RADIOACTIVITY OF ^{210}PO IN THE ENVIRONMENTAL SAMPLES FROM KAPAR, MALAYSIA

Asnor Azrin Sabuti[1], Zaharudin Ahmad[2] and Che Abd Rahim Mohamed*[3]

[1]School of Environmental and Natural Resource Sciences, Faculty of Science and Technology Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia
[2]Malaysian Nuclear Agency, Ministry of Science, Technology and Innovation, 43600 Bangi, Selangor, Malaysia
[3]Marine Ecosystem Research Centre, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia
** carmohd@ukm.my; mohamed6566@yahoo.com*

ABSTRACT

Various environmental samples (seawater, TSS, sediment, rainwater and fly ash) from eight different stations near Kapar coastal area were analyzed. The ^{210}Po activity concentrations in liquid samples (seawater and rainwater) varied between 0.34 ± 0.03 mBq L^{-1} to 22.44 ± 0.53 mBq L^{-1}. Whereas the concentrations in particulate samples (TSS, sediment and fly ash) varied between 43.79 ± 2.31 Bqkg^{-1} to 364.48 ± 5.43 Bqkg^{-1}. Results also showed the radioactivity in Kapar coastal is higher than most of Malaysian coast, reaching a factor of seven. This condition is mainly due to the operation of a coal-fired power plant nearby. This study also clarify the variability of ^{210}Po in environment was strongly influenced from rainfall events especially during wet seasons.

ABSTRAK

Pelbagai sampel persekitaran (air laut, TSS, sedimen, air hujan dan abu terampai) daripada lapan stesen yang berbeza berdekatan persisir pantai Kapar telah dianalisis. Kepekatan aktiviti ^{210}Po di dalam sampel cecair (air laut dan air hujan) adalah berubah-ubah antara 0.34 ± 0.03 mBq L^{-1} hingga 22.44 ± 0.53 mBq L^{-1}. Manakala kepekatannya di dalam sampel partikel (TSS, sedimen dan abu terampai) berubah-ubah antara 43.79 ± 2.31 Bqkg^{-1} hingga 364.48 ± 5.43 Bqkg^{-1}. Keputusan juga menunjukkan keradioaktifan di kawasan persisir pantai Kapar adalah tinggi berbanding kebanyakan kawasan persisir pantai di Malaysia, menjangkau tujuh kali ganda. Keadaan ini berpunca daripada operasi penjanaan elektrik menggunakan arang batu yang berdekatan. Kajian ini juga turut menerangkan perubahan ^{210}Po di persekitaran adalah kuat dipengaruhi oleh kehadiran hujan terutamanya ketika musim basah (musim hujan).

Keywords: ^{210}Po; environmental samples; coal-fired power plant; rainfall events; wet seasons

INTRODUCTION

Research on radioactivity in environment has been very important in the last decades for its importance as a scientific basis for legislative measures to reduce the industrial and agricultural impact on the environment. The effect of radioactive exposure to human has been well discussed in the chemistry composition to the global climate. The enhancement of radioactivity can be contributed by both natural and human activities. Natural processes are mainly from cosmic-ray interactions and radioactive decay products of radon, which emanates from continental surfaces into the atmosphere (Poet *et al.,* 1972). Whereas, the threats from radioactivity through activities like mining, power generations and nuclear weapons are very harmful to mankind.

The use of coal as a fuel source for generating power is one of the source of radioactivity, where non-combustible trace elements are concentrated in the ash and disperse to the atmosphere and surrounding environment. Beck & Miller (1980) reported, based on mean ash content in U. S. coals of 13.4 percent, we would expect the specific activity of this ash to average 7.5 times that of the coal, i.e., 4.5 $pCig^{-1}$ for ^{238}U and each of its decay products (including ^{210}Po). They have also concluded that some of the more volatile trace elements are preferentially re-condensed on smaller particles. This results in an enrichment of fly ash relative to bottom ash, and particularly on the smaller fly ash particles, which are less efficiently removed by the emission control equipment.

