Jurnal Sains Nuklear Malaysia, 2016, 28(1): 1-8
ISSN: 2232-0946

^{210}Po, ^{210}Pb AND ^{210}Po/^{210}Pb IN SEDIMENT CORE FROM SURROUNDING SUNGAI LINGGI ESTUARY

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ABSTRACT

This study was performed to observe the variation in the distribution of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb activity ratio through their vertical profile of the sediment cores taken at surrounding Sungai Linggi estuary. Five sediment cores were taken in February 2011 and were cut to an interval of 2 cm layer. Activity concentrations of ^{210}Po and ^{210}Pb were determined using alpha radiochemical analysis and gamma direct measurement, respectively. Generally, the measured activity of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb were in the ranges of 22.73 – 139.06 Bqkg^{-1} dw., 37.88 – 176.24 Bqkg^{-1} dw. and 0.23 – 1.34, respectively. The variation in the distribution profile for the radionuclides are believed to be influenced by human activities such as agriculture, fertilizer, vehicles, burned fuel fossil and forest, industrial and others via river input from land-base. Other factor is due to organic matter content played important role as the geochemical carrier to transport those radionuclides at study area. It was proved that has a strong correlation between the radionuclide distribution and the sediment composition of organic matter. Furthermore, in those ranges reflected that ^{210}Pb activities were higher than ^{210}Po with an activity ratio average of 0.79. This is probably due to dramatic increase of excess ^{210}Pb supplied from atmospheric deposition, in situ decay of ^{226}Ra and as a result of diagenetic remolibilazation of ^{210}Pb in deeper layes of the sediment column. Thus, those factors are major contributions on the variation of ^{210}Po and ^{210}Pb in the sediment core at surrounding Sungai Linggi estuary.

Keywords: Distribution, ^{210}Pb, ^{210}Po, ^{210}Po/^{210}Pb activity ratio, sediment core

INTRODUCTION

Naturally occurring ^{210}Po ($t_{1/2}$ = 138 days) is the decay product of ^{210}Pb ($t_{1/2}$ = 22.3 years) via ^{210}Bi ($t_{1/2}$ = 5 days) in the ^{238}U decay series and widely distributed in the earth's crust, rivers, oceans, and the atmosphere. Amongst natural radionuclides, ^{210}Po is highly radioactive and radiotoxic, with alpha emitter (particle energy of 5.30 MeV) play a significant role in state radiological effects connected with accumulation of these radionuclides in organisms. The main natural source of polonium in environment is the radioactive decay of uranium (^{238}U), radium (^{226}Ra) and radon (^{222}Rn).The natural concentration of polonium in environment can be enhanced due to human activity (industry, fossil fuel combustion, phosphate fertilizers in agriculture, domestic and industrial sewage) (Daish et al., 2005). In sea water and freshwater, polonium exists in dissolved forms and also is connected with suspended matter. The residence time of polonium in seashore waters is between 18 and 30 days. Remobilization from sediments of polonium in the marine environment is faster in oxygen-free conditions (Carvalho, 1997). Also in rivers, polonium behavior and concentration is differentiated and depends on the geological structure of riverbed (Shaheed et al., 1997).

Lead-210 (^{210}Pb) is also a naturally occurring radioisotope of the ^{238}U daughter decay series, which results from the intermediate decay of ^{226}Ra ($t_{1/2}$ = 1602 years) to the noble gas, ^{222}Rn ($t_{1/2}$ = 3.83 d), by alpha disintegration (Goldberg, 1963). Diffusing into the atmosphere at a constant flux, ^{222}Rn attaches to aerosols that return to the earth as precipitation or dry deposition. This atmospheric addition of ^{210}Pb is in excess of the amount supplied by the in situ decay of ^{226}Ra. Background or supported ^{210}Pb is assumed to be in equilibrium with the decay of ^{226}Ra without the negligible loss of radionuclides. Lead-210 is highly particle reactive and is readily scavenged by organic matter and clay size particles, but under anoxic conditions, ^{210}Pb can be released back to the water column (Benoit, 1988).

