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EFFECTS OF PHYSICO-CHEMICAL SOIL PROPERTIES ON THE ADSORPTION AND TRANSPORT OF ^{137}Cs IN RENGAM AND SELANGOR SOIL SERIES

(Kesan Sifat Fizikokimia Terhadap Penyerapan dan Pengangkutan ^{137}Cs Dalam Tanah Siri Rengam dan Selangor)

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Abstract

In this study, the adsorption of ^{137}Cs in soil samples were quantified using the distribution coefficient (K_d-value). The distribution coefficients (K_d) of ^{137}Cs in Rengam and Selangor soil series were determined by a batch technique. The Malaysian soil series (Rengam and Selangor soil series) were collected systematically at three different depths (0-20 cm, 20-40 cm and 40-60 cm) at two different sites in Malaysia. The batch K_d tests were used with deionized water that was spiked with ^{137}Cs tracer to the soil sample and the activities of ^{137}Cs in the supernatant solution were measured by a low background but high efficiency well-type HPGe detector. Several physicochemical soil properties were also characterised for each soil type. Pearson's correlation and stepwise multiple regression models were applied at 0.05 level of significance throughout all analysis to determine the relationships and influences between distribution coefficients (K_d-value) of ^{137}Cs with physicochemical soil properties of each soil type. The calculated K_d-value for Rengam and Selangor soil series at several depth were determined to be in the range of 202 to 1739 ml.g^{-1} and 3389 to 5919 ml.g^{-1} respectively. The results indicate that the stepwise multiple regression model incorporating pH and porosity influence the K_d-value of ^{137}Cs in Rengam Soil Series and exhibits an R^2 equal to 0.922 indicating that 92.2% of total variation has been explained by the regression model. The regression model also reveals that cation exchange capacity, bulk density, porosity and free manganese oxide (Mn^{2+}) have influence on the K_d-values of ^{137}Cs in Selangor soil series and exhibits an R^2 equal to 0.997 indicating 99.7 % of total variation. Therefore, the sorption coefficients in relation to the environmental factors including physicochemical properties can be used to predict and design the radionuclide transport and safety assessment models.

Keywords: K_d-value, ^{137}Cs, physicochemical soil properties

Abstrak

Dalam kajian ini, penyerapan ^{137}Cs dalam sampel tanih telah dikuantitikan dengan menggunakan Kaedah Pekali Taburan (Nilai K_d). Pekali Taburan (K_d) ^{137}Cs bagi siri tanih Rengam dan Selangor telah ditentukan menggunakan teknik kelompok. Siri tanih Rengam dan Selangor telah dikumpulkan secara sistematik pada tiga kedalaman yang berbeza (0-20 cm, 20-40 cm dan 40-60 cm) di dua lokasi yang berlainan di Malaysia. Nilai K_d ditentukan menggunakan air ternyahion yang ditambahdengan penyurih ^{137}Cs kepda sampel tanih. Aktiviti ^{137}Cs di dalam larutan supernatant dibilang menggunakan pengesan HPGe yang mempunyai latar beakang yang rendah tetapi kecekapan yang tinggi. Beberapa sifat fisikokimia tanih juga telah dicirikan bagi setiap jenis tanih. Korelasi Pearson dan Model Regresi Berganda Berperingkat turut digunakan pada tahap signifikan 0.05 untuk keseluruhan analisis bagi menentukan hubungan dan pengaruh antara pekali taburan (nilai K_d) ^{137}Cs dengan sifat fisikokimia bagi setiap jenis tanih. Nilai K_d yang telah dikira untuk masing-masing siri tanihRengam dan Selangor dicatatkan dalam julat 202 hingga 1739 ml g^{-1} dan 3389 hingga 5919 ml g^{-1}. Keputusan menunjukkan bahawa model regresi berganda berperingkat menggabungkan pH dan keliangan mempengaruhi nilai K_d ^{137}Cs bagi tanih siri Rengam dan mempamerkan R^2 bersamaan dengan 0.922 yang mana menunjukkan 92.2 % dari jumlah variasi telah diterangkan oleh model regresi. Model regresi juga menunjukkan bahawa kapasiti

