

^{210}Po in Paddy Soils and Rice Grains in Chau Doc, An Giang, Vietnam: Activity Concentration, Soil-to-Rice Transfer, and Health Hazard

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ABSTRACT

Introduction: Soil-to-rice transfer factors (TF) are widely used to predict radionuclides' activity concentrations in rice plants. Since rice (*Oryza sativa* L.) is one of the most popular crops in Vietnam, the radiological risks due to ^{210}Po contaminating rice grains and the TF must be concerned. **Methods:** Alpha spectrometry measured the activity concentration of ^{210}Po in rice and soil samples collected in Chau Doc, An Giang province. TF for the uptake of ^{210}Po from soil to rice plant has been calculated the ratio of the dry weight concentration in the plants to the dry weight concentration in the specified soil layer. Assessment of radiological risks due to ingestion of rice grains was based on estimation of the effective dose. **Results:** The activity concentrations of ^{210}Po were in the ranges of 63.77 – 117.75 Bq kg_{dry weight}⁻¹ and 7.38 – 14.16 Bq kg_{dry weight}⁻¹ in soils and rice grains, respectively. Based on the radiation dose assessment, the accumulation of ^{210}Po in rice grains was not considered for public health. The average TF values of ^{210}Po were 0.12, 0.11, and 0.37 for grains, straws, and roots, respectively. Our TF values for rice were higher than the corresponding values found by other studies and the IAEA TRS-472 report. **Conclusion:** The analytical method for ^{210}Po determination using alpha spectrometry can be applied for further studies in the field of environmental radioactivity. The measured TF values can be used to predict the radioactivity level of ^{210}Po in rice plants cultivated in other fields in the Mekong Delta region.

Key words: ^{210}Po , *Oryza sativa* L., Transfer factors

INTRODUCTION

In the topsoil, the natural radionuclides belonging to uranium and thorium series contaminate with various levels. According to the UNSCEAR report, the worldwide activity concentrations of ^{40}K , ^{226}Ra , ^{232}Th , and ^{238}U in soils were in the ranges of 140-850, 17-60, 11-64, and 16-110 Bq kg⁻¹, respectively¹. High background radiation was found in some regions near uranium mines or in some specific soils and rocks. In Vietnam, natural radioactivity was found within the recommended level. Huy et al. (2012) have measured natural radioactivity in 528 soil samples. The mean values of activity concentrations were 412 Bq kg⁻¹ (range from 10 to 1085 Bq kg⁻¹), 43 Bq kg⁻¹ (range from 15 to 122 Bq kg⁻¹), and 60 Bq kg⁻¹ (range from 16 to 129 Bq kg⁻¹) were found for ^{40}K , ^{226}Ra , and ^{232}Th respectively². Other studies in this field focused on four radionuclides ^{40}K , ^{226}Ra , ^{238}U , and ^{232}Th ³⁻⁵. However, there have been very few studies that measured and assessed the activity concentration of ^{210}Po in soil. ^{210}Po is one of the most popular natural radionuclides focused on in environmental studies. It is an alpha-

emitter natural isotope, a member of the ^{238}U decay series with a relatively short half-life (138.4 days). With the high dose conversion factor (DCF), ^{210}Po is one of the most dangerous radionuclides⁶. Moreover, this radionuclide easily contaminates the surface of fabric, metal, and human skin. On the other hand, with the high soil-to-plant transfer factor (TF), it is possible that ^{210}Po can cooperate with food crops, meat, and milk⁷. Therefore, besides the study on ^{210}Po concentration in food crops and its relative human risks, ^{210}Po TF must be estimated.

Since rice (*Oryza sativa*) is the most important crop in Vietnam, radioactive elements in rice grains can cause potentially dangerous impacts on humans. In 2020, rice paddy production in Vietnam amounted to around 42.7 million metric tons⁸. Rice production in Vietnam in the Mekong and Red River deltas is important to the food supply in the country and national economy. About 52% of Vietnam's rice is produced in the Mekong River Delta and 18% in the Red River Delta⁹. However, there are few studies on the radiological risk due to rice consumption in Vietnam. Especially, the TF of ^{210}Po for rice plants has not been

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carried out.

The aim of this study is to find an effective method for determining ^{210}Po activity in soil and rice samples, to assess the radiological health risks due to consumption of rice grains in the Mekong Delta, which is the most significant agricultural region in Vietnam; and to estimate the soil-to-rice transfer factor for ^{210}Po in the rice ecosystem in the study area.