Lead-210 is one of the most radiotoxic daughters of ^{226}Ra. Lead-210 decays by β– emission to ^{210}Bi ($t_{1/2}$ = 5.0 d), which again undergoes decay by β– emission to ^{210}Po. Due to its comparatively long half-life, ^{210}Pb is the most abundant of the radioactive lead isotopes. The existence of convenient radioactive lead isotopes in the natural series brought their early use as an environmental tracer in natural processes. Lead-210 is often applied and widely used in sedimentation studies and dating of sediments as well as in marine pollution studies (Appleby, 1992). Further more, the vertical profiles of ^{210}Pb in sediment cores is effectively utilized to evaluate sediment dynamics that include sediment transport, focusing/erosion, mixing in lacustrine and coastal marine systems (Baskaran et al., 2014). It is essential that accurate values for Pb-210 activities determined for sediment samples in order to set up the geochronology for pollutants deposited in sediments (Tanner et al., 2000).

Polonium-210 and ^{210}Pb is the last member in the radioactive decay series of ^{238}U and their abundant in the environment resulted from ^{222}Rn decay in the atmosphere (Narayana et al., 2006). Both of these radionuclides initially originated from the lithosphere and then enter into the marine environment through several natural processes such as erosion, river input, fallout from the atmosphere, deposition, *in situ* decay, decomposition and dispersion (Stralberg et al., 2003). Source of ^{210}Po and ^{210}Pb from the atmosphere radon decay, which are released from ^{226}Ra found in the rocks and soil is the main source into the marine environment (Cochran et al., 1994).

The activity ratio of ^{210}Po/^{210}Pb is a useful parameter to identify the origin of a material particle either biogenic or lithogenic in coastal areas (Sanchez-Cabeza et al., 1998). Apart from the difference half-life, those radionuclides also have different behavior whereas ^{210}Po has higher affinity for organic substances. Thus, this will affect the distribution ^{210}Po and ^{210}Pb in sediments as a result of turbulence in the organic and inorganic materials particle sedimentation. Furthermore, biogeochemical processes which are controlling the distribution of radionuclide might be cause the ratio of ^{210}Po/ ^{210}Pb more than unity (Jia et al., 2003).

Due to these naturally occurring radionuclides of ^{210}Po and ^{210}Pb are also important tools as geochemical tracers in the marine environmental processes. Moreover, geochemical studies of ^{210}Po and ^{210}Pb in sediment core are helpful to understand a continuum of insight into marine environmental processes. Thus, we performed this study with our purposes were mainly to observe the variation in the distribution of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb activity ratio through their vertical profile of the sediment cores taken at surrounding Sungai Linggi estuary.

MATERIALS AND METHODS

Sampling locations were selected around the Sungai Linggi estuary which are covered a latitude of 02° 22.22' N – 02° 24.04'N and longitude of 101° 57.49' E – 101° 59.57' E (Fig. 1). Five sediment cores were collected in different zones of the estuary to achieve the objectives of the study. The cores were collected on 25 January 2011 using a gravity corer with the inner tube of 50 cm length and 7.5 cm diameter. This sampling was performed in intertidal areas, at points with minimum swell influence in order to discount the presence of any physical mixing on a significant scale. Cores were manually sliced every 2 cm (0 – 2 cm intervals) and kept in a zipped plastic bag before brought to the laboratory for further analyses. In the laboratory, detail of the samples preparation was performed according to Zal U'yun and Yii Mei Wo (2013).