pertukaran kation, ketumpatan pukal, keliangan dan mangan oksida bebas (Mn^{2+}) dapat mempengaruhi nilai K_d ^{137}Cs dalam tanih siri Selangor dan mempamerkan R^2 bersamaan 0.997 yang menunjukkan bahawa 99.7 % daripada jumlah variasi. Maka kajian ini mendapati pekali penyerapan yang berkaitan dengan faktor sekitaran termasuklah sifat fisikokimia boleh digunakan untuk meramal dan merekabentuk pengangkutan radionuklid dan model penilaian keselamatan.

Kata kunci: Nilai K_d, ^{137}Cs, sifat fizikokimia tanah

Introduction

Cs is one of the important radionuclides for several reasons: it shows the almost unlimited solubility in the inventory of radioactive waste is significant [1]. A radioisotopes of cesium, ^{137}Cs is relatively resistant and provides important problems radioecologically [2]. The main sources of soil contamination are global fallout of ^{137}Cs from the atmosphere as a result of atmospheric testing of nuclear weapons, surface and underground nuclear explosions and accidental releases from nuclear facilities. In addition to these main sources of ^{137}Cs soil contamination, there have been few cases of local contamination which could increase the radiation dose to the population in surrounding areas[3]. In addition, because of its chemical similarity to K, Cs is easily assimilated by terrestrial and aquatic organisms [1]. The study of sorption of pollutants in soil is of great interest in terms of ecological and agricultural. It is known that ion exchange reactions occur mainly between Cs ions and clay minerals. The bioavailability of Cs^+ in natural systems depends largely on the adsorption properties of the solid phase [4].Radionuclides present in soil have the potential to be released or leached to the groundwater. In order to understand the way in which radioactive contamination may be transferred from soil to plants, animals and finally to man, it is useful to have a basic knowledge on both the physical and chemical properties of soils and the mechanisms by which radionuclides are passed from the soils to the food chain or the means that lead to the contamination of water supplies [5]. The primary objective of the present study is to determine the effects of physico-chemical soil properties on adsorption of ^{137}Cs in Rengam and Selangor soil series . Such study could assist future environmental study such as environmental radiological impact assessments (RIA) in the future. One of the important factors that needs to be considered in RIA is the possible release or leaching of radionuclides into ground water.

The Rengam Series is a member of the Rengam Family which is a fine, kaolinitic, isohyperthermic, red-yellow Tipik Lutualemkuts. It typifies this family which is developed over coarse grained acid igneous rocks [6].The Selangor soil series is clay dominated, often derived from Entisols. This clay is typically found near the surface of marine alluvial plains typical of the west coast of peninsular Malaysia that have for many years been drained for the cultivation of crops [6]. The complex sorption interaction can be expressed as a distribution coefficient (K_d), which is the ratio of the amount of the radionuclide sorbed by the solid (g g^{-1}) divided by the concentration in the equilibrium solution (g ml^{-1}). The K_d -value is commonly used as a means of assessing the mobility of radionuclides in the environment and for comparing adsorption data obtained from different sources [7].The parameter known as (K_d-value) is defined as the ratio of the concentration of the radionuclide bound to soil to the concentration in the pore water. The value of K_d depends on the type of radionuclides, physico-chemical soil properties and mineralogy of the soil, The K_d-value of the radionuclides needs to be determined in order to estimate the transport or leaching potential used to assess associated risks.

Materials and Methods

Soil sampling and preparation

The studied soil were Rengam and Selangor series, Typic Kandiudult and Typic Endoaquepts, respectively [6]. Undisturbed soil samples were taken from the designated sampling stations shown in 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.

