Specific activity of natural and anthropogenic radionuclides and radiological hazard assessment in surface soil samples collected along the Andaman sea coast in southern region of Thailand

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Abstract. The specific activities of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic radionuclides (^{137}Cs) in 314 surface soil samples collected from three provinces (Phuket, Phang-Nga and Krabi) along the Andaman sea coast in southern region of Thailand were studied and evaluated. Experimental results were obtained by using a high-purity germanium (HPGe) detector and gamma spectrometry analysis system. It was found that the mean values of specific activities of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs were 2859.63 ± 209.83, 157.10 ± 8.06, 137.16 ± 7.26 and 4.88 ± 2.34 Bq/kg, respectively. Furthermore, four radiological hazard indices which are absorbed dose rate in air (D), radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}) and annual effective dose rate (AED_{out}) in the area under consideration were also calculated and came out to be 277.11 ± 16.96 nGy/h, 575.16 ± 34.67 Bq/kg, 1.55 ± 0.09 and 0.34 ± 0.02 mSv/y, respectively. The experimental results were also compared and found to be comparable with national and global radioactivity measurements and evaluations. Moreover, the radioactive contour maps of the investigated area were also created and presented in this paper.

1. Introduction

Naturally occurring radioactive material (NORM), which potentially includes all radioactive elements found in the environment. The acronym TENORM, or technologically enhanced NORM, is often used to refer to those materials which the amount of radioactivity has actually increased or concentrated as a result of industrial processes. The issues related to NORM and TENORM attracted a lot of public interest and mass media in the late 1970s. Long-lived radioactive elements such as uranium, thorium and potassium and any of their decay products, such as radium and radon are examples of NORM [1-3]. These elements are always present in the Earth's crust and atmosphere. Many scientists and researchers around the world are studying and measuring the level of NORM concentration in soil and beach sand samples in some countries such as in Brazil, India, China, Turkey, France and Thailand [4-11]. Along the Andaman sea coast in the southern region of Thailand, the most common activities

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related to TENORM are industrial related to tin mining and its subsequent processing. For this reason, some areas especially in Phuket, Phang-Nga and Krabi provinces could be affected by an increase in naturally occurring radioactive materials [12].

Caesium-137 (^{137}Cs) is a radioactive alkali metal created and globally distributed by the atmospheric testing of atomic weapons during the period 1945-1963. This fission radioactive material was deposited on the earth's surface as fallouts associated with precipitation. The other prime source of ^{137}Cs has been accidental releases by accidents of nuclear power plant (e.g., Three Mile Island in 1979, Chernobyl in 1986, and Fukushima Dai-Ichi in 2011). Baseline monitoring of ^{137}Cs in the landscape is used to assess the present health risk from gamma (γ)-radiation emitted following ^{137}Cs decay and to monitor for new releases of ^{137}Cs. With a half life of 30.17 years, ^{137}Cs has a long residence in the environment and is a health hazard if found in sufficient concentrations.

Furthermore, on Sunday, 26 December 2004, the 2004 Indian Ocean earthquake was an undersea megathrust earthquake, with an epicentre off the west coast of Sumatra, Indonesia. The resulting tsunami hit Thailand's southwestern coast along the Andaman Sea, causing death and destruction. The hardest hit areas in terms of loss of life and property destruction were in Phuket, Phang-Nga and Krabi provinces because they were the most developed and the most densely populated areas along the coast. Much of Phuket province’s west coast was heavily damaged by the tsunami, and most homes, hotels, restaurants and other structures on low ground required significant repairs or rebuilding. Some areas, including Khao Lak in Phang-Nga province, were almost completely wiped out by the waves. Beyond the heavy toll on human lives, the Indian Ocean earthquake has caused an enormous environmental impact that will affect the region for many years to come [13-14]. It has been reported that severe damage has influence on ecosystems such as mangroves, coral reefs, forests, coastal wetlands, vegetation, sand dunes and rock formations, animal and plant biodiversity and groundwater. In addition, the spread of solid and liquid waste and industrial chemicals, water pollution and the destruction of sewage collectors and treatment plants threaten the environment even further, in uncountable ways. The environmental impact will take a long time and significant resources to evaluate [15].

