Activity Levels of ²¹⁰Po in the Coastal Area of Kapar, Malaysia, Close to a Coal-Fired Power Plant

(Aras Aktiviti ²¹⁰Po di Kawasan Pantai Kapar, Malaysia yang Berhampiran Stesen Jana Elektrik Arang Batu)

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ABSTRACT

The activity concentration of ²¹⁰Po from six different samples consisting of raw charcoal, surface sediment, rainwater (suspended solids (SS_{rw}) and dissolved phase (D_{rw}) and estuarine water (suspended solids (SS_{ew}) and dissolved phase (D_{ew})), were analyzed. The activity concentration of ²¹⁰Po in solid samples was between 7.63 ± 0.67 and 744.28 ± 21.12 Bqkg⁻¹ and in dissolved samples varied between 0.34 ± 0.03 and 86.33 ± 6.51 mBqL⁻¹. On average, ²¹⁰Po activity in SS_{rw} sample was the highest, at nearly three times its original form (charcoal). SS_{ew} and surface sediment samples were similarly distributed between 15th March and 1st August samplings, but were relatively lower than charcoal and SS_{rw} samples. The natural meteorological variability also enhanced ²¹⁰Po distribution and dispersion to a few kilometers from the coal-fired power plant.

Keywords: ²¹⁰Po; coal-fired power plant; meteorological variability; solid and dissolved samples

ABSTRAK

Kepekatan aktiviti ²¹⁰Po daripada enam sampel yang berbeza merangkumi arang batu mentah, permukaan sedimen, air hujan (pepejal terampai (SS_{rw}) dan fasa terlarut (D_{rw})) dan air estuari (pepejal terampai (SS_{ew}) dan fasa terlarut (D_{ew})) telah dianalisis di dalam kajian ini. Kepekatan aktiviti ²¹⁰Po di dalam sampel pepejal berada dalam julat 7.63 ±0.67 dan 744.28 ±21.12 Bqkg⁻¹. Manakala pada sampel terlarut pula beubah-ubah di antara 0.34 ±0.03 dan 86.33 ±6.51 mBqL⁻¹. Secara purata, aktiviti ²¹⁰Po di dalam sampel SS_{rw} adalah yang tertinggi, hampir tiga kali ganda daripada bentuk asalnya (arang batu). Sampel SS_{ew} dan permukaan sedimen mempunyai taburan yang sekata di antara persampelan antara 15 Mac dan 1 Ogos, namun secara relatifnya adalah rendah berbanding sampel-sampel arang batu dan SS_{ew}. Pengedaran dan penumpukan juga berbeza-beza bergantung kepada variasi iklim di lokasi kajian. Kajian ini menunjukkan penumpukan ²¹⁰Po di kawasan kajian kelihatan tinggi semasa monsun barat daya berbanding monsun timur laut, yang disebabkan oleh keadaan meteorologi masing-masing. Seterusnya, perubahan meteorologi secara semula jadi juga meningkatkan taburan dan edaran ²¹⁰Po kepada beberapa kilometer daripada stesen janaelektrik arang batu tersebut.

Kata kunci: ²¹⁰Po; perubahan meteorologi; polonium; sampel pepejal dan terlarut; stesen janakuasa arang batu

INTRODUCTION

²¹⁰Po is a non-conservative radionuclide with a physical half life of 138.4 days. This radionuclide is also an intermediate member of the naturally occurring ²³⁸U decay series and is recognized as a tracer for natural processes in the atmosphere and marine environment (Al-Masri et al. 2006; Skwarzec & Fabisiak 2007). ²¹⁰Po is a particle-reactive radionuclide and tends to associate with particulate phases (Yang & Lin 1992). It also has a potential use as a tracer of particle/water transport and particulate scavenging (Swarzenski et al. 1999). In the marine environment, the radioactive disequilibrium of ²¹⁰Po and ²¹⁰Pb is likely to be the result of the preferential uptake of polonium by planktonic organisms and other biogenic particulate material (Skwarzec & Fabisiak 2007). Therefore, the behavior of ²¹⁰Po is closely associated

with zooplankton metabolic activity and the density of zooplankton in surface waters (Skwarzek et al. 2003; Tateda et al. 2003).

Research on radioactivity in the environment has been of great importance over the last decades. This research itself is very significant as a scientific basis for legislative measures, which in turn leads to a reduction in the industrial and agricultural impact of radioactivity on the environment. The effect of radioactive exposure on humans has been well-documented in the chemistry composition. The enhancement of radioactivity can be contributed to by both natural and human activities (anthropogenic). Natural processes are mainly derived from cosmic-ray interactions and radioactive decay of radon products, which emanate from continental surfaces into the atmosphere (Baskaran & Shaw 2001; Papastefanou 2006; Poet et al. 1972). In contrast, the human threats originate from a multitude of sources, such as: mining, waste management problems, an increase in the number of power plants, underground testing of bombs and nuclear weapons. All of these are very harmful to mankind (Flues et al. 2002; Garland & Wakeford 2007; Papastefanou 1996).

One of the sources of radioactivity and noncombustible trace elements in the environment originates from the use of coal as a fuel source for the generation of electricity. This is the reason why radionuclides are concentrated in the ash and dispersed to the atmosphere and surrounding environment. The U.S Geological Survey (USGS) noted the radioactive significance of coal and fly ash in 1997 (USGS 1997). Based on Beck and Miller's (1980) report, the mean ash content in US coal was found to contain the specific activity of natural radionuclide to an average of 7.5 times of the coal, that is, 167 Bqkg⁻¹ for ²³⁸U and each of its decay products (including ²¹⁰Po). In fact, each of its coal-fired power plants managed to enhance the natural radioactivity to an average of five to seven times (Beck & Miller 1980; Flues et al. 2002; Papastefanou 1996). Previous reports also concluded that some of the more volatile trace elements are preferentially re-condensed in smaller particles (Beck & Miller 1980; Swarzenski et al. 1999). The enrichment of radioactivity was dominated in fly ash relative to bottom ash, and particularly in the smaller fly ash particles, which are less efficiently removed by the emission control equipment.