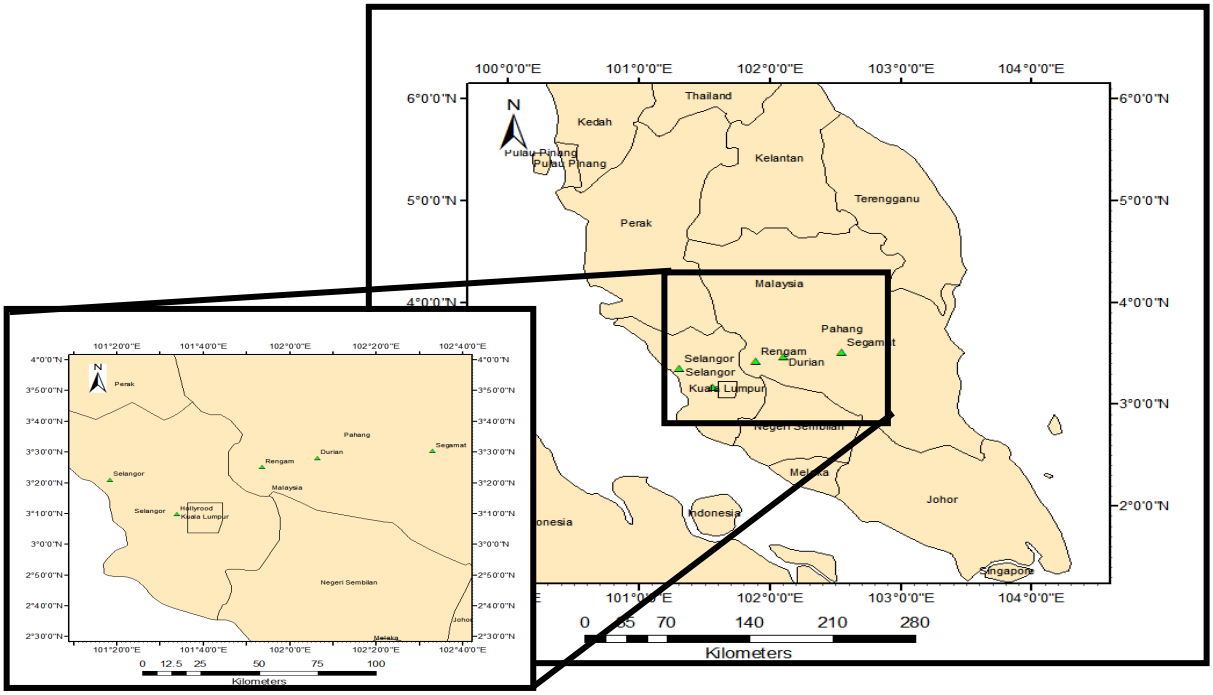

Figure 1. Location of soil sampling stations of Rengam and Selangor soil series

Determination of physico-chemical soil properties and soil clay mineralogy

Triplicate sieved soil samples from each of three depths were used for determination of the following physico-chemical soil parameters: bulk density, porosity and soil particle size distribution. Porosity was calculated as the difference between the soil particle density and bulk density, while particle size distribution was determined by the Pipette method, together with dry sieving [8]. The texture of the soils was obtained by plotting their percentages of sand, silt and clay and categorized using the triangular texture map. Soil bulk densities (g/cm^3) were derived by dividing the mass of the dried soil cores by their volumes [9].

The pH was measured using a pre calibrated lab-top pH meter equipped with a glass electrode in an aqueous suspension of 10 g of dry soil and 25 ml of distilled water (1:2.5,w/w), shaken for 30 minutes in a conical flask [10]. Exchangeable acid ions (Al^{3+} and H^+) were measured in 1.0 M KCl extract by titration. Exchangeable base cations were measured in 1.0 M ammonium acetate extract by Flame Atomic Absorption Spectrophotometer (FAAS, model Perkin Elmer 3300) and cation exchange capacity (CEC) was obtained by summation of the acid and base cations. The base saturation was calculated by the equation [sum cations / CEC] × 100]. The extractable free Fe- and Mn-oxides contents were determined by the dithionite-citrate method [11]. Sulphate contents were measured on a sediment water extract [1 g :10 mL] after two hours of shaking and filtration using the barium sulphate turbidometric procedure [12]. The specific surface area was determined by the ethylene glycol monoethyl ether (EGME) method [13]. Organic carbon was estimated using the Walkley and Black method and organic matter (OM) was calculated by multiplying the values of organic carbon by 1.72 [14].

