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Depth Profile of ^{137}Cs Fallout in Soil in Cameron Highlands

Zaini Hamzah[1], Che Yasmin Amirudin[1]* and Ahmad Saat[2]

[1]*Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia*
[2]*International Education College, Universiti Teknologi MARA, 40200 Shah Alam, Selangor, Malaysia*

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ABSTRACT

^{137}Cs is one of the important man-made radionuclide introduced into the environment through nuclear weapons testing and nuclear accidents. In Cameron Highlands meaningful activity concentration of ^{137}Cs radionuclide has been measured. The present study embarked on the determination of ^{137}Cs activity concentration in soils of the sloping area. This would enable the distribution profile in the soil be investigated. Soil profile samples were collected by using scrapper plate in the sloping area which covers top, centre and bottom of the hills and a core in the forest for comparison purpose. The scrapper is 40 cm long and able to obtain slice of 2 cm interval of soil with diameter of 15.2 cm. Soil samples were oven dried, ground, sieved, homogenize and seal properly into a plastic container. The ^{137}Cs activity concentration was measured by using low background coaxial hyper pure germanium detector gamma spectrometer based on ^{137}Cs 661.66 KeV gamma ray. Results showed there were trend of low activity on the top of the hill and subsequently increases downhill. In all location, two trends of profiles were observed starting from the depth of 0 cm to 18 cm and 18 cm to 30 cm which indicating the presence of two sources of ^{137}Cs. The ^{137}Cs activity concentrations range detected at top location was from 0.05 Bq/kg to 1.53 Bq/kg, centre was 0.22 Bq/kg to 2.11 Bq/kg, bottom was from 0.00 Bq/kg to 2.03 Bg/kg and forest was 0.00 Bq/kg to 0.96 Bq/kg. The basic parameters of the soil such as pH (top; 3.72, centre; 4.26, bottom; 4.02), density (top; 1.45 g/m^3, centre; 1.41 g/m^3 , bottom; 1.59 g/m^3) and organic matter content (top; 17.08 %, centre; 16.75 %, bottom; 20.32 %) also being tested. These results show that the relative activity concentration of ^{137}Cs at the top is lowest, while at the bottom is highest, representing the presence of downward transport. However, identical pattern of profiles were observed at the three points of the slopes.

| Depth Profile | ^{137}Cs method | Gamma Spectrometer | Cameron Highlands |

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1. INTRODUCTION

^{137}Cs has been introduced into the environment as a result of nuclear bomb tests and nuclear power plant accidents [1]. Bomb-derived ^{137}Cs has been used to document soil degradation in different environment of northern and southern hemispheres. An additional input of Chernobyl-derived ^{137}Cs from Chernobyl accidents in 1986 was superimposed on the bomb-derived ^{137}Cs pattern within eroded areas. Based on the field measurement, there is a similarity of the depth distribution of bomb-derived ^{137}Cs with Chernobyl-derived ^{137}Cs for the main soil type [2]. Due to its relatively long physical half-live (30.2 years) and bioavailability, ^{137}Cs is one of the great concern fission products [3]. It is also has a long residence in the environment and is a hazard if found in sufficient concentrations [4]. Cesium is an alkali metal with chemistry similar to that of sodium, potassium and other elements in group 1 in the periodic table [5]. Much of the success of ^{137}Cs as a tracer is attributable to its low solubility and its ability and readily absorbs to small soil particles.

The positive charge fallout cesium ($^{137}Cs^+$) once reaching the earth surface is readily adsorbed onto the cation exchange sites on the surface soil particles and remains in this state almost in all terrestrial environments [6]. The raise used of artificial radionuclide ^{137}Cs in the current year has been seen by geomorphorlogists and soil scientist in determination of patterns and rate of soil redistribution [7]. ^{137}Cs contamination is positively correlated with altitude, so the upland, most susceptible sites, is ironically mostly contaminated too [8]. The significant of ^{137}Cs study in this area could provide the baseline data since not many studies in this research field in Malaysia. This paper presents the study of ^{137}Cs activity concentration in the soil, identifying distribution pattern of soil and comparing the ^{137}Cs depth profile in a hill of agricultural area and forest in Cameron Highlands, Pahang

