



The effect of ^{137}Cs Sorption on a Tropical Soil

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Abstract

Understanding the behaviour of ^{137}Cs in the soil subsurface is important to avoiding health-associated risks of radionuclides in our environment. For this, we studied the tropical Malaysian soil to determine the factors affecting ^{137}Cs sorption level; we evaluated sorption level using distributed coefficients and soil physico-chemical properties on 45 soil samples from 5 sites. We found the range 202 L kg^{-1} to 5919 L kg^{-1} for the distribution coefficients and the geometric mean of 1039 L kg^{-1} . Further, our results show that the main factor contributing ^{137}Cs sorption in the tropical Malaysian soil is cation exchange capacity (CEC).

Keywords: Sorption, ^{137}Cs , physico-chemical soil properties, soil solution distribution coefficient, Malaysian soil.

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1. Introduction

In the tropical Asia, the rapid economic development is generating an increase in the construction of nuclear power plants. These plants are believed to be needed to support the growing power demands of the countries in the region. However, there are serious well-known problems with this approach to power generation. One of the problems is the risk of a nuclear accident and the potential health impact on the environment. Because of its long radiological half-life of 30 years and high fission yield from ^{235}U and ^{239}Pu , ^{137}Cs is considered to be the most important radioactive nuclide (radionuclide) released in the event of a nuclear accident. Thus, for a risk assessment of a nuclear accident, any information predicting the environmental behaviour of ^{137}Cs in the soil subsurface is important to avoiding the health-associated risks of radionuclides.

Some studies have been conducted on the effect of soil minerals on Cs adsorptions on soils. Houmani et al. conducted a similar study for ^{226}Ra [11], we extend their work to understand the effects of Cs adsorption on tropical soils.

According to Staunton and Levacic [21], the entry of radiocesium into the human food chain is strongly controlled by its adsorption on the soil. Within the soil, clay minerals are known to be the dominant adsorbents of Cs [4]. In particular, the frayed edge sites of illite and vermiculite can fix Cs strongly [3]. These studies were conducted on temperate soils. But soil mineralogy is subject to originated materials and weather conditions. Tropical soils are mainly acid soils and their mineralogy is dominated by 1:1 mineral such as Kaolinite, iron or aluminium oxides.

Thus, we evaluated the sorption behaviour of ^{137}Cs on Malaysian soil to understand the factors affecting ^{137}Cs sorption in tropical soils. We collected and evaluated soil

samples from Durian, Holyrood, Rengam, Segamata, and Selangor. All of these sites are located in Malaysia.

In the following sections, we describe our study in detail.

2. Theory and Experimental

In this section, we give the details of our study. In particular, we describe our method for collecting and preparing our samples and our analyses of the samples. We refer to the relevant theories as we describe our experiment.

2.1 Sample collection and preparation

As mentioned earlier, we collected different pristine soil samples from five locations. We collected:

- 1) Haplic Acrisol (siltic) from Durian, Malaysia;
- 2) Haplic Acrisol (aluminic) from Holyrood, Malaysia;
- 3) Haplic Nitisols (dystric) from Rengam, Malaysia;
- 4) Acric Ferralsols (rhodic) from Segamat, Malaysia; and
- 5) Haplic Gleysols (dystric, clayic, drainic) from Selangor, Malaysia.

We first cleared the detritus (grass and litter) and dug the soil with a spade to the depth of 1 m. We then collected soil samples from depths of 0–20 cm, 20–40 cm, and 40–60 cm. In addition, at each depth, we took soil cores (7.6 cm diameter and 4 cm deep) for bulk density determination. Furthermore, at each depth considered, we collected about 500 g of each soil sample in a sealed plastic bag and transferred the bags to the laboratory for subsequent analysis. The samples were then air-dried at room temperature, pounded using a wooden mortar, and sieved through a 2 mm mesh for further use or analysis.

2.2 Physical and chemical Analysis

We examined the sieved soil samples from the three depths to determine their physico-chemical properties. We determined the bulk density, porosity, soil-particle-size

distribution, pH, exchangeable acid cations (Al^{3+} and H^+), exchangeable base cations (Ca^{2+} , Mg^{2+} , K^+ and Na^+), cation exchange capacity (CEC), extractable iron, manganese, and sulphate content of the samples. We also determined the specific surface area, total organic carbon (TOC), and organic matter content of the soil samples.

We calculated soil porosity as the difference between the soil particle density and bulk density; while particle-size distribution was determined using the Pipette method and dry sieving [6]. We obtained the texture of the soils by plotting their percentages of sand, silt and clay; and categorized them 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 [2]. The pH of each sample 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 [13].