For identification of clay minerals in the bulk soil samples, X-ray diffraction (XRD) analysis was performed using a Siemens D500 powder X-ray diffractometer using CuKα radiation, and operated at 40 kV and 30 mA. The oriented specimens were scanned from 3 to 35 degrees 2θ. All samples were prepared for both qualitative and quantitative determination by X-ray diffraction analysis according to the general procedure of Moore and Reynolds [15]. About 1g of soil was placed into 10-ml measuring cylinder. About 5 ml of distilled water was then added in to it and was mixed thoroughly using a glass rod. After 15-20 minutes, the upper solution was then pipetted (using an

eyedropper) and placed slowly on the clean glass slide so that the liquid covered the entire surface of the slide. It was assumed, according to Stoke's Law, that the solution that was sampled contained the clay particles of less than 2 μm. The slide was dried at the room temperature. High drying temperature was not used to avoid any damage to poorly crystallised clay minerals in soils. Several treatments have been undertaken to verify the clay minerals in the soil.

Laboratory sorption experiments

The sorption of ^{137}Cs on Rengam and Selangor soil series were studied by batch technique using at least three replicates. The solution-soil ratio was maintained as 20:1 throughout the experiments in accordance with ASTM D4319-93 [16]. Twenty 20 ml of 1788.570 Bq ml^{-1} ^{137}Cs was spiked into deionised water. The spiked solutions were mixed with 1.0 ± 0.1 g of the soil in 50-cm^3 polyethylene centrifuge tubes 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±2 ^{0}C using an end-over-end shaker. The mixture was 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 though a 0.45-μm micropore filter. The 5ml aliquots of the filtered supernatant were placed in 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 resolutions at the 1332 keV energy of ^{60}Co of 2.3 keV (FWHM).

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

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

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 ± 0.01 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 [17]:

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

where, σ is statistical coverage factor (= 1.645) (P ≤ 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}.

Data analysis

In the data analysis, the soil's physical and chemical properties were considered as independent factors while the observed adsorption of ^{137}Cs was considered as the one dependent factor. The coefficient of correlations and stepwise multiple regressions were calculated to determine the independent factors which were significantly controlled with the dependent factor. The statistical significance was considered at significant level 0.05 (α = 0.05). Therefore, we considered that the regression models and coefficients correlations of the 5% or less chance of error (p ≤ 0.05). Data analysis was performed using the statistical software Statistical Package for Social Sciences (SPSS) Ver.15.0. Assuming that the data were normally distributed, significant differences were considered at $p \leq 0.05$. Pearson's correlation and a stepwise multiple linear regression technique was used to derive the relationship and influences between physico-chemical soil properties and distribution coefficients (K_d-value).

Results and Discussion

The results of K_{d-} experiments on two Malaysian soil types are shown in Table 1 and Figure 1 . The range of K_d-Cs values of these Malaysian soil series were determined to be between 202 ml g^{-1} to 1739 ml g^{-1} and 3389 ml g^{-1} to 5919 ml g^{-1} for Rengam and Selangor soil series, respectively. The results also showed that the K_d-value of ^{137}Cs dependeds on the depths of soil. The results showed that the K_d-value of ^{137}Cs is higher as the depth of soil increased for Rengam soil series. However, for the Selangor soil series, the K_d-Cs-value was found to decrease as the soil depth increased. The remarkable difference in K_d values of the Rengam and Selangor soil series can be attributed to be influenced by number of factors including soil texture, pH, porosity etc (Table 2 and 3).

