Statistical Perspective and Pollution Indicator

in Mengkabong Mangrove Sediment Sabah

Miroslav Radojevic

Faculty of Engineering and Computer Science, The University of Nottingham Malaysia Campus

Jalan Broga, 43500 Semenyih, Selangor Darul Ehsan, Malaysia

Tel: 60-3-8924-8165   E-mail: Miroslav.Radojevic@nottingham.edu.my

Sarva Mangala Praveena (Corresponding Author)

School of Science and Technology, Universiti Malaysia Sabah

Locked Bag No. 2073, 88999 Kota Kinabalu, Sabah, Malaysia

Tel: 60-88-325-772   E-mail: smpraveena@gmail.com

Mohd Harun Abdullah

School of Science and Technology, Universiti Malaysia Sabah

Locked Bag No. 2073, 88999 Kota Kinabalu, Sabah, Malaysia

Tel: 60-88-325-875   E-mail: harunabd@ums.edu.my

Abstract

The role of tidal processes on intertidal surface sediments is frequently stated but rarely investigated due to the methodological restrictions.   This study provides a basis for describing the tidal effects on the mangrove sediment. It reports investigations into the chemical composition of mangrove surface sediment in Mengkabong lagoon, Sabah, Malaysia.   The levels of 16 parameters were determined in 33 surface sediment samples at high and low tides. The studied parameters showed high values at high tide compared to low tide.   The tidal process showed there is a high contribution of seawater that brings additional ions such as Na, Mg and K into the mangrove system at high tide whereas at low tide, lower contribution of seawater was observed.   Spearman correlation coefficient was used for the statistical characterizations of mangrove surface sediment at high and low tides.   Relationships between physicochemical parameters, metals, granulometric fractions, organic matter and base cations at high and low tides were observed.   Geoaccumulation Index calculations showed current sediment quality of Mengkabong mangrove sediments could be classified as having background concentrations of Al, Cu, Fe, and Zn, and as unpolluted with regard to Pb. These findings are important for future research which will explain the chemistry that lies in mangrove sediment.

Keywords: Mangrove, Mengkabong lagoon, tides, Spearman correlation coefficient, Geoaccumulation Index

1. Introduction

Mangrove sediments have been extensively studied around the world (India, Australia, Brazil, Malaysia, Arab, China, Thailand, etc.). Heavy metal cycling is a serious problem addressed in mangrove environments elsewhere (Marchand *et al*., 2006; Pekey, 2006).   High concentrations of heavy metals are derived from anthropogenic inputs such as industrial activities around estuaries as well as discarded automobiles, batteries, tires, waste water disposal, etc. (Shriadah, 1999; Bloom and Ayling, 1977).

Tidal current activity is mainly confined to mangrove channels. Outside the channels, mainly on the upper tidal-flats, tidal current velocities decrease and sediment entrainment is frequently ascribed to wave action. The role of tidal processes on intertidal surface sediments is frequently stated but the differences at these stages have seldom been investigated, apparently because of methodological constraints (Malvarez *et al*., 2001). The methodological constraints were such as determination of tidally influenced areas, correction of hydraulic gradient error based on tidal fluctuation, data requirements to determine tidal range at one location at coastal sites, definition of problems and development of conceptual model (Zektser and Dzhamalov, 2006).   Moreover, one of the most difficult tasks in coastal wetlands

studies is to use proper spatial scales from regional to habitat levels when selecting site criteria that will promote some specific goal or endpoint (Twilley *et al*., 1999).

In attempting to determine the extent of pollution in mangrove sediment and better estimate anthropogenic inputs using the heavy metals load in sediment, the Geoaccumulation Index (I_{geo}) was employed. I_{geo} was introduced by Muller (1979) to assess metal pollution in sediments. I_{geo} includes seven grades describing degrees of enrichment above the background value ranging from unpolluted to very polluted sediment quality. The highest grade (class six) reflects 100-fold enrichment above the background values (Singh *et al*., 2003).

The Mengkabong lagoon in Tuaran District, where the current research was carried out, has experienced a 15% decrease in mangrove cover from 1991 to 2000. In 1991 the mangroves covered 12.6 km^2 while in 2000 they covered 10.7 km^2. Most of the mangroves have been lost due to the spread of development such as housing, storm water road run-off, agricultural activities, aquaculture projects and the surrounding industrial zone, Kota Kinabalu Industrial Park (KKIP) (Environmental Indicator Report, 2003). One of the problems with further development in the area is very little inflow of water from inland areas. This means very limited flushing of sediment and effluent in the lagoon. The sediment is effectively filling up the bay and it is becoming shallower. The whole lagoon is a sensitive ecosystem and further development should be severely limited when detailing plans for that part of KKIP within Tuaran District (Town and Regional Planning Department, 2003). Due to the proximity at KKIP and the increasing development in the area surrounding the mangrove forest, it was decided to assess the distribution of heavy metals at high and low tides, enrichment and current pollution status of the mangrove sediments.

The objectives of the present study are: (1) to determine the physicochemical characteristics (pH, salinity and electrical conductivity), granulometric fractions (clay, silt and clay), heavy metals (Fe, Cu, Zn, Pb and Al) and base cations (Na, K, Mg and Ca) of Mengkabong Bay surface sediment at high and low tides; (2) to determine the statistical characteristics of Mengkabong lagoon surface sediment using the Spearman correlation coefficient; and (3) to assess the extent of metal pollution using Geoaccumulation Index (I_{geo}).

2. Materials and Methods

2.1 Sediment Sampling and Analysis

The study was conducted in Mengkabong mangrove forest on the west coast of Sabah which is 40 km from Kota Kinabalu. The study area spreads from latitude 06^{0}06'N to 06^{0}11'N and longitude 116^{0}08'E to 116^0 13'E. Mangrove sediments were sampled randomly, based to the accessibility of the mangrove forest, and taken in triplicates with an auger at 33 stations from March 2006 to November 2006 (Figure 1) at low and high tide. The exact position of each sampling site was recorded using Global Positioning System (GPS). Mangrove surface sediments were chosen for this study as this layer controls the exchange of metals between sediments and water (El Nemr *et al*., 2006).

