

Investigation of Correlation Between Suspended Sediment Discharge and Activity Concentration of Natural Radionuclides in Langat River Basin

Wei-Koon Lee

School of Civil Engineering
College of Engineering

Universiti Teknologi MARA (UiTM) Shah Alam, Malaysia
leewei994@uitm.edu.my

Tuan Asmaa Tuan Resdi

School of Civil Engineering
College of Engineering

Universiti Teknologi MARA (UiTM) Shah Alam, Malaysia
asmaresdi@gmail.com

Abstract—Investigation of the correlation between natural radionuclide activity concentrations (RC) in river sediment and suspended sediment discharge (SD) is carried out for Langat river basin over a period of 4 years. Three river stations with suspended sediment discharge rate derived from validated sediment rating equation are paired with three radionuclide observation stations by geographical proximity. Long-term correlation of the quarterly RC observation with maximum SD is generally poor. However, for specific time window, the data trend correlation ranges from moderate to good, indicative of the influence of SD on the transport process of NORM within the river basin. Examination of the seasonal variation shows that Station 2 exhibits good correlation of RC and SD in the months of March and August/September, corresponding to the pre-storm seasons, with R^2 value as high as 0.9799. Despite the limitation of the undated RC observation and the use of hindcasted SD data, the present study shows the increase of radionuclide activity concentration of Ra-226, Ra-228 and K-40 in river sediment can be associated with the suspended sediment discharge rate and hence, the river streamflow especially during the pre-storm season.

Keywords— Correlation, Langat river, natural radionuclides, NORM, suspended sediment

I. INTRODUCTION

Naturally occurring radioactive material (NORM) which exists in almost all geological materials on earth includes radioactive nuclei produced by atmospheric cosmic ray interactions and ionising radiation from terrestrial radionuclides. Measurement of NORM is important to provide the background concentration level for long-term monitoring purpose. Example of radionuclides measured include Ra-226, Ra-228 and K-40 which are relatively stable with extremely long half-life. The three series contribute primarily to the Earth's radioactivity sources [1] and have been used as tracers in the study of erosion and accumulation process [2].

The natural radioactivity in river sediment is dependent on its formation and transport processes. Surface migration plays an important role in the dispersion and fate of these radionuclides. Soluble radionuclides can be transported easily by rainfall runoff into the fluvial network and thus dispersed in the basin and eventually ended up in the receiving water body [3]. The transport process is thus driven by soil loss events and varies seasonally with rainfall as well as anthropogenic activities such as land clearing in the river basin [4].

Langat river basin in Malaysia is strategically located next to the Klang Valley which is the most developed area in the country. The spill over effect of Klang Valley urban sprawl has caused increased development in the basin [5]. Basin soil loss and erosion has been attributed to the sediment discharge observed in Langat River and its tributaries [6]. Significant increasing trend in the streamflow of middle-Langat has been reported to be closely related to elevation, variation in landscape metrics, and the change in land use [7]. Based on a recent study by [8], average annual soil loss at the Langat watershed was 67 ton/ha with 11% of the basin area classified as high or very high risk.

According to [9], activity concentrations in middle Langat basin for K-40 shows long-term fluctuation whereas Ra-226 and Ra-228 show short-term fluctuation. [10] further pointed out that the increased activity concentrations in the down-river direction suggests a coupled catchment loss and river sediment relative gain. It can thus be hypothesized that the concentration of radionuclides in the river sediment can be correlated to the sediment discharge rate.

In this paper, our primary objective is to investigate the correlation between the suspended sediment discharge and the measured concentration of natural radionuclides in Langat River basin. The location is selected based on the availability of radionuclide concentration monitoring data by Malaysian Nuclear Agency. The sediment discharge data for the same time period is derived from the projection presented by [9] using validated sediment rating curve equation. For this purpose, streamflow and past sediment discharge data from Malaysian Drainage and Irrigation Department are used.

