

DISTRIBUTION AND SPECIATION OF HEAVY METALS (Cd, Cu AND Ni) IN COASTAL SEDIMENTS OF DUMAI SUMATERA, INDONESIA

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ABSTRACT

Heavy metal concentrations in sediment collected from Dumai coastal waters have been studied by using sequential extraction technique to determine the distribution and geochemical speciation (EFLE, acid reducible, oxidisable organic and resistant) of Cd, Cu and Ni. The results showed that the highest concentrations of total Cd was in Cargo Port area and the lowest in Penyembal, whilst for Cu and Ni the highest concentrations were in Ferry Port and the lowest in Batu Panjang. The total concentrations of Cd, Cu and Ni ranged from 0.65 – 1.82, 1.84 – 13.16 and 7.68 – 17.98 µg/g dry weight, respectively. Higher metal concentrations were detected in the eastern and central parts of Dumai city center where most of anthropogenic activities are concentrated. However, most of the concentrations of Cd, Cu and Ni were still below the ERL and ERM values. Only at few stations, especially in the eastern and central parts of Dumai, showed Cd concentrations exceed the ERL but still well below the ERM values. Metal concentrations in 78.26% (Cd) and 91.30% (Cu and Ni) of the sampling stations were dominated by resistant fraction indicating natural origin of these metals. Non resistant fractions, however, were found higher than resistant fractions at Pelintung, Pertamina, Cargo Port, Penyembal and Batu Panjang for Cd; Penyembal and Batu Panjang for Cu and Pelintung and Guntung for Ni. These findings indicated that anthropogenic inputs of Cd, Cu and Ni occurred in these stations. For all sampling site groups, Cd, Cu and Ni were mostly accumulated in the resistant fraction (55.28 – 58.31%; 65.02 – 91.84% and 50.08 – 66.88% of the total concentrations respectively) which indicated that the mobility and anthropogenic inputs of these metals in Dumai coastal waters were quite low.

Key words : Heavy metal pollution, metal speciation, sediment, Dumai

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INTRODUCTION

Heavy metal contamination in coastal sediment has been an increasing ecotoxicological problem because the coastal area often receives anthropogenic and industrial wastes. These wastes, including heavy metals, which are discharged from

sources such as sewage and industrial effluents may accumulate in the suspended particulate matter and settle on the bottom sediment (Gomez-Parra *et al.*, 2000). Studies on heavy metals in sediment in Indonesian waters still very limited and are restricted in Java Sea (Everaarts, 1989),

Jakarta Bay (Williams *et al.*, 2000) and Central Java (Takarina *et al.*, 2004).

In sediments, heavy metals are present in different geochemical forms which determine their mobilization capacity and bioavailability (Yu *et al.*, 2001). Assessment of heavy metal contamination of the coastal environment based on total metal is not sufficient for an understanding of their environmental behaviour. This is because only a fraction of the total metal is available for biological processes. It would also poses the problem as it does not provide information on which fraction is due to anthropogenic activities (Morillo *et al.*, 2004; Ramirez *et al.*, 2005).

Dumai is one of the main entrance gates for marine transportation in the Straits of Malacca to Sumatera and like many other developing regions in Indonesia, Dumai coastal environment is subjected to negative impacts of the industrial development and anthropogenic activities (Anonymous, 2002; 2004). In the year 2001 there were 66 metals and chemical industries, 22 various industries and 203 agriculture and forest based industries in their operation in Dumai. There were also 6 major oil ports and more than 10 commercial ports where more than 480 tankers and commercial boats and ferries loaded and unloaded in Dumai port each month (Anonymous, 2002). Dumai is also likely to receive impacts from the Straits of Malacca which is one of the busiest international shipping lanes in the world (Abdullah *et al.*, 1999; Chua *et al.*, 2000). Around 900 tankers and commercial vessels and 11 million barrels oil flows pass through the Malacca Strait's waters every day (Gunadi, 2004). According to The Ministry of Communication Indonesia (2004), averagely 70.000 ships annually where 29% of which are super tankers passing the straits. Beside that, the volume of cargo transported in both directions through the Malacca Straits increased substantially in the last two decades. The increasing marine traffic in the Straits poses the Straits in an increasing threat of marine pollution (Nontji, 2004).

Studies on the speciation and distribution of heavy metals in sediments not

only can provide information on the degree of contamination, but also the actual environmental impact on metal bioavailability and mobility as well as their origin (Yap *et al.*, 2002; 2003a,b; 2005; 2006; Morillo *et al.*, 2004; Ramirez *et al.*, 2005, Acevedo-Figueroa *et al.*, 2006). Since heavy metal concentrations in the sediments of Dumai Sumatera has not been reported in the literature, the objective of this study was to determine the baseline concentrations and its distribution of Cd, Cu and Ni in the surface sediment collected from the coastal area of Dumai Sumatera, Indonesia, to evaluate the level of concentration in the context of similar data reported from other geographical areas and to estimate the anthropogenic portions of the metals by comparing between resistant and nonresistant fractions of the metals in the surface sediments.

MATERIALS AND METHODS

Sediment samples were collected in May 2005 from twenty three sampling stations along the coastal area of Dumai, including south and north part of Rupert Island (**Figure. 1**). In order to figure out the distribution of metal contamination in Dumai coastal waters, these stations were divided into five (5) site groups based on the anthropogenic activities: I = East part of Dumai (Station 1-3: newly developed international harbour, proposed industrial park, busy waterway and exposed to the Straits of Malacca); II = Central Dumai (Station 4-11: Oil refinery, tanker berth, densely populated area, cargo and commercial ports and river estuary); III = West part of Dumai (Station 12-16: agriculture area, river estuary, cement factory and fishing village); IV = South part of Rupert (Station 17-19: fishing village, jetties and commercial ports with restaurants) and V = North part of Rupert

(Station 20-23: fishing village and exposed to the Malacca Straits). Samples from each station were collected by using an Ekman grab. The top 3 to 5 cm of each sediment samples was placed in polyethylene plastic bag and they were kept in an ice box. Samples were brought back to laboratory and were oven dried to a constant dry weight at 80°C and sieved through a 63 µm stainless steel sieve. Geochemical fractions of Zn and Pb in the sediments were obtained by using the modified SET (Sequential Extraction Technique) as described by Badri and Aston (1983) and Yap *et al.* (2002). The four fractions considered, the extraction solutions and the conditions employed were:

1). EFLE (Fraction 1): About 10 g of sample was continuously shaken for 3 h with 50 ml 1.0 M ammonium acetate (NH₄CH₃COO), pH 7.0 at room temperature.

2). Acid-reducible (fraction 2): The residue from (1) was continuously shaken for 3 h with 50 ml 0.25 M hydroxyl-ammonium chloride (NH₂OH.HCl) acidified to pH 2 with HCl, at room temperature.

3). Oxidisable-organic (Fraction 3): The residue from (2) was first oxidized with 15 ml hydrogen peroxide (R&M Chemicals 35%) in a water bath at 90 °C. After cooling, the metal released from the organic complexes was continuously shaken for 3 h with 1.0 M ammonium acetate (NH₄CH₃COO) acidified to pH 2.0 with HCl at room temperature.