Figure 1

The sediments were kept cool in an icebox during transportation to the laboratory. The physicochemical parameters (pH, electrical conductivity and salinity) were measured on 1:2 soil to water ratio extracts as soon as the samples reached the laboratory. The pH, electrical conductivity and salinity electrodes were calibrated before the measurements were taken. For other analyses, the surface sediments were air-dried, homogenized using pestle and mortar, passed through a 2-mm mesh screen and stored in polyethylene bags. Methods and references for various analyses are given in Table 1. For the determination of heavy metals, the samples were digested using aqua regia. Approximately 2g of each sample was digested with 15 mL of aqua-regia (1: 3 HCl: HNO$_3$) in a Teflon bomb for 2h at 120 ^{o}C. After cooling, the digested samples were filtered and kept in plastic bottles before the analysis. Radojevic and Bashkin (1999) stated that aqua regia is adequate for extraction of total metals in soil sample and is widely used in most soil analyses laboratories. Base cations (Na, K, Ca and Mg) were determined by the measurement of exchangeable cations using ammonium acetate. Heavy metals and base cations were analyzed using AAS with air/acetylene (Cu, Fe, Pb, Zn, Na, K, Ca and Mg) and nitrous oxide-acetylene (Al) at specific wavelengths (Atomic Absorption Spectrometer Perkin Elmer 4100).

Table 1

3. Results and Discussion

3.1 Descriptive Statistics

The descriptive statistics of physico-chemical properties (pH, salinity, electrical conductivity) granulometric fraction, organic matter, heavy metals (Al, Cu, Fe, Pb and Zn) and base cations (Ca, Mg, Na and K) at low and high tide are given in Table 2.

Table 2

Berner (1981) recorded hundreds of pH measurements of estuarine sediment in Galician Rias Baixas, NorthWest Spain, within the range from pH 6 to pH 8. Similarly, 90% of pH values in the present study fall between pH 6 and pH 8 at

low and high tide thus reflecting the strongly buffered nature of pH values in sediment. According to Mitsch and Gosselink (2000) mangrove sediment salinity elsewhere varies with season and type of mangrove. Moreover, salinity is a function of the height and duration of tides. High salinity is found where tidal exchange occurs. With regard to organic matter present in mangrove sediment, a study done by Koop-Jakobsen and Giblin (2002) explained that during low tide, air exposure of the sediment increases the rate of degradation of organic matter. The granulometric fractions of mangrove sediment at high and low tide are quite similar in composition. It is dominated by the high percentage of sand followed by silt and clay. Eisma (1998) explained that in an intertidal estuary, the dominant sediment type is sand. The sediment with high composition of sand and low clay fraction is formed in agitated waters in the subtidal and intertidal zone. Tides are the predominant transport agent that influences the granulometric fractions in the mangrove environment. Coastlines with strong wave activity and tides are characterized by the concentration of sand, silt and clay (Eisma, 1998).

The elevated concentration of metals during high tide compared to low tide (Figure 2) is due to redox conditions in sediment columns (Akpan *et al.*, 2002). According to Marchand *et al.* (2004) estuarine sediments are usually in reduced condition and have pore water with high concentration of metals. Alloway (1995) explained that metals in interstitial water are the mobile fraction. de Lacerda (2004) showed that the mobile fraction of metals tends to migrate in the sediment through interstitial water until it comes in contact with oxygen. Thus precipitation of hydrous metal oxides will occur. The precipitates of these metals are no longer soluble and are therefore incorporated into the sediment. Wolanski (2006) detailed that when the sediment submerged at high tide, Fe oxides are converted into hydrated forms and provide a large surface area for reactions of metals ion, leading the bioavailability of heavy metals.

The tides control the water-flows that carry the sediments in the mangrove forests, wherein the water from the estuary spills over to inundate the forests at high tide and drains back to the estuary at low tide. During low tide, H_2S and nutrients are released into the overlying water (Walpersdorf, 2008). Sulfide formed as the product of bacterial sulphate reduction usually undergoes rapid transformation in coastal sediments. Hydrogen sulfide may readily precipitate with Fe^{2+} to form iron sulfides. The exact oxidation/reduction transformations involving various reduced sulfur pools in sediments are not yet fully understood. Most sedimentary sulphide is usually recovered as acid volatile sulfur (AVS = H_2S + FeS) and chromium reducible sulfur (CRS .= S^o + FeS_2) pools (Kristensen *et al.*, 1991).

Bryd *et al.* (1990) in Guem Estuary, Korea, found that dissolved lead, cadmium, zinc, manganese and copper concentrations are highest at low salinities in the water column. This has been attributed to the flocculation and dissolution of aluminosilicate phases and colloidal iron oxyhydroxides. The increase in the concentrations of these metals at high salinity indicates the possibility that pore waters enriched in remobilized metals are mixed into the water column and sediment. The anaerobic condition allows the sulphate reduction to occur and leads to sulphide precipitation. Important chemical controls on the change of trace metals in the water column are the oxidation and reduction of sulfur and iron in surface sediments. This was shown by Marchand *et al.* (2006) in French Guiana mangrove forest. Marchand *et al.* (2006) stated that in estuarine sediments, copper and manganese have been identified as pyrite co-precipitates because of pyrite dissolution. In contrast, cadmium, chromium, lead, and zinc are believed to be attached by other sulfides or oxides because only significant amount of these metals dissolved. This is because these trace metals are involved in sorption or co-precipitation with amorphous FeS (Calloy *et al.*, 2002). Trace metals may be incorporated into sulfide minerals by adsorption, precipitation or ion exchange. The limited studies of metal adsorption on sulfide minerals suggest that the adsorption is dominated by the surface hydroxyl groups. The surface interactions of metals with sulfide minerals are likely to influence the fate and transport of metals in anoxic environments (Morel and Hering, 1993).