The radioactivity emissions enhancement from coal-fired power plants has become a major issue among environmental scientists over the past decades. The research previously undertaken monitored the quantity fly ash, in relation to radioactivity, released into the atmosphere, where the efficiency of the plant's emissions control equipment is noted as a variable influential factor (electrostatic precipitators). Fundamentally, much research has been conducted to assess the total amount of ash (mainly bottom ash) generated by the plants, which was dependent on the type and amount of coal burned. Beck and Miller (1980) concluded that a typical modern 1000 MW plant would consume approximately 2.3×10^9 kg year⁻¹ of coal and produce around 3×10^8 kg year⁻¹ of ash. In addition, Papastefanou (1996) also found that about 11 672 MBq of ²²⁶Ra per year are discharged into the environment from four different coal-fired power plants in Greece, totaling 3.62 GW of electrical energy.

In order to address the above questions and predict the mobility of radioactive elements during the coal fuel-cycle, it is necessary to determine the concentration, distribution and form of radioactive elements in both coal and fly ash. The purpose of this study was to determine the changes of ²¹⁰Po activity in the Kapar coastal area which is located in close proximity to a coal-fired power plant. It should be noted that the study of radioactivity monitoring from an industrial or power plant in a coastal area has rarely been undertaken in Malaysia.

METHODS

SITE DESCRIPTION

The study was carried out in a coal-fired power plant facility and its surrounding area. These areas are located in Kapar, Malaysia (Figure 1). The coal-fired power plant, named Sultan Salahuddin Abdul Aziz Power Plant (SSAAPP), situated in the Kapar region, has been operating since 1985. This power plant has a capacity of 2420 MW 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 demand of the country. It uses three different fuels to generate power; 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 originates from Sarawak, Malaysia. The power plant is equipped with modern ESP (Electrostatic Precipitators) and its efficiency is over 99.2% (TNB Generation 2003). Table 1 and Figure 1 show the details of the sample collections.

Kapar is located 56 km from Kuala Lumpur and is only a few kilometers away from Port Klang. Their coast of Kapar is characterized by a semi-diurnal tidal area with humid tropical weather. This location is predominantly surrounded by mudflats with mangrove forests growing along the shorelines. The eastern and southern part of the study area, are locations which are heavily occupied by oil palm plantations, manufacturing industries, residential areas and shipping ports.

SAMPLE COLLECTION AND INSTRUMENTATIONS

The study was undertaken in order to track the influence of the coal-fired power plant on the distribution of ²¹⁰Po activities in the atmosphere and coastal system of Kapar. For this reason, the samples taken ranged from raw charcoal materials to sediment. Bulk rainwater including suspended solid (SS_{rw}) and dissolved phase (D_{rw}) samples was collected using a plastic pail with an approximate area of 660 cm² together with rain gauge indicator. The samples of estuarine water containing suspended solid (SS_{ew}) and dissolved phase (Dew) were collected using a Niskin water sampler and stored in 25 L polypropylene bottles. After immediate acidification to pH2 using concentrated nitric acid (HNO₃) the collected samples were stored. The fractions between dissolved and suspended solid particles were then separated using glass fiber filter paper (Whatman; pore size 0.45 µm). The surface sediment samples were collected using a Petite Ponar grab, keep-in zip-locked plastic bags and stored at freezing temperature.

The sampling on atmospheric deposition samples (marked as site C-1 to C-4) was conducted from 7th of February to 30th of August 2007. The four sampling locations for atmospheric deposition samples were placed at 200 m, 400 m, 1,000 m and 2,000 m, respectively, from the chimney (Table 1 and Figure 1). In contrast, the

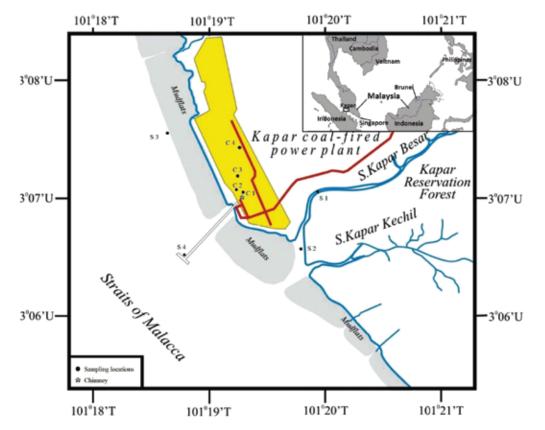


FIGURE 1. Map showing the sampling stations for atmospheric deposition and estuarine environments as display by capital letter of C and S, respectively. Where station C1, C2, C3 and C4 are 200 m, 400 m, 1000 m and 2000 m from the chimney (*), respectively, as atmospheric sampling site but S1 – S4 is sampling sites for estuarine environments

Sample type	Site number	Sampling location	Distance from chimney (meters)	Sampling area (Salinity, psu)
Atmospheric deposition	C 1	03° 07' 01" N 101° 19' 18" E	200	n.a
	C 2	03° 07' 06" N 101° 19' 32" E	400	n.a
	C 3	03° 07' 19" N 101° 19' 08" E	1000	n.a
	C 4	03° 07' 41" N 101° 19' 09" E	2000	n.a
Estuarine waters	S 1	03° 07' 03" N 101° 19' 49" E	2200	Upper estuary (0.1 - 17)
	S 2	03° 06' 27" N 101° 19' 44" E	2000	Middle estuary (18 - 25)
	S 3	03° 07' 34" N 101° 18' 31" E	2500	Water discharge (28 - 29)
	S 4	03° 06' 37" N 101° 18' 42" E	2500	Jetty (27 - 28)

TABLE 1. Information on each sampling location for this study

n.a: not available

estuarine samples (marked as site S-1 to S-4), which were also taken from four locations, were only collected twice: on 8th of March and 1st of August 2007. The reason being that they were to represent the two different monsoon periods, northeast and southwest, respectively. The distance from the chimney was between 2,000 and 2,500 m for all estuarine sampling locations so as to standardize the possible direct influence from SSAAPP.

