Preliminary survey of radioactivity level in Thai medicinal herb plants

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Abstract. In this research, the natural radioactivity concentrations and their respective annual effective dose of the naturally occurring radionuclides $^{226}\text{Ra}$, $^{228}\text{Ra}$ and $^{40}\text{K}$ in selected medicinal herb plants were investigated. Seven kinds of popular Thai medicinal herb plants had been studied: turmeric, ginger, safflower, moringa, gotu kola, garlic and alexandria senna. The radiological risk associated with the use of these medicinal plants was assessed. The activity concentrations of $^{226}\text{Ra}$, $^{228}\text{Ra}$ and $^{40}\text{K}$ were determined using the gamma-ray spectrometry technique. The radioactivity concentrations were found to range from less than 0.20 to 6.67 Bqkg$^{-1}$ for $^{226}\text{Ra}$, less than 0.10 to 9.69 Bqkg$^{-1}$ for $^{228}\text{Ra}$, and from 159.42 to 1216.25 Bqkg$^{-1}$ for $^{40}\text{K}$. Gotu kola showed the highest activity concentrations of $^{226}\text{Ra}$ and $^{228}\text{Ra}$, while ginger showed the highest activity concentration of $^{40}\text{K}$. The total annual effective dose due to ingestion of these herb plants were found to range from 0.0028 to 0.0097 mSvy$^{-1}$ with an average value of 0.0060±0.0001 mSvy$^{-1}$. The results conclude that the Thai medicinal herb plants samples from this research are considered safe in terms of the radiological hazard.

1. Introduction

Thailand has practiced traditional medicine for more than a thousand years with the known record that date back to the Sukhothai period (1238-1377). The knowledge of traditional medicine based on the usage of Thai medicinal herbs has been gradually developed, systematized, revised, recorded, and passed on from generation to generation throughout the country’s history.\[1\] Thai medicinal herbs, known as Sa-mun-phrai in the local language, have been used to treat various symptoms and ailments. After the World Health Organization began to promote national traditional heritage in 1977, the Foundation for the Promotion of Thai Traditional Medicine was established, and over the years the interest and importance of herbal healing has continued to grow.\[2\]

Raw parts of plants and their extracts are used in medicinal products all over the world. It is estimated that 25% of modern medicines are derived from medicinal plants, of which most are flowering plants. About 80% of the world population (especially in developing countries) use herbal medicine plants as their primary source of healthcare \[3\]. There are over 250,000 flowering plant
species that serve as resources for the production of new drugs. Plant research has increased globally and the large body of collected evidence shows the immense potential of medicinal plants used in various traditional systems [4].

Various environmental conditions can affect the medical properties and effectiveness of medicinal plants, and may also have a direct influence on their growth in the place of their occurrence and cultivation [5]. An emerging problem in many developing countries is the industrial pollution that threatens the health of the local environment and can lead to the contamination of vegetation by heavy metals, pesticides, and radioactivity [6]. The contamination level in the medicinal plants should therefore be monitored and analysed.

According to the International Food Safety Authorities Network [7], plants used as food commonly have $^{40}$K, $^{232}$Th and $^{238}$U and their progenies. It is expected that similarities would be found in plants used for medicinal purposes since plants are the primary pathway of natural radionuclides to enter the human body via the food chain. In a variety of concentrations, the Naturally Occurring Radioactive Materials (NORMs) have always been present in every part of the earth and in the tissue of all living beings. Natural radionuclides such as $^{238}$U, $^{232}$Th and $^{40}$K are found in every constituent of the environment (air, water, soil, and food) and in humans, and thereby subjecting human beings to certain degree of radiation exposure. [8] The estimation of risk to humans from medicinal plants through ingestion requires a quantitative understanding of the interrelated pathways by which the radionuclides are eventually ingested by humans [9]. The assessment of radioactive contamination of medicinal plants not only contributes to determination of the quality of the plant material, but also provides useful information on the safety of their consumption by humans.

2. Methods and Materials

Samples of medical herb plants were collected from various local markets and drugstores in Thailand. All samples had been certified and licensed by the Thai Food and Drug Administration under the Ministry of Public Health. They included 7 kinds of popular Thai medicinal herb plants (24 samples) used extensively for treating various diseases or complementary medicine. Table 1 lists the medicinal plants selected for the present study, as well as their common names, scientific names, medical uses, and the particular part of the plant used. Each sample had been dried, blended and packaged inside a capsule. All samples have patented license and been certified by the Food and Drug Administration, Ministry of Public Health, Thailand.

2.1. Sample preparation

The Thai medicinal herb plant samples, which were already in powder form, were removed from inside of their plastic capsules. They were dried at a temperature of 80 °C in an oven for 6 hours. The dry mass value was used to determine the radionuclide concentration. Each dried sample, about 100 g, was put in a cylindrical plastic container (2.5 inch in height and 3 inch in diameter). All samples were sealed and kept for at least 30 days in order to ensure the radioactive secular equilibrium before taken for measurement by gamma spectrometric method [10].

