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CORRELATION BETWEEN ^{137}Cs AND ^{40}K CONCENTRATION IN SOIL AND TEA TREE IN LUONG MY FARM, HOA BINH PROVINCE, VIETNAM

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Abstract. *In this work, the correlation between ^{137}Cs and ^{40}K concentrations in tea tree and soil in Luong My farm, Tan Thanh district, Luong Son commune, Hoa Binh province was experimentally investigated. The measurements were carried out using gamma spectroscopy with a high purity germanium (HPGe) detector. The results showed that ^{40}K is uniformly distributed in the soil depth while ^{137}Cs is mainly located in 10 cm of the soil surface. Soil-plant transfer factor (TF) for ^{40}K varies in the range of 0.491 to 0.623 and that for ^{137}Cs is in range of 0.384 to 0.510. The concentration of these two radionuclides in tree parts is opposite to each other, while ^{40}K is concentrated in the leave, ^{137}Cs is mainly found in the root.*

Keywords: Gamma spectroscopy; soil-plant transfer factor; tea tree; ^{40}K; ^{137}Cs.

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I. INTRODUCTION

In the enviromental radioactivity study, ^{137}Cs and ^{40}K are radionuclides of the most interest. ^{40}K is primordial radioisotope and exists in soil, rock and in all living organisms, even in the human body. Potassium is found in all part of a tree, particularly in trunk, branch, and shoot. It influences the metabolic process and enhances enzyme activity and increases the accumulation of glucid and amino acid. Potassium also helps to increase the water retention ability of the plant cell [1]. ^{137}Cs is the fission product of ^{235}U and other fissionable isotopes in a nuclear reactor and nuclear weapon. This isotope induces long-term radioactive effect due to its long half-life (30.17 years) and it also has high bioavailability [2]. It is the alkali metal with low hydration and having chemical similarities to potassium. Cesium mainly presented in solution in free hydrated cations thich means that Cs^{+} has little or no tendency to form soluble compound. Cesium is easily absorbed by tree root and then transferred to other tree parts above soil surface [3].

Radionuclides in the terrestrial, atmosphere and water can induce bad effect to human health after entering the human body by inhalation or ingestion of food, in which vegetables play an important role [4]. Therefore, study soil-plant transfer process is a meaningful work. Plant uptake of radionuclide from the soil is evaluated by using transfer factor which is defined as the ratio between the activity of radionuclides in plant (in Bq.kg^{-1} dry plant) and activity of radionuclide in soil (in Bq.kg^{-1} dry soil) [5,6].

The transfer processes of potassium and cesium from soil to plant show competition and attracting many researches and some proved that among alkali metal and NH_4^+, K^+ is an important cation that is competitive with Cs^+ in uptake [7,8]. Researches carried out in some plants of semi-natural grass field at Friuli-Venezia Giulia, Italy even showed that potassium screens soil-plant uptake of cesium [9].

Soil-plant transfer of cation is influenced by the soil's features such as organic, clay content and its pH. Implementation of NH_4^+ or decreasing pH will make TF of ^{137}Cs increased and vice verse, this fact is explained as appearance of H^+ will make liberation of the metal and in the soil gel, increasing their solubility and enter to the tree root. ^{137}Cs is known as an element which is bound tightly in the clay, hence clay content increased will screen soil-plant transfer of the ^{137}Cs. Besides, the study carried out in radish from Japan shows that more organic content in the soil will enhance the transfer of the ^{137}Cs from soil to plant [10,11].

Competition in absorption ^{137}Cs and ^{40}K of the plant is reasonable which imply to contrary accumulation of these two isotopes in the plant different part. In the plant, ^{40}K is mainly concentrated in the leaf which is the main part of a tree participating in the photosynthesis process and it prevents ^{137}Cs transfer to the leaf, hence ^{137}Cs is mainly found in the root. Furthermore, in tree parts, potassium has an influence in controlling and adjusting endosmosis of plant cell in early development stage then carbohydrate will play the role so that concentration of potassium is decreased and that of ^{137}Cs is increased. Competition of ^{40}K and ^{137}Cs in the soil-plant transfer is in the aspect of atomic radius, soluble possibility. Many studies have shown that soil-plant transfer factor for ^{137}Cs is smaller than that of ^{40}K.

In our study, soil-plant transfer for tea tree in Luong My farm, Hoa Binh province was investigated. Furthermore, in order to evaluate absorption efficiency of cesium in comparison to potassium, we used a quantity so-called Cs/K discrimination factor (DF), values of DF below unity indicate that K is more efficiently absorbed than Cs. Most reported Cs/K DFs in plants exposed to the nutrient solution are below 1 [3]. In addition, the correlation of concentration of these two radionuclides in soil and plants was assessed by using the Spearman's rank correlation coefficient. This quantity has a value between -1 and 1. A correlation coefficient of 0 (or near 0) means that the two variables have no relation to each other; conversely if a coefficient of -1 or 1 means the two variables have an absolute relationship. If the value of the correlation coefficient is negative ($\rho < 0$), it means that when one variable increases, the other decreases (and vice versa); If the correlation coefficient value is positive ($\rho > 0$), it means that when this variable increases, the other variable increases, and when it decreases, the other variable decreases [12].

