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Impacts of TENORM from fertilizers on soil and vegetables and the effective dose rate due to ingestion of vegetables at the agricultural zone in Vietnam

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Abstract

In this work, the impacts of TENORM (Technologically Enhanced Naturally Occurring Radioactive Materials) from fertilizers on soil and vegetables were estimated. We investigated both the activity concentration of the natural radionuclides and the annual effective dose rate due to the ingestion of vegetables in the crops using fertilizers at the agricultural zone of Hoc Mon, Ho Chi Minh City, Vietnam. The results show that there have not yet been signs of radioactive residues from using conventional fertilizers in agricultural land after a crop at the surveyed area and time. The radiological impact of surveyed vegetables was negligible to the public health.

Keywords Annual effective dose rate · Fertilizer · HPGe · Radioactivity · Soil · TENORM · Vegetable

Introduction

Radioactive isotopes are found all over the human environment: in fossil fuel, soil, rocks, water, air, phosphate ore, vegetation, and within the human body itself. The radioactive substances and radiation that can reach the Earth are also caused by the interaction of cosmic rays with elements in the atmosphere. Also, with the development of the global economy, the advancement of nuclear technology has created an enhanced radiation background through atomic 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, the massive amounts of consumption every year in crops can redistribute radioactive trace elements in soils. Vegetables are terrestrial foods. The migration of naturally occurring radioactive material (NORM) and technology-enhanced naturally occurring radioactive material (TENORM) in soils could enhance radioactive nuclides in vegetables. People may be exposed through the ingestion of vegetables that contains radionuclides resulting from fertilizers and soils. Radioactive nuclides can contaminate plants in many different ways. In the process of growing, plants will receive nutrients from the surrounding environment: 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.

The previous studies of the natural radioactivity in phosphate rock, in NPK fertilizers, showed that the soil might be contaminated with both natural radionuclides and micronutrients [1, 2]. Indeed, the enhancement in natural radioactivity level for soils and vegetables due to the usage of phosphate fertilizers in agricultural lands were found in Bolca et al. [3]. The natural radioactivity, dose assessment, and uranium uptake of some crops in Khan Al-Zabeeb, Jordan was studied by Al-Kharouf et al. [4].

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 [5]. In 2016, Al-Hamarneh and his colleagues studied radioactivity and

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transfer factor (TF) 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 [6].

In this work, the natural radionuclide activity from fertilizers and their residues on soil and vegetables after crops at the agricultural zone of the Hoc Mon District, Ho Chi Minh City, Vietnam, were evaluated. Besides, the annual effective doses due to the ingestion of these vegetables, and the radiological impacts of vegetable ingestion on humans were assessed at the surveyed zone.

Materials and method

Materials

Vegetables were grown at the farm of Xuan Thoi Thuong zone, Hoc Mon District, Ho Chi Minh City, Vietnam (Fig. 1). Xuan Thoi Thuong is located in the southwest of the Hoc Mon District. Xuan Thoi Thuong has an area of 18.09 km^2 and is a fertile land with many canals and water sources, which are favourable for growing fresh vegetables. It is one of the extensive vegetable baskets of Ho Chi Minh City. Therefore, it is necessary to evaluate the quality of fresh vegetables thorough evaluation for the radioactivity concentration of natural radionuclides, which are still accumulated in vegetables after harvesting.

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

To evaluate the effects of radioactivity in fertilizer on soil and vegetable after harvesting, we cultivated Ipomoea Aquatica on 13 plots. Table 1 shows different types of fertilizer for each ($F1$ to $F12$ and $F13$ for non-use of fertilizers was used to be the reference plot). The same fertilizer amount of 0.1 kg/m^2 was used for each plot. Soil samples at 0–20 cm from the surface were selected because the root density is usually found in these soil layers for leafy vegetables [7]. At each plot, five topsoil samples were collected and mixed for a representative sample. They were taken from the respective plots before planting (B) and after harvesting (A) (denoted by $S1$ to $S13$). The Ipomoea Aquatica samples of 10 kg were collected at the 13 plots (denoted by $V1$ to $V13$). To ensure statistics in evaluating the radioactivity level for the 13 types of the concerned fertilizer, three fertilizer samples of 200 mg from the same type were mixed to have a representative sample of each kind of fertilizer. These samples were prepared and analyzed by using a gamma spectrometer with an HPGe detector to evaluate the natural radioactivity of ^{238}U, ^{226}Ra, ^{232}Th, ^{210}Pb, and ^{40} K.

