Assessment of contamination of natural and anthropogenic radionuclides in rice samples collected from Songkhla province, Thailand

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Abstract. Study on assessment of contamination of natural and artificial radionuclides in agricultural products samples are very important to all human being as a consumer. In the present study, we have measured and evaluated the specific activities of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic (^{137}Cs) radionuclide in rice samples. The 30 rice samples were collected from general and department stores at Songkhla province in the south of Thailand. The high-purity germanium (HPGe) detector and gamma-ray spectrometry analysis system which was set-up in advanced laboratory in Thailand Institute of Nuclear Technology (public Organization) or TINT were employed to perform all of measurements and analysis for this study. The frequency distribution of specific activities of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs for this study were also studied and found to be asymmetrical distribution with the skewness of 1.29, 1.43, 2.32 and 0.82, respectively. For this reason, the median values of specific activities of ^{40}K, ^{226}Ra and ^{232}Th which were 620.04 ± 44.30, 3.73 ± 0.54 and 2.44 ± 0.54 Bq/kg respectively, should be selected and also used to calculate some related radiological hazard indices in this study. Furthermore, the excess lifetime cancer risk (ELCR) would be also evaluated and presented. Moreover, the results of present study were taken to compare with some data and studies in Thailand and global measurement and calculations. It was found that the outcomes satisfied the standards of UNSCEAR and IAEA.

1. Introduction

A few decades ago, there are some concerns with radioactivity buildup in food, vegetables and fruits around the world [1-3]. Radioactive materials from soil and fertilizers utilized to the soil for enrichment would be removed to vegetables and plants. Owing to the increase of up to one order of magnitude or more in the concentration of radionuclide levels in food, it is evaluated that the use of phosphate fertilizer has doubled the extended exposure of humans to radioactive materials from ingestion of food [4]. Hence, many food items contain tiny amounts of uranium (U), thorium (Th), potassium (^{40}K), and their progeny products. Anthropogenic radionuclides such as ^{137}Cs can be presented in soil in consequence of nuclear accidents [5-7]. For this reason, there is a high probability for natural (^{226}Ra, ^{232}Th and ^{40}K) and anthropogenic (^{137}Cs) radionuclides entering plants and later into foods consumed by humans. Radionuclides mostly enter the human body by way of ingestion and inhalation. The ingested radionuclides could be accumulated in some parts of the body [8]. Usually, Thai people like to eat rice as a staple food for three meals a day. One of the most well known main dishes among Thais and foreigners is "fried rice". The fired rice or "Kao Pad" is generally consist of rice with pork, beef or chicken meat, vegetables, egg and then stir fire together with some typical spice source. In addition, some other common dishes such as noodles, use rice flour as the main ingredient. In Thailand, not many researches has been carried out so far to assess the amount of the concentration of some concerned radionuclides present in the normal Thai main staple food, which has caused difficulty to decide whether there is a risk for Thai population due to the ingestion of radioactive materials. Hence, it is very importance to create background levels and specific activities of natural and anthropogenic radionuclides in local foods in order to secure the community from future ingestion. However, there are only few researches that have been conducted to assess the concentrations of interested radionuclides of Thai rice. Therefore, the main objectives of this study were to study and evaluate the specific activity of gamma emitting radioactive elements and some related radiological hazard indices and the excess lifetime cancer risk (ELCR) present in rice samples which collected randomly from some local and department stores in Songkhla province, Thailand. Moreover, the study results should be used for discovering the effects caused by the consumption on the Thai community.

2. Materials and methods

2.1. Sample collection and processing

All of 30 rice samples which are Jasmine rice (16 samples), Hom Prathum rice (8 samples) and white rice (6 samples) were collected randomly from some local and department stores in Maueng district in Songkhla province, Thailand. Basically, we do not know which provinces and regions of Thailand have been cultivated for all rice samples. Each sample was collected, treated and prepared by following the International Atomic Energy Agency (IAEA) sampling and preparation of vegetables samples [9]. For example, the rice samples would be grinded with a blender machine and then spread on plastic or aluminium trays and let to dry at room temperature for a few days. Then, all rice samples would be sieved through a 2 mm mesh-sized sieve to remove some coarse portion. A slow-airflow, temperature (60°C) for 5 hours in drying closet will accelerate the drying process without loss of radionuclides from the rice samples. Each sample (about 154 cm^3 which approximately 139-158 gms) was placed in a PVC cylindrical container of diameter 7.0 cm and height 7.0 cm. The containers were sealed tightly with a thick cellophane tape around their necks to avoid any gas escape from them, and kept for a minimum period of 4 weeks to ensure equilibrium between ^{222}Rn and its progeny products.

