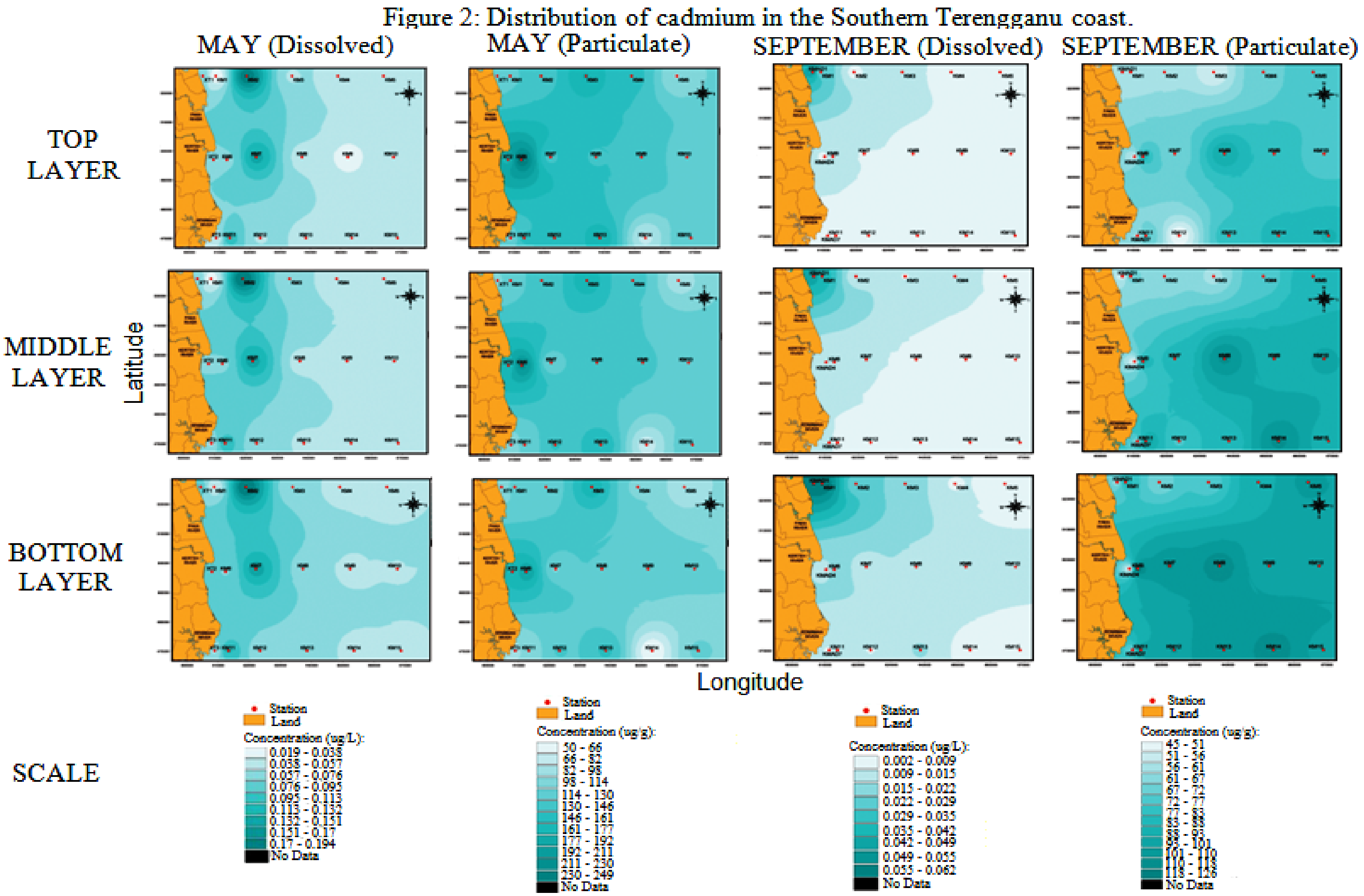


Figure 2: Distribution of cadmium in the South Terengganu coast.

Figure 3: Distribution of manganese in the Southern Terengganu coast.

