

# DIFFERENT TECHNIQUES OF EXCESS $^{210}\text{Pb}$ FOR SEDIMENTATION RATE ESTIMATION IN THE SARAWAK AND SABAH COASTAL WATERS

PERBEZAAN TEKNIK  $^{210}\text{Pb}$  LEBIHAN UNTUK PENGANGGARAN KADAR PEMENDAPAN SEDIMEN DI PERAIRAN SARAWAK DAN SABAH

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## Abstract

Sediment core samples were collected at eight stations in the Sarawak and Sabah coastal waters using a gravity box corer to estimate sedimentation rates based on the activity of excess  $^{210}\text{Pb}$ . The sedimentation rates derived from four mathematical models of CIC, Shukla-CIC, CRS and ADE were generally shown in good agreement with similar or comparable value at all stations. However, based on statistical analysis of independent sample t-test indicated that Shukla-CIC model was the most accurate, reliable and suitable technique to determine the sedimentation rate in the study area.

## Abstrak

Sampel teras sedimen telah diambil di lapan stesen di pesisir pantai Sarawak dan Sabah dengan menggunakan peneras kotak graviti untuk menganggar kadar pemendapan sedimen berdasarkan aktiviti  $^{210}\text{Pb}$  berlebihan. Kadar pemendapan sedimen yang diterbitkan daripada empat model matematik CIC, Shukla-CIC, CRS dan ADE menunjukkan nilai yang hampir sama dan setanding di semua stesen. Bagaimanapun, berdasarkan analisis statistik ujian-t sampel bebas menunjukkan model Shukla-CIC adalah teknik yang paling tepat, boleh digunapakai dan sesuai untuk menentukan kadar pemendapan sedimen di kawasan kajian.

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**Keywords:** Model;  $^{210}\text{Pb}_{\text{ex}}$ ; Sedimentation rate; sediment core; Coastal; Sabah and Sarawak

## INTRODUCTION

As we know marine environments will represent an important value for human activities because sedimentation of sediment is a major problem affected the marine environment (Panayotou 2002). Therefore, the study of sediments in coastal areas has great importance to understanding the interaction between human activities and marine system. Furthermore, the estimation of sedimentation rate is importance to develop a continuum of insight into marine environmental processes (Lu and Matsumoto 2005).

The understanding of recent sedimentary process in marine environments has been greatly in past decades through the application of short-live U-Th-series and bomb produced radioisotopes to sediment geochronology (Dukat and Kuehl 1995). For example,  $^{210}\text{Pb}$  with half-life of 22.3 years is radioactive members of the naturally occurring  $^{238}\text{U}$  decay series has been used intensively as one of the key tracers to study the process of sediment deposition in marine environments (Uğur and Yener 2001). Moreover, it also proved invaluable in the determination of sediment accumulation rates on about a 100-year time scale

(Koide et al. 1972; Koide et al. 1973; Krishnaswami et al. 1971). The  $^{210}\text{Pb}$  method was initiated by Goldberg (1963), then applied to lake sediments by Krishnaswamy et al. (1971) and subsequently introduced to marine sediments by Koide et al (1972). It has been very popular in estimating the sedimentation rate of marine sediment (Huh and Su 1999).

In shallow ocean areas,  $^{210}\text{Pb}$  is supplied by the *in situ* decay of  $^{222}\text{Rn}$  ( $t_{1/2} = 3.8$  days) from dissolved  $^{226}\text{Ra}$  in seawater and from atmospheric deposition of  $^{210}\text{Pb}$  to sea surfaces. In facts, bottom sediments also contain  $^{210}\text{Pb}$  from the *in situ* decay of  $^{226}\text{Ra}$  (supported  $^{210}\text{Pb}$ ;  $^{210}\text{Pb}_{\text{supp}}$ ), the additional  $^{210}\text{Pb}$  is known as excess or unsupported  $^{210}\text{Pb}$  ( $^{210}\text{Pb}_{\text{ex}}$ ) (Zaborska et al. 2007). The activity of  $^{210}\text{Pb}_{\text{ex}}$  is higher at the surface and exponentially decreases with depth in the sediment core over time since deposition while  $^{210}\text{Pb}_{\text{supp}}$  activity remains constant (Shukla 2002; Zaborska et al. 2007). A sedimentation rate is determined for a sediment core based on the down-core activities of  $^{210}\text{Pb}_{\text{ex}}$  (total minus supported).