MATERIALS AND METHODS

Description of the paddy field

This work was carried out in a paddy field in Chau Doc, An Giang province, Vietnam (coordinates $10^{\circ}41'N$, $105^{\circ}7'E$). The map of the study area is shown in Figure 1. As a city located in the south of Vietnam, it has a tropical climate, with a high average humidity of about 80%. The year is divided into two distinct seasons. The rainy season usually lasts from May to November with high rainfall of about 1400 – 1800 mm, and the dry season lasts from December to April. The soil belongs to the fluvisols type; it consists mainly of sediment from the Mekong River. In the investigated paddy field, rice is cultivated 3 seasons per year, and no crop rotation is applied. The growing season normally lasts about 100 days. To ensure sufficient water, the field is maintained in flooded conditions. It means that the soil is covered by water during the growing period.

Sample collection and preparation

The samples were collected at five sites in about 1000 m^2 of the paddy field (Figure 1). At each site, all parts of the rice plants were collected in 1 m^2 at the harvest time (December 2019). In addition, five soil samples were collected at each site for radioactivity analysis. The soil samples were taken at the top layers of soils (0–20 cm) using a steel tube with 70 cm length and 110 mm diameter. The soil pH was measured directly at the sampling time. The rice and soil samples were placed in the plastic bag and taken into the laboratory for further analysis. The rice samples were washed with pure water and separated into root, stem, and grain parts at the laboratory. Both rice and soil samples were dried at 105°C to constant weight, then crushed to a powder and sieved to 0.2 mm grain.

Activity analysis

An alpha spectrometry (Canberra Inc.) equipped with passivated implanted planar silicon (PIPS) detector was used to measure the activity concentration of ^{210}Po in the rice and soil samples. Based on the calibration with the ^{210}Po standard source, the energy resolution was lower than 37 keV (FWHM) with

a detector-to-source spacing equal to the detector diameter. The detector efficiency was higher than 25% achievable with close detector-to-source spacing. The background of the measurement chamber was lower than 1 count per hour. The Genie 2000 Alpha Analyst analyzed the alpha spectrum of ^{210}Po .

To be adaptable with the measurements, the samples were prepared by the procedure described in Figure 2. Rice and soil samples were initially digested in concentrated HNO_3 , HCl , and H_2O_2 . For rice samples, the dry sample was treated with 3 ml concentrated HNO_3 and 50 ml H_2O_2 30%. The solution was then heated to dryness to reduce the organic matter in the sample. This step may repeat 2 – 3 times. The residue was then treated with 50 ml concentrated HCl and 50 ml concentrated HCl . The solution was heated at 50°C until dry. To ensure the sample was completely digested, the step may repeat 2 – 3 times. The pH = 1.5 was adjusted for the solution by adding a volume of 0.5 M HCl .

About 0.5 g of ascorbic acid was added to the solution collected after the digestion process to absorb Fe^{3+} ions and improve the efficiency of the Po precipitation. The experimental setup for co-precipitation of polonium on the cooper disk was performed in Figure 3. A Teflon shelf kept a flat cooper disk (2.5 cm in diameter). The rotation time was set up at 4 h, and the rate of the magnet was 300 rpm. The temperature of the water was adjusted to about 70°C .

After the co-precipitation process, the cooper disk was taken out from the solution and dried under an infrared lamp. The sample was then measured immediately by the alpha spectrometer. The procedure was repeated for the remaining solution to determine the chemical yield of the Po extraction method. The chemical yield H was calculated by Eq. (1).

$$H = 1 - \frac{D_1}{D_2} \quad (1)$$

where, D_1 and D_2 are the count rates of the ^{210}Po energy peak in the first time and second time of Po extraction respectively.

The activity concentration of ^{210}Po in the sample was calculated using Eq. (2)

$$A = \frac{N}{m \times \varepsilon \times H \times t} \quad (2)$$

where A is the activity concentration of ^{226}Ra in the sample (Bq kg^{-1}), N is the count of the alpha energy peak of ^{210}Po , t is the counting time (s), m is the sample mass (kg), ε is the efficiency of the spectrometer calibrated with standard sources, and H is the chemical yield of the sample preparation procedure calculated by Eq (1).