II. STUDY AREA

A. Langat River and Sub-basin

Langat river basin in Malaysia is an inter-state river basin situated mainly in the state of Selangor and partially in Negeri Sembilan within the latitude $2^{\circ} 40'N$ to $3^{\circ} 20'N$ and longitude $101^{\circ} 10'E$ to $102^{\circ} 00'E$. The basin also includes most of Putrajaya Federal Territory and a small fraction of Kuala Lumpur Federal Territory.

Langat River basin is the largest river basin in Selangor. The total catchment area is approximately 2,394 km². The Langat River which originates from Hulu Langat area is joined by three major tributaries. The sub-basins are: Semenyih River (319.7 km²) which flows from the northeast, Beranang River (305.8 km²) from the east and Labu River (247.5 km²) from the southeast (Fig. 1).

The basin can be sub-divided into three (3) distinct zones, namely, the mountainous zone located at the northeast region of Hulu Langat, the hilly area in the middle part of the basin and the flat alluvial plane located at the southwest near the river mouth which connects to the Straits of Malacca. The river sediment composition changes gradually from boulder/gravel in the mountainous zone, to sand and silt in the hilly areas, whereas most of flat alluvial plane area is characterized by peat with clay and silt soil.

B. Basin Land Use and Developments

According to [5], the built-up area in Langat River basin in year 1996 is about 23,018 km². It had increased by 40.70% over the period of 20 years to 119,587 km² in 2016. This indicates that the basin has been experiencing high development pressure over the period, with substantial increase in developed land area (from 7.85% to 40.7%), and significant reduction in agricultural area (60.22% to 25.7%). There remains 17.7% of forest land, and 11.2% of wetland and swamps. A small fraction of the basin area is used for quarry and mining activities. Other than that, the upper Sungai Langat area is utilised for recreational purposes.

C. Basin Climate

The study area has a warm tropical climate, with annual average minimum and maximum temperatures of 19°C and 33°C respectively. The annual variation in rainfall in the basin ranges between 1,792 to 2,757 mm (based on 40 years data from year 1950 to 1990) with an annual average of 2,250 mm [11].

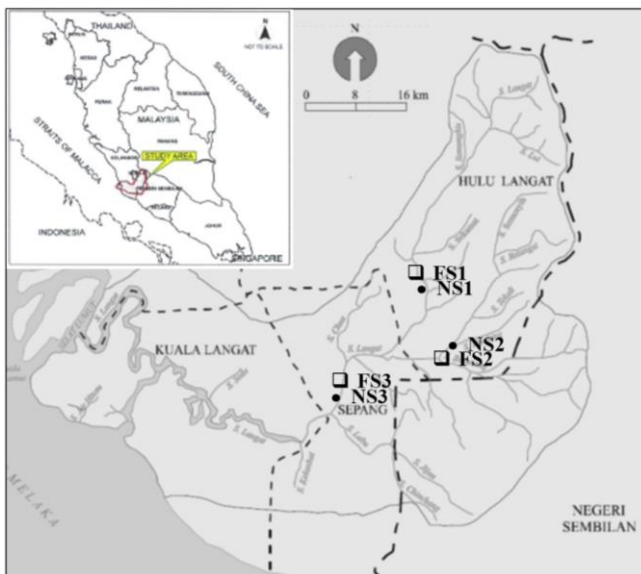


Fig. 1. Langat River basin showing locations of the data stations.

D. Basin River Water Quality

There are two dams on the upstream of Langat River basin, i.e. Langat Dam and Semenyih Dam. Water quality of the Langat River in the upstream falls under class II which indicates good water quality. However, the water quality drops to class III or IV at the lower reaches [6]. Study shows that the water quality is affected primarily by the presence of, amongst others, total suspended solids (TSS), which may be attributed to land clearing, agriculture, industries, sand mining, illegal dumping sites etc. in the wake of the rapid urban expansion.