The sediment water interface is the zone where the water layer interacts with the sediment. The pore or interstitial water is the water that occupies the free space in the sediment. Its composition reflects the sediment chemistry (Rand, 1995). Bouillon *et al.* (2007) showed that exchange of pore-water, combined with the physical conditions related to the tidal cycle, were important in explaining the sediment chemistry at high and low tide. Sediment pore-water represents the mediator fluid in the exchange of the components between sediment and water. A steep gradient in ionic strength in sediment pore water destabilizes the colloidal materials causing it to stick together (flocculate) (Andrews, 2004). Tidal flooding can bring additional ions of Na, Mg, Ca and K into the system and enable ion exchange and alteration of the sediment (Preda and Cox, 2000). The oxic condition during high tide (de Lacerda *et al.*, 2004) makes the mobile fraction of metals tend to migrate to the sediment. According to Hussein and Rabenhorst (2001), dominant cation exchange in soils and sediment involves ions of unequal charge (polymeric Al, Al^{3+}, Na^+). In coastal environments, impacted by tidal inundation, the selectivity coefficient is no longer constant and becomes a function of ionic strength of the soil solution and surface cations composition. Increasing soil salinity will increase selectivity of the colloidal complex for Al. Clay and silt minerals, the most abundant inorganic colloids in estuarine waters, have surface negative charge that is partly balanced by adsorbed cations. The cations from seawater and heavy metals are attracted to the negative charge on silt and clay surfaces. The cations form a mobile layer in a solution closest to clay minerals. The adjacent particles will approach each other and aggregate. This simple explanation is

vastly complicated by the presence of hydroxides and oxyhydroxide coating on the particles.

Table 3 shows the comparison with previous studies done elsewhere.   A study conducted by Zhou *et al*. (2004) in the Pearl River Estuary found that the concentration of Cu, Zn and Pb were high.   This indicates that heavy metal pollution is serious due probably to the higher degree of industrialization during the last two decades in the Pearl River delta region.   Ramanathan *et al*. (1999) observed high concentration of Fe in mangrove sediments.   This might be the result of the textural and mineralogical characteristics of the mangrove sediments.   Similar observations were made by Ray *et al*. (2006), Marchand *et al*. (2006) and Singh *et al*. (2003).   In Punta Mala Bay (Panama), Izmit Bay (Turkey) and Mai Po (Hong Kong), mangrove areas received high inputs of heavy metals from untreated domestic sewage, storm water road run-off as well as inputs from shipping and agricultural activities.   (Defew *et al*., 2005; Pekey, 2006; Che, 1999).   Unlike, studies done by Shriadah (1999) in the Arabian Gulf, Emmerson *et al*. (1997) in Blackwater estuary (UK), Spencer and Macleod (2002) in Medway Estuary (UK) and Rate *et al*. (2000) in Swan River Estuary (Australia), showed that the heavy metal in mangrove sediments tend to be low in contamination indicating that the mangrove sediment were in unpolluted condition.

Table 3

3.2 Spearman Correlation Coefficient Results

Tables 4 and 5 show the values of Spearman correlation coefficient for the studied mangrove surface sediment. According to Binkley and Fisher (2000), the cations that balance the charge of the anions interact strongly with the solid phase of the soil, principally with the cation exchange complex.   Sodium is an important parameter in some soils, particularly the near coastline, where inputs of marine salts are important. An exchange complex of a soil dominated by H^+ or Al^{3+} maintains a low soil pH, and progressive replacement of the H^+ by so-called base cations leads to high pH. According to Hsue and Chen (2000), seawater plays an important role in buffering the pH change.   The process of tidal clash occurs in the Mengkabong mangrove forest where the influx of seawater from the high tide results in major inputs of selected cations which are then adsorbed by the sediment (clay, silt and sand).   A significant correlation (0.798; $p < 0.01$) was observed between salinity and electrical conductivity at high tide, demonstrating the contribution of seawater during high tide compared to low tide in Mengkabong lagoon sediment.   According to Church (1989), seawater contains 3.5% of salinity of which 90% is fully ionized ions.

Table 4

Table 5

Tidal range and flooding frequency therefore determine direction and quantity of nutrient and organic matter exchange between mangroves and the ocean (Dittmar and Lara, 2001).   Sparks (1995) explained that soil organic matter complexes occur with clay minerals and stabilize the soil organic matter.   Organic matter acts as a metal carrier and plays an important role in the metal distribution patterns (Lin and Chen 1998; de Groot *et al*. 1982).

Pekey (2006) and Shriadah (1999) stated that most contaminants are transported as fine-grained suspended matter which has large surface area.   Grain size plays a significant role in determining elemental concentrations in sediment (Pekey, 2006).   Several authors have studied the relationship between heavy metals and the granulometric fractions. Some studies have found good correlations between heavy metals and granulometric fractions while in some other cases, the relationship was less clear (Pekey, 2006; Shriadah, 1999).   Heavy metal ions accumulate in estuarine sediments because of the deposition of metal-enriched allochthonous particles (parents materials; found in a place other than where they and their constituents were formed) or the adsorption of dissolved heavy-metal ions from the water column (Rodrigo, 1989).

It is possible that the differential adsorptive potential is capable of explaining the heavy metals variability. In some cases, the different size fractions of the sediment may differ in their adsorptive potentials because of differences in mineralogy. For instance, quartz is an exceedingly poor scavenger of metal ions. However, some clay minerals have a great affinity for cations because of the presence of iron and manganese oxides.   According to Sparks (1995), adsorption potential increased exponentially with a decrease in particle size.   The low adsorption potential in finer sediment (as in this study) is a result of the tendency of these particles to flocculate in saline waters.   According to Rodrigo (1985) finer sediments will form large particle-aggregates with a correspondingly low total surface-area-to-volume ratio.   According to Velde (1995), salt flocculation occurs at salinities of 0.05 to 0.1%.   Salt flocculation plays an important role in fine grained materials.   The origin of organic matter and binding strength of the organic matter control flocculation and formation of flocs.   Milligan and Loring (1997) also mentioned that organic matter is crucial in formation of flocculated suspensions.