2. EXPERIMENTAL

2.1 Study Area

The study area is located in Cameron Highlands, Pahang. Its forms the smaller district located at North West of Pahang in the upper left corner inside the Pahang state. The place is situated on the main mountain range of Peninsular Malaysia [9]. Cameron Highlands counts roughly 71,000 hectare of lands form which 79 percent is

*Corresponding author at
E-mail addresses: zainihamzah@live.com (Zaini Bin Hamzah)

still forested which makes 21 percent developed. The specific study area was in agricultural area where it is the second major land used after forestry with 16.4 percent of the total land area [9]. It is one of the areas with high altitude in Malaysia thus; the chance to receive fallout of ^{137}Cs radionuclide is relatively high. Figure 1 showed the sampling points in the study area. The details information on sampling locations using global positioning system is shown in Table 1.

2.2 Sampling and Sample Preparation

Soil samples were collected in sloping agricultural hill area (top, center and bottom) and in the forest namely PT, PC, PB and FR respectively. For each of the sampling point, bulk and depth incremental soil samples were taken. The scrapper plate with PVC pipe 40 cm long and 15.2 cm diameter was used for depth profile samples while a bulk samples were collected using hand auger with 20 cm long for basic parameter tests. The samples were taken to the laboratory for oven dried at 60^{o}C until constant weight, then ground using Fritsch pulverizes (400 rpm rotational speed, 2 repetitions, 5 minutes) and sieved through 250 µm sieve. A 150 g of samples was kept and sealed properly in the plastic container with diameter of 6 cm and 4 cm height.

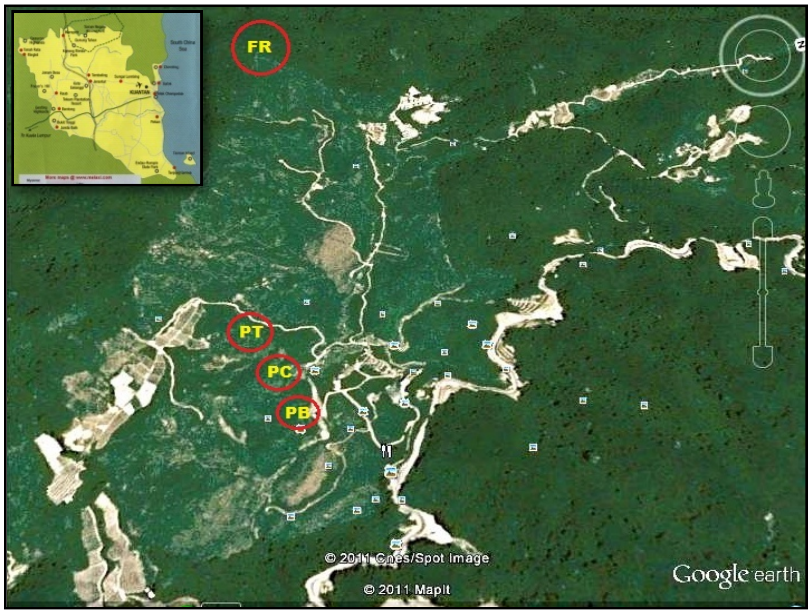

Fig. 1 Google maps of the study location in the agricultural area. Inserted map shows the state of Pahang, Malaysia.

