

Sedimentation, Heavy Metals Profiles and Cluster Analysis of a Former Tin Mining Lake

Zaini Hamzah, Ahmad Saat, Abdul Khalik Wood, and Zaharidah Abu Bakar

Abstract—Pb-210 method has been applied to sedimentation study on an abandoned tin mining lake in Malaysia. In recent years the lakes areas have been converted into more economically important areas with activities such as agriculture, recreational and residential. The sediment profiles at locations in the lakes are expected to be impacted by the discharge of these near shores and on lakes activities. The Pb-210 depth profiles of the cores enable the respective sediment age and sedimentation rate to be determined. The sedimentation rate was found to be $0.57 \pm 0.10 \text{ cm.y}^{-1}$. Age wise, the sediment were about 40 – 50 years old. For metals profiles, generally synchronous higher concentrations were observed in the younger section of the cores. This coincides with the period of the intensification of anthropogenic activities in the area. Mean metal concentrations for all cores for As, Cr, Mn, Zn, U-238 and Th-232 were 48.9 ppm, 41.1 ppm, 407.9 ppm, 199.1 ppm, 30.6 ppm and 85.7 ppm respectively. These results are twice as high compared to results of a control location on undisturbed land. Discharges from such activities containing heavy metals get their way into the lake through surface deposition by rain water, and remobilized from the depositional areas to the sediment coring sites. U-238 and Th-232 may be attributed to the minerals contained in the tin tailings left in the area. Cluster analysis of the metals indicated they are of both anthropogenic and natural origins.

Index Terms—Sediments, heavy metals, Pb-210 method, radionuclides.

I. INTRODUCTION

Sedimentation and chronology study on water and catchments system is important and significant as sedimentation rate is an imperative parameter that determines the water system dynamics. Both natural and anthropogenic factors determined the rate and character of accumulated sediments, while the morphology of the lake basin determines the prevalence of sediment erosion, transportation and accumulation zones [1], [2]. Chronology information of sediment cores are useful in determining the inventory, history and causes of contamination, and checking whether the buried contaminants are migrating, accumulating or

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degenerating [3].

Pb-210 dating method makes use of the steady supply of Pb-210 isotopes in a given location. The method was first used for sediment study by Krishnaswami *et al.* [4], however the theoretical background was described in details in a number of articles [5],[6]. Previous applications show that the method performs best on sedimentation study in relatively older (more than 100 years) [3] and quiescent sediment deposition areas such as salt marsh, bays [7] and natural lakes ([8]).

Heavy metals are released into the environment via airborne contaminants, rural land use activities, sewage sludge, mine waste, industrial waste, wastewater, pesticides and fertilizers applications. Thus, in general industrialization and human activities clearly the main contributors to heavy metals discharged into our ecosystem, such as rivers, lakes, estuaries and marines [9], [10], [11]. These metals subsequently enter into the human food chain either directly or indirectly. Some heavy metals are potentially harmful (Cd, Hg, Pb) but some are essentially important to human health (Fe, Ca, Mg), however high concentration could affect human health.

The lakes within the precinct of Universiti Teknologi MARA Training Centre in Kampung Gajah, Perak are part of the man-made small fresh water lakes system formed by the tin mining dredging activities that ceased operating about 40 to 50 years ago. Over the last decade intensive human activities such as housing, agriculture, fish and animals rearing had taken place around and within the lakes system. These activities might facilitate erosion and pollution inputs into the lakes. Thus increasing research interest has been focused to study such lakes.

Profiles of sediments could facilitate us to acquire information on contamination of lakes such as heavy metal discharge, atmospheric deposition, past drainage basin or land-use history [12], [13]. Sediments composed of fine sand and silt, and due to its stable condition metals to be more easily entrapped in the sediment. Moreover, the fine-grained sediment will acted as effective carrier of heavy metals [9]. The present study explored the vertical profile distributions of Pb-210 and heavy metals including uranium and thorium in sediments of a representative fresh water former tin mining lake, to evaluate the sediment chronology and extent of pollution contributed by these metals.

