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Natural Radioisotopes of Pb, Bi and Po in the Atmosphere of Coal Burning Area

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Abstract

This paper is discussing the changes of natural radionuclides ^{210}Pb, ^{210}Bi and ^{210}Po in atmospheric samples (rainwater and solid fallout) caused by Sultan Salahuddin Abdul Aziz coal-fired Power Plant (SSAAPP) operation. We also describe the seasonal changes of ^{210}Pb, ^{210}Bi and ^{210}Po to the monsoon seasons in Peninsular Malaysia. Bulk atmospheric trap was used to collect atmospheric samples for five months (7 Feb 2007 to 27 July 2007) and placed within the SSAAPP area. The natural radionuclide activity levels in the atmosphere were affected by local meteorological conditions to impact their variance over time. As a result, the natural radionulides were increased from the ambient value in atmospheric particles (solid fallout), which related to coal combustion by-product releases into atmosphere. In contrast, this was giving relatively lower or in the same magnitude from most places of radionuclides in rainwater samples. Degree of changes between ^{210}Pb, ^{210}Bi and ^{210}Po affected by high temperature combustions were found to be different for each nuclide due to their respective volatility. ^{210}Po in rainwater and solid fallout were considerably low during early inter-monsoon period which mainly controlled by the rainfall pattern. On the other hand, ^{210}Pb and ^{210}Bi in solid fallout were recorded higher concentrations which associated to drier conditions and more particulate content in air column during southwest monsoon. The mean activity ratio of $^{210}Bi_{RW}/^{210}Pb_{RW}$ and $^{210}Po_{RW}/^{210}Pb_{RW}$ are 0.47 ± 0.04 and 0.52 ± 0.17, respectively. Whereas for $^{210}Bi_{SF}/^{210}Pb_{SF}$ and $^{210}Po_{SF}/^{210}Pb_{SF}$ are 0.52 ± 0.05 and 0.71 ± 0.13, respectively. Some results showed high activity ratios, reaching to 1.87 ± 0.08 for ^{210}Bi/^{210}Pb and 4.58 ± 0.55 for ^{210}Po/^{210}Pb, of which due to additional of ^{210}Bi and ^{210}Po excess. These ratios also indicating that ^{210}Pb and ^{210}Bi could potentially come from the same source, compared to ^{210}Po which varied differently, showing evidence it came from different source. The excess for ^{210}Pb, ^{210}Bi and ^{210}Po in solid fallout samples was more significant compared to rainwater samples.

Keywords: natural radionuclide; ^{210}Pb; ^{210}Bi and ^{210}Po; coal-fired power plant; seasonal changes; monsoon; rainwater; solid fallout

1. Introduction

The naturally occurring radionuclides (^{210}Pb, ^{210}Bi and ^{210}Po) are useful tracers for studying the processes occurred in the atmospheric systems (e.g., Poet *et al*., 1972; Moore *et al*., 1973; Tsunogai & Fukuda, 1974). In the natural environment, ^{210}Pb, ^{210}Bi and ^{210}Po are produced by radioactive decay from their progenitor, ^{222}Rn, which emanates primarily from land surface. ^{210}Pb, ^{210}Bi and ^{210}Po are considered as short half lives ($t_{½}$) of uranium progenies. The $t_{½}$ of ^{210}Pb is 22.3 years, ^{210}Bi is 5.01 days and ^{210}Po is 138 days. These radionuclides are particle reactive and strongly bound onto particles (Junge, 1974). Previous atmospheric studies for these radionuclides ranging is from the residence time of particulate and aerosol, prediction of mixing and cycling behaviour of particle reactive pollutants, tracing magma degassing, monitoring air radioactivity and many more (e.g., Daish *et al*., 2006; Papastefanou, 2006; Flury and Völkle, 2008).

Most of the ^{210}Pb and its progenies in atmosphere is formed as a decay product of ^{222}Rn emanating from soil. Their concentration levels in atmosphere have been depending by many factors, including concentrations of aerosol, origin of the air masses (Carvalho, 1995), size distribution of aerosol particle, size and phase of the hydrometer (e.g., raindrop or snow) (Martin, 2003), duration, amount and intensity of precipitation (Caillet *et al*., 2001; Martin, 2003; Hirose *et al*., 2004), meteorological conditions and stability of the atmosphere (Baskaran and Shaw, 2001; Sabuti, 2009) and altitude (Baskaran *et al*., 1993).

In addition, significant amounts of natural radioactivity are being continuously injected into atmosphere and environments via human activities. This also includes thermal power plant using coal as fuel (UN-

SCEAR 1982). Coal combustion processes produces significant quantities of dusts containing radionuclide contributed into the atmosphere. The natural radionuclides released from this industry have potential to influence human health particularly through respiration and ingestion process (UNSCEAR, 1993).

Regarding the above statements, the relevance tracing of ^{210}Pb, ^{210}Bi and ^{210}Po concentrations towards studying the fate of fly ash could give important information on their behaviours affected by meteorological changes in tropical region. Thus, this study was conducted on collecting information of ^{210}Pb, ^{210}Bi and ^{210}Po activities in atmospheric samples caused by the operation of coal-fired power plant.

2. Site Description

The study was conducted at the coastal area of Kapar, located in west coast of Peninsular Malaysia (Latitude: 03º 07', Longitude: 101º 19') where a coal-fired power plant is operated (Fig. 1, Table 1). The coal-fired power plant (local name is Sultan Salahuddin Abdul Aziz Power Plant, here stated as SSAAPP thereof) is operated by a private company, Kapar Energy Ventures Sdn. Bhd. The construction was completed on 1981, originally planned to supply electricity throughout the western coast of Peninsular Malaysia. SSAAPP is generating 2420 MW of electric power per year, consuming 2.5×10^9 kg of raw charcoal annually. SSAAPP is the biggest power plant in Malaysia, contributing 23% of the electricity demanded by the country. It uses three different sources to generate the electricity; whereby charcoal is the main fuel and natural gas and medium fuel oils are used as additional sources. The raw charcoal is primarily imported from Australia, Indonesia, South Africa and China although a small quantity does originate from Sarawak, Malaysia. About 760 000 tons of coal are stored in the coal yard area at a time and used between 10 000 to 15 000 tons every day. The natural gas and medium fuel oil are supplied by local oil and gas company, Petronas Berhad through gas pipeline and also by shipping (TNB Generation, 2003).

According to UNSCEAR (1982) on worldwide scale, about 3×10^9 kg of coal is required to produce 1000 MW of electrical power per year. But, this will depend on the carbon content for every grade of fossil fuel been used. The SSAAPP releases a significant amount of dusts and gaseous forms of emissions, in which containing radioisotopes (Mohamed *et al*., 2006). There are three stacks with 200 m in heights and is equipped with modern electrostatic precipitators (ESP) and its efficiency is over 99.2% (TNB Generation, 2003).