For this reason, the area in the southwestern coast of Thailand or the Andaman sea coast (Phuket, Phang-Nga and Krabi provinces) should be the best place to study and evaluate the specific activities of NORM/TENORM (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic radionuclides (^{137}Cs). This paper reports the range and mean values of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic radionuclides (^{137}Cs) in surface soil samples collected from three provinces (Phuket, Phang-Nga and Krabi) along the Andaman sea coast in the southern region of Thailand. By using the resulting data, radiological hazard could be studied and evaluated for the investigated area and compared to national and global radioactivity measurement and evaluations. Moreover, the radioactive contour maps in the monitored area could be also created and discussed.

2. Experimental

2.1 Sample collection and preparation

Surface soil samples (314 samples) were collected (from a ground surface down to 10-15 cm in depth) from 19 districts of Phuket (3), Phang-Nga (8) and Krabi (8) provinces in the southern region of Thailand. The geographical locations of the study areas were determined using the geographical positioning system (GPS) and located between latitude 7^{o} 28’ and 9^{o} 21’ N and longitude 98^{o} 12’ and 99^{o} 24’ E. After collection, each sample was dried up at room temperature and sieved through a 2 mm mesh-sized sieve to remove stones, pebbles and other macro-impurities. All samples were oven dried at a temperature of 100 $^{\circ}$C for 3 hours before the analysis to remove the moisture. The homogenized sample was placed in a 290 cm^3 PVC containers. The container was sealed hermitically and externally using a cellophane tape and kept aside for about a month to ensure equilibrium between ^{226}Ra and its daughters before being taken for gamma spectrometric analysis.

2.2 Measurement and analysis

The specific activities of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic(^{137}Cs) radionuclides in all surface soil samples were determined by employing a high-purity germanium detector (HPGe, CANBERRA Model GC 2018) and gamma spectrometry analysis system at Nuclear and Material Physics Laboratory, Department of Physics, Faculty of Science, Thaksin University. The detector was enclosed in a massive 10 cm thick lead shielding. Gamma ray radioactive standard sources ^{60}Co, ^{137}Cs and ^{133}Ba were used to calibrate the measurement system up to about 2 MeV. Geometric efficiency for beach sand matrices in the container was determined by KCl, IAEA/RGU-1, IAEA/RGTh-1 and IAEA/SL-2 reference materials (International Atomic Energy Agency IAEA, Vienna, Austria). The spectra were analyzed using the program GENIE 2000. The specific activity of ^{40}K was determined from its 1460.8 keV γ-line. The specific activities of ^{226}Ra and ^{232}Th were determined by their decay products ^{214}Pb (351.9 keV) and ^{208}Tl (583.2 keV), respectively. The specific activity of ^{137}Cs was also determined from its 661.7 keV γ-line. The counting time interval was 10,800 s. The background spectrum was recorded immediately after or before the sample counting.

2.3 Equations and formulae

By using the specific activity values of ^{40}K, ^{226}Ra, and ^{232}Th from this study, the absorbed dose rate (D) in outdoor air at 1 m above the ground was calculated using the conversion factors published in [7] and is given below

$$D = 0.461C_{Ra} + 0.623C_{Th} + 0.0414C_{K}, \quad (1)$$

where C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively. Furthermore, the radium equivalent activity (Ra_{eq}) was calculated through the following relationship [5, 16]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \quad (2)$$

Moreover, the external hazard index (H_{ex}) was also evaluated by using the equation which was defined as [5]:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_{K}/4810 \leq 1 \quad (3)$$

Using the absorbed dose rates in air (D) obtained from the specific activities of natural nuclides in beach sand, adopting the conversion factor of 0.7 Sv/Gy to convert from the absorbed dose in air to the effective dose received by adults and considering that people in Thailand, on average, spend approximately 20% of their time outdoors, the annual effective dose rate (AED_{out}) were calculated using the following equation [6 -7]:

$$AED_{out}\ (mSv/y) = D\ (nGy/h) \times 8760\ h \times 0.2 \times 0.7\ (Sv/Gy) \times 10^{-6} \quad (4)$$

All four calculated and average values were evaluated and compared with the recommended values proposed by UNSCEAR [1-3].

3. Results and discussion

3.1 Specific activities of radionuclides in southern Thailand

Specific activities of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic (^{137}Cs) radionuclides in 314 surface soil samples collected from the investigated areas could be analysed and evaluated from the measured gamma ray energy spectrum. Hence, the ranges and average values of those specific activities were also calculated and shown in table 1.

