

## Behavior of $^{210}\text{Po}$ and $^{210}\text{Pb}$ in Shallow Water Region of Mersing Estuary, Johor, Malaysia

Noor Affizah Bujang Saili and Che Abd Rahim Mohamed

*School of Environmental and Natural Resource Sciences, Faculty of Science and Technology,  
Universiti Kebangsaan Malaysia, Bangi Selangor 43600, Malaysia*

### Abstract

$^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities were determined in dissolved and particulate phases in order to understand the behavior of both natural radionuclides in shallow water regions such as Mersing Estuary. Strong statistical correlations between the distribution coefficient values of polonium and lead in dissolved phases with SPM proved that the natural nuclides of polonium and lead have a high affinity to suspended particle materials in the water column, where the SPM acts as a carrier to transport and remove natural isotopes of polonium and lead from their geochemical behavior. However a low statistical correlation ( $r=0.414$ ) found between chlorophyll-a with an activity ratio of  $^{210}\text{Po}/^{210}\text{Pb}$  and SPM implies that the enrichment of  $^{210}\text{Po}$  was not associated with the abundance of chlorophyll-a. But a strong correlation between soluble reactive phosphorus (SRP) and  $^{210}\text{Po}$  in the dissolved phase proved that the contribution of phosphate element as catalysis increasing the activity levels of  $^{210}\text{Po}$  at coastal waters.

**Keywords:**  $^{210}\text{Po}$ ;  $^{210}\text{Pb}$ ; total suspended particulate matter; shallow water region

### 1. Introduction

The activities of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in the marine environment depend on their chemical specification, reaction with surface mineral, dissolved or suspended organic matter and colloids in the water column (Chabaux *et al.*, 2008).  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  mobility in the water column are accomplished by an aggregation of colloidal material and adsorption onto particulate matter (Smoak *et al.*, 1996). On the other hand,  $^{210}\text{Po}$  is more of a biological removal and is actively involved in the biogeochemical cycle compared to  $^{210}\text{Pb}$  (Tsunogai and Nozaki, 1971). The removal of  $^{210}\text{Pb}$  is probably by the scavenging of particulate matter from the sea surface into the deep sea and absorption via sediment-water interface interaction (Nozaki *et al.*, 1997).

According to Balls (1988), the distribution of metal concentration in the water column depends on the amount of total suspended particulate matter (SPM) loading, where a major fraction of nuclides exists in particulate form at high concentrations level of SPM (Baskaran and Santschi, 1993) and about 99% of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are distributed in the particulate phase, especially in the shallow water region of Kuala Selangor (Theng and Mohamed, 2005). However, Wei *et al.* (2012) also found that the distribution coefficients of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are correlated with the particle content in turbid waters except at the offshore region. Therefore, in order to understand  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  behavior in the shallow water region, it is very

important to quantify their activities to investigate the interaction between these natural radionuclides with the large amount of particles in the water column which are affected by physical processes (e.g., Smoak *et al.*, 1996; Hong *et al.*, 1999).

Mersing River was selected as a study area as its water depth is less than 10 m, and it is a small catchment area of about 232 km<sup>2</sup> toward the South China Sea. It regularly undergoes the mixed tide phenomenon and is dominated by a semi-diurnal tide with a wide range of salinity from 1.38 to 32.98 psu. According to Mohd Ekhwan *et al.* (2011) in an analysis between 2005 until 2009, the sedimentation rate in Mersing River is very high (approximately 3.26 tons/ha/yr). Various human activities especially palm oil agriculture, fisheries landing and development expose the river to a very high level of SPM loading which potentially influences the natural radionuclides in the ecosystem. Thus, the objective of this study is to study the relationship between  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  with SPM in the water column in this shallow water region.

### 2. Methodology

#### 2.1. Sample Collection

Samples were conducted on 6 February, 5 April, 4 July and 9 October 2010, along the Mersing River toward the southern South China Sea. About 20l of water samples were collected using the Niskin water

Table 1: Coordinates and description of study area at Mersing River

| Area      | Station | Latitude (N) | Longitude (E) | Description                                      |
|-----------|---------|--------------|---------------|--|
| Riverine  | 1       | 02° 25.062'  | 103° 48.436'  | Next to bridge and road                          |
|           | 2       | 02° 25.348'  | 103° 48.694'  | Nymph area                                       |
|           | 3       | 02° 25.579'  | 103° 48.996'  | Nymph area                                       |
|           | 4       | 02° 25.558'  | 103° 49.559'  | Point of 2 rivers and close to palm oil estate   |
| Estuarine | 5       | 02° 25.604'  | 103° 50.002'  | Jetty for small boats landed                     |
|           | 6       | 02° 26.149'  | 103° 50.396'  | Jetty for fish landed and tourism transportation |
|           | 7       | 02° 26.690'  | 103° 50.962'  | Toward the sea                                   |
|           | 8       | 02° 27.189'  | 103° 51.404'  | Toward the sea                                   |
|           | 9       | 02° 28.149'  | 103° 52.419'  | Toward the sea                                   |

sampler from nine selected stations (Table 1 and Fig. 1). In the laboratory, the water samples were filtered immediately through a pre-weighed membrane filter paper (0.45 µm pore size, 47 mm diameter) for collect total suspended particulate matter (SPM) and dissolved phase for further analyses such as chlorophyll-a, phosphate and natural radionuclides. After that the dissolved phase acidified with concentrated HNO<sub>3</sub> (< pH2). The <sup>209</sup>Po tracer, Fe-carrier and Pb-carrier were then spiked into the samples for further analyses.

2.2. Chemical Analysis

<sup>210</sup>Po and <sup>210</sup>Pb were analyzed through a sequential extraction. The <sup>210</sup>Po extraction from the water samples was well described by Theng and Mohamed (2005) including sample preparation, co-precipitation by Fe-carrier, <sup>210</sup>Po spontaneous plating and counting using Alpha Analyst Spectroscopy system with a silicon-surface barrier detector by Canberra, Inc., with Apex-Alpha software. After <sup>210</sup>Po spontaneous plating, the