The interpretation of coal and fly ash due to its radioactive significance has been made by U.S Geological Survey (USGS) in 1997 (U.S.G.S, 1997). The coal is largely composed of organic matter, but it is the inorganic matter in coal – minerals, trace elements – that have been cited as possible causes of health, environmental and technological problems associated with the use of coal. Some trace elements in coal are naturally radioactive. These radioactive elements include uranium (U), thorium (Th), and their numerous decay products, including radium (Ra) and radon (Rn). Although these elements are less chemically toxic than other coal constituents such as arsenic, selenium or mercury, questions have been raised concerning possible risk from radiation. In order to accurately address these questions and predict the mobility of radioactive elements during coal fuel-cycle, it is important to determine the concentration, distribution and form of radioactive elements in coal and fly ash.

Beck & Miller (1980) had concluded the radioactivity emissions enhancement from coal fired power plant has been a major issue. First, the amount of fly ash associated with amount of radioactivity released to the atmosphere were depends on the efficiency of the plant's emissions control equipment. Furthermore, the total amount of ash generated by the plant depends on the type and amount of coal burned. A typical modern 1000 MW plant will consume about 2.3×10^9 kg $year^{-1}$ of coal and produce about 3×10^8 kg $year^{-1}$ ash. The radioactivity of the coal also varied from different mine site. So that, the values of radioactivity releases from different power plant are vary from each other.

Therefore, the study of radioactivity emission, such as ^{210}Po is important and it is highly radioactive and chemically toxic element. ^{210}Po is non-conservative radionuclide in ^{238}U decay series products through the disintegration of ^{222}Rn. The physical half life of ^{210}Po is 138.4 days. The main source of this nuclide in environment which is comes from the earth's crust in ^{238}U decay series. This radionuclide is also intermediate members of the naturally occurring ^{238}U decay series and is recognized as tracers for naturally processes in the atmosphere. ^{210}Po is a particle-reactive radionuclide and tend to associate with particulate phases (Yang & Lin, 1992) and have a potential utility as tracers of particle/water transport and particulate scavenging (Swarzenski *et al.,* 1999). In marine environments, the radioactive disequilibrium of ^{210}Po and ^{210}Pb is probably the result of

preferential uptake of polonium by planktonic organisms and other biogenic particulate material (Heyraud & Cherry, 1983). Therefore, behavior of ^{210}Po is closely associated to zooplankton metabolic activity (Cherry *et al.,* 1975) and density of zooplankton in surface waters (Tateda *et al.,* 2003).

The purpose of the study is to determine the distribution of ^{210}Po concentrations in coastal area in which under an influence from a coal-fired power plant.

SAMPLING AND ANALYTICAL PROCEDURE

Kapar coastal is a semi-diurnal coastal area, where fishery activities are done in a small scale. It is located 56 km from Kuala Lumpur and only a few kilometers from Klang Port. The Sultan Salahuddin Abdul Aziz Power Plant (SSAAPP) has been operating since 1985 with capacity extent to 2420 MW with coal consuming 2.5 mtpa (million tonne per annum) of raw charcoal, as a main fuel and is the biggest power plant in Malaysia that contribute 23% of electricity demand to the country (TNB Generation 2003). Coal were imported from Australia, Indonesia, South Africa, China and Sarawak, Malaysia. The power plant is equipped with ESP (Electrostatic Precipitators) and its efficiency is up to 99.2 % (TNB Generation 2003). Table 1 and Figure 1 provide the details of the sample collections.