2.2. Gamma-ray spectrometry with high-purity germanium (HPGe) detector

All rice samples were taken to measure and analyze by using a gamma spectrometry analysis system with a measuring time 10,800 s. The HPGe detector (EG&G ORTEC Model GEM 20 P4) and gamma spectrometry analysis system at advanced laboratory, Thailand Institute of Nuclear Technology (Public Organization) (TINT) were employed with sufficient lead shielding which reduced the background by a factor about 95 %. The specific activities of various radionuclides of interest in 30 rice samples were determined in Bq/kg using the counted gamma-ray spectra. The gamma-ray photopeaks corresponding to 351.9 keV (^{214}Pb), 583.2 keV (^{208}Tl), 1460.8 keV (^{40}K) and 661.7 keV (^{137}Cs) were evaluated in achieving of the specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in the rice samples, respectively. The IAEA-473 Milk Powder reference materials (International Atomic Energy Agency (IAEA), Vienna, Austria) was used to determine the geometric efficiency for rice sample matrices in the container.

2.3. Frequency distribution of specific activities

The frequency distribution of specific activities of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic (^{137}Cs) radionuclides in 30 rice samples for the present study were studied and analyzed by using the SPSS computer program. The results would be presented and shown in the subsection 3.1.

2.4. Calculation of related radiological hazard indices and excess lifetime cancer risk

Three related radiological hazard indices which are annual radionuclide intake (D_a) in Bq/yr, annual effective dose (D_{eff}) in Sv/yr**,** total annual effective dose (D^{total}) in Sv/yr and excess lifetime cancer risk (ELCR) [10] were evaluated and presented for the studied area by using the appropriate medium values of specific activities of ^{40}K, ^{226}Ra and ^{232}Th which were analyzed, determined and chosen from section 3.1. In addition, the annual consumption of rice for Thai population in 2020 (83 kg/yr) [11], the dose conversion factor for ^{40}K, ^{226}Ra and ^{232}Th as shown in ICRP 1996 [12], the lifetime span of Thai population in year 2019 (75 years) [13] and the mortality risk coefficient for ^{40}K, ^{226}Ra and ^{232}Th [14] were also used to evaluate all results in this study. Moreover, the comparison between the results with some research data in Thailand and global recommended values were shown in subsection 3.2

3. Results and discussions

3.1. Frequency distribution of specific activities and statistic values

The frequency distribution of specific activities of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs in 30 rice samples collected from Songkhla province were studied, analyzed and presented in the following figures 1– 4 by using the SPSS computer program. In addition, all statistic values which were calculated from this study, were also presented in table 1.

3.2. Radiological hazard indices values and comparison

From section 3.1, it was found that the frequency distribution of specific activities of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs in 30 rice samples collected from the studied area, were asymmetrical distribution with the skewness of 1.29, 1.43, 2.32 and 0.82, respectively. For this reason, the median values of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs which were 620.04 ± 44.30, 3.73 ± 0.54, 2.44 ± 0.54, and 1.94 ± 0.37 Bq/kg for this present study, were the appropriate medium value and should be chosen for calculation four radiological hazard indices and the ELCR value in this study. Moreover, some researches data in Thailand, foreign countries, Office of Atoms for Peace (OAP) annual report data and the recommended values informed by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and IAEA were chosen to compare with the studied results and their average values as presented in table 2.

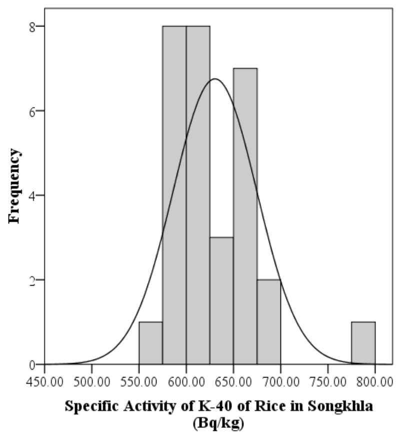

Figure 1. Frequency distribution of Specific activities of ^{40}K in 30 rice samples of Songkhla province

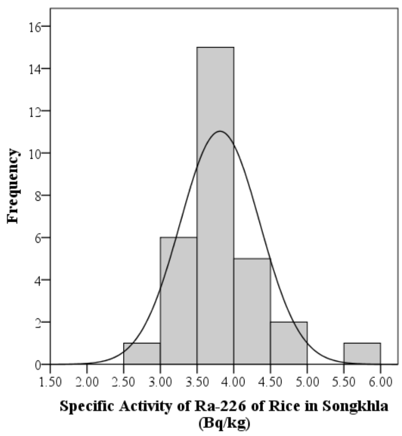

Figure 2. Frequency distribution of Specific activities of ^{226}Ra in 30 rice samples of Songkhla province