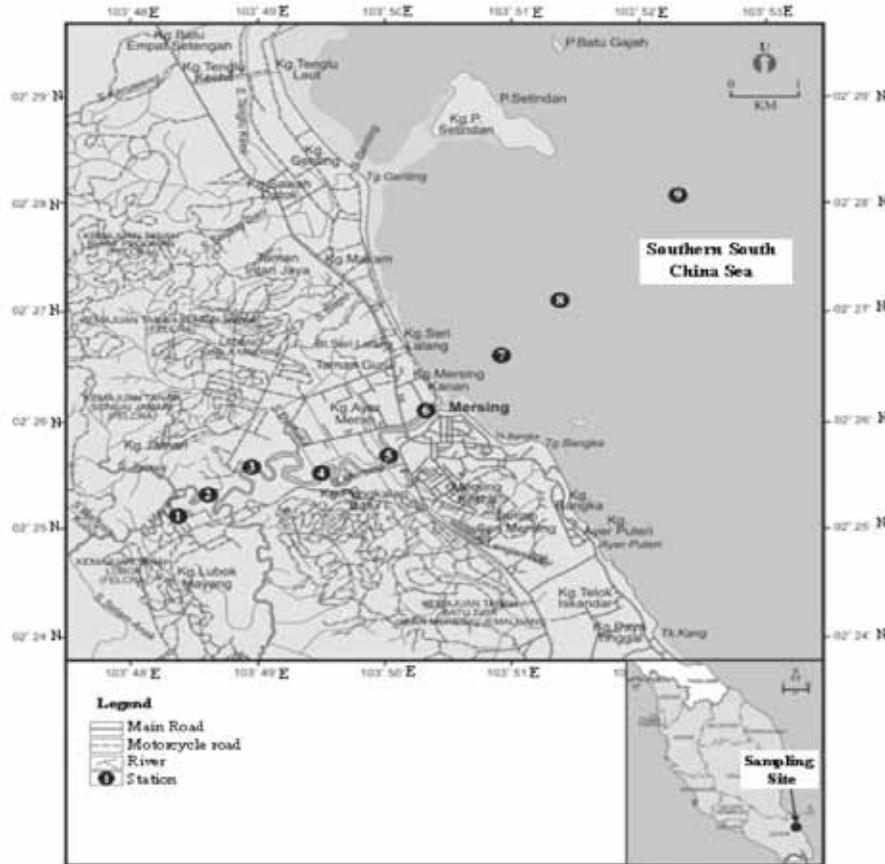


Figure 1. Map of sampling stations along the Mersing river toward the South China sea

solution proceeded to  $^{210}\text{Pb}$  extraction using electro-analytical analysis as described by Sabuti and Mohamed (2010). Finally, lead was separated from the samples as a lead sulfate co-precipitate (Blanco *et al.*, 2004; Kim *et al.*, 2001) and counted using gross alpha-beta spectrometry by Tennelec S5 XLB with Eclipse software after an equilibrium with their daughter.

Similar to suspended particulate matter; the  $^{209}\text{Po}$  tracer and Pb-carrier were spiked into the dried sample for sequential extraction. Concentrated  $\text{HNO}_3$ ,  $\text{HCl}$ ,  $\text{HClO}_4$  and  $\text{HF}$  at a 3:2:2:1 ratio was mixed with the sample in a Teflon beaker for total digestion.  $\text{HClO}_4$  was added and heated until dryness if the sample was not totally digested yet. After dryness, the fully digested residue was re-dissolved in 0.5 M  $\text{HCl}$  and ascorbic acid for  $^{210}\text{Po}$  spontaneous plating. The  $^{210}\text{Pb}$  extraction was proceed sequentially after  $^{210}\text{Po}$  spontaneous plating as described by Sabuti and Mohamed (2010).

Overall, the internal chemical yields were typically 60 to 90%. The reference material, NIST SRM-4357 (Ocean Sediment) was used to verify the performance of the analytical procedure. The obtained recovery value was 88 to 94%.

Soluble reactive phosphorus (SRP) was directly measured using a UV-spectrophotometer after filtration. The phosphor-molybdate complex formed after 8 minutes reacted with a mixture reagent of ammonium molybdate,  $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$  + sulphuric acid  $\text{H}_2\text{SO}_4$  + potassium antimonyl ttrate,  $\text{K}(\text{SbO})\text{C}_4\text{H}_4\text{O}_6\cdot 0.5\text{H}_2\text{O}$  to ascorbic acid,  $\text{C}_6\text{H}_8\text{O}_6$  at 3:1 ratio (Huang and Zhang, 2009).

### 3. Results and Discussion

#### 3.1. Variation of Total Suspended Particulate Matter (SPM)

The total suspended particulate matter (SPM) in Mersing River varied greatly between 4 to 175 mg/L (mean= $22.43\pm 15.71$  mg/L). Statistically, there was no significant difference of SPM ( $p=0.056$ ) concentrations among the nine selected stations. However, there was a significant variation ( $p=0.000$ ) between the different sampling times. The highest peak of SPM was found during the first sampling (6 February 2010) which was during the northeast monsoon. Strong winds from the southern South China Sea toward the eastern Peninsular of Malaysia might trigger huge waves that break at the coastal region. This phenomenon produces turbulence which might interrupts the water-sediment interface in shallow regions and cause resuspension. According to (Baskaran and Santschi, 1993), sediment resuspension in estuarine water significantly influences the  $^{210}\text{Pb}$  activity with respect to SPM concentrations. Therefore,

the variation of SPM loading in the water column might affect the variation of natural radioactivity such as  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in dissolved and particulate phase. It seems to be a common trait in other shallow coastal water regions (Baskaran and Santschi, 1993; Carvalho, 1995; Hong *et al.*, 1999; Theng and Mohamed, 2005).

In this study, the variation of  $^{210}\text{Po}$  ( $r=0.805$ ) and  $^{210}\text{Pb}$  ( $r=0.679$ ) activity in the dissolved phase is significantly associated to SPM concentrations. Fig. 2 proves that the SPM concentration in Mersing River plays an important role in the distribution of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity and mobility.  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the dissolved phase varied from 0.76 to 16.12  $\text{Bq/m}^3$  and 0.16 to 10.56  $\text{Bq/m}^3$ , respectively. Meanwhile, the activity of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the particulate phase ranged from  $2.04\times 10^4$  to  $65.64\times 10^4$   $\text{Bq/m}^3$  and  $5.24\times 10^4$  to  $159.26\times 10^4$   $\text{Bq/m}^3$ , respectively. By comparison, the distribution of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the water column of Mersing River which is located at the east coast of Peninsular Malaysia toward the southern South China Sea was higher than that of the Kuala Selangor Estuary on the west coast toward the Strait of Malacca (Theng and Mohamed, 2005), even though the SPM concentration was much higher. This might be due to effective resuspension as a result of the monsoon effect which exposed the open water area compared to the closed passage of the western coast of Peninsular, which is sheltered by Sumatra Island. Besides that, the activities of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in the marine environment depend on their chemical specification, reaction with surface mineral, dissolved or suspended organic matter and colloids in the water column (Chabaux *et al.*, 2008).