4). Resistant (Fraction 4): The residue from (3) was digested in a combination of concentrated nitric acid (AnalaR grade, R&M Chemicals 65%) and perchloric acid (AnalaR grade, R&M Chemicals 70%).

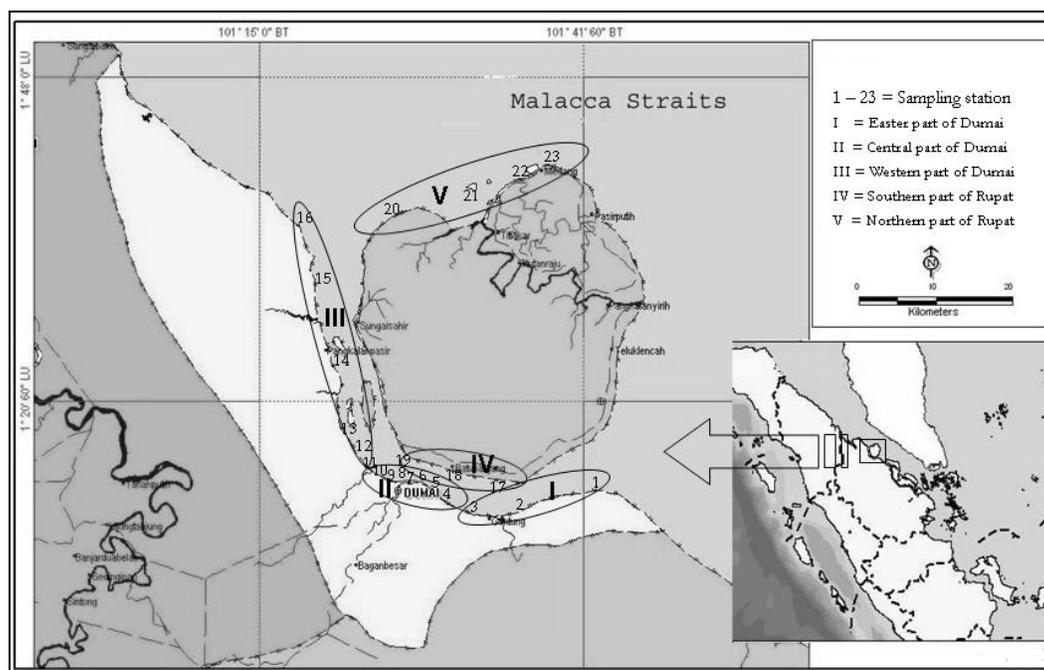


Fig 1. Map of Dumai and the position of sampling stations

Table 1. Mean concentration of Cd in each geochemical fraction of Dumai coastal sediment (Concentration in $\mu\text{g/g d.w} \pm \text{SD}$; $n = 3$)

| No. | Station | Lat. (N) | Long.(E) | EFLE | Acid reducible | Oxidisable organic | Resistant | Total (100%) |
|-----|------------------|-----------|------------|-------------------|-------------------|--------------------|-------------------|--------------|
| 1 | Pelintung | 01°40'23" | 101°41'49" | 0.22±0.01 (15.18) | 0.12±0.02 (8.36) | 0.40±0.03 (28.15) | 0.69±0.24 (48.31) | 1.43 |
| 2 | Guntung | 01°38'35" | 101°34'55" | 0.23±0.02 (15.36) | 0.12±0.02 (8.08) | 0.23±0.05 (15.56) | 0.90±0.13 (60.99) | 1.48 |
| 3 | Mundam | 01°40'12" | 101°30'03" | 0.12±0.04 (8.71) | 0.10±0.05 (7.20) | 0.25±0.03 (18.46) | 0.90±0.16 (65.64) | 1.38 |
| 4 | Pertamina | 01°41'10" | 101°28'52" | 0.14±0.06 (16.57) | 0.17±0.01 (20.68) | 0.16±0.01 (18.43) | 0.37±0.08 (44.31) | 0.84 |
| 5 | Ferry port | 01°41'15" | 101°27'55" | 0.33±0.09 (24.39) | 0.16±0.08 (11.60) | 0.15±0.07 (10.70) | 0.72±0.19 (53.31) | 1.36 |
| 6 | Cargo port | 01°41'19" | 101°27'03" | 0.38±0.02 (20.80) | 0.09±0.01 (4.92) | 0.59±0.76 (32.61) | 0.76±0.19 (41.67) | 1.82 |
| 7 | Sg. Dumai | 01°41'21" | 101°26'14" | 0.19±0.05 (19.72) | 0.10±0.03 (10.43) | 0.13±0.01 (13.23) | 0.56±0.34 (56.62) | 0.98 |
| 8 | Dockyard | 01°41'36" | 101°25'38" | 0.26±0.08 (25.54) | 0.07±0.04 (7.24) | 0.07±0.02 (6.49) | 0.61±0.30 (60.73) | 1.00 |
| 9 | Fishing port | 01°42'03" | 101°24'43" | 0.20±0.01 (20.69) | 0.08±0.01 (8.05) | 0.13±0.02 (13.29) | 0.57±0.18 (57.97) | 0.98 |
| 10 | Purnama | 01°42'34" | 101°24'14" | 0.15±0.04 (18.10) | 0.03±0.00 (3.30) | 0.10±0.00 (12.21) | 0.55±0.35 (66.38) | 0.83 |
| 11 | Sg. Mesjid | 01°43'41" | 101°23'42" | 0.17±0.04 (16.96) | 0.07±0.02 (7.50) | 0.14±0.04 (14.31) | 0.60±0.11 (61.22) | 0.98 |
| 12 | Bangsai Aceh | 01°44'40" | 100°22'54" | 0.17±0.05 (24.87) | 0.04±0.02 (5.73) | 0.12±0.02 (17.53) | 0.36±0.09 (51.87) | 0.69 |
| 13 | Lubuk Gaung | 01°45'36" | 101°22'16" | 0.10±0.04 (11.49) | 0.05±0.03 (5.08) | 0.09±0.02 (9.88) | 0.67±0.42 (73.55) | 0.90 |
| 14 | Penyembal | 01°47'27" | 101°21'43" | 0.14±0.03 (21.89) | 0.12±0.01 (18.55) | 0.16±0.02 (25.38) | 0.22±0.08 (34.18) | 0.65 |
| 15 | Basilam Baru | 01°51'26" | 101°21'10" | 0.08±0.02 (10.25) | 0.10±0.03 (12.86) | 0.10±0.03 (13.76) | 0.48±0.21 (63.13) | 0.76 |
| 16 | Penempul | 01°59'51" | 101°19'55" | 0.12±0.01 (15.14) | 0.12±0.04 (14.37) | 0.08±0.03 (9.43) | 0.50±0.14 (61.06) | 0.82 |
| 17 | Terkul | 0°141'04" | 101°34'43" | 0.15±0.01 (14.16) | 0.13±0.01 (12.10) | 0.13±0.01 (12.18) | 0.64±0.13 (61.56) | 1.05 |
| 18 | Batu Panjang | 01°42'19" | 101°30'41" | 0.11±0.02 (16.40) | 0.07±0.02 (10.69) | 0.16±0.01 (23.82) | 0.33±0.00 (49.09) | 0.67 |
| 19 | Tg. Kapal | 0°143'12" | 101°27'46" | 0.11±0.01 (15.09) | 0.13±0.02 (17.69) | 0.06±0.00 (7.74) | 0.43±0.14 (59.48) | 0.72 |
| 20 | Mombol | 02°04'12" | 101°28'32" | 0.20±0.01 (18.82) | 0.14±0.02 (13.31) | 0.08±0.01 (7.13) | 0.64±0.34 (60.73) | 1.06 |
| 21 | Pulau Babi | 02°05'51" | 101°33'26" | 0.16±0.01 (17.80) | 0.13±0.03 (14.34) | 0.09±0.02 (9.50) | 0.54±0.18 (58.36) | 0.92 |
| 22 | Tg. Medang luar | 02°07'31" | 101°39'10" | 0.23±0.01 (22.26) | 0.07±0.00 (7.06) | 0.17±0.10 (15.88) | 0.57±0.16 (54.80) | 1.04 |
| 23 | Tg. Medang dalam | 02°06'37" | 101°37'40" | 0.19±0.07 (21.27) | 0.05±0.03 (5.97) | 0.17±0.11 (19.33) | 0.47±0.05 (53.43) | 0.87 |