A study done by Rodrigo (1985) showed low heavy metals adsorption in the Saltwater Creek estuary sediment compared to the Avon-Heathcote estuary (New Zealand).   This decrease was due to the flocculation and adsorption potential.   However, it does not affect the overall relationship between heavy metals and granulometric fractions as only a small quantity of very fine sediment is present in the Saltwater Creek, New Zealand (only about 2% of surface

sediment consists of very fine silt and clay). The small quantity of silt and clay (4-7%) in the Mengkabong mangrove forest sediment related to low adsorption potential and flocculation in saline waters is consistent with the study done by Rodrigo (1985). This is supported by correlation coefficient (r) values at high tide and low tide.

Inter-element relationship between metals provides interesting information on metal sources (Manta *et al*., 2003). In the present study, heavy metals and base cations generally show both positive and negative values among different pairs of variables. According to Ray *et al*. (2006) and Jonathan *et al*. (2004), the significant correlation of metals with Fe indicates the adsorption of these metals on to the oxyhydroxides with Fe. Ramanathan *et al*. (1999) stated that the high concentrations of Fe in mangrove sediments were the results of textural and mineralogical characteristics. It is important to note that clays and feldspars are essential species present in the sand and silt. Furthermore, the study of Jonathan *et al*. (2004) in the Gulf of Mannar, India explained the adsorption of these base cations and metals on the aluminophyllosilicate minerals. Firstly, because they are normally present at concentrations higher than the trace metals, they may occupy most of the surface binding sites even though they form less stable surface complexes. Secondly, it may have a significant effect on the amount of natural organics sorbed onto the solid surface (Hart, 1982). Feldspars are the major components and dominate the composition of estuarine sediments of coastal plains (Preda and Cox, 2000). Furthermore, cation forming elements such as Cu, Zn, etc. are more likely to be involved in oxidation and reduction reactions with each other as part of complex cycle (Kilham, 1990). Anthropogenic enrichment for heavy metals occurs naturally in silt and clay-bearing minerals of terrestrial and marine geological deposits. The natural occurrence of heavy metals complicates the assessment of potentially contaminated estuarine sediments. The measureable concentrations of metals do not automatically indicate anthropogenic enrichment in the lagoon. Therefore, heavy metal enrichment assessment must be conducted in detail (Zhou *et al*., 2004) using sediment indexes.

3.3 Geoaccumulation Index (I_{geo})

I_{geo} proposed by Muller (1979) has been applied to the set of sediment data from the present study to assess the sediment quality in Mengkabong mangrove forest (Table 6). I_{geo} values can be used more effectively and more meaningfully in explaining the sediment quality (Karbassi *et al*., 2006). The I_{geo} index showed that all the heavy metals are in Class 0 and Class 1 (Table 2) at high and low tide. Buccolieri *et al*. (2006) also reports similar findings in the Gulf of Taranto, Italy. This suggests that the mangrove sediment of Mengkabong is unpolluted. The input of metals into the sediments that are located seawards tends to be low in terms of concentration of most of the elements and this could be due to the mixing of enriched particulate material with relatively clean marine sediments (Soto-Jimenez and Paez-Osuna, 2001).

Table 6

4. Conclusions

Physicochemical characteristics of the surface sediment of Mengkabong Bay were determined including: pH, salinity and electrical conductivity, granulometric fractions (clay, silt and clay), heavy metals (Fe, Cu, Zn, Pb and Al) and base cations (Na, K, Mg and Ca) at high and low tides. The studied parameters showed high values at high tide compared to low tide. The tidal process showed there is a high contribution of seawater that brings additional ions such as Na, Mg and K into the mangrove system at high tide whereas at low tide, lower contribution of seawater was observed. The tides control the water-flows that carry the sediments in the mangrove forests, wherein the water from the estuary spills over to inundate the forests at high tide and drains back to the estuary at low tide. This simple explanation of conditions at high and low tides is vastly complicated by the presence of hydroxides and oxyhydroxide coating on the particles. The Spearman correlation rank was used for the statistical characterizations of mangrove surface sediment at high and low tides. The associations between physicochemical parameters, organic matter, granulometric fractions, heavy metals (Cu, Fe, Pb, Zn and Al) and base cations (Na, K, Mg and Ca) at high and low tide indicates the important contribution of seawater to sediment chemistry. The I_{geo} value of heavy metals showed that the Mengkabong mangrove sediments have background concentrations of Al, Cu, Fe, and Zn and are unpolluted with respect to Pb. These findings are important for future research which will explain the chemistry that lies in mangrove sediment.

Acknowledgements

References

Andrews, J. E. (2004). *An Introduction to Environmental Chemistry*. Blackwell Publishing. Oxford.

Akpan, E. R., Ekpe, U. J., & Ibok, U. J. (2002). Heavy Metal Trends in the Calabar River, Nigeria. *Environmental Geology, 42*, 47-51.

Alloway, B. J. (1995). *Heavy Metals in Soil*. 2nd Edition. Chapman and Hall, London.

APHA. (1995). *Standard Methods for the Examination of Water and Waste Water*. American Public Health Association.

Washington, D.C.

Berner, R. A. (1981). A New Geochemical Classification of Sedimentary Environments. *Journal of Sedimentology Petrology, 51*, 359-365.

Binkley, D., & Fisher, R. F. (2000).   *Ecology and Management of Forest Soils*. John Wiley and Sons, New York.

Bouillon, S., Middelburg, J. J., Dehairs, F., Borges, A. V., Abril, G., Flindt, M. R., Ulomi, S., & Kristensen, E. (2007). Importance of Intertidal Sediment Processes and Porewater Exchange on the Water Column Biogeochemistry in a Pristine Mangrove Creek (Ras Dege, Tanzania).   *Biogeosciences, 4,* 311–322.

Bloom, H., & Ayling, G.M. (1977). Heavy Metals in the Derwent Estuary. *Environmental Geology, 2*, 3–22.

Bryd, J. T., Lee, K. W., Lee, D. S., Smith, R. G., & Windom, H. L. (1990).   The Behaviour of Trace Metals in the Geum Estuary, Korea.   *Estuaries, 13*, 8-13.