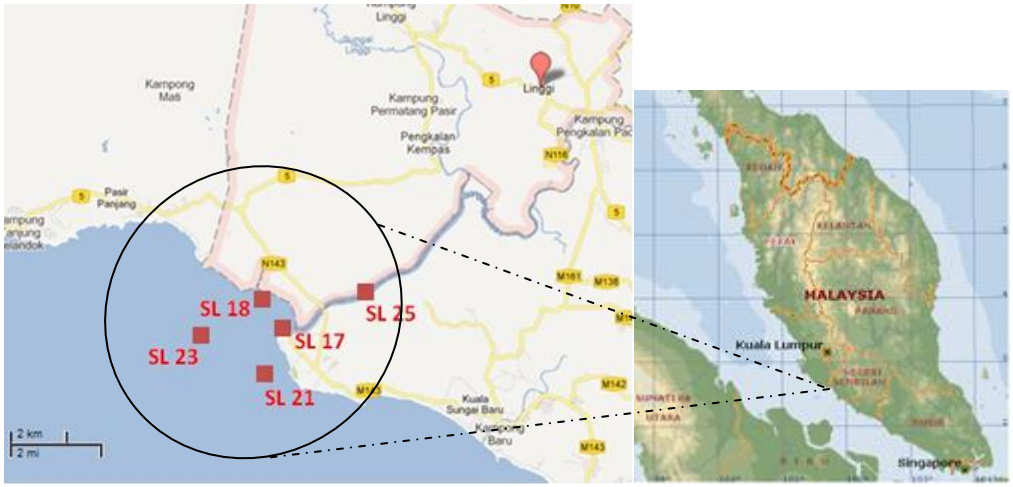

Figure 1: Map showing the locations of sediment core collected in this study

Alpha Spectrometry

The measurements were carried out after the application of a radiochemical method to isolate sequentially the polonium isotopes in aliquots of core sections according to Nita et al. (2013). Polonium was plated by spontanneous deposition onto silver planchets (Jweda and Baskaran, 2011) and was measured by an Alpha Spectrometer. This radiochemical method for purification of the polonium fraction was originally in-house developed in Radiochemistry and Environmental Laboratory (RAS), Malaysian Nuclear Agency.

Gamma Spectrometry

Gamma measurements of ^{210}Pb were carried out using an HpGe detector (Canberra) as reported by Zal U'yun and Yii Mei Wo (2013) in the other previous paper. Its activity was measured directly via 46.54 keV energy peak.

QA/QC

The accuracy and precision of the radiochemical method were estimated using certified reference materials. The polonium yield in the sediment samples ranged from 85% to 98%. The results of

^{210}Po and ^{210}Pb activity concentration in sediment samples are given with 2σ of uncertainty calculated for 95% confidence intervals.

RESULTS AND DISCUSSION

The ranges of activity concentration for ^{210}Po, ^{210}Pb, their ratio and content of organic matter (OM) in sediment core samples collected from Sungai Linggi estuary are presented in Table 1. Generally, the activity concentrations of ^{210}Po measured in sediment cores were varied with the ranges from 22.73 Bqkg^{-1}dw. – 139.06 Bqkg^{-1}dw. Refer to those ranges, it was proven by one-way ANOVA analysis that there has significantly difference of ^{210}Po activity concentrations (p = 0.000) in the sediment core depending on sampling stations. This finding indicated the highest activity concentration of ^{210}Po was measured in Sungai Linggi estuary from SL25 (91.71 – 139.06 Bqkg^{-1}dw.) and the lowest from SL23 (31.53 – 62.58 Bqkg^{-1}dw.). In other world, the highest activity concentration of ^{210}Po in following order of sampling station are SL 25 > SL 17 > SL 18 > SL 21 > SL 23. This indicates the sampling station SL 25, SL 17 and SL 18 has received more input of ^{210}Po from the mainland due to human activities mainly using a phosphate fertilizer in agriculture. This also might be due to transported polonium eluted from soil and contained in dry and wet atmospheric precipitation before deposit onto sediment (Skwarzec and Jahnz, 2007). In the case of the higher activity concentration of ^{210}Po at SL 25 which located in the river channel is due to the principle source of polonium from rocks and soil erosion as well as precipitation. Moreover, the downstream river contains polonium substances eluted from soil by water (Dahlgaard, 1996; Shaheed et al., 1997).