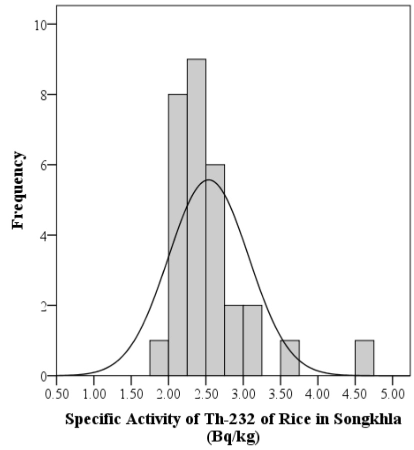

Figure 3. Frequency distribution of Specific activities of ^{232}Th in 30 rice samples of Songkhla province

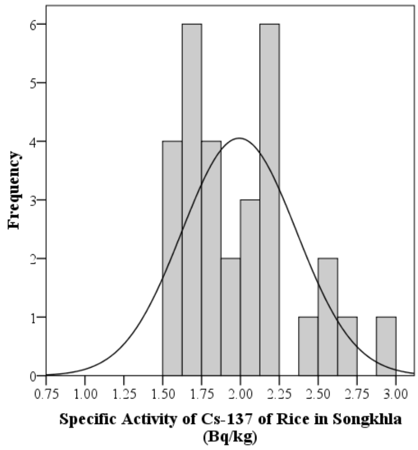

Figure 4. Frequency distribution of Specific activities of ^{137}Cs in 30 rice samples of Songkhla province

Table 1. Statistic values of the frequency distribution of specific activities of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs in 30 rice samples collected from Songkhla province, Thailand.

Statistic values	Analyzed values			
	^{40}K	^{226}Ra	^{232}Th	^{137}Cs
Mean (Bq/kg)	630.34	3.81	2.54	1.99
Median (Bq/kg)	**620.04**	**3.73**	**2.44**	**1.94**
Mode (Bq/kg)	568.95	3.36	2.09	1.55
Std deviation	44.30	0.54	0.54	0.37
Skewness	1.29	1.43	2.32	0.82
Kurtosis	2.75	3.83	7.01	-0.02
Minimum value (Bq/kg)	568.95	2.90	1.97	1.55
Maximum value (Bq/kg)	777.62	5.69	4.60	2.91

Table 2. Related radiological hazard indices and ELCR values for the present investigation and compare to some research studies in Thailand, foreign counties, UNSCEAR and IAEA.

Literatures	D_a (kBq/yr)	D_{eff} (Sv/yr) × 10^{-5}			D^{total} (Sv/yr) × 10^{-5}	ELCR × 10^{-3}
		D_{eff} (^{40}K)	D_{eff}(^{226}Ra)	D_{eff}(^{232}Th)		
Rice samples in Nakhon Nayok [15]	355.70 ± 3.44	218.40 ± 2.13	66.37	24.72	309.49 ± 2.13	17.46 ± 0.15
Sungyod rice in Phatthalung [16]	97.52 ± 14.19	59.81 ± 8.59	18.41 ± 5.60	8.95 ± 3.17	87.17 ± 17.36	4.80 ± 0.78
Jasmin rice in Songkhla [17]	37.82 ± 12.87	20.38 ± 6.60	13.11 ± 4.51	102.99 ± 47.53	136.48 ± 58.64	2.61 ± 0.97
Hom Pratum rice in Songkhla [17]	22.68 ± 13.17	9.48 ± 6.00	13.27 ± 6.04	158.89 ± 75.23	181.64 ± 87.28	2.28 ± 1.18
Organic rice in Phatthalung [18]	50.53 ± 2.69	30.81 ± 1.61	14.87 ± 2.63	7.03 ± 0.00	52.71 ± 4.23	2.63 ± 0.18
Rice in Penang Malasia [19]	6.19 ± 1.73	3.71 ± 1.03	3.30 ± 1.28	1.97 ± 0.59	8.98 ± 2.90	0.36 ± 0.11
Canned rice in Iraq [20]	7.18 ± 3.28	4.00 ± 1.98	6.48 ± 0.95	11.56 ± 1.11	22.04 ± 4.04	0.54 ± 0.17
Amber rice samples in Iraq [21]	2.11	0.87	13.80	5.06	19.73	0.46
Rice samples in Egypt [10]	3.10	1.85	1.39	1.47	4.72	0.18
Rice samples in Saudi Arabia [22]	11.50	7.10	0.93	0.38	8.41	0.53
Rice samples in Italy [23]	10.30	6.14	6.74	5.35	18.22	0.65
Rice samples in India [24]	13.13	6.22	7.13	65.48	78.83	1.15
Rice samples in Ghana [25]	9.41	5.37	10.97	8.27	24.61	0.73
Rice in Songkhla*	**51.98 ± 3.77**	**31.91 ± 2.28**	**8.67 ± 1.25**	**4.66 ± 1.03**	**45.23 ± 4.57**	**2.53 ± 0.20**
UNSCEAR [26-28]	32.29	15.95	155.71	22.91	194.57	5.31
IAEA [29]	-	-	-	-	100	-