Based on the periodic changes in regional wind flow patterns, the climate of Peninsular Malaysia is described by four seasons, namely two monsoon seasons and two inter-monsoon seasons (MMD, 2008) (Table 2). The southwest monsoon season is usually established from May to September while the northeast

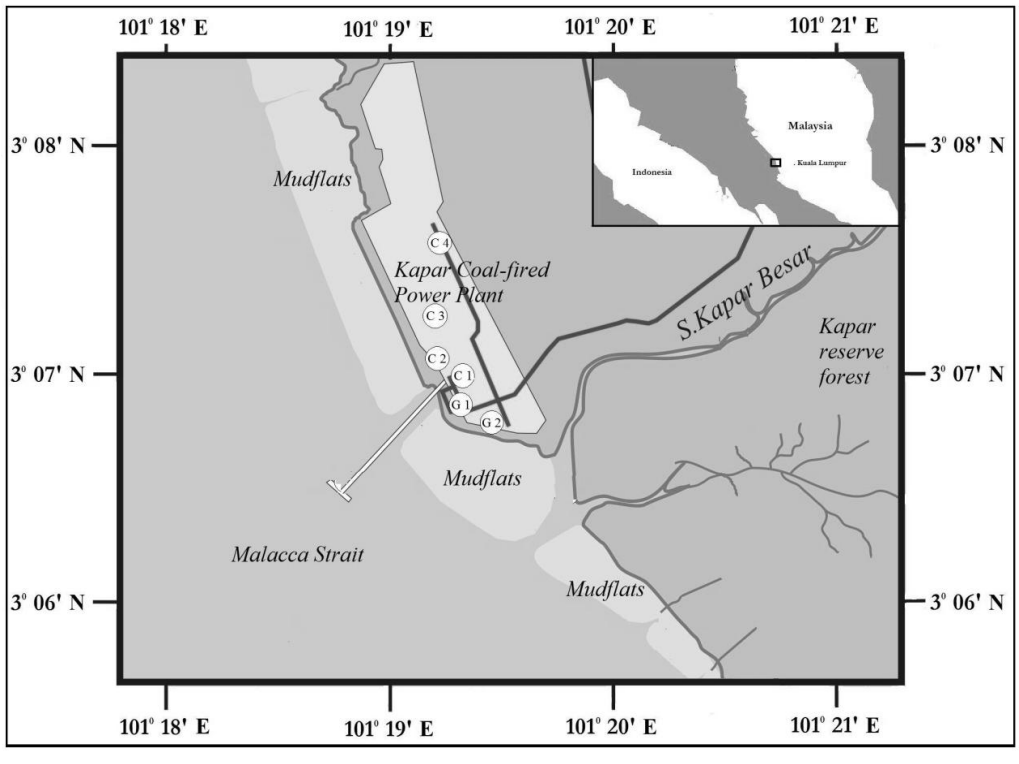

Figure 1. The location of Sultan Salahuddin Abdul Aziz Power Plant, Kapar and circled number are referred to sampling stations.

Table 1. Sampling location codes with Latitude-Longitude coordinate and some information for each location

Location code	Latitude Longitude	Distance from chimney (m)
G 1	03° 06' 48" N 101° 19' 40" E	600
G 2	03° 06' 43" N 101° 19' 47" E	800
C 1	03° 07' 01" N 101° 19' 18" E	200
C 2	03° 07' 06" N 101° 19' 32" E	400
C 3	03° 07' 19" N 101° 19' 08" E	1000
C 4	03° 07' 41" N 101° 19' 09" E	2000

monsoon season usually commences from November to March. On the other hand, there are two inter-monsoon seasons between the southwest and northeast monsoons. During the southwest monsoon, the whole country especially of the west coast of the Peninsula experiencing a drier period. Whereas during the northeast monsoon, the eastern part of the Peninsula receive heavy rainfall with winds may reach 30 knots or more due to strong surges of cold air from the north called cold surges (MMD, 2008). During this period, the west coast of the Peninsula are almost free from this influence due to the fact that this region are sheltered by the mountain ranges (the Titiwangsa Range). In contrast, the two inter-monsoon seasons often result in heavy rainfall that usually occurs in the form of convective rains. During these inter-monsoon seasons, the west coast is generally wetter than the east coast (MMD, 2008).

Driven by these climate variabilities, the west coast of the Peninsula usually received maximum rainfalls in October and November and minimum rainfall in February (MMD, 2008). While in Fig. 2 shows the maximum rainfalls during March-April-May and minimum rainfalls during June-July.

3. Materials and Methods

This study was conducted during the inter-monsoon to southwest monsoon season from 7 February to 27

Table 2. Summary of the sampling and climatic variation

Sampling no.	Sampling period	Sampling duration (days)	Climate description in study area[1]
NE 1	7 Feb – 23 Feb	16	❖ End of the northeast monsoon season
NE 2	24 Feb – 15 Mar	19	❖ Very occasional and limited rainfall activity ❖ Weak wind from the northeast ❖ Increasing atmospheric temperature ❖ Warm and dry period
IM 3	16 Mar – 4 Apr	19	❖ Inter-monsoon season
IM 4	5 Apr – 19 Apr	14	❖ More frequent rainfall activity of light to moderate intensity
IM 5	20 Apr – 4 May	14	❖ Weak wind with no particular direction ❖ Relatively humid period
SW 6	5 May – 17 May	12	❖ Southwest monsoon season
SW 7	18 May – 6 Jun	19	❖ Very occasional rainfall activity
SW 8	7 Jun – 22 Jun	15	❖ Occasional strong wind from the northeast
SW 9	23 Jun – 27 Jul	34	❖ Significant influence from Sumatran Winds ❖ Dry period

[1]From MMD (2008)

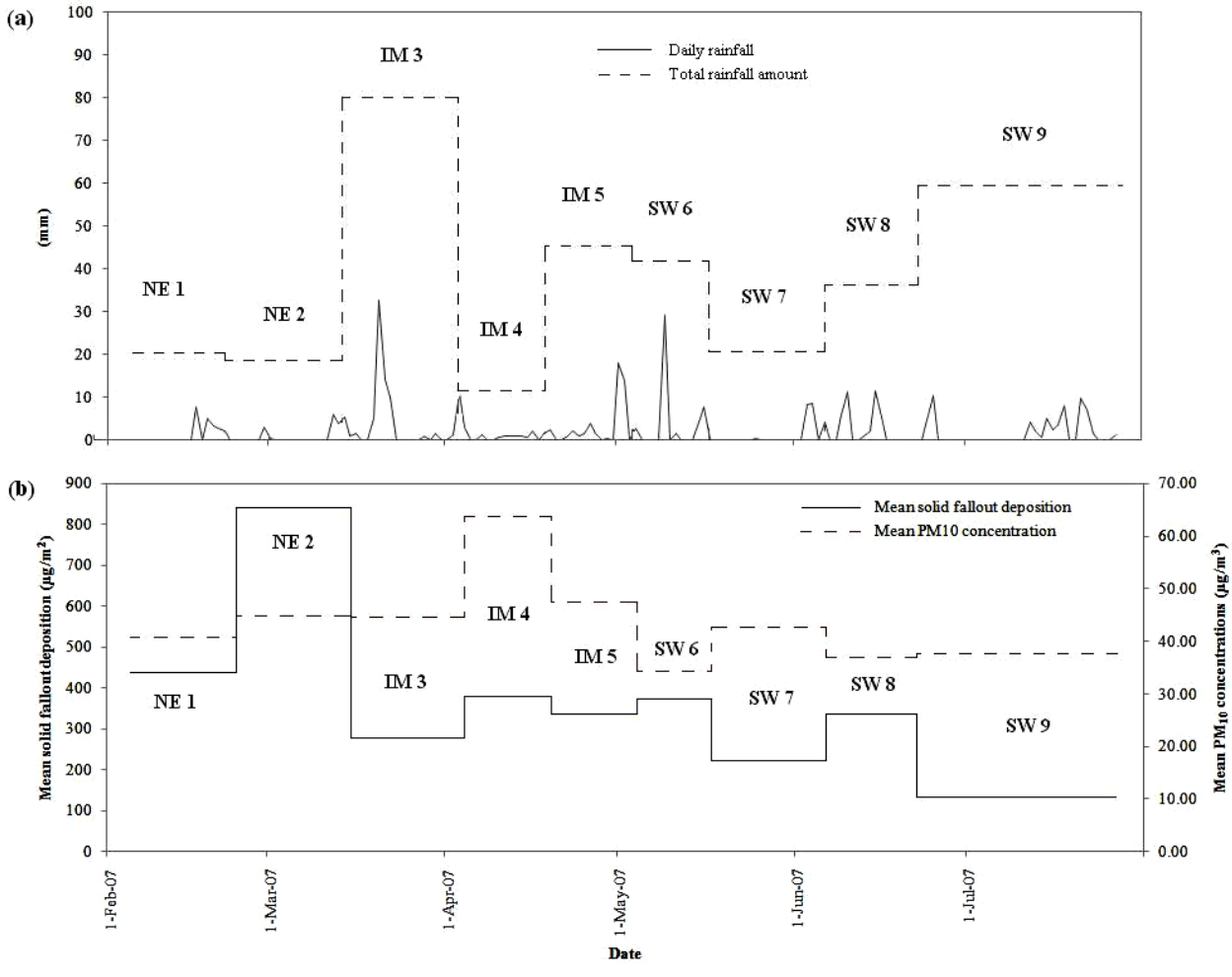

Figure 2. Distribution of daily rainfall within sampling periods

July 2007. The atmospheric sampling was perform via atmospheric traps collecting bulk deposition of rainwater and solid fallout were installed with an approximately 660 cm² area of HDPE pail placed 15 m above the ground to avoid as much as possible any soil derived contribution. The period of every collection was 14 to 20 days depending on the rainfall event (Table 2). Rain gauge was used for determining the amount of rainfall for each event. All collected sample was stored in a polypropylene container and delivered into laboratory for analysis. The fractions of dissolved and solid were separated using the Whatman filter papers with 0.45 μm pore size.

