Specific activities and radiological hazard assessment in beach sand samples in Songkhla province, Thailand after Fukushima Dai-Ichi nuclear power plant accident in Japan

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Abstract. According to the Fukushima Dai-Ichi Nuclear Power Plant Accident in Japan on March 11, 2011, specific activities of natural (40K, 226Ra and 232Th) and anthropogenic (137Cs) radionuclides in 210 beach sand samples which were collected from Maharat, Sai Kaew, Samila, Chalatat, Na Thab, Sakom and Soi Sawan beaches in Songkhla Province along the eastern coast of Thailand, have been studied and evaluated. The specific activity levels of 40K, 226Ra, 232Th and 137Cs in the area were found in the range 110.13 – 4574.12, 6.99 – 451.87, 3.75 – 665.76 and 0.58 – 9.91 Bq/Kg, respectively. Furthermore, the median values of specific activities of 40K, 226Ra, 232Th and 137Cs were calculated and compared with other research data in Thailand as well as global measurements and evaluations. Moreover, four radiological hazard indices for the studied area were also assessed by using the median values of specific activities of 40K, 226Ra and 232Th.

1. Introduction
The Great Tohoku earthquake that occurred off the Japanese northeast coast on March 11, 2011 and the resulting tsunami that damaged the Fukushima Dai-ichi nuclear power plant (FDNPP) resulted in several explosions at the reactors between March 12 and 15. These explosions led to significant releases of radionuclides into the atmosphere [1]. The famous nuclear power plant accident was obviously occurred with a broader disaster. This accident resulted in a release of radioactive materials into the atmosphere and marine environment and evacuation of residents living near the nuclear power plants. This accident has raised concerns around the world about the safety of nuclear power generation [2]. Hence, several studies around the world have been performed to evaluate the specific activities of some anthropogenic radionuclides after the FDNPP accident [3–8]. The radioactivity monitoring result suggests that the Fukushima-derived radionuclides spread in the north part of the Northern Hemisphere atmosphere [9, 10]. In ASEAN countries, Vietnam is the first country which interested in studying the effect from the FDNPP disaster. Radionuclides from the reactor accident at the FDNPP were observed in the surface air at stations in Hanoi, Dalat, and Ho Chi Minh City (HCMC) in Vietnam, about 4500 km southwest of Japan, during the period from March 27 to April 22, 2011 [4]. No studies to date in Thailand were performed and measured the radioactivity in some environment samples from dry deposition of radionuclides from the FDNPP accident.
Naturally Occurring Radioactive Materials (NORM) and Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) consist of materials, usually industrial wastes or by-products enriched with radioactive elements found in the environment, such as uranium, thorium and potassium and any of their decay products. Since 1970s, NORM and TENORM have been attracted to scientists and researchers around the world to monitor and measure the level of radioactivity in both physical and biological environment samples for evaluating the radiation dose for people who use or live in the study area. Besides, there are many scientists and researchers in several countries have studied the levels of natural background radiation by in situ measurements or by analysis of specific activities of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in surface soil or beach sand samples [11–15]. Moreover, oil and gas industry areas should be monitored and evaluated to NORM and TENORM concentration.

Songkhla province is known for its commercial, agro industrial and socio-economic activities in the southern region of Thailand. Majority of the social, economic, and industrial activities in the south of Thailand takes place in this province. Located about 950 kilometers from Bangkok, Songkhla province, with a population of about 1.4 million, is a major seaside province, connecting with Kedah State of Malaysia. Hat Yai district is the best-known business center in the southern region of Thailand. It plays an important role in the southern economy and serves as the junction of communication and a gateway welcoming Malaysian and Singaporean tourists. Consequently, Songkhla is regarded as the capital province of the south of Thailand. The province is strategically located along the eastern coast in the lower southern region of Thailand and also connected to the Gulf of Thailand. There are many activities of oil and gas industry for Thailand kingdom in the Gulf of Thailand. According to the connection with the Pacific Ocean, the Gulf of Thailand might have accumulated some of the radioactive contaminations from the FDNPP accident in Japan. As a result, Songkhla province, especially in some popular beaches for Thai and foreign tourists, should be the best location for NORM/ TENORM and fissionable materials evaluation in the southern region of Thailand.