Our work is organized as follow: Sec. I is Introduction. Research object and method are presented in Sec. II. Results of our work are described in Sec. III and Sec. IV is the conclusion of our work.

II. RESEARCH OBJECT AND METHOD

II.1. Sampling area

Luong My tea farm was selected for our study. This is a low semi-mountainous area having altitude above sea level of 50-80 m, with slope is about 2–3%. It has a low mountain band which was formed by magma stone, limestone, and terrigenous sediment. The climate of this area is typically monsoon tropical. Winter-spring usually is from November to March and summer-autumn is from April to October and the average rain level is about 1.760 mm [5]. The sampling area is quite flat, the height difference between sampling position in comparison to sea level is ignorable. (see Fig. 1).

Fig. 1. Mapping of the sampling location, Luong Son, Hoa Binh, Vietnam.

II.2. Tea tree of Luong My farm

As mentioned, the object of our research is tea tree, its leaf is popularly used to make a drink in Vietnam. Tea tree is usually found in tropical and near-tropical climate. It is a neutral one in the young stage while an adult tea tree adapts very well with sunshine. Under shadow, tea leaf has dark green colour and less shoot due to weak photosynthesis. The scattered light in a high mountain area has better influence on tea quality than direct light. In the foggy, wet and cool weather together with the temperature difference between day and night are a good condition for having high-quality tea leaf. Usually, most suitable rainfall for growing up of tea tree is about 1500-2000 mm, air humidity is about 80–85% is good for tee root growth. For tea trees planted by seed, it normally has tap-root, lateral and absorbed. Tap-root usually has length of 1 m, depending on the soil character and processing method, manured manner, tea tree age and it's species. Tap-root does not exist in tea tree which is planted by using tea branch. The lateral and absorbed root are distributed mainly in the depth from 5 to 50 cm and their horizontal distribution is about two times of tree shadow area (see Fig. 2). For the tea tree which is planted by using a branch, the lateral root is well developed. In the natural growth condition, tea trunk is a single and straight, its branches are arisen continuously to form a branch and shot system [13]. In our work, tea trees selected for study are over 20 years old which were planted in the period from 1981 to 1995. In this farm, tea tree branches are cut and tree is fertilized twice per years, one in early season about on February and other on November, at the last of the season.

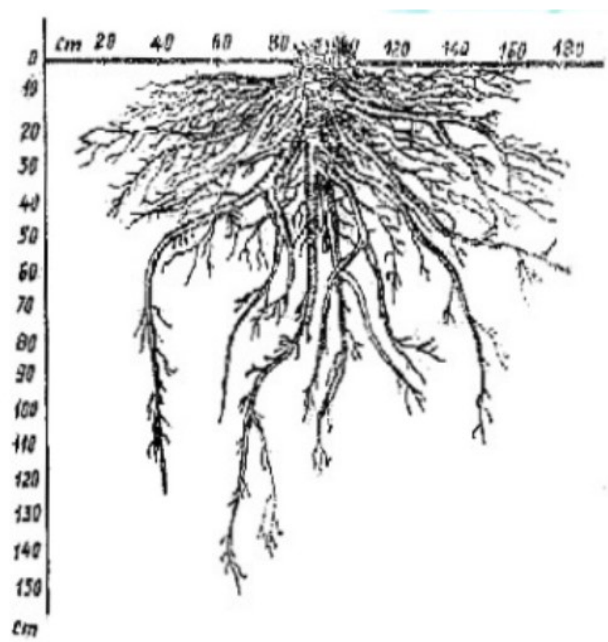

Fig. 2. Root system of mature tea tree [13].

II.3. Sampling method and position

In this work, 14 tea trees and corresponding 14 surrounded soil samples were collected from 14 different positions. Distance between sampling positions is about 700-1500m. Soil was sampled in 4 points within the area of 1 m^2 around the tea tree and in 5 depth layers of 10 cm,

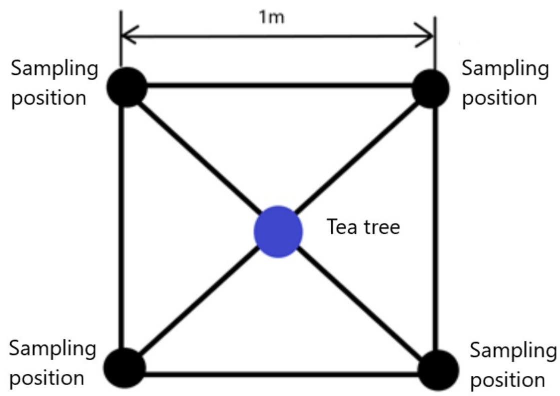

Fig. 3. Soil sampling scheme.