In terms of the temporal variations which the study area experienced during P-1 (7th of February to 15th of March) samplings showed it to be a relatively dry period, characterized by a very low amount and intensity of rainfall (Figure 2). The highest rainfall intensity ever recorded during that period, 4.46 mm h⁻¹, occurred on 16th of February which was also at the end of northeast monsoon for year 2006/07. During this period, the winds from the northeast were relatively weak and tended to behave erratically. The atmospheric temperature also increased slightly, leading to warmer and drier weather (Table 2 and Figure 2).

The second period (P-2) from 16th of March to 5th of May was the transitional period between the northeast and southwest monsoon, during which time more rainfall activity was experienced in the afternoon. The rainfall intensity was normally light to moderate and only occasionally heavy and very heavy. The two highest rainfall intensities recorded during that period were 18.63 and 10.29 mm h⁻¹. These occurred on 20th of March and 1st of May, respectively. The wind was reported been fickle and weak (MMD 2008), while the atmospheric humidity was relatively higher (Table 2 and Figure 2).

The third period (P-3) from 6th of May to 6th of June was slightly shorter than the other periods due to the additional effect of the Sumatran Winds. This period was actually the same as the fourth period (P-4), which is included in the southwest monsoon season for year 2007. The Sumatran winds occurred at the beginning of the southwest monsoon in May (MMD 2008) and are the normal winds which originate from the southwestern of Peninsular Malaysia and blow through the Sumatra island region. As a result, much of the air masses originating from Sumatra were also transported to our study area. Fundamentally, the west coast region of the Malaysia Peninsula is the main area receiving the majority of the air mass input from Sumatra. Nevertheless, the southwest monsoon normally brings a relatively drier season to the Kapar area with occasional rainfall activities occurring in early May and June, in the middle of July and at the end of August.

MEASUREMENT OF ²¹⁰Po

The radiochemical procedures proposed by Narita et al. (1989) were followed, where the dissolved samples fraction were put in pail to be spiked with 0.1 dpm of Polonium-209 tracer and 25 mg mL⁻¹ of ferum carrier. The aqueous was precipitated with ammonium hydroxide (NH_4OH) to form iron(III)hydroxide $[(Fe(III)(OH)_3]]$ precipitation and redissolved with diluted HCl for further radiochemical separations.

Period	Duration	Condition
1 st (P-1)	7 February – 15 March	 End of the northeast monsoon Very occasional and limited rainfall activity Weak wind from the northeast Increasing atmospheric temperature Warm and dry period
2 nd (P-2)	16 March – 5 May	 Monsoon transition More frequent rainfall activity of light to moderate intensity Weak wind with no particular direction Relatively humid period
3 rd (P-3)	6 May – 6 June	 Beginning of the southwest monsoon Very occasional rainfall activity Occasional strong wind from the northeast Significant influence from Sumatran Winds Dry period
4 th (P-4)	7 June – 30 August	 The southwest monsoon Weak wind from the southwest During June normally very dry period experienced During July also relatively dry but usually intensified rainfall received in middle of the month During early August very dry period experienced but more intensified rainfall activity towards the end of month

 TABLE 2. Summary of meteorological and climatic variation during sampling events obtained from the Malaysian Meteorology Department

Where P-1, P-2, P-3 and P-4 is post-northeast monsoon, monsoon transition, pre-southwest monsoon and southwest monsoon, respectively.

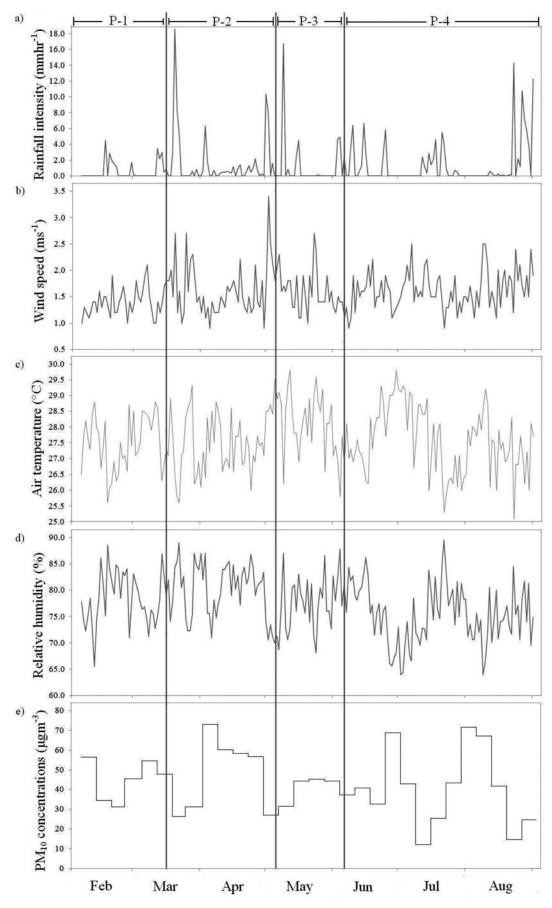


FIGURE 2. Meteorological parameters status during this study. Data was obtained from Meteorological Department of Malaysia (MMD 2008), (a) fainfall intensity, (b) wind speed, (c) air temperature, (d) Pelative humidity and (e) pm₁₀ concentrations

Meanwhile, particulate samples fraction were totally digested with concentrated HNO₃, HCl and HF to dissolve all particles into aqueous form in Teflon beaker after spiked with 1.3 dpm of Polonium-209 tracer. All digestion procedures were using hotplates and taken to dryness below 80 °C to prevent polonium from vaporized. Finally, dried samples were re-dissolved with diluted HCl for radiochemical separation procedures.

The determination of ²¹⁰Po was based on the spontaneous deposition of ²¹⁰Po on a silver disk (Flynn 1968). Before deposition, hydroxylamine hydrochloride and ascorbic acid were added as chelating agent and reduce Fe(III) into Fe(II), respectively. Polonium was spontaneously plated on silver discs using 50 mL of 0.5 M HCl solution at 70–80 °C for 3 h. The activity of ²¹⁰Po was measured using the Alpha Analyst Spectroscopy system with a silicon-surface barrier detector by Canberra, Inc., with Apex-Alpha software.

