

Seasonal Variation of ^{210}Po in Different Salinity: Case of Kuala Selangor River, West Coast Peninsular Malaysia

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Abstract: The relationship, input and output of ^{210}Po has been investigated in the dissolved ($<0.4\ \mu\text{m}$) and particulate ($>0.4\ \mu\text{m}$) samples collected from six stations in the Kuala Selangor River, where each station selected had a different salinity; ranging from the low salinity of surface layer water to the coastal oceanic water. The sampling discussed in this paper was undertaken on three dates; 22 May 2005 (during the dry season), 19 November 2005 (during the wet season) and 26 March 2006 (at the end of the wet season). Water samples were collected to measure the activities of ^{210}Po after spontaneous deposition on a silver disc; with their activity determined through the use of an alpha spectrometry analyzer. The mean activity of total ^{210}Po , which was found to alter according to the sampling dates, was noted as $57.83 \pm 12.26\ \text{dpm L}^{-1}$, $22.90 \pm 3.21\ \text{dpm L}^{-1}$ and $11.83 \pm 4.13\ \text{dpm L}^{-1}$ for the samples from 22 May 2005, 19 November 2005 and 26 March 2006, respectively. The highest concentration of activity of ^{210}Po was obtained during the dry season, whilst the lowest activity found was during the end of the wet season. In March 2006, at the end of the wet season, the behavior of ^{210}Po was found to behave in the manner of a conservative element. From the calculation model, approximately 82.14 % of the polonium input was found in the dissolve phase and around 144.94 % of the removed polonium remained in the particulate phase.

Keywords: radionuclide; polonium; coastal water; particulate; salinity

1. Introduction

Kuala Selangor is a fishing village located 67 km southeast of Kuala Lumpur. It has become a tourist location and is also an established habitat for fireflies which are associated with the red mangrove system along the river bank. The Kuala Selangor River is in fact one of the major river systems in the Selangor state, draining into the Malacca Straits. It is 500 m wide at the mouth with a water depth of about 2.5m during low tide. At high tide the water depth can reach up to about 10m. The coastal zone is characterized by a semi diurnal, macro-tidal regime with a mean spring tidal range of about 4.0m depth (Tee and Mohamed, 2005). The Kuala Selangor River also receives a lot of suspended particles from the land which increase the removal of reactive elements from the water column as is cited by Broecker and Peng (1982).

Uranium and thorium decay series serve as excellent tracers for understanding the nature and time scale of geochemical cycling. This process will involve the adsorption of reactive elements into particles from water, desorption of passive elements from suspended particles and sediment, and the mixing of water within the delta region. Natural radionuclides can be effective as tracers to elucidate the different processes controlling the distribution of elements among the dissolved and particulate phases in the aquatic systems.

In the natural ^{238}U decay series, polonium-210 (^{210}Po) is the final alpha-emitting daughter nuclide and is the most common of the 27 polonium isotopes in the natural environment. ^{210}Po enters the marine environment via the natural radioactive decay of ^{222}Rn , which is particularly particle reactive and largely associated with aerosols and decay of ^{226}Ra in solution through the wet and dry

atmospheric deposition of ^{210}Bi , ^{210}Pb and ^{210}Po (Turekian *et al.* 1977). Throughout the decay of radiation, ^{210}Po is received by humans through the consumption of food and contributes more to the total radiation dose than environmental levels of anthropogenic radionuclides, such as ^{137}Cs and $^{239,240}\text{Pu}$ (Pentreath and Allington 1988). Marine organisms are highly prone to accumulating ^{210}Po in their soft organ tissue and are popular with Malaysians as a seafood source. Carvalho (1995) calculated that 70% of the ^{210}Po dietary intake by the Portuguese population was from seafood.

^{210}Po in ^{238}U decay series is a non-conservative radionuclide product from the disintegration of ^{210}Pb . In the water column it is also produced by the in-situ disintegration of ^{226}Ra . In shallow water column, especially in river water, this contribution is almost negligible compared to the atmospheric flux, but it is greater in deep and open ocean water (Broecker and Peng, 1982). Both radionuclides of ^{210}Po and ^{210}Pb are intermediate members of the naturally occurring ^{238}U decay series and are recognized as tracers for natural processes in the atmosphere. ^{210}Po contains 27 isotope members from the ^{238}U decay series and is also the final alpha-emitting part of a daughter nuclide. The physical half life of ^{210}Po is 138.4 days, which means it can potentially be used as a tracer for biogeochemical processes such as: primary production, geochronology, environmental science and degradation of particles. It is also particle-reactive with varying affinities, both in terms of efficiency and the type of the matter to which it is associated.

The specific activity of ^{210}Po in seawater, suspended particulates and marine organisms from previous study is well-documented (i.e., Tee *et al.* 2004; Tee and Mohamed, 2005). However, to-date no study has quantified in-seasonal temporal variations of ^{210}Po from river to saline water. If

^{210}Po does behave in a similar manner to other trace metals, then it can be assumed that seasonal changes arising from phytoplankton blooms and physico-chemical parameters such as temperature and salinity and biological variables (for example, the reproductive cycle of zooplankton) are likely to affect the specific activity of ^{210}Po . Atmospheric fallout of ^{210}Po , and to a smaller degree in ^{210}Pb sources, itself precipitates on the surface of seas and oceans and is the basic way that ^{210}Po enters the marine environment. Fundamentally, the proportion of the dissolved forms of ^{210}Po in seawater is dependent on seasonal, as well as on chemical and biological factors (Rangarajan *et al.* 1976; Schell 1977; Spencer 1970). Many studies show that effluent discharges from rivers running into coastal seawaters may enhance and elevate the radioactivity in sediment, water and marine biota (Barisic *et al.* 1992; Carvalho 1995; Marovic and Sencar 1995; Martinez *et al.* 1994; McCartney *et al.* 2000).