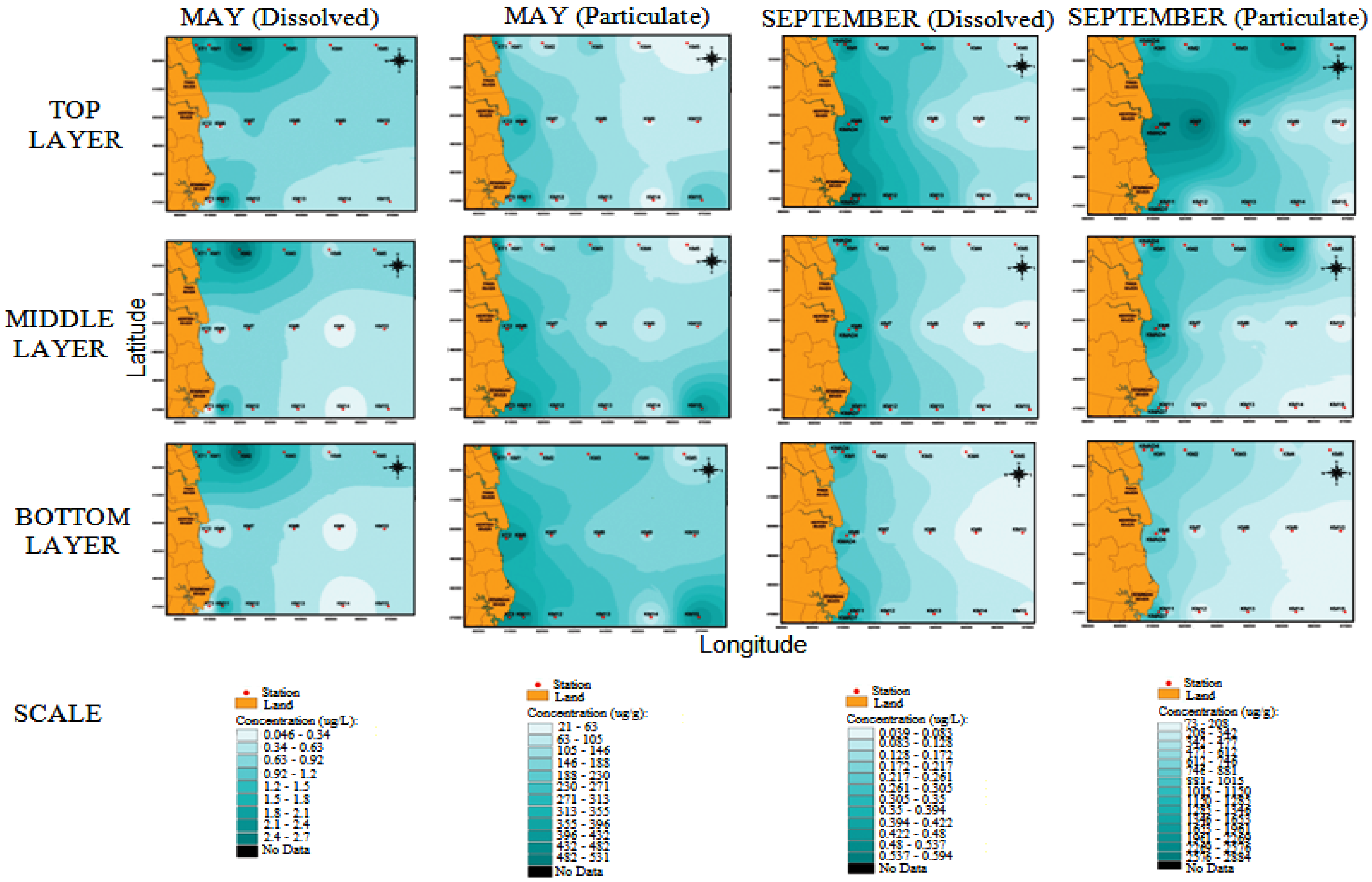


Figure 3: Distribution of manganese in the South Terengganu coast.

Figure 4: Distribution of lead in the Southern Terengganu coast.

