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ANTHROPOGENIC INPUTS OF CHEMICALS IN THE MANGROVE INTERTIDAL ECOSYSTEM SUNGAI PULOH, MALAYSIA

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ABSTRACT: *Mangrove ecosystems are increasingly threatened by anthropogenic activities, leading to the accumulation of heavy metals in coastal sediments. This study investigated the levels, distribution, and potential sources of copper (Cu) and zinc (Zn) in mangrove intertidal sediments of Sungai Puloh (SGP) and Sungai Tengi (SGT) along the west coast of Peninsular Malaysia. Surface sediment samples were collected from eight stations and analyzed using the Sequential Extraction Technique (SET) to determine metal partitioning into exchangeable, acid-reducible, oxidisable-organic, and resistant fractions. Total metal concentrations were measured using atomic absorption spectrophotometry, while total organic matter (TOM) was determined via the loss-on-ignition method. Mean concentrations of Cu ranged from 48.83 to 85.49 µg/g in SGP and 8.30 to 35.80 µg/g in SGT, whereas Zn ranged from 279.51 to 321.55 µg/g in SGP and 99.75 to 1138.38 µg/g in SGT. Geochemical fractionation revealed that a significant proportion of Cu and Zn in SGP occurred in non-resistant fractions (24.4–29.8% for Cu and 59.8–65.9% for Zn), indicating higher anthropogenic influence compared to SGT. Elevated Cu levels in SGP were attributed to industrial effluents and maritime activities, while high Zn concentrations in SGT were linked to domestic waste and agricultural runoff. A positive correlation between TOM and metal concentrations suggests that organic matter plays a role in metal retention and mobility within sediments. Overall, the findings indicate that anthropogenic inputs significantly influence heavy metal contamination in these mangrove ecosystems, with potential ecological and human health implications. Continuous monitoring and improved management strategies are recommended to mitigate further contamination.*

KEYWORDS: Anthropogenic influence, Cu and Zn, Mangrove sediment, Total organic matter.

INTRODUCTION

Today’s mangroves have suffered a lot of neglect and abuse resulting from human activities. Intense anthropogenic activities have recently led to the contamination of marine ecosystems (Galloway et al., 2002). Despite their importance, mangrove ecosystems have been exploited and subjected to inappropriate management practices in terms of land reclamation and unsustainable forestry, as well as aquaculture initiatives (West et al., 1983; Eong, 1995). As a consequence of proximity to urban development, they have experienced significant direct contaminant input. Among the major pollutants from anthropogenic inputs are heavy metals (MacFarlane, 2002). Mangrove forests are very important tropical coastal tidal ecosystems and grow on nutrient-rich muddy substrates that are low in oxygen and that undergo variations in salinity (Shaeffer-Novelli, 1999). The important functional role of mangrove forest communities and their transitional position between marine and terrestrial environments have led to these ecosystems being the object of study within a variety of scientific disciplines such as biology, ecology, geology, oceanography, and pedology. However, scientists, including pedologists, often refer to the substrate on which mangrove vegetation develops as sediment rather than as soil (Clark et al., 1998). Sediments are defined as deposits of solid material on the earth's surface, from any medium (air, water, ice) (Krumbein & Sloss, 1963).

Anthropogenic sources of heavy metals in sediments.

Mangrove sediments have been shown to have a high capacity to accumulate materials discharged to the near-shore marine environment (Harbison, 1986). This is mainly due to the anaerobic as well as rich sulfide and organic matter content that favors the retention of water-borne heavy metals.) Organic matter in sediments is composed of carbon and nutrients in the form of carbohydrates, proteins, fats, and nucleic acids. Most sediment organic matter is derived from plants and animal detritus, bacteria, or planktons found in situ or derived from natural and anthropogenic sources. Sewage and effluents from processing plants, pulp and paper mills, and fish farms are examples of organic –rich wastes of anthropogenic origin.