Buccolieri, A., Buccolieri, G., Cardellicchio, N., Dell'Atti, A., Leo, A. D., & Maci, A. (2006). Heavy Metals in Marine Sediments of Taranto Gulf (Ionian Sea, Southern Italy). *Marine Chemistry, 99*, 227-235.

Calloy, S., O'Day, P. A., Esser, B., & Randall, S. (2002). *Speciation and Fate of Trace Metals in Estuarine Sediments under Reduced and Oxidized Conditions, Seaplane Lagoon, Alameda Naval Air Station (USA)*. U.S. Department of Energy Office of Scientific and Technical Information, P.O. Box 62 Oak Ridge. http://www.llnl.gov/tid/lof.

Che, O. (1999). Concentration of 7 Heavy Metals in Sediments and Mangrove Root Samples from Mai Po, Hong Kong. *Marine Pollution Bulletin*, *39*, 269-279.

Church, A. H. (1989). The Ionic of the Sea. *The Phytologist*. *68*, 239-247.

de Groot, A. J., Zschuppe, K. H., & Salomons, W. (1982).   Standardization of Methods of Analysis for Heavy Metals in Sediments. *Hydrobiologia*, *92*, 689-695.

de Lacerda, L.D., Santelli, R.E., Duursma, E.K., & Abrao, J.J. (2004).   *Environmental Geochemistry in Tropical and Subtropical Environments,* Springer-Verlag, New York.

Defew, L. H., Mair, J. M., & Guzman, H. M. (2005).   An Assessment of Metal Contamination in Mangrove Sediments and Leaves from Punta Mala Bay, Pacific Panama. *Marine Pollution Bulletin*, *50*, 547-552.

Dittmar, T., & Lara, R. J. (2001).   Driving Forces behind Nutrient and Organic Matter Dynamics in a mangrove tidal creek in North Brazil.   *Estuarine, Coastal and Shelf Science**, 52,* 249-259**.

Eisma, D. (1998).   Intertidal *Deposits: River Mouths, Tidal Flats, and Coastal Lagoons*.   CRC Press, Boca Raton, Florida.

El Nemr, A., Khaled, A., & Sikaily, A. E. (2006).   Distribution and Statistical Analysis of Leachable and Total Heavy Metals in the Sediments of the Suez Gulf. *Environmental Monitoring and Assessment*, *118*, 89-112.

Emmerson, R. H. C., O'Reilly-Wiese, S. B., Macleod, C. L., & Lester, J. N. (1997). A Multivariate Assessment of Metal Distribution in Inter-tidal Sediments of the Blackwater Estuary, UK.   *Marine Pollution Bulletin*, *34*, 960-968.

Environmental Indicator Report. (2003). Environment Protection Department, Sabah, Malaysia.   Syarikat Bumi Yakin.

Hart, B. T. 1982. Uptake of Trace Metals by Sediments and Suspended Particulates: A Review. *Hydrobiologia, 91*, 299-313.

Heiri, O., Lotter, A. F. & Lemcke, G. (2001).   Loss on Ignition as a Method for Estimating Organic and Carbonate Content in Sediments: Reproducibility and Comparability of Results.   *Journal of Paleolimnology*, *25*, 101-110.

Hsue, Z., & Chen, Z. (2000). Monitoring the Change of Redox Potential, pH and Electrical Conductivity of the Mangrove Soils in Northern Taiwan. *Proceeding Natural Science Council*, *24*, 143-150.

Hussein, A. H., & Rabenhorst, M. C. (2001). Tidal Inundation of Transgressive Coastal Areas: Pedogenesis of Salinization and Alkalinization. *Soil Science Society America Journal*, *65*,536–544.

Jonathan, M. P., Ram-Mohan, V., & Srinivasalu, S. (2004).   Geochemical Variations of Major and Trace Elements in Recent Sediments off the Gulf of Mannar, the Southeast Coast of India.   *Environmental Geology*, *45*, 466-480.

Kamaruzzaman, B. Y., Ong, M. C., Norhizam, H. A. G., & Willison, K. Y. S. (2006). The Pollution Status of the Selected Heavy Metal in Mengabang Telipot River Surrounding Kolej Universiti Sains and Teknologi Malaysia (KUSTEM), Terengganu. *International Conference on Coastal Oceanography and Sustainable Marine Aquaculture*. 2-4 May 2006, Kota Kinabalu, Sabah, Malaysia.

Karbassi,   A. R., & Shankar, R. (2005). Geochemistry of Two Sediment Cores from the West Coast of India. *International Journal Environmental Science and Technology*, *1(4),* 307-316.

Kilham, P. (1990). Mechanism Controlling the Chemical Composition of Lakes and Rivers. Data from Africa.

Limnology Oceanography, *35*, 80-83.

Koop-Jakobsen, K. & Giblin, A. (2002).   Northeast Tidal Flushing of Ammonium from Intertidal Salt Marsh Sediments: The Relative Importance of Adsorbed Ammonium. *The Biological Bulletin*, *203*, 258-259.

Kristensen, E., Holmer, M., & Bussarawit, N. (1991).   Benthic Metabolism And Sulphate Reduction In A Southeast Asian Mangrove Swamp. *Marine Ecology Progress Series, 73*, 93-103.

Lin, J., & Chen, S. (1998). The Relationship between Adsorption of Heavy Metal and Organic Matter in River Sediment. *Environment International*, *24*, 345-352.

Malvarez, G. C., Cooper, J.A.G., & Jackson, D.W.T. (2001).   Relationships between Wave-Induced Currents and Sediment Grain Size on a Sandy Tidal-Flat.   *Journal of Sedimentary Research*. *71,* 705-712.

Mantha, D. S., Angelone, M., Belaanca, A., Neri, R., & Sprovieri, R. (2003). Heavy Metal in Urban Soils: A Case Study from the City of Palermo (Sicily), Italy. *Science of Total Environment*, *300*, 229-243.

Marchand, C., Baltzerb, F., Lallier-Verge`sa, E., & Albe´rica, P. (2004).   Pore Water Chemistry in Mangrove Sediments: Relationship with Species Composition and Developmental Stages (French Guiana). *Marine Geology*, *208*, 361-381.