Table 1. Mean of distribution coefficients (K_d-value) for ^{137}Cs in Rengam and Selangor soil series

Depths (cm)	Rengam		Selangor	
	Range	Mean value K_d-value in (ml g^{-1}) ± S.D*	Range	Mean value K_d-value in (ml g^{-1}) ± S.D*
0-20	202 - 210	206 ± 4	5001 - 5919	5345 ± 499
20-40	326 - 367	341 ± 22	4608 – 5375	5099 ± 426
40-60	1676 - 1739	1716 ± 35	3389 – 3636	3526 ± 125

*S.D: Standard deviation

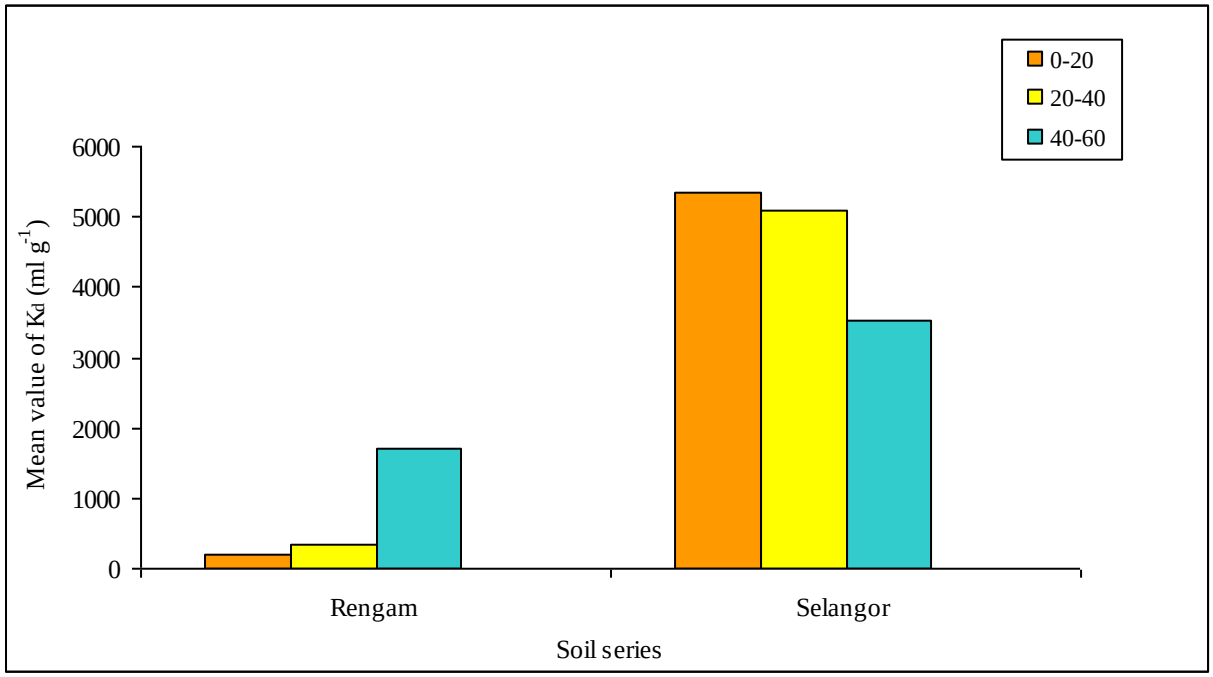

Figure 2.^{137}Cs adsorption onto Rengam and Selangor soils series

Table 2. Physico-chemical and mineralogical characteristics of Rengam soil series

