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ANTHROPOGENIC AIRBORNE DEPOSITIONS OF ^{210}Po, ^{210}Pb AND ^{210}Po/^{210}Pb IN THE MOSSES AND SURFACE SOILS AT THE VICINITY OF A COAL-FIRED POWER

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ABSTRACT

Anthropogenic airborne depositions of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb in the mosses and surface soils collected at the vicinity of a coal-fired power plant were studied. The purpose of the study was to determine activity concentrations of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb for assessing their variation accumulation in the mosses and surface soils collected at the vicinity of a coal-fired power plant. Other purposes were to determine their concentration factor (CF) in relation to track the potential source of those radionuclides and to identify most suitable moss species as a biological indicator for atmospheric deposition contaminants. In this study, different species of moss Leucobryum aduncum, Campylopus serratus, Syrrhopodon ciliates and Vesicularia montagnei were collected in May 2011 at the area around 15 km radius from Tanjung Bin coal-fired power plant located in Pontian, Johor. The activity concentrations of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb in mosses were in the range of 76.81 ± 4.94 – 251.33 ± 16.33 Bq/kg dry wt., 54.37 ± 3.38 – 164.63 ± 11.64 Bq/kg dry wt. and 1.10 – 2.00, respectively. Meanwhile the ranges for those radionuclides in the surface soil were 33.53 ± 2.10 – 179.67 ± 12.15 Bq/kg dry wt., 20.55 ± 1.33 – 106.62 ± 6.64 Bq/kg dry wt. and 1.61 – 2.44, respectively. Corresponding high ability of Leucobryum aduncum to accumulate more ^{210}Po and ^{210}Pb, wide geographical distribution, most abundant and high CF, therefore, the findings can be concluded this species was the most suitable as a biological indicator for atmospheric deposition contaminants such as ^{210}Po and ^{210}Pb. Furthermore, it is clear the accumulation of ^{210}Po and ^{210}Pb in mosses might be supplied from various sources of atmospheric deposition such as coal-fired power plant operation, industrial, agriculture and fertilizer activities, burned fuel fossil and forest; and other potential sources. Meanwhile, the main source of ^{210}Po and ^{210}Pb in surface soil is supplied from the in situ decay of radon and radium.

Keywords: Accumulation, atmospheric deposition, mosses, ^{210}Pb, ^{210}Po, surface soil

INTRODUCTION

Mosses have been widely used as bio-monitors for assessing the atmospheric deposition of heavy metals and radionuclides. Due to the lack of root system, they depend on surface absorption of nutrients and accumulate fallout radionuclides from atmosphere. Therefore, they are often used for bio-monitoring pollution. In these plants, the accumulation degree is much higher than in vascular plants growing in the same habitats. Some species of moss have been widely used in nationwide monitoring and surveys (Bargagli et al.,1995; Berg et al., 1995; Figueira et al., 2002).

The operation of the coal-fired power plant is a subject of public concern due to coal-fired is one of the major. Sources of increased natural radioactivity in the atmosphere in which it can be affected the surrounding environment. Therefore, few factors such as the concentration in coal, the ash content, the combustion temperature, the partitioning between bottom ash and fly ash and the efficiency of the emission control device are taking into account of the amounts of natural radionuclides released to the atmosphere (Ugur et al., 2004). On the other hand, despite stack filtration and other trapping methods, soils in the environment of coal-fired power plants are often found to be enriched in such ash-borne particles (Flues et al., 2002; Ugur et al., 2003).