Total  $^{210}\text{Po}$  activity significantly varied between stations at Mersing River ( $p=0.001$ ). However, there was no significant difference in total  $^{210}\text{Pb}$  activity ( $p=0.569$ ). This implies that the distribution concentration of  $^{210}\text{Pb}$  along the Mersing River was approximately constant compared to  $^{210}\text{Po}$ . The constant activity of  $^{210}\text{Pb}$  might emanate from the in-situ production of supported  $^{210}\text{Pb}$  parent decay in the sediment and its being dissolved in the water column during resuspension. Meanwhile,  $^{210}\text{Po}$  might contribute anthropogenically due to human activities such as palm oil agriculture, fish landing, boat traffic and domestic waste from terrestrial sources. In order to determine the accumulation of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the water column, the inventory of radionuclide of this study was calculated from the measured activity with a decay correction to the sampling date as follows:

$$I = A_o \times d \quad \text{-----} \quad (1)$$

Where  $I$  is the inventory of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the water column ( $\text{Bq/m}^2$ ),  $A_o$  is the activity concentration of radionuclide ( $\text{Bq/m}^3$ ) and  $d$  is the depth of the samples taken during sampling (m).

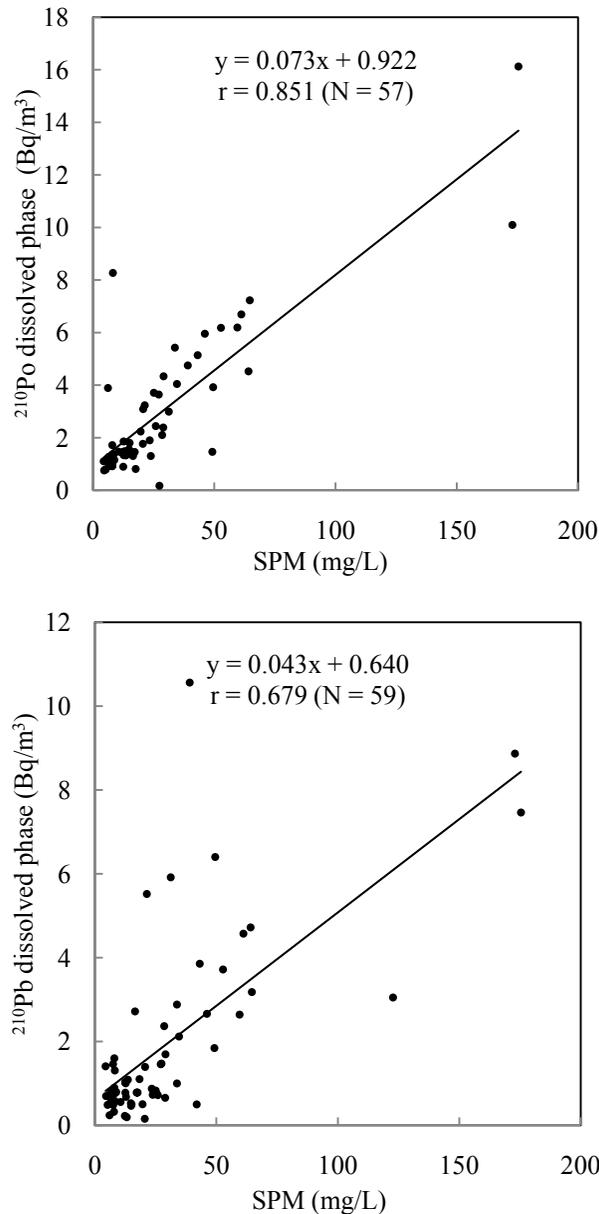


Figure 2. Correlation of <sup>210</sup>Po (Bq/m<sup>3</sup>) and <sup>210</sup>Pb (Bq/m<sup>3</sup>) in the dissolved phase with respect to SPM (mg/L).

Fig. 3 shows the mean inventory of <sup>210</sup>Po and <sup>210</sup>Pb in the dissolved and particulate phases with respect to the variation of mean SPM at the different times of sampling. <sup>210</sup>Po and <sup>210</sup>Pb in the dissolved phase pattern were comparable with the SPM variation which was significantly high during the northeast monsoon. According to Mohamed *et al.* (2006), <sup>210</sup>Po and <sup>210</sup>Pb fluxes in the water column at the southern South China Sea received large input sources from neighboring areas such as from Indo-China by atmospheric transport and the northern South China Sea and the western Pacific by ocean water circulation which were highest compared to other locations in the Straits of Malacca. This indicates that the northeast monsoon significantly plays a critical role in the influence of <sup>210</sup>Po and <sup>210</sup>Pb accumulation

with respect to SPM concentration in the Mersing River.

Meanwhile, peak activity of <sup>210</sup>Po and <sup>210</sup>Pb in the particulate phase was highlighted during the inter-monsoon (April) and southwest monsoon (July), respectively. This condition might be manipulated by the input of each radionuclide during sampling time. Particularly, the <sup>210</sup>Pb adheres to aerosol and is efficiently washed from the atmosphere by precipitation (Preiss *et al.*, 1996; Yamamoto *et al.*, 1998). It is important to establish that higher precipitation occurs during the southwest monsoon even though it is well known as the dry season. According to WMO (2010), 2010 was the one of the top 3 warmest years since the beginning of instrumental climate records in 1850. Moreover, the strong La Niña phenomenon in the tropical Pacific

