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Grass/Soil Concentration Ratios (CRs) of Natural ^{226}Ra, ^{232}Th, and ^{40}K in *Vetiveria zizanioides* (Vetiver Grass) in Thailand

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This work investigated the grass/soil concentration ratios (CRs) of natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in vetiver grass collected from three different regions of Thailand using an HPGe (high-purity germanium) gamma spectrometer. The sampled grass blades were divided into three equal parts to investigate the dependence of grass/soil CRs on the positions of grass blades. The results showed that ^{40}K had the highest grass/soil CRs due to being an essential nutrient for most plants, with ranges of 1.12–7.92, in comparison with ^{226}Ra and ^{232}Th that had ranges of 0.32–1.48 and 0.20–1.31, respectively. In addition, the results indicated that the natural radionuclides investigated were uniformly transferred and accumulated throughout grass blades ($p < 0.05$), which implied the independence of CRs on the section of the leaf. Furthermore, it was found that the soil samples collected from the central region of Thailand had the highest activity concentrations in all radionuclides, mainly due to a primary composition of dark clay, leading to the highest contents of potassium and calcium (Total K and Total Ca, respectively), and the highest cation exchange capacity (CEC) that led to high concentrations of nutrients and radionuclides. Based on the results from this work, the vetiver grass presented potentials to be utilized as radionuclide absorbers in polluted soil and water due to its high CRs in comparison with other reported plants.

Keywords: activity concentration, concentration ratios, gamma spectroscopy, natural radionuclides, vetiver

INTRODUCTION

Vetiver grass (*Vetiveria zizanioides*) is a perennial tussock grass that is native to tropical countries in South and Southeast Asia, which has later expanded into the tropics of both hemispheres. Vetiver grass has a long, massive, and complex root system with large biomass, making it a suitable tool for dryland restoration by reducing soil erosion and surface water runoff (Chen *et al.* 2004; Donjadee *et al.* 2010; Donjadee and Tingsanchali 2013; Gnansounou *et al.* 2017). Vetiver grass also has qualifications for use in economic and environmental-friendly phytoremediation in heavy-metal contaminated soil and water by transferring and accumulating contaminants in its roots and blades, which could reduce considerable amounts of heavy metals in soil and water of concern (Antiochia *et al.* 2007; Aggangan *et al.* 2019; Banerjee *et al.* 2019; Suelee *et al.* 2019). Examples of reports on the capabilities of vetiver grass in soil and water phytoremediation include Chen *et al.* (2004), which showed that planted vetiver grass could adsorb 98, 54, 41, and 88% of the initially applied Pb, Cu, Zn, and Cd, respectively, whereas Roongtanakiat *et al.* (2007)

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indicated that vetiver grass had removal efficiencies of heavy metals in industrial wastewater of 33.7, 27.6, 52.7, 8.9, and 87.5% for Mn, Fe, Zn, Pb, and Cu, respectively. Hence, as these two reports suggested, planting vetiver grass could be an effective tool to improve/restore soil or water conditions and also to prevent the risk of heavy metals entering into groundwater and nearby water resources.