2.2. Measurement of the samples

The gamma-ray spectroscopy system consisted of a high purity germanium (HPGe) detector with a relative efficiency of 60% and 1.85 keV energy resolution (FWHM) at the 1332 keV energy peak (of $^{60}$Co isotope); a multichannel analyzer (MCA) with 4096 channel (ORTEC’s system); and a detector shield with adequate size of cavity to accommodate large sample measurement. The shield was made of lead with 10 cm thickness, and lined inside with graded absorber made of Cd (~1.6 mm.) and Cu (~0.4 mm.).

2.3. Quality assurance and Quality control

Quality assured standard materials procured from IAEA were used for the calibration of the detector. In order to have the density of the reference material similar to that of the dried sample powder, the
reference material power, IAEA-RGU-1 and IAEA-RGTh-1, were diluted and mixed with the laboratory grade boric acid powder with the ratio of 1:3. The weight of diluted reference standard powder used for the detector calibration is about 100 g similar to the weight of the herb sample. All of standards and samples were filled in the cylindrical plastic container, which has an identical size of 2.5 inch in height and 3 inch in diameter, and the container cap was sealed with tape and silicone. Thus, the geometrical factor of the standard will be the same as the sample when they were measured.

Based on the measured gamma-ray peaks emitted by the daughter radionuclides in the $^{232}$Th and $^{238}$U decay series, and the peak of $^{40}$K, the concentrations of $^{226}$Ra, $^{228}$Ra, and $^{40}$K were determined. The calculations relied on the establishment of secular equilibrium in the samples, due to the much smaller lifetime of the daughter radionuclides in the decay series of $^{232}$Th and $^{238}$U. The gamma-rays of $^{228}$Ac (338.4 and 911 keV) were used to determine the $^{228}$Ra concentration. The gamma-rays of $^{214}$Pb (351.3 keV) and $^{214}$Bi (609.3 keV) were used to determine the $^{226}$Ra. The 1461 keV was used to determine the concentration of $^{40}$K in different samples. Each sample was counted for 172,800 sec. (48 hours) in the same geometry setting.

QA/QC included efficiency checks performed weekly and monthly background counts. In addition, comparison measurements were carried out to find the reliability of the measurements. Some of the samples were sent to the Radiation Measurement Group, Bureau of Technical Support for Safety Regulations, Office of Atoms for Peace, Thailand for the analysis of radioactivity by gamma spectrometry. The results of these comparison measurements showed good agreement. The deviation in the results of two laboratories was within 10%. Our laboratory has also participated in the inter-comparison exercises and proficiency testing for gamma spectrometry organized by the IAEA since 2012. The results from these inter-comparisons demonstrated satisfactory performance with respect to the quality of the analytical results generated by the laboratory.

2.4. Calculation of radionuclides and annual committed effective dose

Following the spectrum analysis, counting rates for each detected photo peak and activity per mass unit for each of the detected nuclides are calculated. The specific activity (in Bq/kg) is given by the expression (1).

$$A_{sp} = \left( \frac{N}{t} - \frac{N_b}{t_0} \right) / \left( I_p \cdot \varepsilon \cdot m \right)$$

(1)

Where, $N$ is the net counts of a given peak, $t = 48$ hour is the counting time for the sample, $N$ is the background of the given peak, $t_0 = 48$ hours is the counting time for the background, $\varepsilon$ is the detection efficiency, $I_p$ is the number of gamma photons per disintegration and $m$ is the mass in kg of the measured sample.

If there is more than one peak in the energy analysis range for a nuclide, an average of the peak activities is made, and the result is then the weighted average nuclide activity. The total uncertainty value is composed of the random and systematic errors in all the factors involved in producing the final nuclide concentration result listed in Table 2.

Having obtained the values for the specific activity concentrations of the individual naturally occurring radionuclides in the medicinal plants, the average annual committed effective dose, $E_{ave}$, for ingestion of naturally occurring radioactive materials (NORMs) in the Thai medicinal herb plants were calculated using the expression (2) given by [11].

$$E_{ave} = I_p \times DCF_{ing} \times A_{sp}$$

(2)

Where $DCF_{ing}$ is the dose conversion factor for ingestion, for each radionuclide (i.e., $2.8 \times 10^{-4}$ mSvBq$^{-1}$, $6.7 \times 10^{-4}$ mSvBq$^{-1}$, and $6.2 \times 10^{-6}$ mSvBq$^{-1}$ for $^{226}$Ra, $^{228}$Ra, and $^{40}$K, respectively, for an adult) [12,13], $I_p$ is the consumption rate from intake of NORMs in medicinal plants, $A_{sp}$ is the activity concentration in the plant sample.
Table 1. Scientific name and uses of selected traditional medicinal plants.