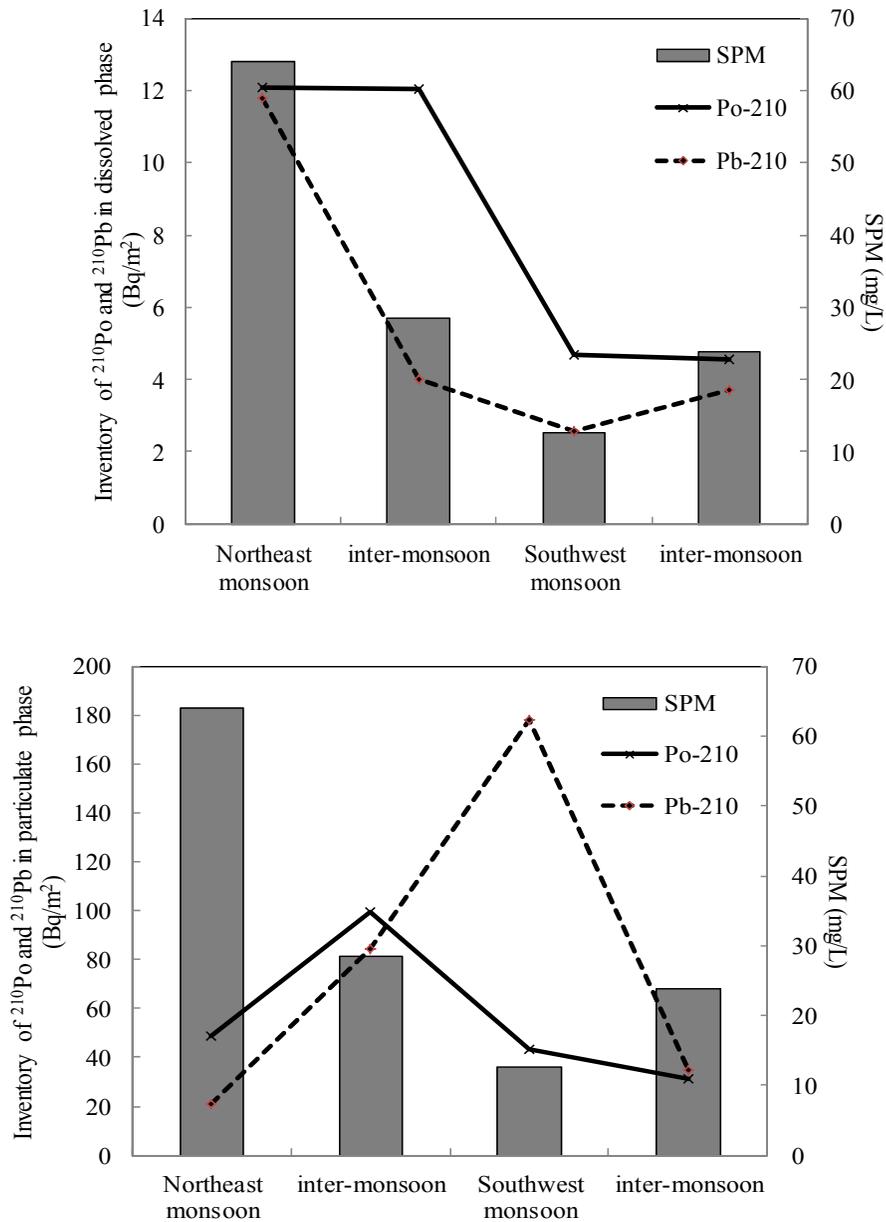


Figure 3. Mean inventory of  $^{210}\text{Po}$  (Bq/m<sup>2</sup>) and  $^{210}\text{Pb}$  (Bq/m<sup>2</sup>) in dissolved and particulate phases with respect to the variations of mean SPM (mg/L) at different sampling times.

in 2010 (WMO, 2011) significantly affected the distribution of rainfall during the seasonal monsoons. By comparison, the mean of rainfall during the northeast monsoon (February), inter-monsoon (April), southwest monsoon (July) and inter-monsoon (October) are 41.8 mm, 117.6 mm, 128.6 mm and 242.2 mm, respectively (MMD, 2010). This data shows that climate changes disrupted the normal global climate. Unfortunately, rainfall data has not yet proven the atmospheric effect on the  $^{210}\text{Pb}$  sources. Therefore, the consideration of in situ production of  $^{210}\text{Pb}$  from parent decay and SPM input via sedimentation and resuspension might play an important role in comparison to atmospheric fallout.

### 3.2. Behavior of $^{210}\text{Po}$ and $^{210}\text{Pb}$ in the water column

Partition coefficients,  $K_d$  parameter is usually utilized in order to further understanding on geochemical characteristics of elements in the aquatic environment (Kim and Yang, 2004; Theng and Mohamed, 2005; Wei *et al.*, 2012; Wei and Murray, 1994). According to (Abril and Fraga, 1996), the mathematical equation has been formulated to describe the behavior of selected contaminants based on physico-chemical aspects of dissolved and particulate phase interaction. In this study, the partition coefficient is defined as follows:

$$K_d = [A_{TSS}] / [A_{diss} \times SPM] \text{ ----- (2)}$$

Where  $A_{TSS}$  is the activity of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the particulate phase (mBq/g),  $A_{diss}$  is the activity of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the dissolved phase (mBq/g) and  $SPM$  is the total suspended particulate matter (Table 2).

The  $K_d$  value for  $^{210}\text{Po}$  ranged from  $0.05 \times 10^6$  L/g to  $3.22 \times 10^6$  L/g,  $0.45 \times 10^6$  L/g to  $74.82 \times 10^6$  L/g,  $1.18 \times 10^6$  L/g to  $93.50 \times 10^6$  L/g and  $1.02 \times 10^6$  L/g to  $49.21 \times 10^6$  L/g, respectively for 6 February, 5 April, 4 July and 9 October 2010. Meanwhile, the  $K_d$  value for  $^{210}\text{Pb}$  varied from  $0.05 \times 10^6$  L/g to  $1.25 \times 10^6$  L/g,  $0.47 \times 10^6$  L/g to  $212.58 \times 10^6$  L/g,  $13.80 \times 10^6$  L/g to  $501.31 \times 10^6$  L/g and  $0.18 \times 10^6$  L/g to  $37.86 \times 10^6$  L/g for each sampling, respectively. Table 2 shows the wide range of  $K_d$  values of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  for each station during the fourth sampling. A high  $K_d$  value for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  indicates that there is a strong adsorption of these radionuclides onto suspended particles in the aquatic environment. The lower  $K_d^{Po}$  and  $K_d^{Pb}$  values were found at Station 5 until Station 7 where boat traffic was quite busy for fish landing and tourism transportation. According to Zuo and Eisma (1993), the  $K_d$  value was inversely proportional to the availability of organic matter via the resuspension process where accumulated organic matter was dispersed from the bed into the water column. In this study, it was considered that the shallow water of Mersing River with respect to boat traffic significantly influenced resuspension (e.g., Ekhwan *et al.*, 2011).

