Study the impacts of TENORM from fertilizer on soil and vegetable and effective dose rate due to ingestion of vegetable at the agricultural zone of Hoc Mon, Ho Chi Minh city, Viet Nam

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Abstract

In this work, the impacts of TENORM (Technologically Enhanced Naturally Occuring Radioactive Materials) from fertilizer on soil and vegetable were estimated. The activity concentration of the natural radionuclides and then the annual effective dose rate due to ingestion of vegetable in the crops using fertilizers at the agricultural zone of Hoc Mon, Ho Chi Minh City, Viet Nam were investigated. The results showed that the soil after harvesting has the reduced total activity, except for the area which has the content increase of ^{238}U, ^{226}Ra and ^{210}Pb from over 45% (for ^{226}Ra) to 73% (for ^{238}U) due to using phosphorus fertilizer containing a large amount of radioactive ^{226}Ra (111.3 Bq kg^{-1}) and ^{238}U (46.5 Bq kg^{-1}). In general, the equivalent radium activity of the soil after a harvest is less than the limit of 370 Bq kg^{-1} given by UNSCEAR 2000 at the surveyed time. It proved that there are no signs of radioactive residues in agricultural land due to using fertilizer in the surveyed area after a crop. The values of the annual effective dose due to ingestion are still less than 1 mSv/y in all cases of surveyed vegetables. It can be concluded that the radiological impact on the surveyed vegetables is negligible.

Keywords: Annual effective dose rate; Fertilizer; HPGe gamma spectrometer; Radioactivity; Soil; Vegetable.

1. Introduction

The human surroundings always exist radioactive isotopes, the radiation emitted from the radioactive isotopes which are contained in fossil fuels, soil, rock, water, air, in some ore containing phosphate, in the vegetation and within the human body itself. The radioactive substances and radiation that can reach Earth are also caused by the interaction of cosmic rays with elements in the atmosphere. In addition, with the development of the global economy, the development of nuclear technology has created an enhanced radiation background through nuclear weapons tests, the operation of nuclear reactors developed to produce electricity, radioactive isotope technologies, etc.

Fertilizers are products from phosphate rock which contain relatively high concentrations of natural radionuclides. Therefore, its usage in large amounts every year in crops can redistribute radioactive trace elements in soils. Vegetables are terrestrial foods. The immigration of naturally occurring radioactive material (NORM) and technology enhanced naturally occurring radioactive material (TENORM) in soils could make enhance of radioactive nuclides in vegetables. People may be exposed through ingestion of vegetables that contains radionuclides resulting from fertilizers and soils. Plants can be contaminated by radioactive nuclides in many different ways. In the process of growing, plants will receive nutrients from soil, groundwater, rainwater, fertilizer. Therefore the radioactive nuclides will be accumulated in the plant. Similarly, the leaf surface may be contaminated by depositing radionuclides from the atmosphere or by irrigating contaminated water.

Bolca et al., 2007 determined the enhancement in natural radioactivity level for soils and vegetables due to the usage of phosphate fertilizers in agricultural lands of the Gediz River basin, Western Turkey. Lambert et al., 2007 showed that the negative side of the fertilizer is that the soil is contaminated with micronutrients and natural radioactivity. According to Khater et al., 2001, phosphate rock containing high natural radioactivity. Truong et al., 2018 shows that the activity concentration of ^{40}K in NPK fertilizer can be up to 13500 Bq/kg. So fertilization is likely to increase the number of natural radionuclides in the soil. Long-term exposure can lead the risk of external and internal radioactive exposure through the use of foods grown on fertilized soil.

In 2008, Al-Kharouf and his colleagues studied natural radioactivity, dose assessment and uranium uptake of some agricultural crops in Khan Al-Zabeeb, Jordan. The results showed that the green parts (leaves, stems, and roots) of cultivated plants tended to accumulate uranium about two times larger than fruit. The maximum dose values from consuming 1 kg of watermelon pulp are 3.1 and 4.7 nSv/y for ^{238}U and ^{234}U respectively. Radium equivalent activity and external hazard index exceeded the allowable limit of 370 Bq/kg and 1 respectively.

In 2014, Asaduzzaman et al. studied the transport of radioactive isotopes of ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y from the soil into root vegetables in some areas of Malaysia. Research results showed that the existence of artificial ^{88}Y isotopes in cassava samples which were grown in the Puchong area. The radioactivity of ^{226}Ra in cassava and sweet potato ranged from 116 to 141 Bq/kg and 49.6 to 81.5 Bq/kg, respectively. The activity of ^{232}Th in cassava in all surveyed areas was relatively lower than those of ^{226}Ra.

In 2016, Al-Hamarneh and his colleagues studied radioactivity and transfer factor of ^{226}Ra, ^{234}U and ^{238}U isotopes from soil to plants for 13 crops at the farms in the North West of Saudi Arabic. The results showed that the maximum transfer coefficient of 0.11 in fruit is for ^{226}Ra; 0.16 for ^{234}U and ^{238}U in the seed. The mean TF value indicated that the roots tend to accumulate Ra and U which are 4 to 6 times higher than for the fruit. In pepper, the TF ratio of fruit and root was lowest (0.07, 0.12, 0.11 of ^{226}Ra, ^{234}U, ^{238}U respectively). This ratio is greatest for potato (0.71, 0.41, 0.4 for ^{226}Ra, ^{234}U, ^{238}U respectively).

In this work, the influences of radioactivity in fertilizer on soil and vegetable after crops at the agricultural zone of the Hoc Mon district, Ho Chi Minh City, Viet Nam were evaluated. Besides, the activity of natural radionuclides in different vegetables which are commonly used in Vietnamese meals was estimated, the annual effective dose due to ingestion of these vegetables was calculated from which the radiological impacts of vegetable ingestion on human were evaluated at the surveyed zone.

2. Materials and Method

2.1. Materials

Vegetables are grown at the farm of Xuan Thoi Thuong zone, Hoc Mon District, Ho Chi Minh City, Viet Nam (Figure 1). Xuan Thoi Thuong is located to the southwest of Hoc Mon District, near Pham Van Hai farm (Binh Chanh district) in the West. This zone has an area of 18.09 km^2. Xuan Thoi Thuong is a fertile land with many canals and water sources, which are favorable for growing fresh vegetables. This is one of the large vegetable baskets of Ho Chi Minh City. Therefore, it is necessary to evaluate the quality of fresh vegetables through evaluation for the radioactivity concentration of natural radionuclides which is co-existing in vegetables after harvesting.