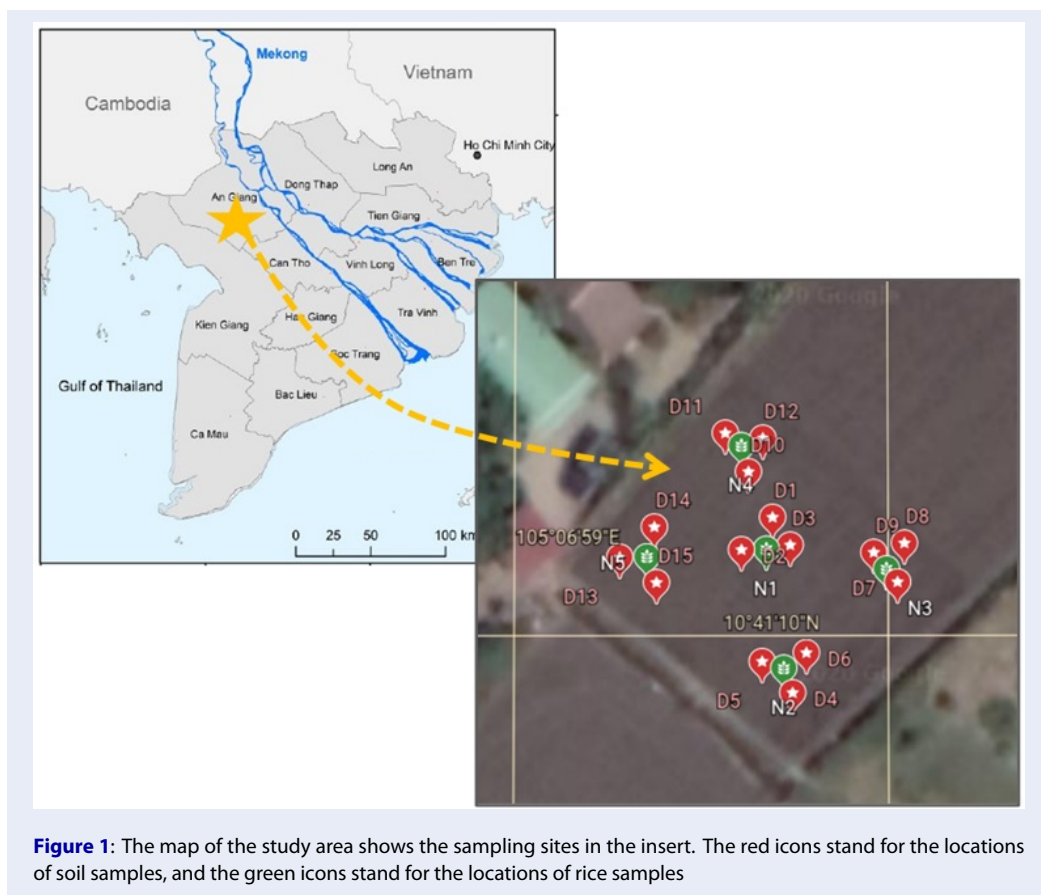


Figure 1: The map of the study area shows the sampling sites in the insert. The red icons stand for the locations of soil samples, and the green icons stand for the locations of rice samples

Dose assessment

According to UNSCEAR (2006), the annual effective dose due to intaking rice grains was calculated by Eq (3).

$$D_E = I \times A \times DCF \tag{3}$$

where D_E is the annual effective dose due to consumption of the rice grains ($Sv\ y^{-1}$), I is the consumption intake rate of rice grains ($100\ kg\ y^{-1}$). The annual intake rate of $100\ kg\ y^{-1}$ was based on the results of our survey in the study area. A is the activity concentration of ^{210}Po in the rice grain sample ($Bq\ kg_{fresh}^{-1}$), and DCF is the dose conversion factor of ^{210}Po according to ICRP (2012)⁶

Soil-to-rice transfer factor

The uptake of radionuclides from soil to rice grains is widely assessed by estimating the soil-to-rice transfer factors (TF)^{3,4,7,10,11}. Since TF can predict the number of radionuclides contaminating the plant based on the activity concentrations of radionuclides in the soil, it is an important factor that is used popularly in

environmental science. According to the IAEA TRS-472 report (2010), TF is defined as the ratio of the radionuclide content in the rice plants to that in the soil as Eq. (4)⁷.

$$TF = \frac{A_{rice}}{A_{soil}} \tag{4}$$

where A_{rice} is the activity concentration of ^{210}Po in dry rice ($Bq\ kg^{-1}_{dry\ weight}$), and A_{soil} is the corresponding activity concentration in dry soil ($Bq\ kg^{-1}_{dry\ weight}$). For rice plant, the soil sample is a top-soil layer (20 cm) which are the upper, outermost layer of soil⁷.