The polonium yield in analyzed samples was typically 60–80%. The system consists of silicone-surface barrier detector placed in standalone nucleus alpha spectrometers having negligible background (0.0003 cpm) and counting efficiency of 11 - 13%. The system was calibrated using mixed standard source of ²³⁰Th, ²³⁹Pu, ²⁴⁴Cu and ²⁴¹Am. A reference material, IAEA-300 (radionuclides in Baltic Sea Sediment), was used to check the performance of the analytical procedure. The obtained recovery value was always greater than 90%.

RESULTS

ACTIVITY CONCENTRATIONS OF ²¹⁰Po IN SOLID SAMPLES

Raw charcoal. Table 3 shows the ²¹⁰Po_{rc} concentrations from two origins (Indonesia and Australia) of raw charcoal (sub-bitumen class), as used by the power plant. In six replicates of each origin, the concentrations lie between 108.35 ± 20.97 and 206.86 ± 22.45 Bq kg⁻¹ with an average of 165.71 ± 56.79 Bqkg⁻¹ for the Indonesian samples and 99.26 ± 16.81 to 173.46 ± 22.89 Bq kg⁻¹ with an average of 145.03 ± 46.25 Bq kg⁻¹ for the Australian samples. These values are notably comparable to previous studies conducted in the US and Europe (Beck & Miller 1980; UNSCEAR 1982), Brazil (Flues et al. 2002) and China (UNSCEAR 1993). The radionuclide concentrations differ between each country of origin, suggesting that the concentrations of ²¹⁰Po in raw charcoal would be different based on their regions and locality to the coal mine. This study did in fact demonstrated that the SSAAPP uses raw charcoal which has a natural radionuclide activity up to four times higher than the lowest concentrations (10 Bqkg⁻¹) recorded in European countries (UNSCEAR 1982), although relatively lower than other countries such as Brazil which generally ranged between 20 and 250 Bq kg⁻¹ (Flues et al. 2002; Flues et al. 2007). Beck and Miller (1980) had previously discovered that the activities of ²¹⁰Pb and ²¹⁰Po in raw charcoal have the tendency to increase according to the excess sources.

TABLE 3. Activity of ²¹⁰Po in selected raw charcoals (²¹⁰Po_{rc}) used during SSAAPP operation

Charcoal origin	Number	Specific activity (Bqkg ⁻¹)
	Ι	150.45 ± 18.83
	II	108.35 ± 20.97
	III	175.66 ± 21.53
Indonesia	IV	206.86 ± 22.45
	V	191.70 ± 23.61
	VI	161.21 ± 24.10
	Average	165.71 ± 56.79
	Ι	160.45 ± 20.33
	II	173.46 ± 22.89
	III	99.26 ± 16.81
Australia	IV	138.33 ± 18.84
	V	145.50 ± 18.50
	VI	153.17 ± 10.64
	Average	145.03 ± 46.25

Suspended solids (SS_{rw}) in rainwater sample The ²¹⁰Po activities in the suspended solids of rainwater (²¹⁰Po_{sSrw}) samples collected from near the SSAAPP between February and August 2007 are shown in Table 4. The SS_{rw} samples, in the form of black particles, were collected for this study from within a 2 km radius and were found to be heavily dominated by the product of coal burning emissions from the power plant. The average ²¹⁰Po_{SSrw} activity recorded in this period was 274.01 ± 117.72 Bq kg⁻¹, which is almost twice from ²¹⁰Po_{rc} concentrations of raw charcoal used by SSAAPP. This finding is in accordance with UNSCEAR (2000), which states that the concentration of natural radionuclides are much higher in suspended solids of rainwater than raw charcoal samples (Table 3). Beck (1989) also found that the enrichment is normally around five to ten times, while Flues et al. (2006) found it to be between two and four times. In relation to these findings, Baxter (1993) concludes the enrichment of radionuclide activities are the result of vaporization and adsorption processes in atmospheric particles, particularly of a finer grain size (Beck & Miller 1980; Karangelos et al. 2004).

Suspended solids (SS_{ew}) in estuarine water The range of ²¹⁰Po activities in suspended solids of estuarine water (²¹⁰Po_{SSew}) was found to be between 43.79 ± 2.31 Bq kg⁻¹ and 151.14 ± 5.96 Bq kg⁻¹, with an average of 86.50 ± 2.41 Bqkg⁻¹ on March 8, 2007 as post northeast monsoon season. While on April 1st, 2007 as southwest monsoon season ranged from 53.51 ± 2.52 Bq kg⁻¹ to 161.21 ± 5.87 Bq kg⁻¹, with an average of 106.00 ± 1.55 Bq kg⁻¹ (Table 5). The previous study of ²¹⁰Po activities in suspended solids in Malaysian estuary by Theng & Mohamed (2005) in Kuala Selangor was between 27 ± 4 and 96 ± 4 Bq kg⁻¹.