Heavy metal contamination of the mangrove ecosystem arises from urban and agricultural runoff, industrial effluents, boating and recreational use of water bodies, chemical spills, sewage treatment plants, leaching from domestic garbage dumps, and mining operations (Peters et al., 1997). As a result, elevated concentrations of heavy metals have been recorded in mangrove sediments, which often reflect long-term pollution caused by human activities (Preda & Cox, 2002). The metal pollutants most commonly entering estuarine systems through industrial sources (based on frequency of detection) are copper (Cu), lead (Pb), and zinc (Zn) (Mills, 1995). These metals consequently occur in high concentrations in many estuaries, with concentrations reaching up to 1000 μg/g Cu, 1000 μg/g Pb, and 2000 μg/g Zn in contaminated sediments (Irvine and Birch, 1998).

Copper is ranked the third most toxic metal to aquatic biota, after mercury and silver (Waldichuk, 1974). In sediments, Cu and Zn are found since they are essential metals for biota, being associated with numerous metalloenzymes and metalloproteins (Thompson, 1990). Elevated levels of Cu and Zn in the environment not only disturb the ecological system but also affect shrimp farming activities, such as the toxicity effect on reduced productivity. Shrimp farming may also increase copper levels in the environment since copper might be used in shrimp farms to control algal growth (Ismail et al., 2004).

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However, high levels of Cu and Zn in the sediment could be potentially toxic, and this may pose a concern because of bioaccumulation once in the food web. Chronic exposure to heavy metals such as Cu, Pb, and Zn is associated with Parkinson’s disease, and metals might act alone or together over time to cause diseases (Gorell et al., 1997).

Malaysia has taken the initiative to protect certain mangrove areas as a protected ecosystem for conservation and maintaining biodiversity. Unfortunately,, Malaysia is one of the fastest-developing countries in the world. As a result of this development, continuous and heavy human activities and continuous chemical input are expected. Mangrove sediments are believed to be the metal repository (Nriagu, 1978), and only a minor fraction of materials escapes into the coastal waters. Bottom sediment is a long-term integrator of geochemical processes; therefore, information on heavy metals in mangrove sediments can establish long-term behavior and sources of these metals.

Sg. Puloh and Sg. Tengi is close to the offshore area of the west coast of Peninsular Malaysia, which is now one of the busiest shipping lanes in the world. As a result, these mangroves may accumulate materials discharged from the nearby sea (straits of Malacca) during tides and solid wastes from the nearby communities, boating and agricultural activities, effluents, and metallic anthropogenic wastes disposed of from nearby smelting industries. Therefore, this research study aims to assess the levels of man-induced heavy metal contamination by copper (Cu) and zinc (Zn) in the mangrove intertidal sediments of Sg. Puloh and Sg. Tengi in West Peninsular Malaysia.

Materials and Methods

Sample Preparation: Sediment samples were collected from the mangroves of Sg. Puloh (SGP N 03°04.786′ E 101°23.903′ e: 8m) which Sg. Tengi (SGT N 03°24.682, E 101°9.971, e: 10m) in Selangor during the January and February dry spell. Four stations were sampled in each of the mangrove sites (SGP 1, SGP 2, SGP 3, and SGP 4) and (SGT1, SGT 2, SGT 3, and SGT 4) respectively. The selection of SGP was based on its exposure to substantial anthropogenic activities, including industrial operations, shipping and port-related activities, aquaculture practices, urban expansion, and domestic effluent inputs. These land-use activities are recognized as important sources of heavy metal enrichment and environmental stress in coastal ecosystems. In contrast, SGT was selected as a relatively pristine mangrove system characterized by limited industrialization, lower population density, reduced human disturbance, and comparatively undisturbed ecological conditions.

The ecological contrast between the two mangrove systems provided an appropriate framework for assessing the influence of anthropogenic pressure on sediment quality, metal distribution, and environmental contamination within mangrove intertidal ecosystems.

All samples were collected during low tide. About 2-3cm surface sediments were collected, placed in a polyethylene bag, transported to the laboratory, and frozen (-10°C) before analysis.