Table 1: Information for each sampling locations for this study

Sample type	Site number	Sampling location	Estimated distance from chimney (meter)	Sampling area (Salinity, psu)
Seawater	S 1	03º 07' 03" N 101º 19' 49" E	n.a	Upper estuary (0.1 - 17)
	S 2	03º 06' 27" N 101º 19' 44" E	n.a	Middle estuary (18 - 25)
	S 3	03º 07' 34" N 101º 18' 31" E	n.a	Water discharge (28 - 29)
	S 4	03º 06' 37" N 101º 18' 42" E	n.a	Jetty (27 - 28)
Rainwater	C 1	03° 07’ 01" N 101° 19’ 18" E	50	n.a
	C 2	03° 07’ 06" N 101° 19’ 32" E	150	n.a
	C 3	03° 07’ 19" N 101° 19’ 08" E	250	n.a
	C 4	03° 07’ 41" N 101° 19’ 09" E	500	n.a

n.a : not available

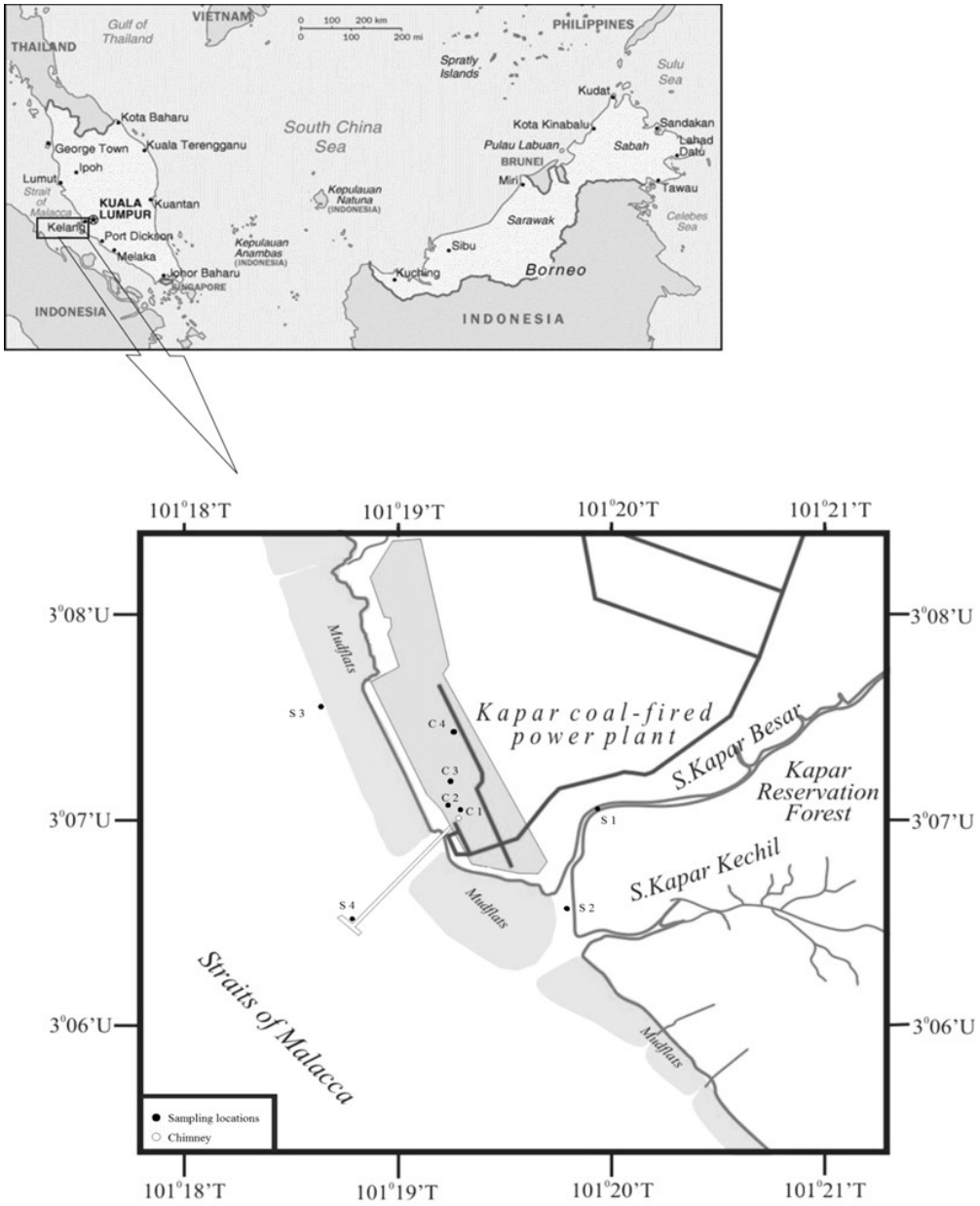

Figure 1: Map showing the sampling locations, where site C occupied as rainwater sampling zone and site S as seawater sampling zone.

