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POTENTIAL USE OF FALLOUT RADIONUCLIDES (^{137}Cs AND ^{210}Pb$_{ex}$) IN ASSESSING SOIL EROSION RATES WITHIN THE LANGAT WATERSHED

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ABSTRACT

Soil erosion is a severe environmental problem arising due to human intervention such as deforestation, urbanization, rapid land use change and overgrazing. Langat watershed is highly risk in erosion potential as it is exposed to land clearing activities due to population growth. The objective of this study is to assess the potential use of ^{137}Cs and excess ^{210}Pb in soil erosion assessment within the Langat watershed. A total of 15 individual sectioned soil cores were collected along the upstream to the downstream of Langat watershed. The net erosion rate based on ^{137}Cs measurement ranged between -8 to -66 t ha^{-1} yr^{-1} with an average of -33 t ha^{-1} yr^{-1}, whereas, based on ^{210}Pb$_{ex}$, the net erosion rate ranged between -5 to -50 t ha^{-1} yr^{-1} with an average of -28 t ha^{-1} yr^{-1}. The sediment delivery ratio estimated by both radionuclides were above 90% indicating that most of the sediment transported out of the watershed. Fallout radionuclides (FRN) method is proven to be a significant alternative to overcome constraints and limitations encountered in conventional approach. Thus, FRN method appeared to be an essential and effective alternative in soil erosion assessment.

Keywords: *^{137}Cs, ^{210}Pb$_{ex}$, fallout radionuclide, Langat, soil redistribution*

INTRODUCTION

Over recent years, there has been a growing concern over water-induced soil erosion and its associated environmental impacts. There is an increased demand to obtain reliable information on soil erosion rates and sediment deposition. Nevertheless, such information is difficult to acquire when using conventional techniques due to validation, accuracy, time-consuming and cost concerns (FAO, 2019a, 2019b). Thus, the potential for using fallout radionuclides have been explored widely in various region and scale (Chaboche et al., 2021; Moustakim et al., 2019; Porto et al., 2018).

Fallout radionuclides, namely, caesium-137 (^{137}Cs) is a man-made radionuclide (half-life = 30.2 ± 0.2 years) and lead-210 (^{210}Pb) (half-life = 22.20 ± 0.22 years) is a naturally occurring radionuclide, are widely used as environmental tracers to study soil redistribution (He and Walling, 1996; Walling, 1999). Soil redistribution consist of soil erosion, transportation and sediment deposition imposed a serious environmental problem globally. ^{137}Cs is originated from nuclear power plant, weapon test or nuclear power plant accident such as Chernobyl in Russia and Fukushima in Japan (Panin et al., 2001; Tagami et al., 2019).

The application of ^{210}Pb is still limited and needs more investigation and validation, even though it has been used extensively in sediment dating. Lead-210 is often used as a complementary radionuclide besides ^{137}Cs due to the low concentration of ^{137}Cs. ^{210}Pb is a natural product of the decay series of uranium-238 (^{238}U). It is derived from radon gas decay, known as the daughter of ^{226}Ra (half-life = 1600 ± 7 years). The diffusion of radon gas from soil, which produces ^{210}Pb into the atmosphere and, subsequently, its fallout provides a continuous input of radionuclides to the soil surface and sediment. The fallout radionuclide is identified as "unsupported" or "excess" ^{210}Pb. Due to its natural existence, ^{210}Pb is often measured as a complimentary radionuclide to ^{137}Cs in measuring soil erosion. Both radionuclides (^{137}Cs and $^{210}Pb_{ex}$) reach the soil surface as fallout and are rapidly absorbed by soil particles. As they clamped onto the soil surface, they acted as a tracer when soil redistribution was controlled by deposition, erosion and transportation processes (Zheng et al., 2007). ^{137}Cs and $^{210}Pb_{ex}$ will provide information on medium-term (40 years) and long-term (100 years) erosion rates and patterns, respectively.