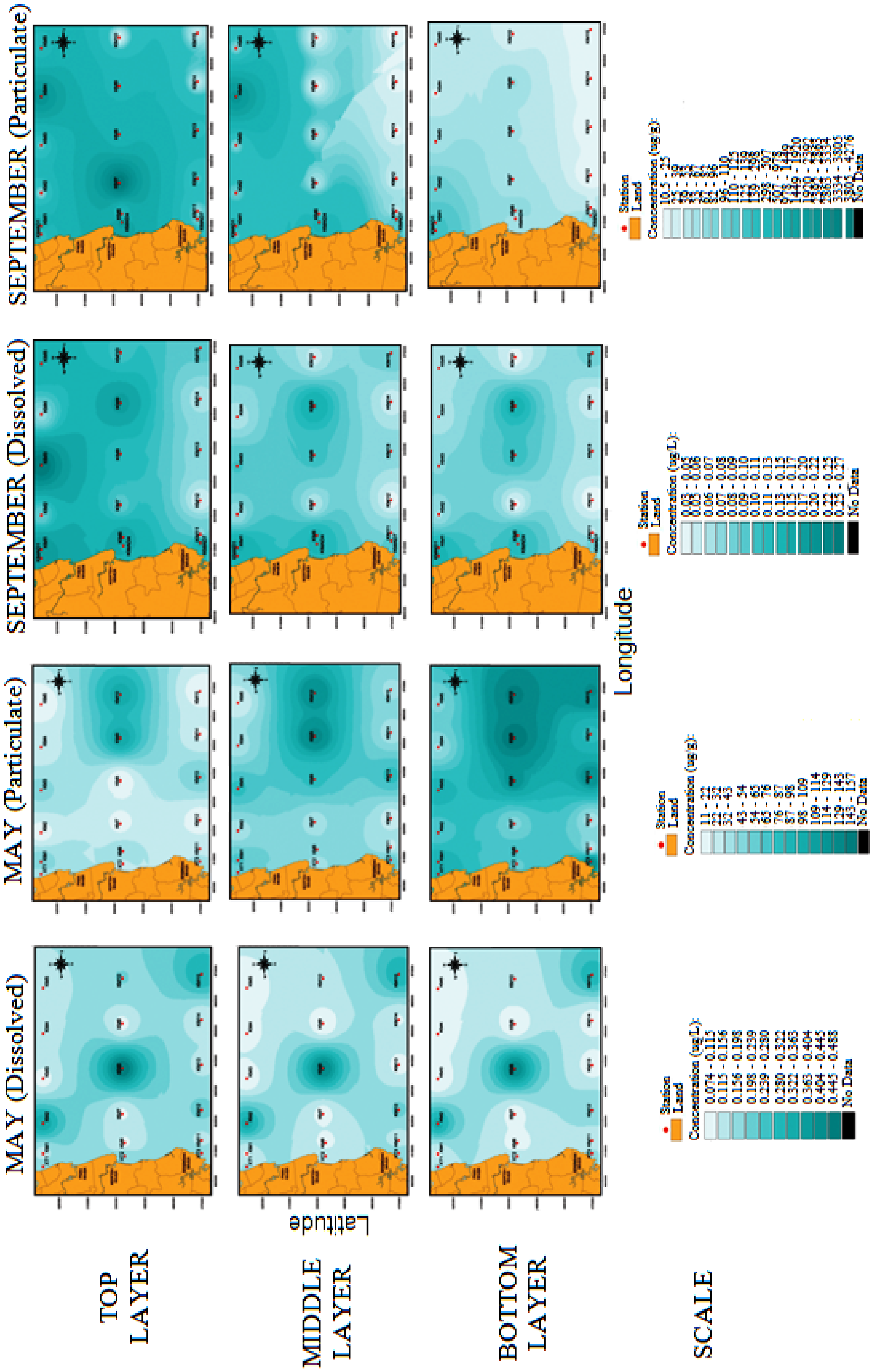


Figure 4: Distribution of lead in the South Terengganu coast.

Table 3: Current flow of the South Terengganu coast.