Polonium-210 (^{210}Po; $t_{1/2}$ =138 days) and ^{210}Pb ($t_{1/2}$ = 22 years) are product of the ^{238}U decay series and derived naturally from the lithogenic minerals in the earth's crust and subsoil. Then, they released into the atmosphere via the decay of ^{222}Rn ($t_{1/2}$ = 3.8 days), the daughter of ^{226}Ra ($t_{1/2}$ = 1,602 years) as a main source in the environment. Polonium-210 and ^{210}Pb return to the earth surface by wet and dry deposition after being attached to airborne particles (Dahlgaard, 1996; Sert et al., 2011).These radionuclides can also build up artificially in the environment as a result of man's activities such as fossil fuel burning power plant, fertilizer industries, waste discharge from phosphate, oil and gas industries; and exhaust gasses of traffic. They also source from the atmosphere as a result of combustion of coal-fired power plant (Carvalho et al., 2010; Godoy et al., 2008; Sert et al., 2011; Villa et al., 2009). Atmospheric fallout of this radionuclide to the soil together with aerosol particles by washout and sedimentation in which can be contaminated the plants and the top layer of soil (Ugur et al., 2003). In fact, most of the natural radioactivity content of ^{210}Po and ^{210}Pb in wild leafy plants, mosses and lichens as a result of the direct deposition of ^{222}Rn daughters from atmospheric precipitation (Hill, 1960).

Nevertheless, no work related to mosses radioactivity has done in Malaysia in particular around coal-fired power plant; however few studies on distribution of heavy metal in mosses had been reported (Low et al., 1985; Mohd Zahari et al., 2012). In the present work, we were studying the several species of moss and surface soil in accumulating of ^{210}Po and ^{210}Pb which are collected at the vicinity of a coal-fired power plant. It is aimed to determine the activity concentrations of ^{210}Po, ^{210}Pb and its activity ratio (^{210}Po/^{210}Pb) for assessing their variation accumulation in the mosses and surface soils collected at the vicinity of a coal-fired power plant. Other purposes were to determine their concentration factor (CF) in relation to track the potential source of those radionuclides and to identify most suitable moss species as a biological indicator for atmospheric deposition contaminants.

MATERIALS AND METHODS

Study Area

Study area is covering the area around 15 km radius from the Tanjung Bin coal-fired power plant situated in Pontian in the southern of Johor, opposite the port of Tanjung Pelepas and beside Tanjung Piai, the southernmost tip of mainland Asia. It is located within latitude N1^{o} 200′ and longitude E103^{o} 320′ (Fig. 1). The area was generally fairly warm and humid all year round, with temperature averaged from 28 to 30^{o}C in daytime and most of the study areas are covered with plantation of palm tree.

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Sampling

Four different species of moss namely *Leucobryum aduncum, Campylopus serratus, Syrrhopodon ciliates and Vesicularia montagnei* were collected from 13 selected sampling station in May 2011 at the area around 15 km radius from Tanjung Bin coal-fired power plant located in Pontian, Johor (Figure 1). At each sampling station, three sub-samples of mosses were taken randomly and combined as a one sample. For soil, surface samples at the depth up to 15 cm were collected at the same sampling point with the mosses.

