Correlation of Natural Radioactivity in Surface Soil and River Sediment along the Middle Reach of Langat River

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Abstract. Concentration of natural radionuclides (Ra-226, Ra-228 and K-40) in samples of catchment surface soil and river sediment at three (3) monitoring stations along the middle reach of Langat River were identified using HPGe detector. Spatial and temporal variations of the radioactivity at 3-month interval were analysed over a period of 9 years. Activity concentrations show sign of long-term fluctuation for K-40, and short-term fluctuation for Ra-226 and Ra-228. Correlation of Ra-226 and Ra-228 is very strong for downstream stations, but weak for the upstream station. Meanwhile, their correlation with K-40 varies considerably possibly due to the high potassium solubility.

1. Introduction
Naturally occurring radioactive material (NORM) in the atmosphere and the earth surface includes radioactive nuclei produced by atmospheric cosmic ray interactions and ionising radiation from terrestrial radionuclides, which exists in almost all geological materials on earth. Over 85% of human radiation exposure [1] can be attributed to the ubiquitous occurrence of these natural radiations in the environment. Despite their low concentration, long-term human exposure through ingestion, inhalation or external contact remains a potential health threat.

Natural radioactivity in soil and sediment are dependent on their formation and transport processes, which is influenced by the chemical and biochemical interactions of the decay products of the uranium and thorium series [2]. Radionuclides may exist in dissolved form, adsorbed form, coprecipitated form, bound with organic matter or in crystal lattices of primary minerals [3]. The extent of sediment sorption of radionuclides depends on both the type and concentration of the radionuclide as well as the surface characteristics of the sediment, contact time and ambient temperature [4]. Their physiochemical properties further dictate the rate of surface migration. Radionuclides in sorption and in dissolved forms is easily transported by surface runoff and river flow, and thus is migrated and dispersed to the downstream of the fluvial network and ultimately coastal water in the estuary. An examination of the long-term spatial and temporal detection of NORM within a catchment and the river system thus allow the distribution system and mechanism to be analysed.

Ra-226 and Ra-228 are the relatively stable progeny from the uranium and thorium decay series respectively. K-40 meanwhile, is a naturally occurring isotope from primodial origin. It has an extremely long half-life and undergoes beta decay to produce either argon or calcium. Together, the three series contribute to the Earth’s radioactivity sources. The main objective of this study is to evaluate the spatial and temporal variation of Ra-226, Ra-228 and K-40 in catchment soil and river sediment along the middle reach of Langat River over a 9-year monitoring period.
2. Study Area

Langat river basin is situated at the boundary of the states of Selangor and Negeri Sembilan within the latitude 2° 40’N to 3° 20’N and longitude 101° 10’E to 102° 00’, with a total catchment area of approximately 2,394 km². The major tributaries in the basin are Semenyih River and Labu River, which converges to Langat River (Figure 1). The basin land use is primarily agricultural (55.13%), followed by forest (19.31%), wetland and swamps (12.73%), urban built-up areas (6.20%), mining (1.61%) and other activities (5.02%). For the area within 1 km distance from the main river, nearly half (47%) of the land use is dominated by cash crop (e.g. rubber plantation and palm oil), 17% is municipal and residential, and 10% is mixed plantation (e.g. orchards, banana, coconuts etc). [5].

Natural radionuclide activity of Ra-226, Ra-228 and K-40 in samples taken from the catchment surface soil and river sediment have been monitored regularly on quarterly basis annually since year 2010 under the program by Malaysian Nuclear Agency (ANM). For this study, three (3) stations located along the western branch of the middle Langat River, from upstream to downstream, namely S5 Kajang (02° 59’ 33.3” N 101° 47’ 8.78” E), S1 Teras Jernang (02° 55’ 13.79” N 101° 45’ 40.77” E), and S4 Dengkil (02° 51’ 19.78” N 101° 40’ 53.89” E), are selected. In addition, another station S3 Sungai Buah (02° 53’ 28.11” N 101° 45’ 21.89” E) in the tributary Semenyih river which meets the main river downstream of S1 but upstream of S4, is also included for cross comparison. The river reach considered is on the western branch of the confluence. A similar study on the eastern branch of the middle Langat River which comprises three (3) stations which ends with S3 has been reported [6].