In response to the FDNPP accident, the 2015 research project to study the increasing of natural and anthropogenic radionuclides in some attractive beaches along the eastern coast of Thailand in Songkhla province have been initiated and performed by collaboration of Thaksin University (TSU), Songkhla Rajabhat University (SRU) and Office of Atoms for Peace (OAP).

This present paper presents the specific activities values of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in 210 beach sands samples collected from seven beaches (Maharat, Sai Kaew, Samila, Chalatat, Na Thab, Sakom and Soi Sawan beaches) which are famous and beautiful beaches in Songkhla province along the eastern coast and in the southern region of Thailand. Furthermore, the distributions and the appropriate medium values of the specific activities and radiological hazard indices for the investigated area were also studied and evaluated. Moreover, the results were also compared with the Office of Atoms for Peace (OAP) annual report data, Thailand and global radioactivity measurement and evaluations.

2. Experimental

2.1. Sample collection and preparation

All of surface beach sand samples (210) were collected from Maharat beach (30) in Sathing Phra district, Sai Kaew beach (30) in Singhanakkhon district, Samila (30) and Chalatat Beaches (30) in Muang Songkhla district, Na Thab beach (30) in Chana district, Sakom (30) and Soi Sawan beaches (30) in Thepha district along the eastern coast of Songkhla province in the south of Thailand. Each sample was dried up at room temperature after collection and sieved through a 2 mm mesh-sized sieve to remove stone, pebbles and other macro-impurities. Before the measurement and analysis, all beach samples were oven dried at a temperature of 100°C for 3 hours for removing moisture. The homogenized sample was placed in a PVC cylindrical container. The container was sealed hermatically and externally using a cellophane tape and kept aside for about a month to ensure equilibrium between $^{226}$Ra and its daughters and $^{228}$Ra and its daughters before being taken for measurement and analysis by using gamma spectrometry technique.
2.2. Measurement and analysis

The specific activities of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in all 210 beach sand 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 Research Unit (NuMPRU), Department of Physics, Faculty of Science, Thaksin University, Songkhla Campus. The detector was enclosed in a massive 10 cm thick lead shielding. Geometric efficiency for beach sand matrices in the container was determined by the KCL, IAEA/RGU-1 and IAEA/RGTh-1 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 146 keV $\gamma$-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 $\gamma$-line. The statistic computer program was employed to analyze the frequency distribution of specific activities of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in all surface beach sand samples for the investigated area. Furthermore, the results were also compared with some research data in Thailand as well as global measurements and evaluations. Moreover, four radiological hazard indices in the study area were also evaluated by using the appropriate medium values of the frequency distribution and the equations as shown in the following section.

2.3. Equations and formulas

The absorbed dose rate (D) in outdoor air at 1 m above the ground was calculated by using the conversion factors published in [16] with all medium values of specific activity of $^{40}$K, $^{226}$Ra, and $^{232}$Th from this study and is given below

$$D = 0.461 C_{Ra} + 0.623 C_{Th} + 0.0414 C_{K}$$

where $C_{Ra}$, $C_{Th}$ and $C_{K}$ are the medium values of specific activity of $^{226}$Ra, $^{232}$Th and $^{40}$K in Bq/kg, respectively. Furthermore, the radium equivalent activity ($R_{eq}$) was calculated through the following relationship [17, 18]:

$$R_{eq} = C_{Ra} + 1.43 C_{Th} + 0.077 C_{K}$$

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

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

The annual effective dose rate ($AED_{out}$) were calculated using the absorbed dose rates in air (D) obtained from the medium values of specific activity of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) radionuclides in beach sand samples, 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 [12, 16]:

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

All four radiological hazard indices and their average values were calculated and compared with the recommended values reported by UNSCEAR [19-21].
3. Results and Discussion

3.1. Specific activity values and ranges of natural and anthropogenic radionuclides

Specific Activity Values and Ranges of natural \(^{40}\text{K}, \ ^{226}\text{Ra}, \ ^{232}\text{Th}\) and anthropogenic \(^{137}\text{Cs}\) radionuclides in 210 beach sand samples collected from Maharat (30), Sai Kaew (30), Samila (30), Chalatat (30), Na Thab (30), Sakom (30) and Soi Sawan (30) beaches in Songkhla Province was measured and calculated by using HPGe detector and gamma spectrometry technique. All results were presented in table 1.

