

SORPTION COEFFICIENTS OF ^{137}Cs ONTO VARIOUS MALAYSIAN SOIL SERIES

(Pekali Serapan ^{137}Cs ke atas Pelbagai Siri Tanih Malaysia)

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Abstract

The ratio of equilibrium radionuclide concentration in the solid phase to the concentration in soil solution i.e. the distribution coefficient (K_d) can be used for quantification of a radionuclide's migration potential in the subsurface. K_d is one of the main parameter that describes soil absorbability for any radionuclide. In this study, distribution coefficients (K_d s) of ^{137}Cs were measured by a batch technique for five Malaysian soil series samples to evaluate the absorbability of ^{137}Cs . The five Malaysian soils series samples i.e. Durian, Rengam, Holyrood, Segamat and Selangor were collected systematically at different depths (0-20 cm, 20-40 cm and 40-60 cm), at five different sites in Malaysia. The calculated K_d values for Durian, Holyrood, Rengam, Segamat and Selangor soil series were in the range of 958 to 3231 ml g^{-1} , 273 to 1349 ml g^{-1} (mean: $853 \pm 444 \text{ ml g}^{-1}$), 202 to 1739 ml g^{-1} (mean: $754 \pm 724 \text{ ml g}^{-1}$), 216 to 546 ml g^{-1} (mean: $388 \pm 113 \text{ ml g}^{-1}$) and 3389 to 5919 ml.g^{-1} (mean: $4657 \pm 918 \text{ ml g}^{-1}$), respectively. Thus this study indicated that the ^{137}Cs K_d values of the soils varies with type and depth of the soil and can be used as a good backfill choice for final disposal sites for retarding migration of ^{137}Cs .

Keywords: K_d – values, ^{137}Cs , Malaysian Soil Series

Abstrak

Nisbah kepekatan radionuklid seimbang dalam fasa pepejal larutan tanah iaitu pekali taburan (K_d) boleh digunakan untuk mengkuantitikan potensi pemindahan radionuklid di dalam sub permukaan. K_d merupakan salah satu parameter utama yang menerangkan keupayaan penyerapan tanah untuk pelbagai radionuklid. Dalam kajian ini, pekali taburan (K_d) ^{137}Cs diukur menggunakan teknik kelompok untuk sampel lima siri tanah Malaysia bagi menilai penyerapan ^{137}Cs . Lima sampel siri tanah i.e Durin, Rengam, Holyrood, Segamat dan Selangor dikumpulkan secara sistematik pada kedalaman yang berbeza (0-20 cm, 20-40 cm dan 40-60 cm) pada lima tapak yang berbeza di Malaysia. Keputusan pengiraan nilai K_d untuk masing-masing siri tanah Durian, Holyrood, Rengam, Segamat dan Selangor berada dalam julat 958 hingga 3231 ml g^{-1} , 273 hingga 1349 ml g^{-1} (min: $853 \pm 444 \text{ ml g}^{-1}$), 202 hingga 1739 ml g^{-1} (min: $754 \pm 724 \text{ ml g}^{-1}$), 216 hingga 546 ml g^{-1} (min: $388 \pm 113 \text{ ml g}^{-1}$) dan 3389 hingga 5919 ml g^{-1} (min: $4657 \pm 918 \text{ ml g}^{-1}$). Oleh itu, kajian ini menunjukkan bahawa nilai K_d ^{137}Cs untuk tanah yang berbeza jenis dan kedalaman boleh digunakan sebagai pilihan yang baik untuk pengambusan (backfill) tapak pelupusan akhir bagi merencat pemindahan keluar ^{137}Cs .