II. METHODOLOGY

A. Study Area

The studied former tin mine fresh water lake is about five hectares in area, located on central Peninsular Malaysia, centre at around N 04° 14.954", E 101° 3.023". It is representing one of the many interconnecting lakes system in

the vicinity. The deepest point is about 9 m, located near the middle of the lake. During sampling in September 2007 the water volume is estimated to be around 10^8 m^3 . The lake is located about 50 m from a main road. Within the lakes water system there are fish and ducks rearing activities. There are water buffalos and cattle farms, as well as recent settlements on land areas within the lakes system. On the studied lake there are two small islands located near the centre

B. Sampling and Sample Preparation

Sediment cores were collected at five points in the lake by using manual gravity corer with PVC core tube of 50 mm inner diameter. These points represent the different base surface morphology of the lake. Five cores of sediments were sampled at each point. Sediment cores obtained were about 20 to 25 cm in length. The cores were sub-sampled by slicing into 2 cm slices. Samples preparation as described earlier [14] where slices of identical depth of the five cores in each point were mixed to form homogeneous representative aggregate at that depth. The aggregates were oven-dried at 60°C until constant mass, pulverized and sieved through $450 \mu\text{m}$ stainless steel sieves. They were then kept in desiccators for one week to reduce water content. The dried samples were kept in air-tight containers for about six months to ensure secular equilibrium of the uranium and thorium series. Control sample collected approximately 5 km from the study lake and undisturbed by mining activities was prepared in the same manner as the core samples. Table I summarizes the sampling locations for samples and control (C2).

TABLE I: SAMPLING LOCATIONS AND WATER DEPTH

Sampling Points	Latitude	Longitude	Water Depth (m)
S1	N 04° 14.941'	E 101° 03.037'	8.67
S2	N 04° 14.863'	E 101° 03.133'	3.69
S3	N 04° 15.008'	E 101° 03.110'	2.05
S4	N 04° 14.990'	E 101° 02.985'	8.05
S5	N 04° 14.844'	E 101° 03.015'	3.70
C2	N 04° 14.907'	E 101° 02.140'	-

C. Method

In this study Pb-210 was determined indirectly by measuring Po-210 using alpha-spectrometry method facility at the Malaysian Nuclear Agency (MNA) Laboratory in Bangi. As an internal standard, known amount of Po-209 was added to known mass (about 0.20 g) of the dried aggregate sediment, and then digested using concentrated HNO_3 and HCl at 90°C . After acid digestion Po-210 was spontaneously deposited for twenty-four hours on clean 2 cm x 2 cm silver disc. Alpha activity of Po-210 was then measured using an EG & G ORTEC alpha-spectrometer.

Heavy metals concentrations were determine using Neutron Activation Analysis (NAA), employing irradiation and measurements facilities of Malaysian Nuclear Agency. Know amount (ca. 0.10 g) of samples and standards were irradiated in the MNA TRIGA MARK II 750 kW research nuclear reactor with thermal neutron flux of $2.0 \times 10^{12} \text{ n.cm}^{-2}\text{s}^{-1}$. Depending on elements to be determined either short (1 minute) or long (6 hours) irradiation and the

corresponding short (20 minutes) or long (3 – 5 days) cooling time procedures were employed. Gamma-ray measurements were carried out using Canberra GEM-series HPGe detector of 1.88 keV resolution and 25.4% relative efficiency at 1332 keV Co-60 gamma-ray, couple to an MCA. Spectrum analyses were carried out Gamma Vision V6.0 software.

III. RESULTS AND DISCUSSION

A. Sedimentation

In Figure 1, the excess Pb-210 activity concentration is plotted against the corresponding sediment depth at the deepest sampling point, S1. Due to the compaction effect on the shallower layers that could introduce serious errors in the calculated values, the first two layers were not considered in the curve fittings. The curve follows a relatively smooth exponential trend despite the variance of about 30% in the estimated exponential equation. Thus it may be stated that the assumptions of the Constant Rate of Supply (CRS) model [5] are fulfilled where the excess Pb-210 activity concentration depth profile will normally follow exponential change as $A_x = A_o e^{-kx}$. Where A_x is activity concentration at depth x (cm), A_o activity concentration at the sediment-water interface, and k is a constant. Knowing the decay constant, λ ($= \ln 2 / 22.26 \text{ yr}^{-1}$) of Pb-210, the constant $k \text{ cm}^{-1}$ can be used to calculate the sedimentation rate $S = \lambda / k$. Hence the age, t (year) of sediment at certain depth of x cm is calculated using $t = x / S$. Finally, the year associated with the related sediment depth is obtained by subtracting t from the sampling date (year).

