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ASSESSMENT OF HEAVY METAL CONCENTRATION IN MARINE SEDIMENTS IN TERENGGANU WATERS

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Abstract: The distribution of heavy metals (Cu, Zn, Cd, Pb) in surface sediments was examined in waters off the coast of Marang, Terengganu. A total of 20 samples were collected using Ponar grab and analysed by inductively coupled plasma-mass spectrometer after closed digestion with acid. The sediments were filtered using a dry sieving method to determine their particle size. The spatial distribution maps on the concentration of selected metals were drawn using the ArcGIS software. Results showed that the average concentration of Cu, Zn, Cd, and Pb were 2.33±0.38 µg/g dry weight, 28.4±3.78 µg/g dry weight, 0.09±0.01 µg/g dry weight and 8.35±1.48 µg/g dry weight, respectively. The level of pollution was also evaluated using the Index of Geoaccumulation (I_{geo}) and Pollution Load Index (PLI). All I_{geo} and PLI values obtained were low, which indicated low or no pollution. Meanwhile, the sediment mean size ranged between -0.77Ø and 3.18Ø, which characterised a sandy type of sediment. Correlation analysis showed a positive correlation between the heavy metals and sediment size. The results indicated that there was a common source of heavy metal pollution in the study area, possibly from shipping activities. Overall, there was no significant heavy metal pollution in the waters off Marang. This finding is important as the data could be used to evaluate the risk of metal contamination and the impact of anthropogenic activities on the marine environment.

Keywords: Heavy metals, sediment, index of geoaccumulation, pollution load index, South China Sea

Introduction

Heavy metal accumulation in river sediments caused by human activities, such as mining and industrial waste discharge, is common in developing countries (Islam *et al.,* 2015), including Malaysia. Nowadays, rivers have become a "sink" for heavy metal accumulation, where their sediments may retain those metals and cause toxicity in the aquatic environment (Li *et al.,* 2013). Heavy metal pollution is also becoming common (Iwuoha *et al.,* 2012) and is among the serious global environmental concerns because they are toxic, non-biodegradable, widespread and accumulative in organisms (Ong *et al.,* 2015; Fu & Wang, 2011). The metals in the water column may be derived from two sources — natural and anthropogenic (Dalman *et al.,* 2006). According to Ghrefat and Yusuf (2006), the weathering of rocks and volcanic activities are examples of natural sources that may introduced heavy metals into the aquatic system. Meanwhile, most of the anthropogenic sources come from human activities, such as agricultural runoff, untreated sewage, aquaculture, mining and industrial waste (Kamaruzzaman *et al.,* 2010).

Marine sediments mostly comprise aluminosilicate minerals and originate from the weathering of rocks and soil erosion (Esen *et al.,* 2010). Heavy metal pollution in the seawater is strongly associated with marine sediments (Horowitz, 1991). The study

of marine sediments is vital to assess such pollution as the sediments have longer residence time in the ocean environment (Lin *et al.,* 2008). When the dissolved heavy metals from natural or anthropogenic sources mix with sea water, they are removed from the water column by adsorbing to marine sediments at a very fast rate (Esen *et al.,* 2010). The adsorption rate of heavy metals will increase with decreasing grain size of the sediments (Esen *et al.,* 2010). Therefore, fine marine sediments will contain higher concentrations of heavy metals compared to those with larger grain size due to the high specific surface adsorption of the smaller particles (Zhang *et al.,* 2007). Thus, heavy metal concentration is significantly higher in fine-grained sediments that are rich in clay minerals (Abrahim *et al.,* 2007), especially surficial sediments that may act as a "pool" or "sink" for such metals (Wang *et al.,* 2008). However, heavy metals in marine sediments will undergo remobilisation as the chemical environment changes (Zoumis *et al.,* 2001). For example, a drop in pH valuedue to ocean acidification will cause the remobilisation of the heavy metals (Yuan *et al.,* 2004).

Marine sediments are useful indicators in the assessment of heavy metal pollution (Nowrouzi *et al.,* 2014). Some metals such as copper (Cu) and zinc (Zn) are essential to the metabolism and growth of living organisms at low concentration, while cadmium (Cd) and plumbum (Pb) are extremely toxic even at low quantities (Jian-Guo *et al.,* 2010; Peng *et al.,* 2008). Therefore, the objectives of this study are to determine the concentration of selected heavy metals, namely Cu, Zn, Cd, Pb in the ocean waters off the coastal district of Marang in the state of Terengganu, Malaysia, to assess the level of pollution in the surficial sediments using the Geoaccumulation Index (I_{geo}) and pollution load index (PLI).