RESULTS

Table 1 summarizes the measurement results on the soil samples. The activity concentrations of ^{210}Po ranged from 63.77 ± 3.38 to $117.75 \pm 4.65\ Bq\ kg^{-1}$ with an average value of $91.52 \pm 3.98\ Bq/kg$. The highest value of $117.75 \pm 4.65\ Bq\ kg^{-1}$ was found in sample D6. This value is 1.85 times higher than the lowest activity concentration value found in sample D3 ($63.77 \pm 3.38\ Bq\ kg^{-1}$). The soil pH values are not

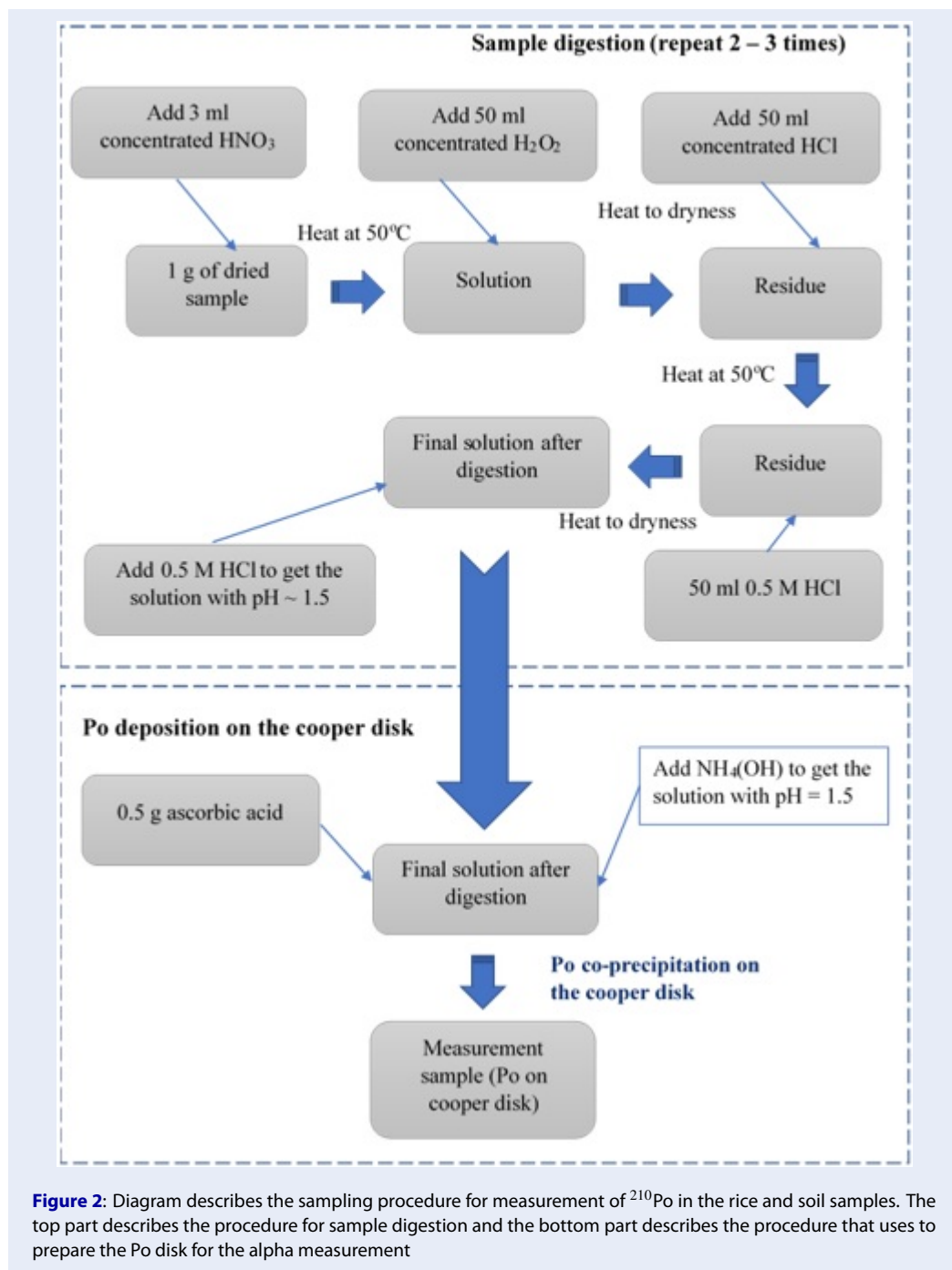


Figure 2: Diagram describes the sampling procedure for measurement of ²¹⁰Po in the rice and soil samples. The top part describes the procedure for sample digestion and the bottom part describes the procedure that uses to prepare the Po disk for the alpha measurement