Note : The values in parentheses represent the fraction in percentage (%)

Before the next fractionation was carried out, the residue used for each fraction was weighed. The residue was rinsed with 20 ml milli-Q water (18.2 Ω). It was then filtered through a Whatman[®] No. 1 filter paper and the filtrate was stored until metal determination. For each fraction of the sequential extraction procedure, a blank was employed using the same procedure to ensure that the samples and chemicals used were free of contaminations.

After filtration, the samples were determined for Cd, Cu and Ni by using an air-acetylene flame Atomic Absorption Spectrophotometer (Perkin-Elmer Model Analyst 800 series). The data are presented in $\mu\text{g/g}$ dry weight basis. Multiple-level calibration standards were analyzed to generate calibration curves against which sample concentrations were calculated. Standard solutions of Cd, Cu and Ni were

prepared from 1000-mg/l (BDH SpectrosoL[®]) stock solution.

To avoid possible contamination, all glassware and equipments used were acid-soaked with 15 % HCl for at least 24 hours and then rinsed with DDW. The quality of the method used was checked with a Certified Reference Material (CRM) for Soil (International Atomic Energy Agency, Soil-5, Vienna, Austria). The agreement between the analytical results for the reference material and its certified values for each metal was satisfactory with the recoveries being about 98%, 96%, and 105 % for Cd, Cu and Ni respectively. During the period of metal analysis, a quality control sample was routinely run through for every 5 – 10 samples.

RESULTS AND DISCUSSION

The mean metal concentrations, their standard deviations and percentages of four geochemical fractions for each station are presented in **Table 1, 2 and 3** for Cd, Cu and Ni, respectively. The total Cd, Cu and Ni concentrations ranged from 0.65 – 1.82 µg/g, 1.84 – 13.16 µg/g and 7.68 – 17.98 µg/g respectively. The mean concentrations of total Cd was found to be the highest in Cargo Port area and the lowest in Penyembal, whilst the highest mean concentrations for Cu and Ni

were found in Ferry Port and the lowest in Batu Panjang. The EFLE fractions for Cd, Cu and Ni ranged from 0.08 – 0.38 µg/g, 0.02 – 0.22 µg/g and 0.18 – 0.94 µg/g respectively. The ‘acid-reducible’ fractions ranged from 0.03 – 0.17 µg/g, 0.01 – 0.10 µg/g and from 0.15 – 1.25 µg/g for Cu respectively. The ‘oxidisable organic’ fractions for Cd ranged from 0.06 – 0.40 µg/g and 0.42 – 2.57 µg/g and 2.04 – 8.21 µg/g for Cu and Ni respectively. The resistant fraction for Cd ranged from 0.22 – 0.90 µg/g, 0.83 – 11.37 µg/g for Cu and for Ni ranged from 5.20 – 11.42 µg/g.

Table 2. Mean concentration of Cu in each geochemical fractions of Dumai coastal sediment (Concentration in µg/g d.w ± SD (n = 3))

| No. | Station | Lat. (N) | Long.(E) | EFLE | Acid reducible | Oxidisable organic | Resistant | Total (100%) |
|-----|------------------|-----------|------------|------------------|------------------|--------------------|--------------------|--------------|
| 1 | Pelintung | 01°40'23" | 101°41'49" | 0.22±0.13 (1.81) | 0.09±0.00 (0.75) | 0.49±0.08 (4.03) | 11.37±3.72 (93.41) | 12.17 |
| 2 | Guntung | 01°38'35" | 101°34'55" | 0.15±0.02 (1.63) | 0.05±0.00 (0.55) | 0.42±0.02 (4.60) | 8.48±2.26 (93.23) | 9.10 |
| 3 | Mundam | 01°40'12" | 101°30'03" | 0.03±0.01 (0.37) | 0.05±0.00 (0.66) | 0.82±0.12 (10.09) | 7.23±1.67 (88.88) | 8.13 |
| 4 | Pertamina | 01°41'10" | 101°28'52" | 0.05±0.02 (0.71) | 0.02±0.00 (0.35) | 1.04±0.09 (15.32) | 5.69±1.33 (83.62) | 6.80 |
| 5 | Ferry port | 01°41'15" | 101°27'55" | 0.18±0.04 (1.37) | 0.06±0.02 (0.49) | 2.17±0.04 (16.48) | 10.75±4.28 (81.66) | 13.16 |
| 6 | Cargo port | 01°41'19" | 101°27'03" | 0.14±0.02 (1.19) | 0.10±0.02 (0.88) | 2.44±0.86 (21.35) | 8.75±4.71 (76.58) | 11.42 |
| 7 | Sg. Dumai | 01°41'21" | 101°26'14" | 0.11±0.02 (1.26) | 0.09±0.01 (1.02) | 2.44±0.99 (27.97) | 6.09±1.43 (69.75) | 8.74 |
| 8 | Dockyard | 01°41'36" | 101°25'38" | 0.13±0.02 (1.66) | 0.04±0.00 (0.47) | 1.42±0.11 (18.22) | 6.22±1.73 (79.65) | 7.81 |
| 9 | Fishing port | 01°42'03" | 101°24'43" | 0.07±0.03 (0.88) | 0.03±0.00 (0.34) | 1.30±0.30 (17.31) | 6.12±0.88 (81.47) | 7.51 |
| 10 | Purnama | 01°42'34" | 101°24'14" | 0.08±0.01 (1.34) | 0.06±0.00 (0.98) | 1.17±0.13 (20.12) | 4.50±0.90 (77.56) | 5.81 |
| 11 | Sg. Mesjid | 01°43'41" | 101°23'42" | 0.03±0.00 (0.59) | 0.07±0.01 (1.27) | 1.21±0.29 (22.18) | 4.13±0.67 (75.96) | 5.44 |
| 12 | Bangsai Aceh | 01°44'40" | 100°22'54" | 0.05±0.02 (1.10) | 0.07±0.01 (1.72) | 1.02±0.06 (23.37) | 3.22±0.98 (73.82) | 4.36 |
| 13 | Lubuk Gaung | 01°45'36" | 101°22'16" | 0.04±0.00 (0.91) | 0.01±0.00 (0.15) | 1.13±0.01 (27.09) | 3.01±0.66 (71.86) | 4.19 |
| 14 | Penyembal | 01°47'27" | 101°21'43" | 0.02±0.01 (0.31) | 0.07±0.00 (1.37) | 2.57±0.04 (49.10) | 2.58±0.81 (49.23) | 5.23 |
| 15 | Basilam Baru | 01°51'26" | 101°21'10" | 0.02±0.01 (0.48) | 0.01±0.01 (0.28) | 1.46±0.18 (38.83) | 2.27±0.25 (60.42) | 3.76 |
| 16 | Penempul | 01°59'51" | 101°19'55" | 0.04±0.01 (0.91) | 0.03±0.00 (0.69) | 1.39±0.06 (28.63) | 3.38±0.44 (69.77) | 4.85 |
| 17 | Terkul | 0°14'04" | 101°34'43" | 0.04±0.01 (0.55) | 0.09±0.00 (1.28) | 1.41±0.04 (19.57) | 5.68±1.15 (78.60) | 7.23 |
| 18 | Batu Panjang | 01°42'19" | 101°30'41" | 0.03±0.00 (1.74) | 0.04±0.00 (2.18) | 0.93±0.39 (50.71) | 0.83±0.03 (45.37) | 1.84 |
| 19 | Tg. Kapal | 0°143'12" | 101°27'46" | 0.03±0.00 (0.54) | 0.04±0.00 (0.69) | 1.26±0.04 (24.11) | 3.89±0.95 (74.67) | 5.21 |
| 20 | Mombol | 02°04'12" | 101°28'32" | 0.12±0.01 (1.75) | 0.03±0.00 (0.46) | 1.67±0.06 (23.59) | 5.25±1.15 (74.20) | 7.07 |
| 21 | Pulau Babi | 02°05'51" | 101°33'26" | 0.07±0.02 (1.28) | 0.01±0.00 (0.12) | 1.74±0.02 (33.81) | 3.34±0.59 (64.79) | 5.16 |
| 22 | Tg. Medang luar | 02°07'31" | 101°39'10" | 0.07±0.00 (0.99) | 0.05±0.01 (0.72) | 1.55±0.48 (22.56) | 5.22±1.69 (75.74) | 6.89 |
| 23 | Tg. Medang dalam | 02°06'37" | 101°37'40" | 0.06±0.04 (1.28) | 0.06±0.02 (1.29) | 1.64±0.49 (36.16) | 2.78±1.00 (61.27) | 4.54 |