![Legend](image)

**Legend**
- S5 Kajang Station
- S1 Teras Jernang Station
- S3 Sungai Buah Station
- S4 Dengkil Station

![Map](image)

**Figure 1.** Sampling stations in middle Langat River.
3. Method
Augur sampling of surface soil and river sediment is carried out at 3-month interval every year from the stations. Each sample weighing 1 kg in mass was first air-dried and then oven-dried at 105°C for up to 3 days to remove all water content. Next, the dried samples were crushed into powder and sieved with 2-mm mesh to produce a homogenized sample. The prepared samples were weighted and sealed with standard electric tape in Marinelli beaker to avoid contact with the air and stored for a minimum period of 21 days. The samples were then 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 final reading of natural radionuclides Ra-226, Ra-228 and K-40 were then correlated and analysed for spatial and temporal variations between the three (3) stations over the monitoring period.

4. Result and Discussion
Figure 2 shows the activity concentration of Ra-226, Ra-228 and K-40 in the catchment surface soil samples (Figure 2a) and river sediment samples (Figure 2b) at each of the four (4) stations monitored over the period from year 2010 to 2018. In Figure 2a, it is observed that there is a marked decrease in Ra-226 concentration moving in the downstream direction from S5 to S1, to S4 from year 2014 onwards. In general, Ra-226 activity concentration peaks in year 2012 and 2017 but drops in year 2015. For river sediment samples, the activity concentration shows no specific trend over time window investigated. Concentrations in the river sediments are generally higher than that observed in the surface soil for Ra-226 and Ra-228, which is more clearly depicted in Figure 3. Furthermore, K-40 is found to have much higher activity concentration compared to Ra-226 and Ra-228 combined.

Figure 3 shows the mean activity concentration of Ra-226, Ra-228 and K-40 in catchment surface soil and river sediment samples, averaged over all the three (3) stations considered from year 2010 to 2018. Our previous investigation of the eastern branch of middle Langat River shows sign of periodic oscillation of K-40 activity concentration in catchment soil samples over a 6-year period. Similar observation can be made here on the western middle Langat River where the concentration peak is recorded in year 2012 and 2017, and the lowest in year 2015. K-40 activity concentration in river sediment also shows identical behaviour with the eastern branch where the highest value is observed in year 2010, and the lowest in year 2015 [6]. Meanwhile, the mean activity concentration of Ra-226 and Ra-228 also show oscillatory trend at much shorter time period. The cyclic pattern observed above needs to be further verified using sufficient long-term data.

Next, the correlation between the radionuclide concentrations in catchment surface soil samples (Table 1) and river sediment (Table 2) samples are examined. At the upstream-most station S5, the correlations are generally very poor except the correlation between Ra-226 and K-40 in the catchment soil sample which is over 0.68 in all three stations. At station S1, high correlation between all the three radioactive elements can be observed. In the downstream-most station S4, good correlation above 0.6 can be observed for all elements in the catchment soil sample, but only between Ra-226 and Ra228 in the river sediment sample (see also Figure 4). K-40 concentrations might not be related to the presence of Ra-226 and Ra-228 due to the high potassium solubility [7-8]. Nevertheless, the present data shows that reasonably good correlation between K-40 and radium in the range of 0.60 to 0.84 can be observed. Overall, Ra-226 and Ra-228 shows strong correlation except at station S5 at the upper catchment region, suggesting difference in the activity ratio of the uranium decay series and the thorium decay series, which are their respective source element.

Table 3 and 4 summarise the mean and range of the ratios of the activity concentrations by station for the different natural radionuclide-pairs in the catchment soil sample and river sediment, respectively. The mean ratio of Ra-226/Ra-228 is most closely related at around 1 in all cases. For Ra-226/K-40 and Ra-228/K-40, the ratios averaged between 0.183 to 0.306. The ratios of Ra-226/Ra-228 reduces for both catchment soils sample and river sediment in the downstream direction.

