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The investigation of 239+240Pu, ⁹⁰Sr and ¹³⁷Cs background radiation levels in soil samples in some provinces in the north of Vietnam

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Abstract: This study was conducted to investigate the background levels of plutonium (239+240Pu), strontium (90Sr), and cesium (137Cs) in the topsoil of some provinces in northern Vietnam. Thirty-one soil samples were collected between October 2020 and January 2022. In the samples, the specific activities of $137Cs$ were measured directly by gamma spectrometry, while the specific activities of, $^{239+240}$ Pu, and 90 Sr were measured by alpha spectrometry, and beta counting, after the radiochemical separation procedures respectively. The concentrations were observed in the range of (0.768–2.129) Bq/kg dry weight (DW) for ¹³⁷Cs, (0.018–0.058) Bq/kg DW for 239+240Pu, and (0.022–0.697) Bq/kg DW for ⁹⁰Sr, with average values are (1.393 \pm 0.196) Bq/kg DW, (0.039 \pm 0.014) and (0.129 \pm 0.011) Bq/kg DW, respectively, which are relatively lower than the reported values in the neighboring country. The average recovery efficiencies for radiochemical separation of $^{239+240}$ Pu and 90 Sr were 63.8% and 52.0%, respectively. The minimum detectable activity (MDA) for ^{90}Sr , $^{239+240}Pu$, and ^{137}Cs are 0.012 Bq/Kg DW, 0.004 Bq/Kg DW, and 0.1 Bq/Kg DW, respectively.

Keywords: *⁹⁰Sr; 239+240Pu; ¹³⁷Cs; Background radiation level, recovery efficiency.*

I. INTRODUCTION

Determining radioactivity in soil is essential because it forms a radioactive pathway in humans, animals, and plants, and indicates radioactive accumulation in the environment [11].

Radioisotopes 239 Pu, 240 Pu, 90 Sr, and 137_{cs} are products of nuclear fission and are present during the fallout from nuclear weapons and nuclear accidents. The halflives of these artificial radioisotopes are very large $(T1/2 \text{ } (^{137}\text{Cs}) = 30,17 \text{ years}; T1/2$ $(^{90}Sr) = 28,79$ years; T1/2 $(^{239}Pu) = 24110$ years; T1/2 $(^{240}Pu) = 6561$ years), which means it can take hundreds of years $(^{90}Sr,$ ¹³⁷Cs) and thousands of years $(^{239+240}Pu)$ for

them to decay to negligible activity. Exposure to water and food contaminated by man-made radionuclides $(^{239+240}$ Pu, 90 Sr, and $137Cs$) may increase the risk of leukemia and bone cancer.

 90 Sr is a highly radioactive fission product released into the environment after atmospheric nuclear tests and nuclear accidents. Therefore, it is an interesting subject for many other types of investigations, from its distribution and behavior in natural systems to its effects on wildlife and humans. As radionuclide activity is an important parameter for predicting radiation exposure, the reliability of exposure prediction depends on the accuracy of the target radionuclide activity determination. Therefore, there is a need for

accurate and reliable methods to determine radionuclide activity in different types of samples [12].

Plutonium (Pu) is a toxic and radioactive element widely present in the environment. It can accumulate in food chains, including in marine biomes. Pu's main source is the global fallout from nuclear weapons testing. In addition, nuclear reactor incidents also contribute to the local pollution of Pu. In an environment consisting of various radioactive isotopes, including ²³⁸Pu, 239 Pu, and 240 Pu, Pu emits high-energy alpha particles. Among them, 239 Pu and 240 Pu are the most abundant isotopes, with long halflives of 24110 and 6560 years, respectively. In contrast, ²³⁸Pu exists at lower concentrations in the environment and has a shorter half-life (87.7 years) [2].

 $137Cs$ is a radioactive isotope of cesium formed by the nuclear fission of ^{235}U and other fissile isotopes in nuclear reactors. It is one of the most problematic fission products in the group with short-medium half-lives because it is easily transported and dispersed in nature owing to its high solubility in water and chemical compounds made from cesium as salts. $137Cs$ react with water to produce a water-soluble compound (cesium hydroxide). The biological behavior of cesium is similar to that of potassium and rubidium. After entering the body, cesium is distributed throughout the body and concentrated in the soft tissues [3].

This study aimed to investigate the specific activities of isotopes $^{239+240}$ Pu, 90 Sr, and $137Cs$ in the soil of the northern provinces of Vietnam. It provides information about the background of these isotopes to the management agency for monitoring and warning of radiation.