*Present study

From table 2, the D_a values of the present study was 51.98 ± 3.77 kBq/yr which was lower than the research data in rice samples from Nahkon Nayok and Sungyod rice from Phatthalung provinces but higher than in Jasmine and Hom Pratum rice samples from Songkhla and Organic rice from Phatthalung provinces, all of international research data and the recommended value (32.29 kBq/yr) as reported by UNSCEAR. The D^{total} value $(45.23 \pm 4.57) \times 10^{-5}$ Sv/yr) from this study which is comprised of D_{eff} (^{40}K), D_{eff}(^{226}Ra) and D_{eff}(^{232}Th), was lower than 194.57×10^{-5} Sv/yr which is reported by UNSCEAR, all of research data in Thailand, but higher than all research data of foreign countries except in India. Furthermore, the D^{total} value was approximately 0.55 times lower than the IAEA dose constraints for public exposure in planned exposure situations and reference level for public exposure in specific existing exposure situations, e.g. exposure due to radionuclides in commodities such as food, drinking water or construction materials (< 1 mSv/yr or 100×10^{-5} Sv/yr). In addition, the ELCR value was also calculated and equal to $(2.53 \pm 0.20) \times 10^{-3}$ which lower than all research data in the Thailand, India and UNSCEAR recommended values but higher than all of international research data. Moreover, we can see that the specific activity of ^{40}K in this investigation which was equal to 620.04 ± 44.30 Bq/kg, should be the main factor of the high value of evaluated related radiological hazard indices and ELCR value for this present study. These high value of the concentration of ^{40}K might be caused from the regular using fertilizer of some group farmers in Thailand with is corresponded to the research in Sri lanka [30]. The results from this study should be repeated and increase more rice samples in different region of Thailand kingdom. Consequently, the concentration level of natural and anthropogenic radionuclides in rice samples for every kind of rice which are produced in Thailand should be randomly measured and monitored in every year of production. Moreover, the results of this study could be considered and used to be the baseline reference of background radiations for the daily food of Thai people and for green food and environment in the future.

4. Conclusions

The evaluated median values of specific activity of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic (^{137}Cs) radionuclides in 30 rice samples collected randomly from Songkhla province were 620.04 ± 44.30, 3.73 ± 0.54, 2.44 ± 0.54 and 1.94 ± 0.37 Bq/kg, respectively. The median values of specific activity of ^{40}K, ^{226}Ra and ^{232}Th were chosen to assess three related radiological hazard indices (D_a, D_{eff}, and D^{total}) and the ELCR value. The results obtained in this study fall within the range of values reported in similar studies conducted nationwide and worldwide. The D_a value and the D_{eff} (^{40}K) was higher than the recommended values 32.29 kBq/yr and 15.95×10^{-5} Sv/yr as reported by UNSCEAR respectively. The D_{eff} (^{226}Ra) and D_{eff} (^{232}Th) values was lower than the recommended values 155.71 and 22.91×10^{-5} Sv/yr as presented by UNSCEAR. Furthermore, the D^{total} value which evaluated from all three of D_{eff} values, was lower than recommended values 194.57×10^{-5} Sv/yr as announced by UNSCEAR and lower than the IAEA dose constraints and reference level for public exposure (< 1 mSv/yr). In addition, the ELCR values was found below the recommended values 5.31×10^{-3} as announced by UNSCEAR. It can be seen that Thai people and consumers of Thai rice will receive the effective annual dose due to some radioactive substances present in Thai rice within the safe range. However, the results of this study show directly the effect of the specific activity of ^{40}K to the ELCR value. The specific activities of ^{40}K might be accumulated from the original paddy soil, water and fertilizers used by groups of rice farmers then transfer to the stem, leave and seed of rice plant. According to the present study results, we should concern to monitor, measure and assess the concentration of natural radionuclides especially ^{40}K in all kinds of rice which usually are the daily food for Thai people in every harvesting year. Hence, this data may contribute a general background level for the rice plant and may also support as a guideline for future measurement and assessment of possible radiological risks to Thai people health. Therefore, we should to study, measure and assess the specific activities of natural and anthropogenic radionuclides in all kinds of rice samples collected from cultivated area around the Thailand Kingdom and also study the transfer factor of those radionuclides from their paddy soil to a rice plant in the next studies and evaluation.

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