Table 1. Ranges and average values of specific activities of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs in 314 surface soil samples collected from Phuket, Phang-Nga and Krabi provinces (Thailand).

Locations	Range and Average Values of Specific Activity (Bq/kg)			
	^{40}K	^{226}Ra	^{232}Th	^{137}Cs
Phuket Province (103)	202.30 – 18803.28[a] 4896.45 ± 288.74[b]	24.12 – 830.82 240.11 ± 10.79	20.23 – 1014.21 210.68 ± 9.89	2.09 – 11.26 5.91 ± 2.94
Phang-Nga Province (97)	251.50 – 15740.34[a] 2879.87 ± 225.50[b]	15.21 – 791.42 165.44 ± 8.44	18.14 – 854.34 160.11 ± 7.92	1.48 – 16.91 5.76 ± 2.49
Krabi Province (114)	120.54 – 6586.33[a] 802.58 ± 115.25[b]	3.05 – 930.59 65.75 ± 4.95	4.83 – 194.07 40.69 ± 3.96	0.95 – 11.05 2.96 ± 1.59
Ranges Average Values	120.54 – 18803.28[a] 2859.63 ± 209.83[b]	3.05 – 930.59 157.10 ± 8.06	4.83 – 1014.21 137.16 ±7.26	0.95 – 16.91 4.88 ± 2.34

[a] Ranges
[b] Average values

From the result in table 1, the ranges and average values of specific activities of all radionuclides in surface soil samples collected from Phuket province were the highest values compared to Phang-Nga and Krabi provinces.

3.2 Comparison of the specific activities to research data in the South of Thailand and worldwide means

The average values of specific activities of natural and anthropogenic radionuclides in 314 surface soil samples collected from the inquired area were compared to data from other research reports, the national and global radioactivity measurements as shown in table 2. The national radioactivity data for Southern Thailand were previously reported by Office of Atoms for Peace, Thailand [23]. The global data were reported by UNSCEAR [3].

Table 2. Comparison of the average value of specific activities of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs in 314 surface soil samples collected from Phuket, Phang-Nga and Krabi provinces (Thailand) with research data in the South of Thailand and worldwide mean.

Data Sources	Specific Activities (Bq/kg)			
	^{40}K	^{226}Ra	^{232}Th	^{137}Cs
Songkhla [17]	3562.14 ± 223.56	107.28 ± 6.74	51.53 ± 5.02	3.05 ± 1.58
Nakhon Si Thammarat [18]	4313.18 ± 148.16	108.55 ± 31.19	73.95 ± 4.15	4.84 ± 2.59
Phatthalung [19]	3573.35 ± 203.89	135.89 ± 6.71	76.34 ± 5.32	4.00 ± 1.92
Chumphon [20]	2135.69 ± 168.87	57.32 ± 5.19	56.98 ± 4.68	2.30 ± 1.38
Surat Thani [21]	2119.10 ± 171.72	75.72 ± 5.75	47.39 ± 4.76	3.85 ± 1.67
Yala [22]	3607.70 ± 235.48	128.94 ± 7.42	85.93 ± 6.13	4.53 ± 2.24
Phuket	4896.45 ± 288.74	240.11 ± 10.79	210.68 ± 9.89	5.91 ± 2.94
Phang-Nga	2879.87 ± 225.50	165.44 ± 8.44	160.11 ± 7.92	5.76 ± 2.49
Krabi	802.58 ± 115.25	65.75 ± 4.95	40.69 ± 3.96	2.96 ± 1.59
South of Thailand [23]	511.04 ± 7.04	171.55 ± 3.13	211.19 ± 1.98	1.13 ± 0.49
Worldwide Mean [3]	400	35	30	-

From the result in table 2, the average specific activity of ^{40}K in surface soil samples collected from Phuket province was the highest but the average from Krabi province was the lowest compared to Songkhla, Nakhon Si Thammarat, Phatthalung, Chumphon, Surat Thani and Yala provinces. The average specific activities of ^{226}Ra, ^{232}Th and ^{137}Cs in surface soil samples collected from Phuket and

