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Research paper

The Evaluation of Brunei Bay Sediment Cores Sedimentation Rate Using ^{210}Pb Radiometric Dating Technique

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Abstract

The use of radioisotopes ^{210}Pb and ^{226}Ra in establishing the geochronology of pollutants in the sediment core and sediment dating is being widely used in the world. The present study was conducted in Brunei Bay region of Malaysian waters to define the sedimentation rate and sediment age as well as to investigate the possible sources of pollutants into this bay. Sediment core samples were cut by layers, dried and analyzed using High Purity Germanium (HPGe) Spectrometer. Results obtained marked the time interval of 1875, 1956, 1962 and 1945 for sediment core B5, B9, B13 and LB consecutively. Sediment core of B9 and LB showed higher sedimentation rate compared to B5 and B13 due to the rapid development of urban and industrial. The increasing of sedimentation rate over the last 25 years was in line with the increasing of human activities surround the bay. Additionally, the health and distribution of mangroves surround Brunei Bay were important to determine the sediment movement which will affect the sedimentation rate in the bay. Overall, by controlling human activities as well as sustaining the mangroves population, could maintain and preserve the natural and unique environment of Brunei Bay.

***Keywords*:** *Brunei Bay, Borneo; radioisotope 210Pb, radioisotope 226Radium; sediment dating; sedimentation rate.*

1. Introduction

Marine sediment serves as a large reservoir for various types of pollution; represents the environmental health and restores the historical chronology of the area. The sediment acts by storing and maintaining the important record for environmental changes study and non-natural disruptions from the past. The most common example is the sources of pollutants stored in sediment layers can be investigated and linked to years through sedimentation rates analysis. The marine environment is very important to benchmark the human activities as the sedimentation occurrence may cause a major problem on the environment [1].

Sediment is a combination of complex materials contributed by rivers, coastal erosion, biological and chemical processes as well as anthropogenic waste. The rapid economic growth and settlement can drive the release of waste materials into the coastal area and accumulate into the sediment. Later, the chemicals and their compounds can re-circulate into the water column through the absorption onto the suspended particulate matter [1-5]. Most particles transformed into various chemical species before reaching the seafloor. The rate of organic particles deposition and decaying is strongly influenced by biological mixing between solid and liquid without bioturbation zone [6].

Radioisotope Lead-210 (^{210}Pb) with half-life of 22.3 years has been widely used as an environmental detector to study the deposition of sediments in different sedimentary environments and marine pollution studies [7]. ^{210}Pb is known as a major detector for particle transport, studying chemical destruction, atmospheric analysis, geochronology and sediment blindness in marine environments [8].

Normally, ^{210}Pb is belongs to ^{238}U family series (Figure 1). Parents with nearly half life are Radon-222 (^{222}Rn, half-life: 3.8 days) being a noble gas, fled to the atmosphere and gives ^{210}Pb with unexpected mobility [9]. The final ^{210}Pb radioactive product of ^{222}Rn returns to the earth through dry or wet precipitation which will settle down into the sediment [10]. This mobility creates an "excess" of ^{210}Pb as compared to the expected secular balance with Radium-226 (^{226}Ra) which forms the basis of the ^{210}Pb dating method (Figure 1). Other than that, ^{210}Pb is introduced into the estuarine through atmospheric deposition, inland and in-situ production from ^{226}Ra in the water columns.

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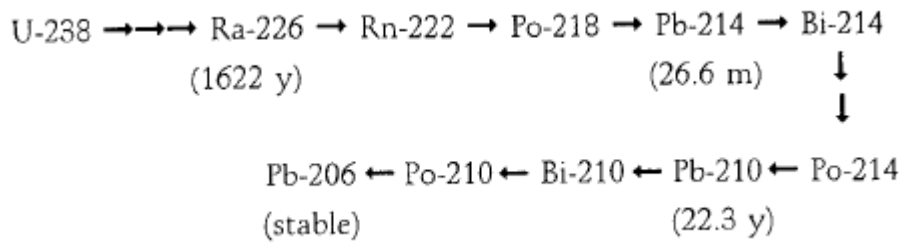


Fig. 1: The decay rate of ^{226}Ra and it daughters which belong to the U238 series [9].