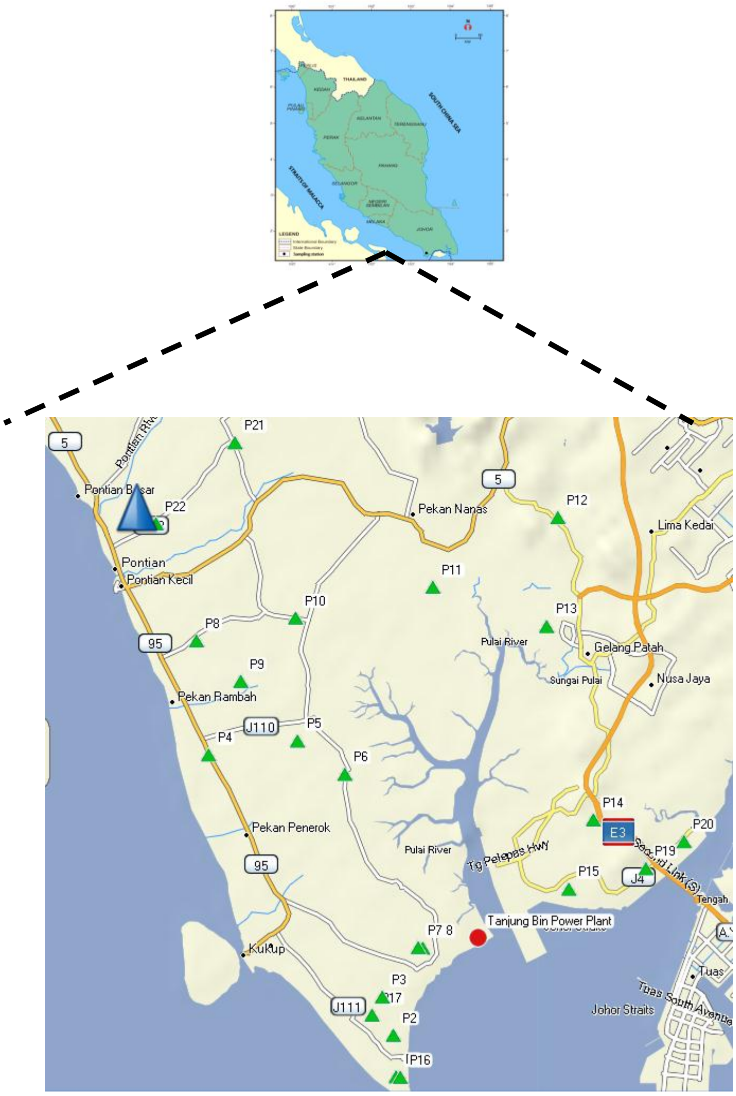

Figure 1: Map showing the locations of moss and surface soil collected in this study

Sample Preparation

The moss samples were sorted to remove as much extraneous material as possible. Detailed information of collected samples such as fresh weight, sample characteristics, date and sampling location were recorded and the samples were brought back to the laboratory for further analysis. The collected moss and soil samples were oven-dried at 60^{o}C for 1-2 days or until constant weight is achieved. Dried samples were then fine-ground, homogenized and sieved (Nita et al., 2013).

Measurement of ^{210}Po

The measurements of ^{210}Po were carried out after the application of a radiochemical method to isolate sequentially the polonium isotopes in digestion solution of mosses and soils according to Nita et al. (2013). This radiochemical method for purification of the polonium fraction was originally in-house developed in Radiochemistry and Environmental Laboratory, Malaysian Nuclear Agency. The alpha activities of ^{209}Po ($t_{1/2}$ = 102 years) and ^{210}Po ($t_{1/2}$ = 138 days) were counted using Alpha Spectrometry System for 24 hours through their alpha particle emission energy of 4.98 and 5.30 MeV, respectively.

Measurement of ^{210}Pb

Measurement of ^{210}Pb was carried out using directly counting of HpGe detector (Canberra) for 54000 seconds according to detail explanation by Zal U'yun and Mei Wo (2013) in their previous paper. Its activity was measured directly via 46.54 keV energy peak.

RESULTS AND DISCUSSION

Activity Concentration of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb in Mosses