Kata kunci: Nilai K_d , ^{137}Cs , siri tanah Malaysia

Introduction

One of the hazardous radionuclides is ^{137}Cs because of its long half-life ($T_{1/2} = 30.2 \text{ yr}$) and high transfer factor by inhalation and ingestion. Knowledge of the mobility of ^{137}Cs in the natural environment is needed for the assessment of the impact of accidental ^{137}Cs releases to the environment. This study focused on improving knowledge on the physical and chemical properties of the soil and the mechanisms by which the ^{137}Cs is uptaken from soil to the food chain or the means that lead to the contamination of the water supplies [1]. The parameter known as (K_d -value) is defined as the distribution of a radionuclide in soil divided by the concentration in the soil

pore water. The value of the K_d depends on the type of radionuclide, the physical, chemical and mineralogical properties of the soil, and the temperature. The K_d -value of the radionuclides needs to be determined in order for the analyst to assess its contribution to the dose to the public. Mollah and Ullah have studied the sorption coefficients (K_d) of ^{137}Cs on soil matrices collected from locations in and around the proposed site for shallow land disposal of low-level radioactive wastes at the Atomic Energy Research Establishment (AERE) campus, Savar, Bangladesh. They found that the K_d for ^{137}Cs ranged from 1278 to 2156 mL g^{-1} [2]. Elejalde et al (1973) have determined the distribution coefficients of ^{137}Cs in 58 soils from 12 points in Biscay (Spain). The highest mean percentages of ^{137}Cs sorption were found in the residual category (69.93%), exchangeable (13.17%) and organic matter (12.54%) fractions. The study also included calculation of the partial K_d 's for chemical species as well as the relationships of the coefficients both between themselves and with the soil parameters. The treatment with ^{137}Cs leads to the sorption of most of the activity, with distribution coefficients K_d in the range from 484 to 42724 $\text{cm}^3 \text{g}^{-1}$ [3]. Soil distribution coefficients (K_d 's) of ^{137}Cs were measured by Nakamaru et al. (2007) for 87 Japanese agricultural soils to evaluate the adsorbability of ^{137}Cs [4]. Soil organic matter (SOM) may affect ^{137}Cs mobility because OM is a reactive colloid. Therefore, tracer studies were carried out to clarify the effect of SOM on ^{137}Cs mobility in different soils. The ^{137}Cs K_d 's ranged from 215 to 43400 L kg^{-1} with a geometric mean of 2210 L kg^{-1} . These results suggest that dissolved organic matter (DOM) increases the mobility of ^{137}Cs but that SOM can decrease the mobility of ^{137}Cs in soil by holding it in exchangeable and SOM-bound forms.

Materials and Methods

Soil sampling and preparation

Undisturbed soil samples were taken from the designated sampling stations shown in Table 1. and Figure 1. First, the detritus (grass and litter) at each sampling station were cleared from the ground surface and the soil was dug with a spade. Soil samples were collected from depths of 0-20 cm, 20-40 cm and 40-60 cm. About 500 g soil samples were collected from each depth. The samples were sealed in plastic bags and taken to the laboratory for further analysis. In the laboratory, the samples were air dried at room temperature, disaggregated using a wooden mortar and pestle and sieved through a 2 mm mesh for further use and analysis. In addition, soil cores (7.6 cm diameter and 4 cm long) were also taken from each depth for bulk density determination.

Table 1. Locations of soil sampling stations

Station No.	Soil series names	Grid References	Locations
1	Durian	03°28'02.9" N 102°06'31.8" E	93 km from Kuala Lumpur to Kuantan , 167 Kuantan to Kuala Lumpur – Kuantan Old Road
2	Holyrood	03°09'45.8" N 101°34'02.4" E	Located at Mature Rubber tree in Rubber Research Institute, Sungai Buloh. 15 km to Batu Tiga , 5 km to Sungai Buloh from Batu Tiga
3	Rengam	03°25'02.4" N 101°53'42.3" E	Kuala Lumpur to Bentong old road , 16 km to Bentong , 60 km to Kuala Lumpur
4	Segamat	03°30'15.9" N 102°32'58.8" E	110 km to Kuantan – Chenor road , near to the At-Taqwa Mosque
5	Selangor	03°20'56.3" N 101°18'29.8" E	Located at Indian Cemetery , Kuantan Village to Bukit Rotan Road