Period	Sampling date	Site	Suspended solid phase (Bqkg ⁻¹)	Dissolved phase (mBqL ⁻¹)
		C 1	7.63 ± 0.67	n.a
	23-Feb-07	C 2	20.71 ± 0.74	13.42 ± 0.20
	25-160-07	C 3	34.52 ± 1.36	16.20 ± 0.38
P-1 Post-northeast monsoon		C 4	n.a	17.44 ± 0.31
	15-Mar-07	C 1	121.38 ± 5.43	22.10 ± 9.36
		C 2	175.42 ± 43.80	n.a
		C 3	73.82 ± 5.28	n.a
		C 4	364.48 ± 16.96	8.55 ± 4.93
	Average during P-1		113.99 ± 32.68	15.54 ± 11.13
		C 1	n.a	4.77 ± 1.88
	04.4.07	C 2	58.54 ± 4.91	7.85 ± 1.42
	04-Apr-07	C 3	150.59 ± 12.18	1.69 ± 0.44
		C 4	n.a	n.a
		C 1	233.61 ± 11.39	7.19 ± 1.80
		C 2	716.68 ± 54.90	6.77 ± 2.32
P-2	19-Apr-07	C 3	345.19 ± 11.82	7.05 ± 1.25
Monsoon transition		C 4	n.a	76.26 ± 4.06
		C 1	185.41 ± 15.22	6.57 ± 0.43
		C 2	152.31 ± 13.55	10.04 ± 0.67
	04-May-07	C 3	258.23 ± 22.39	6.29 ± 0.36
		C 4	n.a	10.80 ± 0.72
	Average duri		262.57 ± 55.80	13.21 ± 9.21
	C 1		448.50 ± 35.11	n.a
		C 2	202.41 ± 17.56	n.a
	17-May-07	C 3	503.93 ± 51.49	18.23 ± 1.52
D 2		C 4	187.89 ± 18.76	22.38 ± 1.51
P-3 Pre-southwest		C 1	196.97 ± 11.37	39.57 ± 8.71
monsoon		C 2	n.a	41.70 ± 9.14
	06-Jun-07	C 3	341.91 ± 28.73	61.39 ± 13.18
		C 4	414.47 ± 21.22	33.32 ± 4.46
	Average during P-3		328.01 ± 71.17	36.10 ± 14.97
	C 1		n.a	n.a
	22-Jun-07	C 1 C 2		
		C 2 C 3	n.a	n.a 10.03 ± 0.41
		C 3	n.a	10.03 ± 0.41 86.33 ± 6.51
	27-Jul-07		n.a	
		C 1	319.79 ± 24.58	12.85 ± 0.69
P-4		C 2	321.28 ± 7.79	5.13 ± 0.34
Southwest monsoon		C 3	176.11 ± 5.20	6.11 ± 0.23
		C 4	224.55 ± 6.92	14.49 ± 0.78
	30-Aug-07	C 1	n.a	12.55 ± 0.52
		C 2	744.28 ± 21.12	7.17 ± 0.58
		C 3	n.a	n.a
	C 4		691.62 ± 22.46	n.a
	Average during P-4		412.94 ± 41.66	19.33 ± 3.19
	Average during this study		274.01 ± 117.72	20.33 ± 22.66

TABLE 4. Activity of ²¹⁰Po in suspended solids (²¹⁰Po_{SSrw}) and dissolved (²¹⁰Po_{Drw}) phases of rainwater samples collected during this study

n.a : not available

The study stated that the main source of radioactivity in that river was from the agricultural activity by oil palm industry, while domestic and industrial waste releases would also important. Therefore, it will be applicable to say that the ²¹⁰Po activities in suspended solids of Kapar coastal region are highly influenced by the deposition of fly ash originating from coal combustions. The deposition of suspended solid or fly ash from coal burning emissions to the Kapar coastal area were notably higher during the second estuarine sampling as southwest monsoon period (P-4), which experienced more dry periods than the first estuarine sampling indicated as post northeast monsoon events (P-1). As a result, ²¹⁰Po_{SSew} activity concentrations in all sampling locations should record higher activities during the dry period during the southwest monsoon.

It was found that only S 1 and S 4 followed the above trend and that in contrary with the S 2 and S 3 profiles which were not similarly distributed. There are possible explanations for such behavior in the selected study areas, which generally due to the geochemical variability for every location. The S 2, which is located in the lower estuary, may have been strongly influenced by an increasing amount of run-off from the land by rivers and streams during post northeast monsoon events (P-1). The watershed and chemical weathering which brings agricultural, industrial and residential wastes during the heavier rainfall of the northeast monsoon are possible reasons for the enhancement of ²¹⁰Po_{SSew} activity at this particular location. These possible sources are also estimated to enhance the ²¹⁰Po_{SSew} activity to a level about twice higher than normally expected. This showed that the influence from geochemical variability is sometime more significant in shaping the natural radionuclide activity concentration in river and estuary of Kapar than by the power plant's effluents or emissions.

For S 3, the changes of ²¹⁰Po_{SSew} activity at this location are expected to fluctuate depending on the level of emissions from the SSAAPP cooling systems. S 3 constantly receives input from the SSAAPP water discharge, which was used to cool systems during SSAAPP operations. The concentrations at this location may not represent the typical concentrations at other sampling sites, irrespective of seasonal variations, since the disruptions arise from water discharge from the power plant.

Surface sediment Normally, natural radionuclide activity or other trace element measurements in sediment samples assess the enhancement of pollution or radioactivity levels in the selected area. Correspondingly, sediment samples are a vital tool. Technically, the seafloor is where every compound or matter is deposited and settle. Therefore, the accumulation of ²¹⁰Po in sediment can be used as an indicator of radioactivity levels from coal burning on the Kapar estuarine systems. Table 5 shows that the level of ²¹⁰Po_{sed} in this study was higher during the second estuarine sampling (southwest monsoon period) than the first estuarine sampling (post northeast monsoon), at 99.43 \pm 3.60 Bq kg⁻¹ to 137.08 \pm 4.68 Bq kg⁻¹, with an average of 117.06 ± 4.16 Bq kg⁻¹ and 65.66 ± 2.70 to 141.80 ± 17.59 Bq kg⁻¹, with an average of 86.06 ± 7.18 Bq kg⁻¹, respectively. Based on the descriptions by the Malaysian Meteorological Department (MMD 2008), the northeast monsoon brings heavy rainfall, particularly to the east coast states of Peninsular Malaysia and western Sarawak, whereas the Southwest Monsoon normally signifies relatively drier weather. The southwest monsoon is comparatively drier throughout the country except for the state of Sabah, Malaysia. During this season, most states experience monthly minimum rainfall (typically 100 - 150 mm) (MMD 2008). Thus, the activity of ²¹⁰Po

