

## SPATIAL VARIATION OF THE RADIOACTIVE ELEMENT POLONIUM-210 IN A COAL BURNING POWER PLANT AREA OF MALAYSIA

**<sup>1</sup>LUBNA ALAM, <sup>2</sup>MAZLIN MOKHTAR, <sup>3</sup>MIR SUJAUL ISLAM**

<sup>1</sup>Institute for Environment and Development (LESTARI), National University of Malaysia, Bangi, 43600, Selangor

<sup>3</sup>Faculty of Civil Engineering & Earth Resources, University Malaysia Pahang, 26300 Gambang Kuantan, Malaysia

E-mail: lubna\_762120@yahoo.com

This study presents the results of Po-210 analysis in water, total suspended solid and sediment samples collected from a coal burning power plant area of Malaysia. Hierarchical cluster analysis has been used to classify the sampling station based on the activity of Po-210. The calculation of flux allowed examining the vulnerability of study location based on the concentrations of Po-210. Among all the stations, ash pond is detected as the most vulnerable location and the less affected area is the inlet. Even though the fluxes of water and sediment are not so high, the coastal sea became the second most vulnerable station because of the higher Po-210 activity in TSS. As a whole, the study clearly indicates the impact of coal burning on the marine environment of Kapar coastal area of Malaysia.

**Keywords-** Polonium, Coal, Water, sediment, TSS

### I. INTRODUCTION

Man and his environment are interconnected to each other while man requires everything from his environment, involving air, water, food, fuel and material resources. Therefore, the beginning of the new millennium seems to be characterized by a steadily increasing attention being paid to the environmental pollution and emphasis has given on marine pollution as the Ocean covers approximately 70% of the earth's total surface area. Additionally, the issue of present and potential radioactive contamination in the marine environment has received considerable attention. The most important radioelements are <sup>40</sup>K and uranium and thorium series known to have a very long half-life and to be abundant in the environment. Po-210 is an alpha emitter within the U-238 decay series and among the natural radionuclide occurring in the ocean, alpha emitters are considered to be the most important because of the radiation exposure. The distribution and behaviour of the natural-series radionuclide polonium-210 in the marine environment has been a focus of study for nearly 50 years. In the beginning, the primary interest in this alpha emitter started off from the moderately high levels of Po-210 measured in some marine biota and the enhanced natural radiation dose they receive from it<sup>1</sup>. Nevertheless, it was noted that Po-210 levels in the sea may become enhanced due to direct anthropogenic inputs from the atmosphere and land runoff, e.g. coal combustion and certain mining, industrial, shipping and agricultural activities, which could result in increased public and occupational radiation exposure<sup>2 3 4 5</sup>. The disequilibria between Po-210 and Pb-210 in seawater and suspended particles have been studied by oceanographers as a possible analogue for assessing particulate organic carbon (POC) flux through the water column<sup>6 7</sup>. However, to adequately answer

such questions about radiation doses as well as to better understand how Po-210 can be best used to measure specific marine biogeochemical processes, an in-depth knowledge of the behaviour of this radionuclide in a variety of marine species and food chains is necessary. Therefore, the aim of this present study is to investigate the spatial variability of Po-210 in a coal burning power plant area of Malaysia.

### II. MATERIALS AND METHODS

The study was conducted at Kapar coastal area (Figure1) which is very adjacent to Sultan Salahuddin Abdul Aziz power station.

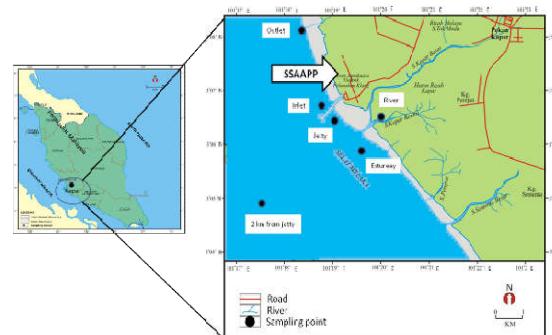


Figure 1. Map of sampling location at Kapar coastal area.