Dissolved phase of rainwater samples were acidified to pH 2 using concentrated nitric acid (HNO_3), and spiked with 20 mg of iron carrier [$FeCl_3.4H_2O$], 25 mg of lead carrier [$Pb(NO_3)_2$], 25 mg of bismuth carrier [$Bi(NO_3)_3.5H_2O$] and, 1.67 mBq of polonium-209 tracer. The aqueous was precipitated with ammonium hydroxide (NH_4OH) to form iron(III)hydroxide [(Fe(III)$(OH)_3$] precipitation and re-dissolved with 0.5 M of HCl for further radiochemical separations.

Meanwhile, solid fallout samples (particulate fraction) were totally digested with 20 ml of concentrated HNO_3, 20 ml of concentrated HCl and 5 ml of concentrated HF to dissolve all solid particles in Teflon® beaker after spiked with 25 mg of lead carrier, 25 mg of bismuth carrier and, 21.7 mBq of polonium-209 tracer. Then samples were heated on hotplate at temperature below 80ºC to prevent the volatilization of polonium (Murray *et al*., 2007) before dissolution with 0.5 M of HCl and followed with radiochemical separation procedure proposed by Narita *et al*. (1989).

The determination of ^{209}Po and ^{210}Po activities were made by auto plating onto polished and pre-cleaned silver foil before immediate counting using an alpha spectrometry (Alpha Analyst Spectroscopy system with a silicon-surface barrier detector by Canberra, Inc.) with chemical yield ranging from 60 – 80%.

Then, bismuth (Bi) was collected via electrodeposition process, forming bismuth oxychloride (BiOCl) precipitation (Narita *et al*., 1989). The BiOCl precipitate was wrapped onto plastic disc and the beta activity of ^{210}Bi was counted immediately using a gross alpha/beta

counting system (Tennelec XLB-5 low background gas-flowing anti-coincidence α/β counter by Canberra, Inc.). Finally, the isotope of lead (^{210}Pb) was also collected via the electrodeposition process by forming lead sulphate ($PbSO_4$) precipitation. The precipitate was wrapped onto plastic disc and count for ^{210}Pb via ^{210}Bi beta activity using the gross alpha/beta counting system after 1 month to allow Bi ingrowths (Ivanovich and Harmond, 1982). The gross alpha/beta counting system has 50% counting efficiency value, and the background for alpha and beta were low with only 0.04 cpm and 0.5 cpm, respectively. The measurement time for each sample is 300 minutes and repeated for three times to ensure the cpm value is consistent. The standard source of beta emitter such as Sr-90 was also applied to calibrate the performance of the instrument in every three months. A standard reference material IAEA-300 was applied to validate the analytical procedure of ^{210}Pb with recovery value ranging from 88.16 ± 4.46 to 109.87 ± 5.62% with average 98.73 ± 5.51%.

In order to determine the actual pattern of air particulate concentration in the study area, which mainly dominated by combustion products of SSAAPP we have collecting information on solid fallout deposition (Table 3). In addition, the PM_{10} concentration provided by SSAAPP is also shown here to give general view on particulate concentrations in SSAAPP air column during our sampling. Through our observation, a good relationship was shown in the highest solid fallout deposition ever recorded (7.29 g/m^2) was collected during drier period of NE 2 sampling with rather low rainfall amount (19.0 mm). Meanwhile, the mean PM10 concentration during IM 4 sampling (63.87 μg/m^3) was the highest fly ash emission from SSAAPP during the study period which could have resulted high solid fallout deposition as well. In contrast, our finding have presented otherwise which led to our assumption was due to a relatively dry period (shown by low rainfall amount, 11.8 mm), had been brought to a long ranged transport by air masses.

4. Results

Rainwater and solid fallout samples were analyzed to investigate the impact of coal combustion processes to the ambient air. In this study, the activity concentrations of natural radionuclides measured in rainwater varied from 5.70 ± 0.14 to 75.29 ± 7.61 mBq/L (average: 34.96 ± 2.68 mBq/L) for ^{210}Pb, 1.18 ± 0.03 to 38.60 ± 1.47 mBq/L (average: 16.46 ± 0.56 mBq/L) for ^{210}Bi, and 5.43 ± 2.97 to 38.05 ± 17.15 mBq/L (average: 18.20 ± 5.77 mBq/L) for ^{210}Po (Table 4). These mean concentration values of lead, bismuth and polonium isotopes are way below from other reported places by Sugihara *et al*. (1999), Caillet *et al*. (2001), Martin (2003), McNeary and Baskaran (2007) and Hirose *et al*. (2004). This comparison shows our results were nearly ten times lower than those studies. In our assumption, this is probably due to volatilization of these radionuclides cased by high temperature combustions and later condensation onto fly ash matrix compared to water vapours. As a result, the rate of washout of the atmosphere in coal-fired power plant area is more significant to atmospheric aerosols and solid particles through dry deposition than wet deposition (i.e., in raindrops).

The natural radionuclide concentrations in solid fallout, however were varied from 170 ± 5 to 1588 ± 71. Bq/kg (average: 560 ± 39 Bq/kg) for ^{210}Pb, 72 ± 2 to 1582 ± 93 Bq/kg (average: 342 ± 17 Bq/kg) for ^{210}Bi, and 133 ± 19 to 456 ± 80 Bq/kg (average: 307 ± 54 Bq/kg) for ^{210}Po (Table 4). These values were extended over two orders of magnitude from the Malaysian natural radionuclide range in soils from 38 to 94 Bq/kg (average

Table 3. The collection of rainfall amount and mean of solid fallout deposition and PM10 concentrations on every sampling collections.

Sampling no.	Total rainfall amount (mm)	Mean solid fallout deposition (g/m^2)	Mean PM_{10} concentration (μg/m^3)
NE 1	20.8	3.67	40.72
NE 2	19.0	7.29	44.77
IM 3	80.8	3.77	44.65
IM 4	11.8	2.25	63.87
IM 5	46.0	6.06	47.40
SW 6	42.6	3.55	34.24
SW 7	21.2	3.88	42.78
SW 8	36.9	5.43	36.96
SW 9	60.4	5.89	37.59

Table 4. The activity concentrations of ^{210}Pb, ^{210}Bi and ^{210}Po in rainwater (RW) and solid fallout (SF) and their average values during this study