III. METHODOLOGY

For this study, three streamflow (SF) stations and three radionuclide concentration (RC) monitoring stations are paired based on their geographical proximity and referred to as Station 1, 2 and 3 respectively (Tab. I).

TABLE I. SUSPENDED SEDIMENT AND RADIONUCLIDE ACTIVITY CONCENTRATION STATION PAIRING

	SF Station	LAT/LON	RC Station	LAT/LON
Station 1	FS1	02° 59' 40" N 101° 47' 10" E	NS1	2° 59' 33.3" N 101° 47' 8.78" E
Station 2	FS2	02° 54' 55" N 101° 49' 25" E	NS2	2° 54' 17.946" N 101° 48' 37.4688" E
Station 3	FS3	2° 59' 34" N 101° 47' 13" E	NS3	2° 51' 19.78" N 101° 40' 53.89" E

Malaysian Nuclear Agency (ANM) carry out quarterly observations of Ra-226, Ra-228 and K-40 at stations NS1 Kajang (2° 59' 33.3" N, 101° 47' 8.78" E), NS2 Rinching Hilir (2° 54' 17.946" N, 101° 48' 37.4688" E), and NS3 Dengkil (2° 51' 19.78" N, 101° 40' 53.89" E). For each station, river sediment samples were collected using auger. The prepared samples were tested on Ortec P-Type co-axial HPGe detector for radioactivity counting in the Radiochemistry and Environment Laboratory for a period of 15 hours to achieve equilibrium for U-238 and Th-232 with their respective progeny.

The radionuclide activity concentration data C is given in Bq/kg, where 1 becquerel (Bq) is defined as the activity of 1 kg of radioactive material in which one nucleus decays per second. Undated quarterly data is available from year 2014 in the month of March, May, September and December for year 2014-2015, and the month of March, May, August and November for year 2016-2018.

The 3 river streamflow stations are: FS1 Sg Langat at Kajang (2° 59' 40" N, 101° 47' 10" E), FS2 Sg Semenyih (2° 54' 55" N, 101° 49' 25" E), and FS3 Sg Langat at Dengkil (2° 59' 34" N, 101° 47' 13" E). These stations are operated and maintained by Department of Irrigation and Drainage Malaysia (DID), Malaysia. Suspended sediment data for the same station is however only available for the period 2006 to 2008.

Study of the available sediment rating curves for the three stations by [12] shows that the best correlation between suspended sediment concentration (SSC) (in ton/m³) and streamflow (SF) (in m³/s) is obtained if SF lead by one day. They conclude that log-transformed linear regression gives the best fit in terms of coefficient of determination (R²) with the lowest percent mean error.

Using the validated SCC-SF correlation derived in [9], the SF data from DID for year 2014 to 2018 is used to derive the SCC, and then the sediment discharge (SD) (in ton/day), where:

$$SD = SF \times SSC \quad (1)$$

Finally, the hindcasted SD data is analysed together with the RC data. Data correlation is investigated using the coefficient of determination (R²). In general, the value of R² approaches unity for strong correlation but approaches zero for poor correlation. Fig. 2 summarises the 2-part

methodology of the study. The present paper focuses on the second part involving the correlation of SD and RC. Further details on the derivation of the daily SD in the first part can be found in [12].