This power station is located at the western coast line of west Malaysia, at the Malacca strait, normally known as the largest power station in Malaysia with generating capacity of 2420 MW and contributes about 23% of the country's energy demand. This power station is the first power station with triple fuel firing capability (gas, oil and coal) in Malaysia. A total of 6 sampling stations were established along the coastal area and one station, ash pond, was selected inside the power station.

Samplings were carried out six times for a period of 15 months from November 2007 to February 2009. Approximately 25 L water samples were collected using polypropylene bottle and sediment samples were collected using Petite Ponar surface grab during each sampling trip. The sediment and water samples transported to the laboratory on the day of sampling. The sediment samples were kept in freezer for further analysis.

In the laboratory, the sediment samples were oven dried at 60°C temperature until dryness. After drying, the sediment samples were weighted again to obtain the dry weight. After that it was demolished to the ground by using mortars (Gelman No. 4012). Then the sediment samples in the form of powder sifted using a sieve (Retsch) in size of 200 µm. The smooth and homogeneous sediment sample stored in zip plastic bags and labeled for further analysis. Around 0.5 g of dried sediment sample was weighted in a beaker with electronic scale (model Dragon 204; Metler Toledo). A known activity of Po-209 tracer (National Institute of Standard & Technology, NIST) was added to determine the chemical recovery of the radiochemical separation. All the samples were digested on hot plate until dryness with sequential of concentrated mineral acids as follows: 30 ml of HNO<sub>3</sub>, 30 ml of HClO<sub>4</sub> and 30 ml of HCl<sup>8</sup>. After digestion, samples were added with 3.75 ml of concentrated HCl and kept it overnight for fully dissolution process. After that, samples were added with distilled water until 10 ml and filtered through a 0.45 µm Whatman GF/C (47 mm) glass fiber filter. Then, polonium isotopes in 90 ml of 0.5 M HCl were spontaneously plated on the silver disc after adding ascorbic acid to prevent Fe deposition for 3 hours<sup>9</sup>. The silver discs were counted in α-spectrometry for 24 hours and its activity corrected to the date of sampling. Meanwhile, capability of radiochemical separation method for Po-210 was verified using IAEA-300 (Radionuclides in Baltic Sea Sediment) reference material issued by IAEA. The mean of analytical recovery was obtained as 80% and the results were under 95% confidence interval (Table 1)

**Table 1 Analytical result for the reference materials IAEA-300 (Radionuclides in Baltic Sea Sediment) along with the certified value for Po-210.**

| SRM replicate | Obtained concentration (Bq/kg) | Certified concentration (Bq/kg) | Recovery (%) |
|---------------|--------------------------------|---------------------------------|--------------|
| 1             | 280.44±10.25                   |                                 | 80           |
| 2             | 345.19±12.14                   |                                 | 92           |
| 3             | 260.22±5.12                    |                                 | 85           |
| 4             | 375.35±15.55                   | 340.5<br>(273.6-361.0)*         | 70           |
| 5             | 295.28±8.85                    |                                 | 62           |
| 6             | 345.65±8.23                    |                                 | 91           |
| Mean          | 317.02±10.02                   |                                 | 80           |

\* 95% confidence Interval

Water samples were filtered to separate the Total Suspended Solid (TSS) through pre-weighted Whatman® cellulose filter paper. This filtered water was acidified with concentrated nitric acid (HNO<sub>3</sub>) and maintained the pH ≤2. Then about 0.1ml of 25mg l<sup>-1</sup> Fe<sup>3+</sup> as carrier and 0.05 ml of 26.704 dpm ml<sup>-1</sup> <sup>209</sup>Po as yield tracer were added into the water samples. After that, Na<sub>2</sub>CO<sub>3</sub> were added into the sample and precipitated with ammonium hydroxide (NH<sub>4</sub>OH). The iron (II) hydroxide [Fe(II)(OH)<sub>2</sub>] precipitate was dissolved with nitric acid and perchloric acid (HClO<sub>4</sub>). After heating 15 minutes, NH<sub>4</sub>OH was added to maintain the pH 8 and centrifuged for separating the solution and colloidal due to obtain solid Fe(OH)<sub>3</sub> precipitate. The precipitated residual was dissolved by HClO<sub>4</sub> and dried at 70°C temperature. After this, it was dissolved by 80 ml 0.5 M HCl and added a small amount of ascorbic acid to reduce the oxidation state of Fe<sup>3+</sup> to Fe<sup>2+</sup>. Silver plate of 2cm×2cm was hooked into the beaker and Po-210 was deposited spontaneously onto the plate at about 70°C temperature for 3 hours. After this the Po-210 activity was determined by alpha spectrometry.