Note : The values in parentheses represent the fraction in percentage (%)

Table 3. Mean concentration of Ni in each geochemical fractions of Dumai coastal sediment (Concentration in $\mu\text{g/g d.w} \pm \text{SD}$ ($n = 3$))

| No. | Station | Lat. (N) | Long.(E) | EFLE | Acid reducible | Oxidisable organic | Resistant | Total (100%) |
|-----|------------------|-----------|------------|------------------|------------------|--------------------|---------------------|--------------|
| 1 | Pelintung | 01°40'23" | 101°41'49" | 0.72±0.05 (3.98) | 0.26±0.07 (1.47) | 8.21±0.43 (45.65) | 8.79±2.23 (48.91) | 17.98 |
| 2 | Guntung | 01°38'35" | 101°34'55" | 0.79±0.13 (5.70) | 0.24±0.04 (1.71) | 6.37±0.14 (46.28) | 6.38±6.10 (46.31) | 13.77 |
| 3 | Mundam | 01°40'12" | 101°30'03" | 0.52±0.11 (3.72) | 0.29±0.06 (2.07) | 5.43±0.10 (39.20) | 7.62±1.77 (55.00) | 13.85 |
| 4 | Pertamina | 01°41'10" | 101°28'52" | 0.29±0.03 (2.79) | 0.36±0.16 (3.52) | 3.20±0.09 (30.98) | 6.47±1.68 (62.71) | 10.31 |
| 5 | Ferry port | 01°41'15" | 101°27'55" | 0.65±0.08 (3.71) | 0.40±0.01 (2.31) | 4.99±1.47 (28.58) | 11.42± 2.96 (65.40) | 17.46 |
| 6 | Cargo port | 01°41'19" | 101°27'03" | 0.31±0.06 (2.47) | 0.39±0.05 (3.08) | 4.23±1.22 (33.35) | 7.75±2.24 (61.10) | 12.69 |
| 7 | Sg. Dumai | 01°41'21" | 101°26'14" | 0.18±0.03 (1.58) | 0.24±0.03 (2.10) | 3.34±0.34 (28.65) | 7.88±2.22 (67.67) | 11.65 |
| 8 | Dockyard | 01°41'36" | 101°25'38" | 0.26±0.11 (1.87) | 0.26±0.11 (1.81) | 3.62±0.26 (25.65) | 9.98±2.30 (70.66) | 14.12 |
| 9 | Fishing port | 01°42'03" | 101°24'43" | 0.27±0.10 (1.86) | 0.41±0.01 (2.83) | 3.98±0.05 (27.28) | 9.94±1.62 (68.03) | 14.61 |
| 10 | Purnama | 01°42'34" | 101°24'14" | 0.21±0.08 (2.01) | 0.24±0.13 (2.27) | 2.83±0.17 (27.09) | 7.18±1.46 (68.63) | 10.46 |
| 11 | Sg. Mesjid | 01°43'41" | 101°23'42" | 0.20±0.06 (2.06) | 0.16±0.10 (1.71) | 2.80±0.18 (29.20) | 6.42±1.56 (67.02) | 9.58 |
| 12 | Bangsai Aceh | 01°44'40" | 100°22'54" | 0.22±0.08 (2.49) | 0.15±0.07 (1.62) | 2.14±0.15 (23.84) | 6.48±2.09 (72.06) | 9.00 |
| 13 | Lubuk Gaung | 01°45'36" | 101°22'16" | 0.20±0.13 (2.08) | 0.19±0.02 (2.07) | 2.09±0.03 (22.27) | 6.91±1.82 (73.57) | 9.40 |
| 14 | Penyembal | 01°47'27" | 101°21'43" | 0.75±0.09 (5.36) | 1.25±0.09 (8.96) | 4.62±0.19 (33.15) | 7.32±2.88 (52.52) | 13.93 |
| 15 | Basilam Baru | 01°51'26" | 101°21'10" | 0.25±0.15 (2.76) | 0.43±0.09 (4.78) | 2.24±0.21 (24.97) | 6.06±3.51 (67.49) | 8.98 |
| 16 | Penempul | 01°59'51" | 101°19'55" | 0.34±0.17 (3.01) | 0.35±0.04 (3.07) | 2.85±0.08 (25.16) | 7.80±1.12 (68.76) | 11.35 |
| 17 | Terkul | 0°14'04" | 101°34'43" | 0.42±0.02 (4.15) | 0.43±0.09 (4.28) | 3.46±0.02 (34.56) | 5.71±2.98 (57.01) | 10.02 |
| 18 | Batu Panjang | 01°42'19" | 101°30'41" | 0.28±0.01 (3.59) | 0.17±0.03 (2.17) | 2.04±0.02 (26.59) | 5.20±0.15 (67.66) | 7.68 |
| 19 | Tg. Kapal | 0°14'31" | 101°27'46" | 0.25±0.02 (2.29) | 0.37±0.02 (3.37) | 2.66±0.01 (24.16) | 7.73±1.56 (70.19) | 11.02 |
| 20 | Mombol | 02°04'12" | 101°28'32" | 0.94±0.01 (6.30) | 0.42±0.05 (2.81) | 3.22±0.05 (21.69) | 10.28±1.91 (69.20) | 14.85 |
| 21 | Pulau Babi | 02°05'51" | 101°33'26" | 0.63±0.11 (5.21) | 0.34±0.06 (2.78) | 2.81±0.10 (23.26) | 8.30±1.54 (68.74) | 12.08 |
| 22 | Tg. Medang luar | 02°07'31" | 101°39'10" | 0.57±0.13 (4.89) | 0.46±0.11 (3.91) | 4.44±1.70 (38.15) | 6.18±1.55 (53.05) | 11.65 |
| 23 | Tg. Medang dalam | 02°06'37" | 101°37'40" | 0.22±0.01 (2.68) | 0.31±0.10 (3.71) | 2.40±0.68 (28.64) | 5.44±1.75 (64.97) | 8.37 |