The relationship between  $K_d^{Po}$  and  $K_d^{Pb}$  with respect to SPM were plotted in Fig. 4 in order to investigate its reactivity to SPM. Overall, based on the data obtained in this study the  $K_d^{Po}$  and  $K_d^{Pb}$  values of each sampling has a strong negative correlation against SPM except for  $K_d^{Pb}$  during the Southwest monsoon and  $K_d^{Po}$  during the inter-monsoon (October). The inverse correlation between  $K_d$  and SPM are commonly found for other nuclides such as  $^{210}\text{Po}$  and  $^{234}\text{Th}$  (Kim and Yang, 2004),  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  (Theng and Mohamed, 2005) and  $^{210}\text{Po}$  (Bacon *et al.*, 1988; Santschi *et al.*, 1979). This type of correlation is also known as “particle concentration effect” and a strong correlation demonstrates that the adsorption efficiency for SPM is very good. These results proved that  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  had an affinity to SPM in Mersing River. These indicate that SPM plays a significant role in controlling the scavenging of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ . The scavenging process by aggregation of colloidal matter and adsorption onto particulate matter influenced the mobilization of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the water column (Smoak *et al.*, 1996).

According to (Wei *et al.*, 2012), there was no systematic trend of  $K_d^{Po}$  and  $K_d^{Pb}$  with SPM in turbid coastal waters. However, this study found that more

than 99% of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  were found in the particulate phase due to the high loading of SPM. According to Stephen (1999), the statistical analyses may suggest a very strong relationship between pH and  $K_d$  term, and when it comes to the actual sorption process, it may be controlled by the iron oxide charge. The same applies to SPM and  $K_d$  term; the  $K_d^{Po}$  and  $K_d^{Pb}$  might be influenced by other covariants; either pH, iron oxide charge, organic matter or anionic constituent such as phosphate, chloride and carbonate. By comparison,  $K_d^{Pb}$  increases proportionally with the presence of organic matter especially humic acid, acid polysaccharides and bovine serum albumin and decreases with an increase of  $\text{AlO}_3$  in sediment (Yang *et al.*, 2011). Thus,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  might be released from surface sediments via resuspension (Tanaka *et al.*, 1983). Mn cycling across the sediment-water interface might play a key role in the enhancement of  $^{210}\text{Pb}$  scavenging at the bottom boundary (Spencer *et al.*, 1980). During Mn cycling,  $^{210}\text{Pb}$  can either be adsorbed onto  $\text{MnO}_2$ -coated particles or coprecipitated with  $\text{MnO}_2$  (Balistreri *et al.*, 1995). This consideration is very significant at the shallow water region like Mersing River. SPM performs as a  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  carrier for their transport and removal based on their geochemical behavior.

In spite of unstable environment conditions and chemical properties of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , a weak correlation was observed between  $K_d^{Po}$  during the inter-monsoon (October) and  $K_d^{Pb}$  during the Southwest monsoon with respect to SPM.  $^{210}\text{Po}$  had a higher biogenic particles reactive than  $^{210}\text{Pb}$  and  $^{234}\text{Th}$  especially in the euphotic zone (Wei and Murray, 1994) and was actively involved in the biogeochemical cycle (Tsunogai and Nozaki, 1971). Based on the lower concentrations of dissolved oxygen during the fourth sampling (October) which ranged from 3.4 to 5.3 mg/L compared to the first to third samplings which ranged from 4.1 to 6.7 mg/L, 4.1 to 7.4 mg/L and 4.7 to 5.8 mg/L, respectively. These results suggest that  $^{210}\text{Po}$  is removed by biogenic particles horizontally due to an almost anoxic environment (Kim and Yang, 2004). Meanwhile,  $^{210}\text{Pb}$  adheres to aerosol and is efficiently washed from the atmosphere by precipitation (Preiss *et al.*, 1996; Yamamoto *et al.*, 1998). The highest inventory of  $^{210}\text{Pb}$  in the particulate phase was during the third sampling (Southwest monsoon) suggesting that  $K_d^{Pb}$  might be affected by atmospheric fallout since the enhanced inventory most likely occurred during high precipitation. Therefore, bio-physico-chemical factors should be considered in order to have a better understanding of the particle behavior of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the shallow water region.

Table 2.  $K_d^{Pb}$ ,  $K_d^{Po}$ ,  $K_d^{Pb}/K_d^{Po}$  and SPM at Mersing River