II. MATERIAL AND METHODS

A. Sampling and sample preparation

In a broad sense, soil includes sediments on the seabed and riverbeds, but here it is only soils from arable and uncultivated land. The sampling site should be free of nearby obstacles (trees and structures) and land use should be considered. In addition, the sampling sites should not have unusual soil or topographical qualities and little vegetation. Sites should be selected for periodic sampling to determine radioactive accumulation. Maps of the sampling locations were sketched or photographed. Sites with flat and grassy terrain, believed to be undisturbed for many years, were selected for sampling. This is a difficult task in Vietnam, where there are almost no grasslands and primeval lands are scarce. For this reason, we prioritized sampling at local meteorological stations. Soil samples were taken from the surface soil layer at a depth of (0-5) cm because artificial radionuclides mostly remain in the surface soil layer [1, 6]. Each sampling location will take samples at five to eight points using specialized sampling equipment and then mix them well. The soil to be measured must consist of particles with diameters of 2 mm or less [10].

Soil samples were collected at several points in the provinces of Hai Phong, Quang Ninh, Cao Bang, Son La, Lao Cai, Lang Son, Hung Yen, and Thai Binh from October 2020 to January 2022. Some provinces in the Northern region of Vietnam that are densely populated and geographically located less than 300 km from China's two Fangcheng nuclear power plants are areas at high risk of being affected by the occurrence of radiation and nuclear incidents. The samples were stored and prepared according to the measurement purpose. Each sample was dried at 105 °C, passed through a 2 mm sieve (removing foreign

material, if any), and homogenized. For the analysis of isotopes $^{239+240}$ Pu and 90 Sr, soil samples after drying were calcined at 500 °C for 4 h to remove organic matter. The sampling locations are shown in Table I.

Radioactive separation of Pu isotopes was performed on sections of 50 g of dry soil, according to the procedure guidelines of the Japan Agency for Science and Technology. After calcination at 500 °C for 4 h to remove organic matter and adding 50 µL of a solution containing 25 mBq of the 242 Pu yield tracer, filtration was performed using 8M HNO₃. Plutonium was separated using a Dower AG1- X8 ion exchange resin (100-200) mesh and then electrically deposited on a stainless-steel plate for alpha spectrometry measurement.

As the 90 Sr isotope is a beta-pure emitter, it requires complex sample processing, which involves chemically separating strontium from other elements by suitable methods (ion exchange or extraction chromatography, precipitation, etc.) and subsequent detection on available instruments. The procedure requires several steps, from isolating small amounts of strontium from complex backgrounds to the purification and detection of strontium, where each step contributes to some degree to the uncertainty of the determination (IAEA 295, 1989; IAEA 1401, 2004). The most difficult step is to separate Sr from Ca owing to their similar chemical properties. In the standard procedure for this purpose, the fuming nitric acid precipitation method is used (IAEA 295, 1989; ISO 13160, 2012) [12]. This analytical method separates alkaline earth elements (Ca, Sr, Ba, Ra) through carbonate precipitation. Calcium was removed via the precipitation of strontium nitrate using concentrated nitric acid. Barium and radium were separated as chromate. $90Y$ was then removed through hydroxide precipitation, which eventually yielded pure 90 Sr [7]. Radioactivity recovery was determined using the marker ${}^{85}Sr$, which was added before sample processing. The Sr precipitate was dissolved, and the solution was stored for 14 d to establish an equilibrium of $\rm{^{90}Sr^{-90}Y}.$

Sample No.	Province	Sample symbol	Longitude	Latitude
		DCT 20.3.1	107.7721	20.9759
\mathfrak{D}	Quang Ninh	DCT 20.3.2	107.7721	20.9759
3		DCT 21.2.1	107.7721	20.9759
$\overline{4}$		DBC 20.3.1	107.0602	20.9617
5		DBC 20.9	107.0602	20.9617
6		DMC 20.9	107.9818	21.5186
7		DMC	107.9818	21.5186
8		DHP 20.9	106.6302	20.8058
9	Hai Phong	DHP 20.2.1	106.6302	20.8058
10		DHP 21.2.2	106.6302	20.8058
11		DBLV 20.3.1	107.7231	20.1308
12		DBLV 20.3.2	107.7231	20.1308
13		DBLV 21.1.2	107.7231	20.1308

Table I. Location of soil sampling points

B. Analytical methods

 $137Cs$ radioactivity in soil samples was measured by using the GMX wide-band gamma detector of EG&G ORTEC with relative resolution and recording efficiency at a beam of 1.332 MeV, corresponding to 1.9 keV and > 40%. The samples weighing 500–700 g were placed in a Marinelli beaker with an inner diameter of 10 cm (probe diameter 9.6 cm). The samples were then placed directly on top of the probe and measured at (80000-86000) second intervals. The gamma spectrum receiver was a DART EG&G ORTEC multichannel analyzer. The spectrum analysis software is Gamma Vision-32. The standard samples used to analyze the radioactivity of $137Cs$ were three standard samples (RGU-1, RGTh-1, and RGK-1) from the International Atomic Energy Agency (IAEA), which were prepared with the same geometry as the measured samples.