To evaluate the 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 consumed in Vietnamese meals were studied. The samples of these vegetables were collected after a crop, and each sample had a fresh weight of about 10 kg.

The edible parts, including the leaf and stem of each sample of vegetables, were washed, dried at the room temperature, ashed at 450 °C for 24 h. Each sample of soil or

Table 1 The list of fertilizer types (commonly used in the Vietnam market) used for surveyed 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
$F13$	No fertilizer used	–

fertilizer was dried at the room temperature, crushed to their particle sizes of less than 0.2 mm. Then samples were dried at 105 °C, packed in cylinder beakers, and sealed for about 40 days to ensure the secular equilibrium between ^{226}Ra radionuclide and its short-lived decay products [8].

Instrumentation and calibration

Activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th, and their daughters and ^{40} K radionuclides from these samples were measured by the gamma spectrometer with high purity germanium (HPGe) detector of ORTEC Industries Inc. (GMX35 P4-70). The activity concentration of a specific radionuclide was determined using the relation given in Eq. (1).

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

where A is the sample activity concentration on the sampling date (Bqkg^{-1}), S is the net peak area, $\varepsilon(E)$ is full energy peak efficiency of the detector, f is the gamma yield of the E gamma energy under consideration, m is the mass of the sample (kg), and t is the live collection 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 [9].

Minimum Detectable Activity—MDA (Bq kg^{-1}) values were also calculated for every interested energy line as follows:

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

where $L_D = 2.71 + 4.66\sqrt{B}$ is the detection limit for a confidence interval of 95%; B is the continuum under the peak [9].

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 the certified IAEA soil standard samples of RGU1, RGTh1, and RGK1. The 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 -Angle 3.0 [10]. The true coincidence summing effects were corrected by the CCCC code [11].

The activity of radionuclide was estimated using the acquiring gamma spectra from itself or via its direct daughter radionuclides (taking the weighted average of activities). Briefly, these are the 46.5 keV gamma for ^{210}Pb; 63.38 keV gamma (^{234}Th) and 1001 keV gamma ($^{234\mathrm{m}}$Pa) for ^{238}U; 295 keV and 352 keV gammas (^{214}Pb), and 609 keV gamma (^{214}Bi) for ^{226}Ra; 338 keV, 795 keV and 911 keV gammas (^{228}Ac) 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 recorded activity of ^{232}Th was based on the assumption that ^{232}Th is in secular equilibrium with its progenies, which is often not true, particularly in the case of the agricultural land.

The MDA 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 [12]. The standard deviations of the activity concentration (A) were calculated from the error propagation of S-the net peak area, $\varepsilon(E)$-full energy peak efficiency of the detector, f-the branching ratio of the E gamma energy under consideration, m-the mass of the sample (kg) given in Loan et al. 2018b [2].

Radium equivalent activity-Ra$_{eq}$ (Bq kg^{-1})

The radium equivalent activity of the sample containing different levels of ^{226}Ra, ^{232}Th, and ^{40} K nuclides are estimated as follows:

$$\mathrm{Ra_{eq}} = 370\left(\frac{A_{\mathrm{Ra}}}{370} + \frac{A_{\mathrm{Th}}}{259} + \frac{A_K}{4810}\right) = A_{\mathrm{Ra}} + 1.43A_{\mathrm{Th}} + 0.077A_K. \tag{3}$$

where A_{Ra}, A_{Th}, and A_K are activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th (^{228}Ac), and ^{40} K nuclides respectively in the samples. The maximum value of Ra$_{eq}$ must be less than 370 Bq kg^{-1} to keep the absorbed dose of less than 1.5 mGy y^{-1} [13].