Coastal water, estuarine and bay have two types of ^{210}Pb supply. One comes from the atmospheric ^{222}Rn and the other is produced by ^{226}Ra decay process in the water column. Generally ^{210}Pb is forcely removed from the water column into the sediment through adsorption onto particulate matter chemically. Other than that, ^{210}Pb activity on the surface of the sediment is the result of interaction between collection, sediment mixing, and radioactive damage. This interaction determines the extent to which particle changes occur between the sediment and extreme water column [8]. ^{210}Pb radioisotope is bound to material and trapped in sediment, where the sedimentation rate transformation changes the ^{210}Pb dilution as it accumulates in sediment [11-12].

In the shallow marine area, ^{210}Pb is introduced via wet and dry precipitation by the release of radon gas in the atmosphere as well as supplied by the ^{222}Rn decay in the sea water. The addition of ^{210}Pb in this water system, so being called as $^{210}Pb_{ex}$ is an excessive or unsupported ^{210}Pb. In fact, sediment contains ^{210}Pb resulting from in-situ affected by ^{226}Ra is known as ^{210}Pb supported ($^{210}Pb_{supp}$). Thus, all ^{210}Pb sources in the sediment are known as ^{210}Pb total, $210Pb_{tot}$ [13-14]. $^{210}Pb_{ex}$ activity is higher on the surface sediment and exponentially decreases with depth in sediment from time to time due to the sedimentation process, while $^{210}Pb_{supp}$ activity is consistent with depth [13, 15]. $^{210}Pb_{supp}$ comes from the inoculation of in situ radionuclide ^{226}Ra and will usually be equilibrium with ^{226}Ra, while $^{210}Pb_{ex}$ mostly derives from atmospheric flux. $^{210}Pb_{ex}$ is determined by reducing the supported concentration of the total concentration at phase [16]. It is commonly used in the calculation of sedimentation rate and total inventory in the sediment core.

In order to establish the geochronology for stored pollutants in the sediment, the ^{210}Pb activity values need to be determined precisely. Numerous research have been carried out to establish chronology and determine sedimentation rates using unsupported ^{226}Ra ($^{226}Ra_{ex}$) in various sedimentary environment [17-23]. However, assumptions need to be created on the behavior of ^{226}Ra when applying $^{226}Ra_{ex}$; primarily related to the sediment water transfer and potentially for post-control [24, 20]. According to [21], ^{226}Ra has a half-life of 1602 years and the potential of dating can be expanded between 100-10,000 years. ^{226}Ra is not directly used in the study of limnochronology, although it is regularly measured ^{210}Pb dating in order to measure the ^{210}Pb supported activity.

The chronological technique is commonly used to estimate the average of sediment rate for more than 100 years in marine and freshwater environment [25-32]. The ^{210}Pb chronolgical method was introduced by Goldberg [32] and was applied to the sediment samples by [33] in the study of marine sediments [23]. $^{210}Pb_{ex}$ dating in sediment also can describe the changes between natural and anthropogenic activities in past [34-35]. Therefore it is the most appropriate approach to determine the chronology of the past 100 years in sediment core to determine changes of the past environment as well as to evaluate the accumulation of materials [36-43].

Apart from that, low gamma transmitting radionuclides such as ^{210}Pb has been widely used in geo chronological studies. In fact, the addition of sediments using $^{210}Pb_{ex}$ profile has been a standard technique to determine the sediments geochemical activity for 100-150 years. Typically, chronology study which is commonly practice is based on two assumptions that underpin the basis of $^{210}Pb_{ex}$ collection. On the other hand, the Constant Initial Concentration (CIC), which assuming that the initial concentration of $^{210}Pb_{ex}$ in the deposited sediment is similar regardless of changes in the sediment collection rates.