Sampling date/ Period	Sampling site	Suspended solids phase (Bq kg ⁻¹)	Sediment (Bq kg ⁻¹)	Dissolved phase (mBq L ⁻¹)
8-Mar-2007 Post northeast monsoon P-1	S 1	43.79 ± 2.31	141.80 ± 17.59	0.62 ± 0.03
	S 2	151.14 ± 5.96	65.66 ± 2.70	1.51 ± 0.04
	S 3	84.91 ± 4.73	68.16 ± 4.07	1.87 ± 0.17
	S 4	66.14 ± 8.07	68.62 ± 3.07	0.34 ± 0.03
	Average	86.50 ± 12.92	86.06 ± 12.95	1.09 ± 0.15
1-Aug-2007 Southwest monsoon P-4	S 1	99.78 ± 3.33	137.08 ± 4.68	16.55 ± 0.31
	S 2	109.50 ± 2.69	99.43 ± 3.60	22.44 ± 0.53
	S 3	53.51 ± 2.52	102.88 ± 3.30	4.69 ± 0.16
	S 4	161.21 ± 5.87	128.83 ± 5.05	9.01 ± 0.30
	Average	106.00 ± 7.69	117.06 ± 8.31	13.17 ± 0.74

TABLE 5. Activity of ²¹⁰Po in suspended solids (²¹⁰Po_{SSew}) and dissolved (²¹⁰Po_{Dew}) phases of estuarine water samples, and surface sediments (²¹⁰Po_{sed}) collected during this study

in this study may have been more accumulated between transition monsoon and southwest monsoon season of year 2007 than in the northeast monsoon of year 2006/07. This may be induced by the relatively warmer and drier southwest monsoon.

ACTIVITY CONCENTRATIONS ²¹⁰Po IN DISSOLVED PHASE

Rainwater In dissolved phase of rainwater (D_{rw}) sample, the activity concentration of $^{210}Po_{Drw}$ ranged from 1.69 \pm 0.44 to 86.33 \pm 6.51 mBq L⁻¹ (Table 4). The ²¹⁰Po_{Drw} activity in rainwater is also important as this measurement allows scientists to assess the dispersion processes and radionuclide deposition from the atmosphere. Rainfall is one of the essential processes which acts as a washing or removal agent for aerosol and air particulates in the atmosphere and deposits them onto maritime or continental surfaces. Nevertheless, radionuclide activities in precipitation samples were found to fluctuate, depending on the amount and period of rainfall (Sugihara et al. 1999), aerosol distribution and size (Balkanov & Sorensen 2001), meteorological factors (Martinez-Aguirre et al. 1991), among other factors. Such that, the concentration of ²¹⁰Po_{Drw} during this study correlated with the emissions from coal burning and the suspended solid or fly ash contents from the power plant.

Estuarine water ²¹⁰Po_{Dew} activity fluctuated at different locations from the upper river down to the coastal waters. The ²¹⁰Po_{Dew} activities at all sampling locations were recorded as being higher during the southwest monsoon period than the post northeast monsoon events. Based on Table 5, the ²¹⁰Po_{Dew} activity during March 8, 2007 sampling was ranging between 0.34 \pm 0.03 and 1.87 \pm 0.17 mBq L⁻¹, with an average of 1.09 ± 0.07 mBq L⁻¹. For August 1st, 2007 sampling, the $^{210}\mbox{Po}_{\mbox{Dew}}$ activity was varied between 4.69 ± 0.16 and 22.44 ± 0.53 mBq L⁻¹, with an average of 13.17 ± 0.33 mBq L⁻¹. In contrast to the solid phase samples, there were very distinct differences between to the two estuarine water samplings undertaken and samples were over 10 times higher. For this reason, it was possible to find the changes in radionuclide concentrations much more clearly in the dissolved rather than the particulate phases.

DISCUSSION

THE SEASONAL CHANGES AND CHARACTERIZATION OF ²¹⁰Po ACTIVITIES BETWEEN FEBRUARY AND AUGUST, 2007

Figure 3 and Table 5 show the ²¹⁰Po activities in two components: atmosphere and estuarine. As mentioned previously, the second estuarine sampling during southwest monsoon period recorded higher ²¹⁰Po activities than the first estuarine sampling on post-northeast monsoon events, except for ²¹⁰Po_{SSew} in S 2 (middle estuary) and S 3 (water discharge). With regard to S 2 and S 3, it can be assumed

that the activity during the second sampling (P-4) was only about half of the activity expected. Therefore, there has to be another possible factor to explain the lower activity level between these two locations.

The most plausible reason is perhaps related to their locations, in which, S 2 is situated at the mouth of the Sungai Kapar Besar, which is a mixing zone between freshwater and saline water, whereas S 3 is situated at the water discharge point from the SSAAPP which leads to a higher quantity of suspended solids as a result of re-suspension of bottom sediment thus explaining the mechanism of ²¹⁰Po re-dessolution from bottom sediment and intertidal mudflats (Carvalho 1997). Sholkovitz et al. (1978) and Theng and Mohamed (2005) had concluded that ²¹⁰Po in the dissolved phase from this mixing zone (like in S 2) is removed from the solution. This behavior is driven by a combination of co-precipitation with humic substances, Fe-Mn hydroxides and by adsorption onto suspended particles (Carvalho 1995; Swarzenski et al. 1999). These processes are deemed likely to enhance ²¹⁰Po activity in particulate forms such as suspended solid matter. Therefore, the concentrations of suspended solids in these regions are very important for adsorbing ²¹⁰Po in the dissolved phase.-

The ²¹⁰Po activities in surface sediment samples (²¹⁰Po_{sed}) may represent their accumulation at each location for 100 - 150 days. This is a reasonable hypothesis based on the ²¹⁰Po profile in suspended solids of rainwater (²¹⁰Po_{SSrw}) (Fig. 3) to profile in surface sediment samples $(^{210}Po_{sed})$ (Table 5). This theory can be verified through the ²¹⁰Po activities in suspended solids of rainwater (²¹⁰Po_{SSrw}) which increased between April and August, 2007, while those in dissolved phase of rainwater (²¹⁰Po_{Drw}) increasing from May to mid June. Such that, higher ²¹⁰Po activities in sediment samples during the southwest monsoon events reflect increments of ²¹⁰Po_{SSrw} and ²¹⁰Po_{Drw} during these periods (transition monsoon to southwest monsoon). Correspondingly, throughout this study, the first estuarine sampling in post-northeast monsoon accumulated ²¹⁰Po activities during the northeast monsoon, whilst the second estuarine sampling in southwest monsoon accumulated them during the southwest monsoon events.