On the other hand, the weight of TSS samples were measure by deducting the filter paper weight. TSS samples with filter paper were oven dried at 60°C temperature and the radiochemical separation and counting of Po-210 were carried out according to the method described for sediment.

In general, Po-210 activities involving tracer addition and counting by alpha spectrometry system calculated by following equation:

$$\text{Activity of Po-210} = (\text{actual Po-210 / actual Po-209}) \times (\text{tracer added} \times \text{tracer activity}) / \text{sample weight} \dots\dots\dots (1)$$

Radionuclide activity at the date of sampling has been calculated according to the following equation:

$$A = A_0 e^{-\lambda t} \dots\dots\dots (2)$$

Where:

A = Current amount of radioactivity

A<sub>0</sub> = Original amount of radioactivity

e = base natural log (approximately 2.718)

λ = the decay constant = 0.693/t<sub>1/2</sub> (where t<sub>1/2</sub> = half-life)

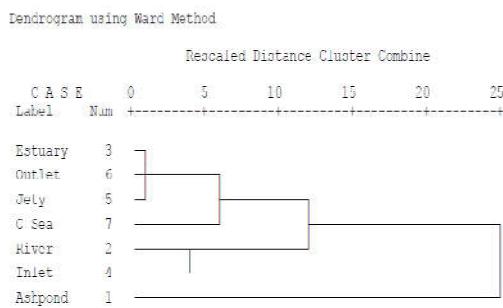
t = the amount of time elapsed from A<sub>0</sub> to A

### III. RESULTS AND DISCUSSION

The analysis of seawater samples revealed that the mean value of Po-210 concentration in dissolved phase collected from different locations during different sampling periods was 0.69±0.34 mBq/l, with the minimum and maximum values of 0.12±0.01 and 1.82±0.04 mBq/l respectively. Po-210 concentrations in the particulate phase ranged from

$24.60 \pm 1.1$  to  $348.29 \pm 15.54$  Bq/kg (dw) with the mean value of  $85.90 \pm 65.62$  Bq/kg (dw). The descriptive statistic shows that the concentration of Po-210 in surface sediment samples ranged from  $13.74 \pm 0.61$  to  $68.59 \pm 3.06$  Bq/kg dry wt with the mean value of  $32.95 \pm 14.85$  Bq/kg dry weight.

Hierarchical cluster analysis was used to detect similar groups between the sampling sites in six sampling periods at Kapar coastal area (Figure 2). The dendrogram revealed that at the rescaled distance of 25 units all the stations were similar, while at around 1 unit of such distance all the stations were distinct from each other. According to the dendrogram, two separate groups have been recognized. One includes ash pond and the other group includes other six stations. The cluster analysis also revealed very close similarities between estuary, outlet and jetty.

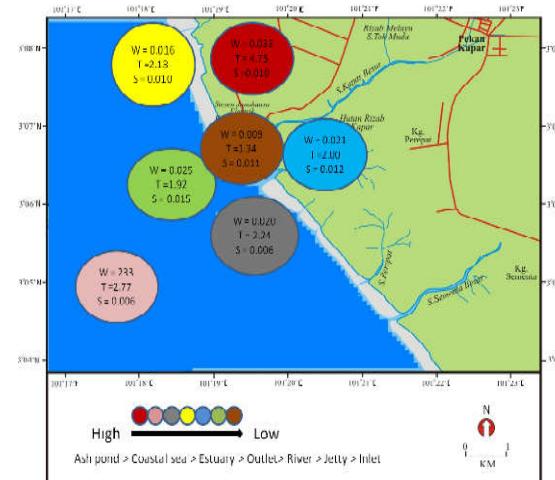


**Figure 2.** Dendrogram for hierarchical cluster analysis of the sampling location based on the concentrations of Po-210 in water, TSS and sediment.