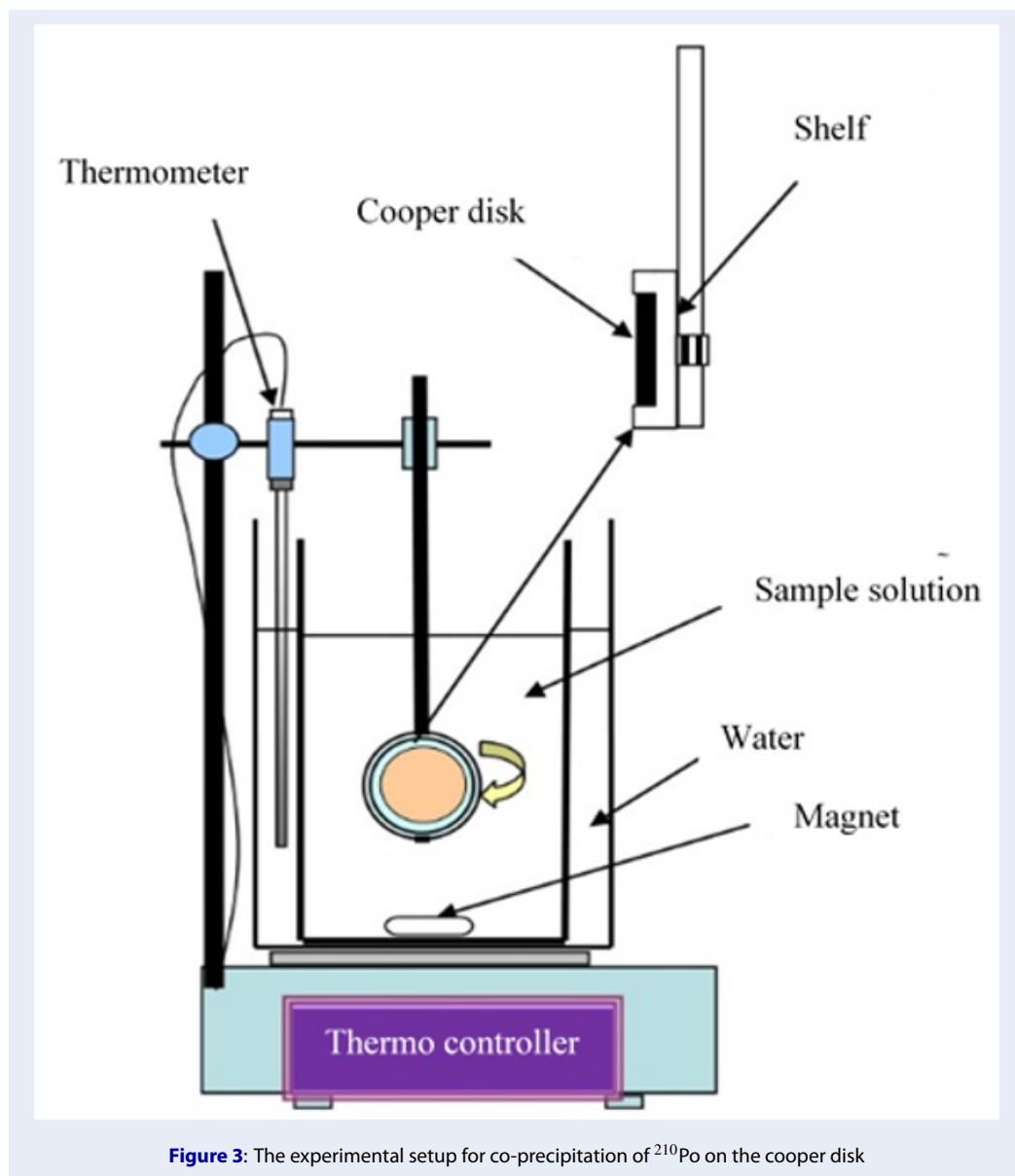


Figure 3: The experimental setup for co-precipitation of ^{210}Po on the cooper disk