Table 1: ^{210}Po, ^{210}Pb, ^{210}Po/^{210}Pb and organic matter in sediment cores of Sungai Linggi estuary

Sampling station	**Activity concentration (Bqkg^{-1} dw.)**		**^{210}Po/^{210}Pb**	**Organic matter (OM) (%)**
	^{210}Po	**^{210}Pb**		
SL 17	62.48 – 123.98	92.85 – 127.30	0.53 – 1.24	10.56 – 13.64
SL 18	39.75 – 104.26	110.45 – 176.24	0.23 – 0.87	7.05 – 12.12
SL 21	22.73 – 69.32	54.21 – 109.77	0.38 – 1.07	6.24 – 8.84
SL 23	31.53 – 62.58	37.88 – 56.98	0.70 – 1.34	4.10 – 5.87
SL 25	91.71 – 139.06	105.84 – 147.73	0.70 – 1.16	9.74 – 13.06

It is interesting to note that ^{210}Po activity concentrations in sediment core decrease in coastal (SL 21 and SL 23), which corresponds to dissolution/desorption of ^{210}Po particulate or sediment particle with increased salinity. Furthermore, the Pearson correlation proved that there exists highly significant correlated between ^{210}Po and OM with R = 0.72 (SL 21) and R = 0.91 (SL 23) (Fig. 2). This correlation suggested that river contribution is the main source of the terrigenous ^{210}Po and organic material delivered to the sediment. In other word, this revealed the distribution of ^{210}Po in the sediments was probably controlled by OM which is played important role as geochemical carrier to transport this radionuclide at study area. Align with decreased of ^{210}Po activities at station SL 21 and SL 23 might be due to diagenetic process or suggesting increasing OM loadings to the water system can be accounted for by steady state diagenesis of ^{210}Po.