| Scientific name     | Common name | Medicinal uses                                                                                       | Parts used |
|---------------------|-------------|-------------------------------------------------------------------------------------------------------|------------|
| 1. Curcuma longa    | Turmeric    | Laxative, tonic, alterative, antipyretic, antibacterial, detergent (an agent that cleanses boils, ulcers, stops bleeding, wounds, etc.), ophthalmicum (remedy for diseases of the eye), antiperiodic (prevents the periodic recurrence of attacks of a disease; as in malaria), aperient (mild or gentle laxative), diuretic, antiseptic, and deobstruent (removes obstructions by opening the natural passages or pores of the body). | Roots      |
| 2. Zingiber officinale Roscoe | Ginger      | Antispasmodic, antiemetic, analgesic, anti-septic, appetizer, aromatic, carminative, condiment, diaphoretic, expectorant, febrifuge, pungent, sialagogue, stimulant Topically: increases blood flow to an area | Roots and rhizomes |
| 3. Carthamus tinctorius L. | Safflower    | Diaphoretic, diuretic, emmenagogue, analgesic, carminative | Flowers |
| 4. Moringa oleifera Lam. | Moringa      | anti-diabetic effects, cardio tonic | Seed |
| 5. Centella asiatica (L.) Urb. | Gotu kola    | Alternative, antipyretic, diuretic, febrifuge, antispasmodic, nervine, sedative, tonic | Whole plant |
| 6. Allium sativum L. | Garlic       | Alternative, anthelmintic, antispasmodic, carminative, cholagogue, digestive, expectorant, febrifuge, antibiotic, antiseptic, stimulant | Bulb |
| 7. Senna alexandrina Mill. | Alexandria Senna | A laxative substance, anthelmintic | Leaves |

3. Results and Discussions
Table 2 shows the radioactivity concentrations (Bq kg\(^{-1}\)) of the natural radionuclides \(^{226}\)Ra, \(^{228}\)Ra and \(^{40}\)K in the medicinal plants selected for this study. All natural radionuclides were determined in 7 kinds of Thai medicinal herb plants in a total of 24 samples. The data showed that all Thai medicinal herb plants had activity concentrations in the ranged of less than 0.20 to 6.67 Bq kg\(^{-1}\) for \(^{226}\)Ra, less than 0.10 to 9.69 Bq kg\(^{-1}\) for \(^{228}\)Ra, and from 159.42 to 1216.25 Bq kg\(^{-1}\) for \(^{40}\)K. Gotu kola had the highest activity concentration of \(^{226}\)Ra and \(^{228}\)Ra, while ginger had the highest activity concentration of \(^{40}\)K.

The variations in the activity concentrations could be due to differences in the geological location of the plants and the radiochemical composition of the soils in which these medicinal plants were grown or cultivated. The levels of activity concentration of natural radionuclides are not normalized across the globe, and each plant has ability to absorb particular elements more than the others [11].

In this paper, the assumption that a unit consumption rate (\(I_p\)) of 1 kg per annum was used. The number is derived from the medical labels which received the certificate and license from The Thai Food and Drug Administration of the Ministry of Public Health. The average total annual committed effective doses due to ingestion of \(^{226}\)Ra, \(^{228}\)Ra, and \(^{40}\)K in the medicinal herb plants were calculated using the corresponding values of activity concentrations for each of the medicinal plants. The results are presented in Table 2 and Figure 1. The results varied from 0.0028 to 0.0097 mSv y\(^{-1}\) with an average value of 0.0060±0.0001 mSv y\(^{-1}\). The highest average was recorded for Turmeric, while Safflower and Alexandria Senna showed the lowest average.

From the viewpoint of the annual effective dose and the activity concentration of \(^{226}\)Ra, \(^{228}\)Ra, and \(^{40}\)K in the medicinal herb plants compared with this study (the conservative comparison), Table 2
showed the results from published work with the annual committed effective dose and the activity concentration of NORMs in the different medicinal herb plants conducted in India and Nigeria [14,15]. The average activity concentrations of both $^{226}$Ra and $^{228}$Ra in our study appear to be lower than the values obtained in South India [14] and Nigeria [15]. However, our study showed lower average activity concentration of $^{40}$K than the South India study, but higher average concentrations than the Nigeria study. Thus, the average annual committed effective dose due to the ingestion of $^{226}$Ra, $^{228}$Ra, and $^{40}$K in Thai medicinal herb plants in this study was slightly higher than the results reported in Nigeria [15] but much lower than that reported in South India [14].