Table 1 The locations of the soil samples (composite and profile)

Code	Locations	Elevation (m)	Description
PT	N 04º 27.088' E 101º 22.043'	1267	Top of the hill
PC	N 04º 27.101' E 101º 22.049'	1211	Center of the hill
PB	N 04º 27.156' E 101º 22.073'	1202	Bottom of the hill
FR	N 04º 27.264' E 101º 21.411'	1443	Forest (undisturbed area)

2.3 Measurements

Gamma spectrometer measurement was carried out using low background hyper pure coaxial germanium (p-type) detector. It is linked to a multi-channel digital analyzer system from EG & G, ORTEC. The gamma peak resolution at 1332 keV ^{60}Co was 1.82 keV and relative efficiency of 25 % with energy range from 40 keV to 10 MeV. The ORTEC Gamma Vision^{TM} Version 6.07 software was used for the spectra analysis and processing [10]. All the samples were counted at 28800 second live time and the spectrum was analyzed using Gamma Vision software for energy peak at 661.66 keV of ^{137}Cs. The efficiency calibration of the detector was made using secondary standard made up of UO_3 and KCl matrix in the same geometry as the sample [10]. The spectrometer efficiency at 661.66 keV obtained was used to calculate the activity concentration of ^{137}Cs in the soil using the equation,

$$A_{Ei} = \frac{N_{Ei}}{\varepsilon_E \times t \times \gamma_d \times M_s} \quad (1)$$

A_{Ei} = Activity concentration (in Bq/kg)
N_{Ei} = Net peak area of a peak at energy E
ε_E = Detection efficiency at energy E
t = Counting live time
γ_d = Number of gammas per disintegratio of this nuclide for a transition at energy E
M_s = Mass in kg of the measured sample

The minimum detectable activity (MDA) of this low background gamma spectrometer at 661.66 keV was 0.03927 Bq/kg calculated using the equation 2 [11];

$$MDA = \frac{\left(4.65\sqrt{N} + 3\right)}{ET} \quad (2)$$

N = background count
T = counting time
E = counting efficiency

The basic parameters cover pH, density (bulk density), texture (pipette method) and organic carbon content (loss of ignition) were also being tests. In-situ measurement using radiation survey meter (LUDLUM rate meter, Geiger Mueller Counter, model 2241) was record in each sampling location to determine radiation dose level.

3. RESULTS AND DISCUSSION

3.1 Depth Distribution Profile of ^{137}Cs

The vertical distribution profile of ^{137}Cs for the study area is showed in Table 2.0. The activity concentrations of ^{137}Cs are observed low in the upper land and high in the bottom area. The accumulation of this radionuclide clearly showed in this sloping area, where there is a movement of ^{137}Cs from the upper site down to lower placed. This reflects the progressive accumulation of soils contains ^{137}Cs at the bottom of the slope [12]. This result agrees with the surface dose reading as shown in Table 3.0. The surface dose reading at bottom of hill has higher the reading as compared to the center and top location. The depth profiles of ^{137}Cs activity concentration in the overall distribution patterns were observed and it seems to have the similar pattern. For the top, center and bottom of the slope, ^{137}Cs activity concentrations were slightly high in the top layer (0 cm to 2 cm) compare to inner layer of the soil profiles.

The ^{137}Cs activity concentrations decrease until 18 cm layer profile. The activity concentrations have increased again when reaching the depth of 20 cm and slowly decrease again down to 30 cm depth. These two trends indicate the presence of two different sources. The first trend of ^{137}Cs vertical profile may came from Chernobyl nuclear power plant accident in April 1986 [13]. The second trend came from environment as a consequence of above ground nuclear weapon testing between 1945 to 1963 [5] as shown in Figure 2.0 to Figure 5.0. The mean value of depth profile ^{137}Cs activity concentrations at TP, TC, and TB were 0.73 Bq/kg, 1.22 Bq/kg and 1.32 Bq/kg respectively.

In the forest soils, most ^{137}Cs survive in the organic layer and migrated downward through leaching of ^{137}Cs in the forest floor in a very slow processed [14]. The first layer of the forest soil was taken after removing the leaves and humus layer which cover the surface of the soil. For the first layer up to 16 cm, the ^{137}Cs activity concentrations decreased gradually. The second trend started at the depth of 18 cm. Decrease of ^{137}Cs in the first cm can be due to soil loss or ^{137}Cs migration in the soil matrix and animal perturbation [15]. It is very low migration depth of ^{137}Cs in forest soil and even many decades after deposition the majority of ^{137}Cs activity concentration is retained in the surface organic soil layers [13]. ^{137}Cs is attributed especially in the forest floor layers and the migration towards mineral horizons is function of mineralization of humus substances [13]. Further information regarding this result can be supported by the basic parameters of soil as presented in Table 4.

