e-Jurnal Sains Nuklear Malaysia, 2023, Vol. 35 (No.2): 9 – 24
eISSN: 2232-0946

DETERMINATION OF ARSENIC TEMPORAL ENRICHMENT IN JURU RIVER'S CORES SEDIMENT USING NUCLEAR ANALYTICAL TECHNIQUES

Yii Mei-Wo

Malaysian Nuclear Agency,
43000 Kajang, MALAYSIA
Correspondence author: yii@nm.gov.my

ABSTRACT

Perai Industrial Area is one of the densest industrial and heavily populated zones in Malaysia. Land pollutants from human activities potentially be released into nearby water resources. Of concern arsenic concentration levels in sediments were investigated in the nearby Juru River. Using KC™ Kajak corer, one sediment core was collected at the river estuary and another one within the river, about 2-3 km away from the estuary. Arsenic concentrations were measured using Neutron Activation Analysis (NAA) technique; whereas the sediment deposition rates were determined through Lead-210 dating, performed using a Gamma-ray Spectrometry System. The enrichment factor for arsenic in the river was calculated using continental crust values and iron concentration as the normalisation metal. The results (9.7 – 25.9 (average 15.7) µg/g, within river; 10.2 – 24.0 (average 14.5) µg/g, estuary) indicated significant enrichment of arsenic in the study area, and the sedimentation rates (0.94±0.11 cm/y, within river; 2.70±0.68 cm/y, estuary) suggested rapid deposition of contaminants.

Keywords: arsenic, Juru river, Neutron Activation Analysis (NAA), enrichment

INTRODUCTION

Nearly 40% of the world's population lives within 100 km of the coast. The exploitation of coastal natural resources during development and the use of coastal areas as receptacles for societal waste have promoted sediment contamination and endangered marine resources (Burke et al., 2001; Cochran & Masque, 2004; United Nation, 2017). Erosion process at river and coastal bring soils and terrestrial chemical and biological wastes into the aquatic environment and combined together as sediment. While weathering is a major source of sediments, rapid economic growth and settlement development can lead to the release of domestic waste materials into coastal areas, resulting in their accumulation in sediments (Joseph et al., 2018). The Perai Industrial Estate is one of the major industrial parks in Penang and Malaysia. Due to heavy industrial development and population growth in the vicinity of the Perai industrial area, it is suspected that industrial and household discharges are being released into the river. The nearby Juru River has been reported as one of the most polluted rivers in the country (Zainudin, 2010).

Land pollutants from human activities transported to rivers and end up in marine ecosystem continuously. Time deposition of such pollutants can be chronologised by using natural radionuclides such as ^{210}Pb. Pb-210 ($T_{1/2}$ = 22.20±0.22 years), a member of the ^{238}U decay series, is present in the sediments through two main routes. Firstly, there is a continuous *in-situ* production of ^{210}Pb from

^{226}Ra ($T_{1/2}$ = 1600±7 years), and this fraction, known as background or supported ^{210}Pb, is assumed to be in radioactive secular equilibrium with ^{226}Ra. On the other hand, ^{222}Rn ($T_{1/2}$ = 3.8235±0.0003 days) which is a noble gas, emanates from the Earth's surface and goes into the atmosphere where it decays into ^{210}Pb, which is removed from the atmosphere back to the Earth's surface (by wet precipitation and/or dry fallout) contributing to the unsupported or excess fraction of ^{210}Pb (Krishnaswami & Lal, 1978). This atmospheric addition of ^{210}Pb is over of the amount permanently supplied by the *in-situ* decay of ^{226}Ra. As time goes by, the activity of excess ^{210}Pb will decay until reaching equilibrium with the supported ^{210}Pb fraction (^{226}Ra). The activity differences between the initial excess ^{210}Pb (at the surface of the sediment core) and the subjacent core sections can be used to estimate the time when the sediment at this section was deposited (Chakrabarty et al., 2006; Likuku, 2006; Aliev et al., 2007; Sanchez-Cabeza & Ruiz-Fernández, 2012; Wan-Mahmood et al., 2016). Historical deposition of anthropogenic radionuclide such as ^{137}Cs ($T_{1/2}$ = 30.05±0.08 years) can be used to complement the deposition profile.

Inorganic pollutants such as arsenic, that are released into river water can be accumulated in the aquatic food chain and sediments (Islam et al., 2017; Li et al., 2017; Yunus et al., 2020), and promote adverse effects on aquatic life (Yi et al., 2011; Elias et al., 2012), and also to humans through fish consumption. The major sources of inorganic anthropogenic pollution are from agriculture activities, animal waste, domestic, industrial, mining and petroleum activities, as well as industrial emissions (Shazili et al., 2006; Zhu et al., 2011; Pandey & Singh, 2017). Arsenic is an odourless, tasteless but toxic element that can cause numerous human health effects. Inorganic arsenic is a known carcinogen that can cause various cancers in the skin, lungs, liver and bladder (Yunus et al., 2020). Exposure to high amounts of arsenic can also bring fatality.

This study is aimed to assess the sediment deposition rates by radionuclides measurement, as well as the concentration and enrichment of arsenic in two sediment cores collected from Juru River, Penang, that might have been promoted by the economic development and population growth in the Perai Industrial area.

MATERIALS AND METHODS

Sample collection and preparation

Sediment cores were collected at Juru River during the dry season in 2017, one within the estuary zone and another 2 - 3 km upriver, in river (Fig. 1) to avoid massive mud transportation into the river sediment by the heavy downpour during the rainy season and the water turbulence that disturbs the sediment surface. The cores were collected using a KC™ Kajak Core Sampler (Fig. 2) with an 8 cm diameter PVC tube. Pre-preparation, handling and treatment of samples were according to the same procedure as reported earlier by Yii et al. (2020a).