20 cm, 30 cm, 40 cm and 50 cm by using special sampling tool (see Fig. 3), then removed stone, tree root and putted into plastic boxes. Tea tree samples with full three parts as root, trunk and leaf were washed and then put in the plastic bag. In 14 sampling positions, 42 samples of the tea tree parts and 70 soil samples in 5 different depth layers were collected. The information about samples and their labels are shown in Table 1.

Table 1. Information of the collected samples.

Position	**Sample label**									
	Tree				**Soil**					
	Leaf	**Trunk**	**Root**	**Whole tree**	**10cm**	**20cm**	**30cm**	**40cm**	**50cm**	**Average of 5 soil layers**
1	L1	T1	R1	C1	D1-10	D1-20	D1-30	D1-40	D1-50	D1
2	L2	T2	R2	C2	D2-10	D2-20	D2-30	D2-40	D2-50	D2
3	L3	T3	R3	C3	D3-10	D3-20	D3-30	D3-40	D3-50	D3
4	L4	T4	R4	C4	D4-10	D4-20	D4-30	D4-40	D4-50	D4
5	L5	T5	R5	C5	D5-10	D5-20	D5-30	D5-40	D5-50	D5
6	L6	T6	R6	C6	D6-10	D6-20	D6-30	D6-40	D6-50	D6
7	L7	T7	R7	C7	D7-10	D7-20	D7-30	D7-40	D7-50	D7
8	L8	T8	R8	C8	D8-10	D8-20	D8-30	D8-40	D8-50	D8
9	L9	T9	R9	C9	D9-10	D9-20	D9-30	D9-40	D9-50	D9
10	L10	T10	R10	C10	D10-10	D10-20	D10-30	D10-40	D10-50	D10
11	L11	T11	R11	C11	D11-10	D11-20	D11-30	D11-40	D11-50	D11
12	L12	T12	R12	C12	D12-10	D12-20	D12-30	D12-40	D12-50	D12
13	L13	T13	R13	C13	D13-10	D13-20	D13-30	D13-40	D13-50	D13
14	L14	T14	R14	C14	D14-10	D14-20	D14-30	D14-40	D14-50	D14

II.4. Sample preparation

The collected soil samples were exposed under sunshine for naturally drying. After that, these samples were dried at 110°C in the oven until weight changing less than 1%. The dried soil samples were removed remaining stone by using a sieve with hole diameter of 1 mm and then ground to achieve the particle dimension of 0.25 mm. Full part tea tree samples were washed carefully to remove the soil and dust by clean water and then were dried at 110^{0}C and also finely ground to obtain particle dimension of 0.5 mm. The processed samples were mixed carefully to ensure homogeneity and packed in a cylinder box with a diameter of 74 mm and height of 30 mm. Weight of soil samples are 180 g and that of tea tree ones are 100 g. All these samples were sealed for 4 weeks to establish secular equilibrium before measurement.

II.5. Sample measurement and data analysis

Samples were measured by the CANBERRA lead shielded low-background gamma spectroscopy with high purity germanium detector HPGe having a resolution of 1.86 keV at 1333 keV photopeak of ^{60}Co and relative efficiency is 15%. In order to accumulate enough statistics and reduce statistical error, measurement time for soil samples was 100.000 seconds while that for the plant samples was 150.000 seconds. In addition to that, the background measurement time was 100.000 seconds. The software Geniee2K was used for data acquisition and spectrum analysis. The activities of ^{40}K and ^{137}Cs in the samples were deduced by our own developed method using only one absolute value of the efficiency at energy of 1460 keV corresponding to characteristics gamma-ray of ^{40}K and relative efficiency curve [14]. The typical measured gamma spectrum of the soil and tea tree samples are shown in Figures 4 and 5.