We measured the exchangeable acid ions (Al^{3+} and H^+) in 1.0 M KCl extract by titration [17]. The exchangeable base cations were measured in 1.0 M ammonium acetate extract by Flame Atomic Absorption Spectrophotometer (FAAS, model Perkin Elmer 3300) [22] and cation exchange capacity (CEC) was obtained by the summation of the acid and base cations. The base saturation was calculated using the equation ($\sum \text{cations} / \text{CEC} \times 100$).

The extractable Fe and Mn-oxides contents were determined by the dithionite-citrate method [19]. The sulphate content was measured on a sediment water extract [1 g :10 mL] after two hours of shaking and filtration using the barium sulphate turbidimetric procedure [8]. The specific surface area was determined by the ethylene glycol monoethyl ether (EGME) method [24]. We estimated organic carbon using the Walkley and Black method; organic matter (OM) was calculated by multiplying the values of organic carbon by 1.72 [22].

2.3 Clay mineralogical analysis

For the identification of clay mineral constituents in the bulk soil samples, we performed X-ray diffraction (XRD) analysis with a Siemens D500 powder X-ray diffractometer using $\text{CuK}\alpha$ radiation and operating at 40 Kv and 30 mA. The oriented specimens were scanned from 3 to 35 degree 2θ at 1 degree. All of the samples were prepared for both qualitative and quantitative analyses by X-ray diffraction analysis according to the general procedure of Moore and Reynolds [14].

We put about 1 g of soil into a 10 ml measuring cylinder and added about 5 ml of distilled water into it; we then mixed the content of the cylinder thoroughly using a glass rod. After 15–20 minutes, the upper solution was then pipetted (using an eyedropper) and placed slowly on a clean glass slide so that the liquid covered the entire surface of the slide. A porous ceramic plate was used under the slide to catch any spills. 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 should not be used because poorly crystallised clay minerals in soils can be damaged by high temperature. Several treatments have been undertaken to verify the clay minerals in the soil.

2.4 Bath sorption test for the measurement of the distribution coefficient (K_d)

We studied the sorption of ^{137}Cs on the topical Malaysian soil using the batch technique with at least three replicates. The solution-soil ratio was maintained as 20:1 (w/v) throughout the experiments, in accordance with ASTM D4319-93 [1]. 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 ^{137}Cs spike by shaking for 24 h at 23 ± 2 $^\circ\text{C}$ 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 through 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 resolution at 1332 keV energy of ^{60}Co of 2.3 keV (FWHM).

The distribution coefficient, K_d (ml g^{-1}), was then calculated using the following equation:

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

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

Treatment of the raw counting data was performed with GENIE 2000 software. 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 [5]:

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

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

2.5 Data treatment and statistical 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 only dependent factor. The coefficient of correlations and stepwise multiple regressions were calculated to determine the independent factors which were significantly controlled by the dependent factor. The statistical significance was

considered at significant level 0.05 ($\alpha = 0.05$). Therefore, we considered that the regression models and coefficient of correlations of the 5% or less chance of error ($p \leq 0.05$). Our data analysis was performed using the statistical software, Statistical Package for Social Sciences (SPSS) Version 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 were used to derive the relationship and influences between the physico-chemical soil properties and distribution coefficients (K_{ds}).

3. Results and discussion

In the previous section, we described the experiments of our study. Here, we discuss the results. We begin with the mineralogy analysis of the soil samples. We then discuss the

distribution coefficients and the soil properties. We conclude with a discussion of the relationship between the distribution coefficients and some soil properties.

3.1 Soil mineralogy analysis

Table 1 shows the soil clay minerals in the studied soils. In the first column, we show the depths at which soil samples were collected. In the other columns, we show the minerals at each soil sample site (shown as *soil series* in the table).

As shown in the table, for all the soil series, the dominant clay minerals are Kaolinite, Quarts, and Gibbsite. Although some soils contain 2:1 type silicate like Montmorillonite or Illite, this mineralogy indicates that these soils are highly weathered tropical soils and the soil cation exchange capacity is influenced by changes in the soil conditions.