A high scavenging rate of ^{210}Po in coastal waters can be attributed to riverine influences, the input of terrigenous material, resuspension and advection processes over the continental shelf (Thunell *et al.*, 1994). The stability of ^{210}Po in soluble forms is also dependent on the presence of organic matter such as humic matter, organic particulate and so on; complex forms with polonium (Bojanowski *et al.* 1981). The impact of fertilizers, such as phosphates, has also been reported to be a principle source of enhancement of the naturally occurring radionuclides in the environment (Othman and Al-Masri 2007).

Earlier studies have concluded that ^{210}Po behaves more like nutrient elements and several studies of oceanic ^{210}Po distribution have shown that scavenging from the water column is enhanced at the continental margins relative to the open ocean (Bacon *et al.* 1976). In line with such findings, the aim of this study is to investigate the seasonal input and removal rates of ^{210}Po with different salinity, focusing on the Kuala Selangor River; in particular the river to seawater region via the brackish estuary water area.

2. Sampling and method

Sampling was conducted on the Kuala Selangor River on 22 May 2005, 19 November 2005 and 26 March 2006 at six stations from the upper to the lower streams (Figure 1). About 10 litres of surface water samples and in-situ water quality parameters were collected using a Van-Dorn water sampler. These were measured with the calibrated portable probe model YSI-6000 series at the six stations from the upper to lower streams of the Kuala Selangor River, respectively. Water samples were stored in 10 L polyethylene containers which had been cleaned with 2 M of nitric acid. Water samples were briefly filtered in the laboratory through the pre-weighted Whatman® cellulose nitrate membrane with a pore size of 0.45 μm and 47mm in diameter. Then acidified in the dissolved phase with concentrated nitric acid to pH 2 whereby 0.5 ml of 25 mg L⁻¹ Fe³⁺ was added as a carrier solution and 0.2 ml of 20 dpm ml⁻¹ ^{209}Po as a chemical yield tracer.

The solution was then stirred until it became homogenized through the addition of 12.5g of a Na₂CO₃ and NH₄OH

solution, to adjust the pH to pH 9. After the solution had stood for 24 hours, the supernatant was siphoned out and the precipitate was transferred into a glass beaker to remove carbonate gases under a controlled temperature of 80°C. After two hours the precipitate was dissolved and re-precipitated with hydrochloric acid before being reheated on the hotplate. This procedure, referred to as the decarbonation process, ensured that all the gaseous carbonate particles were released entirely from the sample. Following the addition of an ammonium solution, precipitates were collected.

The particulate samples were digested by mixing nitric acid and perchloric acid (10:1) together, spiked with 0.5 ml of ^{209}Po (20 dpm/ml) as a chemical recovery. After digestion the sample was kept at room temperature, it was then filtered through pre-weighted Whatman® cellulose nitrate membrane with a pore size of 0.45 μm and 47mm in diameter so that the aqueous liquid could be collected. Following this, the aqueous samples were placed and kept on a hotplate until completely dry.

For the next stage, the precipitate from the dissolved samples and residue from the particulate samples were re-dissolved in 80 ml of 0.5 M HCl with small amounts of ascorbic acid to reduce the oxidation state of Fe³⁺ to Fe²⁺ (Nozaki & Tsunogai 1973). Spontaneous deposition polonium was placed on a silver disc (measuring 2cm x 2cm) for 2 hours at 80°C. The activities of ^{209}Po and ^{210}Po were determined using the Alpha Spectrometry (EG&G ORTEC) with errors based on the 1-sigma counting statistics. Quality control procedures were applied using the standard reference material IAEA-300 Radionuclides In Baltic Sea Sediment with efficiency $\geq 95\%$.

3. Results

Analytical Results

The results of our investigations into the dissolved and particulate phases for ^{210}Po , *in-situ* parameter with respect to the set sampling dates and six sampling stations are shown in Table 1. The average total activity of ^{210}Po (dissolved + particulate) indicated by 57.83 ± 12.26 dpm L⁻¹, 22.90 ± 3.21 dpm L⁻¹ and 11.83 ± 4.13 dpm L⁻¹ respectively, showed that the sequence went from high to low activity through the sampling dates of May 2005, November 2005 and March 2006. For all of the total dissolved and particulate phases, findings demonstrated that over 99% of the ^{210}Po in coastal water was found in the particulate phase, as supported by Tee (2005).

The activities of ^{210}Po in the dissolved phase in the Kuala Selangor River, estuary and coastal area ranged from $(8.79 - 82.95) \times 10^{-5}$ dpm L⁻¹, $(5.31 - 22.04) \times 10^{-5}$ dpm L⁻¹, $(1.08 - 11.33) \times 10^{-5}$ dpm L⁻¹, respectively, from sampling on May 2005, November 2005 and March 2006. These activities mirrored more or less the same profile along the river, although there were different levels of salinity on the sampling dates of May and November 2005 (not for March 2006) (Figure 3A, B, C). The concentration activity of ^{210}Po in the particulate phase ranged from 10.25 - 42.60 dpm g⁻¹, 3.46 - 24.78 dpm g⁻¹, 1.42 - 14.17 dpm g⁻¹ as obtained from the sampling in May 2005, November 2005 and March 2006,

respectively. This trend was similar to that of the dissolved phase, where all the profile trends crosses in station 4 could be accredited to the Kuala Selangor River estuary undergoing the same mixing rate circumstances and therefore maintaining small range activities during the whole of the sampling period (Figure 3).

4. Discussion

Distribution of ^{210}Po in study area

The increasing concentration levels of ^{210}Po found in the study area have related to the pH value from the inland river water where polonium is usually found to be soluble in most mineral acids and silica (Al-Masri *et al.* 2006). As a consequence, the urea contents of biotic organism or humic acid from the river can be considered to influence the end-member of oceanic ^{210}Po activities in Station 6 on the sampling dates in the year 2005 (Figures 3A & 3C). It also shows that the lowest pH values and salinity values, ranging between 15 psu < ^{210}Po < 20 psu, at the end-member river as being caused by the mixing process (Table 1).