The objective of this paper is to assess the potential use of fallout radionuclides in assessing soil erosion rates within the Langat watershed. Rapid changes in land use activities in the Langat watershed vicinity would induce significant soil redistribution. Therefore, an erosion study was needed to explore the rates and patterns of soil erosion within a larger watershed based on a tropical climate. The findings of this study will provide an understanding of the erosion system and scientific basis to implement an effective countermeasure for the increasing erosion potential.

MATERIALS AND METHODS

Study Area

This study was conducted at the Langat watershed with a catchment area of 2287 km^2 (Figure 1). Langat River originated from Mount Nuang, Titiwangsa Range with a total length of 141 km (LUAS, 2011, 2015) and flows through three states namely Selangor, the Federal Territory of Putrajaya and Negeri Sembilan, towards the Straits of Malacca near Banting town, Kuala Langat. The Langat watershed has a tropical climate with mean annual temperature varies from 23°C to 31°C (based on years 1979-2014). Maximum rainfall normally occurs in November and minimum rainfall occurs in February with an annual precipitation at 2061 mm (DID, 2010).

The main tributaries include Semenyih River, Beranang River and Labu River with two important dams, namely, Langat dam and Semenyih dam. The Langat watershed is regarded as one of the most critical water catchment areas providing domestic and industrial water to the population within the Langat River.

In this study, the sampling area was divided into fifteen stations along the Langat watershed as shown in Figure 1 and the description of each sampling station is given in Table 1. Sampling activities included a field survey (coordinate and land use) were conducted from February to July 2019 (upstream, middle stream and downstream).

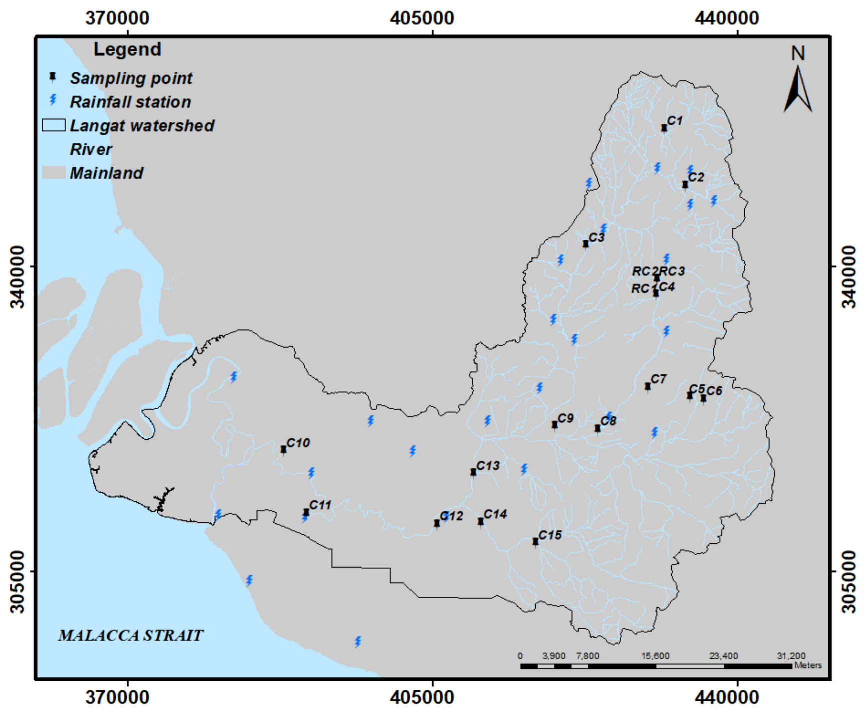

Figure 1: Sampling location along the Langat watershed

Table 1: Area description of sampling station

Stations	Area description
Reference point (RC1, RC2 and RC3)	Forestry area near Forest Reserve Ulu Langat, Forest Reserve Gunung Nuang and Forest Reserve Sungai Tekala
C1	Forest area near Pangsun River
C2	Fruit farm near Lui River
C3	Forest area
C4	Palm oil plantation near main road
C5	Rubber plantation near Broga road
C6	Cleared land near hill side
C7	Grassland near residential area
C8	Rubber plantation near Rinching
C9	Palm oil plantation near main road
C10	Palm oil plantation near Langat River
C11	Bushes near Langat Riverbank
C12	Palm oil plantation near paper factory
C13	Bushes near Langat Riverbank
C14	Sandy soil near Hartalega factory
C15	Palm oil plantation between Batang Nilai River and Labu River