Fig 1. The sampling location at Hoc Mon District, Ho Chi Minh City, Vietnam.

To assess the effects of radioactivity in fertilizer on soil and vegetable after harvesting, we cultivated Ipomoea Aquatica on 13 plots. Each was supplied by a type of fertilizers given in Table 1 (F1 to F12, F13 is for non-use of fertilizers - the reference plot). The same fertilizer amount of 0.1 kg/m^2 was used for each plot. Soil samples (symbolized by S1 to S13) before planting (B) and after harvest (A) were collected. Similarly, after cultivation the Ipomoea Aquatica samples at 13 areas (symbolized by V1 to V13) were collected. These samples were prepared and analyzed by using gamma spectrometer with HPGe detector to access natural radioactivity of ^{238}U, ^{226}Ra, ^{232}Th, ^{210}Pb, ^{40}K in them, from that the transport of radioactivity in fertilizer on crops and residues if available after a crop were evaluated.

Table 1. The list of fertilizer types used for Ipomoea Aquatica crops

Sample	Fertilizer	N-P-K
F1	Korea DAP	NPK 18 – 46 – 0
F2	Super phosphate fertilizer (Long Thanh)	NPK 0 – 20 – 0

F3	Fused phosphate (Van Dien)	NPK 0 – 17 – 0
F4	997 TVL (Dau trau)	NPK 18 – 18 – 6
F5	999 TVL (Dau trau)	NPK 20 – 10 – 6
F6	Advanced Fertilizer	NPK 12-12-17-9 TE
F7	NPK 20-20-0	NPK 20-20-0
F8	Versatile (Dau trau)	NPK 17-12-7 +TE
F9	AVS (Con co vang)	NPK 20-20-15
F10	Viet Nhat	NPK 16-16-8 +13S
F11	Typical	NPK 14-13-13 -6
F12	Co bay	NPK 12-12-17 +2MgO
F13	The reference sample - No fertilizer used.	-

To evaluate effective dose rate from internal exposure due to ingestion of vegetables which were cultivated in the surveyed zone, the vegetable samples of Turnip (denoted by Tur), Basil (Bas), Amaranthus Tricolor (Amat), Ipomoea Aquatica (Ipo), Amaranthus (Ama), Mustard (Mus), Serrate leaf (Ser), Malabar spinach (Mas), Sui choy (Sui), Jute plant (Jute), Sweet potato leaf (Spo) which are commonly used in Vietnamese meals were studied. The samples of these vegetables were collected after crop. The fresh weight (g), ash weight (g) of these samples of vegetables were given in Table 2.

Each sample of vegetable was dried at room temperature, ashed at 450^{o}C in 24 hours. Each sample of soil or fertilizer was dried at room temperature, crushed to their particle sizes of less than 0.2 mm. Then samples were dried at 110^{o}C for 8 hours. Then they were packed in cylinder beakers with 20 mm in sample height and 76 mm in sample diameter. Samples were also sealed for about 30 days in order to ensure that a secular equilibrium between ^{226}Ra and its decay products of a short half - life has been reached.

Table 2. Fresh weight (g), ash weight (g) of the vegetable samples

No	Vegetables	Fresh weight (g)	Ash weight (g)
1	Turnip	7500	63.40

2	Basil	8400	133.27
3	Amaranthus Tricolor	5000	42.14
4	Ipomoea Aquatica	10000	116.10
5	Amaranthus	11500	65.91
6	Mustard	7000	94.02
7	Serrate leaf	8500	113.50
8	Malabar spinach	7400	133.53
9	Sui Choy	6500	72.00
10	Jute plant	2500	36.01
11	Sweet potato leaf	5000	66.77
12	Turnip radish	7500	63.40
13	Turnip leaf	5500	90.05
14	Ipomoea aquatica root	5100	55.79
15	Ipomoea aquatica stem	10000	108.47

2.2. Instrumentation and calibration

Activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th, and theirs daughters and ^{40}K radionuclides from these samples were measured by the gamma spectrometer using HPGe detector of GMX35 P4-70 and were calculated by the equation:

$$A = \frac{S}{\varepsilon(E) \times f \times m \times t \times K_c \times K_w} \quad (1)$$

where A is the sample activity concentration on the sample date (Bq/kg), S is the net peak area, $\varepsilon(E)$ is full energy peak efficiency of detector, f is the branching ratio of the E gamma energy under consideration, m is the mass of the sample (kg) and t is the collection live time (s), K_c is the correction factor for the nuclide decay during counting and K_w is the correction factor for the nuclide decay from the time the sample was obtained to the start of acquisition (Canberra Industries Inc., 2004).

The minimum detection activity MDA (Bq/kg) values were also calculated for every interested energy line by equation (2).

$$MDA = \frac{L_D}{\varepsilon(E) \times f \times m \times t \times K_c \times K_w} \quad (2)$$

where $L_D = 2.71 + 4.66\sqrt{B}$ is the detection limit for a confidence interval of 95%; B has calculated continuum under the peak (Canberra Industries Inc., 2004).

The Full Energy Peak Efficiency (FEPE) of the detector was calibrated by measurements of gamma spectra emitted from the radionuclides of uranium, thorium series and potassium in certified IAEA soil standard samples of RGU1, RGTh1 and RGK1. Self-absorption effect of gamma rays caused by the difference of composition and density between analyzed samples and standard samples were corrected by using the efficiency calculation software of Angle 3.0 (Ortec Industries Inc., 2012). The true coincidence summing effects were corrected by CCCC code (Vidmar, 2010).

The FEPE calibration and the calculation of the radioactivity were validated by the certification from annual test of IAEA (IAEA-TELworld wide proficiency tests).

The activity of radionuclide was estimated using gamma spectrometry by acquiring gamma spectra from itself or via its direct daughter radionuclide taking the weighted average of activities. In details, these are the 46.5 keV gamma for ^{210}Pb; ^{234}Th (63.38 keV gamma) and ^{234m}Pa (1001 keV gamma) for ^{238}U; ^{214}Pb (295 keV and 352 keV gammas), and ^{214}Bi (609 keV gamma) for ^{226}Ra; ^{228}Ac (338 keV, 795 keV and 911 keV gammas) for ^{232}Th; the ^{212}Pb, ^{212}Bi, ^{40}K activities were estimated by their 238 keV, 727 keV and 1460 keV gammas respectively. The MDA (minimum detection activity) values were also calculated for every interested gamma energy. The calculated values of activity were compared with these respective MDA values before giving the final results.