Table (1): The mineralogy of soil used for this study

	Soil series				
	Durian	Holyrood	Rengam	Segamat	Selangor
Order	Ultisols		Oxisols		Inceptisols
soil depth (cm)					
0 - 20	Kaolinite, Quartz and Gibbsite	Illite and Quartz	Kaolinite, Quartz and Orthoclase	Kaolinite, Andesine, Chlorite and a few Quartz	Kaolinite, Quartz and Goethite
20 - 40	Kaolinite, Quartz, Illite, Muscovite	Quartz and Calcite	Kaolinite, Quartz and Albite	Illite, Kaolinite, Andesine and Apatite	Kaolinite, Montmorillonite and Quartz
40 - 60	Kaolinite, Quartz, Muscovite	Illite, Quartz, Gibbsite and Chlorite	Kaolinite, Quartz and Orthoclase	Kaolinite, Biotite, Goeyhite and Andesine	Kaolinite, Quartz and Goethite

3.2 K_d ranges of ^{137}Cs and soil properties

In Table 2, we show the K_d ranges of ^{137}Cs and the soil physical properties across the five sites shown in the table as *soil series*. Each row shows the K_d range for a given soil physical property with the maximum K_d shown in the row labelled *maximum* and the minimum K_d for the soil property is shown as *minimum* in the table. The geometric mean of the K_{ds} is given as *mean* in the table. The first row of the table gives the K_d range and the geometric mean for all the physical properties in each soil sample. The number of soil samples analysed for each site is shown as *n* in the table.

For all the soils analysed, K_{ds} range from 202 L kg⁻¹ to 5919 L kg⁻¹ and the geometric mean is 1039 L kg⁻¹. This range is narrower than the values reported for Japanese soil (215 L kg⁻¹ – 43400 L kg⁻¹, geometric mean of 2210 L kg⁻¹) [15] and those reported for temperate environments (2.7×10^2 L kg⁻¹ – 3.6×10^5 L kg⁻¹) [12]. Moreover, the geometric mean of the K_{ds} for Malaysian soil was lower than the values known for temperate soils. For the soil physical properties, we found a big difference between the soil series for soil-particle-size distribution. Holyrood soils and Rengam soils are sandy soils; the other soils have high clay content.

Table (2): Soil classification, K_d range of ^{137}Cs , and soil physical properties for tested the soils

		Soil Series					
		Durian (n=9)	Holyrood (n=9)	Rengam (n=9)	Segamat (n=9)	Selangor (n=9)	All soils (n=45)
Soil Classification**		Typic Hapludult	Typic Paleudult	Typic Kandiudult	Rhodic Acrucox	Sulfic Endoaquept	
K_d ^{137}Cs range	mean*	1935	716	495	373	4572	1039
	Maximum	3231	1349	1732	546	5919	5919
	Minimum	958	274	202	217	3390	202
Sand%	mean*	10	80	52	6	10	19
	Maximum	14	85	57	13	14	85
	minimum	8	72	47	2	7	2
Silt%	mean*	27	3.4	14	20	18	14
	maximum	42	9.0	17	23	26	42
	minimum	20	2.0	11	18	14	2
Clay%	mean*	61	16	34	73	71	44
	maximum	72	20	40	78	78	78
	minimum	46	13	26	64	60	13
Bulk Density (g/cm)	mean*	1.01	1.02	1.05	0.98	1.03	1.02
	maximum	1.05	1.09	1.12	1.06	1.11	1.12
	minimum	0.96	0.95	0.97	0.87	0.96	0.87
Porosity (g cm ⁻³)	mean*	58	60	59	63	59	60
	maximum	59	63	61	67	61	67
	minimum	57	59	57	60	55	55
Specific surface Area (m ² /g)	mean*	46	39	77	93	111	68
	maximum	56	68	98	103	128	128
	minimum	33	13	46	81	98	13

*Geometric mean was used because the K_d values showed log-normal distribution.

** Soil Taxonomy [20]

Table 3 is similar to Table 2. While Table 2 is about the physical properties of the studied soils, Table 3 is concerned with the chemical properties. For the chemical properties, all of the studied soils are acid soils and the pH ranges between

3.4 and 4.8; Selangor soil shows a particularly low pH range. For the other soil properties, no big difference was observed between the soil series.

Table (3): Soil classification, K_d range of ^{137}Cs , and soil chemical properties for the tested soil