Sampling	Layer	Current flow (m/s)
May 2007	Top	0.18 ± 0.12
	Middle	0.34 ± 0.10
	Bottom	0.38 ± 0.20
September 2007	Top	0.14 ± 0.05
	Middle	0.63 ± 0.30
	Bottom	0.73 ± 0.31

China Sea off the South Terengganu coast are shown in Table 3. Throughout the period of study, the highest concentration of both dissolved and particulate metals were recorded during May 2007 whereas the lowest concentration of metals was found in September 2007 (Table 3). Overall, dissolved Cd and Pb concentration was low at the surface layer and higher at the bottom water. Meanwhile dissolved Mn showed an opposite pattern where there was high concentration of Mn on the surface and lower at the bottom waters. Particulate Pb and Mn were high at the bottom and lower at the surface waters whereas particulate Cd was low at the surface and high at the bottom waters.

Dissolved Cd (Figure 2) and Mn (Figure 3) distribution in the sampling region showed high concentrations at stations in the vicinity of Dungun River estuary and at near-shore stations. The concentration of dissolved metals was high at station KM2 whereas the concentration of these metals at other stations varied only in small amounts between stations. Particulate Mn and Pb (Figure 4) demonstrated very high concentrations at the northwest region of the study area whilst particulate Cd concentration was low at the same area.

High concentration of metals at near-shore stations, especially off the Dungun River estuary, are probably as a result of river flush-out during water tidal phenomenon as each transect is located parallel to the Dungun River, Paka River, Kerteh River and Kemaman River estuaries. This result is an agreement with Guieu *et al*. (1996) as the outflow of trace metals by Lena River is identified as the point-source of Cu, Ni, Cd, Fe, Pb and Zn into the Laptev Sea, Russia. Furthermore, anthropogenic input from land-based activities include fertilizer production, vegetation, petroleum, metal mining as well as industrial activities are also a common source of these metals towards the South China Sea due to high population of people living at the coast of Terengganu linked to urbanisation areas along the coastline (Zuhairi, 2010; Secretary of Terengganu State, 1999; Utoomprurkporn and Snidvongs, 1999; Shamsuddin, 1996). The variable and seemingly-random distribution of dissolved Pb during all sampling periods has shown elsewhere to be contributed by petroleum industries and metals mining (Swaine, 2000).

Results of Pearson correlation show more elements were correlated during September 2007 compared to May 2007 (Table 4). This data suggest that the ocean current turbulence led to re-suspension of the surface sediment and the release of some metals from the sediment particle towards the water column as the currents were stronger in September 2007 in contrast to May 2007 (Table 4). This is in line with the study carried out by Owens (1997) which showed that removal of metals to dissolved phase occurs in the maximum-turbidity zone which is similar to Mersey Estuary, North West England as dissolved Pb was high with maximum turbidities (Martino *et al.*, 2002).

The partitioning of metals between dissolved and particulate phase in the aquatic environment depends on parameters like dissolved oxygen, pH, salinity, specific metal ion, metal concentration, nature of particle and particle concentration (Munksgaard and Parry, 2001). The percentage of metals partitioning between dissolved and particulate phase in the South China Sea of the Southern Terengganu coast measured in this study is shown in Figure 5.

During May 2007, high proportions (> 80%) of all metals studied were found in the dissolved phase, whereas in September 2007, 39% of Cd was dominant in particulate phase. The majority of the other elements were found in the dissolved phase (> 70%). The implication of the trace metals were dominantly by the dissolved phase was close to the results obtained by William (2008) at Terengganu River estuary as more than 50% of metals are in the dissolved form. In Kalloni Bay, Gavriil and Angelidis

Table 4: Comparison of regional dissolved (μg/L) and particulate (μg/g) trace metal concentration range in seawater.