2.3. The annual effective dose due to ingestion of terrestrial food

The annual effective dose due to ingestion of terrestrial food, f, containing radionuclide, i, is given by Saueia et al.,2006.

$$E_V = \sum_i C_{V,i} U_V FCD_{ing,i} \quad (3)$$

E_V is the annual effective dose (Sv/y) due to ingestion of terrestrial food; $C_{V,i}$ is activity concentration of radionuclide i in the edible part of plants (Bq/kg); U_v is ingestion rate (kg/y); $FCD_{ing,i}$ is the dose conversion factor (Sv/Bq). The methodology of UNSCEAR, 2017 employs dose coefficients for an adult member of the public and are the committed effective doses to 70 years of age per unit intake of radionuclides given by ICRP, 2012.

The upper limit of the digestion rate of vegetables for Vietnamese people is 272 (g/capita/day) (National Institue of Nutrition, 2010). The values of $FCD_{ing,v}$ are presented in Table 3 (UNSCEAR, 2017).

Table 3. The dose conversion factor for adults to age 70 years from internal exposure $FCD_{ing,i}$

Radionuclide i	Progeny	$FCD_{ing,i}$ (Sv/Bq)
^{232}Th	^{228}Ra, ^{228}Ac, ^{228}Th, ^{212}Pb	2.3×10^{-7}
^{238}U	^{234}Th, ^{234m}Pa	4.5×10^{-8}
^{226}Ra	-	2.8×10^{-7}
^{214}Pb	-	1.4×10^{-10}
^{214}Bi	-	1.1×10^{-10}
^{210}Pb	-	6.9×10^{-7}

3. Results and discussions

3.1. Radioactivity in fertilizer samples

Table 4 presents the radioactivity of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb in fertilizer samples. The results show that there is much difference in radioactive activity in 12 NPK chemical fertilizers. It depends on the existence of chemical elements such as N, P, K and other nutrient content in the fertilizer sample and soil. The radioactivity concentration varies from 1.2 Bq kg^{-1} to 598.6 Bq kg^{-1} for ^{238}U , from 0.7 Bq kg^{-1} to 111.3 Bq kg^{-1}. for ^{226} Ra, from 44.9 Bq kg^{-1} to 6390.7 Bq kg^{-1} for ^{40}K, from 3.5 Bq kg^{-1} to 11.7 Bq kg^{-1} for ^{232}Th and from 0 Bq kg^{-1} to 50.4 Bq kg^{-1} for ^{210}Pb.

Table 4. The radioactivity concentration of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb in the fertilizer samples

Samples	**^{238}U**	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{210}Pb**
F1	598.6 ± 3.0	4.7 ± 0.3	3.9 ± 0.2	44.9 ± 2.9	<8.76
F2	49.1 ± 4.6	87.9 ± 4.3	7.1 ± 0.3	75.8 ± 4.8	46.6 ± 2.9
F3	46.5 ± 4.2	111.3 ± 5.4	9.4 ± 0.5	168.1 ± 10.4	<8.76

F4	33.4 ± 5.1	2.0 ± 0.2	4.7 ± 0.4	2009.3 ± 122.5	<8.76
F5	37.6 ± 4.7	5.5 ± 0.4	8.6 ± 0.5	1259.3 ± 3.1	<8.76
F6	1.2 ± 0.1	3.1 ± 0.3	10.5 ± 0.5	3364.4 ± 205.1	<8.76
F7	62.2 ± 3.7	5.1 ± 0.3	5.5 ± 0.5	240.7 ± 14.8	<8.76
F8	45.6 ± 5.4	1.8 ± 0.1	<1.34	1738.7 ± 106.0	<8.76
F9	24.4 ± 8.1	0.7 ± 0.1	3.5 ± 0.3	6390.7 ± 389.4	<8.76
F10	25.8 ± 5.3	2.4 ± 0.2	11.7 ± 0.6	1888.6 ± 115.2	<8.76
F11	19.7 ± 6.9	3.6 ± 0.2	7.5 ± 0.5	2918.5 ± 177.8	<8.76
F12	49.1 ± 2.2	91.9 ± 2.8	10.6 ± 0.6	74.7 ± 2.5	50.4 ± 3.1
Min	1.2 ± 0.1	0.7 ± 0.1	1.34	44.9 ± 2.9	<8.76
Max	598.6 ± 3.0	111.3 ± 5.4	11.7 ± 0.6	6390.7 ± 389.4	50.4 ± 3.1

It is noticed that the radioactivity of ^{238}U in the surveyed fertilizer of NPK almost are higher than the radioactivity of ^{226}Ra. The cause of this difference is attributed to the differences between production technologies, mining, processing and origin of phosphate ore. The concentration of ^{226}Ra will be significantly lost if the content of P_2O_5 is enriched by over 30% due to the chemical and thermal treatment and finish in phosphogypsum industrial waste (IAEA 2013).

The activity of radionuclide ^{226}Ra in the group of superphosphate (F2, F3, F12) is higher than other NPK fertilizers, the activity varies from 87.9 to 111.3 Bq kg^{-1}, with the average activity of 99.6 Bq kg^{-1}. It is explained that superphosphate in fertilizer of F1, F3, F12 (see Table 1) contains a large amount of phosphogypsum, so there is still a significant presence of radioisotope ^{226}Ra (IAEA 2013).

3.2. Radioactivity in the soil before planting and after harvesting

To assess the effect of residual fertilizer on cultivation, we conducted for soil sampling at the beginning of planting and after harvesting. The analytical results are given in Table 5 and Table 6. From the obtained results, the ratios of radioactivity in soil before planting and after harvesting were estimated. The results are shown in Figure 2.