The total concentration of ^{210}Pb in the sediment is usually determined by gamma spectrometry or alpha spectrometry through the radionuclide daughter count when the balance is assured. Other than that, there are less common method such as liquid thinning and beta counting [25, 44-46]. There are few common mathematical model have been used in radioisotopes dating namely CF:CS (Flux Always; Realization Sedimentation), CIC (Constant Initial Concentration) and CRS (Supply Continuity Rate). As shown by Appleby (2001), CF:CS model assumes that the $^{210}Pb_{ex}$ concentration throughout the sediment core is identical to the initial concentration of ^{210}Pb as the $^{210}Pb_{ex}$ value is constant along the sediment core.

The present study was carried out during the period of 2013 in the Brunei Bay of Malaysian waters. The aims of this study were to define the sedimentation rate and sediment age of the collected sediment cores. Besides that, the present study also intended to investigate the possible pollutant sources into Brunei Bay area since this bay is well known with its unique and diverse ecosystem [47-48].

2. Methodology

2.1. Study Area

Brunei Bay is a sheltered marine water system which links Brunei and Malaysian territories of Sabah, Sarawak and Labuan. This bay is located on the Northwest coast of Borneo Island with the bay opening to the Southeast part of the South China Sea. The temperature of Brunei Bay varies from 20°C up to 36°C due to climate changes between the northeast and southwest monsoon yearly. Additionally, this bay is being dominated by mangrove and nipah forest, palm oil plantation, logging and dense local settlements [47-48]. Other than that, Brunei Bay has a unique and diverse ecosystem due to a variety wildlife which has been inhabit the bay area namely dolphin, sea-cow, turtles, crocodiles, *Proboscis* monkey and many more. Figure 2 shows the sampling location for sediment core samples. Four locations were selected to represent natural preserved area as well as high human activities areas.