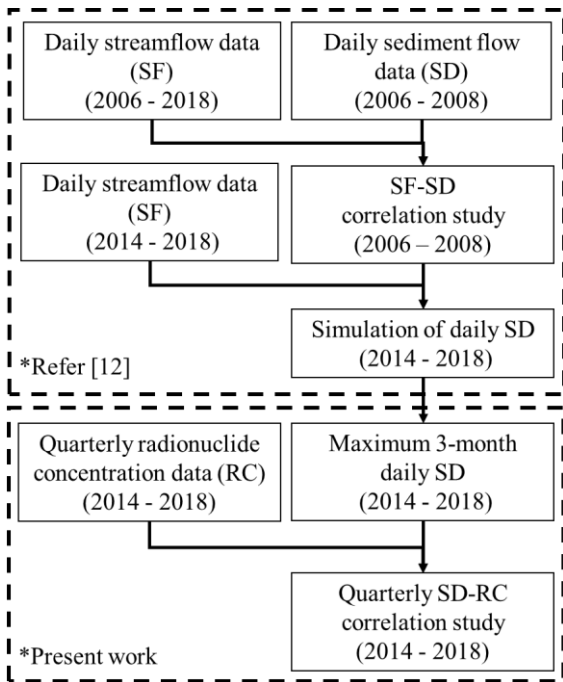


Fig. 2. Summary of methodology

IV. RESULTS AND DISCUSSIONS

A. Long-term Correlation

The average and the maximum daily sediment discharge derived from Eq. (1) within a 3-month time window before and after the date of radionuclide measurement is chosen for the correlation analysis. In general, the correlation of the

observed radionuclide activity concentration with the average quarterly suspended sediment discharge is very poor and is thus not further discussed here. Meanwhile, the correlation of the observed radionuclide activity concentration with the maximum suspended sediment discharge in the 3-month period shows marginally better correlation as summarised in Tab. II. This may be attributed to the fact that the occurrence of high sediment discharge event contributes to the accumulation of NORM from the basin source into the river system.

To further examine the correlation, we identify the specific time window where the fluctuations of RC and SD follow identical trend. Fig. 3 shows the plots of the activity concentration of Ra-226, Ra-228 and K-40 together with SD over the selected time periods. Tab. III summarises the time period and the R^2 value of the respective case.

For Station 1, RC for Ra-226 and RA-228 are correlated to SD over a very short time period of 5 months (December 2015 to May 2016) from the observation data. However, K-40 exhibit much stronger correlation ($R^2 = 9814$) from August 2017 to May 2018. Station 2 shows moderate correlation of the RC of all three radionuclides with SD from May 2014 to March 2016, which comprises eight data points over a 22-month period. For Station 3, correlation of K-40 to SD is much inferior to correlation of Ra-226 and Ra-228 to SD. The latter shows moderate R^2 values over a period of up to 27 months (August 2016 to November 2018).

TABLE II. LONG-TERM R^2 CORRELATION OF RADIONUCLIDE ACTIVITY CONCENTRATION TO SUSPENDED SEDIMENT DISCHARGE (2014-2018)

	<i>Ra-226</i>	<i>Ra-228</i>	<i>K-40</i>
<i>Station 1</i>	0.0655	0.0405	0.2313
<i>Station 2</i>	0.0608	0.0143	0.0398
<i>Station 3</i>	0.0226	0.0427	0.1219