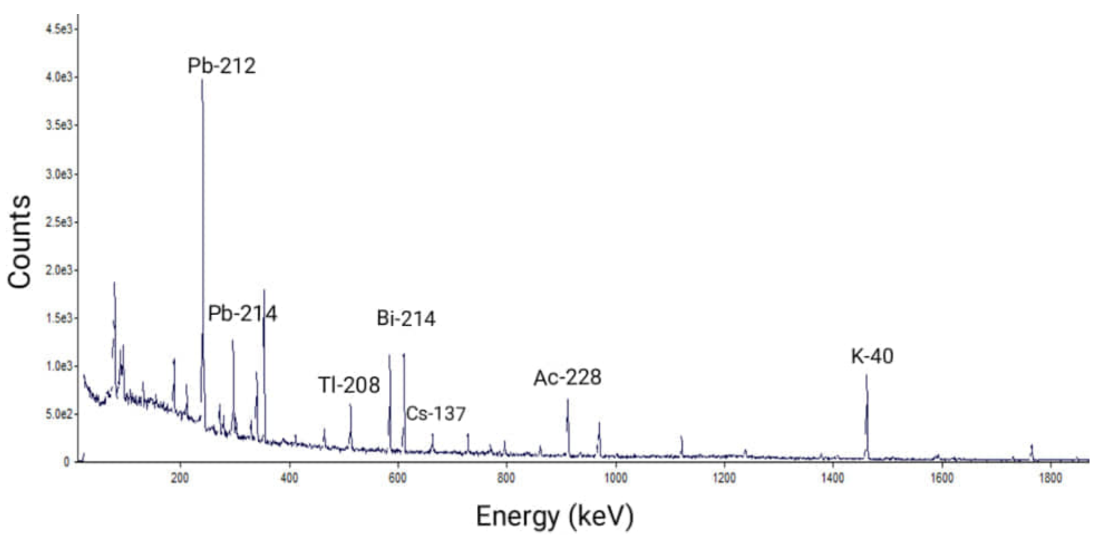

Fig. 4. Gamma spectrum of soil sample at position 1 (layer 0-10cm).

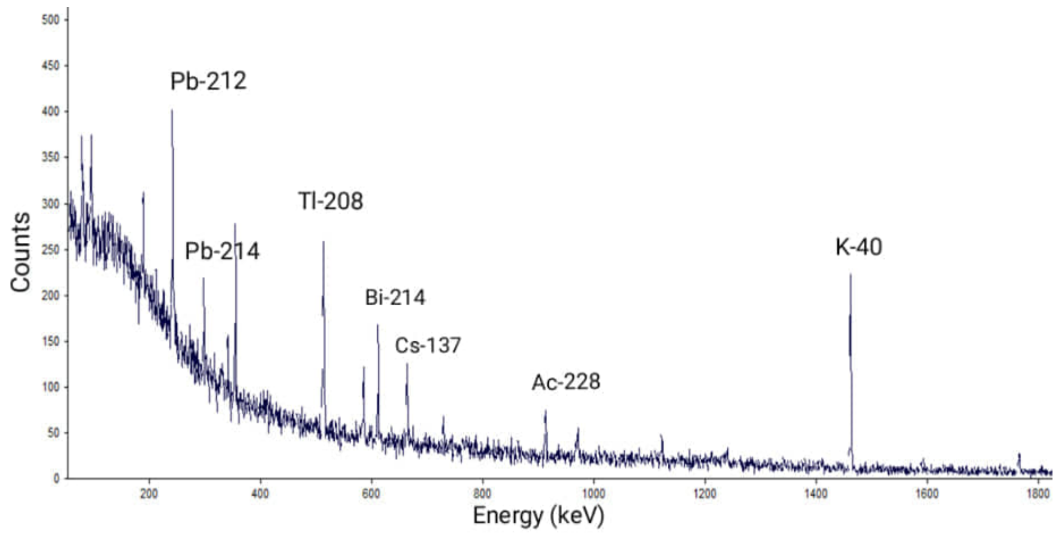

Fig. 5. Gamma spectrum of root sample at position 1.

Soil-plant transfer of radionuclides is evaluated using transfer factor (TF) which is defined as the ratio between the activity of radionuclides in the plant (in Bq.kg^{-1} dry plant) and activity of radionuclide in soil (in Bq.kg^{-1} dry soil) [5,6].

In addition to that, we have used discrimination factor (DF) Cs/K to determine the contrary correlation in the soil-plant transfer of Cs and K, DF is defined as follows [3]:

$$DF = \frac{C_{Cs-P}/C_{K-P}}{C_{Cs-S}/C_{K-S}}. \tag{1}$$

Here, C_{Cs-P} and C_{K-P} are concentration of ^{137}Cs and ^{40}K in the plant, C_{Cs-S} and C_{K-S} are that of ^{137}Cs and ^{40}K in the soil.

Furthermore, the analysis included investigating the distribution of ^{137}Cs and ^{40}K in soil layers, the correlation between ^{137}Cs and ^{40}K activities in the soil and tea tree parts were performed (see next session).

To evaluate the relationship between K and Cs activity in soil and tea, Spearman's rank correlation coefficient was calculated.

Uncertainties of all measurements were calculated taking into account random and systematic components of uncertainty, i.e. uncertainties due to sample preparation, efficiency calibration, measurement of sample and nuclear data [15]. The total uncertainty was calculated by uncertainty propagation equation.The uncertainties were presented at the 95% confidence level.

III. RESULTS AND DISCUSSION

Many experimental studies showed that the soil-plant transfer of radionuclide depends very much on the physical and chemical properties of the soil. Therefore, we have first analyzed soil to evaluate the soil character by the laboratory of the University of Science, Vietnam National University, Hanoi and the obtained results are shown in Table 2.

Table 2. The physical and chemical properties of the soil at tea farm Luong My.