Materials and Methods

Marang town is located in a coastal district with the same name in the state of Terengganu on the east coast of Peninsular Malaysia. The district is adjacent to the South China Sea and the main economic activities are fishing and tourism. The resort island of Pulau Kapas is located off the district's coast, and shipping traffic is quite heavy between the island and mainland.

Sampling was carried out aboard the *UMT Discovery VII*, a research vessel belonging to Universiti Malaysia Terengganu, on September 21, 2018. Sediment samples were collected using a Ponar grab at 20 stations shown in the map of the study area in Figure 1. The outermost layer of the sediment samples was removed to avoid metal contamination from the wall of the grab, and the inner part was placed in plastic containers. The plastic containers were kept in an ice chest under low temperature (Niu *et al.,* 2009; Kamaruzzaman *et al.,* 2010; 2011). The coordinates of each station were recorded using the Global Positioning System (GPS).

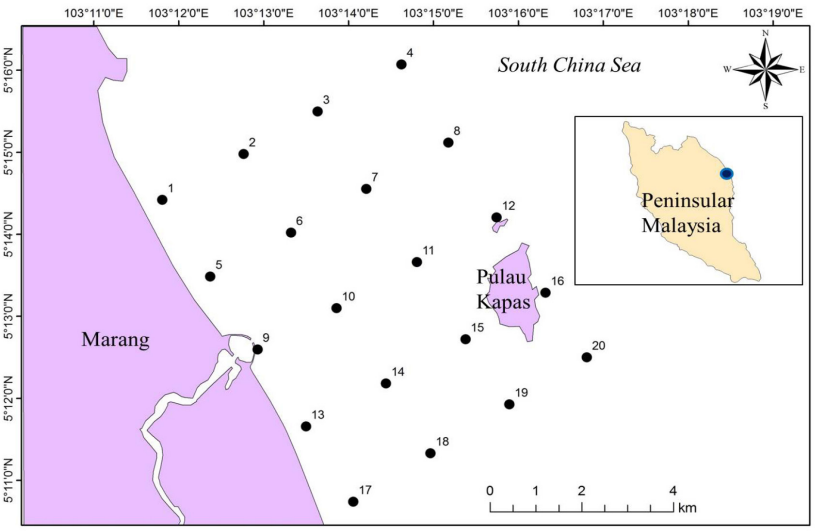

Figure 1: Map of sampling stations located in the study area off the coast of Marang

All glass and plastic apparatus were soaked in 10 % HNO_3 overnight and rinsed with Mili-Q water (Merck KGaA, Darmstadt, Germany) before use (Ong *et al.,* 2013). The Teflon beakers used for sample digestion were boiled in 65 % for one hour to prevent contamination. After boiling, the beakers were dried in an oven at 80°C for one hour.

The sediment samples were defrosted at room temperature and dried in an oven at 60°C until the weight became constant. The dried samples were ground using mortar and pestle (Gargouri *et al.,* 2011) before being homogenised and stored in clean polypropylene containers.

The sediment samples were first processed using the "Teflon Bomb" digestion method and total heavy metal content was determined using inductively coupled plasma-mass spectrometry (ICP-MS) as described by Ong *et al.* (2013; 2015). Briefly, 50 mg of the homogenised samples with grain size of less than 63µm was mixed with 1.5 mL of concentrated hydrofluoric acid (HF), and HCl in sealed Teflon vessels. The Teflon vessels were heated at 100°C for eight hours. After cooling at room temperature, the solution in the vessels was transferred into a polypropylene tube and diluted to 10 mL with deionised water. The digested and diluted samples were then loaded into the ICP-MS system to determine the concentration of Cu, Zn, Cd, and Pb. The Standard Reference Material 1646a Estuarine Sediment was used for the recovery test.

To analyse the sediment particle size, 200 g of samples were allocated for the dry sieving technique. A sieve shaker with mesh sizes ranging from 63 µm to 4,000 µm, was used to filter the samples. After 10 minutes, the remaining sediment samples trapped at each mesh were weighed. The GRADISTAT particle size analysis software (Kenneth Pye Associates Ltd, Wokingham, UK) was used to determine the characteristics of the sediments.

Results and Discussion

The Standard Reference Material 1646a Estuarine Sediment was used to assess the accuracy of the experiment. The recovery percentages of Cu, Zn, Cd, Pb are shown in Table 1. Cd seemed to have the highest recovery percentage at 101.35 %, while Zn had the lowest percentage at 88.96 %.