Table 5. The radioactivity concentration (Bq/kg) of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb in soil samples before planting

Sample	**^{238}U**	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{210}Pb**
S1	49.08 ± 2.53	27.77 ± 2.87	26.19 ± 2.40	69.67 ± 2.84	78.30 ± 2.83
S2	48.31 ± 3.11	26.34 ± 3.14	26.25 ± 3.00	69.70 ± 3.44	77.32 ± 3.12
S3	39.04 ± 2.73	28.25 ± 3.39	24.95 ± 2.93	68.50 ± 3.27	70.49 ± 2.83
S4	54.99 ± 3.31	30.96 ± 3.47	27.36 ± 3.26	76.48 ± 3.48	75.00 ± 2.92
S5	30.21 ± 2.25	26.80 ± 2.86	22.56 ± 2.76	71.52 ± 3.10	75.90 ± 3.00
S6	41.52 ± 2.77	25.91 ± 3.11	19.24 ± 5.60	79.41 ± 3.67	60.92 ± 2.51
S7	33.37 ± 2.43	27.50 ± 3.39	24.16 ± 2.93	66.43 ± 3.21	63.52 ± 2.54
S8	48.41 ± 3.17	29.32 ± 3.24	26.74 ± 3.34	80.80 ± 3.79	72.66 ± 2.99
S9	39.69 ± 2.65	26.58 ± 3.32	24.82 ± 3.09	80.56 ± 3.59	81.19 ± 3.18
S10	37.79 ± 2.70	29.30 ± 3.12	26.84 ± 3.04	75.45 ± 3.51	77.94 ± 2.98
S11	31.86 ± 2.51	26.56 ± 4.75	27.32 ± 2.93	77.06 ± 3.66	76.60 ± 3.07
S12	34.72 ± 2.62	27.90 ± 3.28	25.23 ± 3.46	78.23 ± 3.61	66.84 ± 2.57
S13	46.17 ± 3.06	23.92 ± 16.78	24.17 ± 3.81	75.21 ± 3.64	78.88 ± 3.28

Table 6. The radioactivity concentration (Bq/kg) of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb in soil samples after harvesting

Sample	**^{238}U**	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{210}Pb**
S1	30.27 ± 2.45	26.45 ± 3.46	23.06 ± 3.26	69.97 ± 3.43	69.54 ± 2.69
S2	41.36 ± 2.57	27.75 ± 3.01	25.25 ± 2.62	65.66 ± 3.06	72.37 ± 2.91
S3	67.63 ± 4.14	41.19 ± 4.40	37.70 ± 4.03	110.60 ± 4.72	101.54 ± 3.76
S4	38.51 ± 2.72	28.75 ± 3.24	23.21 ± 3.12	63.37 ± 3.20	58.98 ± 2.38
S5	39.10 ± 2.72	28.12 ± 2.96	18.74 ± 3.41	69.08 ± 3.15	70.76 ± 2.93
S6	29.17 ± 2.23	25.83 ± 3.45	23.61 ± 2.67	70.48 ± 3.21	61.14 ± 2.57
S7	37.74 ± 2.51	27.74 ± 2.82	19.94 ± 3.82	70.34 ± 3.18	69.59 ± 2.83
S8	31.38 ± 2.49	26.99 ± 3.48	20.01 ± 3.43	67.13 ± 3.30	69.02 ± 2.82
S9	28.19 ± 2.41	28.39 ± 3.22	18.59 ± 3.71	70.97 ± 3.35	76.93 ± 3.35

S10	34.98 ± 2.61	26.04 ± 3.10	24.89 ± 3.55	81.31 ± 3.64	64.44 ± 2.61
S11	34.87 ± 2.78	27.07 ± 3.81	26.23 ± 3.36	71.30 ± 3.64	76.28 ± 3.13
S12	46.32 ± 2.94	25.03 ± 3.26	27.99 ± 3.12	74.45 ± 3.36	60.35 ± 2.42
S13	39.48 ± 2.86	28.31 ± 3.03	20.51 ± 3.60	68.87 ± 3.31	64.98 ± 2.88

Figure 2 shows the ratios of radioactivity concentration in the soil after harvesting and before planting for using different fertilizers.

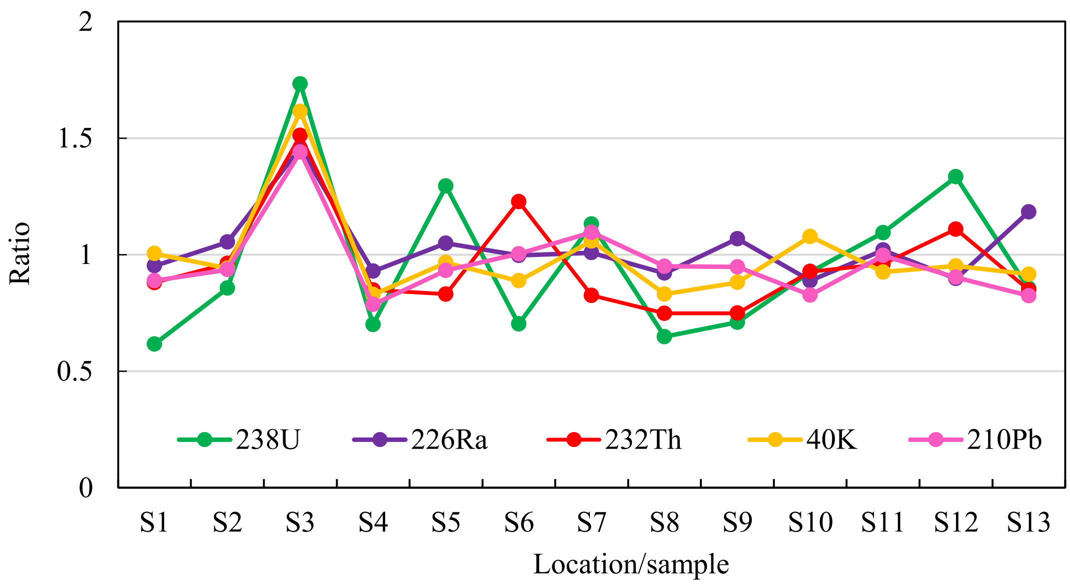

Fig 2. The ratios of radioactivity concentration in the soil after harvesting and before planting for using different fertilizers.

The analytical results show that soil samples after crops with using different fertilizer have the radioactivity of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb varying in the range from 0.5 to 1.5 times compared with the one in soil samples before planting. The trend varies depending on the type of radioisotope and the absorption mechanism of the vegetable.