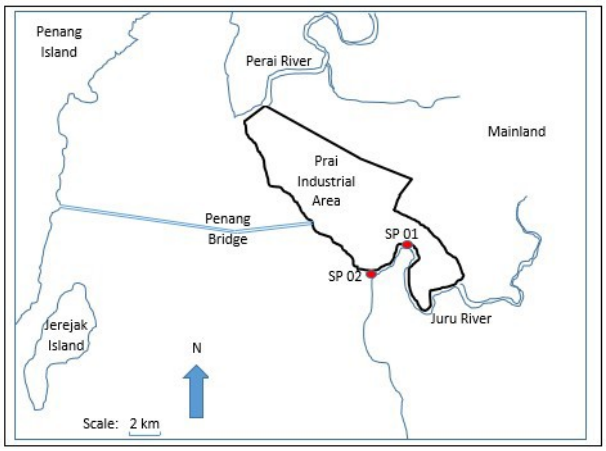


Figure 1. Sampling location

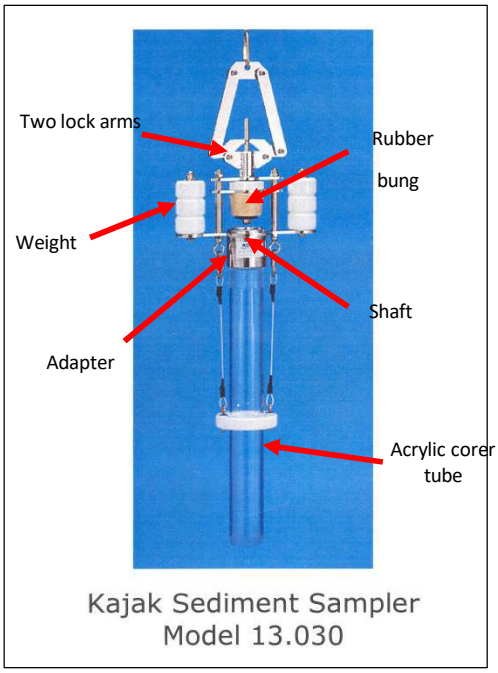


Figure 2. Sampling corer

Table 1. Sampling location details

Date	Station	Coordinate	Core length (cm)
Feb 2017	SP 01, Juru River	5.3464 °N 100.4189 °E	74
Feb 2017	SP 02, Juru River	5.3395 °N 100.4048 °E	78

Radionuclides analysis

Pb-210, ^{226}Ra and ^{137}Cs activities were determined by gamma-ray spectrometry (Fig. 3) consisting of a high-purity germanium (HPGe) setup and a multichannel analyser (16,384 channels). This 25% relative efficiency p-type closed end coaxial well-detector from CANBERRA™ operates at 2,000 HV bias supply (detector: 62 mm diameter, 49 mm, and 5 mm distance from a window; well: 35.5 mm depth, 23.5 mm diameter, active volume 8.8 cc), is designed to provide FWHM resolution of 1.85 keV at 1332 keV gamma-ray line of ^{60}Co and 820 eV at 122 keV gamma-ray line of ^{57}Co. The detector is shielded in an 11 cm thick chamber made of lead, cadmium and copper to reduce cosmic radiation. It was calibrated with the same procedures as reported earlier by Yii et al. (2009) using a customised gamma multi-nuclide standard source (source no. 1290-84 and 1755-30 from Isotope Products Laboratories, USA) comprised of ^{210}Pb, ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co in the same counting geometry. Fine ground dry sediments from each section of the core were sealed in a 6 mL cylindrical HDPE container with thick PVC tape to inhibit radon from escaping

(Yii et al., 2016). The net weight of each sample was recorded and divided by the sample volume to obtain sample density. All samples were stored for at least 30 days to establish secular equilibrium between ^{226}Ra and their respective progenies before gamma counting (Dowdall & O'dea, 2002; Yang et al., 2005).

Samples were counted for 15 hours and corrected for density and sampling date. Background counts were determined by counting similar geometry containers filled with inert material over the weekend. The counting times were long enough to ensure a 2π counting error of less than 10% (Ahmed & El-Arabi, 2005; Arogunjo et al., 2005; El-Reefy et al., 2006). The ^{226}Ra were measured through the gamma transitions of its progenies; ^{214}Pb (295.21 and 351.92 keV) and ^{214}Bi (609.31 keV, 1120.29 keV and 1764.49 keV). Meanwhile, ^{210}Pb was measured directly *via* energy 46.5 keV peak and ^{137}Cs was measured directly *via* energy 661.62 keV peak (Mishra & Sadasivan, 1971; Yang et al., 2005; El-Reefy et al., 2006). The sample activities were calculated using the equation reported by Yang et al. (2005) and Chen et al. (2005). The minimum detectable activity (MDA) for the radionuclides of interest was quantified as follows: ^{226}Ra (1 Bq/kg), ^{210}Pb (5 Bq/kg) and ^{137}Cs (1 Bq/kg). Reference materials IAEA-Soil-6 (^{226}Ra, 69.56 – 93.43 Bq/kg; ^{137}Cs, 51.43 – 57.91 Bq/kg at 30th January 1983) and IAEA-412 (^{210}Pb, 85.0 – 91.4 Bq/kg at 1st January 2019) were used for quality control evaluation. The results of samples were accepted if values obtained for the reference material fell within these 95% confidence interval as mentioned in the certificate.

Figure 3. Radioactivity measurement (from left to right): samples, well detector, gamma spectrometry system.

Trace elements analysis

Trace elements present in sediment samples were analysed using the NAA method (Fig. 4). Neutron Activation Analysis (NAA) is a non-destructive analytical technique where digestion or extractions, are not required, and therefore the integrity of the sample is not changed by the addition of any foreign materials for irradiation. Thus, the problem of reagent introduced contamination is completely avoided (Elias et al., 2014). The physical phenomenon upon which NAA is based in the properties of the nucleus, radioactivity and the interaction of radiation with matter. The neutron source from the reactor interacts with the target nucleus (samples – soil, sediment, or plant) by non-elastic collision; a compound nucleus is formed in a highly excited state, and a target nucleus (new nucleus) becomes radioactive and will further get de-excited by emitting decay gamma rays (IAEA 1990). The intensities of the gamma rays which can be quantified using the Gamma Spectrometry system are proportional to their concentrations.

For the NAA analysis, approximately 0.1 g powdered duplicate sediment aliquots were stored separately in heat-sealed polyethylene vials. Reference material IAEA-Soil-7 (Arsenic ranged between 12.5 – 14.2 µg/g at 95% confidence interval) was used as quality control material. All

samples were irradiated (up to 6 hours at a rotating rack) with a thermal neutron flux of 4.0×10^{12} n cm^{-2} s^{-1} obtained from the TRIGA Mark II research reactor located at the Malaysian Nuclear Agency which operated at 750 kW using pneumatic transport facility. Irradiated samples were left aside to allow the decay process before gamma-ray spectrometry counting, the cooling duration varied from 2 to 4 days before performing the first counting, and 3 to 4 weeks for the second counting. The radioisotope of the elements counted and their characteristics are shown in Table 2 (Ashraf et al., 2018; Elias et al., 2018).