With the known ability of vetiver grass to absorb heavy metals from soil and water, it is interesting to investigate the accumulation of natural radionuclides – especially ^{226}Ra, ^{232}Th, and ^{40}K – to its grass structure by determining the activity concentrations and grass/soil CRs in their grass blades (Hancock *et al.* 2006; Ahl *et al.* 2009; Alsaffar *et al.* 2015; Saenboonruang *et al.* 2018a, b). As mentioned, one important quantity that could describe and show such potential was the CRs, which could be determined by finding the ratios of activity concentrations of radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in plants to those in soils. The high values of CRs (usually greater than 1) would imply that the plant could transfer and accumulate high amounts of radionuclides in their plant system (Bettencourt *et al.* 1988). For example, Chakraborty *et al.* (2013) reported that the average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in grass samples collected from Chittagong City, Bangladesh, were 22.13 ± 2.30, 38.47 ± 2.72, and 451.90 ± 24.89 Bq kg^{-1}, respectively, and in soil samples collected at the same site of grass samples were 1.26 ± 0.11, 3.66 ± 0.31, and 134.95 ± 3.68 Bq kg^{-1}, respectively, leading to grass/soil CRs of 0.056, 0.089, and 0.275, respectively. As another example, Jazzer and Thabayneh (2014) showed that the average grass/soil CRs of ^{226}Ra, ^{232}Th, and ^{40}K in the West Bank, Palestine, were 1.26, 1.12, and 1.20, respectively, which suggested the potential to utilize grass as an absorber of radionuclides in a polluted environment as their CRs were greater than 1 (the grass could accumulate radionuclides with higher concentrations than those in soil, which could potentially reduce radioactivity in polluted soil). It should be noted that the clear differences between the CR values in the two mentioned works were probably due to differences in grass types, activity concentrations in the soil, the physicochemical properties such as pH, total minerals, and CEC of the soil, geological structures, and the distribution of radionuclides in the soil (Tshivhase *et al.* 2015). However, despite the global availability of radioactivity concentration and CR values of ^{226}Ra, ^{232}Th, and ^{40}K in other plants, the information of those in vetiver grass – especially in Thailand – was considered inadequate as Thailand contained vast bio/landscape-diversity and widely utilized vetiver grass in steep-slope hazard areas (Chomchalow 2011).

As aforementioned, this work determined the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in vetiver grass in Thailand using HPGe gamma spectroscopy and their corresponding grass/soil CRs. The obtained results would then be compared with previous literature values from other plants and locations around the globe. It should be noted that the first three sets of soil and vetiver grass samples investigated in this work were selected from Pathumthani, Suratthani, and Srisaket provinces – located in the central, southern, and eastern Thailand, respectively – where vetiver grass had been widely planted throughout the areas.

MATERIALS AND METHODS

Study Area, Sampling, and Preparation

The sampling locations are shown in Figure 1. Vetiver grass sample # 1, which would be referred to as grass # 1 in this work, was collected from rice fields of the Land Development Department, Ministry of Agriculture and Cooperatives, in Pathumthani Province located in central Thailand (14°02′N, 100°45′E), on the low alluvial flats of the Chao Phraya River where large numbers of industrial parks such as Nava Nakorn Industrial Promotion Zone, Bangkadi Industrial Park, and Techno Thani are located. Vetiver grass sample # 2, which would be referred to as grass # 2, was collected from a palm-tree farm in Suratthani Province located in southern Thailand (9°31′N, 99°10′E),

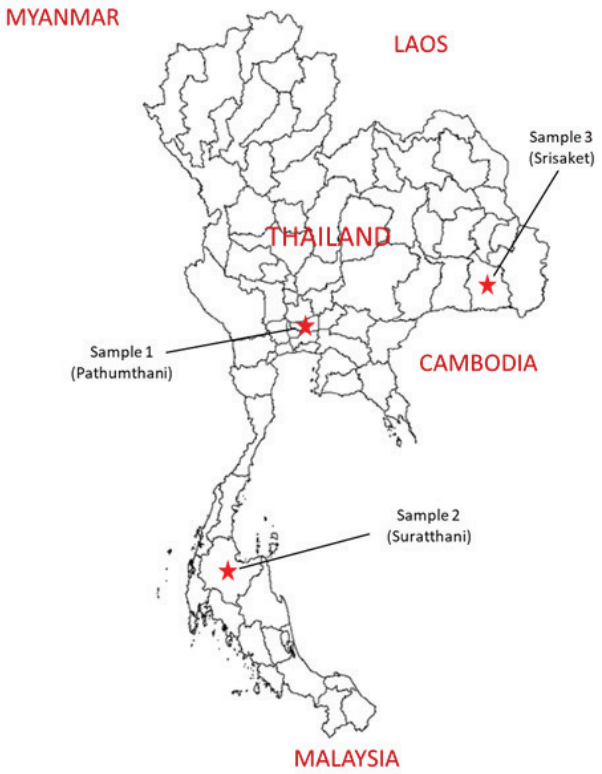

Figure 1. Locations of sampling locations.