Table 1. Specific Activity Values and Ranges of \(^{40}\text{K}, \ ^{226}\text{Ra}, \ ^{232}\text{Th}\) and \(^{137}\text{Cs}\) in Bq/kg in 210 beach sand samples for Maharat, Sai Kaew, Samila, Chalatat, Na Thab, Sakom and Soi Sawan beaches in Songkhla Province.

| Study Areas          | \(^{40}\text{K}\) Values (Bq/kg) | \(^{226}\text{Ra}\) Values (Bq/kg) | \(^{232}\text{Th}\) Values (Bq/kg) | \(^{137}\text{Cs}\) Values |
|----------------------|-----------------------------------|------------------------------------|-----------------------------------|---------------------------|
| Maharat beach (30)   | 648.99 – 1508.70                  | 18.85 – 111.51                     | 14.60 – 122.94                    | N.A.                      |
| Sai Kaew beach (30)   | 145.51 – 1154.30                  | 6.99\(^a\) – 74.53                | 9.68 – 83.99                      | 0.79 - 2.31               |
| Samila beach (30)     | 384.56 – 4574.12\(^b\)            | 14.09 – 433.53                     | 9.13 – 448.78                     | N.A.                      |
| Chalatat beach (30)   | 167.85 – 1904.85                  | 7.85 – 451.87\(^b\)               | 13.45 – 665.76\(^b\)             | 0.58\(^a\) - 4.52         |
| Na Thab beach (30)    | 110.13\(^a\) – 2307.65            | 9.25 – 258.87                      | 11.68 – 235.12                    | 1.28 - 4.91               |
| Sakom beach (30)      | 122.06 – 1925.11                  | 12.97 – 120.74                     | 6.47 – 179.63                     | 1.18 - 9.91\(^b\)         |
| Soi Sawan beach (30)  | 512.00 – 3955.88                  | 20.11 – 83.18                      | 3.75\(^a\) – 94.68               | 1.77 - 9.30               |

Ranges (210)          | 110.13 - 4574.12                  | 6.99 - 451.87                      | 3.75 - 665.76                     | 0.58 - 9.91               |

\(^a\) Minimum values \(^b\) Maximum values

3.2. Frequency distributions of specific activities

The frequency distribution of specific activities of \(^{40}\text{K}, \ ^{226}\text{Ra}, \ ^{232}\text{Th}\) and \(^{137}\text{Cs}\) in 210 beach sand samples collected from all seven beaches in Songkhla province (Thailand), were studied and analyzed by using a statistical computer program. The results of Chalatat, Sakom and Soi Sawan beaches were chosen and presented in the following figure 1– figure 12. It was found that the frequency distribution of specific activities of \(^{40}\text{K}, \ ^{226}\text{Ra}, \ ^{232}\text{Th}\) and \(^{137}\text{Cs}\) in all beach sand samples collected from Songkhla province, were asymmetrical distribution.

3.3. Median value of specific activities

According to the asymmetry of frequency distributions of \(^{40}\text{K}, \ ^{226}\text{Ra}, \ ^{232}\text{Th}\) and \(^{137}\text{Cs}\) in all (210) beach sand samples as shown in figure 1– figure 12, the median values should be selected and used for the corresponding radiological hazard evaluation in the investigated area. All of these results were also compared with the Office of Atoms for Peace (OAP) annual report data, Thailand and global radioactivity measurements and evaluations as presented in table 2.