Table 1: Results of the recovery test for Cu, Zn, Cd, and Pb

Heavy metal	Measured values (µg/g dry weight)	Certified values (µg/g dry weight)	Recovery percentage (%)
Cu	9.28 ± 0.71	10.01 ± 0.34	92.7
Zn	43.5 ± 2.1	48.9 ± 1.6	89.0
Cd	0.15 ± 0.01	0.148 ± 0.007	101
Pb	11.2 ± 1.9	11.7 ± 1.2	95.7

The distribution patterns of the heavy metals in the study area are shown in Figure 2. The concentrations consisted of varied ranges, namely 0.77–5.96 µg/g dry weight for Cu, 11.6–76.1 µg/g dry weight for Zn, 0.03–0.19 µg/g dry weight for Cd and 2.34–22.5 µg/g dry weight for Pb. The average concentrations of Cu, Zn, Cd, and Pb were 2.33 ± 0.38 µg/g dry weight, 28.43 ± 3.78 µg/g dry weight, 0.09 ± 0.01 µg/g dry weight and 8.35 ± 1.48 µg/g dry weight, respectively.

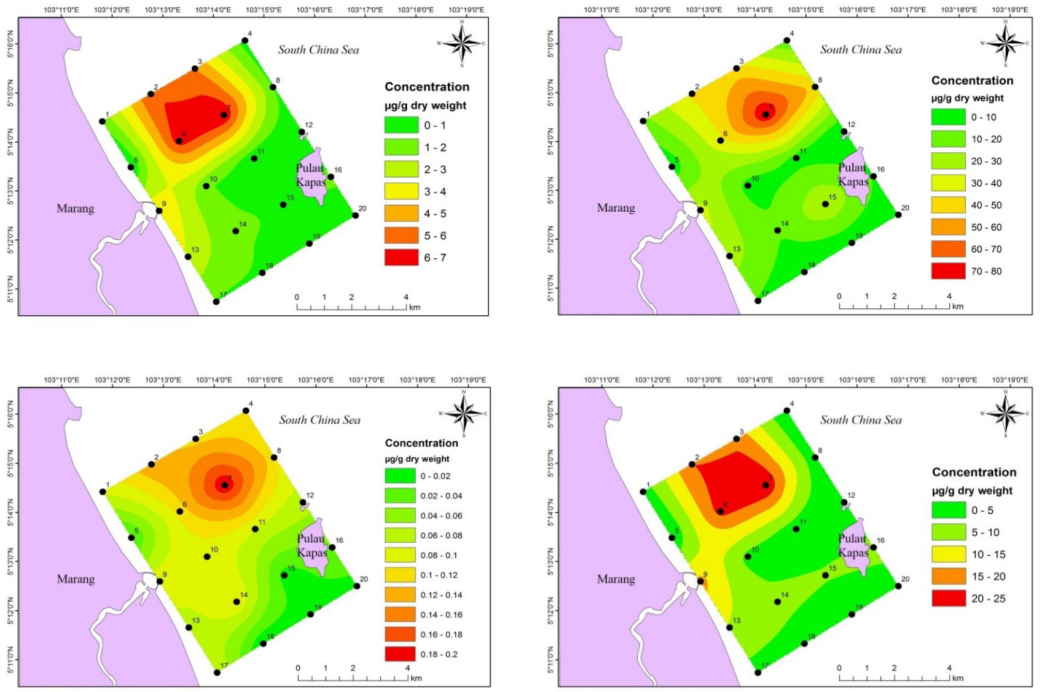

Figure 2: The distribution of Cu, Zn, Cd and Pb concentrations in the study area

Based on Figure 2, the distribution of heavy metals was mainly concentrated at Station 7. The concentrations of Cu, Zn, Cd, and Pb were the highest in that particular station. Besides that, there was also a "trail" of high heavy metal concentration that could be seen clearly from the estuary of Marang River to Station 7. This indicated s that the Marang River could probably be the source of the heavy metal contamination that came from untreated industrial and domestic waste discharged upstream of the river (Chen *et al.,* 2007). Kishe and Machiwa (2003) and Muniz *et al.* (2004), in their findings, also stated that heavy metal pollution in the marine environment mainly came from the discharge of untreated industrial and domestic waste.