on the central coastal plain of the Thapi River consisting of grassland and rubber/coconut plantations. Vetiver grass sample # 3, which would be referred to as grass # 3, was collected from a mango farm in Srisaket Province located in eastern Thailand (14°32′N, 104°08′E) in the valley of the Mun River (a tributary of Mekong River). To determine grass/soil CRs, a core sampler was used to collect soil samples (20–30 cm deep) that were within a 20-cm radius of the vetiver grass samples. For each location, three replicate grass and soil samples (with at least 100 m apart from each other) were measured to determine average values of the activity concentrations and grass/soil CRs. All samples were collected in December 2019.

In all grass samples, each blade was approximately 100 cm long and was divided into three parts (top, middle, and bottom) with an average length of 33 cm in each part (Figure 2) in order to determine dependencies of grass/soil CRs on leaf section. All vetiver grass samples were thoroughly washed with water to remove surface contaminants, then open-air-dried for 7 d, oven-dried at 105 °C for 4 h, ground into fine powders using a stainless-steel ball grinder, and oven-dried again at 105 °C until constant weights were achieved (approximately 1 d). Soil

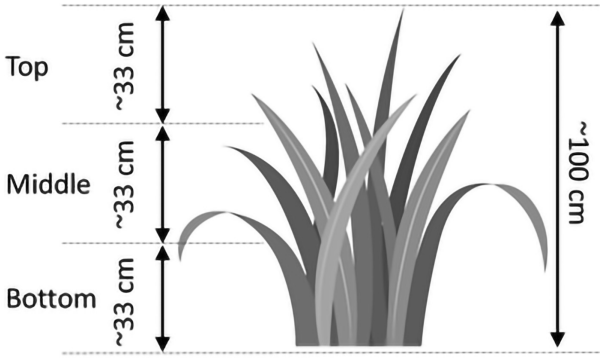

Figure 2. Image showing parts (top, middle, and bottom) of vetiver grass used in this work, with an average length of 33 cm in each part.

samples were collected at ~ 20–30 cm deep, and stones/gravels/roots were carefully removed by hand before the soil was open-air-dried in sunlight for 7 d, ground, and oven-dried at 105 °C for 12 h. The soil samples were then sieved to collect only particle sizes of less than 250 µm (60 mesh), after which 150 g of vetiver grass and 250 g of soil samples were transferred to 500 mL cylindrical beakers, which were sealed with masking tape to avoid radon (Rn) escaping. The sealed samples were stored in a dark container for at least 30 days to allow the radium (Ra) and its progenies to reach secular equilibria prior to gamma spectroscopy (Jevremovic *et al.* 2011). Images of the prepared soil samples are shown in Figure 3, which shows that soil # 1 (Pathumthani Province) (Figure 3a) and soil

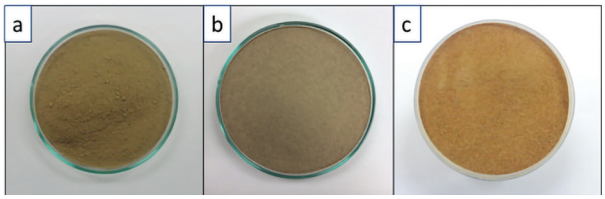

Figure 3. Soil samples from a) Pathumthani, b) Suratthani, and c) Srisaket.

2 (Suratthani Province) (Figure 3b) mostly composed of dark clay (7.5Y 7/4 and 10.0YR 6/4 in Munsell color system, respectively), while soil # 3 (Srisaket Province) (Figure 3c) mostly composed of brown sand (10.0YR 7/8 in Munsell color system).