Note : The values in parentheses represent the fraction in percentage (%)

The comparison between resistant and nonresistant fractions of metals in the coastal sediments of Dumai is shown in **Figure 2a, 2b and 2c** for Cd, Cu and Ni, respectively. Resistant fractions ranged from 34.18% in Penyembal to 73.55% in Lubuk Gaung for Cd; from 45.37% in Pelintung to 93.41% in Batu Panjang for Cu and from 46.31% in Guntung to 73.57% in Lubuk Gaung for Ni. Metal concentrations in sediments at most sampling stations were dominated by resistant fractions (Cd: 78.26%, Cu: 91.30% and Ni: 91.30%). Only at stations Pelintung, Pertamina, Cargo

port, Penyembal and Batu Panjang showed that non resistant fractions were higher than resistant fractions for Cd. Meanwhile, higher percentages of non resistant fractions when compared to resistant fractions of Cu were found in Penyembal and Batu Panjang, whilst for Ni was found in Pelintung and Guntung. This indicated that anthropogenic inputs of Cd, as represented by nonresistant fraction, occurred in more stations as compared to Cu and Ni.

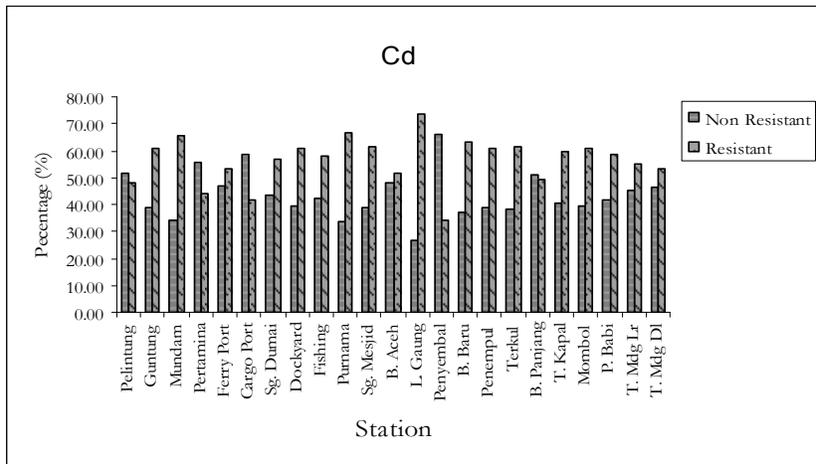


Fig 2a. Resistant and non resistant fractions of Cd

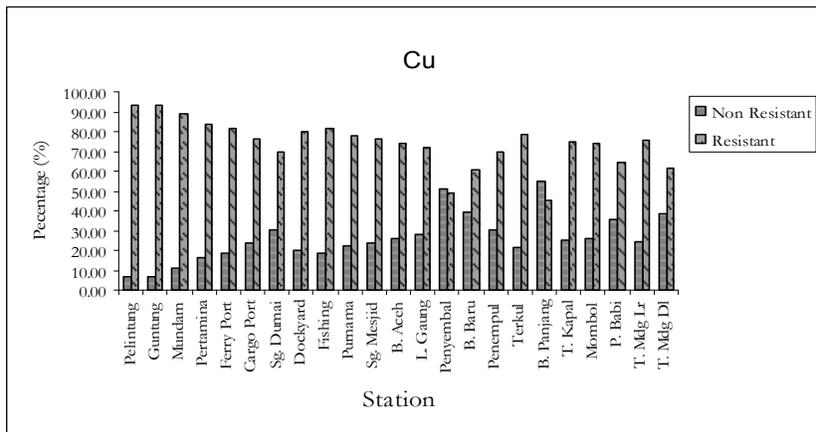


Fig 2b. Resistant and non resistant fractions of Cu

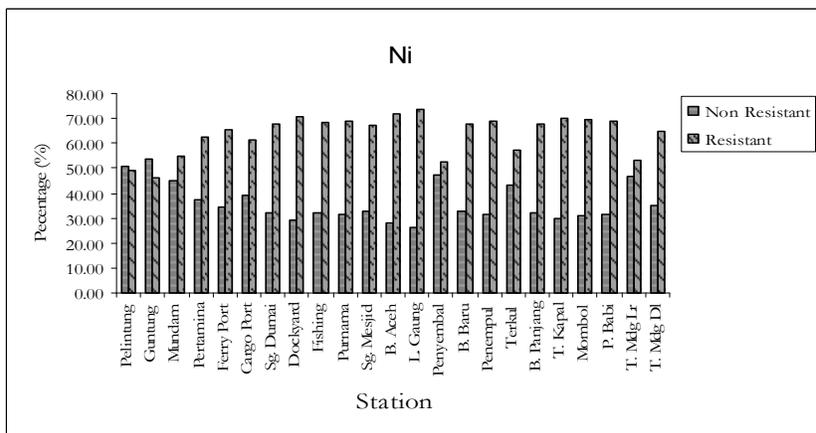


Fig 2c. Resistant and non resistant fractions of Ni

The EFLE fraction contributed rather higher portion for Cd but only a small portion of the total Cu and Ni of sediments from all stations, suggesting poor bioavailabilities of these metals. Highest percentages of the fractions were found in dockyard station (25.54%), Pelintung (1.81%) and Mombol (6.30%) and the lowest were detected in Mundam (8.71%), Penyembal (0.31%) and Sungai Dumai (1.58%) for Cd, Cu and Ni respectively. Yap *et al.* (2005) found that less than 10% of Cu in sediments from the west coast of Peninsular Malaysia were in the EFLE fraction. Although the percentage of this fraction was low when compared to the other fractions, this is very important in ecotoxicological point of view because this is the fraction which is bioavailable to sediment-ingesting animals and therefore can

pose hazardous effects to the environment (Morillo *et al.*, 2004; Yap *et al.*, 2002; 2005; Ramirez *et al.*, 2005).

Figure 3a, 3b and 3c compare the non-resistant and resistant fractions in the sediments of all five sampling site groups. Resistant fractions covered 55.28 – 58.31% of the total Cd at all site groups; Cu covered 65.02% - 91.84% and Ni covered 50.08% – 66.88% of the total Pb at most site groups. None of the site group showed higher percentage of nonresistant fraction which indicated that there was no significant anthropogenic input of Cd, Cu or Ni to the Dumai coastal waters. Contribution of each fraction considered in this study for each site group is shown in **Figure 4a, 4b and 4c**. Overall distribution of geochemical fractions of Cd, Cu and Ni in the coastal sediments of Dumai is shown in **Figure 5**.