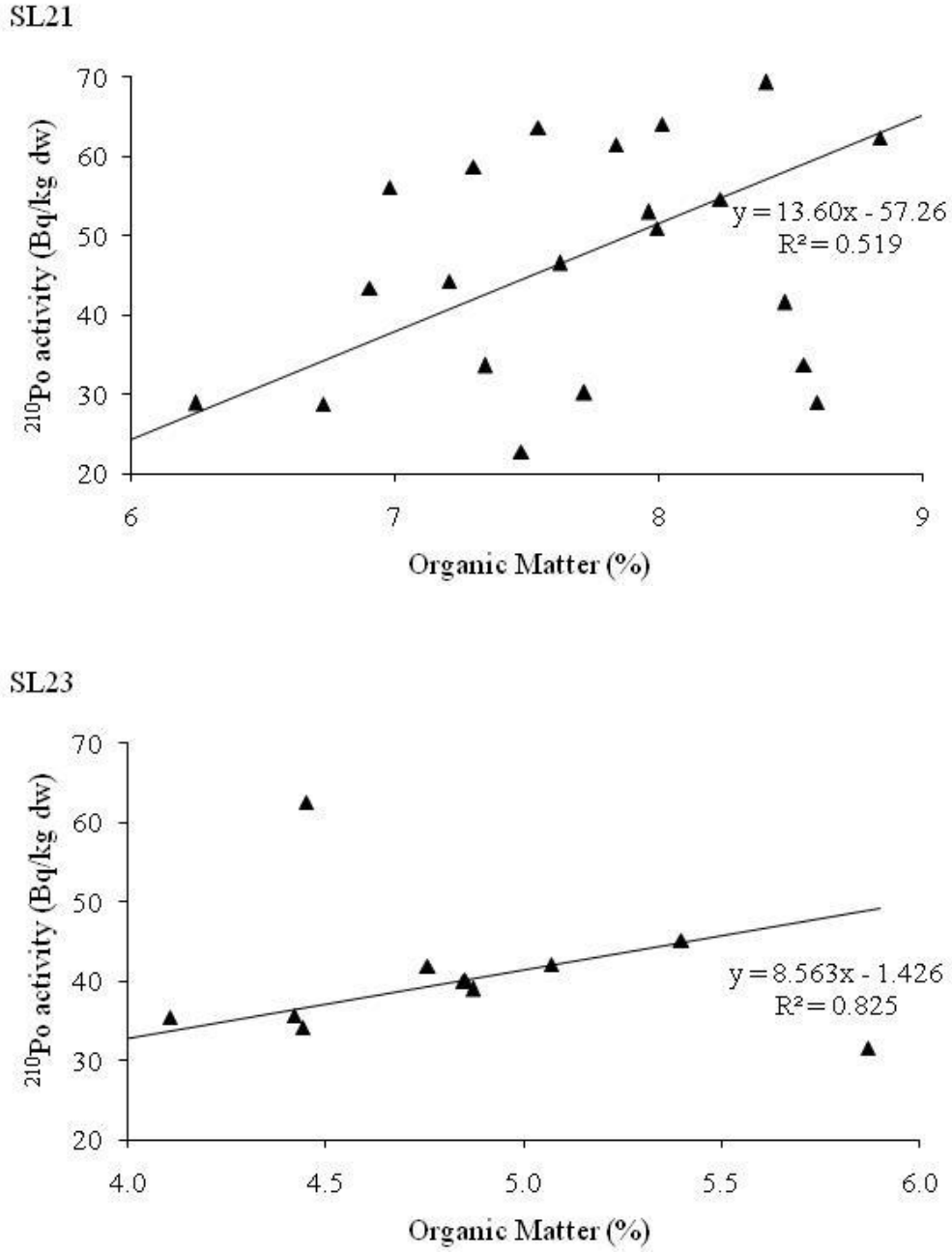

Figure 2: Highly significant correlation of ^{210}Po activity with the organic matter at station SL 21 and SL 23

Meanwhile, activity profiles of ^{210}Pb in sediment core at all sampling stations were ranged from 37.88 Bqkg^{-1}dw. to 176.24 Bqkg^{-1}dw. Align with that, an analysis of One-way ANOVA proved that there have significant different activities (p = 0.000) of ^{210}Pb in sediment core collected in Sungai Linggi estuary. It is observed that station SL18 and SL 23 was highest and lowest activity of ^{210}Pb, respectively. This indicating the sampling station SL18 has received more input of ^{210}Pb from the mainland due to human activities mainly using a phosphate fertilizer in agriculture, vehicles, burned fuel fossil and forest. The lower activity of ^{210}Pb at station SL 23 probably due to this radionuclide

from the mainland may be dissolute and dilute when the riverine suspended sediment contacted with seawater. Meanwhile, the different of activity concentrations of ^{210}Pb observed at the different sampling stations could be in connection with various geochemical conditions and sediment characteristics. Even though both (^{210}Po and ^{210}Pb) are different behavior and geochemical properties, but both radionuclides have a strong positive correlation (R = 0.76) at station SL 23. This might be probably due to both radionuclides are supplied from the same source or same environment (Fig. 3). Thus, this relationship revealed that ^{210}Po was supplied by *in situ* decay from its parent ^{210}Pb.