		Soil Series					All soils (n=45)
		Durian (n=9)	Holyrood (n=9)	Rengam (n=9)	Segamat (n=9)	Selangor (n=9)	
Soil classification**		Typic Hapludult	Typic Paleudult	Typic Kandudult	Rhodic Acrudox	Sulfic Endoaquept	
K_d ^{137}Cs range	mean*	1935	716	495	373	4572	1039
	maximum	3231	1349	1732	546	5919	5919
	Minimum	958	274	202	217	3390	202
pH	mean*	4.0	4.2	4.1	4.4	3.5	4.0
	maximum	4.0	4.7	4.2	4.8	3.8	4.8
	minimum	3.8	4.0	4.0	4.2	3.4	3.4
Total organic carbon (%)	mean*	0.80	0.68	1.04	0.49	0.58	0.69
	maximum	1.09	2.43	1.69	0.57	2.66	2.66
	minimum	0.42	0.18	0.69	0.43	0.11	0.11
CEC (meq/100g soil)	mean*	6.56	4.86	5.85	7.17	7.84	6.37
	maximum	7.29	5.66	6.83	8.64	11.69	11.69
	minimum	5.15	4.11	5.27	5.53	4.76	4.11
Base saturation (%)	mean*	9.7	6.4	8.0	20.1	22.1	11.7
	maximum	16.3	14.6	15.5	40.8	38.1	40.8
	minimum	2.5	2.8	4.6	10.5	14.4	2.5
Fe oxides ($\mu\text{g.g}^{-1}$)	mean*	6829	4268	5570	8539	2902	5259
	maximum	7866	5897	6683	9135	3727	9135
	minimum	5353	2524	4741	8094	2459	2459
Mn oxides ($\mu\text{g.g}^{-1}$)	mean*	147	168	122	322	196	180
	maximum	220	214	191	362	252	362
	minimum	108	134	72	260	157	72

*Geometric mean was used because the K_d -values showed log-normal distribution.

** Soil Taxonomy [20]

3.3 Relationships between K_{ds} of ^{137}Cs and soil properties

The correlation coefficients and statistical significance of the relationships between the K_{ds} of ^{137}Cs and soil properties are shown in Table 4. A row of the table shows a soil property in the first column of the table and the corresponding correlation coefficient and statistical significance are shown in the second column and the third column respectively.

Table (4): Correlation coefficients between K_d -Cs and soil properties

	R^2 value (n=45)	Statistical significance
Sand	-0.173	**0.5%
Silt	0.038	no
Clay	0.207	**0.5%
Bulk Density	0.048	no
Porosity	-0.177	**0.5%
Specific surface area	0.116	*5%
pH	-0.594	***0.05%
TOC	0.020	no
CEC	0.362	***0.05%
Base Saturation	0.064	no
Fe oxides	-0.282	***0.05%
Mn oxides	-0.016	no

In Fig. 1-5, we show the relationships between K_{ds} of ^{137}Cs and some soil properties: Fig. 1 gives the graph for K_{ds} and Sand%; Fig. 2 is about K_{ds} and Clay%; Fig. 3 is about K_{ds} and

pH; Fig. 4 is on K_{ds} and CEC; while Fig. 5 is for K_{ds} and Fe oxides. For each graph, we show the K_d values on the Y axis and the mineral content on the X axis.