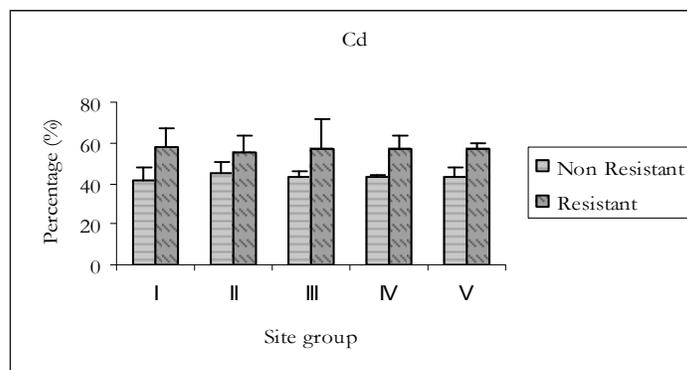


Fig 3a. Resistant and non resistant fractions of Cd

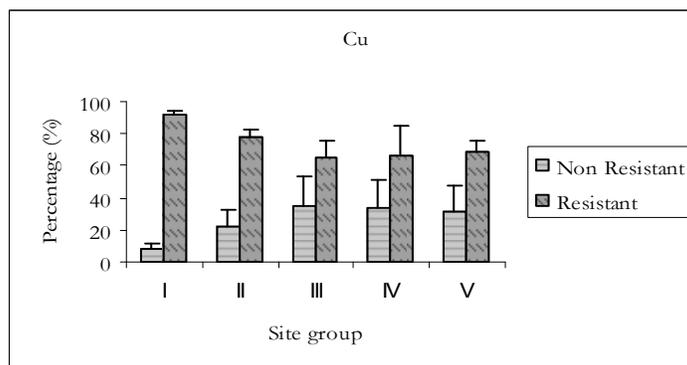


Fig 3b. Resistant and non resistant fractions of Cu

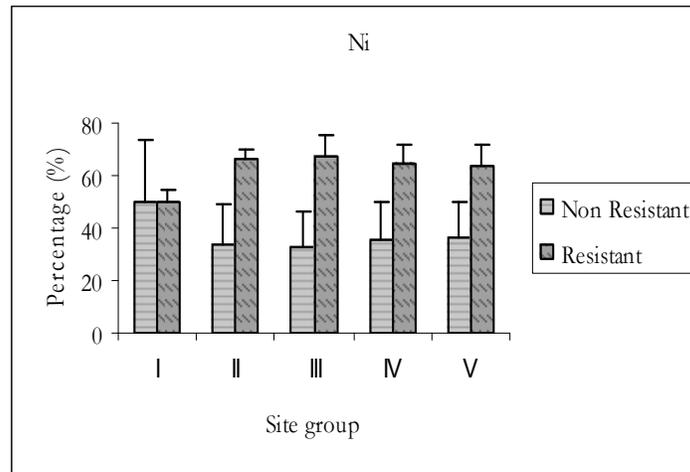


Fig 3c. Resistant and non resistant fractions of Ni

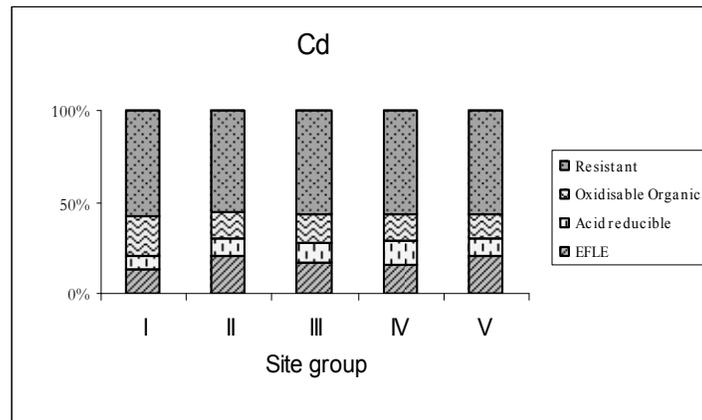


Fig 4a. Distribution of geochemical fractions of Cd at each site group

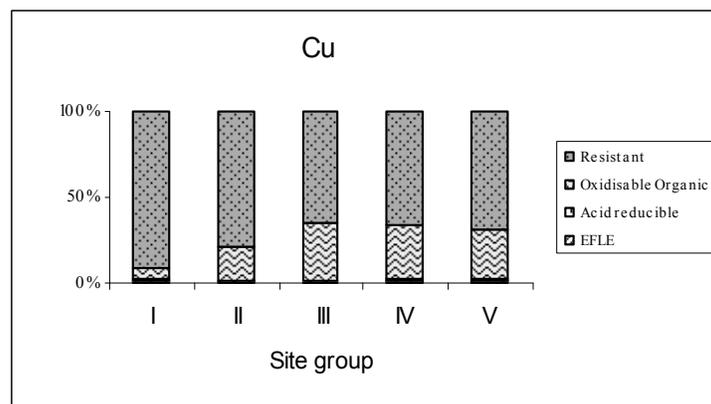


Fig 4b. Distribution of geochemical fractions of Cu at each site group

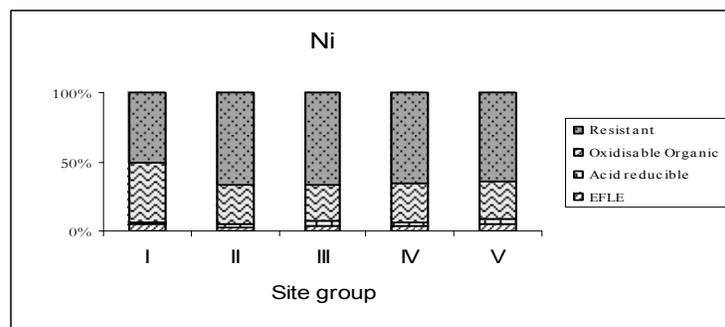


Fig 4c. Distribution of geochemical fractions of Ni at each site group

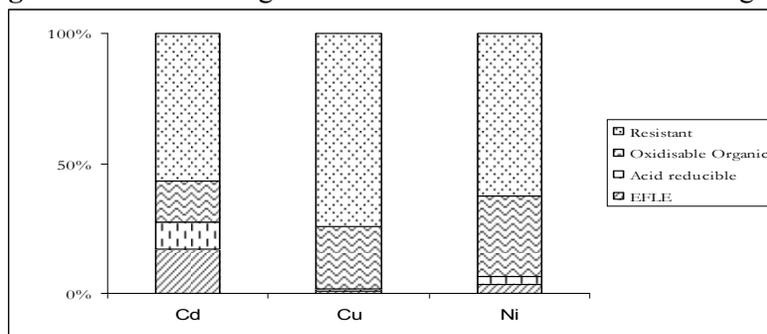


Fig 5. Overall distribution of geochemical fractions of Cd, Cu and Ni in coastal sediment of Dumai

Within the site groups, the percentage of Cd in the EFLE fraction (also known as the labile fraction) in the central part of Dumai (site group II) was found to be the highest (20.35%) suggesting that the central part of Dumai is a potential source of contamination for Cd. Higher percentages of Cu in this fraction were found at the north part of Rupert Island (1.32%) as well as at the east and central part of Dumai (1.27% and 1.12%) compared to other stations (0.74 – 0.94%). As for Cu, EFLE fractions for Ni were also found to be higher in the north part of Rupert Island and in the east part of Dumai (4.77% and 4.47%, respectively). This indicated that the source of these metals originated from the central and eastern part of Dumai coast as well as from the Straits of Malacca in the North part of Rupert Island.