During this investigation, a different mathematical model was performed to calculate the sedimentation rate based on vertical profiles of  $^{210}\text{Pb}_{\text{ex}}$  in sediment cores versus depth (cm). There are four mathematical models such as the constant rate of supply model (CRS), the constant initial concentration model (CIC), Shukla-CIC model and advection-diffusion equation (ADE). The CRS model was initiated by Appleby and Oldfield (1978) and Oldfield and Appleby (1984) assumes a constant  $^{210}\text{Pb}$  flux at water-sediment interface and requires both the integrated activity and the differential activity to yield a variable sedimentation rate (Shukla 2002). The CRS model is used when the supply of  $^{210}\text{Pb}_{\text{ex}}$  is constant and the sediment deposition rate is variable (Appleby and Oldfield 1978; Robbins 1978). The CIC model has been applied by Robbins and Edgington (1975), Durham and Joshi (1980) among others assumes both a constant mass flux and a constant activity flux at the water-sediment interface; and requires only differential activity to yield a constant sedimentation rate (Shukla 2002). It also appropriate when initial activity of  $^{210}\text{Pb}_{\text{ex}}$  is constant and there is no mixing of surface sediments (Robbins and Edgington 1975). This implies deposition of sedimentary material characterized by constant  $^{210}\text{Pb}_{\text{ex}}$  activity such that either both  $^{210}\text{Pb}_{\text{ex}}$  activity and mass of deposited material to the sediment surface are constant or both vary at the same rate (Zaborska et al. 2007). The ADE was given by Robbins (1978) that incorporates the diffusion coefficient and mass flux seems to have been recently solved by Shukla (1996; 1997), assuming constant porosity in a core and a homogeneous layer at the water-sediment interface. In connection with the assumption by Shukla (2002) based on the solution of ADE, new mathematical model had been formulated and well known as Shukla-CIC model.

The objective of this paper are to evaluate the mathematical models which have been used to calculated the sedimentation rate based on  $^{210}\text{Pb}_{\text{ex}}$  activities profile and to analyze many data sets based on old and new models to quantify sedimentation rate as accurately as possible.

## EXPERIMENTAL

### Field procedures

Sediment cores from eight stations along Sarawak and Sabah coastal waters were collected in 2004 (Fig. 1 and Table 1) using gravity box corer with 65 cm length and 10 cm diameter tubes. Cores were sliced every 2 cm (0-2 cm intervals) and kept into sample container before bring to the laboratory for further analyses. Briefly, all the sediment samples were weight, dried in an oven at 60 °C until a constant weight and re-weigh for calculation of porosity. Then, dried sediments were ground into fine particles for homogenous. Sediment sub-samples were also retained for analyses of grain size and organic carbon content.

### Laboratory procedures

Radiochemical separation of  $^{210}\text{Po}$  was performed on 0.5 g of dried sediments samples which spiked with 0.05 – 0.1 g of 0.5 Bq/g  $^{209}\text{Po}$  tracer as analytical yield and digested on a hot plate until dryness with 30 ml of  $\text{HNO}_3$ , 5 ml of  $\text{HClO}_4$  and 10 ml of  $\text{HCl}$  (Tee et al. 2003; Wood et al. 1997). Then, polonium isotopes were spontaneously plated on the silver disc after adding ascorbic acid to prevent Fe deposition and  $^{210}\text{Pb}$  activities were determined from its granddaughter ( $^{210}\text{Po}$ ) with alpha spectrometry during secular

equilibrium stage (Al-Masri et al. 2002; Henderson et al. 1999; San Miguel et al. 2004; Wood et al. 1997). The  $^{210}\text{Pb}_{\text{supp}}$  activity was calculated as the average of several constant  $^{210}\text{Pb}$  determinations in deep sediment layers (below the zone of  $^{210}\text{Pb}$  exponential decline) (Zaborska et al. 2007).