Table 2. The elements and radionuclides measured using Neutron Activation Analysis (NAA)

Elements	Radionuclides	Half-life	γ-ray Energy (keV)
As	^{76}As	26.4 hours	559
Fe	^{59}Fe	45.1days	1099,1292

The gamma-ray detector used for the NAA analysis was a closed end coaxial 3" x 3" HPGe detector connected to a multichannel analyser that was previously calibrated from low to high energies by using a mixed standard solution including ^{241}Am (59.5 keV), ^{109}Cd (88.1 keV), ^{57}Co (122.1, 136.5 keV), ^{133}Ba (81.0, 303.0, 356.0, 384.0 keV), ^{137}Cs (661.7 keV), ^{60}Co (1173.2, 1332.5 keV) and ^{88}Y (898.0, 1836.1 keV) (Saion et al. 2007; Alnour et al. 2014). The gamma-ray spectrometry counting process of the irradiated samples, mixed standard solution and reference material was performed for one hour each. The results of the samples were accepted when the values obtained for the reference material fell within the 95% confidence interval as mentioned in the certificate.

Figure 4. NAA metal analysis (from left to right): Samples, neutron irradiation, gamma counting

Enrichment factor (EF)

The enrichment factor (EF) was calculated based on Equation (1) to evaluate the potential contamination caused by trace elements (Elias et al., 2018):

$$EF = \frac{(M/R)_{measure}}{(M/R)_{CC}} \qquad (1)$$

where $(M/R)_{measure}$ is the concentration ratio of the element of interest (M) to the reference element (R) measured in the sample, and $(M/R)_{CC}$ is the concentration ratio in the continental crust (Wedepohl, 1995).

Iron (Fe) was used as a reference element to normalise the trace element concentrations to compensate for variations in particle size and mineralogy (Loring & Rantala, 1992). Iron was selected as a reference element for normalisation in the EF calculation as no significant correlations were observed between Fe and the trace element concentrations found. Deely & Fergusson (1994) reported that this is due to iron distribution not being related to other elements. The natural concentration of iron is

usually relatively high, and therefore the estuarine sediment not expected to be substantially enriched by the anthropogenic source (Niencheski et al., 1994; Abrahim & Parker, 2008). The EF value will be less than 2.0 if the element found in the sediment originated predominantly from the lithogenous materials, whereas EFs are much greater than 2.0 indicating that the element is of anthropogenic origin (Szefer et al., 1996). EF values were classified according to Abdullah et al. (2020) as shown in Table 3.

Table 3. The indication status of enrichment factor (Abdullah et al., 2020)

Enrichment factor (EF) value	**Enrichment status**
< 2	Depletion to minimal enrichment
between 2 to 5	Moderate enrichment
between 5 to 20	Significant enrichment
between 20 to 40	Very high enrichment
more than 40	Extremely high enrichment

RESULTS AND DISCUSSION

Radionuclide activities

The activity concentrations for ^{137}Cs, ^{210}Pb and ^{226}Ra in each core are illustrated in Fig. 5 and summarised in Table 4. The ^{137}Cs activities are below the detection limit of 1 Bq/kg for all sediment cores. Meanwhile, ^{226}Ra activity ranges (Bq/kg) are highly variable among cores, with the highest values observed in the core collected upriver (Table 4). These values are higher than those found in sediment cores from previous studies in Asia (e.g. east coastal area of Peninsular Malaysia, 46.2 – 121.5 Bq/kg (Wan-Mahmood et al., 2005) and Vietnam coast, 23.1 – 40.2 Bq/kg (Nguyen et al., 2009); the Economic Exclusive Zone of the east coast of Peninsular Malaysia, 16 – 46 Bq/kg (Wan-Mahmood & Yii, 2012), or elsewhere, e.g. Gulf of Mexico, 11.8 – 97.3 Bq/kg (James et al., 1998); Santos - Cubatão drainage basin, 28.2 – 80.0 Bq/kg (Sanders et al., 2010); Estuary of Coatzacoalcos River, 15.57±1.4 Bq/kg (Ruiz-Fernandez et al., 2012)), but much lower than values reported for Krka River estuary, 45 – 662 Bq/kg (Cuculic et al., 2006)). A single factor ANOVA analysis revealed that ^{226}Ra activities are significantly different among cores: where SP 01 (97.0±32.8 Bq/kg) > SP 02 (53.9±9.9 Bq/kg).

Cores SP 01 (the upriver core) shows high variability of ^{226}Ra activities as shown in Fig. 5. This variability could potentially be attributed to the influx of radioactivity from the mainland, which is likely a result of human activities, such as the use of phosphate fertiliser in agriculture. Transported radionuclides eluted from soil and contained in dry and wet atmospheric precipitation (^{210}Pb) are deposited together onto sediment (Li et al., 1977).

Activities of ^{210}Pb are also found higher than those previously reported in the region such as in Kuala Muda area (north west Peninsular Malaysia), 11.9 – 78.8 Bq/kg (Yii et al., 2010), Linggi River Estuary (south west Peninsular Malaysia), 37.9 – 176.2 Bq/kg (Wan-Mahmood et al., 2016), and Brunei Bay, 2.6 – 32.3 Bq/kg (Joseph et al., 2018). Again, the single factor ANOVA analysis revealed that there are significant differences between cores for the ^{210}Pb activities where SP01 (154.7±51.5 Bq/kg) > SP02 (100.2±20.0 Bq/kg).