Moreover, shipping was also one of the sources of heavy metal pollution. Ship traffic in the study area could be considered heavy due to fishing and tourist activities around Pulau Kapas. The antifouling paint on boats and ships used to prevent the growth of organisms like barnacles and algae contained high amounts of Cu and Zn (Bothner *et al.,* 1998; Orlić & Tang, 1999). Therefore, the elevated concentration of heavy metals in the study samples could also be caused by the washout of antifouling paints from boats and ships (Guerra-García & García-Gómez, 2005). The use of cheap leaded fuel in outboard engines was also a main contributor of Pb pollution in the study area (Nasr *et al.,* 2006).

Although the Marang River might be the main source of heavy metal pollution, Station 9 and its surrounding areas — which were directly adjacent the estuary — did not record the highest concentration of such metals. This observation could be explained by Boström *et al.* (1974), which stated that heavy metals in the marine environment could be transported by prevailing currents. Therefore, the heavy metals from Marang River — and Pulau Kapas to some extent — were probably carried away by the sea current and concentrated on the sediments in Station 7 and its surrounding areas (Morillo *et al.,* 2004).

Figure 3 shows the average percentage of clay, silt and sand in the sediment samples. Due to the uniqueness of the estuary ecosystem, fine sediments, such as silt and clay, would usually be trapped in the area and rarely washed out to sea (Duck & Wewetzer, 2001). However, this was not observed at the Marang River.

Figure 4 shows that clay and silt were mostly found at Station 6 and Station 7, which were quite far from the estuary. This could be attributed to the sea current and tidal waves, which were influenced by the monsoon. The sea surface circulation at the study area was known to vary according to monsoon (Akhir *et al.,* 2014).

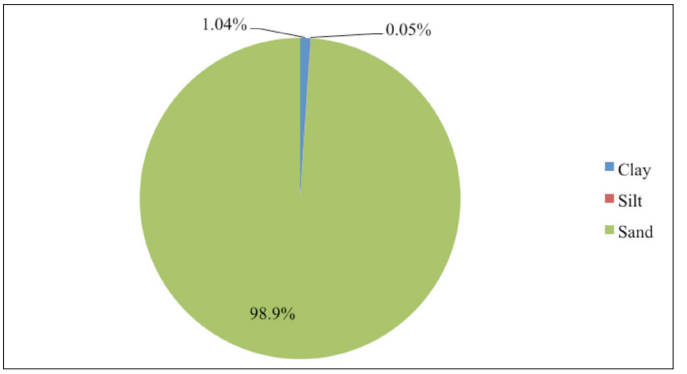

Figure 3: Average percentage of clay, silt and sand in the study area

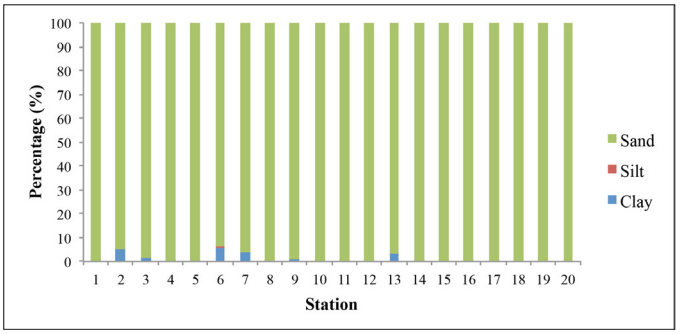

Figure 4: Percentage of sand, silt and clay from sediment samples obtained at each station

According to Akhir and Yong (2011), the sea current would flow northward along Peninsular Malaysia during the southwest monsoon, which occurred from May to September. This was due to prevailing south westerly winds generated in the South China Sea (Raj, 1982). Since the samplings in Station 6 and 7 were conducted in October 2018, it was possible that the sediments had been carried by the northward current from the estuary to Station 6 and Station 7. Moreover, the time taken for sediments with smaller grain sizes, such as clay and silt, to sink into the ocean was very long (George *et al.,* 2007). Hence, the fine sediments would only sink entirely into the seabed after travelling some distance.

In Figure 5, all the selected metals showed positive correlation with the mean sediment size. Pb had the highest correlation (r=0.857), followed by Cu (r=0.846), Zn (r=0.575) and Cd (r=0.406). The positive correlation indicated that the concentration of heavy metals was higher in fine sediments than coarse ones. This was in line with the theory of smaller grain size adsorbing a higher concentration of heavy metals due to a larger surface area to volume ratio (Martincic *et al.,* 1990).