The surface seawater samples were collected using Niskin water sampler and stored in 25 L polypropylene bottles. Meanwhile, the sediment samples were collected using Petite Ponar surface grab. Whereas the collection rainwater and fly ash samples were made using an approximately 660 cm² opening of a plastic pail placed 1 m to 15 m above the ground. Rain gauge also used for determining the rainfall events. Sample collected was store in the polypropylene container and acidified to pH 2 using nitric acid (HNO_3). The fractions between dissolved and suspended solid particles were separated using filter paper (Whatman GF/C; pore size 0.45 μm). The procedure of

determination of ^{210}Po activity concentrations was done using a published method proposed by Narita et al. (1989). ^{209}Po and ^{210}Po activities were determined by alpha spectrometry (Canberra Alpha Analyst Spectroscopy system, with Apex-Alpha software).

RESULTS

Radioactivity of Kapar environmental samples

The activity concentrations of ^{210}Po in environmental samples for are shown in Table 2 and Table 3. Overall, the activities of ^{210}Po in dissolved phase were very low relative to the solid phase. In brief, the activities of ^{210}Po in seawater samplings were in the range of 0.34 ± 0.03 mBqL^{-1} to 22.44 ± 0.53 mBqL^{-1} for dissolved samples, 43.79 ± 2.31 Bqkg^{-1} to 161.21 ± 5.87 Bqkg^{-1} for suspended solids and 65.66 ± 2.70 Bqkg^{-1} to 141.80 ± 17.59 Bqkg^{-11} for sediment samples. Meanwhile, activities of ^{210}Po in rainwater samplings were in the range of 1.36 ± 3.72 mBqL^{-1} to 22.10 ± 9.36 mBqL^{-1} for dissolved and 73.82 ± 5.28 Bqkg^{-1} to 321.28 ± 7.79 Bqkg^{-1} for fly ash samples. The results from fly ash samples show two times higher than suspended solids and sediment samples, unlike dissolved samples for seawater and rainwater samples, which had a similar range. Thus, the variability and changes of ^{210}Po activities in solid samples seems to be higher than dissolved samples.

Table 2: ^{210}Po activity concentrations from seawater samples

Sample	Sampling date (Mean rainfall) (Season)	$^{210}Po_{ES}$ (mBqL^{-1})	$^{210}Po_{TSS}$ (Bqkg^{-1})	$^{210}Po_{SED}$ (Bqkg^{-1})
S 1	March, 8	0.62 ± 0.03	43.79 ± 2.31	141.80 ± 17.59
S 2	(0.06 mm day^{-1})	1.51 ± 0.04	151.14 ± 5.96	65.66 ± 2.70
S 3	(Dry)	1.87 ± 0.17	84.91 ± 4.73	68.16 ± 4.07
S 4		0.34 ± 0.03	66.14 ± 8.07	68.62 ± 3.07
S 1	August, 1	16.55 ± 0.31	99.78 ± 3.33	137.08 ± 4.68
S 2	(0.15 mm day^{-1})	22.44 ± 0.53	109.50 ± 2.69	99.43 ± 3.60
S 3	(Wet)	4.69 ± 0.16	53.51 ± 2.52	102.88 ± 3.30
S 4		9.01 ± 0.30	161.21 ± 5.87	128.83 ± 5.05