The relative proportion of particulate ^{210}Po was especially high in May due to a lack of rainfall and hot weather in this season. The volume of water was more dependent on the evaporation process and thus the concentration of suspended solids increased to a greater volume. The concentration of radionuclides in May is considered to be influenced by the input and removal processes (dry deposition, wet deposition) of the atmosphere on the activity concentration. Additionally, it was found that the coagulation process significantly changes the activity of ^{210}Po (Grundel and Porstendorfer 2004).

Furthermore, each monsoon season brings different weather and atmospheric aerosol conditions. The classification size of aerosol also could affect the concentration of ^{210}Po . Figure 2 shows the scattered values of polonium from the different sampling dates and the correlation regression line obtained, which had almost the same increase for May and November 2005, with a lower one for 23 November 2006. Demonstrating that the enrichment concentration activity of ^{210}Po was the highest in May, followed by November 2005, and the lowest in March 2006 at the end of the wet season. The high enrichment level of polonium can be argued to be a result of a correspondingly high concentration of parents (i.e., radon and radium) in the study area which may have been transported by agriculture activities (e.g., phosphate fertilizer substance) into the river system (Othman and Al-Masri 2007). Conversely high activity of polonium in the particulate phase, especially in the low saline seawater, can be argued to be a consequence of the particle reactivity of polonium binding with lithogenous particle sources (Melieres *et al.* 2003). Additionally, the regression coefficient (R^2) value indicated a negative value (-0.7421) when compared with the other sampling date in the dry season. This negative value signifies a very weak binding force between the dissolved phase and particulate phase where the concentration activity was lower as it was the final part of the wet season in March, when the Kuala Selangor River was loaded with rainwater.

Input and removal rate

Figure 3 clarifies the relationship between the polonium and salinity values obtained during this study. This figure was also used to calculate the percentage input and removal of ^{210}Po along the Kuala Selangor River (Table 2). The dashed lines represent the theoretical mixing lines (TML) connected directly from the zero salinity value of end members to the coastal salinity values of end members. If the trend plotted, such as TML, is in a straight line, this indicates a lesser input or removal of ^{210}Po . This model applies during a steady state where there is no change in the river water and seawater contents flow into the mixing condition (Figure 3). The higher line of the TML (A_a) indicates the input of ^{210}Po from other sources, whilst the lower line (A_b) gives the removal of ^{210}Po occurring in the study area. Test results show that the trend for the activity of polonium against the salinity value crossing the TML line in Figure 3, representing the really conservative behaviour of ^{210}Po . It also indicates that both input and removal were occurring in the mixing zone. As a consequence, the interception of the y-axis with the dashed line can be used to estimate the input or removal rate of polonium using Li and Chan's formula (1979), as given below:

$$I = (A_{a,b} - K_r) S_r \quad \text{----- (1)}$$

Where I is the input or removal rate (dpm d^{-1}), $A_{a,b}$ define the activities of ^{210}Po (dpm L^{-1}) where the a,b indicates the y-axis intercept of the observed mixing curve profile, K_r represents ^{210}Po activity in the lowest salinity (dpm L^{-1}) and S_r is the river runoff volume per day as occurred in Kuala Selangor ($\text{m}^3 \text{d}^{-1}$). According to the Figure 3E, a lesser sloping of the TML line plotted along the river stations (i.e., Stations 4, 5 & 6) can be summarized as $I = (A_a - \text{TML}_{\text{intercept}}) S_r$ instead of equation (1). This is because the stations 4, 5 and 6 indicated the stable values of input and output along them and therefore the K_r value will (instead of by the $\text{TML}_{\text{intercept}}$ line) be indicated by the TML line intercepting the y-axis although it will cross station 3.

Figures 3A and 3C show May and November 2005 have an input (A_a) and removal (A_b) values of polonium in the dissolved phase. Only the particulate phase had a removal value (Figures 3B and 3D). It was found that the removal of ^{210}Po in the uppermost water surface of the dissolved phase was due to the volatilization across the sea-air interface (Peter *et al.* 1999). For both of the dissolved and particulate phases, there was a high level of activity of polonium from the end riverine member through to the coastal water samples, this fluctuated widely from the dashed theoretical mixing lines (TML) in all cases apart from the sampling undertaken in March 2006. This result suggests that ^{210}Po behaved in a conservative manner, especially in the particulate phase samples. However, such a result is not surprising given that ^{210}Po in the Kuala Selangor River is affected by the northeast monsoon which leads to the high flow rate of the river water and the constant activity value of polonium in the river stations (Figures 3E and 3F).

Table 2 shows the input and removal rate for the dissolved and particulate phases for the various sampling dates. The calculated results, given in percentages, were used to estimate the input rate and divided by the riverine end member (the lowest salinity of sampling station) along the

river to the coastal water region as given in the equation below:

$$\text{Input/removal (\%)} = \frac{\text{input, removal rate}}{\text{riverine end member}} \quad \text{----- (2)}$$

The results demonstrate that the total input or removal of polonium in all the dissolved and particulate samples underwent removal behaviour, except in the dissolved phase for May 2005 and March 2006, with an input of 116.65 % and 82.14 % respectively, depending on the riverine input rate. These figures are lower than from the Jiulong River in China where data represented the removal rate of 167% in the dissolved phase (Yang *et al.* 2003). The main input sources along the Kuala Selangor River can be observed from the Figure 3A, specifically from the low salinity area and the mouth of the estuary. Near the estuary the salinity was found to be between 15 psu and 20 psu where samples were also undergoing mixing conditions. It was therefore observed that the removal occurred in this area and increased again in the coastal water area. Likewise that a higher concentration may correspond to strong inversion layers occurring during the dry season, which in turn will lead to an enrichment of radioactive nuclides in the lower atmosphere and minimum values indicating a higher turbulence in the atmosphere (EL-Hussein *et al.* 2001). Research undertaken at the Tagus River in Portugal showed that ^{210}Po is transported into coastal areas and accumulated in estuarine mixing areas (Carvalho 1997). There is no doubt that the input of ^{210}Po also comes from rivers as is shown by the results of the samples taken from station 5 (Figure 3A).