Sampling Strategy

Soil erosion rates were estimated using ^{137}C and $^{210}Pb_{ex}$ are based on the comparison between individual and reference inventory obtained from a representative local fallout (Mabit et al., 2008). In this study, a relatively undisturbed, flat, stable site, and at the highest altitude is considered to minimize soil erosion (Khodadadi et al., 2021). Three stations were considered as reference points namely, Forest Reserve Ulu Langat, Forest Reserve Gunung Nuang and Forest Reserve Sungai Tekala (Table 1). The reference site and individual points should receive the same amount of precipitation and have similar geomorphological parameters (Nouira et al., 2003).

At each station, a stainless-steel soil corer with 40 cm long and 10.5 cm diameter was used to collect segmented core at 2 cm increments. The maximum depth is limited to 30 cm depth due to stony soil at certain sampling points. The location of the study area which included latitude, longitude and elevation were determined by a Global Positioning System (GPS).

A total of 15 individual soil cores were collected within the Langat watershed following the same procedure as the reference point. The individual cores were collected based on different types of land use. There were two core samples collected from secondary forest, five from palm oil plantation, two from rubber plantation, three from bushes/riverbank, one each from fruit farm, cleared land and grassland, respectively.

Sample Preparation and Analyses

Soil samples were oven dried at 60ºC, disaggregated, and passed through a 2 mm sieve. Particle size analysis was carried out by using a particle size analyser (Model: Microtrac-X100 from USA). Besides, organic matter and soil texture were estimated using standard soil analysis method (Miyazawa et al., 2000). After sample pre-preparation, a readily weighted sample was packed into a 6 ml cylindrical container or in 30 ml plastic container and sealed for at least 21 days (International Atomic Energy Agency, 2014) to establish the secular equilibrium between ^{226}Ra and ^{222}Rn (half-life = 3.8235 ± 0.0003 days).

Measurement of ^{137}Cs and unsupported ^{210}Pb activities were undertaken simultaneously by high-purity germanium (HPGe) gamma spectrometry system (Model: Canberra from USA). The sample was counted for over 50 000s with an analytical precision at ± 10% at the 95% level of confidence. The detector is a closed end coaxial well-detector operated on 2000 HV bias supply. The p-type detector with the FWHM resolution of 0.82 keV at 122 keV gamma line of ^{57}Co and 1.85 keV at 1.3 MeV gamma line of ^{60}Co, with the relative efficiency of 25% (Yii and Wan Mahmood, 2020). The activity of ^{137}Cs in the soil sample was obtained from the count rate at 662 keV peak energy. The unsupported ^{210}Pb activity concentration in the sample was calculated from difference between the total ^{210}Pb (46.5 keV peak energy) and the ^{226}Ra (measured via its daughter ^{214}Bi, 609.3 keV peak energy) (Montes et al., 2019; Rabesiranana et al., 2016). Activity concentration for each radionuclide was estimated using the equation below:

$$A = \frac{\left[\left(\frac{C_1}{T}\right)-\left(\frac{B}{T_b}\right)\right]}{E.w.\gamma} \qquad \text{Eq..(1)}$$

Where, A is the radionuclide activity concentration (Bq kg^{-1}), C_1 is the sample peak area (count), T is the sample counting time (s), B is the peak area for sample background (count), T_b is the background counting time (s), E is the efficiency of the equipment (%), w is the sample weight (g) and γ is the emission probability (%).

Gamma spectrometry detector was calibrated using standard multi-nuclide standard source purchased from Isotope Products Laboratories, USA (Yii and Wan Mahmood, 2020). The minimum detectable activity (MDA) was estimated to be 5 Bq kg^{-1} for total ^{210}Pb, 1 Bq kg^{-1} for ^{137}Cs and 1 Bq kg^{-1} for ^{226}Ra.