Determination of Activity Concentrations

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were determined using a coaxial HPGe gamma spectrometer (Model No. GEM30P4-76-RB, USA), having a relative efficiency of 35%, a peak-to-Compton ratio of 66:1, and a resolution (FWHM) of 1.76 keV at 1.33 MeV of ^{60}Co. The detector had a crystal diameter and length of 58.8 mm and 58.7 mm, respectively. A cylindrical lead shield with a fixed bottom and a movable cover was used to reduce external and background radiation. The HPGe detector was connected to a full-featured, 16k channel integrated multichannel analyzer (ORTEC; Model DSPEC jr 2.0, USA). The gamma rays emitted from the samples, as well as the baseline gamma background from a blank sample, were analyzed using the Gamma Vision software. The calibration of the energy was performed using standard gamma sources consisting of the isotopes ^{133}Ba (81.0, 276.4, 302.9, 356.0, and 383.9 keV), ^{109}Cd (88.0 keV), ^{137}Cs (661.7 keV), ^{57}Co (122.1 and 136.5 keV), ^{60}Co (1173.2 and 1332.5 keV), ^{54}Mg (834.9 keV), ^{22}Na (1274.5 keV), and ^{65}Zn (1115.5 keV).

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the samples were calculated using Equation 1:

$$A = (CPS \times 1000)/(\varepsilon_\gamma \times P_\gamma \times W) \quad (1)$$

where A is the activity concentration (Bq kg^{-1}), CPS is the net count per second obtained by subtracting the total counts with background counts at the respective energy, ε_γ is the efficiency of the gamma spectrometer at the respective gamma energy, P_γ is the probability of the transition of the radionuclide of interest for the respective gamma energy, and W is the weight of the sample (g). All measurements were performed for a 100,000-s counting time period. The ε_γ values were determined using reference materials – IAEA-375 (radionuclides and trace elements in soil), IAEA-372 (radionuclides in the grass), and IAEA-447 (natural and artificial radionuclides in moss-soil) – contained in beakers with similar geometry

as the samples. The values for ε_γ and P_γ for gamma energy lines used in the activity concentrations calculations are shown in Table 1 (Saenboonruang *et al.* 2018a). It should be noted that the minimum detection activity (MDA) in this work was measured to be 0.3 Bq kg^{-1}; 0.1 Bq kg^{-1}; and 3.0 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The method of measuring MDA was based on Done and Loan (2016).

Determination of Grass/Soil CRs

The values of the grass/soil CRs were calculated as the ratio of the activity concentrations of radionuclide in the vetiver grass (A_{grass}) to that in the corresponding soil sample (A_{soil}), as shown in Equation 2:

$$CR = A_{grass}(\text{Bq kg}^{-1}, \text{dry weight})/A_{soil}(\text{Bq kg}^{-1}, \text{dry weight}) \quad (2)$$

The grass/soil CRs of ^{226}Ra, ^{232}Th, and ^{40}K were summarized as geometric means (GM) and geometric standard deviations (GSD) (Saenboonruang *et al.* 2018a, b).

Soil Characterizations

Since the concentration of radionuclides in soil depends on various characteristics, the CEC and Total K and Total Ca for each soil sample were determined. To measure the CEC of the soil samples, 1 g of soil was mixed with 18 mL of CH_3COONH_4 (1N) and continuously stirred for 1 h. After the stirring, the sample was allowed to settle and filtered. The soil sample was then rinsed using alcohol and left in a desiccator for the alcohol to completely evaporate. Na^+, K^+, Ca^{2+}, and Mg^{2+} were analyzed using atomic absorption spectrometry (AAS) (Perkin-Elmer Analyst 800, USA) and the sum of the equivalent concentrations of these ions was used for the determination of CEC in the soil samples (Kim *et al.* 2017; Maity and Pandit 2014). The equations for the CEC calculations could be found in Culman *et al.* (2019). For the measurement of Total K and Total Ca, the detailed methods of sample preparation could be found in Arslan and Tyson (1999). In summary, 1 g of soil samples was mixed with 70 mL of 70% HNO_3 and 30 mL of 50% HF in closed PTFE beakers and heated on a hot plate at 140 °C for at least 6 h under the fume hood until the dissolution was completed and all acid was removed. The sample was then heated on a hot plate again at 70 °C for 5 min to remove volatile silicon tetrafluoride. The dried sample was then dissolved in 40 mL of concentrated HNO_3 and transferred to 1-L flasks for the measurement of Total K and Total Ca using AAS (Perkin-Elmer Analyst 800, USA).