Every year the Kuala Selangor River experiences a varying level of rainfall during the southwest and northeast monsoons. The southwest monsoon usually occurs in the later half of May or early June and ends in September, whilst the northeast monsoon usually commences in early November and ends in March. The seasonal cycle of freshwater discharge from the river dominates the surface distribution of water properties in the sea, especially during the rainy season when the river input is at its largest. Moreover, the haze problem has also been found to affect air quality on the western coast of the Malaysian Peninsular. This is as a result of air pollution from forest burning in Sumatera, Indonesia which has been occurring every year between August to October since the year 1997 (Koe *et al.* 2001). On top of the ^{210}Po formed by the decay of ^{210}Pb contained in the atmosphere, additional amounts are emitted directly from the Earth as a consequence of forest fires (Moore *et al.* 1974). It is therefore, unsurprising that the sampling from May and November 2005 indicated a high concentration of ^{210}Po (Table 1). Whilst the lowest activities of ^{210}Po obtained in March 2006 correlated to a lower level of polonium deposition from the atmosphere at the end of the wet season.

Rainwater also acts to remove the ^{210}Po from the river water all the way to the coastal waters, giving a similar constant level of concentration in the Malacca Strait. According to the TRMM Online Visualization and Analysis System (TOVAS and TRMM 2006), estimations of rainfall accumulated during sampling period for May 2005,

November 2005 and March 2006 sampling dates were 150–200 mm, 100–200 mm and 200–250 mm, respectively. These results prove that the difference between discharged river water to coastal can be classified by the rainy season in the Malaysian Peninsular. Another reason is that all the river water removed ^{210}Po accumulated at the mouth of the estuary of the Kuala Selangor River, which resulted in station 2 showing the highest value of ^{210}Po with an input of 82.14 % (Figure 3E).

Particulate phase

The calculated removal value of ^{210}Po in the particulate phase was 100%, 137.13% and 144.94% for the Kuala Selangor River on May 2005, November 2005 and March 2006, respectively. All the particulate phase of ^{210}Po in the Kuala Selangor River underwent removal after the rainfall season. This was particularly the case for the March 2006 sampling due to ^{210}Po being rapidly scavenged from the dissolved phase onto the particulate phase. Most of the ^{210}Po flowing through the estuary is exported to the coastal bottom sediments and suspended matter, rather than in the soluble dissolved phase. This case is similar to that of the Tagus estuary in Portugal where about 67% of the dissolved radionuclide inputs were trapped by particles and can increase the concentration activities of radioactivity in the sediment. Another 33% of the dissolved ^{210}Po in waste discharges is transferred to the sea in the soluble phase, after several cycles of the adsorption-reticulation process in the estuary (Carvalho 1997). This particulate matter in the river water is deposited and removed from the sediment, and association with small particles leads it be further transported to the coastal waters by the current. As a consequence, Figure 3 (B, D, F) shows that the concentration of ^{210}Po removed from the low salinity area to end oceanic member concentration. There is no doubt that acidic environments may enhance the adsorption and uptake of ^{210}Po by organisms. Additionally, the salinity reading for the coastal waters experienced rising pH values and ^{210}Po was rapidly removed from the high salinity area (Germain *et al.* 1995).

5. Conclusion

^{210}Po monitoring was carried out three times during both the wet and dry season on the Kuala Selangor River during November 2005, May 2005 and March 2006, respectively. Activities of ^{210}Po showed almost the same value for all the sampling dates which is approximately at the mean value of 9.73 dpm g⁻¹ and 4.33 dpm g⁻¹ respectively, with respect to the particulate and dissolved phases. The findings of this investigation indicated that ^{210}Po activities are non-conservative during the mixing of river with seawater for the sampling dates in May and November 2005, but that ^{210}Po tended towards conservative behaviour in March 2006 whilst experiencing the high rainfall of the wet season, both for the dissolved and particulate fraction. The highest input was found in the dissolved phase in May 2005 at 116.65% with removal at 144.94% in the particulate phase during the March 2006 sampling. The results also showed the continuous enrichment of ^{210}Po in the Kuala Selangor River as follows: May 2005 in the dry season, going to November 2005 in wet season and then March 2006 at the end of the wet season.