Estimation of Erosion Rates

Soil erosion rate was estimated based on the comparison between inventory at a specific point to the reference point. The radionuclide inventory was estimated using the equation below:

$$I_a = \sum A_i \rho_i H_i \qquad \text{Eq...(2)}$$

Where, I_a is the radionuclide inventory, A_i is the sample activity of the i^{th} sample at depth increment (Bq kg^{-1}), ρ_i is the bulk density of i^{th} sample (kg m^{-3}) and H_i is the depth of the i^{th} sample at depth increment (m). The radionuclide inventory was denoted by unit Bq m^{-2}.

The conversion model developed by Walling et al. (2001) was applied to estimate soil erosion rates for the sampling sites. Difference conversion models were used for ^{137}Cs and ^{210}Pb$_{ex}$ measurement which are proportional model and mass balance model 2 (MBM2), respectively. A software package based on an Excel Add-in is available to convert the radionuclide inventory to soil redistribution rates (International Atomic Energy Agency, 2014).

Based on the study by Porto et al. (2014), the ^{137}Cs fallout input is assumed to be completely mixed within the cultivated layer and the soil reduction is directly proportionate to the decline of ^{137}Cs in the soil profile. The average annual soil loss rate, Y (t ha^{-1} yr^{-1}) can be given as (International Atomic Energy Agency, 2014) :

$$Y = 10 \, \frac{BdX}{100TP} \qquad \text{Eq....(3)}$$

Where, X is the percentage decline in total ^{137}Cs inventory (given as A_{ref} – A/A_{ref} x 100), d is the ploughing depth (m), T is the time elapsed since the initiation of ^{137}Cs accumulation (year) which is assumed at 1963, B is the bulk density of the soil (kg m^{-3}), P is the particle size correction factor, A_{ref} is the local ^{137}Cs reference inventory (Bq m^{-2}), and A is the total measured inventory at the individual sampling point (Bq m^{-2}).

As for ^{210}Pb$_{ex}$, the MBM2 can be expressed as Walling et al. (2001):

$$\frac{d(A)}{dt} = (1-\Gamma)I(t) - (\lambda + (\frac{PR}{m}))A(t) \qquad \text{Eq….(4)}$$

Where, A(t) is the ^{210}Pb$_{ex}$ inventories (Bq m^{-2}); R is the soil erosion rate (kg m^{-2} year); t is the time since the onset of ^{210}Pb$_{ex}$ fallout (year); I(t) is the annual deposition flux at the time t (Bq m^{-2} year); m is the cumulative mass depth representing the average plough depth (kg m^{-2}); λ is the decay constant for ^{210}Pb$_{ex}$ (year^{-1}); P is the particle size factor; Γ is the proportion of the freshly deposited ^{210}Pb$_{ex}$ removed by erosion before mixing into the plough layer; Γ is expressed as $\Gamma = p\,\gamma\,(1-e^{-R/H})$ where γ is the proportion of the annual ^{210}Pb$_{ex}$ input susceptible to removal by erosion, and H is the relaxation mass depth of the initial distribution of fallout $^{210}Pb_{ex}$ in the soil profile (Rabesiranana et al., 2016; Walling et al., 2001). The soil erosion rates based on ^{137}Cs and $^{210}Pb_{ex}$ measurement were spatially mapped using the spatial analyst tool in ArcGIS ver. 10.3.

RESULTS AND DISCUSSION

The mean inventories of ^{137}Cs and $^{210}Pb_{ex}$ for three segmented soil cores from the reference site were estimated. The mean reference inventories for ^{137}Cs and $^{210}Pb_{ex}$ were estimated at 160 ± 5 Bq m^{-2} and 2040 ± 5 Bq m^{-2}, respectively. The mean reference inventory for ^{137}Cs is similar to the inventory estimated by Gharibreza et al. (2013a), however, it is five times lower than study by Zainudin and Wan Ruslan (2012). This is proven that the spatial variability in fallout deposition occurred in some areas (Yang et al., 2011).