very different between sample sites. We also found that the TDS of irrigation water was from 300 to 301 ppm and the soil temperature was from 30 to 33°C. Table 2 summarizes the activity concentrations of ^{210}Po measured in grain, straw, and root parts of rice samples collected in the study area. The activity concentrations of ^{210}Po were found from 3.56 ± 0.78 to $9.12 \pm 1.26 \text{ Bq kg}_{\text{fresh}}^{-1}$, with the mean value of $6.52 \pm 1.04 \text{ Bq kg}_{\text{fresh}}^{-1}$ in straw samples. They were 4.89 ± 0.90 to $9.38 \pm 1.25 \text{ Bq kg}_{\text{fresh}}^{-1}$, with a mean value of $7.19 \pm 1.08 \text{ Bq kg}_{\text{fresh}}^{-1}$. To compare the partitions of ^{210}Po in various parts of the rice plant, the activity concentration in the root part of sample 1 was measured. We found that the partitions in the order

root > grain > straw.

The TF for rice components was estimated by the ratio between activity concentration of ^{210}Po in rice and corresponding soil, as discussed in the previous section. For this purpose, the average activity of each soil group was calculated. The results are shown in Table 3. For five study sites in this work, the TF was from 0.054 ± 0.012 to $0.178 \pm 0.026 \text{ Bq kg}^{-1}$, with the mean value of $0.110 \pm 0.018 \text{ Bq kg}^{-1}$ for rice straw samples. For rice grain samples, the TF were from 0.081 ± 0.015 to $0.160 \pm 0.024 \text{ Bq kg}^{-1}$, with the mean value of $0.12 \pm 0.019 \text{ Bq kg}^{-1}$. For rice root sample 1, we found that the TF was 0.597 ± 0.367 . The comparison between the TFs estimated for vari-

Table 1: Activity concentrations of ²¹⁰Po in soil samples

Group	Sample No.	pH	Activity (Bq kg _{dry} ⁻¹)
1	D1	6.4	85.93 ± 3.86
	D2	6.4	82.72 ± 3.79
	D3	6.6	63.77 ± 3.38
2	D4	6.5	77.11 ± 3.55
	D5	6.5	102.04 ± 4.17
	D6	6.3	117.75 ± 4.65
3	D7	6.4	76.70 ± 3.59
	D8	6.3	82.99 ± 3.74
	D9	6.2	113.21 ± 4.36
4	D10	6.3	106.28 ± 4.57
	D11	6.7	90.45 ± 3.92
	D12	6.4	90.89 ± 4.09
5	D13	6.4	91.73 ± 3.85
	D14	6.5	94.70 ± 3.94
	D15	6.5	96.53 ± 4.20
Average value		6.4	91.52 ± 3.98

Table 2: Activity concentrations of ²¹⁰Po in rice samples

Sample No.		Activity concentration in fresh sample (Bq kg ⁻¹)	Activity concentration in dry sample (Bq kg ⁻¹)
Straw	L1	9.12 ± 1.26	13.77 ± 1.91
	L2	3.56 ± 0.78	5.37 ± 1.18
	L3	3.34 ± 0.75	5.04 ± 1.13
	L4	7.46 ± 1.13	11.26 ± 1.71
	L5	9.12 ± 1.26	13.77 ± 1.91
	Mean	6.52 ± 1.04	9.84 ± 1.57
Grain	H1	8.22 ± 1.16	12.42 ± 1.75
	H2	8.22 ± 1.16	12.42 ± 1.75
	H3	4.89 ± 0.90	7.38 ± 1.35
	H4	5.24 ± 0.93	7.91 ± 1.40
	H5	9.38 ± 1.25	14.16 ± 1.89
	Mean	7.19 ± 1.08	10.85 ± 1.63
Root	R1	22.26 ± 1.98	33.62 ± 2.98

ous rice components is shown in Figure 4.

DISCUSSION

Assessment of the analytical method

Many methods were employed to determine ^{210}Po in the environmental samples (e.g., gamma spectrometry, alpha spectrometry, inductively coupled plasma mass spectrometry,...). In the radiation detection field, the measurements of this radionuclide are mostly based on gamma or alpha spectrum analysis. However, the results derived from gamma rays analysis are not accurate due to radon release from the samples. On the other hand, the method is only performed with many environmental samples⁵. Our method using the analysis of alpha rays of ^{210}Po can resolve these problems. Moreover, since the direct measurement of alpha rays, the analysis does not take much time to achieve the secular radioactive equilibrium. Based on the application of this analytical method for all samples in this study, the average efficiency of the sampling procedure reaches $95 \pm 4\%$.