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Metal analysis

The geochemical fractions were analyzed using the Sequential Extraction Technique (SET) as described by Badri and Aston (1983).

The SET (Sequential Extraction Technique)

The four fractions considered, extraction solutions, and conditions employed are as follows:

1. **EFLE (easily freely leachable and exchangeable**): About 10g of the sediment sample is weighed in a clean conical flask and shaken for 3 hours with 50ml of 1.0M ammonium acetate (NH_4CH_3COO), pH 7.0 (in a shaker of about 100rpm) at room temperature. The EFLE liberates the metals that are loosely bound to other substances .
2. **Acid – reducible fraction**: The residue from (1) is weighed and shaken for 3 hours with 50ml of 0.2Mhydroxylammonium chloride ($NH_2OH.HCl$), acidified to pH 2.0 with hydrochloric acid (HCl) at room temperature. This stage releases the metals from acid reducible substances, e.g., SO_3.
3. **Oxidisable -Organic:** The residue from (2) is weighed and first oxidized with 15ml of 30% hydrogen peroxide (H_2O_2) in a water bath at 90-95℃, until dried. After cooling, the heavy metals released from the organic complexes are continuously shaken for 3 hours with 1.0 M ammonium acetate (NH_4CH_3OO), acidified to pH 2.0 with HCl at room temperature. According to the experiment, this fraction contains a greater percentage of the non-resistant fractions. The H_2O_2 is used because it is a powerful and versatile oxidant. It hydrolyses formaldehyde, carbon disulfides, carbohydrates, organophosphorus and nitrogen compounds, and various water-soluble polymers. By its oxidant ability, it liberates metals from these organic complexes.
4. **Resistant fraction**: The residue from (3) is weighed and digested in a combination of concentrated nitric acid and perchloric acid as in the direct aqua regia method. This is the fraction that can neither be easily leached, acid-reduced, nor oxidized,

The direct aqua regia method (Ismail, 1993) was employed for the heavy metal analysis. About 1g of each dried sample is weighed and digested in a mixture of concentrated nitric acid (HNO_3 AnalaR grade, BDH 69%) and perchloric acid (AnalaR grade 60%) in a ratio 4:1. The samples are then placed in the digesting block, and the temperature was first set at a low temperature of 40°C for 1hr and later increased to 140°C for at least 3hrs (Ismail et al., 1993 and Yap, et al., 2002) After digestion, the samples were diluted to a 40ml volume with double distilled water and filtered using the Whatman No.1 filter paper into acid-washed polyethylene sample bottles. Then the samples were determined for Cu and Zn by using an air-acetylene flame Atomic absorption Spectrophotometer (Analyst 800 model, by Perkin-Elmer) and presented in µg/g dry weight basis (DW). Calibration curves were generated by analyzing Multiple-level Calibration standards. Standard solutions of Cu and Zn were prepared from 1,000mg/l (BDH SpectrosoL®) stock solution. In order to avoid contamination, all glassware was soaked in acid wash (10% HCL) for at least 24 hours and later rinsed with double-distilled water. A Certified

Reference Material (CRM) for soil (International Atomic Energy Agency, Soil- 5, Vienna, Austria) was used to check the quality of this method. The analytical results for reference material and its certified values showed satisfactory agreement with the recoveries being 99.4% Cu and 105% for Zn. A quality control sample was analyzed in every eight samples during the AAS metal analysis. The Total Organic Matter (TOM) was determined by using a high temperature method (Mucha et al., 2003).