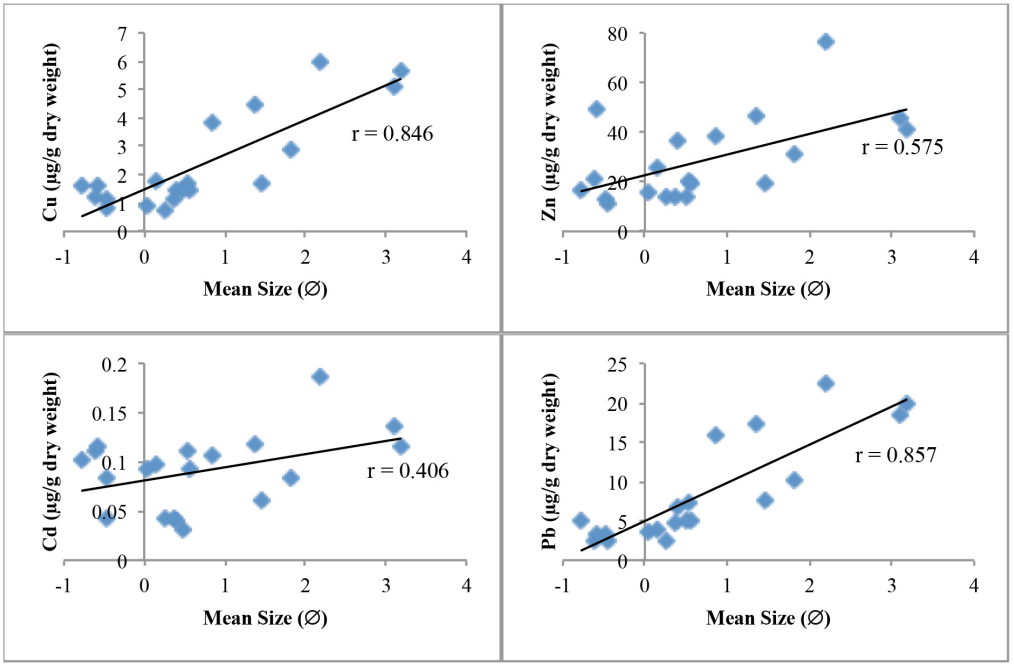

Figure 5: Correlation between selected metals (Cu, Zn, Cd, Pb) and sediment mean size

Table 2 shows the correlation between the heavy metals in the study area. All the metals showed a positive correlation with each other, which indicated that the natural and anthropogenic sources were similar (Amin *et al.,* 2009). Among the metals, Cu had a stronger relationship with Pb and Zn. This showed that Cu, Pb and Zn in the study area likely came from the same sources, such as antifouling paints and leaded fuel used in ships and outboard engines. The metals might also have come from benthic organisms that had similar uptake and release mechanisms in the sediments (Broman *et al.,* 1991).

Table 2: Correlation between heavy metals concentration

	Cu	Zn	Cd
Zn	0.825	1	
Cd	0.701	0.749	1
Pb	0.983	0.798	0.646

I_{geo} was introduced by Muller (1969) to describe heavy metal contamination in soil sediments. The formula used to calculate I_{geo} is stated in Equation 1 (Eq. 1).

$$I_{geo} = \log_2[\frac{C_n}{1.5B_n}] \qquad \text{(Eq.1)}$$

where C_n is the measured concentration of metal n in the sediment, while B_n is the background value for metal n. The background values used were referred to the concentration of metals in the Upper Continental Crust reported by Wedepohl (1995). The calculated I_{geo} value was used to determine the level of pollution at the sampling sites. There were seven classes of pollution as shown in Table 3 (Muller, 1969; Nowrouzi & Pourkhabbaz, 2014).

Table 3: Pollution categories based on the Geoaccumulation Index (I_{geo})

Class	Value	Classification
0	<0	Unpolluted
1	0 – 1	Unpolluted to moderately polluted
2	1 – 2	Moderately polluted
3	2 – 3	Moderate to strongly polluted
4	3 – 4	Strongly polluted
5	4 – 5	Strong to extremely polluted
6	>5	Extremely polluted

The I_{geo} values of the selected metals in this study are shown in the interpolation maps of the values in Figure 6. Based on Table 3, the sediment at Station 7 could be categorized as unpolluted to moderately polluted with Cd as the I_{geo} value was 0.286 (Class 1). The sediments from other stations were considered unpolluted with Cd since the I_{geo} values were less than 0 (Class 0). Furthermore, all stations were also considered unpolluted with Cu, Zn and Pb since the I_{geo} values were less than 0.