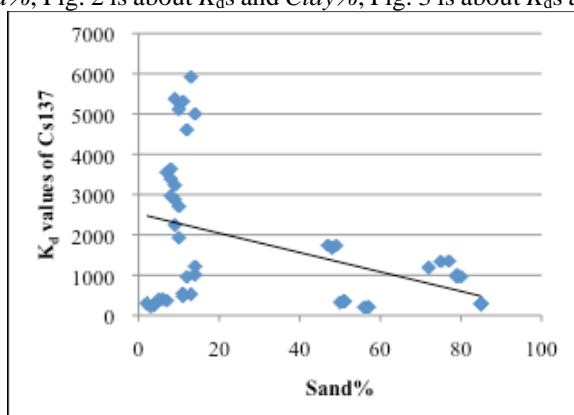


Fig (1): Relationship between K_d values of ^{137}Cs and Sand%

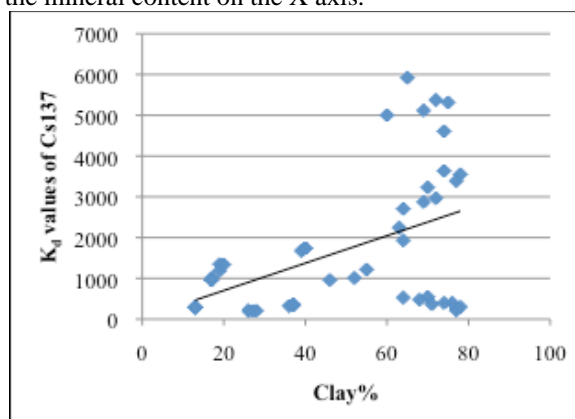


Fig (2): Relationship between K_d values of ^{137}Cs and Clay%

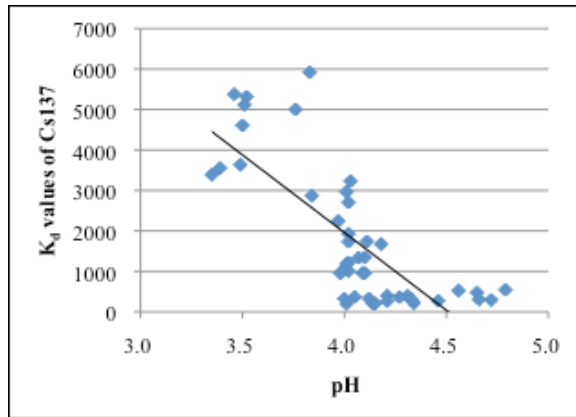


Fig (3): Relationship between K_d values of ^{137}Cs and pH

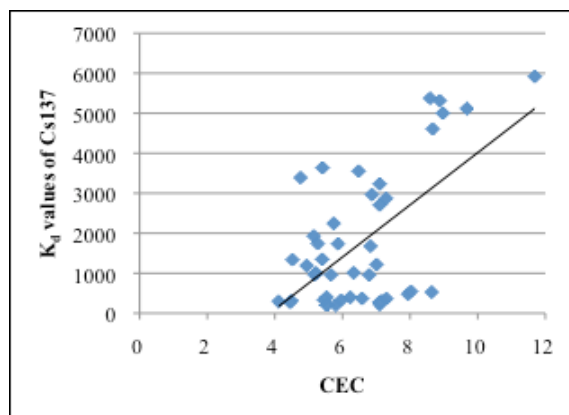


Fig (4): Relationship between K_d values of ^{137}Cs and CEC

As can be observed from the figures, among all the soil properties considered, strong positive correlations with the K_d s were obtained for *CEC* and *Clay%*; while strong negative correlations with the K_d s were obtained for *pH* and *Fe oxides*. The positive correlations of *CEC* and *Clay%* with ^{137}Cs K_d s indicate that the ^{137}Cs sorption in acid tropical soil is mainly due to the surface charge of clay mineral. We recorded high value of ^{137}Cs K_d s for Segamat soil series. This can be explained by the high clay mineral content and corresponding high CEC value (shown in Table 2 and Table 3). Moreover, high values for *Fe oxides* is the typical property of highly weathered acid soil; and the negative correlations of *Fe oxides*

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and *pH* with ^{137}Cs K_d s suggest that highly weathered acid tropical soil contains high ^{137}Cs sorption capacity.

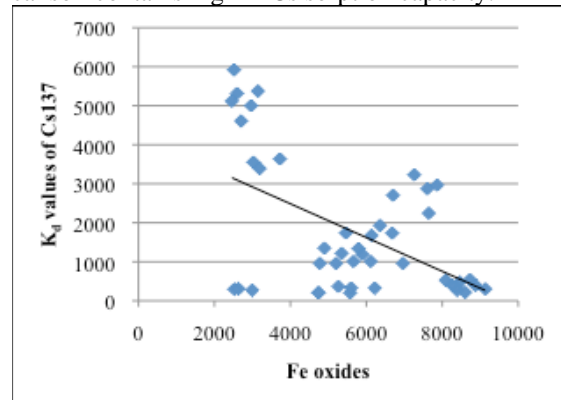


Fig (5): Relationship between K_d values of ^{137}Cs and Fe oxides

As shown in Table 1, all of the soils studied have variable charge minerals and their surface charges are decreased by low *pH* conditions. Therefore, the negative correlation of *pH* with ^{137}Cs K_d s cannot be explained by the direct effect of *pH* on ^{137}Cs sorption. Many previous reports indicate the inhibition of ^{137}Cs sorption by the competition with potassium or other alkaline metals. It is well-known, however, that the decrease in exchangeable K causes the release of the nonexchangeable K from phyllosilicate minerals in the rhizosphere [9–10, 17], and the decrease in K concentration increases Cs sorption in soil [7, 15]. Thus, we can conclude that in strongly weathered tropical soils, the decrease in major alkaline metals like K or Na causes an increase in ^{137}Cs sorption capacity.

4. Conclusion

We have presented our study of the tropical Malaysian soil. Our experimental results show some contrasting results to those of similar studies conducted for the temperate soils. For the acid tropical soils in Malaysia, we found the clay mineral amount and its CEC to be the major factors affecting ^{137}Cs sorption. The ^{137}Cs sorption level tends to be high in strongly weathered acid tropical soil. This is likely to be the result of the decrease in alkaline metals with the weathering process. We believe that our study will be useful to those wishing to understand the impact of ^{137}Cs sorption on tropical soils.

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