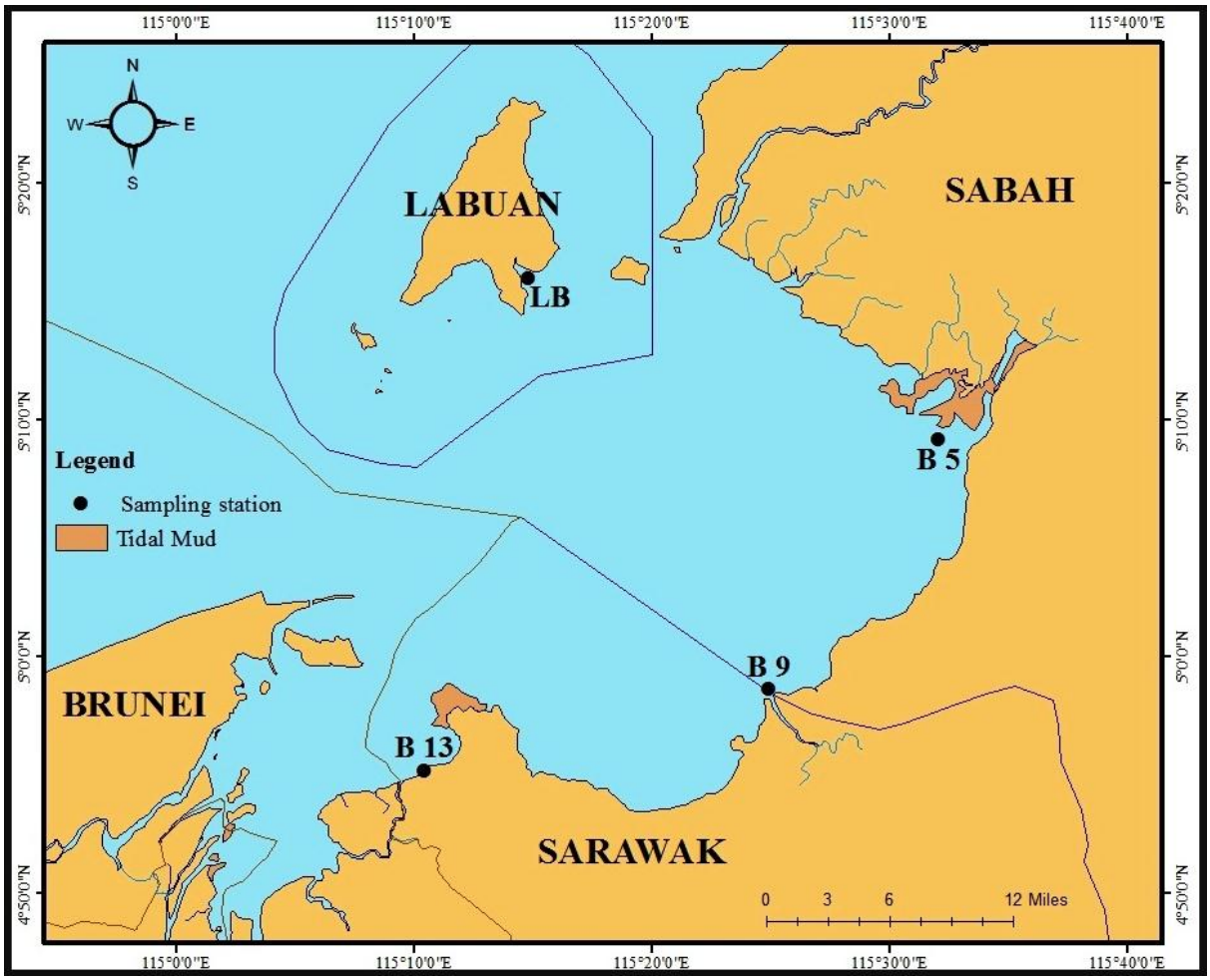


Fig. 2: Sampling location at Brunei Bay

2.2. Analytical Method

The measurements of ^{210}Pb and ^{226}Ra were performed using the High Purity Germanium (HPGe) Spectrometer CANBERRA Model 747 Lead Shield and HPGe Specyrometer BE5030 Model (Broad Energy Ge) with a crystal (diameter: 80.5 mm, thickness: 31.0 mm) from Canberra USA. There is no standard guideline for the optimum thickness of the sediment core section due to some literature indicates that the thickness between 2 and 7 cm has been used. However, the thickness of sediment core with 1 cm does not show very precise results [49]. In the present paper, we intended to maintain an average value of 3 cm for B5, B9 and B13 sediment core thickness. However for LB sediment core, the core length was start from 2cm from the top until 40cm and 3cm downward core.

Some studies did not count the upper part of the core (first sedative layer) in calculating the sediment age as they consider it is non-solid sediment layer. Therefore the difference of 10 years is acceptable for this appointment method [50-51]. In this study, the core surface sediment (0-3cm) was counted and the sedimentation rate calculation was started from layer 0-3cm for B5, B9 and B13, meanwhile LB start with sediment layer of 0-2cm.

Next, the sample were dried, firmly crushed into fine particle (< 63 mm) and stored in polypropylene vacuum bottles. The bottles were then sealed with thick PVC tape to prevent the degradation of radioisotope ^{226}Ra. All samples were stored for a period of more than 30 days to establish a secular balance between 226Ra and their radioactive progeny before gamma ray count [52-54].

Finally the radioisotope value for ^{210}Pb and ^{226}Ra were calculated for 86,400s using both HPGe Spectrometer with 25% detector efficiency and their activity were corrected up to date of sample collection. The counting time is long enough to make sure the 2σ counting error less than 10%. ^{226}Ra was measured through photopeaks of it progenies. The ^{210}Pb activities were measured directly through the peak energy of 46.54 and 661.62 keV.

2.3. ^{210}Pb radioactive Calculation

In the present study, the sedimentation rate was assessed by a CIC $^{210}Pb_{ex}$ model [55]. The $^{210}Pb_{ex}$ activity is obtained by subtract the $^{210}Pb_{supp}$ activity value out of the total ^{210}Pb activity per sedimentary core layer. The $^{210}Pb_{supp}$ value is determined by using ^{226}Ra activity, assuming that the activity of $^{210}Pb_{supp}$ and ^{226}Ra are in a secular balance [40]. Calculated ln ($^{210}Pb_{ex}$) data obtained in this study was plotted against sediment depth. The straight line obtained can be used to calculate the sediment deposition (cmyr^{-1}) by using below equations:

$$A(^{210}Pb_{ex}) - A_0(^{210}Pb_{ex})e^{-\lambda h/s} \quad (1)$$

$$A(^{210}Pb_{ex}) - A_0(^{210}Pb_{ex})e^{-\lambda H/S} \quad (2)$$

where $^{210}Pb_{ex}$ is calculated by subtracting ^{226}Ra activity from the total ^{210}Pb; A ($^{210}Pb_{ex}$) and A0 ($^{210}Pb_{ex}$) are $^{210}Pb_{ex}$ activity at two different depth on the sediment core (Bqkg^{-1}); s is the suction rate (cmyr^{-1}); S is the mass of the suction (gcm^{-2}yr^{-1}); λ is the degradation constant of ^{210}Pb (0.0311yr^{-1}), h is the cumulative depth (cm); H is the cumulative depth of mass (gcm^{-2}). Sediment layer below than 30cm shows similar ^{210}Pb activity to the activity supported by ^{226}Ra. The $^{210}Pb_{ex}$ radioactivity value observed in sediments above 30 cm shows a vertical profile decreasing in concentration with depth [56].

3. Results and Discussion

Based on Table 1 and Figure 3, the B5 core sample showed a high ^{210}Pb and ^{226}Ra activities at sediment surface layer to 27cm layer with 19.66-32.23 Bqkg^{-1} and total organic value of 0.535-4.59%. Meanwhile ^{210}Pb and ^{226}Ra activities in core B9 sample from 0-36cm depth were range from 14.14 to 25.44 Bqkg^{-1} with the total value of organic carbon between 0.000-1.37 %. The core B 13 samples did not show any significant ^{210}Pb and ^{226}Ra activities due to a relatively short sediment core with a depth of 24cm and gave 0.88-5.75 % of total organic. In the meantime, LB core sample showed a mixed pattern of ^{210}Pb and ^{226}Ra activities with total organic carbon content between 1.25-5.81% (Table 1 and Figure 3).

Table 1: Range value of ^{210}Pb and ^{226}Ra activities, sedimentation rates and total organic carbon in sediment cores of Brunei Bay.

Station	^{210}Pb activity, Bq/kg	^{226}Ra activity, Bq/kg	Sedimentation rates / year	Total organic carbon, %
B 5	12.33-32.33	11.87-31.75	0.48	0.54-4.60
B 9	8.55-27.63	11.5-21.21	0.88	0.00-1.37
B 13	2.58-19.76	6.66-17.38	0.47	0.88-5.75
LB	2.59-29.00	7.17-25.1	2.13	1.25-5.81



Fig. 3: Vertical profile of ^{210}Pb and ^{226}Ra activities (Bqkg^{-1}) in the sediment cores of Brunei Bay.

Based on the calculated sedimentation rate, sediment core B5 with 66cm deep indicates a time interval of 1875, sediment core B9 with an interval of 1956 with core depth of 54cm. In the meantime, sediment core B13 only found at 1962 interval at 24cm depth while LB samples provided a time interval in 1945 at sediment core of 145cm.

Generally, the ^{210}Pb profile in the sediment cores of Brunei Bay has indicated mixing activities. It is likely to be described by the relationship with intense biological bioturbation that occurs in the sediment layer [57]. Changes in oxygen may also affect the remobilization of ^{210}Pb which in turn causes the mixing of the ^{210}Pb profile [58]. Other than that, the mixing effect is commonly caused by a strong physical and bioturbation process of biology [59-60].

In the present study as described in the methodology, the sediment rate is assessed using CIC $^{210}Pb_{ex}$ model. According to [55], the value of $^{210}Pb_{ex}$ activity is obtained by deducting the $^{210}Pb_{supp}$ activity from the total ^{210}Pb activity for each sedimentary core layer. Figure 4 shows a plot of data Ln (^{210}Pbex) against the depth of the sediment core (cm). A straight line has been produced and used to calculate the sediment rate in a year (cm / yr). The sedimentation rates were found to be between 0.47 to 2.13 cm / yr as shown in Table 1. The sedimentation rate for station B5 (0.48 cm / yr) and B13 (0.47 cm / yr) is low compared to station B9 (0.88 cm / yr) and station LB (2.13 cm / yr).