Phang-Nga provinces were higher than in Songkhla, Nakhon Si Thammarat, Phatthalung, Chumphon, Surat Thani and Yala provinces. Furthermore, the average specific activities of ^{226}Ra, ^{232}Th and ^{137}Cs in surface soil samples collected from Krabi province were comparable with all data of Songkhla, Nakhon Si Thammarat, Phatthalung, Chumphon, Surat Thani and Yala provinces. We could also see that the average specific activities of ^{40}K in Phuket, Phang-Nga and Krabi provinces were always higher than the research data from the south of Thailand and worldwide mean. Moreover, the average specific activities of ^{226}Ra and ^{232}Th were lower than the research data from the south of Thailand (except only the average specific activity of ^{226}Ra in Phuket province which was higher) but higher than the worldwide means. The average specific activity of ^{137}Cs in surface soil samples collected from all three investigated provinces were higher than the research data from the south of Thailand, previously reported.

3.3 Radiological hazard evaluation and comparison

By using the specific activity values of ^{40}K, ^{226}Ra and ^{232}Th as shown in table 1 and table 2, the gamma absorbed dose rate in air (D), the radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}) and the annual effective dose rate (AED_{out}) could be evaluated by using equations (1) to (4), compared to UNSCEAR data and presented in table 3.

Table 3. Gamma absorbed dose rate in air, radium equivalent activity, external hazard index, and annual effective dose rate of 314 surface soil samples collected from Phuket, Phang-Nga and Krabi provinces (Thailand).

Locations	D (nGy/h)	Ra_{eq} (Bq/kg)	H_{ex}	AED_{out} (mSv/y)
Phuket	52.12 – 1793.31	113.13 – 3728.99	0.31 – 10.07	0.06 – 2.20
(103)	446.92 ± 23.19	922.99 ± 47.37	2.49 ± 0.13	0.55 ± 0.03
Phang-Nga	35.84 – 1319.88	75.07 – 2709.96	0.20 – 7.32	0.04 – 1.62
(97)	295.24 ± 18.16	616.14 ± 37.13	1.66 ± 0.10	0.36 ± 0.02
Krabi	16.81 – 575.97	33.83 – 1249.45	0.09 – 3.38	0.02 – 0.71
(114)	89.16 ± 9.53	186.35 ± 19.50	0.50 ± 0.05	0.11 ± 0.01
Ranges	16.81 – 1793.31	33.83 – 3728.99	0.09 – 10.07	0.02 – 2.20
Average values	277.11 ± 16.96	575.16 ± 34.67	1.55 ± 0.09	0.34 ± 0.02
UNSCEAR [1-3]	55	370	1	0.48

From table 3, we saw that the average values of all four radiological hazard indices in Phuket province were higher than the UNSCEAR recommended values. In Phang-Nga province, the gamma absorbed dose rates in air (D), the radium equivalent activity (Ra_{eq}) and the external hazard index (H_{ex}) were higher than those recommended values but the annual effective dose rate (AED_{out}) in this area was lower. There was only one radiological index which was the absorbed dose rates in air in Krabi province that was higher than the referenced value. Furthermore, the average values of the gamma absorbed dose rates in air, the radium equivalent activity and the external hazard index in all three provinces were 5.04, 1.55 and 1.55 times higher than the UNSCEAR recommended values. Moreover, the calculated annual effective dose rate with average value 0.34 mSv/y was lower than the worldwide average value (0.48 mSv/y).

3.4 Contour maps of radiological hazard indices

Furthermore, the contour maps of all four radiological hazard indices which were the gamma absorbed dose rates in air (D), the radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}) and the annual effective dose rate (AED_{out}) from 314 surface soil samples collected from Phuket, Phang-Nga and Krabi provinces (Thailand) were constructed by using a computer program and shown in figure 1 - 4.

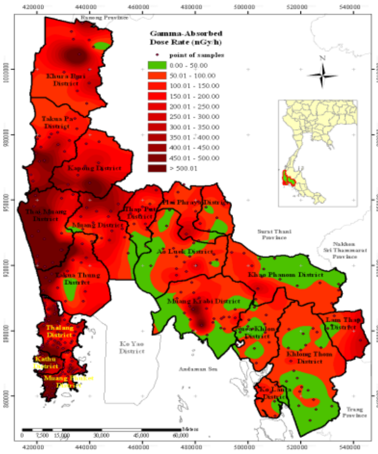

Figure 1. The contour map of the absorbed dose rates in air (D) from 314 surface soil samples collected from Phuket, Phang-Nga and Krabi provinces (Thailand).