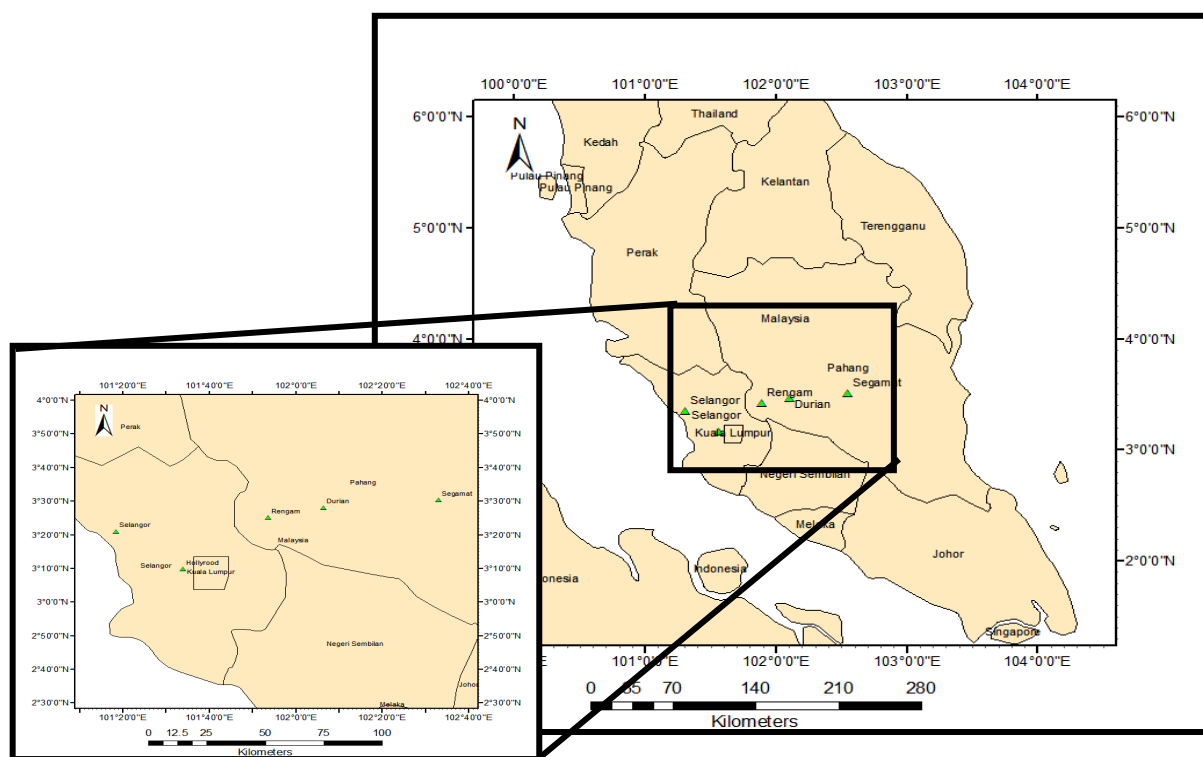


Figure 1. Location of soil sampling stations of five Malaysian soil series

Laboratory sorption experiments

The sorption of ^{137}Cs on five Malaysian soil series was studied using the batch technique on at least three replicates. The solution-soil ratio was maintained as 20:1 throughout the experiments in accordance with ASTM D4319-93 [5]. Twenty ml of 1788 Bq ml^{-1} ^{137}Cs tracer was spiked into deionised water. This solution was then mixed with $1.0 \pm 0.1 \text{ g}$ of each soil in 50 cm^3 polyethylene centrifuge tube with screw caps and septum sealed. Suspensions were initially preconditioned, using deionised water with on ^{137}Cs spike by shaking for 24 h at $23 \pm 2^\circ\text{C}$ using an end-over-end shaker. The spiked slurries were shaken for 7 days to achieve the equilibrium of ^{137}Cs adsorption, the solution phase was separated by centrifugation at 3000 rpm, and the supernatant was filtered through a $0.45\text{-}\mu\text{m}$ micropore filter. The 5 cm^3 of the supernatant was pipetted into small glass vials. The activity concentration measurements of ^{137}Cs in the vials was performed with a Canberra p-type HPGe well detector (GCW 2523) with active volume of 155 cm^3 , a relative photopeak efficiency of 25%, and a resolution at the 1332 keV energy of ^{60}Co of 2.3 keV (FWHM).