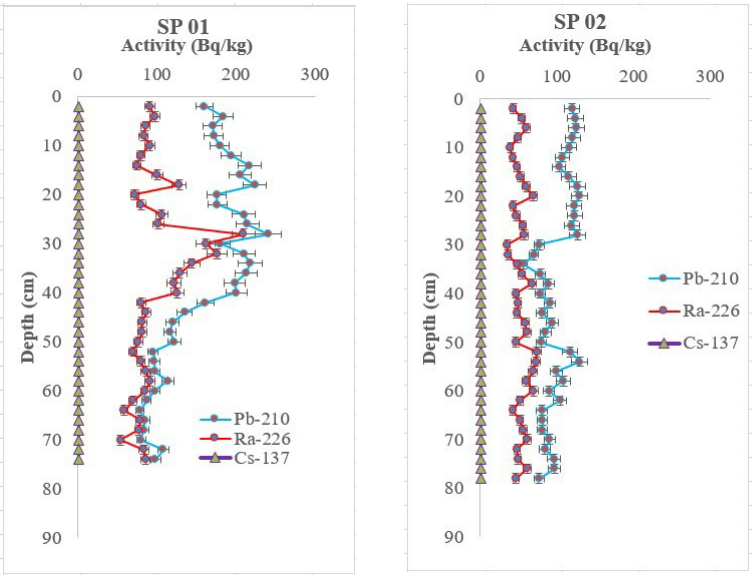


Figure 5. Activity concentrations of ^{137}Cs, ^{210}Pb and ^{226}Ra in the sediment core at Juru River

Figure 5 above clearly shows that the activities of ^{210}Pb are mostly found higher than ^{226}Ra along both cores, and the equilibrium between both radionuclides is only achieved at cores SP01(after 50 cm). The excess ^{210}Pb ($^{210}Pb_{ex}$) activities were estimated from the difference between ^{210}Pb and ^{226}Ra activities (Table 4). It has been shown that ^{210}Pb may be associated with biogenic particles and high ^{210}Pb activities may be derived from the lithogenic inclusion (Bralower & Thierstein, 1987). Lead-210 is highly particle reactive and is readily scavenged by organic matter and clay size particles (Ontiveros-Cuadras et al., 2012), although under anoxic conditions, ^{210}Pb can be released back to the water column (Benoit, 1988).

Table 4. Radioactivity concentrations of ^{226}Ra, ^{210}Pb, their ratios and sedimentation rate at Juru River

Sampling Station	**Activity concentration (Bq/kg dw.)**			**^{210}Pb excess (Bq/kg dw.)**	**Sedimentation accumulation rate (cm/y)**
	^{137}Cs	**^{226}Ra**	**^{210}Pb**		
SP 01	< MDA	54.0 – 209.9	78.8 – 241.0	5.9 – 142.0	0.94±0.11
SP 02	< MDA	36.0 – 74.3	56.5 – 129.9	8.4 – 79.3	2.70±0.68

The logarithmic values of $^{210}Pb_{ex}$ activity depth profiles in both cores show significant departures from linearity (Fig. 6), characteristic of a profile resulting only from radioactive decay, in aquatic environments where a steady state sedimentation process occurs. In most cases, such divergences could be the result of strong variations in the sediment accumulation process, sediment sources and mixing (UNESCO, 1978). As described in Yii et al. (in press), $^{210}Pb_{ex}$ activities in the cores of this study are strongly influenced by the variations in grain size, and organic matter content.

Despite all the complex features observed in the $^{210}Pb_{ex}$ activity depth profiles, both of them showed significant ($p<0.05$) decreasing trends with depth, with high slope values, which would account for high accumulation rates (Alongi et al., 2005); although, owing to the non-monotic $^{210}Pb_{ex}$ depth profiles, it is very difficult to derive an age model from them. However, attempts are made to approximate a preliminary mean sedimentation rate for each core, by using the constant flux-constant

sedimentation (CF:CS) model (Krishnaswamy et al., 1971), which assumes a constant $^{210}Pb_{ex}$ flux to the sediment surface and a sediment accumulation rate (Sanchez-Cabeza & Ruiz-Fernandez, 2012). Thus, the logarithm of excess ^{210}Pb (ln $^{210}Pb_{ex}$) data obtained in this study was plotted against the depth of the sediment core; and the slope of the regression between both variables was used to estimate the mean sediment accumulation rate (SAR, in cm/y). The mean SAR values obtained (Table 4) ranged from 0.94±0.11 cm/y in SP01 to 2.70±0.68 cm/y in SP02. The highest value recorded at station SP 02 corresponds to the estuarine area of Juru River, where reportedly affected by heavy loads of waste water discharges (Zainudin, 2010).

The mean sedimentation rate interval found in this study of 0.94 – 2.70 cm/y is comparable to those reported by Joseph et al. (2018) for Brunei Bay where the sedimentation rates reported range between 0.47 – 2.13 cm/y and higher sedimentation was attributed to rapid urban and industrial development. The study results are also comparable to the sedimentation rates found at the Linggi River estuary between 0.70 – 1.97 cm/y (Wan-Mahmood & Yii, 2013). However, the sedimentation rate found in this study is much higher than in some other areas as reported by Cheevaporn et al. (1994) at Bang Pakong River, Thailand (0.47 – 0.72 cm/y), and by Xu et al. (2008) at Nile River Delta, Egypt (0.42 – 0.56 cm/y).