Unlike ^{137}Cs, limited reference inventories or fallout flux of $^{210}Pb_{ex}$ have been reported in Malaysia. However, it has been documented that $^{210}Pb_{ex}$ was successfully estimated in Lake Bera, Malaysia (Gharibreza et al., 2013b), Zambia (Walling et al., 2003) and Spain (Gaspar et al., 2017). The global annual deposition for $^{210}Pb_{ex}$ reference inventory was reported in the range of 766 to 12, 233 Bq m^{-2}. The mean reference inventory (2040 ± 5 Bq m^{-2}) reported in this study is in the range of global deposition.

The individual soil cores were divided into fifteen corers numbered from C1 to C15 according to their location from upstream to downstream of the Langat watershed. The concentrations of ^{137}Cs and $^{210}Pb_{ex}$ in cultivated soils were relatively uniform throughout the soil profile. These individual soil cores had experienced net erosion due to rapid land use change since 1970s (Rapport et al., 2002).

The mean of inventories estimated from fifteen individual points for both radionuclides were lower than the reference inventory, thus, it is observed that erosion has occurred at the study area (Khodadadi et al., 2021). The mean of inventories for ^{137}Cs measured for 15 individual cores collected ranged between 43 ± 6 Bq m^{-2} to 139 ± 19 Bq m^{-2} with an average of 88 ± 13 Bq m^{-2}. On the other hand, the mean inventories for $^{210}Pb_{ex}$ ranged between 429 ± 13 Bq m^{-2} to 2312 ± 26 Bq m^{-2} with an average of 1080 ± 21 Bq m^{-2}.

The net erosion rates calculated using Eq. (3) ranged from -8 to -66 t ha^{-1} yr^{-1} with an average of -33 t ha^{-1} yr^{-1} based on ^{137}Cs measurement. The average sediment delivery ratio based on ^{137}Cs measurement is 95%. Moreover, based on $^{210}Pb_{ex}$ measurement, the net erosion rates calculated using Eq. (4) was ranged from -5 to -50 t ha^{-1} yr^{-1} with an average of -28 t ha^{-1} yr^{-1}. The average sediment delivery ratio estimated based on $^{210}Pb_{ex}$ is 90%. A negative signed indicated the soil loss at the Langat watershed.

The spatial distribution of ^{137}Cs inventories in the Langat watershed was highly variable (Figure 2). In general, the distribution of ^{137}Cs inventories presented a contrast between the upstream to the downstream of the watershed. The highest ^{137}Cs inventories were found at the Labu sub-basin and some of area at the Banting town. In contrast, lower ^{137}Cs inventory was recorded on the middle stream near Semenyih, Buah and Dengkil sub-basin. Subsequently, the spatial distribution of soil erosion rates contrasts with the distribution pattern of the ^{137}Cs inventories. It was noted that, the highest erosion rate was found in the middle stream near Semenyih, Buah and Dengkil sub-basin, whereas the lowest erosion rates occurred near Labu sub-basin towards Banting town (Figure 3).

Based on $^{210}Pb_{ex}$ measurement, the highest inventory was found at the northern part of the Langat watershed. The northern part of Langat watershed was surrounded by the steep slope and mountainous area. The dense forest on the steep slope protects the soil surface from soil erosion, thus, the higher inventory of $^{210}Pb_{ex}$ were found (Gaspar et al., 2013). The lowest inventories were noted from the middle stream towards the downstream to the Malacca Strait (Figure 4). Similar to the ^{137}Cs measurement, the spatial distribution of soil erosion rates was slightly different than the inventories. The highest erosion rate occurred from the middle stream towards the Malacca Strait. On the other hand, the lowest erosion risk is located to the north of the Langat watershed (Figure 5).

The spatial distribution for $^{210}Pb_{ex}$ inventories has a different pattern than ^{137}Cs inventories in the Langat watershed. $^{210}Pb_{ex}$ is more sensitive to recent changes in soil erosion rates compared to ^{137}Cs measurements due to its constant fallout inputs and the shorter half-life of ^{210}Pb (Porto et al., 2016; Walling et al., 2003).