	Physical and chemical characteristics of research land							
Sample notation	Sand (%)	Limon (%)	Clay (%)	Humus (m_e/100 g soil)	Ca^{2+} (m_e/100 g soil)	Mg^{2+} (mg/kg)	Fe^{3+}	pH
D1	22.7	36.5	40.8	3.48	4.04	2.87	84.1	5.86
D2	23.7	38.2	38.1	3.57	4.05	2.98	85	6.21
D3	28.6	36.1	35.3	2.0	6.79	4.23	62	5.96
D4	26.1	36.7	37.2	2.1	6.68	4.13	63.1	6.05
D5	25.6	34	40.4	2.11	4.72	3.28	70	6.12
D6	23.3	32.1	44.6	2.0	4.67	3.36	69.2	5.97
D7	18.9	36.5	44.6	2.36	4.34	3.02	79	5.85
D8	19.7	37.1	43.2	2.35	4.2	3.05	78	6.08
D9	26.4	35.2	38.4	3.10	4.02	2.99	80.2	5.89
D10	22.1	34.9	43	3.22	4.1	2.89	78.9	6.14
D11	19.8	40.1	40.1	3.41	4.13	2.78	80.2	6.23
D12	20.1	36.2	43.7	3.0	3.98	3.02	75.1	5.75
D13	25.6	39.8	34.6	3.55	4.51	3.1	76.8	5.92
D14	18.8	37.5	43.7	3.05	4.46	3.14	77.8	6.03

With these results, we could conclude that tea tree collected from 14 different positions were grown in the similar conditions.

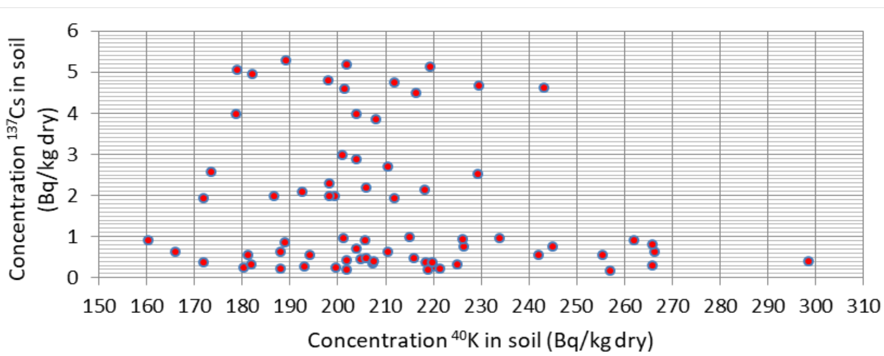

Fig. 6. Scatterplot of ^{40}K versus ^{137}Cs concerntrations in soil (Bq/kg dry).

In all soil and tea tree samples, artificial radionuclide ^{137}Cs was determined. The measured activity of ^{40}K in the soil layers varied from 172 to 298 Bq/kg while the activity of ^{137}Cs was found in a wide range, the ratio between maximum value (5.28 Bq/kg) and minimum one (0.17 Bq/kg) is 31. Although these radionuclides are both alkali, while ^{40}K is the primordial radionuclide, ^{137}Cs is fall out one which is consequence of the nuclear bomb testing or nuclear power plant accident. The measured activities of ^{40}K together with ^{137}Cs in the soil are shown in Fig. 6 and as can be seen, the concentrations of these two isotopes are scattered, it means that there is no dependence between them in the soil.

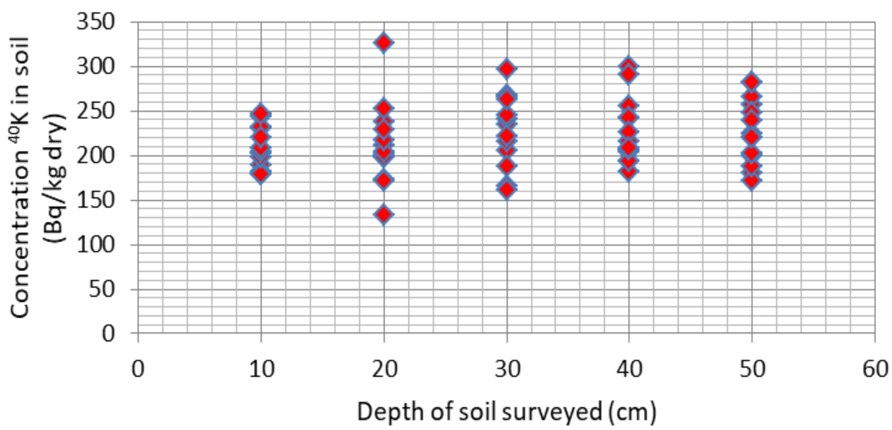

Fig. 7. Concentration of ^{40}K in soil layers.