\[\text{Figure 1 - Figure 12}\]
3.4. Radiological hazard assessment

Furthermore, four radiological hazard indices which are gamma absorbed dose rate (D), radium equivalent activity (Ra\textsubscript{eq}), external hazard index (H\textsubscript{ex}) and annual external effective dose rate (AED\textsubscript{out}), were also evaluated and presented in table 2 for Maharat, Sai Kaew, Samila, Chalatat, Na Thab, Sakom and Soi Sawan beaches in Songkhla Province by using the median values of specific activities of \(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th. Moreover, the results were also compared with some research data in Thailand and UNSCEAR as shown in the same table.
The ranges of specific activities of natural (\(^{40}\text{K}, \ ^{226}\text{Ra}, \ ^{232}\text{Th}\)) and anthropogenic (\(^{137}\text{Cs}\)) radionuclides in 210 beach sand samples collected from Maharat, Sai Kaew, Samila, Chalatat, Na Thab, Sakom and Soi Sawan beaches in Songkhla Province were 110.13 – 4574.12 Bq/kg for \(^{40}\text{K}, 6.99 – 451.87\) Bq/kg for \(^{226}\text{Ra}, 3.75 – 665.76\) Bq/kg for \(^{232}\text{Th}\) and 0.58 – 9.91 Bq/kg for \(^{137}\text{Cs}\) as shown in table 1. The frequency distribution of specific activities of \(^{40}\text{K}, ^{226}\text{Ra}, ^{232}\text{Th}\) and \(^{137}\text{Cs}\) in beach sand samples in every location were studied and found to be asymmetrical distribution as shown in figure 1 – figure 12. Hence, the median values analyzed from all frequency distributions, were chosen to evaluate the radiological hazard indices for the investigated area. Furthermore, specific activities of \(^{40}\text{K}\) in all beaches were mostly higher than some research in Thailand, OAP research data and worldwide mean but lower than in Phuket and Phang Nga province. The specific activities of \(^{226}\text{Ra}\) and \(^{232}\text{Th}\) were in the same order of research data in Thailand and worldwide mean but lower than OAP research data. Moreover, specific activities of \(^{137}\text{Cs}\) in the investigated area were in the same range of all research data in Thailand and OAP research data as presented in table 2. According to Songkhla province is one of the province which is located in the eastern coast of Thailand and connected to the Pacific Ocean. The study area, Maharat, Sai Kaew, Samila, Chalatat, Na Thab, Sakom and Soi Sawan beaches which are located on the eastern coast of Songkhla province, certainly have some experiences of NORM and TENORM activities e.g. oil and gas industry wastes and FDNNP accident. For this reason, the values of gamma absorbed dose rate (D) in all beaches were higher than about 2.9 – 6.6 times to some research data in Thailand and UNSCEAR but mainly lower than in Patong and Naiyang beaches in Phuket province. The values of radium equivalent activity in all beaches were higher than about 2.7 – 6.2 times to some research data in Thailand but mostly lower than in Patong and Naiyang beaches in Phuket province which is less than 370 Bq/kg, which are the acceptable value for safe use. The values of external hazard index obtained in this study were ranged from 0.27 ± 0.01 to 0.61 ± 0.06.

### Table 2. Comparison of median values of specific activity of \(^{40}\text{K}, ^{226}\text{Ra}, ^{232}\text{Th}\) and \(^{137}\text{Cs}\) in Bq/kg and radiological hazard indices in 7 beach sand samples in Songkhla province with OAP annual report data, Thailand and global radioactivity measurement and evaluations.