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. [14].

$$E_V = \sum_i C_{V,i} U_V \mathrm{FCD}_{\mathrm{ing},i} \tag{4}$$

E_V is the annual effective dose (Sv y^{-1}) due to ingestion of terrestrial food; $C_{V,\,i}$ is activity concentration of radionuclide i in the edible part of plants (Bq kg^{-1}); U_v is ingestion rate (kg y^{-1}); FCD$_{\mathrm{ing},\,i}$ is the dose conversion factor (Sv Bq^{-1}). The methodology of UNSCEAR, 2017 [15] employs dose coefficients for an adult member of the public and the committed effective doses to 70 years of age per unit intake of radionuclides given by ICRP, 2012 [16].

Note: The upper limit of the ingestion rate of vegetables for Vietnamese people is 272 g day^{-1} (per capita) [17].

The values of $\mathrm{FCD}_{\mathrm{ing},\,v}$ are given by UNSCEAR, 2017 [15].

Results and discussions

Natural activity concentration in fertilizer samples

Table 2 presented the activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th (^{228}Ac), ^{40} K, ^{210}Pb in fertilizer samples. The results showed that there is a difference in radioactivity concentrations in 12 types of NPK chemical fertilizers. The variation depends on the existence of chemical elements such as N, P, K, and other nutrient content in the fertilizer samples. The activity concentrations of fertilizer samples vary from 1.2 Bq kg^{-1} to 598.6 Bq kg^{-1} for ^{238}U, from 1.8 Bq kg^{-1} to 111.3 Bq kg^{-1} for ^{226} Ra, from 45 Bq kg^{-1} to 6391 Bq kg^{-1} for ^{40} K, from 0 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. Similar results can also be found in previous work [18].

It is noticed that the activity concentrations of ^{238}U in the surveyed fertilizer samples of NPK are mostly higher than the activity of ^{226}Ra. It is attributed to the differences between production technologies, mining, processing, and origin of phosphate ore. The activity 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 [19].

The activity concentrations of ^{226}Ra in the group of superphosphate (*F*2, *F*3, *F*12) are higher than these in other NPK fertilizers; their activities vary from 87.9 to 111.3 Bq kg^{-1}, with the average activity of 97.0 Bq kg^{-1}. This confirms the significant presence of radioisotope ^{226}Ra these samples in phosphogypsum samples [19].

Activity concentration of natural radionuclides in the soil before planting and after harvesting

We investigated soil samples before planting and after harvesting to evaluate the effect of residual fertilizer on cultivation. The analytical results of the activity concentration of natural radionuclides were given in Table 3. Following that, the ratios of activity concentration in the soil before planting and after harvesting were estimated and shown in Fig. 2.

Figure 2 shows the ratios of these activity concentrations in the soil samples after harvesting and before planting using different fertilizers.

The analytical results showed that soil samples using different fertilizers after crops contained ^{238}U, ^{226}Ra, ^{232}Th, ^{40} K, ^{210}Pb radionuclides with the activity concentrations in the range from 0.5 to 1.5 times compared with the ones in soil samples before planting. The trend varies with the type of radioisotope and the absorption mechanism of the vegetable. In general, with a moderate amount of fertilizer, the soil after one crop has almost no radioactive residue from fertilizer, except for the soil sample at the S3 location, which is fertilized with phosphorus fertilizer (*F*3, Tables 1 and 2) containing a large amount of radioactive ^{226}Ra (111.3 Bq kg^{-1}) and ^{238}U (46.5 Bq kg^{-1}). As a result, the radioactivity concentrations of ^{238}U, ^{226}Ra, and ^{210}Pb in soil samples at the S3 location increased from 45% (for ^{226}Ra) to 73% (for ^{238}U).

To evaluate the total effect of natural radioactivity in uranium, thorium series, ^{40} K, and ^{210}Pb, the equivalent radium activity, Raeq for soil samples collected before planting and after harvesting were calculated, then the radioactive residues from the fertilizer on the surveyed agricultural land were estimated. The results are presented in Table 4.