The acid reducible fraction found in this study ranged from 3.30 - 20.68 and 0.12 – 2.18% and 1.47 – 8.96 % for Cd, Cu and Ni, respectively. This finding is still comparable to other studies who found less

than 10% of this fraction (Yap *et al.*, 2005 and Ramirez *et al.*, 2005).

Highest percentages of oxidisable organic (bound to organic) Cd (20.72 %) and Ni (43.71 %) in site group I was probably due to organic matter inputs from the surrounding area. Other input may come from Pelintung River and also from the central part of Dumai (where human activities are concentrated) which were carried out by the currents during high tide and deposited in the eastern part of Dumai. However Cu was found to be highest in site group III (33.40%) where two rivers (Sungai Mesjid and Sungai Buluhala) drain their water to the coastal area. Morillo *et al.* (2004) also found a higher percentage of oxidisable fractions of the metal studied in stations close to the river mouth which might be related to the organic matter. Higher percentages of metals in this fraction, which related to the organic inputs from anthropogenic activities, have also been reported in other studies (Yap *et al.*,

2003a; 2005; Cuong and Obbard, 2006). These metals might be associated with several forms of organic materials and they might occur in the forms of stable complexes and metal sulphides particles (Tokalioglu *et al.*, 2000; Morillo *et al.*, 2004; Cuong and Obbard, 2006).

The resistant fractions for Cd, Cu and Ni in coastal sediments of Dumai covered more than 50% of total amount of those metals and reflected that Dumai coastal sediments were not heavily contaminated. The resistant fractions of these metals in sediments were probably, according to Badri and Aston (1983), due to natural sources such as chemical weathering of igneous and metamorphic rocks and decomposition of biota detritus. Anthropogenic inputs of these metals were considered to be less as their fractions (as represented by non resistant fractions) were still lower than the resistant fractions or natural sources. Metals from anthropogenic inputs according to Badri and Aston (1983) and Yap *et al.* (2002; 2003a) were assumed to be trapped within silicate minerals and incorporated into the crystalline lattice positions of the minerals and therefore the resistant fraction is low in biological availability.

The nonresistant fractions of heavy metals found in the sediments were certainly of much concern. Besides posing an impact to the living organisms, this nonresistant fraction is most likely due to anthropogenic inputs rather than natural origins (Yap *et al.*, 2002). The levels of nonresistant fraction of Cd, Cu and Ni found in the sampling sites could be mostly due to man-induced activities such as shipping (sea based), human settlement, industries and other oil-related activities (land based). In addition, natural processes such as aerial deposition that brings small particles due to industrial activities and forest fires, which are often occurred in the surrounding Dumai area, should also to be taken into account. However, the nonresistant fractions of Cd, Cu and Ni in the sediments of most of stations were considered low. Yap *et al.* (2002; 2003b) reported that nonresistant

fraction contributed about 50% of the total Cu while Cd contributed 24 – 71% nonresistant fraction of the total concentration indicating anthropogenic inputs of this metal into the Straits of Malacca. Higher percentages of nonresistant than resistant fraction of Cd in sediments were also reported in other studies. Nonresistant Cd contributed 50 – 70% of the total in sediment from Spain (Morillo *et al.*, 2004); 57% in sediment from Suez canal (Abd. El-Azim and El-Moselhy, 2005); 50 – 70% in sediment from Singapore (Cuong and Obbard, 2006) and 82% in sediment from Yangtze estuary China (Fang and Wang (2006). However, nonresistant fraction for Cu and Ni contributed about 16 - <50% of the total Cu and Ni concentration and suggested that these mostly related to natural origin (Yap *et al.*, 2003b; Morillo *et al.*, 2004; Abd. El-Azim and El-Moselhy, 2005; Cuong and Obbard, 2006 and Fang and Wang (2006).

Among the three metals studied, the nonresistant fraction of Cd in sediment was greater than Cu and Ni and the mobility of these metals, as represented by EFLE fraction, decreased in the order of Cd > Ni > Cu. This also indicated that Cd is more available for exchange or released into the marine environment. The present finding was in agreement with the previous study in Singapore (Cuong and Obbard, 2006).

Concentrations of total metal analyzed at each site group are shown in (**Table 4**). The highest concentrations of all metals were found in samples from the East Dumai (site group I) with 1.43 µg/g, 9.80 µg/g and 15.20 µg/g for Cd, Cu and Ni respectively. Stations at west part of Dumai (site group III) being the lowest for Cd and Cu (0.76 µg/g and 4.48 µg/g) and south part of Rupert island (site group IV) was the lowest for Ni (9.57 µg/g). High concentrations of metals at the east and central part of Dumai was probably due to the current direction in the Rupert Straits which bring the water mass from the northwest to the south east in the Straits of Malacca during high tide. In Rupert Straits,

the water flows from the north to the south direction, then turns eastward and meets the Malacca Straits waters again. During low tide the current will move back in the opposite direction following the same path as in high tide (Anonymous, 2002). The waters in the centre of Dumai coastal area also receive effluents from the anthropogenic activities which were carried by Sungai

Dumai, Sungai Mesjid and some sewer outlets from the city center and oil refineries. Higher concentration of metals in Pelintung might also be attributed to the recent development of 3500 hectares Pelintung Industrial Park and the construction of its 1000 m international harbour.

Table 4. Mean metal concentrations in coastal sediments of Dumai according to site groups

| Site groups | Metal concentration ($\mu\text{g/g}$) | | |
|--------------------|---|------|-------|
| | Cd | Cu | Ni |
| I (East Dumai) | 1.43 | 9.80 | 15.20 |
| II (Central Dumai) | 1.10 | 8.34 | 12.61 |
| III (West Dumai) | 0.76 | 4.48 | 10.53 |
| IV (South Rupal) | 0.81 | 4.76 | 9.57 |
| V (North Rupal) | 0.97 | 5.91 | 11.74 |

The total metal concentrations in the present study were comparable to other studies reported in literature (**Table 5**). Cd concentrations were still within the range found in other studies. The present result were higher than Java Sea (Everaarts, 1989), offshore west coast of Peninsular Malaysia (Yap *et al.*, 2003), Chile sandy beaches (Ramirez *et al.*, 2005), Karachi coast (Qori *et al.*, 2005), Hugli river estuary India (Sarkar *et al.*, 2004), Kranji and P. Tekong Singapore (Cuong and Obbard, 2006). However it still much lower than Arabian Gulf (Shriadah, 1999), Coruga and Gaderu India (Ray *et al.*, 2006), Coastal sediment India (Reddy *et al.*, 2004), Tees estuary (Jones and Turki, 1997). Concentrations of Cu were comparable or even much lower that concentrations found in other geographical areas (**Table 4**), but higher than samples from Karachi coast (Qori *et al.*, 2005), Tg. Piai, Malaysia (Yap *et al.*, 2006). Ni concentrations in the present study were also comparable and lower than reported values in Table 4, but still higher than sample from Chile sandy beaches (Ramirez *et al.*, 2005) and Karachi coast (Qori *et al.*, 2005), Tg. Piai, Malaysia (Yap *et al.*, 2006), Ponggol estuary, Singapore

(Nayar *et al.*, 2004) and Sg. Buloh, Singapore (Cuong *et al.*, 2005).