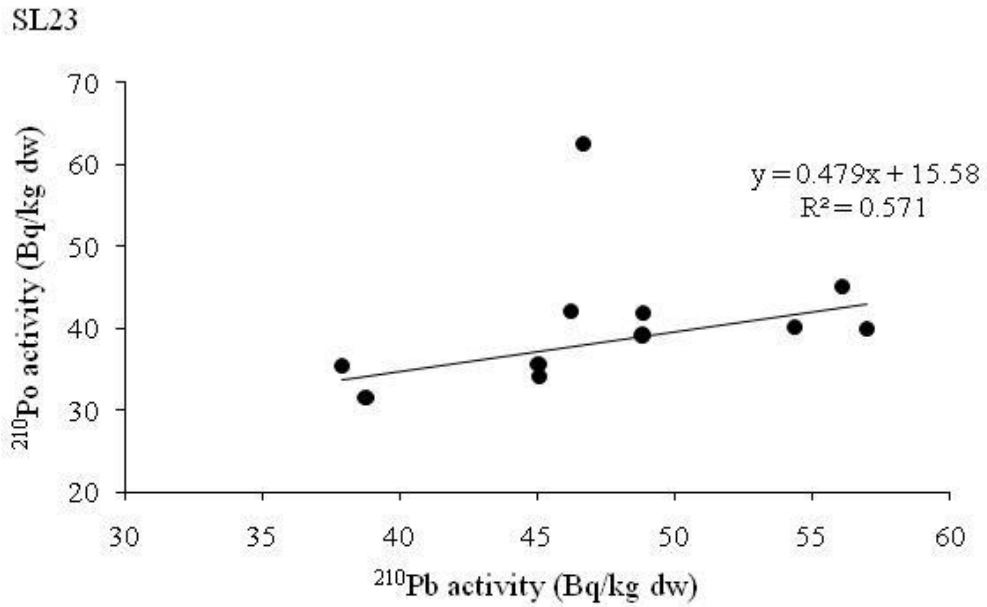

Figure 3: Strong Significant correlation between ^{210}Po with its parent ^{210}Pb in sediment core at station SL 23

Activity ratios of ^{210}Po/^{210}Pb in sediment cores taken at Sungai Linggi estuary are given in Table 1. Generally, the activity ratios measured were varied with the ranges from 0.23 to 1.34. Refer to those ranges, it was proven by One-way ANOVA analysis that there has significantly difference of ^{210}Po/^{210}Pb activity ratios (p = 0.000) in the sediment cores depending on sampling stations. This variation of activity ratios could be linked to their different chemical reactivity, biological enrichments, and volatility (Kim et al., 2011). Furthermore it is related to the sampling location and the anthropogenic (human) activities on the surrounding areas, whose effluents may changing the concentrations of naturally occurring radionuclides in the environment especially ^{210}Po and ^{210}Pb. Higher activity ratios i.e more than unity (^{210}Po/^{210}Pb $\geq$ 1) at some stations probably due to rapid and active regenerated of ^{210}Po by its parent, ^{210}Pb compared to production of ^{210}Pb via ^{226}Ra decay. Furthermore, this high ratio interesting to note that enrichment of ^{210}Po produced from efficient transport in water column prior to deposit on seabed. Obviously the ^{210}Po/^{210}Pb activity ratio is low (0.23) might be linked to the strong affinity of ^{210}Po with the particulate matter. On the other hand, this is probably due to dramatic increase of excess ^{210}Pb supplied from atmospheric deposition, in situ decay of ^{226}Ra and as a result of diagenetic remolibilazation of ^{210}Pb in deeper layer of the sediment column. For the activity ratio less than unity (^{210}Po/^{210}Pb < 1) that can be seen at station SL 21probably important to note that most of the excess ^{210}Pb at the water-sediment interface is derived from atmospheric deposition at the air-water interface (Baskaran, 2011).

Generally, the variation in the distribution profile for the radionuclides of ^{210}Po, ^{210}Pb and its activity ratio of ^{210}Po/^{210}Pb are influenced by human activities such as agriculture, fertilizer, vehicles, burned fuel fossil and forest, industrial and others via river input from land-base. Furthermore, radionuclides are wide dispersed in the hydrosphere and are influenced by physical, biological and chemical (Dahlgaard, 1996; Shaheed et al., 1997).

CONCLUSIONS

In this work, it can be concluded that the variation in the distribution sediment core profile for the radionuclides of ^{210}Po, ^{210}Pb and its activity ratio of ^{210}Po/^{210}Pb in Sungai Linggi estuary are believed to be influenced by human (anthropogenic) activities such as agriculture, fertilizer, vehicles, burned fuel fossil and forest, industrial and others via river input from land-base. Other factor is due to organic matter content played important role as geochemical carrier to transport those radionuclides at study area.

ACKNOWLEDGMENTS

The authors would like to thank the Malaysian Nuclear Agency for the support in allocating fund through PQRD seed-fund and staff of RAS in assisting throughout this study.

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