Location	Cd		Mn		Pb		Reference
	Dissolved	Particulate	Dissolved	Particulate	Dissolved	Particulate	
Kemaman coast, Terengganu	0.01-0.06	<100-400	-	-	0.01-0.29	700-1100	Shamsuddin, 1996
South China Sea off Sabah, Sarawak & Brunei Darussalam	1.124 – 153.988	112.4 – 3259.6	-	-	4.144 – 310.8	< 4144	Utoomprurkporn & Snidvongs, 1999
Chukai river estuary, Kemaman	0.13 – 0.21	-	-	-	1.20 – 1.84	-	Kamaruzzaman *et al.*, 2002
Mekong river delta	3.37 – 10.12	-	-	-	103.6 – 105.67	19 - 42	Cenci, 2004
Northern Taiwan coast	0.006 – 0.028	-	1.7 – 4.84	-	0.018 – 0.086	-	Fang *et al.*, 2006
Kranji and Pulau Tekong, Singapore	0.013-0.109	0.16-0.73	-	-	0.009-0.062	01.10-6.08	Cuong *et al.*, 2008
Dungun to Kemaman coast, Terengganu	0.013 – 0.909	11.34 – 267.51	-	-	0.036 – 1.174	9.16 – 65.99	Zuhairi. 2010
Southern Terengganu coast	0.002 – 0.19	45 - 249	0.04 – 2.71	21 – 2885	0.03 – 0.49	10 – 4277	This study

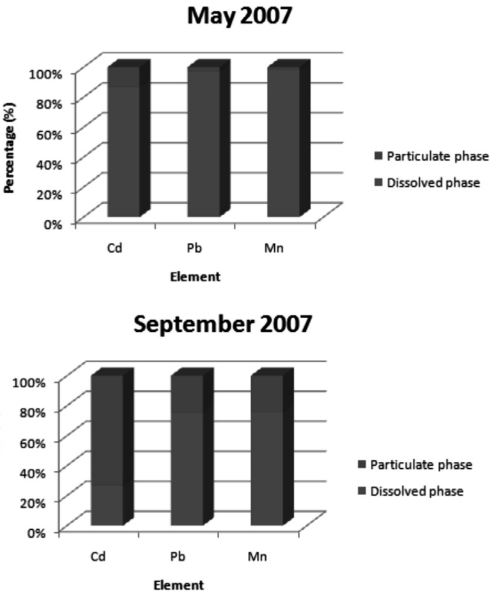


Figure 5: Percentage of dissolved and particulate trace metals as for the total value of each metal during the sampling periods.

(2005) found that dissolved phase is the major carrier for Cd and Pb.

The partitioning of a metal between the particulate (> 0.45 μm) and dissolved (< 0.45 μm) phases is commonly quantified in term of the distribution coefficient K_D (Munksgaard and Parry, 2001; Nguyen *et al.*, 2005).

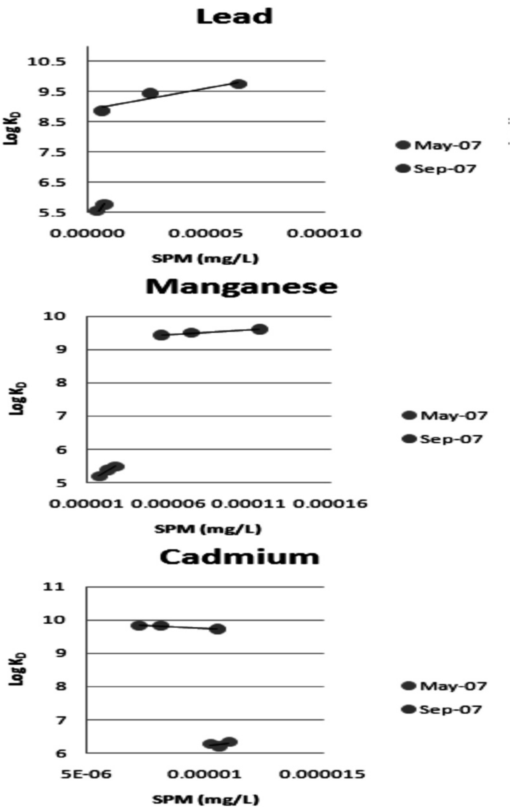


Figure 6: Partitioning coefficeins (Log K_D) of leaf, manganese and cadmium as a function of suspended particulate matter concerntration in the South Terengganu coast.

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

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 coefficients in the South China Sea of the Southern Terengganu coast decreased following the sequence of Cd > Pb > Mn. The highest mean log K_D was for Cd confirming that this metal has the strongest affinity for suspended particulate matter whilst Pb and Mn with the lowest log K_D have the lowest affinities for suspended particulate matter (Bibby and Webster-Brown, 2005).