Rengam soil series				
Parameter	**Unit**	**Depths (cm)**		
		0-20	**20-40**	**40-60**
Bulk density	(g/cm^3)	0.98±0.02	1.06±0.02	1.10±0.03
Porosity*	(%)	60.28±0.49	59.58±0.91	57.37±0.44
Sand	(%)	57±0.58	50±0.6	48±1
Silt	(%)	16±0.58	13±1.2	12±1.15
Clay	(%)	27±1	37±0.6	40±0.58
Soil texture		Sandy clay loam	Sandy clay	Sandy clay
Specific surface area	(m^2/g)	77.2±7	88.1±9.5	94.1±0.9
pH (H_2O)		4.10±0.08	4.06±0.06	4.10±0.08
Total organic carbone	(%)	1.89±0.20	0.65±0.06	0.45±0.13
Organic matter	(%)	5.59±0.59	1.93±0.17	1.33±0.38
Cation exchange capacity	meq / 100 g soil	5.62±0.15	6.00±0.57	5.99±0.79
Cation exchange capacity of clay	CEC / 100 g clay	20.85 ± 0.95	16.36 ±1.54	15.12 ± 2.20
Base Saturation	(%)	13.00±2.65	7.67±2.08	5.33±0.58
Free Iron oxide (Fe^{3+})	$\mu g.g^{-1}$	5021.1±481.5	5696.4±486.7	6094.4±611.4
Free Manganese oxide (Mn^{2+})	$\mu g.g^{-1}$	142.1±28.3	121.6±43.8	120.2±62.8
Sulphate (SO_4^{2-})	$\mu g.g^{-1}$	35 ± 0	47.5 ± 2.5	22.5 ± 2.5
Main cay mineral type (in order of predominance)		Kaolinite	Illite	Montmorillonite

*Porosity calculated based on soil particle density of 2.65 g /cm^3 and soil bulk density value; meq: milli-equivalents.

Table 3. Physico-chemical and mineralogical characteristics of Selangor soil series

Selangor soil series				
Parameter	**Unit**	**Depths (cm)**		
		0-20	**20-40**	**40-60**
Bulk density	(g/cm^3)	0.98 ± 0.02	1.07 ± 0.06	1.05 ± 0.04
Porosity*	(%)	60.39 ± 0.74	57.87 ± 0.57	58.16 ± 2.84
Sand	(%)	12 ± 2.08	10 ± 1.53	8 ± 0.58
Silt	(%)	23 ± 2.65	16 ± 2.89	16 ± 1.73
Clay	(%)	65 ± 4.51	74 ± 1.53	76 ± 2.08
Soil texture		Clayey	Clayey	Clayey
Specific surface area	(m^2/g)	101.8 ± 3.1	113.7± 14.6	121.3 ± 3.2
pH (H_2O)		3.70 ± 0.17	3.49 ± 0.03	3.41 ± 0.07

Total organic carbone	(%)	4.26 ± 0.10	2.70 ± 0.10	1.93 ± 0.29
Organic matter	(%)	12.65 ± 0.29	8.00 ± 0.30	5.73 ± 0.87
Cation exchange capacity	meq / 100 g soil	8.79 ± 0.31	8.71 ± 0.15	5.55 ± 0.87
Cation exchange capacity of clay	CEC / 100 g clay	15.65 ± 2.07	11.83 ± 0.11	7.27 ± 1.06
Base Saturation	(%)	32.33 ± 1.23	15.67 ± 2.08	21.67 ± 3.79
Free Iron oxide (Fe^{3+})	$\mu g.g^{-1}$	2647.3 ± 277.0	2815.6 ± 292.5	3315.4 ± 365.7
Free Manganese oxide (Mn^{2+})	$\mu g.g^{-1}$	196.0 ± 24.9	227.3 ± 24.9	169.6 ± 11.1
Sulphate (SO_4^{2-})	$\mu g.g^{-1}$	145 ± 5	247.5 ± 7.5	92.5 ± 7.5
Main cay mineral type (in order of predominance)		Kaolinite	Illite	Montmorillonite

*Porosity calculated based on soil particle density of 2.65 g /cm^3 and soil bulk density value; meq: milli-equivalents.