The radioactivity in soil samples after harvesting (S13 sample – an area without fertilizer, Table 6) has changed slightly and the trend has decreased by about 10% for ^{40}K. It is explained by the fact that plants have used a quantity of the nutrient element for their growth and development.

Particularly, the radioactivity of ^{238}U, ^{226}Ra and ^{210}Pb in area S3 have increased from 45% (for ^{226}Ra) to 73% (for ^{238}U). The reason is that the phosphorus fertilizer (F3, Table 1) used in the area containing a large amount of ^{226}Ra (111.3 Bq kg^{-1}) and ^{238}U (46.5 Bq kg^{-1}) (see Table 4).

To assess the total effect of natural radioactivity in uranium, thorium series, ^{40}K and ^{210}Pb, we calculated the total activity of A_{tot} including equivalent radium activity, Raeq (contributed by ^{226}Ra, ^{232}Th, ^{40}K) and the contribution of ^{210}Pb to soil samples collected before planting and after harvesting, then the impact of the radioactive residues from the fertilizer on the surveyed agricultural land. Results are presented in Table 7.

In general, the soil after harvesting has a reduced total activity, which means that during the absorption and development process the plant has absorbed radioisotopes, so with a moderate amount of fertilizer, the soil after one crop has almost no radioactive residue from fertilizer, except for position S3, the content of ^{238}U, ^{226}Ra and ^{210}Pb increased from over 45% (for ^{226}Ra) to 73% (for ^{238}U). The reason is that the soil is fertilized with phosphorus (P3, Table 7.1) containing a large amount of radioactive ^{226}Ra (111.3 Bq kg^{-1}) and ^{238}U (46.5 Bq kg^{-1}). Note that the equivalent radium activity of the soil after harvest is less than the limit of 370 Bq kg^{-1} given by UNSCEAR 2000. It shows that there are no signs of radioactive residues in agricultural land due to fertilizer in the surveyed area.

Table 7. The total radioactivity (Bq/kg) of soil samples before planting and after harvesting for using the different fertilizers

Soil sample	Before planting (B)	After harvesting (A)	z-score (U-test)	Conclusion
S1	148.89 ± 5.30	134.35 ± 6.40	1.75	-
S2	146.56 ± 6.17	141.28 ± 5.62	0.63	-
S3	139.69 ± 6.09	205.16 ± 8.18	-6.42	up
S4	150.97 ± 6.51	125.80 ± 6.01	2.84	-
S5	140.47 ± 5.73	131.00 ± 6.42	1.10	-
S6	120.46 ± 8.95	126.16 ± 5.76	-0.54	-
S7	130.68 ± 5.96	131.26 ± 6.77	-0.06	-
S8	146.44 ± 6.51	129.79 ± 6.65	1.79	-
S9	149.47 ± 6.38	137.37 ± 7.06	1.27	-
S10	151.43 ± 6.13	132.33 ± 6.50	2.14	-
S11	148.16 ± 7.04	146.35 ± 6.89	0.18	-
S12	136.84 ± 6.47	131.14 ± 6.04	0.64	-
S13	143.15 ± 17.95	127.92 ± 6.64	0.80	-

Note: Using U-test to evaluate if radioactivity of soil sample after harvesting increases comparing with the one before using fertilizer? Null hypothesis was supposed that radioactivity of soil sample after harvesting does not to change or has a downtrend. The z-score of less than -2 prove that radioactivity of soil sample after harvesting has uptrend in the 95% confidence interval.

3.3. Determination of the radioactivity of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb in ipomoea aquatica samples using the different types of fertilizer (ash sample)

The natural radioactivity in the Ipomoea Aquatica samples which were grown in different fertilizer conditions ranged from 0.95 ± 0.15 to 10.91 ± 1.07 Bq kg^{-1} for ^{238}U, from 2.00 ± 0.33 Bq kg^{-1} to 7.26 ± 1.79 Bq kg^{-1} for ^{226}Ra, from 1.02 ± 0.58 Bq kg^{-1} to 4.19 ± 0.93 Bq kg^{-1} for ^{232}Th, from 14.56 ± 2.80 Bq kg^{-1} to 34.34 ± 8.25 Bq kg^{-1} for ^{210}Pb and from 1335.41 ± 40.62 Bq kg^{-1} to 3210.46 ± 97.04 Bq kg^{-1} for ^{40}K as shown in Table 8.

Results showed that ^{40}K accounts for the highest proportion among the radioisotopes, the second is ^{210}Pb, while ^{232}Th is found at the lowest activity. This can be explained by the fact that potassium is the plant's nutrient. Plants absorb potassium from the soil with various amounts, according to their metabolism. The highest activity of ^{40}K (3210.46 ± 97.04) Bq kg^{-1} in the V12 sample is explained by the fertilizer used in this location (F9, Table 1) was NPK type (20-20-15) which has the highest potassium content (6390.7 ± 389.4 Bq kg^{-1}). The lowest activity of ^{40}K (1335.41 ± 40.62) Bq kg^{-1} in the V1 sample is caused by the fact that there is no potassium in fertilizer sample used (F1, Table 1), but there are too much nitrogen and phosphor, then there is a competitive absorption in plant for these elements and potassium available in soil. In the meanwhile, the activity of ^{40}K (1629.49 ± 49.61) Bq kg^{-1} in V13 (no using fertilizer) shows that the plant absorbs potassium available in soils without any significant competition with other elements.