Marchand, C., Lalliet   Verges, E., Baltzer, F., Alberic, P., Cossa, D., & Baillif, P. (2006). Heavy Metals Distribution in Mangrove Sediments along the Mobile Coastline of French Guiana. *Marine Chemistry*, *98*, 1-17.

Milligan, T. G., & Loring, D. H. (1997). The Effect of Flocculation on the Size Distributions of Bottom Sediment in Coastal Inlets: Implications for Contaminant Transport. *Water, Air and Soil Pollution*, *99*, 33-42.

Mitsch, W. J., & Gosselink, J. G. (2000). *Wetlands*. 3rd Edition, John Wiley and Sons, New York.

Morel, F. M. M., & Hering, J. G. (1993).   *Principles and Applications of Aquatic Chemistry*.   John Wiley and Sons, New York.

Muller, G. (1979). Schwermetalle in den sedimenten des Rheins-VeraÈnderungenseit. *Umschau*, *79*, 778-783.

Pekey, H. (2006).   Heavy Metals Pollution Assessment in Sediments of the Izmit Bay, Turkey. *Environmental Monitoring and Assessment*, *123*, 219-231.

Preda, M., & Cox, M. E. (2000). Sediment-Water Interaction, Acidity and Other Water Quality Parameters in a Subtropical Setting, Pimpama River, Southeast Queensland.   *Environmental Geology*, *39*, 319-329.

Radojevic, M., & Bashkin, V. N. (1999).   *Practical Environmental Analysis*.   Royal Society of Chemistry, Cambridge.

Ramanathan, A. L., Subramaniam, V., Ramesh, R., Chidambaram, S., & James, A. (1999).   Environmental Geochemistry of the Pichavaram Mangrove Ecosystem (Tropical), Southeast Coast of India. *Environmental Geology*, *37*, 223-233.

Rand, G. M.   (1995). *Fundamentals of Aquatic Toxicology—Effects, Environmental Fate and Risk Assessmen.* Taylor and Francis Publishers, Washingston.

Rate, A. W. W., Robertson, A. E., & Borg, A. T. (2000).   Distribution of Heavy Metals in Near-Shore Sediments of the Swan River Estuary, Western Australia.   *Water, Air and Soil Pollution*, *124*, 155–168.

Ray, A. K., Tripathy, S. C., Patra, S., & Sarma, V. V. (2006).   Assessment of Godavari Estuarine Mangrove Ecosystem through Trace Metals Studies.   *Environment International*, *32*, 219-223.

Rodrigo, A. G. (1989). Surficial Sediment Heavy Metal Associations in the Avon-Heathcote Estuary, New Zealand. *New Zealand Journal of Marine and Freshwater Research, 23*, 255-262.

Shriadah, M. M. A. (1999). Heavy Metals in Mangrove Sediments of the United Arab Emirates Shoreline (Arabian Gulf). *Water, Air and Soil Pollution*, *116*, 523-534.

Singh, A. K., Hasnain, S. I., & Banerjee, D. K. (2003).   Grain Size and Geochemical Portioning of Heavy Metals in Sediments of the Damodar River- A Tributary of the Lower Ganga, India. *Environmental Geology, 39*, 90-98.

Soto-Jimenez, M. F., & Pacz-Osuna, F. (2001). Distribution and Normalization of Heavy Metal Concentrations in Mangrove and Lagoonal Sediments from Mazatlan Harbour (SE Gulf Of Calfornia).   *Estuarine, Coastal and Shelf Science*, *53*, 259-274.

Sparks, D. (1995). *Environmental Soil Chemistry*. Academic Press Limited, California.

Spencer, K. L., & Macleod, C. L. (2002). Distribution and Partitioning of Heavy Metals in Estuarine Sediment Cores and Implications for the Use of Sediment Quality Standards. *Hydrology and Earth System Sciences, 6*, 989-998.

Solomon, W. & Forstner, U. 1984. *Metals in the Hydrocycle*. Springer-Verlag, New York.

Town and Regional Planning Department (TRPD) (2003). *Project Sabah*, 2003.   Environmental Local Planning (ELP), Kota Kinabalu, Sabah.

Twilley, R. R., Rivera-Monroy, V. H., Chen, R., & Botero, L. (1999).   Adapting an Ecological Mangrove Model to Simulate Trajectories in Restoration Ecology.   *Marine Pollution Bulletin*, 37, 404-419.

Vanloon, G. W., & Duffy, S. J. (2005). *Environmental Chemistry: A Global Perspective*.   2nd Edition.   Oxford University Press Inc.

Velde, B. (1995). *Origin and Mineralogy of Clays*. Springer, New York.

Walpersdorf, E., Werner, U., Polerecky, L., & Franke U. (2008).   Intertidal *and Coral Lagoon Sediments* . http://www.mpi-bremen.de/en/Sediments_2.html. 7 February 2008.

Wolanski , E. (2006). The Environment in Asia Pacific Harbours, Springer, Dordrecht.

Zektser , I. S., & Dzhamalov, R. G. (2006).   *Submarine Groundwater*.   CRC Press, Boca Raton, Florida.

Zhou, H., Peng, X., & Pan, J. (2004).   Distribution, Source and Enrichment of Some Chemical Elements in Sediments of the Pearl River Estuary, China.   *Continental Shelf Research*, *24*, 1857-1875.