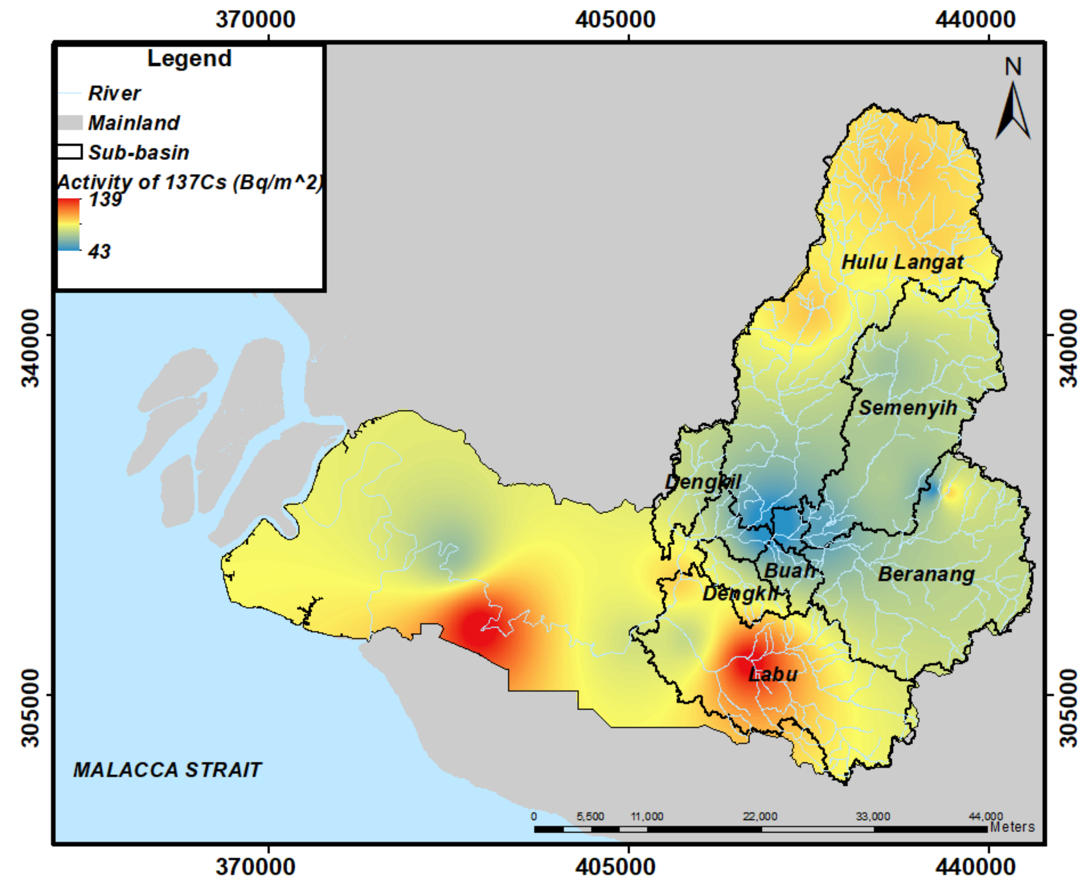

Figure 2: Spatial distribution of ^{137}Cs inventories in Langat watershed

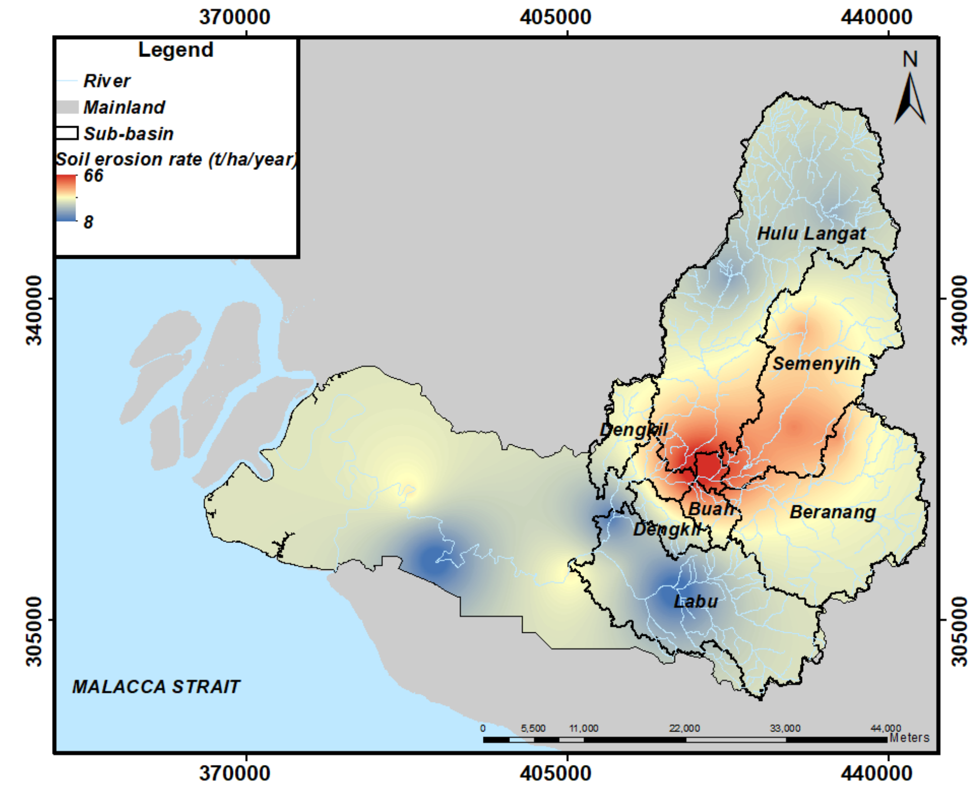