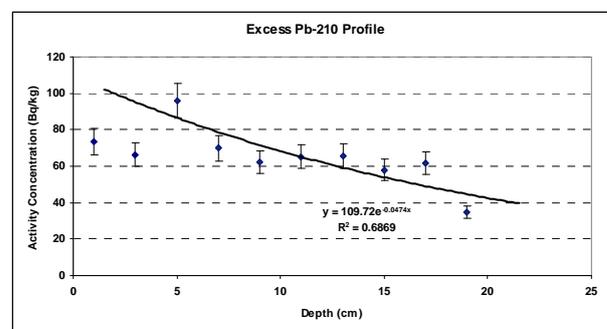


Fig. 1. Excess Pb-210 profiles in S1.

The least-squared fitting in Figure 1 gives a k value of 0.0474 cm^{-1} and this gives the sedimentation rate of the core to be 0.567 cm y^{-1} . This value represents the average sedimentation rate over the time period of accumulation depth of the sediment. On the basis of the measured activity concentrations and the cross-sectional area ($X \text{ m}^2$) of the core, the excess Pb-210 inventory and flux can be calculated by

using $I = \frac{1}{X} \sum_i m_i A_i$ and $\phi = \lambda I$ respectively, where m

(kg) is the mass of the respective core slices. In this study they were found to be $16468 \pm 128 \text{ Bqm}^{-2}$ and $512 \pm 23 \text{ Bq m}^{-2} \text{ y}^{-1}$ respectively. This flux is much higher than the regional atmospheric input ($50 \text{ Bq m}^{-2} \text{ y}^{-1}$) and the global average of $183 \text{ Bq m}^{-2} \text{ y}^{-1}$, suggesting that the sampling area experiences

higher in-situ supply of Pb-210. The main sources of excess Pb-210 are inflow [5] from other connected lakes system, and sediment focusing related to the Lake Basin morphology, slope action and random redistribution [1]. Based on the location of the studied lake in the interconnected lake-water system both sources above are acceptable explanations. Although the natural fall-out is very low in this region, being a former tin mining area the area is rich with uranium and thorium bearing minerals such as monazite and ilmenite. Uranium and thorium are sources of radioactive radon gases, Rn-222 and Rn-219. These gases decay in chains through Pb-210 before reaching stability. Therefore, it is favorable to assume that these gases being emanated from the soil in the area, and their daughters, including Pb-210, are accumulated in the lake either through dry deposition or together with the rainfall. However, since this is not the objective of the present study it will not be explored in details here.

The sedimentation rate obtained from Figure 1 was used to estimate the age of the sediment layers and corresponding sediment dates. The results are shown in Table II. For the length of the core study, the age estimated by the CRS model of Pb-210 method was found to be about 40 years. Not forgetting the uncertainties attributed to the modification of the method to suit the study of recent sediment, the 40 years age is in agreement to the time when the mining activities stopped in the area at around late 1960's. This date can be assumed as the beginning of the natural sedimentation process. As mentioned earlier the compaction or loss of surface part of the sediment core is the main difficulty and the source of uncertainty in applying this method to recent lake sediment. This fact emphasizes the importance of coring techniques as well as slicing.

TABLE II: SEDIMENT AGE, AND DATE IN RELATION TO DEPTH.