Table 8. The radioactivity of ^{238}U, ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K, in Ipomoea Aquatica samples using the different types of fertilizer

Ipomoea aquatica sample	^{238}U	^{226}Ra	^{232}Th	^{210}Pb	^{40}K
V1	2.77 ± 0.32	3.04 ± 0.48	4.07 ± 1.88	17.41 ± 5.46	1335.41 ± 40.62
V2	3.64 ± 0.47	5.97 ± 1.29	2.25 ± 0.54	26.33 ± 6.83	2217.83 ± 67.15

V3	< 0.24	2.00 ± 0.33	3.35 ± 0.75	27.83 ± 5.12	2384.21 ± 71.88
V4	1.32 ± 0.16	4.05 ± 0.82	1.74 ± 0.40	20.44 ± 6.15	2038.82 ± 61.49
V5	0.95 ± 0.15	3.94 ± 0.82	2.84 ± 0.77	34.34 ± 8.25	2175.93 ± 65.98
V6	7.30 ± 0.77	4.79 ± 1.03	3.55 ± 0.75	14.56 ± 2.80	2644.21 ± 79.75
V7	4.98 ± 0.7	7.26 ± 1.79	4.01 ± 5.95	31.58 ± 8.81	1747.34 ± 53.49
V8	4.39 ± 0.6	3.22 ± 0.55	2.1 ± 0.43	24.81 ± 6.94	2128.81 ± 64.49
V9	7.08 ± 1.02	4.46 ± 0.83	1.54 ± 1.77	21.58 ± 8.68	3210.46 ± 97.04
V10	< 0.24	2.78 ± 0.46	1.02 ± 0.58	29.49 ± 6.82	1974.46 ± 60.05
V11	6.64 ± 0.73	5.78 ± 1.28	3.48 ± 0.77	22.14 ± 4.26	1712.95 ± 52.04
V12	6.47 ± 0.67	3.29 ± 0.77	1.41 ± 0.19	24.41 ± 6.64	1889.75 ± 57.27
V13	3.34 ± 0.45	2.09 ± 0.63	2.51 ± 0.79	23.04 ± 6.22	1629.49 ± 49.61
Min	< 0.24	2.00 ± 0.33	1.02 ± 0.58	14.56 ± 2.80	1335.41 ± 40.62
Max	7.30 ± 0.77	7.26 ± 1.79	4.07 ± 1.88	34.34 ± 8.25	3210.46 ± 97.04

In addition, the plant absorbs more ^{210}Pb radioisotope than the other three isotopes such as thorium, radium and uranium. Although ^{210}Pb exists only in F2 and F12 fertilizer samples, the ^{210}Pb superiority in vegetables is explained by radioactive deposition from the air due to pollution. Besides, plants absorb a lot of ^{210}Pb from the soil through the root system.

The high activities of ^{210}Pb in V5, V7, V10 samples (34.34 Bq kg^{-1}, 31.58 Bq kg^{-1}, 29.49 Bq kg^{-1} respectively) were explained by using F5, F7, F10 fertilizers which have a high content of P_2O_5 (10%, 20% and 16% respectively).

The results also showed that in all of the surveyed Ipomoea Aquatica samples, plants absorbed more ^{226}Ra than ^{238}U and ^{232}Th. This can be explained by ^{226}Ra being a member of the ^{238}U radioactive chain, so ^{226}Ra is present in all uranium-containing environments, but ^{226}Ra usually exists in the form of water-soluble chemical compounds more than ^{238}U, it makes plant is easy to absorb. This was also found in the work of Verkhovskaya et al. 1969, Menzel, 1965.

3.4. The activity concentration of natural radionuclides in vegetables

To evaluate effective dose rate from internal exposure due to ingestion of vegetables which were cultivated in the surveyed zone, the vegetable samples which are commonly used in Vietnamese meals were grown using fertilizer as usual. The samples of these vegetable were then collected after crop (see Table 2). The activity of natural radionuclides in these vegetables was analyzed. The values were given in Table 9 and were illustrated in Figure 2, Figure 3 and Figure 4.

Table 9. The activity concentration of natural radionuclides in fresh vegetables

Samples	Fresh activity concentration (Bq/kg)								
	^{238}U	^{226}Ra		^{232}Th	^{212}Pb	^{212}Bi	^{208}Tl	^{40}K	^{210}Pb
		^{214}Pb	^{214}Bi	(^{228}Ac)					
Turnip (Tur)	0.828 ± 0.064	0.388 ± 0.012	0.356 ± 0.012	1.129 ± 0.057	0.080 ± 0.005	0.291 ± 0.055	0.078 ± 0.005	79.777 ± 2.566	1.201 ± 0.073
Basil (Bas)	0.136 ± 0.034	0.684 ± 0.027	0.720 ± 0.024	1.386 ± 0.059	0.257 ± 0.011	0.398 ± 0.123	0.310 ± 0.025	148.128 ± 4.742	0.353 ±0.136
Amaranthus Tricolor (Amat)	≤ 0.003	0.143 ± 0.009	0.123 ± 0.011	0.279 ± 0.021	0.035 ± 0.002	≤ 0.145	0.051 ± 0.006	84.466 ± 2.703	0.614 ±0.158
Ipomoea aquatica (Ipo)	0.172 ± 9.023	0.232 ± 0.009	0.222 ± 0.010	0.529 ± 0.035	0.079 ± 0.005	0.174 ± 0.052	0.060 ± 0.004	105.973 ± 3.405	0.144 ± 0.015
Amaranthus (Ama)	0.033 ± 0.004	0.269 ± 0.008	0.257 ± 0.010	0.571 ± 0.038	0.084 ± 0.004	0.170 ± 0.052	0.075 ± 0.004	68.077 ± 2.174	0.061 ±0.007
Mustard (Mus)	0.663 ± 0.076	0.332 ± 0.014	0.323 ± 0.016	0.748 ± 0.053	0.112 ± 0.008	0.110 ± 0.050	0.049 ± 0.004	122.267 ± 3.960	0.028 ±0.003
Serrate leaf (Ser)	0.045 ± 0.006	0.299 ± 0.012	0.295 ± 0.013	0.673 ± 0.044	0.090 ± 0.006	0.233 ± 0.065	0.117 ± 0.008	111.038 ± 3.581	0.140 ±0.014
Malabar spinach (Mas)	0.084 ±0.021	0.287 ± 0.017	0.261 ± 0.019	0.639 ± 0.029	0.110 ± 0.006	0.353 ± 0.090	0.117 ± 0.012	130.226 ± 4.164	≤ 0.025
Sui Choy (Sui)	≤ 0.005	0.433 ± 0.018	0.440 ± 0.018	0.795 ± 0.035	0.178 ± 0.007	0.276 ± 0.071	0.120 ± 0.012	94.218 ± 3.014	≤ 0.011
Jute plant (Jute)	0.131 ±0.030	0.816 ± 0.027	0.757 ± 0.031	1.155 ± 0.055	0.290 ± 0.012	0.285 ± 0.102	0.214 ± 0.023	110.876 ± 3.590	0.600 ±0.179
Sweet potato leaf (Spo)	0.014 ± 0.011	0.872 ± 0.026	0.827 ± 0.028	2.356 ± 0.120	0.346 ± 0.016	0.519 ± 0.109	0.270 ± 0.018	148.096 ± 4.774	0.323 ±0.033

The results showed that there is an accumulation of natural radionuclides in uranium series, thorium series, ^{40}K and especially of ^{210}Pb in surveyed vegetable samples with the different activity concentrations. The activity concentration of ^{40}K radionuclides in vegetable samples were always higher than those of other radionuclides (about 100 times), the most notable values of 148.13 ± 4.74; 148.10 ± 4.77; 130.23 ± 4.16; 122.27 ± 3.96 Bq/kg were found in basil, sweet potatoes, Malabar spinach, mustard respectively; the lowest value of 68.08 ± 2.17 Bq/kg was in Ipomoea Aquatica sample. It is explained that potassium plays an important role in the growth of plants and therefore increases productivity and quality for crops.