The equivalent radium activities of the soil samples after harvesting is less than the limit of 370 Bq kg^{-1} given by

Table 2 The natural activity concentrations (Bq kg^{-1}) in the fertilizer samples

Samples	^{238}U	^{226}Ra	^{232}Th (^{228}Ac)	^{40} K	^{210}Pb
*F*1	598.6 ± 3.0	4.7 ± 0.3	3.9 ± 0.2	45 ± 3	< 9.0
*F*2	49.1 ± 4.6	87.9 ± 4.3	7.1 ± 0.3	76 ± 5	46.6 ± 2.9
*F*3	46.5 ± 4.2	111.3 ± 5.4	9.4 ± 0.5	168 ± 10	< 9.0
*F*4	33.4 ± 5.1	2.0 ± 0.2	4.7 ± 0.4	2009 ± 123	< 9.0
*F*5	37.6 ± 4.7	5.5 ± 0.4	8.6 ± 0.5	1259 ± 3	< 9.0
*F*6	1.2 ± 0.1	3.1 ± 0.3	10.5 ± 0.5	3364 ± 205	< 9.0
*F*7	62.2 ± 3.7	5.1 ± 0.3	5.5 ± 0.5	241 ± 15	< 9.0
*F*8	45.6 ± 5.4	1.8 ± 0.1	< 1.3	1739 ± 106	< 9.0
*F*9	24.4 ± 8.1	0.7 ± 0.1	3.5 ± 0.3	6391 ± 389	< 9.0
*F*10	25.8 ± 5.3	2.4 ± 0.2	11.7 ± 0.6	1889 ± 115	< 9.0
*F*11	19.7 ± 6.9	3.6 ± 0.2	7.5 ± 0.5	2919 ± 178	< 9.0
*F*12	49.1 ± 2.2	91.9 ± 2.8	10.6 ± 0.6	75 ± 3	50.4 ± 3.1
Min	1.2 ± 0.1	1.8 ± 0.1	< 1.3	45 ± 3	< 9.0
Max	598.6 ± 3.0	111.3 ± 5.4	11.7 ± 0.6	6391 ± 389	50.4 ± 3.1

Table 3 The activity concentrations (Bq kg^{-1}) of ^{238}U, ^{226}Ra, ^{232}Th (^{228}Ac), ^{40} K, ^{210}Pb in soil samples before planting (*B*) and after harvesting (*A*)

Sample/ location	^{238}U		^{226}Ra		^{232}Th (^{228}Ac)		^{40} K		^{210}Pb	
	B	*A*	*B*	*A*	*B*	*A*	*B*	*A*	*B*	*A*
*S*1	49 ± 3	30 ± 3	28 ± 3	26 ± 3	26 ± 2	23 ± 3	70 ± 3	70 ± 3	78 ± 3	70 ± 3
*S*2	48 ± 3	41 ± 3	26 ± 3	28 ± 3	26 ± 3	25 ± 3	70 ± 3	66 ± 3	77 ± 3	72 ± 3
*S*3	39 ± 3	68 ± 4	28 ± 3	42 ± 4	25 ± 3	38 ± 4	69 ± 3	111 ± 5	70 ± 3	102 ± 4
*S*4	55 ± 3	39 ± 3	31 ± 3	29 ± 3	27 ± 3	23 ± 3	76 ± 3	63 ± 3	75 ± 3	59 ± 2
*S*5	30 ± 2	39 ± 3	27 ± 3	28 ± 3	23 ± 3	19 ± 3	72 ± 3	69 ± 3	76 ± 3	71 ± 3
*S*6	42 ± 3	29 ± 3	26 ± 3	26 ± 3	19 ± 6	24 ± 3	79 ± 4	70 ± 3	61 ± 3	61 ± 3
*S*7	33 ± 2	38 ± 3	28 ± 3	28 ± 3	24 ± 3	20 ± 4	66 ± 3	70 ± 3	64 ± 3	70 ± 3
*S*8	48 ± 3	31 ± 3	29 ± 3	27 ± 3	27 ± 3	20 ± 3	81 ± 4	67 ± 3	73 ± 3	69 ± 3
*S*9	40 ± 3	28 ± 2	27 ± 3	28 ± 3	25 ± 3	19 ± 4	81 ± 4	71 ± 3	82 ± 3	77 ± 3
*S*10	38 ± 3	35 ± 3	29 ± 3	26 ± 3	27 ± 3	25 ± 4	75 ± 4	81 ± 4	78 ± 3	64 ± 3
*S*11	32 ± 3	35 ± 3	27 ± 5	27 ± 4	27 ± 3	26 ± 3	77 ± 4	71 ± 4	77 ± 3	76 ± 3
*S*12	35 ± 3	46 ± 3	28 ± 3	25 ± 3	25 ± 3	28 ± 3	78 ± 4	74 ± 3	67 ± 3	60 ± 2
*S*13	46 ± 3	39 ± 3	27 ± 3	28 ± 3	24 ± 4	21 ± 4	75 ± 4	69 ± 3	79 ± 3	65 ± 3