Sampling no.	Station	Rainwater (mB q/L)			Solid fallout (Bq/kg)		
		$^{210}Pb_{RW}$	$^{210}Bi_{RW}$	$^{210}Po_{RW}$	$^{210}Pb_{SF}$	$^{210}Bi_{SF}$	$^{210}Po_{SF}$
NE 1	G 1	54.52 ± 0.85	24.66 ± 0.26	14.00 ± 0.24	3037.67 ± 80.19	2254.49 ± 29.43	169.18 ± 21.64
	G 2	27.86 ± 0.30	15.47 ± 0.20	23.45 ± 0.59	5081.44 ± 73.45	5328.94 ± 202.40	306.17 ± 10.38
	C 1	50.77 ± 0.71	12.02 ± 0.15	n.a	195.15 ± 2.83	289.20 ± 6.80	n.a
	C 2	39.32 ± 0.59	8.58 ± 0.10	13.42 ± 0.20	69.65 ± 1.11	17.13 ± 0.20	20.71 ± 0.74
	C 3	32.06 ± 0.40	33.62 ± 0.43	16.20 ± 0.38	114.40 ± 2.31	131.56 ± 2.71	34.52 ± 1.36
	C 4	43.78 ± 1.11	22.06 ± 0.29	17.44 ± 0.31	1032.05 ± 15.51	1472.22 ± 40.70	n.a
	Average	41.39 ± 1.64	19.40 ± 0.58	16.90 ± 0.76	1588.39 ± 71.10	1582.26 ± 93.48	132.65 ± 18.92
NE 2	G 1	60.26 ± 0.72	6.77 ± 0.04	n.a	734.65 ± 26.82	271.24 ± 2.16	568.46 ± 31.32
	G 2	35.14 ± 0.43	8.04 ± 0.05	n.a	1152.97 ± 36.69	n.a	597.98 ± 24.39
	C 1	36.72 ± 0.32	12.18 ± 0.10	22.10 ± 9.36	1492.37 ± 184.47	144.78 ± 3.63	n.a
	C 2	60.59 ± 0.76	18.81 ± 0.13	n.a	278.32 ± 17.54	n.a	175.42 ± 43.80
	C 3	23.38 ± 0.42	9.44 ± 1.87	n.a	238.39 ± 12.80	n.a	n.a
	C 4	21.81 ± 0.15	13.35 ± 0.01	8.55 ± 4.93	866.75 ± 30.01	149.73 ± 1.41	n.a
	Average	39.66 ± 1.20	22.93 ± 0.59	15.33 ± 10.96	793.91 ± 127.22	188.58 ± 5.27	447.29 ± 115.81
IM 3	G 1	49.65 ± 6.18	1.33 ± 0.02	n.a	96.77 ± 1.55	87.01 ± 1.28	193.79 ± 14.25
	G 2	21.69 ± 0.45	0.93 ± 0.01	7.42 ± 1.53	528.20 ± 13.26	211.15 ± 2.68	234.87 ± 17.68
	C 1	42.61 ± 2.49	1.14 ± 0.01	4.77 ± 1.88	181.80 ± 3.76	135.86 ± 3.57	121.38 ± 5.43
	C 2	46.99 ± 5.93	0.84 ± 0.01	7.85 ± 1.42	187.17 ± 10.41	33.86 ± 0.44	58.54 ± 4.91
	C 3	33.27 ± 2.14	1.65 ± 0.02	1.69 ± 0.44	29.23 ± 0.30	15.76 ± 0.18	150.59 ± 12.18
	Average	38.84 ± 7.71	1.18 ± 0.03	5.43 ± 2.97	204.63 ± 13.75	96.73 ± 3.58	151.83 ± 24.79
IM 4	G 1	40.73 ± 0.68	12.08 ± 0.15	8.62 ± 1.91	286.42 ± 6.28	111.80 ± 1.30	n.a
	G 2	62.34 ± 2.92	8.21 ± 0.11	32.51 ± 1.66	498.85 ± 22.92	694.51 ± 43.44	194.78 ± 10.33
	C 1	64.34 ± 1.67	29.34 ± 0.35	7.19 ± 1.80	303.77 ± 7.31	100.28 ± 1.86	233.61 ± 11.39
	C 2	62.69 ± 2.40	40.18 ± 0.58	6.77 ± 2.32	390.64 ± 10.98	552.14 ± 7.62	716.68 ± 54.90
	C 3	59.77 ± 0.93	24.76 ± 0.55	7.05 ± 1.25	168.50 ± 2.69	35.27 ± 0.65	345.19 ± 11.82
	C 4	161.84 ± 11.84	117.02 ± 1.91	76.26 ± 4.06	983.89 ± 27.43	135.15 ± 1.43	364.48 ± 16.96
	Average	75.29 ± 7.61	38.60 ± 1.47	23.07 ± 11.90	438.68 ± 31.00	271.53 ± 19.26	370.95 ± 44.51

Sampling no.	Station	Rainwater (mB q/L)			Solid fallout (Bq/kg)		
		$^{210}Pb_{RW}$	$^{210}Bi_{RW}$	$^{210}Po_{RW}$	$^{210}Pb_{SF}$	$^{210}Bi_{SF}$	$^{210}Po_{SF}$
IM 5	G 1	22.57 ± 0.33	2.37 ± 0.03	21.57 ± 2.26	1537.04 ± 26.30	165.95 ± 3.28	721.10 ± 147.67
	G 2	12.13 ± 0.17	9.05 ± 0.12	6.02 ± 0.35	92.35 ± 3.49	n.a	168.84 ± 5.95
	C 1	10.57 ± 0.12	5.54 ± 0.06	6.57 ± 0.43	994.26 ± 52.48	184.98 ± 4.25	185.41 ± 15.22
	C 2	11.80 ± 0.23	11.54 ± 0.16	10.04 ± 0.67	219.10 ± 6.00	152.58 ± 3.07	152.31 ± 13.55
	C 3	18.07 ± 0.35	10.79 ± 0.16	6.29 ± 0.36	128.22 ± 2.93	n.a	258.23 ± 22.39
	C 4	10.39 ± 0.11	2.79 ± 0.04	10.80 ± 0.72	630.72 ± 9.64	558.77 ± 10.88	n.a
	Average	14.26 ± 0.53	7.01 ± 0.23	10.22 ± 1.79	600.28 ± 46.55	265.57 ± 10.96	297.18 ± 75.97
SW 6	G 1	80.01 ± 1.82	32.84 ± 0.47	25.61 ± 1.59	848.60 ± 26.07	297.29 ± 4.07	448.50 ± 35.11
	C 3	52.41 ± 1.44	40.82 ± 0.87	18.23 ± 1.52	689.27 ± 31.19	279.73 ± 6.02	503.93 ± 51.49
	C 4	52.87 ± 1.78	20.91 ± 0.32	22.38 ± 1.51	199.20 ± 3.71	191.04 ± 3.99	187.89 ± 18.76
	Average	61.76 ± 3.03	31.52 ± 0.94	22.07 ± 2.74	579.02 ± 33.45	256.02 ± 8.44	380.11 ± 61.92
SW 7	G 1	63.92 ± 2.75	12.04 ± 0.31	41.34 ± 7.43	587.14 ± 6.06	246.90 ± 3.45	1044.65 ± 97.69
	G 2	23.80 ± 0.40	19.35 ± 0.44	10.98 ± 1.12	507.61 ± 15.39	140.14 ± 2.64	416.10 ± 22.60
	C 1	29.98 ± 0.92	3.03 ± 0.04	39.57 ± 8.71	178.34 ± 2.58	104.01 ± 1.55	196.97 ± 11.37
	C 2	25.26 ± 0.50	1.25 ± 0.02	41.70 ± 9.14	116.62 ± 2.02	240.03 ± 6.30	319.79 ± 24.58
	C 3	16.93 ± 0.26	4.12 ± 0.05	61.39 ± 13.18	134.88 ± 2.24	130.18 ± 2.95	341.91 ± 28.73
	C 4	31.96 ± 0.72	15.67 ± 0.24	33.32 ± 4.46	60.47 ± 0.83	24.39 ± 0.38	414.47 ± 21.22
	Average	31.98 ± 2.07	9.24 ± 0.41	38.05 ± 17.15	264.18 ± 11.81	147.61 ± 6.95	455.65 ± 79.73
SW 8	G 1	4.13 ± 0.05	0.54 ± 0.01	11.10 ± 0.46	243.11 ± 2.85	37.84 ± 0.44	n.a
	G 2	8.17 ± 0.09	7.56 ± 0.07	10.31 ± 0.42	328.97 ± 4.39	143.44 ± 1.43	n.a
	C 1	5.75 ± 0.06	23.86 ± 0.36	12.85 ± 0.69	14.67 ± 0.20	40.59 ± 0.41	n.a
	C 3	4.50 ± 0.05	8.81 ± 0.14	10.03 ± 0.41	110.25 ± 1.77	58.35 ± 0.57	n.a
	C 4	5.97 ± 0.07	12.45 ± 0.21	86.33 ± 6.51	152.94 ± 1.79	77.31 ± 0.77	n.a
	Average	5.70 ± 0.14	10.64 ± 0.37	26.12 ± 3.05	169.99 ± 5.09	71.51 ± 1.65	n.a
SW 9	G 1	3.57 ± 0.05	14.08 ± 0.37	7.76 ± 0.39	811.84 ± 17.67	268.41 ± 3.30	n.a
	G 2	5.66 ± 0.10	-	7.36 ± 0.24	709.58 ± 7.97	315.34 ± 4.44	170.67 ± 4.71
	C 2	-	8.81 ± 0.13	5.13 ± 0.34	223.10 ± 2.41	136.30 ± 1.76	321.28 ± 7.79
	C 3	8.08 ± 0.11	7.25 ± 0.10	6.11 ± 0.23	115.51 ± 2.59	57.61 ± 0.62	176.11 ± 5.20
	Average	5.77 ± 0.15	7.60 ± 0.39	6.59 ± 0.64	398.35 ± 14.82	196.15 ± 5.34	223.15 ± 12.57