Furthermore, the flux in dissolve phase of water ranged from 0.003 to 0.051 Bq/m<sup>2</sup>/day and showed a wide variation of two orders of magnitude from place to place (Table 2). The flux of TSS obtained from 0.68 to 9.69 Bq/m<sup>2</sup>/day whereas, in sediment sample ranged from 0.003 to 0.023 Bq/m<sup>2</sup>/day. Based on the calculated value of flux, the spatial variability of Po-210 in the coastal area of Kapar is presented in Figure 3. The mean flux of Po-210 in water and TSS samples of ash pond was considerably higher than that of other stations because the burnt ash from power plant which is deposited in the ash pond. Therefore the main source of high Po-210 in the pond is the burnt coal of the power plant. On the other hand the higher activity of Po-210 observed in the pond could be attributed to its lentic nature where the Po-210 attached to aerosols get deposited on water surface to higher degree in contrast to that of the running waters where there is a possibility of the deposited radionuclide being carried away owing to its lotic condition<sup>10</sup>. The higher background activity of Po-210 in pond water may also be due to the contribution from microorganisms. The mean flux of Po-210 in water sample of estuary was higher than

the sample of river. This elevated activity at estuary is because of the terrestrial input via river and also the impact of coal burning.

On the contrary, measurement of TSS samples shows a large difference with generally much higher level of Po-210 activities at the station of coastal sea. Moreover, the station coastal sea demonstrating lower dissolved Po-210 flux comparatively to the river. In this case it is assumed that most of the Po-210 was bound to the particulate phase because of the depth and stability of water flow. Additionally, the stability of the Po-210 activity in soluble form also depends on the presence of organic matter. So it can be assumed that in the river and estuarine water the Po-210 activities are higher because of the input of terrigenous materials. Interestingly, the flux of Po-210 in sediment of coastal sea was comparatively lower. This is because of the lotic character of the coastal sea where the Po-210 can not sink properly to the sediment. On the basis of average flux, the study area has been classified for vulnerability (Figure 3). In general, among all the stations, the ash pond is detected as the most vulnerable area and the less affected area is the inlet. Even though the fluxes of water and sediment are not so high, the coastal sea became the second most vulnerable station because of the higher Po-210 activity in TSS.



**Figure 3.** Spatial variation of Po-210 concentration in Kapar coastal area.

The three most important stations of study area are inlet, jetty and outlet, which are very adjacent to the power plant, demonstrating very dramatic changes in Po-210 flux. It has been seen that the higher Po-210 fluxes were associated with jetty which is the transfer path of coal in the power plant. The outlet is also demonstrating the higher activity where the hot water discharges from the power plant. In areas where hydrothermal activity is strong, enhanced scavenging activity of Po-210 was observed<sup>11</sup>. Among these three stations the Po-210 flux is lowest at inlet where

there is a continuous flow of water. On the other hand, in case of sediment, the activity of Po-210 was higher at jetty which is deeper than other stations. As a result, it can be concluded that this higher flux is because of the depth where the sediment is less influenced by the surface water circulation and the Po-210 can be deposited properly.

**Table 2 : Calculated values of flux ( $\text{Bqm}^{-2}\text{day}^{-1}$ ) based on the concentration of Po-210 in water, TSS and sediment.**