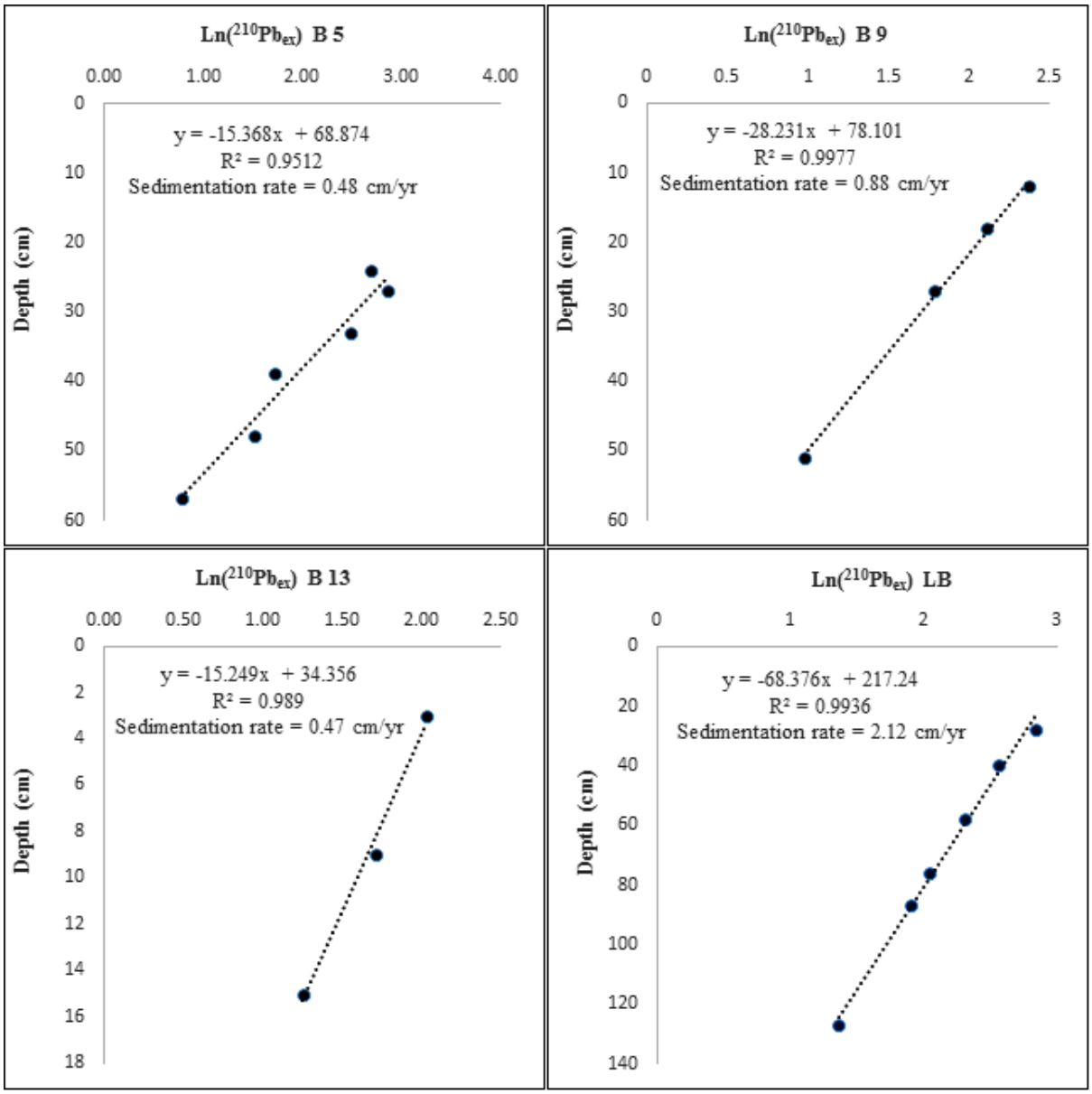


Fig. 4: Graph Ln (Pb_{ex}) versus depth (cm) for sediment cores in Brunei Bay.