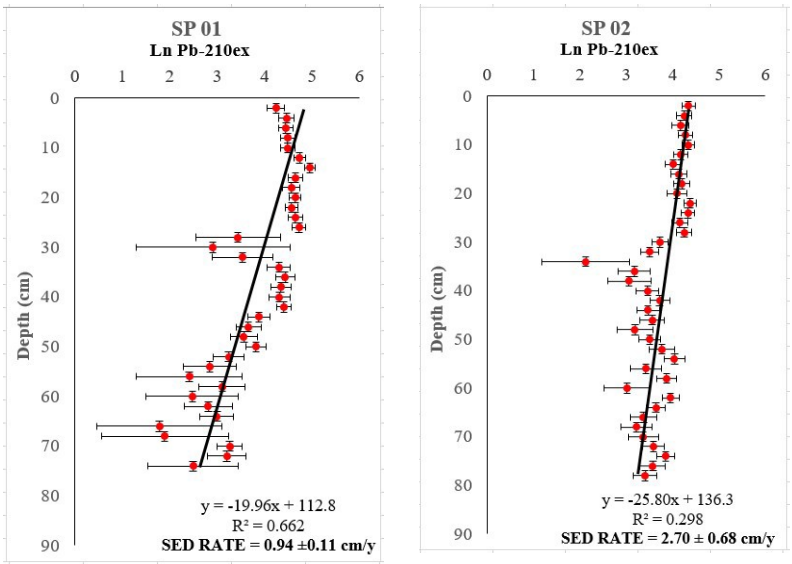


Figure 6. Sedimentation rates in core SP01 and SP02

Trace elemental concentrations

Table 5 below summarises the concentration of arsenic and iron found in the study area. Arsenic (As) concentrations in both cores are less than 30 µg/g with a mean of around 15 µg/g. Overall, the concentration ranges for arsenic in this study are comparable to most of the studies reported by other researchers in the same area/region. For instance, studies at Juru River conducted earlier by Wood et al. (1993) reported a range from 0.90 – 12.3 µg/g (As), while Abdullah et al. (2020) reported a range between 1.07 – 12.16 µg/g (As) for surface sediment of Bukit Merah Reservoir, which are half of the current study. A review made by Yunus et al. (2020) reported that most researchers found arsenic concentration level in the sediments at several locations in Peninsular Malaysia ranged between 6.13 – 42.30 µg/g, with an exceptional case where a study in Port Klang's sediment found the arsenic concentration reaching 475.26 µg/g. The study by Yii et al. (2020b) regarding the metal concentration in surface sediment collected along the Juru River indicated that contaminants were released from a

diffuse source upstream at the sampling site.

Table 5. Concentrations and enrichment factor (EF) ranges of arsenic and iron (µg/g) in sediment cores from Juru river

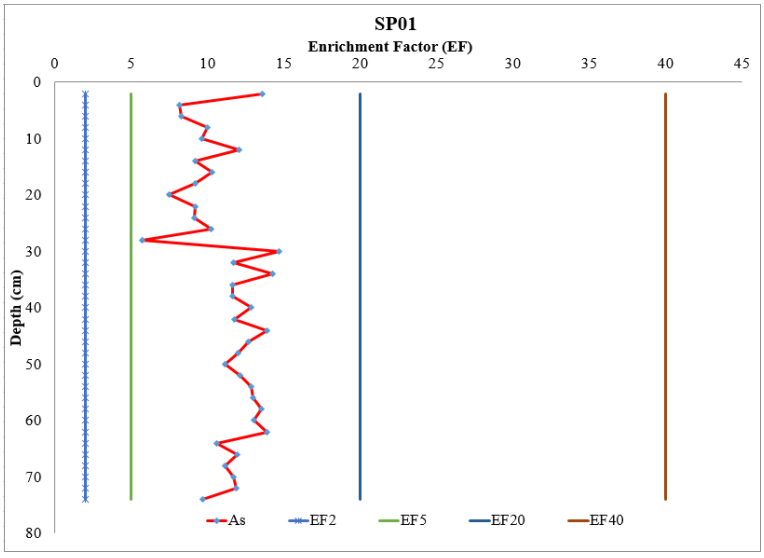
Element	CC[1]	Location	Concentration Range[2]	EF range
As	**1.7**	**SP 01**	9.7 – 25.9 (15.7)	5.7 – 14.7 (11.3)
		SP 02	10.2 – 24.0 (14.5)	6.8 – 17.5 (10.0)
Fe	**43200**	**SP 01**	25250 – 52560 (36366)	-
		SP 02	33900 – 42100 (37062)	-

[1]CC = Continental crust values as published by Wedepohl in 1995.
[2]Determined by Neutron Activation Analysis (NAA)
Note: Values given in the parenthesis are the average values

Enrichment factors

The enrichment factors obtained in this study (Table 5 and Fig. 7) indicated that sediments are enriched by arsenic in the entire cores collected from SP01 and SP02 with significant enrichment. For core SP 01, the EFs of arsenic showed enrichment throughout the sediment core, especially at the bottom part of the core. Meanwhile, for core SP 02, arsenic also showing significant enrichment especially when reaching the peak at the middle part of the core, while the top and bottom portions of the core are less enriched. Both cores having enrichment with an average EF value of about 10.0.

In summary, the EFs obtained for the arsenic in the sediments from the Juru River are quite significant. The Perai Industrial area is densely populated and is comprised of many heavy industries, and the most probable sources of pollution might be from the manufacturing industries, agriculture and sewage emissions. Other human activities such as the use of arsenical herbicides mining, metallurgy and wood preservation can also introduce arsenic into the water bodies eventually deposited into sediment (Sarmani, 1989; Shazili et al., 2006; Yunus et al., 2020).



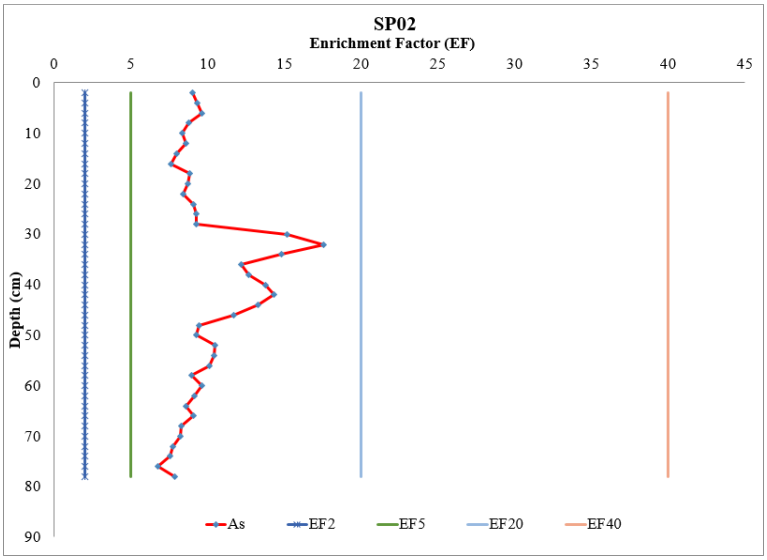


Figure 7. Elemental Enrichment Factor versus depth for core SP01 and SP02

CONCLUSION

Two sediment cores were collected at the Juru River located in Penang's Perai Industrial Area in February 2017 and analysed using nuclear techniques, with the purpose to evaluate the sediment accumulation rates and to assess the enrichments by arsenic at the study area. Concentrations of arsenic were quantified using Neutron Activation Analysis (NAA) technique. Using gamma spectrometry, ^{210}Pb and ^{226}Ra activity profiles were established for each core; however, owing to the complexity of the excess ^{210}Pb activity profiles obtained, it was not possible to obtain an accurate age model from them. Nonetheless, preliminary apparent sediment accumulation rates were estimated, and were found to range between 0.94 – 2.70 cm/y, with the highest sedimentation rate recorded in the core collected at the estuary of Juru River (SP02). Arsenic was found significantly enriched in the study area but the results are still comparable to other studies, therefore further monitoring and mitigation strategies for the pollutant are strongly recommended to improve environmental sustainability.