| Sampling date |             | Flux ( $\text{Bqm}^{-2}\text{day}^{-1}$ ) |       |       |          |
|---------------|-------------|---|-------|-------|----------|
|               |             | Stations                                  | Water | TSS   | Sediment |
| Nov-07        | Ash pond    | NA  | NA    | NA    |          |
|               | River       | 0.023                                     | 1.35  | 0.007 |          |
|               | Estuary     | 0.025                                     | 2.22  | 0.005 |          |
|               | Inlet       | 0.009                                     | 1.44  | 0.009 |          |
|               | Jetty       | 0.030                                     | 2.48  | 0.012 |          |
|               | Outlet      | 0.022                                     | 1.69  | 0.006 |          |
|               | Coastal sea | 0.015                                     | 4.14  | 0.006 |          |
| Feb-08        | Ash pond    | 0.025                                     | 2.36  | 0.004 |          |
|               | River       | 0.006                                     | 0.68  | 0.009 |          |
|               | Estuary     | 0.007                                     | 0.73  | 0.005 |          |
|               | Inlet       | 0.003                                     | 0.72  | 0.012 |          |
|               | Jetty       | 0.022                                     | 1.22  | 0.008 |          |
|               | Outlet      | 0.006                                     | 0.80  | 0.011 |          |
|               | Coastal sea | 0.004                                     | 1.39  | 0.007 |          |
| May-08        | Ash pond    | 0.051                                     | 9.69  | 0.01  |          |
|               | River       | 0.026                                     | 3.82  | 0.005 |          |
|               | Estuary     | 0.026                                     | 5.61  | 0.003 |          |
|               | Inlet       | 0.012                                     | 3.03  | 0.007 |          |
|               | Jetty       | 0.030                                     | 3.50  | 0.012 |          |
|               | Outlet      | 0.023                                     | 5.83  | 0.006 |          |
|               | Coastal sea | 0.019                                     | 5.85  | 0.004 |          |
| Aug-08        | Ash pond    | 0.026                                     | 4.22  | 0.01  |          |
|               | River       | 0.024                                     | 2.85  | 0.023 |          |
|               | Estuary     | 0.025                                     | 2.02  | 0.005 |          |
|               | Inlet       | 0.010                                     | 1.11  | 0.018 |          |
|               | Jetty       | 0.026                                     | 1.18  | 0.021 |          |
|               | Outlet      | 0.02                                      | 1.66  | 0.018 |          |
|               | Coastal sea | 0.014                                     | 2.10  | 0.004 |          |
| Nov-08        | Ash pond    | 0.038                                     | 3.76  | 0.014 |          |
|               | River       | 0.026                                     | 1.76  | 0.013 |          |
|               | Estuary     | 0.022                                     | 1.46  | 0.008 |          |
|               | Inlet       | 0.013                                     | 0.94  | 0.011 |          |
|               | Jetty       | 0.023                                     | 1.41  | 0.016 |          |
|               | Outlet      | 0.015                                     | 1.38  | 0.008 |          |
|               | Coastal sea | 0.011                                     | 1.57  | 0.006 |          |
| Feb-09        | Ash pond    | 0.026                                     | 3.6   | 0.013 |          |
|               | River       | 0.023                                     | 1.54  | 0.012 |          |

|             |       |      |       |
|-------------|-------|------|-------|
| Estuary     | 0.016 | 1.4  | 0.009 |
| Inlet       | 0.007 | 0.8  | 0.011 |
| Jetty       | 0.017 | 1.72 | 0.02  |
| Outlet      | 0.009 | 1.4  | 0.013 |
| Coastal sea | 0.014 | 1.54 | 0.009 |

## CONCLUSION

In this study, the spatial variation of naturally occurring radionuclide, Po-210, in the coastal area of Kapar has been examined. The dendrogram produced by hierarchical cluster analysis recognized two separate groups of study location at Kapar coast area based on the concentration level of Po-210 on water, TSS and sediment samples. The first group included the ash pond and the second group encompassed inlet, outlet, jetty, river, coast sea and estuary. At the same time, close similarities observed in between estuary, outlet and jetty of the study area. Based on the calculation of flux comparatively higher concentration of Po-210 has been recorded at the coastal area of Kapar, which might be the impact of coal burning. Among the seven stations studied in this study, ash pond is detected as the most vulnerable location which is because of the burnt coal dumping.

## ACKNOWLEDGMENTS

This research is supported by the research projects GGPM-2014-010 and TRGS/1/2015/UKM/02/5/2.

