CONTAMINATION OF ^{226}Ra RADIONUCLIDE IN WELL WATER, ASSOCIATED HEALTH RISKS AND LONG-TERM BEHAVIORS IN AGRICULTURAL SOILS: A CASE STUDY IN PHU YEN PROVINCE, VIETNAM

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Abstract: Sixty samples of well water were collected in a coastal area of Phu Yen province, Vietnam for ^{226}Ra analysis. Activity concentrations of ^{226}Ra (A-^{226}Ra) were found from 1.7 to 159 mBq L^{-1} with the average value of 5.44 mBq L^{-1}. The results showed that A-^{226}Ra in well water samples related to the topographical conditions of sampling locations and types of well. The values of A-^{226}Ra, effective dose and excess lifetime cancer risk did not exceed their recommended values. Long-term change of A-^{226}Ra in agricultural soils due to well water irrigation was based on model assessment. The results showed that A-^{226}Ra increased continuously under the cultivation process including water irrigation and fertilizer application. However, it got an equilibrium value after a certain period of the agricultural cultivation. This period correlated with A-^{226}Ra in irrigation water and A-^{226}Ra partition lost by processes of volatilization, leaching, radioactive decay and plant uptake.

Keywords: *^{226}Ra radionuclide, well water, soil model, soil-plant ecosystem*

1. INTRODUCTION

Contamination of ^{226}Ra in well water caused a potential health risk to population directly by drinking water or indirectly by ingestion of food crops. Many previous research carried out the ^{226}Ra measurement in ground water including well water [1-3]. Most of these studies were focused on evaluating the levels of radioactivity and the associated radiation dose in case of ingestion as well as searching for correlations with soil and bedrock radioactivity and its geochemical parameters. The calculation of the radiological dose was simple based on the activity concentration, the consumption intake rate of water and the dose conversion factor from ICRP [4]. However, the influences of ^{226}Ra in well water used for agricultural irrigation on the activity of ^{226}Ra in the agricultural soils have not been studied. In this study, we propose a modelling method for assessment the long-term change of ^{226}Ra activity in agricultural soil under the irrigation of the well water. The model was discussed in our previous publications [5, 6].

2. THE MAIN PART OF REPORT

2.1. Study area

Tuy An is one of the districts that nearby the East Sea, the map of Tuy An district is shown in Fig. 1a. The study area could be divided to two regions based on their topographics. They are a swamp region located in the east of An My district and near the East Sea and a hilly region located in the west of An My district. In Tuy An, a significant part of drinking water is from ground water. Besides that, ground water is irrigation water for vegetable cultivation in some regions (Fig. 1c). Ground water is exploited by two types of well. Dug wells are the wells which have the depth from 5 to 10 m, while drill wells have the depth from 10 to 40 m.