Soil pH affects all chemical, physical and biological soil properties. Number of researchers has studied in soils, showed that variations in soil pH were natural, due to chemical characteristics of different type of soil [16]. The results of soil samples analysis show that the pH of all samples is less than 5 (pH<5) i.e. within the range 3.18 – 4.16. It has the trend that higher altitude showed lower soil pH as compared to the bottom part of the hill. Lower pH will help ^{137}Cs ions to move freely and easy being washed by the runoff water. Furthermore, the soil density (1.0 g/m^3) and porosity especially in the undisturbed area of the forest together with its soil composition has contributed to the adsorption of ^{137}Cs.

These indicate no human activities in the high elevation area compare in the bottom area (density 1.59 g/m^3). If the bulk density becomes too high, it can limit plant root growth. The specific bulk density that will adversely affect plant root growth and development depends on many factors including parent materials, soil texture, the crop being growth and management history [17]. Soil texture in the hill location is sandy clay loam soil particles. Soil texture directly affects the porosity of soil, which in turn, determined its water-retention, flow characteristic and nutrient-holding capacity. Heavy clay soils normally have higher percentage of smaller pores and higher water holding capacity. On the other hand, sandy soils have relatively higher percentage of larger pores with lower water holding capacity under relatively dry conditions [18].

Table 2.0: Activity concentrations of ^{137}Cs at the slope of the hill and the forest

Layer (cm)	Activity Concentration (Bq/kg)			
	Top	Center	Bottom	Forest
00 – 02	1.53 ± 0.16	1.82 ± 0.17	2.03 ± 0.19	0.49 ± 0.08
02 – 04	1.22 ± 0.14	1.75 ± 0.16	1.76 ± 0.18	0.10 ± 0.04
04 – 06	1.25 ± 0.14	2.11 ± 0.18	1.67 ± 0.17	0.07 ± 0.08
06 – 08	0.60 ± 0.10	2.06 ± 0.17	1.08 ± 0.14	<MDA
08 – 10	0.76 ± 0.11	2.12 ± 0.19	1.92 ± 0.18	0.47 ± 0.08
10 – 12	0.63 ± 0.10	1.23 ± 0.13	1.62 ± 0.17	0.38 ± 0.07
12 – 14	0.58 ± 0.09	0.82 ± 0.11	1.58 ± 0.16	0.36 ± 0.07
14 – 16	0.15 ± 0.05	0.71 ± 0.10	1.22 ± 0.15	0.34 ± 0.07
16 – 18	0.05 ± 0.03	0.93 ± 0.11	0.87 ± 0.13	0.55 ± 0.09
18 – 20	0.92 ± 0.12	0.61 ± 0.09	0.53 ± 0.10	<MDA
20 – 22	0.62 ± 0.09	1.39 ± 0.14	1.90 ± 0.18	0.21± 0.05
22 – 24	0.54 ± 0.09	0.45 ± 0.08	<MDA	<MDA
24 – 26	0.98 ± 0.12	0.79 ± 0.11	0.91 ± 0.12	0.27 ± 0.06
26 – 28	0.68 ± 0.10	<MDA	0.79 ± 0.11	<MDA
28 – 30	0.42 ± 0.08	0.22 ± 0.05	0.54 ± 0.08	0.19 ± 0.05

Table 3.0: Surface dose reading on the hill slope and the forest

Location	Surface dose (μSv/hr)	
	0 meter	1 meter
PT	0.180	0.150
	0.160	0.150
	0.180	0.170
Average	**0.173**	**0.157**
PC	0.198	0.155
	0.154	0.194
	0.153	0.168
Average	**0.168**	**0.172**
PB	0.210	0.240
	0.210	0.210
	0.200	0.170
Average	**0.207**	**0.207**
FR	0.100	0.120
	0.120	0.120
	0.130	0.120
Average	**0.120**	**0.120**