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References

- [1] Al-Masri, M. S., Al-Karfan, K., Khalili, H., and Hassan, M. (2006). Speciation of ^{210}Po and ^{210}Pb in air particulates determined by sequential extraction. *Journal of Environmental Radioactivity* **91**, 103-112.
- [2] Bacon, M. P., Spencer, D. W., and Brewer, P. G. (1976). $^{210}\text{Po}/^{226}\text{Ra}$ and ^{210}Po o/ ^{210}Pb disequilibria in seawater and suspended particulate matter. *Earth and Planetary Science Letters* **32**, 277-296.
- [3] Barisic, D., Lulic, S., and Milatic, P. (1992). Radium and uranium in phosphate fertilizers and their impact on the radioactivity of water. *Water Res.* **26**, 607-611.
- [4] Bojanowski, R., Pempkowiak, J., and Szefer, P. (1981). Occurrence of organically bound polonium-210 in Baltic and Vistula river waters. *Stud.at. Oceanol.* **34**, 93-112.
- [5] Broecker, W. S., and Peng, T. H. (1982). 'Tracers in the Sea.' (Eldigio Press: New York)
- [6] Carvalho, F. P. (1995). ^{210}Pb and ^{210}Po in sediments and suspended matter in Tagus estuaries Portugal, local enhancement of natural levels by wastes from phosphate ore processing industry. *Sci. Total Environ.* **159**, 201-214.
- [7] Carvalho, F. P. (1997). Distribution, cycling and mean residence time of ^{226}Ra , ^{210}Pb and ^{210}Po in the tagus estuary. *The Science of the Total Environment* **196**, 151-161.
- [8] EL-Hussein, A., Mohamemed, A., EL-Hady, M. A., Ahmed, A. A., Ali, A. E., and Barakat, A. (2001). Diurnal and seasonal variation of short-lived radon progeny concentration and atmosphere temporal variations of ^{210}Po and ^7Be in Egypt. *Atmospheric Environment* **35**, 4305-4313.
- [9] Flynn, W. W. (1968). The determination of low levels of polonium-210 in environmental materials. *Analytical Chimica Acta* **43**, 221-227.
- [10] Germain, P., G, L., and S, S. (1995). Transfer of polonium-210 into *Mytilus edulis* (L.) and *Fucus vesiculosus* (L.) from the baie de Seine (Channel coast of France). *The Science of the Total Environment* **164**, 109-123.
- [11] Grundel, M., and Porstendorfer, J. (2004). Differences between the activity size distributions of the different natural radionuclide aerosols in outdoor air. *Atmospheric Environment* **38**, 3723-3728.
- [12] Koe, L. C. C., Arellano, A. F., and McGregor, J. L. (2001). Investigating the haze transport from 1997 biomass burning in Southeast Asia: its impact upon Singapore. *Atmospheric Environment* **35**, 2723-2734.
- [13] Li, Y. H., and Chan, L. H. (1979). Desorption of Ba and Ra-226 from riverborn sediments in the Hudson estuary. *Earth and Planetary Science Letters* **43**, 343-350.
- [14] Marovic, G., and Sencar, J. (1995). ^{226}Ra and possible water contamination due to phosphate fertilizer production. *J. Radioanal. Nucl. Chem* **200**, 9-18.
- [15] Martinez, A. A., Garcia, L. M., and Ivanovich, M. (1994). The distribution of U, Th, and ^{226}Ra derived from the phosphate fertilizer industries on an estuarine system in south west Spain. *J. Environ. Radioact* **22**, 155-177.
- [16] McCartney, M., Davidson, C. M., Howe, S. E., and Keating, G. E. (2000). Temporal changes in the distribution of natural radionuclides along the Cumbrian coast following the reduction of discharges from a phosphoric acid production plant. *J. Environ. Radioact.* **49** (3), 279-291.
- [17] Melieres, M. A., Michel, P., and Sandrine, R. (2003). Surface air concentration and deposition of lead-210 in French Guiana: two years of continuous monitoring. *Journal of Environmental Radioactivity* **66**, 261-269.
- [18] Moore, H. E., Poet, S. E., Martell, E. A., and Wilkening, L. H. (1974). Origin of Rn-222 and its long-lived daughters in air over Hawaii. *Journal of Geophysical Research* **79**, 5019-5024.
- [19] Nozaki, Y., and Tsunogai, S. (1973). A simultaneous determination of lead-210 and polonium-210 in sea water. *Analytical Chimica Acta* **64**, 209-216.
- [20] Othman, I., and Al-Masri, M. S. (2007). Impact of phosphate industry on the environment: A case study. *Applied Radiation and Isotopes* **65**, 131-141.
- [21] Pentreath, R. J., and Allington, D. J. (1988) Dose to man from the consumption of marine seafoods: a comparison of the naturally occurring ^{210}Po with artificially produced radionuclides. In 'Proceedings of the 7th International Congress of IRPA'. Sydney pp. 1582-1585
- [22] Peter, W. S., Brent, A. M., Kai, S., and James, F. T. (1999). ^{210}Pb and ^{210}Po , manganese and iron cycling across the O₂/H₂S interface of a permanently anoxic fjord: Framvaren, Norway. *Marine Chemistry* **67**, 199-217.
- [23] Rangarajan, C., Gopalakrishnam, S., and Eapen, C. D. (1976) Global variation of ^{210}Pb in surface air and precipitation. In 'Health and Safety Laboratory Environment Quarterly, HASL-298'. (Ed. EP Hardy) pp. 163-182. (U.S.E.R.D.A.: New York)
- [24] Schell, W. R. (1977). Concentration, physico-chemical states and mean residence time of ^{210}Pb and ^{210}Po in marine and estuarine waters. *Geochim. cosmochim. Acta* **41**, 1019-1031.
- [25] Spencer, D. W. (1970). 'Distribution of lead-210 and polonium-210 between soluble and particulate phases in sea water.' (Woods Hole Oceanogr. Inst., USA EC: Massachusetts)
- [26] Tee, L. T., and Mohd, C. A. R. (2005). Activities of ^{210}Po and ^{210}Pb in the water column at Kuala Selangor, Malaysia. *Journal of Environmental Radioactivity* **80**, 273-286.
- [27] Tee, L. T., Zaharuddin, A., and Mohd, C. A. R. (2004). Activity concentration of ^{210}Po in the soft parts of cockle (*Anadara granosa*) at Kuala Selangor, Malaysia. *Journal of Radioanalytical and Nuclear Chemistry* **262**, 485-488.
- [28] Thunell, R. C., Pilskaln, C. H., Tappa, E., and Sautter, L. R. (1994). Temporal variability in sediment fluxes in

the San Pedro Basin, southern California bight. *Continental Shelf Research* **14**, 333-352.

[29] TOVAS, and TRMM (2006) <http://lake.nascom.nasa.gov/tovas/3B42RT/index>. In.

[30] Turekian, K. K., Nozaki, Y., and Benninger, L. K. (1977). Geochemistry of atmospheric radon and radon products. *Annual Review of Earth and Planetary Sciences* **5**, 227-255.

[31] Yang, W. F., Chen, M., Huang, Y. P., Qiu, Y. S., Zhang, L., Xing, N., and Li, Y. P. (2003). The input/removal rates of size-fractionated ^{210}Po in the Jiulongjiang estuary and its implications. *Chinese Science Bulletin* **48**, 2362-2365.