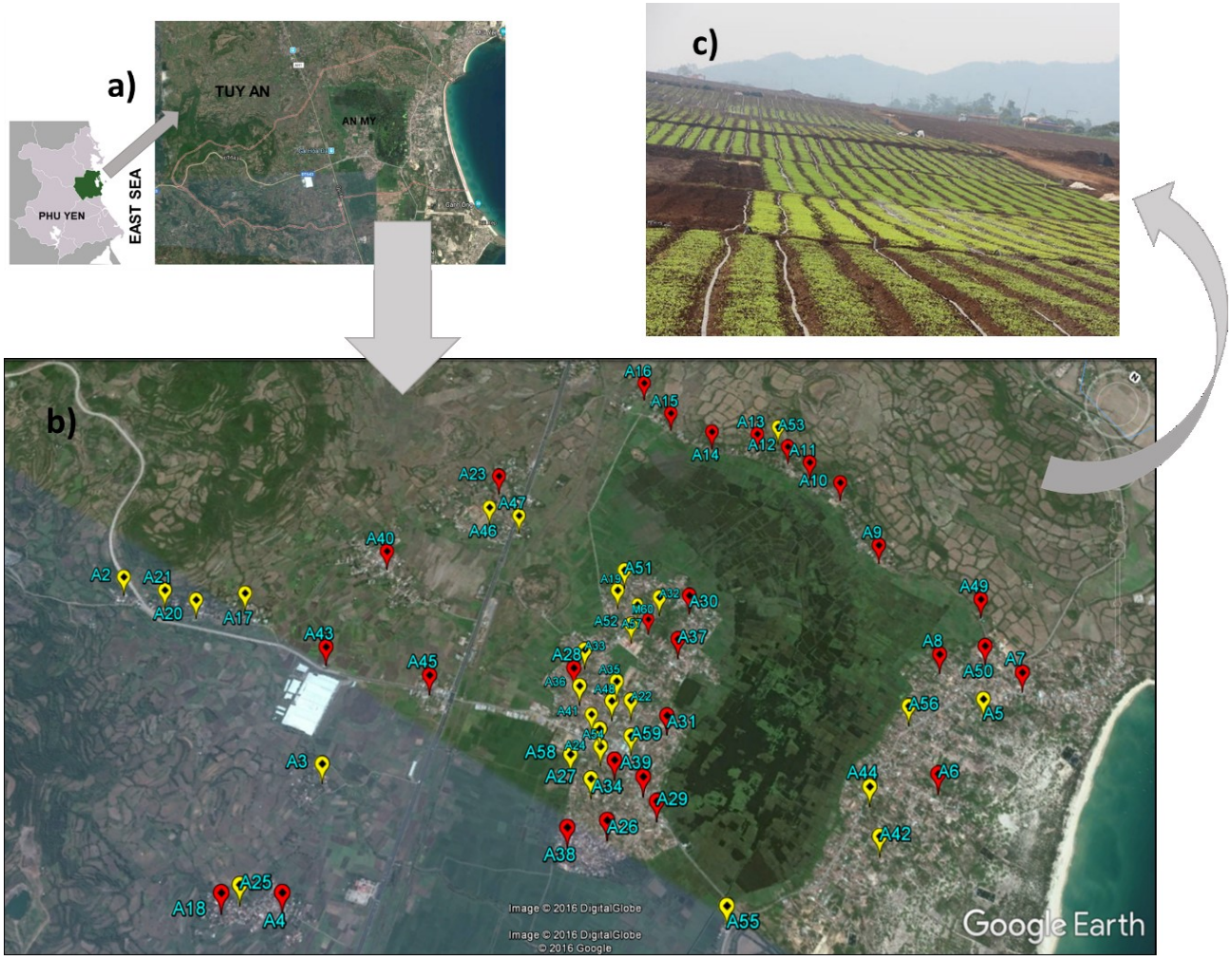

Fig. 1. a) Map of Phu Yen province shows the study area in the insert, b) Map of study area shows the sampling points (The red icons present the locations of dug wells while the yellow icons present the locations of drill wells), and c) Picture of the vegetable farm located in the study area.

2.2. Research subjects and methodology

In this study, 60 well water samples from two types of wells were collected from 60 different locations throughout An My, Tuy An district, Phu Yen provice, Vietnam. The sampling locations are presented in Fig.1b. About 2 L of well water collected from 60 wells was used for analysis. For ^{226}Ra analysis, the preparation procedure of well water samples is described in our previous publication [7]. In this study, the measurement of ^{226}Ra in well water samples was performed by an alpha spectroscopy (ORTEC) equipped with Passivated Implanted Planar Silicon detectors (PIPS). The method of radium co-precipitation with MnO_2 on the stainless steel disks was applied for sample preparation. In oder to assess the health risks to population, the concepts of annual effective dose (D_{eff}) and excess lifetime cancer risk (ELCR) were employed [4]. The calculations of D_{eff} and ELCR was explain in our publications [8, 9].

The soil model was supported by Canadian Centre for Environmental Modelling and Chemistry (CEMC). It is well known as a simple assessment of the relative potential for degrading reaction, evaporation, and leaching of a pesticide applied to a surface soil. The soil model was based on the fugacity approach which was described in two publications Mackay and Stiver (1991) and Mackay (2001) [10, 11]. However, this method is only applicable for chemicals with a measurable vapor pressure. For most nonvolatile chemicals, such as heavy metal an equivalence approach is used instead [12]. The details of the equivalence approach were described in previous studies [12-14]. The combination fugacity approach and equivalence approach in the Quantitative Water-Air-Sediment Interaction (QWASI) model was applied for ^{226}Ra and ^{210}Po radionuclides in aquatic system [15, 16]. Similarly that, fate

of ^{137}Cs was modeled by the CEMC soil model in our previous publications [5, 6]. For modeling, the concentration and the leaching rate of ^{226}Ra in the topsoil were important output values that were necessary for further calculations.