The sedimentation rate for sediment core of B9 and LB were high. It can be explained by various activities that can be found within B9 and LB areas. The sources of sediment input into the B9 area is likely to be high contributed by the rapidly growing agricultural and logging activities. The increased of urbanization and population development resulted in increased of municipal waste is a possible major sources in contributing to sediment rates process of LB area. Those activities included were drainage, land use for disposal, hydrological variation, land use development, flood water turbidity due to drainage work, coastal reclamation, drilling work for municipal and industrial as well as land [61-62].

Determination of sediment rates is an important tool for explaining the origin of particle materials, particle transport, sediment mixing and sedimentation quantities [63-65]. However, the content of organic matter, iron and manganese granulometry as well as topography and meteorological topography such as floods and storms are the major factors affecting sedimentation in marine environments [66]. ^{210}Pb radioactive nucleus which is naturally used in environmental research is known as particlereactive in marine environments [67].

According to the Department of Irrigation Malaysia, major floods recorded in the Brunei Bay area were in the year of 1956, 1965, 1976, 1985, 1999 and 2007. The sediment rates calculated in B5, B9 and B13 were increased during these event while LB area was not affected by the events as the distance factor played a major role (Figure 3). Additionally, Lawas River also counted as major contributor to the sedimentation with 440m river mouth wide. Sedimentation rates also been referred to the amount of organic and inorganic material deposited through rainwater and river water at all times. The sediment distribution and sediment deposits in the river can cause water degradation as well as changes in the physical and chemical criteria of the water.

Nonetheless, the percentage of total organic carbon in all core samples studied showed a weak relationship with ^{210}Pb activity. It is shows that ^{210}Pb has no relation with biogenic particles and high ^{210}Pb sources is probably derived from the lithogenic (detrital) inclusion as described by [68]. This is in agreement with others reported as lower total organic carbon may be associated with high sediment rate in marine sediment [68, 69-75]. In [76] has described that the relationship between total organic carbon and sediment rate may influenced by the storage conditions as a positive correlation was observed under the oxidized state.

The deterioration of organic matter depends primarily on underwater redox conditions [77-84]. When sediment rate is slow (below 5 cm/s), the organic matter has enough time to interact with oxygen within the water column. Therefore different redox

conditions may control the correlation of total organic carbon and sedimentation rate in the sediment. Generally, it is believed that when sediment rate is lower than 5 cm/s, the redox variant has a significant control over the relationship between total organic carbon, sedimentation rate and the correlation curve. This is in agreement with the findings by Stein [76] as under a strong reduction condition, there is no correlation between total organic carbon and sedimentation rate in the sediment.

Figure 5 shows the value of redox potential is decreasing sediment with depth. It is similar to those found by [85] as this condition can cause some ^{210}Pb to be transported from the original depth and associated with iron and manganese digenesis.