Figure 1. Map showing location of the eight sampling stations in the Sarawak and Sabah coastal waters

Table 1. Sampling location, coordinate, water depth and distance of station to land obtained in this study

Location	Area	Station	Latitude, °N	Longitude, °E	Water depth (m)	Distance from station to mainland (Nautical miles)
Sarawak Coastal waters	Lunau	SR 01	03° 43.7'	110° 01.9'	74.0	113.0
	Kuching	SR 02	04° 00.0'	110° 38.3'	82.0	83.0
	Pulau Bruit	SR 03	04° 23.1'	111° 26.3'	94.0	94.0
	Bintulu	SR 04	04° 52.0'	112° 15.3'	92.0	113.0
	Miri	SR 05	04° 51.1'	113° 29.9'	85.0	32.0
Sabah coastal waters	Tuaran	SB 02	06° 32.7'	115° 53.9'	58.5	28.0
	T. Merudu	SB 03	07° 24.7'	116° 46.9'	70.4	9.5
	Kinabatangan	SB 05	04° 44.9'	118° 38.8'	74.3	10.8

### Sedimentation rate calculation

The principal calculation of sedimentation rate in marine sediment was generally used the following an equation (Tee et al. 2003):

$$A_x = A_0 e^{-\lambda t}; \text{ where } t = x/s$$

$$A_x = A_0 e^{-\lambda(x/s)}$$

The sedimentation rates at eight stations of the Sarawak and Sabah coastal waters were estimated using difference mathematical model based on the activity of  $^{210}\text{Pb}_{\text{ex}}$  derived from above an equation as follows:

i. CIC model

In the CIC model, the dependence between  $^{210}\text{Pb}_{\text{ex}}$  activity after time  $t$  and  $^{210}\text{Pb}_{\text{ex}}$  activity at the sediment surface is given by:

$$A_x = A_0 e^{-\lambda(x/s)}$$

Where  $A_x$  is the  $^{210}\text{Pb}_{\text{ex}}$  activity at layer  $x$  (cm below sediment – water interface),  $A_0$  is the  $^{210}\text{Pb}_{\text{ex}}$  activity of the surface layer,  $\lambda$  is the  $^{210}\text{Pb}$  decay constant (0.031/year) and  $s$  is the sedimentation rate which derived from:

$$s = -\lambda a$$

where  $a$  is a slope of the line derived from a linear regression analysis of the  $\ln(^{210}\text{Pb}_{\text{ex}})$  versus depth profile.

ii. Shukla – CIC model (Shukla, 2002)

The sedimentation rate was estimated using the Shukla's software approached from CIC model were corrected for compaction (calculation was done based on measurements of sediment porosity).

iii. CRS model

$$A_x = A_0 e^{-\lambda t}; \text{ where } t = x/s$$

$$s = \lambda x / [\ln(A_{c0}/A_{cx})]$$

where  $A_{cx}$  is the cumulative  $^{210}\text{Pb}_{\text{ex}}$  activity in sediments of depth  $x$ ,  $A_{c0}$  was calculated as the average of several cumulative  $^{210}\text{Pb}_{\text{ex}}$  activity in deep sediment layers (below the zone of  $^{210}\text{Pb}_{\text{ex}}$  exponential decline),  $s$  is the sedimentation rate (cm/year),  $x$  is the depth of sediment layer (cm),  $t$  is the age of sediment (years) and  $\lambda$  is the  $^{210}\text{Pb}$  decay constant (0.031/year).

iv. ADE model

$$s = \lambda x / [\ln(A_0/A_x)] - D_b/x [\ln(A_0/A_x)]$$

where  $\lambda$  is the  $^{210}\text{Pb}$  decay constant (0.031/year),  $x$  is the depth interval between two levels (cm),  $A_0$  and  $A_x$  activity of  $^{210}\text{Pb}_{\text{ex}}$  at an upper sediment level and at a lower level with the distance  $x$  below  $A_0$ ,  $D_b$  is the bioturbation coefficient ( $\text{cm}^2\text{y}^{-1}$ ). If mixing is negligible ( $D_b = 0$ ), then the above equation can be simplified (Schulz & Zabel 2006):

$$s = \lambda x / [\ln(A_0/A_x)]$$

## RESULTS AND DISCUSSION

### Sediment features

The sediment cores collected at eight stations of Sarawak and Sabah coastal waters exhibited low values of porosity at Station SR 04 but comparable at other stations. The organic carbon content was lower at Station SR 05, SB 02 and SB 03 compared to other stations. Meanwhile, all stations of study area showed different grain size fraction of sediment (Table 2).