Activity concentrations of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb in four different moss species of *Leucobryum aduncum, Campylopus serratus, Syrrhopodon ciliates dan Vesicularia montagnei* are given in Table 1. The ranges of ^{210}Po and ^{210}Pb activity concentration in all species of mosses were found to be varied with 76.81 ± 4.94 – 251.33 ± 16.33 Bq/kg dw. and 54.37 ± 3.38 – 164.63 ± 11.64 Bq/kg dw., respectively. We suggest that various sources of atmospheric deposition such as coal-fired power plant operation, industrial, agriculture and fertilizer activities (include agricultural-waste burning), burned fuel fossil and forest; and other potential sources could serve as an additional source of ^{210}Po and ^{210}Pb accumulated in mosses at study area. Meanwhile, activity ratios of ^{210}Po/^{210}Pb were ranged of 1.10 to 2.00. This variation could be linked to the differences in accumulation properties of moss species. The different of activity concentrations of ^{210}Po and ^{210}Pb observed at the different sampling stations could be in connection with various ecological conditions and individual moss characteristics (Nita et al., 2013). In line with that, it was observed that moss species of *C. serratus* and *L. aduncum* have the highest activity concentrations of ^{210}Po (*C. Serratus*: 147.93 – 251.33 Bq/kg dw.; *L. aduncum*: 113.43 – 206.38 Bq/kg dw.), ^{210}Pb (*C. Serratus*: 73.85 – 164.63 Bq/kg dw.; *L. aduncum*: 78.27 – 131.12 Bq/kg dw.) and ^{210}Po/^{210}Pb (*C. Serratus*: 1.36 – 2.00.; *L. aduncum*: 1.14 – 1.95). Eventhough *L. aduncum* was observed to have the lower capture efficiency of radionuclide compared to *C. Serratus*, but it was present at all sampling station except P14 and P17 (Table 1). In addition, it was supported in other report that species commonly present everywhere and abundantly growth particularly in the peat area due to the geographical and the climatic conditions. This due to the precipitation of rainwater affects on the accumulation, mobility and leaching of ^{210}Po and ^{210}Pb in *L. aduncum* (Poikolainen, 2004).

On the other hand, *L. aduncum* had high water retention properties of the leucocytes (Tan and Ho, 2008; Chantanaorrapint, 2010). They concluded the different life forms of bryophytes were able to store different amounts of interception water and cushions had a high one. Upon wetting, the leaves become widely spreading provides a large surface area for the deposition of fallout nuclides. Due to this open field, it was not well protected from any direct radionuclide deposition particularly for

^{210}Poand ^{210}Pb (Chantanaorrapint, 2010). Thus, these unique characterizations of *L. aduncum* were supported to explain this species capability to accumulate more ^{210}Po and ^{210}Pb and it was considered as the most suitable bio-indicator for ^{210}Po and ^{210}Pb deposition in study area.

Activity Concentration of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb in Surface Soils

The activity concentrations of ^{210}Po, ^{210}Pb and ^{210}Po/^{210}Pb in surface soil were ranged from 33.53 ± 2.10 – 179.67 ± 12.15 Bq/kg dw., 20.55 ± 1.33 – 106.62 ± 6.64 Bq/kg dw. and 1.61 – 2.44 (Table 1). Refer to those ranges, the activity concentrations of ^{210}Po and ^{210}Pb in the surface soil were found to be higher at some sampling stations might be due to utilised of fertilizer in agriculture activities at surrounding area. This also probably was related to the releases of radionuclides from the operation of coal-fired power plant which were transported via wind, rain and other factors that could affect the results to some extents. Meanwhile, the lowest activity concentration of ^{210}Po and ^{210}Pb would be attributed to the fact the sampling site is suitable for agriculture, so almost all the soils of sampling areas are mixed up by tillage therefore this can be reduced the ^{210}Po and ^{210}Pb activity concentration on the surface layer (Nita et al., 2013). Furthermore, the soil of the study area is classified as peat soil which may influence the adsorption of metals and radionuclides.

The high ^{210}Po/^{210}Pb in surface soil which more than unity at all sampling station could be explained that ^{210}Po is supplied from the in situ deposition of radon decay and its daughters in the soil itself (Nita et al., 2013). Other source of ^{210}Po in surface soil might be supplied from human activities such as coal-fired power plant operation, industrial, agriculture and fertilizer activities, burned fuel fossil and forest. Meanwhile, in the case whereas the lower activity of ^{210}Pb in the soils may be attributed to the mixing of soil, soil erosion and also the washout of ^{210}Pb from the surface soil during the flood in raining season, exposed to weathering process as well as cultivation process.