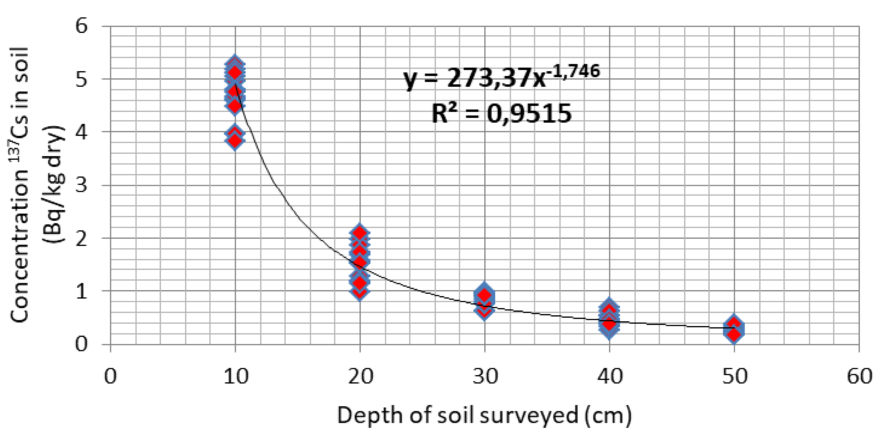

Fig. 8. Concentration of ^{137}Cs in soil layers

Analyzing the concentration depth profile shown that ^{40}K is distributed quite uniformly in different soil layers (see Fig. 7) while concentration of ^{137}Cs has a decreasing tendency by depth (see Fig. 8), ^{137}Cs concentrate mainly in the first 10 cm soil layer and decreased fast from 20 cm depth. Average activity of ^{137}Cs at 10 cm layer is higher than that at 20, 30, 40 and 50 cm: 2.0, 5.7, 9.9 and 17.6 times respectively. The concentration of ^{137}Cs at 50 cm depth layer is very small, the average value corresponding to 14 investigated positions is 0.27 Bq/kg dried soil. For tea trees, as mentioned before their three parts leaf, trunk and root were processed separately. The measured concentration of ^{40}K and ^{137}Cs in the above parts of the tea tree are shown in Table 3.

Table 3. ^{40}K and ^{137}Cs concentration in different parts of the tea tree (Bq/kg dry).

^{40}K							
R1	75.11±2.2	T1	78.33±3.1	L1	184±5.6	C1	112.48
R2	78.14±1.9	T2	83.02±2.1	L2	194.34±3.9	C2	118.50
R3	78.74±2.3	T3	81.01±3.2	L3	182±5.9	C3	113.91
R4	68.74±1.9	T4	77.45±2.2	L4	198.2±3.8	C4	114.79
R5	51.79±2.1	T5	78.91±2.3	L5	188.24±3.9	C5	106.31
R6	69.32±2	T6	84.08±1.8	L6	199.27±6.4	C6	117.5
R7	58.11±2.8	T7	75.73±2.9	L7	172.4±5.4	C7	102.08
R8	61.37±2.1	T8	67.61±3.1	L8	186.1±5	C8	105.02
R9	82.4±2.5	T9	80.6±2	L9	196.4±4.4	C9	119.8
R10	75.6±3	T10	98.3±2.2	L10	178±1.9	C10	117.3
R11	85.3±2	T11	89.4±2.2	L11	188±3.1	C11	120.9
R12	69.4±2.5	T12	90.5±3.1	L12	186.5±4.5	C12	115.4
R13	90.8±2.3	T13	81.1±2.6	L13	188.2±4.5	C13	120.03
R14	87.5±3.9	T14	71.6±2	L14	198.7±3.4	C14	119.26
^{137}Cs							
R1	1.05±0.2	T1	0.78±0.06	L1	0.43±0.03	C1	0.753
R2	1.17±0.3	T2	0.49±0.02	L2	0.49±0.01	C2	0.716
R3	1.12±0.6	T3	0.74±0.4	L3	0.54±0.3	C3	0.8
R4	1.07±0.3	T4	0.72±0.02	L4	0.44±0.02	C4	0.743
R5	1.21±0.3	T5	0.73±0.04	L5	0.46±0.04	C5	0.8
R6	1.04±0.4	T6	0.46±0.06	L6	0.66±0.02	C6	0.72
R7	1.55±0.2	T7	0.77±0.01	L7	0.46±0.04	C7	0.926
R8	1.18±0.6	T8	0.89±0.03	L8	0.44±0.02	C8	0.83
R9	0.97±0.2	T9	0.57±0.02	L9	0.52±0.01	C9	0.686
R10	1.12±0.1	T10	0.48±0.02	L10	0.54±0.03	C10	0.713
R11	1.09±0.2	T11	0.61±0.02	L11	0.51±0.01	C11	0.736
R12	1.01±0.02	T12	0.64±0.04	L12	0.51±0.03	C12	0.72
R13	1.09±0.5	T13	0.64±0.03	L13	0.43±0.02	C13	0.72
R14	1.03±0.4	T14	0.55±0.02	L14	0.52±0.02	C14	0.7