Plutonium activity was measured by using a Canberra Alpha Soloist alpha spectrometer at a resolution of 20 keV. Efficiency of 241 Am at 10 mm distance: 25%. Alpha background: 0.016 cpm. The energy measurements ranged from 3 to 10 MeV. The sample chamber was vacuum-sealed and the distance from the sample to the detector was varied from 1 mm to 41 mm. Vacuum pump Model ALPHA-PPS-115 of ORTEC. DART ORTEC, including amplification and MCA. The computer was installed with ORTEC MAESTRO-32 spectrum acquisition software. The measurement time ranged from 240,000 to 400,000 s.

The activity of 90 Sr was measured using a low-background beta counting system (XLB: CANBERRA-S5XLBGPF, USA). The detector was a proportional counting tube with a beta background of 0.81 ± 0.09 cpm and a beta counting efficiency of 45%. The computer was installed with Eclipse LB 3.3 control software. The counting time was 120 min. Quality control of the analytical results was performed by using the International Atomic Energy Agency's Soil4/2000 certified reference material, and the deviation between the tested and certified values was within approximately 5% [7, 8].

The recovery of plutonium was determined by alpha spectrometry, and the recovery of strontium was determined by gamma spectrometry, a method based on the comparison of the activity of the tracer isotope introduced into the sample initially and its activity in the template separated at the final stage (template to determine performance). These activity values were determined under the same measurement conditions and with decay correction. In all cases, the tracer radioisotope introduced to

determine the separation efficiency should initially be absent from the sample. Radiation should not affect the measurement of radioisotope activity to be analyzed. The recovery efficiencies of Sr and plutonium are presented in Tables II and III, respectively.

III. RESULTS AND DISCUSSION

The specific activities of the artificial radionuclides ^{137}Cs , $^{239+240}Pu$, and ^{90}Sr measured by direct gamma spectrometry, alpha spectroscopy, and beta counting in the studied soil samples are given in Tables II and III. The minimum detectable activity (MDA) for isotopes 90 Sr, $^{239+240}$ Pu, and 137 Cs was 0.012 Bq/kg DW, 0.004 Bq/Kg DW; 0.1 Bq/Kg DW.

Sample	Sample symbol	⁸⁵ Sr recovery efficiency,	Activity ⁹⁰ Sr, Bq/kg	Activity $137Cs$, Bq/Kg
No.		$\frac{0}{0}$	DW	DW
1	DBLV 20.3.1	39.92 ± 2.15	0.167 ± 0.015	0.960 ± 0.054
$\overline{2}$	DMC 20.9	64.66 ± 3.09	0.061 ± 0.005	1.048 ± 0.064
3	DLS 02	38.30 ± 2.11	0.040 ± 0.003	1.511 ± 0.158
$\overline{\mathcal{L}}$	DBC 20.9	63.08 ± 3.36	0.697 ± 0.061	2.038 ± 0.081
5	DHP 20.9	59.66 ± 2.81	0.104 ± 0.009	1.694 ± 0.081
6	DLC 20.4.1	57.32 ± 3.42	0.203 ± 0.017	0.786 ± 0.058
$\overline{7}$	DHY 13	29.17 ± 1.52	0.095 ± 0.008	1.358 ± 0.075
8	DTB 22.2	49.86 ± 2.37	0.183 ± 0.014	1.123 ± 0.214
9	DTB 02	46.13 ± 2.24	0.033 ± 0.003	1.933 ± 0.188
10	DHY 03	47.41 ± 2.25	0.073 ± 0.006	1.447 ± 0.207
11	DHY 02	59.16 ± 2.72	0.089 ± 0.007	1.204 ± 0.451
12	DTB 01	27.52 ± 1.48	0.055 ± 0.005	1.586 ± 0.245
13	DTB 27.2	56.38 ± 3.28	0.022 ± 0.002	1.259 ± 0.259
14	DHY 09	63.23 ± 2.92	0.056 ± 0.005	1.143 ± 0.276
15	DTB 30.1	59.43 ± 2.74	0.106 ± 0.008	1.678 ± 0.320
16	DTB 31.1	78.42 ± 3.50	0.100 ± 0.008	1.145 ± 0.305
17	DTB 35.1	55.56 ± 2.59	0.124 ± 0.010	1.116 ± 0.298