Statistical Analysis

Statistical analysis was performed using the IBM Statistical Package for the Social Sciences (SPSS) v.24.0 software. The reported values were expressed as mean ± standard deviation and the analysis of variance was used to determine whether the results were statistically different at the significance level of $p < 0.05$.

RESULTS AND DISCUSSION

Tables 2 and 3 show the activity concentrations and grass/soil CRs of ^{226}Ra, ^{232}Th, and ^{40}K in vetiver grass and their corresponding soils, respectively. The activity concentrations of ^{226}Ra were in the range of 4.1–5.8 Bq kg^{-1} DW in the grass and 3.1–15.7 Bq kg^{-1} DW in soil, the activity concentrations of ^{232}Th were in the range of 2.6–5.2 Bq kg^{-1} DW in the grass and 2.8–16.6 Bq kg^{-1} DW in soil, and the activity concentrations of ^{40}K were in the range of 281.2–584.2 Bq kg^{-1} DW in the grass and 51.7–446.2 Bq kg^{-1} DW in soil. In addition, the grass/soil CR results also indicated that ^{40}K had the highest grass/soil *CRs* with the range of 1.12–7.92 compared to the grass/soil CR values of ^{226}Ra and ^{232}Th, which had the ranges of 0.32–1.48 and 0.20–1.31, respectively. Furthermore, there were no differences in the activity concentrations and grass/soil CRs among the different parts of the leaf blades (top, middle, and bottom) in each sample ($p < 0.05$). Hence, based on the results obtained from this work, vetiver grass transferred and accumulated natural radioactivity uniformly throughout the grass blade.

The higher activity concentrations and grass/soil CRs of ^{40}K were expected as K is an essential and the most abundant inorganic mineral nutrient due to its function

Table 1. Gamma energy lines and their corresponding ε_γ and P_γ values used in radioactivity calculation.

Radionuclide	Transition isotope	Energy (keV)	ε_γ		P_γ
			Soil	Vetiver	
^{226}Ra	^{214}Pb	351.9	0.188	0.050	0.3560
	^{214}Bi	609.3	0.041	0.021	0.4549
^{232}Th	^{212}Pb	238.6	0.120	0.041	0.4660
	^{208}Tl	583.2	0.023	0.008	0.8500
^{40}K		1460.8	0.007	0.008	0.1066

Table 2. Activity concentration (dry weight; DW) of ^{226}Ra, ^{232}Th, and ^{40}K in vetiver grass (top, middle, and bottom of the leaf blade) and the respective collection soil, where ± represents the standard deviation of the mean.

Grass #	Part	Activity concentration (Bq kg^{-1} DW)		
		^{226}Ra	^{232}Th	^{40}K
1	Top	5.1 ± 0.7	3.3 ± 0.7	501.2 ± 25.1
	Middle	5.2 ± 0.7	3.3 ± 0.3	584.2 ± 33.0
	Bottom	5.8 ± 0.3	4.4 ± 0.4	509.3 ± 43.8
	Soil	12.5 ± 0.2	16.6 ± 0.2	446.2 ± 41.4
2	Top	5.0 ± 0.3	3.7 ± 0.5	285.0 ± 4.1
	Middle	5.0 ± 0.4	5.2 ± 0.1	299.7 ± 13.1
	Bottom	5.3 ± 0.5	3.6 ± 0.3	281.2 ± 9.3
	Soil	15.7 ± 0.2	7.1 ± 0.1	225 ± 2.5
3	Top	4.1 ± 0.4	2.6 ± 0.1	394.0 ± 17.3
	Middle	4.5 ± 0.3	2.7 ± 0.6	394.5 ± 14.2
	Bottom	4.3 ± 0.3	2.6 ± 0.2	409.8 ± 10.0
	Soil	3.1 ± 0.1	2.8 ± 0.3	51.7 ± 4.1