*Mean rainfall are based from record of past 14 days rainfall data collection
$^{210}Po_{DISS}$: dissolved ^{210}Po
$^{210}Po_{TSS}$: ^{210}Po in suspended solids
$^{210}Po_{SED}$: ^{210}Po in sediment

By comparing the ^{210}Po activity concentrations in dissolved samples to the previous study from Theng & Mohamed (2005), this study showed almost 20 times higher. But, the ^{210}Po activity concentrations in suspended solids were only two times higher. Based from this aspect, a significant contribution from each different locality could be more important. It is because, the major input to each region could be different from each other. So that, from this study, the input from coal-fired power plant could be important into enhancing the radioactivity. For a comparison, the Kuala

Selangor coastal area is a fishermen village and also a good site for tourism industry as it is known for the firefly habitat along the mangrove area. On the other side, the Kapar coastal area has a much more radioactivity sources due to the activity of coal-fired power plant nearby. In fact, the previous study by Beck & Miller (1980) showed a coal-fired power plant manages to enhance the natural radioactivity to average five or seven times.

Table 3: ^{210}Po activity concentrations from rainwater samples

Sample	Sampling date (Mean rainfall) (Season)	$^{210}Po_{RW}$ ($mBqL^{-1}$)	$^{210}Po_{ASH}$ ($Bqkg^{-1}$)
C 1	March, 15	22.10 ± 9.36	121.38 ± 5.43
C 2	(0.11 mm day^{-1})	1.36 ± 3.72	175.42 ± 43.80
C 3	(Dry)	3.14 ± 3.71	73.82 ± 5.28
C 4		8.55 ± 4.93	364.48 ± 16.96
C 1	July, 27	12.85 ± 0.69	319.79 ± 24.58
C 2	(0.28 mm day^{-1})	5.13 ± 0.34	321.28 ± 7.79
C 3	(Wet)	6.11 ± 0.23	176.11 ± 5.20
C 4		14.49 ± 0.78	224.55 ± 6.92

*Mean rainfall are based from record of past 14 days rainfall data collection
$^{210}Po_{DISS}$: dissolved ^{210}Po
$^{210}Po_{ASH}$: ^{210}Po in fly ash

DISCUSSIONS

The seasonal variations of ^{210}Po activity concentrations in the coastal and estuary

Based from the results obtained, all sampling site recorded higher activity during the wet season than during the dry season, as shown in Figure 2. Thus, the relationship between ^{210}Po activity and the rainfall amount were observed. The seasonal variation, referring to the availability and the intensity of rainfall events, was strongly controlling the activity concentrations of ^{210}Po in dissolved levels, in which increased significantly during dry to wet season (March 8 to August 1) (Figure 2.a). These results could be suggesting a theory that the particulate reactive ^{210}Po have a rapid exchange to the dissolved phase during heavier and intensified rainfall. The other explanation could also possibly due to the supply of polonium from agricultural activities such as oil plantations and farming, which come together during seasons of heavy rain. This fact is true as the activity concentration of ^{210}Po is more associated with biogenic particles, such as agricultural products (Germain et al., 1995; Cherry & Heyraud, 1981). So that, during wet season, more polonium input in dissolved samples can be estimated.

In the marine environment, ^{210}Po is mainly produced from the decay of ^{210}Pb. It means that, the suspended solids samples (refer as TSS) in surface layer are reflecting their main source is coming from atmospheric deposition (Tan et al. 2007; Ugur et al. 2002). The input of fly ashes to the coastal area can be assumed to be high during dry season, March 8 (mean rainfall: 0.06 mm day^{-1}) sampling than August 1 (rainfall: 0.15 mm day^{-1}). But in contrast, from Figure 2.b, the activities of $^{210}Po_{TSS}$ were not constantly distributed as the S 1 and S 4 were not same as S 2 and S 3. It showed that, for the influence of river run off during wet season in increasing the agricultural input was more

significant that the influence from fly ash input to the variability of $^{210}Po_{TSS}$ activity concentration in S1. Meanwhile, the high activity concentration of $^{210}Po_{TSS}$ at the S 4 during wet season than the dry season may be controlled by other source such as fallen fragments of particulate coal during the loading of coal from ship to the power plant.