Concentration Factor (CF)

Variability in the accumulation of contaminants in particular radionuclides from media such as water, soil and sediment to plant is an important source of uncertainty in generic models for the prediction of transfer factor of contaminants to the plant and assist to identify the suitable bio-indicator. The level of radioactive or other contaminants in plant (i.e: moss, lichen, vascular plants etc.) is commonly defined in terms of a concentration factor (CF) where:

$$CF = \frac{\text{Activity concentration per kg of the plant}}{\text{Activity concentration per kg of media (eg. soil or sediment or water)}}$$

It is interesting to note that the mosses showed a significantly higher level of ^{210}Po and ^{210}Pb activity concentration compared to soil at almost sampling stations. This finding could be explained as mosses grow on other media (i.e. soil, tree bark etc.) and they depend on the soil only for support and not for nutrients. They accumulate their nutrients from atmospheric moisture and dust particles. As a result of this, the radon daughters which are attached to the dust particles are absorbed and accumulated by the mosses over a period, resulting in a higher activity level of ^{210}Po and ^{210}Pb in the mosses (Karunakara et al., 2000). On the other hand, the value of CF for ^{210}Po and ^{210}Pb were varied with the ranges of 0.78 – 3.42 and 1.08 – 4.74, respectively. Refer to those ranges the moss species of *L. aduncum* has a highest value of CF for ^{210}Po and ^{210}Pb compared to *C. Serratus*, *S. Ciliates* and *V. montagnei.* This indicated this species more ability to capture and accumulate airborne particulates which are contained ^{210}Po and ^{210}Pb by both passive and active processes through an extra cellular ion-exchange process (Knight et al.,1961). Therefore, this species of *L.*

aduncum has some advantages compared to other species due to its higher accumulating capacity, thus explains the increment of ^{210}Po and ^{210}Pb activity concentration in this moss species. Corresponding high CF of ^{210}Po and ^{210}Pb in *L. aduncum* therefore, the findings can be concluded this species was the most suitable as a biological indicator for atmospheric deposition contaminants such as ^{210}Po and ^{210}Pb.

Table 1: Activity concentration of ^{210}Po, ^{210}Pb, $^{210}Po/^{210}Pb$ and CF in the moss and surface soil at the vicinity of a coal-fired power plant