Equation (1) expresses the long-term change of A-^{226}Ra (C Bq kg^{-1}) in the agricultural soil due to various processes.

$$C = \frac{\left(C_{water} + C_{fertilizer}\right)\left(1 - e^{-(L_V + L_L + L_D + L_U)\times t}\right)}{L_V + L_L + L_D + L_U} \quad (1)$$

where, $C_{fertilizer}$ is A-^{226}Ra in fertilizer (Bq d^{-1}), C_{water} is the daily supply for irrigation (Bq d^{-1}) and t is the time in day. In this study, UPA fertilizer-a type of NPK fertilizers was used for long-term assessment of ^{226}Ra in the agricultural soil because A-^{226}Ra in UPA fertilizer is significantly high (139 Bq kg^{-1}) [7]. In the considered scenario, the application rate of the fertilizers was taken to be 80 kg UPA ha^{-1} $year^{-1}$, the application rate of well water was taken to be 5 L m^{-2} d^{-1} (actual application rates may differ depending on the plant and soil properties), and the homogenous distribution of radionuclides was assumed in the upper 20 cm of the soil (1 × 1 × 0.2 m) [6]. The soil's apparent density was taken as 1.3 g cm^{-3}. Four factors L_V, L_L, L_D and L_U (Bq d^{-1}) are the partitions of ^{226}Ra lost by volatilization, leaching, radioactive decay and plant uptake. L_V, L_L and L_D are calculated by the soil model. L_U was based on the soil-to-plant transfer factor (TF) [17] and the cultivation yield of vegetables. In the considered scenario, the cultivation yield of vegetables was assumed 5 kg for the cultivation time of 30 days.

For simulation behaviors of ^{226}Ra in the agricultural soils, soil adsorption coefficient (K_d) and organic carbon-water partition coefficient (K_{OC}) were from IAEA TRS-472 [18]. For simulation, some agricultural soil samples (the depth of 0-20 cm) was collected from the study area for determination of the soil characteristics. The average values of soil water content (23%), soil air content (27%), clay content (16%) and organic matter content (11.8%) were used for model input. Other general properties of soil were available in the model [10,11].

2.3. Results

The comparison between A-^{226}Ra in water samples collected from 30 dug wells in the study area was shown in Fig. 3a. According to the plot, the activities of ^{226}Ra were varied from 1.7 ± 0.3 to 85 ± 5 mBq l^{-1} with the mean value of 4.29 ± 0.13 mBq l^{-1}. Particularly, the low levels of A-^{226}Ra (<10 mBq l^{-1}) concentrated in well water samples collected from wells 4, 5, 6, 9, 11, 12, 15, 18, 19, 20, 21, 24 and 27. The high levels of A-^{226}Ra (>30 mBq l^{-1}) distributed in well water samples collected from wells 1, 2, 3, 13 and 25.

The comparison between A-^{226}Ra in water samples collected from 30 drill wells in the study area was shown in Fig. 3b. According to the plot, the activities of ^{226}Ra are widely varied from 4.9 ± 0.8 to 159 ± 9 mBq l^{-1} with the mean value of 12.9 ± 0.3 mBq l^{-1}. Particularly, the high levels of A-^{226}Ra (>100 mBq l^{-1}) concentrate in well water samples collected from wells 1, 2 and 3. The low levels of A-^{226}Ra (<10 mBq l^{-1}) concentrated in well water samples collected from wells 12, 15, 16, 19, 20 and 24.

According to the simulation results, L_V, L_L, L_D and L_U were 5.82 × 10^{-4} % C_{water}, 9.47 × 10^{-3} % C_{water}, 5.93 × 10^{-5} % C_{water} and 1.15 × 10^{-5} % C_{water}, respectively. The long-term change of A-^{226}Ra in agricultural soil is presented in Fig.4. The results showed that A-^{226}Ra increased continuously under the cultivation process (including water irrigation and fertilizer application). However, it got a equilibrium value (C_{eq}) after the certain period of the agricultural cultivation. The period of time that A-^{226}Ra in agricultural soil reach the equilibrium value (T_{eq}) correlated with A-^{226}Ra in irrigation water (C_{water}) (Fig. 4a) and the ^{226}Ra partition lost by three processes of volatilization, leaching, radioactive decay and plant uptake $L_V + L_L + L_D + L_U$ (Fig.4b). The correlation between T_{eq} and C_{water} is shown in Fig.4c.