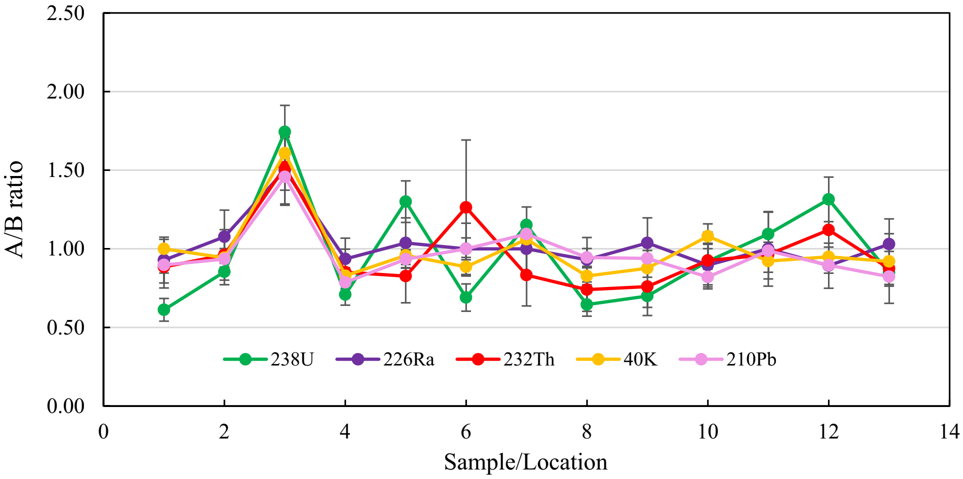

Fig. 2 The ratios of radioactivity concentrations in the soil after harvesting and before planting for using different fertilizers

UNSCEAR, 2000 [13]. It shows that there are no signs of radioactive residues in agricultural land due to fertilizer in the surveyed area.

Determination of the radioactivity of ^{238}U, ^{226}Ra, ^{232}Th, ^{40} K, ^{210}Pb in Ipomoea Aquatica samples (dry weight basis) using different fertilizers

Table 5 shows the natural radioactivity concentration in the Ipomoea Aquatica samples (in dry weight basis) which were grown in different fertilizer conditions ranged from 1.0 ± 0.2 to 10.9 ± 1.1 Bq kg^{-1} for ^{238}U, from 2.0 ± 0.3 Bq kg^{-1} to 7.3 ± 1.8 Bq kg^{-1} for ^{226}Ra, from 1.0 ± 0.6 Bq kg^{-1} to 4.2 ± 0.9 Bq kg^{-1} for ^{232}Th, from 14.6 ± 2.8 Bq kg^{-1} to 34.3 ± 8.3 Bq kg^{-1} for ^{210}Pb and from 1335 ± 41 Bq kg^{-1} to 3210 ± 97 Bq kg^{-1} for ^{40} K.

Among the radioisotopes, ^{40} K is the highest proportion and the second is ^{210}Pb, while ^{232}Th is found to be the lowest radioactivity concentration in Ipomoea Aquatica samples. It can be explained by the fact that potassium is a macronutrient for plants and plants absorb potassium from the soil to a certain extent, base to their metabolism. Thus, the highest radioactivity concentration of ^{40} K (3210 ± 97) Bq kg^{-1} in the *V*12 sample is explained that the fertilizer used in this location (*F*9, Table 1) was NPK (20-20-15), which has the highest potassium content (6391 ± 389) Bq kg^{-1}. The lowest activity of ^{40} K (1335 ± 41) Bq kg^{-1} in the *V*1 sample is caused by the fact that there is no potassium content in the fertilizer sample used (*F*1, Table 1). Because of the high content of nitrogen and phosphorus, competitive absorptions in plants might occur between these elements and potassium available in the soil. In the meanwhile, the activity of ^{40} K (1629 ± 50) Bq kg^{-1} in *V*13 (no using fertilizer) shows that the plant absorbs potassium available in soils without any significant competition with other elements.