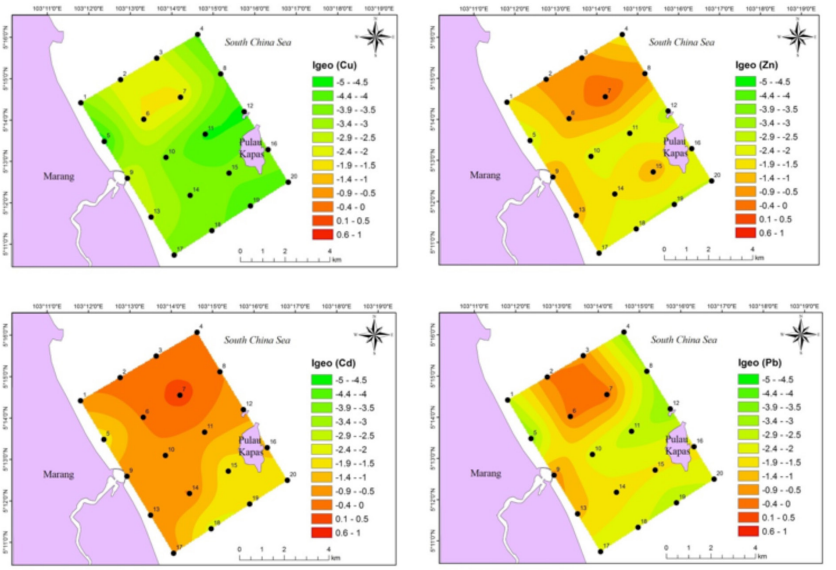

Figure 6: Interpolation maps of values of Cu, Zn, Cd and Pb at each station

The PLI proposed by Tomlinson *et al.* (1980) in Equation 2 (Eq. 2) is used to calculate its value:

$$PLI = (CF_1 \text{ x } CF_2 \text{ x } CF_3 \text{ x } \ldots\ \ldots\ \ldots \text{ x } CF_n)^{\frac{1}{n}} \quad \text{(Eq. 2)}$$

where n is the number of metals. The contamination factor (CF) is defined in Equation 3 (Eq. 3).

$$\text{CF} = \frac{\textit{Metal concentration in the sediments}}{\textit{Background value of the metal}} \quad \text{(Eq. 3)}$$

The background value of the metals was also referred to Wedepohl (1995). PLI value > 1 was considered polluted, while PLI value of < 1 was considered no pollution (Tomlinson *et al.,* 1980; Harikumar *et al.,* 2009). Lower PLI values indicated less pollution from anthropogenic sources (Chakravarty & Patgiri, 2009).

Figure 8 shows the PLI values at each station using their interpolation maps. Among the stations, only Station 7 was considered polluted with heavy metals because its value was more than 1 (1.13).

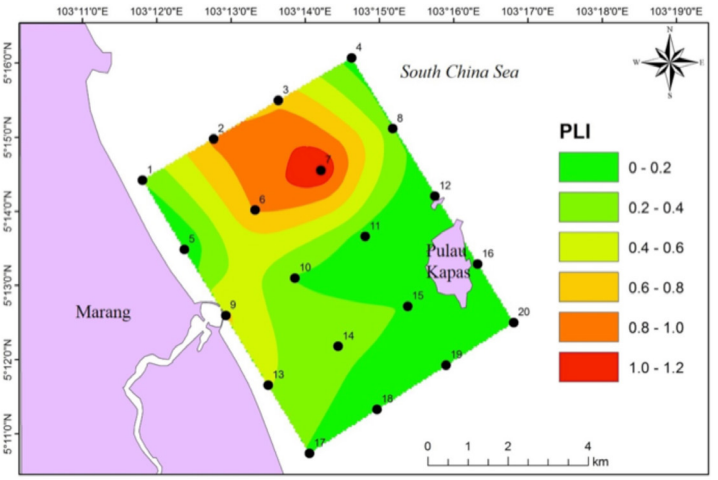

Figure 8: Interpolation map of PLI values at each station

Conclusion

Generally, marine sediments in the waters off Marang could be considered unpolluted. This was because all heavy metal concentrations were below their background values. However, only Cd in Station 7 was classified as Class 1, which indicated an unpolluted to moderately polluted level based on the I_{geo} value obtained in this study. Furthermore, the PLI values at all stations, except Station 7, were less than 1, which indicated no pollution. Although this study found no significant pollution of heavy metals in the waters off Marang, the data could be used to enhance the safety of fishing products, where local fishermen could be cautioned against fishing in areas where high concentration of the metals were detected.

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