| Area      | Station | $K_d^{Po}$ ( $\times 10^6$ L/g) |       |       |       |       |        | $K_d^{Pb}$ ( $\times 10^6$ L/g) |       |       |       |       |       | $K_d^{Pb}/K_d^{Po}$ |       |       |        |        |       | SPM ( $\times 10^{-3}$ g/L) |        |        |       |       |       |
|-----------|---------|---------------------------------|-------|-------|-------|-------|--------|---------------------------------|-------|-------|-------|-------|-------|---------------------|-------|-------|--------|--------|-------|-----------------------------|--------|--------|-------|-------|-------|
|           |         | 6-Feb                           | 5-Apr | 4-Jul | 9-Oct | 6-Feb | 5-Apr  | 4-Jul                           | 9-Oct | 6-Feb | 5-Apr | 4-Jul | 9-Oct | 6-Feb               | 5-Apr | 4-Jul | 9-Oct  | 6-Feb  | 5-Apr | 4-Jul                       | 9-Oct  | 6-Feb  | 5-Apr | 4-Jul | 9-Oct |
| Riverine  | 1S      | 2.12                            | 21.15 | 62.23 | 9.92  | 0.58  | 29.79  | 196.01                          | 3.42  | 3.68  | 0.71  | 0.32  | 2.90  | 49.20               | 23.81 | 4.38  | 16.49  | -      | 25.06 | 7.99                        | 17.57  | 28.51  | 14.79 | 4.56  | 10.53 |
|           | 1B      | -                               | 5.03  | 27.40 | 13.11 | -     | 24.59  | 99.32                           | 15.02 | -     | 0.20  | 0.28  | 0.87  | -                   | -     | 7.99  | 17.57  | -      | -     | -                           | -      | -      | -     | -     |       |
|           | 2S      | 3.22                            | 22.32 | 93.50 | 15.00 | 1.25  | 35.65  | 501.30                          | 20.44 | 2.57  | 0.63  | 0.19  | 0.73  | 28.51               | 14.79 | 4.56  | 10.53  | 28.51  | 14.79 | 4.56                        | 10.53  | 28.51  | 14.79 | 4.56  |       |
|           | 2B      | 1.25                            | 7.38  | 9.53  | 49.22 | 0.24  | 9.24   | 68.31                           | 2.18  | 5.16  | 0.80  | 0.14  | 22.53 | 49.51               | 28.95 | 13.57 | 27.39  | 49.51  | 28.95 | 13.57                       | 27.39  | 49.51  | 28.95 | 13.57 |       |
|           | 3S      | 2.82                            | 3.17  | 5.72  | 6.60  | 0.59  | 6.82   | 234.52                          | 24.47 | 4.78  | 0.47  | 0.02  | 0.27  | 21.35               | 20.65 | 12.34 | 7.96   | 21.35  | 20.65 | 12.34                       | 7.96   | 21.35  | 20.65 | 12.34 |       |
|           | 3B      | 2.31                            | 2.13  | 15.58 | -     | 0.56  | 2.44   | 211.05                          | 0.18  | 4.14  | 0.87  | 0.07  | 0.00  | 31.23               | 34.61 | 7.55  | 122.74 | 31.23  | 34.61 | 7.55                        | 122.74 | 31.23  | 34.61 | 7.55  |       |
|           | 4S      | 0.59                            | 0.87  | 6.29  | 3.44  | 0.13  | 1.52   | 228.84                          | 12.51 | 4.44  | 0.57  | 0.03  | 0.27  | 39.05               | 59.52 | 13.08 | 12.63  | 39.05  | 59.52 | 13.08                       | 12.63  | 39.05  | 59.52 | 13.08 |       |
|           | 4B      | 1.01                            | 5.78  | 1.79  | -     | 0.47  | 13.75  | 35.11                           | 5.49  | 2.13  | 0.42  | 0.05  | -     | 43.15               | 8.17  | 23.39 | 41.89  | 43.15  | 8.17  | 23.39                       | 41.89  | 43.15  | 8.17  | 23.39 |       |
|           | 5S      | 0.33                            | 0.70  | 3.76  | -     | 0.38  | 0.47   | 42.94                           | 7.92  | 0.86  | 1.50  | 0.09  | -     | 61.12               | 52.72 | 17.16 | 18.37  | 61.12  | 52.72 | 17.16                       | 18.37  | 61.12  | 52.72 | 17.16 |       |
|           | 5B      | 0.11                            | 0.45  | 1.18  | 1.02  | 0.07  | 0.83   | 32.68                           | 5.79  | 1.70  | 0.55  | 0.04  | 0.18  | 172.90              | 64.60 | 19.59 | 25.89  | 172.90 | 64.60 | 19.59                       | 25.89  | 172.90 | 64.60 | 19.59 |       |
| Estuarine | 6S      | 0.05                            | 1.12  | 4.75  | -     | 0.05  | 2.70   | 171.77                          | 3.90  | 0.99  | 0.41  | 0.03  | -     | 175.40              | 29.07 | 20.52 | 33.78  | 175.40 | 29.07 | 20.52                       | 33.78  | 175.40 | 29.07 | 20.52 |       |
|           | 7S      | 0.59                            | 0.61  | 3.10  | 2.31  | 0.59  | 0.84   | 13.79                           | 18.19 | 1.01  | 0.73  | 0.22  | 0.13  | 33.78               | 46.12 | 15.11 | 14.86  | 33.78  | 46.12 | 15.11                       | 14.86  | 33.78  | 46.12 | 15.11 |       |
|           | 8S      | -                               | 25.45 | 11.69 | 2.77  | -     | 48.01  | 32.37                           | 25.74 | -     | 0.53  | 0.36  | 0.11  | -                   | 6.50  | 7.48  | 7.85   | -      | 6.50  | 7.48                        | 7.85   | -      | 6.50  | 7.48  |       |
|           | 8B      | -                               | 35.76 | 6.25  | 4.48  | -     | 156.43 | 32.45                           | 21.92 | -     | 0.23  | 0.19  | 0.20  | -                   | 5.99  | 12.81 | 8.19   | -      | 5.99  | 12.81                       | 8.19   | -      | 5.99  | 12.81 |       |
|           | 9S      | -                               | 23.22 | 5.67  | 1.98  | -     | 82.52  | 18.90                           | 37.86 | -     | 0.28  | 0.30  | 0.05  | -                   | 7.88  | 8.78  | 6.16   | -      | 7.88  | 8.78                        | 6.16   | -      | 7.88  | 8.78  |       |
|           | 9B      | -                               | 74.82 | 5.65  | 8.21  | -     | 212.58 | 15.16                           | 22.54 | -     | 0.35  | 0.37  | 0.36  | -                   | 5.23  | 12.46 | -      | -      | 5.23  | 12.46                       | -      | -      | 5.23  | 12.46 |       |