FUTURE WORKS

To have a better understanding of the elemental deposition history at the study area, for future analysis, thin slide of sediment (0.5 cm – 1.0 cm instead of 2 cm that is being practiced now) can be performed to get a better estimation of sediment accumulation rate. Various analytical techniques can be applied as a comparison as well.

ACKNOWLEDGEMENTS

The author would like to express his special appreciations to the International Atomic Energy Agency for providing research funding under research grant (K41016 – RC 20884), to the project Technical Officer, Ms. Martina Rozmaric Macefat and Member States participating the K41016 project for their

guidance. He would also like to thank the personnel of Radiochemistry and Environment Group (RAS), and Analytical Chemistry Group (ACA) from Malaysian Nuclear Agency for their constant support and cooperation throughout this study.

REFERENCES

Abrahim, G.M.S. & Parker, R.J. (2008). Assessment of heavy metal enrichment factors and the degree of contamination in marine sediments from Tamaki Estuary, Auckland, New Zealand, *Environ. Monit. Assess.* 136:227–238.

Ahmed, N.K. & El-Arabi, A.G.M. (2005). Natural radioactivity in farm soil and phosphate fertilizer and its environmental implications in Qena governorate, upper Egypt, *J. Environ. Radioact.* 84(1):51–64.

Aliev, R.A., Bobrov, V.A., Kalmykov, St.N., Melgunov, M.S., Vlasova, I.E., Shevchenko, V.P., Novigatsky, A.N. & Lisitzin, A.P. (2007). Natural and artificial radionuclides as a tool for sedimentation studies in the Arctic region, *J. Radioanal. Nucl. Chem.* 274: 315–321.

Alnour, I.A., Wagiran, H., Ibrahim, N., Hamzah, S., Wee, B.S. & Elias, M.S. (2014). New approach for calibration the efficiency of HPGe detectors. In: AIP Conf. Proc. 1584:38–44.

Alongi, D.M., Pfitzner, J., Trott, L.A., Tirendi, F., Dixon, P. & Klumpp, D.W. (2005). Rapid sediment accumulation and microbial mineralization in forests of the mangrove Kandelia candel in the Jiulongjiang Estuary, China, *Estuar. Coast. Shelf Sci.* 63(4):605–618.

Arogunjo, A.M., Ofuga, E.E. & Afolabi, M.A. (2005). Levels of natural radionuclides in some Nigerian cereals and tubers, *J. Environ. Radioact.* 82(1):1–6.

Ashraf, A.R., Saion, E., Gharibshahi, E., Yap, C.K., Kamari, H.M., Elias, M.S. & Rahman S.A. (2018). Distribution of heavy metals in core marine sediments of coastal East Malaysia by instrumental neutron activation analysis and inductively coupled plasma spectroscopy, *Appl. Radiat. Isot.* 132:222–231.

Abdullah, M.I.C., Sah, A.S.R.M. & Haris, H. (2020). Geoaccumulation Index and Enrichment Factor of Arsenic in Surface Sediment of Bukit Merah Reservoir, Malaysia, *Trop. Life Sci. Res.* 31(3): 109–125.

Benoit, G. (1988). The biogeochemistry of ^{210}Po and ^{210}Pb in fresh waters and sediments, PhD Dissertation, Massachusetts Institute of Technology, Cambridge, Massachusetts, PB--89-198097/XAB.

Bralower, T.J. & Thierstein, H.R. (1987). Organic carbon and metal accumulation in Holocene and mid-Cretaceous marine sediments: Paleoceanographic significance, *Geol. Soc. Spec. Publ.* 26(1):345–369.

Burke et al. (2001). World Resources Institute (WRI). Washington DC, Paul Harrison and Fred Pearce, AAAS Atlas of Population and Environment 2001, AAAS. University of California Press, Berkeley, 216.

Chakrabarty, A., Jha, S.K., Gothankar, S.S., Tripathi, R.M., Khan, A.H. & Puranik, V.D. (2006). Sedimentation rate of Nagarjuna Sagar Dam using a natural radiotracer. In: Application of Radiotracers in Chemical, Environmental and Biological Sciences, S., Lahiri, D., Nayak, & A. Mukhopadhyay, (eds.). Vol. 2, Saha Institute of Nuclear Physics, Kolkata, 148–150.

Cheevaporn, V., Jacinto, G.S. & Sandiego-Mcglonne, M.L. (1994). History of heavy metal contamination in Bang Pakong river estuary, Thailand, *J. Sci. Soc. Thailand.* 20:9–22.

Chen, S.B., Zhu, Y.G. & Hu, Q.H. (2005). Soil to plant transfer of ^{238}U, ^{226}Ra and ^{232}Th on a uranium mining-impacted soil from south eastern China, *J. Environ. Radioact.* 82(2):223–236.

Cochran, J.K. & Masque, P. (2004). Natural radionuclides applied to coastal zone process. In: Marine Radioactivity, H.D. Livingston, (ed.). Elsevier, Amsterdam, 1–21.

Cuculic, V., Cukrov, N., Barisic, D. & Mlakar, M. (2006). Uranium in sediments, mussels (Mytilus sp.) and seawater of the Krka river estuary, *J. Environ. Radioact.* 85(1):59–70.

Deely, J.M. & Fergusson, J.E. (1994). Heavy metal and organic matter concentration and distributions in dated sediments of a small estuary adjacent to a small urban area, *Sci. Total Environ*. 153(1–2):97–111.

Dowdall, M. & O'dea, J. (2002). Ra-226/^{238}U disequilibrium in an upland organic soil exhibiting elevated natural radioactivity, *J. Environ. Radioact.* 59(1):91–104.

Elias, M.S., Hamzah, M.S., Rahman, S.A., Wee, B.S. & Salim, N.A.A. (2012). Assessment of sediment quality collected from Tunku Abdul Rahman Park, Sabah, *Nucl. Sci. J. Malays.* 24(1):59–70.