Table II. Results of specific activity analysis of ¹³⁷Cs, ⁹⁰Sr isotopes, and recovery efficiency of ⁸⁵Sr tracer

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Sample No.	Sample symbol	Activity, Bq/kg DW	Recovery efficiency, %
1	DHP 20.2.1	0.037 ± 0.012	105.41 ± 35.14
2	DHP 21.2.2	0.039 ± 0.010	98.24 ± 22.54
3	DLS 20.1.1	0.056 ± 0.027	20.94 ± 9.36
$\overline{4}$	DBLV 21.1.2	0.030 ± 0.012	23.53 ± 8.89
5	DBLV 20.3.2	0.034 ± 0.014	47.15 ± 17.82
6	DCB 20.3.1	0.018 ± 0.009	29.55 ± 13.21
7	DBC 20.3.1	0.058 ± 0.020	69.39 ± 23.13
8	DCT 20.3.2	0.048 ± 0.016	74.71 ± 23.63
9	DCT 21.2.1	0.042 ± 0.012	76.07 ± 21.10
10	DSL 21.2.1	0.046 ± 0.012	61.31 ± 15.33
11	DMC	0.023 ± 0.006	95.16 ± 21.83

Table III. Results of specific activity analysis of ²³⁹⁺²⁴⁰Pu isotope and recovery efficiency of ²⁴²Pu tracer

Fig. 1. Alpha spectrum of ²³⁹⁺²⁴⁰Pu and ²⁴²Pu tracer isotopes in soil samples

Fig. 3. Gamma spectrum of the tracer isotope ⁸⁵Sr in the soil sample

The analytical results showed the presence of ^{137}Cs , ^{90}Sr , and $^{239+240}Pu$ in the topsoil through the measurement results. The analysis results of these isotopes were all in units of Bq/kg dry weight. The specific

activities ranged the 90 Sr from 0.022 to 0.697 Bq/kg DW, with an average value of $(0.129 \pm$ 0.145) Bq/kg DW (Table II). The recovery efficiency was measured by using the isotope tracer ⁸⁵Sr, with an average recovery value of

52.0%. Soil samples with low recovery efficiency are characterized by a dark brown color, many types of clay containing phyllosilicate minerals, rich in oxides and hydroxides of silicon and aluminum, causing difficulty in the decomposition process and reducing recovery efficiency.

The specific activities of $^{239+240}$ Pu ranged from 0.018 Bq/kg DW to 0.058 Bq/kg DW, with an average value of (0.039 ± 0.012) Bq/kg DW (Table III). DHP 20.2.1 sample is used to measure the repeatability of the analytical method by processing and measuring the sample 3 times to determine the coefficient of variation (CV%), the coefficient of variation of the sample is as 4.1% (<11%, AOAC-2012). From this, it can be seen that the method used to analyze $239+240$ Pu has good repeatability. The recovery efficiency was measured by an isotope tracer 242 Pu, with an average recovery value of 63.8%. A few samples (DLS 20.1.1, DBLV 20.1.2) exhibited a low recovery of approximately 20%. The low recovery was due to interference from (organic substances and iron) in the sample. In particular, the high percentage of Fe in DLS 20.1.1, and DBLV 20.1.2, inhibited the oxidation of Pu3+ to Pu4+, thus affecting the capture of Pu on the Ion exchange column [10].

 $137Cs$ was determined by direct gamma measurements and had specific activities ranging from 0.786 to 2.129 Bq/kg DW, with an average value of (1.393 ± 0.370) Bq/kg DW (Table II).

Compared with previous domestic studies, the results of analyzing the specific activities of radioactive isotopes $^{239+240}$ Pu, 90 Sr, and $137Cs$ in this study are similar to the values reported in southern Vietnam by Sieu et al. and were smaller than those reported by Quang et al. with soil samples collected in northern and southern Vietnam. This difference may be due to the sampling depth. This study collected samples at a depth of (0-5) cm while the study by Quang et al. collected samples from (0-10) cm. The average Specific activity results and standard deviation in the study by Sieu et al are (0.81 \pm 0.56) Bq/kg DW for ¹³⁷Cs, (0.039 \pm 0.047) Bq/kg DW for $^{239+240}$ Pu and (0.39 \pm 0.17) Bq/kg DW for with 90 Sr [9]. The average Specific activity results and standard deviation in the study by Quang et al are (4.66 ± 3.65) Bq/kg DW for ^{137}Cs , (0.094 \pm 0.049) Bq/kg DW for ²³⁹⁺²⁴⁰Pu, and (0.56 \pm 0.28) Bq/kg DW for ^{90}Sr [7, 8].