Figure 3: Spatial distribution of estimated net erosion rates by ^{137}Cs

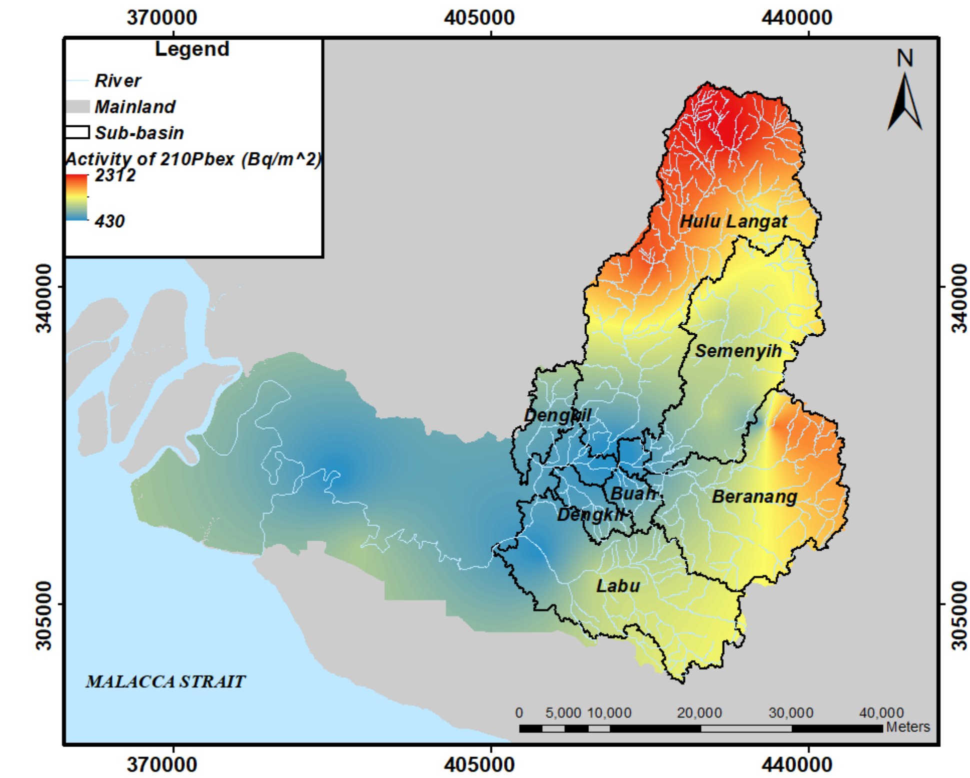

Figure 4: Spatial distribution of $^{210}Pb_{ex}$ inventories in Langat watershed