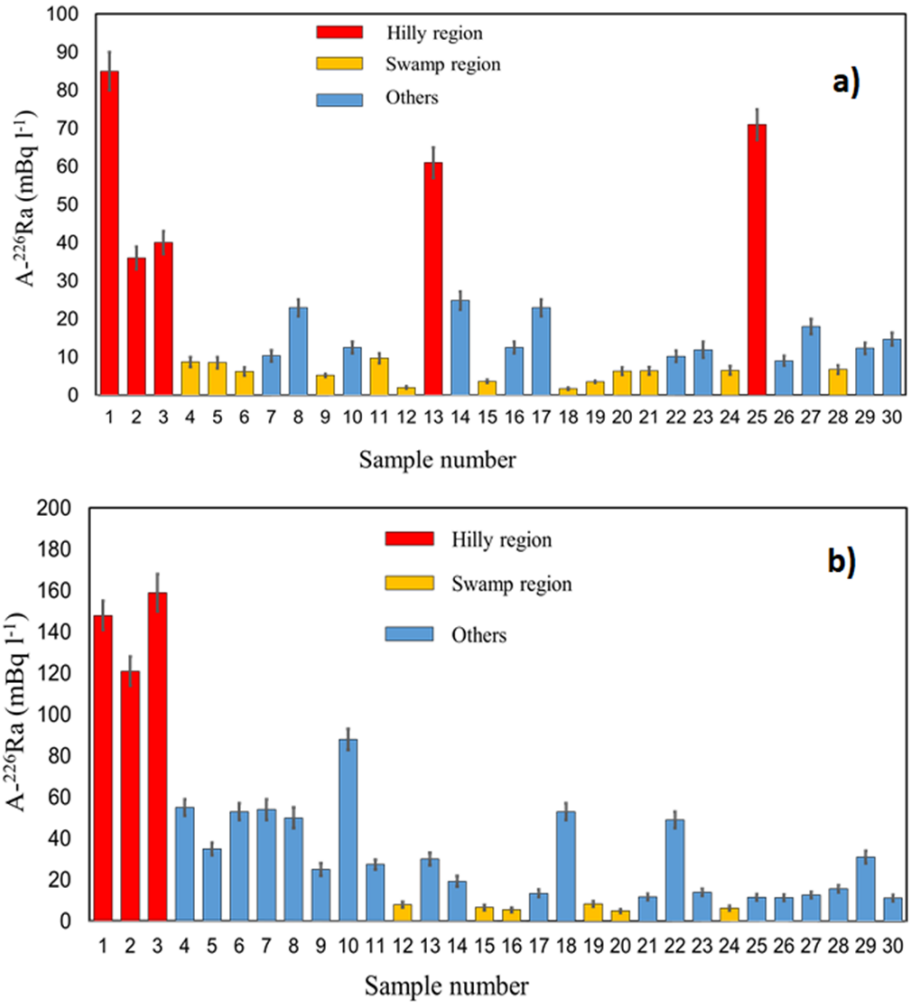

Fig. 3. Comparison between A-^{226}Ra in water samples collected from dug wells (a) and drill wells (b) in the study area

The predictions of T_{eq} and C_{eq} for the study area are shown in Fig.5. Using the well water samples for irrigation increase A-^{226}Ra which are available in the agricultural soils. With the consumption values of C_{soil} and $C_{fertilizer}$ as discussed above, the addition activities of ^{226}Ra in the agricultural soil (0-20 cm) were in the range from 0.77 to 188 Bq m^{-2} with the average value of 112 Bq m^{-2}. The periods of time to get C_{eq} values were in the range from 240 to 770 days with the average value of 710 days. Finally, long-term water irrigation and fertilizer application increase A-^{226}Ra in the agricultural soil from 0.0096% (dug well sample 18) to 2.36% (drill well sample 3) with the average value of 1.4%.