Based on the results, it can be seen that all sampling sites recorded higher activity during southwest monsoon than post-northeast monsoon, as shown in Figure 3 and Table 5. Thus, the relationship between $^{210}Po_{Dew}$ activities and rainfall intensity were observed. The seasonal variation, referring to availability and intensity of rainfall events, controls the activity concentrations of $^{210}Po_{Dew}$ which gradually increased during the dry to wet period. These results suggest that the particulate reactive ^{210}Po has a rapid exchange to the dissolved phase during heavier and intensified rainfall.

Moreover, heavier rainfall activity leads to greater runoff from the land (from rivers, streams and underground water sources) and stronger ocean currents and tidal effects (Eisenbud & Gesell 1997; Monte et al. 2003). These factors



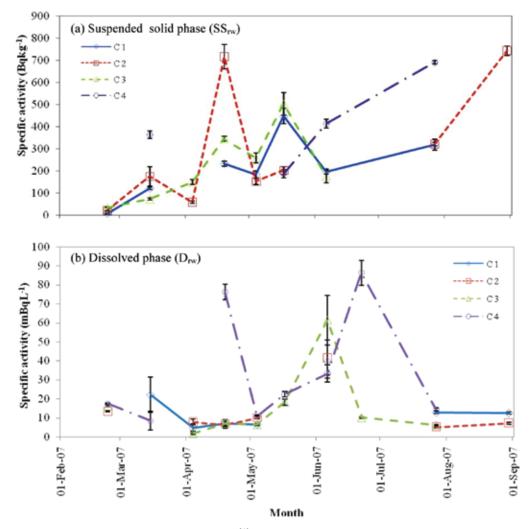


FIGURE 3. Distribution average activity of ²¹⁰Po in rainwater samples at various time and location of sampling events from February to September 2007 (a) suspended salid phase and (b) discoved phase

tend to enhance the mechanical and chemical weathering processes (Mas et al. 2007) and subsequently, lead to an increased concentration of ²¹⁰Po_{Dew} activity during higher rainfall activities in August. At the same time, heavier rainfall activity is also likely to enhance the re-suspension of bottom sediments, which result in higher concentrations of ²¹⁰Po in water.

With respect to ²¹⁰Po_{Drv}, the activity concentrations of ²¹⁰Po in the atmosphere positively correlated with a higher mean rainfall. Martin (2003) found higher ²¹⁰Po activity in the atmosphere was affected by the enhancement of biogenic aerosol or particulate content during the rainy season. However, Martin (2003) also concluded a negative correlation between rainfall intensity and the concentration of insoluble particulate matter or dust. Theses finding clearly prove the activity concentrations of ²¹⁰Po in the atmosphere is correlative to the amount of dust and rainfall.

THE TRANSFORMATION OF ²¹⁰Po FROM COAL BURNING ACTIVITY TO COASTAL ENVIRONMENT

This section, discusses the journey of ²¹⁰Po from coal combustion to emissions in the form of fly ash or suspended solids, before it is deposited onto the seafloor. For this reason, only the solid phase samples were selected. The ²¹⁰Po activities in the raw charcoal samples used by SSAAPP were very similar for both the Indonesian and Australian charcoals, with an average of 165.71 ± 56.79 Bq kg⁻¹ and 145.03 ± 46.25 Bq kg⁻¹, respectively (Table 3 and Figure 4). During P-1, the direct product of coal burning emissions (210Possrw) was recorded as being relatively lower than its predecessor, with an average of 113.99 ± 32.68 Bq kg⁻¹ (Table 4). The contents of ²¹⁰Po_{SSew} and ²¹⁰Po_{sed} within a 2,500 m radius of SSAAPP were also recorded with low ²¹⁰Po activities in relation to the suspended solids in rainwater samples, with 86.50 ± 12.92 Bq kg⁻¹ and 86.06 \pm 12.95 Bq kg⁻¹, respectively (Table 5). Nevertheless, the

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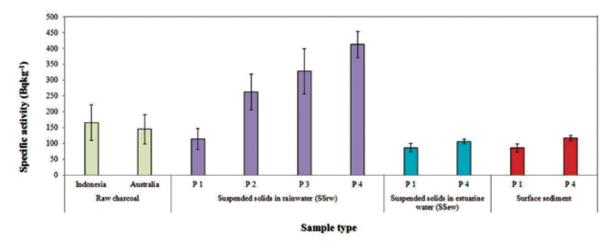


FIGURE 4. ²¹⁰Po activities in each of the solid samples analyzed during this study which reproduced from Table 3, Table 4 and Table 5 data to show temporal changes of 210Po from P-1 to P-4

patterns showed that ²¹⁰Po activities among suspended solids in rainwater, suspended solids in estuarine water and surface sediment samples were almost consistent and altered very little, lies between 86 and 117 Bq kg⁻¹.

With reference to the ²¹⁰Po activities in surface sediment samples ($^{\rm 210}{\rm Po}_{\rm sed})$ as discussed before, the first estuarine sampling during post-northeast monsoon was in essence collecting ²¹⁰Po sources throughout November 2006 and January 2007. These were much likely to have been affected by the northeast monsoon whilst ²¹⁰Po sources for the second estuarine sampling (southwest monsoon events) were collected between April and August 2007. For the record, the climate in the Kapar region during post-northeast monsoon was characterized by high rainfall intensity and atmospheric relative humidity, rather strong northeastern winds and a lower atmospheric temperature (MMD 2008). Even though our records demonstrate that the period February to March 2007 was associated with low rainfall activities and much slower winds from the northeast, which resulted in higher atmospheric temperatures, the aforementioned factors could be important causes as to why ²¹⁰Po activities were lower during this period of time as opposed to any other. These factors, which may also have been driven by the amount of coal burning emissions, are all major contributors for lower ²¹⁰Po activities during the first estuarine sampling.