*n.a: data not available

67 Bq/kg), (UNSCEAR, 2000). This show, the activity levels of lead, bismuth and polonium iotopes in coal combustion by-products such as fly ash are significantly higher than the corresponding activity concentrations in coals and soil (earth's crust). In the combustion process, there is approximately a magnitude of enhancement of the concentration from coal to ash. In addition, our results are relatively comparable to previous researches in coal fired power plant area (Beck & Miller, 1980; UNSCEAR, 1982; UNSCEAR, 1993; Yener and Uysal, 1996; Bem *et al*., 2002; Karangelos *et al*., 2004; Flues *et al*., 2007; Papastefanou, 2009).

5. Discussions

5.1. Impact of coal-fired power plant on ambient air

Natural radionuclides in coal samples are in same order of in-situ ambient soils but the combustion process will enrich the concentration of elements by a factor of two to five especially in finer fly-ash (Mishra, 2004). Based on the previous published information (Beck & Miller, 1980), the calculated ^{210}Pb and ^{210}Po in fly ash escape from the coal-fired plant is five times higher than the raw coal yard (e.g.: Beck and Miller, 1980; UNSCEAR, 1982; UNSCEAR, 1993; Yener and Uysal, 1996; Bem *et al*., 2002; Karangelos *et al*., 2004; Flues *et al*., 2007; Papastefanou, 2009). The same indication in our data shows clearly the natural radionuclide activity levels were higher than the average levels of their parent, ^{226}Ra in Malaysian soil as been discussed previously (Table 4).

In addition, Beck and Miller (1980) had observed the influence of air activity concentration at ground level is usually extended over about 1 km for short (50 m height) stacks and over 5 – 10 km for taller (150 m height) stacks. It is reported that the stack height of the coal-fired power plant also affected the diffusion of radionuclide. The influence of air activity concentration at ground level is extended over about 1 km for the short stacks (50 m height) and over 5 – 10 km for taller stacks (150 m height) (Beck and Miller, 1980). According to previous reports, this is mainly due to combustion of fossil fuel which containing natural radionuclide

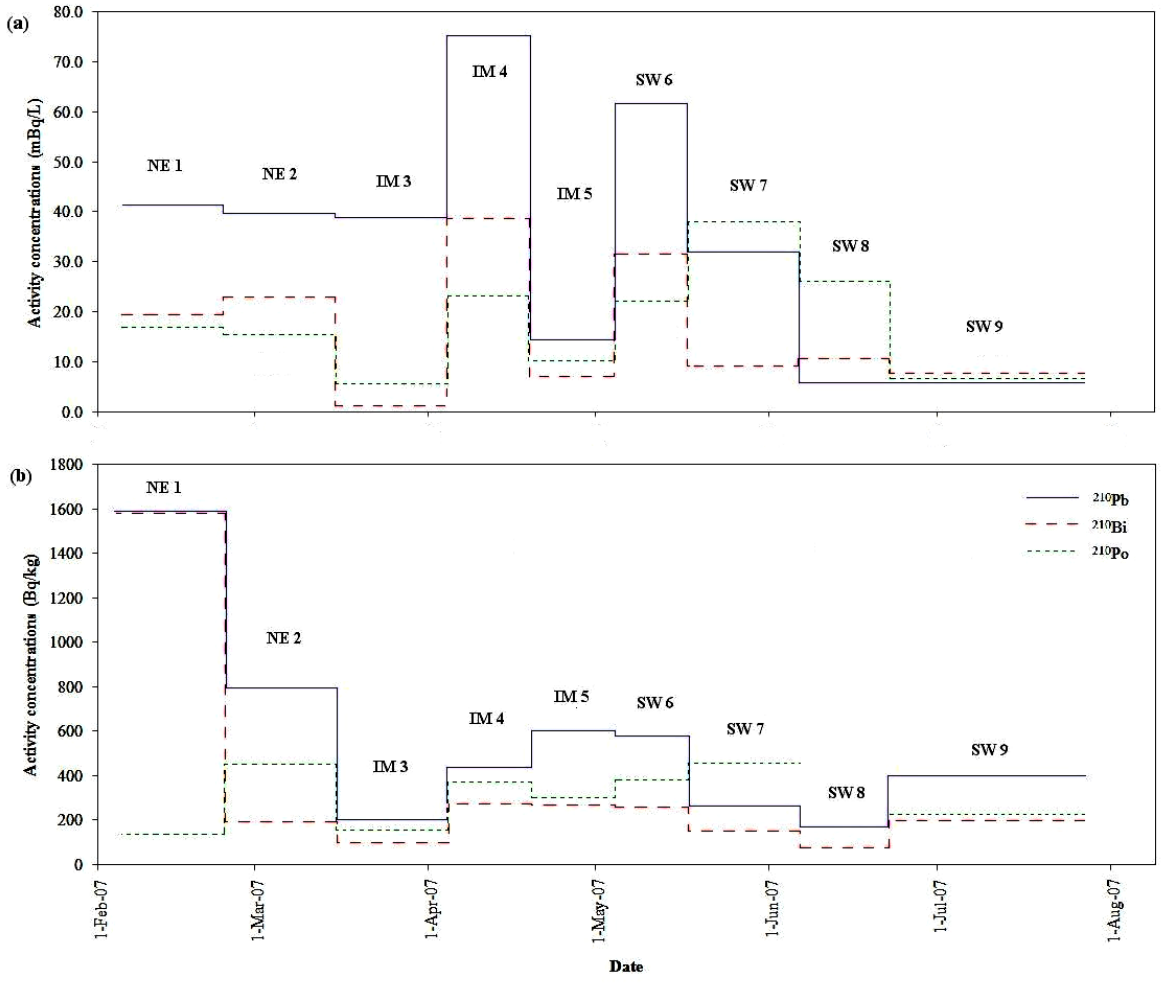

Figure 3. Fluctuations of the mean ^{210}Pb, ^{210}Bi and ^{210}Po activity concentrations in (a) rainwater (dissolved) and (b) solid fallout (particulate) samples from 7 February until 27 July 2007.