2.4. Discussion

According to the results, minimum, maximum and average values of A-^{226}Ra in grill water samples are higher than them in dug water samples in the study area. The high levels of A-^{226}Ra were found in well water collected in the hilly region since the high contributions of plutonic and basalt rocks. These rocks are from the rapid cooling of basaltic lava exposed at or very near the surface of the earth. Therefore, the activity concentrations of natural radionuclides in these rocks are significantly higher than other types of soil and rock [19, 20]. The low levels A-^{226}Ra were found in well water collected in the swamp region. The reason was explained by the dilution of the surface water. Our average value of A-^{226}Ra in well water (5.44 mBq l^{-1}) is lower than those measured in well water (drill well water, dug well water, bore well water, spring water or ground water) in most of countries such as Karnataka State, India (33.05 mBq l^{-1}) [21], Finland (41 mBq l^{-1}) [22], Iran [23]; Paraná State, Brazil [24] and Yemen (Assalamia, Alhomira and Dempt) [25]. However, our range of A-^{226}Ra nearly equals to the ranges found in Fujian Province, China [26], Syria [27] and Sudan [28]. A-^{226}Ra was various in order drill well water > dug well water > drinking water and tap water in both Phu

Yen province and Ho Chi Minh city [1]. ^{226}Ra concentrates in well water as a result of the leaching of ^{226}Ra and its parent radionuclides (RaThU) from the soil boundary. Therefore, A-^{226}Ra in well water depends on the activity of RaThU in soil and ground water. The increase of RaThU in soil and ground water along with the depth of the soil profile [29], causes A-^{226}Ra higher in drill well water than dig well water. Moreover, construction materials of dig well prevent RaThU escape from the soil boundary.