The results show that ^{40}K concentration in the root varies from 51.7 to 90.8 Bq/kg dry, this value is in range of (67.6-98.3 Bq/kg dry) for the trunk and in the range of (172-199.2 Bq/kg dry) for the leaf. While all that of ^{137}Cs are (0.97 - 1.21 Bq/kg dry), (0.46 - 0.89 Bq/kg dry) and (0.43-0.66 Bq/kg dry) for root, trunk and leaf respectively. By comparing the concentrations of 40K and 137Cs in three different parts of tea we can see that ^{40}K concentrates most in leaf (see Fig. 9a), in contrary ^{137}Cs was found most in root (see Fig. 9b). Therefore, we can conclude that there is a clear contrary in accumulation of ^{40}K and ^{137}Cs in different parts of tea tree. It however, in order to evaluate the soil-plant transfer of ^{40}K in comparison to ^{137}Cs in these tea trees, we calculated the transfer factor (TF), the obtained TF values of ^{40}K and ^{137}Cs are in range of (0.491 - 0.623) and (0.384 - 0.510) respectively. It means that tea tree in our investigation area, Luong My farm absorbs ^{40}K better than ^{137}Cs.

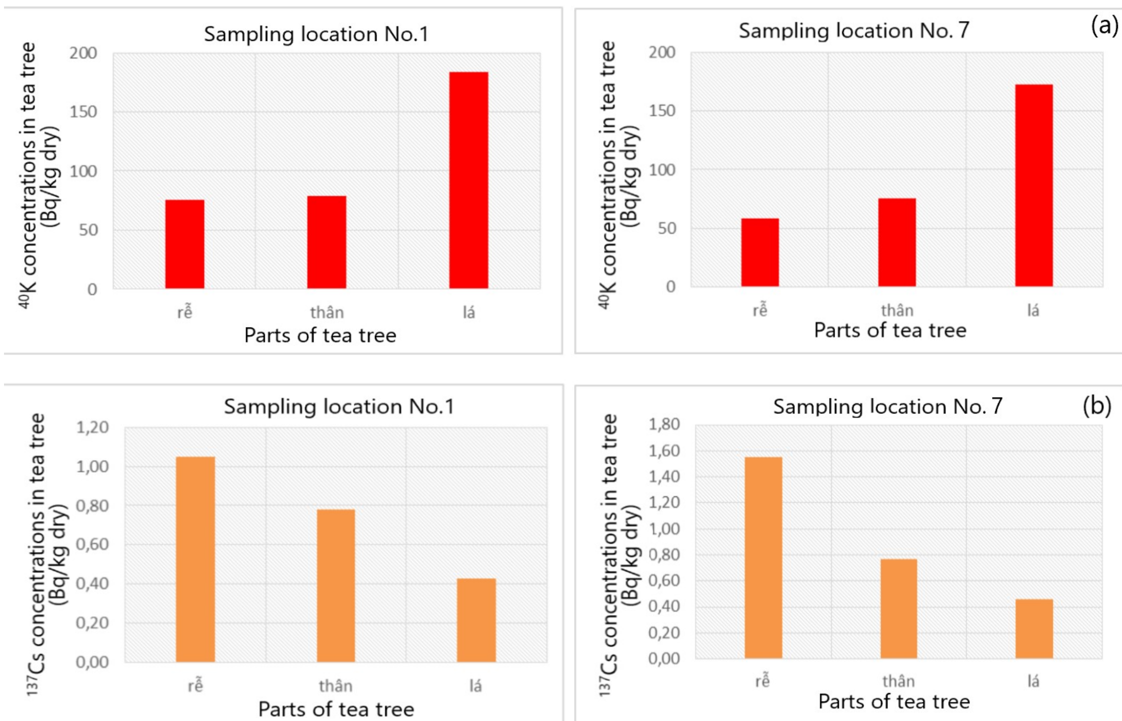

Fig. 9. Concentration of ^{40}K(a) and ^{137}Cs in tea tree parts (b).

We also investigated ^{137}Cs concentration in the tree and it's soil-plant transfer factor TF(Cs) as a function of ^{40}K concentration in the soil (see Fig. 10 and Fig. 11) and calculate the Spearman's rank correlation coefficient, the obtained results are r_s = -0.47 and -0.75, respectively. As can be seen in Fig. 10 and Fig. 11, ^{137}Cs concentration in the tree and TF(Cs) is decreased with increasing of ^{40}K concentration in the soil. However, ^{40}K concentration is changed in a narrow range, the ratio between maximum and minimum values is only 1.2 hence by using results as in Fig. 11, the influence of ^{40}K in the soil to TF(Cs) is not very clear. In order to further evaluate the role of ^{40}K in preventing the absorption of ^{137}Cs of tea tree, we calculated DF Cs/K (see Table 4). In 14 investigated positions, DF Cs/K is smaller than unit. It means that tea tree absorbs ^{40}K better than ^{137}Cs. At the positions 5, 7 and 11, DF Cs/K is near unity which corresponds to low concentration of ^{40}K in the soil (190.6, 192.5 and 193.8 Bq/kg dry respectively). In contrary, at the position DF Cs/K much smaller than unity, ^{40}K concentration is higher.