The concentration of in rice grain samples and health hazard

Many studies in the world have focused on the activity concentrations of radionuclides in rice grains because they are relevant to the human risks due to the intake of rice. Based on the results in Table 2 and the method for risk assessment in the method section, we estimated that the average effective dose due to ingestion of rice is $0.17 \pm 0.03 \text{ mSv y}^{-1}$. The values were found between $0.12 \pm 0.02 \text{ mSv y}^{-1}$ (grain sample H3) and $0.23 \pm 0.03 \text{ mSv y}^{-1}$ (grain sample H5). These results were derived with an assumption that the annual intake rate of 100 kg y^{-1} was based on our survey results in the study area. The DCF of ^{210}Po is $2.4 \times 10^{-7} \text{ Sv Bq}^{-1}$, according to the ICRP document⁶. The highest value of sample H5 does not exceed the limitation dose of 10 mSv y^{-1} recommended by ICRP (2007) for foodstuffs¹². However, it does not indicate that rice consumption is negligible for public health since the term of foodstuffs is general, and the recommended dose considers all environmental radionuclides. Some studies measured the activity concentration of ^{210}Po in rice. In 2014, Rani et al. measured the ^{210}Po activity in rice grains in Kanyakumari city, India. They found that the average activity was $0.21 \pm 0.04 \text{ Bq kg}^{-1}$ which caused the annual effective dose of $45.99 \mu\text{Bq y}^{-1}$ due to rice consumption¹³. In northern Vietnam, the average activity of ^{210}Po found in rice grains was $2.0 \pm 0.6 \text{ Bq kg}^{-1}$ ¹⁴. This value is five times lower than our average activity measured in the study area.

The concentration of in rice soil samples

The concentrations of ^{210}Po in surface soils vary across a wide range, depending on the uranium concentration in the area's bedrock, the depth of the soil sampled, climatic conditions, and soil properties¹⁵. In addition, phosphate fertilizers commonly contain significant concentrations of ^{210}Pb , so that rice cultivation causes a high concentration of ^{210}Po in the paddy soil^{16,17}. In Brazil, the activity concentrations of ^{210}Po in surface soil were between 27 and 74 Bq kg^{-1} ¹⁸. In Kaiga (India), it was found in the range of $17.1 - 228.2 \text{ Bq kg}^{-1}$, with an average value of 83.3 Bq kg^{-1} ¹⁹. In Spain, They were found between $16 - 780 \text{ Bq kg}^{-1}$ ²⁰. In a review article, Parfenov reported that the worldwide activity of ^{210}Po was from 8 to 220 Bq kg^{-1} ²¹. Compared to these studies, the recent activity levels of ^{210}Po in the paddy soil in the study area are than theirs.

Transfer of ^{210}Po from soil to various rice parts

Most studies on the TFs of radionuclides have focused on the transfer to rice grains since the grain part is used as human food crops. Compared with other studies in the world, our recent values of ^{210}Po are higher than theirs. Soil to plant transfer is influenced by several factors: the physicochemical characteristics of the radionuclides, the form of the fallout or the waste, the time after fallout, soil properties, the type of crop, and the soil management practices⁷. The difference between our TFs of ^{210}Po and the TFs found in other studies is due to the soil properties and rice cultivation conditions. There are some studies focused on the TF of ^{210}Po in rice ecosystems. In Japan, the TFs of ^{210}Po in white rice grains were found between 6.2×10^{-5} and 3.6×10^{-3} with the geometric mean of 4.7×10^{-4} . In brown rice, they were between 9.6×10^{-5} and 8.6×10^{-3} with the geometric mean of 4.7×10^{-4} . The study found that the wide range of TFs was due to the differences in soil type, rice type, and climate conditions¹⁴. Vandehove et al. (2009) found the average value of TF_{Po} for rice grains was 1.68×10^{-2} ¹⁰. According to the IAEA TRS-472 report, the TF of polonium for rice grains was from 9.4×10^{-3} to 1.7×10^{-2} , with a mean value of 1.3×10^{-2} . Compared with these studies, our recent TFs are higher than the reference values. Based on Table 3, we also found that ^{210}Po contamination in rice grains is higher than in rice straws.