Table 3. Grass/soil CRs of ^{226}Ra, ^{232}Th, and ^{40}K in vetiver grass (top, middle, and bottom of the leaf blade), where values are shown as GM (GSD).

Grass #	Part	CRs		
		^{226}Ra	^{232}Th	^{40}K
1	Top	0.40(1.14)	0.20(1.22)	1.12(1.05)
	Middle	0.41(1.13)	0.20(1.10)	1.31(1.06)
	Bottom	0.46(1.05)	0.26(1.10)	1.14(1.10)
	GMs	0.43(1.08)	0.22(1.18)	1.19(1.09)
2	Top	0.32(1.05)	0.52(1.14)	1.26(1.01)
	Middle	0.32(1.07)	0.72(1.00)	1.33(1.05)
	Bottom	0.33(1.09)	0.50(1.08)	1.25(1.03)
	GMs	0.32(1.03)	0.57(1.22)	1.28(1.03)
3	Top	1.33(1.09)	1.31(1.96)	7.61(1.04)
	Middle	1.48(1.07)	0.90(1.25)	7.63(1.04)
	Bottom	1.39(1.08)	0.87(1.07)	7.92(1.02)
	GMs	1.40(1.05)	1.00(1.26)	7.72(1.02)

as a stabilizer in metabolism (Dreyer and Uozumi 2011; Chandrashekatra and Somashekarappa 2016). As a result, K was able to be transferred and accumulate in all parts of vetiver grass. On the other hand, the activity concentrations and the grass/soil CRs of ^{226}Ra and ^{232}Th were much lower than ^{40}K, mostly due to their high affinity with soil and being non-essential nutrients for plants (Saenboonruang *et al.* 2018a, b). However, ^{226}Ra had slightly higher activity concentrations than ^{232}Th, which could be attributed to the fact that ^{226}Ra is chemically similar to Ca, which is considered an important nutrient; hence, ^{226}Ra could be transported and accumulated along with Ca. In addition, This an immobile element due to its water insolubility, which would further reduce its ability to be transported with water into the grass (Vandenhove *et al.* 2009).

Another interesting result shown in Tables 2 and 3 was that soil # 1 and soil # 3 had the highest and lowest activity concentrations, respectively, for all the radionuclides studied. These observed behaviors could have been due to differences in soil properties and characteristics (CEC, Total K, and Total Ca). For example, soil # 1 was collected from central Thailand where agricultural sites and farms are abundant and was mostly composed of dark clay (Figure 3a), which resulted in higher nutrients than other soil types, as shown in Table 4 (Total K and Total Ca in soil # 1 were higher than those in soil # 2 and soil # 3). On the other hand, soil # 3 – which was collected from northeastern Thailand, where the weather is usually dry and arid and was mainly composed of brown sands (Figure 3c) – had the lowest levels of nutrients (Lesturgez *et al.* 2006; Janjirawuttikul *et al.* 2010). The ability of soil to attract, retain, and exchange nutrients could also be determined by considering the CEC shown in Table 4. The results clearly showed that soil # 1 exhibited the highest CEC value, resulting in a high ability to attract and retain important cationic nutrients such as K^+ and Ca^+. As a result, nutrients as well as the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil # 1 were the highest among the soil samples (with soil # 1 containing a ^{40}K concentration approximately twice and nine times higher than those in soil # 2 and soil # 3, respectively). Interestingly, it was found that the accumulation of natural radionuclides investigated in this work – especially ^{40}K – in grass # 3 were still comparable with values from other samples despite its low concentrations in soils (average value of 399.4 Bq kg^{-1} DW in grass # 3, compared with 531.6 Bq kg^{-1} DW and 288.6 Bq kg^{-1} DW in grass # 1 and grass # 2, respectively). The high accumulation of natural radionuclides in grass # 3 and their low concentrations in soil samples consequently led to the highest values of grass/soil CRs in grass # 3.