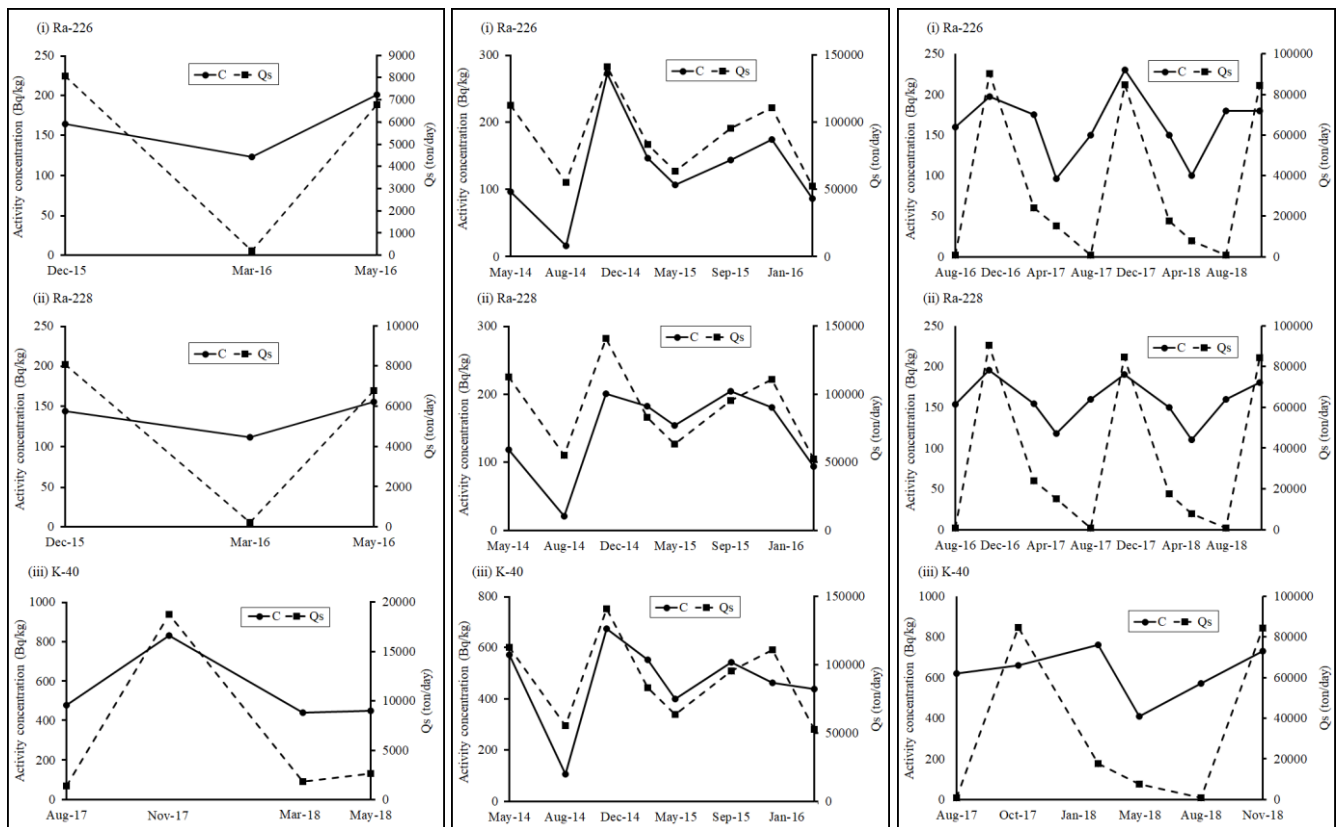


Fig. 3. Radionuclide activity concentration C and derived suspended sediment discharge Q_s for (from left to right): Station 1, 2 and 3

TABLE III. CORRELATION OF RADIONUCLIDE ACTIVITY CONCENTRATION TO SUSPENDED SEDIMENT DISCHARGE OVER SELECTED TIME WINDOW

Radionuclide	Station 1		Station 2		Station 3	
	Time window	R^2	Time window	R^2	Time window	R^2
Ra-226	Dec 2015 - May 2016	0.6341	May 2014 - Mar 2016	0.6796	Aug 2016 - Nov 2018	0.4260
Ra-228		0.8394		0.4230		0.5489
K-40	Aug 2017 - May 2018	0.9814		0.5636	Aug 2017 - Nov 2018	0.2372

Although the observed correlation between RC and SD is only limited in time window compared to the 4-year data period, the behaviour is suggestive of the influence of SD, and thus SF, on the transport process of NORM within the river basin. The quality of the input data certainly has a strong bearing on the above analysis. One of the limitations of the present investigation is the undated RC measurement where only the month of measurement is indicated. Besides, the SD data used is derived from hindcast due to unavailable field measurement.

B. Seasonal Correlation

For seasonal correlation, we consider the RC and SD at specific time period of the year. Tab. IV summarises the R^2

values for each of the station and the respective radionuclide. It is observed that both Station 1 and 2 shows moderate ($R^2 \sim 0.5$) to excellent correlation ($R^2 > 0.9$) in the month of March. For Station 2, reasonably good correlation ($R^2 \sim 0.7$) also occurs for data in the month of August/September. However, the R^2 value for Station 3 are generally below 0.6, suggesting lack of good correlation between the RC and SD for all the radionuclides.