| Locations                        | Specific Activity (Bq/kg) | Radiological Hazard Indices |
|----------------------------------|---------------------------|-----------------------------|
|                                  | \(^{40}\text{K}\) | \(^{226}\text{Ra}\) | \(^{232}\text{Th}\) | \(^{137}\text{Cs}\) | D (mSv/h) | Ra\(_{eq}\) (Bq/kg) | IL\(_{eq}\) (mSV/yr) |
| Patong beach (Phuket province)   | 3538.09 ± 959.56         | 29.72 ± 11.48              | 32.71 ± 7.51             | 3.28 ± 1.03             | 180.56 ± 49.70 | 348.93 ± 96.11 | 0.94 ± 0.26 |
| Naiyang beach (Phuket province)  | 1648.27 ± 702.98         | 14.62 ± 6.54               | 18.10 ± 4.22             | 1.19 ± 0.66             | 86.25 ± 34.75  | 167.42 ± 66.70 | 0.45 ± 0.18 |
| Takina Pa and Thai Muang beach   | 1069.99 ± 119.43         | 40.39 ± 4.30               | 41.15 ± 3.65             | N.A.                    | 88.55 ± 9.20  | 181.62 ± 18.71 | 0.49 ± 0.05 |
| (Phang Nga province)             |                           |                             |                         |                        |               |                 | 0.11 ± 0.01 |
| Ao Nang beach (Krabi province)   | 330.73 ± 40.00           | 4.60                        | 5.78                     | N.A.                    | 19.41         | 38.33            | 0.10         | 0.02 |
| Nopparat Thara beach             | 307.60 ± 53.11           | 5.31                        | 5.28                     | N.A.                    | 18.47         | 36.55            | 0.10         | 0.02 |
| Pakmeng beach (Trang province)   | 57.3                      | 23.1                        | 6.9                      | N.A.                    | 17.32         | 37.38            | 0.10         | 0.02 |
| Chaweng beach (Sarut Thani province) | 373.30 ± 188.55       | 18.85                        | 23.53                    | N.A.                    | 38.80         | 81.24            | 0.22         | 0.05 |
| Maharat beach (Songkhla province) | 895.97 ± 122.52         | 39.28 ± 3.27               | 42.06 ± 2.34             | N.A.                    | 81.40 ± 8.04  | 168.42 ± 16.05 | 0.45 ± 0.04 |
| Sai Kaew beach                    | 726.91 ± 115.41          | 21.79 ± 2.50               | 15.69 ± 1.72             | 1.52 ± 0.47             | 49.91 ± 7.00  | 100.20 ± 13.85 | 0.27 ± 0.01 |
| Samila beach                     | 868.51 ± 118.42          | 30.50 ± 4.22               | 23.31 ± 2.22             | N.A.                    | 64.54 ± 8.33  | 130.71 ± 16.51 | 0.35 ± 0.04 |
| Chalatat beach (Songkhla province) | 488.05 ± 105.21         | 28.58 ± 3.84               | 30.11 ± 2.06             | 1.90 ± 0.65             | 52.14 ± 7.41  | 109.21 ± 14.89 | 0.29 ± 0.04 |
| Na Thab beach* (Songkhla province) | 1044.54 ± 122.25       | 23.97 ± 3.02               | 23.76 ± 2.04             | 2.04 ± 1.17             | 69.10 ± 7.72  | 138.38 ± 15.34 | 0.37 ± 0.04 |
| Sakom beach*                     | 1028.92 ± 130.68         | 31.53 ± 3.18               | 24.98 ± 2.00             | 3.17 ± 0.95             | 72.69 ± 8.12  | 146.47 ± 16.09 | 0.40 ± 0.04 |
| Soi Sawan beach* (Songkhla province) | 1869.32 ± 183.33   | 36.73 ± 3.88               | 31.61 ± 2.68             | 4.00 ± 1.10             | 114.02 ± 11.05 | 225.87 ± 21.83 | 0.61 ± 0.06 |
| OAP (Southern region of Thailand[22]) | 511.04 ± 7.04       | 171.55 ± 3.13              | 211.19 ± 1.98            | 1.13 ± 0.49             | N.A.          | N.A.            | N.A.        |
| Worldwide mean [21]              | 400                       | 35                          | 30                        | N.A.                    | N.A.          | N.A.            | N.A.        |
| UNSCEAR [19,21]                  | N.A.                      | N.A.                        | N.A.                     | N.A.                    | 55            | 370.1           | 0.48        |

*Present study
which were less than unity. The calculated annual effective dose rate for all beaches were also ranged from 0.06 ± 0.01 to 0.14 ± 0.01 mSv/y which lower than the worldwide average value of 0.48 mSv/y as presented by UNSCEAR.

4. Conclusions
The median values of specific activity of natural radionuclides ($^{40}$K, $^{226}$Ra, $^{232}$Th) in 210 beach sand samples collected from the monitored area (Maharat, Sai Kaew, Samila, Chalatat, Na Thab, Sakom and Soi Sawan beaches in Songkhla province) are at the typical level of radioactivity from natural background radiation. Furthermore, the median values of specific activity of $^{137}$Cs in the study area were in the same level of research data in Thailand and OAP after the Fukushima Dai-ichi nuclear power plant accident. Moreover, there are three (Ra$_{eq}$, H$_{eq}$ and AED$_{out}$) from four values of radiological hazard indices were lower than the recommend values which was reported by UNSCEAR. This means that the FDNPP accident was insignificant for the population living and spending sometimes in the investigated area. The results agree with the discussion “the effect of this radioactive cloud from FDNPP accident on low latitude areas seems to be rather minor [23].

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