Table 1. Analytical methods used in this study

Parameter	Method	Reference
pH	Glass electrode	Mettler Toledo Delta 320 Manual
EC	Glass electrode	Eutech/Oakton ECTestr [TM] 11 Plus Manual
Salinity	Glass electrode	Eutech/Oakton SaltTestr [TM] 11 Plus Manual
Organic Matter	Loss on Ignition (LOI)	Heiri *et al*. (2001), Radojevic and Bashkin (1999)
Granulometric analysis	Pipette Method	Radojevic and Bashkin (1999)
Total Heavy Metals	AAS	APHA (1995)
Base Cations	AAS	APHA (1995)

Table 2. Descriptive Statistics for each parameter at high and low tide (n=33).   All elemental concentrations are in mg kg^{-1} except for Na, K, Mg, Ca, Fe and Al. (Sa=salinity, EC=Electrical Conductivity, OM=Organic matter, HT = High Tide, LT = Low tide)

Parameter		Min	Max	Mean	SD
pH	HT	5.4	7.6	6.5	0.4
	LT	4.5	7.2	6.1	0.6
Sa (%)	HT	1.5	7.4	4.5	2.2
	LT	0.1	1.5	0.5	0.4
EC(mS cm^{-1})	HT	2.1	11.4	6.8	2.6
	LT	0.4	4.8	2.6	1.0
OM (%)	HT	6.4	11.7	9.0	1.3
	LT	1.4	5.2	2.5	0.8
Sand (%)	HT	91.9	95.1	93.6	0.8
	LT	91.7	97.3	95	1.7
Silt (%)	HT	2.5	5.2	3.6	0.8
	LT	0.4	6.4	2.7	1.8
Clay (%)	HT	2.2	4.2	2.8	0.4
	LT	1.3	2.9	2.4	0.4
Na (g kg^{-1})	HT	14.2	83.5	47.5	1.0
	LT	2.5	92.7	41.6	0.4
K (g kg^{-1})	HT	5.7	16.0	9.4	2.5
	LT	2.4	10.6	7.8	1.8
Mg (g kg^{-1})	HT	2.0	9.2	5.3	1.8
	LT	1.0	7.6	3.8	1.9
Ca (g kg^{-1})	HT	2.6	52.9	21.3	41.7
	LT	1.5	47.7	16.2	15.1
Fe (g kg^{-1})	HT	3.4	14.2	7.7	2.7
	LT	1.4	18.4	6.8	4.0
Cu (mg kg^{-1})	HT	4.1	49.0	28.0	14.0
	LT	2.1	44.0	19.0	13.0
Zn (mg kg^{-1})	HT	24.0	93.0	57.0	17.0
	LT	12.0	73.0	41.0	17.0
Pb (mg kg^{-1})	HT	24.0	69.0	52.0	11.0
	LT	34.0	47.0	41.0	3.0
Al (g kg^{-1})	HT	4.4	3.5	14.8	8.2
	LT	2.4	2.4	9.5	6.0

Table 3. Comparison of metal levels (mg kg^{-1}) in various mangrove environments

LOCATION	Al	Cu	Fe	Pb	Zn
Present Study	2411-3539 3	2-49	1434-183 60	24-69	12-93
Telipot mangrove, Terengganu, Malaysia[1]	-	3.53	-	8.60	27.15
Pearl River Estuary, China[2]	-	46.5	-	59.26	150.06
Pichavaram, India[3]	46100	43.4	8080	10.4	62.2
Izmit Bay, Turkey[4]	-	39.2	-	77.1	345
Punta Mala Bay, Panama[5]	-	56.3	9827	78.2	105
Mai Po, Hong Kong[6]	-	20-75	-	58.2	53.3
Arabian Gulf, United Arab Emirates[7]		8.32		37.3	9.05
Kaw River, French Guiana[8]		2.8	7890	1.3	25.1
Ganges River, India[9]	-	55	40456	22	102
Beeleigh, Blackwater Estuary,UK[10]	19643	77	33302	60	190
Medway Estuary, UK[11]	-	42	30282	67	138
Mean sediment[12]	72, 000	50	41, 000	19	95

[1]Kamaruzzaman et al. (2006); [2] Zhou et al. (2004); [3] Ramanathan et al. (1999); [4] Pekey (2006);

[5] Defew et al. (2005); [6] Che (1999); [7] Shriadah (1999); [8] Marchand et al. (2006); [9] Singh et al. (2003)

[10] Emmerson et al. (1997); [11] Spencer and Macleod (2002); [12] Solomon & Forstner (1984)

Table 4. Spearman correlation coefficient for Mengkabong surface sediments at high tide (n=33)

	pH	Sa	EC	OM	Fe	Cu	Zn	Pb	Al	Ca	Mg	K	Na	Clay	Silt	Sand
pH	1															
Sa	0.065	1														
EC	-0.070	0.798**	1													
OM	-0.177	0.108	0.255	1												
Fe	-0.112	-0.042	-0.117	-0.091	1											
Cu	-0.025	-0.100	-0.175	0.127	0.110	1										
Zn	-0.215	-0.380*	-0.307	-0.338*	-0.003	-0.344*	1									
Pb	0.013	0.006	-0.019	0.054	0.163	0.215	-0.175	1								
Al	0.233	-0.213	-0.172	-0.228	0.011	-0.218	0.345*	0.192	1							
Ca	-0.102	0.267	0.252	-0.077	0.194	-0.104	-0.129	-0.017	0.017	1						
Mg	-0.120	-0.155	-0.135	0.075	-0.024	0.103	-0.077	0.071	0.214	0.369*	1					
K	-0.134	-0.185	-0.256	0.036	0.231	0.332*	-0.006	-0.062	-0.135	0.116	0.280	1				
Na	0.199	-0.073	-0.164	-0.194	0.233	-0.165	0.069	0.021	-0.037	0.119	-0.091	0.055	1			
Clay	-0.191	0.398*	0.469**	0.335*	0.150	0.180	-0.227	0.057	-0.317	0.053	0.145	0.070	0.071	1		
Silt	0.377*	-0.332*	-0.404*	-0.251	0.096	-0.075	0.255	-0.021	0.209	-0.308	-0.295	0.155	0.200	-0.290	1	
Sand	-0.177	0.151	0.184	0.091	-0.159	0.019	-0.195	-0.048	-0.060	0.279	0.255	-0.138	-0.224	-0.218	-0.842**	1

(Sa=salinity, EC=Electrical Conductivity, OM=Organic matter)