Determination of Total Organic Matter

About 10g of dried sediment sample was put into a crucible and heated in a furnace at 500˚C for 4hrs It was then put in a desiccator and weighed.

$$\%TOM = \frac{\text{weight of sediments (before drying - weight of sediment after drying)}}{\text{Weight of sediment before drying}} \times 100\%$$

RESULTS AND DISCUSSION

Table I: Mean metal levels and total organic matter in mangrove sediments of Sg Puloh and Sg Tengi

Stations	Site description	mean conc. Cu(µg/g)	mean conc. Zn(µg/g)	TOM%
SGP 1	Industrial effluence	85.49±4.47	317.9±9.13	18
SGP 2	near roadside	81.39±3.19	321.55±13.78	12
SGP 3	under jetty	48.83±4.15	279.51±10.11	13
SGP 4	river mouth	52.96±2.28	291.58±5.66	11
SGT 1	solid wastes	35.8±4.32	1138.38±22.15	16
SGT 2	near road side	8.3±1.01	664.3±12.35	11
SGT 3	under the jetty	29.66±2.12	99.75±3.80	14
SGT 4	river mouth	28.14±2.11	685.51±14.46	3

Table II Mean Cu concentrations (µg/g) in geometrical fractions of sediment samples.

Stations	EFLE	Acicd-reducible	Oxidisable-organic	Resistant	∑Non-Resistant	Total (100%)
SGP 1	0.55(0.6)	0.2(0.2)	25.12(29.0)	60.83(70.2)	25.86(29.8)	86.69
SGP 2	0.41(0.5)	0.06(0.1)	23.07(27.8)	59.61(71.7)	23.54(28.3)	83.15
SGP 3	0.46(0.9)	0.29(0.6)	11.62(22.9)	38.37(75.6)	12.37(24.4)	50.74
SGP4	0.34(0.6)	0.14(0.3)	13.41(24.5)	40.79(74.6)	13.89(25.4)	54.68
SGT 1	0.23(0.7)	0.06(0.2)	4.46(1.5)	30.16(86.4)	4.75(13.6)	34.91
SGT 2	0.47(5.5)	0.14(1.7)	1.44(17.0)	6.42(75.8)	2.05(24.2)	8.47

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SGT 3	0.38(1.1)	0.15(0.5)	1.95(6.4)	28.23(92.0)	2.48(8.0)	30.7
SGT 4	0.45(1.5)	0.09(0.3)	1.55(5.2)	27.9(93.0)	2.09(7.0)	29.99

***The values in parentheses are the percentage of the fraction out of the total concentration**

Table III Mean Zn concentrations (µg/g) in geometrical fractions of sediment samples

Stations	**EFLE**	**Acicd-reducible**	**Oxidisable-organic**	**Resistant**	**∑Non-Resistant**	**Total (100%)**
SGP 1	29.85(9.3)	83.06(26.0)	93.11(29.2)	113.2(35.5)	206.0(64.5)	319.2
SGP 2	17.51(5.4)	83.05(25.7)	92.86(28.7)	130.0(40.2)	193.4(59.8)	323.4
SGP 3	20.27(7.3)	82.24(29.5)	81.08(29.1)	94.80(34.1)	183.59(65.9)	278.39
SGP4	23.05(8.0)	80.88(27.5)	82.29(28.0)	107.72(36.6)	186.37(63.4)	294.1
SGT 1	0.29(0.1)	5.1(0.4)	437.13(38.45)	694.33(87.5)	442.52(38.92)	1136.85
SGT 2	0.46(0.1)	24.28(3.6)	195.16(29.4)	443.86(66.9)	219.9(33.1)	663.76
SGT 3	0.24(0.2)	4.53(4.5)	18.90(18.9)	76.58(76.4)	23.67(23.6)	100.25
SGT 4	0.21(.03)	0.6(0.1)	205.67(30.0)	479.72(69.9)	206.48(30.1)	686.2