SPM : Total suspended particulate matter

“S” : Surface water samples

“B” : Bottom water samples

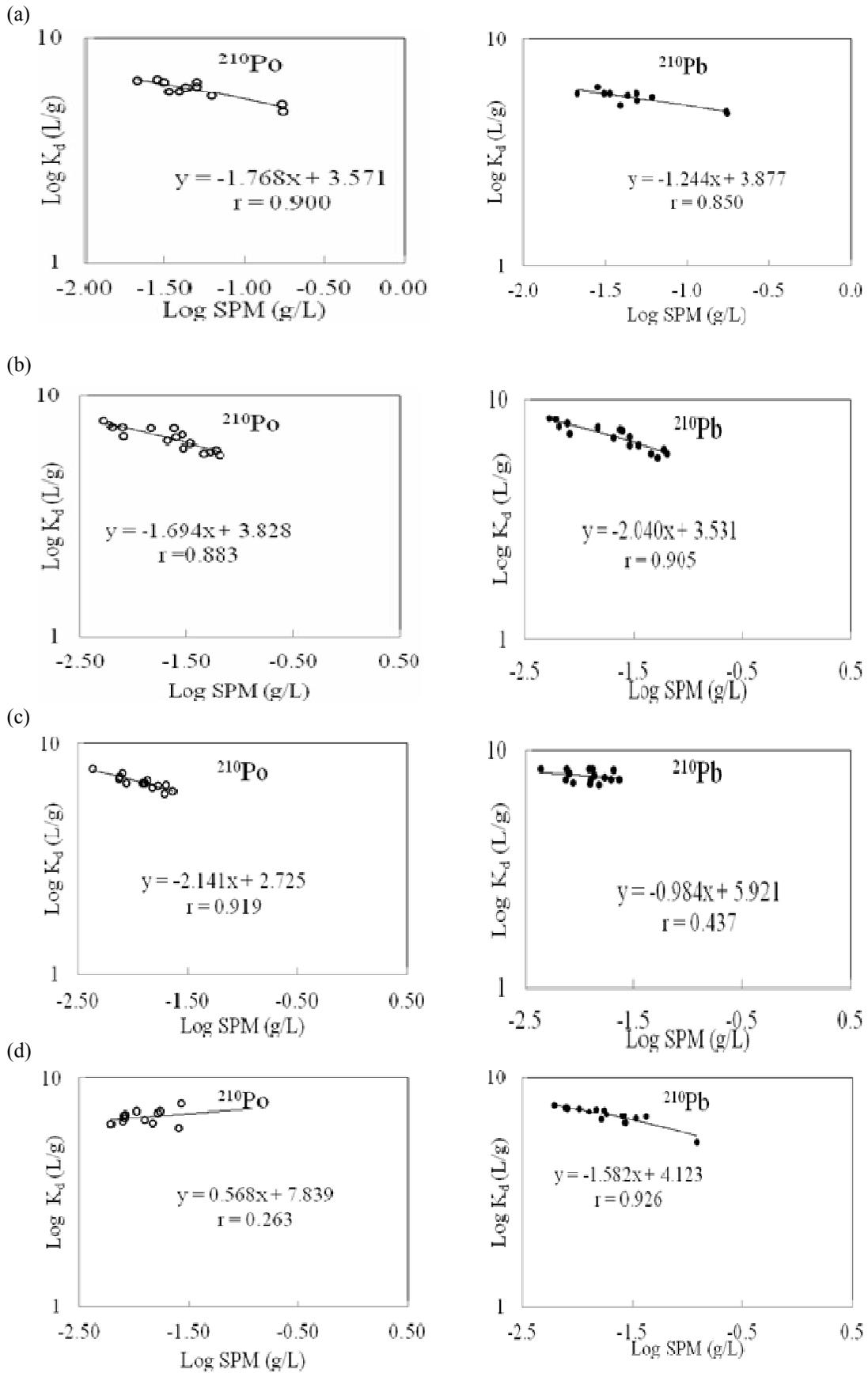


Figure 4. Correlation between  $\log K_d$  (L/g) and  $\log \text{SPM}$  (g/L) at the Mersing River on (a) 6 February 2010, (b) 5 April 2010, (c) 4 July 2010 and (d) 9 October 2010

Table 3. Correlation of <sup>210</sup>Po, <sup>210</sup>Pb, SRP, chlorophyll-a and SPM in water column

|                   |                     | <sup>210</sup> Po | <sup>210</sup> Pb | SRP    | Chl-a  | SPM |
|-------------------|---------------------|-------------------|-------------------|--------|--------|-----|
| <sup>210</sup> Po | Pearson Correlation | 1                 |                   |        |        |     |
|                   | Sig. (2-tailed)     |                   |                   |        |        |     |
|                   | N                   | 156               |                   |        |        |     |
| <sup>210</sup> Pb | Pearson Correlation | .378**            | 1                 |        |        |     |
|                   | Sig. (2-tailed)     | .000              |                   |        |        |     |
|                   | N                   | 155               | 157               |        |        |     |
| SRP               | Pearson Correlation | .681**            | .316*             | 1      |        |     |
|                   | Sig. (2-tailed)     | .000              | .012              |        |        |     |
|                   | N                   | 61                | 63                | 63     |        |     |
| Chl-a             | Pearson Correlation | .418**            | .390**            | .335** | 1      |     |
|                   | Sig. (2-tailed)     | .001              | .002              | .007   |        |     |
|                   | N                   | 61                | 63                | 63     | 63     |     |
| SPM               | Pearson Correlation | .805**            | .679**            | .697** | .425** | 1   |
|                   | Sig. (2-tailed)     | .000              | .000              | .000   | .001   |     |
|                   | N                   | 57                | 59                | 59     | 59     | 59  |

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

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

### 3.3. Biological Productivity of Mersing River

Chlorophyll-a content in water column has been utilized as an indicator of assessing abundance of phytoplankton biomass (Nozaki *et al.*, 1997). The simultaneous measurements of chlorophyll-a, <sup>210</sup>Po and <sup>210</sup>Pb in water column were performed in this study with purpose to study the biogeochemistry behavior of these well known as particles reactive nuclides at shallow

water region. <sup>210</sup>Po is preferentially removed by biogenic particles and actively involved in the biogeochemical cycle compared to <sup>210</sup>Pb (Tsunogai and Nozaki, 1971). In the aquatic environment, <sup>210</sup>Po is assimilated by phytoplankton due to its sulfur-like quality even though it is a non-essential element for their metabolism (Fisher *et al.*, 1983). In this study, the relationship between <sup>210</sup>Po, <sup>210</sup>Pb, SRP, chlorophyll-a and SPM were displayed in Table 3. It should be noted that <sup>210</sup>Po, <sup>210</sup>Pb, SRP, and

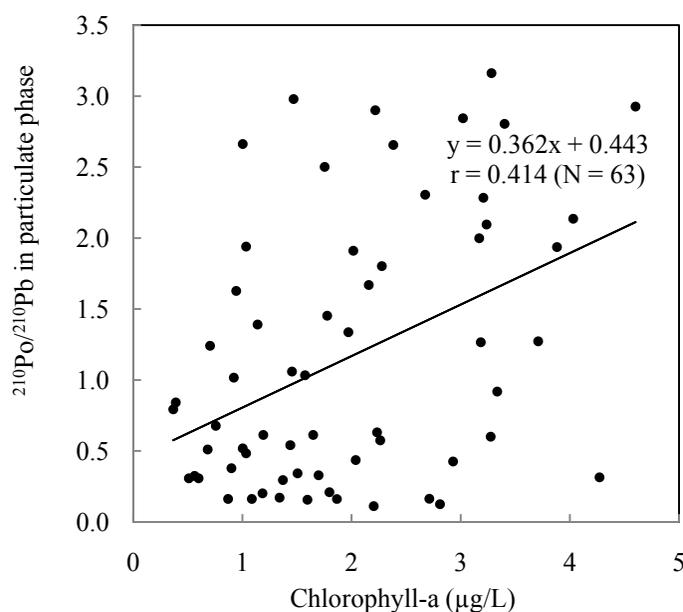


Figure 5. Correlation between <sup>210</sup>Po/<sup>210</sup>Pb in particulate phase and chlorophyll-a (µg/L).

SPM show a significantly ( $p < 0.01$ ) lower correlation with respect to chlorophyll-a indicating a low primary production as the particulate reactive elements and SPM seem to have a low affinity to biogenic organic matter in the Mersing River water column. The low correlation ( $r = 0.414$ ) between  $^{210}\text{Po}/^{210}\text{Pb}$  in the particulate phase and chlorophyll-a in Fig. 5 proved that the enrichment of  $^{210}\text{Po}$  were not associated with the abundance of chlorophyll-a. This might be due to the turbidity effect by resuspension.