Figure Headings

Figure 1: Map showing the sampling stations at Kuala Selangor from the sea to river region.

Figure 2: A plot of dissolved versus particulate of ^{210}Po in the surface water of the Kuala Selangor River for the sampling dates: 22 May 2005, 19 November 2005 and 26 March 2006.

Figure 3: The activities of ^{210}Po in the dissolved and particulate phase versus salinity profile.

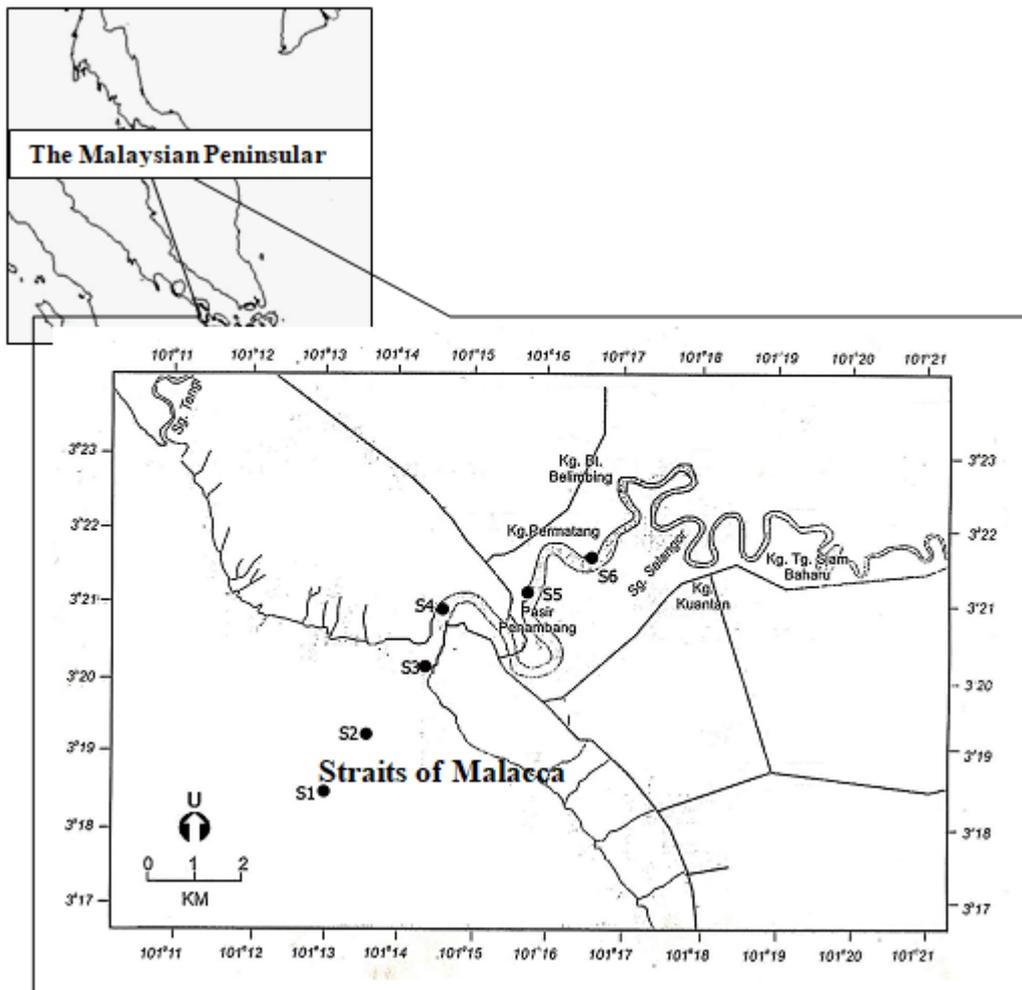


Figure 1: Map showing the sampling stations at Kuala Selangor from sea to river region.