In order to estimate possible environmental consequences of the analyzed metals at the studied sites, concentrations of total Cd, Cu and Ni were compared to the Sediment Quality Guidelines of Effect Range Low (ERL) and Effect Range Median (ERM) as proposed by Long *et al.* (1995; 1997). The present results showed that Cd concentrations in most of the stations were still below the ERL value (1.2 $\mu\text{g/g}$) and ERM (9.6 $\mu\text{g/g}$). Only at Station Pelintung, Guntung, Mundam, Ferry Port and Cargo Port exceed the ERL but still well below ERM value. However, concentrations of Cd in all stations were above the background concentration of non-contaminated sediment (0.17 $\mu\text{g/g}$) as suggested by Salomon and Forstner (1984). Concentrations of Cu and Ni in sediments at all stations were still below the ERL values (34.0 $\mu\text{g/g}$ and 20.9 $\mu\text{g/g}$) and the ERM values (270.0 $\mu\text{g/g}$ and 51.6 $\mu\text{g/g}$) and even still below the background concentration of non-contaminated sediment (33 $\mu\text{g/g}$ for Cu and 52 $\mu\text{g/g}$ for Ni) as suggested by Salomon and Forstner (1984). Although

most of the metal concentrations were still below the ERM values, follow up monitoring should be continued, especially at the eastern

and the center part of Dumai waters where most of anthropogenic activities are concentrated.

Table 5. Comparison of total metal (Cd, Cu and Ni) concentrations ($\mu\text{g/g}$) in coastal sediments of Dumai with other locations

| Location | Cd | Cu | Ni | Reference |
|---------------------------------|------------|-----------------|--------------|---------------------------------------|
| Tg. Piai, Malaysia | 0.72-1.19 | 3.43-3.81 | 10.10-11.00 | Yap <i>et al.</i> , 2006 |
| Kranji and P. Tekong, Singapore | 0.06-0.19 | 7.70-17.90 | 17.10-26.10 | Cuong and Obbard, 2006 |
| Espinho/Mondego Cape, Portugal | - | 117.00-848.00 | 17.00-89.00 | Vidinha <i>et al.</i> , 2006 |
| Guanabara Bay, Brazil | - | 2.00-18840.00 | 1.00-3515.50 | Baptista-Nito <i>et al.</i> , 2006 |
| Bristol Channel, UK | 0.05-0.92 | 23.90-46.30 | 18.80-26.60 | Duquesne <i>et al.</i> , 2006 |
| Mandovy estuary, India | - | 11.50-77.50 | - | Alagarsamy, 2006 |
| Estuarine lagoon, Puerto Rico | 0.10-1.80 | 22.00-105.00 | - | Acevedo-Figueroa <i>et al.</i> , 2006 |
| Suez Canal, Egypt | 1.45-3.06 | - | 14.22-54.18 | Abd. El-Azim and El-Moselhy, 2005 |
| Taranto Gulf, Italy | - | 42.40-52.30 | 47.90-60.70 | Buccolieri <i>et al.</i> , 2006 |
| Coruga and Gaderu, India | 6.00-17.00 | 34.00-58.00 | 7.50-52.00 | Ray <i>et al.</i> , 2006 |
| Mangrove area, Singapore | 0.18-0.27 | 7.06-32.00 | 7.44-11.65 | Cuong <i>et al.</i> , 2005 |
| Karachi coast, Pakistan | 0.01-0.14 | 0.01-0.10 | 0.03-0.04 | Qori <i>et al.</i> , 2005 |
| Danube, Serbia | - | 27.00 | 32.30 | Relic <i>et al.</i> , 2005 |
| Sandy beaches, Chile | 0.80-1.09 | 1259.00-1896.00 | 5.67-13.60 | Ramirez <i>et al.</i> , 2005 |
| Ponggol estuary, Singapore | 0.24 | 34.65 | 6.07 | Nayar <i>et al.</i> , 2004 |
| Hugli river estuary, India | 0.10-0.20 | 21.50-64.10 | 26.50-44.50 | Sarkar <i>et al.</i> , 2004 |
| Coastal sediment, India | 8.57-45.85 | 85.17-312.87 | 60.26-221.50 | Reddy <i>et al.</i> , 2004 |
| Semarang coast, Indonesia | - | 33.00-72.00 | 1.00-29.00 | Takarina <i>et al.</i> , 2004 |
| South west coast, Spain | 0.19-2.50 | 41.00-336.00 | 10.00-61.00 | Morillo <i>et al.</i> , 2004 |
| Gulf of Suez, Egypt | 2.26-4.40 | 1.84-10.25 | - | El-Moselhy & Gabal, 2004 |
| Offshore west coast, Malaysia | 0.10-1.42 | 0.25-13.80 | - | Yap <i>et al.</i> , 2002; 2003a |
| Intertidal west coast, Malaysia | 0.03-1.98 | 0.40-315.00 | - | Yap <i>et al.</i> , 2002; 2003b |
| Sonora Coast, Mexico | 0.50-2.51 | 1.50-27.00 | - | Garcia-Rico <i>et al.</i> , 2003 |
| Lami coast, Fiji | - | 5.00-1980.00 | - | Gangaiya <i>et al.</i> , 2001 |
| Jakarta Bay, Indonesia | - | 3.00-128.00 | 17.80-36.10 | Williams <i>et al.</i> , 2000 |
| Seberang Prai, Malaysia | 0.27-4.68 | 9.99-63.44 | - | Ismail and Asmah, 1999 |
| Patagonian Coast, Argentina | Nd-7.50 | 2.40-20.10 | - | Gill <i>et al.</i> , 1999 |
| Tees Estuary, UK | 0.30-9.80 | 18.00-262.00 | 21.00-80.00 | Jones and Turki, 1997 |
| Victoria Harbour, Hong Kong | 2.61-3.33 | 45.20-3790.00 | - | Wong <i>et al.</i> , 1995 |
| Bintulu coast, Malaysia | 1.00-5.00 | 7.00-13.00 | - | Ismail, 1993 |
| Java Sea, Indonesia | 0.03-0.61 | 6.00-54.00 | - | Everaarts, 1989 |
| Dumai coast, Indonesia | 0.65-1.82 | 1.84-13.16 | 7.68-17.98 | Present study |

CONCLUSION

This study establishes a baseline for heavy metal concentration and speciation in sediment along the coastlines of Dumai. Higher concentrations of metals were found mostly in the eastern part of Dumai coastal sediments. Local hydrodynamic and coastal topography, especially current direction in Rupat Strait may play an important role in transporting contaminated water from the central part of Dumai and Malacca Straits and deposited in the eastern part of Dumai coastal waters.

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