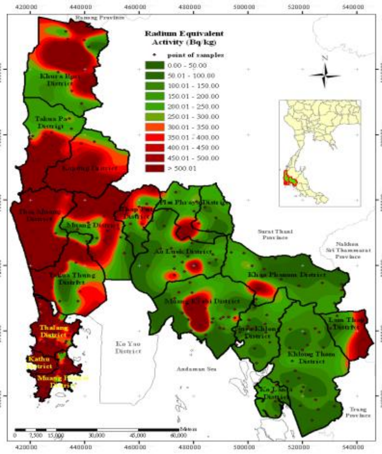

Figure 2. The contour map of the radium equivalent activity (Ra_{eq}) from 314 surface soil samples collected from Phuket, Phang-Nga and Krabi provinces (Thailand).

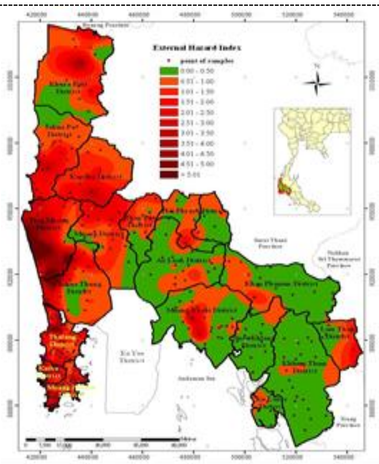

Figure 3. The contour map of the external hazard index (H_{ex}) from 314 surface soil samples collected from Phuket, Phang-Nga and Krabi provinces (Thailand).

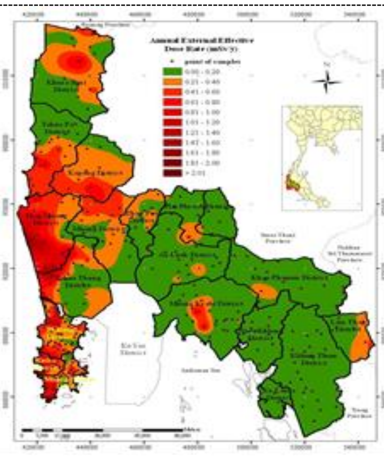

Figure 4. The contour map of the annual effective dose rate (AED_{out}) from 314 surface soil samples collected from Phuket, Phang-Nga and Krabi provinces (Thailand).

4. Conclusion

Specific activities of natural (^{40}K, ^{226}Ra, ^{232}Th) and anthropogenic (^{137}Cs) radionuclides in 314 surface soil samples collected from three provinces (Phuket, Phang-Nga and Krabi) along the Andaman sea coast in southern region of Thailand ranged from 120.54 – 18803.28, 3.05 – 930.59, 4.83 – 1014.21 and 0.95 – 16.91 Bq/kg with the average values of 2859.63 ± 209.83, 157.10 ± 8.06, 137.16 ±7.26 and 4.88 ± 2.34 Bq/kg, respectively. Furthermore, four radiological hazard indices which were the gamma absorbed dose rate in air (D), the radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}), and the annual effective dose rate (AED_{out}) in the monitoring areas were evaluated to be 277.11 ± 16.96 nGy/h, 575.16 ± 34.67 Bq/kg, 1.55 ± 0.09 and 0.34 ± 0.02 mSv/y, respectively. It was found that the average value of the gamma absorbed dose rate in air obtained in this study (277.11 nGy/h) was 5.04 times higher than the global average (55 nGy/h). The average value of the radium equivalent activity (575.16 Bq/kg) was 1.55 times higher than 370 Bq/kg, which was acceptable for safe use [1-3]. The average values of the external hazard index obtained in this study was found to be 1.55 which was higher than unity. We could see that three out of four radiological hazard indices were greater than the values recommended by UNSCEAR [1-3], especially in Phuket and Phang-Nga provinces. This indicated a possibility that the radiation hazard might be a concern for the population living in the investigated area. According to the calculations, the average annual effective dose rate in the area was 0.34 mSv/y, which was still lower than the worldwide average value (0.48 mSv/y). For this reason, the radiation hazard was considered insignificant for the population living in the studied area. Nonetheless, from the contour maps, we could see that populations living in the northern and western parts, especially in Phang-Nga and Phuket provinces, could be exposed to a higher background radioactivity than those in the central and eastern regions of the investigated area.

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