### Table 2. Radioactivity concentration of $^{226}$Ra, $^{228}$Ra, and $^{40}$K (in Bq kg$^{-1}$) and annual committed effective dose (mSv y$^{-1}$) in the Thai medicinal herb plant samples (dry weight)

| Medicinal plants (number of Sample) | Average of Activity concentration (in Bq kg$^{-1}$) | Average Annual effective dose (mSv y$^{-1}$) |
|-------------------------------------|-----------------------------------------------|---------------------------------------------|
|                                     | $^{226}$Ra | $^{228}$Ra | $^{40}$K |                                     |
| Present study                       |          |          |        |                                      |
| 1. Turmeric (8)                     | 1.94±0.62 | 3.90±0.36 | 749.87±8.79 | 0.0097±0.0004 |
| 2. Ginger (3)                      | 2.98±0.56 | 4.31±0.50 | 669.35±12.03 | 0.0092±0.0003 |
| 3. Safflower (2)                    | 2.4±1.10  | 2.74±0.43 | 115.74±3.55 | 0.0028±0.0004 |
| 4. Moringa (3)                      | 1.01±0.45 | 3.13±0.33 | 368.40±5.82 | 0.0046±0.0003 |
| 5. Gotu kola (3)                    | 3.86±0.21 | 8.38±0.76 | 396.53±5.37 | 0.0086±0.0004 |
| 6. Garlic (2)                       | 0.51±0.24 | 0.13±0.08 | 370.43±6.31 | 0.0042±0.0005 |
| 7. Alexandria Senna (3)             | 1.03±0.11 | 1.09±0.27 | 271.95±7.46 | 0.0028±0.0003 |
| **Average**                         | **1.97±0.34** | **3.38±1.19** | **420.33±3.77** | **0.0060±0.0001** |
| South India study [14]              |          |          |        |                                      |
| 1. Justica adhatoda                 | 11.27±1.59 | 5.07±2.15 | 6831.40±490.28 | 0.1067±0.0136 |
| 2. Ficus racemosa                   | 4.51±0.23  | 8.72±0.44 | 832.95±59.69  | 0.0084±0.0005 |
| 3. Eupatorium odoratum              | 11.15±0.66 | 4.87±0.79 | 1305.00±94.74 | 0.0436±0.0041 |
| 4. Ziziphus rugosa                  | 5.98±2.56  | 5.10±0.31 | 143.99±10.83  | 0.0091±0.0016 |
| 5. Citrus limon                     | 6.08±0.73  | 5.67±1.00  | 1910.00±139.09 | 0.0148±0.0012 |
| 6. Bauhinia acuminate               | 2.66±0.21  | 2.42±0.36  | 3724.00±27.44 | 0.0580±0.0032 |
| 7. Mimosa pudica                    | 5.35±0.34  | 4.14±0.44  | 320.86±2.40   | 0.0235±0.0019 |
| 8. Mesua ferrea                     | 3.73±0.17  | 4.43±0.26  | 93.79±7.19    | 0.0075±0.0007 |
| **Average**                         | **6.34±0.85** | **5.05±0.63** | **1895.25±163.33** | **0.0340±0.0043** |
| Nigeria study [15]                  |          |          |        |                                      |
| 1. Cashew nut tree                  | 40.08±4.12 | 38.69±0.71 | 74.59±2.19    | 0.0069±0.0004 |
| 2. Neem tree                        | 19.22±2.12 | 31.69±0.66 | 324.18±8.69   | 0.0049±0.0002 |
| 3. African copaiba                  | 30.69±2.56 | 30.47±0.24 | 123.34±6.22   | 0.0055±0.0003 |
| 4. Moringa                          | 13.71±1.96 | 39.48±0.90 | 184.59±3.43   | 0.0047±0.0003 |
| 5. Guava                            | 10.79±4.24 | 36.53±1.10 | 219.98±9.28   | 0.0043±0.0005 |
| 6. Umbrella tree                    | 18.18±3.52 | 41.05±0.92 | 145.59±7.19   | 0.0051±0.0003 |
| 7. Shea butter tree                 | 42.47±3.76 | 27.76±1.02 | 129.78±5.63   | 0.0064±0.0004 |
| **Average**                         | **25.02±0.95** | **35.10±0.29** | **171.72±2.60** | **0.0054±0.0001** |

### 4. Conclusion
The average annual effective dose determined in this study due to the ingestion of natural radionuclides in Thai medicinal herb plants is far below the average annual effective dose of 0.3 mSv y$^{-1}$ received per person worldwide due to the internal exposures other than radon and thoron that is reported UNSCEAR 2000 report [13]. The study has found insignificant annual committed effective dose due to the use of these Thai medicinal herb plants into Plant Medicine, therefore the radiological hazard associated with intake of the natural radionuclides in the medicinal plants is trivial.
Figure 1. The variation of the annual committed effective dose from natural radionuclides $^{226}$Ra, $^{228}$Ra, and $^{40}$K) in the Thai medicinal herb plant samples.

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