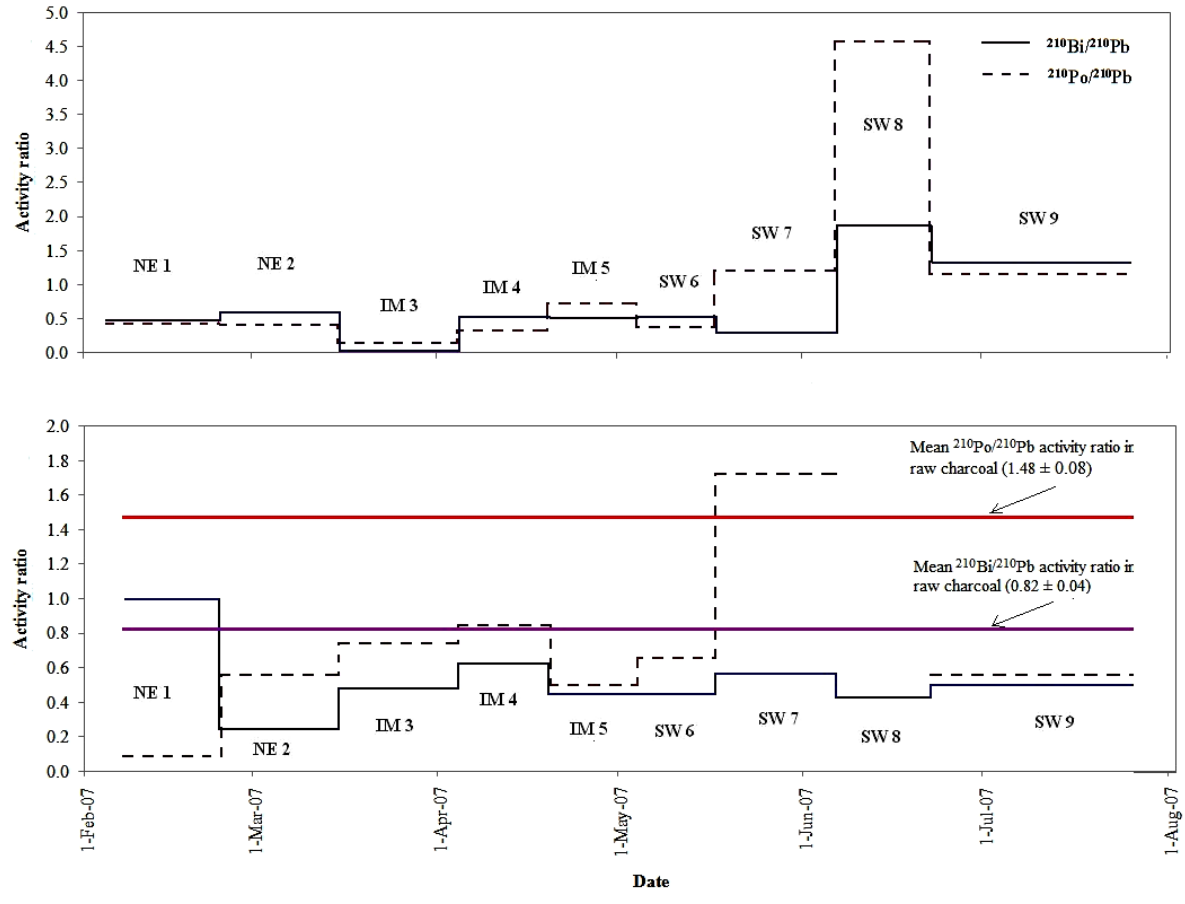

Figure 4. Fluctuations of the mean activity ratios (^{210}Bi/ ^{210}Pb and ^{210}Po/ ^{210}Pb) in (a) rainwater (dissolved) and (b) solid fallout (particulate) samples from 7 February until 27 July 2007.

decay series such as uranium-thorium series may have an impact on environmental radioactivity level in the vicinity of power plants (Beck, 1989; Papastefanou, 1996; Realo *et al*., 1996). Fly ashes escaping from the chimney usually containing natural radionuclide that are concentrated a few times in comparison with their content in raw charcoal, in which enhancing natural radioactivity caused by solid fallout from combustion of coal (Bem *et al*., 2002).

5.2. Temporal variations of ^{210}Pb, ^{210}Bi and ^{210}Po activity concentrations in rainfall and solid fallout samples in coal-fired power plant area.

Fig. 3 are showing the fluctuations of mean activity ^{210}Pb, ^{210}Bi and ^{210}Po in rainwater (dissolved phase) and solid fallout (particulate phase) samples from 7 February until 27 July 2007. From this figure, the meteorological variability was affecting the levels of these radionuclides. The annual average values may also vary from year to year due to variations in coal burned, annual average meteorology parameters, and other factors.

Early part of this study in NE 1 and NE 2 was coming through the end of northeast monsoon season where in western part of Peninsular Malaysia was experiencing occasional rainfall events from the initially warm and dry January (Fig. 2(a)). The concentrations of Pb, Bi and Po were relatively high in both samples, rainwater and solid fallout. But, the trend was gradually decreased towards IM 3 (Fig. 3). This figure giving an evident that this region has received grater radionuclides sources during the northeast monsoon. The activity ratios of ^{210}Bi/^{210}Pb and ^{210}Po/^{210}Pb showed in both samples were below 1.0 (Fig. 4 and Table 5), in which is also supporting this evident where contribution from other sources leading to excess activities for each radionuclides are not significant.

While the activity concentrations of ^{210}Pb, ^{210}Bi and ^{210}Po in rainwater and solid fallout were considerably low during early inter-monsoon period (IM 3) in March, it was then increased from late inter-monsoon period to early southwest monsoon period in April (IM 5) until June (SW 7) (Fig. 3). According to Malaysian Meteorological Department (MMD 2008), this was a south west monsoon period where the climate in west coast of Peninsular Malaysia was relatively dry. But however, rainfall does sometimes occur with high intensity joined by thunderstorm during the evening. The enhanced activities of radionuclides during low

Table 5. The mean activity ratios of $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ in rainwater (RW) and solid fallout (SF) on every sampling collections.

Sampling no.	$^{210}Bi_{RW}/^{210}Pb_{RW}$	$^{210}Po_{RW}/^{210}Pb_{RW}$	$^{210}Bi_{SF}/^{210}Pb_{SF}$	$^{210}Po_{SF}/^{210}Pb_{SF}$
NE 1	0.47 ± 0.02	0.41 ± 0.02	1.00 ± 0.07	0.08 ± 0.01
NE 2	0.58 ± 0.02	0.39 ± 0.28	0.24 ± 0.04	0.56 ± 0.17
IM 3	0.03 ± 0.01	0.14 ± 0.08	0.47 ± 0.04	0.74 ± 0.13
IM 4	0.51 ± 0.06	0.31 ± 0.16	0.62 ± 0.06	0.85 ± 0.12
IM 5	0.49 ± 0.02	0.72 ± 0.13	0.44 ± 0.04	0.50 ± 0.13
SW 6	0.51 ± 0.03	0.36 ± 0.05	0.44 ± 0.03	0.66 ± 0.11
SW 7	0.29 ± 0.02	1.19 ± 0.54	0.56 ± 0.04	1.72 ± 0.31
SW 8	1.87 ± 0.08	4.58 ± 0.55	0.42 ± 0.02	n.a
SW 9	1.32 ± 0.08	1.14 ± 0.11	0.49 ± 0.02	0.56 ± 0.04

precipitation periods possibly due to enhanced ^{222}Rn emanation in dry season and followed by the important scavenging of ^{210}Pb in reduced rainfall events (Caillet *et al*., 2001). The activity ratios of $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ in both samples were also increased (Fig. 4). But the $^{210}Po/^{210}Pb$ activity ratios during IM 4, only fly ash's sample was over 1.0, and interestingly, the dissolved phase recorded very small values, below 0.5. In this south west monsoon period, the climate in west coast of Peninsular Malaysia was relatively dry (Fig. 2(a)). However, rainfall does sometimes occur with high intensity joined by thunderstorm during the evening (MMD 2008). This could support the association between ^{210}Po in both phases (rainwater and solid fallout) availability in the air column with influence from rainfall activities. The peak for ^{210}Po in solid fallout was high during intensified rainfall. This period was the starting of southwest monsoon and the rainfall activities reduces. So, this could suggest ^{210}Po activity concentration, like ^{210}Pb and ^{210}Bi in solid phase being washout by rainfall. However, the trend for rainwater samples was only increases starting from May and peaked at early of June. The trend was eventually peaked during less rainfall activity as shown at SW 7 and SW 8 (Table 2 and Fig. 2(a)). This could explain that, the concentrations of ^{210}Po in dissolved phase were accumulated in the cloud during dry season of inter-monsoon period.