In the Southern Terengganu coast, a positive log K_D with suspended particulate matter (Figure 6) were found in May 2007 for all trace metals, but throughout September 2007, an inverse relationship of partitioning coefficient and suspended particulate matter was found for Cd, the exception being for Mn and Pb as the log K_D were positively correlated with the suspended particulate matter (Figure 6).

The apparent positive correlation for the metals was likely due to desorption reactions, where suspended particulate matter concentrations were low, caused by the competition with Cl^- ions, thus lowering K_D at low suspended particulate matter concentrations. Similar positive correlation of log K_D with suspended particulate matter was found in Waikato and Kaipara River in New Zealand, Sagami and Wakasa Bay in Japan as well as Danshuei River estuary, Taiwan (Bibby and Webster-Brown, 2005; Jiann *et al.*, 2005; Takata *et al.*, 2010).

The inverse relationship of log K_D with level of suspended particulate matter (Figure 6) was found in the North Australian coast (Munksgaard and Parry, 2001), Lake Balaton, Hungary (Nguyen *et al.*, 2005) and Port Jackson estuary (Sydney Harbour), Australia (Hatje *et al.*, 2003). The decrease in log K_D values with level of suspended particulate matter called the "particle concentration effect" (PCE), were attributed to heterogeneity effects of particle size and composition, including the presence of colloidal organic matter (Benoit *et al.,* 1994; Tang *et al.*, 2002). PCE could be obtained if the concentrations of colloids increase in proportion to the quantity of suspended macro-particles, and then a decrease in the apparent partition coefficient with increasing concentration of suspended particulate matter (Benoit *et al.*, 1994).

A comparison with values measured elsewhere is shown in Table 1. Dissolved and particulate Cd and Mn concentration in this study is generally comparable and lower than many locations in the South China Sea region (Table 1). Levels of dissolved and particulate lead found in the South China Sea off the South Terengganu coast are higher compared to levels (Table 1) reported for Northern Taiwan coast (Fang *et al.*, 2006), Kemaman coast (Shamsuddin, 1996) as well as Kranji and Pulau Tekong, Singapore (Cuong *et al.*, 2008). Values of dissolved lead in the South China Sea off the South Terengganu coast are lower compared to data reported for South China Sea off Sabah, Sarawak and Brunei (Utoomprurkporn and Snidvongs, 1999), Mekong River delta (Cenci, 2004), Chukai river estuary, Kemaman (Kamaruzzaman *et al.*, 2002) and Dungun to Kemaman coast (Zuhairi, 2010).

Conclusions

The present study provides the first information on trace metals in the South China Sea off the Southern Terengganu coast. This Class-100 clean method is reliable and efficient and can be applied on-board ship during the sampling trip. Throughout the study, concentrations of all metals were generally in the range of reported values for the region except for Pb which were higher at a small number of sampling points. Dissolved and particulate metal distributions in the South China Sea off the Southern Terengganu coast showed high concentrations in the vicinity of Dungun River estuary. Concentrations at near-shore stations were higher than at the offshore stations.

The Northeast monsoon plays an important role in the distribution of particulate trace metals as shown by the measured values of particulate phase metals and calculated K_D values. The majority of all metals measured throughout the study were in the dissolved phase. It is concluded that these metals were more dominant in the particulate phase as a result of greater re-suspension of surface sediment into the water column. The proportions of metals in dissolved and particulate forms are supported by K_D (distribution coefficient) calculations from the data obtained. The distribution coefficient (K_D) for metals between the particulate and dissolved phase is similar to reported values for the region. The order of mean log K_D found in the South China Sea of the Southern Terengganu coast was Cd > Pb > Mn. A positive correlation of log K_D with concentration of suspended particulate matter were found in May 2007 for all trace metals but throughout September 2007, an inverse relationship of partitioning coefficient and level of suspended particulate matter were found for all trace metals, except for Mn and Pb where the log K_D was positively correlated with concentration of suspended particulate matter. During November 2007, log K_D of Cr and Mn were negatively correlated with concentration of suspended particulate matter whereas a positively correlation of log K_D with concentration of suspended particulate matter were obtained for Al, Cd, Cu, Fe, Ni, Pb and Zn.

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