Depth (cm)	Sediment Age (year)	Sediment Date
1	2	2005
3	5	2002
5	8	1999
7	11	1996
9	14	1993
11	17	1990
13	20	1987
15	23	1984
17	26	1981
19	29	1978
21	32	1975
23	35	1972
25	38	1969
27	44	1963

B. Metals Profiles

From Figure 2, generally the results of the present study indicated that all metals concentrations in the sediment cores at all sampling point were more than twice higher than those from the control core, C2. The near uniform trend of metals, uranium and thorium concentrations in the control core profile is an indication of insignificant accumulation of elements over time in the core. However the trend profiles of metals shows synchronously higher concentration in core depth down to about 8 cm. According to previous study on the sediment age of the lake, this correspond to about a decade of sedimentation [15], that coincides with the

beginning of anthropogenic activities in and around the lakes system. As for uranium and thorium the same trend is also observable except for cores S4 and S5 (Figure 2(e) and (f)) where higher concentrations were found at older sections of the sediment cores

Arsenic. The As profile for all cores (Figure 2(a)) showed higher concentration in the younger sediment, near at the surface layers that may be attributed atmospheric deposition from fuel combustion during past mining activities or road traffic and agricultural activities. S2 is located about 50 m from the main road. Previous studies showed atmospheric deposition was mainly linked to high concentration of As, Zn, Co and Pb [15], [16], [17]. It is generally known that As was found in contaminated tin-ore in the form of arsenious oxide and removed by roasting and washing from the ore. The by product, arsenious oxide, might originated from previous mining activities and remobilized into the lake. The similar trend portrayed by As and Mn (Figure 2(c)) could be interpreted as remobilization has taken place [19]. The overall mean of As concentration in all cores is 48.9 ± 9.6 ppm, which is higher than the Severe Effect Level (SEL) of 33 ppm of the Sediment Quality Guidelines of New York Department of Environmental Conservation (NYDEC), where pronounced disturbance of the sediment dwelling community can be expected [19].

Chromium. Figure 2(b) showed increasing trend in less deep sediment for Cr concentration in sediments at coring points. However, the profile of S5 sediment core showed an almost constant trend from the upper layer to the bottom of sediment. The presence of Cr in the sediment lake might be due to industrial emission, lithogenic origin (mine extraction), naturally origin or atmospheric deposition [16]. One notable observation here is that the overall mean concentration of 41.1 ± 3.6 ppm is lower than the SEL of 110 ppm, however higher than the NYDEC Lower Effect Limit (LEL) of 26 ppm. This can cause toxicity to some aquatic species in the vicinity [19].

Manganese. Mn showed an almost identical profile trend as As (Figure 2(c)). Generally, concentration of Mn showed decreasing trend to the upper layer and high at about 10 cm depth. Only S2 and S5 showed significantly high concentration of Mn observed in the upper layer of sediment. Mn could also be attributed to remobilization from into the lake from former ore processing activities. The overall average of Mn in all cores is 407.9 ± 68.3 ppm. This is lower than the LEL concentration of 460 ppm [19] [12].

Zinc. Smelting operation, usage of fertilizer and pesticides in agriculture, soil erosion due to human activities, fossil fuel and land development could contribute to high concentration of Zn in sediment lake [7, 8, 9]. The studied lake is located near to duck farming activity and main road. This factor as well as the other factors mentioned earlier seems to be consistent in contributing to increasing concentration of Zn in lake sediment, especially the younger sediments (Figure 1(d)). The overall mean of Zn concentration in all cores is 199.1 ± 17.6 ppm, which is higher than LEL of 120 ppm but lower than SEL of 270 ppm of the Sediment Quality Guidelines of New York NYDEC [19].

Uranium and Thorium. Two contrasting patterns of profile were observed for U and Th (Figures 2(e) and 2(f)). One at locations S1, S2 and S3 that showed almost similar pattern to

other metals, while S4 and S5 showed an opposite pattern. The U and Th concentration at the surface layers of the sediment may be due to anthropogenic input from fertilizer used in agriculture. The overall mean of U and Th concentration in all cores are 30.6 ± 6.1 ppm and 85.7 ± 10.0 ppm respectively, which is much higher than the average earth crust composition of 3.5 ppm and 11.0 ppm [19] is expected, due to the high concentration of U and Th in monazite and ilmenite minerals that are abundance in former tin mining areas. Over the years, weathering and erosion remobilized the minerals into the lake [20]. This may explained the observation that higher concentration in older layer of sediments in S4 and S5. The concentration of Th is always higher than U in the lake sediment due to the fact that Th that occurs naturally is among the least soluble and immobile trace element in natural water. Relatively U is more soluble than Th thus has greater mobility in water. A study on the ratio of Th/U of the five cores showed the average values ranged between 2.20 – 4.21. These agreed well to the world average ratio of Th/U of approximately 3.5 – 4.0.