Sampling Station	**Moss Species**	**Moss**			**Soil**			**Concentration Factor, CF (Unitless)**	
		^{210}Po (Bq/kg dw.)	**^{210}Pb (Bq/kg dw.)**	**$^{210}Po/^{210}Pb$**	**^{210}Po (Bq/kg dw.)**	**^{210}Pb (Bq/kg dw.)**	**$^{210}Po/^{210}Pb$**	**^{210}Po**	**^{210}Pb**
P4 - Sg. Karang	*L. aduncum*	126.66 ± 8.67	98.88 ± 6.18	1.28	105.28 ± 7.20	57.53 ± 3.91	1.83	1.20	1.72
P5 – Kg. Peradin	*L. aduncum*	173.69 ± 13.79	88.98 ± 5.91	1.95	112.09 ± 7.93	65.75 ± 4.77	1.71	1.55	1.35
	C. serratus	147.93 ± 11.75	73.85 ± 4.99	2.00				1.32	1.12
P6 – Kg. Buluh Kasap	*L. aduncum*	143.69 ± 9.83	120.10 ± 8.71	1.20	183.93 ± 12.01	75.38 ± 4.94	2.44	0.78	1.59
P7 – Tg. Bin 1	*L. aduncum*	129.10 ± 8.96	79.32 ± 5.27	1.63	72.90 ± 5.06	30.64 ± 2.01	2.38	1.77	2.59
	S. ciliates	108.61 ± 7.43	71.53 ± 4.69	1.52				1.49	2.33
P8 – Pt. Tegong	*L. aduncum*	113.43 ± 7.30	78.27 ± 5.29	1.45	137.61 ± 9.31	72.74 ± 5.14	1.89	0.82	1.08
	C. serratus	199.61 ± 13.03	130.99 ± 9.67	1.52				1.45	1.80
	S. ciliates	129.83 ± 8.12	77.26 ± 5.10	1.68				0.94	1.06
P10 – Pt. Gantong	*L. aduncum*	134.80 ± 9.03	91.14 ± 5.88	1.48	103.90 ± 6.82	52.26 ± 3.41	1.99	1.30	1.74
	S. ciliates	120.97 ± 8.93	78.82 ± 5.17	1.54				1.16	1.51
P11 – Kg. Jeram Batu	*L. aduncum*	206.38 ± 13.54	130.11 ± 8.15	1.59	143.85 ± 9.37	75.04 ± 4.84	1.92	1.44	1.73
	C. serratus	192.64 ± 12.54	125.88 ± 8.36	1.53				1.34	1.68
	S. ciliates	125.61 ± 8.08	85.54 ± 5.78	1.47				0.87	1.14
P13 – Bt. Tempurung	*L. aduncum*	130.27 ± 8.81	97.46 ± 6.59	1.34	38.08 ± 2.49	20.55 ± 1.33	1.85	3.42	4.74
	V. montagnei	102.01 ± 7.53	83.62 ± 6.06	1.22				2.68	4.07
P15 – Kg. Pok Kecil	*L. aduncum*	112.88 ± 7.26	86.66 ± 5.68	1.30	66.21 ± 4.38	34.11 ± 2.21	1.94	1.71	2.54
P17 – Kg. Perpat	*V. montagnei*	152.82 ± 9.55	117.80 ± 8.33	1.30	53.65 ± 3.89	28.39 ± 1.94	1.89	2.45	4.15
P18 – Tg. Bin 2	*L. aduncum*	114.05 ± 7.55	77.35 ± 5.29	1.47	78.49 ± 5.12	36.45 ± 2.37	2.15	1.45	2.12
	C. serratus	159.40 ± 10.78	111.19 ± 7.23	1.43				2.04	3.05
	S. ciliates	125.51 ± 9.27	92.26 ± 5.95	1.36				1.60	2.53
P19 – Kg. Kupang	*L. aduncum*	100.81 ± 6.61	88.07 ± 5.68	1.14	49.39 ± 3.10	29.86 ± 1.70	1.65	2.04	2.95
	V. montagnei	76.81 ± 4.94	54.37 ± 3.38	1.41				1.56	1.82
P21- Pt. Selangor	*L. aduncum*	204.87 ± 13.44	131.12 ± 8.46	1.56	179.67 ± 12.15	106.62 ± 6.64	1.69	1.14	1.23
	C. serratus	251.33 ± 16.33	164.63 ± 11.64	1.53				1.40	1.54

CONCLUSIONS

In this study, the activity concentrations of ^{210}Po ^{210}Pb and ^{210}Po/^{210}Pb in all species of mosses were found to be varied with the range of 76.81 ± 4.94 – 251.33 ± 16.33 Bq/kg dw., 54.37 ± 3.38 – 164.63 ± 11.64 Bq/kg dw. and 1.10 – 2.00, respectively. Meanwhile the ranges for those radionuclides in the surface soil were 33.53 ± 2.10 – 179.67 ± 12.15 Bq/kg dw., 20.55 ± 1.33 – 106.62 ± 6.64 Bq/kg dw. and 1.61 – 2.44, respectively. This variation can be concluded that these radionuclides might be supplied from various sources of atmospheric deposition such as coal-fired power plant operation, industrial, agriculture and fertilizer activities, burned fuel fossil and forest, *in situ* decay of radon and radium; and other sources. Corresponding high ability of *Leucobryum aduncum* to accumulate more ^{210}Po and ^{210}Pb, wide geographical distribution, most abundant and high CF, therefore, the findings can be suggested this species was the most suitable as a biological indicator for atmospheric deposition contaminants such as ^{210}Po and ^{210}Pb.

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