Table 4.0: Basic parameter of soil samples in hill location and forest

Code	Texture (%)				pH	Density (g/m^3)	Organic Carbon Content (%)
	Clay	Silt	Sand				
PT	23.42	15.57	61.01	Sandy clay loam	3.72	1.45	17.08
PC	24.02	17.10	58.88	Sandy clay loam	4.36	1.41	16.75
PB	23.50	15.25	61.25	Sandy clay loam	4.02	1.59	20.32
FR	23.60	15.89	60.51	Sandy clay loam	3.18	1.00	10.51

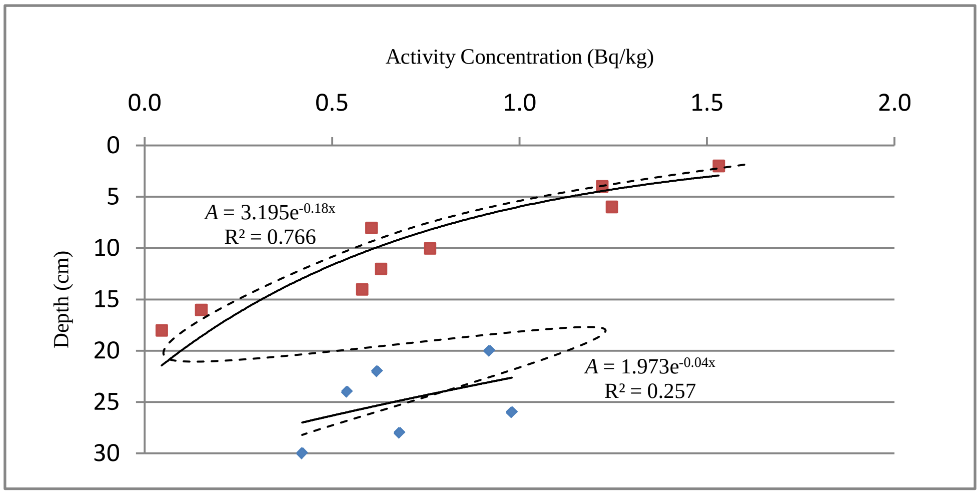

Figure 2.0: Depth distribution of ^{137}Cs in top of the hill

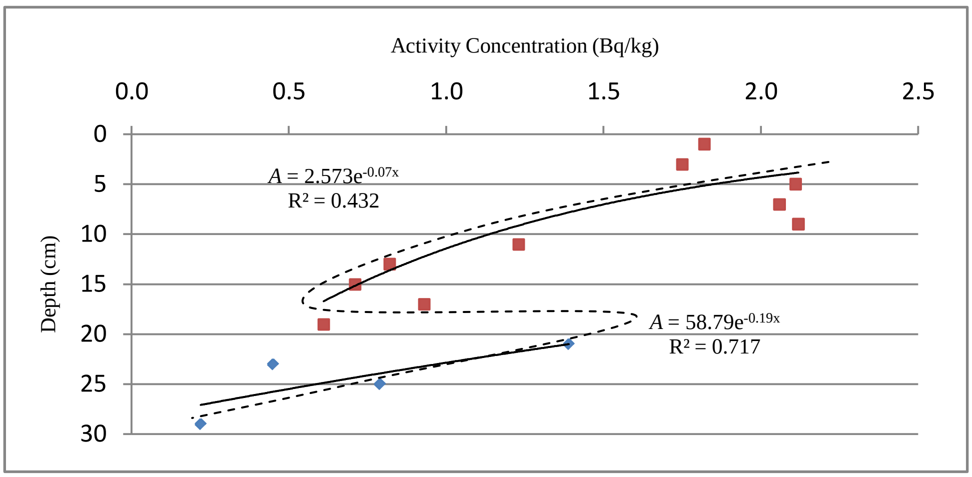

Figure 3.0: Depth distribution of ^{137}Cs in center of the hill

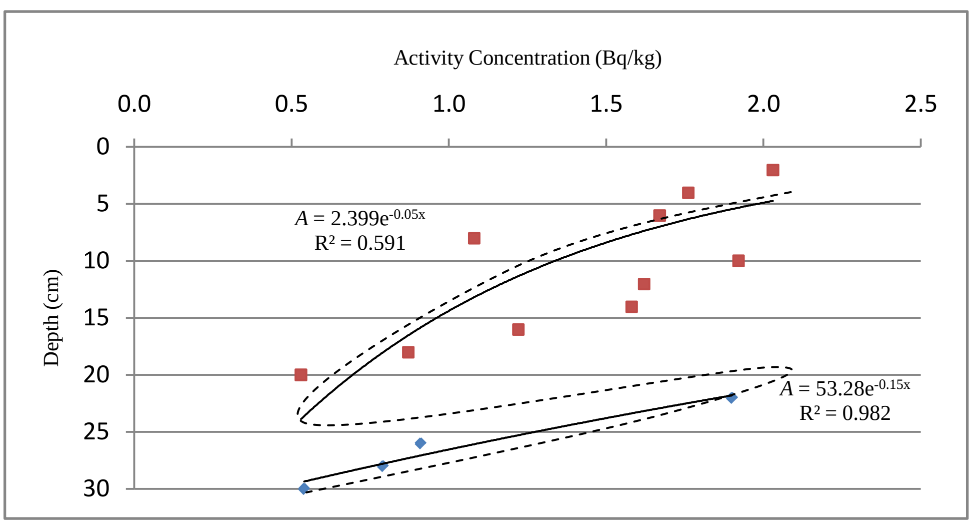

Figure 4.0: Depth distribution of ^{137}Cs in bottom of the hill