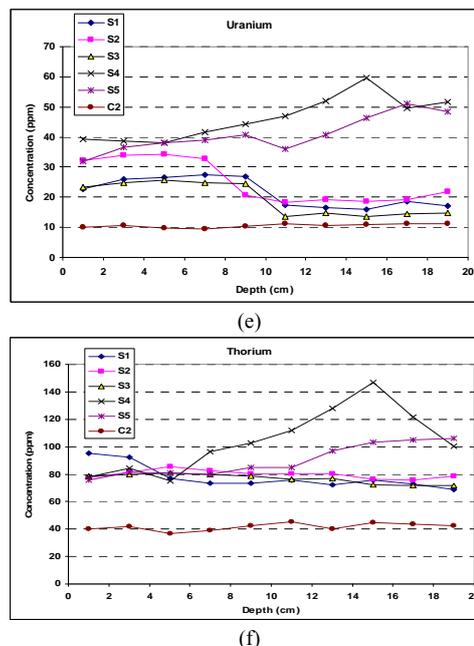
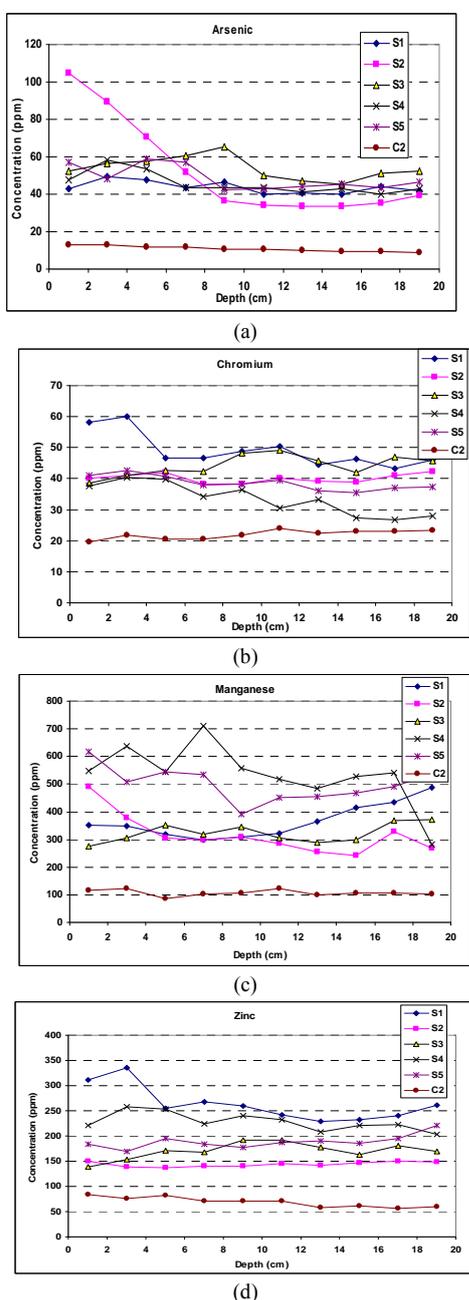


Fig. 2. Depth profiles of As, Cr, Mn, Zn, U and Th in sediment cores and control site.