Also, the plant has an uptake of more ^{210}Pb radioisotopes than the other three isotopes such as thorium, radium, and uranium. Although ^{210}Pb exists only in F2 and F12 fertilizer

Table 4 The equivalent radium activity (Bq kg^{-1}) of soil samples before planting (*B*) and after harvesting (*A*) using the different fertilizers

Soil sample	(*B*)	(*A*)	*z*-score (*U*-test)	Trend
*S*1	71 ± 4	65 ± 6	0.79	–
*S*2	69 ± 5	69 ± 5	0.05	–
*S*3	69 ± 5	104 ± 7	− 3.80	Up
*S*4	76 ± 6	67 ± 6	1.14	–
*S*5	65 ± 5	60 ± 6	0.58	–
*S*6	60 ± 9	65 ± 5	− 0.55	–
*S*7	67 ± 5	62 ± 6	0.67	–
*S*8	74 ± 6	61 ± 6	1.56	–
*S*9	68 ± 6	60 ± 6	0.94	–
*S*10	73 ± 5	68 ± 6	0.70	–
*S*11	72 ± 6	70 ± 6	0.17	–
*S*12	70 ± 6	71 ± 6	− 0.10	–
*S*13	67 ± 6	63 ± 6	0.43	–

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

Table 5 The activity concentrations (Bq kg^{-1}) of ^{238}U, ^{226}Ra, ^{232}Th, ^{210}Pb, and ^{40} K, in Ipomoea Aquatica samples, using different types of fertilizer

Ipomoea aquatica sample	^{238}U	^{226}Ra	^{232}Th (^{228}Ac)	^{210}Pb	^{40} K
*V*1	2.8 ± 0.3	3.0 ± 0.5	4.1 ± 1.9	17.4 ± 5.5	1335 ± 41
*V*2	3.6 ± 0.5	6.0 ± 1.3	2.3 ± 0.5	26.3 ± 6.8	2218 ± 67
*V*3	< 0.2	2.0 ± 0.3	3.4 ± 0.8	27.8 ± 5.1	2384 ± 72
*V*4	1.3 ± 0.2	4.1 ± 0.8	1.7 ± 0.4	20.4 ± 6.2	2039 ± 61
*V*5	1.0 ± 0.2	3.9 ± 0.8	2.8 ± 0.8	34.3 ± 8.3	2176 ± 66
*V*6	7.3 ± 0.8	4.8 ± 1.0	3.6 ± 0.8	14.6 ± 2.8	2644 ± 80
*V*7	5.0 ± 0.7	7.3 ± 1.8	4.0 ± 3.0	31.6 ± 8.8	1747 ± 53
*V*8	4.4 ± 0.6	3.2 ± 0.6	2.1 ± 0.4	24.8 ± 6.9	2129 ± 64
*V*9	7.1 ± 1.0	4.5 ± 0.8	1.5 ± 0.8	21.6 ± 8.7	3210 ± 97
*V*10	< 0.2	2.8 ± 0.5	1.0 ± 0.6	29.5 ± 6.8	1974 ± 60
*V*11	6.6 ± 0.7	5.8 ± 1.3	3.5 ± 0.8	22.1 ± 4.3	1713 ± 52
*V*12	6.5 ± 0.7	3.3 ± 0.8	1.4 ± 0.2	24.4 ± 6.6	1890 ± 57
*V*13	3.3 ± 0.5	2.1 ± 0.6	2.5 ± 0.8	23.0 ± 6.2	1629 ± 50
Min	< 0.2	2.0 ± 0.3	1.0 ± 0.6	14.6 ± 2.8	1335 ± 41
Max	7.3 ± 0.8	7.3 ± 1.8	4.1 ± 1.9	34.3 ± 8.3	3210 ± 97

samples, the ^{210}Pb superiority in vegetables is explained by radioactive deposition from the air. Besides, there is an uptake of a lot of ^{210}Pb from the soil through the root system.