CONCLUSION

The current study was carried out on five selected cultivated sites located in a paddy field in the Chau Doc

Table 3: Estimations of TF values for various components of the rice samples collected in the study area

Sample No.	TF	
Straw	L1	0.178 ± 0.026
	L2	0.054 ± 0.012
	L3	0.055 ± 0.013
	L4	0.117 ± 0.019
	L5	0.146 ± 0.021
	Mean value	0.110 ± 0.018
Grain	H1	0.160 ± 0.024
	H2	0.125 ± 0.018
	H3	0.081 ± 0.015
	H4	0.082 ± 0.015
	H5	0.150 ± 0.021
	Mean value	0.120 ± 0.019
Root	R1	0.367 ± 0.036

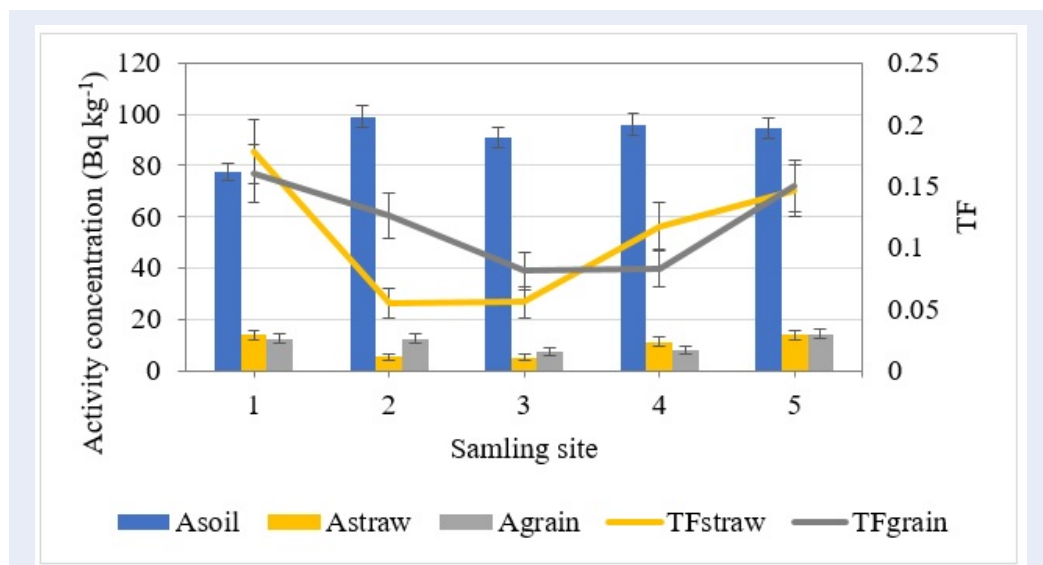


Figure 4: Comparison between activity concentrations of ²¹⁰Po in soil, straw, and grain samples and TF values. In this figure, Asoil, Astraw, and Agrain are activity concentrations of ²¹⁰Po in soil, straw, and grain, respectively. TFstraw and TFgrain are the TF values for straw, and grain respectively.

City, An Giang Province. ^{210}Po activity was measured in surface soil and rice samples by alpha spectrometry equipped with PIPS detector. An analytical method has been proposed to co-precipitate polonium on the copper disk to adapt with the measurement. Based on this method for the standard source in this study, the efficiency of the sampling procedure reaches $95 \pm 4\%$. The average concentrations of ^{210}Po were $91.52 \pm 3.98 \text{ Bq kg}^{-1}$ and samples $0.120 \pm 0.019 \text{ Bq kg}^{-1}$ were found in soil and rice grain samples. The analytical results on the collected rice samples show that the accumulation of ^{210}Po in the grain part is higher than it in the straw part. Our activity concentrations in rice and soil are quite high compared to other studies in the world and the north of Vietnam. However, the effective dose rate due to the consumption of rice grains causes an insignificant radiological risk to the local population. The estimation of the transfer characteristics of ^{210}Po from paddy soil to various parts of the rice plant shows the high uptake rates of this radionuclide in the study area.

LIST OF ABBREVIATIONS

TF: Soil-to-rice transfer factor

DCF: Dose Conversion Factor

ICRP: International Commission on Radiological Protection

UNSCEAR: United Nations Scientific Committee on the Effects of Atomic Radiation

IAEA: International Atomic Energy Agency

COMPETING INTERESTS

The authors declare that they have no conflicts of interest.

AUTHORS CONTRIBUTIONS

All authors contributed to the experiments and the preparation of the manuscript. Van Thang Nguyen carried out the analysis of radionuclide in the environmental samples and preparing the manuscript; Nguyen Phong Thu Huynh calculated the activity concentration, and estimated the dose and the transfer factor; Cong Hao Le was responsible for language editing and response to the reviewers.

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