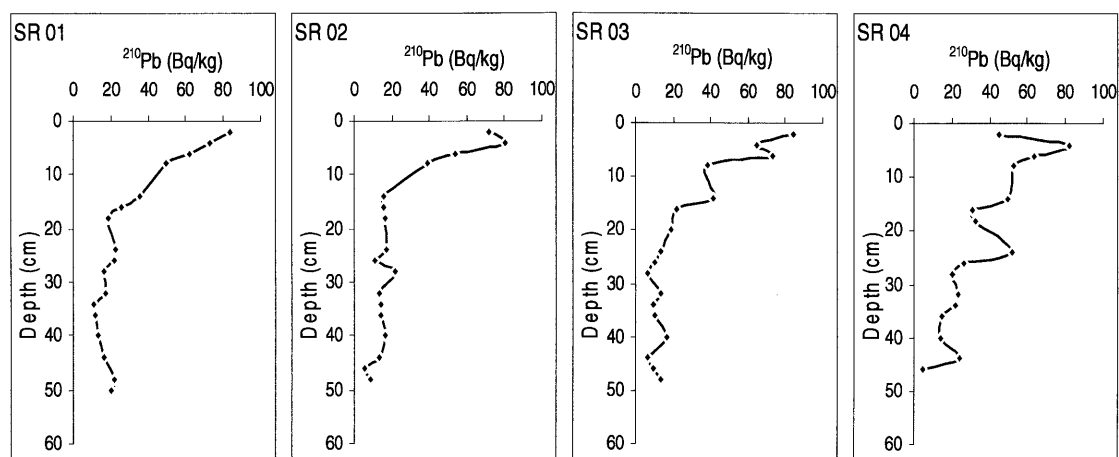
Table 2. Porosity, organic content and grain size of sediment obtained in this study

Station	Porosity (%)	Organic carbon content (%)	Grain size (% fraction)		
			> 63 $\mu\text{m}$ (sand)	4 - 63 $\mu\text{m}$ (silt)	< 4 $\mu\text{m}$ (clay)
SR 01	58 – 73	1.27 – 2.98	2.29	66.50	31.21
SR 02	48 – 73	2.09 – 3.59	3.47	64.64	31.89
SR 03	61 – 75	1.76 – 3.63	0.51	54.32	45.17
SR 04	29 – 75	2.43 – 4.30	6.61	65.87	27.53
SR 05	47 – 73	0.96 – 2.12	5.29	59.24	35.47
SB 02	55 – 71	2.13 – 3.19	15.64	66.38	17.98
SB 03	56 – 72	1.24 – 2.52	8.94	55.88	35.18
SB 05	71 – 86	2.91 – 4.70	1.30	45.53	53.17

### Vertical profiles of $^{210}\text{Pb}$ and $^{210}\text{Pb}_{\text{ex}}$

The vertical profiles of  $^{210}\text{Pb}$  and  $^{210}\text{Pb}_{\text{ex}}$  were generally shown a similar condition and non linear decreased with increasing depth at all sampling stations (Fig. 2 and 3). Additionally, their distributions were normal decay pattern, where fluctuated or uniform activities at the surface layer (< 10 cm depth) as sediment mixing were caused by biological (bioturbation) and physical processes (wave and tidal current), decreased activities with increasing depth as decaying zone (10 – 20 cm depth) and constant activity in the deepest layer (>20 cm depth) (Gokmen et al. 1996). However, the profile of  $^{210}\text{Pb}$  and  $^{210}\text{Pb}_{\text{ex}}$  were indicated obviously fluctuate along sediment core at Station SR 03, SR 04, SR 05 and SB 02. In this situation probably due to strong sediment mixed by biological and physical processes occurred at past time.

Generally, the activity concentrations of  $^{210}\text{Pb}$  and  $^{210}\text{Pb}_{\text{ex}}$  were extremely high which aligned with respect to the high organic carbon content and clay (< 4  $\mu\text{m}$ ) in the sediment core at Station SB 05 (Table 2). Clay and organic carbon content may control  $^{210}\text{Pb}$  and  $^{210}\text{Pb}_{\text{ex}}$  activity concentrations since  $^{210}\text{Pb}$  is tendency to associate with fine particle as clay due to its higher specific surface compared to coarse particles of > 4  $\mu\text{m}$  and indicating the affinity of  $^{210}\text{Pb}$  and  $^{210}\text{Pb}_{\text{ex}}$  with organic carbon content for incorporation into the sediment. Also, this indicated that organic carbon was the main geochemical carrier of this radionuclide in the sampling stations.