In our data, the highest concentrations of ^{210}Pb and ^{210}Bi were observed during relatively low rainfall as observed at NE 1, NE 2, IM 4 and SW 7 (Fig. 2(a) and Fig. 3). Afterwards, with increasing precipitation, the concentrations quickly decreased to 205 ± 14 Bq/kg for ^{210}PbSF and 97 ± 4 Bq/kg for ^{210}BiSF. It illustrates the scavenging of radionuclides associated with aerosols below the clouds due to washout mechanism and this phenomana usually dominates during the beginning of the rain events (Caillet *et al*., 2001).

According to Fig. 5, significant difference is shown for ^{210}Bi and ^{210}Po from their parent, ^{210}Pb ehere the correlation coefficients value (R^2) of radionuclides activities in rainwater samples were low at 0.392 and 0.112 for $^{210}Pb_{RW}$ versus $^{210}Bi_{RW}$ and $^{210}Pb_{RW}$ versus $^{210}Po_{RW}$, respectively. Meanwhile, no relationship for the activity in solid fallout samples, with correlation coefficients value (R2) for $^{210}Pb_{SF}$ versus $^{210}Po_{SF}$ was 0.016. This is proving that there was no significant link in term of possible sources between $^{210}Pb_{SF}$ and $^{210}Po_{SF}$ in SSAAPP solid fallout samples. In contrast, a strong and positive connection between $^{210}Pb_{SF}$ and $^{210}Bi_{SF}$ was observed, with correlation coefficients value (R^2) was 0.822. This could explain that lead and bismuth in solid fallout are produced almost in similar magnitude from possible sources in SSAAPP.

5.3. Relationships between rainfall and solid fallout for ^{210}Pb, ^{210}Bi and ^{210}Po

The relationship between mean amounts of solid fallout depositions compared to total rainfall amount collected over each sampling periods was showing an inverse trend (Fig. 2). The decreasing solid fallout concentrations is maybe due to fly ash contents from the SSAAPP does not stand in air column for long period, especially during higher rainfall events (wet seasons), where dust suspension was considerably low because of wet surface. This was also be concluded by Ambe and Nishikawa (1987), as a negative correlations between rainfall intensity and concentration of insoluble particulate matter or dusts. The negative correlation between dust input and rainfall from this study showed good combination to prove the relationship of radionuclide concentrations and amount of solid fallout and rainfall.

On the other hand, based from our observation the activity concentrations of ^{210}Pb and ^{210}Bi were positively correlated with the mean solid fallout depositions in

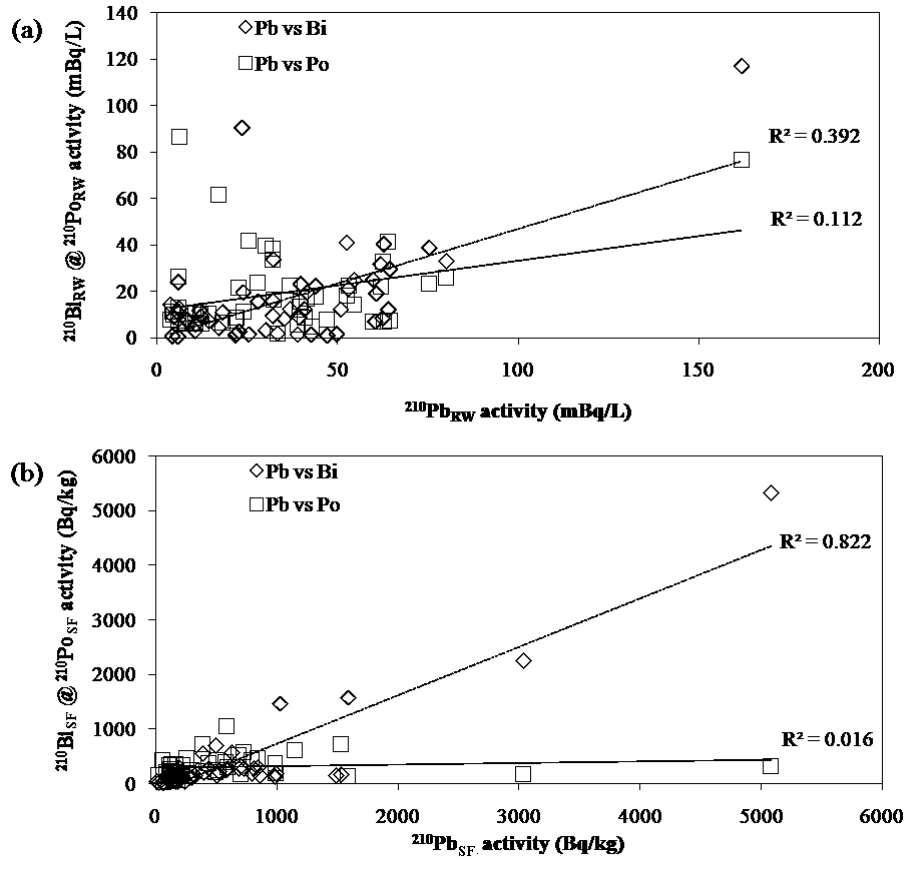

Figure 5. The relationship between ^{210}Bi and ^{210}Po to ^{210}Pb in every sample of (a) rainwater and (b) solid fallout shown by the coefficient of correlation (R^2).

SSAAPP (Fig. 6). Martin (2003) suggested that this evidence would be expected to be associated with insoluble dust particles rather than soluble particles (i.e., sea salt and condensed gases) or dissolved form (raindrops). Meanwhile, ^{210}Po in both phases had very least correlation with the solid fallout depositions as shown in Fig. 6. This evident shows that ^{210}Po by its nature not attached with insoluble dust particles, but in contrast is more correlated with rainfall episodes. This data giving us some understanding that during dry seasons, the possibility of getting high radionuclide concentration due to the availability of dust in the air column will be higher. In our observation, the dust loadings at SSAAPP area are giving more dry deposition rates in the wet season and less dust suspension rates when the ground is wet. During dry seasons where more dust loadings was obtained with high radionuclide concentrations compared to wet season. This can be concluded as negative correlations between radionuclide concentration and amount of rainfall, and positive correlation of radionuclide activities with amount of dust loadings. Such relationships have been commonly observed and explained in rainfall as a major depositional pathway of these radionuclides (Turekian *et al*., 1983; Olsen *et al*., 1985). In previous observation, correlation of rainfall amount with ^{210}Po seems better than with ^{210}Pb, likely due to a relatively greater contribution of ^{210}Pb from dry deposition (Olsen *et al*., 1985). Nonetheless, significant activity contribution from dust which originally from the coal-fired burning, giving that these results shows higher concentration than other publications (eg., Moore *et al*., 1973; Martin, 2003; Papastefanou, 2006; Al-Masri *et al*., 2006).

5.4. Behaviour and properties of ^{210}Pb, ^{210}Bi and ^{210}Po in solid fallout samples affected by SSAAPP

The activity concentrations of studied natural radionuclide in the solid fallout samples were mainly followed the series of Pb>Bi>Po. Except for some samples, the series of Po>Pb>Bi; Pb>Po>Bi; Bi>Po>Pb and Bi>Pb>Po were showed (Table 4). In comparison, the mean activities of ^{210}Pb, ^{210}Bi and ^{210}Po in raw charcoal that was used by this power plant were significantly lower than the combustion products in the form of solid fallout with a sequence of Po>Pb>Bi (Table 6).