The relationship between chlorophyll-a and SRP with respect to SPM was plotted in Fig. 6. SRP concentration in the water column is moderately associated with SPM concentration. However, chlorophyll-a has a low correlation with SPM. These indicate that chlorophyll-a has a low affinity to SPM in the Mersing River. On other hand, palm oil agriculture activities probably contribute significantly to SRP concentration in the water column via sedimentation and river bank erosion. The relatively

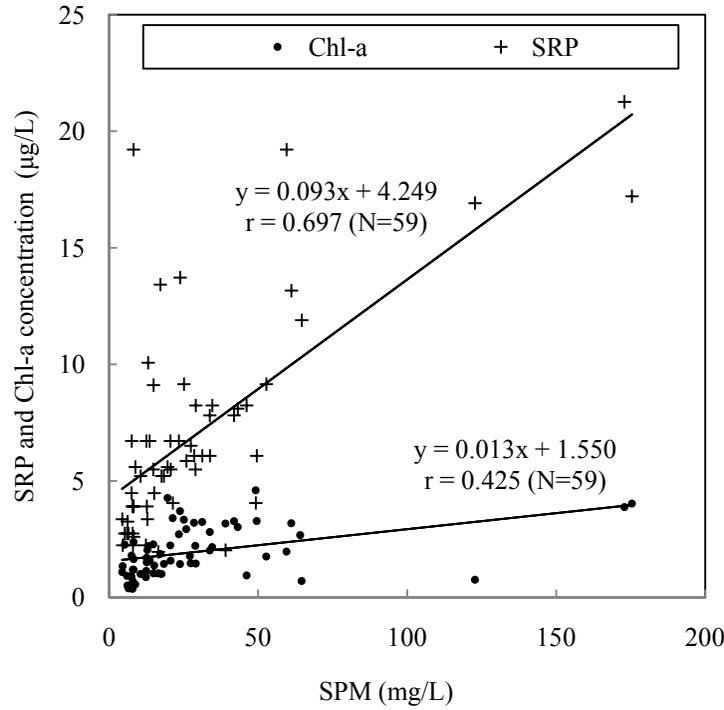


Figure 6. Correlation between SRP ( $\mu\text{g/L}$ ) and chlorophyll-a ( $\mu\text{g/L}$ ) and SPM ( $\text{mg/L}$ ) in the water column.

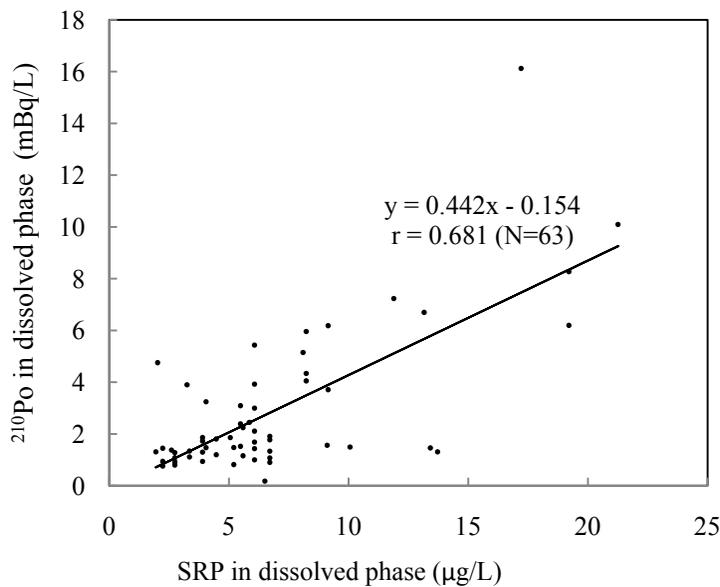


Figure 7. Correlation between  $^{210}\text{Po}$  (mBq/L) and SRP ( $\mu\text{g/L}$ ) in the dissolved phase.

high inventory of  $^{210}\text{Po}$  at station 4 might be due to fertilizer waste input from the nearest palm oil agricultural area via the two small river run-offs (Saili, 2013). Fig. 7 shows the strong correlation ( $r=0.681$ ) between SRP and  $^{210}\text{Po}$  in the dissolved phase, significantly proving that the contribution of phosphate concentrations potentially increase  $^{210}\text{Po}$  radioactivity (Carvalho, 1995; 1997). These results indicate that  $^{210}\text{Po}$  has a stronger affinity to the total suspended particulate matter than chlorophyll-a (Table 3). Nevertheless, the data obtained in this study was insufficient to support this assumption and to evaluate lithogenic particle domination. A study of particle size on transport settling and sedimentation along pathways might be useful in order to investigate particle dynamics source-to-sink to get a clear geochemical mechanism (Fahl and E-M, 2007; Honjo, 1982; Liu *et al.*, 2009; Nakatsuka *et al.*, 2004). Therefore, this study suggests that a high concentration of SPM in Mersing River is caused by resuspension from bottom sediment.

#### 4. Conclusions

Resuspension of bottom sediments caused by natural and man-made activities are important processes that manipulate SPM input into the Mersing River. Those processes are significantly play a critical role in the influence of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  accumulation and transportation with respect to SPM concentration in the water column. A strong correlation between  $K_d^{Po}$  and  $K_d^{Pb}$  with SPM proved that  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  has a high affinity to SPM. The SPM also performs as a  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  carrier for their transport and removal based on their geochemical behavior. Low correlation between chlorophyll-a with  $^{210}\text{Po}/^{210}\text{Pb}$  ratio indicates that the enrichment of polonium was not associated with the abundance of chlorophyll-a as indicator for biological productivity in marine environments. But a strong correlation between SRP and  $^{210}\text{Po}$  in the dissolved phase proved that the contribution of phosphate potentially increasing the activity of  $^{210}\text{Po}$ , and further intensive study is needed to investigate this correlation especially Malaysia coastal waters.

#### Acknowledgements

The authors would like to thank all the laboratory members for their help during the sampling period and with sample analysis. We would also like to thank the Ministry of Science, Technology and Innovation (MOSTI), National Oceanography Directorate (NOD) for providing the research grant NOD/R&D/01/002.

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Received 18 March 2014

Accepted 25 April 2014

#### Correspondence to

Che Abd Rahim Mohamed  
 School of Environmental and Natural Resource Sciences,  
 Faculty of Science and Technology,  
 Universiti Kebangsaan Malaysia,  
 Bangi Selangor 43600,  
 Malaysia  
 E-mail: mohamed6566@yahoo.com, carmohd@gmail.com