The distribution of ^{40}K activity concentrations in different vegetables in Figure 3 showed that the different vegetables have different uptakes of ^{40}K.

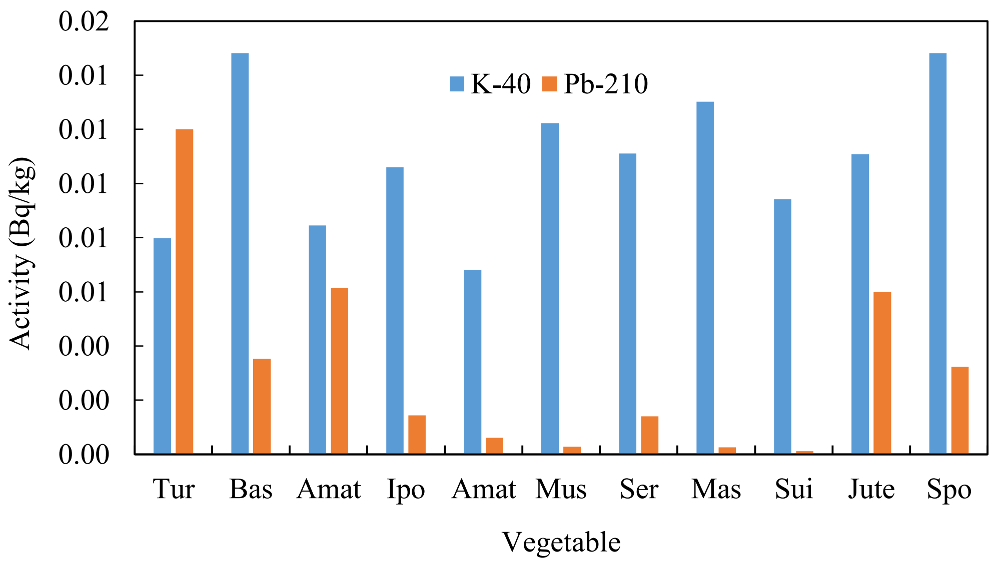

Fig. 3. Activity/100 of ^{40}K and activity ^{210}Pb in surveyed vegetables.

Figure 4 showed the activity concentrations of radionuclides in the ^{232}Th series for surveyed vegetables. The variation of activity concentration of ^{228}Ac ^{212}Pb, ^{212}Bi, ^{208}Tl in each vegetable sample proved that there is no secular equilibrium between the ^{232}Th radionuclides and their progenies of ^{228}Ac ^{212}Pb, ^{212}Bi, ^{208}Tl. The vegetables have more ^{228}Ac uptake than ^{212}Pb, ^{212}Bi, ^{208}Tl. The highest ^{228}Ac activity value of 2.356 Bq/kg is found in Sweet potato leaf. The activity concentration of ^{212}Pb, ^{212}Bi, ^{208}Tl are also not the same in the different vegetables indicating there is an uptake competition for different isotopes in the same vegetable.

It can be seen from Figure 4 that the ^{214}Pb and ^{214}Bi activty concentration are concentrated in basil, jute plant, and sweet potato leaf samples. The ^{214}Pb and ^{214}Bi activty concentration have relatively similar values in the same vegetable sample. In the meanwhile, the distribution of ^{238}U in vegetables is not the same as the distribution of ^{214}Pb, ^{214}Bi radionuclides. The vegetables have the different uptakes of ^{238}U. The ^{238}U activity has a high value of 0.828

Bq/kg and 0.663 Bq/kg for turnip sample and mustard respectively. It indicates that a secular equilibrium does not happens between ^{238}U radionuclides and their ^{226}Ra progenies.

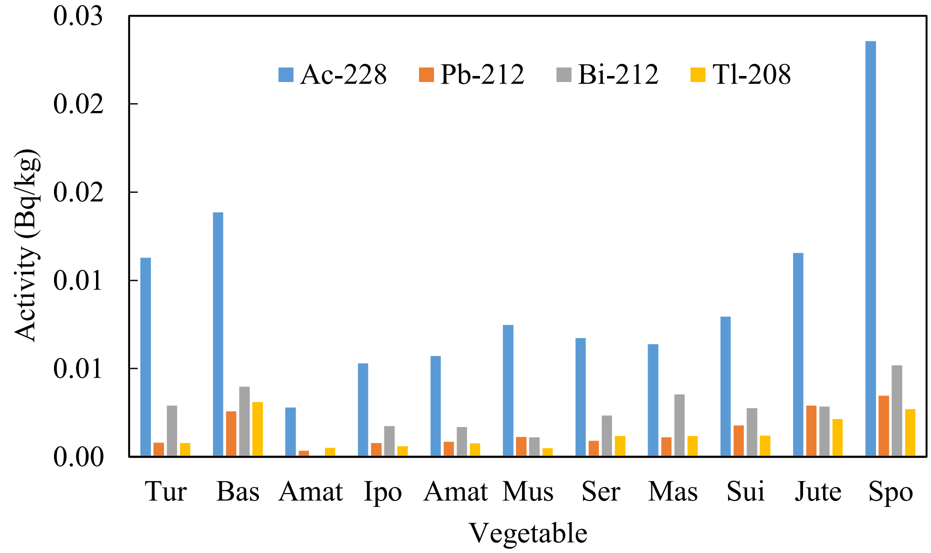

Fig. 4. The activity of radionuclides in ^{232}Th series in surveyed vegetables.