The specific activity analysis results of radioisotopes $^{239+240}$ Pu, 90 Sr, and 137 Cs were relatively lower than those reported in neighboring countries. Specifically, the radioisotopes $^{239+240}$ Pu, 90 Sr, and 137 Cs were analyzed in Inner Mongolia, China. The average Specific activity results and standard deviation are (9.21 \pm 8.95) Bq/kg DW for ¹³⁷Cs, (0.41 ± 0.44) Bq/kg DW for ²³⁹⁺²⁴⁰Pu, and (4.32) \pm 2.48) Bq/kg DW for ⁹⁰Sr, which are all within the global fallout range [4].

The specific activities of ^{90}Sr , ^{137}Cs , and 239+240Pu in topsoil were also determined in Korea. The average Specific activity results and standard deviation are (3.82 ± 2.63) Bq/kg DW for ⁹⁰Sr, (33.2 \pm 16.1) Bq/kg DW for ¹³⁷Cs, and (0.80 ± 0.41) Bq/kg DW for ²³⁹⁺²⁴⁰Pu [6].

The average Specific activity results and standard deviation are (81.11 ± 95.43) Bq/kg DW for ^{137}Cs , (1.637 \pm 1.924) Bq/kg DW for ²³⁹⁺²⁴⁰Pu in Montenegro [1], and (6.2 ± 1.2) Bq/kg DW for 90 Sr, (88 \pm 31) Bq/kg DW for $137Cs$, (0.57 \pm 0.13) Bq/kg DW for $239+240Pu$ in Jordan. [5] Specific activity ranges and average values of ^{137}Cs , $^{239+240}Pu$, and ^{90}Sr isotopes in the soil of Vietnam and other countries are shown in Table IV.

		Specific activity, Bq/kg dry weight			
References	Locations	137 _{Cs}	$239+240$ Pu	90 Sr	
	Northern Vietnam	$0.786 - 2.129$ ^(*)	$0.018 - 0.058$	$0.022 - 0.697$	
This work		1.393 ± 0.370	0.039 ± 0.012	0.129 ± 0.145	
[9]	Southern Vietnam	$< 0.10 - 3.14$	$< 0.001 - 0.207$	$< 0.08 - 0.79$	
		0.81 ± 0.56	0.039 ± 0.047	0.39 ± 0.17	
	Northern and central Vietnam	$0.94 - 14.95$	$0.03 - 0.28$	$0.16 - 1.14$	
[7, 8]		4.66 ± 3.65	0.094 ± 0.049	0.56 ± 0.28	
$[4]$	China's Inner Mongolia	$0.26 - 28.3$	$0.05 - 1.26$	$1.2 - 7.6$	
		9.21 ± 8.95	0.41 ± 0.44	4.32 ± 2.48	
		$7.86 - 70.1$	$0.18 - 1.85$	$1.10 - 13.5$	
[6]	Korea	33.2 ± 16.1	0.80 ± 0.41	3.82 ± 2.63	
	Montenegro	$1.8 - 413$	$0.036 - 8.2$		
$[1]$		81.11 ± 95.43	1.637 ± 1.924		
		$7.50 - 576$	$0.28 - 1.01$	$2.8 - 11.4$	
$[5]$	Jordan	88 ± 31	0.57 ± 0.13	6.2 ± 1.2	

Table IV. Specific activity ranges and average values of ¹³⁷Cs, ²³⁹⁺²⁴⁰Pu, and ⁹⁰Sr isotopes in the soil in Vietnam and other countries

(*) Value in the upper row: Specific activity range

Lower row value: (Average specific activity) \pm (Standard deviation)

IV. CONCLUSIONS

The study conducted sampling and analysis of specific activities of artificial radioactive isotopes, including 90 Sr, $239+240$ Pu, and $137Cs$, in topsoil samples from some northern provinces of Vietnam. The specific activity averages of the isotopes $\frac{90}{9}$ Sr, $239+240$ Pu, and 137 Cs in the topsoil samples is (0.129 ± 0.145) Bq/kg DW, (0.039 ± 0.012) Bq/kg DW, (1.393 ± 0.370) Bq/kg DW, respectively. These results are similar to those measured in southern Vietnam and smaller than those measured in neighboring countries and around the world. Despite this limitation, the data obtained from this study will be useful as baseline data for ^{90}Sr , $239+240$ Pu, and 137 Cs specific activities in Vietnamese soils.

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