***The values in parentheses are the percentage of the fraction out of the total concentration**

The result from Table I above shows the mean concentration levels of Cu in µg/g dry weight, ranging from 48.83 ±0.15 to 85.49 ±0.47 for Sg. Puloh., and 8.3 ±0.01 to 35.8 ±0.32 for Sg. Tengi. While the mean concentration levels of Zn range from 279.51 ±1.1 to 321.55 ±2.78 for Sg. Puloh and 99.75 ±0.80 to 1138.38 ±1.15 for Sg. Tengi. The Total organic matter by dry weight ranged from 11 – 18% in Sg. Puloh and 3 –16% in Sg.Tengi, and is comparable with (0.6-19%), which is reported by William et al 2002. Total metal concentrations are computed by the summation of the four geochemical fractions. It ranged from 50.74 - 86.69 µg/g Cu and from 278.39 - 319 µg/g Zn in SGP (Table II), also it ranged from 8.47 - 34.91 µg/g Cu and from 100.25 - 1136.85 g/g Zn in SGT (Table III). The elevated levels of Cu in SGP could be a result of discharge from nearby sea (Harbison, 1986), industrial effluents, and boating (Peters et al., 1997). The elevation of Zn in SGT could be a result of leaching from domestic garbage dumps and agricultural runoff from the nearby communities directly to the mangrove (Peters et al., 1997 , Chew et al., 2002). The elevated values of Cu in SGP 1 and SGP 2 may be from industrial effluence and subsequent oxidation of sulphides between tides, which mobilizes the previously trapped metals (Clark et al., 1998; Tam and Wong, 2000). SGT 1 showed elevated Zn concentrations, which is a result of leaching from the refuse dumps and domestic discharge from adjacent areas through the monsoon drainage system (Mohkeri, 2002). High input of organic matter into Sg. Tengi probably contributed to these values. SGT 1 probably received high organic matter input from human activities. MacFarlane et al. (2003) reported that the concentrations in mangrove sediments of SE Australia ranged from 1 - 102µg/g and 10 -287µg/g (Table IV) for Cu and Zn, respectively. These values are comparable to values obtained in the SGP of this study. Also, this present study is comparable with that of Ismail et al. (1993), who reported that the background levels of heavy metal concentration of west coast peninsular Malaysia were <10 µg/g for Cu and 50 - 1400 µg/g for Zn. However, the wide difference in Cu could be attributed to rapid development and increased human activities within the past 15years. Moreover, the reports of Thomas and Fernandez (1997), of mangrove sediments in India, showed very high Cu (303µg/g) when compared to this study; this could possibly be a result of more industrial pollution and higher human activities due to population.

Figure 1. Comparison of mean Cu concentration levels of non-resistant fractions (%) between SGP and SGT using a paired *t-test*

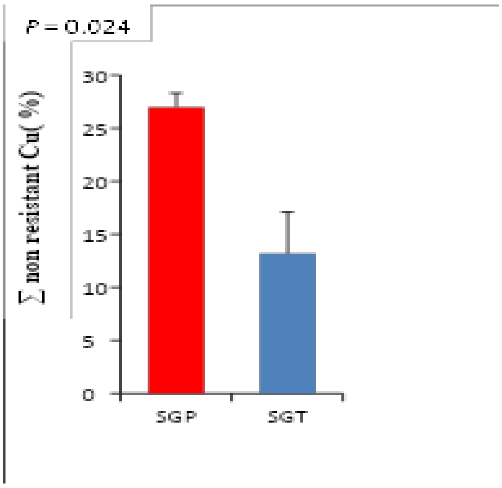
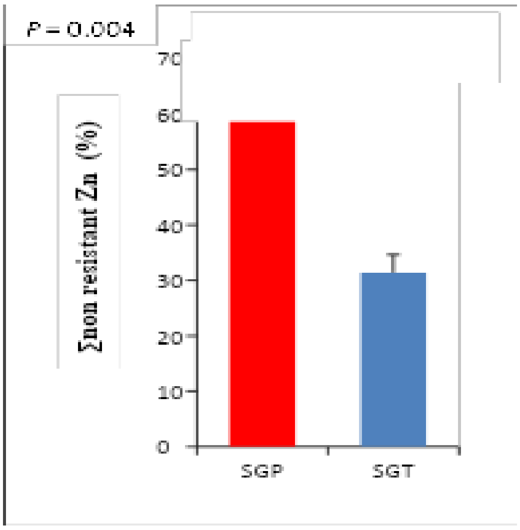