The results also showed that in all of the surveyed Ipomoea Aquatica samples, plants absorbed more ^{226}Ra than ^{238}U and ^{232}Th. It can be explained that ^{226}Ra being a member of the ^{238}U radioactive chain presents in all uranium-containing environments, and ^{226}Ra usually exists in the form of water-soluble chemical compounds more than ^{238}U. Finally, it makes the plant is easy to absorb ^{226}Ra, which was also found in the work of Menzel and Verkhovskaya et al. [20, 21].

The activity concentration of natural radionuclides in common vegetables

To evaluate the effective dose rate from internal exposure due to the 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 vegetables were then collected after a crop. The activity of natural radionuclides in these vegetables was analyzed. The values were given in Table 6.

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 showed higher than those of other radionuclides (about 100 times). The most important values of 148 ± 5; 148 ± 5; 130 ± 4; 122 ± 4 Bq kg^{-1} were found in basil, sweet potatoes, malabar spinach, mustard, respectively; the lowest value of 68 ± 2 Bq kg^{-1} was in the amaranthus 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 showed different uptakes of ^{40} K. The variation of activity concentration of ^{228}Ac ^{212}Pb, ^{212}Bi, ^{208}Tl in each vegetable sample proved the non-secular equilibrium between the ^{232}Th radionuclides and their progenies of ^{228}Ac ^{212}Pb, ^{212}Bi, ^{208}Tl. The vegetables uptake more ^{228}Ac than ^{212}Pb, ^{212}Bi, ^{208}Tl. The highest ^{228}Ac activity value of 2.36 Bq kg^{-1} is found in sweet potato leaf. The radioactivity 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.

The ^{214}Pb and ^{214}Bi radioactivity concentrations are concentrated in basil, jute plant, and sweet potato leaf samples. The ^{214}Pb and ^{214}Bi radioactivity concentrations 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 different uptakes of ^{238}U. The ^{238}U activity has a high value of 0.83 Bq kg^{-1} and 0.66 Bq kg^{-1} for turnip sample and mustard, respectively.

Table 6 The activity concentration of natural radionuclides in fresh vegetables (wet weight basis)

Samples	Activity concentration (Bq kg^{-1})								
	^{238}U	^{226}Ra		^{232}Th	^{212}Pb	^{212}Bi	^{208}Tl	^{40} K	^{210}Pb
		^{214}Pb	^{214}Bi	(^{228}Ac)					
Turnip (Tur)	0.83 ± 0.06	0.39 ± 0.01	0.36 ± 0.01	1.13 ± 0.06	0.08 ± 0.01	0.29 ± 0.06	0.08 ± 0.01	80 ± 3	1.20 ± 0.07
Basil (Bas)	0.14 ± 0.03	0.68 ± 0.03	0.72 ± 0.02	1.39 ± 0.06	0.26 ± 0.01	0.40 ± 0.12	0.31 ± 0.03	148 ± 5	0.35 ± 0.14
Amaranthus Tricolor (Amat)	≤ 0.01	0.14 ± 0.01	0.12 ± 0.01	0.28 ± 0.02	0.04 ± 0.01	≤ 0.15	0.05 ± 0.01	84 ± 3	0.61 ± 0.16
Ipomoea aquatica (Ipo)	0.17 ± 9.02	0.23 ± 0.01	0.22 ± 0.01	0.53 ± 0.04	0.08 ± 0.01	0.17 ± 0.05	0.06 ± 0.01	106 ± 3	0.14 ± 0.02
Amaranthus (Ama)	0.03 ± 0.01	0.27 ± 0.01	0.26 ± 0.01	0.57 ± 0.04	0.08 ± 0.01	0.17 ± 0.05	0.08 ± 0.01	68 ± 2	0.06 ± 0.01
Mustard (Mus)	0.66 ± 0.08	0.33 ± 0.01	0.32 ± 0.02	0.75 ± 0.05	0.11 ± 0.01	0.11 ± 0.05	0.05 ± 0.01	122 ± 4	0.03 ± 0.01
Serrate leaf (Ser)	0.05 ± 0.01	0.30 ± 0.01	0.30 ± 0.01	0.673 ± 0.04	0.09 ± 0.01	0.23 ± 0.07	0.12 ± 0.01	111 ± 4	0.14 ± 0.01
Malabar spinach (Mas)	0.08 ± 0.02	0.29 ± 0.02	0.26 ± 0.02	0.64 ± 0.03	0.11 ± 0.01	0.35 ± 0.09	0.12 ± 0.01	130 ± 4	≤ 0.03
Sui Choy (Sui)	≤ 0.01	0.43 ± 0.02	0.44 ± 0.02	0.80 ± 0.04	0.18 ± 0.01	0.28 ± 0.07	0.12 ± 0.01	94 ± 3	≤ 0.01
Jute plant (Jute)	0.13 ± 0.03	0.82 ± 0.03	0.76 ± 0.03	1.16 ± 0.06	0.29 ± 0.01	0.29 ± 0.10	0.21 ± 0.02	111 ± 4	0.60 ± 0.18
Sweet potato leaf (Spo)	0.01 ± 0.01	0.87 ± 0.03	0.83 ± 0.03	2.36 ± 0.12	0.35 ± 0.02	0.52 ± 0.11	0.27 ± 0.02	148 ± 5	0.32 ± 0.03
Min	0	0.14 ± 0.01	0.12 ± 0.01	0.28 ± 0.02	0.04 ± 0.01	0	0.05 ± 0.01	68 ± 2	0
Max	0.83 ± 0.06	0.87 ± 0.03	0.83 ± 0.03	2.36 ± 0.12	0.35 ± 0.02	0.52 ± 0.11	0.31 ± 0.03	148 ± 5	1.20 ± 0.07