The different series sequence may perhaps have been influenced by the different physicochemical properties, different behaviour of radionuclide and enrichment at every different stage of the combustion

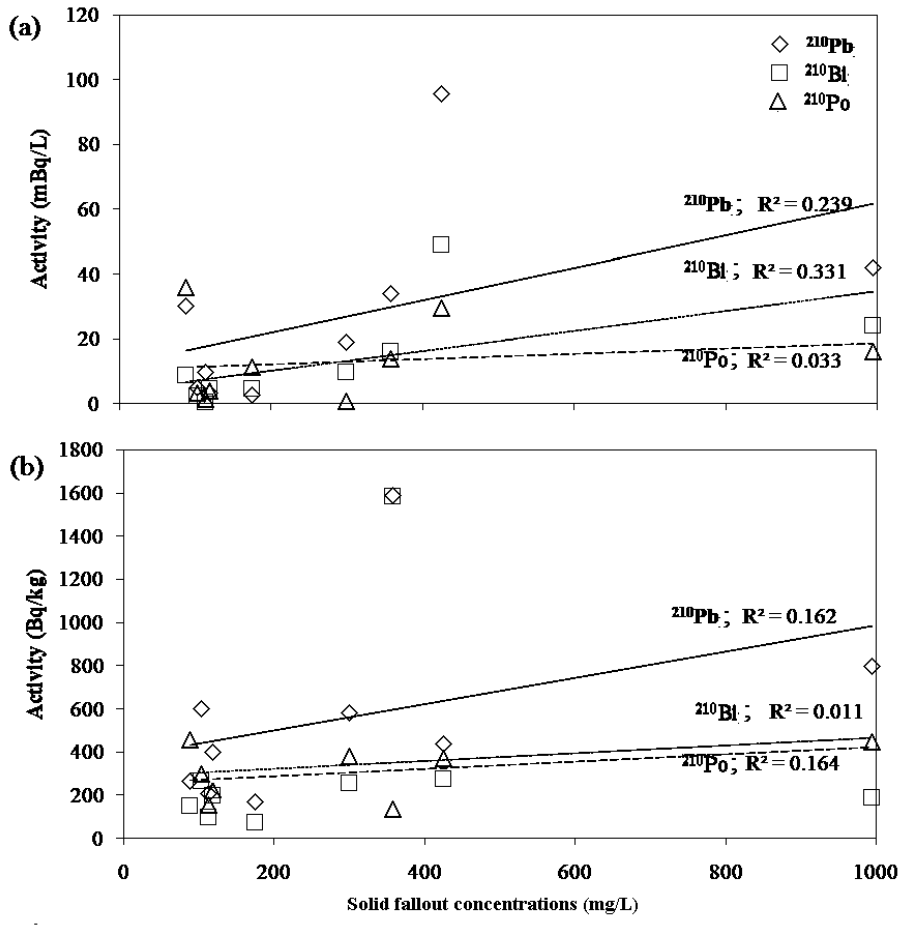

Figure 6. The positive relationship between the mean ^{210}Pb, ^{210}Bi and ^{210}Po activity to the mean solid fallout depositions in every sample of (a) rainwater and (b) solid fallout samples, shown by the coefficient of correlation (R^2).

process (Flues *et al*., 2006). ^{210}Pb which is more volatile leaves the boiler in gaseous form with the flue-gas and condenses as the temperature of the flue-gas drops. The difference in the enrichment of ^{238}U, ^{226}Ra and ^{210}Pb in the ashes results in secular radioactive disequilibrium in the ashes (Coles *et al*., 1978). Owing to these factors, there was a significant disturbance of radioactive equilibrium within ^{210}Pb, ^{210}Bi and ^{210}Po in the fly-ash samples (Fig. 4). The ratios of ^{210}Bi/^{210}Pb and ^{210}Po/^{210}Pb were calculated to evaluate the secular disequilibrium in rainwater and solid fallout of the SSAAPP. Table 5 is showing the activity ratios of ^{210}Bi/^{210}Pb and ^{210}Po/^{210}Pb, with respective average 0.47 ± 0.04 and 0.52 ± 0.17 for rainwater and 0.52 ± 0.05 and 0.71 ± 0.13 for solid fallout. These values are well described by previous findings by Coles *et al*. (1978), Beck (1989), Zelsinski and Budahn (1998) and Karangelos *et al*. (2004), as secular disequilibrium between the parents and daughters.

Natural radionuclide of lead, bismuth and polonium are particulate reactive, the availability of aerosol components and particulate matter in air column may be a major factor for the changes of these radionuclide activities, which contributed more excess of ^{210}Po (Papastefanou, 2006). Anand and Rangarajan (1990) also had proved about 17% of ^{210}Po and ^{210}Pb were derived from atmospheric ^{222}Rn decay, while 25% both of them were derived from soil and the rest was from other sources especially from human activities. Some variation of activity in rainfall events were also showed from their relation to the input of dust loadings, especially ^{210}Po.

Table 6. The mean activity of $^{210}Pb_{RC}$, $^{210}Bi_{RC}$ and $^{210}Po_{RC}$, from 12 replicates of raw charcoal used by the SSAAPP and their activity ratios

Mean activity concentration (Bq/kg)			Activity ratio	
^{210}Pb	^{210}Bi	^{210}Po	^{210}Bi/^{210}Pb	^{210}Po/^{210}Pb
105.28 ± 4.45	86.35 ± 2.93	155.37 ± 4.93	0.82 ± 0.04	1.48 ± 0.08

In all data collected from this study showed, almost 13.04% from $^{210}Bi/^{210}Pb$ activity ratios in dissolved phase in rainwater were over 1.0. So, from our understanding there must have some contribution from variety of natural and pollutant sources. But, almost 20.93% of $^{210}Bi/^{210}Pb$ activity ratios in particulate phase (solid fallout) were over 1.0. This observation is telling that ^{210}Bi and ^{210}Pb activities in particulate phase were more affected by the excess components (e.g., from outside sources). Similarly to the $^{210}Po/^{210}Pb$ activity ratios that over 1.0 recorded higher number for particulate phase than dissolved phase. More than 40% of $^{210}Po/^{210}Pb$ activity ratios in particulate phase were over 1.0, whereas only 30% in dissolved phase. This is showing opposite reflection to the $^{210}Bi/^{210}Pb$ activity ratios as mentioned above. But, the behaviour of ^{210}Po has slight difference to its parent, ^{210}Pb and ^{210}Bi, in which ^{210}Po has positive correlation to the rainfall. From this study, the difference might be their efficiency and scavenging behaviour due to the rainfall activities, which should be explain in future.

6. Conclusions

In attempt to evaluate the escaping radioactivity from SSAAPP, we discovered that the activities of ^{210}Pb, ^{210}Bi and ^{210}Po in surrounding area have potentially increases the ambient natural background radiation levels. The natural radionuclide is estimated to be enhanced more than five times which caused by coal combustion. In addition, combustion processes in coal-fired power plant has affecting the fractionation of these radionuclide in rainwater and most importantly solid fallout samples. But, the atmospheric concentration levels of natural radionuclides in this region will also continuously change by climate variability between monsoonal differences. ^{210}Po activities, in particular for dissolved phase, were accumulated in the cloud during dry period, in which ^{210}Po has positive correlation to the rainfall. Meanwhile, the activity concentration of $^{210}Pb_{SF}$ and $^{210}Bi_{SF}$ were positively correlated with the particulate amount in air column which can be driven by coal burning emissions.

Finally, this investigation should be continued to confirm whether airborne ^{210}Pb concentrations in Kapar region will consistently vary seasonally, or mean annual concentrations remained relatively constant over a long period of time or not.

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