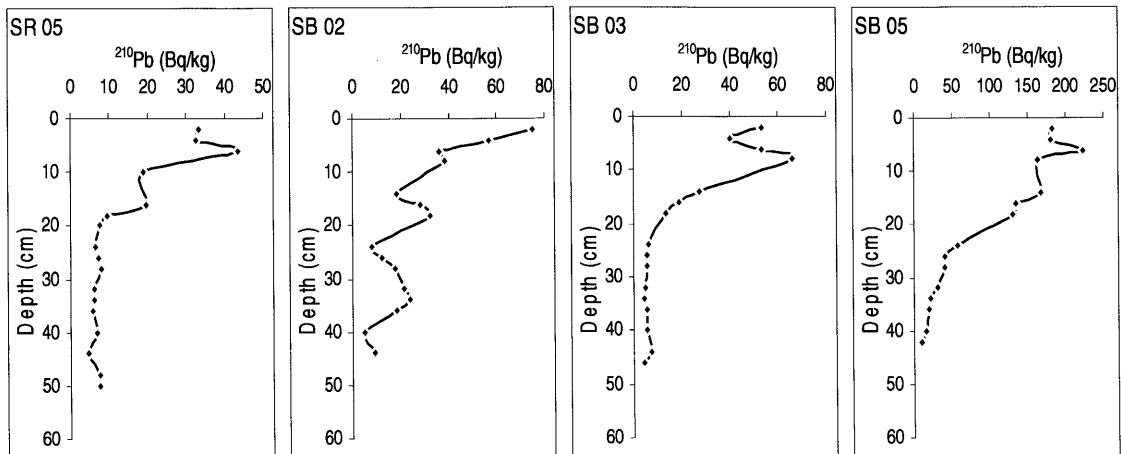


Figure 2. Vertical profiles of  $^{210}\text{Pb}$  in sediment cores at Sarawak and Sabah coastal waters