However, the concentrations of $^{210}Po_{TSS}$ in suspended solids (TSS) were higher than the sediment samples in S 2 sampling site with 151.14 ± 5.96 $Bqkg^{-1}$ and 109.50 ± 2.69 $Bqkg^{-1}$ for March 8 and August 1, respectively. Also, the activity concentrations of ^{210}Po in suspended solids from this study were found to be slightly variable from each sampling site. One reason is may be caused by variable sizes of particles in sediments in S 2. Carvalho (1995) was suggesting, this behaviour may reflect from the finer grain size of suspended matter in this region compared to other sampling sites.

Meanwhile, the site S 3 gave variable results for ^{210}Po in dissolved and particulate samples. The low activity concentrations of dissolved ^{210}Po in waste water discharge from the power plant were associated with relatively higher activity concentrations of ^{210}Po in suspended solids and sediment. Furthermore, the particulate matter in waste water discharges is likely to deposit near the discharge point. Thus, the sediment samples in the mouth of the outlet have high concentration of ^{210}Po activity. These concentrations may not represent typical concentrations like other sampling site, since the interruption arises from power plant water discharge.
Radionuclide activity measurement in sediment samples can be use as assessment of radionuclide levels in the selected area. The results showed from Figure 2.c, the release of ^{210}Po to the Kapar estuarine systems has been increasing during wet season, which may be controlled by the larger amount of rainfall that contributed more ^{210}Po sources from wider area. Except for S 1, the activity of ^{210}Po was slightly higher during dry season and relatively high concentrations than other sampling site. The high value of ^{210}Po level in sediments may correspond to the accumulation of radionuclides from industrial and residential wastes from Kapar town and also accumulation of agricultural wastes from palm oil plantation nearby. It also may due to the extreme enrichment of insoluble coal or fly ash from the power plant to this area.

The seasonal variations of ^{210}Po activity concentrations in rainwater

The activity concentration of ^{210}Po in rainwater samples as well as the fly ash content varied consistently with sampling time and condition. The rainwater and aerosol samples represent the activity of natural radionuclide in the atmosphere. Figure 3.a showed, most of the ^{210}Po activity concentrations in rainwater (dissolved) and fly ash samples were higher during wet season, based on July 27 sampling (mean rainfall: 0.28 mm day^{-1}) compared to dry season of March 15 sampling (mean rainfall: 0.11 mm day^{-1}), except for rainwater sample for C 1 and fly ash sample for C 4. These values are showing a good assumption, an increase of ^{210}Po activity concentration in atmosphere with increase rainfall activity. This is also evidence that higher ^{210}Po activity in atmosphere due to the enhancement of biogenic aerosol or particulate content during wet seasons. Ambe & Nishikawa (1987) concluded as negative correlations between rainfall intensity and concentration of insoluble particulate matter or dusts, a good combination to prove the relationship of particulate reactive radionuclide (such as ^{210}Pb and ^{210}Bi) concentration and amount of dust and amount of rainfall. But in ^{210}Po case, the activity concentrations of ^{210}Po in atmosphere were positively correlated with rainfall intensity.