Table 4. Values of CEC and Total K and Total Ca for all soil samples.

Parameters	Soil #1	Soil #2	Soil #3
CEC (cmol kg^{-1})	32.30	5.20	4.00
Total K (%)	0.37	0.04	0.03
Total Ca (%)	0.137	0.031	0.002

Table 5 compares the current grass/soil CRs with previous reports. The comparisons indicated that the CRs for

Table 5. Reported values of grass/soil CRs of ^{226}Ra, ^{232}Th, and ^{40}K from the current study compared with values from literature values.

Location	Plant type	CRs			Reference
		^{226}Ra	^{232}Th	^{40}K	
Thailand	Grass	0.32–1.40	0.22–1.00	1.19–7.72	Current study
Turkey	Grass	0.40	1.44	0.99	Cengiz (2018)
Palestine	Grass	1.26	1.15	1.20	Jazzar and Thabayneh (2014)
Bangladesh	Grass	0.056	0.089	0.275	Chakraborty *et al.* (2013)
Slovenia	Grass	0.42	–	–	Strok and Smodis (2013)
Spain	Grass	0.17	0.058	–	Tome *et al.* (2003)
Taiwan	Grass	–	–	0.67–2.84	Wang *et al.* (1997)
India	Wheat	0.009–0.016	0.006–0.024	0.14–3.10	Pulhani *et al.* (2005)
Thailand	Green Chiretta	0.34	–	2.80	Saenboonruang *et al.* (2018a)
Malaysia	Taro	0.037	–	0.44	Solehah *et al*. (2016)
Malaysia	Long bean	0.051	0.003	2.43	Solehah *et al*. (2016)

^{226}Ra in this work were in agreement with reports of grass samples from Turkey, Palestine, and Slovenia; for ^{232}Th were in agreement with reports from Palestine; and for ^{40}K were in agreement with Turkey, Palestine, and Taiwan. However, the results showed that CRs of natural radionuclides in this work were higher than those reported in Bangladesh and Spain. The differences in grass/soil CRs could be explained by the differences in grass types; activity concentrations in the soil; the physiochemical characteristics such as CEC, Total K, and Total Ca of the soils; geological structures; and the distribution of radionuclides in the soil (Tshivhase *et al.* 2015). Furthermore, Table 5 also indicated that the CRs of vetiver grass in this work were higher than other previously reported CRs of other plant types, implying the high capability of vetiver grass to transfer and accumulate natural radionuclides that could reduce activity concentrations in contaminated soils.

CONCLUSIONS

The activity concentrations and grass/soil CRs of natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in vetiver grass collected from three regions (central, northeastern, and southern) of Thailand were investigated. The results showed that ^{40}K had the highest activity concentrations and grass/soil CRs among the three radionuclides due to K being essential to plants, whereas Ra and Th have a high affinity to soil particles and are non-essential. In addition, all parts of the vetiver grass uniformly transferred and accumulated natural radionuclides in the top, middle, and bottom of the grass blade, implying the independence of radionuclide concentrations and grass/soil CRs on leaf sections. Moreover, it was found that the grass samples – regardless of the concentration in the soil samples or of the location – could accumulate radionuclides in their blades with approximately the same concentrations. Hence, based on the overall results obtained from this work, vetiver grass presented potentials to be utilized as a natural radionuclide absorber, in a similar way to its known application in the phytoremediation of heavy metals contaminating soil and water.

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