K_d (ml g^{-1}) was then calculated using equation 1:-

$$K_d (\text{ml/g}) = \frac{(C_i - C_f)}{C_f} \cdot \frac{V}{M} \quad (1)$$

where C_i (Bq ml^{-1}) is initial activity of the radiotracer in the aqueous phase, C_f (Bq ml^{-1}) is activity of radiotracer in the final supernatant, V is solution volume (20 ml), and M is soil dry weight ($1.0 \pm 0.01 \text{ g}$).

Treatment of the raw counting data was performed with the GENIE 2000 program. The activity (cps) of the ^{137}Cs photopeak (661.6 keV) was determined. Based on the background and blank counts, the minimum detectable activity (MDA) for this geometry was derived using Currie's Method formula [6]:

$$MDA = \frac{\sigma\sqrt{B}}{\varepsilon PTW} (\text{Bq.kg}^{-1}), \quad (2)$$

Where, σ is statistical coverage factor ($= 1.645$) ($P \leq 0.05$), B is background radiation of the radionuclide of interest, ε is counting efficiency of the detector, P is absolute transition probability of γ -decay, W is dried sample weight in g, and T is counting time in seconds. The MDA for ^{137}Cs determined was 2.087 Bq kg^{-1} .

Results and Discussion

The results of the experiments regarding distribution coefficients of ^{137}Cs in the five Malaysian soil types are shown in Table.2 and Figure.2. The calculated K_d values for Durian, Holyrood, Rengam, Segamat and Selangor soil series (all three depths included) were in the mean of $(2126.22 \pm 887.91 \text{ ml g}^{-1})$, $(853.36 \pm 444.98 \text{ ml g}^{-1})$, $(754.98 \pm 724 \text{ ml g}^{-1})$, $(388.88 \pm 113.99 \text{ ml g}^{-1})$ and $(4657 \pm 918 \text{ ml g}^{-1})$, respectively. The results also showed that the K_d -values depend on the depths of the soil. The K_d -values increase with depth except for Segamat and Selangor soil series. The results of this study indicated that 97.96 - 99.39%, 93.19 - 98.54% , 91.00 - 98.86% ,91.57 - 96.47% and 99.41 - 99.66% of added ^{137}Cs tracer was strongly adsorbed by the Durian, Holyrood, Rengam, Segamat and Selangor soil series, respectively. The soil series in this study showed higher K_d -value in upper layer.

Table 2. Mean distribution coefficients (K_d -value) for ^{137}Cs in five of Malaysian soil series

Depths (cm)	Soil series				
	Durian	Holyrood	Rengam	Segamat	Selangor
	K_d -value in (ml g^{-1}) $\pm$ S.D*				
0-20	1061.91 ± 137.16	292.54 ± 17.32	206.61 ± 4.26	517.03 ± 35.39	5345.99 ± 499.67
20-40	2293.49 ± 389.84	975.29 ± 28.54	341.54 ± 22.90	386.94 ± 18.73	5099.78 ± 426.56
40-60	3023.26 ± 186.09	1292.25 ± 88.89	1716.79 ± 35.23	262.68 ± 42.89	3526.39 ± 125.52