C. Correlation and Cluster analysis

Correlation analysis of metals within the sediment lake are presented in Table III. In the table, Al, Co and Fe are also included for comparison purpose, although their profiles were not analysed in the present paper. Correlation between Al with As and Cr showed strong and good correlation respectively. Al showed least correlation with Co, Fe, Mn, Zn and Th. While As have good correlation with Co, Cr, Fe, Mn, Zn, and Th. Co have good correlation with Cr, Fe, Zn and Th. Cr have good correlation Fe, Zn, and LOI. Fe have good correlation Mn, Zn and Th. Mn have strong correlation only with Th. Zn have good correlation with Th. Th showed strong correlation with Mn and good correlation with As, Co, Fe, and Zn. While least correlation between Th with Al, Cr and U. Correlation analysis for U showed no correlation with Al, Co, Cr, Fe and Zn and low correlation with As, Mn and Th. Although loss-on-ignition (LOI) is not shown in the figure, generally good correlations between % LOI were observed with metals element excepted for U showed low correlation. This finding showed organic matter play an important role in metals concentration within sediment lake and corresponds to the previous studies [18], [21], [22].

TABLE III: CORRELATION COEFFICIENT (R^2) OF AL, AS, CO, CR, MN, ZN, TH, AND U IN LAKE SEDIMENT

Element	As	Co	Cr	Fe	Mn	Zn	Th	U
Al	0.77	0.33	0.53	0.25	0.26	0.23	0.64	0.06
As		0.52	0.61	0.56	0.56	0.49	0.60	0.24
Co			0.55	0.81	0.34	0.53	0.24	0.00
Cr				0.55	0.13	0.73	0.49	0.00
Fe					0.54	0.56	0.17	0.03
Mn						0.20	0.23	0.23
Zn							0.51	0.09
Th								0.13

Good correlation between Co, As, Cr, Zn, and Th to each other could be suggests from same source of contamination. This correlation suggests contamination input is mainly due to atmospheric deposition, human settlement and agriculture activity. While good correlation between As, Co, Cr, Th and Zn with either major elements (Al, Fe and Mn) indicated a natural origin of these elements in the sediment lake. Thus, presence of metals elements in the sediment lake is suggests from mainly from natural and anthropogenic input. This is due to profile of metals element within the sediment cores as discussed earlier showed increasing trend at the surface sediment.

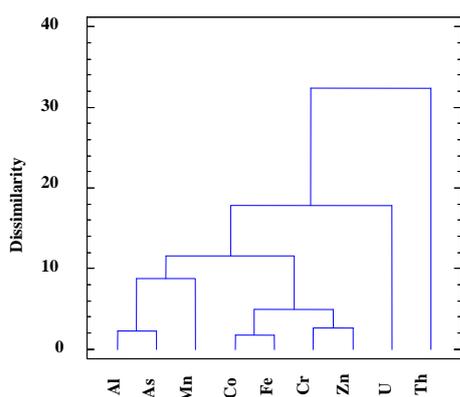


Fig. 3. Dendrogram analysis mean of metal elements within the sediment lake

Figure 3 showed metals diagram of cluster analysis for mean of metals elements within the lake sediment from pointa S1, S2, S3, S4 and S5. Cluster analysis for mean elements could be grouped into three clusters or groups. Al, As and Mn have good similarity and are clustered in one group, Co, Cr, Fe, Zn and U are clustered into another group, while Th is individual forms and joined together with the other two group of metals. Strong similarity between Al with As and Mn showed relationship of these metals come from same origin. It can be seen if Al is taken as an indicator of detritus matter, both groups of element is from lithogenic input. However, strong similarity of Zn, Cr, Co and Fe also indicated strong anthropogenic input. Since As and Zn are pollution indicator, it might from fuel combustion during metals smelting or vehicle exhaust. The relation of U with both of two groups showed U possibly from natural and anthropogenic input such as in agriculture activities. Th showed less similarity but still correlated with the other elements.

IV. CONCLUSIONS

The study has found that most of the anthropogenic metals showed an almost regular pattern of depth profile concentrations. Higher concentrations were observed at relatively younger layers (about 10 years old) that that may be attributed to the impact of recent intensification of anthropogenic activities in the area. However natural weathering and erosion may also contributed to the recent accumulation. The presence of natural and anthropogenic

sources of heavy metals input was confirmed by the co-relational and cluster analysis of the metals.

V. ACKNOWLEDGMENT

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