Table 4 reports the regression result between K_d-value of ^{137}Cs of Rengam soil series and physico-chemical soil parameters. The final ^{137}Cs K_d model uses two independent variables, porosity and pH and exhibits an R^2 equal to 0.922 indicating that 92.2% of total variation has been explained by the regression model. The final regression model is:

$$K_d\text{-value of } ^{137}Cs = 16377.562 - 485.848 \times \text{porosity} + 3200.708 \times \text{pH} \quad (3)$$

Base on the objective of this study, correlation analysis was followed by stepwise multiple regression analysis in order to verify the correlation. From the final equation model described above, the porosity is a negative influencing parameter on K_d-Cs-value and pH has positive influence on K_d-Cs-value in Rengam soil series. One of largely influencing parameter related to porosity is a factor of particle (grain) size. Statistical data reveals that there is a negative relationship between porosity with clay and porosity with bulk density. Clay soils exhibit a variety of micropores and macropores which may hold water contents and control the filtration rates. [18]. The Cs sorption is related to the pH of the soil because of its nature of charge density at specific value. The equation A determines the pH factor as a significant parameter to evaluate the sorption. It is reported that the maximum Cs sorption was observed at higher pH, when the negative charge density on the surface of absorbents was highest [19]. Therefore, The pH influence found agreed with the findings reported by Kami-Ishikawa et al for the clayey nature of soils.As the acidic conditions are concerned, the availability of cations may create competition with other cations for the available sorption sites [20].

Table 4 presents the regression model between physic chemical soil properties and K_d-value of ^{137}Cs of Selangor soil series. The final ^{137}Cs K_d model was established using four independent variables including cation exchange capacity, bulk density, porosity and free manganese oxide (Mn^{2+}) which exhibits an R^2 equal to 0.997 indicating 99.7 % of total variation as mentioned in equation B. The final regression model is:

$$K_d\text{-value of } ^{137}Cs = -9203.798 + 393.586 \times \text{CEC} + 4219.826 \times \text{B.D} + 90.337 \times \text{porosity} + 5.013 \times \text{Mn} \quad (4)$$

where CEC is cation exchange capacity, B.D is bulk density.

Table 4. Regression summary for K_d-value of ^{137}Cs of Rengam and Selangor soil series respectively

Physico-chemical variable	**Regression parameters**		
	Regression coefficient	**t-statistic**	***p*-value**
Model 1			
Constant	16377.56		
Porosity	- 485.85	-8.29	0.001
pH (H_2O)	3200.71	2.58	0.042
Model 2			
Constant	-9203.798		
Cation exchange capacity(CEC)	393.586	16.43	0.001
Bulk density (B D)	4219.826	4.79	0.009
Porosity	90.337	5.81	0.004
Free manganese oxide (Mn^{2+})	5.013	3.09	0.036

It can be concluded from the final equation model described as Eq 4, the cation exchange capacity, bulk density, porosity and free manganese oxide (Mn^{2+}) have positive effect on K_d-Cs-value in Selangor soil series. It is fact that the cation exchange capacity influence on the adsorption of ^{137}Cs due to its ion exchange characteristics. The CEC value strongly depends on the clay content and thus the relationship between the K_d-value and CEC will be similar to but weaker than that for the K_d value and the clay content [21].The bulk density and porosity have influence on the adsorption of ^{137}Cs due to their relation with the permeability of the contents to perform sorption. The small pore size and high density may not allow the contents for migration to create the negative influence and vice versa. The free manganese oxide (Mn^{2+}) influence positively on the adsorption of ^{137}Cs due to its ability to adsorb the Cs. It may provide the binding sites for the incorporation of Cs to migrate through the channels in the soil. Hasany and Chaudhary (1984) have reported the adsorption of ^{137}Cs on MnO^2 under different conditions of physical parameters [22].

Conclusion

The study results suggest that Cs-K_d was affected by several soil properties, such as pH, porosity in Rengam soil series and bulk density, cation exchange capacity and free manganese oxide (Mn^{2+}) in Selangor soil series. The study signified that the Rengam and Selangor soil series have good sorption for ^{137}Cs radionuclide. The distribution and migration of Cs in the soil depends in particular on its physicochemical properties and the movement of soil water with associated contents. The transport of the Cs in the soil is determined by the amount of absorbed Cs and other soil properties including texture, porosity and organic contaminants. The adsorption properties have also relation with the type and composition of the clay mineral content of the soil. This study has revealed some trends of sorption in relation to the nature of soil texture and other physicochemical properties and the data could be used in radionuclide transport and safety assessment models.

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