For the suspended solid of rainwater samples, the average $^{210}Po_{SSrw}$ activities showed a distinct increment from monsoon transition (P-2) to southwest monsoon (P-4) of almost three times from the level at post-northeast monsoon (Figure 4). The monsoon transition (P-2) in Figure 3a, demonstrated that $^{210}Po_{SSrw}$ activities increased from the middle of March until the end of April for all locations, apart from C-4. The increment was affected by higher PM₁₀ concentrations and atmospheric temperatures (Figure 2) during April, in which more coal combustion

emissions were released. This result shows that the enrichment of ²¹⁰Po in suspended solid is associated with the concentration of particulate matter in air columns (Carvalho 1995; Martin 2003), which are induced by coal combustion emissions from the SSAAPP, particularly during warmer atmospheric temperatures. In contrast, during pre-southwest monsoon (P-3), one of the principal causes of higher ²¹⁰Po_{SSrw} activities was noted to be extreme weather. In this study, extreme weather was referred to an extremely dry or wet period, depending on the rainfall availability and atmospheric temperature. Figure 2 shows a dry period during the middle of May until June with no rainfall occurrence. However, it should be noted that when rainfall does occur, the intensity was eventually high and predominantly classified as heavy rainfall. Such that, it is possible to stipulate that the increasing atmospheric temperature during a dry period is one of the key factors of ²¹⁰Po enrichment in atmospheric samples. This is essential due to higher temperatures causing more ²²²Rn and its daughters' to emanate from earth, depending on the volatility of each radionuclide (Karangelos et al. 2004). P-4, which was actually the continuation of P-3, also experienced extreme weather. In this instance, higher ²¹⁰Po_{SSrw} activities were driven by higher SSAAPP emissions during July and August, according to the PM₁₀ concentrations documented, which reached 68.91 and 71.57 µgm⁻³, respectively (Figures 3 and 2(e).)

Figure 4 also presents a slight increment of ²¹⁰Po activities in suspended solids and surface sediment samples during P-4. This result demonstrates the enrichment by ²¹⁰Po in suspended solids of rainwater which increased from P-1 to P-4. Consequently, it can be verified that ²¹⁰Po enrichments in suspended solids of estuarine water and surface sediment in the Kapar estuarine and coastal region were significantly affected by the SSAAPP's coal burning emissions.

DISTRIBUTION AND DISPERSION OF ²¹⁰Po WITHIN THE SSAAPP REGION

One important question could be asked with respect to how coal combustion emissions release radionuclides and increase the ambient natural background radiation levels in both air and soil, thus potentially increasing the exposure of radiation to surrounding populations. Depending on the chimney's height (Beck & Miller 1980), the natural meteorological variability is able to disperse fly ash particles or suspended solids over a few kilometers. It is expected that the actual average values may vary from time to time due to variations in the coal burned, and other related factors. Therefore, in relation to the meteorological conditions in this study, it can be concluded that the majority of the suspended solids, as too radionuclide dispersion, were all transported over a greater distance. This assumption has been made based on the ²¹⁰Po activities in suspended solids of rainwater and dissolved phase of rainwater profiles versus distance from the power plant (Figure 5). Only during P-1 for both samples, and during P-2 and P-4 for rainwater samples, were ²¹⁰Po activities relatively low and occurring in a radius area about 1 km from the source.

Beck and Miller (1980) observed that the influence of air activity concentration at ground level is predominantly extended at approximately 1 km and above for short (50 m) chimneys and over 5-10 km for taller (150 m) chimneys. Beck (1989) also noted that old plants with a 50 m chimney presented an increase in soil radioactivity of up to 1,000 m. Observations of greyish-coloured soil between 200 and 600 m indicate possible contamination of the soil in the vicinity of the coal-fired power plant. Flues et al. (2002) documented a two- to three-fold increase in natural radionuclide concentrations, mainly for the uranium series (²³⁴Th, ²²⁶Ra and ²¹⁰Pb), in soils within 1 km of power facilities. But, also noted that at a distance greater than 1 km, statistically significant increases were not observed. This information is highly relevant and should be carefully deliberated over by the authorities when planning a newer and larger coal-fired power plant in the region.

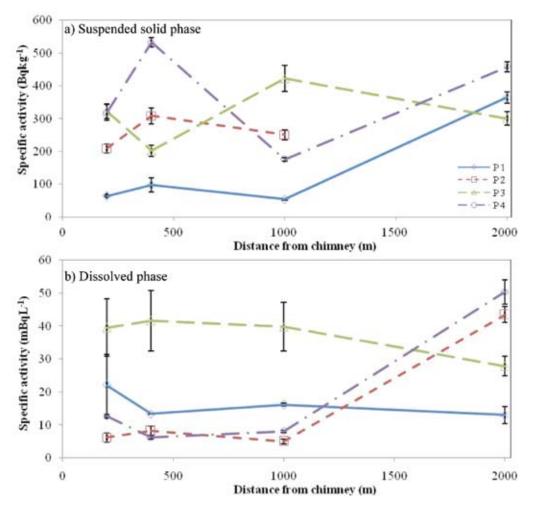


FIGURE 5. Distribution pattern of an average activity of 210Po collected from rainwater samples at various distance from chimney (a) suspended solid phase and (b) discolved phase

CONCLUSION

The specific activities of ²¹⁰Po in various environmental samples were significantly affected by the deposition of suspended solids in rainwater discharged from the coal-fired power plant of SSAAPP. Moreover, the ²¹⁰Po content in suspended solids of rainwater was heavily enriched from its original form (charcoal) by nearly three times the quantity. Distribution and accumulation also varied depending on climatic variations at the survey sites. This study showed that ²¹⁰Po accumulation in the study site was noticeably higher during the southwest monsoon than the northeast monsoon, owing to respective meteorological conditions. A further finding was that most of the suspended solids in rainwater containing ²¹⁰Po originating from SSAAPP burning emissions were transported over a distance of a few kilometers.

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