Elias, M.S., Hamzah, M.S., Rahman, S.A., Wee, B.S., Salim, N.A.A., Sanuri, E., Talib, A., Yatim, J.M., Devi, P., Baharuddin, A.H., Put, E.E., Harun, M.K.A. & Awang, M.K. (2014). Development of Fingerprint Characteristic for forensic Investigation of Elemental Pollution in Sediment From Linggi Area, Tech. Rep. ScienceFund Project (04-03-01-SF0142), NUKLEARMALAYSIA/L/2014/102. Malaysian Nuclear Agency, Kajang, 11.

Elias, M.S., Ibrahim, S., Samuding, K., Rahman, S.A. & Yii, M.W. (2018). Assessment of toxic elements in sediments of Linggi river using NAA and ICP-MS techniques, *MethodsX* 5:454–465.

El-Reefy, H.I., Sharshar, T., Zaghloul, R. & Badran, H.M. (2006). Distribution of gamma-ray emitting radionuclides in the environment of Burullus Lake: I. Soils and vegetation, *J. Environ. Radioact.* 87(2):148–169.

International Atomic Energy Agency (1990). Practical aspects of operating a neutron activation analysis laboratory, TECDOC-564. IAEA, Vienna, 252.

Islam, M.A., Al-Mamun, A., Hossain, F., Quraishi, S.B., Naher, K., Khan, R., Das, R., Tamim, U., Hossain, S.M. & Nahid, F. (2017). Contamination and ecological risk assessment of trace elements in sediments of the rivers of Sundarban mangrove forest, Bangladesh, *Mar. Pollut. Bull.* 124(1):356–366.

James, W.D., Boothe, P.N. & Presley, B.J. (1998). Compton suppression gamma-spectroscopy in the analysis of radium and lead isotopes in ocean sediments, *J. Radioanal. Nucl. Chem.* 236:261–266.

Joseph, B., Adiana, G., Shazili, N.A.M., Ong, M.C., Juahir, H., Shaari, H., Yii, M.W., Kamarudin, M.K.A., Gasim, M.B., Maulud, K.N.A. & Islam, M.S. (2018). The Evaluation of Brunei Bay Sediment Cores Sedimentation Rate Using ^{210}Pb Radiometric Dating Technique, *Int. J. Res. Eng. Technol.* 7(3.14):107–114.

Krishnaswami, S. & Lal, D. (1978). Radionuclide alimnochronology. In: Lakes, Chemistry, Geology, Physics, A. Lerman, (ed.). Springer Verlag, New York, 153–177.

Krishnaswamy, S., Lal, D., Martin, J. & Meybeck, M. (1971). Geochronology of lake sediments, *Earth Planet. Sci. Lett.* 11(1):407–414.

Li, H., Ye, S., Ye, J., Fan, J., Gao, M. & Guo, H. (2017). Baseline survey of sediments and marine organisms in Liaohe Estuary: heavy metals, polychlorinated biphenyls and organochlorine pesticides, *Mar. Pollut. Bull.* 114(1):555–563.

Li, Y. H., Mathieu, G.G., Biscaye, P. & Simpson, H.J. (1977). The flux of ^{226}Ra from estuarine and continental shelf sediments, *Earth Planet. Sci. Let.* 37(2):237–241.

Likuku, A.S. (2006). Factors influencing concentrations of ^{210}Pb and ^{7}Be over the city of Edinburgh (55.9^{o}N, 03.2^{o}W), *J. Environ. Radioact.* 87(3): 289–304.

Loring, D.H. & Rantala, R.T.T. (1992). Manual for the geochemical analyses of marine sediments and suspended particulate matter, *Earth-Sci. Rev.* 32(4):235–283.

Mishra, U.C. & Sadasivan, S. (1971). Gamma spectroscopic measurements of soil radioactivity, *Int. J. Appl. Radiat. Isot.* 22:256–257.

Nguyen, T.N., Nguyen, T.B., Nguyen, V.P., Le, N.S., Truong, Y., Mai, T.H., Nguyen, T.L., Nguyen, M.S., Phan, S.H., Le, N.C., Dang, D.N., Nguyen, Q.L., Nguyen, H.Q. & Tran, T.M. (2009). Radionuclides concentration in marine environmental samples along the coast of Vietnam, *Nucl. Sci. J. Malays.* 21(2):12–20.

Niencheski, L.F., Windom, H.L. & Smith, R. (1994). Distribution of particulate trace metal in Patos Lagoon estuary (Brazil), *Mar. Pollut. Bull.* 28(2):96–102.

Ontiveros-Cuadras, J.F., Ruiz-Fernández, A.C., Sanchez-Cabeza, J.A., Wee-Kwong, L.L. & Pérez-Bernal, L.H. (2012). Geochemical fractionation of ^{210}Pb in oxic estuarine sediments of Coatzacoalcos River, Gulf of Mexico, *J. Radioanal. Nucl. Chem.* 292**:**947–956.

Pandey, J. & Singh, R. (2017). Heavy metals in sediments of Ganga River: Up and down stream urban influences, *Appl. Water Sci.* 7:1669–1678.

Ruiz-Fernandez, A.C., Sanchez-Cabeza, J.A., Alonso-Hernandez, C., Martinez-Herrera, V., Perez-Bernal, L.H., Preda, M., Hillaire-Marcel, C., Gastaud, J. & Quejido-Cabezas, A.J. (2012). Effect of land use change and sediment mobilization on coastal contamination (Coatzacoalcos River, Mexico), *Continental Shelf Res.* 37:57–65.

Saion, E., Wood, A.K.H., Sulaiman, Z.A., Alzahrany, A.A., Elias, M.S. & Wee, B.S. (2007). Determination of heavy metal pollution in depth profile of marine sediment samples from the Strait of Malacca, *Fresenius Environ. Bull*. 16(10):1279–1287.

Sanchez-Cabeza, J.A. & Ruiz-Fernandez, A.C. (2012). Pb-210 sediment radiochronology: An integrated formulation and classification of dating models, *Geochim. Cosmochim. Acta.* 82:183–200.