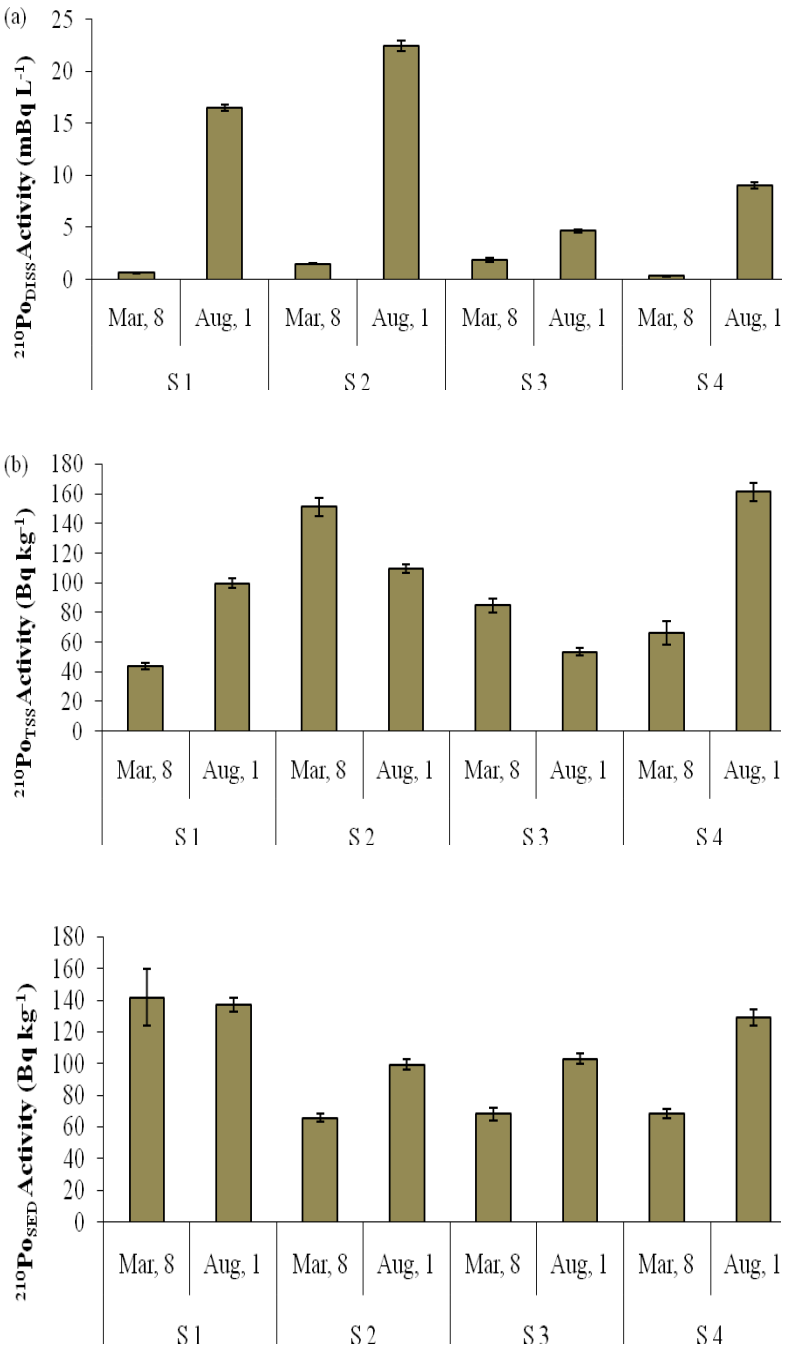

Figure 2: Activity concentrations of ^{210}Po from Kapar coastal area in (a) water, (b) suspended solids (TSS) and (c) sediment samples through different sampling location and date.