Fig. 4 plots the RC and SD for the specific months with reasonably good correlation as identified from Tab. IV. From the plots, it can be seen that the 5 data points for RC and SD follow identical trend in the said month (or season) annually over the 4-year period.

TABLE IV. SEASONAL R^2 CORRELATION OF RADIONUCLIDE ACTIVITY CONCENTRATION TO SUSPENDED SEDIMENT DISCHARGE

Month	Station 1			Station 2			Station 3		
	Ra-226	Ra-228	K-40	Ra-226	Ra-228	K-40	Ra-226	Ra-228	K-40
Mar	0.5551	0.4108	0.1397	0.9752	0.5588	0.9799	-0.0154	-0.8439	0.0533
May	0.3918	-0.1471	0.0182	0.1277	0.0003	0.1617	-0.0071	0.0818	0.203
Aug/Sep	0.023	0.294	-0.0892	0.3942	0.7028	0.0652	0.5302	0.3784	-0.5484
Nov/Dec	-0.1961	-0.5298	0.5318	-0.0001	-0.0109	0.0875	-0.0016	-0.0165	0.1252

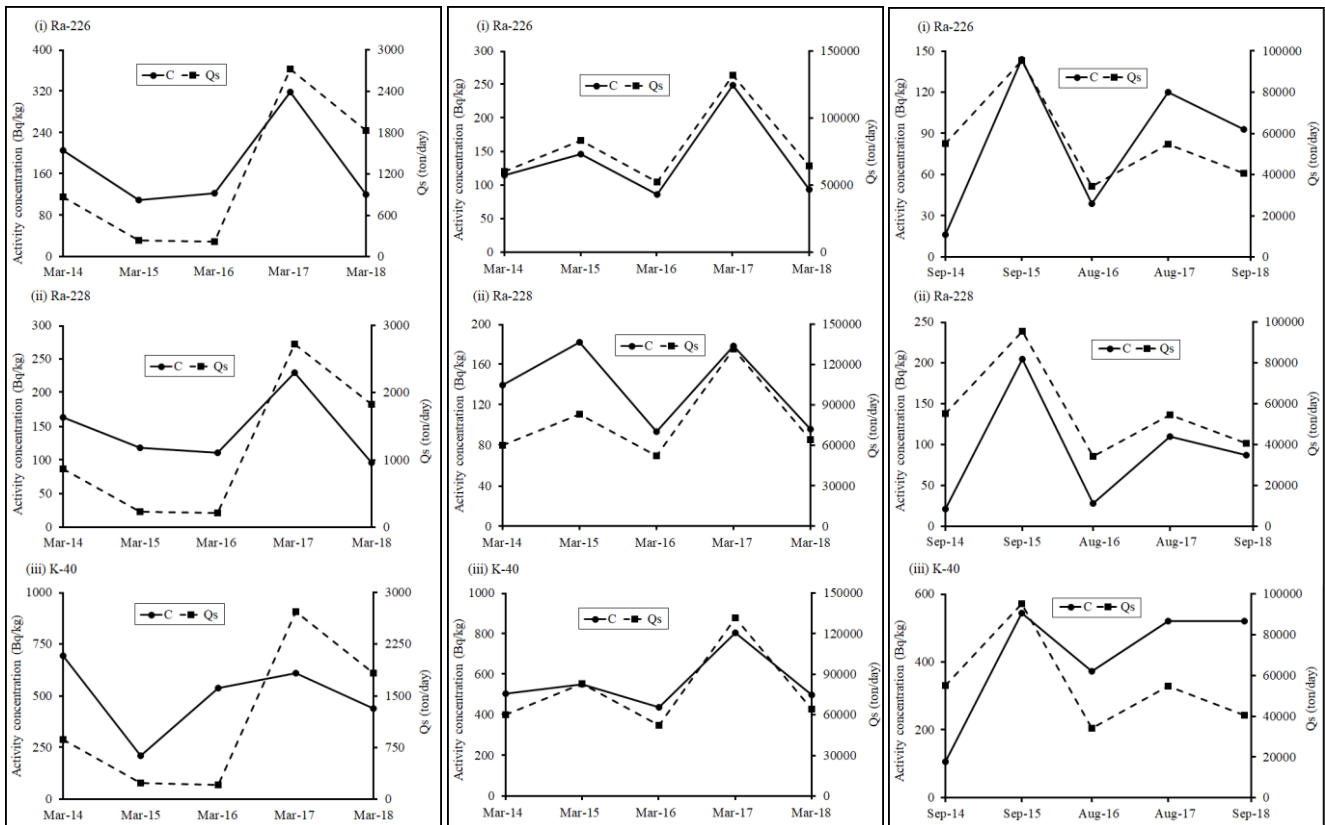


Fig. 4. Radionuclides activity concentration C and derived suspended sediment discharge Q_s for: Station 1 in the month of March (left), Station 2 in the month of March (middle) and August/September (right).

It is worth noting that Station 2 exhibits good correlation twice annually in both the months of March and August/September. Langat River basin which is located on the west coast of Peninsular Malaysia, is subjected to both the southwest monsoon (May to September) and northeast monsoon (November to February) annually. Fig. 5 shows the average monthly rainfall of the basin [13]. The month of March and August/September both coincides with the season before peak rainfall occurs, whereas the month of May and November/December is past the peak rainfall season in the basin. The good correlation prior to the stormy seasons may suggest