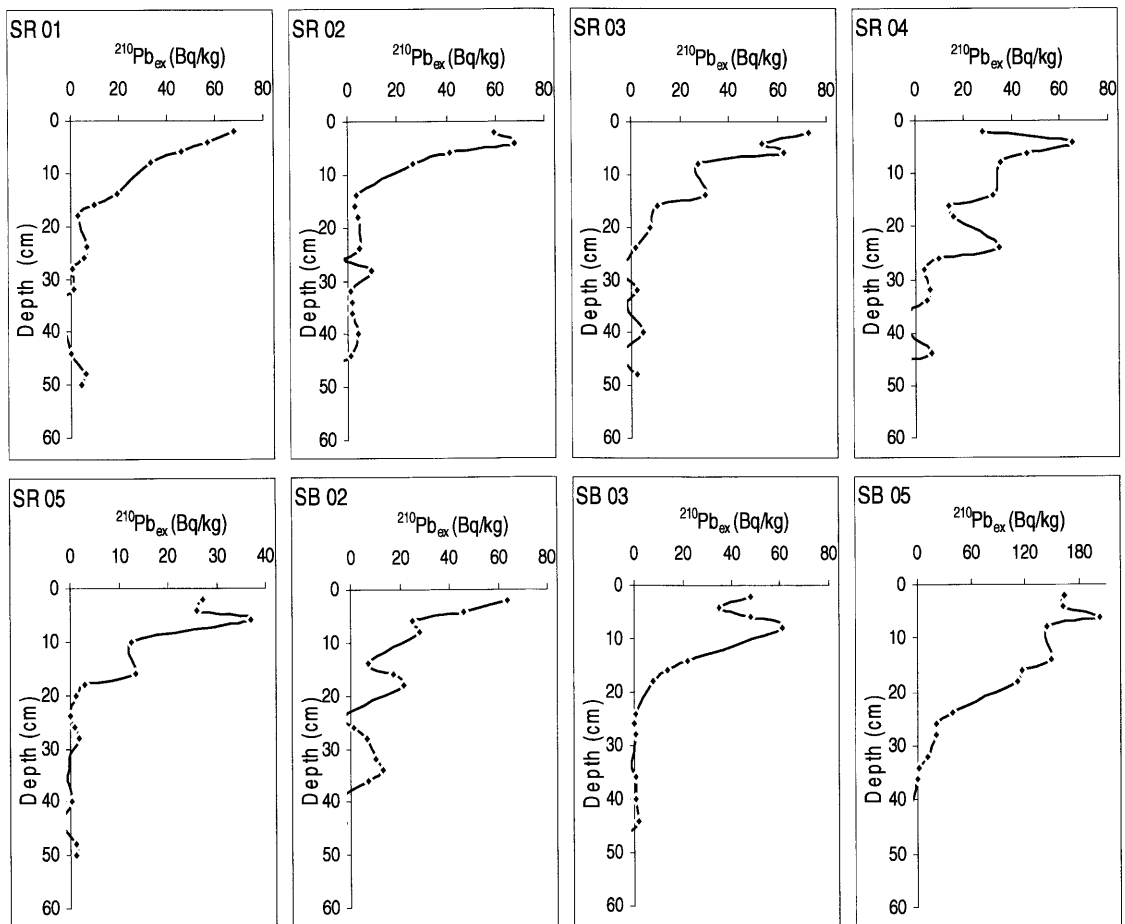


Figure 3. Vertical distributions of  $^{210}\text{Pb}_{\text{ex}}$  in sediment cores at study area

### Sedimentation rate

Estimation of sedimentation rates derived for all stations using four different mathematical models i.e CIC, Shukla-CIC's, CRS and ADE models are presented in Table 3. The sedimentation rates derived by using

four models were generally shown best agreement with similar value or comparable ranging from 0.22 to 0.32 cm/year at Station SB 03 and SB 05 (Table 3).

However, the sedimentation rate value derived by Shukla-CIC model showed big different with other models especially at Station SR 03, SR 04, SR 05 and SB 02. In this case due to strong sediment mixing by bioturbation (biological) and physical process observed along the decay region in the sediment core at those stations (Fig. 2). Consequently, obviously fluctuated on activities of  $^{210}\text{Pb}_{\text{ex}}$  at the decay region, thus, it was difficult to fit a curve slope to determine the sedimentation rate. Therefore, the estimation of sedimentation rate was not accurate. Additionally, less accuracy in calculation of porosity may cause influence the accuracy of sedimentation rate.

Table 3. Sedimentation rates based on four different mathematical models

Station	Sedimentation rate (cm/year)			
	CIC model	Shukla-CIC model	CRS model	ADE model
SR 01	0.30	0.38	0.27	0.29
SR 02	0.42	0.44	0.40	0.30
SR 03	0.42	0.43	0.25	0.34
SR 04	0.38	0.81	0.40	0.42
SR 05	0.43	0.48	0.18	0.29
SB 02	0.53	0.59	0.32	0.29
SB 03	0.27	0.25	0.20	0.27
SB 05	0.29	0.31	0.29	0.32

Generally, estimation of sedimentation rates based on Shukla-CIC's model is to be considerably accurate, compared with other models. It was approved by statistic analysis of independent sample t-test (Table 4), Shukla-CIC:CIC models and Shukla-CIC: CRS models, both have a significant accuracy and suitable ( $p < 0.05$ ), but other pairs showed their accuracy and suitable is not significant ( $p > 0.05$ ). Furthermore, refer to mean value derived from independent sample t-test, the accuracy and suitable of four model can be summarized in the following order i.e. Shukla-CIC > ADE > CRS > CIC. Thus, in this study, Shukla-CIC model found more accurate, reliable and suitable to estimate sedimentation rate at coastal waters.

Table 4. Analysis statistic of independent sample t-test to evaluate a suitable model

Model	Shukla-CIC	CIC	CRS	ADE	Mean
Shukla-CIC		p = 0.019 t = 2.644	p = 0.024 t = 2.525	p = 0.056 t = 2.087	0.4613
CIC	p = 0.019 t = -2.644		p = 0.900 t = -0.128	p = 0.210 t = -1.314	0.2838
CRS	p = 0.024 t = -2.525	p = 0.900 t = 0.128		p = 0.298 t = -1.080	0.2888
ADE	p = 0.056 t = -2.087	p = 0.210 t = 1.314	p = 0.298 t = 1.080		0.3263

## CONCLUSION

Sedimentation rates in Sarawak and Sabah coastal waters were successfully determined using four suggested models through the activity concentration of  $^{210}\text{Pb}_{\text{ex}}$ . The sedimentation rates derived from four models were generally shown in good agreement with similar or comparable value at all stations. However, based on statistical analysis of independent sample t-test indicated that Shukla-CIC model was the most accurate, reliable and suitable technique to determine the sedimentation rate in the sediment cores at Sarawak and Sabah coastal waters.

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