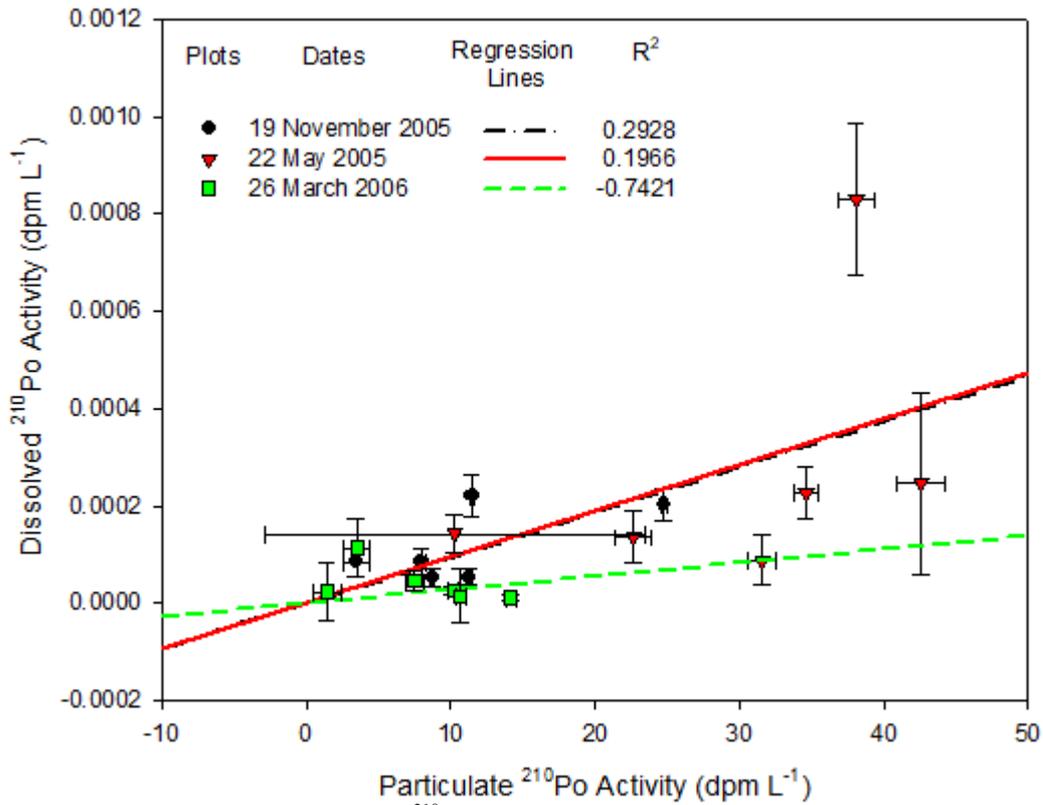
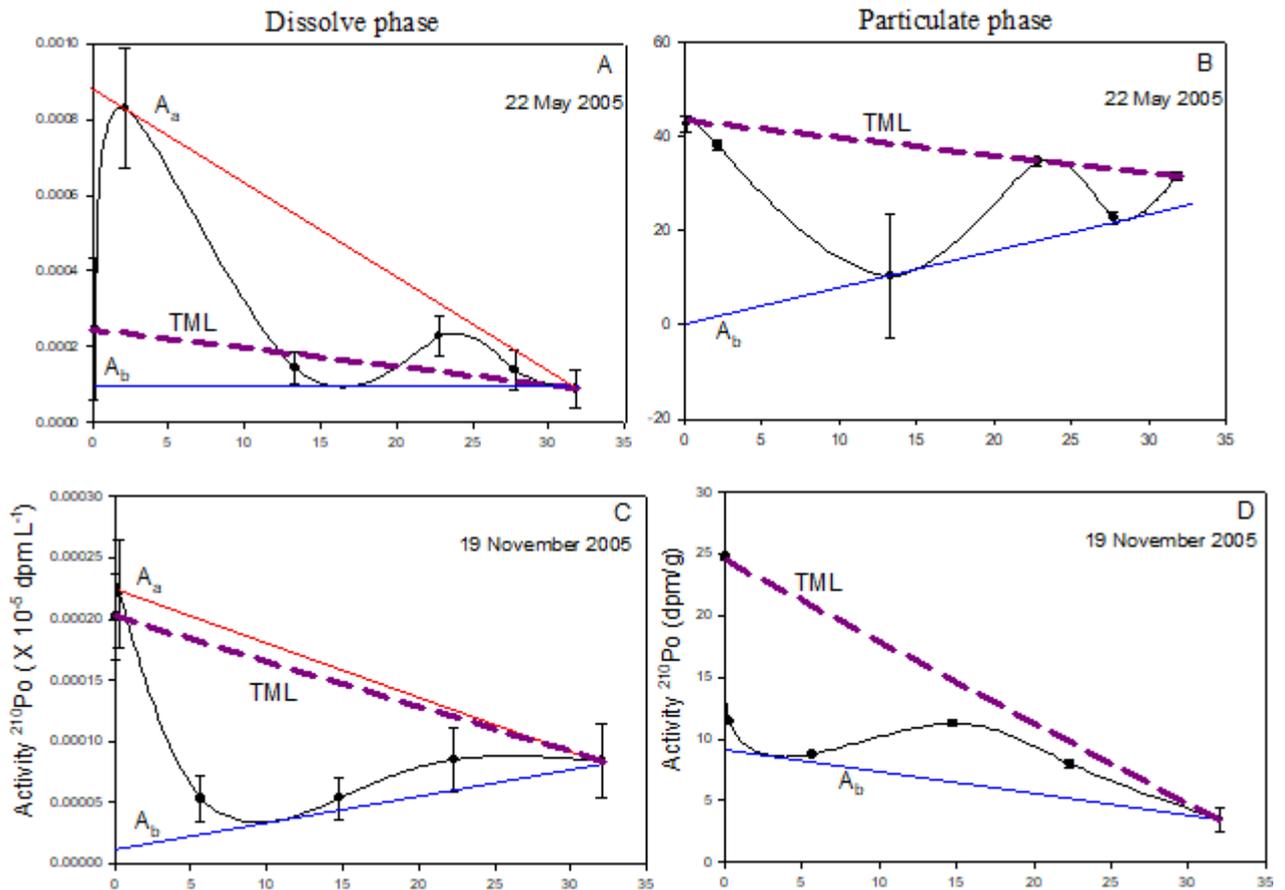


Figure 2: A plot of dissolved versus particulate of ²¹⁰Po in the surface water of the Kuala Selangor River for the sampling dates: 22 May 2005, 19 November 2005 and 26 March 2006



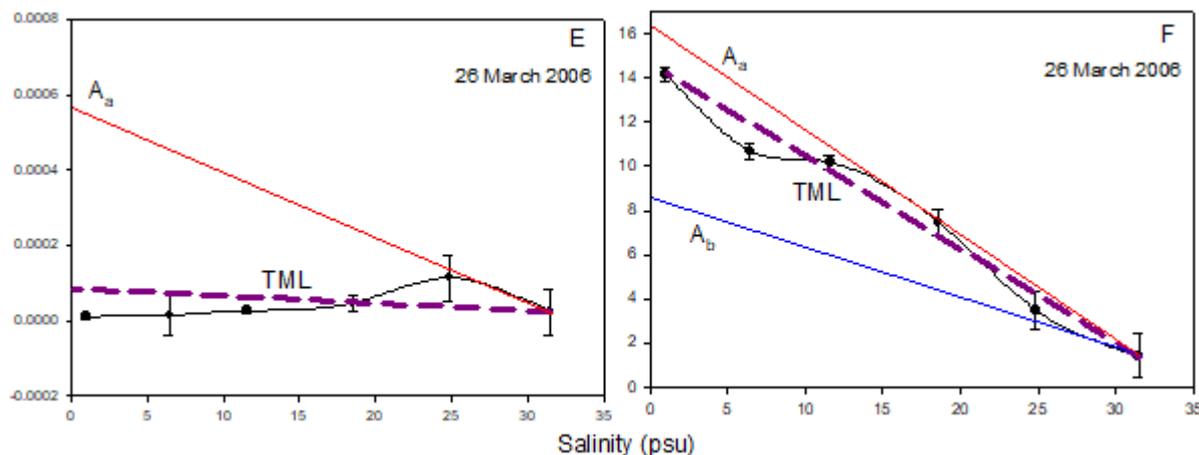


Figure 3: The activities of ²¹⁰Po in the dissolved and particulate phase versus salinity profile.