Figure 5 shows the frequency distribution of the ratios of the different elements in catchment soil and river sediment sample. The ratio of Ra-226/Ra-228 peaks at 0.93 in catchment sample, and 1.23 in river sediment, where the former shows a larger spread from 0.66 to 1.38. Meanwhile, the ratios of Ra-226/K-40 and Ra-228/K-40 show considerable fluctuation between 0.09 to 0.39.
a. Surface soil sample  
b. River sediment sample

**Figure 2.** Activity concentration of Ra-226, Ra-228 and K-40 in middle Langat River.

**Table 1.** The correlation between radionuclides in surface soil sample.

| Radionuclide | S5   | S1   | S4   |
|--------------|------|------|------|
| Ra-226       | 1    | 0.087| 0.839|
| Ra-228       | 1    | 0.133| 1    |
| K-40         | 1    | 1    |      |

**Table 2.** The correlation between radionuclides in river sediment sample.

| Radionuclide | S5   | S1   | S4   |
|--------------|------|------|------|
| Ra-226       | 1    | 0.112| -0.309|
| Ra-228       | 1    | 0.559| 1    |
| K-40         | 1    | 1    |      |
Figure 3. The mean activity concentration of radionuclides in surface soil and river sediment sample.
Figure 4. The correlation between radionuclides in Station S4.

Table 3. Ratio of radionuclides in surface soil sample.

| Stations | Ratio       | S5 | S1            | S4             |       |
|----------|-------------|----|---------------|----------------|-------|
|          | Mean | Range  | Mean | Range        | Mean | Range  |
| Ra-226/ Ra-228 | 1.098 | 0.723 - 1.422 | 0.979 | 0.898 - 1.091 | 0.890 | 0.661 - 1.111 |
| Ra-226/ K-40   | 0.234 | 0.193 - 0.297 | 0.183 | 0.153 - 0.297 | 0.184 | 0.090 - 0.258 |
| Ra-228/ K-40   | 0.223 | 0.169 - 0.370 | 0.187 | 0.169 - 0.370 | 0.205 | 0.136 - 0.299 |

Table 4. Ratio of radionuclides in river sediment sample.

| Stations | Ratio       | S5 | S1            | S4             |       |
|----------|-------------|----|---------------|----------------|-------|
|          | Mean | Range  | Mean | Range        | Mean | Range  |
| Ra-226/ Ra-228 | 1.170 | 0.939 - 1.406 | 1.099 | 0.977 - 1.235 | 1.000 | 0.955 - 1.111 |
| Ra-226/ K-40   | 0.306 | 0.241 - 0.395 | 0.260 | 0.218 - 0.337 | 0.267 | 0.244 - 0.319 |
| Ra-228/ K-40   | 0.261 | 0.221 - 0.293 | 0.237 | 0.207 - 0.273 | 0.268 | 0.240 - 0.329 |
5. Conclusion
In this study, the temporal and spatial trends in the observed activity concentration of three (3) natural radionuclides, namely K-40, Ra-226 and Ra-228 in catchment soil samples and river sediment samples at three (3) stations along Langat River in Middle Langat River Basin have been reported. The data shows indication of long-term periodic fluctuation especially for K-40. Radioactivity correlation between activity concentration in catchment soil and river sediment sample for the same elements is very poor and omitted herein. Meanwhile, correlation between Ra-226 and Ra-228 is the strongest for Station S1 and S4 but weak for Station S5, whereas correlation between K-40 and radium range substantially from excellent (S1 and S4) to very poor (S5). Activity concentration ratios show that K-40 is up to 4 times higher than both Ra-226 and Ra-228, which have almost identical values. Moving in the downstream direction, the ratios of Ra-226/ Ra-228 tend to reduce for catchment soils sample. Overall, high-quality long-term data is required to further investigate the spatial and temporal variations of the activity concentrations of the above radionuclides.

Figure 5. Frequency distribution of Ra-226/Ra-228, Ra-226/K-40 and Ra-228/K-40 activity concentration ratios in middle Langat River.

a. Surface soil sample  
b. River sediment sample
6. References

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