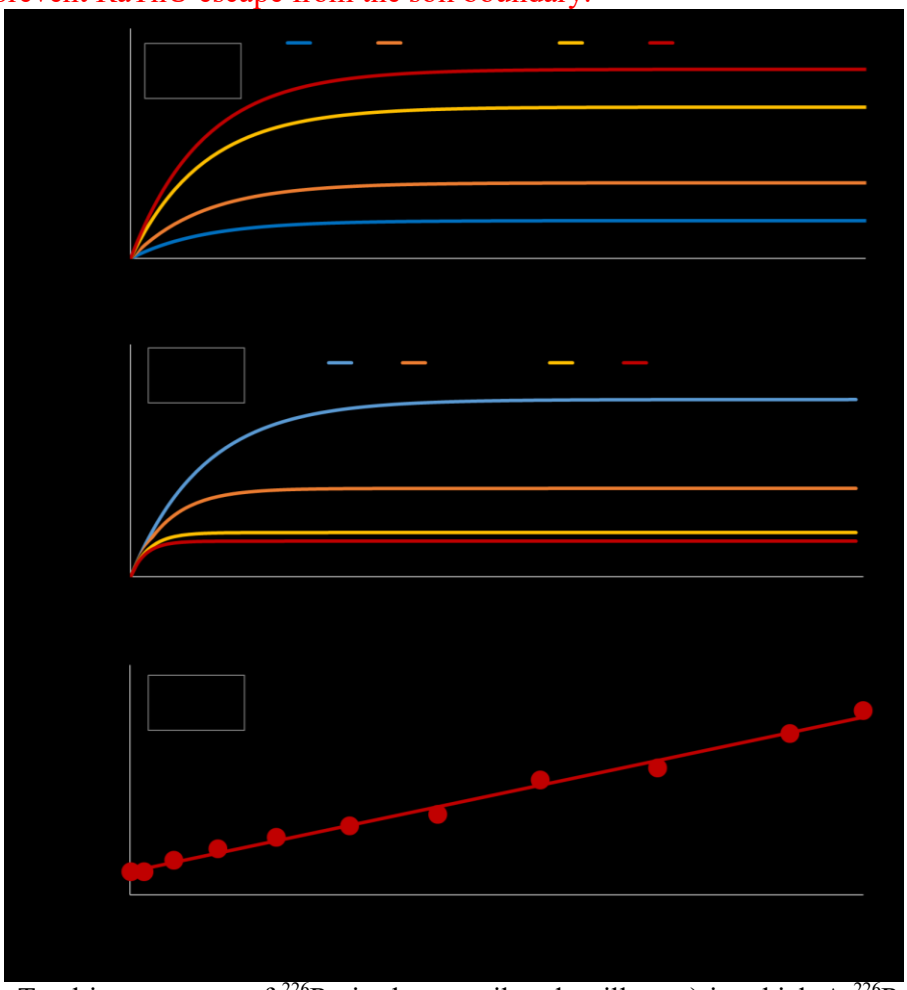

Fig. 4. Total increase rate of ^{226}Ra in the topsoil under tillage a) in which A-^{226}Ra in irrigation water change from 1 to 5 Bq m^{-3}, b) in which the total losing rates of ^{226}Ra activity due to various processes change from 1 to 5%, and correlation between time periods that A-^{226}Ra in agricultural soil reach equilibrium values (T_{eq}) and A-^{226}Ra in irrigation water (C_{water})

For water samples of dug wells, the dose levels are widely varied from 0.34 to 17.3 µSv y^{-1} with the mean value is 0.88 µSv y^{-1}. For water samples of drill wells, the dose values are widely varied from 1.0 to 32.4 µSv y^{-1} with the mean value is 2.64 µSv y^{-1}. For all well water samples, the average value of annual effective dose is 1.11 µSv y^{-1}. There are 8 well water samples (A1, A2, A3, A4, A5, A7, A17, A20, A21, A22, A25, A40, A44 and A51) where the doses exceed the world average dose (8 µSv y^{-1}) due to ^{226}Ra in drinking water. However, all values of annual effective dose are much lower than the average radiation dose from drinking water and ingestion of food (300 µSv y^{-1}) [30, 31]. For water samples of dug wells, the ELCRs are widely varied from 1.2 × 10^{-6} to 6.1 × 10^{-5} with the mean value is 1.3 × 10^{-5}. For water samples of drill wells, the ELCRs are widely varied from 3.5 × 10^{-6} to 1.1 × 10^{-4} with the mean value is 2.7 × 10^{-5}. For all well water samples, the average value of ELCRs is 2.1 × 10^{-5}. All values of ELCRs were found lower than the world's average (0.29 × 10^{-3}) [32].

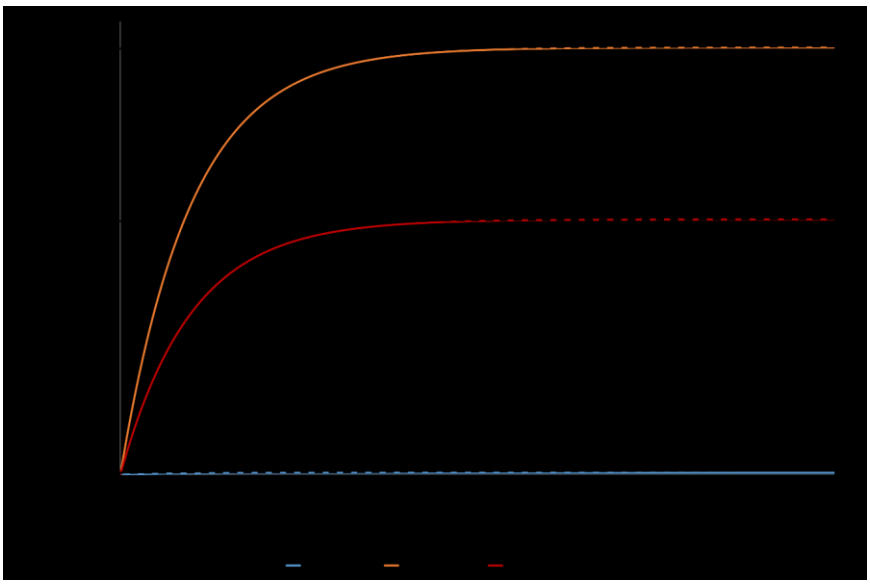

Fig. 5. Prediction of T_{eq} and C_{eq} for minimum, maximum and average values of C_{water}