## REFERENCES

- [1] 1. Cherry, R. D.; Shannon, L. V., The Alpha Radioactivity of Marine Organisms. *Atomic Energy Review* 1974, 12, 3-45; (b) Stewart, G. M.; Fowler, S. W.; Fisher, N. S., The bioaccumulation of U- and Th-series radionuclides in marine organisms. In *Radioactivity in the Environment*, Baxter, M. S., Ed. Elsevier: 2008; Vol. 13.
- [2] 2. Germain, P.; Leclerc, G.; Simon, S., Transfer of polonium-210 into *Mytilus edulis* (L.) and *Fucus vesiculosus* (L.) from the baie de Seine (Channel coast of France). *Science of the Total Environment* 1995, 164 (2), 109-123.
- [3] 3. Kim, G.; Hong, Y.-L.; Jang, J.; Lee, I.; Hwang, D.-W.; Yang, H.-S., Evidence for anthropogenic  $^{210}\text{Po}$  in the urban atmosphere of Seoul, Korea. *Environmental Science & Technology* 2005, 39, 1519-1522.
- [4] 4. McDonald, P.; Baxter, M. S.; Scott, E. M., Technological enhancement of natural radionuclides in the marine environment. *Journal of Environmental Radioactivity* 1996, 32, 67-90.
- [5] 5. Othman, I.; Al-Masri, Impact of phosphate industry on the environment: a case study. *Applied Radiation and Isotopes* 2007, 65, 131-141.
- [6] 6. (a) Stewart, G. M.; Cochran, J. K.; Miquel, J. C.; Masque, P.; Szlosek, J.; y., R.; Baena, A. M.; Fowler, S. W.; Gasser, B.; Hirschberg, D. J., Comparing POC export from  $^{234}\text{Th}/^{238}\text{U}$  and  $^{210}\text{Po}/^{210}\text{Pb}$  disequilibrium with estimates from sediment traps in the northwest Mediterranean. *Deep Sea Research* 2007, 54 1549-1570; (b) Stewart, G. M.; Moran, S. B.; Lomas, M. W., Seasonal POC fluxes at BATS estimated from  $^{210}\text{Po}$  deficits. *Deep Sea Research* 2010, 57, 113-124.

- [7] 7. Verdeny, E.; Masque, P.; Garcia-Orellana, J.; Hanfland, C.; Cochran, J. K.; Stewart, G. M., POC export from ocean surface waters by means of  $^{234}\text{Th}/^{238}\text{U}$  and  $^{210}\text{Po}/^{210}\text{Pb}$  disequilibria: a review of the use of two radiotracer pairs. Deep Sea Research 2009, 56, 1502-1518.
- [8] 8. (a) Wood, A. K. H.; Ahmad, Z.; Shazili, N. A. M.; Yaakob, R.; Carpenter, R. O. Y., Geochemistry of sediments in Johor Strait between Malaysia and Singapore. Continental Shelf Research 1997, 17 (10), 1207-1228; (b) Theng, T.; Ahmad, Z.; Mohamed, C., Estimation of sedimentation rates using  $<sup>210</sup>\text{Pb}$  and  $<sup>210</sup>\text{Po}$  at the coastal water of Sabah, Malaysia. Journal of Radioanalytical and Nuclear Chemistry 2003, 256 (1), 115-120.
- [9] 9. Al-Masri, M. S.; Al-Bich, F., Polonium-210 distribution in Syrian phosphogypsum. Journal of Radioanalytical and Nuclear Chemistry 2002, 251 (3), 431-435.
- [10] 10. Hameed, P. S.; Shaheed, K.; Somasundaram, S. S. N.; Iyengar, M. A. R., Bioaccumulation of  $^{210}\text{Pb}$  in the Kaveri River ecosystem, India. Journal of Environmental Radioactivity 1997, 37 (1), 17-27.
- [11] 11. Kadko, D.; Bacon, M. P.; Hudson, A., Enhanced scavenging of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  by processes associated with the East Pacific Rise near  $8^\circ 45' \text{N}$ . Earth and planetary science letters 1987, 81 (4), 349-357.

★ ★ ★