Figure 2. Comparison of mean Zn concentration levels of non-resistant fractions (%) between SGP and SGT using paired *t-test*



Geochemical fraction analyses of Cu and Zn in the mangrove sediments, as shown in tables II and III, depicted that the levels of Cu and Zn in the non-resistant fraction of sediment from Sungai Puloh are more consistent that of Sungai Tengi. It ranged from 24.4 to 29.8% for Cu and 59.8 to 65.9% for Zn. A high percentage of Cu and Zn is from the acid-reducible and oxidisable-organic fractions, especially samples from Sungai Puloh. This supports the conclusion of Ismail et al. (2004) that a high percentage of non-resistant mainly contributed by anthropogenic sources. Also, Yap (2003) observed that non – resistant fractions are considered as input from polluted waters. Therefore, very high percentage levels of Zn in the non-resistant fraction in Sungai Puloh is indicative that there is imminent anthropogenic pollution. When the mean of non- resistant percentage levels was compared using the paired *t-test* (Fig 1 and 2), Cu and Zn show significantly higher levels in samples from Sungai Puloh than Sungai Tengi ($p<0.05$), respectively. This could possibly be a result of industrial effluent that empties into Sg Pulloh.

The binding capacity of heavy metals towards organic matters was different in the sum of % non-resistant fractions of Cu, which showed a stronger positive Pearson correlation of 0.266($p>0.05$) than with that of Zn (0.225($p>0.05$) for SGT. This is also similar to SGP, where total organic matter showed a stronger relationship with Cu (0.682, $P > 0.05$) than that of Zn (0.374, $P> 0.05$). This supports the reports of some authors (Mat & Maah, 1994; Badri and Aston, 1983; Tessier et al., 1979) that suggested that there is a major form of Cu-organic complexes in sediment. Furthermore, the oxidizable-organic fractions represent a greater percentage of non – resistant fractions. High organic carbon content (Mac-Farlane et al., 2003), acid volatile sulfides (Chapman et al., 1998; SAB, 1995), and high salinity (Greger, 2004) affect heavy metal accumulation in the mangrove ecosystem. The positive relationship between the total organic matter and non- resistant fractions (which is mainly anthropogenic) suggests that man is constantly contaminating the mangrove ecosystem through his everyday activities.

Figure 3. Comparison of TOM% and Non-Resistant fraction% of Cu and Zn in SGP.