that the gradually increasing basin runoff tend to carry more 'fresh' sediment containing NORM into the river network during these times which are well detected in field radionuclide measurements. In the post-storm seasons, the high streamflow rate may have caused dilution effect, hence less consistent observation of the radionuclides.

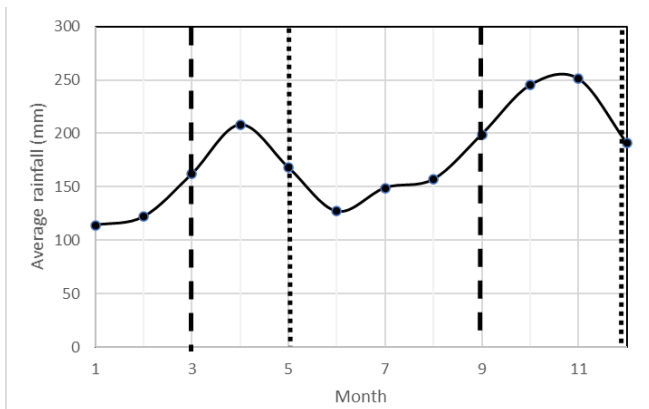


Fig. 5. Average monthly rainfall in Langat River basin, showing time of radionuclide measurements during pre-storm (dashed-line) and post-storm (dotted-line) seasons.

V. CONCLUSION

This paper presented an investigation of the correlation between radionuclide activity concentrations (RC) of Ra-226, Ra-228 and K-40 to the suspended sediment discharge (SD) in Langat River basin over a period of 4 years from 2014 to 2018. The missing SD data is hindcasted from validated sediment discharge rating curve in [12] using the available streamflow (SF) data. Four annual field measurement of RC is available for the months of March, May, August/September and November/ December. Long-term analysis of RC and SD shows very weak correlation, but which improves considerably in specific time window. The observation suggests that the RC and SD are closely related but the trend may be partly obscured due to slightly inferior data quality. Further examination shows that Station 2 shows good correlation of RC and SD in pre-storm seasons. We conclude that the activity concentration of NORM in river sediment can be expected to increase with streamflow runoff but may not be detected if dilution effect is strong.

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