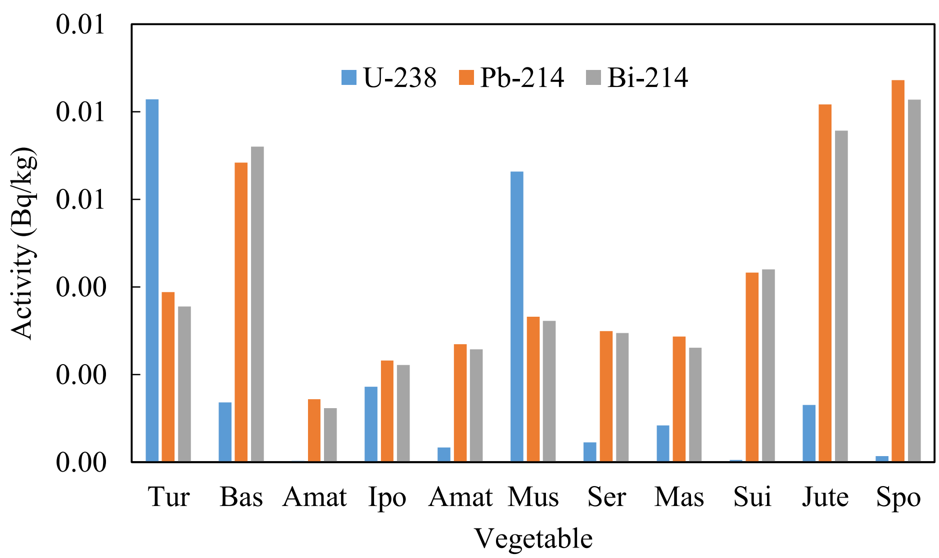

Fig. 4. The activity of ^{238}U, ^{214}Pb, ^{214}Bi in surveyed vegetables.

The ^{210}Pb radionuclides were also found in most of the vegetables (Figure 3) with relatively low activities such as Turnips, Amaranthus, Jute plant and Sweet potato leaf. The highest value of 1.201 Bq/kg is for Turnips. It is explained by the deposition of the ^{210}Pb radionuclide from the atmosphere into leaves and stems of plants and soil environment, by the potential soil pollution from around industry zones, therefore by ^{210}Pb uptake of plants from the soil environment.

3.5. Calculation of the effective dose rate due to ingestion of vegetables (terrestrial food)

The values of E_V from different vegetables were calculated based on the formula (3) and given in the Table 10. They are based on activity concentrations of ^{232}Th and its progeny of ^{228}Ac, ^{228}Ra, ^{228}Th, ^{212}Pb; ^{238}U and its progenies of ^{234}Th, ^{234m}Pa; ^{226}Ra; ^{214}Bi; ^{214}Pb and ^{210}Pb which are given in the Table 9 and the dose conversion factors which are in Table 3.

Table 10. The annual effective dose rate (mSv/y) due to ingestion of surveyed vegetables

Vegetables	E_V (mSv/y)
Turnip	0.122 ± 0.007
Basil	0.057 ± 0.011
Amaranthus Tricolor	0.052 ± 0.011
Ipomoea Aquatica	0.029 ± 0.002
Amaranthus	0.025 ± 0.002
Mustard	0.031 ± 0.002
Serrate leaf	0.033 ± 0.002
Malabar spinach	0.024 ± 0.001
Sui Choy	0.031 ± 0.001
Jute plant	0.090 ± 0.014
Sweet potato leaf	0.099 ± 0.006

It can be seen that turnip, jute plant and sweet potatoes leaf cause rather high internal exposure due to ingestion than the others. The values of the annual effective dose due to ingestion are 0.122 mSv/y, 0.090 mSv/y and 0.099 mSv/y for turnip, jute plant and sweet potato leaf respectively. However, these values for all cases of surveyed vegetables are less than the world average of 0,29 mSv/y (for total ingestion exposure of natural radioactivity) [UNSCEAR 2000, Table 31, Annex B]. It can be concluded that the radiological impact of surveyed vegetables is negligible.

Conclussion

- The activity of radionuclide ^{226}Ra in the group of superphosphate is higher than other NPK fertilizers. It is explained that superphosphate in fertilizer contains a large amount of phosphogypsum, so there is still a significant presence of radioisotope ^{226}Ra (IAEA 2013).

- Almost the surveyed soil samples after crops with using different fertilizer have the radioactivity of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb varying in the range from 0.5 to 1.5 times compared with the one in surveyed soil samples before planting. The trend varies depending on the type of radioisotope and the absorption mechanism of the vegetable. Particularly, the radioactivity of ^{238}U, ^{226}Ra and ^{210}Pb in the area using phosphorus fertilizer (S3) have increased from 45% (for ^{226}Ra) to 73% (for ^{238}U).
- In general, the soil after harvesting has a reduced total activity, which means that during the absorption and development process the plant has absorbed radioisotopes, so with a moderate amount of fertilizer, the soil after one crop has almost no radioactive residue from fertilizer, except for position S3, the content of ^{238}U, ^{226}Ra and ^{210}Pb increased from over 45% (for ^{226}Ra) to 73% (for ^{238}U) due to using phosphorus fertilizer (P3, Table 7.1) containing a large amount of radioactive ^{226}Ra (111.3 Bq kg^{-1}) and ^{238}U (46.5 Bq kg^{-1}).
- Note that the equivalent radium activity of the soil after harvest is less than the limit of 370 Bq kg^{-1} given by UNSCEAR 2000 at the surveyed time. It shows that there are no signs of radioactive residues in agricultural land due to fertilizer in the surveyed area after a crop.
- There is an accumulation of natural radionuclides in uranium series, thorium series and ^{40}K in surveyed vegetable samples with the different activity concentrations because of uptake and metabolites of plants with soil environment and the deposition of radionuclides from the atmosphere into leaves and stems of plants.
- There is no radioactivity equilibrium between ^{238}U and its progenies, ^{232}Th and its progenies in surveyed vegetable samples as an uptake competition for different radioisotopes in the same vegetable.
- The values of the annual effective dose due to ingestion for all cases of surveyed vegetables are less than the world average of 0.29 mSv/y (given by UNSCEAR 2000). It can be concluded that the radiological impact of surveyed vegetables is negligible.

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