**Significant at the 0.01 level ($p < 0.01$)
* Significant at the 0.05 level ($p < 0.05$)

Table 5. Spearman correlation coefficient for Mengkabong surface sediments at low tide (n=33)

	pH	Sa	EC	OM	Fe	Cu	Zn	Pb	Al	Ca	Mg	K	Na	Clay	Silt	Sand
pH	1															
Sa	0.259	1														
EC	-0.306	-0.220	1													
OM	0.041	0.007	0.033	1												
Fe	0.211	-0.011	0.255	0.033	1											
Cu	-0.020	0.048	-0.079	-0.079	0.021	1										
Zn	-0.378*	0.025	0.248	0.282	0.458**	0.072	1									
Pb	-0.168	-0.011	0.112	0.124	0.025	0.033	-0.071	1								
Al	0.115	-0.263	0.093	-0.009	0.531**	-0.087	0.345*	-0.105	1							
Ca	0.221	-0.198	-0.017	0.265	-0.079	0.037	-0.184	0.262	-0.007	1						
Mg	0.443*	0.058	0.155	0.304	0.093	0.054	-0.147	-0.020	0.061	0.560**	1					
K	0.005	0.320	-0.096	0.176	0.385*	0.213	0.415*	0.086	0.026	0.089	0.075	1				
Na	-0.003	-0.023	-0.021	-0.127	0.025	-0.052	-0.257	0.222	-0.154	0.212	0.084	0.143	1			
Clay	0.129	0.244	-0.159	-0.010	-0.097	0.291	-0.246	-0.183	-0.374*	-0.103	0.009	0.003	0.076	1		
Silt	-0.338*	-0.122	0.305	-0.055	0.404*	-0.083	0.510**	0.025	0.516**	-0.339*	-0.274	0.190	0.046	-0.219	1	
Sand	0.301	0.072	-0.291	0.088	-0.412*	0.022	-0.441**	0.007	-0.485**	0.369*	0.273	-0.195	-0.018	0.139	-0.977**	1

(Sa=salinity, EC=Electrical Conductivity, OM=Organic matter)

**Significant at the 0.01 level ($p < 0.01$)
* Significant at the 0.05 level ($p < 0.05$)

Table 6. Index of Geoaccumulation (I_{geo}) for heavy metals in sediment of Mengkabong mangrove forest

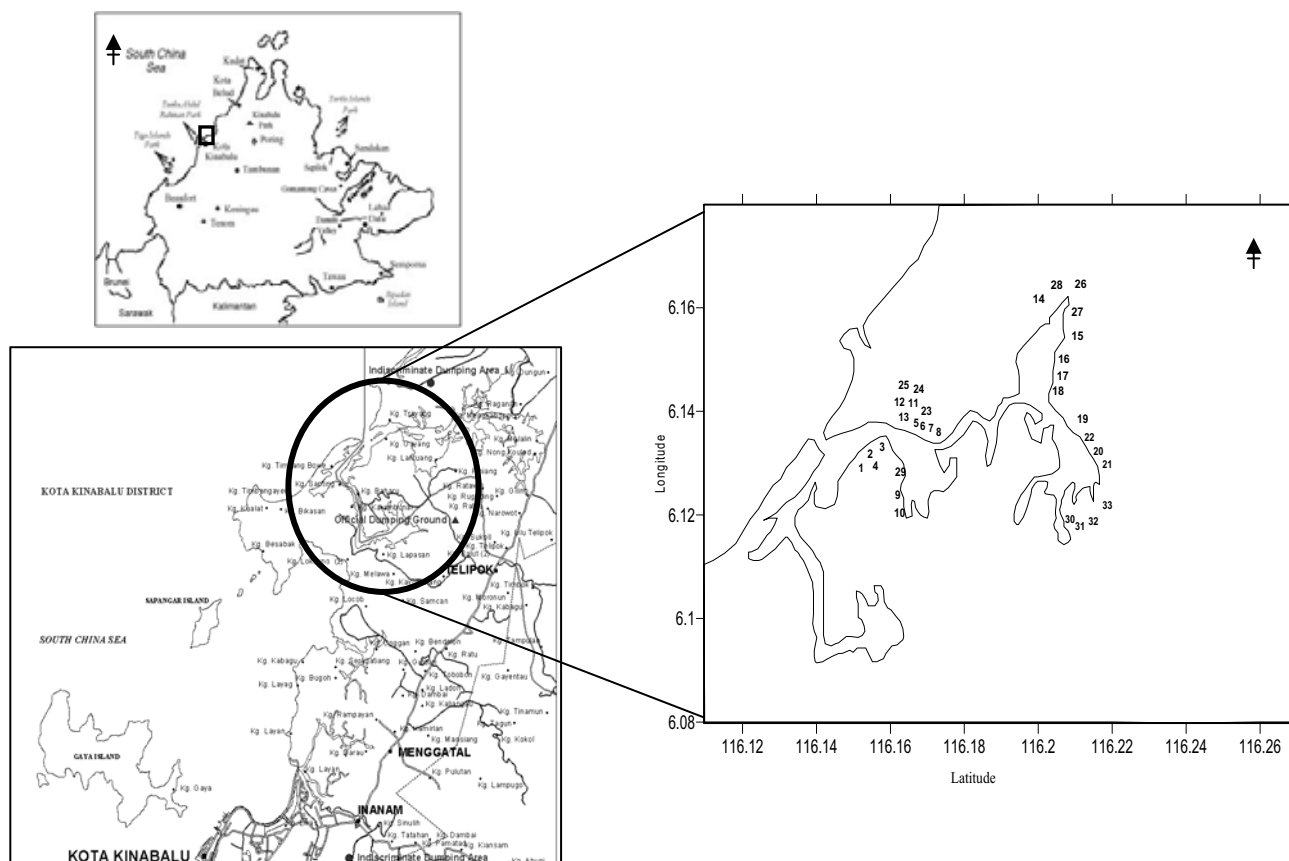| Heavy Metals (I_{geo} class for Mengkabong lagoon sediment)	Pollution Intensity	Geoaccumulation index
Al, Cu, Fe, Zn	Background concentration	0
Pb	Unpolluted	0-1
	Moderately to unpolluted	1-2
	Moderately polluted	2-3
	Moderately to highly polluted	3-4
	Highly polluted	4-5
	Very highly polluted	>5

Figure 1. Location of 33 surface sediments taken in Mengkabong Bay