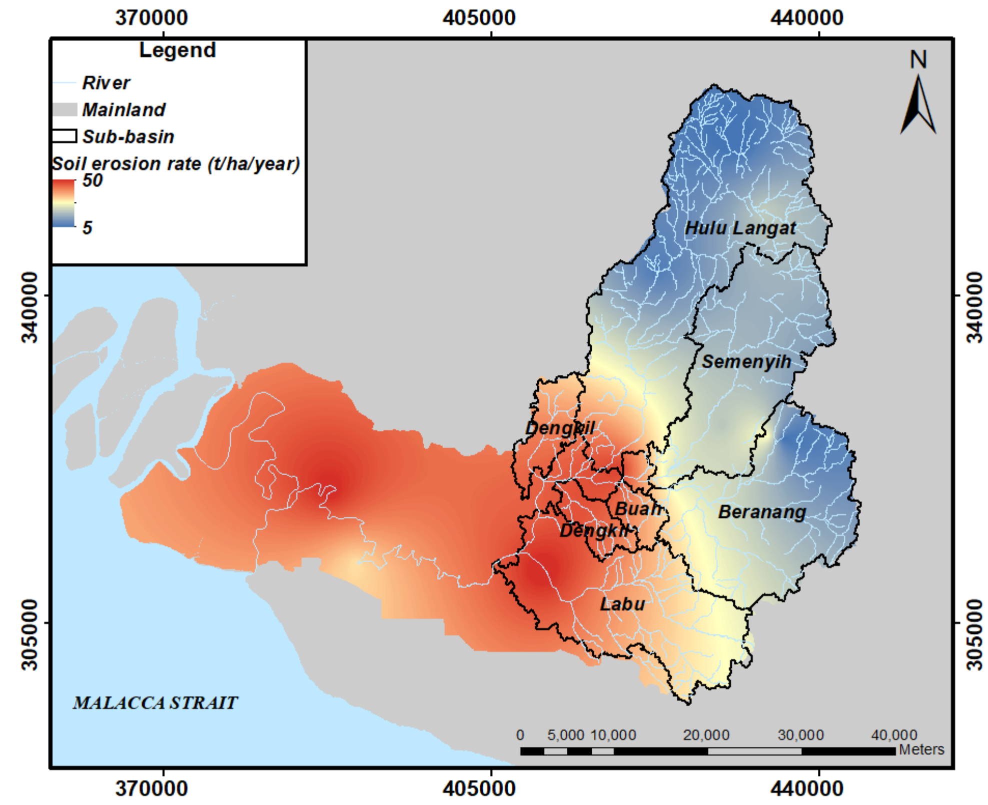

Figure 5: Spatial distribution of estimated net erosion rates by $^{210}Pb_{ex}$

CONCLUSION

The spatial distribution of fallout radionuclides inventories (^{137}Cs and $^{210}Pb_{ex}$) provided the basis for estimating soil erosion rates at the Langat watershed. This contribution proved the potential use of fallout radionuclides to quantify the medium- and long-term soil erosion rates and provides quantitative information on the relationship between erosion and inventory.

Based on the result obtained, it is proven that the radionuclide inventories are inversely proportional to the soil erosion rate. This study also illustrates the importance of establishing reference inventory within the local catchment. Individual inventory was relatively lower compared to the reference inventory, indicating erosion occurred at the specific point.

The combination of fallout radionuclides measurements with GIS software highlighted the soil redistribution processes occurring on the different geomorphic components identified within the Langat watershed. This potential combination encouraged the ability to upscale this study to a large watershed and help to strategies for a cost-effective method of assessing soil redistribution with limited sampling points.

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