Sanders, C.J., Sanders, L.M., Smoak, J. M., Patchineelam, S., Machado, W. & Luiz-Silva, W. (2010). Radium-226 and lead-210 ratios along a sediment core profile implying fertilizer industry source. In: Conference AGU meeting of the Americas, August 2010, Foz do Iguacu.

Sarmani, S.B. (1989). The determination of heavy metals in water, suspended materials and sediments from Langat River, Malaysia, *Hydrobiologia.* 176:233–238.

Shazili, N.A.M., Yunus, K., Ahmad, A.S. & Abdullah, N. (2006). Heavy metal pollution status in the Malaysian aquatic environment, *Aquat. Ecosys. Health.* 9(2):137–145.

Skwarzec, B. & Jahnz, A. (2007). The inflow of polonium ^{210}Po from Vistula river catchments area, *J. Environ. Sci. Health Part A.* 42(14):2117–2122.

Szefer, P., Szefer, K., Glasby, G.P., Pempkowiak, J. & Kaliszan, R. (1996). Heavy–metal pollution in surficial sediments from the Southern Baltic sea off Poland, *J. Environ. Sci. Health.* 31(10):2723–2754.

UNESCO, (1978). UNESCO/SCOR Workshop on The Biogeochemistry of Estuarine Sediments, UNESCO, New York.

United Nation, (2017). Factsheet: People and Oceans. In: The Ocean Conference, United Nations, 05–09 June 2017, New York.

Wan-Mahmood, Z. & Yii, M.W. (2012). Marine radioactivity concentration in the Exclusive Economic Zone of Peninsular Malaysia: ^{226}Ra, ^{228}Ra and ^{228}Ra/^{226}Ra, *J. Radioanal. Nucl. Chem.* 292:183–192.

Wan-Mahmood, Z. & Yii, M.W. (2013). Sedimentation rate in the Sungai Linggi estuary using excess ^{210}Pb and ^{137}Cs, *J. Radioanal. Nucl. Chem.* 298:1727–1732.

Wan-Mahmood, Z., Ahmad, Z., Ishak, A. K., Yii, M. W., Mohamed, N., Sharib, J., Ishak, K., Razali,

K. N. & Mahmud, M. (2005). Kajian Awal Ke Atas Taburan Radionuklid Tabii Di Perairan Pantai Timur Semenanjung Malaysia, *Malaysian J. Anal. Sci.* 9(2):325–337. (in Malays)

Wan-Mahmood, Z., Yii, M.W. & Ishak, A.K. (2016). Po-210, ^{210}Pb and ^{210}Po/^{210}Pb in sediment core from surrounding Sungai Linggi estuary, *Nucl. Sci. J. Malays.* 28(1):1–8.

Wedepohl, K.H. (1995). The composition of the continental crust, *Geochim. Cosmochim. Acta* 59(7):1217–1232.

Wood, A.K., Muhammad, N., Mahmood, C.S., Ahmad, Z., Shazili, N.A., Law, A.T. & Yaakob, R. (1993). Corer sampling and the use of neutron activation analysis in evaluating pollution at the Juru waterway, Penang, *Nucl. Sci. J. Malays.* 11(2):105–128.

Xu, Z., Salem, A., Chen, Z.Y., Zhang, W.G., Chen, J., Wang, Z.H., Sun, Q. L. & Yin, D.W. (2008). Pb-210 and Cs-137 distribution in Burullus lagoon sediments of Nile river delta, Egypt: sedimentation rate after Aswan High Dam, *Front Earth Sci-PRC.* 2**:**434–438.

Yang, Y.X., Wu, X.M., Jiang, Z.Y., Wang, W.X., Lu, J.G., Lin, J., Wang, L.M. & Hsia, Y.F. (2005). Radioactivity concentrations in soils of the Xiazhuang granite area, China, *Appl. Radiat. Isot.* 63(2):255–259.

Yi, Y.J., Yang, Z.F. & Zhang, S.H. (2011). Ecological risk assessment of heavy metals in sediment and human health risk assessment of heavy metals in fishes in the middle and lower reaches of the Yangtze River basin, *Environ. Pollut.* 159(10):2575–2585.

Yii, M.W., Ahmad, Z. & Ishak, A.K. (2009). Distribution of naturally occurring radionuclides activity concentration in East Malaysian marine sediment, *Appl. Radiat. Isot.* 67(4):630–635.

Yii, M.W., Ruiz-Fernandez, A.C., Wan-Mahmood, Z. & Paulus, W. (*in press*). Deposition history of Arsenic, Barium, Chromium and Zinc at Perai Industrial Area of Penang, Malaysia–A case study using nuclear techniques, TECDOC. IAEA, Vienna, (*in press*).

Yii, M.W., Paulus, W. & Wan-Mahmood, Z. (2020a). Time deposition of copper, nickel and manganese at Perai river, Penang. In: Seminar R&D 2020, 16–19 November 2020, Nuklear Malaysia, Bangi.

Yii, M.W., Wan-Mahmood, Z., Elias, M.S. & Abdullah, Y. (2020b). The distribution of heavy metals and natural radionuclides within the surface sediments of the Juru river, Penang. In: AIP Conf. Proc. 2295:020006.

Yii, M.W., Wan-Mahmood, Z., Sharib, J. & Ahmad, Z. (2010). Comparison of ^{210}Pb level in Kuala Muda tsunami affected marine sediment core measured using two different techniques, *Nucl. Sci. J. Malays.* 22(1):29–41.

Yii, M.W. et al. (2016). Ambient radioactivity and radiological studies in the vicinity of Lynas Rare-Earth Processing Plant, Gebeng Industrial Estate, Kuantan, Pahang, Tech. Rep. ScienceFund Project (06-03-01-SF0189), NUKLEARMALAYSIA/L/2016/196. Malaysian Nuclear Agency, Kajang, 145.

Yunus, K., Zuraidah, M.A. & John, A. (2020). A review on the accumulation of heavy metals in coastal sediment of Peninsular Malaysia, *Ecofeminism and Climate Change.* 1(1):21–35.

Zainudin, Z. (2010). Benchmarking River Water Quality in Malaysia, *Jurutera.* 12–15.

Zhu, L.M., Xu, J., Wang, F. & Lee, B. (2011). An assessment of selected heavy metal contamination in the surface sediments from the South China Sea before 1998, *J. Geochem. Explor.* 108(1):1–14.