A-^{226}Ra transferred from well water to vegetables due to irrigation dose not contribute significantly to the total radiological dose. However, the results prove that A-^{226}Ra increases due to the application of well water. The long-term change of A-^{226}Ra in the agricultural soil due to various processes is shown in Fig.4. In Fig.4a, C_{eq} were 98.8, 197.7, 296.5, 395.4, 494.2 and 593.1 Bq m^{-2} with A-^{226}Ra in C_{water} were 1, 2, 3, 4 and 5 mBq L^{-1}, respectively. Besides that, T_{eq} increase with the increase of A-^{226}Ra in irrigation water. In order to study influences of the ^{226}Ra partition lost by three processes of volatilization, leaching, radioactive decay and plant uptake (p) on T_{eq} and C_{eq}, we investigated the total increase rates of ^{226}Ra in agricultural soil with p values from 1 to 5%. The simulation results are shown in Fig. 4b. The increase of p value caused the decrease of T_{eq} and C_{eq}. We found out that C_{eq} were 1010, 760, 370 and 270 Bq m^{-2}, T_{eq} were 190.7, 95.4, 63.6 and 47.7 mBq L^{-1} with the p values were 1, 2, 3, 4 and 5%, respectively. The correlation between T_{eq} and C_{water} is shown in Fig.4c. To find the correlation, the simulations were carried out with the C_{water} from 1 to 500 Bq m^{-3}. The results showed that T_{eq} and C_{eq} were in ranges from 970 to 1110 days and from 110.4 to 357.1 Bq m^{-2}, respectively. The linear correlation (R^2=0.99) was found between T_{eq} and C_{water} in this study.

3. CONCLUSION

We determined A-^{226}Ra in 60 well water samples in An My, Tuy An district Phu Yen province, Vietnam. The health risks to population due to exposure of well water were assessed via annual effective doses and excess lifetime cancer risks. Long-term change of ^{226}Ra in agricultural soils was based on model assessment. Some important conclusions are listed below:

- Levels of ^{226}Ra depended on topographical conditions and types of well.
- Total dose and excess lifetime cancer risk did not exceed the world's average values.
- In agricultural soils, ^{226}Ra activity increased and reached equilibrium value (C_{eq}).
- Time to reach C_{eq} correlated with ^{226}Ra activity in well water.

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NỒNG ĐỘ ^{226}Ra TRONG NƯỚC GIẾNG, CÁC TÁC ĐỘNG LIÊN QUAN ĐẾN SỨC KHỎE NGƯỜI DÂN VÀ SỰ THAY ĐỔI THEO THỜI GIAN TRONG ĐẤT NÔNG NGHIỆP: TRƯỜNG HỢP NGHIÊN CỨU Ở HUYỆN TUY AN, PHÚ YÊN, VIỆT NAM

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Tóm tắt:

Tổng cộng có 60 mẫu nước giếng được thu thập ở khu vực ven biển của huyện Tuy An, Phú Yên được sự dụng để phân tích hoạt độ ^{226}Ra. Hoạt độ ^{226}Ra đo được trong khoảng từ 1,7 đến 159 mBq L^{-1}, với giá trị trung bình là 5,44 mBq L^{-1}. Kết quả cho thấy, hoạt độ ^{226}Ra trong nước giếng phụ thuộc vào địa hình tại các điểm lấy mẫu và loại giếng nước. Hoạt độ ^{226}Ra, liều hiệu dụng và nguy cơ gây ung thư từ các mẫu nước giếng nằm trong các giới han an toàn của thế giới. Sự thay đổi hoạt độ ^{226}Ra trong đất nông nghiệp theo thời gian dài do tưới nước giếng được đánh giá bằng chương trình mô phỏng. Kết quả cho thấy hoạt độ hoạt độ ^{226}Ra tăng liên tục trong lớp đất bề mặt dưới tác động của các quá trình tưới tiêu và bón phân. Tuy nhiên, hoạt độ này đạt giá trị bão hòa sau một thời gian canh tác nhất định. Thời gian này phụ thuộc vào hoạt độ ^{226}Ra trong nước tưới và hoạt độ ^{226}Ra mất do các quá trình bay hơi, khuếch tán, phân rã phóng xạ và hấp thu của cây trồng.

Từ khóa:
Đồng vị ^{226}Ra, nước giếng, mô hình mô phỏng, liều hiệu dụng, hệ sinh thái đất - cây trồng