It indicates that a secular equilibrium does not happen between ^{238}U radionuclides and their ^{226}Ra progenies.

The ^{210}Pb presents in most of the vegetables, such as turnips, amaranthus, jute plant, and sweet potato leaf with relatively low concentration with the highest value of 1.20 Bq kg^{-1} in 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.

Besides, sweet potato leaf absorbed more natural radionuclides as ^{226}Ra, ^{232}Th, and their progenies than the other vegetable samples, especially for amaranthus tricolor.

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 (4) and given in Table 7.

It can be seen that turnip, jute plant, and sweet potatoes leaf cause rather high internal exposure due to ingestion than the others. The biggest values of the annual effective dose due to ingestion are 0.122 mSv y^{-1} for turnip. These values for all cases of surveyed vegetables are lower than the world average of 0.290 mSv y^{-1} (for total ingestion exposure of natural radioactivity) (UNSCEAR, 2000, Table 31, Annex B) [13]. It can be concluded that the radiological impact of surveyed vegetables is negligible to the public health.

Table 7 The annual effective dose rate (mSv y^{-1}) due to ingestion of surveyed vegetables

Vegetables	E_V (mSv y^{-1})
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

Conclusions

- All the surveyed soil samples after a crop using different fertilizers have the radioactivity concentrations 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 ones in surveyed soil samples before planting. The trend depends on the type of radioisotope and the absorption mechanism of the vegetable. Particularly, the radioactivity concentrations of ^{238}U, ^{226}Ra, and ^{210}Pb in the area using phosphorus fertilizer have increased from 45% (for ^{226}Ra) to 73% (for ^{238}U).
- The soil samples after harvesting have a reduced equivalent radium activity. During the absorption and devel-

opment process, the plant has absorbed radioisotopes. Therefore, with a moderate amount of fertilizer, the soil after a crop has almost no radioactive residue from fertilizer, except for the area using phosphorus fertilizer.

- The equivalent radium activity of all soil samples after harvesting is less than the limit of 370 Bq kg^{-1}. The values of the annual effective dose due to ingestion for all cases of surveyed vegetables are less than the world average of 0.290 mSv y^{-1}. There are no signs of radioactive residues in agricultural land due to fertilizer in the surveyed area after a crop.
- It can be concluded that the radiological impact of surveyed vegetables is negligible.

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Compliance with ethical standards

Conflict of interest The authors declare no conflict of interest.

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