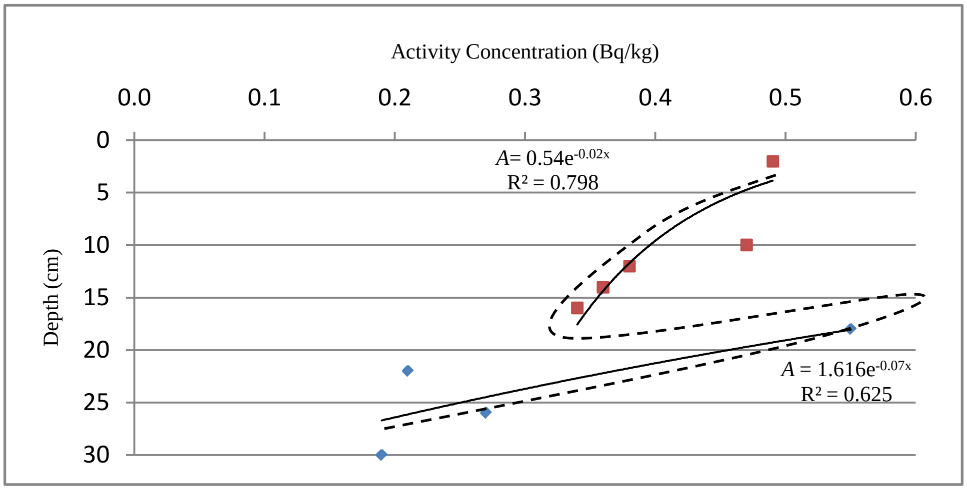

Figure 5.0: Depth distribution of ^{137}Cs in the forest

4. CONCLUSION

The activity concentrations of ^{137}Cs in soil for PT, PC, PB and FR shown the similar trend i.e. two depth redistribution trends. The obviously trends observed represent the first trend which indicating the sources from Chernobyl accident in 1986. While the second trend indicating the nuclear weapon test sources during 1950's to 1970's. This conclusion was supported by the basic soil parameters. The next stage of this study aim to cover more study area in this agriculture area thus will have complete set data and finally the soil erosion study can be done.

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