Table Headings

Table 1. List of dissolved, particulate phase of ²¹⁰Po and the *in-situ* parameter for each sampling site for the sampling dates in May 2005, November 2005 and March 2006.

Table 2. The input/removal rate and percentage of dissolved and particulate phase (SPM) ²¹⁰Po on surface water of the Kuala Selangor River during sampling in May 2005, November 2005 and March 2006.

Table 1: List of dissolved, particulate phase of ²¹⁰Po and the *in-situ* parameter for each sampling site during sampling in May 2005, November 2005 and March 2006.

	Sampling date	Station 1	Station 2	Station 3	Station 4	Station 5	Station 6
Coordinate location		03° 17' 28 N	03° 18' 31 N	03° 21' 10 N	03° 20' 04 N	03° 21' 09 N	03° 22' 02 N
		101° 12' 52 E	101° 13' 02 E	101° 14' 10 E	101° 15' 25 E	101° 15' 23 E	101° 15' 47 E
Dissolved ²¹⁰ Po (dpmL ⁻¹ X 10 ⁻⁵)	22-05-2005	8.79 ± 5.10	13.66 ± 5.23	22.82 ± 5.35	14.25 ± 4.07	82.95 ± 15.70	24.66 ± 18.64
	19-11-2005	8.36 ± 2.99	8.47 ± 2.64	5.37 ± 1.73	5.31 ± 1.91	22.04 ± 4.38	20.17 ± 3.44
	26-03-2006	2.35 ± 6.11	11.33 ± 6.09	4.66 ± 1.97	2.67 ± 0.87	1.45 ± 5.56	1.08 ± 0.72
Particulate ²¹⁰ Po (dpm g ⁻¹)	22-05-2005	31.59 ± 0.97	22.66 ± 1.23	34.64 ± 0.84	*10.25 ± 13.17	38.1 ± 1.19	42.6 ± 2.08
	19-11-2005	3.46 ± 0.95	7.98 ± 0.30	11.25 ± 0.24	8.74 ± 0.13	11.48 ± 0.13	24.78 ± 0.43
	26-03-2006	1.42 ± 0.98	3.49 ± 0.90	7.48 ± 0.59	10.19 ± 0.35	10.67 ± 0.33	14.17 ± 0.31
Temperature (OC #0)	22-05-2005	29.97	30.11	30.94	30.83	30.82	30.58
	19-11-2005	29.21	29.68	29.98	29.85	29.13	29.04
	26-03-2006	30.5	29.9	30.12	29.51	29.77	28.93
Salinity (psu)	22-05-2005	31.81	29.83	30.77	13.31	0.76	0.24
	19-11-2005	32.06	22.32	14.78	1.65	0.21	0.12
	26-03-2006	31.45	26.84	17.55	6.44	6.58	1.00
TDS (mg L ⁻¹)	22-05-2005	31.8	30.04	30.91	14.48	0.99	0.34
	19-11-2005	32.01	23.11	15.83	1.98	0.24	0.16
	26-03-2006	31.5	27.31	18.62	7.41	7.56	1.3
DO (%)	22-05-2005	79.7	78.2	92	64.2	71.5	63.1
	19-11-2005	106.6	95.8	78.4	75.3	86.9	82.4
	26-03-2006	113.5	96.4	93.9	78.3	77.5	82.1
pH	22-05-2005	7.99	7.77	7.76	7.13	6.4	6.23
	19-11-2005	8.09	7.44	7.14	6.33	3.01	3.4
	26-03-2006	7.88	7.84	7.55	7.24	6.36	6.16
Turbidity (NTU)	22-05-2005	124.4	71.2	197.9	74.4	323.3	354.1
	19-11-2005	8.8	51.3	63.1	101.5	121	49.7
	26-03-2006	4.3	11.4	55.8	61.7	43.8	147.3

* A high standard deviation caused by the high background of the instruments used

Table 2: The input/removal rate and percentage of dissolved and particulate phase (SPM) ²¹⁰Po on surface water of the Kuala Selangor River during sampling in May 2005, November 2005 and March 2006

Date	Phase	Riverine end member (lowest salinity, K _r) X S _r	Intercept y-axis		Input/removal X S _r		Input/removal (%)		
			A _a (input)	A _b (removal)	I _a (input)	I _b (removal)	input	removal	total
22-May-05	Dissolved	2.47 X 10 ⁻⁴	8.81 X 10 ⁻⁴	-0.0001	6.34 X 10 ⁻⁴	-3.47 X 10 ⁻⁴	257.20	-140.54	116.65
	Particulate	42.60	-	0	-	-42.60	-	-100.00	-100.00
19-Nov-05	Dissolved	2.02 X 10 ⁻⁴	2.30 X 10 ⁻⁴	-0.00001	2.85 X 10 ⁻⁵	-2.12 X 10 ⁻⁴	14.03	-104.96	-90.92
	Particulate	24.78	-	-9.2	-	-33.98	-	-137.13	-137.13
26-Mar-06	Dissolved	1.00 X 10 ⁻⁴	5.60 X 10 ⁻⁴	-	4.60 X 10 ⁻⁴	-	82.14	-	82.14
	Particulate	14.17	16.4	-8.6	2.23	-22.8	15.76	-160.70	-144.94

- * The negative value indicates the removal of ^{210}Po mean
- * Dissolved phase (dpm L^{-1}), particulate phase (dpm g^{-1})
- * S_r = river runoff volume per day (constant value)
- * - = non occurred