*S.D: Standard deviation

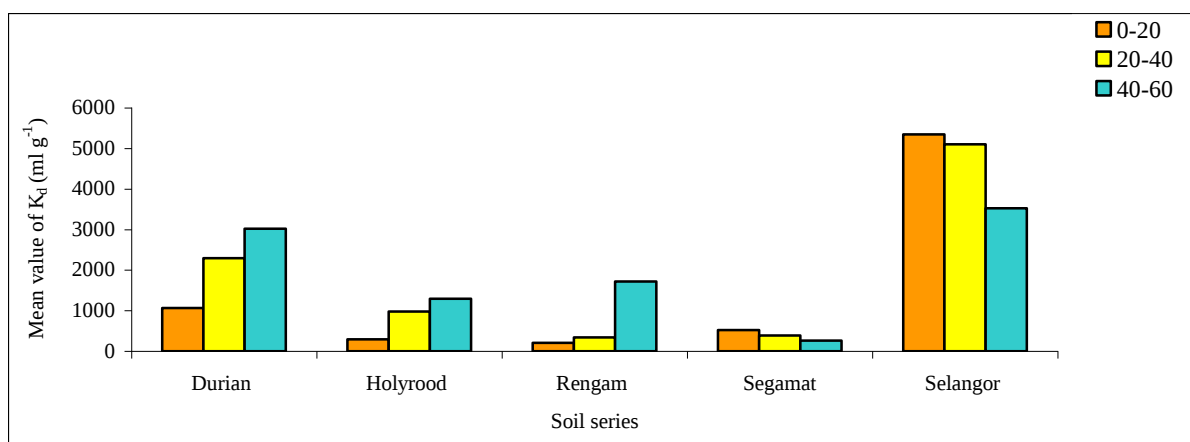


Fig 2. ^{137}Cs adsorption onto five of Malaysian soils series.

Given the above results, it can be concluded that further investigation is required in order for a better relationship between K_d -value and physico-chemical soil properties to be established. Soils have the tendency to absorb and distribute the ^{137}Cs depending upon the texture and physicochemical properties. The investigation was made to measure the absorption capacity of the soil in batch method to accommodate ^{137}Cs . Selangor soil exhibits the maximum sorption coefficient value with a steep decrease towards depth, which might be attributed to the decrease in porosity. Other soils did not exhibit the remarkable sorption except Durian soil, and Segamat soil was found with minimum sorption coefficient value. The behaviour of the different soils was presenting different trends with respect to their depth. Selangor and Segamat soils show that the K_d values decrease with the increase in depth, however other three soils presents more sorption coefficient with the depth. The variability in the values of sorption coefficients can be attributed to the type of soil and its composition. Zidan et al (2010) have reported the texture of Selangor soil as a clayey one with concentration of clay mainly combination of Kaolinite, illite and monmorillonite ranging from 65%, 74% and 76% at the depths of up to 20, 40 and 60 cm respectively [7]. ^{137}Cs distribution or sorption coefficients may need some type of interactions to clayey nature of the specific particles. It was also observed that Durian soil's composition is clayey too but with less concentration as compared to the Selangor soil. The nature of the other soils including Holyrood and Rengam is not Clayey but sandy which did not allow the Cs to incorporate properly for channelling to migration [8]. In case of Segamat soil, minimum sorption of ^{137}Cs was observed even the texture of soil was not sandy but clayey [9]. The lack of sorption capacity of the clay towards Cs incorporation can be attributed to the type or concentration of the clay contents. Instead of being Calyey in texture, the Segamat soil was not able to allow Cs for remarkable sorption and migration values which might be attributed to different nature and composition of the clay than Selangor soil. Bachhuber et al. (1982) have reported the rates of distribution, migration and retardation factors of the distribution coefficients of ^{137}Cs in the various horizons using batch method. They have made their findings of the distribution coefficients based on the accumulated organic matter in the soil [10]. Chang and Hsu have reported the adsorption of ^{137}Cs on bentonite clay which is an evidence regarding existence of some interactions between clay and ^{137}Cs [11].

Conclusion

The distribution and migration of ^{137}Cs in the soil depends in particular on its adsorption properties and the movement of soil water. The movement of water carrying the subject matter in the soil is determined by the amount of precipitation and other soil properties including texture, porosity and organic contaminants. The adsorption properties are also determined by the proportion, type and composition of the clay mineral content of the soil. This study has revealed some trends of sorption in relation to the nature or type of clay. The sorption was not supported by soil containing sandy texture but enhanced sorption was observed with the soil containing a combination of clay including kaolinite, illite and montmorillonite i.e Selangor soil. The obtained results indicate that these types of soil could be considered as good backfill choices for final disposal sites for retarding migration of ^{137}Cs .

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