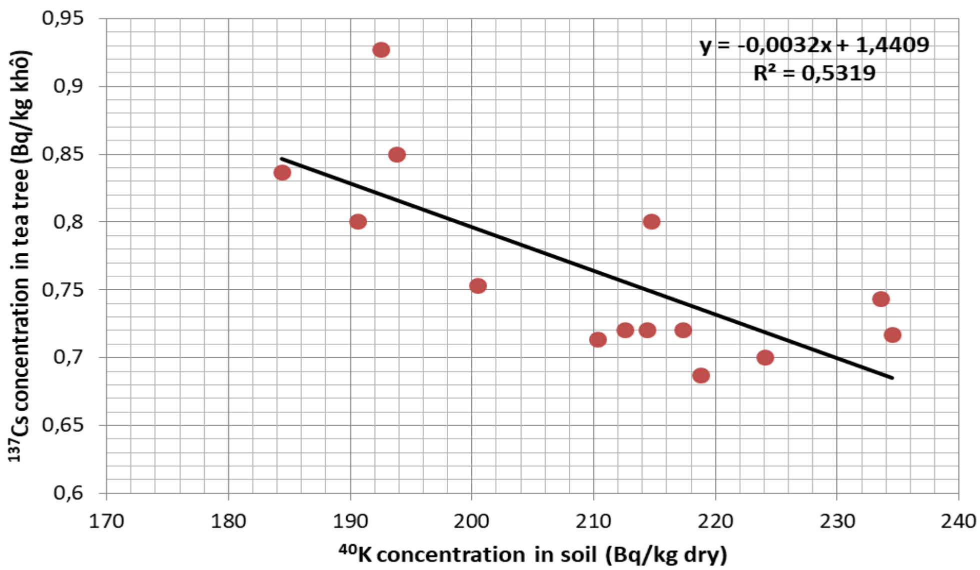

Fig. 10. ^{137}Cs concentration in tea tree as a function of ^{40}K soil concentration with coefficient of determination R^2=0.5319

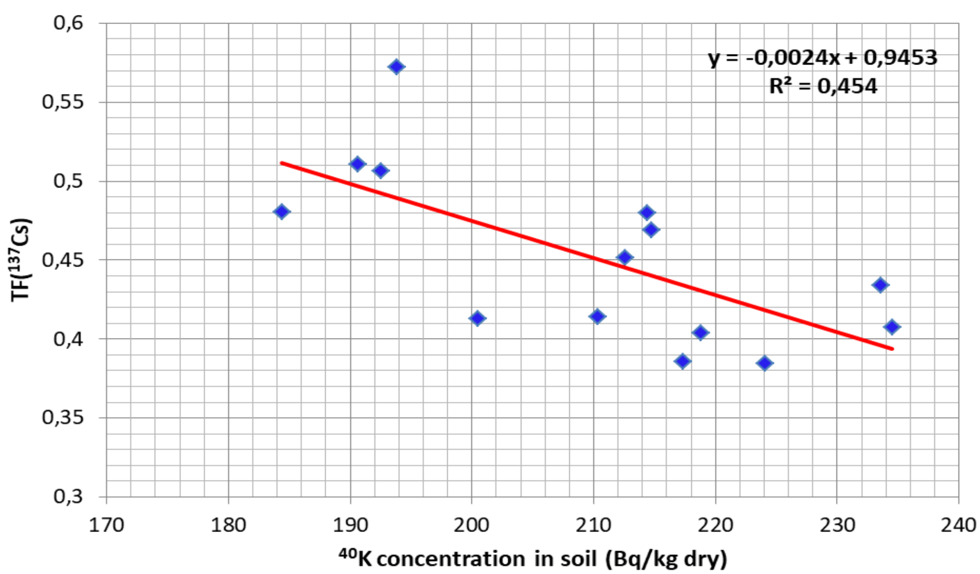

Fig. 11. TF(^{137}Cs) as a function of ^{40}K soil concentration with coefficient of determination R^2=0.454

Table 4. Discrimination factor (DF) Cs/K.

Investigated positions	1	2	3	4	5	6	7
DF Cs/K	0.736	0.806	0.884	0.882	0.916	0.713	0.955
Investigated positions	8	9	10	11	12	13	14
DF Cs/K	0.843	0.738	0.742	0.917	0.832	0.857	0.723

IV. CONCLUSIONS

In our study, we can conclude that there is not a correlation between ^{40}K and ^{137}Cs in the soil. While ^{40}K is distributed uniformly in different soil layers, ^{137}Cs is decreased by depth. Soil-plant transfer of ^{40}K and ^{137}Cs is quite contrary, ^{40}K was mainly found in the leaf but ^{137}Cs was found most in the root. Analyzed data showed that ^{40}K prevents soil-plant transfer of ^{137}Cs for tea tree.

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