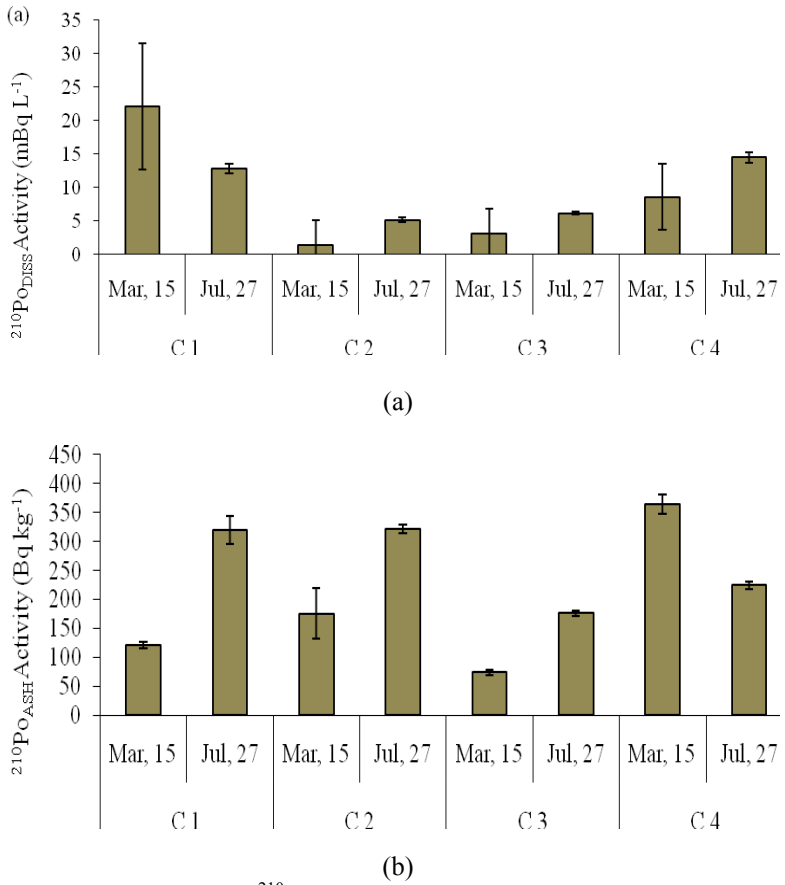

Figure 3: Activity concentrations of ^{210}Po from Kapar Power Station in (a) rainwater and (b) fly ash samples through different sampling location and date.

Based from Figure 3 were also possible to estimate the ^{210}Po dispersion from chimney point. Thus, it showed high ^{210}Po activities in C 1 for dissolved samples and C 2 for fly ash samples. The trend is suggesting inside 150 m radius from the coal-fired power plant area, a possibility to get high concentration of ^{210}Po in dissolved samples were also high. Meanwhile for ^{210}Po in solid samples were high inside 250 m area. This is perhaps due to the significant influence from the power plant operation and coal burning process. In contrast, the lowest activity is around 250 m, which may consider being the least affected area. Later, the ^{210}Po activity concentrations were increasing from 500 m extended to about 10 km, as concluded by Flues et al. (2002). These curve trends were cohesive and quite interesting to get possible explanation referring to the depletion of ^{210}Po activities around 250 m area. We are suggesting the height of chimney could also be important to disperse the ^{210}Po, which bounded into the fly ash. As mentioned by Beck & Miller (1980), the influence of deposition of fly ash from 150 m tall chimney is only started from 1 km, while the maximum levels of fly ash deposition occur at 6 km. This means, the depleted values around 250 m could possibly because the height of chimney, did not affected this area as much as it affected the longer distance. And inside 250 m, the high ^{210}Po concentrations were resulted by higher polluted air containing dust and fly ash due to the coal combustion, coal and dust transportation and soil re-suspension inside the power plant area.

CONCLUSIONS

Based from the comparison to the previous researches from estuary ecosystems, the ^{210}Po activity concentration has been interestingly increased in average to five or seven times due to the operation of coal-fired power station. Moreover, the high concentration of ^{210}Po and perhaps, other natural radionuclide is mainly associated with high content of fly ash from coal combustion to the atmosphere and marine systems. Then, the influence of rainfall events, referred as wet and dry season, had also contributed to the dispersion of ^{210}Po activity. The rainfall events could affect the amount of fly ash deposition from the power plant as well as its transportation to other places. During wet season, the influences from excess supply of polonium from agricultural activities and biogenic particles through the rain have been more important. Whereas during dry season, the fly ash has been directly influenced the activity of ^{210}Po in atmosphere as well as marine environments. In terms of pollution effects from the power station, dry season contributed more ^{210}Po activity in atmosphere. Meanwhile, due to various sources from wider area such as river run-off, groundwater run-off and atmospheric deposition has been contributed higher ^{210}Po activity in the sediments.

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