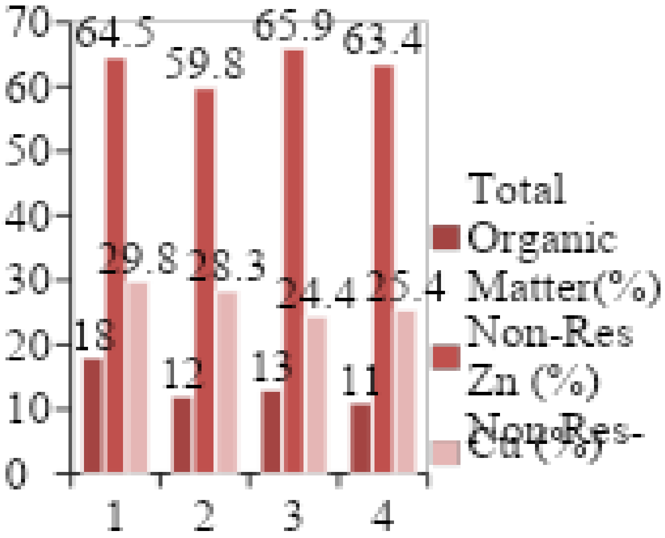


Figure 4. Comparison of TOM% and Non-Resistant fraction% of Cu and Zn in SGT

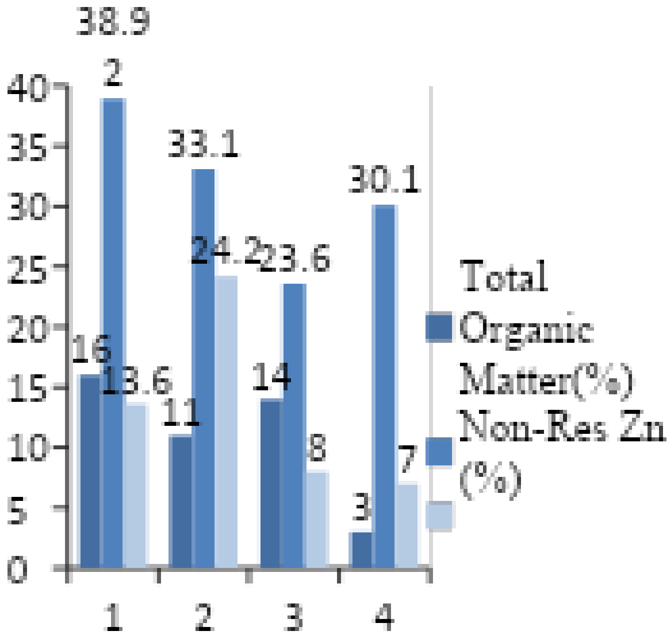


Table IV. Comparison of this study with the distributions of heavy metals Copper and Zinc (µg/g) in mangrove sediments of different countries based on published literature (G.R.MacFarlane et al., 2007).

Location	**Cu**	**Zn**	**Authors**
Ting Kok, Hong Kong	13	55	Chen, et al. (2003)
Western Australia	16	34	Alongi et al.(2003)
Shenzhen, China	36	106	Peng et al. (1997)
Karachi, Pakistan	12--56	35 -67	Zahir et al.(2004)
SE Australia	1-102	10-287	MacFarlene et al. (2003)
N.Australia	25-143	46-210	Seanger et al. (1990)
India	303	764	Thomas and Fernandez (1997)
Mai Po, Hong Kong	41.9-49.8	227.2-321.2	Ong Che, (1999)
Deep Bay, Hong Kong	80	240	Tam and Wong (2000)
Brisbane River, Australia	3.1-30.2	40.8-144	Mackey et al. (1992)
S.Buloh, Singapore	7.06	51.24	Dang et al. (2005)
S.Khatib, Singapore	32	120	Dang et al. (2005)
West Peninsular, Malaysia	48.83-85.49	279.51-321.55	This study
	8.3-35.8	99.75-1138.38	This study

CONCLUSION

This study provides clear evidence that mangrove intertidal sediments in Sungai Puloh and Sungai Tengi act as effective sinks for heavy metals, particularly copper (Cu) and zinc (Zn), with contamination largely influenced by anthropogenic activities. The elevated concentrations of Cu in Sungai Puloh and Zn in Sungai Tengi reflect site-specific pollution sources, including industrial effluents, maritime activities, domestic waste disposal, and agricultural runoff. The association between total organic matter and metal distribution highlights the critical role of organic-rich sediments in regulating metal sequestration and potential remobilization under changing environmental conditions. This dual function positions mangrove sediments as both sinks and latent sources of contamination, with implications for sediment–water exchange and trophic transfer. Overall, the findings emphasize that increasing human activities along coastal zones are contributing to the progressive accumulation of heavy metals in mangrove ecosystems. Given the ecological importance of mangroves and their role in supporting biodiversity and coastal livelihoods, several management and policy measures are recommended to minimize heavy metal contamination in the mangrove ecosystems of Sungai Puloh and Sungai Tengi. Regulatory agencies should strengthen monitoring and enforcement of industrial and domestic waste discharge into adjacent coastal waters, particularly in areas surrounding Sungai Puloh where anthropogenic activities are more intense. Industries, aquaculture facilities, and maritime operators should be required to adopt effective waste-treatment and pollution-control measures before effluent release. Furthermore, future research should integrate ecological risk assessment, trophic transfer studies, and human health evaluations to provide a more comprehensive understanding of the long-term consequences of heavy metal contamination in mangrove environments.

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