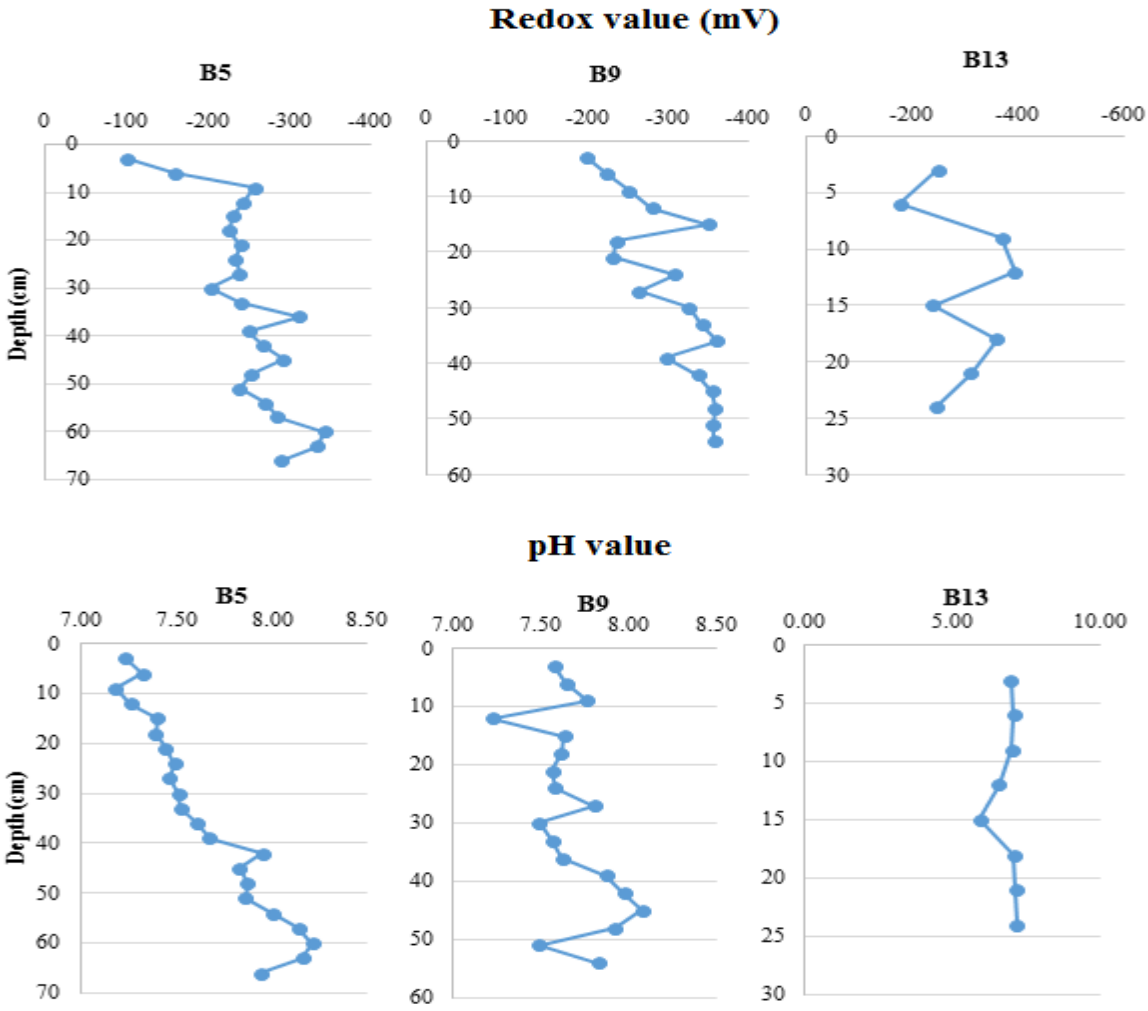


Fig. 5: The distribution of pH and redox value in the sediment cores of Brunei Bay.

Sediment core of B5 and B13 showed low sedimentation rates and this is likely due to the presence of abundant mangrove trees on the coastal and river banks. Mangrove trees are important in stabilizing coastline and estuary as well as retaining from tidal bore and soil erosion [77]. On the other hand, B9 and LB were surrounded by less mangrove areas and most of the mangroves have been exploited and turned into industrial, saline, aquaculture, aquaculture, and nursery areas.

4. Conclusion

Sediment core of B5, B9, B13 and LB have indicated a timeinterval of 1875, 1956, 1962 and 1945 respectively. Sediment rates are high in areas with industrial, high population, municipalities, agriculture, high human activity and closed to the river mouth. It was proven by the high sediment rates in LB and B9 areas with high developed industrial and urban activities. Besides that, the sustainability and health of mangrove trees were shown to play an important role in the movement and diverting the sediments from rivers and shores. All areas of sediment rates studies have decreased toward down core. The sediment rates has increased over the last 25 years due to the rapid increasing of industrial and the urban development. The sedimentation studies of sediment are very important to understand past events, especially in terms of anthropogenic inclusion or contribution from human activity to the environment.

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